**Atmospheric
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Sensitivity studies of dust ice nuclei effect on cirrus clouds with the Community Atmosphere Model CAM5

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Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

Abstract

In this study the effect of dust aerosol on upper tropospheric cirrus clouds through heterogeneous ice nucleation is investigated in the Community Atmospheric Model version 5 (CAM5) with two ice nucleation parameterizations. Both parameterizations consider homogeneous and heterogeneous nucleation and the competition between the two mechanisms in cirrus clouds, but differ significantly in the number concentration of heterogeneous ice nuclei (IN) from dust. Heterogeneous nucleation on dust aerosol reduces the occurrence frequency of homogeneous nucleation and thus the ice crystal number concentration in the Northern Hemisphere (NH) cirrus clouds compared to simulations with pure homogeneous nucleation. Global and annual mean shortwave and longwave cloud forcing are reduced by up to $2\,\mathrm{W\,m}^{-2}$ due to the presence of dust IN, with the net cloud forcing change of -0.2 to $-0.4\,\mathrm{W\,m}^{-2}$ (cooling). Comparison of model simulations with in situ aircraft data obtained in NH mid-latitudes suggests that homogeneous ice nucleation may play an important role in the ice nucleation at these regions with temperatures of 205–230 K. However, simulations overestimate observed ice crystal number concentrations in the tropical tropopause regions with temperatures of 190–205 K, and overestimate the frequency of occurrence of high ice crystal number concentration ($>200\,\mathrm{l}^{-1}$) and underestimate the frequency of low ice crystal number concentration ($<30\,\mathrm{l}^{-1}$) at NH mid-latitudes. These results highlight the importance of quantifying the number concentrations and properties of heterogeneous IN (including dust aerosol) in the upper troposphere from the global perspective.

1 Introduction

Cirrus clouds composed of ice crystals cover roughly 30 % of the globe (Wang et al., 1996; Wylie and Menzel, 1999), and have been shown to play an important role in modifying the global radiative balance through scattering shortwave radiation and absorbing and emitting longwave (LW) terrestrial radiation (Corti et al., 2005; Kay and Gettelman,

2009; Liou, 1986; Lohmann and Roeckner, 1995; Ramanathan and Collins, 1991). Cirrus clouds also modulate water vapor in the upper troposphere and lower stratosphere (Gettelman et al., 2002; Korolev and Isaac, 2006; Krämer et al., 2009).

Although cirrus clouds are an important player in the global climate system, there are still large uncertainties in the understanding of cirrus cloud processes (DeMott et al., 2003; Lohmann et al., 2008; Barahona and Nenes, 2011). Compared to droplet activation in warm clouds, ice nucleation in cold clouds is more complicated. There are multiple ice nucleation mechanisms identified depending on aerosol surface properties and environmental conditions. Cirrus clouds can be formed by the homogeneous freezing of cloud droplets and aerosol solution particles at low temperatures ($< -37\,^\circ$C) and high relative humidities over ice (RH$_i$, 140–160 %) (e.g., Koop et al., 2000). Cirrus clouds can also be formed by the heterogeneous nucleation of ice on aerosol particles (Pruppacher and Klett, 1997), called ice nuclei (IN).

Various insoluble or partly insoluble aerosol particles can act as IN in deposition, contact, immersion, and condensation "freezing" modes, such as mineral dust, metallic particles, and primary biological particles (Cziczo et al., 2004; DeMott et al., 2000; Szyrmer and Zawadzki, 1997). Although very scarce, heterogeneous IN could significantly impact the ice crystal number concentration compared to pure homogeneous freezing by initiating ice nucleation earlier, depleting available water vapor and ultimately hindering the occurrence of homogeneous freezing (Chen et al., 2000; DeMott et al., 1994; Kärcher and Lohmann, 2003; Liu and Penner, 2005; Barahona and Nenes, 2009a,b). The relative importance of heterogeneous IN depends on the concentration of IN as well as the cooling rate (DeMott et al., 1994; Kärcher and Lohmann, 2003). Till now the importance of heterogeneous IN on global upper tropospheric ice number concentrations remains unclear, partially due to the scarcity of IN measurements and instrument artifacts of in situ ice crystal number measurements (Heymsfield, 2007; McFarquhar et al., 2007). Recent measurements indicate that heterogeneous IN might play a more important role than was previously thought in the Tropical Tropopause Layer (TTL) with extreme low temperatures (less than $-70\,^\circ$C) after correcting the

measurement artifacts (Jensen et al., 2010) and after identifying new classes of particles and phase states that can act as IN (Abbatt et al., 2006; Murray et al., 2010).

There have been significant advancements in the development of ice nucleation parameterizations for large-scale models. Homogeneous freezing of aqueous solutions can be relatively well represented by theory (Koop et al., 2000). For heterogeneous nucleation, empirical fits of IN observations are often used (Cooper, 1986; Cotton et al., 1986; DeMott et al., 1998; Meyers et al., 1992). Heterogeneous ice nucleation rates can also be calculated using the classical nucleation theory (CNT) (Fletcher, 1962; Pruppacher and Klett, 1997; Khvorostyanov and Curry, 2000, 2004). Phillips et al. (2008) derived empirical formulations for parameterizing atmospheric IN that account for contributions from different chemical species of aerosol in addition to temperature and ice supersaturation, based on observations from several field campaigns obtained from the Colorado State University (CSU) Count Flow Diffusion Chamber (CFDC). Using a larger dataset obtained from the same CFDC, DeMott et al. (2010) derived an empirical formulation of the number concentration of IN in mixed-phase clouds as a function of temperature and number concentration of aerosol particles with sizes larger than 0.5 μm. Key parameters (contact angle and activation energy) in CNT can also be constrained using laboratory data for different types of aerosols (Chen et al., 2008), and possibly ambient observations (Curry and Khvorostyanov, 2012).

Determining the ice crystal concentration in cirrus clouds require (in addition to a nucleation theory) knowledge of the supersaturation that develops in clouds; this is typically accomplished by considering a dynamical parcel framework (e.g., Pruppacher and Klett, 1997) and determining ice crystal number from the maximum supersaturation that develops. Kärcher and Lohmann (2002a,b) first derived a formulation for homogeneous freezing of sulfate solution droplets based on this approach The parameterization was extended in Kärcher et al. (2006) to consider the competition of heterogeneous ice nucleation on mineral dust and black carbon with homogeneous nucleation. Liu and Penner (2005) presented a parameterization for ice number concentration for large-scale models based on parcel model numerical simulations, and

Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

considered the competition between homogeneous and heterogeneous nucleation at temperature less than −35 °C. Heterogeneous nucleation was based on the CNT. The ice crystal concentration is related to updraft velocity, temperature, and aerosol concentration. Barahona and Nenes (2009a,b) provided a framework for ice nucleation parameterizations by solving analytically the parcel model equations. These parameterizations provide expressions for ice crystal size distribution and concentration, explicitly considering the effects of aerosol size and number, updraft velocity, and deposition coefficient, and the competition between homogeneous nucleation and heterogeneous nucleation. The IN nucleation "spectrum" (i.e., the number of IN as a function of parcel supersaturation) can have any forms and be derived from CNT or empirical data.

Using the Liu and Penner (2005) parameterization implemented in the Community Atmospheric Model version 3 (Liu et al., 2007), Liu et al. (2009) investigated the impacts of anthropogenic aerosol (black carbon and sulfate) on ice clouds and climate, and found that the effect of anthropogenic black carbon on cirrus clouds through ice nucleation depends critically on its ice nucleation efficiency assumed in the model and thus the relative importance of homogeneous and heterogeneous ice nucleation. Using the European Centre/Hamburg (ECHAM) general circulation model with the Kärcher et al. (2006) ice nucleation parameterization, Hendricks et al. (2011) studied the effects of heterogeneous IN (mineral dust and black carbon) on cirrus clouds, found that the annual zonal mean cloud ice number concentration is reduced by up to 20 % in the tropics and 1–10 % in the midlatitudes due to the heterogeneous IN. Significant reduction (up to 10 %) in the mean ice water content are modeled, which leads to the reduction of annual and zonal mean water vapor mixing ratio of up to 5 % at cirrus levels.

