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2008 Environ. Res. Lett. 3 045022

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Cirrus cloud formation and ice supersaturated regions in a global climate model

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Received 6 July 2008
Accepted for publication 28 November 2008
Published 22 December 2008
Online at stacks.iop.org/ERL/3/045022

Abstract
At temperatures below 238 K, cirrus clouds can form by homogeneous and heterogeneous ice nucleation mechanisms. ECHAM5 contains a two-moment cloud microphysics scheme and permits cirrus formation by homogeneous freezing of solution droplets and heterogeneous freezing on immersed dust nuclei. On changing the mass accommodation coefficient, $\alpha$, of water vapor on ice crystals from 0.5 in the standard ECHAM5 simulation to 0.006 as suggested by previous laboratory experiments, the number of ice crystals increases by a factor of 14, as a result of the delayed relaxation of supersaturation. At the same time, the ice water path increases by only 29\% in the global annual mean, indicating that the ice crystals are much smaller in the case of low $\alpha$. As a consequence, the short wave and long wave cloud forcing at the top of the atmosphere increase by 15 and 18 W m\(^{-2}\), respectively. Assuming heterogeneous freezing caused by immersed dust particles instead of homogeneous freezing, the effect is much weaker, decreasing the global annual mean short wave and long wave cloud forcing by 2.7 and 4.7 W m\(^{-2}\). Overall, these results provide little support, if any, for kinetic growth limitation of ice particles (i.e. a very low $\alpha$).

Keywords: cirrus clouds, ice supersaturated regions, climate modeling

1. Introduction

Cirrus clouds can form by homogeneous and heterogeneous ice nucleation mechanisms at temperatures below 238 K. They cover on average 30\% of the Earth surface (Wylie and Menzel 1999) and thus are important modulators of the radiation budget. Thin cirrus are semi-transparent in the solar radiation spectrum, allowing the majority of the solar radiation to be transmitted to the surface. Because of their cold temperatures, they emit the absorbed infrared radiation at much lower temperatures than the Earth’s surface and thus cause a warming of the Earth–atmosphere system (Chen \textit{et al} 2000). For thick cirrus, the reflected short wave radiation balances the emitted long wave radiation.

While homogeneous freezing of supercooled aqueous phase aerosol particles is rather well understood (Koop \textit{et al} 2000), knowledge about heterogeneous ice nucleation is still in its infancy. A change in the number of ice crystals in cirrus clouds could exert a cloud albedo effect in the same way that this effect acts for water clouds. The cloud albedo effect refers to the change in the radiative forcing at the top-of-the-atmosphere (TOA) caused by an enhancement in cloud albedo from anthropogenic aerosols that leads to more and smaller cloud droplets for a given cloud water content (Twomey 1977). In addition, a change in the ice water content could exert a radiative effect in the infrared. The magnitude of these effects in the global mean has not yet been established.
In recent years, physically based parametrization schemes of cirrus formation for use in global models have been developed (Kärcher and Lohmann 2002, Liu and Penner 2005, Kärcher et al 2006, Liu et al 2007, Tompkins et al 2007, Penner et al 2008). They led to significant progress in understanding underlying mechanisms of aerosol-induced cloud modifications. A general circulation model (GCM) study concluded that a cloud albedo effect based solely on ubiquitous homogeneous freezing is small globally (Lohmann and Kärcher 2002). This effect is expected to be also true in the presence of heterogeneous IN that cause solution droplets to freeze at relative humidities with respect to ice (RH$_i$) close to homogeneous values (above 130–140%) (Kärcher and Lohmann 2003). Efficient heterogeneous IN, however, would be expected to reduce RH$_i$, so that the climate effect may be larger (Liu and Penner 2005). In situ measurements reveal that organic-containing aerosols are less abundant than sulfate aerosols in ice cloud particles, suggesting that organics do not freeze preferentially (Cziczo et al 2004). A model study explains this finding by the disparate water uptake of organic aerosols, and suggests that organics are unlikely to significantly modify cirrus formation unless they are present in very high concentrations (compared with sulfate-rich particles) at low temperatures (Kärcher and Koop 2005).

Recent high-altitude aircraft measurements indicated the presence of rather large, thin hexagonal plate ice crystals near the tropical tropopause in very low concentrations. These large ice crystals are suggested to have grown on particles that acted as ice nuclei at low supersaturations with respect to ice (Jensen et al 2008). Very recently, Zobrist et al (2008) based on laboratory experiments showed that mixed organic/inorganic aerosols may form glasses at temperatures prevalent in the upper troposphere, and that this may greatly reduce their uptake of water and hence their homogeneous ice formation ability.

