Responses to 2nd Referee's Comments

General comments

This paper presents important results showing the potential impact of mineral dust on cirrus clouds using two ice nucleation schemes in CAM5. Suggestions are made for presenting the climate impact results more clearly. The paper is well written and organized with figures of high quality. It is definitely worthy of being published in ACP.

→Reply: We thank the reviewer for the encouraging comments.

It is always helpful to relate model results to observations. The observational study by Haag et al. (2003, ACP) and perhaps other INCA papers are relevant to the results presented in Figs. 2-4. Using RHi distributions, Haag et al. showed that NH midlatitude cirrus likely resulted from a combination of homo- and heterogeneous nucleation processes, whereas SH mid-latitude cirrus were dominated by homogeneous nucleation. These INCA results appear to support the LP parameterization more than the BN scheme, and this should be mentioned.

→Reply: We thank the reviewer for pointing out the study by Haag et al. (2003) as an observational constrain of ice nucleation mechanisms. Following the reviewer's comment, we added the following sentences in the revised manuscript when we discuss the results presented in Figures 2-4: "Using the probability distributions of RHi measured during the INCA campaign, Haag et al. (2003) showed that NH mid-latitude in situ cirrus clouds were likely formed from a combination of homogeneous and heterogeneous nucleation processes, whereas SH mid-latitude cirrus clouds were dominated by homogeneous nucleation. These INCA results appear to support the LP05 parameterization more than the BN09 scheme with the heterogeneous IN spectrum of Phillips et al. (2008)".

Since differences between the LP and BN schemes result primarily from differences in the way ice nuclei are predicted from the aerosol size distributions (note dust concentrations are roughly equal for both LP and BN simulations), some discussion regarding the differences between the CNT method and PDA08 method appears warranted.

→Reply: Following the reviewer's comment, we added a discussion regarding the differences between the CNT method and PDA08 method in section 1 (Introduction) in the revised manuscript: "CNT is a statistical method which parameterizes the rate of heterogeneous ice nucleation as a function of ambient conditions such as temperature and vapor pressure, and properties of the ice nuclei (such as size, contact angle of ice germ on the substrate, and activation energy). The empirical method of Phillips et al. (2008) was developed from the IN measurements by the Colorado State University (CSU) Continuous Flow Diffusion Chamber (CFDC), and derived IN number concentration as a function of surface area densities of aerosol species (mineral dust, black carbon and hydrophobic organics) in addition to air temperature and ice supersaturation. Ice crystal number concentration predicted from CNT is much higher than that from Phillips et al. (2008), when both methods were tested in the same cloud parcel model framework for the same assumed aerosol distribution (Eidhammer et al., 2009)".

As discussed in Section 2.3, first paragraph, the subgrid variability of the updraft velocity (w) in CAM5 used for driving the LP05 parameterization has an upper threshold limit of 20 cm s-1. This was done to decrease ice particle number concentrations Ni, which in turn increased ice particle effective size De, bringing De into closer agreement with measurements during the development of CAM5. This improved the realism of ice cloud-radiation interactions in CAM5. However, it did not change the over-prediction of w by the Bretherton-Park moist turbulence-convection scheme. Thus, w tends to be near 20 cm s-1 much of the time in CAM5, and its mean value is likely high relative to observations. This in turn may produce anomalously high supersaturations with respect to ice (RHi), and this high RHi bias may induce a homogeneous nucleation bias. That is, it may take relatively high concentrations of ice nuclei (IN) to prevent RHi in cirrus cloud updrafts from reaching threshold RHi values at which homogeneous nucleation occurs. These points should be mentioned in the paper.

As the reviewer points out, the subgrid variability of the updraft velocity (*w*) in CAM5 used for driving the ice nucleation parameterizations has an upper threshold limit of 20 cm s^{-1}. As a result, *w* tends to be near 20 cm s^{-1} much of the time in CAM5. Our analysis of *w* measured in the INCA and SPartICus campaigns shows that the measured mean *w* can be higher than this *w* upper limit of 20 cm s^{-1} used in CAM5 (Zhang et al., in preparation, 2012). This upper limit is also lower than that seen in the limited observations in the tropical anvil cirrus of 30-50 cm s^{-1} (Jensen et al., 2009). However, our modeled updraft velocity can be too high at high altitudes (e.g., near the tropopause) (Hoyle et al., 2005). We agree with the reviewer that *w* plays a critical role on the ice supersaturation, the occurrence of homogeneous nucleation, and thus the relative importance of homogeneous and heterogeneous ice nucleation in cirrus clouds. Following the reviewer's comment, we added some discussions in the revised manuscript: "The sub-grid variability of updraft velocity needed for driving the parameterization is derived from the square root of turbulent kinetic energy (TKE) [*Bretherton and Park*, 2009] with an assumed maximum threshold value of 0.2 m s^{-1} [*Gettelman et al.*, 2010]. We find that *w* tends to be near 0.2 m s^{-1} (the upper limit) much of the time in CAM5. Our analysis of updraft velocity measurements in the Interhemispheric Differences in Cirrus Properties From Anthropogenic Emissions (INCA) and Small Particles in Cirrus (SPartICus) campaigns indicates higher mean updraft velocities than the upper threshold limit of 0.2 m s^{-1} [*Zhang et al.*, Characteristics of vertical velocity in cirrus clouds and its impact on ice nucleation, in preparation, 2012]. This upper threshold limit is also lower than that seen in the limited observations in the tropical anvil cirrus of 0.30-0.5 m s^{-1} [*Jensen et al.*, 2009]. However, the model may predict too high subgrid variability of updraft velocity at high altitudes (e.g., near the tropopause) [*Hoyle et al.*, 2005]. We note that too high *w* may produce anomalously high ice supersaturations, which may induce a homogeneous ice nucleation bias. The effects of subgrid updraft velocity on ice nucleation in cirrus clouds will be investigated in a future study."

