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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Possible influence of anthropogenic aerosols on cirrus clouds and anthropogenic forcing" *by* J. E. Penner et al.

J. E. Penner et al.

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We are grateful for the evaluations of the reviewers, which have allowed us to improve and clarify the manuscript. Below we address each of their comments. The reviewer comments are in italics and our response is in regular type.

Review 1:

Unfortunately it turns out that the sign of the forcing depends on the model configuration. In fact, in section 4 I have the impression that every single sensitivity test proves the computed radiative forcing highly volatile.

As shown in Table 4a, for the 3-mode model, which we now explicitly state is the better of the two model configurations (based on Reviewer 2 comments and our assessment of the Aitken aerosol number concentrations), the net forcing only changes sign if it is very small (as is the case for sulfate forcing). The results from the mass-only model, do change sign, but we think it is important to show these differences because mass-only models are still used in many applications. Also, both the KL parameterization and the LP parameterization are part of the scientific literature, and it is important to show differences. Indeed, it is also important to show the uncertainty associated with using different model configurations. Nevertheless, we now only quote the figures in the abstract that are supported by the 3-mode (i.e. best) model results.

This alone makes it very probable that the authors had not payed due attention to questions of statistical and physical significance. The reported results may be noise, even if some of them seem plausible.

We have modified our discussion of the cases to emphasize that the 3-mode model is, in fact, in better agreement with observations and contains a better description of the relevant physics. In addition, we state that the KL parameterization is also emphasized since it includes an explicit calculation of the physics of competing freezing mechanisms in a parcel updraft. As we note below, one of the reasons for performing an off-line simulation is that questions of statistical significance do not occur. Variations in cloud cover, and liquid and ice water content due to variations in the &8221;weather&8221; of the model do not occur in the off-line treatment.

2 Major comments 1) In Figure 1 the authors have cirrus clouds at the ground, and in figure 2 they show crystal number concentrations at 140 hPa in the mid-latitudes and at the poles, i.e. far above the tropopause. What is that? Noise? How far apart must the curves in figs 2 and 3 be to be significantly different?

The clouds at the ground should not be termed &8220;cirrus&8221;, but they are cold (<238K) stratus clouds that form in these locations in the model, so we have used the ice number parameterizations to calculate number concentrations there. The clouds above the tropopause have very small cloud fraction and ice water contents (<0.1 mg/m^3 for the grid average ice water content). They are present at these altitudes in

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the CAM3 model, so we include them in our calculations, though their effect on forcing is very small. This explanation is now included in the discussion of Fig. 1. Questions of significance do not apply to these calculations, since they are not subject to variations in ice presence and amount or to changes in temperature or other meteorological variability. This fact is now included in our motivation for using off-line simulations in section 2.

2) The description of the method (parts of section 1 and section 3) is not clear enough. While reading I had never the impression I understand the strategy and the how they ran the models. For instance, it is unclear what 8220;fixed off8211;line meteorological fields8221; (p. 13907, l. 1) are and how they are used to model cirrus formation and evolution. Is it so that model output (every 6 hrs) is taken to represent fixed conditions for cirrus formation and subsequent evolution, that the cloud is abandoned after 6 hrs or earlier when the next model output is taken, and so on? When in the cloud evolution are the number concentrations recorded? Section 3 should be rewritten, such that the selected procedure gets clear.

We have altered the manuscript to put the description of how the meteorological fields (with their fixed cloud water and ice water contents) are calculated and used in the offline simulations in a single place in the manuscript (new section 2), which makes it clear what we have done. In addition the following text has been added:

The monthly average aerosol concentration fields from this model are used together with fixed, off-line meteorological fields to calculate ice crystal number concentrations (Ni) in cirrus clouds. The off-line meteorological fields include the cloud fraction and ice water content and were calculated from a simulation using the CAM3 NCAR general circulation model (Collins et al., 2004; 2006). These fields were saved every six hours, and so are generally consistent with the aerosol fields which were developed from this same meteorology. The off-line methodology is used to calculate Ni and the radiative impact of anthropogenic aerosols on cirrus clouds every hour. This methodology is similar to that employed by Chen and Penner (2005) in their study of aerosol effects on

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liquid phase clouds. The meteorological data for cloud fraction, ice water content, and liquid water content were interpolated to hourly data and were fixed in all simulations. This radiative forcing estimate is similar to the so-called Twomey effect of aerosols on warm, liquid phase clouds, since the feedbacks associated with the subsequent effects of aerosols on the sedimentation of ice crystals are not allowed to change the occurrence of ice nor is the ability of ice nuclei to form additional cirrus at lower RHi accounted for (Liu et al., 2007; 2008; Haag and Kärcher, 2004). One advantage of avoiding these feedbacks is that the radiative forcing calculated here corresponds to that defined by the Intergovernmental Panel on Climate Change (Forster et al., 2007). While treatment of such feedbacks is desirable in global climate models, this treatment requires the development of a model with a sub-grid scale parameterization of cloud fraction and supersaturation that can accurately calculate the changes to cloud occurrence associated with changes to aerosols and different ice nucleation modes. Many current global climate models do not yet have this capability. A second advantage of our approach is that small changes to ice number concentrations and to radiative forcing are not subject to variations associated with weather variations as would occur if the changes to ice number concentrations were allowed to change the cloud fields through changes in ice sedimentation rates and other processes. Thus, the difference in forcing associated with different aerosol emissions are all statistically significant, even if they are only carried out for only a single year.

