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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