The impact of aerosols and gravity waves on cirrus clouds at midlatitudes

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[1] We use a Lagrangian microphysical aerosol-cloud model to simulate cirrus clouds along trajectories at northern hemisphere midlatitudes. The model is constrained by recent in situ observations in terms of aerosol size distributions, freezing relative humidities, cooling rates, and cirrus particle sedimentation rates. Key features include competition between insoluble and volatile aerosol particles and temperature perturbations induced by high-frequency gravity waves. Recent analyses of field measurements have revealed the crucial roles both factors play in cirrus formation. We show that most cirrus form in synoptic cold pools, but with microphysical properties determined by mesoscale variability in vertical velocities. Heterogeneous ice nuclei (IN) present in concentrations probably typical for northern midlatitude background conditions (<0.01–0.03 cm$^{-3}$) significantly modify cirrus properties but do not control cirrus formation. The key effect of IN on cirrus clouds is a reduction of the number of ice crystals. This indirect aerosol effect results in reduced cloud albedo due to increased effective radii and decreased ice water contents, as well as in nonlinear changes of cirrus occurrence, optical extinction, and fraction of clouds that are subvisible. The nonlinear dependence of the three latter quantities appears when IN concentrations rise above a threshold concentration of some 0.01 cm$^{-3}$, the exact value depending on the cloud formation temperature, cooling rate, and IN freezing relative humidity. In such conditions, IN become the controlling factor in cirrus formation, diminishing the role of homogeneous freezing. Ice nuclei with freezing thresholds near ice saturation are capable of introducing strong changes of cloud properties, even at low concentrations. Optically thin and subvisible cirrus are particularly susceptible to IN. The presence of a small number of IN (0.001 cm$^{-3}$) can significantly increase their occurrence frequencies. If such clouds predominantly form on IN, they might be affected by anthropogenic activities. Changes in upper tropospheric cooling rates and ice-forming aerosols in a future climate may induce changes in cirrus occurrence that are comparable in magnitude to observed decadal trends in global cirrus cover.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; KEYWORDS: cirrus clouds, ice nuclei, gravity waves


1. Introduction

[2] Ice crystals in cirrus clouds form by freezing of atmospheric aerosol particles. Several modes of ice nucleation exist [Pruppacher and Klett, 1997], broadly categorized into homogeneous and heterogeneous modes. Changes in the freezing mode alter the number and size of the nucleated ice crystals and thus changes in cirrus cloud properties.

[3] By determining temperature and cooling rates, dynamical processes impact cirrus macrophysical properties and microphysical structure [Lynch et al., 2002]. A recent analysis of temperatures, vertical winds, and total crystal concentrations taken from the Interhemispheric Differences in Cirrus Properties From Anthropogenic Emissions (INCA) field experiments emphasized the key role of mesoscale variability in vertical velocities, or cooling rates, in controlling cirrus properties [Kärcher and Ström, 2003]. Similar findings concerning the distributions of ice crystal concentrations and cooling rates in cirrus have been reported based on an analysis of the Subsonic Aircraft: Contrail and Cloud Effects Special Study (SUCCESS) field measurements [Luo et al., 2004].

[4] The analysis of INCA measurements of relative humidity over ice (RHI) provided compelling evidence for the existence of a heterogeneous freezing mode in midlatitude cirrus clouds [Haag et al., 2003]. The authors pointed
out that the cirrus did not exclusively form on heterogeneous ice nuclei (IN), and that homogeneous freezing was the dominant mechanism by which the ice crystals had formed. Direct measurements of IN during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers—Florida Area Cirrus Experiment (CRYSTAL-FACE) revealed the important role mineral dust particles could play in nucleating ice crystals at RHI close to saturation [DeMott et al., 2003a].

[5] In view of the strong dynamical control of cirrus cloud formation, Kärcher and Ström [2003] argued that it may be difficult to separate the effects of aerosol changes and dynamical changes on cirrus cloud cover and microphysical properties in observations. This difficulty served as one motivation to conduct the present modeling study, which builds on the experimental evidence from INCA, SUCCESS, and CRYSTAL-FACE noted above. (1) Variability in vertical velocities in the range 10–100 cm s\(^{-1}\) caused by gravity wave-induced, mesoscale temperature fluctuations explains the observed high number densities (0.1–10 cm\(^{-3}\)) of homogeneously formed ice crystals in cirrus. (2) Freezing in cirrus clouds can be initiated by less than 0.01–0.03 cm\(^{-3}\) of IN nucleating ice around RHI = 130% or at even lower values.

[6] By using these and other observational constraints in a microphysical aerosol/cirrus model as a guide, we explore the range of possible effects of aerosol and dynamical changes on cloud microphysical and optical properties and on the frequency of cloud occurrence. While we believe that the general picture of cirrus formation given here is well supported by available observations, we cannot rule out that yet unexplored details of ice nucleation and dynamical variability will impact the scenarios described in this work. What follows is a comprehensive trajectory model study considering the combined impact of aerosol properties, freezing mechanisms, and atmospheric dynamics on the near-global distribution and properties of cirrus clouds. Current global atmospheric models are not capable of describing this whole suite of processes. More direct measurements of IN and dynamical variability along with improved parameterization schemes are necessary to enable such global model studies in the future.

[7] Cirrus changes are predicted on a global (hemispheric) scale by employing domain-filling trajectories from a weather forecast model coupled to an aerosol-cirrus microphysical model, covering the northern midlatitudes during fall in the year 2000 (section 2). Results in terms of total ice crystal number density and mean size, cloud ice water content, cloud optical extinction, and spatial coverage are presented and discussed in section 3. Besides investigating the indirect aerosol effect on cirrus, we also delineate important implications for subvisible cirrus forming in the tropopause region. A summary of our findings and the conclusions are given in section 4.

2. Model Description

2.1. Global Fields, Trajectories, and Microphysics

2.1.1. Temperature, Water Vapor, and Relative Humidity

[8] The aerosol-cirrus microphysical model APSC (Advanced Particle Simulation Code) [Kärcher, 2003], the global thermodynamic and wind fields taken from the European Centre for Medium-Range Weather Forecasts (ECMWF), and the trajectory model FLEXPART [Stohl et al., 2003] have been described in more detail in our preceding study [Haag et al., 2003; Haag, 2004]. We summarize the main issues.

[9] The trajectory calculations were driven with operational ECMWF data, covering the time period September to November 2000. The ECMWF operational forecast model had a spectral truncation of T511 (equivalent to ~40 km horizontal resolution) and 60 vertical levels. In FLEXPART 160,000 particles (air parcels) are initially distributed homogeneously in the atmosphere, according to the distribution of atmospheric mass, and are then followed throughout the simulation. While we do not claim that trajectories are accurate over the entire duration of the simulation, we use them in a statistical sense where accuracy is required only for the trajectory segments between the reinitializations of the RHI values (see below in this section).

