Dear Editor,

We thank reviewer 3 for the review and detailed comments. The evaluation has helped us to improve the quality of the manuscript, and our reply to the respective comments is as follows. Also accordingly we have revised the manuscript and the modifications are highlighted.

Thanks once again,

-Authors

**Anonymous Referee #3**

**Specific Comments**
Abstract
1. The finding highlighted in the abstract that inferred single contact angles do not vary substantially with particle size or temperature seems to contradict some number of other studies.

Reply: The reviewer is correct in pointing out that particle size is important for the deposition ice nucleation. In this study we only looked at four different particle sizes: 100, 300, 400, and 500 nm. For these sizes, we observed the onset $RH_{ice}$ and single contact angles varied between 115 to 132% and ~18 to 24 degrees, respectively (Table 1 and 2). These results are in agreement with many past studies and also with a deposition ice nucleation parameterization (Wang and Knopf, 2011 and references therein). Please refer to Fig. S1 (c.f. in the supplementary section).

In the revised manuscript, we have specified the range of single onset contact angles (in the abstract section) and cited the Fig. S1 (in the section 3.1).

The revised text in the abstract reads as follows.

"Results show that onset single contact angles vary from ~18 to 24 degrees, while the PDF parameters are sensitive to these environmental conditions (i.e., temperature and dust size)".

The revised text in section 3.1 reads as follows.

"Under the conditions that we carried out our experiments, the onset $RH_{ice}$ varied from 115 to 132% and the respective single contact angles varied between ~18 and 24 degrees. The dependency of these contact angles on the onset $RH_{ice}$ is in agreement with the previous study (Wang and Knopf, 2011) that parameterized the deposition ice nucleation as a function of $RH_{ice}$ (Fig. S1; c. f. in the supplement section). The deposition ice nucleation onset has been shown to be insensitive to the experimental temperatures (-25 to -35 degC) in past studies (e.g. Kanji and Abbatt, 2006; Kulkarni and Dobbie, 2010) for dust particles".

2. The cloud model simulation results are stated without introducing the fact that a model was used and why.

Reply: The particular sentence has been revised as “Cloud modeling simulations were performed to understand the sensitivity of cloud properties (i.e. ice number concentration, ice water content, and cloud initiation time) to the representation of contact angle and PDF distribution parameters”.

Introduction Pages 2486 to 2487 : A limitation of parametric methods is stated, but nothing is said in this regard about CNT. The assumptions made in applying CNT are that atmospheric IN can be described by the theoretical construct, either via single contact angles or some distribution of such, and that all ice nuclei behave in a purely stochastic fashion. In fact, the role of time is not even mentioned in this section. It is not correct to ignore these facts, which are quite difficult to validate experimentally. Furthermore, some confusion is present here because the application of CNT is distinctly different than the approach taken by Möhler et al. (2006), where it is assumed that particles activate without time dependence (similar to Connolly et al.). Finally, I believe that the Niedermeier et al. reference given is an application of CNT involving probability distributions of contact angles, and characteristic active sites, and offers an alternate approach to the contact angle-PDF approach that should be better acknowledged.

Reply: The parameters that are not included into the CNT are described in the introduction section. The revised sentence in section 1 reads as follows. “It should be noted that parameters such as the magnitudes of elastic strain, aerosol surface irregularities, and active sites might affect CNT calculations, but are not accounted into the CNT approach and are ignored in this study as uncertainties of these parameters are very large”.

Role of time in the CNT calculations could be important. In our CNT calculations, we have used the particle residence time within ice chamber (~ 12 seconds). Later the CNT predicted active fractions and experimental activated fractions are used to calculate the PDF parameters. CNT calculations are time dependent and are acknowledged in section 1. Following sentence is added. “For the CNT calculations the time is restricted to the particle residence time within the ice nucleation chamber (~12 seconds; see Sect. 2)”.

Niedermeier et al. (2012) methodology to model the heterogeneous ice nucleation observations are described in sections 1 and 4. Following text is added to describe their methodology.

“CNT was further modified by Niedermeier et al. (2011), where instead of a distribution of contact angles over the entire dust sample, they described a conceptual model that treats each particle consisting of a distribution of surface sites or properties of IN. They concluded that ice nucleation treatment models that are based on the stochastic theory might be influenced by the heterogeneity of surface properties depending upon the time and freezing temperatures”.