3) Why is it required to use different parameterizations (KL, LP, KLm) when the only effect of this seems to be the variation in the threshold for heterogeneous nucleation? If this variation is all that is intended here, why cannot that be achieved with LP alone, isn8217;t this just a free parameter? This would also render the presentation in section 3 clearer. Different parameterizations certainly involve further subtle differences that may cause effects not considered in the paper. I do not understand this strategy.

One could use a single parameterization if the only difference that we are investigating was the heterogeneous ice nucleation threshold. But, in fact, the reason for includ-

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ing the LP parameterization is that it is computationally far more efficient than the KL parameterization. But, one needs to be assured that the LP parameterization is not too different from a parcel model calculation. To make this clear, we have added the following text to the manuscript:

To calculate the impacts of different aerosol sources on ice crystal number concentrations, we ran the coupled IMPACT/CAM model for 5 years and used the monthly average concentrations averaged over the last 3 years for calculations of Ni and the radiative forcing due to anthropogenic aerosols. We used two recently developed physically-based ice nucleation parameterizations (Liu and Penner, 2005; Kärcher et al., 2006) that account for the competition between homogeneous and heterogeneous nucleation to determine the ice nuclei concentrations associated with anthropogenic aerosols (see Sect. 4). The former parameterization is computationally efficient, and may therefore be useful in coupled climate/aerosol simulations. The latter parameterization calculates the supersaturation as part of an adiabatic parcel model parameterization for the probability distribution of sub-grid scale updrafts and aerosol concentrations at each grid point and is computationally demanding. Here, we emphasize the use of the Kaercher et al. (2006) since it includes an explicit representation of the relevant physics that determines the supersaturation when different aerosol types freeze in an updraft, but also show results for the Liu and Penner (2005) parameterization since it may be useful in coupled model calculations (Liu et al., 2007; 2008, see Sect. 5).

3 Minor comment 1) In the introduction (p. 13906, l. 20 ff) it should not only be stated that Hendricks did this and that, but also their main conclusions should be reported and then the expected progress of the present paper over the state of Hendricks (2005) should be stated.

We have modified the introduction. Our calculation method is significantly different than that of Hendricks et al., so the discussion of our results in comparison with theirs is now entirely in the Section 6: Discussion and conclusions.

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Review 2:

Major comments: 1) I agree with the referee 1 that the sensitivity studies seem a bit arbitrary. I would also question their rational for using the KL scheme, which, in the paper, is motivated by its 140% RHi threshold. However, in the Kaercher et al. (2006) paper also the 130% RHi threshold is used for heterogeneous nucleation. Thus, comparing these 2 schemes might make sense because the assumptions behind the competition between homogeneous and heterogeneous freezing are different, but that needs to be stated. I would second referee 1 that you could vary the RHi threshold within one parameterization if that is what you use the two different parameterizations for. Otherwise if you compare them because of different assumptions, more detail on these schemes needs to be added.

We did not intend to motivate the KL parameterization by the difference in thresholds between it and the LP scheme. In fact, we did not use a threshold of 140

2) I have a problem with your comparison of the 3-mode scheme with a mass-only scheme, which seems to yield larger differences in forcing. I am in particular worried that these estimates are treated equally in terms of their likelihood. I would argue that the 3-mode scheme should be superior and therefore its results should be more reliable.

We agree that the 3-mode scheme is the superior scheme, and have changed the manuscript to emphasize this scheme more in our presentation. Thus, the forcing quoted in the abstract is now only from the 3-mode scheme. In Section 2, we explicitly state:

These aerosol simulations were carried out using both the 3-mode version of the aerosol model in which both aerosol mass and number concentrations of sulfate aerosols are calculated as well as with a version of the model that only predicts aerosol mass. As we show below, the predictions of the 3-mode model, particularly for the Aitken number concentration, are more realistic than those of the mass-only model.

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Moreover, the physical processes determining the sulfate aerosol number concentrations in the upper troposphere (through nucleation, condensation, and coagulation) are represented in the 3-mode model, whereas the number concentration is assumed in the mass only model (by assuming a fixed size distribution for the sulfate aerosols). Nevertheless, understanding the differences between the predictions of the mass-only model and those of the 3-mode model are of interest for comparison with coupled climate model simulations that have relied on the mass-only model for computational efficiency.

Also, at the very end of the article the authors state the cirrus scheme has been used online in another publication. This yields larger positive forcings than discussed here. This to me questions the whole paper. Online simulations should be more reliable because the ice crystals can then sediment, which has been omitted in the offline simulations. Thus, what is the point in presenting simulations that likely overestimate the impact of anthropogenic aerosols on cirrus clouds? This point should be made much clearer.

