Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2015-969-RC1, 2016

Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Classical nucleation theory of immersion freezing: Sensitivity of contact angle schemes to thermodynamic and kinetic parameters" *by* L. Ickes et al.

Anonymous Referee #1

Received and published: 12 February 2016

General comment:

In order to find an appropriate (CNT-based) parameterization for immersion freezing (induced by mineral dusts) in GCMs, the authors tested various combinations of different descriptions for interfacial free energy σ_w, activation energy $\Delta g^{\#}$ as well as different possibilities to include contact angle (single contact angle, contact angle distribution and temperature dependent contact angle). To do so, the different schemes are fitted against laboratory heterogeneous freezing data for different dusts (spanning several temperature, particle surface and time ranges).

I have to admit that after first reading I have been in two minds about recommending this paper for publication in ACP. On the one hand the authors e.g. vividly demon-

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strate that contact angle values gained for one substance largely depend on the values/parametrizations used for σ_w and $\Delta g^{\#}$ so that contact angle values obtained in different studies for a given substance do not necessarily agree. This is important if these contact angle values are used in GCMs but connected with other σ_w and $\Delta g^{\#}$ values/parameterizations. On the other hand e.g. I doubt the physics behind some of the presented parameterization schemes (see specific comments). However, due to the importance for the heterogeneous ice nucleation community I would recommend this paper for publication after the following comments have been addressed.

Specific comments:

Page 1, line 10-11: I do not understand the meaning of the sentence starting with “We show that additional…”. Please clarify. What is “J_{imm}”?

Page 4, line 12-13: I agree that in case of heterogeneous ice nucleation the freezing curve is shifted to higher T compared to the homogeneous case. But does the curve necessarily has to be less steep? Looking on Fig. A3 in the paper of Hoose and Möhler (2012) it can be seen that the heterogeneous nucleation rate is steeper for higher temperatures (depending on the parameterizations used in CNT). So what causes the heterogeneous freezing curve to be less step compared to the homogeneous case?

Chapter 2.1.3: I have a problem with the interpretation of the temperature dependent contact angle scheme. In a physical sense the contact angle is “determined by the condition of mechanical equilibrium, i.e., there must be no net force component along the solid surface” (Pruppacher and Klett, 1997, P. 136). Due to the decrease of the interfacial free energies with temperature does the contact angle then should decrease with decreasing temperature for a given particle (let’s assume homogeneous surface conditions)? Here, the contact angle increases with decreasing T. I would interpret this behavior in that way that particles with larger contact angles (higher energy barrier) can be activated with decreasing temperature. Is this in agreement to your description?

Chapter 3.1: I have some trouble with those schemes fitting $\Delta g^{\#}$. In lines 27 to 30 you mention that an aerosol type specific $\Delta g^{\#}$ value is physically questionable. On the other hand this statement is reversed on the next page saying that the particle itself might influence the diffusion of water molecules to the ice cluster. I agree that the attachment of water molecules to the ice cluster is influenced due to the presence of the ice nucleating particle i.e., the INP "blocks" water molecules since the ice cluster is just a cap and not spherical (in terms of CNT) as for the homogeneous case. But I think that n_s in CNT takes care of this. Are there any mechanisms which could confirm your hypothesis of aerosol-type specific $\Delta g^{\#}$? Is there a specific term in CNT which would take care of this? Or does $\Delta g^{\#}$ represent here just a fitting parameter without physical meaning?

On page 8, line 19-20 you mention that a single contact angle is not able to represent the experimental results for mineral dust. This has also been shown in e.g. Lüönd et al. (2010), Welti et al. (2012), etc. But is this a general finding? What about other substances like biological particles?

Page 9, line 18-20: I do not understand why this procedure has been performed. Some further explanations here would be great.

Page 10, line 26-27: The curve in Fig. 4a does not approach -100%...

Page 10, line 30: It looks like that the compensation in Fig. 4b is not completely linear.

Chapter 6 and Appendix C (especially Fig. 9), concerning particle size dependence: Do you know the reason for the change of the contact angle distribution with particle size for the kaolinite sample? In general, does the ice nucleation ability of a given substance has to scale with particle size? How pure is the used kaolinite sample as well as the other samples and is it possible that the chemical composition of the samples change with size and therefore the ice nucleating ability? Is there any bias in the measured frozen fractions due to multiple charged and therefore larger particles? A recent paper by Hartmann et al. (2016) shows that due to commonly used particle generation

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methods multiple charged particles (they also used a FLUKA kaolinite sample in their study) can be present which bias the determined frozen fraction.

Conclusions: I am wondering why the alpha-pdf scheme is worse compared to the other two schemes when trying to represent the measured frozen fractions as a function of temperature for the various dusts and dust sizes. But in contrast, the time dependence of freezing can only be reasonably represented by the alpha-pdf scheme. In the latter case this was shown for a given dust (kaolinite) and one size only. The questions arises again: Is there an influence of size dependent composition or multiple charged particles (see comment above) which bias the fitting as a function of T and size?

In general, the conclusion is too vague. From my point of view, this study is a good contribution in order to find appropriate parameterizations based on CNT for GCMs. But I would suggest to clearly state the limits of parameterizing the immersion freezing behavior of mineral dusts due to e.g., limited amount of data available, bias due to possible multiple charges/size-dependent particle composition issues, etc. and that further experimental studies as a function of temperature and time are needed (only one sentence, the last one on page 15, is not sufficient).

Appendix: In general, there is no warm or cold temperature. The temperature can only be high or low. In some cases, plots/tables are shown based on f, others are based on contact angle α. It is difficult to directly inter-compare the results. Could you please refer to only one parameter or both f and α?

Technical notes:

Page 9, line 4: the schemes are mixed up here: CNT #7 is the $\alpha(T)$ scheme and CNT #5 and #6 are the α-pdf schemes.

Page 12, line 18: Should it read "…, which was found to not represent…"?

Page 15, line 6: ‘have been compiled’ instead of ‘are compiled’?

In the upper left plots of Fig. 7 and 8 the highest temperature value (i.e., 260K) is truncated.

Figure 8, lower right panel: The mean freezing temperature is about 237K. Does this “agree” with the high mean contact angle of 0.3?

Caption of Fig. 8: There is a ‘decreasing’ missing in the sentence starting with: ”contact angle gets larger with *decreasing* T because. . . ”

On page 33, line 5-6. There is the verb missing in the sentence “. . . FF is fitted or the active site density directly.”

Fig. 11: The color coding is less than ideal. It is difficult to see which of the lines correspond to which study.

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