Responses to 1st Referee's Comments

The present manuscript discusses effects of mineral dust aerosol on cirrus through heterogeneous ice nucleation. Investigations based on global simulations with the CAM5 model are presented. Cirrus clouds are the most frequent cloud type in the upper troposphere. They have large effects on the Earth's radiation budget and are of large relevance for the climate system. The present knowledge on cirrus microphysical properties and the role of aerosols in cirrus formation is still very uncertain. Hence, the manuscript is of high relevance for atmospheric and climate science and is well suited as a contribution to ACP.

As an important result, the study reveals that homogeneous freezing activity is reduced due to competition with heterogeneous ice nucleation on mineral dust aerosol. The effect induces changes in cirrus microphysical properties and global mean cloud forcing. A clear advantage of the study is that uncertainties of these results are estimated by application of two different ice nucleation parameterizations and by comparison with measurements. The manuscript is clearly written and well structured. The applied methods are described thoroughly and, in most cases, the results obtained are discussed carefully.

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

Unfortunately, the statistical significance of the simulated effects of mineral dust shown in Figure 8 of the manuscript is not proven. In the case of Figure 9, it is not clear whether the differences between the model runs shown are larger than model noise. Since a fully coupled general circulation model is applied, the differences can also be due to feedbacks of the cirrus changes on model dynamics, rather than being directly related to cloud microphysical effects. To some extent, they could just be related to changed 'weather' and could average out if a longer, decadal simulation would be analysed. Even if the differences found by the authors are larger than the interannual variability of the discussed quantities they can still be affected by feedbacks. To cope with this problem, a robust statistical analysis of the aerosol-induced differences (e.g. by means of a student's t-test) is necessary. This needs to be urgently addressed in the paper.

Following the reviewer's comment, we have added the Student's t-test in Figure 8 for the differences between the model runs. Figure 9 is for the zonal mean distributions of climatological means of cloud properties, not the differences between model runs. So the statistical significance is not shown for this figure. We added a new figure (Figure 10) which shows the zonal mean distributions of net cloud forcing (CF = SWCF +

LWCF) differences between the model runs. The interannual variability of CF differences are indicated as the ranges of the two standard deviations (2-σ) estimated from the CF differences of each of 5 years at different model latitudes. We also calculated the standard deviations of CF in the combined simulations (e.g., LP) and found that it is slightly smaller than the standard deviation of CF differences between the simulations (LP – LPhom), but with a very similar pattern as a function of latitude.

We note that since we used the prescribed sea surface temperatures in our model simulations, 5-year runs plus 3-month spin-up are sufficient to average out the model noise based on our previous experience with CAM5 (i.e., Meskhidze et al., 2011; Gettelman et al., in press, 2012). Furthermore, as a test, we ran CAM5 for 10 years plus 3-month spin up for two simulations (LP and LPhom), we found the results are very similar to those from 5-year runs.

The manuscript aims to discuss climate impacts (title of section 5). However, with fixed sea surface temperatures and a simulation of only five realizations (5-year runs), a climate impact cannot be quantified. The simulations can just be used to discuss perturbations of cloud microphysics and related radiative flux changes, which could trigger possible climate change. This however has to be demonstrated by means of longer-term coupled atmosphere-ocean simulations. I would suggest changing the title of section 5 (e.g. 'Atmospheric effects') and other formulations in the manuscript accordingly.

We agree with the reviewer that the full climate impacts (i.e., including "slow" responses due to the sea surface temperature change) need to be investigated with the coupled atmospheric-ocean model simulations, which is beyond the scope of this paper. Following the reviewer's comment, we changed the title of section 5 to "Atmospheric effects". We also changed the wordings related to "climate impact" in the manuscript accordingly.

Specific Comments:

Page 13124, lines 5-6: I do not see that the mentioned parameterizations provide information about the ice crystal size distribution. In my understanding, the parameterizations provide initial ice crystal number concentrations which serve as *input for the microphysical cloud scheme (2-moment) of the large-scale model. The formulations should be skipped/corrected.*

Following the reviewer's comment, we removed "size distribution" in the revised manuscript.

Section 2.2: The description of the model representation of the Bergeron-Findeisen process is hardly understandable. It should be rendered more precisely or skipped, just referring to Gettleman et al. (2010).

