Author comments for referee #1 and referee #2

Reply to the comments by anonymous referee #1

We appreciate the referee's valuable comments.

General comments:

Scientific Significance:

The manuscript investigates if using a PDF of contact angles is a better approach for the parameterization of freezing as compared to using a single contact angle in CNT. While many measurements show that this approach could be more appropriate is it not clear that this also applies in a global climate model (GCM) with a 30-minute timestep.

Reply: see our reply below for the time dependence of CNT, and the applicability of CNT in a GCM with a 30-min time step. We would like to note here that the approach of a PDF of contact angles has a less time dependency than the single contact angle approach in CNT.

Scientific Quality:

The research question is very interesting, especially concerning the parameterization. The study of the importance of the specific freezing pathways is relevant and plausible to follow. However, concerning the parameterization, the manuscript lacks at some points carefully thinking. The approach and the applied methods may not be suitable in the context of a GCM. The discussion of the results sometimes misses this critical reflection and consequences of certain interpretations are sometimes difficult to understand if not missing.

Reply: see our reply below for the applicability of CNT in a GCM with a 30-min time step.

Details:

- It seems rather difficult to study a time-dependence of a process without having subtimestepping. What are the consequences resulting from treating the time dependencies of the frozen fraction in a timestep of 30 minutes? You should add sensitivity tests to check whether it is appropriate to use such a crude time-resolution. Does it make sense to study an intermediate approach between singular approach and CNT if the timestep cannot resolve the CNT behavior?

Reply: Following the reviewer's comment, we performed sensitivity tests to check whether it is appropriate to use a classical-theory-based parameterization with a crude time step of 30 minutes in the CAM5 model.

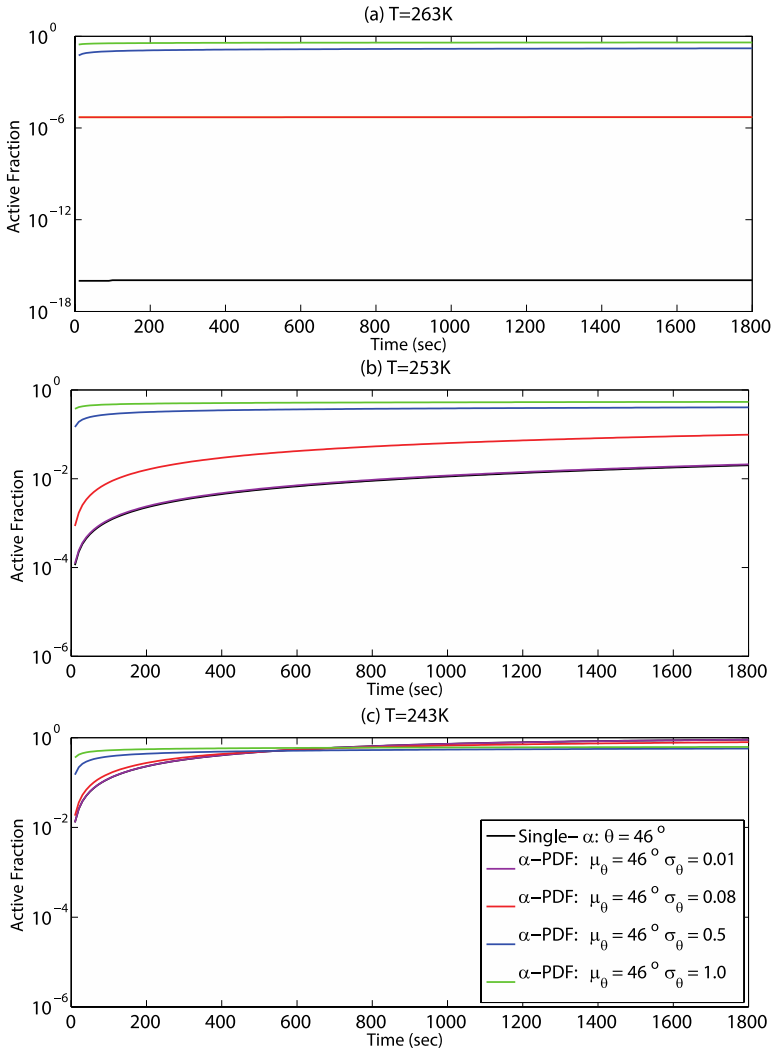

Figure S1. Calculated change in the active fraction with time at different temperatures for 300nm monodisperse particles and for different contact angle distributions.

Figure S1 shows the active fraction with different contact angle distributions as a function of integration time at different temperatures. It can be seen that at T = 263 K the active fractions in all contact angle distributions are almost constant with time, indicating very weak dependence of the active fraction on time at warm temperature (the active fraction in the α-PDF model with σ = 0.01 is about 0.499×10^{-5} and the active fraction in the α-PDF model with σ = 0.08 is about 0.516×10^{-5}, so these two lines are overlapped). At T = 253 K and T = 243 K, the active fractions in the single-α model and the α-PDF

models with σ = 0.01 and 0.08 increase with time (the α-PDF model with σ = 0.01 is very similar as the single-α model), but the active fraction in the α-PDF model with σ = 0.08 is a little insensitive to time than that in the single-α model. With increasing standard deviation in the α-PDF model, the active fractions become weaker dependent on time, especially for the weakest time dependence with σ = 1.0. As the single-α model can be thought as the special α-PDF model with σ = 0, and increasing the standard deviation reduces the time dependence, we can conclude the single-α model has stronger time dependence than the α-PDF model, which is consistent with Welti et al., 2012. Although the single-α model has stronger time dependence, if we use the following diagnostics, originally developed by Ervens and Feingold (2013), to determine the sensitivity of the active fraction to time in detail, we will find the active fraction in the single-α model is still only weakly dependent on time.