Besides nucleation effects, ice growth impedances were recently found to lead potentially to high supersaturations within cirrus clouds (Peter et al 2006, 2008). The role of physico-chemical processes affecting the accommodation coefficient of water vapor on ice and of water vapor on aerosols is presently unclear. These effects could be related to unknown intrinsic properties of the ice substance at the extreme temperatures at high tropical cirrus levels, or due to natural or anthropogenic substances on the ice surface (Wood et al 2001). Magee et al (2006) suggested that assuming an accommodation coefficient $\alpha$ between 0.0045 and 0.0075 at low supersaturations ($S_i < 1.2$) was necessary to match their laboratory data of ice crystal growth at temperatures between $-40$ and $-60{^\circ}C$. The mechanisms leading to this surprising low values of $\alpha$ remain unclear.

The properties and global distributions of ice supersaturated regions (ISSRs) were discovered during recent years (Gierens et al 1999, Spichtinger et al 2003, Gettelman et al 2006). ISSRs are potential formation regions of cirrus and persistent contrails. ISSRs are rather ubiquitous (Spichtinger et al 2003) in the upper tropical troposphere, with frequencies of occurrence even exceeding 50% of the time at 150 hPa. Immel et al (2008) evaluated lidar data over Northern Germany and concluded that 50% of the air at 11–12 km was supersaturated with respect to ice. For a physical treatment of the formation of cirrus clouds in a GCM, ISSRs must be represented correctly in terms of geographical frequency of occurrence as well as in terms of probability distributions of ice supersaturation.

In this paper, we discuss the impact of heterogeneous freezing versus homogeneous freezing and of variations in the deposition coefficient on cirrus cloud properties and ice supersaturation in the ECHAM5 GCM.

2. Model description

The ECHAM5 GCM (Roeckner et al 2003) used in this study includes a two-moment aerosol scheme ECHAM5-HAM that predicts the aerosol mixing state in addition to the aerosol mass and number concentrations (Stier et al 2005). The size-distribution is represented by a superposition of log-normal modes including the major global aerosol compounds sulfate, black carbon, organic carbon, sea salt and mineral dust.

The stratiform cloud scheme consists of prognostic equations for the water phases (vapor, liquid, solid), bulk cloud microphysics (Lohmann and Roeckner 1996), and an empirical cloud cover scheme (Sundqvist et al 1989). The microphysics scheme includes phase changes between the water components and precipitation processes (autoconversion, accretion, aggregation), evaporation of rain, melting of snow and sedimentation of cloud ice (Lohmann et al 2007). We abandoned the saturation adjustment scheme for cirrus clouds. Thus instead of depositing all water vapor above saturation with respect to ice onto the existing ice crystals, we solve the depositional growth equation (Lohmann and Kärcher 2002). This enables to permit supersaturations with respect to ice. Cirrus clouds are then formed by homogeneous freezing once the homogeneous freezing threshold (Koop et al 2000) is exceeded. We limit ice supersaturation to water saturation. Thus, supersaturations in excess of water saturation occur only because of numerical noise and because other processes that increase RH$_i$ can occur after the calculation of the depositional growth. The calculation of the fractional cloud cover including the threshold relative humidity limits, above which a cloud cloud form, has not changed. This implies that once a cirrus forms, the entirely grid box is assumed to be cloudy.

The cloud microphysics scheme also includes prognostic equations for the number concentrations of cloud droplets and ice crystals in stratiform clouds and has been coupled to the aerosol scheme ECHAM5-HAM (Lohmann et al 2007). In the standard simulation that assumes cirrus cloud formation solely by homogeneous freezing (E5-hom, see table 1), an accommodation coefficient $\alpha$ for depositional growth of water vapor onto ice crystals of 0.5 is employed (Kärcher and Lohmann 2002). The number of ice particles forming via homogeneous freezing depends mainly on the vertical velocity, which is given as the sum of the grid-box mean vertical velocity and a subgrid-scale contribution as obtained from the turbulent kinetic energy (Lohmann and Kärcher 2002). We neglect aerosol size effects for cirrus cloud formation as they were
shown to be small (Kärcher and Lohmann 2002, Lohmann et al. 2003).

The version of the model used here includes some new features. Ice crystals in mixed-phase clouds with temperatures between 0 and $-35^\circ C$ are now regarded as plates as this is a more realistic shape for ice crystals in the mixed-phase cloud regime that rapidly grow to larger sizes due to the Bergeron–Findeisen process than assuming spherical crystals (Pruppacher and Klett 1997). Small ice crystals, on the other hand, are more likely spherical (Korolev and Isaac 2003). Thus, for cirrus crystals formed at temperatures below $-35^\circ C$ mainly via homogeneous freezing assuming spheres seems appropriate.

