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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Understanding cirrus ice crystal number variability for different heterogeneous ice nucleation spectra" *by* S. C. Sullivan et al.

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Thank you for your thorough and insightful remarks. Our more recent work has shown that the updraft representation affects the nucleated ice crystal number concentration greatly within CAM5, and we have incorporated a more detailed discussion of the input dynamics in our simulations. A more appropriate and up-to-date measurement-model comparison has also been added. Responses to all the comments raised are provided in italics.

This paper evaluates the ice concentrations (and sensitivities to input parameters) produced by several ice nucleation parameterizations within the framework of global-

model simulations. I have significant concerns about the modeling framework and discussion, particularly concerning the importance of vertical velocity that is given little attention in the paper.

Major comments:

A number of detailed modeling studies have shown that both the number of ice crystals produced by homogeneous freezing and the competition between homogeneous freezing and heterogeneous nucleation depend strongly on the assumed vertical wind speed (cooling rate) driving the . . . the performance of the nucleation parameterizations as *they are operating in CAM* rather than indicating the competition between nucleation modes in the real atmosphere.

Section 2.2 has been expanded to include more discussion of how the updraft velocities are specified. We use a Gaussian distribution of updrafts with a mean value close to zero and the standard deviation from the CAM 5.1 values, which are coming from the turbulent kinetic energy in the Bretherton and Park 2009 moist turbulence scheme. This is the most thorough way that we have used so far to account for the effect of sub-grid vertical motion variability on formed hydrometeor number. To comment on how these values compare to reality, the supplementary material now contains a comparison of the CAM5 updraft velocities (the standard deviation within our Gaussian distribution) and the MMCR values from SPARTICUS in both 1997 and 2007. The comparisons are done at the same altitude, longitude, and latitude and over year-long periods for consistency. We use the daily-averaged CAM5.1 updraft velocities, which agree better with observations. Other studies also examine the vertical velocity distribution for SPARTICUS data or for CAM simulations [Muhlbauer et al. 2014, Shi et al. 2015], and we mention these.

In the discussion of the comparison between measured and simulated ice concentrations, the authors note that sedimentation and autoconversion will reduce ice concentrations. Are the ice concentrations shown simply what comes out of the nucleation parameterizations, or are they mean ice concentrations in the model including cloud aging processes such as sedimentation, mixing, and autoconversion? If the former case is true, then the comparison with measured ice concentrations is inappropriate. Recent studies have shown that sedimentation produces broad regions of relatively low ice concentrations (Spichtinger and Gierens, 2009, ACP; Jensen et al., 2012, JGR; Jensen et al., 2013, JGR; Murphy 2014, ACP). Sedimentation can reduce mean ice concentrations by up to an order of magnitude compared to concentrations immediately after ice nucleation events. Small-scale dynamics and entrainment further reduce ice concentrations as cirrus age (e.g., Jensen et al. 2011, JGR; Dinh et al. 2014, ACP).

The ice concentrations shown are just the output of the nucleation parameterization; the sensitivities are of this nucleated ice crystal number to different inputs. For clarity, this is stated in the appendix definitions (e.g., "nucleated ice crystal number sensitivity to coarse mode dust diameter"), the captions of Figures 2 and 3 ("Annually-averaged output nucleated ice crystal number" and "Distributions of simulation output, i.e. of the annually-averaged output nucleated ice crystal number . . . are shown"), and the description of Figure 2 in Section 3.1 ("Figures 2a through d show the annually-averaged potential nucleated ice crystal number for each grid cell, given the vertical velocity and aerosol profile"). We use data from more recent cirrus measurement campaigns, at altitudes for which secondary ice formation should be insignificant, and consider only crystal numbers in the smallest size bins. While we do not expect a one-to-one comparison with these values, this comparison still indicates reasonable agreement, given that these data have not been used to tune model parameters and that the measurement uncertainty may be around an order of magnitude itself. These distributions are now shown in Figure 3 and discussed in Section 3.1 and the response to comment 5 below. The issues about sedimentation and entrainment are now included in Section 3.1, using the suggested references.

Specific comments:

1. Page 21672, Line 25: There are better citations than Gettelman (2002) for the points

made here. Chen et al. (2000, J. Clim.) would be appropriate for the climatic effects of cirrus. For the general issue of dehydration of air entering the stratosphere, Brewer (1949, QJRMS) and Jensen et al. (1996, GRL) would be appropriate.

Thank your pointing this out. Citations have been changed.

2. Somewhere in the paper, it would be appropriate to cite the Cziczo et al. (2013, Science) observational evidence for common occurrence of heterogeneous ice nucleation.

The second paragraph of the introduction discusses different nucleation regimes. The Cziczo et al. citation is incorporated here.

3. Page 21677, Line 17: TAPENADE is not defined.

TAPENADE is not an acronym, so does not require a definition (www-sop.inria.fr/tropics/tapenade.html).

4. Section 2.1: The authors should add some discussion about the errors associated with approximations made in the BN parameterization based on comparisons with detailed cloud models. Perhaps these comparisons were made in earlier papers that can be cited here.