$$S(X) = \frac{\partial P}{\partial lnX} \qquad (1)$$

where P is the active fraction, X can be any of parameters from temperature, particle size, contact angle or time. Here, we only consider X is time. At T = 253 K, from t = 10s (P = 0.00011) to t = 1800s (P = 0.02), $S(X)$ is 0.0038. At T = 243K, from t = 10s (P = 0.013) to t = 1800s (P = 0.9044), $S(X)$ is 0.172. The very small values of $S(X)$, which are consistent with the values in Ervens and Feingold (2013), indicate that the active fraction is the least sensitive to time. Ervens and Feingold (2013) performed many sensitivity tests to investigate relative importance of temperature, particle size, contact angle and time for classical nucleation theory. They used Eq. (1) to explore the sensitivity of the frozen fraction to the above four parameters. From Figure 1(a) to Figure 1(d) of their paper, they found from comparison of $S(X)$ that among the four parameters P is the least sensitive to time. Ervens and Feingold (2013) concluded that a change in T (temperature) of ~1 K has a similar impact on P (the active fraction) as θ (contact angle) changes of $\Delta\theta = 2^o$ whereas a similar change is only caused by an increase in D_{IN} (particle diameter) by one

order of magnitude or in *t* (time) by three orders of magnitude. They hence suggested that it seems feasible to develop more physically (CNT) based relationships instead of those empirically based relationships in large-scale models. Therefore, the overestimate of the frozen fraction due to a crude time step of 30 min is negligible compared to the uncertainties in temperature and mean contact angle.

- Using a contact angle distribution leads to freezing of the smallest contact angles first. In the following timestep these contact angles should not be available a second time. This would mean that one would either need to track the contact angle with time in the model or need to implement a time dependent contact angle distribution. Not doing so might only shift the contact angle to smaller sizes and enables freezing at higher temperatures. Instead depletion of small contact angles should slow down the freezing process. This is at the moment not represented in the model. Without any sensitivity studies showing that it is appropriate to use the same contact angle distribution every time step it seems not reasonable to do so. Please add sensitivity studies or explanations why this assumption is justified.

Reply: As described in p 7149 line 23 – p 7150 line 7, during each model time step (30 minutes) cloud borne aerosols will be updated, which means that fresh particles as cloud condensation nuclei (CCN) will be nucleated into cloud droplets. These newly formed cloud droplets containing cloud borne aerosols will be used as input for the immersion freezing in the current time step and make up the nucleated particles in the last time step to keep the distribution of contact angles unchanged. As is shown in Fig. S1, at T = 263 K and T = 253 K, the active fractions in the single-α model and the α-PDF model with σ = 0.01 are much smaller than 100%. In the subsequent time step, newly-formed cloud droplets are sufficient to make up those depleted amount of cloud droplets. What is more, after the Wegener-Bergeron-Findeisen process sets in, further ice nucleation will be

suppressed. Overall, In this case we actually benefit from the long time step because the clouds and the environmental conditions change significantly from time step to time step, such that starting from fresh is not a bad assumption. In particular, entrainment of new IN, temperature changes and the shutdown of ice nucleation through the Bergeron process are thought to be important. Therefore, based on the above explanation, assuming a constant distribution of contact angles among time steps only causes a small artifact. This artifact at high temperatures would be smaller than that at low temperatures. Another point is that new parameterizations implemented in this study reduce nucleated ice crystals compared to the default Meyers et al. scheme (see Table 4 in our paper), which means that the depletion of cloud-borne aerosols has a smaller effect on model results than the default scheme.

- Looking at the fit results (Table 2) the differences are not so fundamental between the single-alpha and the alpha-PDF. The error in between the measurements (for example CSU106 and CSU108) is larger. The argumentation why the alpha-PDF has smaller RSMEs is not plausible when looking at Fig. 1 because no noticeable difference can be seen between both curves in the temperature range where the data points are. I suggest adding error bars to the data points and to revisit the argumentation.

Reply: Since CFDC cannot provide observation data at warm temperatures (>-15^{o}C) and the number of data points for CSU106 and ZINC106 is 5 and 3 respectively, significant differences between the single contact angle model and alpha-PDF model are hard to see, as the reviewer mentions. However, some differences can still be found. With the increasing temperature, the active fraction predicted by the single contact angle model decreases much more rapidly than that predicted by the alpha-PDF model, and the alpha-PDF model agrees with the observation data better at warm temperatures. Larger errors in the single contact angle model compared to those in the alpha-PDF model are

mainly from the warmest data points (CSU106 for T=-18.5^{o}C and ZINC106 for T=-27.7^{o}C). Unfortunately, the errors associated with observation data are not available. RMSEs in CSU108 are much larger than those in CSU106 may be from the larger uncertainty of the data point at T = -25.3 K in CSU108. Due to few data points in CSU108 and the smallest RMSEs in CSU106, we use the CSU106 fit results to do the simulation in CAM5.