Assuming ice crystals to be plates at temperature above $-35^\circ C$ also changes the calculation of the ice crystal effective radius $r_i$ which is now given as described in Pruppacher and Klett (1997) for a plate of type P1a (in $\mu m$):

$$
 r_i = 0.5 \times 10^4 \left( \frac{IWC}{0.0376N_i} \right)^{0.302}
$$

(1)

where $N_i$ is the ice crystal number concentration in $m^{-3}$ and IWC is the ice water content in $g m^{-3}$. At temperatures below $-35^\circ C$ we revert to the formula used in ECHAM4 (Lohmann 2002):

$$
 r_i = 1.61r^3 + 3.56 \times 10^{-6}\rho_i
$$

(2)

where the mean volume radius $r_i$ is obtained from the cloud ice mixing ratio in the cloudy part of the grid box $q_i$ in $kg kg^{-1}$ and the ice crystal number concentration $N_i$:

$$
 r_i = \left( \frac{3q_i\rho_a}{4\pi\rho_iN_i} \right)^{1/3}
$$

(3)

where $\rho_a$ = air density and $\rho_i$ = ice density, here assumed to be 925 $kg m^{-3}$. This change was necessary because the empirical formula that has been used in previous versions of ECHAM5

$$
 r_i = 83.8 \times IWC^{0.216}
$$

(4)

would not have been able to yield a cloud albedo effect for cirrus clouds. A negative cloud albedo effect can arise for cirrus clouds formed by heterogeneous freezing with fewer but larger crystals and thus a larger effective radius and smaller optical depth, which reflect less solar radiation back to space than a cirrus cloud formed by homogeneous freezing (Kärcher et al. 2006). By using equations (1)–(3) this effect can be considered.

The fall velocity of ice crystals has been changed to one appropriate for monodisperse crystals, that vary their shapes with increasing size following Spichtinger and Gierens (2008a). Assuming monodisperse ice crystals is consistent with the other formulations for microphysical processes in the model and allows us to use the same fall velocity for ice crystal mass and number ($v_{mf} = v_n$).

Spichtinger and Gierens (2008a) assume that small crystals have a droxtal (=quasi-spherical) shape (aspect ratio $\approx 1$), the majority of the particles has columnar shape and larger particles are hexagonal cylinders with aspect ratio larger than 1. According to Heymsfield and Iaquinta (2000), the terminal velocity for columnar shaped ice crystals can be described as a function of the mass as

$$
 v_{mf} = v_n = \gamma \left( \frac{m}{m_0} \right)^\beta
$$

(5)

with $m$ in kg and $m_0 = 1$ kg. For a realistic treatment of the droxtal/columnar shape of ice crystals over a broad range of sizes the following coefficients are used (Spichtinger and Gierens 2008a):

<table>
<thead>
<tr>
<th>$m$ $&lt; m_1$</th>
<th>$m_1$ $\leq m &lt; m_2$</th>
<th>$m_2$ $\leq m &lt; m_3$</th>
<th>$m &gt; m_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma$ (m s$^{-1}$)</td>
<td>735.4</td>
<td>63292.4</td>
<td>329.8</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.42</td>
<td>0.57</td>
<td>0.31</td>
</tr>
</tbody>
</table>

with the transitions at $m_1 = 2.15 \times 10^{-13}$ kg, $m_2 = 2.17 \times 10^{-9}$ kg, $m_3 = 4.26 \times 10^{-9}$ kg.

2.1. Set-up of the simulations

The ECHAM5 simulations have been carried out in T42 horizontal resolution ($2.8125^\circ \times 2.8125^\circ$) and 19 vertical levels with the model top at 10 hPa and a timestep of 30 min. All simulations use climatological sea surface temperature and sea-ice extent. They were simulated for five years after an initial spin-up of three months using aerosol emissions for the year 2000 (Dentener et al. 2006).

The reference simulation E5-hom is conducted such that the global annual mean TOA radiation budget is balanced to within 1 W m$^{-2}$ and that the values of the short wave and long wave cloud forcings are within the uncertainty of the radiative flux measurements of $\pm 5$ W m$^{-2}$ as reported by Kiehl and Trenberth (1997). This required changing the critical radius above which aerosols could possible act as cloud condensation nuclei in stratiform clouds from 35 to 30 nm. The number

<table>
<thead>
<tr>
<th>Simulation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>E5-hom</td>
<td>Simulation with ECHAM5-HAM coupled to the two-moment cloud microphysics scheme for stratiform clouds (Lohmann et al. 2007) assuming that all cirrus clouds form by homogeneous freezing with an accommodation coefficient of water on ice $\alpha$ of 0.5.</td>
</tr>
<tr>
<td>E5-het</td>
<td>Same as E5-hom, but assuming that particles that contain immersed dust particles are available as immersion freezing nuclei initiating freezing at 130% RH; no homogeneous freezing.</td>
</tr>
<tr>
<td>E5-homhet</td>
<td>Same as E5-hom, but assuming immersion freezing of dust in those grid boxes where the immersion dust nuclei concentration exceeds 11 $l^{-1}$ and homogeneous freezing elsewhere.</td>
</tr>
<tr>
<td>E5-homhet10</td>
<td>Same as E5-homhet, but limiting immersion freezing of dust to immersion dust nuclei concentration $&gt;101^{-1}$.</td>
</tr>
<tr>
<td>E5-alpha</td>
<td>Same as E5-hom, but decreasing $\alpha$ from 0.5 to 0.006.</td>
</tr>
</tbody>
</table>

Table 1. Sensitivity simulations.
Figure 1. Annual zonal mean latitude versus pressure cross sections of the number concentration of solution droplets that limits the ice crystal number concentration formed by homogeneous freezing in simulation E5-hom, the number concentration of immersed dust nuclei limiting heterogeneous freezing in simulation E5-het, the combined number of solution droplets/IN in simulations E5-homhet and E5-homhet10 (top row) and the ice crystal number concentrations within the cloudy part of the grid box in simulations E5-hom, E5-het, E5-homhet and E5-alpha (bottom row).

concentration of solution droplets, which is the upper bound on the number of ice crystals that can be formed by homogeneous freezing is shown in figure 1. It exceeds 100 particles cm\(^{-3}\) in the cirrus level.

