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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling of cirrus clouds – Part 2: Competition of different nucleation mechanisms" *by* P. Spichtinger and K. M. Gierens

Anonymous Referee #1

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General comments

This is a very well written and fascinating paper. While parcel and box models have given excellent insights into the important interplay of nucleation processes in cirrus clouds, the utility of the present study is to introduce new insights and implications that are only resolved through the inclusion of additional dimensionality and the sedimentation processes that ensue following ice crystal growth. The finding of conditions that promote long-lasting cloud supersaturations is unique and important considering the unknown variability of ice nuclei number concentrations in a changing climate. My primary scientific criticism, and it may be a minor one, is that I believe that a consider-

ation of the dynamics of ice nucleation on natural ice nuclei could impact conclusions. In particular, heterogeneous ice formation is seldom observed to occur as a threshold phenomenon like homogeneous freezing. Therefore, it would have been interesting to consider a case where the ice nuclei number concentration increases over a range of RH toward the homogeneous freezing condition rather than ensuing instantly at one RH that is below the homogeneous limit. I only suspect that this makes a difference. My sole technical criticism is that the figures are unreadable without enlarging them greatly.

Specific comments

Introduction:

1) Page 9062, lines 29-30: While Vali (1985) is a good general reference on ice nucleation mechanisms, I do not find it a good one for homogeneous freezing of haze particles. Ascribing homogeneous freezing conditions based on Koop et al. (2000) would seem to require the qualifier “on average" and should state a temperature below which the RH mentioned applies. Is it in excess of 45% below -38°C?

2) Page 9063, line 2: It is stated that ice nuclei are rare in the upper troposphere, but none of the references given here help in supporting that statement since they do not include measurements of ice nuclei. Should this at least say “expected to be rare at most times"? More useful would be to provide support for the range of values you have selected. This turns out to be critical to the conclusions. Furthermore, some models of heterogeneous ice nucleation in use to do not limit values to these levels.

3) Page 9063, line 8: I am not sure that I understand how a box model differs from a parcel model, in particular some of the ones referenced that suggested the same conclusions. Can you explain?

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4) Page 9065, line 2-5: As mentioned in my general comment, I could argue quite strongly I think, based in fact on the results in this paper showing the impact of the RH onset for threshold heterogeneous nucleation, that a nucleation process that occurs with increasing contribution over a broad RH range could be important to consider. None of the laboratory studies references I am aware of, and Moehler et al. (2006) given here is one of those, suggests that heterogeneous nucleation in this T, RH regime occurs as a simple RH threshold phenomenon. Also for a freezing process (versus deposition in Moehler et al.), Marcolli et al. (2007) indicate disagreement with a threshold condition process. Niether do the atmospheric measurements referenced (DeMott et al. 2003) support this. Shall you simply say this was not investigated?

Setup:

1) Page 9067, line 3: I did not see the definition of the term “ISSR" anywhere. I think I know what it means, but a definition would help.

Discussion of results:

1) Page 9071, lines 26-27: The result noted does not seem especially clear in comparing Fig. 4 to Fig. 5.

2) Page 9072, discussion starting line 12: This is an example of where I wonder how a more realistic implementation of heterogeneous nucleation might impact results and conclusions. In fact, ice formation may occur through the whole RH range up to the point of homogeneous freezing.

3) Page 9075, lines 10-11: I really do not understand this stated impact of the presumably “relatively low nucleation rate" at warmer cirrus temperatures. The homogeneous nucleation rate is always relatively high once proper conditions are achieved. Is the

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noted effect really controlled by nucleation rate or by the depletion rates/competition for water vapor?

4) Page 9076, lines 15-23: This section argues for no impact of fluctuations on heterogeneous nucleation, but does not acknowledge that a more realistic implementation of nucleation could have an impact because it is no longer a simple number phenomenon. It is unclear to me that your conclusions still follow.

5) Page 9077, line 14: “Apart from mechanisms related to relaxation" And these are?

6) Page 9078, lines 21-25: Can you be specific about what exotic mechanisms are you referring to? For example, the impact of nitric acid uptake by ice? Also, I was curious what a comparison of your simulation results to observations of RH distributions say about the expected concentrations of ice nuclei in the upper troposphere. Finally, I was also curious if, in light of your results, the parameterization of ice nucleation in some models might explain why they simulate cirrus little differently using heterogeneous nucleation as compared to homogeneous freezing. In particular, if one assumes that high concentrations of ice nuclei are possible, the results you obtained might never be found.

Technical corrections

1) Throughout the paper, is “boxmodel" a word? I suspect it should be “box model".

2) Page 9063, line 20: I would suggest “nevertheless" for the word “yet".

3) Page 9064, line 15: I would suggest “common" for the word “dual".

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4) Page 9071, lines 15-19: The statement “increase rate of the supersaturation" appears here and later. I suggest rewriting this statement for clarity as, “The increasing rate of the supersaturation in response to lifting becomes more pronounced as soon as the ... Hence the overshooting over the homogeneous nucleation threshold increases and more crystals are formed homogeneously when the heterogeneous nucleation event is past."

5) Page 9071, line 26: “in-cloud"

6) Page 9073, line 3: “affects"

7) Page 9075, lines 15-19: I suggest rewriting these statements for clarity as, “However, the increasing rate of the supersaturation in response to lifting is retarded by the few heterogeneous ice crystals such that fewer ice crystals are produced homogeneously."

8) Page 9076, lines 10-11: I suggest rewriting this sentence as, “These crystals now retard the increasing rate of supersaturation in response to lifting ..."

9) Page 9076, lines 4: “surface area"

10) Page 9080, lines 15-17: I suggest “This impeding effect ... is strong even when RHi_{het} is just a little below RHi_{hom}."

11) Figure 6 caption: Please restate in the figure caption the meaning of the blue line in the first column of figures.

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12) Figure 8 caption: Please restate here that these results are for purely homogeneous freezing.

13) Figure 12: I think the caption is incorrect in describing the plots a-e. It appears that they are for different ice nuclei concentrations, not for different time periods. The time periods are distinguished within each plot.

Reference:

Marcolli, C., S. Gedamke, T. Peter, and B. Zobrist (2007), Efficiency of immersion mode ice nucleation on surrogates of mineral dust, Atmos. Chem. Phys., 7, 5081-5091.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9061, 2008.