[10] In the combined FLEXPART/APSC simulations, we only considered particles initially lying within the global midlatitude northern hemisphere (20°–80°N). The particles can leave and reenter this domain and exhibit large vertical displacements. We ran the APSC model on 10,125 sections of these particle tracks lying above 400 hPa and considered only data points with temperatures below 235 K to analyze the properties of cirrus clouds. All the results shown in this work are taken from data sets confined to a region from 2 km below to 1 km above the local tropopause (hereafter referred to as tropopause region). In this way, depending on the location and time, upper tropospheric and lower stratospheric air masses are sampled in the model.

[11] Figure 1 shows the 3-monthly mean fields of RHI (top panel), temperature T (middle panel) and H\(_2\)O mixing ratio (bottom panel) calculated by FLEXPART using the synoptical ECMWF results. These quantities describe the large-scale moisture and temperature distributions that eventually control the regions in which cirrus clouds form and persist, as detailed later in section 3.1 and Figure 5.

[12] Moist tropopause regions are predicted east of Scandinavia and north of 60°N owing to the low temperatures, and over Greenland, northern Canada, and Alaska. Part of the high RHI might be caused by topographically induced, enhanced vertical velocities, especially in the lee of Greenland and Scandinavia. Enhanced RHI is also predicted along the storm tracks over the North Atlantic around 60°W extending toward Europe and over the west Pacific between 180°W and 140°W. While we cannot exclude that the moist region north of 60°N is partly influenced by temperature errors in the model, the moist regions in the storm tracks are connected to cyclonic activity and have been described in other studies [e.g., Eckhardt et al., 2004].

[13] The ECMWF model provides upper tropospheric RHI fields below ice saturation that are in good agreement with in situ data [Ovarlez and van Velthoven, 1997]. However, the ECMWF model does not predict ice supersaturation, in contrast to observations [Spichtinger et al., 2003]. Our method to predict realistic RHI fields in cirrus-forming regions despite this deficiency is as follows. At RHI values above 95%, we keep the H\(_2\)O mixing ratio fixed on each trajectory and let RHI vary according to changes of T, air pressure, and the calculated rates of H\(_2\)O vapor...
deposition onto and evaporation from aerosol and cirrus ice particles. The H$_2$O mixing ratio is reinitialized from the actual ECMWF fields after the cirrus cloud vanishes, typically forced by synoptic warming. If cirrus formation does not occur within 6 hours after RHI first increased above 95%, the H$_2$O mixing ratio is likewise reinitialized.

The trajectories follow the motions caused by atmospheric waves sufficiently long to be resolved in the employed version of the ECMWF model. However, smaller-scale gravity waves, non-hydrostatic waves, and deep convection are not explicitly resolved. The trajectories reflect these processes only to the extent that the physical
parameterizations feed back on the grid-scale winds, generally underestimating mesoscale variability.

In our case, sampling the ECMWF vertical winds along the trajectories leads to the dotted distribution of vertical velocities shown in Figure 2, with a mean of 1 cm s$^{-1}$ or an equivalent adiabatic cooling rate of 0.35 K h$^{-1}$. This does not agree with available in situ measurements. As an example, we show the updraft distribution (dashed) taken during INCA over Scotland, giving an average of 26.2 cm s$^{-1}$ indicative of mesoscale variability.

To account for high frequency gravity wave signatures in the temperatures finally used to drive the APSC, we superimpose rapid oscillations to the synoptic temperatures, with random amplitudes (up to 2 K) and a mean period (1,200 s) guided by the INCA observations. While the distribution of amplitudes is uniform, the periods are normally distributed with a standard deviation of 400 s. The resulting distribution (solid) in Figure 2 has a slightly higher mean updraft speed (32.2 cm s$^{-1}$) than the measurements indicate. The corresponding distribution of cooling rates (not shown) with mean values 1–10 K h$^{-1}$ are very similar to those inferred from SUCCESS measurements [Luo et al., 2004] and from tropical observations [Jensen and Pfister, 2004]. Later we will vary the mean period of the temperature waves to mimick faster or slower cooling.

The temperature oscillations and their effect on RHI are not be discernable in the mean fields shown in Figure 1, as they average out on longer timescales. Examples of fluctuations of $T$ and RHI and their impact on cirrus properties are presented in section 2.2 (see Figure 4). The ad-hoc treatment of mesoscale temperature fluctuations in the APSC model is in rough agreement with the data and appears to be sufficient for our purposes. However, it would be useful to improve this part of the model in future work, in particular in case studies.

2.1.2. Aerosol/Cirrus Microphysics

We use the APSC along each trajectory. The code includes detailed particle microphysics and an approximate treatment of the loss of ice crystals from the tropopause region through sedimentation. The time step was held variable to obtain accurate numerical solutions. We sampled the microphysical information discussed in this work uniformly every 6 min.

Cirrus clouds form directly from aerosol particles by homogeneous and heterogeneous freezing [Koop et al., 2000; Kärcher and Lohmann, 2003]. Ice crystals are assumed to be hexagonal columns with an aspect ratio of 3 for radii of volume-equivalent spheres above 12.5 μm, becoming increasingly spherical at smaller sizes, and completely spherical below 7.5 μm. Only water vapor was allowed to interact with aerosol particles and ice crystals, and no equilibrium assumptions are made in the model with respect to particle nucleation and growth. The ice crystals grow by vapor diffusion, with the diffusional growth rate including gas kinetic and ventilation corrections as well as a shape-dependent capacity factor.

The dry aerosol size distribution consists of two lognormal modes, with a total number density of 400 cm$^{-3}$ (0.1 cm$^{-3}$), a mean number radius of 18 nm (420 nm), and a geometric standard deviation of 1.45 (1.3), respectively. In case HOM, all particles were allowed to freeze homogeneously and are assumed to consist of aqueous sulfuric acid. On the basis of sensitivity studies carried out with the APSC, we expect no significant changes of our results upon using other aqueous aerosol compositions. In particular, the total number of ice crystals nucleated from liquid particles is insensitive to variations of the homogeneous freezing threshold or details of the nucleation rate. In the simulations labeled MIX, the particles in the smaller mode are still assumed to be liquid, but the larger mode particles have a variable total number density $n$ and act as IN with variable freezing relative humidities.

Nominal aerosol freezing thresholds are defined such that one particle in equilibrium with ambient H$_2$O with $r = 0.25$ μm freezes in 1 s at a certain value of RHI. The freezing thresholds of liquid particles depend on $T$ and are generally higher than 145%. Two cases with IN freezing thresholds of 130% and 105% (mimicking nearly perfect IN) are considered.

