

1 Response to reviewers

2

3 I thank both reviewers for their detailed reviews. In addition to the changes they
4 requested, the mass shading in Figure 4 has been corrected. The draft version had
5 erroneous color shading. The model calculations were not affected.

6

7 Reviewer 1 (E. Jensen):

8

9 1) Emphasize the importance of small departures from the Koop et al. approximation.

10

11 *This has been added to the abstract and to the top panel of Figure 1.*

12

13 2) Provide more detail about the temperature fluctuations in the model.

14

15 *This has been done.*

16

17 3) Figure 2 is plotted versus frostpoint temperature – add nucleation temperature?

18

19 *A sentence has been added to the figure caption.*

20

21 4) Page 10707, do newer laboratory measurements provide evidence for variations in
22 supersaturation threshold with aerosol composition?

23

24 *I've looked and haven't found anything that directly addresses this question. I did add a
25 reference to the Zobrist et al. (2003) paper on freezing of high molecular weight
26 polymers that indicates a variation in water freezing activity with molecular weight.*

27

28 5) Page 10708, the Kraemer study included outflow cirrus and used FSSP instruments
29 susceptible to shatter. Also, the sampling of cold cirrus is very sparse... the most one can
30 conclude from the measurements is that there is a lack of compelling evidence for a
31 strong increase of ice concentration with decreasing temperature.

32

33 *The outflow cirrus possibility was already mentioned; I've added a mention of shatter. As
34 requested I've removed the limits from Figure 2. Statements about the observed
35 temperature dependence have been softened: the abstract has been changed to say "not
36 observed" instead of the stronger "opposite of observed."*

37

38 6) There seem to be two mechanisms for generation of fallstreaks discussed in the
39 manuscript: (1) the diversity of ice concentrations produced by nucleation at different
40 altitudes because of the differences in temperature histories, including layers with low ice
41 concentrations that permit growth of large crystals that lead to fallstreaks; and (2) the
42 "mother cloud" effect whereby ice crystals diffuse out of the high concentration layer
43 into supersaturated air below where growth rates are rapid and large crystals result.

44

1 *I appreciate this clarification from the reviewer. The abstract and discussion have been*
2 *changed to more clearly differentiate these mechanisms and two more (entrainment and*
3 *the classic heterogeneous nucleation).*

4
5 7) Page 10710. In ATTREX, the layers with high ice concentrations were embedded
6 within deep layers with low ice concentrations.

7
8 *Comment added.*

9
10 8) Page 10711: Thin layers in ATTREX were remarkable; this model provides an
11 explanation

12
13 *I agree but don't want to push the agreement too far because the 1-dimensional model*
14 *has imposed cooling profiles that are inherently arbitrary. I think that it would take a 3-D*
15 *model with dynamically consistent temperature fluctuations to prove this agreement.*

16
17 9) Page 10711: Not convinced that curved fall streaks show that shear cannot be
18 simulated with a 1-D model.

19
20 *I partly agree. Curved fall streaks show that shear is present. The point made by*
21 *Spichtinger and Gierens (2009) is not that wind shear per se cannot be simulated in one*
22 *dimension but that the shear interacts with and modifies the small-scale dynamics. This is*
23 *another aspect to my reply to point 8 above about needing a three-dimensional model for*
24 *some questions.*

25
26 Citations:

27
28 *The suggested citations have been added to the paper. I made some extra model runs with*
29 *an accommodation coefficient that varied with the saturation ratio as suggested by Zhang*
30 *and Harrington (2014) and it made almost difference to the initial ice number; this is*
31 *now noted in section 3.*

32
33
34 Reviewer 2 (anonymous):

35
36 Main points 1) It is well known that sedimentation is of major importance for shaping
37 cirrus clouds in the vertical.... Thus, the qualitative results about the role of
38 sedimentation in comparison with the observations seem not really new. ...Since all
39 events were tracked, I would like to see a quantitative statistical analysis about the
40 frequency of occurrence of such events...For the stratiform cirrus clouds
41 in the model study, the vertical extensions of nucleation zone and sedimentation
42 dominated vertical layer could be estimated and used for a more quantitative
43 comparison with observations.

44 *I do not claim to have found the importance of sedimentation, and I think this is stated in*
45 *the discussion. The new result is the interaction of the variability of temperature histories*
46 *with sedimentation to add importance to those histories that only produce a few crystals.*

1 *The frequency of occurrence of the events is shown in Figure 1 for the parcel. The 1D*
2 *model uses the same microphysical code and temperature histories. Comparing the*
3 *vertical extent of the sedimentation dominated vertical layer to observations would be*
4 *interesting but can't be done because the initial humidity vertical profile is an*
5 *assumption in the 1D model.*

6
7 2) Lack of model description and setup.

8
9 *More detail has been added to the model description; see also response to reviewer 1.*
10 *The reviewer is incorrect in thinking that the model was developed for polar*
11 *stratospheric clouds; the parcel model was developed for cubic ice over a range of*
12 *temperatures and the one-dimensional model was developed for cirrus.*

13
14 Minor points: 1) Description and realism of temperature fluctuations.

15
16 *The temperature fluctuations are now explained more in the text. The reviewer is correct*
17 *that temperature fluctuations "are not just noise but they stem from dynamic features."*
18 *Unfortunately, a one-dimensional model or parcel model cannot resolve those dynamics*
19 *and must therefore use imposed fluctuations. The fractal fluctuations are quite realistic in*
20 *terms of reproducing the spectrum and autocorrelations found in the atmosphere.*

21
22 2) Ice nucleation: better reference the well-known effect of modification and/or
23 suppression of homogeneous nucleation events due to previous heterogeneous nucleation
24 events.

25
26 *Most of the suggested references have been added; two didn't quite fit. For example, one*
27 *of the papers modeled measurements now known to be affected by shatter.*

28
29 3. Accommodation coefficient: There is a recent review including new measurements on
30 the role of the accommodation coefficient for phase transitions vapour-ice (Skrotzki et al.,
31 2013). They report values in order of 0.5 to unity for the accommodation coefficient.

32
33 *This paper is now referenced. Based on it I have rerun the simulations (and changed all*
34 *figures) with slightly higher values of the accommodation coefficients (0.2 and 0.4 for I_c*
35 *and I_h , respectively). I'm not quite willing to go up near unity on the basis of this study*
36 *when studies such as Magee et al. (2006) still support much lower values. It makes me*
37 *wonder if some of the discrepancy in the literature is due to some experiments measuring*
38 *the accommodation coefficient on disordered ice and some on hexagonal ice.*

39
40 4. Interpretation of low ice crystal number concentrations: It is not correct that
41 low ice crystal number concentrations are hard to obtain at low temperatures.
42 Spichtinger and Krämer (2013) have shown that under certain conditions this
43 work quite well. In addition, former studies by Lin et al. (1998) showed that for
44 wavy structures phase shifts might lead to low number concentrations, even in a
45 high velocity regime.