- *The fit for the alpha-PDF does not seem reasonable because the frozen fraction is not approaching 0 at the warmest temperatures. This is physically wrong and will cause some mistakes in the calculation of IN concentrations especially at the warmest temperatures.*

Reply One reason for the non-zero of frozen fraction at warmest temperatures is that even at warmest temperatures, some particles with smallest contact angles (although their number concentrations are very low) still can nucleate. Small mean contact angle and large standard deviation can produce the tail of the distribution with a contact angle of 0^{o} or very close to 0^{o}. With the increasing standard deviation, this phenomenon can become more obvious (see Figure S1 for T = 263 K). In Niedermeier et al. (2014) the frozen fraction (when the number of surface sites (n_{site}) equals 1, the soccer ball model is the same as the alpha-PDF model) is obviously not 0 at very warmest temperatures with largest standard deviation (see sold line (n_{site}=1) in Figure 1 in their paper). The other reason is that at warmest temperatures the frozen fraction will become insensitive to the temperature change in the alpha-PDF model (Ervens and Feingold, 2013). Thus the active fraction will not decrease obviously with the increase of temperature. With comparison of Figure 1(a) and Figure 1(e) in Ervens and Feingold (2013) when introducing the alpha-PDF model, activation of particles with the smallest contact angles results in this phenomenon at warmest temperatures.

To address this non-physical behavior with the alpha-PDF model at the warmest temperatures for dust and soot, we have applied a cut off of 0 for the active fractions at temperatures larger than -10 ^{o}C, repeated all the simulations and updated our results in the revised manuscript. The results from the alpha-PDF model change slightly, and the main conclusions remain the same.

- If in the case of the alpha-PDF parameterization freezing starts already earlier (at small contact angles) it is reasonable that freezing occurs at lower altitudes. Please check if this is a physical phenomenon or due to the too high frozen fractions at the warmest temperatures (see above). Please verify.

Reply: As indicated previously, we have applied a cut off of 0 for the active fraction at temperatures larger than -10 ^{o}C for all our simulations and updated our results in the revised manuscript.

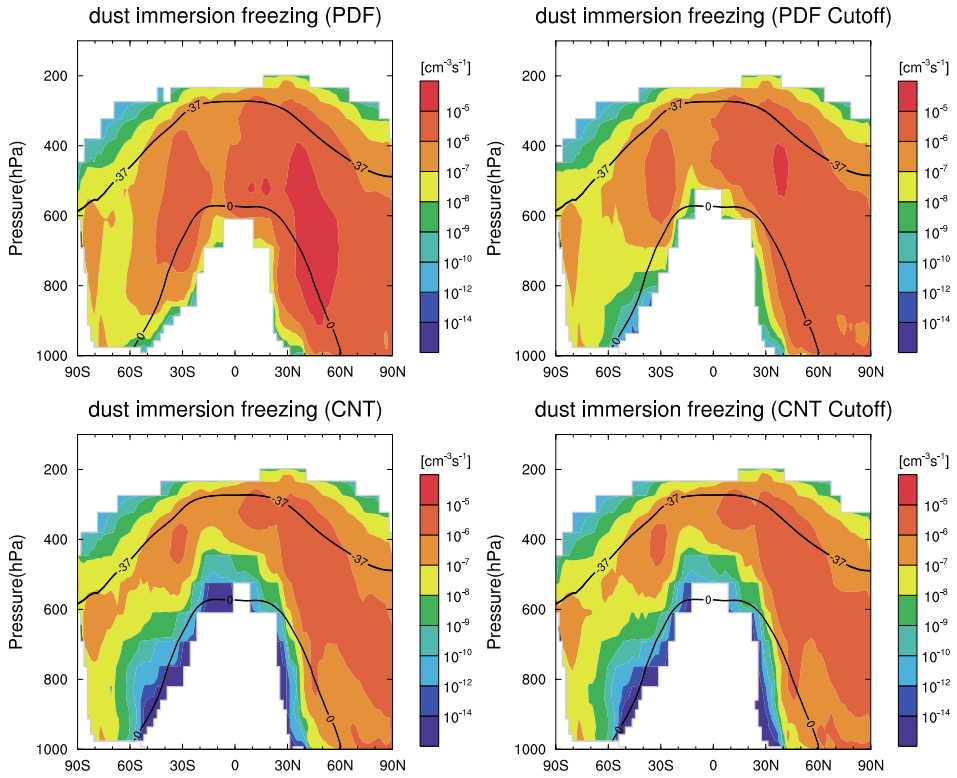

Figure S2. Zonal and annual mean immersion freezing rates in different simulations. Isotherms of 0 ℃ and -37 ℃ are plotted

Figure S2 shows the comparison of four simulations (the PDF simulation without/with a cutoff and the CNT simulation without/with a cutoff). We can see the freezing rate in the PDF simulation with a cutoff at warm temperatures decreases compared to that in the PDF simulation without a cutoff. As the reviewer suspected, the reason is the unphysically high active fraction at temperatures warmer than -10 ^{o}C in the original simulations. However, compared to the CNT simulations, the main increase of the freezing rate in the PDF simulation with a cutoff still locate at low latitudes (with warm temperatures) although this increase is not as pronounced as before. It can be seen from Fig. 8 in Welti et al. (2012) paper that in the case of the alpha-PDF parameterization freezing starts already earlier (at small contact angles) and decreases much more slowly with increase of temperature, which causes the freezing occurs at low altitudes. A larger standard deviation can lead to earlier freezing and a slower increase of the freezing rate with decreasing temperature (see Figure 9(b) in our paper or Figure 6 in Niedermeier et al. 2014). The larger standard deviation means higher frequency of smaller contact angles, which induces earlier freezing and a higher freezing rate at warmest temperatures. The single contact angle model can be seen as a special alpha-PDF in which the standard deviation equals 0. Based on the result of the PDF simulation with a cutoff and the above explanation, we will use "due to a slower increase of the active fraction with decreasing temperature in the alpha-PDF model, the freezing rate in the PDF simulation is significantly higher at low altitudes compared to the CNT simulation" to replace the original conclusion in the revised version.

