Sensitivity studies of cirrus clouds formed by heterogeneous freezing in the ECHAM GCM

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[1] Cirrus clouds can form by homogeneous and heterogeneous ice nucleation mechanisms at temperatures below 235 K. Here we evaluate the effectiveness of heterogeneous freezing versus homogeneous freezing using a newly developed parameterization of heterogeneous freezing that is restricted to immersion freezing as the most likely pathway for heterogeneous ice formation in cirrus conditions [Kärcher and Lohmann, 2003]. In addition to a reference simulation considering homogeneous nucleation with temperature-dependent freezing thresholds, we discuss two idealized model experiments. We conduct a scenario that hypothetically assumes that the aerosol particles available for homogeneous freezing could act as freezing nuclei commencing freezing at 130% with respect to ice and contrast that by a scenario that only considers black carbon and mineral dust as immersion nuclei with the same freezing relative humidity of 130%. These idealized simulations serve to delimit possible climate responses. If the number of freezing nuclei is limited by the number of black carbon and dust aerosols, then heterogeneous freezing results in fewer ice crystals than formed by homogeneous freezing. These fewer ice crystals grow more readily to precipitation size and with that increase the global mean precipitation, decrease the ice water path, and trap less outgoing longwave radiation at the top of the atmosphere.

INDEX TERMS: 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 3354 Meteorology and Atmospheric Dynamics: Precipitation (1854); 3367 Meteorology and Atmospheric Dynamics: Theoretical modeling; KEYWORDS: cirrus clouds, heterogeneous freezing, climate modeling


1. Introduction

[2] Cirrus clouds can form by homogeneous and heterogeneous ice nucleation mechanisms at temperatures below 235 K. While homogeneous freezing of supercooled aqueous phase aerosol particles is rather well understood, understanding heterogeneous ice nucleation is in its infancy. Koop et al. [2000] showed that nucleation rates of supercooled aqueous aerosol particles can be parameterized as a function of temperature and the water activity of the aerosol particles; the latter can reasonably well be approximated by the ambient relative humidity for the purpose of global model applications.

[3] Haag et al. [2003b] showed an excellent agreement of adiabatic parcel model simulations and measurements of the maximum relative humidity inside cirrus and outside close to cirrus clouds taken in and near young cirrus in the region over Punta Arenas, Chile, in the Southern Hemisphere. These data were taken as part of the Interhemispheric Differences in Cirrus Properties From Anthropogenic Emissions (INCA) field experiment [Ovarlez et al., 2002]. The simulations indicate that the observed cirrus clouds formed predominantly by homogeneous freezing. Thus the maximum relative humidities outside clouds can be interpreted as the supersaturations required for homogeneous nucleation. These freezing threshold relative humidities are considerably lower in the region over Prestwick, Scotland, in the Northern Hemisphere, whereas the maximum supersaturations observed within clouds are comparable to those detected in the Southern Hemisphere. Consequently, assuming pure homogeneous freezing may overestimate the freezing threshold and considering pure heterogeneous ice nucleation with an ice nuclei number sufficient to prevent homogeneous freezing from occurring may underestimate the maximum supersaturations inside clouds. If both homogeneous and heterogeneous ice nucleation take place, then the relative humidity decreases outside clouds. At the same time, if the number of ice nuclei that form heterogeneously is not sufficient to deplete the supersaturation fast enough, it...
builds up the homogeneous freezing threshold inside clouds.

[4] In the climate modeling framework, Lohmann and Kärcher [2002] started to account for cirrus formation by homogeneous freezing using a parameterization derived from the change of supersaturation as a function of time and validated by adiabatic parcel model simulations [Kärcher and Lohmann, 2002a] employing the water activity concept by Koop et al. [2000]. In the light of the findings described above, it is important to evaluate the global impact of heterogeneous ice nucleation mechanisms on cirrus cloud formation and lifetime. At the moment, we have no parameterization that can describe the observed competition between heterogeneous and homogeneous ice formation so that the simulations described in this study will be limited to either pure homogeneous or heterogeneous ice formation. It is, however, necessary to start with comparing pure homogeneous freezing with pure heterogeneous ice formation in order to understand their differences in terms of cirrus cloud properties and their impact on cloud radiative properties. Only in this framework we will later on be able to understand the competition between homogeneous and heterogeneous ice nucleation in a global modeling framework.

