Interactive comment on “Parameterizing ice nucleation rates for cloud modeling using contact angle and activation energy derived from laboratory data” by J.-P. Chen et al.

J.-P. Chen et al.

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We greatly appreciate reviewer 2 for his/her thorough review. The comments are very constructive and the suggestions can help enhance the readability of our manuscript. We have followed all of them in revising our manuscript. Please note that we made some adjustment in selecting experimental data for deposition nucleation on mineral dust to exclude possible events of "condensation freezing" that was mentioned by the authors but we failed to notice. The resulting contact angles are somewhat smaller, but this does not affect our major conclusions. All relevant data and discussions have been updated.

Specific Comments:
A. The title is misleading in that the work described has no direct connection to cloud modeling. Cloud modeling motivates the study, but no descriptions of or results from atmospheric modeling are presented. The work stands very well on its own. I suggest deleting the words for cloud modeling from the title.

Reply: We gladly agree to use the more concise and appropriate title: "Parameterizing ice nucleation rates using contact angle and activation energy derived from laboratory data"

B. The overall treatment, though classical and correctly done, makes numerous assumptions. For instance, the use of a contact angle to describe the interaction of the ice germ with the solid substrate assumes that the ice germ is a spherical cap. How would the treatment differ if the ice germ were shaped like an hexagonal prism (assumed by Hobbs, 1974, p. 473, among others)? Such nuances may not be apparent to many readers, so the authors need to state all major assumptions explicitly and preferably early in the paper (perhaps at the beginning of Section 2.2).

Reply: In addition to the modifications that made according to reviewer’s point C below, we also added the following discussions in Section 2 to point out the assumptions on the ice germ: The (classical) description is the simplest and most fundamental notion for viewing heterogeneous nucleation as derived from the phenomenon of wettability and its manifestation in the contact angle. On an insoluble substrate, the germ of the new phase is assumed to be a spherical cap with the contact angle characterizing the relationship between the three interfacial energies involved. While this model is based on the formation of a liquid germ from the vapor, it is also adopted as the basis for heterogeneous nucleation of solids from a gaseous of aqueous parent phase. Ice germs may have crystalline structure with hexagonal shape or prismatic shape (cf. Hobbs 1974, p. 473), so it is difficult to define ice germ’s contact angle. But it must also be realized that ice germs may actually be so small that descriptions in terms of simple geometric forms may not be appropriate either. In addition, nucleation takes place on specific locations (sites) on the substrate surfaces, which is a clear indication
of the dominant roles of specific surface features such as steps or dislocations. But even so, the statistical model described above would still work if the preferred sites for embryo growth can be considered equal and randomly distributed. There is very little theoretical guidance on how to formulate descriptions of the interaction energy between such sites and the germs of ice. In any case, the contact angle for ice germs discussed here should be considered as an "apparent parameter" and further discussion on this will follow.

C. Section 2.2 contains numerous equations, but little physical context. A few extra sentences describing the conceptual basis of the equations would help readers who are not experts in nucleation theory. If the pertinent conceptual model is described at the beginning of this section, identification of the implied assumptions (point B above) would follow naturally. When reworking this section, please reevaluate which equations need to be displayed and numbered, and which could be blended into the text. Some equations (e.g., 8 and 12) are redundant in form, differing only in the magnitude of a parameter or two. One such equation could be displayed, the second one alluded to. In general, this section would read more clearly if the equations for freezing nucleation were contrasted qualitatively with those for deposition nucleation. For instance, point out precisely how Eq. (13) differs from Eq. (5).

Reply: Originally, we wish to make the paper more concise by neglecting details of the nucleation theory that can be found in textbooks. But we agree that a few extra sentences will enable novice or even experienced readers to read through the content without constantly referencing textbooks. As the reviewer nicely pointed out for us, this would also allow us to avoid repetition of similar equations. We have revised the manuscript accordingly.

D. The sources of the experimental data used for the statistical analyses are discussed in general terms in Section 2.1, and they are referenced in Table 1. Still, it is unclear exactly how those data are processed and applied to the procedures outlined in Section 3. Please review these sections and modify them as necessary to ensure the maximum
clarity; make sure that sufficient information exists to enable a motivated reader to reproduce the analyses, at least in principle.

Reply: We have put in additional descriptions in Section 2.1 on how the experimental data are processed. Hope these would be sufficient for the readers.

E. The caveats and qualifying remarks discussed in Section 4.4 and touched on again in the second-to-last paragraph of the paper are crucially important. The main point is that the contact angle and the activation energy are apparent parameters emerging from the analyses. When one knows so little about the mechanism of ice nucleation and acknowledges that the classical theory is tentative, one must back away from interpreting the parameters in physical terms. The contact angle could have physical meaning under some circumstances, but one can hardly expect the surface of the nucleus to be energetically uniform. Ice most likely forms, rather, at so-called active sites, where crystalline defects or contaminants exist. I would like to see this paragraph in the last section expanded somewhat to emphasize such points. Mention of the fact that the analyses yield apparent parameters should also be made earlier in the text, as well as in the abstract.

Reply: These are also good suggestions. We have incorporated these comments in the last section, as well as mentioning the "apparent parameters" earlier in the text (see reply to point B above) and in the abstract.

Technical Corrections:
We appreciate the reviewer’s effort of thorough editing. All corrections are incorporated into the revision.

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