Liquid particles do not limit the number of cirrus ice crystals owing to their high number density. Depending on the cooling rates at the point of ice formation, particles with very different sizes may contribute to cirrus formation. After each cirrus event, the aerosol size distribution is reinitialized. The concentrations of IN are only poorly known from measurements and are varied within the range 0.001–0.1 cm$^{-3}$ in the cirrus simulations, with values $<0.01$ cm$^{-3}$ regarded to be more typical for tropopause conditions. In collecting the few available direct measurements of IN, it seems that IN concentrations below 0.01–0.03 cm$^{-3}$...
appear to characterize northern midlatitude background conditions [Haag et al., 2003; DeMott et al., 2003a, and references therein].

2.1.3. Treatment of Sedimentation

In section 3 we perform statistical analyses of cloud parameters in the tropopause region. In this regard, the following method to treat the vertical redistribution of ice crystals should provide a reasonable estimate for the actual loss of ice particles and ice water due to sedimentation, a process known to be notoriously difficult to model, especially when using a trajectory approach.

We reduce the ice crystal number density every time step $\Delta t$ by the factor $\exp[-\Delta t/(\Delta z/v)]$ in each size bin. The sedimentation timescale is given by $\Delta z/v$, with the size- and shape-dependent ice crystal terminal fall speed $v$ and the vertical thickness $\Delta z = 1$ km. The value of $\Delta z$ is chosen to bring calculated and observed frequency distributions of the total number densities of ice crystals, $n_i$, measured during INCA [Kärcher and Ström, 2003] into good agreement.

In the following, we justify the use of the parameter $\Delta z = 1$ km to determine the sedimentation losses. We first seek to find a good agreement of the distributions of $n_i$ observed over Chile (label SH), where freezing was consistent with our case HOM [Haag et al., 2003]. The left panel in Figure 3 depicts this frequency distribution of $n_i$ as thick solid stair steps. A model result using an unimodal aerosol size distribution based on case HOM is shown as a dotted distribution; here, only particles from the small mode are present. More ice crystals at high concentrations exist, indicating that the distribution of cooling rates in the model contains a larger fraction of very high values than observed.

More importantly, the dotted model curve decreases monotonously toward low $n_i$, while the SH measurements develop a secondary maximum around $n_i = 0.02$ cm$^{-3}$. We can roughly account for this maximum in the model by adding the few aerosol particles from the large mode, see thin solid stair steps. In fact, a large particle mode was also observed during INCA [Kärcher and Ström, 2003]. Compared to the dotted distribution, high $n_i$-values are reduced, while low $n_i$-values occur more frequently. This is caused by an aerosol size effect in the homogeneous freezing process, which becomes notable for large freezing particles in combination with rather high cooling rates and low temperatures [Kärcher and Lohmann, 2002].

To assess the effect of the parameterized loss of ice crystals from the tropopause region, we finally show the frequency distribution with bimodal aerosol but without sedimentation as dashed stair steps. Compared to the thin solid distribution and the SH observations, the frequency of occurrence of $n_i$-values near 0.1 cm$^{-3}$ is dramatically underestimated while the number of cases with concentrations $1-10$ cm$^{-3}$ is reduced. If the ice crystals are not allowed to fall out of the formation layer, particularly those with small concentrations and hence large sizes, the probability to nucleate high concentrations is reduced owing to increased deposition of H$_2$O on preexisting ice crystals.

We have seen that we can achieve a satisfactory agreement between the INCA-SH observations in case HOM when we account for both, a bimodal freezing aerosol spectrum and sedimentation. Do we also find a good agreement with the observations taken over Scotland (label NH) shown as thick solid stair steps in the right panel of Figure 3? In this case, we know that heterogeneous IN were present, most likely with concentrations $\leq 0.01$ cm$^{-3}$. We show the result of the model case MIX 0.01 as thin solid stair steps. For $n_i > 20$ cm$^{-3}$, a few data were taken during

Figure 3. Observed and modeled distributions of $n_i$ with various assumptions for freezing aerosols and sedimentation of ice crystals. Best agreement with measurements is achieved for a bimodal aerosol size distributions (as observed) and an average vertical thickness of 1 km to compute sedimentation timescales for falling ice crystals. In the observed distributions, the leftmost peaks are generated by the lower detection limit of the instrument measuring $n_i$; this has not been taken into account in the modeled distributions.
convective episodes with very high cooling rates which are not properly represented in the model. As the modeled cooling rate spectrum has been fitted to reproduce the mean value of $n_i$, it averages out the high $n_i$-values compared to the NH measurements.

[29] On the other hand, the model is in fair agreement toward low $n_i$-values. Note that in the observed distribution, the high occurrence frequency of 0.067 in the 0.01 cm$^{-3}$-bin and zero in the next bin to the left is a discretization artifact that could be smoothed out by using other bin widths. In sum, we may regard case MIX 0.01 (with an IN freezing threshold of 130%) as being close to the INCA-NH observations, although lower IN concentrations or multiple IN types all freezing at or above 130% would also be consistent with the data.

2.2. Exemplary Simulations

[30] Typical results of combined FLEXPART/APSC simulations along single trajectories are shown in Figure 4. In case HOM-S (top panel), all particles were allowed to freeze...
homogeneously, and mesoscale gravity waves are absent (only synoptic cooling). Three cloud events occur on day 2 of the simulation. Owing to the moderate cooling rates of the order 20–30 K d−1, only ~0.01 cm−3 ice crystals form at temperatures near 230 K. During the existence of the ice phase, RHI decreases from the freezing threshold toward ice saturation, but never reaches saturation. The small concentrations and hence large sizes of the ice particles lead to a quite rapid decrease of the simulated crystal number density owing to efficient sedimentation. The time evolution of RHI during the cloud-free phases exhibits an instantaneous decrease three times on days 1 and 3 of the simulation. This is caused by reinitializations of the H2O mixing ratio, as detailed in section 2.1.1.

[31] In case HOM (middle panel), the aerosol freezing properties are unchanged, but rapid temperature oscillations are superimposed to the synoptic temperatures. Compared to case HOM-S, the rapid cooling rates lead to much higher ice crystal concentrations between 0.1–1 cm−3 (solid ni-curves) than in case HOM-S. The difference in ni is caused by the wave nature of T(t) and by its statistical distribution. High values of ni lead to smaller ice particle sizes and less efficient sedimentation. This causes the decreases of ni in the HOM cloud events to be slower than those in the HOM-S events. Note that the times when clouds form also differ between HOM and HOM-S.

[32] Cloud onset time, crystal concentrations, and cloud life time also change upon changing the stochastic sequence of cooling and warming, keeping the mean wave periods and fluctuation amplitudes unchanged. This is illustrated by the dashed ni-curves in the middle panel, showing the resulting new evolution of ni. (The time evolutions of T and RHI are not shown for this case.) Although we may expect similar distributions of cloud properties when averaged over a sufficiently large number of cloud events, this exercise shows that it is difficult to predict single cloud properties when the exact sequence of cooling rates is not known. In other words, predicting cloud properties will require a statistical evaluation including an ensemble of stochastic trajectories.

