

First of all, we would like appreciate the anonymous reviewer's comments and suggestions. In response to the reviewer comments, we have made relevant revisions in the manuscript. Listed below are our answers and the changes made to the manuscript according to the questions and suggestions given by the reviewer. Each comment of the reviewer is listed (black) and followed by our responses (blue).

Anonymous Referee #2

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Review of Lee and Penner: Aerosol effects on ice clouds: can the traditional concept of aerosol indirect effects be applied to aerosol-cloud interactions in cirrus? The influence of cirrus clouds on the radiative budget is an important field of research. Furthermore it is not clear yet how heterogeneous freezing modifies the microphysical and optical properties. The presented paper contributes to our understanding of the processes involved. However there are some points that have to be clarified before publication.

Major comments:

It is not clear to me which aerosol species are considered in your study and how/if they act as cloud IN for immersion/contact and deposition freezing. In section 2.3. you state that for contact and immersion freezing dust and BC is used. For deposition freezing only dust is considered? Why is BC not considered for deposition freezing?

BC aerosols can be neglected in the background troposphere because dust is observed there to be the most important species for ice nucleation (DeMott et al 2003, PNAS; Phillips et al 2008, JAS). To confirm this, we repeated simulations excluding BC from all of heterogeneous-freezing pathways. Then, we also repeated simulations including BC in all of heterogeneous-freezing pathways. From these repeated simulations, we find that the qualitative nature of results here did not vary with the inclusion or exclusion of BC.

On the other hand, in section 3 you describe the background aerosol concentration which you obtain from the CAM-UMICH. Here you say that BC/OM from fossil fuel, BC/OM from biomass, sea salt and dust are considered as well. Are these species also taken into account for the heterogeneous freezing modes? Please clarify which species are considered and how they can act as IN.

Sea salt and sulfate act only as CCN and they determine the haze particle (which is unactivated CCN) number and mass for homogeneous freezing of haze particles.

BC/OM aerosols are assumed to act as BC aerosols for contact freezing.

The following is added:

(LL303-307 in p11)

BC/OM aerosols are assumed to act as BC aerosols for contact, immersion, and condensation-freezing activation of IN. Dust and BC aerosols are assumed to act only as IN. Sulfate and sea salt aerosols are assumed to act only as CCN and thus nuclei for haze particles; some of these haze particles go through homogeneous freezing.

2) In section 4.5 you state that changes in homogeneously frozen ice crystals are the main reason for the different IWP in the PI and PD simulations. In figure 3 you show the background aerosol number concentration which lies in the range of approx. 10 to 30 cm⁻³. The simulated CINC shown in figure 6 is approx. two orders of magnitudes lower, in the order of 0.1 cm⁻³ (if the unit of l-1 is correct here). Normally the number of homogeneously frozen ice crystals is not limited by the available background aerosol concentration as also in the upper troposphere there are enough aerosols available for freezing. Thus, in your PI simulations there are already enough aerosols available and I don't see the reason for the strong difference between PI and PD. Could you maybe add some explanation?

As mentioned in section 4.5, the homogeneous freezing is not about the freezing of activated droplets but about the freezing of unactivated droplets or haze particles (i.e., particles located on the left side of Kohler curve before the peak). Also, as mentioned in section 4.5, haze particles form on CCN (but not on IN) and thus their numbers are controlled by CCN numbers; the number of haze particles is proportional to that of CCN and, hence, the number of haze particles homogeneously frozen is proportional to the number of CCN for a given environmental condition as described in Phillips et al. (2007, JAS). In figure 3, most of differences between the PI and PD aerosols are made by CCN and these differences lead to those in the number of haze particles and thus their homogeneous freezing as shown in Figure 7. These differences in homogenous freezing of haze particles lead to those in CINC and in feedbacks among CINC, supersaturation and dynamics, which in turn lead to those in IWP.