In this study, the ice nucleation parameterization of Barahona and Nenes (2009b, hereafter as BN09) was implemented in the Community atmosphere Model version 5 (CAM5), which is the atmospheric component of the Community Earth System Model version 1 (CESM1). Together with the existing Liu and Penner (2005, hereafter as LP05) ice nucleation parameterization in CAM5, we investigate the sensitivity of cirrus

clouds and climate to different formulations of heterogeneous ice nucleation (through its competition with homogeneous freezing). Modeled ice crystal number concentration and RH_i are compared with available observational data to understand the ice nucleation mechanisms in the upper troposphere. Sensitivity tests for pure homogeneous and pure heterogeneous nucleation were performed to examine the importance of heterogeneous IN from dust aerosol. The paper is organized as follows. Model description is presented in Sect. 2. Section 3 compares modeled ice crystal number concentrations from different parameterizations. Section 4 compares model results with observational data. Section 5 presents climate impacts of dust IN and conclusions are in Sect. 6.

2 Model description

2.1 CAM5

This study uses CAM5, the latest released version of CAM (Neale et al., 2010). Compared to CAM version 3 (Collins et al., 2006a,b), several important physics components have been updated. CAM5 includes a two-moment stratiform cloud microphysics scheme (Morrison and Gettelman, 2008; Gettelman et al., 2008). The model radiation scheme has been updated to the Rapid Radiative Transfer Model for GCM described by Iacono et al. (2008). The deep convective scheme has been modified following Neale et al. (2008). The shallow convective scheme follows that of Park and Bretherton (2009). The moist boundary layer scheme is based on Bretherton and Park (2009). The cloud condensation parameterization uses a new liquid cloud macrophysics closure described by Park et al. (2012). The aerosol treatment in the model uses the modal aerosol module (MAM) (Liu et al., 2011). MAM treats the internal mixing within a mode and external mixing between modes of major aerosol species (sulfate, black carbon, primary organic carbon, secondary organic carbon, mineral dust and seas salt), and predicts mass mixing ratio of different aerosol species in each mode and number mixing ratio of each mode.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2.2 Ice cloud scheme in CAM5

Gettelman et al. (2010) modified cloud microphysics and macrophysics closure related to ice phase for CAM5 as compared to the CAM version 3.5 used in MG08. This includes: (1) condensation closure was modified to perform saturation adjustment
5 only for liquid water. This allows the supersaturation over ice in ice clouds. Water vapor deposition onto ice crystals in mixed-phase and ice clouds is treated with process rate calculations. Sublimation occurs when water vapor is sub-saturated over ice; (2) the Bergeron-Findeisen process is treated for mixed-phase clouds as follows: cloud condensation calculated by grid-scale macrophysics closure is assumed to be initially
10 cloud liquid. During the same time step, vapor deposition occurs if ice is present, and would deplete cloud liquid given the assumption that water vapor is saturated with respect to water in mixed-phase clouds; (3) the diagnostic fractional cloudiness scheme for ice clouds is still based on Slingo (1987), but calculated using the total water, not just water vapor; and (4) ice nucleation in MG08 is changed as discussed in Sect. 2.3.

15 ## 2.3 Ice nucleation parameterizations in the standard CAM5

Along with vapor deposition, ice nucleation provides a source for cloud ice in MG08. Loss terms include aggregation, autoconversion, accretion, melting and sedimentation. The empirical temperature-based IN parameterization from Cooper (1986) in MG08 was replaced in CAM5 with an explicit ice nucleation scheme with links to the
20 aerosol concentration and type (Gettelman et al., 2010). Ice nucleation is based on Liu et al. (2007), which includes homogeneous nucleation on sulfate, heterogeneous immersion freezing on mineral dust and competition between the two mechanisms at temperatures below $-37\,^\circ$C (Liu and Penner, 2005). Although LP05 includes the immersion nucleation on soot it is not considered in this work as it is still highly uncer-
25 tain (Kärcher et al., 2007). In the LP05 parameterization, nucleated ice crystal number concentration is computed based on temperature, relative humidity, sulfate and dust number concentration, and sub-grid updraft velocity. The sub-grid variability of updraft

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

velocity needed for driving the parameterization is derived from the square root of turbulent kinetic energy (TKE) (Bretherton and Park, 2009) with an assumed maximum value of $0.2\,m\,s^{-1}$ (Gettelman et al., 2010). The limiting IN number concentration above which homogeneous nucleation is prevented is derived from parcel model simulations as a function of temperature and vertical velocity, and used in the parameterization of the competition between homogeneous and heterogeneous ice nucleation.

In mixed-phase clouds with temperatures between 0 and −35 °C, contact freezing of cloud droplets through Brownian coagulation with insoluble IN (assumed to be mineral dust) (Young, 1974), deposition/condensation nucleation (Meyers et al., 1992), and immersion freezing of cloud droplets (Bigg, 1953) are included. Ice multiplication through Hallet-Mossop secondary ice production due to accretion of drops by snow is included following Cotton et al. (1986). More detailed description can be found in Gettelman et al. (2010).

We note that there are a few changes between this study and Gettelman et al. (2010) in CAM5 for the LP05 parameterization: (1) we use all Aitken mode sulfate aerosol for homogeneous nucleation while Gettelman et al. used a portion of the Aitken mode sulfate with diameter greater than 0.1 μm; (2) accumulation mode dust (0.1 to 1.0 μm diameter) in addition to coarse mode dust (1.0–10.0 μm diameter) is used in the ice nucleation; and (3) we extend the LP05 parameterization for ice nucleation in pure ice clouds with temperatures between 0 and −37 °C where only heterogeneous nucleation occurs. This change is to be consistent with BN09 parameterization which includes ice nucleation for pure ice clouds at temperatures less than −37 °C as well as temperatures between 0 and −37 °C. We note that the third modification has a small impact on model results (not shown).

2.4 BN09 parameterizations in CAM5

For this study, we implemented the BN09 framework in CAM5. This framework explicitly considers the competition between homogeneous and heterogeneous freezing, and has the flexibility of using different IN "nucleation spectra" either from empirical

correlations derived from observation data or from CNT. The "nucleation spectrum" describes the number of ice crystals nucleated from IN as a function of ice supersaturation. In the version of the BN09 code used in this study, there are four options for "nucleation spectrum". The first one (MY92) is derived from in-situ measurements of IN concentrations for temperature between 250 and 266 K and ice supersaturation between 2 and 25 % following Meyers et al. (1992). The second one (PDG07) is also based on in-situ measurement described by Phillips et al. (2007). The third one (PDA08), presented by Phillips et al. (2008), is a more comprehensive formulation that takes the surface area contribution from different types of aerosol (i.e., dust and soot) with sizes larger than 0.1 µm diameter and freezing modes (i.e., deposition and immersion) into account. The last one is derived from a semi-empirical form of CNT. The PDA08 is used in this study as it uses fewer tuning parameters. Barahona et al. (2010) carried out sensitivity studies using a global chemical transport model with four different nucleation spectra and found that when the competition between homogeneous and heterogeneous freezing is considered, global mean ice crystal number concentration vary by up to a factor of twenty depending on the nucleation spectrum used.

