

Interactive
Comment

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.

Anonymous Referee #2

Received and published: 28 August 2008

General Comments/Suggestions:

This paper very nicely lays out a framework for extracting fundamental parameters of nucleation theory from experimental data. The approach is sound and offers direct applicability to problems in physical chemistry, as well as cloud physics. The paper is well organized and should be of wide interest to readers of ACP. The comments and suggestions offered here are designed to help the authors refine the description of the technique and reduce ambiguities in a few places.

Specific Comments:

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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.

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).

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).

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.

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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.

Technical Corrections:

Numerous small corrections need to be made to correct grammatical errors. Please be patient making the changes, then proofread the paper carefully.

- a. Page 14420, Abstract, Line 11: Change comma to a semi-colon.
- b. P. 14420, L. 19: Comma before thus.
- c. P. 14420, L. 25: Change at to in. Make environment plural.
- d. P. 14421, L. 11: Delete hyphen in supersaturation.
- e. P. 14421, L. 17: Here and throughout the paper, a comma is needed after i.e.
- f. P. 14422, L. 8: Insert before properties: lack of knowledge about the.
- g. P. 14422, L. 16: measurements should be plural.
- h. P. 14422, L. 24: Delete s from resolves.
- i. P. 14422, L. 25: Replace were with have been.

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- j. P. 14423, L. 1: Insert a before solute.
- k. P. 14423, L. 2: Replace measured with conducted.
- l. P. 14423, L. 20-21: This single-sentence paragraph needs to be developed more fully. A second sentence could mention the four types of IN to be considered in the paper, then follow-on sentences could what categories were not included, and why not.
- m. P. 14423, L. 25: Replace did with have.
- n. P. 14424, L. 3: Make environment and adjective (environmental).
- o. P. 14424, L. 11-12: Explain in greater detail the nature of Fig. 7 in the paper by Field (what is plotted) and how nucleation rates were determined from the figure.
- p. P. 14424, L. 16-17: Define all acronyms.
- q. P. 14424, L. 18: The concept of adiabatic cooling in a cloud chamber must be justified. Walls can have enormous impacts on the cooling rates and the liquid water contents. Explain the extent to which the study here depends on the assumption of adiabaticity and how much error would result if the expansion were not adiabatic.
- r. P. 14424, L. 19: Delete mode. How is it known that the crystals formed by deposition nucleation?
- s. P. 14425, L. 15: Delete mode and the.
- t. P. 14426, L. 11: Replace carried with was measured.
- u. P. 14426, L. 26: Comma needed after temperatures.
- v. P. 14427, L. 22: The citation to Mason shows 1974 here and a couple of other places in the manuscript. However, I believe the year is 1971. Please correct all instances.
- w. P. 14428, L. 7: The word molecule show be plural.

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x. P. 14428, L. 9: Insert the word the before embryo.

z. P. 14429, L. 14 and Eqs. (8) and (12): The mathematics is not wrong, but technically what is here called bulk free energy of phase change is more appropriately termed a chemical potential difference between phases; The Greek letter mu could then be used (for chemical potential) and so decrease the number of times that the letter g is used in the paper.

aa. P. 14432, L. 15 (Eq. 17a): The term called y is ambiguous as written. Consider writing it as the algebraic sum of logarithms.

bb. P. 14433, L. 4 (Eq. 17b): Please check the sign of term a.

cc. P. 14435, L. 7: Insert for after suffice.

dd. P. 14435, L. 8: Insert the after including.

ee. P. 14435, L. 15: Replace the latter with which.

ff. P. 14436, L. 24: Add a citation and reference for Antonoffs rule.

gg. P. 14437, L. 23: The word value should be plural.

hh. P. 14438, L. 15: This part of the sentence should read but it enlarges them.

ii. P. 14439, L. 10: Add d to oppose (i.e., opposed, not oppose).

jj. P. 14439, L. 26 + next line: Insert does after supersaturation; delete s from becomes. (i.e., supersaturation does the activation energy become important.)

kk. P. 14440, L. 3: Replace implicates with implies.

ll. P. 14440, L. 21: Make agent plural.

mm. P. 14440, L. 3: Replace little with few.

nn. P. 14441, L. 23: Replace comparing with compared.

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oo. P. 14442, L. 9: Clarify what one fold means.

pp. P. 14443, L. 16: Add page number to the book citation.

qq. P. 14445, L. 9: Add comma after water. Replace the two words enhances and depresses with enhancing and depressing, respectively.

rr. P. 14446, L. 24: Insert the before natural.

ss. P. 14446, L. 27: Insert them after applying.

tt. P. 14448, L. 22: Rename Section 5 Summary and Conclusions.

uu. P. 14449, L. 16: Make show plural.

vv. P. 14449, L. 27-28: It would help future researchers to learn also what specific types of data would be sufficient for verification. (Offer positive suggestions, not just negative ones.)

ww. P. 14450, L. 4: Replace that with those.

References

Hobbs, P. V. (1974). Ice Physics. Oxford, Clarendon, 837 pp.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14419, 2008.

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