[5] The homogeneous freezing parameterization developed previously [Kärcher and Lohmann, 2002a, 2002b] uses temperature, aerosol size and number density, and updraft speeds empirically corrected for subgrid-scale variability to determine the number concentration of newly formed ice crystals, \( n_i \), in a given model time step \( \Delta t \). The evaluation of \( n_i \) is based on a nucleation rate expression that links the rate with temperature and water activity in the freezing aerosol [Koop et al., 2000]. The latter is approximated by the ambient relative humidity to obtain an approximate but accurate analytical solution. Within \( \Delta t \), the parameterization assumes ongoing adiabatic cooling with a constant updraft speed until the freezing conditions are met, otherwise it returns without effect. The climate model then uses \( n_i \) and integrates prognostic equations for the cloud ice mass mixing ratio and ice crystal number concentrations, as described in section 2.1.

[6] In this study we use the parameterization of heterogeneous ice formation as described in Kärcher and Lohmann [2003]. It works similar to its homogeneous counterpart, but describes a different nucleation mode using a modified expression of the activity-based homogeneous nucleation rate. The freezing rate is given as the product of the nucleation rate and the surface area of the ice nuclei that trigger freezing. We restrict ourselves to immersion freezing as the most likely heterogeneous ice nucleation mechanism for cirrus formation at temperatures below \( \sim 238 \) K [DeMott et al., 1997]. The exact nature (immersion mode or deposition mode) of the heterogeneous ice nucleation process is not important for our sensitivity studies. Instead, the key factors controlling the impact of heterogeneous freezing in the model are the freezing threshold relative humidity and the total concentrations of available ice nuclei, both of which will be varied.

[7] As for homogeneous freezing, immersion freezing is formulated in terms of the water activity. However, the water activity actually used to compute the heterogeneous ice nucleation rate is shifted toward smaller values such that heterogeneous freezing occurs at a prescribed relative humidity lower than the homogeneous freezing threshold at a given temperature. The shift may depend on temperature, as suggested by the experimental results of DeMott et al. [1999] and Zuberi et al. [2002], but could also depend on other variables required to characterize the heterogeneous freezing process [Kärcher and Lohmann, 2003]. This approach has the advantage that (1) it reduces to homogeneous freezing for pure particles, (2) is simple to comprehend and straight forward to apply, and (3) can be fitted to any experimental data. In most practical cases, the volume fraction of the immersed solid particle is not an important factor in determining the freezing threshold for mixed particles. The climate model and the experimental setup are described below followed by the results of the sensitivity tests with different freezing scenarios and a summary and outlook.

2. Climate Model Description and Experimental Design

2.1. Treatment of Aerosols and Clouds in ECHAM4

[8] The ECHAM4 general circulation model (GCM) [Roeckner et al., 1996] used in this study is described in Lohmann et al. [2003]. Prognostic aerosol variables are the mass mixing ratios of sulfate, methane sulfonic acid, hydrophilic and hydrophobic organic carbon, hydrosoluble and hydroporphobic black carbon, submicron and supermicron dust \((0-1 \, \mu \text{m} \text{ and } 1-2 \, \mu \text{m})\), and submicron and supermicron sea salt \((0-1 \, \mu \text{m} \text{ and } 1-10 \, \mu \text{m})\). Transport, dry and wet deposition, and chemical transformations of the aerosols and gaseous precursors are calculated on-line in the GCM [Feichter et al., 1996; Lohmann et al., 1999a].

