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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Studies of heterogeneous freezing by three different desert dust samples" *by* P. J. Connolly et al.

Anonymous Referee #2

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General Comments:

This paper presents heterogeneous freezing of ice by three different mineral dust aerosol samples. The general efficacy of mineral dust with regard to ice formation is currently of much interest for implications to ice and mixed phase cloud formation. The paper extends further to describe a parameterization that can be used in atmospheric cloud models where cooling rates are approximately 1 K/min by using ice active surface site densities which are based on the surface area of the IN particles. I think the paper and topic would be of interest to readers of ACP and I therefore recommend it for publication after the comments below have been addressed/clarified.

Specific comments

1) The authors mention on page 467, line 1: 8220;8230;.temperature range appropriate for heterogeneous freezing8221;

While presumably the authors mean the temperature range above T>235 K (where homogeneous freezing is not a competing pathway for ice formation), the specific temperature range should be stated here.

I am ambivalent about using phrase 8220;temperature range appropriate for heterogeneous freezing8221;. This would imply that heterogeneous freezing does not occur below 235 K. If the authors mean that it is not a significant pathway for ice formation at T < 235 K, then a reference should be provided to support this claim.

2) On page on page 471, line 20-25, the authors state that one of their assumptions is that ns (IASSD) is constant for all dust sizes.

This is however not true and the authors have implicitly acknowledged the reason (surface structure and composition both vary with size due to the heterogeneity of mineral dust) for this. However, is this insignificant enough to ignore?

If this assumption is acceptable, what are the reasons it was deemed acceptable (page 471, line 24)?

Is this a valid assumption, especially in light of previous studies (see e.g Archuleta et al. [2005]) that have shown a strong dependency of RH of ice formation on IN size at a given temperature? This further extends to there being present different contact angles (of ice germs) with varying aerosol size. The influence of temperature/RH at which a particle will form ice will also be affected by different contact angles. Lower temperatures (higher RH) will be required to activate sites with large contact angles and vice versa. Perhaps the paper could benefit from contact angle calculations to show how these would change with aerosol size in the temperature range studied.

3) In section 4.3 Page 479 Line 16: When comparing modeled and observed ice concentrations, are the observations coming from just the CPI? If this is indeed the case,

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wouldn8217;t this be an underestimate of ice crystal concentration since the CPI only images particles larger than 10 microns. This would mean that the CPI numbers are already underestimates of the true ice crystal concentrations and therefore any differences between observed and modeled concentrations are in effect greater than implied in the paper. This would also imply that when the model is in agreement with the CPI numbers, it actually underestimates the ice crystal concentrations.

4) Section 5, Page 486: The authors mention (by citing Krueger et al [2004]) that atmospheric processing of Ca containing compounds such as calcite and dolomite may react with nitric acid to form nitrate salts. However, I don8217;t see how this (or any soluble coating formation onto dust particles) would serve as a possible reason to explain the observation of glaciation (high ice crystal concentration) at warm temperatures. To the contrary, one would expect that due to the transformation into soluble species or a soluble coating, water uptake would lead to solution coatings and therefore freezing temperatures would be lowered. This observation has also been supported in many previous studies that have showed that soluble coatings on dust particles (e.g Eastwood et al. [2009]) have decreased freezing efficiency compared to freezing of uncoated dust particles. Thus dust processing that result in soluble coatings or solubilizing of the dust will likely serve to reduce the number of ice crystals at warmer temperatures.

Perhaps this reason should be removed as a possible explanation for differences in observation of ice in the laboratory vs. field.

There is the possibility however that chemical ageing due to oxidation of the mineral surfaces by trace gasses such as ozone may lead to enhanced ice formation.

Technical corrections:

1) Page 465, Line 9 8230;.modes of ice nucleation, i.e. condensation-8230;8230; The parentheses can be removed and a comma after ice nucleation.

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2) Pg 466 Line 20: the word -nucleation- should be added after heterogeneous deposition. Line 22: There should be a comma after 8220;8230;.instrumental error8221; and the word 8220;this8221; should be removed. Line 24: The word 8220;and8221; should be added after the word 8220;8230;.increasing,8230;.8221; Line 28: The word 8220;will8221; should be removed.

3) Page 469 Line 3: a-spherical should be one word

4) Page 473 Line 11: 8220;the8221; should be removed. i.e 8220;in eq. (4)8230;8221;

5) Page 475 Line 19: Should add the word 8220;to8221; 8220;8230;Eq. (6) is set to zero8230;.8221;

6) Page 480 Line 12: Add a comma after ice concentration.

7) Page 485 Line 18-20: The sentence sounds confusing especially the latter part 8220;8230;8230;as noted from the confidence limits, which are shown bar the 8220;error8221; bars.8221; Do the authors mean 8220;8230;..by the 8220;error8221; bars.8221;? Line 21: Has 8220;Ac8221; been defined before in the paper?

8) Page 487 Line 1: 8220;into8221; should be replaced with 8220;of8221; and the word 8220;onto8221; should be added between 8216;nucleation8217; and 8220;three8221;. Line 13: should read 8220;..to get good agreement between modeled and experimental data.8221;

9) Page 488 Line 4: 8220;ice concentrations8221; should read 8220;ice crystal concentrations8221;

Figures and Captions

1) Figure 2 (caption): The word 8220;shows8221; and the phrase 8220;and-is-used8221; should be removed. 2) Figure 4 (caption): part c) 0C should read 0°C 3) Figures 5, 6, 8-11, and 14. The cloud particle images are very interesting and would be useful to readers, but the shape of the crystals would probably better relayed

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Interactive Comment

by showing just a few particles that are more representative of the images currently shown. This would allow for publication of larger images and a more clear indication of what the particles look like.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 463, 2009.

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