46

1 *Both papers are already cited as showing that it is possible to create low ice crystal*
2 *number concentrations at low temperatures from homogeneous freezing. This paper goes*
3 *beyond that to show (Figure 1) that the vast majority of temperature histories produce*
4 *high numbers. Yet the few that produce low numbers are still important.*

5
6 5. Mixing of different scenarios in the discussion: In the discussion, different scenarios
7 were mixed in a confusing way. For instance, the comparison with cold stratospheric
8 conditions including NAT particles is misleading. ... The comparison to liquid clouds is
9 also not really meaningful, since the conditions for pure ice clouds are quite different.

10
11 *The comparison to stratospheric NAT clouds is there because the Fueglistaler et al.*
12 *paper identified a mechanism of sedimentation that also applies to cirrus. I've added a*
13 *sentence to try to clarify the mechanisms. I agree that conditions for ice clouds are*
14 *different – the point of mentioning liquid clouds is to say that they are very different.*

15
16 6. Cirrus clouds in a supersaturated environment: This issue is strongly related to the
17 former point. Ice formation requires high supersaturation (in contrast to droplet
18 formation), thus from theory it is very clear that ice clouds are embedded into a
19 supersaturated environment. This was shown in many former measurements, thus some
20 articles of the relevant literature should be cited.

21
22 *I've moved the citation of Diao et al. to a separate sentence where I also cite several of*
23 *the observational studies suggested by the reviewer.*

24

25

26

27 Document with tracked changes follows for the editor:

1 **Rare temperature histories and cirrus ice number**
2 **density in a parcel and one-dimensional model**
3

4 Short: Cirrus ice crystal number density

5

6 **D. M. Murphy**

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8 Boulder, CO USA

9 Correspondence to: D. M. Murphy (daniel.m.murphy@noaa.gov)

10

11 **Abstract**

12

13 A parcel and a one-dimensional model are used to investigate the temperature
14 dependence of ice crystal number density. The number of ice crystals initially formed in a
15 cold cirrus cloud is very sensitive to the nucleation mechanism and the detailed history of
16 cooling rates during nucleation. A possible small spread in the homogeneous freezing
17 threshold due to varying particle composition is identified as a sensitive nucleation
18 parameter. In a parcel model, the slow growth rate of ice crystals at low temperatures
19 inherently leads to a strong increase in ice number density at low temperatures. This
20 temperature dependence is not observed. The model temperature dependence occurs for a
21 wide range of assumptions and for either homogeneous or, less strongly, heterogeneous
22 freezing. However, the parcel model also shows that random temperature fluctuations
23 result in an extremely wide range of ice number density. A one-dimensional model is
24 used to show that the rare temperature trajectories resulting in the lowest number

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1 densities are disproportionately important. Low number density ice crystals sediment and
2 influence a large volume of air. When such fall streaks are included, the ice number
3 becomes less sensitive to the details of nucleation than it is in a parcel model. The one-
4 dimensional simulations have a more realistic temperature dependence than the parcel
5 mode. The one-dimensional model also produces layers with vertical dimensions of
6 meters even if the temperature forcing has a much broader vertical wavelength. Unlike
7 warm clouds, cirrus clouds are frequently surrounded by supersaturated air.
8 Sedimentation through supersaturated air increases the importance of any process that
9 produces small numbers of ice crystals. This paper emphasizes the relatively rare
10 temperature trajectories that produce the fewest crystals. Other processes are
11 heterogeneous nucleation, sedimentation from the very bottom of clouds, annealing of
12 disordered to hexagonal ice, and entrainment.
13
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1 **1 Introduction**

2

3 Cirrus clouds cover large areas of the Earth (Wang et al., 1994). They reflect a significant
4 amount of sunlight, trap infrared heat, and affect the local heating rates and circulation of
5 the upper troposphere (Liou, 1986). These radiative effects are sensitive to the number
6 density of ice crystals (Fu and Liou, 1993; McFarquhar et al., 2000). There are significant
7 uncertainties in ice nucleation in cirrus clouds, that is, the mechanisms by which ice
8 crystals form on pre-existing aerosol particles.

9

10 The most basic distinction is between homogeneous and heterogeneous freezing.
11 Homogeneous freezing originates from a water solution droplet. Koop et al. (2000)
12 showed that to at least a first approximation homogeneous freezing depends only on
13 water activity and not on the identity of the solute. At equilibrium the water activity of
14 the droplet is equal to the ambient relative humidity, so homogenous freezing should
15 depend only on relative humidity, an important simplification. However, as will be
16 shown below, the number of ice crystals is significantly changed by even small
17 departures from the Koop et al. approximation that all solutes behave the same.

18

19 Heterogeneous freezing is initiated at a solid surface or other interface. The solid may
20 interact with the gas phase for deposition freezing, be inside a droplet for immersion
21 freezing, or be in other configurations. Analysis of the residuals left from evaporated ice
22 crystals shows that heterogeneous freezing is often the dominant mechanism in high

1 altitude cirrus (Cziczo et al., 2013) or that residuals are not fully consistent with either
2 simple heterogeneous or simple homogeneous freezing (Froyd et al., 2010).
3

4 Heterogeneous freezing requires particles with specific compositions, so a key aspect is
5 the availability of such particles. A few heterogeneous nuclei that form ice at low
6 supersaturation can reduce the number of ice crystals compared to homogeneous freezing,
7 whereas many heterogeneous nuclei can enhance the number. Issues such as the
8 competition between homogeneous and heterogeneous freezing and the effect of cooling
9 rate have been studied extensively (DeMott et al., 1997; Jensen and Toon, 1997; Kärcher
10 and Lohmann, 2003; Ren and MacKenzie, 2005; Kärcher et al., 2006; Barahona and
11 Nenes, 2009; Spichtinger and Gierens, 2009c; Spichtinger and Cziczo, 2010). Extremely
12 few heterogeneous nuclei form ice that sediments without a significant effect on the
13 further evolution of the cloud.

14
15 Small-scale temperature fluctuations also strongly affect the number of ice crystals. Ice
16 formation depends on the cooling rate as well as absolute humidity because water uptake
17 by the ice crystals quenches the supersaturation that allows nucleation (Jensen and Toon,
18 1994). Even though their amplitude is small, the speed of small-scale temperature
19 fluctuations creates high cooling rates that can greatly enhance the number of ice crystals
20 (Murphy and Gary, 1995; Hoyle et al, 2005; Jensen et al., 1998; 2013a). The shape and
21 crystal structure of the ice crystals also affects their number and especially the size
22 distribution (Murphy, 2003; Sheridan et al., 2009).