[9] We convert the mass of each aerosol component into a particle number concentration assuming that the size distribution of the total aerosol can be approximated as a four-mode lognormal distribution. Mode radii and standard deviations of the respective modes are locally prescribed. For the two smallest size categories, variations in size distribution with altitude are considered as observed by DeReus et al. [2001] and Schröder et al. [2002]. Carbonaceous aerosols fall into the smallest size class in accordance with the aerosol climatology described by Hess et al. [1998]. They are assumed to have a mode radius increasing from \(0.03 \, \mu \text{m} \text{ below } 4 \, \text{km} \) [DeReus et al., 2001] to \(0.05 \, \mu \text{m} \text{ above } 10 \, \text{km} \) [Schröder et al., 2002] and a standard deviation between 1.55 and 1.8. Black carbon particles which can have a chain-like fractal geometry are approximated by volume equivalent spheres showing particle number-to-mass ratios typical for atmospheric black carbon. Ammonium sulfate, methane sulfonic acid and submicron seasalt fall into the second smallest category with a vertically uniform standard deviation of 1.5 and a mode radius decreasing from \(0.08 \, \mu \text{m} \text{ below } 4 \, \text{km} \) [DeReus et al., 2001] to \(0.05 \, \mu \text{m} \text{ above } 10 \, \text{km} \) [Schröder et al., 2002]. The third size class is a small coarse mode with a mode radius of 0.45 \(\mu \text{m} \text{ and a standard deviation of 1.5 everywhere. It is assumed to be composed of sea salt and smaller size dust in ECHAM. The fourth size class is comprised of the larger size dust with a mode radius of 1.9 \(\mu \text{m} \text{ and a standard deviation of 2.15 [Hess et al., 1998]. The minimum number of aerosol particles assumed to always be present is \(2 \, \text{of } 9\).}]}\)
10 particles cm\(^{-3}\) in accordance with observations taken at midlatitudes and in the tropics [Minikin et al., 2003].

[10] The prognostic cloud variables are the mass mixing ratios of cloud liquid water and cloud ice for large-scale clouds and their number concentrations, as described in Lohmann and Kärcher [2002] and Lohmann et al. [2003]. Fractional cloud cover is diagnosed from relative humidity following Sundqvist et al. [1989]. The improved calculation of the effective longwave cloud fraction and the maximum-random overlap of clouds by Räisänen [1997] is used in this study.

[11] Cloud ice can originate from depositional growth during large-scale ascent or from detrainment of deep convective clouds. The homogeneous and heterogeneous ice nucleation rates of cirrus clouds formed by large-scale ascent require that the grid box is supersaturated with respect to ice. Hence subgrid-scale fluctuations in cloud cover cannot be resolved here. This simplified assumption will not be necessary any longer once we incorporate a more sophisticated method to allow for subgrid-scale fluctuation of temperature and humidity, which is not yet available in the case of cirrus clouds. In the case of detrainment, the detrained ice water is added to the ice water content of the large-scale ice clouds. The large-scale ice crystal number concentration is assumed to be unaffected. In case that detrainment occurs within a grid box that is not supersaturated, the detrained ice is sublimated and the ice crystal number concentration is set to a minimum value of 10\(^{-2}\) cm\(^{-3}\).

[12] In our reference simulation (simulation HOM; section 2.2), the number of newly frozen ice crystals \(n_i\), at temperatures below \(-35\) °C is obtained from the parameterization of homogeneous freezing by Kärcher and Lohmann [2002b] that takes into account the effects of the aerosol size distribution on \(n_i\). \(n_i\) is a function of the vertical velocity, temperature, and supersaturation with respect to ice. The parameterization scheme considers a multimodal aerosol size structure, with an arbitrary number of modes each of which is characterized by a lognormal distribution. Consequently, the parameterization is called by ECHAM with the number mode radius of the freezing particles prior to freezing, along with a total number density and a geometric width for each mode.

[13] In order to reduce the numerical expense of the parameterization, we consider a bimodal aerosol size distribution instead of the four-modal size distribution discussed above. Therefore the two smallest size classes of the four-modal distribution were merged since they show very similar particle sizes in the upper troposphere. The mode radius and standard deviation of the merged mode is averaged from the corresponding size distribution parameters of the two underlying modes weighted by their particle number concentrations. The second mode used to drive the parameterization corresponds to the third size class of the four-modal distribution. As mentioned above, this size mode consists of super-micron sea salt and smaller size dust. The largest size dust hardly reaches the upper troposphere and is therefore neglected for homogeneous freezing. Note that also very fine sulfate particles present in the nucleation and Aitken mode size ranges are generally neglected here since such fine particles are of secondary importance for cloud formation.

