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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of heterogeneous ice nucleation by natural dust and soot based on a probability density function of contact angle model with the Community Atmospheric Model version 5" *by* Y. Wang et al.

Anonymous Referee #1

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The Manuscript "Impact of heterogeneous ice nucleation by natural dust and soot based on a probability density function of contact angle model with the Community Atmospheric Model version 5" written by Y. Wang, X. Liu, C. Hoose and B. Wang outlines the relative importance of different freezing modes for heterogeneous freezing. Additionally differences in Classical Nucleation Theory (CNT) based parameterization concerning the contact angle and its implications for freezing are investigated. For that purpose a new CNT based freezing parameterization is implemented into CAM 5 using a probability density function of contact angles (alpha-PDF) instead of a single

contact angle. The needed parameters are estimated by data fitting. The manuscript contributes new information concerning the question how to parameterize freezing in global climate models, but it is questionable if the applied method is appropriate.

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.

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.

Details:

- It seems rather difficult to study a time-dependence of a process without having sub-timestepping. 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?

- 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.

- 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.

- 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.

- 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.

- 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.

- 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)?

- 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?

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.

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.

– 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?

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

Specific comments:

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

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

– 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

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

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

– p 7151 line 4: Add precisely what is changing

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

– 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)

– 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

– 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?

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

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

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

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

– 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.

– 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?

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

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.

– p 7144 line 2f: split sentence in two

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

References 1.) Chen, J.-P., Hazra, A., and Levin, Z.: Parameterizing ice nucleation rates using contact angle and activation energy derived from laboratory data, Atmospheric Chemistry and Physics, 8, 7431–7449, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 7141, 2014.