In the sensitivity experiment E5-het (table 1), homogeneous freezing of super-cooled soluble/mixed aerosol particles is suppressed, and only heterogeneous freezing takes place. In order to calculate the heterogeneous freezing rate we apply a shift in the water activity to the homogeneous nucleation rate following Kärcher and Lohmann (2003). This means we evaluate the heterogeneous freezing rate at 130\% RH\(_{\text{f}}\). The number of ice crystals that results from the freezing rate is then limited by the number of immersed dust particles. Using a simple threshold above which heterogeneous freezing commences seems justified based on cloud resolving simulations that investigated the impact of varying freezing thresholds (Spichtinger and Gierens 2008b). As there is currently no consensus on the freezing ability of black carbon, (Mohler et al 2005, Dymarska et al 2006, Kärcher et al 2007), we limit the number of heterogeneous ice nuclei to the number of immersed dust particles in the accumulation and coarse mode as described by Hoose et al (2008a). As shown in figure 1, its number concentration is at most 2 cm\(^{-3}\) in the cirrus level.

In simulation E5-homhet, we allow both homogeneous and heterogeneous freezing in a simplified way following Abbatt et al (2006). Here heterogeneous freezing occurs in those grid boxes where the immersion dust nuclei concentration exceeds 1 l\(^{-1}\) and homogeneous freezing occurs elsewhere. This simple treatment seems justified as homogeneous and heterogeneous freezing may seldom occur simultaneously (Spichtinger and Gierens 2008b). The choice of 1 l\(^{-1}\) is motivated by observations of (Richardson et al 2007) concluding that typical IN concentrations are between 1 and 10 l\(^{-1}\) in the free troposphere over the United States, which provides an upper bound. Barahona and Nenes (2008) show that an IN concentration greater >0.1 l\(^{-1}\) is necessary to see an effect on homogeneous freezing. As we are not able to allow heterogeneous and homogeneous freezing in the same grid box at this point, we needed an IN concentration that is realistic, but yet large enough, that the available supersaturation can be removed within one model timestep. Thus, limiting heterogeneous freezing to grid boxes where more than 1 l\(^{-1}\) is available seems a reasonable choice. We investigated the impact of this choice by adding a sensitivity study in which we increased this threshold to 10 l\(^{-1}\) (simulation E5-homhet10).

Figure 1 shows the number concentrations of solution droplets respective IN that were potentially available for homogeneous or heterogeneous freezing depending on the immersed dust nuclei concentration as described above. When the 10 l\(^{-1}\) threshold is used, fewer cirrus clouds experience heterogeneous freezing causing the particle numbers to be higher. The resulting ice crystal number concentrations in E5-homhet almost mimic those in simulation E5-het. In both simulations ice clouds are restricted to lower altitudes, i.e. warmer temperatures than in simulation E5-hom.

The accommodation coefficient of water vapor on ice crystals \(\alpha\) is used to calculate the initial ice crystal number concentration upon nucleation and enters the calculation of depositional growth. In simulation E5-alpha \(\alpha\) has been decreased from 0.5 to 0.006 (cf table 1). This was suggested
to be the most likely value from a laboratory experiment at −50 °C (Magee et al. 2006).

3. Model evaluation

The validation of the coupled aerosol–cloud microphysics scheme in stratiform clouds has been described in Lohmann et al. (2007). Here we focus on the validation of ice supersaturated regions (ISSRs) and cirrus clouds.

Ice crystal number concentrations have been obtained from field experiments. Here we compare with in situ data taken during the INCA campaign that compared cirrus in the Northern Hemisphere (NH) with the Southern Hemisphere (SH) at equivalent latitudes and seasons (Gayet et al. 2004). The INCA measurements were carefully quality checked. The median (and the 25 and 75 percentile) ice crystal number concentrations were 2.2 (0.8–4.7) cm⁻³ for the NH and 1.4 (0.6–3.0) cm⁻³ for the SH. Simulated values were averaged over the same geographical area and altitude range, i.e. over 50°N–60°N, 10°W–5°E, 200–400 hPa in the NH and 60°S–50°S, 85°W–75°W, 200–400 hPa in the SH.