[14] In ECHAM, we obtain the mesoscale updraft velocity \(w\) needed for ice crystal formation as the sum of the grid mean vertical velocity and a turbulent contribution expressed in terms of the turbulent kinetic energy (TKE) [Lohmann and Kärcher, 2002]:

\[
w = \bar{w} + c \sqrt{TKE},
\]

where \(c = 0.7\). The choice of the specific dependence on TKE is discussed in Lohmann et al. [1999b]. The square root of TKE is used a measure of the turbulent vertical velocity. The coefficient \(c\) should be less than unity, since we are only interested in the upward component of the vertical velocity. We choose 0.7 as the average amplitude of a positive sine curve. The model is, however, not that sensitive to the choice of \(c\) as discussed by Lohmann and Kärcher [2002]. This choice of the turbulent contribution in conjunction with the underlying advective transport scheme leads to a reasonable representation of the relative humidity in ice-supersaturated regions and of grid-box averages of the total ice crystal number density in cirrus, as shown by Lohmann and Kärcher [2002] and Kärcher and Ström [2003].

2.2. Design of the Model Simulations

[15] All simulations were conducted in T30 horizontal resolution with 19 vertical levels and a 30 minute time-step over a period of 5 years after an initial spin-up of 3 months using climatological sea surface temperatures and sea ice extent. In the reference simulation, HOM, cirrus formation is based on homogeneous freezing including aerosol size effects [Kärcher and Lohmann, 2002b]. To study heterogeneous freezing, we conduct two sensitivity studies. In the first scenario, we assume that all the aerosols available for homogeneous freezing freeze effectively at 130% relative humidity with respect to ice (simulation HOM130%). That is, we only replaced the temperature-dependent freezing threshold for homogeneous freezing by a constant value of 130% relative humidity. With that, this experiment represents an upper limit for the number of freezing nuclei. In the other simulation we assume that only black carbon and mineral dust trigger heterogeneous freezing as shown by experimental results of DeMott et al. [1999] and Zuberi et al. [2002] using the same freezing relative humidity of 130% (simulation HET130%). The freezing relative humidity of 130% is taken for simplicity. In principle, we could have employed the explicit temperature dependence found in DeMott et al. [1999] and Zuberi et al. [2002], but it is unclear how to extrapolate the data for black carbon to lower temperatures. However, reducing the threshold freezing relative humidity to 105% or enhancing it to 150% changes the vertically integrated ice crystal number by only 1% because the limiting factor is the scarcity of the number of freezing nuclei. Because the model cannot resolve subgrid-scale fluctuations in cloud cover, the effect on the global mean cloud coverage is small. Thus even taking the observed freezing thresholds would hardly change the results. The ice crystal number is more sensitive to changes in the freezing threshold in the case of homogeneous freezing. These scenarios are covered in the following discussions because a simulation HOM150% is similar to the reference scenario HOM and the difference between
HOM and a simulation comparable to HOM100% was discussed by Lohmann and Kärcher [2002]. Therefore we subsequently focus only on simulations HOM, HOM130%, and HET130%.

3. Results

[16] We validated the ECHAM4 model including the parameterization of homogeneous freezing with aerosol size effects (simulation HOM) in Lohmann et al. [2003] showing that ECHAM captures the observed frequency of occurrence of ice water content, ice water path, effective ice crystal radius and ice crystal number concentration from radar data at the Atmospheric Radiation Measurements (ARM) site in Oklahoma. We also showed that the ice crystal number concentration as a function of vertical velocity and relative humidity with respect to ice for Southern and Northern Hemisphere data from the INCA campaign [Ström et al., 2002] is reasonably well reproduced. However, because the GCM cannot capture the fine-scale structure of the vertical velocity that is present in the observations, it misses the details of the observed ice crystal distribution and especially underestimates high ice crystal concentrations [Kärcher and Ström, 2003]. Haag et al. [2003b] showed that ECHAM4 captures the observed frequency distribution of supersaturation with respect to ice as obtained from the MOZAIC (Measurement of Ozone on Airbus in-Service Aircraft) measurements [Gierens et al., 1999]. Therefore we feel confident to now investigate the impact of the heterogeneous freezing on cirrus clouds.