- Where does the freezing at T > -15°C originate from if you only consider freezing of dust and BC as shown in Fig. 7? Plotting it like this does not make sense- the frequency

of freezing has to be evaluated per temperature-bin, using an annual mean temperature does not make sense.

Reply: Niemand et al. (2012) reported the Aerosol Interactions and Dynamics in the Atmosphere (AIDA) cloud chamber measurement of natural dust at temperatures of -13 and -16 $^{\circ}$C with active fractions of 10^{-4} and 10^{-5}, which agree with our fitted active fractions from the alpha-PDF parameterization (Figure 1). Since there are no measured active fractions reported above -10 $^{\circ}$C, we set a cut off of 0 for active fractions at temperatures larger than -10 ^{o}C in all the new simulations, and updated all the figures (including Fig. 7).

Following the reviewer comment, we use 3-hourly outputs of temperature and frequency for plotting Fig. 7 instead of using an annual mean temperature, although we find that the results do not change much.

- The calculation of the IN(10s) concentrations in Section 4.5/Fig. 10 is not clear. Is the calculation done based on interstitial aerosol concentrations from the simulations at the same location and pressure as the measurements? Is the temperature then not taken out of the simulation but assumed to be the processing temperature of the measurement? If so is the relative humidity not taken into account and how is the information gained in which freezing modes the freezing occurred? Does your simulated aerosol concentration agree with the measured ones during the field measurements? What are the consequences of using the model results and comparing it to measurements with a resolution of 10 s, which is 180 times shorter than the model time step (if not calculated)?

Reply: As noted at beginning of Section 4.5, the interstitial aerosol concentrations from the simulations are used at the same location and pressure as the measurements. The

temperature is assumed to be the processing temperature of the instrument. In the same way, the relative humidity is assumed to be equal to the processing conditions in the instrument. It is assumed in our calculations that 100% of the relative humidity with respect to water (RH_w) is used for the immersion freezing, and 98% RH_w for deposition freezing. Thus immersion/condensation and deposition nucleation modes are taken into account (in Fig. 10 in our paper, we thus sum up immersion and deposition nucleation), which is consistent with the observed dominant ice nucleation modes in the CFDC. The time step of 10s was used in our calculations for a direct comparability to the observations.

Unfortunately, we do not have simultaneous aerosol measurements in these locations. In Fig. 12 we show the relationship of the IN concentrations and aerosol number concentrations with diameter larger than 0.5 μm. It can be seen that generally our model results fit the observations well especially compared to DeMott et al. (2014). In Fig. 10(a)-(c), the model reproduces a similar magnitude and pattern of IN concentrations as observations in these locations. Due to the good agreements of both IN concentrations with observations (Fig. 10) and the relationship between IN concentrations and aerosol number concentrations with diameter larger than 0.5 μm (Fig. 12), we assume that the simulated aerosol number concentrations with diameter larger than 0.5 μm in these locations (Fig. 10(a)-(c)) would agree with observations. The large underestimates of IN concentrations in Barrow, Alaska (Fig. 10(d)) can corroborate the above explanation, as in this region aerosol concentrations are significantly underestimated by global modes in general (Liu et al., 2012; Wang et al., 2011).

- *Fig. 11: There are locations where the background colors do not fit the measurements, especially in the Pacific at 258 K. This should be discussed more, also concerning the significance/validity of this comparison in general. Besides the model predicts many IN*

at 264 K → How can ice form at these high temperatures if only soot and dust are accounted for as IN?

Reply: Following the reviewer's comment, we added the following discussion in the revised manuscript. In near-surface-air over marine regions, compared to dust IN, marine biogenic IN (types of marine biogenic particles include marine microorganisms, exopolymer secretions/colloidal aggregates, glassy organic aerosols, crystalline hydrated NaCl and frost flowers) are most likely to play a dominant role in determining IN concentrations at high temperatures. Thus over the Southern Ocean at 258 K, especially near the Antarctic coast, the model greatly underestimates IN(10s) concentrations (Burrows et al., 2013). Another region where the model significantly underestimates IN(10s) concentrations at 258 K is over the Pacific. In the remote marine boundary layer of equatorial Pacific Ocean, due to ocean upwelling, ship-based measurements found that atmospheric IN concentrations were associated with high concentrations of biogenic materials (Rosinski et al., 1987). Since we have applied a cut off of 0 for the active fraction at temperatures larger than -10 ^{o}C for all our new simulations and temperatures in some observations are warmer than -10 ^{o}C, the plot at 264 K is removed in the revised manuscript.

Presentation Quality:

The paper is well structured, the abstract and conclusion summarizes the paper in a clear way. Some argumentations are difficult to follow (see also specific comments). Comments concerning the number and quality of the figures can be found in the specific comments. I would recommend to shorten the title and to choose a more appropriate short title.

Reply: Following the reviewer's comment, we change to use a short title as "Different contact angle distributions for heterogeneous ice nucleation in the Community Atmospheric Model version 5".

Details:

− *Section 4.5/Fig. 10: The information about the IN(10s) concentration compared to measurement results in Fig. 10 is contradicting. At p 7157 line 19-20 it seems that the IN(10s) concentration is calculated based on interstitial aerosol concentrations from the simulations, later on at p 7158 line 1-2 it seems that the IN(10s) concentration is sampled at the grid boxes of the model.*

Reply: The IN(10s) concentration is a diagnosed variable which is calculated based on interstitial aerosol concentrations from the simulations as noted at p 7157 line 19-20. At p 7158 line 1-2, the word "sampled" may have confused the reviewer. We have changed "sampled" to "diagnosed based on interstitial aerosol concentrations from the simulations at the measurement locations" at p 7158 line 1-2.