23

1 **2 Model description**

2

3 A parcel model and a one-dimensional model are used for the calculations presented here.

4 The parcel model is an extension of that in Murphy (2003). It tracks nucleation and

5 growth of ice crystals from an initial aerosol size distribution. Ice is tracked in 20 size

6 bins logarithmically spaced between 90 nm and 80 μm. Ice growth includes free

7 molecular, transition, and continuum flow, the Kelvin effect for small particles, and the

8 heat of deposition or evaporation. The model does not include asymmetric growth (Zhang

9 and Harrington, 2014). Ice is initially formed as stacking-disordered (formerly called

10 cubic) ice and anneals to hexagonal ice. Aggregation is not included. Most model

11 parameters are the same as in Murphy (2003). The annealing rate from stacking-

12 disordered to hexagonal ice was reduced by a factor of 10 because subsequent data

13 suggest a lower, albeit more complicated, rate (Murray and Bertram, 2006). The mass

14 accommodation coefficient of water vapor on ice is not known well with recent studies

15 supporting values from less than 0.01 to 0.7 (Magee et al., 2006; Skrotzki et al., 2013)

16 and possibly a range depending on the saturation ratio and crystal axis (Zhang and

17 Harrington, 2014). Here it is set to 0.2 for hexagonal ice and 0.4 for stacking-disordered

18 ice on the grounds that as a metastable phase it must be kinetically easier to form than

19 hexagonal ice. Sensitivity tests were conducted for many of the parameters, including the

20 accommodation coefficient.

21

22 The one-dimensional model tracks ice crystal formation events rather than size bins. The

23 main reason to track events is that it turns out that small amounts of sedimentation are

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1 important for the evolution of the model cloud. By tracking events the ice crystals can
2 sediment by fractions of the vertical model spacing and there is therefore no numerical
3 vertical diffusion of ice crystals (Jensen et al., 2010; Sölch and Kärcher, 2010). Model
4 physics such as the nucleation rate and ice growth equations were the same in the parcel
5 and one-dimensional models. The parcel model includes kinetically-limited water uptake
6 and loss by aerosols.
7

8 Water vapor, temperature and other state parameters were tracked at a fixed vertical
9 spacing. At each time step an event was generated if the integrated probability of
10 nucleation exceeded 0.02 per liter. Thereafter the ice crystal diameter and sedimentation
11 of that particle event were tracked as continuous variables whereas the water vapor for
12 growth (evaporation) was taken from (given to) the nearest bins according to the position.
13 For example, if a growing ice crystal event was exactly in the middle of a bin all the
14 water was taken from that bin whereas if it was at a bin boundary half the water was
15 taken from each bin.

16
17 A detail is that the nucleation probability must be integrated over time and vertical
18 distance in order to make nucleation events independent of the size of a time step or the
19 water vapor grid spacing. When the integral of the nucleation probability reaches a
20 threshold, an event is generated at the most probable location and the integrated
21 nucleation probability reduced by one at that location. As a simple example, consider
22 only three vertical bins in which, because of imposed cooling and water vapor, the
23 probability of nucleation has slowly accumulated to 0.3 per 50 liters in the top and

1 bottom bins and 0.5 in the middle bin. Because the total of 1.1 exceeds one, a nucleation
2 event is generated in the most probable (middle) bin. Following this, the nucleation
3 probability is 0.3 in the top and bottom bins and negative 0.5 in the middle bin. A few
4 time steps later the integrated probability might be 0.6 in the top bin, 0.0 in the middle
5 bin, and 0.5 in the bottom bin and the next event will take place in the top bin. During
6 rapid nucleation, the total probability can increase in a single time step by more than one
7 ice crystal per 50 liters. In that case ice events were initiated in more than one vertical bin
8 and the surface area and mass of the subsequent ice were weighted according to the
9 probability. A typical model run tracked perhaps 20,000 formation events. Ice crystals
10 were formed as stacking-disordered ice and annealed stochastically to hexagonal ice
11 depending on integrated probabilities analogous to those for ice nucleation. For
12 computational reasons, every few time steps ice events with essentially identical sizes
13 and (sedimented) vertical positions were combined.

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15

16 **3 Parcel model results**

17

18 Parcel model simulations were run for frost point temperatures ranging from 185 to 230
19 K. At each temperature 80 simulations were run with various seeds for the random
20 number generator that initializes the fractal small-scale temperature fluctuations. The
21 fluctuations have a Hurst exponent of about 0.7. This gives a realistic short-term
22 autocorrelation. As in Murphy (2003), temperature fluctuations with periods shorter than
23 2 minutes were removed. The amplitude of a fractal depends on the period. For the

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1 simulations here some representative amplitudes were standard deviations of 0.045 K
2 over 10 minutes, 0.20 K over one hour, and 1.33 K over 10 hours. In terms of a power
3 spectrum, one may compare the one and 10 hour variances: $\log_{10}((0.2/1.33)^2) \approx -1.65$
4 for a slope of about $-5/3$. A 3.2K cooling with a 12-hour, half-sine pattern was added to

5 the temperature fluctuations; otherwise many random fluctuations would never generate a
6 model cloud. The same set of temperature fluctuations were repeated at each initial
7 temperature. All simulations were recalculated for a variety of model parameters and
8 assumptions about homogeneous and heterogeneous ice nucleation processes.

9
10 One striking result is that various temperature histories generated a very wide range of ice
11 crystal number densities for the same model assumptions. Figure 1 shows a histogram of
12 the maximum ice number density with homogeneous freezing for temperature histories
13 with ice saturation of 1.25 at 196 K (frost point of 197.4 K) along with a subset of the
14 model temperature trajectories. Jensen et al. (2010) also found wide distributions of ice
15 number density depending on the relative phase of waves creating temperature
16 fluctuations. Low ice number densities can be generated by temperature histories that
17 spend a very short amount of time above the ice nucleation point (Spichtinger and
18 Krämer, 2013). Wide distributions of number density are also evident in various portions
19 of one- and two-dimensional model fields (Lin et al., 2005; Spichtinger and Gierens,
20 2009b).