3) Section 4.6: You describe the influence of the changed IWP on the SW and LW radiation. Besides the IWP/LWP the effective radius of the ice crystals (or ice crystal number concentration) also influences the optical depth of clouds and therefore their radiative impact. Fusina et al, 2007 show that the transition of ice clouds from a cooling to a warming regime strongly depends on the ice crystal number concentration/size of the crystals. As in the PD simulations the IWP and CINC changes it would be interesting to see how the effective radius of the crystals and the corresponding optical depth of the clouds change. Could you maybe add a figure of these essential variables for the radiation or give some statements about their changes from PI to PD?

The following is added:

(LL530-536 in 19)

The averaged effective size (optical depth) over cloudy areas is 30 μm (0.43) and 33 μm (0.29) for the high- and low-aerosol runs, respectively. The percentage difference in the effective size between the high and low-aerosol runs is ~ 3 times smaller than that in IWP. Hence, the difference in IWP accounts for the difference in the optical depth more than that in the effective size and the radiation difference is associated more with the IWP variation than the effective-size variation between the high- and low-aerosol runs.

Minor comments:

1) Abstract, line 5: Please add CINC after IWP as the CINC strongly influences the radiation (see Fusina et al, 2007, Zhang 1999).

Done

2) Page 10432, line 19: How are the shape parameters and the characteristic diameter of the distribution chosen? Could you please add some information?

The following is added:

(LL131-134 in p5)

Simulations described in the following sections show that cloud mass in the simulated cirrus clouds is composed only of cloud ice, snow and aggregates. For cloud ice, snow, and aggregates, $v(D_n)$ is 0.18 (20 μm), 0.18 (400 μm), and 0.5 (400 μm), respectively.

3) Page 10436, line 17 ff. Are the different shapes of the crystals also applied to the radiation code? Could you add some information how many different crystal types are involved?

The following is added:

(LL158-167 in p6)

For the radiative properties of cloud ice, a generalized effective size is inferred from the mean ($\langle \rangle$) size of the equivalent spherical diameter (D_i) with a look-up table for $\frac{D_{ge,i}}{\langle D_i \rangle}$, where

$D_{ge,i}$ is a generalized effective size of cloud ice. This look-up table was generated by integrating numerically the formula for the generalized effective size of cloud ice given by Fu (1996) for a range of values of $\langle D_i \rangle$. Crystals are assumed to be columnar (McFarquhar et al., 1999) with a ratio of length to width, L/D , specified from aircraft observations (Ono, 1969; Auer and Veal, 1970) in the manner of Fu and Liou (1993), obeying a gamma size distribution. This axial ratio varies between 1 and almost 5 for $D = 20 - 160 \mu\text{m}$.

(LL655-669 in p23)

The dependence of crystal optical properties on the crystal habit is a function of the aspect ratio of ice particles (Fu, 2007); the difference in the crystal optical properties is proportional to that in the aspect ratio characterizing the crystal habit. This study assumed the columnar shape of ice crystals for the characterization of the optical properties and thus calculation of radiative fluxes, following Phillips et al. (2007). The columnar shape has an aspect ratio of $\sim 0.3-0.5$, which corresponds to the lower range of aspect ratio of ice particles. Fu (2007) showed the increase in reflected SW by cirrus clouds by $\sim 2\%$ when the crystal habit changes from the columnar shape to the plate or dendrites having aspect ratio around 0.7-1.0, corresponding to the upper range of the aspect ratio. Wendisch et al. (2007) showed that that change in the habit leads to $\sim 1.4 - 2.0\%$ increases in outgoing LW. These changes in SW and LW bring only less than 5% change to the offset of varying reflected SW by varying outgoing LW between the high- and low-aerosol runs shown in Table 2. This demonstrates that the qualitative nature of results of this study does not depend on crystal optical properties varying with the crystal habit.

4) Page 10438, line 15: For which aerosols do you apply a bi –or trimodal log-normal size distribution? Please clarify.