− *In section 4.4 (p 7157 line 2-3) it is mentioned that changes in the mean contact angle do not have an impact on the temperature dependence of the active fraction. In the conclusions (p 7162 line 8-11) this statement is the other way around. Which one is right?*

Reply: These two statements may confuse the reviewer. We want to state that changes in the mean contact angle will not change much the slope of variations of active fraction with temperature, but the smaller (larger) mean contact angle will lead to larger (smaller) active fraction. Therefore, in the revised version, in section 4.4 (p 7157 line 2-3), we

replaced the original sentence by "indicating that changes of the mean contact angle do not change much the slope of variations of active fraction with temperature" in line 2. In the conclusions (p 7162 line 8-11), we used "we find that though the change of mean contact do not change the slope of variations of active fraction with temperature, it still can change the active fraction at a given temperature" to replace "we find that … (2012)". We added "judged from the absolute changes of the active fraction (not from its temperature dependence), the mean contact angle has a larger impact on the active fraction than that of standard deviation, which is consistent with the cloud-resolving model results by Kulkarni et al. (2012)" after "… may be depressed" in line 15.

– *The concept of the transition from singular to stochastic behavior should be explained in more detail.*

Reply: Following the reviewer's comment, we added "(do not change the slope of the curve)" after "temperature dependence of the active fraction" at p 7157 line 3. In addition, we used "with the increase of standard deviation, a broader distribution of contact angles will be allocated to aerosol particles. Since the different contact angle on each particle results in the different freezing temperature of each particle, the temperature range in which droplets freeze becomes broader. For example, for σ = 0.01, droplets freeze within a narrow temperature interval of about 10 ^{o}C, while for σ = 0.08, freezing occurs over a temperature range of about 18 ^{o}C. The change of the activated fraction with temperature (Figure 9(b)) becomes smoother with increase of the standard deviation, which indicates the "recovery" of the singular behavior (Niedermeier et al., 2011, 2014; Welti et al., 2012) and weakening of the time dependence of the stochastic behavior (see Figure S1 for the change of the time dependence with increase of the standard deviation)" to replace "Increasing … 2012)" at p 7157 line 5-8 (Section 4.4).

Specific comments:

− p 7143 line 5: what is hidden behind etc? Name all processes.

Reply: In the revised version, we deleted "and etc", and added "convective detrainment of cloud ice".

− p 7143 line 28: Why e.g. if all nucleation modes suggested by Vali are mentioned?

Reply: All nucleation modes suggested by Vali are mentioned. In the revised version, we deleted "e.g.," in line 28.

− p 7144 line 17-19: "Thus, at the given supercooling, if an ice germ reaches the critical germ radius, the droplet will freeze immediately. Otherwise the droplet should still keep liquid state irrespective of the time." → formulation is not so clear, the reference of otherwise is not obvious

Reply: We replaced these two sentences with "Thus, the freezing of a droplet is only determined by whether the temperature is below the characteristic temperature of the immersed IN in the droplet" in the revised version.

− p 7145 l line 3 and p 7145 line 10: missing reference: Chen et al. 2008

Reply: In the revised version, we added this reference in line 10.

− p 7151 line 2: it would be interesting to know the mineralogical composition of the dust

Reply: In the revised version, we added "which generally contain quartz, feldspars and clay minerals in different compositions (Linke et al., 2006)" behind "Saharan dust" in line 2.

– *p 7151 line 4: Add precisely what is changing*

Reply: In the revised version, we changed "some formulas" to "expressions of J_{het} and f"

– *p 7151 line 6: Why is the activation energy aerosol dependent? What is the physical reasoning behind this?*

Reply: In the revised version, as for activation energy, we added "for the transfer of a water molecule across the water-ice boundary" between "energy" and "is" in line 6. We agree that there are different approaches in the literature to parameterize this value, and that the approach of Chen et al (2008) to use the activation energy as a second fit parameter might be unphysical. However, as the activation energy is set to the same value for the single-alpha and alpha-PDF simulations, we argue that its value is not relevant for our analysis.

– *p 7155 line 13-16: from Fig. 4 no difference can be seen. Add also the same figure for the CNT simulation and/or the difference of both simulations (instead of Fig. 4)*

Reply: Following the reviewer's comment, we added this figure about the immersion freezing by dust for the PDF simulation to the figure for the CNT simulation for the comparison in the revised version.

– *p 7157 line 6-7: It should be elaborated why the enlarged temperature range of rapid increase of active fraction leads to stronger temperature dependence and to a weaker time dependence*

Reply: See the above reply.

– *p 7162 line 6-7: Where is the assumption/statement coming from that in case of the single-alpha model the freezing rate is constant in time?*

Reply: This holds as long as temperature and humidity are constant and the potentially ice nucleating particles are not depleted. We have added the reference "(Hoose et al., 2010)" at the end of sentence.