Their approach provides an alternate methodology to the contact angle-PDF approach is acknowledged as follows (added in section 4).

“In the future, new approaches such as ice-active surface site density approaches (e.g. Connolly et al., 2009; Niedermeier et al., 2011) that offer an alternative method to develop the CNT based time-dependence parameterizations should be explored and incorporated in the modeling studies (e.g. Ervens and Feingold, 2012) to understand the importance of such approaches towards simulating cloud properties".

Methodology

Page 2488, line 12: Friedman et al. called this device the compact ice chamber (CIC). Now it is the PNNL AML ice chamber. Define AML. I suggest sticking with one name.

Reply: Corrected. It is CIC. AML is defined.

Page 2489, lines 3 to 5 and lines 15 to 16: 1 micron seems too small of a size to set as a cutoff for ice crystals if one is generating 500 nm particles, or even 300 nm particle using a DMA. It is impossible that there will not be some multiply charged particles this large at the exit. Statements are made regarding such particles, but are only explicitly documented for 100 nm particles out to triplet sizes. For 300 and 500 nm particles, 10

Reply: Referee is correct in pointing out that triple charged particles from 500 nm might bias the ice signal detected by the OPC, such that one might inadvertently count dust particles as the ice crystals. This possibility is actually taken into account when we report the measurements. We always ran the blank experiments where the CIC is set to $RH_{ice}$ = 100% (no supersaturation) conditions, size selected aerosols are sampled, and the particles at the exit of the CIC are counted. This is considered as a background count, and the average of these counts (if > 0) are automatically substracted from the counts when we are running the experiment ($RH_{ice}$ > 100%).

For 300 and 500 nm size particles, the contribution from multiple charged particles (double and triple charged particles) was less than 10%. The following table describes the size of multiple charged particles. Only 100 nm has significant contribution and these numbers are described in the paper. Multiple charged particles for 400 and 500 nm size were based on the Baron and Willeke (2001) calculations.

| Size (nm) | Double charged particles | | Triple charged particles | |
|---|---|---|---|---|
| | Size (nm) | Contribution (%) | Size (nm) | Contribution (%) |
| 100 | 152 | 36 | 197 | 16 |
| 300 | 506 | 10 | 705 | 10 |
| 400 | 696 | 10 | 982 | 10 |
| 500 | 880 | 10 | 1250 | 10 |

The contribution from multiple charged particles is described in the revised manuscript. The new sentence reads as follows.

"For 300, 400, and 500 nm diameter size particles, the contribution of these multiple charged particles was less than 10%. For sizes 100 and 300 nm particles, the multiple charge calculations were based on the routine experimental measurements, whereas for 400 and 500 nm particles the calculations are based on the Baron and Willeke (2001) calculations"

Page 2490, lines 6 to 7: Please clarify if residence time was varied in any experiments to prove the applicability of the CNT time dependence. If not, simply say so as a preface to all of these calculations. It is an assumption made, the validity of which has not been tested.

Reply: The particle residence time within the experiment (in the ice nucleation chamber or CIC) was fixed. CNT calculations were based on this time. This is acknowledged in the introduction section. It reads as follows, "For the CNT calculations the ice nucleation time is restricted to the particle residence time within the ice nucleation chamber (~12 seconds; see Sect. 2).

The 'time' information is not assumed, but is based on the total particle residence time within the ice chamber.

Page 2491, lines 12 to 14: The threshold condition used to define onset single contact angles seems rather artificial given the nature of the actual data shown. This is just an observation.

Reply: Onset single contact angles were determined at the value of $RH_{ice}$ measured at $F_{ice}$ = 1%. For these calculations the onset $RH_{ice}$ values are directly read off from the PDF fitted curve of active fraction verses $RH_{ice}$.
This is described in section 2.2. See equation (5) and related discussions.

Pages 2491 to 2493, section 2.3: I suggest that this section needs some attention to detail, as it is very confusing how many models are used and what tests or actual simulations are done.

Reply: Sorry for the confusion. Actually there is only one cloud model used. The first set of tests is only the offline (box) module testing of the PDF- $\theta$ and onset- $\theta$ based CNT parameterizations under different measurement conditions. This has been clarified in section 2.3 (paragraph 1 and 2). The second set of tests is based on the cloud model simulations (paragraph 3).