3.1. Activated Aerosol Fraction and Number of Crystals Formed by Homogeneous and Heterogeneous Freezing

[17] The number concentrations of hygroscopic aerosol particles (AP) that can freeze homogeneously in simulation HOM and the ice nuclei concentration (IN) from simulation HET130% are shown in Figure 1. It shows that the ice nuclei concentration amounts to between 0.1 and 5% of the hygroscopic aerosol number concentration in the zonal mean. The fraction is highest in the lower troposphere because the dust and black carbon number concentration decrease more rapidly with height than some of the aqueous species like sulfate and MSA that are formed by gas-to-particle conversion within the atmosphere. This figure also shows that the ice crystal concentration (IC) is limited by the number of ice nuclei at colder temperatures. Recall that we assume all black carbon and dust particles to be capable...
to serve as IN. Observations suggest that this results in an upper limit assumption on IN numbers encountered in the atmosphere [DeMott et al., 2003; Haag et al., 2003a]. It is remarkable that even with these upper limit IN concentrations the ice crystal nucleation rate is frequently limited by the number of available IN. This indicates the potential of heterogeneous ice nucleation to induce cirrus cloud features different from those obtained assuming pure homogeneous nucleation. In the case of homogeneous freezing, the crystal nucleation rate is only limited by the number of hygroscopic aerosols in the tropical upper troposphere where vertical velocities are large and the number of aerosols is small. Because of this limitation, ECHAM4 studies showed that the Mount Pinatubo eruption enhanced the ice crystal number concentration formed by homogeneous freezing in this region [Lohmann et al., 2003].

3.2. Ice Water Content and Ice Crystal Number Concentrations

The impact of the different nucleation schemes on the zonal mean ice crystal number concentration and ice water content is shown in Figure 3. The ice water content is averaged over cloudy and cloud-free regions and periods, whereas the number of ice crystals is averaged only over cloudy regions and periods. Maxima in ice water content and ice crystal number concentrations in simulation HOM are associated with topographically induced updrafts, with anvil cirrus outflow from deep convection in the tropics and with extratropical cyclones. Arctic ice clouds are also characterized by a high ice crystal number concentration of up to $2 \text{ cm}^{-3}$ but at lower ice water contents. The vertically integrated ice crystal aggregation and accretion rate is not that much smaller in the Arctic than at midlat-
itudes (not shown) but the nucleation rate is smaller in the Arctic than in midlatitudes (Figure 2), so that the high ice crystal concentrations are likely caused by advection from midlatitudes. Ice crystal number concentrations of this order of magnitude have been observed in the Arctic by the forward scattering spectrometer probe (FSSP) [Lohmann et al., 2001], but there is an ongoing debate as to how reliably ice crystal measurements can be inferred from the FSSP, as, for instance, discussed by Field et al. [2003].

When homogeneous freezing commences at 130% relative humidity with respect to ice, i.e., almost unlimited ice nuclei are assumed, then the number of ice crystals is increased by up to 3 cm$^{-3}$, which is consistent with Kärcher and Lohmann [2003], and the ice water content is slightly increased. If the number of freezing nuclei is limited by the number of dust and black carbon aerosols, the ice crystal number concentrations are lowered by more than 1 cm$^{-3}$ in the upper troposphere. The largest decrease of more than 10 cm$^{-3}$ occurs in the tropics at 150 hPa. These fewer particles grow to larger sizes and with that start to sediment serving as seeds to initiate accretion with ice crystals and cloud droplets in lower level clouds. This increases the global mean precipitation rate (Table 1) and decreases the ice water content by up to 1 mg kg$^{-1}$.