Following the reviewer's comment, we changed the model description regarding the Bergeron-Findeisen process to be "(2) the Bergeron-Findeisen process is treated for mixed-phase clouds to deplete cloud liquid given the assumption that water vapor is saturated with respect to water in mixed-phase clouds". We have cited Gettelman et al. (2010) for details.

Page 13126, line 17: It should be mentioned here that aggregation provides a sink for ice particle number only and conserves the ice water content, while the other sinks mentioned affect both quantities.

Thanks for pointing out this. Following the reviewer's comment, we added the sentence "Aggregation reduces the ice particle number only and conserves the ice water content, while the other sinks mentioned above affect both quantities".

Section 3, paragraph 3: Mineral dust aerosol effects on cirrus are the major subject of this study. Hence, a more detailed description of the model representation of mineral dust cycles (sources, initial size distribution assumptions, sinks) would be appropriate here (or in section 2.1), rather than just referring to other litereature.

We thank the reviewer for the comment. We added a description of the model representation of mineral dust cycles in section 2.1. It reads as "Mineral dust is emitted in the accumulation mode with the size (diameter) range of 0.1-1 μm and coarse mode with the size range of 1-10 μm following the emission scheme of Zender et al. (2003). Once in the atmosphere, mineral dust is transported horizontally and vertically, and is removed by the dry and wet deposition. On the annual average, ~62% of global emitted dust mass is removed by the dry deposition (mainly by the gravitational settling), while wet deposition removes ~85% dust in the accumulation mode (Liu et al., 2012)".

Page 13134, lines 1-2: This should be re-worded carefully. Homogeneous freezing seems to dominate Ni, but this does not mean that heterogeneous nucleation is unimportant. It could still have important effects during many cloud events, especially at low cooling rates.

We thank the reviewer for the comment. We changed the sentence to "This indicates that homogeneous nucleation is often the dominant ice nucleation mechanism in this temperature range. Considering the large fluctuation of ice crystal number concentration at a given temperature, heterogeneous nucleation may still have important effects during many cloud events, especially at low cooling rates".

Section 4, comparison with MOZAIC data: It should be discussed whether the MOZAIC data could be biased towards cloud-free conditions since pilots might avoid passages through thick cirrus layers. It should also be discussed whether the all-sky model data could be biased towards low supersaturations owing to the humidity relaxation during the comparatively large time steps of large-scale models.

It is a good point. We added these discussions in the revised manuscript: "We note that MOZAIC data could be biased towards cloud-free conditions since pilots might avoid passages through thick cirrus layers", and "The all-sky model data can be biased towards low supersaturations owing to the water vapor deposition on ice crystals to remove any supersaturations inside clouds for CAM5 with a large model time step of 20 mins".

Page 13136, lines 2-4: 'These comparisons suggests that homogeneous nucleation may play an important role . . .'. I do not see this from the comparison. The het simulations generate high concentrations (larger than 100/L) more frequently than observed and fit even better to the observations than the hom cases. Only for concentration between 10-100/L the BN-het case shows too low frequencies. Here homogeneous freezing seems to be very important. As a potential reason for the discrepancies between model and observations also possible shortcomings in the representation of cooling rates in the model should be mentioned.

The reviewer is correct that the heterogeneous simulations generate high concentrations (larger than 100/L) more frequently than observed and agree better with observations than the homogeneous simulations. We feel that a solid conclusion on the dominant role of heterogeneous nucleation mode during the SPartICus is still premature due to the discrepancies between model simulations and observations. The shortcomings in the representation of model physics include (1) the possible underestimation of the growth processes of ice crystals by aggregation and/or rimming in CAM5 with a coarse spatial resolution; (2) the treatment of the subgrid updraft velocity (cooling rate) which is critical for the occurrence of homogeneous nucleation as the reviewer indicates; and (3) the treatment of convective detrainment which influences modeled ice crystal number. We have included the discussion in the revised manuscript: "Although LPhet and LP modeled histograms agree best with the observations, a solid conclusion on the dominant role of heterogeneous nucleation mode during the SPartICus is still premature due to the discrepancies between model simulations and observations. Future analysis will evaluate the model representation of subgrid updraft velocity (cooling rate) which is critical for the occurrence of homogeneous nucleation, and separate the in situ cirrus cases from the convective anvil cirrus cases in the comparison of model results with observations."

