

Dear Editor,

We thank reviewer 1 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 #1

General comments:

The paper is concerned with deposition ice nucleation. How do you know that the particles triggered deposition ice nucleation. Can this be validated? I assume for lower temperatures this might be correct but at -25 C maybe not.

Reply: Deposition ice nucleation mechanism is defined as the ice formation mechanism where water vapor directly deposits onto the particle surface to form the ice. These required conditions are simulated in our experiments by running the experiments at water sub-saturated conditions. Thus controlling the humidity and temperature conditions, we can only test the deposition ice nucleation efficiency of various aerosols. Previous studies have shown deposition ice nucleation at -25 degC temperatures, and also diffusion ice growth calculations show that ice crystal can grow to size larger than 1 micrometers (the ice crystal detectable limit in the present study) within ~12 seconds (maximum particle residence time within the ice nucleation chamber). This confirms that we are observing ice crystals formed only by deposition ice nucleation mechanism.

Instead of “deposition freezing” maybe use “deposition ice nucleation” since no liquid water is present in this nucleation process.

Reply: Corrected.

This manuscript could benefit from additional analysis of the experimental data with respect to classical nucleation theory as given in more detail below. This is something the authors might consider.

Reply: The comment is addressed below (see reply under P. 2494, l. 16 comment heading).

Specific comments:

p. 2490, l. 16: What is the number density of water molecules at the ice nucleus/water interface you applied?

Reply: We have used 10^{21} molecules per m^3 (Fletcher, 1962). A fixed pre-exponential constant was used and therefore, also considering comment from other reviewer, equation (4) discussions are removed.

p. 2492, l. 17: Is it justified to use the integrated number of dust particles. This would indicate that dust size dependence is neglected. However, the ice nucleation experiments indicate differences in activated fraction with different sizes, in particular at higher RH. I assume you are using the fitted dust size distribution in your model (Fig. 3)? What ice nucleation activity do you assign to particles larger 0.5 micrometer? Please elaborate here.

Reply: The integrated number of dust particles ($= 10.7 \text{ L}^{-1}$) given in the text is the sum of the fitted dust size distribution shown in Fig. 3. What was used in the model is the dust size distribution, meaning that dust size dependence is included. For larger particles, the ice nucleation probability is higher based on the CNT theory due to their larger surface area. We do not specifically assign ice nucleation activity to particles larger 0.5 micrometer.

p. 2493, l. 1: Please give a reference why you can assume that deposition ice nucleation occurs at $T < -22\text{C}$?

Reply: Here we wanted to say that the cirrus cloud was observed at temperatures colder than -22 degC . Sentence is revised to remove “since the cloud temperatures are colder than -22 degC ” for clarity. The new sentence reads as follows “The cirrus cloud case observed from the U.S. Department of Energy’s Atmospheric Radiation Measurement (ARM) program at the South Great Plain site in March 09, 2000 (Comstock et al., 2007) is chosen to test deposition ice nucleation”.

p. 2493, l. 2: Figs 4 and 5: Why is an active fraction of 1 not reached?

Reply: Ice nucleating properties of aerosols are hypothesized to depend upon many factors: size, insolubility, epitaxial and lattice structure properties, active sites, aging and processing of aerosol particles (Pruppacher and Klett, 1997). The relative importance of these factors is unknown. We find that only few dust particles have these favorable factors and majority of the particles do not satisfy ice nuclei requirements, and therefore we do not obtain 100% activation. This is also observed in many past studies (e.g. Welti et al. 2009; DeMott et al. 2011).

p. 2493, l. 20: Tables 1 and 3 do not show any uncertainties. For this reason one cannot be convinced or it cannot be stated that there is no significant difference of the data.

Reply: The uncertainties are ± 3.0 degree and ± 0.04 in mean and standard deviation parameters, respectively. Following the reviewer’s comment, we added these uncertainty errors in the revised manuscript (section 3.1; Table 1 and 2 captions).

p. 2494, l. 12: The sentence beginning “It might be that the. . .” reads awkward and should be reworded. Looking at previous literature which could be cited here, one can see that the onset for mineral dust does not change significantly with temperature within the deposition ice nucleation regime.

Reply: This paragraph is revised for clarity. See last paragraph of the section 3.1.

Referee is correct. Past studies have shown that onset nucleation efficiency of dust particles does not change significantly with temperature. This is also added in the revised manuscript (last paragraph of the section 3.1). Following sentences are added to the manuscript.

“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”.

p. 2494, l. 16: This discussion is based on classical nucleation theory and should be made clear. For example “According to classical nucleation theory larger size particles should nucleate at . . .” (reference). As a matter of fact, I believe, the authors have all the parameters to calculate expected change in RH onset when changing the particle size. Assuming a similar J_{het} (you obtained similar contact angles) you can change $S_{v,l}$ in Eq. 3 when employing a different particle size to reproduce J_{het} (or contact angles). This would be a valuable addition when doing this for all your data. It will give you an idea if classical nucleation theory can capture the observations.

Reply: We agree with the reviewer’s comment. We changed the sentence and the new sentence reads as follows, “The lack of size dependency observed here could be due to the experimental RH_{ice} uncertainty based on CNT”.