The new two paragraphs read as follows.
"Two sets of simulations are carried out to examine the sensitivity of modeled cloud properties to different representations of contact angles in CNT.
First, we ran offline module tests for the PDF-$\theta$ and onset- $\theta$ based CNT parameterizations to calculate the ice number concentration ($N_{ice}^{model}$) using a dust size distribution from Cziczo et al. (2006) (Fig. 3), temperature, $RH_{ice}$, the integration time step (2 s), and the PDF parameters (Table 1). In the module tests for the PDF- $\theta$ parameterization, we calculated $F_{ice}^{mod}$ and $F_{ice}$ from equations (3) and (5), respectively. We chose the dust size distribution from the literature study of Cziczo et al. (2006) with total dust concentrations ($N_0$) of about 10.7 $L^{-1}$. The $N_0$ was calculated by integrating the observed dust size distribution (Fig. 3)".

-Page 2491, lines 21-22: Was it a stratiform or a cirrus case? I assume this refers to the present paper or is it just mentioning the paper in which the CRM was described?

Reply: The cloud is stratiform evolving cirrus. The cited papers (Khain et al., 2004; Fan et al., 2009) describe the model. We have added the paper for the stratiform/cirrus case: Comstock et al., 2007. The revised sentence reads as follows. “In the second set of simulations, we conducted sensitivity studies using a cloud resolving model (CRM) that has the dynamical framework of a large-eddy simulation model and a spectral-bin microphysical scheme (Khain et al., 2004; Fan et al., 2009) to simulate a stratiform/cirrus case (Comstock et al., 2007)”.

-Page 2492, line 12, “Two sets of model simulations. . .”: So in these simulations you are not simulating the cirrus case? Are the initial conditions the same? It is confusing.

Reply: Sorry for the confusion. Actually there is only one cloud model used. The first set of tests is only the offline (box) module test of CNT with the single contact angle and PDF-contact angle under different measurement conditions. This has been clarified in section 2.3.

-Page 2492, lines 16 to 17: Is this dust size distribution for the same cloud simulated? Or is this an arbitrary choice, and why?

Reply: The initial dust size distribution is kind of an arbitrary choice, since we do not have any observations about it and the CRM model does not include chemistry and aerosol modules to simulate it. The choice of dust size distribution does not matter much to our tests here because our purpose is to test the sensitivity of cloud properties to the CNT parameterizations in the deposition mode only (not to simulate the actual case) as stated in section 3.3.

Lines 21 to 22: Is this using the same model?

Reply: As clarified now, the second set of tests used a CRM model called SAM. The first set of tests is only offline (box) module test, not CRM model simulations.

-Page 2492, line 5: It would be good to know something about the sounding. What is the temperature range in which the cloud forms?

Reply: In our simulations, the cloud generally forms at the temperature range of -27 to -14 degC. This has been added to the first paragraph of section 3.3. New sentence reads as follows, “Clouds generally form at the temperature range of -27 to -14 degC in our simulations”.
The temperature profile from the sounding has been added to the first panel of Fig. 8.

Results and discussion
Page 2493, lines 10 to 11: The results indicate dependencies of ice active fraction on RH and temperature, but they also suggest the possibility of a variable, purely aerosol interference at all temperatures and sizes. The ice nucleation signal starts directly from ice saturation, unlike some other recent studies of such aerosols, including the Welti et al. paper. Has this been carefully characterized as ice and rejected as an artifact of the detection method used?
What if the ice cut size is pushed to something larger that should still capture all nucleated and grown ice, such as 2-3 microns. If this background goes away, it could be smaller ice crystals, but it could also imply aerosol interference that requires correction.
Even the PDF scheme cannot reproduce that initial tail can it?
Might not this explain the difficulty with the PDF parameter fits in general, and the

insensitivity of single contact angle to size and temperature?
I suspect that a more conservative approach to detecting assured ice would help a great deal.

