Immersion freezing by natural dust based on a soccer ball model with the Community Atmospheric Model version 5: climate effects

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Abstract

We introduce a simplified version of the soccer ball model (SBM) developed by Niedermeier et al (2014 Geophys. Res. Lett. 41 736–741) into the Community Atmospheric Model version 5 (CAM5). It is the first time that SBM is used in an atmospheric model to parameterize the heterogeneous ice nucleation. The SBM, which was simplified for its suitable application in atmospheric models, uses the classical nucleation theory to describe the immersion/condensation freezing by dust in the mixed-phase cloud regime. Uncertain parameters (mean contact angle, standard deviation of contact angle probability distribution, and number of surface sites) in the SBM are constrained by fitting them to recent natural dust (Saharan dust) datasets. With the SBM in CAM5, we investigate the sensitivity of modeled cloud properties to the SBM parameters, and find significant seasonal and regional differences in the sensitivity among the three SBM parameters. Changes of mean contact angle and the number of surface sites lead to changes of cloud properties in Arctic in spring, which could be attributed to the transport of dust ice nuclei to this region. In winter, significant changes of cloud properties induced by these two parameters mainly occur in northern hemispheric mid-latitudes (e.g., East Asia). In comparison, no obvious changes of cloud properties caused by changes of standard deviation can be found in all the seasons. These results are valuable for understanding the heterogeneous ice nucleation behavior, and useful for guiding the future model developments.

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1. Introduction

In mixed-phase clouds, the primary ice formation is via heterogeneous ice nucleation with participation of some special types of aerosol particles (e.g., mineral dust, soot and biological particles) through serving as ice nuclei (IN) (Hoose and Möhler 2012). Mineral dust and soot particles often induce
ice nucleation at temperatures lower than $-10^\circ C$, while biological particles like bacteria can induce ice nucleation above $-10^\circ C$ (e.g., Hartmann et al 2013). In order to represent the heterogeneous ice nucleation process in atmospheric models, heterogeneous ice nucleation parameterizations have been developed on the basis of either singular (or deterministic) approach or stochastic approach.

The ‘singular approach’ neglects thermal fluctuations and assumes the instantaneous ice nucleation (i.e., without considering time dependence) when temperature reaches the freezing point (Phillips et al 2008, DeMott et al 2010, Vali 2014). The second one, ‘stochastic approach’ described by Bigg (1953), conversely holds that the heterogeneous ice nucleation behavior is time-dependent, implying that the ice nucleation is a time evolution process (Vali 1994, Hoose et al 2010, Wang et al 2014, Herbert et al 2014). In order to bridge these two seemingly contradictory extremes, Niedermeier et al (2011) introduced a soccer ball model (SBM), which applied the classical nucleation theory (CNT), to explore the transition from stochastic behavior to singular behavior of ice nucleation process by changing the three parameters in SBM (i.e., mean contact angle, standard deviation of contact angle probability distribution, and number of surface sites). Lüönd et al (2010) and Ervens and Feingold (2012) utilized a probability density function of contact angles ($\theta$-PDF) model and an active site model to combine the key features of these two hypotheses regarding the heterogeneous ice nucleation. The $\theta$-PDF and active site models as they applied, to some extent, can be understood as two special cases of the SBM.

Large-scale atmospheric models are now implementing ice nucleation parameterizations that link either IN number concentration or ice nucleation rate to the aerosol properties in order to treat aerosol-cold cloud interactions. Hoose et al (2010) introduced a CNT-based scheme of heterogeneous ice nucleation by mineral dust, soot, and biological particles in the Community Atmosphere Model version 3 (CAM3)-Oslo. However, this heterogeneous ice nucleation scheme is based on a single contact angle model so that it may overestimate the stochastic behavior of heterogeneous ice nucleation. Wang et al (2014) implemented a $\theta$-PDF model in CAM5 to consider the singular behavior of heterogeneous ice nucleation by allowing the variation of contact angle. The $\theta$-PDF model still assumes that each particle surface is homogeneous (with the same contact angle) and surface properties differ only among different particles. Thus, the effects of the inhomogeneity of each particle surface on heterogeneous ice nucleation can not be taken into account. More recently, Niedermeier et al (2014) developed a simplified version of SBM to increase its computational efficiency because the original SBM developed by Niedermeier et al (2011) uses the Monte Carlo technique, which hampers its application in large-scale atmospheric models due to the computational costs. We note that the SBM is a unified contact angle ($\theta$) model, which encompasses the single-$\theta$, $\theta$-PDF and the active site model. The $\theta$-PDF model is a simplified version of the SBM by assuming number of surface sites to be 1. Therefore the implemented SBM in CAM5 allows us to investigate the effect of number of surface sites.