This study shows that the LP scheme produces results that agree with observations better than those predicted by the BN scheme. However, could this be an artifact of the treatment of w in CAM5 as described above? If there is a high RHi bias and this bias were removed, might the BN scheme show more sensitivity to IN? Reducing w would make it easier for IN to prevent RHi from reaching threshold values for homogeneous nucleation initiation. Would the PDA08 IN spectra then have a greater influence on Ni, producing greater differences between BNhom and BN results? If so, might this bring the BN results into greater agreement with observations?

We agree with the reviewer that the treatment of subgrid updraft velocity (*w*) in CAM5 plays a critical role on the modeled RHi, which will affect the occurrence of homogeneous nucleation and thus the relative importance of homogeneous versus heterogeneous nucleation. We find that the mean updraft velocities measured during the INCA and SPartICus campaigns are higher than *w* predicted in CAM5. However, modeled *w* can be higher than that seen in the tropopause (*Hoyle et al.*, 2005). We agree with the reviewer that if there is a high RHi bias in CAM5 resulted from the overestimation of *w*, and after the bias was removed, IN predicted from PDA08 will have a greater role on the overall Ni, and BN scheme will show more sensitivity to IN, producing larger differences between BNhom and BN results. Following the reviewer's comment, we added these points in the revised manuscript when we discuss the differences of ice crystal number concentrations between BNhom and BN in Figure 4: "We note that the relative importance of ice nucleation mechanisms depends on the in-cloud updraft velocity. If there was a high updraft velocity bias in CAM5, reducing the bias would make it easier for IN to prevent RHi from reaching the threshold values for the initiation of homogeneous nucleation, and thus increase the importance of IN from PDA08 on the overall Ni. This will produce greater differences between BNhom and BN results and improve the agreement of BN results with the INCA observations."

The results shown in Fig. 8 and Fig. 9 in Section 5 are very interesting. Regarding Fig. 8, it might strengthen the findings to cite other studies having similar results. Regarding Fig. 9, the details are interesting but cannot be clearly seen in this format. It is suggested to break this into two figures; Fig. 9a and 9b, with larger panels to clearly show the details. In addition, what many are interested in is the net cloud radiative forcing (SWCF + LWCF) or net CRF for each simulation as a function of latitude, as well as the net CRF differences between simulations (e.g. net CRFLP – net CRFLPhom, net CRFLPhet – net CRFLP). This, for example, would show the potential cooling effect that heterogeneous nucleation may have on our present and future climate, and issues like these are driving climate research. The net CRF plots could constitute Fig. 10.

Following the reviewer comment, we cited Liu et al. (2009) and Hendricks et al. (2011). Both studies found the similar results as shown in Figure 8 about the changes of cloud ice water content, temperature, cloud cover and specific humidity in the upper troposphere when the ice crystal number concentration in cirrus clouds is changed.

Regarding Figure 9, we would like to thank the reviewer for the good suggestions. We have broken Figure 9 into Figure 9a (for SWCF, LWCF, net CF, and CLDHGH) and Figure 9b (for IWP, ACTREI and CDNUMI) in the revised manuscript. We added net cloud forcing (CF) for each simulation in Figure 9a. We added new Figure 10 for the net CF differences between simulations (LP – LPhom, LPhet – LP, BN – BNhom, and BNhet – BN) to show the potential effects of heterogeneous ice nucleation on present and future climate.

The Lohmann et al. (2008, ERL) study also addresses "simple" competition effects between heterogeneous nucleation from dust aerosol and homogeneous nucleation using the ECHAM5 GCM. They find much stronger net CRF cooling effects than in this study (-2.0 W m-2 vs. -0.3 W m-2 found here). This should definitely be mentioned, and if possible reasons given for the differing results.