Reply: As mentioned above, we perform blank experiments to remove any artifact from counting dust particles as ice crystals. The reported activated fractions are corrected to such artifacts.
If we push the ice threshold limit to 2 to 3 micrometers, we will observe lower ice number concentration. There will be very few ice particles above 3 micrometers, and if the concentration falls below background count limit or concentration, then we always report zero active fraction.
We agree that PDF scheme cannot reproduce the initial tail, however the least root mean square error (RMSE) fit ensures we obtained the best fit (PDF parameters) between observed and modeled active fraction magnitudes.
Onset single contact angles were determined at the value of $RH_{ice}$ measured at $F_{ice}$ = 1%. For these calculations the onset $RH_{ice}$ values are directly read off from the PDF modeled fitted curve, see Fig. 2. This is described in section 2.2. The lower active fraction experimental data points that are not reproduced by the PDF model are lower than threshold $F_{ice}$ = 1% and therefore does not explain the insensitivity of single contact angle to size and temperature. As noted above increasing the ice threshold limit is not desirable.

Page 2494, line 2 to 3: I find the discussion here to be overly confident in the fact that the PDF parameter scatter is simply the result of active site variations. Such results are not very evident in data shown in Welti et al. (2009) and Jones et al. (2011).

Reply: Other referee also points out that the scatter in PDF parameters cannot be completely explained by the active site theory. We repeat the similar reply here and have removed the active site discussions in the revised manuscript.

We define active sites as the favored sites for the ice to nucleate. In this section we are trying to explain the variability in the measurements, e.g. in Fig. 4 for the activation spectra of dust particles, using active sites concept. In this figure we can observe that the ice nucleating properties of dust particles are function of temperature, $RH_{ice}$, and size. To explain the variation in the activation spectra, at any measurement condition, we think each dust particle has different surface property than the other or the distribution of active sites from particle to particle is different. However, we do not have any measurements of active sites to support this premise. Therefore considering these limitations, we removed the active site argument from the discussion section.

PDF parameters are used to parameterize the activation spectra of dust particles (also see Wheeler and Bertram, 2012). If the activation properties of dust particles vary (see Fig. 4) then the PDF parameters also vary. This is the reason why we see different PDF parameters for different temperature and particle sizes (Table 1 and 2). This is added in the 2$^{nd}$ paragraph of section 3.1.

Page 2494, lines 6 to 8: How many experiments are represented here for each PDF fit? Does this discussion imply that insufficient repeats were done in this study?

Reply: The experiments were not repeated. It is not sure if repeating the experiment would lead to better PDF parameters. But if we assume the surface properties of each dust particle is different than the other then repeating the experiments at one measurement condition multiple times would help to build a statistical active fraction data. However, as mentioned in the above reply we do not have any measurements on the active sites so it would be difficult to validate this assumption. In the revised manuscript we have removed active site argument for clarity.

Section 3.2, page 2495: I am just not sure that the sensitivity studies run offline here deserve even as much space as they are given. Much of this is intuitive on the basis of CNT, and has been explored for CNT going back many years. Nevertheless, the point that the onset of ice formation by the single contact angle method is nearly a step function affecting all of the ice nuclei at one RH, with consequent potential impact on cloud microphysical and radiative properties, is a point worth repeating (see, e.g., Eidhammer et al. 2009). You might comment.

Reply: Correct, the module test of single contact angle approach show the step function, affecting all of the particles, at one $RH_{ice}$, to activate (Fig. 6). This observation is an agreement with the Eidhammer et al. (2009) study for the simulation where they used CNT based parameterization (from theory: Khvorostyanov and Curry, 2000, 2004). They suggest that steeper curves can produce more efficient immersion freezing of haze droplets. This will glaciate the clouds quickly and would alter the total ice crystal number concentration. Further, they suggest that these rapid freezing rates would influence the cloud properties (thickness, lifetime) and amount of precipitation. Therefore, it is possible that the steep rise of activation observed in our study can also be having implications on the cloud properties (as pointed out by the reviewer). Following sentence is added to the section 3.2 (last paragraph).
"It was observed that nearly all particles activate at one particular $RH_{ice}$ predicted by the single contact angle parameterization method, and therefore might also influence cloud properties (Eidhammer et al., 2009)

Section 3.3, page 2495, line 21-22: Probably better stated as "The cloud depth decreases as the contact angle increases."

Reply: Corrected.

Page 2495, line 26: Ice nucleation being very efficient goes along with a low contact angle doesn't it? Again, this seems intuitive. And why is there a need to flex calculations for small contact angles if they do not represent real dust?

