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Interactive comment on “Laboratory measurements and model sensitivity studies of dust deposition ice nucleation” by G. Kulkarni et al.

Anonymous Referee #3

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General Comment

This paper presents some interesting data, albeit with apparent issues, and explores their parameterization using classical nucleation theory (CNT). It is a useful contribution, though I find the paper not especially informative in the exercise to explore what is fairly well known from basic CNT calculations. I recommend revision to clarify a number of points regarding assumptions made in data analysis and application of CNT. Whether or not the revisions are minor or major depend on responses to questions below.

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Abstract

1. The finding highlighted in the abstract that inferred single contact angles do not vary substantially with particle size or temperature seems to contradict some number of other studies.

2. The cloud model simulation results are stated without introducing the fact that a model was used and why.

Introduction Pages 2486 to 2487 : A limitation of parametric methods is stated, but nothing is said in this regard about CNT. The assumptions made in applying CNT are that atmospheric IN can be described by the theoretical construct, either via single contact angles or some distribution of such, and that all ice nuclei behave in a purely stochastic fashion. In fact, the role of time is not even mentioned in this section. It is not correct to ignore these facts, which are quite difficult to validate experimentally. Furthermore, some confusion is present here because the application of CNT is distinctly different than the approach taken by Möhler et al. (2006), where it is assumed that particles activate without time dependence (similar to Connolly et al.). Finally, I believe that the Niedermeier et al. reference given is an application of CNT involving probability distributions of contact angles, and characteristic active sites, and offers an alternate approach to the contact angle-PDF approach that should be better acknowledged.

Methodology

Page 2488, line 12: Friedman et al. called this device the compact ice chamber (CIC). Now it is the PNNL AML ice chamber. Define AML. I suggest sticking with one name.

Page 2489, lines 3 to 5 and lines 15 to 16: 1 micron seems too small of a size to set as a cutoff for ice crystals if one is generating 500 nm particles, or even 300 nm particle using a DMA. It is impossible that there will not be some multiply charged particles this large at the exit. Statements are made regarding such particles, but are only explicitly documented for 100 nm particles out to triplet sizes. For 300 and 500 nm particles, 10

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Page 2490, lines 6 to 7: Please clarify if residence time was varied in any experiments to prove the applicability of the CNT time dependence. If not, simply say so as a preface to all of these calculations. It is an assumption made, the validity of which has not been tested.

Page 2491, lines 12 to 14: The threshold condition used to define onset single contact angles seems rather artificial given the nature of the actual data shown. This is just an observation.

Pages 2491 to 2493, section 2.3: I suggest that this section needs some attention to detail, as it is very confusing how many models are used and what tests or actual simulations are done.

-Page 2491, lines 21-22: Was it a stratiform or a cirrus case? I assume this refers to the present paper or is it just mentioning the paper in which the CRM was described?

-Page 2492, line 12, “Two sets of model simulations...”: So in these simulations you are not simulating the cirrus case? Are the initial conditions the same? It is confusing.

-Page 2492, lines 16 to 17: Is this dust size distribution for the same cloud simulated? Or is this an arbitrary choice, and why? Lines 21 to 22: Is this using the same model? Earlier it was mentioned that SAM is a CRM.

-Page 2492, line 5: It would be good to know something about the sounding. What is the temperature range in which the cloud forms?

Results and discussion

Page 2493, lines 10 to 11: The results indicate dependencies of ice active fraction on RH and temperature, but they also suggest the possibility of a variable, purely aerosol interference at all temperatures and sizes. The ice nucleation signal starts directly from ice saturation, unlike some other recent studies of such aerosols, including the Welti et al. paper. Has this been carefully characterized as ice and rejected as an artifact of the detection method used? What if the ice cut size is pushed to something larger than

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should still capture all nucleated and grown ice, such as 2-3 microns. If this background goes away, it could be smaller ice crystals, but it could also imply aerosol interference that requires correction. Even the PDF scheme cannot reproduce that initial tail can it? Might not this explain the difficulty with the PDF parameter fits in general, and the insensitivity of single contact angle to size and temperature? I suspect that a more conservative approach to detecting assured ice would help a great deal.