We thank the reviewer for pointing us to the Lohmann et al. (2008) study. Following the reviewer's comment, we mentioned the Lohmann et al. study in the revised manuscript. We added some discussion on the possible reasons why they found much stronger net CRF cooling effects (-2.0 W m^{-2}) than in our study (-0.3 W m^{-2}). In their study they assumed that the homogeneous nucleation will be completely switched off (i.e., only heterogeneous nucleation will occur) when the IN concentration is higher than 1 L^{-1}, and homogeneous nucleation occurs elsewhere (no transition between pure homogeneous and pure heterogeneous nucleation). In our study, when the IN concentration is about 20 L^{-1} (as calculated from Phillips et al. 2008 in BNhet shown in Figure 2), homogeneous nucleation still plays a dominant role in the combined simulation for the BN09 parameterization (Figure 4). Only when the IN concentration is higher than 100 L^{-1} in the northern hemisphere (as calculated in LPhet shown in Figure 2), the heterogeneous nucleation then becomes very important in the combined simulation for the LP05 parameterization (Figure 4). Thus the much lower IN threshold value (1 L^{-1}) assumed in Lohmann et al. (2008) to completely switch off the homogeneous nucleation can be the main reason for the much larger net CF cooling effects of IN in the Lohmann et al. (2008) study than this study. This is confirmed from the significant reduction of Ni on the global scale in their combined simulation as compared to their pure homogeneous nucleation simulation and the similar Ni in their combined simulation as that in their pure heterogeneous nucleation simulation shown in Figure 2 of Lohmann et al. (2008). The global annual mean vertically integrated Ni is reduced by 47% in their combined simulation as compared to their pure homogeneous nucleation simulation (Table 2 of Lohmann et al., 2008), while in our study the reduction is only 18% for the LP parameterization and the reduction is only seen in the northern hemisphere (Figure 2 and Table 2 of this study). This indicates a much greater (and a dominant) role of heterogeneous nucleation in the combined simulation of Lohmann et al. (2008) than that in this study. We have added the above discussion in the conclusion (section 6) part of the revised manuscript.

Specific comments:

1. Page 13134, line 28: Suggest changing 170% to 180% based on results in Fig. 6.

Done.

2. In Fig. 7, the LP histogram is more correlated with the observations than the BN histogram, LP is better matched with LPhet than LPhom, and BN is better matched with BNhom than BNhet. Does this imply that heterogeneous nucleation was the dominant nucleation mode during SPARTICUS?

The reviewer is correct that the LP histogram is more correlated with the observations than the BN histogram, and LP is better matched with LPhet than LPhom, and BN is better matched with BNhom than BNhet. From the model results, heterogeneous nucleation plays a very important role in the combined LP simulation, but not in the combined BN simulation during SPartICus. Although LP and LPhet modeled histograms agree best with the observations, we feel that a solid conclusion on the dominant role of heterogeneous nucleation mode during the SPartICus is still premature because (1) other ice microphysical processes such as aggregation and rimming of ice crystals which can reduce ice crystal number concentration may be underestimated in CAM5 with a coarse spatial resolution; (2) there may be biases with the subgrid updraft velocity (cooling rate) which is critical for the occurrence of homogeneous nucleation; and (3) the convective detrainment complicates the model to observation comparison of inferring the dominant in situ ice nucleation mechanism. Future analysis will evaluate the model representation of subgrid updraft velocity (cooling rate), and separate the in situ cirrus cases from the convective cirrus anvil cases in the comparison of model results with observations. We have added these discussions in the revised manuscript.

3. SWCF in Fig. 9: Why are all of the simulations excepting BNhet exceeding the observed SWCF in the tropics? Is this a cirrus cloud coverage issue or more likely a problem with the treatment of low and mid-level clouds?

It is more likely a cirrus cloud issue. As seen from comparison with observations in Figure 5, all of the model simulations except BNhet overestimate the ice crystal number concentration at low temperatures in the tropics. This may result in too strong SWCF as shown in Figure 9. We have emphasized this in the revised manuscript.

4. Effective radius in Fig. 9: Why is Re evaluated only at the tops of cirrus clouds rather than a vertically integrated average?

Since there is a strong vertical variation of Re within clouds and often satellite observations often show the Re at the tops of the cirrus clouds, we show here the Re at the cirrus cloud top, similar to Gettelman et al. (2010).

5. Page 13139, lines 26-28: This contradicts the points raised in comment 2 above, which may indicate that heterogeneous nucleation dominated during SPARTICUS.

As we replied to comment 2 above, although LP and LPhet modeled histograms agree the best with observations, we feel that a solid conclusion on the dominant role of heterogeneous nucleation mode during the SPartICus is still premature. We changed the wording from “dominant” to “important” in that sentence (Page 13139, line 25) to be “…and thus homogeneous nucleation may play an important role in the ice formation”, and we modified the tone of the following sentences to be: “However, the large fluctuation (by more than one order of magnitude) of observation data at a given temperature also suggest the role of heterogeneous nucleation affecting or completely inhibiting the homogeneous nucleation" in the revised manuscript.

6. Page 13140, lines 11-14: While the net global cooling is -0.3 W m-2 for the LP simulations, the latitudinal dependence on this net cooling is also worth mentioning. Please indicate what this net cooling is for the tropics and the extra-tropics, separately for each hemisphere in the extra-tropics. This information could also be in a table.

Following the reviewer's comment, we added the new Table 3 to show the net cloud forcing change in the tropics (30 °S to 30 °N), SH and NH mid-latitudes (30-60 °S and 30-60 °N) and SH and NH extra-tropics (60-90 °S and 60-90 °N) for the LP and BN simulations due to the dust IN effect. We mentioned these values in the text as well.

References

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