The parameterization's approximations are discussed in detail in the Barahona and Nenes 2008 and Barahona and Nenes 2009ab manuscripts. We summarize these and add appropriate citations in the second paragraph of Section 2.1. Parameterization output matches that of a detailed parcel model to within 5% for a wide range of cirrus formation conditions, so we do not anticipate substantial error, relative to the parcel framework. This is also mentioned in the manuscript.

4. Section 2.4: Are the parameters in the classical nucleation theory spectrum for dust and black carbon based on field or laboratory measurements? Or are they purely theoretically derived?

The spectrum is the CNT formulation as described in Barahona and Nenes 2009. Contact angles come from the laboratory data in Chen et al. 2008. Nucleation efficiencies were discussed in Personal correspondence but are in agreement with the laboratory data from Möhler et al. 2006 and field data presented in Pruppacher and Klett (2007). The values and sources are listed in Table 1.

5. Figure 2e: This Figure is missing recent field campaign observations made with improved instrumentation (e.g. SPartICus, MACPEX, and ATTREX; see Jensen et al., 2013, JGR, and Jensen et al., 2013, PNAS). These datasets provide much more extensive data (better statistics) than those included in Krämer et al. (2009) and are less susceptible to shattering artifacts that can inflate the measured ice concentrations.

A more thorough, up-to-date measurement-model comparison is now given in Figure 3 and discussed in Section 3.1. SPARTICUS FSSP and MACPEX VIPS and 2DS are data are used within 20 hPa of the simulated altitude for January 2010 and April 2011 respectively. We use the same uniformity criterion as Jensen et al. 2013, in which the values span at least 45 s, and use only values for the FSSP 3.80, 5.85, 8.30, 11.45, and 14.25 μm-centered bins; the VIPS 10-20 μ m bin; and the 2DS 5-15 mum bin. These smallest size bin values are assumed to be the best possible measurement proxy for the newly-nucleated crystal numbers that we are modeling. Attempts to reduce shattering artifacts in these datasets, both by specially-designed probe tips and by post-processing with inter-arrival time algorithms, are explicitly mentioned.

The data is publicly available, and it is relatively easy to generate ice concentration statistics from the data. The temperature variability in ice concentrations apparent in the Krämer et al. (2009) dataset is very likely simply a result of limited sampling statistics. The ATTREX data shows that larger ice concentrations do occur in cold TTL cirrus on some occasions (Jensen et al., 2013, PNAS).

A focus on the temperature dependence of ice crystal number concentrations is removed. The model-measurement comparison is done instead with probability distributions of N$_i$ in the smallest size bins of each instrument relative to the probability distribution of our simulated, newly-nucleated N$_i$. No ATTREX data are used in the model-measurement comparison because these are taken at much higher altitudes than those at which we perform our simulations.

6. Page 21682, Line 15: There are more appropriate citations for the issue of surprisingly low ice concentrations at low temperatures. Specifically, Krämer et al. (2009) and Jensen et al. (2010, ACP) were the first to note this issue.

This citation changed to Krämer et al. 2009, Jensen et al. 2010, and Barahona and Nenes 2011.

7. Figure 2: It looks like the ice concentrations are relatively high poleward of about 40$\deg$ in the PDA13 simulations and in the northern extratropics in the CNT and AIDA simulations. Are these results simply caused by a lack of available IN resulting in predominance of homogeneous freezing? It is noteworthy that the ice concentration geographic variability is quite large, and I am not aware of any observation evidence for such large variability. Perhaps this goes back to the issue raised above of whether the ice concentrations shown are just after nucleation events or mean values at all cloud ages. Cloud aging processes would tend to wash out the large variability just after ice nucleation.

The well-documented spatial variability in INP concentrations (e.g., DeMott et al. 2010, PNAS; Murray et al. 2012, Chem. Soc. Rev.) most likely causes the observed variability in crystal number, since we are looking at nucleated ice crystal numbers. Other studies like Barahona et al. 2010, JGR (Figure 4) have also seen significant spatial variability in modeled, nucleated N$_i$, both within and between different nucleation spectra. Cloud aging processes and spatiotemporal averaging will reduce this variability but not completely eliminate it. A sentence about N$_i$ spatial variability, including this last point, has been included in Section 3.1.

8. Figures 2-4: In each of these multi-panel figures, it would make comparison much

easier if the same color scale were used for all four panels.

Figures were done initially with varying color scales because it tends to suppress field structure; however, figures 2 and 3 (ice crystal number concentrations and contributions) were replotted with the same color scale for all panels. The values in Figure 4 are too different to use the same color bars.

9. Section 3.1: What are the relative contributions from homogeneous and heterogeneous ice nucleation in these simulations? Later in the paper, ice nucleation tendencies are used to infer relative contributions from different modes of nucleation, but would it be possible to simply show side-by-side figures with the ice concentrations from homogeneous and heterogeneous ice nucleation?

