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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The two faces of cirrus clouds" *by* D. Barahona and A. Nenes

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We thank Dr. Murray for his thorough, constructive and thoughtful comments.

1) My primary comment is concerned with the conclusion that heterogeneous nucleation cannot explain the low ice numbers in cold cirrus. This conclusion is arrived at by varying the updraft rates (without gravity waves) up to 100 cms-1. Slow updrafts are representative of synoptic cooling and faster ones are representative of the phase of gravity waves in which there is upward movement of air. They show that for glassy aerosol at updrafts larger than 15 cms-1 insufficient ice crystals nucleate to prevent the saturation rising to the homogeneous threshold which leads to high ice densities. This is in good agreement with Murray et al. (Murray et al., 2010) who showed that only a small number of ice crystals formed below 15 cms-1 and a much larger numbers form above this threshold as homogeneous nucleation becomes viable. This simplified

model employing a constant updraft over the duration of the model run is appropriate for modelling synoptic cooling, but it is not sensible to approximate the effect of a gravity wave as a constant cooling. A parcel of air influenced by a gravity wave will only experience rapid updraft for a time within the cycle of the wave. The mean amplitude in temperature associated with these waves, according to B&N, is up to a degree. This would correspond to just 10-20% in RHi. Hence the model employing a constant cooling over many degrees (and many10's % in RHi) is not representative of a gravity wave.

The ice crystal concentration (N_c) is strongly influenced by $\frac{dS}{dt}$ in the vicinity of the maximum saturation ratio S_{max} and only weakly dependent on S (Barahona and Nenes, 2008; Kärcher and Lohmann, 2002; Spichtinger and Gierens, 2009). The results presented in Figure 2 are not influenced by the initial conditions selected as it is not necessary for the wave to cool over many degrees; it would be enough to increase $\frac{dS}{dt}$ in the vicinity of S_{max} which is quite likely given the ubiquitous character of temperature fluctuations in the upper troposphere (Bacmeister et al., 1999; Sato, 1990). The parameterization used in generating Figures 1 and 2 (Barahona and Nenes, 2009) take this into account as it calculates the size of the ice crystals "backwards", that is, from the point of the maximum supersaturation to the point of freezing and therefore is not influenced by initial conditions.

This is now clarified in the text.

A more accurate picture is a synoptic cooling with gravity wave driven temperature perturbation superimposed on it (as is described later in the paper when only homogeneous nucleation is considered). In this case and in the presence of glassy aerosol, ice particles would be produced as soon as S > 1.2 during the cold phase of a wave. These particles would then have time to grow and deplete water vapour during the warm phase of the wave. The extent of the depletion would depend on the number and size of the crystals and the time that they exist in a supersaturated environment. The number of ice particles which nucleate in the next cold phase of the wave would de-

pend on a number of factors including the extent of depletion. Given a sufficient synoptic cooling rate homogeneous nucleation may be possible in addition to heterogeneous nucleation. This synoptic cooling with the effect of a gravity wave superimposed is very different to a situation in which there is a constant and very rapid cooling.

Observations suggest that the gravity wave spectrum of the upper troposphere is not described by a single wave but instead by the superposition of gravity waves of different source (Bacmeister et al., 1999). The approach described by the reviewer may therefore leave out important interactions in the formation of cirrus. We have proposed a new approach (as presented in Section 3) that considers the full spectrum, and arguably describes a more complete picture of cirrus.

Hence, on the basis of their constant cooling model run B&N should not conclude that heterogeneous nucleation cannot produce low ice number densities based on this model.

We did not say that heterogeneous freezing of glassy aerosol cannot produce low crystal numbers, but that it is not necessary. This is important, especially given the prevailing view that pure homogeneous freezing is always associated with high N_c. Thus the section makes an important point towards justifying the new approach and we prefer to keep it. We have rewritten several parts of the section to clarify the objective of the section.

2) Title: I suggest replacing 'two faces' with 'two dynamical states'. When I first saw the title I thought the paper was about the two crystalline phases of hexagonal ice. After reading the abstract I then understood, but the authors may well miss interested readers!

This is a good point. We have changed the title to avoid confusion.

3) It would be helpful if the authors would explicitly state which region(s) of the atmosphere they are interested in. They refer to cold cirrus, but many of the citations are

focused specifically on the TTL and the low ice density issue is certainly a TTL issue. I think the paper is primarily about the TTL, but this is not stated clearly.

The introduction has been expanded where it is made clear that cold thin cirrus are typically found at the TTL.

4) In the abstract and conclusions it is stated very boldly that our understanding of cirrus in climate change is reshaped. Are the types of cloud discussed here, subvisible, cirrus in the TTL region, really that critical for climate to warrant such a bold statement? other cirrus types in lower and warmer regions are far more important for climate.

Our conclusions are not limited to TTL cirrus. We tested our model over a wide range of temperature and cloud formation conditions not only those of TTL-cirrus. In our analysis, two main states in the evolution of cirrus are identified and it is the surrounding dynamic conditions what determines which state is preferred. Neither the cirrus states nor the view presented in this work have been proposed before.

5)*P30858. 'It is well known that . . ..primarily by homogeneous'. I do not think this is true. Many studies show that heterogeneous nucleation is also important. In fact, DeMott et al. (2003) which is cited to back up this statement show that there are a lot of heterogeneous nuclei in cirrus regions.*

True, however homogeneous freezing is likely the main mechanism of ice crystal production in cirrus between 200 K and 235 K. This is because N_c (typically around 0.1-1 cm^{-3}) in cirrus is typically much higher than measured N_{IN}(typically around 0.01-0.1 cm^{-3}), the homogeneous freezing threshold is generally reached (DeMott et al., 2003; Krämer et al., 2009), and ammonium sulfate aerosol is widely available in the upper troposphere. We acknowledge however that heterogeneous freezing can impact cloud formation in polluted and dust-rich regions and have been rephrased the statement accordingly.