We performed sensitivity tests to calculate variation in onset RH_{ice} as dust particle size is varied keeping all other parameters constant. For larger size particles (500 nm) we see decrease in onset RH_{ice} by ~2% compared to smaller size particles (100 nm). These details are added into the revised manuscript. See last paragraph of the section 3.1. It reads as follows.

“The lack of size dependency observed here could be due to the experimental RH_{ice} uncertainty based on CNT. For example, larger size particles could nucleate at lower onset RH_{ice} than smaller size dust particles due to the differences in surface area. This premise was verified using CNT, where the sensitivity of onset RH_{ice} to the dust particle size was investigated keeping all other parameters in equations (1) to (3) constant. We observed that for 500 nm particles, the onset RH_{ice} was decreased by ~2% compared to 100 nm particles. The difference between these two onset RH_{ice} magnitudes is still less than the experimental RH_{ice} uncertainty ($\sim\pm 3\%$ in present study), and therefore we would not be able to distinguish the size effect at onset conditions”.

Section 3.2:

This paragraph may need further explanation (text or equations):

- a) Why does the onset approach result in $N_{ice}=N_0$ but only in the regime of water saturation. The experiments indicate that the onset occurs in subsaturated conditions?
- b) The PDF approach yields $N_{ice}<N_0$ at water saturation. However, it also yields ice particles at lower RH compared to the onset approach. Shouldn’t the total (accumulated) number of ice crystals derived from the PDF approach be compared to initial number N_0 ?

- c) For what reason is the water saturation regime at all discussed?
d) Figure 6, 100nm, upper panel: Why does the onset approach produces 10-3 L-1 at 140% RH_{ice}?

Reply: a) To avoid confusion we removed the water saturation regime in Fig. 6 and in the discussions. Figure 6 is updated.

b) No, because in the model we do not integrate over the activation spectrum to calculate modeled N_{ice} . The modeled N_{ice} is function of measurement conditions (size, temperature, RH_{ice}). See section 2.2.

c) We do not have the discussion about the water saturation regime.

d) Water saturation regime discussions are removed. The new figure (Fig. 6) does not have that.

p. 2496, l. 25: Maybe state the ice nucleation mechanisms which are not included.

Reply: Following the reviewer comment the sentence is revised. The new sentence reads as follows, “Other three heterogeneous ice nucleation mechanisms (condensation, immersion, and contact) are not included in the simulations. In addition, the homogeneous freezing of aerosols probably need to be included as shown in Sölch and Kärcher (2011)”.

p. 2497, l. 12: “The particles . . .” please reword. The particles do not nucleate; they might induce ice nucleation, etc.

Reply: For clarity a few points from this paragraph are merged to the last paragraph, and the original paragraph is removed.

p. 2497, l. 16: What changes in cloud radiative forcing are expected? Please elaborate and give references.

Reply: Sentence was incomplete. And as mentioned above this paragraph is been removed.

p. 2497, l. 25: “In the last case . . .” please reword. The particles do not nucleate; they might induce ice nucleation, etc.

Reply: Sentence is revised. New sentence reads as follows, “In the last case (Fig. 9c), it is evident that as more particles serve as an IN, under favorable ice nucleation conditions (low temperature and high RH_{ice}), it would lead to the larger N_i and IWC ”.

p. 2498, l. 25: What do you mean with “errors in the IN measurements”? How do you want to minimize potential error sources.

Reply: We meant if ice detection thresholds were different among the different studies this might lead to errors in the IN measurements. Since this has already been described as one source of error for the disagreement above, “errors in the IN measurements” are removed in the revised manuscript.

The error sources are highlighted in Kanji et al. (2011) and briefly added at the beginning of this paragraph.

Technical corrections:

Reply: Completed.

p. 2485, l. 22: Change “comes” to “come”.

Reply: Done

p. 2486, l. 29: Omit “the” in front of “climate”.

Reply: Done

p. 2487, l. 1: Omit “the” in front of “CNT”.

Reply: Done

p.2489, l. 14: Insert “the” before”DMA”.

Reply: Done

p. 2489, l. 25: I assume “with” should be “the”.

Reply: Yes, corrected.

p. 2490, l. 13: Change “further” to “lastly”.

Reply: Done

p. 2491, l. 12: Change table sequence: table 3 should be table 2 and vice versa.

Reply: We changed the sequence of Tables 2 and 3.

p. 2491, l. 15: Insert “off” after “read”.

Reply: Done

p. 2493, l. 20: Please check table numbers.

Reply: Corrected.

Tables: include uncertainty as mentioned above.

Reply: Done

Table 3: There is a “0” missing for experimental temperatures.

Reply: Added

Figures:

Fig. 1: Omit “and” before “size”.

Reply: Done

Fig. 2: Reword: “Active fraction of ATD particles, 400 nm in diameter, is given as a function of RH_{ice} ”.

Reply: Completed. New sentence reads as follows “Active fraction of ATD particles, at -35 degC and 400 nm in diameter (Table 2), is given as a function RH_{ice} ”

Fig. 8: A title for the color bar is missing.

Reply: Added.

References:

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