Under the BN09 framework, the number of ice crystals nucleated is a function of aerosol number concentration, temperature, updraft velocity, and accommodation coefficient for water vapor deposition on ice crystals. Ice crystal number is derived from solving equations for temperature, ice supersaturation, and ice water content for an ascending air parcel with initial condition that is saturated with respect to ice. For consistency, the same sub-grid updraft velocity is used to drive both BN09 and LP05. BN09 is applied only for pure ice clouds (i.e., cirrus clouds with temperatures less than −37 °C as well as pure ice clouds with temperatures between 0 and −37 °C when only heterogeneous nucleation takes place). In mixed-phase clouds, we use the same heterogeneous ice nucleation parameterizations in Gettelman et al. (2010), as documented in Sect. 2.3.

3 Modeled ice crystal number concentrations

We run CAM5 with prescribed present-day sea surface temperatures (SST) and aerosol and precursor emissions from Lamarque (2010) for 5 yr plus 3-month spin up, and results from the last 5-yr are used in the analysis. All simulations have been carried out at 2.5 × 1.9 horizontal resolution with 30 vertical levels. In the control case we use the LP05 and BN09 parameterizations in CAM5 including the competition between homogeneous and heterogeneous nucleation (i.e., "combined simulations", denoted below as LP and BN, respectively). Two additional sets of simulations were performed: pure homogeneous freezing (hereafter as LP-hom and BN-hom, respectively), and pure heterogeneous freezing (hereafter as LP-het and BN-het, respectively), for LP05 and BN09 parameterizations by turning off their representation of heterogeneous or homogeneous nucleation, respectively. Table 1 gives the list of simulations carried out. We note that ice formation in mixed-phase clouds discussed in Sect. 2.3 keeps the same between all these simulations.

The annual and zonal mean mass and number concentrations of mineral dust in the accumulation mode with the size (diameter) range of 0.1–1 µm and in the coarse mode with the size (diameter) range of 1–10 µm are shown in Fig. 1. About 90 % of the total dust mass is in the coarse mode, while the accumulation mode dust dominates the total dust number concentration (Liu et al., 2011). Accumulation mode dust number concentration is in the range of 0.1–1.0 cm^{-3} in the NH upper troposphere (100–300 hPa), where coarse mode dust number concentration is one order of magnitude lower. Dust is uplifted from the NH subtropics and midlatitude sources to the middle and upper troposphere and transported towards the Arctic in the upper troposphere. The dust number concentration in the upper troposphere agrees reasonably well with the model result by Hendricks et al. (2011). As we noted in Sect. 2, dust number concentration is used as an input for the heterogeneous nucleation formulations in LP05 and BN09.

An offline (box model) comparison between the LP05 and BN09 parameterizations for the pure homogeneous nucleation case done by Barahona and Nenes (2008)

showed that the two parameterizations give similar ice number concentrations at cirrus temperatures less than $-40\,°C$ and updraft velocity less than $1\,m\,s^{-1}$. The comparison of in-cloud ice crystal number concentration (Ni) for LP-hom and BN-hom is shown in Fig. 2 (middle panel). We can see that distribution patterns of Ni from BN-hom and LP-hom cases are similar, with higher Ni in the Southern Hemisphere (SH) and in the tropics. Both LP-hom and BN-hom cases predict Ni higher than $500\,l^{-1}$ in the tropical tropopause and in SH. Ni is reduced at warmer temperatures in the middle troposphere (300–500 hPa) with values of $20–100\,l^{-1}$, consistent with the nucleation rate reduction with increasing temperatures for homogeneous ice nucleation (Koop et al., 2000; Pruppacher and Klett, 1997) and aggregational growth of small ice crystals during the gravitational sedimentation from above. In the lower troposphere (700–850 hPa) Ni increases to $100–200\,l^{-1}$ in storm track regions due to the ice nucleation in mixed-phase clouds.

To compare BN09 and LP05 under the same condition, we turn off the soot contribution to IN number in BN09 (i.e., only dust particles can act as IN in both LP05 and BN09). An offline (box model) comparison showed that heterogeneous IN number concentration from CNT-based spectrum is typically a few times higher than that from the PDA08 (Barahona and Nenes, 2009a) with the same input of aerosols. As shown in Fig. 2 (lower panel), Ni calculated from BN-het with PDA08 is in the range of $10–50\,l^{-1}$ in the upper troposphere regime, much lower than that ($100–300\,l^{-1}$) in the NH mid-latitudes from LP-het. The higher Ni ($100–200\,l^{-1}$) in the subtropical and mid-latitudes of NH in LP-het is associated with the Saharan and Asian dust. The different Ni between LP-het and BN-het is due to the different formulations of IN number concentration (i.e., the CNT method in LP-het and the PDA08 in BN-het) even though they have the input of almost the same dust number concentrations between the two simulations (not shown).

Ice crystals formed from heterogeneous nucleation can inhibit the subsequent homogeneous nucleation through water vapor consumption in the cloud parcel. Offline comparison by Barahona and Nenes (2009b) showed that the limiting heterogeneous

IN number (N_{lim}) that would completely inhibit homogeneous nucleation is lower in LP05 parameterization than that in BN09 parameterization (i.e., homogeneous nucleation is more easily prevented in LP05 parameterization) under the same IN number concentration. Therefore we expect that the occurrence frequency of homogeneous nucleation will be lower with the LP05 parameterization having both higher heterogeneous IN number concentration and lower N_{lim} than the BN09 parameterization when both homogeneous and heterogeneous nucleation are allowed to occur and compete. As a result Ni will be lower with LP05 in regions where there is sufficient heterogeneous IN to inhibit the homogeneous nucleation. Indeed, this is the case. As shown in Fig. 2 (upper panel), Ni in the NH upper troposphere is significantly reduced (by up to a factor of 2) in LP compared to LP-hom due to higher heterogeneous dust IN concentration (100–300 l^{-1}) there from LP05 (lower panel), clearly indicating the inhibition of homogeneous nucleation by heterogeneous nucleation. There are no obvious changes between BN and BN-hom due to the lower heterogeneous dust IN number concentration (10–50 l^{-1}) in the upper troposphere which does not have a strong impact on the homogeneous nucleation there, consistent with the study by Barahona et al. (2010).

Figure 3 shows the frequency distribution of homogeneous nucleation occurrence events in CAM5 (i.e., count as 1 if homogeneous nucleation occurs, otherwise 0 at each 3-dimensional grid) for combined homogeneous and heterogeneous nucleation case and pure homogeneous nucleation case. We can see that the frequency of occurrence of homogeneous nucleation is similar for LP-hom and BN-hom case (lower panel). Heterogeneous IN reduces the frequency of homogeneous nucleation significantly for LP05 parameterization (upper panel), due to its higher heterogeneous IN number concentration and lower N_{lim}, while the reduction is evident only in the NH high-latitudes for BN09.

Figure 4 shows the percentage contribution of ice crystals from heterogeneous ice nucleation to total ice number diagnosed in the ice nucleation parameterizations for the LP and BN cases. We can see that homogeneous nucleation is the dominant contributor to Ni in the upper troposphere in the tropics and in the SH, and contribution from

heterogeneous nucleation is much smaller there ($< 10\,\%$). In both LP and BN simulations, heterogeneous IN concentration is not high enough to completely turn off the occurrence of homogeneous freezing there. In the NH heterogeneous nucleation becomes more important especially for the LP case. The contribution of heterogeneous nucleation to total ice crystal number can reach over 50 % for the LP case due to the larger IN number from the CNT. The contribution from heterogeneous nucleation is less than 10 % (and mostly less than 1 %) in the upper troposphere for the BN case.