Reply: Referee is correct in pointing out that particles with low contact angles are efficient IN. In this work we wanted to understand the sensitivity of cloud properties to contact angles and therefore these simulations are carried out for reference. Past studies (e.g. Kanji and Abbatt, 2006) have shown the onset $RH_{ice}$ can be as low as 105% for various types of dust particles. The respective contact angle can be 8 degrees (Fig. S1). In our work we varied the contact angles from 5 to 30 degrees. Thus, we investigated the influence of possible contact angle distribution on modifying the cloud properties.

Page 2497, lines 5 to 6: A similar comment as the previous one, as this is another point that should be self-evident in applying simple CNT calculations. If it is necessary to get to higher RH for ice nucleation to occur, then it should translate into a later time and/or higher altitude of cirrus formation, correct? Does the model provide special insight or simply verification of such facts?

Reply: The CRM model simulations did show that the cirrus cloud formation is delayed and the cloud base is higher as the contact angle increases (Fig. 8). However, the cloud top height is not at higher altitude since higher altitude does not necessarily mean higher $RH_{ice}$. The last sentence of the 2nd paragraph in section 3.3 has been modified to reflect this point. It reads as follows.
"In general, cloud formation is delayed and cloud base height is higher with the increase of contact angle (Fig. 8). But higher cloud top height does not necessarily lead to higher $RH_{ice}$"

Page 2497, line 9: I think suppression is the wrong word here. I thought Friedman et al. showed no ice nucleation activity. What is meant by suppressed?

Reply: We wanted to say poor ice nucleating properties of the particles. Sentence is revised. New sentence reads as follows.
"Although our version of CNT does not directly include information of aerosol chemical composition and coating, the simulations with the larger contact angles can be associated with the particles that have poor ice nucleating properties".

Page 2497, lines 11-14: Again, here is a case of putting the "cart before the horse" Such behavior seems predetermined by existing theoretical equations. The model simply verifies the obvious impact. I suggest stating expectations upfront based on the nature of CNT calculations, and then show that these behaviors are realized in the model.

Reply: For clarity we removed the repetition. Sentence is added to the previous paragraph.

Page 2498, line 1: I am surprised at this blanket statement on the lack of sensitivity to N0. The N0 used in this study is a very small number. One can see in the figure that at 10 times N0, the ice number did not go up by 10 through the cloud depth. It thus seems possible that higher N0 will eventually lead to negative feedbacks, and so without knowing just how high N0 is or should be allowed to be specified, there will be a problem in predicting aerosol effects on clouds.

Reply: We meant that the results of the sensitivity of cloud properties to contact angle and PDF parameters should not be qualitatively changed by $N_0$. We agree with the reviewer that to examine aerosol effects on clouds for specific cases, we have to know $N_0$. The cloud properties definitely are sensitive to $N_0$. The sentence has been modified as "…the results on cloud sensitivity to contact angle and PDF parameters are not expected to change qualitatively at the higher $N_0$ since the fraction of activated IN is determined by the PDF parameters, not the $N_0$. Certainly, $N_0$ will change cloud properties of a single case. It is a necessary quantity to be known to examine aerosol effects on clouds".

Page 2498, line 16, 26 to 28: In reference to the comments on ice detection threshold and IN measurement errors, is the thought that you are missing ice? I think just the opposite, and I think there are things to do with the data (e.g., moving the ice detection threshold size) to explore this issue further.

Reply: It is possible that we are underestimating the total number of ice crystals by setting the ice threshold size at 1 micrometer. Because for example in the case of 300 nm particles, assuming no multiple charged particles are present, ice crystals that are formed within the ice chamber from size 300 to 1000 nm are not counted. Moving the ice detection threshold, to say 3 micrometers, will further reduce the ice crystal concentration.

Summary and future work

Point 1: As I mentioned earlier in these comments, I detect that there is reluctance to say this approach appears incorrect. If so, it should be said that this approach could lead to large errors.

Reply: We observed that cloud properties are sensitive to the two different ice nucleation parameterizations. Recent study (Wheeler and Bertram, 2012) also showed the sensitivity of these two parameterizations towards ice nucleation measurements. They suggested that single contact angle approach is not suitable to parameterize the heterogeneous ice nucleation. We agree with the referee that caution should be taken while implementing such schemes in cloud models. The following text is added to section 4, "Cloud properties are observed to be sensitive to the magnitude and representation method of contact angles, implying that accurate representation of contact angle is crucial to simulate cloud properties in the model".