21
22 The absolute number of ice crystals in the model is sensitive to several of the assumed
23 parameters, especially the accommodation coefficient. However, the results here

1 emphasize trends with temperature and probability distributions due to temperature
2 fluctuations. Such trends and distributions are much less sensitive to model parameters.
3
4 Figure 2 shows results from the parcel model for a range of temperatures. The
5 homogeneous nucleation calculations show many more ice crystals at low temperatures.
6 This is because there is less water vapor at lower temperatures causing slower growth of
7 the ice crystals. With slower growth, more time elapses before the ice surface area grows
8 enough to reduce the supersaturation and suppress further nucleation. With a longer
9 nucleation event, more ice crystals form. This is a very fundamental temperature
10 dependence and occurs when a variety of model parameters are changed (blue lines). The
11 slope of the temperature dependence is similar to Kärcher (2002, Fig. 3) or Spichtinger
12 and Gierens (2009a), but represents the median of a wide distribution rather than a single
13 vertical velocity.

14
15 The inset in Figure 2a explores which are the most sensitive parameters for ice crystal
16 number density. The sensitivity to the upper side of the default parameters is almost a
17 mirror image of those shown. Ice number is extremely insensitive to the absolute
18 nucleation rate. Ice number becomes sensitive to the accommodation coefficient of water
19 at values below about 0.2. Model runs with an accommodation coefficient that became
20 small near saturation ratios of 1.0 (simplified from Zhang and Harrington, 2014) made
21 almost no difference to ice number; the number is determined mostly by the
22 accommodation coefficient near the nucleation threshold. Saturation-dependent
23 accommodation coefficients become important in other situations. Ice number is

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1 moderately sensitive to displacing the homogeneous nucleation point by a small amount
2 of supersaturation. “Faster T fluctuations” means that the fractal temperature fluctuations
3 were smoothed at one rather than 2 minutes.

4
5 Of interest is the sensitivity to the slope of the nucleation rate. The Koop et al. (2000)
6 nucleation rate is a very steep function of relative humidity at the nucleation threshold but
7 is not quite a step function. Artificially increasing or lowering the slope with
8 supersaturation has a much larger impact on the number of ice crystals than the
9 nucleation rate itself. This is potentially important on the side of reducing the slope. If the
10 Koop et al. result that water activity controls freezing is only a first approximation, then
11 particles with different composition should freeze at slightly different supersaturations.

12 High molecular weight polymers have a larger freezing point depression than simple
13 molecules (Zobrist et al., 2003). A small range of freezing activities would be equivalent
14 to a reduced slope with temperature, and the parcel model shows that the ice number is
15 quite sensitive to this. Because the Koop et al. (2000) nucleation rate is extremely steep,
16 the 0.67 slope case shown in Figure 2a means that the relative humidity at freezing is the
17 same within about 1% for particles of various compositions. This is equivalent to a

18 spread in freezing point depression of less than 0.1 K, well within the scatter of
19 experimental data. Yet that reduction in slope reduced the number of ice crystals by about
20 a factor of two. Larger spreads in the water activity at freezing (not shown) led to much
21 larger reductions in the number of ice crystals.

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1 Figure 2b shows results from several heterogeneous cases. As expected, the ice number
2 density is sensitive to the number of heterogeneous nuclei. Also as expected, if there are
3 too few nuclei at very low temperature then the heterogeneous nuclei do not deplete the
4 water vapor enough to prevent homogeneous freezing (Jensen and Toon, 1997; Kärcher
5 and Lohmann, 2003). Figure 2b also compares cases where the heterogeneous nuclei are
6 essentially identical or freeze over a range of relative humidities (“diverse nuclei” in the
7 figure). Over these and other assumptions about heterogeneous freezing, the number of
8 ice crystals always increased at lower temperatures.

9

10 Figure 2c shows some published results on observed ice number density. There are only a
11 few studies since the recognition that ice shattering on aircraft probes invalidates much of
12 the older data (Jensen et al., 2009). There are several points of comparison. First, the
13 results here support the contention in Jensen et al. (2013b) that the high ice number
14 densities observed in thin layers near 190 K can be explained by homogeneous freezing.
15 Second, it is extremely hard in a parcel model to reproduce the low end of the observed
16 ice number densities below about 190 K, regardless of the number of heterogeneous ice
17 nuclei. It might be possible if the accommodation coefficient were unity and other
18 parameters were adjusted, but then the model could not match observations at warmer

19 temperatures. Third, some of the higher ice number densities observed in Krämer et al.
20 (2009) above about 210 K cannot be reproduced by the parcel model. Two possible
21 explanations are outflow cirrus that are not described by the parcel model and ice shatter
22 artifacts, especially at warmer temperatures where there are large crystals more likely to
23 hit the FSSP probe they used (Krämer et al., 2009).

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2 Fourth and most important, neither homogeneous nor heterogeneous freezing in a parcel

3 model can reproduce the observed slope in either Krämer et al. (2009) or Jensen et al.

4 (2012). No simple tuning of the parcel model parameters can change the modeled

5 increase in ice number at low temperature. The slope is a fundamental consequence of

6 less water vapor at lower temperatures. The only way to reverse the slope would be if

7 some parameters such as the number of ice nuclei or the accommodation coefficient were

8 themselves functions of temperature. Even so, the temperature dependence of the

9 parameters would have to be very strong to overcome the basic increase in ice number at

10 lower temperature.

11

12 **4 One-dimensional model**

13

14 The failure of a parcel model to reproduce the observed temperature dependence of ice

15 crystal number suggests that there may be important ice cloud microphysics not captured

16 by a parcel model. The most obvious candidate is sedimentation of ice crystals. At least a

17 one-dimensional model is necessary to investigate sedimentation.

18

19 Figure 3 shows the configuration of the one-dimensional model runs. An initial saturation

20 ratio of 1.1 is imposed except for a subsaturated region near the bottom of the model

21 domain so that falling ice crystals can evaporate. Then one of the same temperature

22 trajectories used in the parcel model is imposed over a 100-meter thick layer with a

23 maximum amplitude at the center of the layer and a smooth transition to zero above and

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1 below that. A slightly supersaturated layer remains below the imposed temperature
2 fluctuation. The right-hand profiles in Figure 3 show the saturation profile just as ice
3 starts to nucleate and grow near the center of the cooled layer, causing a dip in saturation
4 ratio at the 200 m vertical tick.

5

6 Figure 4 shows sample results from the one-dimensional model for one of the imposed
7 temperature profiles. There are differences but more importantly similarities when
8 different assumptions about ice nucleation are made.

9

10 One difference is that the cloud in homogeneous cooling forms later in time because it
11 requires a higher ice supersaturation. Second, the number of ice crystals formed
12 (contours) is much lower when heterogeneous ice nuclei are present. Third, the
13 homogeneous freezing case is less striated than the heterogeneous cases.

14

15 There are important similarities. Despite a range of more than a factor of 20 in peak ice
16 number, the peak ice mass densities are within 40% for the different types of ice
17 nucleation in the three panels. For all cases, layers and fall streaks are generated that are
18 far narrower than the vertical scale of the imposed cooling (Figure 3). On this plot of
19 altitude versus time, the slopes of the fall streaks at around 15000 seconds are also similar,
20 meaning that the fall speeds are similar.