4 Comparison of model results with observations

Krämer et al. (2009) collected an extensive aircraft observational dataset in the temperature range of 183–240 K. We compare model simulations against this dataset for pure homogeneous, pure heterogeneous and combined cases with LP05 and BN09 parameterizations. We output modeled in-cloud Ni every 3 h and bin them into 1 K interval of temperatures from 190 to 240 K (i.e., in the cirrus cloud regime) in the upper troposphere from 30° S to 75° N. The range between 25th and 75th percentiles of modeled Ni within each temperature bin is shown in Fig. 5. The most distinctive feature of Fig. 5 is that observed Ni tends to increase with increasing temperature for the whole temperature range. Ni is in the range of ~ 10–$60\,l^{-1}$ at temperatures below 205 K. There have been studies to suggest that under these very low temperatures (i.e., near the tropical tropopause layer) aerosols rich with organic matter (e.g., Froyd et al., 2009) may become glassy, prevent homogeneous ice nucleation, and only allow heterogeneous nucleation to happen (Jensen et al., 2010; Murray et al., 2010). Barahona and Nenes (2011) also suggested that low Ni would result from the emergence of dynamical equilibrium states, favored at low temperatures. There is no obvious Ni trend in the temperature range between 205 K and 230 K, and Ni is higher than that in the lower temperatures and within the range of $20\,l^{-1}$ to a few hundred per liter. Above 230 K, Ni increases with temperature and is frequently higher than $1000\,l^{-1}$. This could be due to the shattering of large ice crystals at warmer temperatures, as

suggested in Field et al. (2003) and Lawson (2011). Simulated Ni decreases with increasing temperature for pure homogeneous nucleation (LP-hom and BN-hom) and combined simulations (LP and BN) especially with BN09 parameterization, consistent with the predominance of homogeneous nucleation. Ni from LP-hom is close to that from BN-hom. Ni from LP is lower than that from LP-hom especially in the NH mid-latitudes with temperatures between 205–230 K by up to a factor of 2, because of the stronger effect of heterogeneous ice nucleation in the LP05 parameterization than in BN09 (Fig. 4). Ni from BN is almost the same as BN-hom due to the low heterogeneous IN concentration from the PDA08 spectrum. Ni from pure homogeneous (LP-hom and BN-hom) and combined simulations (LP and BN) overestimates observed Ni by a factor of 10–100 in the temperature range of 190–205 K, while in the temperature range of 205–230 K, Ni from combined simulations is closer to the observed values. Compared to pure homogeneous and combined nucleation simulations, pure heterogeneous nucleation simulation with the BN09 parameterization (BN-het) shows better agreement with the observation in the temperature range of 190–205 K, although it is not clear if heterogeneous IN spectrum (PDA08) used in BN-het can be extrapolated to such low temperature regime. Ni from pure heterogeneous nucleation simulations (LP-het and BN-het) is too low (20–40 l^{-1}) in the temperature range of 205–230 K compared to the observational data. LP-het which is based on CNT gives a higher Ni than BN-het which is based on PDA08 in the whole temperature range. The above comparison is consistent with the assertion that homogeneous ice nucleation is not able to explain the very low Ni in the low temperature range of less than $\sim$ 205 K near the tropical tropopause, and pure heterogeneous nucleation may dominate there (Jensen et al., 2010). The low Ni in cirrus clouds at these temperatures may also be due to the occurrence of "dynamical equilibrium" states between ice production and sedimentation loss (Barahona and Nenes, 2011). However, pure heterogeneous nucleation on dust with both LP05 and BN09 parameterization would not be able to explain observed Ni concentrations ($\sim$ 100 l^{-1}) in the temperature range of 205–230 K, even in the LP-het

simulation with CNT. This indicates that homogeneous nucleation is the dominant ice nucleation mechanism in this temperature range.

Ice supersaturation is the driver of ice nucleation, and the level and distribution thereof gives insight on the relative importance of freezing mechanisms (e.g., Haag et al., 2003). We compare the probability distribution frequency (PDF) of relative humidity with respective to ice, RH_i, in 200–500 hPa from 30° S to 60° N from instantaneous model output with pure homogeneous, pure heterogeneous and combined nucleation for LP05 and BN09 parameterizations, respectively. The modeled RH_i is compared in Fig. 6 with humidity observations in the upper troposphere from the Measurements of Ozone, Water Vapor, Carbon Monoxide and Nitrogen Oxides by Airbus In-Service Aircraft (MOZAIC) project (Gierens et al., 1999) and from the Atmospheric Infra-Red Sounder (AIRS) satellite (Gettelman et al., 2006a). Both observed PDFs of RH_i from MOZAIC and AIRS show exponential decreases with increasing of RH. Note that MOZAIC may be subject to compressional heating artifacts (inflating the RH), and AIRS with a large RH uncertainty ($\sim 20\%$) suffers from its 2–3 km vertical weighting function that may reduce the RH and the frequency of occurrence of thin supersaturated layers (Gettelman et al., 2006b). The peak in frequency at 100 % RH_i in the simulations is due to cloud points that are not sampled by MOZAIC or AIRS. Modeled supersaturation for pure homogeneous and combined nucleation with the BN09 parameterization (BN-hom and BN) is higher than AIRS and lower than MOZAIC. Results from BN-hom and BN are similar. Model simulations with the LP05 parameterization (LP-hom and LP) underestimate the occurrence frequency of RH_i larger than ~ 120–140 %. As shown in Fig. 6, with the decrease of Ni from pure heterogeneous nucleation simulations (LP-het and BN-het), occurrence frequency of high ice supersaturation is significantly increased. The low Ni (20–50 l^{-1}) in the upper troposphere from BN-het simulation could not consume water vapor efficiently by depositional growth of ice crystals and thus occurrence frequency of ice supersaturation from this simulation is larger than that from MOZIAC at $RH_i \sim 150$–170 %. We note that low Ni (and

thus small surface area of ice crystals) has been used to explain the persistent high supersautration in cold cirrus clouds (Murray et al., 2010; Peter et al., 2006).

Number concentration and size distribution of ice particles were routinely measured over the DOE Atmospheric Radiation Measurement (ARM) South Great Plains site (36.6° N, 97.5° W) from December 2009 to June 2010 during the Small Particles in Cirrus (SPartICus) campaign (http://campaign.arm.gov/sparticus/). The suite of cloud particle probes included a 2D-S probe (Lawson et al., 2006), which was equipped with probe tips that reduced shattering and a post-processing algorithm designed to remove residual shattered particles (Lawson, 2011). SPartICus provides relatively long-term statistics ($\sim$ 150 h of in situ data) of concentrations of ice particles in cirrus. A recent comparison between historical and SPartICus measurements in deep cirrus shows that SPartICus data typically contain lower concentrations of small ($< \sim 50\,\mu m$) ice particles, particularly near cloud base where large particles contribute the most to shattering (Lawson, 2011). Figure 7 shows the PDF of in-cloud ice number concentrations from SPartICus data obtained during January to June 2010.[1] The most frequently observed N_i is in the range of about 10–200 l^{-1}, although ice number concentrations larger than 1000 l^{-1} are also observed in a few occasions.

To compare with the aircraft measurements collected over the SGP area during the SPartICus campaign, we sampled instantaneous ice crystal number concentrations at the SGP site (interpolated using values in four GCM grid-boxes closest to the site) every three hours from January to June from 5-yr simulations for different cases listed in Table 1. As most of the aircraft measurements were collected above 500 hPa, only samples above this level are considered in the analysis. All LP05 model simulations overestimate observed frequencies for N_i higher than $\sim$ 100 l^{-1}, and underestimate observed frequencies for N_i less than 20–40 l^{-1}. This is also the case for BN and BN-hom. BN-het produces two modes in the PDF: one at 10 l^{-1} and another at 200–400 l^{-1}. The lower N_i mode is due to the heterogeneous ice nucleation, while the higher mode is due

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[1]The SPartICus data used in this analysis are considered preliminary. However, final processing of the data is not expected to significantly alter the results shown here.

Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

to the convective detrainment, which also contributes to the occurrence of Ni at 200–400 l^{-1} for other cases (not shown). These comparisons suggest that homogeneous nucleation may play an important role in cirrus clouds to produce the high frequency of Ni higher than 100 l^{-1} over the ARM SGP site during the SPartICus. However model simulations underestimate the frequencies of occurrence for Ni less than 40 l^{-1} except for the BN-het simulation. This may indicate that CAM5 with large horizontal and vertical grid sizes may underestimate the growth processes of ice crystals by aggregation and/or rimming, which can reduce the ice number concentration in clouds (Spichtinger and Gierens, 2009a,b).

5   Climate impacts

In this section, we will examine the impact of heterogeneous IN on climate by comparing simulations between the pure homogeneous nucleation and the combined homogeneous and heterogeneous nucleation simulations. Figure 8 shows latitude-pressure cross sections of annual mean Ni (grid-box average), IWC (grid-box average), cloud cover, temperature, and specific humidity from the LP simulation and differences of these variables between LP-hom and LP. There is a significant increase in Ni in LP-hom compared to LP owing to the significant number of heterogeneous IN in the subtropics and midlatitudes from dust sources in North African and Asian deserts (see Fig. 1). Compared to LP, IWC in the upper troposphere in LP-hom is increased by 20 % and reduced downward due to smaller ice crystal sizes and slower sedimentation from the ice-producing layers. The strong increase in Ni enhances cloud cover in the tropics and the midlatitudes by 2–5 %, with a corresponding reduction of lower-level cloud cover by 1–2 %. Upper tropospheric temperature increases by 0.5–1.0 K due to the longwave radiative heating from the increased presence of cirrus. Upper tropospheric water vapor generally increases by 10–20 %. The higher tropical tropopause temperature increases the water vapor saturation pressure, and allows more water vapor to be transported to the lower stratosphere (Rosenfield et al., 1998; Liu et al., 2009). Thus

specific humidity in the lower stratosphere is increased by ∼ 10 %. In comparison, the difference in Ni between BN-hom and BN is much smaller with maximum changes in the tropical tropopause about $50\,l^{-1}$. IWC is changed by a few percents. Compared to BN, cloud cover in BN-hom increases by 1–3 % in the tropical tropopause with virtually no change in temperature there. Upper tropospheric and lower stratospheric water vapor in BN-hom is slightly reduced by less than 5 % (not shown).

Figure 9 shows the annual and zonal mean shortwave cloud forcing (SWCF), longwave cloud forcing (LWCF), ice water path (IWP), high cloud cover (CLDHGH), cloud-top ice effective radius (ACTREI), and column cloud ice number concentration (CDN-UMI) from the model simulations carried out in this study. Modeled SWCF and LWCF are compared with satellite retrievals from the Clouds and the Earth's Radiant Energy System (CERES) (Wielicki et al., 1996), and CLDHGH with combined CloudSat and CALIPSO cloud fractions (Gettelman et al., 2010). Overall, SWCF from combined nucleation simulations (LP and BN) agrees well with the observations, except in the tropics and subtropics where the SWCF is too strong. Noticeable differences in SWCF between different ice nucleation simulations are only in the tropics and subtropics. This is expected since SWCF in the extratropics is dominated from middle and low-level clouds. The SWCF differences reach $\sim 10\,\mathrm{W\,m^{-2}}$ between different simulations for both LP05 and BN09 parameterizations, with pure homogeneous nucleation simulations having the strongest (more negative) SWCF, followed by the slightly weaker SWCF from the combined nucleation simulations and then weakest SWCF from the pure heterogeneous nucleation simulations. This is consistent with the lowest Ni from pure heterogeneous nucleation simulations, the smallest IWP, and the weakest SWCF. Similar differences between different simulations occur for LWCF, but expand to the extratropics, since cirrus clouds have an important contribution to LWCF globally. LWCF from different model simulations underestimates the observations in the midlatitudes. The lowest Ni from the pure heterogeneous nucleation (LP-het and BN-het) results in the largest ice crystal size and highest sedimentation rate and thus the smallest IWP. The same is true for CLDHGH from BN-het which is lower by ∼ 10 % than other

two simulations (BN and BN-hom) in the SH extratropics due to the lowest Ni from BN-het simulation. There is no obvious difference in the cloud-top ice effective radius except for BN-het simulation which has a larger ice effective radius due to the lower Ni. The cloud-top ice effective radius is similar among the three simulations for LP05, although differences are much larger at lower cloud levels. Column ice number concentration from the pure homogeneous nucleation simulation is higher than that from the combined nucleation simulation especially in the NH midlatitudes for the LP05 parameterization, and both simulations are a few times higher than that from the pure heterogeneous nucleation simulation for both LP05 and BN09 parameterizations.

Table 2 gives the global and annual means of LWP, IWP, SWCF, LWCF, total and high cloud cover, and column ice number concentrations from different simulations. Comparing combined (LP) and pure homogeneous nucleation (LP-hom) simulations, heterogeneous dust IN reduces the column ice number concentration by 20 %. This reduces global mean SWCF (less negative) by $2.0\,\mathrm{W\,m^{-2}}$ and LWCF (less positive) by $2.4\,\mathrm{W\,m^{-2}}$, respectively for LP05. The net change in global mean cloud forcing (CF) is $-0.4\,\mathrm{W\,m^{-2}}$ (cooling). Global mean IWP is reduced by $1.2\,\mathrm{g\,m^{-2}}$ with a slight decrease of global mean LWP by $0.5\,\mathrm{g\,m^{-2}}$. The total and high cloud cover reduces by 0.1–0.2 % due to the compensation of increase and decrease at different vertical levels. The SWCF and LWCF in the pure heterogeneous nucleation simulation (LP-het) are much weaker (by $7\sim9\,\mathrm{W\,m^{-2}}$ on the global mean) compare to the above two simulations, due to a factor of ~4 lower column ice number concentration. High-cloud cover is also lower by 4 % on the global mean. The column ice number concentration with BN-het is half of that with LP-het. Comparing combined simulation (BN) and pure homogeneous nucleation (BN-hom), heterogeneous dust IN has a negligible effect on the global mean SWCF, and reduce the global mean LWCF by $0.3\,\mathrm{W\,m^{-2}}$ (cooling). The SWCF and LWCF in the pure heterogeneous nucleation simulation (BN-het) are lower than the BN and BN-hom simulations by $9–10\,\mathrm{W\,m^{-2}}$.

6   Discussion and conclusions

In this study the impact of heterogeneous dust IN on upper tropospheric cirrus clouds and climate are investigated using CAM5 with two ice nucleation parameterization frameworks (LP05 and BN09). Both parameterizations consider the homogeneous and heterogeneous nucleation and competition between the two mechanisms. The heterogeneous IN concentration from LP05 is based on the CNT, and is higher than that from BN09, which is based on the PDA08 spectrum. The difference is larger in the NH upper troposphere (by up to a factor of ten) where mineral dust particles from deserts are more abundant. Because of the inhibition of homogeneous nucleation by heterogeneous dust IN, the frequency occurrence of homogeneous nucleation in the upper troposphere is substantially reduced in the NH compared with that from the pure homogeneous nucleation simulation in LP05. The impact of heterogeneous dust IN is much smaller in BN09 because of the lower dust IN concentration as predicted by the PDA08. As a result, the overall ice crystal number concentration is reduced by up to a factor of 2 in NH in the combined nucleation simulation compared to the pure homogeneous nucleation simulation for LP05. Contribution of ice crystals nucleated from heterogeneous nucleation to total nucleated ice number concentration can reach over 50 % in the NH from the LP05 parameterization compared to less than 10 % globally for the BN09 parameterization.