– *Table 1: Why is the activation energy in the case of deposition freezing negative? Please elaborate and add the physical explanation.*

Reply: In the case of deposition freezing, $\Delta g^{\#}$ (activation energy) is the energy of desorption per molecule, which stands for the surface with the outward flux of desorbed molecules. Instead, J_{het} specifies the flux of water molecules to the germs. Actually in J_{het} for the deposition freezing, there should be no a negative sign before $\Delta g^{\#}$ (see Eq. 9-8b in Pruppacher and Klett, 1997). However, in order to use the unified formula for both immersion and deposition freezing (Eq. 1), a negative sign is added before $\Delta g^{\#}$ *in*. (as in Chen et al., 2008). Thus the fit result for the activation energy in the case of deposition freezing is negative to offset this negative sign.

– *Fig 1: there is an error in the legend. ZINC 106: Obs and ZINC 106: single and alpha need to be switched.*

Reply: We have corrected this error in the revised version, and thank the reviewer for the careful reading.

− *Fig 1: the measurement points of ZINC 106 should be larger (difficult to recognize below the curves).*

Reply: We have enlarged the measurement points including CSU106 and ZINC 106 in the revised version.

− *Fig 1: there should be error bars included for the data points (in x- and y-direction).*

Reply: Unfortunately, no uncertainties to the data points are available to us.

− *Fig 2 and Fig 3 both show interstitial dust and soot particles, but the plots do not look similar. Why? If something different is plotted it should be made clear in the legend. If the same is plotted please delete the redundant figure.*

Reply: The reason why the two plots in Figures 2 and 3 for interstitial dust and soot particles do not look similar is because of the different scales with different colors in two plots. Following the reviewer's comment, we deleted the redundant Figure 3(c) in the revised version.

− *Fig 3: If soot is mainly coated (a) compared to (b), why is it not cloud-borne then? Is the reason that the particles are too small?*

Reply: Coated and uncoated soot and dust number concentrations in Fig. 3 are only calculated from the predicted total interstitial aerosol number concentrations. Because the size of soot is much smaller than dust, the one monolayer criterion results in a larger fraction of coated soot particles than of coated dust particles.

− *Fig. 1 and Fig. 9: It would be better to have the y-axis ranging from 0 to 1.*

Reply: As differences among observation data points are more than two orders of

magnitude and the largest active fraction is less than 0.1, y-axis needs to be logarithmic in order to show the data points clearly. Since we make y-axis logarithmic, the minimum value of y-axis canot be set to 0. So, in the revised version, we decide to keep the same logarithmic y-axis and still show x-axis from 0 ℃ but not plot any line between 0 and -10 ℃ to illustrate the cut-off.

Technical corrections:

– p 7144 line 2: change word order: For immersion freezing, a supercooled cloud droplet containing an ice nucleus nucleates by subsequent cooling at a certain degree of supercooling.

Reply: In the revised paper, we changed word order following your suggestion.

– p 7144 line 2f: split sentence in two

Reply: We split this sentence into two as: "During the time an immersed aerosol particle spends at constant environmental temperature, water molecules within supercooled water stay in the thermal fluctuation state of capturing and losing molecules to produce the clusters." and "This process resemble the structure of ice.".

– p 7156 line 28- p 7158 line 3: verb missing: However, the temperature range in which ice fraction rapidly increases does not become broader

Reply: In p 7156 line 28, we followed your correction. In p 7158 line 3, we added "are sampled" before "at the same pressure" in line 3.

References:

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Reply to the comments by anonymous referee #2

We appreciate the referee's valuable comments.

General comments: In this study, the authors improved heterogeneous ice nucleation parameterization for mix-phase clouds in CAM5 by implementing a classical-nucleation-theory-based parameterization (CNT), and further improved it by extending it from a single contact angle model to a PDF of contact angle model. The paper represents a significant advancement in parameterizing heterogeneous ice nucleation in global climate models and the authors have done a careful job on implementing the parameterization in CAM5. In particular, I applaud the authors efforts on a) refitting the CNT in single alfa mode and PDF of alfa mode to constrain the key uncertain parameters in these parameters from observations; b) utilizing the cloud-bore aerosol capability in CAM5 modal aerosol treatment and treating the cloud-born and interstitial aerosols separately in their heterogeneous ice nucleation parameterization; c) evaluating IN concentrations using available observations around global. The implementation in CAM5 makes it possible to examine how natural and anthropogenic aerosols affect mixed-phase clouds and further climate. Though I agree with the reviewer #1 on the challenge in representing the time-dependent behavior in climate models with long time step, this challenge is generally true for any time-dependent processes treated in climate models with long time step, especially those related to cloud microphysics, such as for droplet activation for liquid clouds and homogeneous freezing/heterogeneous freezing in cirrus clouds. I would suggest the authors to add some further discussion/review on how climate models treat time-dependent processes, which will help to put this study in context (for example, how Hoose et al. (2010) implemented CNT in their climate model). The paper is also well written and is a great addition to the literature dealing with heterogeneous nucleation and their parameterizations in climate models. I would strongly recommend the publication of this paper after some further clarifications are made:

Reply: Please see our reply to the reviewer #1 for the time dependence of CNT, and the applicability of CNT in a GCM with a 30-min time step. We would like to note here that the approach of a PDF of contact angles has a less time dependency than the single contact angle approach in CNT. In the revised version, following the reviewer's comment, we added more detailed discussions about the time dependence in Section 3.

Specific comments:

1. lines 15 to 23, page 7143: This part reads awkward, and needs rewording. In particular, "On the other hand" does not fit well.