21

22 A final similarity is that in this model the fall streaks are often generated in layers that are
23 slightly displaced from the layers that produce large numbers of ice crystals. This is most

1 visible in the diverse nuclei (bottom) panel but there is also a nearly complete but less
2 visible displacement between high number and fall streak layers in the homogeneous
3 freezing (top) panel. Only with a few good ice nuclei do the most productive nucleation
4 events generate fall streaks, and even then the fall rate of the number density (contours) is
5 perhaps half the fall rate of the mass (color).

6

7 In hindsight there must often be a difference between layers and times that produce high
8 number density and those that produce fall streaks. The high number density ice crystals
9 are small and usually don't fall fast enough to reach supersaturated regions where they
10 can continue to grow. It is worth thinking about how this relates to the probability
11 distributions of the parcel model in Figure 1. Because the imposed cooling has different
12 amplitudes at different altitudes each layer will reach a given nucleation threshold at a
13 different point in the time history. Some will reach it during a rapid jump in temperature,
14 some at a slow point. It is similar to using different temperature histories. Figure 1 shows
15 that there are always a few temperature histories that produce low ice numbers, even in
16 homogeneous freezing that usually creates high number densities.

17

18 These rare freezing events that produce fall streaks are disproportionately important
19 because the falling crystals influence a much larger volume of air than those that don't
20 fall. Exactly how much more important will depend on the initial vertical temperature and
21 humidity profiles. The model does give a key result: these rare low number density
22 events can still generate fall streaks even if they are between or falling through high

1 | number density layers. Conversely, high number density layers can form in between
2 | layers with lower number, a feature that has been observed (Jensen et al., 2013b).
3 |

4 | Figure 5 shows ice number densities from the one-dimensional model analogous to the
5 | parcel model in Figure 2. The ice number densities within the upper, cooled layer follow
6 | the same patterns as the parcel model. This is true for both the temperature dependence of
7 | each case and the relative magnitudes of the various cases. The absolute values are lower
8 | in the one-dimensional model because the vertical averaging includes both high number
9 | density layers and layers with few ice crystals.

10 |

11 | The averaging layer below the cooled region is looking at fall streaks. Here the number
12 | densities are much lower. The heterogeneous cases have a slightly smaller temperature
13 | dependence: a factor of about 5 over the temperature range instead of a factor of 10. The
14 | homogeneous case is completely different below the imposed cooling than within it. It is
15 | not clear if the maximum near 200 K is robust or a feature of the initial profile in the
16 | model. But the absence of very high number densities below 200 K should be robust.

17 | With very little water available at cold temperatures, only regions with few particles can
18 | grow large enough crystals to produce fall streaks. The sensitivity to the accommodation
19 | coefficient is also much smaller in the fall streaks than in the upper layer.

20 |

21 | The one-dimensional model produced remarkably fine vertical structure from a much
22 | smoother temperature profile. A vertical resolution for water vapor concentration of
23 | about 2 meters was found to be necessary even though the falling ice crystals were

1 tracked to fractions of the grid spacing. The generation of fine vertical structure in cirrus
2 clouds has been seen previously. The sedimenting NAT crystals in Fueglistaler et al.
3 (2002) required fine vertical resolution at the base of the cloud. They used 1 cm (!)
4 vertical resolution near cloud base, although no tests were done to see if such fine
5 resolution was necessary (S. Fueglistaler, private communication, 2013). Lin et al.
6 (2005) found that 1-meter vertical spacing was necessary in a cirrus cloud model with
7 broad uplift. Spichtinger and Gierens (2009a) and Jensen et al. (2012) found that 10
8 meters was necessary, again without the boundary conditions changing that rapidly in the
9 vertical.

10

11 There are two reasons for the fine vertical structure. First, ice formation is very sensitive
12 to the cooling rate as a nucleation threshold is crossed, so small variations in temperature
13 history can produce large variations in ice number density. Second,
14 sedimentation acts to amplify small vertical differences in ice crystal size into much
15 larger fall streaks.

16

17 The one-dimensional model is still missing some important physics. Simply looking at
18 cirrus clouds shows that fall streaks are horizontally patchy and curved, neither of which
19 can be captured in a one-dimensional model. Wind shear is important to cirrus clouds
20 (Spichtinger and Gierens, 2009b). Radiation-induced turbulence affects the generation of
21 large particles and fall streaks (Gu and Liou, 2000) and the radiative properties are
22 themselves sensitive to ice crystal size (Stackhouse and Stephens, 1991). Neither process
23 is easy to capture in one dimension.

1

2 The vertical profiles of saturation and cooling used here in the one-dimensional model
3 are obviously arbitrary. Trying to find a more realistic profile is limited both by the
4 inherent limitations of working in one dimension and a lack of observations. There are
5 few observations of upper tropospheric temperatures on time scales of a minute and
6 horizontal scales well below a kilometer. Even when an aircraft can measure a slice of
7 temperature profiles, translating the observed temperatures into those experienced by an
8 air parcel depends on unmeasured phase relationships between waves of different
9 frequencies (Bacmeister et al., 1999). Similar issues can arise for ground-based remote
10 sensing of a single point.

11

12

13 **5 Discussion and conclusions**

14

15 The one-dimensional model results presented here are in many respects similar to a
16 model of nitric acid trihydrate (NAT) crystals in the polar stratosphere (Fueglistaler et al.,
17 2002). In an environment with much slower crystal growth than even the coldest cirrus
18 cloud, Fueglistaler et al. found that sedimentation out of a “mother cloud” into a
19 supersaturated layer could produce a few large NAT crystals. The number of sedimenting
20 NAT crystals was insensitive to assumptions about the nucleation mechanism. The reason
21 was the supersaturated region below the cloud. Any crystal that managed to make its way
22 into this layer grew and accelerated down from the cloud. The physics of those falling
23 crystals was determined by mixing at the bottom of the cloud and the growth conditions

1 below the cloud, not the nucleation conditions in the cloud. This work suggests that a
2 similar process often occurs for cirrus clouds. Besides the bottom of the cloud, this work
3 suggests that the variety of temperature histories leads to some cloud layers with low ice
4 number density, and these layers also produce falling crystals. Model runs at a pressure
5 and temperature characteristic of polar stratospheric clouds show similar processes to the
6 tropopause conditions shown here.