Point 2: This could start with "As expected on the basis of well-known CNT ice nucleation sensitivities to contact angle. . ."

Reply: Revised. New sentence reads as follows "As expected on the basis of CNT, cloud properties were observed to be sensitive to the contact angles".

Point 3: Just to repeat a point above, I believe that it would be instructive to vary N0 over a broader range. So cloud properties depend on the PDF parameters and N0 in the CNT approach. This is important, as N0 is not always well known, nor are large particles always only dust.

Reply: Agree, for the reason we have performed simulations where $N_0$ was varied (Table 3). The sensitivity tests and the corresponding results are described in section 3.3.

Point 4. Onset contact angles are calculated, not observed. Rephrase.

Reply: Corrected. The new sentence reads as follows, "Calculated onset single contact angles are consistent with the literature data, but …"

Page 2500, lines 4 to 5: "Therefore, for the purpose of applying CNT to describe atmospheric ice nucleation. . .”

Reply: Revised. New sentence reads as follows, “Therefore, for the purpose of applying CNT to represent atmospheric ice nucleation, experimental methods applied to obtain the contact angles need to be standardized in the future (Cantrell and Heymsfield 2005).”.

Page 2500, lines 15 to 17: Please make it explicitly clear how fundamentally different these approaches (e.g., Connolly et al.) are compared to CNT. It involves the basic assumption on the time dependence of ice nucleation, and this is what must ultimately be explored somehow.

Reply: In the introduction section we have described the Connolly et al. (2009) approach. Following text is added to the revised manuscript.

“Connolly et al. (2009) developed a new parameterization based on their laboratory heterogeneous ice nucleation data. Unlike the CNT approach, their model is based on the singular theory, or deterministic approach. In this approach it is assumed that particles have multiple nucleation sites where ice could form and the ice formation rate is determined by the most efficient nucleation site. Such that in deposition ice nucleation experiments, as soon as any of those nucleation sites reach the characteristics $RH_{ice}$, the ice will form immediately and if this characteristic $RH_{ice}$ is held constant, then no further ice nucleation events should occur, suggesting there is no time dependence”.

Table 1: Are these results for one experiment in each case? Please clarify.

Reply: Yes, these results are for one experiment in each case. The table caption is revised. It reads as follows. “One experiment was performed at each temperature and dust size”.

Table 3: Is -3 a typo? Should it be -30?

Reply: It was typo. Corrected.

Figure 5: These results also do not appear to have been corrected for possible aerosol influence. Were multiplets characterized for kaolinite generation?

Reply: Multiple charged particles are also observed in kaolinite dust sample (400 nm diameter) experiments and their contribution was less than 10%. The revised text reads as follows. “For 300, 400, and 500 nm diameter size particles, the contribution of these multiple charged particles was less than 10%. For sizes 100 and 300 nm particles, the multiple charge calculations were based on the routine experimental measurements, whereas for 400 and 500 nm particles the calculations are based on the Baron and Willeke (2001) calculations”.

Referee is correct in pointing out that multiple charged particles might bias the ice signal detected by the OPC, such that one might inadvertently count dust particles as the ice crystals. This is actually is considered when we report the measurements. We always ran the blank experiments where the CIC is set to $RH_{ice}$ = 100% (no supersaturation) conditions and count the particles at the exit of the CIC. This is considered as a background count, and the average of these counts (if > 0) are automatically subtracted from the counts when we are running the experiment ($RH_{ice}$ > 100%).

Figure 6: Most results show 1 to 1.5 order of magnitude increase of active fraction over these RH ranges, while most of the PDFs suggest > 3 orders of magnitude increase. Thus, I wonder if all of the data have been well represented by the PDF approach, although I think omitting possible aerosol noise would help.

Reply: The least RMSE fit ensures we obtained best fit (PDF parameters) between observed and modeled active fraction magnitudes. This also ensures that data have been well represented by the PDF approach.
The reported activated fractions are corrected for the artifacts from multiple charged particles that might produce aerosol noise, see the above reply.

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