Modeled values in simulation E5-hom are 1.7 cm⁻³ in the NH versus 0.8 cm⁻³ in the SH, 1.0 versus 0.3 cm⁻³ for simulation E5-het, 1.1 versus 0.3 cm⁻³ for simulation E5-homhet, 1.0 versus 0.5 cm⁻³ for simulation E5-homhet10 and 25.6 versus 20.4 cm⁻³ for simulation E5-alpha. Ice crystal number concentrations in E5-hom fall within the 25–75 percentile values in both hemispheres. In the simulations employing heterogeneous freezing, ice crystal number concentrations barely fall within the 25 percentile in the NH, but are below the 25 percentile value in the SH. On the contrary, simulation E5-alpha overestimates the ice crystal number concentration by at least a factor of 10 in both hemispheres (cf also figure 1). Overall, the simulated ice crystal number concentrations are closest to the observed ones in E5-hom suggesting that homogeneous freezing with a high deposition coefficient is the most plausible mechanism for cirrus formation.

An overview of the global mean cloud properties is given in table 2. The reference simulation E5-hom is very similar to the previously described version ECHAM5-RH (Lohmann et al. 2007) and compares reasonably well with observations. We compare the model with four years of TOVS satellite data. TOVS ice water path data are available for the subgroup of cirrus with visible optical depth (τ) between 0.7 and 3.8, hereafter called ‘semi-transparent cirrus’ (Stubenrauch et al. 2004). The ice water path of semi-transparent cirrus (IWPsemi) can exceed the total ice water path (IWPtot) because IWPsemi is sampled only over cloudy regions whereas IWPtot is an average over cloudy and cloud-free regions. All model simulations underestimate the ice water path both in semi-transparent cirrus and the total ice water path. IWPtot has been much higher in a previous version of ECHAM5 (Lohmann et al. 2007), where the effective ice crystal size just depended on the ice water mass (equation (4)). In that version the ice crystals were much larger so that a similar optical depth and thus long wave cloud forcing could be obtained for a much larger IWPtot. However, as shown in table 2, the effective ice crystal size for semi-transparent cirrus is already overestimated in all simulations with α = 0.5, thus having even larger crystals as in the previous ECHAM5 version does not seem to be realistic either. The simultaneous underestimation of IWPsemi and overestimation of Rssemi in the simulations with α = 0.5 suggests that the semi-transparent cirrus are, on average, less optically thick than suggested by the TOVS data. One has also to keep in mind though that current IWP retrievals are still very uncertain (Buehler et al. 2007).

The most striking difference between E5-alpha and E5-hom is the factor 14 higher number concentration in ice crystals in E5-alpha. This results from the slower depositional growth that cannot deplete the supersaturation effectively. In fact, the total water vapor mass (WVM) has increased by 3 kg m⁻² in the global mean. As a result of the slower
Figure 2. Annual zonal means of ice water path (IWPsemi) of semi-transparent cirrus (visible optical depth ($\tau$) between 0.7 and 3.8), their effective radii, vertically integrated ice crystal number concentration, total cloud cover and short wave and long wave cloud forcing from the different model simulations described in table 1 and from observations described in table 2. Dotted black lines refer to TOVS data for IWPsemi and effective radius (Stubenrauch et al 2004), to ISCCP for total cloud cover and to ERBE for the short wave and long wave cloud forcing. The dashed line refers to surface observations for total cloud cover and to TOVS data for the long wave cloud forcing (Susskind et al 1997, Scott et al 1999). Cloud cover from MODIS is included as the dotted–dashed line.

depletion of supersaturation, more ice crystals are nucleated. These ice crystals share a similar amount of condensing water, hence they remain smaller and have smaller gravitational settling velocities, thus increasing the total amount of cloud ice that remains within the atmosphere (IWPtot in table 2). In turn, this reduces the global mean precipitation rate ($P_{tm}$) by 0.42 mm d$^{-1}$ in E5-alpha as compared to E5-hom. The more numerous and much smaller ice crystal scatter more radiation back to space, thus enhancing the short wave cloud forcing by 14.7 to $-68.2$ W m$^{-2}$. At the same time, more long wave radiation is trapped in the Earth–atmosphere system, increasing the long wave cloud forcing by 18.2–47.3 W m$^{-2}$. Changes in liquid water clouds triggered by these changes in cirrus clouds are much smaller and thus not shown.

While the increase of IWPtot in E5-alpha as compared to E5-hom is to be expected as a consequence of the less efficient dehydration by the smaller ice particles, the reason for the reduction in IWPsemi is less obvious. The reason is that cirrus in E5-alpha do not only reveal higher ice number densities, but also higher visible optical depth $\tau$. Therefore, many more clouds no longer satisfy the criterion $0.7 < \tau < 3.8$ for the semi-transparent cirrus subclass.