Larger positive cloud radiative forcing in a coupled simulation is expected if the addition of heterogeneous nuclei increases the cloud amount (cloud fraction and ice water path). Moreover, an increase in cloud amount is anticipated when IN are increased since these IN cause freezing at lower relative humidities than homogeneous ice nuclei. Thus, we anticipate that this should occur when the parameterization of ice number is included in a coupled to a global model. Nevertheless, these feedbacks are not included in the IPCC definition of radiative forcing, so that separate calculations without these feedbacks need to be available in the literature. We have revised the paper to better motivate the off-line simulations (as noted in our response to Referee 1) and to discuss the fact that feedbacks can explain much of the differences between the off-line and on-line calculations. Also, I meant to call into question the coupled model estimates of cloud forcing rather than the off-line simulations of radiative forcing, since the estimates of the probability of high supersaturations in the coupled model do not agree with observations. However, at the present time, the reasons for differences in the two simulations is not entirely clear and part of on-going calculations. We have revised our discussion of these points as follows:

Their calculated ice crystal number concentrations are significantly smaller than those found here and their calculated forcing between PD and PI simulations is positive (of order 0.5 to 0.7 Wm^{-2}) rather than only slightly positive ($\sim$0.2 Wm^{-2} with the LP parameterization and mass only model) as found here. Smaller ice crystal number concentrations are expected since coagulation and sedimentation of ice crystals are included as feedbacks in the coupled climate simulation. Moreover, an increase in cloud amount is anticipated when IN are increased since these IN cause freezing at lower relative humidities than homogeneous ice nuclei (e.g. Haag and Kaercher, 2004), so the larger cloud forcing in the coupled model simulation might also be anticipated. Nevertheless, the maximum values of RHi produced by the current version of the coupled model do not simulate the highest values recorded in the MOZAIC observations (Liu et al., 2007). This low bias may cause heterogeneous nucleation to inappropriately dominate much more frequently than it does in the off-line simulations reported here. Understanding the cause of the differences in these simulations is part of our on-going research.

1) What is the rational for varying the updraft speed but not the RHi threshold?

The maximum RHi experienced in a parcel model is determined by the cooling rate, which is determined by the updraft speed. Subgrid scale variations in updraft speed (or some method to determine the 8220;average8221; updraft for nucleation8212;as used in some warm cloud nucleation parameterizations, i.e. that in Lohmann et al., 1999) must be included to properly simulate nucleation. The motivation for using the KL and LP parameterizations as well as a better description of these methods and their difference is now better explained (see above).

2) Why are the authors comparing their 3-mode scheme with a mass-only scheme?

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As noted above, the purpose is to see if the results are significantly different, since the mass only scheme is computationally efficient, but is less accurate for prediction of Aitken nuclei and does not include the physics needed to predict number concentration of sulfate aerosols.

If the purpose is to reduce the high accumulation mode number concentrations, then why not vary the collision kernel?

This motivating sentence has been removed. However, we note that collision kernels are determined by the physics of the size of the particles undergoing coagulation. In our opinion, they should not be tuned to simply change results. We now emphasize that the number concentrations of ice crystals are mainly determined by the Aitken number concentration and this motivates the fact that the 3-mode model is the better of the two models.

At the end of the paper the authors compare their results with the results by Liu et al., which uses the mass-only scheme. If that is their motivation, it should be made clearer.

The motivation is to re-emphasize the differences between a coupled model simulation and that reported here and to partially explain the differences in these two types of simulations.

3) Why are you doing offline simulations? Is that to rule out any impact on water clouds?

This rules out changes due to all cloud feedbacks so that the forcing estimates correspond to those defined by the IPCC.

4) P. 13911, line 8: do you really mean $T > Tcrithet$ or is that a typo? If not, I do not understand this as you probably do not allow T to be larger than 238K for cirrus formation

This is changed to state that $T > 238$.

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5) How do you allow for homogeneous and heterogeneous freezing in between the temperature thresholds? Do you calculate both freezing types and then take a linear combination?

This is exactly what we do for the LP parameterization, as stated in the original manuscript.

If so, that is not what the KL parameterization suggests. It suggests to only calculate homogeneous nucleation if heterogeneous nucleation with subsequent growth to ice crystal size is not sufficient to deplete the available supersaturation.

This is correct. As we explain now, in the KL parameterization,: The parameterization accounts for the continuous competition between homogeneous and heterogeneous nucleation at any temperature and allows homogeneous nucleation to occur in the rising parcel even if some heterogeneous nucleation has already taken place as long as the supersaturation is able to continue to grow.

6) P. 13914, lines 26-28: This is something that was also found in Lohmann and Kaercher (2002), and which can be understood because there you have the high updraft velocities so that the ice crystal number concentration in pre-industrial times might be limited by the number of sulfate aerosols.

Reference to the finding of Lohmann and Kaercher (2002) has been added. However, the updraft velocities do not explain the finding that the ice crystal number concentrations are limited by the number of sulfate aerosols, since the updraft velocities in the present calculation are the same at all locations. The limitation at high altitude tropical regions is caused by the very low temperatures there.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13903, 2008.