

Interactive comment on “On the ice nucleation spectrum” by D. Barahona

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Thank you for the positive assessment. The comments are addressed below.

1 General Comments

Chapter 4.2: Did you check your model against laboratory data for homogeneous ice nucleation as you did for deposition nucleation?

This would be a good comparison. Unfortunately no data on homogeneous freezing of cloud droplets as a function of the droplet size distribution was found.

From the description of the model for deposition nucleation it was not clear to me whether the surface of an individual particle is

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homogeneous being related with a single contact angle but contact angles could differ from particle to particle or whether the particle is divided into sites of finite surface area each associated with a given contact angle. From my impression I would say the first case is applied. However the valid case should be clarified more clearly.

The NPDF is the distribution of the number of ice germs per particle normalized to a reference state. It is not a distribution of contact angles. Whether ice germs are formed at different rates in different parts of the particle surface or at the same rate in the whole particle is irrelevant, because the NPDF is only concerned with how many there are on the particle. Assumptions on the structure of the surface refer only to the reference state, which by convenience was assumed to be an smooth homogeneous particle described by a single contact angle.

Section 2 have been rewritten to make this clear.

In the abstract, on Page 29619, line 7–14 and Page 29624, line 7–15 you write that low contact angles feature singular and large contact angles feature stochastic nucleation behaviour. Can you confirm this suggestion through calculations for the frozen fraction as function of nucleation time for fixed temperatures? E.g., Broadley et al., ACPD (2011) determined a cooling rate independence on nucleation for droplets containing low surface areas of illite which is consistent with singular description. But additional measurements at a constant temperature clearly feature time dependence being in agreement with the stochastic view on nucleation. They suggest a multi component model that could reconcile both the nucleation measurement at constant temperature with the cooling rate independence.

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This is a very interesting point. What needs to be considered to reconcile the results of Broadley et al., ACPD (2011) with the results presented here is that cooling rate independence at variable S_i (here S_i is used instead of T) and constant nucleation probability does not necessarily imply temporal independence at constant S_i and variable nucleation probability. This is because each time-variable experiment at constant S_i represents a single point in the f_f vs. S_i curve. This is shown in the revised paper where a new section on temporal effects on deposition ice nucleation has been added.

The referred conclusion has been modified accordingly to emphasize that when studied as a function of S_i , efficient IN display characteristics typically associated with singular behavior. This however may not imply a lack of temporal dependency when studied at constant S_i .

In contrast to your conclusion, Niedermeier et al., ACP (2011) showed that a steep increase in the frozen fraction vs. temperature curve features stochastic behaviour while the time dependence is weaker for frozen fraction curves with shallower slopes.

On the contrary, the conclusions of Niedermeier et al. are consistent with this study. The key point is that an steep f_f vs. T profile may or may not imply a lack of temporal dependency at constant T (here S_i is used instead of T). So the conclusion of Niedermeier et al. is valid, but not absolute. Large dispersion in surface properties (high σ_φ) does imply a weaker temporal dependency (since $f_f \sim \frac{\ln \phi}{\sqrt{\pi} \sigma_\varphi}$) and would lead to a shallow f_f vs S_i curve for inefficient IN. For efficient IN however the effects of dispersion are masked by the high sensitivity of f_f to S_i . Thus regardless of σ_φ , the f_f vs. S_i curve is steep, however when analyzed at constant S_i temporal effects on f_f can still be weak if σ_φ is high. This is now thoroughly discussed in the revised paper.

Your theory suggests that ``inferring the aerosol ice nucleation properties from measurement of S_{i_onset} at $f_f = 0.01$ may carry

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significant error.''' and that the determination of Si_{onset} at $f_f = 0.5$ would be appropriate. But for the characterization of the nucleation process I would suggest determining the entire frozen fraction curve ...

Good point. This has been emphasized in the revised paper.

I agree on the statement of referee 2 by saying "in what respect can the formalism presented here be considered as less idealized" compared to existing models? More explanation is necessary to illustrate the fundamental difference to other descriptions.

As mentioned above, details of the microstructure of the particles are not hardwired into the NPDF. I have extended the introduction to better explain the features of current models and rewritten Section 2 including more details on the physical basis behind the theory.

2 Specific Comments

Pages 29605–606, I do not understand the step from Eq. (3) to Eq. (4). Should there be a $d\varphi$ in front...? Is it included in $n(\xi)$? A clarification would be fine.

This is explained by the requirement that the number concentration of particles in each φ class must be equal in the ξ and φ spaces, i.e., $n(\xi)d\xi = n(\varphi)d\varphi$. This is now clarified

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in the paper.

Page 29606, line 19: It could be mentioned that Eq. (4) is also used for the total derivative of Eq. (5)

Equation (6), i.e., the total derivative, has been removed as it is not used further in the paper.

Page 29610, I do not understand the transition from Eq. (18) to (20). Can you explain it in more detail?

To make this more clear, I have introduced a general definition of τ_{nuc} and used it to arrive to Eq. (20) in a more elegant way.

Page 29612, line 14-16: 'Active site' was introduced by Fletcher (1969) to be characterized through $\cos(\theta) = 1$, i.e. $f = 0$, i.e. an energy barrier of zero. How does this fit with your specification ...

This paragraph has been removed. The relation between active sites and nucleation by surface adsorption, including the time-dependent approximation of Fletcher (1969), is discussed in Section 2.

Page 29615, Eq. (35) and (36): I do not understand why Eq. (36) can be written like this with the presented definition of the parameter c .

This involves taking the constants terms out of the functionality in Eq.(34) and keeping only terms involving the ratio $\frac{\theta}{\theta_0}$. The equation has been rederived in a more clear fashion.

Pages 29620-621, the dust paragraph and figure 6: First

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of all, a link to Fig. 6 is not given in the text at all. For comprehensible reasons you did not include data from Welts et al. (2009) and M\"ohler et al. (2006) where $Sw > 1$. Therefore I would suggest marking this region in Fig. 6 where $Sw > 1$ because then condensation/immersion freezing will take place and therefore the freezing behaviour could change compared to your model outputs for that region in the figure.

S_i at water saturation is 1.6 and 1.7 at $T = 223$ K and $T = 210$ K, respectively. These points would fall outside the plotting region. The reference to Figure 6 has been added to the text.

Page 29620, line 9–11: I do not understand the meaning of this sentence, can you please clarify?

It emphasizes that the depicted relation in Fig. 4 does not imply a physical dependency of $\bar{\theta}$ on S_i . The sentence has been clarified.

Page 29607, line 8: delete one ``the'' right before ``homogeneous nucleation rate: ''

Page 29609, line 11–12: Maybe you ought to write:

``Using Table 1 with ...:''

Page 29611, line 8: The temperature cannot be warm or cold, just high or low.

Page 29612, line 2: It is the ice germ radius.

Page 29618, line 19–23: Maybe it is worth mentioning here that Tonset ($ff = 0:01$) does not change with increasing $varr$. Therefore...

All have been corrected.