In this study, we make the first to introduce the SBM (Niedermeier et al 2014) in a start-of-the art climate model CAM5 and to explore the impacts of parameters in the SBM on simulated mixed-phase clouds. The effects of singular versus stochastic approach of heterogeneous ice nucleation on cloud properties and climate are investigated via changing the SBM parameters. The CAM5 model is briefly discussed in section 2. Section 3 presents CAM5 results and sensitivity of cloud properties and climate to the SBM parameters. Discussion and conclusions are given in section 4.

2. CAM5

A two-moment stratiform cloud microphysics scheme (Morrison andGettelman 2008 (MG08), Gettelman et al 2010) is used in CAM5. Number concentrations and mass mixing ratios of cloud droplets and ice crystals are predicted. The treatment of ice nucleation in cirrus clouds follows Liu et al (2007). In mixed-phase clouds, deposition nucleation, immersion and condensation freezing are parameterized with Meyers et al (1992), which does not provide a link to IN properties. The contact freezing of cloud droplets by coarse mode dust is represented based on Young (1974).

In order to represent aerosol processes and properties in the atmosphere, a modal aerosol module (Liu et al 2012) is implemented in CAM5. This aerosol module treats several major aerosol species (including mineral dust and black carbon (BC)), which are internally mixed in a single mode but externally mixed between different modes. The number concentration of aerosol in different modes and mass mixing ratios of aerosol species in these modes are predicted. Other parameterization schemes in CAM5 include a deep convection scheme following Zhang and McFarlane (1995), and a shallow convection scheme following Park and Bretherton (2009).

3. Results

3.1. Representation of immersion/condensation freezing by natural dust in CAM5

The representation of immersion/condensation freezing by natural dust in the updated CAM5 uses the SBM based on Niedermeier et al (2014), to replace the Meyers et al (1992) scheme. In the SBM, there are three uncertain parameters that need to be constrained by the observation data. They are mean contact angle ($\mu_\theta$), standard deviation of contact angle probability distribution ($\sigma_\theta$), and the number of surface sites ($n_{\text{surf}}$). Here, we use the observation data of Saharan dust to determine these three uncertain parameters. Compared to pure minerals (i.e., kaolinite, illite and montmorillonite) or commercially available Arizona Test Dust, Saharan dust, which is an internal mixture of different minerals (e.g., quartz, feldspars and clay minerals), more physically represents the
heterogeneous freezing ability of dust in the atmosphere (Hoose and Möhler 2012).

We constrain the SBM (Niedermeier et al. 2014) with the Colorado State University Continuous-Flow Diffusion Chamber (CFDC)-HAPER version I (CSU CFDC-IH) experiments with relative humidity with respect to water (RHw) of 106% and 108% (hereafter, CSU106 and CSU108) and the Zurich Ice Nuclei Chamber (ZINC) experiments with RHw of 106%, 108% and 110% (hereafter ZINC106, ZINC108, and ZINC110) (DeMott et al. 2011). Best-fit parameters are obtained by the least square method in these five experiments for the active fractions as a function of temperature, and listed in table 1. As seen from table 1, values of the mean contact angle ($\mu_\theta$) and the standard deviation ($\sigma_\theta$) in all experiments are same as those in Wang et al. (2014), which constrained the $\theta$-PDF model with the same observation datasets. We note that the number of surface sites ($n_{site}$) is smaller than 1 with CSU106 and ZINC110, which is reasonable (Niedermeier et al. 2014). The physical meaning of $n_{site}$ smaller than 1 is that not every particle features a site. The fit curves with these five experiments can be found in the supplementary figure S1. In addition, when we implement the SBM in CAM5, we apply a cut off of 0 for the active fractions at temperatures higher than $-10^\circ$C for the immersion/condensation freezing by dust. The reason is that Saharan natural dust is reported in recent CFDC observations to have onset temperatures ranging from about $-10^\circ$ to $-15^\circ$C (Phillips et al. 2012, Hoose and Möhler 2012).