The differences between E5-hom and E5-het are comparatively smaller. Here the ice crystal number concentration decreases by 50%, causing the ice crystals to be larger and to sediment more rapidly, thus increasing the global mean precipitation. This reduces the short wave cloud forcing by 2.7 W m$^{-2}$, i.e. exerts a negative cloud albedo effect as discussed above. The reduced cloud cover allows more radiation to be emitted to space, thus reducing the long wave cloud forcing by 4.7 W m$^{-2}$. The changes are a bit larger than the changes between homogeneous and heterogeneous freezing obtained with the previous version of ECHAM, the ECHAM4 GCM (Lohmann et al 2004). Given that the changes in ice water path and ice crystal number concentration between homogeneous and heterogeneous freezing are comparable in ECHAM4 and ECHAM5, the larger changes in the cloud radiative forcing are due to the different cloud optical properties described above. Simulation E5-homhet and E5-homhet10 fall in between E5-hom and E5-het, being closer to simulation E5-het in most aspects.

Annual zonal means of the ice water path of semi-transparent cirrus (IWPsemi), their effective radii, the vertically integrated ice crystal number concentration, total cloud cover, short wave and long wave cloud forcing from simulations E5-hom, E5-het, E5-homhet and E5-alpha as compared to observations are shown in figure 2. Recall that both IWPsemi and the effective ice crystal radii are conditionally sampled over cloudy events only. The TOVS data are less reliable polewards of 60° because of poor data coverage.
Figure 3. Frequency of occurrence of RH, in the Northern Hemisphere mid-latitudes (30°N–60°N) (left panel) and in the tropics (30°S–30°N) (right panel) in two different levels from MOZAIC aircraft data, MLS satellite data and from one year of instantaneous data from the simulations E5-hom, E5-het, E5-homhet, E5-homhet10 and E5-alpha. All data are normalized to unity at RH = 100%.

While TOVS suggests smaller IWP\textsuperscript{semi} and crystal size in mid-latitudes than in the tropics, IWP\textsuperscript{semi} and crystal size hardly vary with latitude in the simulations. The crystal size in semi-transparent cirrus in simulations E5-hom, E5-het and E5-homhet approaches the observed one in the tropics. Thus, our assumption of crystal shape seems to be most appropriate in the tropics. IWP\textsuperscript{semi} is underestimated by a factor of two in simulations E5-hom, E5-het and E5-homhet. This might partly be an artifact of our coarse vertical resolution and the lack of the fractional cloud cover in the vertical, which limits optical thin clouds. Simulation E5-alpha underestimates IWP\textsuperscript{semi} and crystal size as compared to TOVS between 60°S and 60°N the most and thus is the least realistic simulation.

The total cloud cover, short wave and long wave cloud forcing of E5-hom, E5-het and E5-homhet agree well with observations, except that the short wave cloud forcing is too negative in the subtropics. This suggests that the marine stratus clouds are too reflective. Note that the short wave cloud forcing from the satellite observations cannot be reliably retrieved over ice-covered surfaces. The long wave cloud forcing is slightly underestimated in the tropics in simulation E5-het. As the ice water path is only 8% lower in E5-het as compared to E5-hom, it suggests that heterogeneous freezing is either producing too few ice crystals and/or that the ice clouds form at too low altitudes in E5-het (cf figure 1). Both mechanisms act to reduce long wave cloud forcing in E5-het as compared to E5-hom (table 2). Simulation E5-alpha is a clear outlier in all the quantities displayed. Its 14 times higher ice crystal number concentration (cf figure 1) causes the crystals to be too small. These small crystals do not sediment efficiently, keeping the water in the atmosphere. This results in a drastic overestimation of both the short wave and long wave cloud forcing. Thus, the small deposition coefficients found in laboratory data do not seem adequate to be applied globally. The data of Magee et al (2006) are suggestive of an RH\textsubscript{i} dependence of \( \alpha \) with higher values of \( \alpha \) at higher RH\textsubscript{i}. Investigating a value of \( \alpha \) that varies with RH\textsubscript{i} is beyond the scope of this study but will be exploited in future.

The frequency of occurrence of different supersaturations with respect to ice in cloud-free regions has been obtained from the MOZAIC (measurement of ozone on airbus in-service aircraft) aircraft data (Helten et al 1998, Spichtinger et al 2004) and Microwave Limb Sounder (MLS) satellite data (Read et al 2001, Spichtinger et al 2003) for Northern Hemisphere mid-latitudes (30°N–60°N) and the tropics (30°S–30°N) and is shown in figure 3. From the MOZAIC data base we used 1 min averaged data, i.e. representing a horizontal resolution of ~15 km along a flight path. The footprint/field of view of MLS is approximately 100×200 km\textsuperscript{2} in the horizontal directions, the vertical resolution is about 2–3 km, depending on the pressure level (Read et al 2001, Spichtinger et al 2003).