7

8 There is a fundamental difference in sedimentation from cirrus clouds compared to warm
9 liquid clouds. The air around a warm cloud is subsaturated whereas the high
10 supersaturations required for ice nucleation mean that the air around a newly formed
11 cirrus cloud is supersaturated. Mixing and entrainment at the base or edges of a warm
12 cloud causes evaporation of water droplets. In contrast, mixing and entrainment at the
13 base or edges of a cirrus cloud causes growth of ice crystals near the edge while
14 simultaneously reducing their number density and hence competition for vapor. This can
15 induce sedimentation from the base and periphery of the cloud, very unlike a warm cloud.

16 Large areas surrounding cirrus clouds in the upper troposphere are supersaturated (e.g.
17 Vay et al., 2000; Ström et al., 2003; Krämer et al., 2009; Diao et al., 2013).

18

19 Sedimentation has several roles in cold cirrus clouds (Spichtinger and Gierens, 2009b).
20 Depending on the environment, sedimentation can limit (Kärcher, 2002) or extend (Luo
21 et al., 2003) the lifetime of very thin cirrus clouds. For some ranges of temperature
22 fluctuations a balance can be achieved between nucleation and sedimentation (Barahona

Dan Murphy 8/18/2014 10:41 AM
Deleted: (Diao et al., 2013).

1 and Nenes, 2011). Here, sedimentation is a mechanism for amplifying the importance of
2 conditions that produce only a few ice crystals.

3

4 The very low ice number densities sometimes observed below 200 K can be produced in
5 fall streaks, as also found by Jensen et al. (2012). The picture developed here of a cold
6 cirrus cloud (below perhaps 210 K) is that the reason aircraft observations usually
7 measure low ice number density is that the falling crystals sweep out a much larger
8 volume than the ones that stay put. Jensen et al. (2013a) suggested that a few
9 heterogeneous nuclei could initiate fall streaks even if there aren't enough such nuclei to
10 suppress homogeneous nucleation. The model here generalizes that: fall streaks are
11 produced by any time or place that makes a few large particles. These regions of few
12 crystals might be from heterogeneous nucleation, temperature blips that happened to
13 produce only a few ice crystals from homogeneous nucleation, or mixing at the bottom or
14 edge of a cloud. The number and mass of ice crystals in such fall streaks is fairly
15 insensitive to how they were produced.

16

17 There may be other observational tests of the importance of sedimentation besides
18 comparing to observed ice number densities. Using statistics on the size of regions
19 containing supersaturation and/or ice crystals, Diao et al. (2013) estimated that mature or
20 evaporating cirrus are about seven times more prevalent than nucleation regions.

21 Although their mature and evaporations classifications are not exactly comparable to fall
22 streaks, it does support the notion that throughout most of a cirrus cloud the number of
23 ice crystals is not determined by nucleation at that spot. A lidar with high spatial

1 resolution could probably observe whether the fall streaks are originating from locations
2 slightly displaced from the high number density layers. Such an observation might
3 constrain the nucleation mechanism: in the model, only nucleation on a few good ice
4 nuclei created fall streaks coincident with the highest number densities.

5

6 At least for homogeneous freezing, the one-dimensional model predicts a wide range in
7 ice number density between various regions of the cloud. In that case the visible
8 reflectance could still be determined by the zones of numerous, small ice crystals that
9 scatter light more efficiently even though other properties such as median ice number
10 density were determined mostly by sedimentation.

11

12 Finally, the results here contain both bad and good news for large-scale modeling of
13 cirrus clouds. The bad news is that the requirement for extremely fine vertical resolution
14 implies that even mesoscale models have far too coarse of resolution to directly model
15 cirrus clouds. The importance of sedimentation from rare temperature trajectories implies
16 that parameterizations based on parcel models (e.g. Kärcher and Lohmann, 2003;
17 Barahona and Nenes, 2009) probably miss essential physics.

18

19 The good news for large-scale modeling is that in the one-dimensional model the ice
20 number density below cloud is much less sensitive to details of the microphysics than in a
21 parcel model. There are new parameters required to describe sedimentation, such as the
22 vertical extent of any supersaturated region below the cloud. But such parameters should
23 be amenable to calculation by a three-dimensional model, although the required vertical

- 1 resolution (perhaps tens of meters) is still more suitable for a local model than a global
- 2 model.

1

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3

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5

6 **References**

7

8 Bacmeister, J. T., Eckermann, S. D., Tsias, A., Carslaw, K. S., and Peter, T., Mesoscale
9 temperature fluctuations induced by a spectrum of gravity waves: A comparison of
10 parameterizations and their impact on stratospheric microphysics. *J. Atmos. Sci.*, 56,
11 1913–1924, 1999.

12 Barahona, D., and Nenes, A., Parameterizing the competition between homogeneous and
13 heterogeneous freezing in ice cloud formation – polydisperse ice nuclei, *Atmos. Chem.*
14 *Phys.*, 9, 5933–5948, 2009.

15 Barahona, D., and A. Nenes, Dynamical states of low temperature cirrus, *Atmos. Chem.*
16 *Phys.*, 11, 3757–3771, 2011.

17 Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diao, M., Zondlo, M. A., Smith, J.
18 B., Twohy, C. H., and Murphy, D. M., Clarifying the dominant sources and
19 mechanisms of cirrus cloud formation, *Science*, 340, 1320-1324, 2013.

20 [DeMott, P. J., D. C. Rogers, and S. M. Kreidenweis, The susceptibility of ice formation](#)
21 [in upper tropospheric clouds to insoluble aerosol components, *J. Geophys. Res.*, 102,](#)
22 [19,575-19,584, 1997.](#)

1 Diao, M., Zondlo, M. A., Heymsfield, A. J., Beaton, S. P. and Rogers, D. C., Evolution of
2 ice crystal regions on the microscale based on in situ observations, *Geophys. Res. Lett.*,
3 40, 3473-3478, 2013.

4 [Froyd, K. D., D. M. Murphy, P. Lawson, D. Baumgardner, and R. L. Herman, Aerosols](#)
5 [that form subvisible cirrus at the tropical tropopause, *Atmos. Chem. Phys.*, 10, 209-218,](#)
6 [2010.](#)

7 Fu, Q., and Liou, K. N., Parameterization of the radiative properties of cirrus clouds, *J.*
8 *Atmos. Sci.*, 50, 2008-2025, 1993.

9 Fueglistaler, S., Luo, B. P., Voigt, C., Carslaw, K. S., and Peter, Th., NAT-rock
10 formation by mother clouds: a microphysical model study, *Atmos. Chem. Phys.*, 2, 93-
11 98, 2002.

12 Hoyle, C. R., Luo, B. P., and Peter, T., The origin of high ice crystal number densities in
13 cirrus clouds, *J. Atmos. Sci.*, 62, 2568-2579, 2005.