Section 5, title: The title should be changed (e.g. 'Atmospheric impacts') since climate change cannot be simulated by means of the simulation set-up chosen (see general comments above). All other formulations about 'simulated climate effects' should be rephrased accordingly.

Following the reviewer's comment, we changed the title of section 5 to "Atmospheric effects". We also changed other wordings about "climate impacts" in the revised manuscript.

Section 5, Figures 8/9: It needs to be shown whether the differences presented in Figure 8 and discussed in section 5 are statistically significant (e.g. by means of a student t-test performed on the base of the results obtained for the individual model years). In Figure 9, the interannual variability should be indicated in order to enable a fair evaluation of the significance of the differences between the model runs shown. See also general comments above.

Following the reviewer's comment, we performed the Student's t-test based on monthly mean model results from individual model years. Differences significant at the 90% level of the Student's t-test are depicted by dots in Figure 8 (right panel).

Figure 9 shows the multi-year averaged zonal mean distributions of cloud properties, not for the differences between the model runs. So the statistical significance is not shown for this figure. We show the interannual variability of the net cloud forcing (SWCF + LWCF) differences between two model runs in the newly-added Figure 10 as the reviewer indicated. This interannual variability is slightly larger than that of the net cloud forcing from the control run (combined nucleation simulation), but both have very similar patterns as a function of latitudes.

Page 13136, lines 25-27: Could the increase in stratospheric water vapour also be due to reduced sedimentation of clouds ice?

Yes. We added in the revised manuscript: "This together with reduced sedimentation of cloud ice allows more water vapor to be transported to the lower stratosphere".

Section 5, last paragraph, table 2: It should be discussed whether the differences between the global mean numbers gained from the different model runs are larger than their interannual variation. Otherwise the differences could just be due to 'model noise'. To proof significance appropriate statistical analysis is necessary here. Differences that are not significant should not be discussed.

Following the reviewer's comment, we calculated the standard deviations based on the model results for individual model years. When the differences between the global mean numbers from different model runs are less than the standard deviations (one sigma) we don't discuss them. We also added the standard deviations in the new Table 3, which gives the differences of net cloud forcing between different simulations.

Conclusions: It should be assured that all results discussed are statistically significant.

Following the reviewer's comment, in the conclusions we assured and emphasized the differences discussed are statistically significant (i.e., larger than the standard deviations).

Technical corrections:

Page 13123, line 25: include period/full stop behind approach.

Done.

Page 13125, line 24: replace 'seas' by 'sea'.

Done.

Page 13126, line 2: explain MG08.

Done. We added "hereafter as MG08" after we first cite "Morrison and Gettelman, 2008" in section 2.1.

Page 13128, line 26: include 'as' before 'in'.

Done.

Page 13129, line 5: include units behind 2.5x1.9.

Done.

Page 13131, line 22: replace 'reduces' by 'reduce'.

Done.

Page 13133, line 16: include 'the' before 'heterogeneous'.

Done.

Page 13134, line 5; and other parts of the manuscript: Replace 'probability distribution frequency' by 'probability distribution' or 'frequency distribution'.

Done. We changed to use "probability distribution" in the revised manuscript.

Page 13135, line 4: Explain DOE.

Done. It stands for "Department of Energy".

Page 13135, line 18: Explain SGP.

Done. We added the acronym "SGP" after we first introduce "Southern Great Plains" above.

References

Gettelman A., X. Liu, D. Barahona, U. Lohmann, and C. Chen, Climate Impacts of Ice Nucleation, *Journal of Geophysical Research*, in press, 2012.

Meskhidze N., J. Xu, B. Gantt, Y. Zhang, A. Nenes, S. J. Ghan, X. Liu, R. C. Easter, and R. Zaveri (2011), Global distribution and climate forcing of marine carbonaceous aerosol. 1. Model improvements and evaluation. *Atmospheric Chemistry and Physics*, *11*, 11689–11705.

Zender, C., H. Bian and D. Newman (2003), Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology, *J. Geophys.Res*, 108(D14): 4416, doi:4410.1029/2002JD002775.