The following is added:

(LL301-303 in p11)

Here, a bi-modal log-normal size distribution is assumed for sulfate and sea salt aerosols. For fossil fuel BC/OM, biomass BC/OM, and dust, a tri-modal log-normal size distribution is assumed.

5) Page 10438, line 29: show the time series of the domain averaged total background (remove time before domain).

Done

6) Page 10439, line 5: You write that periodic boundary conditions are used on the horizontal boundaries. I thought that the model is nudged towards the ECMWF – analyses? Could you please explain what you mean here?

The large-scale forcing is imposed as the large-scale advective tendencies. The CSR domain is considered to be small compared to large-scale disturbances. Hence, the large-scale forcing is approximated to be uniform over the model domain and large-scale terms are defined to be functions of height and time only. For example, the large-scale advective tendency of water vapor mixing ratio is $(\partial \bar{q} / dt)_{LS} = -\bar{V} \cdot \nabla \bar{q} - \bar{w}(\partial \bar{q} / \partial z)$. Here, bars indicate observed large-scale values. This tendency term is included water vapor prediction equation with a relaxation time of 1 hr. The imposition of large-scale temperature tendency follows the same methodology as that of water vapor mixing ratio. The wind field nudging is also done following the similar methodology to that of water vapor mixing ratio.

The forcing is to maintain the horizontally-averaged fields in simulations close to the observed fields. But it is not to remove the differences in cloud-scale circulations between high- and low-aerosol runs. The large-scale forcing does not entirely control the amount of water deposited the model produces; it only determines the net total water, energy and momentum supplied to or removed from the domain. We do in fact see differences between the high and low aerosol cases. Although feedbacks from these differences onto the large-scale flow cannot be captured by this design, the controlled large-scale forcing isolates the effects of microphysics in an imposed large-scale flow. This enables one to see more clearly the particular effects of the aerosol changes on microphysics.

The details of the forcing can be found in Krueger et al. (1996: GEWEX Cloud Systems Study Working Group 4: First Cloud-Resolving Model Intercomparison Project CASE 2).

The temperature, humidity and wind fields affected by the large-scale forcings advect with winds and these fields can move out of the domain. The purpose of this study is to examine aerosol-cloud interactions for a given large-scale forcings or large-scale environment. This also means that we want to conserve energy, water, and momentum budget imposed by the large-scale forcings. In other words, we want the net domain energy, water and momentum variations to be controlled by the large-scale forcings only (but not by boundary conditions); this way, we are able to say energy, water and momentum are imposed on model domain only by large-scale environment or large-scale forcings. This is why we used a periodic boundary condition which enables energy moving out of one side of domain can move into the domain through the other side of the domain by allowing the temperature, humidity and wind fields moving out of one side of the domain to move into the domain through the other side of the domain. This enables the conservation of energy, water and momentum imposed by the large-scale forcings.

7) Page 10439, section 4.1: I'm not sure if it makes sense to show figure 4. It does not contain important information that could not be described in the text. You maybe could remove this figure and just mention the content in the text.

The following in the old manuscript (LL 11-14 in p10439) is removed with Figure 4:

Figure 4 depicts the temporal evolution of cloud-top and cloud-base height (upper two lines are for cloud-top and lower two lines for cloud-base) from 10 minutes after the cloud formation to the end of simulation. Figure 4 indicates that cloud depth is ~ 2 km.

8) Page 10440, equation 7: Should the sedimentation also be a sink of cloud ice or is the budget equation over the whole vertical domain such that even the cloud ice which has sedimented to the ground is still counted?

Integration of the sedimentation rate of cloud ice over whole domain and time period as represented by Eq. (6) results in 0, since no cloud ice reaches the surface. If a precipitable hydrometeor (e.g., aggregates) reaches the surface, for the budget equation of aggregates (but not cloud ice), the amount of aggregates reaching the surface (or the surface cumulative precipitation of aggregates) should be included as a separate term. However, no aggregates reach the surface in this study and, thus, for the budget equation of aggregates, the term of surface precipitation will not be included.

