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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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Review

for the manuscript:

Laboratory measurements and model sensitivity studies of dust deposition ice nucleation by G. Kulkarni et al.

submitted to Atmospheric Chemistry and Physics – Discussion

The submitted manuscript describes measurements of the ice nucleation abilities of Arizona Test Dust (ATD) and Kaolinite particles in the deposition freezing mode. The resulting dataset was used to derive functions describing the surface heterogeneity of the used samples by fitting distributions functions for the contact angle based on classical nucleation theory (CNT). The resulting distribution functions were then used in model studies to test their applicability in cloud resolving model simulations. The approach to parameterize the surface heterogeneity of the dust samples is derived from previous studies but for immersion freezing. However, only recently, a paper by Wheeler & Betram (ACP, 2012) has been published that uses a similar approach for deposition freezing. In contrast to the submitted article, Wheeler & Bertram used four different models to test which model fits the data best. This publication is not cited here but it should and I recommend an extensive comparison with the data and derived distribution functions published there. It surprises me that the authors argue, that the step-wise appearance of their results with the distribution function in Figure 2 are the result of the non-linearity of CNT. CNT does not have functions that produce a periodicity like observed in Figure 2. This is also not the case for previous studies for immersion freezing and the already mentioned paper by Wheeler & Bertram, where the simulated functions are monotonous. The two data sets for ATD and Kaolinite in the present study seem to be very comparable (Figure 5), it is therefore surprising that the fit parameters for the PDF differ so much for both particle types. It would also help to provide root mean square error (sums) for the results from the fits. These observations together let me conclude that there might be a flaw in the data analysis routine. The current state of the paper does not provide enough information to draw a real conclusion about the data quality. I therefore recommend to reject the paper in its current state and encourage the authors to re-submit it after major revisions of the paper. In a new version the authors should take care to describe experiment and data analysis more precisely and carefully.

Some specific remarks for corrections will follow below.

General readability: The text should be carefully sub-edited for better readability. Especially the proper use of indefinite and definite articles like “the” according to english grammar should be checked. Often they are missing, the wrong type is used or an

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article is used where it is inappropriate. Some examples are given below. Secondly, I recommend not to use symbols (e.g.like T as a placeholder for the word temperature) in the text. Please use the written words instead. Also, please refer to equations like ... in equation (1) . . . and not just . . . in (1) ... Abstract

Line 4: What is an onset single angle? Introduction

Page 2485, line 14: ...barrier for ICE nucleation.

Page 2486, line 7: THE contact angle.... The following sentence:

There are different definitions of the contact angle for different nucleation modes. The surface energies listed here define the contact angle for immersion freezing, for deposition nucleation, no liquid water is involved, hence no surfaces between liquid water and other phases play a role here. Since the paper focuses on deposition nucleation, the definition for deposition nucleation would be more appropriate here and it should be explicitly mentioned for which nucleation mode the definition is given.

Line 12: . . . contact angleS derived from....

Line 14: ...even was applied to A global climate model.....

Lines 16/17: Elastic strain, aerosol surface irregularities, and active sites are NOT parameters of the standard CNT, so how should they be constrained then in CNT?

Line 22: Is there a reason why the authors did not consider these approaches here?

Line 29: . . . ice nucleation in climate models (delete THE).

This sentence the previous and the following two sentences on the next page do not make a lot of sense to me. It seems that the authors mix up parameters in parameterizations for climate models with CNT. It is not clear what kind of parameters they suggest are missing and if they are missing in CNT or the climate models. These sentences should be re-written to clarify what the authors intend to say. In addition, there is already a paper on deposition nucleation using the approaches from Lüönd et al.

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(2010): Wheeler and Bertram ACP (2012). This study should be also discussed here and the differences between the present study and the study of Wheeler and Bertram.

Page 2487, lines 7 and 13: delete “THE” after ...using

Page 2488, line 14: Did you mean vertical instead of horizontal? Or did you mean the parallel arrangement of the two plates instead of the orientation of the plates themselves. Please re-write this sentence to make this clearer.

Line 16: The principle of A continuous . . ..

Line 17: ...ensures THAT aerosol particles WHICH are placed . . ..