14 Gensch, I. V., Bunz, H., Baumgardner, D. G., Christensen, L. E., Fahey, D. W., Herman,
15 R. L., Popp, P. J., Smith, J. B., Troy, R. F., Webster, C. R., Weinstock, E. M., Wilson,
16 J. C., Peter, T., and Krämer, M., Supersaturations, microphysics, and nitric acid
17 partitioning in a cold cirrus cloud observed during CR-AVE 2006: an observation-
18 modeling intercomparison study, *Environ. Res. Lett.*, 3, 1-9, 2008.

19 Gu, Y., and Liou, K. N., Interactions of radiation, microphysics, and turbulence in the
20 evolution of cirrus clouds, *J. Atmos. Sci.*, 57, 2463-2479, 2000.

21 [Jensen, E. J., and Toon, O. B., Ice nucleation in the upper troposphere: Sensitivity to](#)
22 [aerosol number density, temperature, and cooling rate, *Geophys. Res. Lett.*, 21, 2019-](#)
23 [2022, 1994.](#)

1 Jensen, E. J., and Toon, O. B., The potential impact of soot particles from aircraft exhaust
2 on cirrus clouds, *Geophys. Res. Lett.*, 24, 249-252, 1997.

3 Jensen, E. J., Toon, O. B., Tabazadeh, A., Sachse, G. W., Anderson, B. E., Chan, K. R.,
4 Twohy, C. W., Gandrud, B., Aulenbach, S. M., Heymsfield, A. J., Hallett, J., and Gary,
5 B., Ice nucleation processes in upper tropospheric wave-clouds observed during
6 SUCCESS, *Geophys. Res. Lett.*, 25, 1363-1366, 1998.

7 Jensen, E. J., Lawson, P., Baker, B., Pilon, B., Mo, Q., Heymsfield, A. J., Bansemer, A.,
8 Bui, T. P., McGill, M., Hlavka, D., Heymsfield, G., Platnick, S., Arnold, G. T., and
9 Tanelli, S., On the importance of small ice crystals in tropical anvil cirrus, *Atmos.*
10 *Chem. Phys.*, 9, 5519-5537, 2009.

11 Jensen, E. J., Pfister, L., Bui, T.-P., Lawson, P., and Baumgardner, D., Ice nucleation and
12 cloud microphysical properties in tropical tropopause layer cirrus, *Atmos. Chem. Phys.*,
13 10, 1369-1384, 2010.

14 Jensen, E. J., Pfister, and L., Bui T. P., Physical processes controlling ice concentrations
15 in cold cirrus near the tropical tropopause, *J. Geophys. Res.*, 117, D11205,
16 doi:10.1029/2011JD017319, 2012.

17 Jensen, E. J., Lawson, R. P., Bergman, J. W., Pfister, L., Bui, T. P., and Schmitt C. G.,
18 Physical processes controlling ice concentrations in synoptically forced, midlatitude
19 cirrus, *J. Geophys. Res.*, 118, 5348–5360, doi:10.1002/jgrd.50421, 2013a.

20 Jensen, E. J., Diskin, G., Lawson, R. P., Lance, S., Bui, T. P., Hlavka D., McGill, M.,
21 Pfister, L., Toon, O. B., and Gao, R., Ice nucleation and dehydration in the Tropical
22 Tropopause Layer, *Proc. Natl. Acad. Sci. USA*, 110, 2041-2046, 2013b.

- 1 Kärcher, B., Properties of subvisual cirrus clouds formed by homogeneous freezing,
2 *Atmos. Chem. Phys.*, 2, 161-170, 2002.
- 3 Kärcher, B., and Lohmann U., A parameterization of cirrus cloud formation:
4 heterogeneous freezing, *J. Geophys. Res.*, 108, 4402, doi:10.1029/2002JD003220,
5 2003.
- 6 [Kärcher, B., J. Hendricks, and U. Lohmann, Physically based parameterization of cirrus](#)
7 [cloud formation for use in global atmospheric models, *J. Geophys. Res.*, 111, D01205,](#)
8 [doi:10.1029/2005JD006219, 2006.](#)
- 9 Koop, T., Luo, B., Tsias, A., and Peter, T., Water activity as the determinant for
10 homogeneous ice nucleation in aqueous solutions, *Nature*, 406, 611-614, 2000.
- 11 Krämer, M., Schiller, C., Afchine, A., Bauer, R., Gensch, I., Mangold, A., Schlicht, S.,
12 Spelten, N., Sitnikov, N., Borrmann, S., de Reus, M., and Spichtinger, P., Ice
13 supersaturations and cirrus cloud crystal numbers, *Atmos. Chem. Phys.*, 9, 3505-3522,
14 2009.
- 15 Lin, R.-F., Starr, D. O., Reichardt, J., and DeMott, P. J., Nucleation in synoptically forced
16 cirrostratus, *J. Geophys. Res.*, 110, D08208, doi:10.1029/2004JD005362, 2005.
- 17 Liou, K.-N., Influence of cirrus clouds on weather and climate processes: A global
18 perspective, *Monthly Weather Rev.*, 114, 1167-1199, 1986.
- 19 Luo, B. P., Peter, Th., Wernli, J., Fueglistaler, S., Wirth, M., Kiemle, C., Flentje, H.,
20 Yushkov, V. A., Khattatov, V., Rudakov, V., Thomas, A., Morrmann, S., Toci, G.,
21 Massinghi, P., Beuermann, J., Schiller, C., Cairo, F., Di Don-francesco, G., Adriani,
22 A., Volk, C. M., Ström, J., Noone, K., Mitev, V., MacKenzie, R. A., Carslaw, K. S.,
23 Trautmann, T., Santacesaria, V., and Stefanutti, L., Ultrathin Tropical Tropopause

1 Clouds (UTTCs): II. Stabilization mechanisms, *Atmos. Chem. Phys.*, 3, 1093-1100,
2 2003.

3 [Magee, N., A. M. Moyle, and D. Lamb, Experimental determination of the deposition](#)
4 [coefficient of small cirrus-like ice crystals near -50 C, *Geophys. Res. Lett.*, 33, L17813,](#)
5 [doi:10.1029/2006GL026665, 2006.](#)

6 McFarquhar, G. M., Heymsfield, A. J., Spinhirne, J., and Hart, B., Thin and subvisual
7 tropopause tropical cirrus: Observations and radiative impacts, *J. Atmos. Sci.*, 57,
8 1841-1853, 2000.

9 Murphy, D. M., and Gary, B. L., Mesoscale temperature fluctuations and polar
10 stratospheric clouds, *J. Atmos. Sci.*, 52, 1753–1760, 1995.