[33] In the model simulation MIX (bottom panel), the aerosols in the larger mode freeze early at RHI = 130% so that competition of the homogeneous nuclei with few IN during ice initiation becomes possible. The dashed and solid curves show ni assuming 0.001 cm−3 and 0.1 cm−3 IN, respectively. The other curves represent T(t) and RHI(t) for the latter case only. The dynamics in case MIX (essentially T(t) along the trajectory) is the same as used in case HOM. The simulation MIX demonstrates how cirrus cloud properties can be modified by the presence of heterogeneous IN. In particular, the freezing onsets and the number of cirrus events change: compared to 3 cirrus events in case HOM, 2 (5) clouds form with different life times and properties in case MIX 0.1 (MIX 0.001). This is because some ice crystals form earlier than in case HOM, thereby alter the evolution of RHI and thus the conditions for subsequent homogeneous freezing.

[34] The case MIX 0.1 (solid ni-curves), for which the corresponding evolution of RHI is shown, serves to illustrate an interesting dynamical feature. In the second cirrus event, a large number of small ice crystals form initially (ni ≈ 15 cm−3 with a mean number radius of ri ≈ 12 µm). The ice particles rapidly take up water molecules in excess of ice saturation, resulting in strongly damped oscillations of RHI that are caused by ongoing temperature fluctuations. In fact, the calculation of the timescale τg for depletion of H2O yields a value around 9 s. The ice particles continue to grow and simultaneously sediment out of the trajectory because the air mass synoptically cools for another 1.5 days. Evaluating τg on day 2 (day 3) with ni = 1 cm−3 (0.15 cm−3) and ri = 17 µm (20 µm), τg takes the value 89 s (540 s). This increase in τg relative to the mean temperature wave period of 1200 s explains the enhanced oscillation amplitudes of RHI with increasing cloud age. For example, at the end of the cloud life time, typical mean RHI amplitudes are of the order ±5%. The RHI amplitudes can be higher for lower values of ni, see the third cirrus event in case HOM (middle panel).

2.3. Comments on Model Uncertainties

[35] A summary of the model experiments studied in section 3 is given in Table 1. These simulations assume that small-scale gravity waves are continuously present without geographical differences and with the same statistical properties. Despite the fact that the presence of waves does mainly affect the nucleation of ice in supersaturated regions, with little impact on the aerosol behavior in unsaturated air, this is a simplification, as the real sources and properties of gravity waves may vary spatially and temporally.

[36] Another simplification concerns the treatment of the freezing aerosol. We assume that IN are present everywhere through the simulation period, and ignore geographical variability within the season investigated. We have not

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### Table 1. Summary of Model Experiments Carried Out in This Work

<table>
<thead>
<tr>
<th>Experiment</th>
<th>τ, s</th>
<th>n0, cm−3</th>
<th>RHIC, %</th>
<th>focc, %</th>
<th>n0c, cm−3</th>
<th>r0, µm</th>
<th>IWC, mg m−3</th>
<th>E, km−1</th>
<th>focc, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>HOM-S</td>
<td>0</td>
<td>–</td>
<td>5.2 (6.1)</td>
<td>8 × 10−3 (0.02)</td>
<td>60.0 (65.8)</td>
<td>0.5 (2.7)</td>
<td>0.02 (0.08)</td>
<td>59</td>
<td></td>
</tr>
<tr>
<td>HOM</td>
<td>1200</td>
<td>0.01</td>
<td>8.5 (9.2)</td>
<td>0.6 (3.6)</td>
<td>13.8 (20.6)</td>
<td>4.0 (10.0)</td>
<td>0.6 (1.3)</td>
<td>15</td>
<td></td>
</tr>
<tr>
<td>HOM 900</td>
<td>900</td>
<td>0.01</td>
<td>9.0 (9.9)</td>
<td>0.8 (10.1)</td>
<td>12.6 (18.8)</td>
<td>4.5 (12.1)</td>
<td>0.7 (2.0)</td>
<td>16</td>
<td></td>
</tr>
<tr>
<td>HOM 1500</td>
<td>1500</td>
<td>0</td>
<td>7.4 (8.5)</td>
<td>0.2 (1.3)</td>
<td>16.7 (25.0)</td>
<td>3.0 (7.6)</td>
<td>0.3 (0.8)</td>
<td>18</td>
<td></td>
</tr>
<tr>
<td>MIX 0.001</td>
<td>1200</td>
<td>0.001</td>
<td>9.1 (9.9)</td>
<td>0.3 (2.1)</td>
<td>15.8 (24.3)</td>
<td>3.0 (9.2)</td>
<td>0.3 (1.2)</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>MIX 0.01</td>
<td>1200</td>
<td>0.01</td>
<td>9.0 (9.5)</td>
<td>0.2 (2.2)</td>
<td>15.8 (24.3)</td>
<td>3.0 (9.2)</td>
<td>0.3 (1.2)</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>MIX 0.1</td>
<td>1200</td>
<td>0.001</td>
<td>7.7 (8.5)</td>
<td>0.07 (2.1)</td>
<td>17.9 (24.8)</td>
<td>2.4 (7.3)</td>
<td>0.2 (0.8)</td>
<td>21</td>
<td></td>
</tr>
<tr>
<td>MIX-IN</td>
<td>1200</td>
<td>0.001</td>
<td>105.0 (117.7)</td>
<td>0.03 (2.9)</td>
<td>11.7 (19.5)</td>
<td>1.3 (7.8)</td>
<td>0.1 (1.0)</td>
<td>40</td>
<td></td>
</tr>
</tbody>
</table>

*The experiments differ by the assumed mean period τ of small-scale temperature oscillations, the number density n0 of heterogeneous ice nuclei, and their freezing threshold relative humidities RHIC. The median (mean) values for frequency of cloud occurrence focc, total ice crystal number density n0c, mean number radius r0, total ice water content IWC, 1µm cloud extinction, and fraction of clouds that are subvisible focc, are derived from the calculated distribution functions representative for the midlatitude northern hemisphere in the full season. Mean and median values might differ significantly in some cases, indicating skewed distribution functions. In such cases, median values are less affected by outliers, and we may consider them more representative than the respective mean values.*
considered the presence of more than one IN type, possibly with different freezing characteristics, contributing to cirrus formation.

These simplifications are only partly motivated by our wish to understand basic processes with a minimum of assumptions. More importantly, it is hardly possible to improve this approach to date, because of the lack of microphysical measurements of IN that could be used to constrain global aerosol properties; even if technically feasible, we have not yet achieved the necessary understanding to properly parameterize wave-induced temperature fluctuations globally.