The following is added:

(LL380-381 in p14)

Since cloud ice does not reach the surface, the surface precipitation of cloud ice is not included in (7).

9) Page 10441, line 10: Could you explain what you mean with this sentence "the absolute value of any variable A is represented by $|A|$ "?

I think $|a|$ is a well-known symbol in mathematics to indicate the absolute value of some variables. Negative values of a variable turns into positive values and positive values of a variable remain positive with this symbol.

The following is added:

(LL402-404 in p14)

The absolute value turns negative values of a variable into positive values. However, the absolute value does not change the sign of positive values.

10) Page 10442, equation (9): Is the crystal shape factor given by $Sh=C/D$? If so, then $Fr_e = \int (C \cdot f_{Re} \cdot f_{gam}(D) dD)$.

Corrected.

11) Page 10442, line 13: Add space after "and eta"

Done

12) Page 10443, line 9-17: In this paragraph you explain the interaction between depositional heating and the intensification of updrafts. Probably, the strength of this interaction depends on the stratification of the atmosphere (the Brunt-Väisälä – frequency). In your case there is a relatively unstable layer between 13.5 and 14.5 km height. Could you please comment on how sensitive your results are to the stratification?

The following is added:

(LL670-680 in p23-24)

The large-scale stability of cloud layer may have an impact on results here. To examine the effect of the stability on results here, the high- and low-aerosol runs were repeated only by varying the stability. The first (second) set of these repeated high- and low-aerosol runs is with increased (decreased) potential-temperature gradient by a factor of 2 in cloud layer. With increased (decreased) stability, increases in deposition and IWP in this repeated high-aerosol run are smaller (larger) than the standard high-aerosol run. However, the more important role of feedbacks among CINC, deposition and dynamics in the IWP response to aerosols than that of conversion of cloud ice to aggregates and sedimentation of hydrometeors is simulated in these repeated simulations. This demonstrates that the qualitative nature of results here does not depend on the stability of cloud layer.

13) Page 10445, line 20: remove "that" in front of 2%.

Done

14) Page 10446, line 2: You could merge the first two sentences to: " Upward shortwave (SW) and longwave (LW) fluxes at the top ... in table 2: And remove "In table 2, SW and LW represents".

Done

15) Page 10446, line 9-11: Please rewrite this sentence as it is not clear.

The sentence pointed out here is revised as follows:

(LL537-541 in p19)

Longwave radiation emitted from the surface is absorbed by clouds more due to higher IWP in the high-aerosol run than in the low-aerosol run. Hence, longwave radiation going above cloud layer and thus reaching and going above the top of the atmosphere (i.e., outgoing longwave radiation) is smaller in the high-aerosol run than in the low-aerosol run (Table 2).

16) Page 10446, line 19-23: This findings seem interesting to me as it shows that it is not possible to calculate changes in the radiative properties of clouds without taking changes in the dynamics

and following from that the IWP into account. Maybe you could highlight a little bit more this result and the difference to the finding of Penner et al., (2009).

The following is added:

(LL640-654 in p22)

Coarse resolutions and a saturation adjustment used in a climate model in Penner et al. (2009) resulted in the different offset of the variation in shortwave radiation by longwave radiation than simulated here. While the change in longwave radiation is smaller than the change in shortwave radiation for simulations here, the change in longwave radiation dominates that of shortwave radiation for cirrus clouds perturbed by aerosol concentration in Penner et al. (2009). Here, the consideration of feedbacks among CINC, deposition, and updrafts has actually reversed the LW impact simulated by Penner et al. (2009). This indicates that the effect of aerosols on radiative properties of cirrus clouds and, hence, the assessment of aerosol indirect effect is highly sensitive to how sub-grid cloud-scale processes are represented in climate models. This also indicates that microphysics parameterizations (to represent sub-grid cloud processes), able to predict particle mass and number, and thereby, surface area, coupled with a prediction of supersaturation, need to be implemented into climate models for a better assessment of aerosol effects on cirrus clouds.