### Table 1. Fit parameters obtained for the SBM model for the immersion/condensation freezing by natural dust. The root mean square errors (RMSE) between the fit curves and the data are given.

| Observation | $\mu_\theta$ ($^\circ$) | $\sigma_\theta$ | $n_{site}$ | RMSE |
|-------------|------------------------|----------------|------------|------|
| CSU106      | 46.0                   | 0.01           | 0.3        | 0.023|
| CSU108      | 47.0                   | 0.01           | 1.7        | 0.223|
| ZINC106     | 62.0                   | 0.04           | 2.22       | 0.076|
| ZINC108     | 61.0                   | 0.01           | 1.32       | 0.073|
| ZINC110     | 59.0                   | 0.02           | 0.76       | 0.083|

3.2. CAM5 results

We use the values of three parameters ($\mu_\theta$, $\sigma_\theta$, and $n_{site}$) constrained with CSU106 for the SBM to parameterize the immersion/condensation freezing in the updated CAM5. The parameterizations for the contact freezing and deposition nucleation have been updated based on Hoose et al. (2010) and Wang et al. (2014). To explore the effects of uncertainties

3.2.1. Evaluation of modeled IN concentrations with CFDC observations. To evaluate the SBM in CAM5, we utilize the observed IN concentrations from the CFDC (Rogers et al. 2001) to compare with model results. To be consistent with the CFDC operation principles, which collect clear-sky aerosol particles with an aerosol inlet and measure the number of ice crystals formed in the chamber at a given temperature or RHw with a residence time of 5–20 s, we diagnose IN concentrations in 10 s (hereafter, IN(10 s)) in immersion/condensation freezing with the input of online modeled aerosols. RHw of 100% is used for the immersion/condensation freezing.

Figure 1 shows the comparison of modeled IN number concentrations, IN(10 s) from the control simulation with CFDC observations. The observations describe the
Table 2. Global and seasonal mean liquid water path (LWP, g m\(^{-2}\)) for simulations. MAM is for March–April–May, JJA for June–July–August, SON for September–October–November, and DJF for December–January–February.

| Simulation | Description | MAM | JJA | SON | DJF |
|------------|-------------|-----|-----|-----|-----|
| CTL        | Updated CAM5 With the SBM and nudged winds, using \(\mu_\theta = 46.0^\circ\), \(\sigma_\theta = 0.01\), \(n_{\text{site}} = 0.3\) | 45.12 | 52.06 | 48.21 | 46.99 |
| MU1        | As in CTL, but with \(\mu_\theta = 36.0^\circ\) | 45.02 | 52.06 | 48.16 | 46.93 |
| MU2        | As in CTL, but with \(\mu_\theta = 26.0^\circ\) | 44.91 | 51.98 | 48.02 | 46.80 |
| MU3        | As in CTL, but with \(\mu_\theta = 16.0^\circ\) | 44.89 | 51.96 | 48.02 | 46.77 |
| MU4        | As in CTL, but with \(\mu_\theta = 56.0^\circ\) | 45.15 | 52.08 | 48.25 | 47.02 |
| SD1        | As in CTL, but with \(\sigma_\theta = 0.02\) | 45.21 | 52.12 | 48.32 | 47.12 |
| SD2        | As in CTL, but with \(\sigma_\theta = 0.05\) | 45.18 | 52.12 | 48.30 | 47.11 |
| SD3        | As in CTL, but with \(\sigma_\theta = 0.05\) | 45.12 | 52.07 | 48.20 | 47.00 |
| NS1        | As in CTL, but with \(n_{\text{site}} = 3\) | 44.97 | 51.98 | 48.08 | 46.84 |
| NS2        | As in CTL, but with \(n_{\text{site}} = 30\) | 44.91 | 51.99 | 48.03 | 46.79 |
| NS3        | As in CTL, but with \(n_{\text{site}} = 300\) | 44.92 | 52.00 | 47.97 | 46.77 |