We compare modeled cloud ice number concentration from different simulations with the two parameterizations with observations. Good agreement only occurs at temperatures lower than 205 K for the pure heterogeneous nucleation simulation with BN09 using the PDA08's heterogeneous IN spectrum. However, pure heterogeneous nucleation simulations give too low ice number concentrations at the temperature range of 205–230 K, and thus homogeneous nucleation may play a dominant role in the ice formation. This is also confirmed from the relatively long-term ($\sim$ 150 h) statistics of cirrus clouds obtained at a NH midlatitude site during the SPartICus campaign, which shows the large frequency (41 %) of high cloud ice number concentrations ($> 100\,\mathrm{l}^{-1}$). The

large fluctuation (by more than one order of magnitude) of observation data at a given temperature also suggest the role of heterogeneous nucleation affecting or completely inhibiting the homogeneous nucleation as well as the importance of aggregation and rimming of ice crystals. Model simulations with homogenous nucleation dominating the ice formation underestimate the occurrence frequency of cloud ice number concentration larger than $20\,l^{-1}$, while overestimates the frequency of higher ice number concentration ($> 100\,l^{-1}$) during the SPartICus.

Heterogeneous dust IN may have a significant global impact on cloud radiative forcing (SWCF and LWCF) by $2\,W\,m^{-2}$, ice water content by 20%, and upper tropospheric water vapor mixing ratio by 10–20% if there is sufficient number of heterogeneous IN ($\sim 200\,l^{-1}$) in the upper troposphere. The net cloud forcing change is $-0.4\,W\,m^{-2}$. However, even with a small number of heterogeneous IN (20–$50\,l^{-1}$) in the upper troposphere the net cloud forcing change, which is dominated by the LWCF change, can be $-0.3\,W\,m^{-2}$. These results highlight the importance of underestimating the ice nucleation mechanisms and of quantifying the number concentrations and properties of heterogeneous IN. We note that the relative importance of heterogeneous IN on upper tropospheric cirrus clouds also depends on the cooling rate (i.e., vertical velocity) of air parcels in clouds. If the vertical velocity is sufficient high (e.g., larger than $0.5\,m\,s^{-1}$) homogeneous nucleation may eventually take place after the heterogeneous nucleation and dominate the overall ice number concentration (e.g., Kärcher and Lohmann, 2003). The evaluation of modeled subgrid vertical velocity with SPartICus remote sensing and in situ aircraft observations will be conducted in our next study.

Model evaluation with observations indicates several areas of future model improvement. The subgrid features of ice supersaturation, and ice nucleation and growth need to be better represented in global climate models with horizontal grid sizes of $100\,km$. These ice microphysics parameterizations need to be linked with the cloud macrophysics scheme in climate models. This can be achieved by implementing a statistical PDF cloud macrophysics scheme (e.g., Kärcher and Burkhardt, 2008) into CAM3 (Wang and Penner, 2010) and CAM5 (M. Wang, personal communication), which will

be evaluated with observation data. Further model improvement would also separate the prediction of cloud ice crystals by sources, such as the ice crystals formed from homogeneous nucleation, heterogeneous nucleation and from convective detrainment (Hendricks et al., 2011). BN09 ice nucleation parameterization will also be modified to account for the impact of pre-existing ice crystals on ice nucleation and to include ice nucleation on glassy aerosols (Murray et al., 2010) and on ammonium sulfate (Abbatt et al., 2006). As we find from this study, major differences in CAM5 simulations with LP05 (the default scheme in CAM5) and the new implemented BN09 are due to their different heterogeneous ice nucleation formulations. We note that BN09 has four IN spectra to choose. The comparison of these IN spectra on ice crystal number concentration and impact on cirrus clouds under the same BN09 framework will be presented in our future study.

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Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

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Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◀ | ▶

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◀ | ▶

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Li-
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Modeling the effect of dust ice nuclei on cirrus clouds with CAM5

X. Liu et al.

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

McFarquhar, G. M., Um, J., Freer, M., Baumgardner, D., Kok, G. L., and Mace, G.: Importance of small ice crystals to cirrus properties: Observations from the Tropical Warm Pool International Cloud Experiment (TWP-ICE), Geophys. Res. Lett., 34, L13803, doi:10.1029/2007gl029865, 2007.

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. CAM5 cases in this study.

Case Name	Description
LP	LP05, combined nucleation
LP-hom	LP05, pure homogeneous nucleation
LP-het	LP05, pure heterogeneous nucleation
BN	BN09, combined nucleation
BN-hom	BN09, pure homogeneous nucleation
BN-het	BN09, pure heterogeneous nucleation

Table 2. Global annual mean cloud properties from sensitivity simulations listed in Table 1 for liquid water path (LWP, $g\,m^{-2}$), ice water path (IWP, $g\,m^{-2}$), shortwave cloud forcing (SWCF, $W\,m^{-2}$), longwave cloud forcing (LWCF, $W\,m^{-2}$), total cloud cover (CLDTOT, %), high cloud cover (CLDHGH, %), and column ice number concentration (CDNUMI, $\times 10^{8}\,m^{-2}$).