Nevertheless, our base cases HOM and MIX 0.01 agree with state-of-the-art cirrus observations in terms of important cloud properties. Hence we regard our simulations as reasonably well defined basic studies, inasmuch as we are mainly interested in the differences in cirrus cloud properties that arise from changes in aerosol composition and vertical air motion. This ensures that we examine realistic cirrus scenarios and not just perform an academic exercise. We vary the key unknown parameters of this problem (IN freezing threshold, total IN concentration, and cooling rates) in order to explore the possible range of cirrus changes induced by such changes in aerosol and dynamical properties.

3. Results and Discussion

3.1. Cirrus Cloud Occurrence Frequency

We have calculated the frequency of occurrence of cirrus clouds in the global midlatitude northern hemisphere. We define on a $7.5^\circ \times 7.5^\circ$ latitude/longitude grid (see

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**Figure 5.** Frequency of cirrus cloud occurrence based on the RHI field directly taken from FLEXPART trajectories based on winds from the ECMWF model (top), and from combined APSC/FLEXPART simulations using the synoptic temperatures (middle) and synoptic temperatures with superimposed small-scale temperature oscillations (bottom). Geographical patterns of cirrus occurrence are similar in all three cases, indicating that the formation of synoptic cold pools is a prerequisite for cirrus formation, independent of the presence of small-scale temperature fluctuations. In contrast to ECMWF, the microphysical model takes into account kinetic limitations during ice evaporation (in cases HOM and HOM-S) and rapid local cooling rates (in case HOM only), both prolonging the cirrus lifetimes.
dictated by the small-scale cooling rates, as exemplified in Figure 4.

[45] Second, the latter argument implies that values of \( n_i \) are higher and \( r_i \) are smaller in case HOM than in HOM-S (see Table 1), and sedimentation losses are strongly reduced in case HOM. On average, within a synoptic cold pool, this causes a substantial increase in the lifetime of the cirrus clouds and therefore a higher frequency of occurrence over the investigated period.

[46] In conclusion, the calculated cirrus cloud lifetimes are too short in the ECMWF model. This does not mean that the overall effects of cirrus are misrepresented in that model; we rather believe that errors introduced by the missing ice microphysics are compensated by proper tuning of sedimentation and precipitation processes, both of which are based on simplified parameterization schemes. It is not our intention to quantify such effects by the comparison of the cases HOM-S and HOM with the ECMWF data. We only point out the importance and need of realistic microphysical parameterizations of cirrus cloud formation in global atmospheric models.

[47] How does the frequency of occurrence change when the distribution of high frequency gravity waves is varied? In what follows, we increase and decrease the mean oscillation period of the temperature fluctuations from 1200 s to 1500 s and 900 s, respectively, keeping the statistical distribution of cooling rates constant. The corresponding mean vertical wind speeds (adiabatic cooling rates) change from 32.2 cm s\(^{-1}\) (11.3 K h\(^{-1}\)) to 22.9 cm s\(^{-1}\) (8.1 K h\(^{-1}\)) and 50.2 cm s\(^{-1}\) (17.7 K h\(^{-1}\)). In another case, we keep the wave period fixed but just alter the time sequence of oscillations.

[48] Figure 6 displays the model response of occurrence frequency upon dynamical changes relative to the base case HOM shown in Figure 5. It is interesting to note that a simple change in the stochastic sequence of temperature waves already leads to changes of the geographical distribution of cloud occurrence relative to case HOM (top panel). The changes, however, are rather small (~1%) and mostly uncorrelated with regions of either low or high cloud occurrence.

[49] The changes in cloud occurrence are more significant (~3% in most regions) when the mean wave period is multiplied by factors of 0.75 (middle panel, faster oscillations and higher cooling rates) and 1.25 (bottom panel, slower oscillations and lower cooling rates), respectively, preserving the shape of the cooling rate distribution. The rather moderate changes in vertical air motions lead to enhanced (reduced) ice crystal number densities (see Table 1), with corresponding impacts on cloud lifetimes. Recall that \( n_i \) is a strong function of the local cooling rate (Figure 4). In some cases with high cloud occurrence, the dynamically-induced changes can reach up to ±4%, for instance around 55°N and 120°E, but the overall pattern of changes is irregular owing to the stochastic nature of the temperature perturbations.

[50] How does the frequency of occurrence change in the presence of heterogeneous ice nuclei? We now focus on the effects of aerosols on cirrus occurrence and discuss Figure 7. Compared to case HOM, the presence of 0.001 cm\(^{-3}\) heterogeneous IN (case MIX 0.001, top panel) increases the cirrus occurrence in most regions by about...
2%. The magnitude of these changes is comparable to case HOM 900, compare with Figure 6. The increase in the cirrus occurrence in case MIX 0.001 is caused by the prescribed low IN freezing threshold of 130%. The probability to reach this threshold value is higher than the probability to reach the homogeneous freezing thresholds. Thus, on average, clouds form more frequently and exist over longer periods in case MIX 0.001 than in HOM.

In the simulations MIX 0.001 and MIX 0.01, the early freezing IN do not significantly suppress homogeneous freezing owing to their relatively low number density. (This point will be elucidated further in section 3.2.) Still, the few IN alter the time evolution of RHI and thus have an impact on subsequent homogeneous freezing events. The depositional growth of the heterogeneously nucleated crystals slows the increase of RHI forced by cooling. This reduces the slope of RHI(t), hence the cooling rate, at the point of homogeneous nucleation, leading to fewer ice crystals compared to case HOM (negative Twomey effect for cirrus clouds, [Kärcher and Lohmann, 2003]). The few IN, however, do not influence the cloud lifetime via enhanced sedimentation losses.

Figure 7 demonstrates that the negative Twomey effect becomes important for IN concentrations higher than 0.01 cm$^{-3}$ in midlatitude cirrus. In the case MIX 0.01, the smaller increase of the cirrus occurrence compared to MIX 0.001 indicates that homogeneous freezing is more often “weakened” in this case. However, the median (mean) value of the frequency of occurrence (see Table 1) is still higher in MIX 0.01 compared to the base case HOM.