Figure 2. Zonal and annual mean immersion/condensation freezing rates in different sensitivity simulations. Isotherms of 0 °C and −37 °C are plotted.
relationships between IN number concentrations and number concentrations of aerosols with diameter larger than 0.5 μm (Na500), as IN number concentrations were found to be correlated well with the number concentrations of coarse-mode aerosols (Georgii and Kleinjung 1967, DeMott et al 2006). In CAM5, we calculate Na500 following Wang et al (2014), diagnose IN(10 s) at multiple temperatures (−21 °C used in figure 1(a) and −27°C in figure 1(b)), and output Na500 and IN(10 s) at all model grids. It can be seen that the modeled IN(10 s) versus aerosol relationships at the two temperatures are in good agreement with observations, especially with DeMott et al (2014) which is developed specifically for ice nucleation by mineral dust.

3.2.2. Sensitivity of modeled cloud properties to the SBM parameters

Different values of uncertain parameters in the SBM will influence the freezing rate (ΔN/Δt, here ΔN, is the change of ice crystal number concentration due to the immersion/condensation freezing over one model time step Δt). In figure 2, we show the immersion/condensation freezing rates in nine SBM sensitivity simulations. The immersion/condensation freezing rates in the CTL and MU4 simulations can be found in the supplementary material. The nonzero immersion/condensation freezing rates in the CTL and MU4 simulations can be found in the supplementary material. The non-zero immersion/condensation freezing rates at T>0 °C and T<−37 °C are due to the zonal and annual averaging. With decreasing mean contact angle (from left to right in the upper panel), the immersion/condensation freezing rate increases, as expected. When the standard deviation is increased from 0.02 to 0.5 (from left to right in the middle panel), no obvious changes of the immersion/condensation freezing rate can be found, which can be attributed to the flattening of active fraction’s dependence on temperature with increasing the standard deviation (see the explanation below). When the number of surface sites changes from 3 to 300 (from left to right in the lower panel), the freezing rate increases correspondingly, which is consistent with Niedermeier et al (2011).