Case Name	LWP	IWP	SWCF	LWCF	CLDTOT	CLDHGH	CDNUMI
LP	46.9	18.3	−53.0	29.5	65.4	42.5	4.82
LP-hom	47.4	19.4	−55.0	31.9	65.6	42.6	5.86
LP-het	45.3	17.2	−46.3	22.6	62.1	38.8	1.34
BN	47.1	17.8	−52.9	28.8	64.5	41.1	5.75
BN-hom	46.9	17.8	−52.9	29.1	64.7	41.6	5.79
BN-het	44.6	15.4	−43.8	18.6	61.0	37.5	0.67
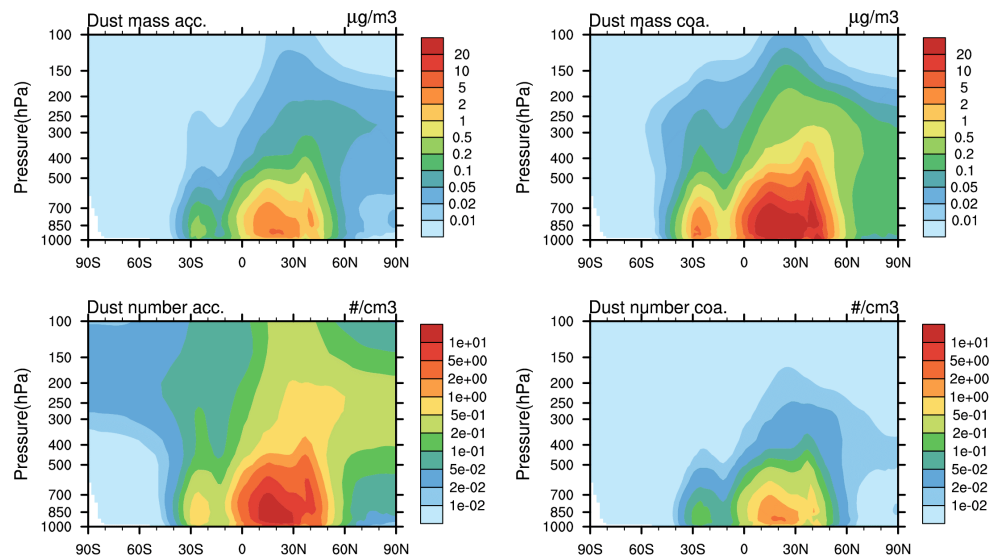
Fig. 1. Annual mean and vertical cross sections of mass (upper) and number (lower) concentrations of mineral dust in the accumulation mode (0.1–1 µm diameter, left) and in the coarse mode (1–10 µm diameter, right). Results are taken from the LP05 combined simulation and represent 5-yr averages.
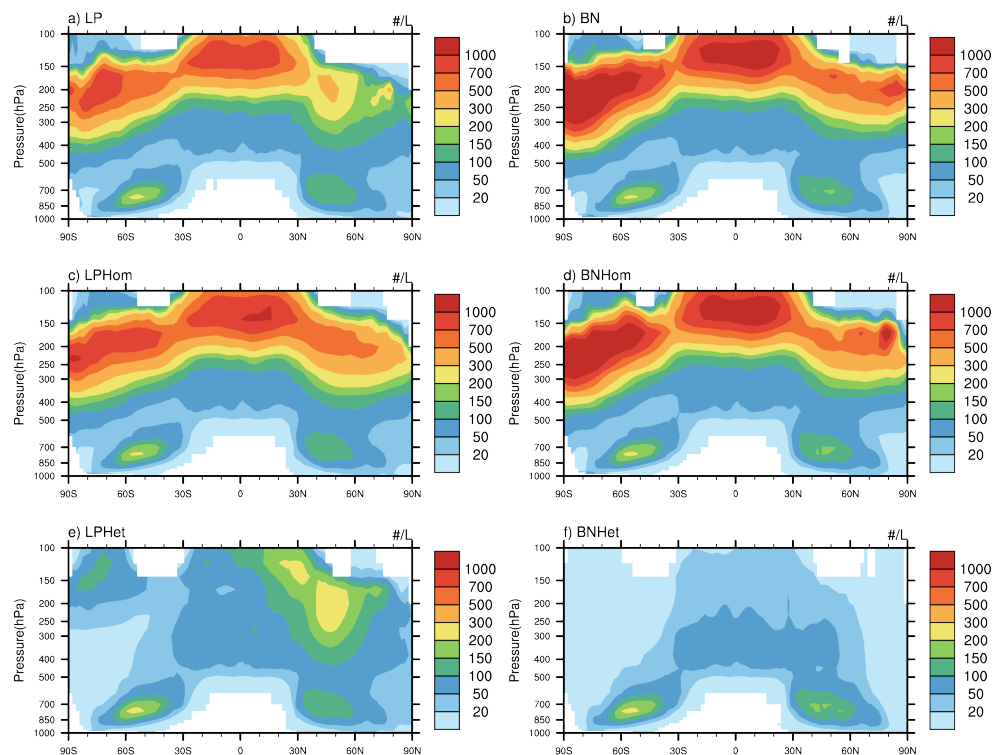
Fig. 2. Comparison of in-cloud ice crystal number concentration with LP05 (left) and BN09 (right) parameterizations for the combined homogeneous and heterogeneous nucleation (upper), pure homogeneous nucleation (middle), and pure heterogeneous nucleation (lower) simulations in CAM5.
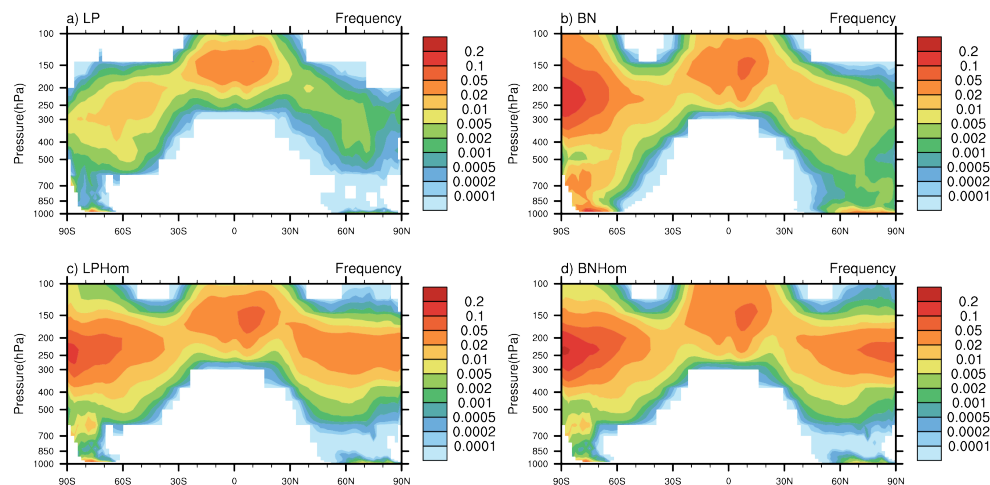
Fig. 3. Frequency distribution of homogeneous nucleation occurrence events in CAM5 for combined homogeneous and heterogeneous nucleation case (upper) and pure homogeneous nucleation case (lower) with LP05 (left) and BN09 (right) parameterizations.
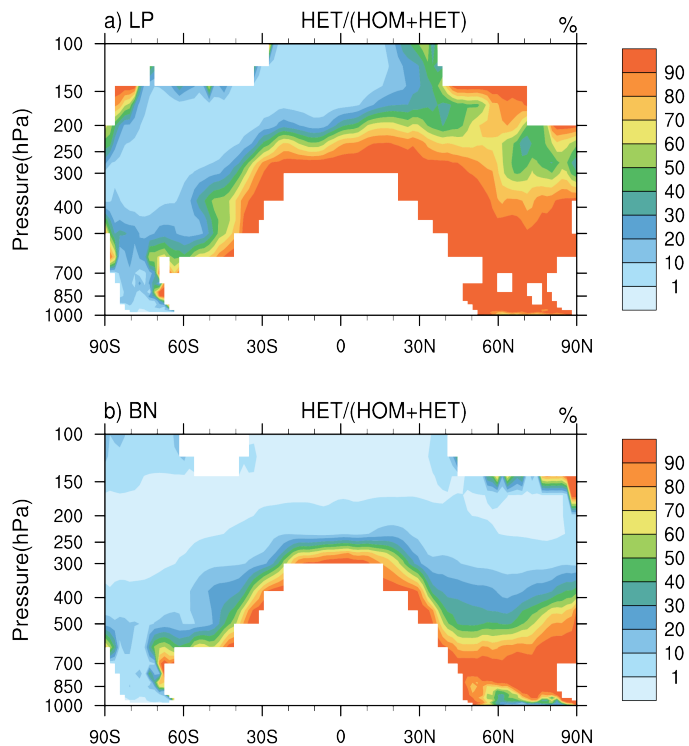
Fig. 4. Percentage contribution from heterogeneous ice nucleation to total ice crystal number concentration diagnosed in the ice nucleation parameterizations for the combined LP and BN cases.
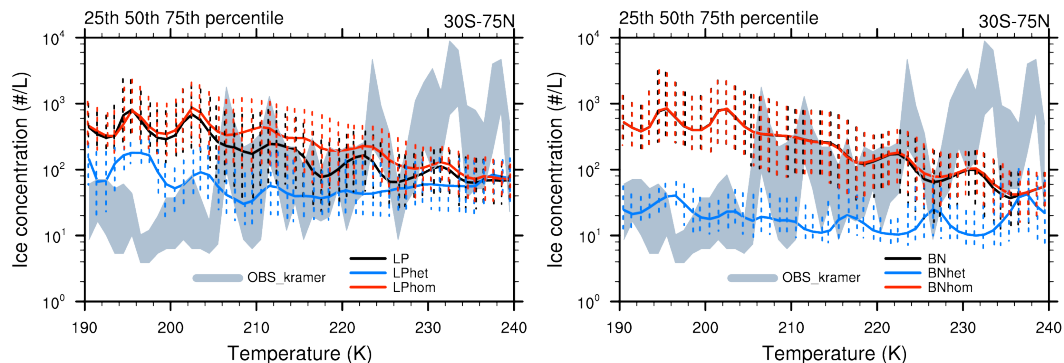
Fig. 5. In-cloud ice crystal number concentration (l^{-1}) versus temperature. Model results are sampled every 3 h over tropical, mid-latitude and Arctic regions including the observation locations reported in Krämer et al. (2009). The 50 % percentile (solid line), 25 % and 75 % percentiles (error bar) are shown for each 1 K temperature bins.
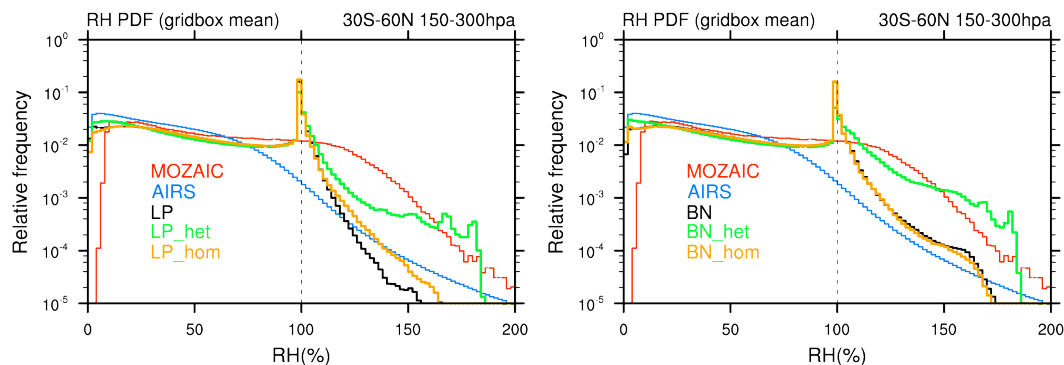
Fig. 6. Probability distribution frequency (PDF) of relative humidity in 200–500 hPa from 30° S to 60° N from pure homogeneous, pure heterogeneous and combined nucleation simulations for LP05 (left) and BN09 (right) parameterizations, in comparison with humidity observations in the upper troposphere from the Measurements of Ozone, Water Vapor, Carbon Monoxide and Nitrogen Oxides by Airbus In-Service Aircraft (MOZAIC) and from the Atmospheric Infra-Red Sounder (AIRS) satellite.