One difference between the MLS and MOZAIC observations is the cloud screening. MLS is relatively insensitive to ice clouds with IWC < 5 mg m\textsuperscript{-3}, thus the relative humidity retrieval is not affected significantly by thin clouds. On the other hand, an effective cloud clearing has been carried out for thick cirrus. Thus, MLS data represents clear air or maybe data contaminated by thin or subvisible cirrus which does not artificially enhance the retrieved RH\textsubscript{i} values (Spichtinger et al 2003). MOZAIC, on the other hand, is almost a point measurement. It cannot distinguish between cloudy and clear air. Data from thick clouds are, however, excluded as the
aeroplanes carrying the instruments avoid big cloud systems. Alternatively AIRS satellite data have been used (Gettelman et al. 2006) for detecting ISSR regions. The number of vertical levels which contain information of ISSRs from AIRS is superior to that from MLS, but the actual vertical resolution is rather comparable in the cirrus levels (AIRS: 100, 150, 200, 250 hPa as compared to 147 versus 215 hPa from MLS). However as there is no cloud screening being conducted for the AIRS data, we decided to stick to the MLS data.

As shown in figure 3, both observational data sets suggest an exponential decrease for RH above 100%. They differ in the slope, with the MLS slope being less steep than the MOZAIC slope. This likely results from the larger field-of-view of the MLS data that misses smaller ISSR regions. As smaller ISSR regions are probably less supersaturated, this would bias the MLS slopes towards higher supersaturations. On the other hand, the scale of the MLS data might be more adequate for the comparison with a global climate model. On the other hand, there are more uncertainties associated with estimating RH from satellite than from the in situ observations of MOZAIC. Thus, both estimates have their merits and limitations.

The exponential RH slope has been shown to be quite variable from different observations and its origin is not exactly known (Spichtinger et al. 2002, 2003, Haag et al. 2003, Gettelman et al. 2006). It has been suggested that the RH slope stems from mixing processes (Gierens et al. 1999). Because of the uncertainties associated with the RH slope, it is most important for the GCM to reproduce an exponential slope at all. In the simulations, we obtained RH by restricting the analysis to grid boxes with less than 0.1 mg kg\(^{-1}\) condensate. However, varying the threshold of cloud condensate in the model simulations does not affect the exponential slope significantly (not shown).

In the Northern Hemisphere between 30° and 60° N, the exponential decrease of RH in simulation E5-hom is not as pronounced as suggested in the MOZAIC data but it matches the MLS slope at altitudes between 130 and 200 hPa at RH below 150%. At lower altitudes RH remains too high almost throughout. The exponential decrease in E5-hom is better matched in the tropics, where it lies between the observational estimates for RH below 155% at both altitude ranges. As the sample size becomes smaller at higher supersaturations, the error increases and the highest values of RH are less certain. Thus achieving a good agreement between the model and the observations at lower RH is more important than matching the RH slope at higher supersaturations.

Of all simulations, E5-alpha shows the best agreement with MOZAIC data at RH below 140% in terms of the RH slope. This could suggest that the ice crystals in all other simulations sediment out too rapidly so that RH cannot be sufficiently depleted. Simulation E5-het clearly fails to agree with observations at RH exceeding 130% because by design it depletes higher RH, whereas they are observed rather frequently (figure 3 and Peter et al. 2006).

In the Northern Hemisphere, simulations E5-homhet and E5-homhet10 are rather similar to simulation E5-het, because there are sufficient immersed dust nuclei to switch from homogeneous to heterogeneous freezing. Such a drastic switch from homogeneous to heterogeneous freezing is, however, not supported by observations. This is an artifact of the simplified homogeneous versus heterogeneous freezing treatment that causes the heterogeneous freezing, when present, to be too effective. In the tropics, the RH slopes of E5-homhet and E5-homhet10 are similar to those of MOZAIC above 130% but with too low frequencies. Also from these comparisons, heterogeneous freezing seems to be the most likely freezing scenario. Limited heterogeneous freezing might take place, but probably in even fewer cases than considered in the E5-homhet10 sensitivity simulation, or in combination with a lower accommodation coefficient.

Geographical distributions of ISSRs as seen by the MLS (Spichtinger et al. 2003) are available at 147 hPa and at 215 hPa and are shown in figure 4. A comparison of MLS data with MOZAIC data in the extratropical Northern Hemisphere (30°–90° N) between 175 and 225 hPa reveals that MOZAIC detects ISSRs on average during 10% of the time, while MLS only detects ISSRs 2% of the time (Spichtinger et al. 2003). Because of the factor 5 difference between MLS and MOZAIC the absolute values of ISSRs in figure 4 should not be compared with each other, but just the general geographical distributions.

The corresponding results from the ECHAM5 model simulations are divided into levels below and above 200 hPa. The same cloud screening as for MLS is applied to the model simulations, i.e. ISSRs are restricted to grid boxes with less than 5 mg m\(^{-3}\) condensate. Because simulation E5.5-homhet10 is rather similar to simulation ECHAM5.5-homhet, it is not shown here.

The observations suggest that the frequency of ISSRs in the tropics can exceed 50% of the time at 147 hPa and can approach 30% at 215 hPa. The higher frequency of occurrence of ISSRs in the tropics is related to deep convection insofar as the region of the maximum frequency of occurrence of ISSRs follows the intertropical convergence zone (Spichtinger et al. 2003). It is not clear yet if deep convection itself produces ISSRs by vertical transport of high water vapor concentrations in the convective cell itself or if ISSRs are formed at the top of the cells from lenses of higher specific humidity left over by former convective activity.