### 3.3. Impact of the Freezing Parameterization on Precipitation and the Radiation Budget

Figure 4 shows the annual zonal mean ice water path, vertically integrated ice crystal number concentration, precipitation and total cloud cover for the simulations HOM, HOM130% and HET130%. Ice water path and the vertically integrated ice crystal number concentration are averages over cloudy and cloud-free periods. Unfortunately, there are no global observations of ice water path or ice crystal number concentration up to now. We include observations of precipitation from the Global Precipitation Data Set [Stendel and Arpe, 1996], and TCC from ISCCP [Rossow and Schiffer, 1999] and Surface-Based Observations [Hahn et al., 1994] (Lower Value).

### Table 1. Global Annual 5-Year Mean Ice Water Path (IWP) in g m$^{-2}$, Vertically Integrated Number of Ice Crystals (Ni) Averaged Over Cloudy and Cloud-Free Periods in 10$^6$ cm$^{-3}$, Shortwave (SW) and Longwave (LW) Radiation at the Top of the Atmosphere in W m$^{-2}$, Precipitation (PR) in mm d$^{-1}$, and Total Cloud Cover (TCC) in Percent for the Simulations HOM, HOM130%, and HET130% Together With Observations (OBS) of the Radiation Budget From ERBE [Ramanathan et al., 2001], Precipitation From the Global Precipitation Data Set [Stendel and Arpe, 1996], and TCC From ISCCP [Rossow and Schiffer, 1999] and Surface-Based Observations [Hahn et al., 1994] (Lower Value)

<table>
<thead>
<tr>
<th></th>
<th>HOM</th>
<th>HOM130%</th>
<th>HET130%</th>
<th>OBS</th>
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<td>6.7</td>
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<td>2.64</td>
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<tr>
<td>TCC</td>
<td>59.8</td>
<td>59.7</td>
<td>59.9</td>
<td>62/67</td>
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Figure 3. Five-year average of the annual zonal mean latitude versus pressure cross sections of ice water content (IWC) (mg kg$^{-1}$) including cloudy and cloud-free periods and ice crystal number concentrations (IC) (cm$^{-3}$) averaged only over cloudy periods for the simulation HOM, and the differences HOM130% - HOM, and HET130% - HOM. Areas with less than 1% of cloud occurrences are not shown.
In simulation HOM an order of magnitude higher ice crystal concentrations are simulated in the tropics than in HET130%. The high ice crystal number concentrations in simulations HOM and HOM130% result in a less efficient precipitation formation. This could be a reason for the smaller than observed global annual mean precipitation of 2.67 mm d$^{-1}$ and 2.65 mm d$^{-1}$, respectively, as compared to the global mean observed precipitation from GPCC averaged over 1986 to 1994, which amounts to 2.78 mm d$^{-1}$.

Even though the global mean precipitation in all simulations is in good agreement with the observations, all simulations tend to underestimate precipitation in the subtropics. Reasons for these differences in precipitation will be further discussed in chapter 4.

Cloud cover is observed from ISCCP and from ground based observations. While ISCCP estimates a global mean cloud coverage of 67%, the surface based observations only estimate a global mean cloud coverage of 62%. All our simulations predict a global mean cloud cover of only 60%, which is mainly caused by underestimating cloud cover in the subtropics (Figure 4). Deviations poleward of 60° may partly be caused by the inability of ISCCP to detect clouds over ice surfaces and the fewer observational stations at high latitudes.

The differences in ice water path and ice crystal number concentration between the different freezing simulations are also reflected in the radiation budget at the top of the atmosphere. The reduction in ice water path and ice crystal number in simulation HET130% as compared to...
simulation HOM130% leads to a higher absorption of solar radiation of 2 W m\(^{-2}\) (Table 1). The net shortwave radiation at the top of the atmosphere in all simulations agrees within 2 W m\(^{-2}\) with observations from the Earth Radiation Budget Experiment (ERBE) of 238 W m\(^{-2}\) [Ramanathan et al., 2001]. On the other hand, the lowest ice water path in HET130% results in 3 W m\(^{-2}\) more outgoing longwave radiation emitted to space than in HOM and HOM130%. Even in the ERBE data the radiation budget at the top of the atmosphere is not balanced, but there is a 3 W m\(^{-2}\) disagreement between the absorbed shortwave and outgoing longwave radiation. Ramanathan et al. [2001] argue that this difference may be real, due to instrument artifacts or a combination of both. In all our simulations this discrepancy is slightly larger. In HET130% this is caused by too much absorption of solar radiation while there is too little outgoing longwave radiation in HOM and HOM130% caused by the higher ice water paths and ice crystal number concentrations in these experiments.