17) Page 10446, section 4.6: Due to the depositional growth latent heat is released which changes the temperature and supersaturation in the cloud layer. The crystal habit is dependent on these variables. Are changes in crystal habit due to these processes calculated and taken into account in the radiation calculation or are these changes way too small?

The following is added to describe the calculation of the effective size of cloud ice and assumption of a crystal habit for the radiation calculation:

(LL158-167 in p6)

For the radiative properties of cloud ice, a generalized effective size is inferred from the mean ($\langle \rangle$) size of the equivalent spherical diameter (D_i) with a look-up table for $\frac{D_{ge,i}}{\langle D_i \rangle}$, where

$D_{ge,i}$ is a generalized effective size of cloud ice. This look-up table was generated by integrating numerically the formula for the generalized effective size of cloud ice given by Fu (1996) for a range of values of $\langle D_i \rangle$. Crystals are assumed to be columnar (McFarquhar et al., 1999) with a ratio of length to width, L/D , specified from aircraft observations (Ono, 1969; Auer and Veal, 1970) in the manner of Fu and Liou (1993), obeying a gamma size distribution. Note that this assumption of columnar crystals is only applied for radiative properties of cloud ice. The axial ratio varies between 1 and almost 5 for $D = 20 - 160 \mu\text{m}$.

The following is added in the summary and discussion to discuss about the effects of crystal habit on radiation and its effects on the results here (LL655-669 in p23) in the new manuscript):

The dependence of crystal optical properties on the crystal habit is a function of the aspect ratio of ice particles (Fu, 2007); the difference in the crystal optical properties is proportional to that in the aspect ratio characterizing the crystal habit. This study assumed the columnar shape of ice crystals for the characterization of the optical properties and thus calculation of radiative fluxes,

following Phillips et al. (2007). The columnar shape has an aspect ratio of $\sim 0.3-0.5$, which corresponds to the lower range of aspect ratio of ice particles. Fu (2007) showed the increase in reflected SW by cirrus clouds by $\sim 2\%$ when the crystal habit changes from the columnar shape to the plate or dendrites having aspect ratio around $0.7-1.0$, corresponding to the upper range of the aspect ratio. Wendisch et al. (2007) showed that that change in the habit leads to $\sim 1.4 - 2.0\%$ increases in outgoing LW. These changes in SW and LW bring only less than 5% change to the offset of varying reflected SW by varying outgoing LW between the high- and low-aerosol runs shown in Table 2. This demonstrates that the qualitative nature of results of this study does not depend on crystal optical properties varying with the crystal habit.

18) Page 10446, line 25: Please rewrite the sentence to something like " Aerosol-cloud interactions in cirrus clouds developing in an environment with large-scale low vertical motion off the".

Done

19) Page 10447, lines 16-27: Could you maybe rewrite this paragraph as it is not that clear to me what exactly you want to say.