In the model case MIX 0.1 we already identify a significant number of freezing events where homogeneous freezing does not occur owing to preexisting IN despite of rapid wave cooling. While in MIX 0.001 and MIX 0.01 the IN have a positive effect on the calculated cirrus occurrence, MIX 0.1 shows a marked decrease of the occurrence...
Figure 7. Calculated changes of the frequency of cirrus cloud occurrence relative to case HOM caused by changes in freezing aerosol properties. Thick contours separate negative and positive changes. The case MIX 0.01 separates two regimes of the indirect aerosol effect. For lower IN concentrations (MIX 0.001), the occurrence frequency increases because cirrus formation conditions are more frequently met without significantly changing the overall cloud properties. For higher IN concentrations (MIX 0.1), the occurrence frequency decreases because homogeneous freezing becomes frequently suppressed and cloud formation is then limited by the available IN concentration leading to larger crystals and increased sedimentation losses. In case MIX-IN with $0.01$ cm$^{-3}$ almost perfect IN, the former effect by far outweighs the latter, drastically increasing cloud occurrence. Here sedimentation losses remain small because the available supersaturation is too small to grow the ice particles to large sizes. See text and Table 1 for more details.
frequency in most regions. In some regions this decrease can reach up to −4% compared to HOM. This effect is amplified by the concomitant reduction of mean ice crystal concentrations, leading to larger mean crystal sizes (see Table 1), and hence increasing sedimentation losses limiting the cirrus lifetimes. Together, this outweighs the effect of the low IN freezing threshold that by itself leads to an increase in cloud occurrence.

[54] We recall that IN concentrations exceeding 0.01 cm$^{-3}$ are not supported by the current upper tropospheric measurements, but may be present locally, e.g., in aircraft or convective plumes. On the basis of our results (further detailed in section 3.2), we anticipate that cirrus formation would be controlled by heterogeneous nucleation in conditions reported by DeMott et al. [2003b], where concentrations of ice-forming Saharan dust particles in a 2 km thick mid tropospheric layer exceeded 1 cm$^{-3}$.

[55] It is an important result of the present work that the Twomey effect can lead to both, increased or decreased cloud cover, depending on the abundance of IN. The IN concentration separating these two regimes is ~0.02 cm$^{-3}$, and must be seen in conjunction with the mean cooling rates (order 10 K h$^{-1}$) and mean temperatures (223 K) used in the simulations. The separation appears at higher IN concentrations in the case of lower temperatures and/or higher cooling rates. We recall that IN concentrations near or below 0.01 cm$^{-3}$ appear to characterize northern hemisphere background conditions.

[56] How does the frequency of cirrus occurrence change in the presence of nearly perfect IN? In the model cases MIX $n_i$ the prescribed nominal IN freezing threshold was 130%. In the simulation MIX-IN (bottom panel in Figure 7), a value of 105% was prescribed, together with an IN concentration of 0.01 cm$^{-3}$. The model response upon this reduction of the IN freezing threshold is very pronounced. We find an increase of the occurrence frequency relative to HOM which is much higher than the other calculated changes displayed in Figures 6 and 7. We explain this finding as follows.

[57] First, the temperature fluctuations are very often strong enough to frequently initiate heterogeneous freezing already if the synthetically cooled air approaches ice saturation. Second, as the very low median (mean) value of $n_i$ in the case MIX-IN (Table 1) reveals, the low freezing threshold leads to a particularly frequent suppression of homogeneous freezing, because in most of the cloud events, the cooling rates are not able to bridge the great gap in RHI between the heterogeneous freezing threshold of 105% and the homogeneous freezing thresholds above 145%. Thus cirrus formation is effectively limited by the few IN present in this case, as expected on the basis of theoretical considerations [Kärcher and Lohmann, 2003]. Third, as discussed later in Figure 10, the ice crystals formed on the near perfect IN stay smaller than in HOM owing to the small supersaturation available for growth. Thus the average sedimentation loss is actually reduced.

[58] This finding is supported by the comparison of the geographical pattern of the changes in occurrence frequency in case MIX-IN with the large-scale geographical distribution of RHI directly inferred from the ECMWF model (compare the bottom panel in Figure 7 with the top panel in Figure 1). As this comparison shows, the increase in the frequency of occurrence in case MIX-IN is directly linked to the regions with enhanced RHI predicted by the ECMWF model, e.g., the regions east of Scandinavia, north of 60°N, and over the North Atlantic around 60°W.

[59] How do these changes compare with global cirrus trends? Cirrus trends are difficult to quantify and depend on the type of observation (ground-based, satellite-based) and the location (over land, over ocean), and could be attributed to a plethora of natural and anthropogenic causes. For the period 1982–1991, mean global trends in cirrus occurrence frequency were 1.7% and 6.2% per decade over land and ocean, respectively [Boucher, 1999, and references therein], with regional decadal trends up to ~13% over some heavy air traffic regions. Monthly mean cirrus and cirrostratus fractions from the ISCCP D2 dataset evaluated in the volcanically unperturbed periods July 1983 to June 1991 and July 1993 to June 1996 show trends between 0.5–3% per decade over the United States of America (with a minimum in summer) and no significant trend over the year over western Europe [Minnis et al., 2004].

[60] Our results suggest that moderate changes in the mean cooling rates and the presence of few IN, especially when they are potent ice-forming agents, can lead to changes of cirrus occurrence and cirrus cloud coverage that are comparable in magnitude to observed decadal trends in global cirrus cover. This issue warrants further studies.

3.2. Cirrus Cloud Microphysical and Optical Properties

[61] The purpose of this section is to support the interpretation of Figures 5–7 by studying in more detail the changes in total ice crystal number density ($n_i$), mean radii ($r_i$), total ice water content (IWC), and optical extinction at 1 μm wavelength ($E$). Figures 8–10 depict the probability distributions of these quantities for the various scenarios discussed in section 3.1. In each figure, $n_i$ is shown in the top left, $r_i$ in the top right, $E$ in the bottom left, and IWC in the bottom right corner. Mean and median values of these distributions are summarized in Table 1.

[62] As outlined in section 2.1.3, the distributions of $n_i$ in cases HOM and MIX 0.01 are in reasonable agreement with field observations. They are shown in Figure 8 as black and red stair steps, respectively. Compared to HOM, the case MIX 0.01 shows an increase in number densities around 0.01 cm$^{-3}$ that is accompanied by a slight reduction at concentrations in the range 1–10 cm$^{-3}$. We reiterate from section 3.1 the causes of this indirect aerosol effect: (1) reduction of the number of homogeneously nucleated ice crystals because early frozen IN deposit H$_2$O vapor and thereby slow the cooling rates; (2) at sufficiently high IN concentrations frequent complete suppression of homogeneous freezing.

[63] While case MIX 0.001 (green) is somewhat less pronounced than MIX 0.01, the negative Twomey effect is very marked in case MIX 0.1 (purple), as we could expect from the discussion of occurrence frequencies (Figure 7). Case MIX 0.001 can still be regarded as consistent with available field measurements, but case MIX 0.1 is certainly not. Likewise, the pure synoptic case HOM-S (blue) is in striking disagreement with the observations, because it leads to total ice particle concentrations far lower than observed.