Sensitivity of cloud properties (e.g., liquid water path (LWP)) to the SBM parameters in the four seasons is shown in figure 3. In order to see the changes more clearly, we compare the MU3, SD3, and NS3 simulations with the CTL simulation. In spring (March–April–May, MAM), decrease of the mean contact angle and increase of the number of surface sites lead to large decrease of LWP in high latitudes of the northern hemisphere (NH) (e.g., Arctic) by up to 3 g m−2. It is because that larger freezing rates in MU3 and NS3 (see figure 2) result in more ice crystals and thus larger ice water path (IWP), which causes the decrease of the LWP. Compared to other seasons, the largest changes in Arctic found in spring can be due to the transport of dust ice nuclei to these regions in spring (Wang et al 2011, Liu et al 2012). These changes to the LWP in the Arctic in spring will affect the cloud radiative properties and surface energy balance, and thus lower-tropospheric stability and surface temperature, which can further influence the Arctic sea ice simulations in the model (Xie et al 2013, Barton et al 2014). In winter (December–January–February, DJF), obvious decreases of LWP caused by changes of mean contact angle and number of surface site occur in NH mid-latitudes where large amounts of mixed-phase clouds exist. The changes to the LWP in the NH mid-latitudes in winter will affect the winter storm tracks (Wang et al 2014) and will also lead to changes in downward longwave radiation at the surface and thus result in changes of nighttime and diurnal surface temperature over East Asia.
Changes of LWP caused by standard deviation are smaller in all seasons. The explanation is given as follows. The changes of the standard deviation can lead to changes of the temperature range in which droplets freeze. If increasing the standard deviation, it inevitably leads to the beginning of nucleation at higher temperature caused by an increase of the occurrence of smaller contact angles and the end of nucleation at lower temperature as well as caused by an increase of the occurrence of larger contact angles. Therefore, when comparing with the smaller standard deviation, the larger standard deviation will result in two different changes of the frozen fractions. As shown in figure 1 in Niedermeier et al. (2014), with comparison of standard deviation $\sigma_\theta$=0.1 and 0.5, changing $\sigma_\theta$ from 0.1 to 0.5 results in an increase of the frozen fraction at $T>-20$ C but a decrease of the frozen fraction at $T<-20$ C. Therefore, these two different signs of changes of frozen fractions in two temperature ranges with changes of the standard deviation will offset each other in mixed-phase clouds and thus results in the least influence on seasonal mean cloud properties. We note that using a cloud model, Kulkarni et al. (2012) also found that the standard deviation of the contact angle distribution with the $\theta$-PDF approach (they made changes of the standard deviation from 0.14 to 0.38) in the cloud-resolving model has the least influence on cloud properties. In addition, no obvious changes of LWP in summer (June–July–August, JJA) could be found for all the three parameters.

4. Discussion and conclusions

In this study, the SBM is implemented in a global climate model to represent the heterogeneous immersion/condensation nucleation by natural dust in mixed-phase clouds. The three uncertain parameters (mean contact angle, standard deviation of contact angle probability distribution, and number of surface sites) in the SBM are constrained with observation data. The modeled IN(10 s) concentrations in CAM5 correlate well with large-size aerosol number concentrations, in agreement with observations.

Sensitivities of modeled cloud properties to the three parameters in SBM are investigated. The findings from this study are new and significant: changes in the surface site number and mean contact angle have a noticeable effect on cloud properties (e.g., cloud LWP) depending on latitudes and seasons, while changes in the standard deviation does not cause significant effects in all seasons. These results are valuable for understanding the heterogeneous ice nucleation behavior, and useful for guiding the future model development.

Table 2 gives the global mean LWP for all simulations in the four seasons (other cloud properties, e.g., IWP, shortwave and longwave cloud forcings can be found in the supplementary tables S1, S2 and S3, respectively). With the change of mean contact angle, the global mean LWP in spring changes by the range of $-0.23$ to $0.03$ g m$^{-2}$. With the change of number of surface sites, the global mean LWP in spring changes by $-0.20$ to $-0.15$ g m$^{-2}$. While in winter, the global mean LWP changes by the range of $-0.22$ to $-0.15$ g m$^{-2}$ with the change of mean contact angle and by $-0.22$ to $-0.15$ g m$^{-2}$ with the change of number of surface sites. In summer, mean contact angle and the number of surface sites have no significant effects on the global mean LWP. Comparing to mean contact angle and the number of surface sites, change of the standard deviation generally has the least influence on the global mean LWP for all the four seasons.

There are two points regarding the application of CNT-based ice nucleation schemes (e.g., SBM) to large-scale models with long model time steps (e.g., 30 min in CAM5). The first one is about the time dependency of CNT. As shown in Wang et al. (2014) and Ervens and Feingold (2013), the active fraction of aerosols to form ice crystals is several orders of magnitude less sensitive to time compared to the other key parameters in CNT (temperature, contact angle, and particle size). Sensitivity studies performed in Ervens and Feingold (2013) showed that a change in temperature of $\sim$1 K causes a similar impact on the active fraction as a change in contact angle of $\Delta \theta = 2^\circ$, whereas a similar change is caused by an increase of time by three orders of magnitude. Second, CAM5 does not consider the aerosol loss due to the droplet freezing. However, this assumption only causes a small artifact on model results as discussed in Wang et al. (2014).

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