Title Page

Abstract   Introduction

Conclusions   References

Tables   Figures

◄   ►

Back   Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
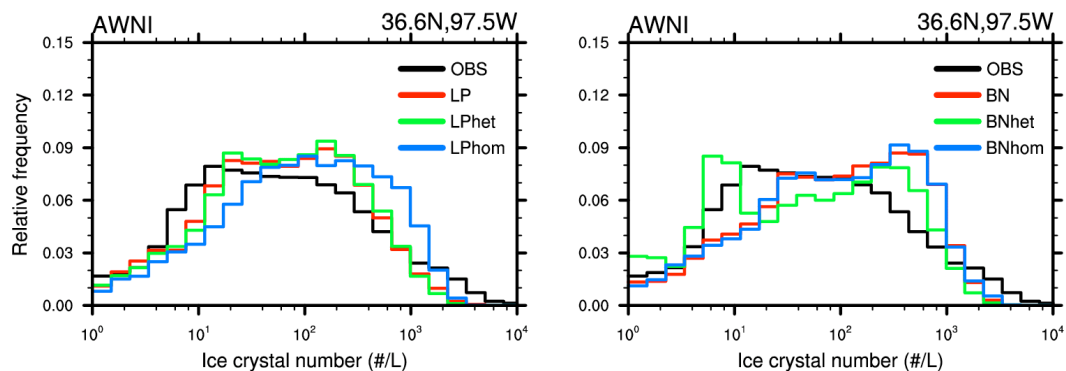
Fig. 7. Probability distribution frequency (PDF) of in-cloud ice number concentration (l^{-1}). Model results are sampled every 3 h and interpolated into the field measurement site location (36°36′ N, 97°29′ W). All data are sampled over 7 km.
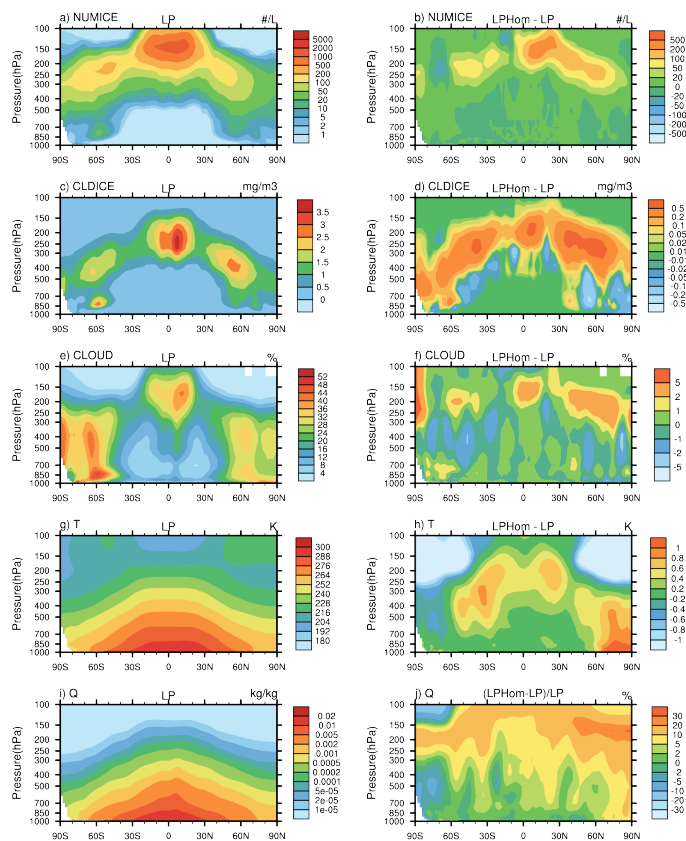
Fig. 8. Latitude-pressure cross sections of annual mean Ni (grid-box average), IWC (grid-box average), cloud cover, temperature, and specific humidity from combined ice nucleation simulation (left) and differences of these variables between pure homogeneous and combined nucleation simulations (right) for the LP05 parameterization in CAM5.
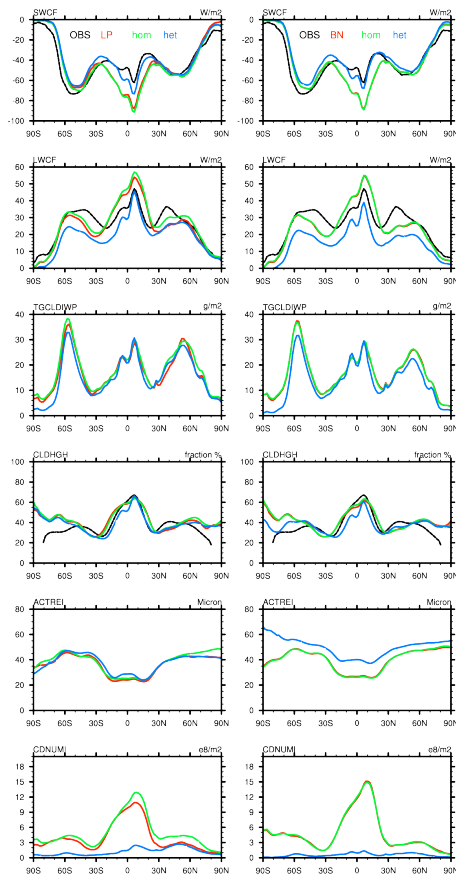
Fig. 9. Annual and zonal mean distributions of shortwave cloud forcing (SWCF), longwave cloud forcing (LWCF), ice water path (IWP), high cloud cover (CLDHGH), cloud-top ice effective radius (ACTREI), and column cloud ice number concentration (CDNUMI) from different model simulations with LP05 (left) and BN09 (right) parameterizations.