In the paragraph pointed out here, we wanted to explain the equivalence between cloud liquid (rain) in warm clouds and cloud ice (aggregates) in cold clouds (with no liquid-phase hydrometeors and thus no "rimed" solid hydrometeors formed by collection processes). As well known, the aerosol second indirect effect was first proposed by Albrecht (1989) based on the observation of warm clouds but not on that of clouds with solid-phase hydrometeors. Here, we wanted to extend this proposed concept of the aerosol second indirect effect from warm clouds to cold clouds only with solid hydrometeors (but not with rimed hydrometeors). For this extension, we focused on the important role of the conversion of cloud liquid to rain in the response of cloud mass to aerosols in warm clouds in the proposed indirect effect by Albrecht (1989). Cloud liquid (or droplets) is at the state of non-precipitable hydrometeors and collisions among droplets make it become large precipitable hydrometeors (i.e., rain in warm clouds). The conversion of non-precipitable hydrometeors (i.e., cloud liquid in warm clouds) to precipitable hydrometeors (i.e., rain in warm clouds) through collisions among droplets decreases with increasing aerosols due to decreasing droplet size and thus collision efficiencies among droplets; this conversion is generally referred to as "autoconversion". This decreasing conversion increases non-precipitable hydrometeors (i.e., cloud liquid in warm clouds), leading to an increase in cloud albedo, which is a core concept of Albrecht's aerosol second indirect effect; more suspended non-precipitable hydrometers increase cloud mass and thus albedo. We believe that this Albrecht's proposed concept can easily be extended to cold clouds with no liquid-phase hydrometeors, since there is also conversion of non-precipitable hydrometeors to precipitable hydrometeors in cold clouds as well. In cold clouds with no liquid-phase hydrometeors, ice crystals or cloud ice corresponds to non-precipitable hydrometeors and aggregates correspond to precipitable hydrometeors. Collisions among ice crystals make them large to be aggregates. In other words, the conversion of ice crystals to aggregates occurs through collision among ice crystals; this is none other than autoconversion. In the paragraph pointed out here, we intend to explain that both rain and aggregates are initiated through autoconversion and thus the effect of aerosols on the formation of rain through autoconversion in warm clouds is equivalent (or analogous) to that on the formation of aggregates through autoconversion in cold clouds with no liquid-phase hydrometeors. In the paragraph here, we also intend to say, with this equivalence, that the traditional understanding of aerosol indirect effect first proposed by Albrecht (1989) is not applicable to cold clouds in case aerosol effects on autoconversion in cold clouds are negligible, since, as mentioned above, changes in autoconversion due to aerosol changes are proposed to control the cloud mass

response to aerosols in the traditional understanding. We believe all of these points are adequately described in the paragraph pointed out here.

20) Page 10447, line 28: Rewrite the sentence to something like " The effect of changes in LW radiation on ice clouds caused by increasing ...".

Corrected.

21) Page 10448, line 1-4: You state that the increase in IWP could enhance the greenhouse effect of ice clouds. However, if the optical depth of the clouds is high enough, the albedo effect (cooling effect) dominates and an additional increase in IWP would then increase this cooling effect. Could you maybe add this dependence on optical depth somewhere?

The words "infrared warming effect" and an associated sentence are removed, since, with increased aerosols, anyway, the increase in reflected solar radiation is larger than the decrease in outgoing longwave radiation; though large portion of this reflection of solar radiation is offset by the decrease in outgoing LW.

The following is added:

(LL612-617 in p21)

However, it should be pointed out that as the optical depth of clouds increases, the increase in outgoing shortwave radiation can be enhanced and, thus, the offset of the increase in outgoing shortwave radiation by the decrease in outgoing longwave radiation can diminish. Hence, in clouds with higher optical depth than clouds simulated here, the offset by the decreased outgoing longwave radiation with the increased aerosols is likely to be smaller than simulated here.

22) Page 10449, line 23: replace Du2.5 is set at " with "DU2.5 is set to". References: These two articles also investigate the Competition of homogeneous and heterogeneous freezing and dynamical influences and you may want to cite them:

1. Replaced.

2. The following is added:

(LL351-355 in p13)

Kärcher and Ström (2003) also reported the dominance of homogeneous freezing over heterogeneous nucleation based on an observation of midlatitude cirrus clouds. This dominance of homogeneous freezing indicates that relative humidity where homogeneous nucleation starts to occur is much larger than that where heterogeneous nucleation starts to occur according to Spichtinger and Gierens (2009).

Spichtinger and Gierens, 2009b, Modelling of cirrus clouds -part 2: Competition of different nucleation mechanisms, *Atmos. Chem. Phys.*, 9(7), 2319-2334.

Kärcher and Ström, 2003, The roles of dynamical variability and aerosols in cirrus cloud formation, *Atmos. Chem. Phys.*, 3. 823-838, 2003.