The heterogeneous fraction fields are now included for all spectra in the Supplementary Information. Discussion of this field is incorporated into Section 3.1. The discussion at the end of Section 3.1 is also edited for clarity.

10. Page 21684, Line 2: Presumably, the authors mean 1 cm^{-3} here rather than 1 L^{-1}.

Yes, thank you for pointing this out.

11. Section 3.2: It is apparent from this section that there are considerable discrepancies between laboratory results of ice nucleation efficacy, particularly for BC. There are also a number of parameters in the INP parameterization that are difficult to constrain. BC and dust concentrations in global model simulations are highly uncertain. It might be worth adding a general note that all of these factors lead to large uncertainties in the ice concentrations predicted by the parameterizations.

Two sentences pointing out the uncertainty in N$_i$ simulations are included at the end of Section 3.2. Given the highly nonlinear response of N$_i$ to INP, uncertainty in INP concentration may be reduced when expressed in terms of uncertainty in nucleated Ni error (e.g., Barahona et al., 2010). Looking at Figure 1, a cirrus system can have similar N$_i$ numbers but very different INP numbers, so we may be able to tolerate higher INP

uncertainty in cirrus modeling studies.

12. Page 21687, lines 11-12 and Figure 4b: The color scale on Figure 4b gives the impression that all of the sensitivities are negative. It would be helpful to indicate where the positive values occur. Perhaps a zero sensitivity contour could be added to the panels b and c of Figure 4.

Zero contours have been added to Figure 4 panels b and c. This is noted in the discussion of Section 3.3.

13. Figure 5: It would help if the axes were labeled.

Good point. Figure remade with axes labeled.

14. Page 21688, Line 23: The text refers to peaks in N$_i$ in Figure 5d, but the figure just shows updraft speed and dust number sensitivities. This sentence should be rewritten to clarify the point.

This sentence reworded to indicate correlation between large updrafts and large dust number sensitivities (rather than large N$_i$): "Figure 4d has primarily positive sensitivities of small magnitude with an occasional, large spike in $\partial N_i/\partial N_{dust,a}$, which always corresponds to a large updraft."

15. Section 3.4: It is not clear what the meaning of mean efficiency is for spectra that produce both positive and negative tendencies, depending on geographic location, temperature, and vertical wind speed. Are the means calculated exclusively in regions where heterogeneous nucleation on dust dominates? The efficiencies would seem to make little sense when both homogeneous and heterogeneous nucleation are occurring or when BC and dust are competing. Wouldn't it be clearer just to show the contributions to ice concentration from homogeneous freezing, heterogeneous nucleation on soot, and heterogeneous nucleation on BC for each of the four parameterizations?

Only positive tendencies are used to construct the nucleation efficiency distributions, *shown in Figure 7 (which was previously Figure 6). This was stated in the original text, but the first sentence of Section 3.4 is modified to make this clearer: "The positive values of* $\delta N_i / \delta N_{INP}$*, for which nucleation is purely heterogeneous, can be understood as nucleation efficiencies". The point is made later in Section 3.4 as well: "Figure 7a shows the distribution of a random sample of 5000 daily-averaged dust number sensitivities, when ice nucleation is purely heterogeneous, i.e. the sensitivity is greater than 0."*

With the above said, we do not state that the sensitivity reflects the inherent nucleation efficiency of a given aerosol group. The sensitivity reflects the nucleation efficiency of an aerosol group, given the particular model state where other aerosol may also be nucleating. In this sense, the sensitivities can be interpreted as efficiencies, even if both dust and BC act as INP. This sort of definition may be quite useful because it can be related to ambient data, e.g. by comparing the distribution of cirrus ice residuals to the nucleating aerosol in the model. This particular definition of efficiency is clarified in the second sentence of Section 3.4

Finally, the direct contributions of dust and black carbon for the Phillips spectra are shown in what was Figure 3, now Figure 4. The AIDA spectrum does not include black carbon, and the BC contribution for the CNT spectrum is negligibly small. The heterogeneously-formed fraction field is included as Supplementary Figure 10.

16. Page 21692, Lines 13 – 14: So, how large is the sulfate sensitivity in the model, and how does it compare to detailed cloud models? Kärcher and Lohmann (2002) showed that the sensitivity of ice concentration from homogeneous freezing to aerosol concentration is relatively weak (at least compared to the sensitivities to cooling rate and temperature).

The sulfate sensitivity is generally on the order of 0.001 $cm^3\ cm^{-3}$ *but can be as large as 0.025* $cm^3\ cm^{-3}$ *for the lowest temperatures in the SH. The sulfate sensitivity magnitude and the Kärcher and Lohmann (2002) analysis are mentioned in Section 3.6.*

17. References: It would seem that there are errors in the reference list. For example, the Hoose and Möhler reference has four numbers following the year. Are these all page numbers? Other references also have more than one number following the year. The authors should carefully proofread the reference list.

Thank you for pointing this out. These numbers were not present in the original proof. We will make sure that they will not be around in the final publication.

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