

Interactive comment on “A comprehensive parameterization of heterogeneous ice nucleation of dust surrogate: laboratory study with hematite particles and its application to atmospheric models” by N. Hiranuma et al.

Anonymous Referee #2

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General Comment

This manuscript makes a very nice contribution to the literature on mineral dust IN parameterization, potentially providing one for a surrogate of most mineral dusts for use in atmospheric models. This parameterization extends to low temperatures relevant for deep convective and cirrus clouds. The paper does a less good job initially justifying why hematite can be that surrogate (it seems like a topic of evaluation of the paper, but this is not presented up front), and it highlights a difference with an existing parameterization that is constrained by field data, without considering that that study already discussed the major disagreement with laboratory surrogate studies.

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A few key things needing attention in my opinion are:

- 1) Justification for using hematite to represent "atmospheric dust" at the start of the paper, or stating this as a point of evaluation in this manuscript.
- 2) Recognizing the fact that P13 is at least inclusive of some atmospheric INP data, so the mystery of it not agreeing with lab data below -40°C is neither a new discovery, nor is it explained by new data presented herein. As an explanation of lower temperature discrepancies with laboratory dust data, P13 offered that coatings are common and may limit deposition nucleation. This may not be the full reason, but this paper needs to better explain or speculate as to why the modeling community should use the new parameterization in preference to P13.
- 3) Relatedly, the parameterization seems to drastically limit ice supersaturation at low temperatures in atmospheric situations, while still producing ample ice crystals. Some discussion of the realism of that result should be added.

Specific Comments

Abstract

Lines 21–24, but also a general comment: The major question this begs is why we should deem that hematite is representative for cirrus, and why we should deem that the lab studies are relevant for atmospheric particles? This seems like a topic in itself. The parameterization you choose to compare to is constrained by measurements of atmospheric IN, and it notes already that the results are inconsistent with cloud chamber data for pure mineral dusts. Hence, it should be understood that this is not a new story, but a remaining mystery of sorts. This paper may shed some additional light on it due to the fact that it is hard to imagine a different situation for any pure dust aerosol, so perhaps the atmospheric measurements had some unresolved issues. Neverthe-

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less, this paper needs to state more clearly what its goals are besides the new dust parameterization.

1. Introduction

Page 16496, lines 8-10: Regarding this statement, I do not understand what is meant by Welti et al. introducing this idea. The concept of a freezing mechanism of solutions on particles at below water saturation has a history in going back to Zuberi et al. (2002), Archuleta et al. (2005) and so forth. Welti et al. referenced this earlier work (exploring it in the context of CNT), as does the present paper later in the manuscript.

Page 16497, lines 7-9: There is no doubt that there is a need for better parameterizations for cloud models, but an issue not mentioned is if one can be certain that laboratory measurements are representative for the atmosphere. So actually, what this seems to argue for are more and better in situ measurements. Apparently, these may not be as straightforward as setting conditions in instruments and measuring INP. At least, I think the authors should be open to all possibilities.

2. Method

Page 16498, line 6: What is the basis for selecting hematite particles as a proxy in this case?

Page 16501, lines 13-14: I am a bit surprised at the selection of use of the aerosol-independent M92 scheme to represent heterogeneous nucleation when applying a dust parameterization for cirrus levels. Can you please state if the values predicted by M92 are capped at some low temperature, or is it extended far beyond its usual valid (where data were represented in the original paper) temperature range? Is there a reason that the immersion freezing parameterization for hematite was not joined to the one for deposition?

Page 16501, lines 21-24: Further clarification is also needed regarding the use of P13. First, does the parameterization allow initialization otherwise in accord with the

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present parameterization in terms of the numbers of dust particles? Second, even if ice nucleation below water saturation is used for mineral dust particles only, does not this create some likely overlap with the M92 scheme? That is, I would expect the P13 formulation to make ice prior to homogeneous freezing temperatures, where the M92 formulation is already generating ice.

Page 16502, line 22: Is a time step of 20s reasonable or sufficient for cirrus simulations?

3. Results

Page 16504, lines 23-25: Could you say a little more about how immersion freezing data is used to “constrain” fitting curves? Because immersion freezing is requisite at the warmer temperatures? Or because you consider SCF as a subset of immersion freezing? Yet, you have not yet introduced the concept of SCF in the paper to this point.

Page 16505, line 9: I suggest replacing “while concurrent” with “during”.

Page 16505, line 14: Considering this sentence ending, “. . .pointed to a new freezing process in this study” I believe that “point” would be more appropriate. However, there seems a basic problem in saying it is a new freezing process, when what is imagined is something much like what has been discussed explicitly in recent papers and has been alluded to in some previous ones. Should it say that these results appear to support the existence of a pore or surface freezing process, as discussed in recent literature (e.g., Marcolli, 2014). Then move on to define it as SCF, rather than PCF.

Page 16506: I must question the semantics of what can be called “atmospheric dust particles” because I am not terribly convinced that the hematite results encapsulated by ns fits show that they are comparable to actual atmospheric dust, but rather to soil dust samples brought into the laboratory. Are any of the referenced studies, for any temperature regime, actual measurements of atmospheric dust, rather than surrogates for such?

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Page 16506, line 9: “are” compared.

Page 16506, line 16: Is “mobile” the right word for such ice nuclei counters? Mobile implies that they move, rather than the fact that they are movable or able to be installed on aircraft. Perhaps “portable”?

Page 16508, lines 3-5: The lower bound was explained, but not the upper. What controls the upper bound, why physically does n_s remain constant up to the water saturation line, and why presumably? It is explained rather obtusely at present. Does it mean simply that experimental data does not exist in this regime?

Page 16509: Various here, the “three” and “four” parameterizations are mentioned. Please clarify.

Page 16510, COSMO simulations: I find a lot missing in the discussion here. First, the averaging used to obtain the results is not totally clear. What defines a cloudy area for the results shown in Figures 7 and 8? Does it imply that areas with very low ice content and low ice concentrations are averaged along with others of higher optical depth or higher ice water content across the domain? The lower ice concentrations in P13, despite the inclusion of homogeneous freezing are striking and somewhat surprising unless the small regions where stronger ice formation occurs (e.g., homogeneous freezing regions) are averaged out, while the higher values in the AIDA-based parameterizations surprise me if they are averaged over all cloudy parcels, even at low ice supersaturations. Next, the results not only show a vast difference between the domain averaged ice crystal concentrations, but the relative picture of maximum supersaturation evident in Figure 8 is drastically different. If I follow this correctly, it would be highly unlikely to find an area supersaturated with respect to ice by more than 20

Page 16510, last sentence: The data in P13 are clearly limited at lower temperatures. That paper acknowledged this issue and also discussed discrepancies between field and laboratory data at temperatures below -35°C , offering some speculation about this. Can you say that your results confirm these things and can you speak to whether or

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not the modeling provides any new insights?

4. Discussions

Page 16511, lines 14-18: I wonder if the discrepancies between the parameterizations noted here do not actually highlight most the need for more high quality atmospheric measurements of INP's in the upper troposphere. This is what stands out to me at least. The laboratory measurements seem consistent.

5. Conclusion

Page 16512, lines 14-16: I did not note anywhere in the paper where a hypothesis for the appearance of an RH-dependent ice nucleation below -60°C was discussed. Can you suggest anything?

Last sentence: Can you comment on whether or not you believe that the stronger ice nucleation at limited ice supersaturations on the basis of the new parameterization is realistic for the atmosphere? How might this be examined?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 16493, 2014.

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