Lines 23 to beginning of next page: The description of what is controlled in the experimental setup is very vague and imprecise. e.g. the linear T gradient establishes by the process of heat transfer but cannot actively be controlled. The only parameters which are actively controlled are the two temperatures of the two walls. This should be clearly stated. The physical process by which the relative humidity profile is established and which controls the RH at the sample position is not correctly explained here. Also, it is not clear which flow setting is used: Is 10 lpm the total flow through the chamber, the total of both sheath flows, or the flow of one of the two sheath flows? Please be precise and clear here.

Page 2489, first paragraph: Precisely, ice nucleates on the surface of an aerosol particle, then the newly formed ice crystal grows but NOT the aerosol particle!

Line 7: Delete THE at beginning of sentence.

Line 10: Please specify what type of aerosol generator was used (e.g. fluidized bed...) and explain the abbreviation DMA.

Line 13: It is not clear if the authors observed the contribution of multiple charges (if yes, how was it done?) or if this is a reference to literature (if so, please cite correctly).

Line 14: ...size particles THE DMA produced.....

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Line 15: Where do the percentages for 152 nm and 197 nm particles come from?

Line 16: 500 nm sizeD particles....

Line 19: How were the RHi corrections been done?

Lines 22-26: The term "modified" does not seem correct to me here. CNT does not make any assumptions about the nature and heterogeneity of a surface that catalyzes ice nucleation. To me the following aspects should be carefully separated: CNT provides a formula based on physical laws to describe a nucleation rate for a certain contact angle of a certain surface area. In contrast, different models exist to describe the surface heterogeneity of a sample of particles. For each surface (fraction) of these models, CNT can be applied in its pure form. This should be made clear in the text. I also recommend to reverse the order of equations 1 – 3 to better represent the relation between CNT and the surface model (parameterization).

Page 2490, line 6: ...residence TIME . . ..

Line 11: Sv,i is the saturation ration not the supersaturation (the following formula is also not correct).

Line 15: Why is this formula given here, when a fixed (non-temperature-dependent) constant from literature is used?

Line 21: I assume this refers to equ. 2 not 1?

Page 2491, lines 6 – 9: As already mentioned earlier, it is not entirely clear to me how the "continuous" fit curve is produced. The mathematical procedure should be better described, also how and which parameter was discretized. Does "further, we did not find any sensitivity" mean, that with increasing number of bins, the fit curve did not change anymore? Please provide RMSE or a similar parameters to describe the quality of the fit.

Line 12: How can equ. 1 be modified? Please provide details.

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Lines 6 – 9, 16: You are discussing results (Fig 2) here already, but the results section only comes later!

Page 2492, lines 17 – 19: Why mention the value of N0 two times here?

Line 23: THE CRM was also....

Page 2493, line 9: ...size OF ATD particles.... Line 10: Wouldn't it make more sense to plot Fice against RHi instead of RHw? The nucleation rate is a function of Si not Sw!!!

Lines 20 – 24 (and next page): How should the existence of active sites be the reason for a scatter in the PDF parameters? Marcolli et al. (ACP, 2007) described a surface model that assumes active sites, but the PDF does not. It is also not argued well, why different PDF's are derived for different temperatures and particle sizes. In a first guess I would assume that the contact angle distribution should be the same for different experiment temperatures as it describes a particle property. It might be different for different sizes if one assumes e.g. that the chemical composition may vary for different sizes but this should be discussed. In general, it appears to me that the authors do not connect their phenomenological observations to the physical concepts behind CNT and the PDF approach well enough. E.g. in lines 1 – 5 (page 2494) the authors argue that the particle to particle variability is the cause for a scatter in PDF parameters but the underlying assumption of the PDF is already a particle-to-particle variability in the contact angle.

Page 2494, line 10: I would be careful with this statement: If the PDF approach correctly describes a particle population, at low activated fractions, only the most efficient particles with the smallest contact angles are activated. So, a contact angle, derived from a low activated fraction does not describe the whole population but only the most active fraction!

Page 2495, line 20: from 5° to 30° both, Ni and IWC, decrease....

Page 2496, lines 5 – 7 and following paragraph and following page: Higher nucleation

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rates for smaller contact angles are a direct consequence of the CNT, so some of the results discussed here are somewhat trivial and do not require simulations to be drawn.

Page 2498: Please add Wheeler & Betram and their results for the PDF model in the discussion here!

Table 3: Some columns are named -3 (temperature) I guess these are -30?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 2483, 2012.

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