11 Murphy, D. M., Dehydration in cold clouds is enhanced by a transition from cubic to
12 hexagonal ice, *Geophys. Res. Lett.*, 30, 2230, doi:10.1029/2003GL018566, 2003.

13 Murray, B. J., and Bertram A. K., Formation and stability of cubic ice in water droplets,
14 *Phys. Chem. Chem. Phys.*, 8, 186-192, 2006.

15 [Ren, C., and A. R. MacKenzie, Cirrus parameterization and the role of ice nuclei, *Q. J. R.*](#)
16 [Meteorol. Soc., 131, 1585–1605, 2005.](#)

17 Sheridan, L., Harrington, J. Y., Lamb, D., and Sulia, K., Influence of ice crystal aspect
18 ratio on the evolution of ice size spectra during vapor depositional growth, *J. Atmos.*
19 *Sci.*, 66, 3732-3743, 2009.

20 [Skrotzki, J. P. Connolly, M. Schnaiter, H. Saathoff, O. Möhler, R. Wagner, M. Niemand,](#)
21 [V. Ebert, and T. Leisner, The accommodation coefficient of water molecules on ice –](#)
22 [cirrus cloud studies at the AIDA simulation chamber, *Atmos. Chem. Phys.*, 13, 4451-](#)
23 [4466, 2013.](#)

1 Sölch, I., and Kärcher, B., A large-eddy model for cirrus clouds with explicit aerosol and
2 ice microphysics and Lagrangian ice particle tracking, *Q. J. R. Meteorol. Soc.*, 136,
3 2074-2093, 2010.

4 Spichtinger, P., and Gierens, K. M., Modelling of cirrus clouds – Part 1a: Model
5 description and validation, *Atmos. Chem. Phys.*, 9, 685-706, 2009a.

6 Spichtinger, P., and Gierens, K. M., Modelling of cirrus clouds – Part 1b: Structuring
7 cirrus clouds by dynamics, *Atmos. Chem. Phys.*, 9, 707-719, 2009b.

8 Spichtinger, P., and Gierens, K. M., Modelling of cirrus clouds – Part 2: Competition of
9 different nucleation mechanisms, *Atmos. Chem. Phys.*, 9, 2319-2334, 2009c.

10 [Spichtinger, P. and D.J.Cziczo, Impact of heterogeneous ice nuclei on homogeneous](#)
11 [freezing events in cirrus clouds. *J. Geophys. Res.*, 115, D14208,](#)
12 [doi:10.1029/2009JD012168, 2010.](#)

13 Spichtinger, P., and Krämer, M., Tropical tropopause ice clouds: a dynamic approach to
14 the mystery of low crystal numbers, *Atmos. Chem. Phys.*, 13, 9801–9818, 2013.

15 [Stackhouse, P. W., and G. L. Stephens, A theoretical and observational study of the](#)
16 [radiative properties of cirrus: Results from FIRE 1986, *J. Atmos. Sci.*, 48, 2044-2059,](#)
17 [1991.](#)

18 [Ström, J., M. Seifert, B. Kärcher, J. Ovarlez, A. Minikin, J.-F. Gayet, R. Krejci, A.](#)
19 [Petzold, F. Auriol, W. Haag, R. Busen, U. Schumann, and H. C. Hansson, Cirrus](#)
20 [cloud occurrence as function of ambient relative humidity: a comparison of](#)
21 [observations obtained during the INCA experiment, *Atmos. Chem. Phys.*, 3, 1807-](#)
22 [1816, 2003.](#)

1 [Vay, S. A., B. E. Anderson, E. J. Jensen, G. W. Sachse, J. Ovarlez, G. L. Gregory, S. R.](#)
2 [Nolf, J. R. Podolske, T. A. Slate, and C. E. Sorens, Tropospheric water vapor](#)
3 [measurements over the North Atlantic during the Subsonic Assessment Ozone and](#)
4 [Nitrogen Oxide Experiment \(SONEX\), *J. Geophys. Res.*, 105, 3745–3755, 2000.](#)

5 Wang, P.-H., McCormick, M. P., Poole, L. R., Chu, W. P., Yue, G. K., Kent, G. S., and
6 Skeens, K. M., Tropical high cloud characteristics derived from SAGE II extinction
7 measurements, *Atmos. Res.*, 34, 53-83, 1994.

8 [Zhang, C., and J. Y. Harrington, Including surface kinetic effects in simple models of ice](#)
9 [vapor diffusion, *J. Atmos. Sci.*, 71, 372-390, 2014.](#)

10 [Zobrist, B., U. Weers, and T. Koop, Ice nucleation in aqueous solutions of poly\[ethylene](#)
11 [glycol\] with different molar mass, *J. Chem. Phys.*, 118, 10254-10261, 2003.](#)

12

1

2 Figure 1. Histogram of ice number density generated by homogeneous freezing in a
3 parcel model. Also shown are the first 8 of the 80 temperature trajectories, all with same
4 amplitude and fractal slope of temperature fluctuations. Doubling the accommodation
5 coefficient of water on the growing ice crystals systematically reduces the number
6 density, as does assuming that there is a very small spread in the water activity at freezing.
7 However, the phase and slope of small-scale fluctuations near the homogeneous freezing
8 threshold creates a histogram that is much wider than the effect of changing this or other
9 model parameters.

10

11 Figure 2. Parcel model results for the temperature dependence of ice crystal number
12 density along with results from observations. In (b), “good” heterogeneous nuclei freeze
13 at an ice supersaturation of 1.3 ± 0.02 whereas “diverse” nuclei are uniformly distributed
14 between supersaturations of 1.3 and 1.65. (c) shows observations described in Gensch et
15 al., 2008, Krämer et al., 2009, Jensen et al., 2010, and Jensen et al., 2013b. Ranges in (c)
16 are approximated from the data in the Jensen et al. papers. In this temperature range
17 homogeneous freezing occurs about 3 K below the frost point.

18

19 Figure 4. One-dimensional model results for three assumptions about ice nucleation and
20 the same imposed cooling. Contours show ice crystal concentration in liter^{-1} . Color shows
21 ice mass concentration with the same scale for all three panels.

22

1

2 Figure 3. Vertical profiles showing how the one-dimensional model was initialized and
3 cooling imposed. The imposed cooling used a smooth vertical profile over 100 meters but
4 fluctuated in time. One can picture the half-sine temperature perturbation wiggling with
5 an amplitude that followed one of the temperature trajectories in Figure 1.

6

7 Figure 5. Ice number densities in the one-dimensional model. The values are the median
8 over 20 temperature histories of the maximum in time averaged over vertical range either
9 in or below the layer with imposed cooling. These ranges are indicated in Figure 4. The
10 axis ranges are the same as in Figure 2.

11