Page 2494, line 2 to 3: I find the discussion here to be overly confident in the fact that the PDF parameter scatter is simply the result of active site variations. Such results are not very evident in data shown in Welti et al. (2009) and Jones et al. (2011).

Page 2494, lines 6 to 8: How many experiments are represented here for each PDF fit? Does this discussion imply that insufficient repeats were done in this study?

Section 3.2, page 2495: I am just not sure that the sensitivity studies run offline here deserve even as much space as they are given. Much of this is intuitive on the basis of CNT, and has been explored for CNT going back many years. Nevertheless, the point that the onset of ice formation by the single contact angle method is nearly a step function affecting all of the ice nuclei at one RH, with consequent potential impact on cloud microphysical and radiative properties, is a point worth repeating (see, e.g., Eidhammer et al. 2009). You might comment.

Section 3.3, page 2495, line 21-22: Probably better stated as “The cloud depth decreases as the contact angle increases.”

Page 2495, line 26: Ice nucleation being very efficient goes along with a low contact angle doesn't it? Again, this seems intuitive. And why is there a need to flex calculations for small contact angles if they do not represent real dust?

Page 2497, lines 5 to 6: A similar comment as the previous one, as this is another point that should be self-evident in applying simple CNT calculations. If it is necessary to get to higher RH for ice nucleation to occur, then it should translate into a later time and/or

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higher altitude of cirrus formation, correct? Does the model provide special insight or simply verification of such facts?

Page 2497, line 9: I think suppression is the wrong word here. I thought Friedman et al. showed no ice nucleation activity. What is meant by suppressed?

Page 2497, lines 11-14: Again, here is a case of putting the “cart before the horse” Such behavior seems predetermined by existing theoretical equations. The model simply verifies the obvious impact. I suggest stating expectations upfront based on the nature of CNT calculations, and then show that these behaviors are realized in the model.

Page 2498, line 1: I am surprised at this blanket statement on the lack of sensitivity to N_0 . The N_0 used in this study is a very small number. One can see in the figure that at 10 times N_0 , the ice number did not go up by 10 through the cloud depth. It thus seems possible that higher N_0 will eventually lead to negative feedbacks, and so without knowing just how high N_0 is or should be allowed to be specified, there will be a problem in predicting aerosol effects on clouds.

Page 2498, line 16, 26 to 28: In reference to the comments on ice detection threshold and IN measurement errors, is the thought that you are missing ice? I think just the opposite, and I think there are things to do with the data (e.g., moving the ice detection threshold size) to explore this issue further.

Summary and future work

Point 1: As I mentioned earlier in these comments, I detect that there is reluctance to say this approach appears incorrect. If so, it should be said that this approach could lead to large errors.

Point 2: This could start with “As expected on the basis of well-known CNT ice nucleation sensitivities to contact angle. . .”

Point 3: Just to repeat a point above, I believe that it would be instructive to vary N_0

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over a broader range. So cloud properties depend on the PDF parameters and N_0 in the CNT approach. This is important, as N_0 is not always well known, nor are large particles always only dust.

Point 4. Onset contact angles are calculated, not observed. Rephrase.

Page 2500, lines 4 to 5: “Therefore, for the purpose of applying CNT to describe atmospheric ice nucleation. . .”

Page 2500, lines 15 to 17: Please make it explicitly clear how fundamentally different these approaches (e.g., Connolly et al.) are compared to CNT. It involves the basic assumption on the time dependence of ice nucleation, and this is what must ultimately be explored somehow.

Table 1: Are these results for one experiment in each case? Please clarify.

Table 3: Is -3 a typo? Should it be -30?

Figure 5: These results also do not appear to have been corrected for possible aerosol influence. Were multiplets characterized for kaolinite generation?

Figure 6: Most results show 1 to 1.5 order of magnitude increase of active fraction over these RH ranges, while most of the PDFs suggest > 3 orders of magnitude increase. Thus, I wonder if all of the data have been well represented by the PDF approach, although I think omitting possible aerosol noise would help.

Reference

Eidhammer, T., P. J. DeMott, and S. M. Kreidenweis, 2009: A comparison of heterogeneous ice nucleation parameterizations using a parcel model framework. *J. Geophys. Res.*, 114, D06202, doi:10.1029/2008JD011095.

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