Reply: We reworded lines 15 to 23, page 7143 as follows: "Natural mineral dust particles are often internally mixtures of different minerals, quartz and other component (Murray et al., 2012). In order to reduce the complexity encountered in natural mineral dusts, many laboratory studies, on the one hand, have often used commercially available minerals, in particular kaolinite, illite and montmorillonite (Hoose and Möhler, 2012; Hoose et al., 2008b). Other laboratory experiments, on the other hand, used commercially available Arizona Test Dust (ATD) as a surrogate for natural mineral dusts (e.g., Knopf and Koop, 2006; Marcolli et al., 2007; Kulkarni et al., 2012). ATD can possibly be more ice nucleation active than natural desert dust, either due to its enhanced roughness resulting from the milling or due to its different mineralogical composition (Möhler et al., 2006)."

2. Line 11, page 7145: "weak time dependence". It will be helpful for readers to further elaborate what "weak time dependence" mean here, as this concept are mentioned several times later in the paper.

Reply: We added "(meaning that the observed ice nucleation fraction increases slowly with increasing time)." behind "weak time dependence". More detailed explanations were added in the corresponding section to answer the reviewer #1.

3. Page 7149, line 25: dt is the model time step. This may warrant further discussions here, as also mentioned by reviewer #1. I noted that the authors have some discussion on this in the last paragraph of Section 5. I agreed with the authors that one way to handle this is to add ice-borne aerosol particles, though this is clearly beyond the scope of the current manuscript. Some further discussions on this will be helpful here. For example, dt may be thought as time scale to replenish IN population in a grid point. Also, how do other climate models handle similar situation.

Reply: We have added more discussion about aerosol scavenging due to droplet freezing in our answers to reviewer #1. Your suggestion is valuable and we will mention that dt may be thought as time scale to replenish IN population in a grid point. Currently, as far as we know, it seems that no major climate model considers aerosol scavenging due to droplet freezing except for the ECHAM5-HAM version described by Hoose et al. (2008a), which considered aerosol processing in clouds in ECHAM by introducing a separate in-crystal aerosol mode. However, due to the added complexity and the computational demands, the standard version of ECHAM doesn't take all aerosol processes in clouds into account. If ice nucleation is to be described with an alpha-pdf model, the scavenged dust particles would have to be additionally tagged with their contact angle, adding even more complexity. CAM5 developers are trying to include ice-borne aerosols recently, but until now it hasn't been completed.

4. Page 7150, line 18: so 2000 bins are used for calculating activation fraction from Eq. (4) for refitting some of uncertain parameters. How about online in CAM5? How many bins are used for calculating activation fraction online in CAM5?

Reply: In CAM5, due to consideration of the computation efficiency, we used 101 bins to calculate the activated fraction. 100 bins were used for fitting in Lüönd et al. (2010). We have made sensitivity tests and find the number of bins only causes around one degree difference of contact angle.

5. Page 7151, line 9: why is the same activation energy as that in the single alfa model used for the PDF alfa model?

Reply: In the formulation by Chen et al (2008), the activation energy is aerosol type and nucleation mode dependent. It is the transfer of a water molecule across the water-ice boundary and should from a theoretical standpoint be independent on the contact angle.

6. Section 3: How are aerosol number concentrations (dust and soot) used in the CNT single-alfa and PDF-alfa model caclucated? For 3-mode treatment, dust and soot are internally mixed with other aerosol species in the accumulation mode.

Reply: In the accumulation mode, interstitial soot (dust) number concentration is calculated with the interstitial mass concentration of soot (dust) in this mode multiplied by different constants based on the soot (dust) size distributions, respectively. Cloud-borne soot (dust) number concentration is calculated from the predicted total cloud-borne number concentration in the accumulation mode weighted by the mass fraction of cloud-borne soot (dust) in this mode. In the coarse mode, interstitial (cloud-borne) dust number concentration is calculated from the predicted total interstitial (cloud-borne) number concentration in the coarse mode weighted by the mass fraction of

interstitial (cloud-borne) dust in this mode.

7. Page 7153, lines 1-15: there are some discussions here regarding coated vs. uncoated. However, how soot or dust particles are counted as coated particles are defined in next paragraph (lines 16-21). Suggest to move the latter before lines 1-15.

Reply: In lines 1-15, our discussion mainly focuses on the total number concentrations of soot and dust. Coated and uncoated aerosols are only mentioned in line 3. In the revised version, we added "(see next paragraph for definitions of coated and uncoated portion)" after "… contact freezing" in line 4, page 7153.

8. Page 7153, line 20: here one monolayer is used to define a coated particle. Any uncertainty on this definition and how this might affect your results? How does lab experiment define coated vs uncoated dust particles?

Reply: Theoretically, using a criterion of more than one monolayer will lead to more uncoated particles and thus lead to increase of deposition and contact freezing rates. In laboratory experiments with coated particles, the resulting coating thicknesses are often poorly constrained and not necessarily evenly distributed over the particle size distribution (see e.g. Cziczo et al, 2009).

9. Page 7154, line 9: fine dust is separately as well in MAM7, which may affect coated vs. uncoated dust number concentrations.

Reply: In MAM7, dust is internally mixed in fine soil dust and coarse soil dust modes. These two modes are both soluble modes. Since sulfate is also internally mixed with dust in these two modes, the coated and uncoated dust number concentrations would be not affected a lot due to MAM7. Conversely, soot in MAM7 is internally mixed in accumulation and primary carbon modes. The primary carbon mode is the insoluble mode

(no sulfate in this mode), which will leads to the large number concentration of uncoated soot. Thus, the deposition and contact freezing rates by soot will increase a lot compared to MAM3.

10. Page 7153, line 12: dNi: how is this calculated? Does this Ni change include all changes in the Ni prognostic equation, such as sources/sinks from ice nucleation, advection, convective detrainment, conversion from ice to snow?