[25] In summary, the differences between the various freezing simulations are rather small and their deviations from observational data generally fall within the observational uncertainty. Kärcher and Ström [2003] showed that the ice crystal number concentrations from simulation HOM are smaller but on the same order of magnitude as the in situ observations taken during the INCA campaign. Given that the ice crystal number concentrations in HET130% are smaller than in HOM and that the atmosphere never has as many ice nuclei freezing at 130% relative humidity with respect to ice as assumed in simulation HOM130%, we conclude that among the scenarios investigated here, simulation HOM is the most realistic scenario in terms of the considered cirrus cloud properties.

### 4. Summary and Outlook

[26] In this paper we evaluated the global impact of homogeneous freezing of supercooled hygroscopic aerosol particles (simulation HOM) versus heterogeneous freezing initiated by immersion nuclei at 130% relative humidity with respect to ice on ice cloud properties, precipitation and the radiation budget. We compared an experiment in which ice nucleation is limited by the number of mineral dust and black carbon aerosols as suggested by laboratory measurements (simulation HET130%) and recent field campaigns [DeMott et al., 2003] with an experiment potentially allowing all supercooled aerosols to initiate freezing at the same freezing relative humidity (simulation HOM130%). Limiting the number of ice nuclei to black carbon and dust translates into fewer ice crystals, more precipitation, and a smaller ice water path than in simulations HOM and HOM130%.

[27] One question is, how many ice nuclei are realistic? Minikin et al. [2003] showed a much higher number concentration of nonvolatile particles in the upper troposphere than present in simulation HET130%. This would suggest that the limitation of ice crystal formation by ice nuclei in simulation HET130% may be an artifact. The discrepancy between the ice nuclei concentration in simulation HET130% and in the observations could result from sea salt, primary organic or metallic aerosol particles in the observations or may be caused by having chosen a mode radius for dust aerosols that is larger than observed. On the other hand, the study by Haag et al. [2003b] concluded based on modeling results that only <0.01 cm\(^{-3}\) particles out of 20 cm\(^{-3}\) measured nonvolatile particles could have served as ice nuclei in order to match the INCA observations over Scotland. This suggests that the atmosphere probably has even fewer ice nuclei than assumed in HET130%.

[28] Clearly, one shortcoming of this study is the neglect of the competition between homogeneous and heterogeneous freezing, which would take place in nature. The only way we could have considered it here would have been an ad-hoc assumption. Instead, we opted to analyze the sensitivity of ice crystal formation to rather simple assumptions about heterogeneous freezing and analyze the competition between homogeneous and heterogeneous freezing once we have derived an appropriate parameterization for that. It is necessary to learn how a GCM behaves under such idealized conditions of heterogeneous freezing in order to interpret future studies that include more complex freezing pathways.

[29] Another shortcoming is the neglect of an adequate parameterization of subgrid-scale cloud cover that takes temperature and humidity fluctuation into account. This is beyond the scope of this study but should be considered in future.

[30] The difficulties in simulating precipitation as discussed in section 3.3 partly arise from the simplified treatment of the nucleation process. A mixture of heterogeneous and homogeneous ice nucleation as, for instance, discussed in Haag et al. [2003b], will allow the few heterogeneousy formed ice crystals to grow to precipitation size particles that can sediment, while the majority of the particles formed by homogeneous freezing will remain aloft. This results in a bimodal ice crystal size spectrum. In order to describe this process adequately in the GCM, we would need separate prognostic equations for ice crystals formed by homogeneous and by heterogeneous nucleation, respectively. This will be done in a future study.

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