6) Use of 'freeze' throughout the paper. Freeze is by definition the transition from *a liquid to a solid. Nucleation of ice and crystallisation of solution droplets is freezing. Deposition nucleation of ice onto a crystalline ammonium sulphate particle is not freezing. I initially thought the authors were talking about immersion mode heterogeneous freezing, but they seem to be misusing the word freeze. This needs to be corrected throughout the manuscript.*

We have corrected this throughout paper and used the word nucleation when referring to the formation of ice on glassy and ammonium sulfate IN.

7) P30861. Ln 7. Delete 'typically'. This is based on very limited measurements.

Done.

8) P30862, ln 1. Insert 'on sulphate' after 'heterogeneous freezing'. There needs to be a stronger distinction between the heterogeneous nucleation on sulphate and on glass.

The statement has been rewritten.

9) 30862, ln 1-3. 30-70% supersaturations. Froyd et al is an inappropriate reference for this, their paper is on the composition of aerosol in the TTL. Also, I do not understand where these numbers come from. Kramer et al.'s figure 3 suggests S = 0.3 - 2.0 below 200 K and fig 7 suggests that the most probably S is _1 until you get to the very lowest temperature. There are other measurements which suggest larger in cloud S. However, the uncertainty in the water data in general is too high to rule out heterogeneous nucleation and should not be done. This section needs to be revised.

The statement is referring to the maximum supersaturation which is close to the freezing threshold of the aerosol population and set by the clear sky cut-off relative humidity observed (Haag et al., 2003). We have rephrased the statement to avoid confusion.

10) 30862, ln 7-15. B&N suggest that heterogeneous nucleation on glassy aerosol is inconsistent with Kramer et al.'s saturation data, because they suggest S would be mostly below 30%. As mentioned above Kramer et al. show that the most likely S between 185 and 200 K to be around 1, and only below this does S increase. Hence,

this does not support B&N's argument.

Predominance of heterogeneous freezing on glassy aerosol would imply S below 30%. If heterogeneous freezing is the only mechanism occurring, clear-sky RH above 130% would be rarely observed (as any additional supersaturation would be removed by the growing crystals). If N_{IN} is too low to efficiently remove supersaturation by heterogeneous freezing then S would still have to remain below S_{hom} for N_c to remain low unless homogeneous freezing is suppressed. The fact that observations (e.g., Krämer et al., 2009; Selkirk et al., 2010) commonly show limiting RH values around 150-180% indicates that heterogeneous freezing does not completely deplete supersaturation and homogeneous freezing likely occurs. This is now clarified in the text.

11) Gravity wave spectrum. How well known is this? I understand that gravity waves are very hard to measure and characterise and our knowledge is limited. What B&N have done in this respect seems sensible, but I worry that the conclusions drawn from this 'shaky foundation' are a little too confident. The paper is written in a very confident style and I think it needs to be accepted that there are still questions and problems and the style needs to be set accordingly

We have already acknowledged that the gravity wave spectrum used is representative and that variations in the spectrum shape affect the evolution of the cloud near orographic sources. However we have varied δTover a wide range to test the sensitivity to the spectrum parameters and it is not likely that our conclusions would be significantly changed by deviations from a Gaussian/Fourier series form.

12) I would have liked to have seen a more comprehensive review of what is known about gravity waves in the TTL region and why this particular parameterisation is suitable.

Excellent point. Done.

13) Conflict with Jensen and Pfister (2004). J&P state that the effect of gravity waves *is to increase the ice number density. Since B&N based their gravity wave spectrum on J&P (at least in part), they need to discuss this apparent discrepancy.*

Our approach is different from the one used in Jensen and Pfister (2004). In this scheme cloudy parcels behave as isolated from each other. Our model takes into account the history of individual parcels allowing previous freezing events to interact with new ones trough changes in the ice crystal population. We have expanded the introduction section in the paper where the results of several studies (including Jensen and Pfister (2004)) is discussed.

14) 30877 ln 11. Why are clouds in the dynamic equilibrium region less sensitive to IN? As far as I can see the effect of changing IN and introducing heterogeneous IN have not been explored in the wave model.

The conclusion comes directly from the tendency of the system in equilibrium to self-regulate. We however agree that the question of IN effects on cirrus evolution is very important, and have expanded our model to include heterogeneous nucleation. Results of simulations with heterogeneous nucleation active are now presented in Figure 8.

15) The 'Pulse –decay' and 'dynamic equilibrium' terminology. I find this terminology a little confusing. If you look at Fig3a and b, the curves for the dynamic equilibrium regime look like pulse decays. I know you are not referring to this, but it is confusing.

The terminology reflects the fate of the cirrus cloud under the different regimes and is mean to distinguish a cloud that is self-regulated from one that decays over time.

16) What are the in-cloud saturation values predicted by your model? How do these compare with the field data. I can only see a plot where S-1 is averaged over the layer.

The cloud layer refers to the cloud itself, so all values are in-cloud values.

Technical comments:

1) P30865. Ln 15. Symbol 'x' should be non-bold and italic. Done

2) Fig 1 caption. 'lines' needs to be 'line' in several places. Done.

3) Fig 8. I wondered if the two axes might be switched on this plot to be consistent with the preceding plots. Done

4) P 30874 ln 13. Are these ammonium sulphate particles solid or liquid? The statement now reads, "composed of deliquesced ammonium sulfate"

5) P30860 ln 29. Should 'depressed' be 'suppressed'? Corrected

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