Reply: dNi is the ice crystal number concentration which is only predicted from the different heterogeneous freezing modes (immersion, deposition and contact freezing). Therefore, it doesn't include all changes in the Ni prognostic equation as referred to by the referee. We explained dNi more accurately in the revised version.

11. Fig. 7: so each data point sampled in Fig. 7 represent one annual mean value at a particular grid point? This needs clarification.

Reply: Yes, it is true that using annual mean temperature value at each grid point is not reasonable. In the revised version, the temperature value and frequency of three freezing modes were sampled every 3 hours at each grid point and then the Fig. 7 was updated, although we found that the results do not change much..

12. Page 7157, line 14-25: I agreed with reviewer 1 that the comparison between observations and model can be challenging here, as aerosol number concentrations can be different, though the same location is chosen.

Reply: Right now, we don't have aerosol data in these locations on hand. However, the comparison between observations and our model results is, to some extent, already shown in our manuscript. In Fig. 12 we show the relationship of the IN concentrations and

aerosol number concentrations with diameter larger than 0.5μm for all gridpoints. We can see that generally our model results fit the observations very well especially with DeMott et al. (2014). On the locations shown in Fig. 10(a)-(c), the modeled IN concentrations reproduce the similar magnitude and pattern as observations. As you said, we should firstly confirm that the simulated aerosol concentration in these locations agree with observations and then further do comparison of IN concentrations. However, due to good agreement of IN concentrations with observations in Fig. 10 and confirmed relationship between IN concentrations and aerosol number concentrations with diameter larger than 0.5μm in Fig. 12, we may derive that the simulated aerosol number concentrations with diameter larger than 0.5μm in these locations (Fig. 10(a)-(c)) should be agreement with observations (if we had observations in these locations). The large underestimates of IN concentrations in Barrow, Alaska (Fig. 10(d)) can confirm the above explanations because in this region the aerosol number concentrations, especially soot, are greatly underestimated (Liu et al., 2012; Wang et al., 2011).

13. Page 7159, line 29: what is the prescribed size distribution for transported dust?

Reply: Transported dust is in coarse mode with mass median diameter 2.524μm and standard deviation is 2.0. In the revised version, we added this information to the manuscript.

14. Figure 11: I applaud the authors effort for collecting these IN observations around global for evaluating their model. For those measurements in 1980s and 1970s, what are the measurement techniques used and how would that affect the comparison here with the model results. For example, 10s residence time is used for comparing with DeMott et al. results. Is that still used for comparing to these old results as well?

Reply: The observations in 1980s and 1970s were made using a filter technique (not a

Continuous Flow Diffusion Chamber (CFDC) technique). The filter based technique involves drawing a known volume of air through a filter and detecting ice nuclei on that filter using a diffusion chamber or droplet freezing methodology. Flow rates through the filter can range from 2.5 to 10 SLPM depending upon the altitude of the aircraft. Flow rates were monitored using a mass flowmeter. The flow rate was recorded on the aircraft data system so that final sample volumes were calculated by integrating the flow rate each 10s (Borys 1989). Therefore, it still make sense that 10s residence time is used for comparing to these old results as well.

15. Page 7160, line 4: the size of sea salt particle is not small.

Reply: Yes, the size of sea salt particle is not small. In Page 7160, line 4, "due to its smaller size" is supposed to describe soot and we moved it behind "soot" in line 4 in the revised version.

16. Section 4.6. I found section 4.6 is very interesting, and it is worth to add some further discussions. For example, while IWP in present day simulation is generally smaller with the new parameterizations compared to CTL, changes in IWP are generally larger. So what might cause this? Is this due to increased dust concentrations (partly due to less efficient wet scavenging) and increased soot concentrations in the PD simulations? As for changes in LWP and LCC, why are they generally larger than in CTL? How column-integrated droplet number concentration changes, and how LWP from stratiform clouds changes? It may be beneficial for readers to add some of these results into the abstract.

Reply: IWP in present day simulation is generally smaller with the new parameterizations compared to CTL because Meyers' scheme, which doesn't link to aerosols, largely overestimates the nucleated ice number concentrations (DeMott et al.,

2010). There may be two reasons that cause changes of IWP between the present-day and preindustrial simulations with the new parameterizations to be generally larger than those in CTL. As you said, one is increased dust concentrations (partly due to less efficient wet scavenging) and increased soot concentrations in the PD simulations. Another reason may be that soot is taken into account in new parameterizations, which enlarges the differences between the present-day and preindustrial simulations. In the revised version, according to your suggestion, we added these results into the abstract and added these two possible explanations in Section 4.6.

17. Page 7162, lines 5-7: This "On the other hand" does not fit well here, and suggest to reword this sentence here.

Reply: We deleted this sentence in the revised version.

Technical corrections:

1. page 7144, line 10: "which includes" → "which include"?

Reply: In the revised paper, we corrected it.

2. page 7149, line 5: "we can" → "we"?

Reply: In the revised paper, we deleted "can".

3. page 7153, line 7: remove "both".

Reply: In the revised paper, we removed "both".

4. Page 7157, line 9: suggest to replace "it results" with something like "increasing the standard deviation results".

Reply: Follow the reviewer's suggestion and we added more explanations here to answer the reviewer #1.

5. Page 7159, line 10: "diagnosed" → "diagnose"?

Reply: In the revised paper, we corrected it.

6. Page 7162, line 26: "their behaviors explored in global models". This sounds not like a complete sentence.

Reply: It was changed to "their behaviors should be explored in global models" in the revised paper.

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