

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 #2

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The authors do a thorough examination of different possible ways of fitting the classical nucleation theory to immersion freezing experiments using various formulations for parameters of the theory. They compare the outcomes using three criteria: 1) How well each CNT formulation reproduces the experimental freezing curves; 2) How good are the size and time dependences of each formulation compared with experimental data (if available)? and 3) Are the values of the fit parameters microphysically reasonable?

I believe that this is a useful paper that should eventually be published: at the moment CNT is the only theoretically based approach that can be used as a basis for parametrizing immersion freezing for global models, and this paper provides valuable information for constructing such parametrizations. At the same time it should be kept

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in mind that CNT is by no means perfect.

I have really just one major comment. The authors make the following statement in the Conclusions: "Criterion 3 is difficult to evaluate coming from a macroscopic level as microphysical knowledge is missing at this point". I don't think the situation is quite that bad. There is information available in the literature that can help at least discuss whether the contact angle distributions are physically reasonable, or consistent with information gained from other studies.

First, there are molecular dynamics papers that have investigated water (both liquid and ice) especially at kaolinite surfaces (e.g. Hu and Michaelidis, *Surface Science* 601, 5378, 2007; *ibid*, 602, 960, 2008; Croteau et al., *J.Phys. Chem A*, 114, 8396, 2010; Solc et al., *Geoderma* 169, 47, 2011). Please check these and discuss how realistic the contact angle distributions derived in your study are.

Secondly, there are observations of freezing microdroplets at different hydrophobic surfaces indicating that the contact angle does not change when freezing occurs (Jung et al., *Langmuir* 27, 3059, 2011; Heydari et al., *J. Phys. Chem. C*, 117, 21752, 2013). This information can be used in the context of Young equations. If you write down Young equations for contact angles of 1) a water cluster on a surface S, 2) an ice cluster on S (against air) and 3) an ice cluster on S, immersed in water, you can figure out what the contact angle of ice immersed in water should be if the contact angle of liquid water on the same surface is known (literature values of water contact angles can be found many minerals).

Thirdly, one can make the following question (this time disregarding the assumption of equal contact angles for water and ice): Are the results in this work (i.e. contact angles of ice immersed in water) consistent (again, in the context of Young equations) with ice contact angles derived from deposition nucleation studies? This question should be answerable with the help of the different interfacial tensions used (water, ice, ice in water) and water contact angles. I suggest that you do these two exercises with the

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Young equations and discuss.

ACPD

Minor:

- Contact angle values in Tables 3 and 5 don't seem consistent with the contact angle distributions in Fig. 9. Should the radian values be multiplied by pi? However, even in that case, the mu-values in Fig. 10 appear a bit strange: they are mostly below 90 degrees, although the modes of the distributions in Fig. 9 are all above 90 degrees, and the distributions appear to be skewed right. How come?
- What happened to Appendix A?
- The order of Figs. 9 and 10 should be changed
- The English of the ms should be checked.

Interactive comment

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2015-969, 2016.

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