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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the ice nucleation spectrum" *by* D. Barahona

Anonymous Referee #1

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In this study, a new formulation of the ice nucleation spectrum is presented which based on statistical view of ice nucleation. It is used to generate parameterizations for homogeneous droplet freezing as well as heterogeneous deposition ice nucleation on dust and soot particles. In general, important conclusions and suggestions for further work (experimental and theoretical) are drawn. Therefore, I recommend publication in ACP once the following comments have been addressed.

General comments

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

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

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. 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.

Your theory suggests that "inferring the aerosol ice nucleation properties from measurement of $S_{\text{i,onset}}$ at $f_{\text{f}} = 0.01$ may carry significant error." and that the determination of $S_{\text{i,onset}}$ at $f_{\text{f}} = 0.5$ would be appropriate. But for the characterization of the nucleation process I would suggest determining the entire frozen fraction curve to get information about the mean freezing temperature/ice saturation as well as the spread in the contact angle distribution, etc. You also said that in the conclusion part but I would suggest pointing out that more clearly.

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.

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Interactive Comment

The derivations are not always clear to me. The respecting equations are noted in the "Specific comments" section.

Specific comments

Pages 29605-606, I do not understand the step from Eq. (3) to Eq. (4). Should there be a $\bar{\varphi}$ in front of the exponential term because $d\varphi = \bar{\varphi}\, d\xi$? Is it included in $n(\xi)$? A clarification would be fine.

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

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 $\bar{\varphi} = \bar{v}_{\mathrm{p}} J_{\mathrm{hom}} \tau_{\mathrm{nuc}}$ and making $\alpha = \beta = (\nu + 1)$, we obtain:"

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

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 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 (Δg) of zero. How does this fit with your specification of $\theta < 1$? In general, do you mean: $\theta < 1$ degree, $\theta < 1$ rad or maybe $f < 1$?

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.

Page 29618, line 19-23: Maybe it is worth mentioning here that T_{onset} ($f_{\mathrm{f}} = 0.01$) does not change with increasing varr. Therefore the stretching effect with increasing varr becomes only obvious if the whole f_{f} curve or in this case T_{50} ($f_{\mathrm{f}} = 0.5$) is considered.

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Pages 29620-621, the dust paragraph and figure 6: First of all, a link to Fig. 6 is not given in the text at all. For comprehensible reasons you did not include data from Welti et al. (2009) and Möhler et al. (2006) where $S_w > 1$. Therefore I would suggest marking this region in Fig. 6 where $S_w > 1$ 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.

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

References

Broadley, S. L., Murray, B. J., Herbert, R. J., Atkinson, J. D., Dobbie, S., Condliffe, E., and Neve, L.: Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust, Atmos. Chem. Phys. Discuss., 11, 22801-22856, doi:10.5194/acpd-11-22801-2011, 2011.

Fletcher, N. H.: Active sites and ice crystal nucleation, J. Atmos. Sci., 26(6), 1266–1271, 1969.

Möhler, O., Field, P. R., Connolly, P., Benz, S., Saathoff, H., Schnaiter, M., Wagner, R., Cotton, R., Krämer, M., Mangold, A., and Heymsfield, A. J.: Efficiency of the deposition mode ice nucleation on mineral dust particles, Atmos. Chem. Phys., 6, 3007–3021, doi:10.5194/acp-6-3007-2006, 2006.

Niedermeier, D., Shaw, R. A., Hartmann, S., Wex, H., Clauss, T., Voigtländer, J., and Stratmann, F.: Heterogeneous ice nucleation: exploring the transition from stochastic to singular freezing behavior, Atmos. Chem. Phys., 11, 8767-8775, doi:10.5194/acp-11-8767-2011, 2011.

Welti, A., Lüönd, F., Stetzer, O., and Lohmann, U.: Influence of particle size on the ice nucleating ability of mineral dusts, Atmos. Chem. Phys., 9, 6705–6715,

doi:10.5194/acp-9-6705-2009, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 29601, 2011.