[64] The changes in $n_i$ are accompanied by changes in $r_i$ and IWC, although less pronounced. However, IWC is not a
good indicator of aerosol effects on cirrus at comparable


temperatures, as its relative changes appear to be smaller

than those of \( n_i \) and \( r_i \) (see Table 1). Our calculated

distributions of IWC in case HOM and all MIX \( n \) cases

are skewed toward low IWC values and compare well with

measured distributions (compare with Figure 7a in Ström et al. [2003]), both in terms of shape of the distributions and average values. Distributions of IWC are more sensitive to

the underlying temperature distributions, as IWC is a strong

function of \( T \). The distribution of IWC in case HOM-S is in

striking disagreement with the in situ observations.

The changes in median \( r_i \) (comparable to the effective mean

radius) from 13.8 \( \mu m \) (HOM) to 17.9 \( \mu m \) (MIX 0.1) are

significant and may decide on whether the net cloud radiative

forcing is positive or negative.

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**Figure 8.** Normalized distributions of total ice crystal number density \( n_i \) (top left), number mean ice crystal radius \( r_i \) (top right), optical extinction at 1 \( \mu m \) wavelength \( E \) (bottom left), and total ice water content IWC (bottom right) for the model experiments HOM-S (blue), HOM (black), MIX 0.001 (green), MIX 0.01 (red), and MIX 0.1 (purple). The median and mean values derived from these distributions are given in Table 1.

[55] The presence of IN modify optically relevant cloud properties? Our simulations emphasize that the median cirrus particle sizes peak in the radius range of 10–20 \( \mu m \), mainly resulting from the small-scale gravity wave forcing of cirrus formation. In this size range, depending on the shape of the ice crystals, the positive infrared and the negative solar cloud forcing are of similar magnitude [Zhang et al., 1999].

The changes in median \( r_i \) (comparable to the effective mean radius) from 13.8 \( \mu m \) (HOM) to 17.9 \( \mu m \) (MIX 0.1) are significant and may decide on whether the net cloud radiative forcing is positive or negative.
In the case HOM-S, the median crystal size is more than three times larger and would possibly lead to a net warming in most cases. The same result would be obtained in any other synoptic situation in the absence of mesoscale gravity waves that drive rapid cooling cycles.

The general shape of the distributions of $E$, especially the tail toward small extinctions (or optical depths) is consistent with lidar observations of optical depth (at 0.532 $\mu$m wavelength) taken during INCA [Immler and Schrems, 2002] and a subvisible cirrus lidar climatology taken over southern France [Goldfarb et al., 2001].

The changes of $E$ between cases HOM and MIX $n$ are more pronounced than those of $r_i$ and IWC and comparable to the changes in the total crystal concentration.

Inspection of Figure 8 reveals that the number of cases with low extinction values (i.e., the left wing of the distribution of $E$) increases when going from HOM to cases MIX $n$. As ice clouds with $E < 0.01$ km$^{-1}$ can be regarded as subvisible, this implies an increasing fraction $f_{svc}$ of subvisible cirrus (SVC), as listed in Table 1.

The value of $f_{svc}$ increases when going from MIX 0.001 to MIX 0.01 because the lifetime of SVCs becomes longer owing to less efficient sedimentation (more but smaller crystals form). The value of $f_{svc}$ decreases when going from MIX 0.01 to MIX 0.1 because the number of IN on which the ice crystals form continues to increase and relatively more events with $E$ values above the visibility threshold appear.

Figure 9. Same as Figure 8, but considering changes in small-scale cooling rates.
Which effects are caused by increases in the mean cooling rate? We now focus on changes in cooling rates and their effect on the distributions of cloud properties. For this purpose, Figure 9 shows the simulation results for cases HOM 900 (faster cooling) and HOM 1500 (slower cooling) in comparison with the baseline case HOM, see also Table 1. When the mean cooling rates are increased, the mean \( r_i \) decreases and total \( n_i \) increases. The total IWC and \( E \) also increase, indicating that the enhanced concentration of crystals overcompensates the effect of smaller average size. Upon increasing the mean cooling rate, a third peak at crystal concentrations around 20–30 cm\(^{-3}\) begins to develop, with concomitant decreases at lower concentrations. A mere change in local vertical wind speeds may therefore considerably alter the distribution of \( n_i \), while changes in the distributions of \( r_i \) and IWC are more subtle. The increase of \( f_{svc} \) when going from HOM to HOM 1500 is expected, as fewer but larger crystals form on average. An interesting feature is the increase of \( f_{svc} \) when going from HOM to HOM 900 although \( n_i \) increases. Freezing events extend over a time which is inversely proportional to the cooling rate [Kärcher and Lohmann, 2002], typically of the order of seconds to minutes. The high frequency oscillations of \( T \) and hence RHI in scenario HOM 900 contain an increased number of cases where freezing is not fully completed during the fastest wave cycles. This leads to fewer nucleated ice particles compared to cases with longer period oscillations.

Figure 10. Same as Figure 8, but considering changes in IN freezing relative humidities.
and hence more subvisible cases. This is another example of the highly nonlinear response of the freezing aerosol system to subtle changes in dynamical conditions driving the supersaturation.

[75] Which effects are caused by lowering the IN freezing relative humidities? Finally, we study in more detail changes in IN properties and their effect on the distributions of cloud properties. Figure 10 shows the simulation results for case MIX-IN (low freezing threshold) in comparison with the baseline cases HOM and MIX 0.01, see also Table 1. (Note that we use the same concentration of IN in both MIX cases.) The figure underlines the strong effect IN have when they freeze slightly above ice saturation, even if present in relatively low concentrations. All distribution functions change considerably in case MIX-IN compared to HOM and MIX 0.01. The fact that IN predominantly freeze is best illustrated by the distribution of \( n_i \), which reveals a marked maximum at the prescribed total IN concentration of 0.01 cm\(^{-3}\) formed at the expense of homogeneous freezing events.

[76] Another obvious modification caused by the almost perfect IN is visible in the distribution of \( E \), where a broad secondary maximum develops at low extinction values. The mean extinction is significantly lower than in cases HOM and MIX 0.01, and the occurrence frequency of SVC increases markedly, as inferred from Table 1.

[77] So, why does \( f_{\text{sec}} \) increase so strongly from 25% to 40% when going from MIX 0.01 to MIX-IN? The arguments are similar to those used to interpret Figure 7 (bottom panel). The final ice crystal sizes tend to be smaller in MIX-IN because far less supersaturation (5%) is available to grow the nascent ice particles after they form. This is supported by the peak in the distribution of \( r_i \) that develops near 1 \( \mu \text{m} \) which is not present in MIX 0.01. Also, compared to MIX 0.01, homogeneous freezing is more frequently weakened or suppressed, as inferred from the diminishing peak at 1–10 cm\(^{-3}\) in the distribution of \( n_i \).

4. Summary and Conclusions

[78] The key findings of this study are summarized as follows.