The simulations E5-hom, E5-het and E5-homhet also suggest a higher frequency of occurrence of ISSRs in the tropics than in the mid-latitudes. In simulations E5-hom, E5-het and E5-homhet maximum frequencies exceeding 30% in the tropics at altitudes above 200 hPa are captured as well. However, ECHAM5 predicts a far higher frequency of occurrence of ISSRs in the storm tracks than suggested by MLS, which is especially pronounced at altitudes below 200 hPa. In simulation E5-alpha, the contrast between a high frequency of occurrence in the tropics and a low frequency of occurrence in high latitudes at altitudes above 200 hPa is less pronounced in worse agreement with observations. Also, this simulation predicts more ISSRs at altitudes below 200 hPa than at altitudes above 200 hPa, which is contrary to the observations and the other model simulations. On the other hand, simulation E5-alpha suggests fewer ISSRs at the higher latitudes, which agrees better with observations.
4. Conclusions

The heterogeneous freezing scheme for cirrus clouds that was developed and tested in ECHAM4 has been incorporated into ECHAM5 (homogeneous freezing was available previously in ECHAM5). In addition, a few modifications with respect to the latest version of the ECHAM5 model with the two-moment cloud microphysics schemes (Lohmann 2008) are included in this version. The main improvement is the inclusion of a consistent ice crystal fall velocity not only for the ice crystal mass but also for its number concentration. The second improvement is a change in the ice crystal shape for clouds warmer than $-35^\circ$C and the parameterization of the effective radius of ice crystals both in cirrus and in mixed-phase clouds.

In this paper we tested its sensitivity with respect to the accommodation coefficient and with respect to heterogeneous
versus homogeneous freezing. The main findings are:

- The ice crystal number concentration and thus the cloud radiative properties are far more sensitive to the deposition coefficient than to the mode of freezing. This might partly be a problem of the design of the simulations with homogeneous and heterogeneous nucleation not being competitive but regionally separated.
- If the deposition coefficient is decreased from 0.5 to 0.006 as suggested by laboratory data and by assuming only homogeneous freezing, the ice crystal number concentration increases by a factor of 14, while the impact on ice water content and cloud cover is much smaller. This changes the global annual mean short wave and long wave cloud forcing by 15 and 18 W m$^{-2}$, respectively. However, the comparison with observations of ice water path and ice crystal size in semi-transparent cirrus, short wave and long wave cloud forcing suggests that a deposition coefficient as low as 0.006 is not appropriate for a global scale model. A similar conclusion has been drawn from adiabatic parcel model studies (Kay and Wood 2008).
- The impact of heterogeneous freezing versus homogeneous freezing is much more moderate than changing the deposition coefficient with global annual mean changes of the short wave and long wave cloud forcing by 2.7 and 4.7 W m$^{-2}$, respectively.
- The large difference between the frequency spectra of supersaturations with respect to ice from MLS and MOZART does not permit a conclusive evaluation about which model version performs best.

Simulations E5-homhet and ECHAM5-homhet10 are clearly simplified treatments of homogeneous and heterogeneous freezing as they ignore the competition between these freezing modes. A physical parameterization that takes this competition into account was developed by Kärcher et al. (2006). In order to apply it in a GCM, different categories of ice crystals are needed as the heterogeneously formed ice crystals will form at a lower RH$_i$. They have a head start and thus grow to a larger size than the homogeneously formed ice crystals. Such a scheme is currently under development and will be implemented in future.

In simulations E5-het, E5-homhet and E5-homhet10, the number concentration of available immersion dust nuclei is calculated every timestep from all dust aerosols in the internal accumulation and coarse mode minus the pre-existing ice crystal number concentration. This is due to the lack of a memory of how many immersion dust nuclei entered a persistent cloud in previous timesteps. Thus, at every timestep only those immersed dust nuclei whose concentration exceeds that of the pre-existing ice crystal number concentration are available for freezing, which might underestimate the potential ice nuclei number concentration. A proper treatment of which aerosols enter cloud droplets and ice crystals has been recently developed for ECHAM5 (Hoese et al. 2008b). It will be applied in future so that we can take the actual number of ice nuclei that were already activated into account and only subtract them from all available dust nuclei in subsequent timesteps.

Acknowledgments

We thank the two anonymous reviewers for their helpful comments and suggestions, Claudia Stubenrauch for providing the TOVS data, Sylvaine Ferrachat and Rebekka Posselt for technical help and the German (DKRZ) and Swiss Computing Centres (CSCS) for computing time. This study contributed towards the Swiss climate research program NCCR Climate. It was partly supported by the EC within the framework of the MC fellowship ‘Impact of mesoscale dynamics and aerosols on the life cycle of cirrus clouds’ and partly by the Integrated Project SCOUT-O3.

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