1. For most cirrus clouds to form, air masses must cool to sufficiently low temperatures so that synoptic-scale regions become supersaturated with respect to ice. These synoptic cold pools define the overall thermodynamic conditions in which cirrus formation takes place. However, cloud microphysical properties are dictated by mesoscale variability in cooling rates present at the point of freezing.

2. Changes in upper tropospheric cooling rates (vertical winds) or changes in freezing properties of aerosols can lead to changes of cirrus cloud occurrence that are comparable in magnitude to observed decadal trends in global cirrus cover.

3. Aerosol particles that nucleate cirrus crystals at 130% relative humidity over ice present in concentrations probably typical for northern midlatitude background conditions (<0.01–0.03 cm\(^{-3}\)) significantly modify certain cirrus properties but do not control cirrus formation. The impact of such ice nuclei (IN) becomes particularly strong and possibly dominates cirrus formation if they freeze near ice saturation, at low temperatures, and/or in weak updrafts.

[80] 4. Heterogeneous IN may cause an indirect aerosol effect on cirrus by reducing the number of ice crystals formed by homogeneous freezing alone. This negative Twomey effect may increase or decrease the frequency of cirrus cloud occurrence, depending on the concentration of IN. Lowering the ice nucleation threshold to ice saturation may lead to particularly strong increases in cloud occurrence.

[81] 5. Heterogeneous IN may lead to increases of cirrus crystal effective radii sufficiently large to modify the net cloud radiative forcing by several W m\(^{-2}\) or perhaps to a change of its sign. The cirrus optical extinction is reduced by IN. The frequency of thin and subvisible cirrus (SVC) cloud occurrence increases even when relatively few (0.001 cm\(^{-3}\)) IN are present, but decreases when their concentration is large. The pronounced tendency to increase thin cloud occurrence becomes larger the earlier the IN freeze.

[82] We conclude this work by adding a few remarks to each point enumerated above.

[83] Point 1 suggests that global models, including weather forecast, chemistry transport, and climate models, predict cirrus coverage correctly to first order using a thermodynamic approach (saturation adjustment). One such study linked the occurrence of SVCs to ECMWF high cloud cover [Bregman et al., 2002]. Our results indicate that the so computed cloud cover tends to be smaller (by up to 5%) than the real value owing to neglected kinetic effects of evaporating ice crystals, and further enhancements may be caused by IN if present. It is of paramount importance to include small-scale temperature fluctuations caused by high frequency gravity waves currently unresolved in global models, to correctly predict cloud physical properties when combined with suitable cirrus parameterizations.

[84] Point 2 emphasizes the role of cirrus changes in a future climate. Climate change may both alter atmospheric dynamics and atmospheric chemistry. The dynamical effects on cirrus are directly tied to changes in atmospheric gravity wave activity, and alterations of temperature and water vapor fields. The effects of IN on cirrus could be brought about by changes in the ice-forming ability of the particles (e.g., via chemical surface ageing in an atmosphere with changing oxidative capacity) as well as by changes in the geographical distribution of IN (e.g., via changes in circulation patterns and convective activity).

[85] Point 3 implies that it will often be difficult to isolate the impact of IN on cirrus evolution in observations, as they only slightly alter overall cloud properties such as total crystal number and ice water content. Likely, they more significantly modify the ice crystal size distributions by producing large crystals already in the formation layer [Kärcher, 2003]. However, large crystals may also be produced by other physical mechanisms. At any rate, more in situ case studies including direct measurements of the chemical composition and concentration of IN are needed to confirm the mechanisms worked out in this paper.

[86] Our results do not rule out cases in which cirrus properties are controlled by efficient heterogeneous IN. This may occur in dust plumes, convective plumes, or aircraft plumes with IN concentrations well above 0.01 cm\(^{-3}\) under similar dynamic conditions. Here, the major experimental challenge is to determine the freezing thresholds of all particles nucleating ice, and a major modeling challenge...
is to predict their global distribution and seasonal cycle. Measurements confirm that mineral dust and metallic particles appear to be common heterogeneous IN [Talbot et al., 1998; Sassen et al., 2003; DeMott et al., 2003b; Cziczo et al., 2004], while the importance of soot particles is less clear. Accurate estimates of emission source regions and strengths are critical to predicting global IN concentrations.

Page 177: Point 4 states that the Twomey effect in cirrus clouds leads to a reduction of cloud particle number, confirming previous single parcel simulations carried out at constant cooling rates [DeMott et al., 1997; Kärcher and Lohmann, 2003] with the help of a near-global, statistical analysis of cirrus clouds over a period of 3 months. We have also shown quantitatively how the mean ice crystal size (hence cloud radiative forcing) depends on the freezing mechanisms.

Page 178: Understanding and predicting cirrus clouds remains to be a highly complex issue. Owing to nonlinear effects in the freezing aerosol system driven by high frequency gravity waves, we find a threshold IN concentration around 0.01 cm\(^{-3}\) over which cirrus occurrence, optical extinction, and the fraction of clouds staying subvisible change nonmonotonously. We recall that this threshold concentration depends on the cloud formation temperature and cooling rate, and on the IN freezing relative humidity. Very potent IN deserve special attention as they are capable of introducing strong changes of cloud properties at low concentrations.

Page 179: Point 5 refers to results showing that slow synoptic cooling leads to large ice crystals, implying that such clouds likely warm the atmosphere. The radiative forcing of midlatitude cirrus formed in slow updraft conditions (5 cm s\(^{-1}\)) has been studied by Jensen et al. [2001]. Their results showed that the addition of IN with freezing thresholds of 133% increased the net radiative forcing (warming) by almost a factor of 2 when increasing the IN concentration from 0.001 cm\(^{-3}\) to 1 cm\(^{-3}\). It would be interesting to repeat these simulations including a more realistic spectrum of cooling rates, and with lower freezing relative humidities.

Page 180: Our results confirm that SVCs can be particularly susceptible to heterogeneous IN [Kärcher, 2002], but demonstrate that SVC coverage does not necessarily increase in proportion to the IN concentration. This adds further complexity to accurately predict cirrus coverage in the tropical tropopause layer [Kärcher, 2004], a region in which cirrus cloud modification through IN might impact the stratospheric moisture budget and hence climate.

Page 181: Satellite observations [Wang et al., 1996; L. Thomason, personal communication, 2004] reveal a seasonal cycle of SVC occurrence in the northern midlatitudes with maxima from March to August, where the upper troposphere is frequently perturbed by vertical transport from the boundary layer, possibly introducing IN into cirrus forming regions. If SVC do form on IN, this may point to a possible anthropogenic influence of such clouds. This idea is supported by the fact that SVC occurrence is lower in southern midlatitudes, a region characterized by comparatively little anthropogenic influence. However, less dynamic forcing may be expected in the southern hemisphere, which may also cause a larger fraction of thin clouds.

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