**Responses to Referees**

The authors wish to thank both reviewers for their useful suggestions and thoughtful comments. Below are our point-by-point responses to reviewers' comments.

**Referee #1**

**Reviewer's comment:** The most important point I would like to make is in regards to the use of hematite particles. Why was Hematite chosen as dust particles for cirrus clouds? Is there prior evidence of its presence in cirrus clouds?

**Authors' response:** We did not select hematite because it is a major atmospheric dust component in cirrus clouds. We chose hematite because it is a good model substance to infer $n_s$ values over a wide $T$ and $RH_{ice}$ range (i.e., page 16500 lines 9-12 and lines 16-21 and Sect. 3.3). The atmospheric relevance was demonstrated by comparing the model hematite results with more relevant desert dust aerosol results (page 16506 lines 20-25).

**Reviewer's comment:** The author refers us to the a paper by *Matsuki et al*. (2010) which examined mineral dust particles from cloud residual and clear sky in Niger. Matsuki et al. shows in Fig 4 in their paper that out of the different types of mineral dust particles that were found in cloud droplets residual, hematite frequency was very small compared to all other mineral dust particles as clay minerals.

**Authors' response:** We cited *Matsuki et al*. (2010) to state that hematite is an example of atmospheric mineral dust particles that can be found as cloud-borne particles, not specifically to infer its presence in cirrus clouds. To clarify our point, we have rephrased "cloud-borne particles" to "cloud-borne particles in shallow stratocumulus clouds" on the page 16506 line 6 in the first manuscript version (now moved to the page 16497 line 21 after "…proxy for atmospheric dust particles).

**Reviewer's comment:** In addition hematite particles are different than most of the atmospheric dust particles (in term of mineralogy and shape), can that also affect their IN ability? Since the atmosphere does not only contain hematite particle but a combination of different dust particles type, why not then combine all dust measurements and try to get an isoline that will represent all of them?

**Authors' response:** In this work, we demonstrated that the formulated hematite $n_s$-isolines are reasonably comparable to that of desert dust samples, previously studied in AIDA (i.e., ATD and SD2; *Möhler et al*., 2006), at $n_s$ ~$10^{-11}$ $m^{-2}$ (Fig. 3A and associated text on the page 16506 lines 20-25; page 16507 lines 20-22). Nonetheless, we feel it is important to point out and re-emphasize that this work is a conceptual study with laboratory-synthesized hematite particles as a model aerosol for deposition ice nucleation over a wide range of $T$ and $RH_{ice}$. In this study, we did not attempt to conclude how much hematite content contributes to ice formation in cirrus clouds. For clarity, we have added the following sentence on the page 16498 line 3 after "…existing parameterizations.":

"It is important to note that the purpose of the current study is to perform a conceptual study with laboratory-synthesized hematite particles as a model aerosol for deposition ice nucleation, over a wide range of $T$ and $RH_{ice}$, but not to quantify how much hematite content contributes to ice formation in cirrus clouds."

**Reviewer's comment:** Another general comment is that this manuscript will substantially benefit if the author will start the paragraphs with words other than figure or we, in addition there are too many places that are written in first body (e.g. page 16505 lines 4 and 29, page 16506 line 9 etc.).

**Authors' response:** The authors appreciate the prior suggestion by the reviewer on means to improve the overall quality of the paper. As per reviewer's suggestion, we have modified the following sentences (starting with words other than "figure" or "we"):

Sect. 2.2. page 16498 line 23-page 16499 line 3: "For this study, a cooling rate of 5 °C $min^{-1}$ was typically applied at the beginning. Then, the cooling rate decreased to <0.1 °C $min^{-1}$ within 400 s for each pumping expansion experiment, which was mainly due to an increasing heat flux from the chamber walls. Afterwards, an almost constant temperature was maintained by the stirred and well-mixed volume of the cold chamber. During the experiment, the pressure in the vessel decreased from 1000 to 800 mb."

Sect. 2.3. page 16500 line 27: "The mean size and surface area of hematite particles were prescribed with an assumption that…"

Sect. 3.1. page 16504 lines 3-6: "The temporal profiles of deposition nucleation experiments from HALO campaigns, including $N_{ice}$, gas $T$, $RH_{ice}$ and $RH_{water}$ measured by the TDL as well as the polarized light scattering properties in near-backscattering direction measured by SIMONE are shown in Figure 1."

Sect. 3.1. page 16504 line 20: "The initial $n_s$-isoline curves in the $T$-$RH_{ice}$ space are illustrated in Figure 2."

Sect. 3.2. page 16505 lines 27-28: "The $n_s$-isolines of hematite particles were compared to previous measurements made using different aerosol species."

Sect. 3.2. page 16506 lines 9-14: "Comparison of the hematite $n_s$-isolines to previous deposition freezing observations are shown in Figure 3. More specifically, previous measurements were performed with natural Saharan desert dust (SD2, *Möhler et al*., 2006), reference Arizona test dust (ATD, *Möhler et al*., 2006; *Welti et al*., 2009), volcanic ash (*Steinke et al*., 2011), soot (*Möhler et al*., 2005), clay minerals (*Welti et al*., 2009; *Koehler et al*., 2010) and organics (*Shilling et al*., 2006; *Wang and Knopf*, 2011)."

Sect. 3.3. page 16507 lines 24-25: "Next, the ice nucleation efficiency of hematite particles was parameterized over a wide range of $T$-$RH_{ice}$."

Sect. 3.4. page 16509 lines 8-9: "The SCAM5 results for monthly mean profiles of the simulated ice crystal number concentrations over the ARM SGP site for four cases are shown in Figure 5."

Sect. 3.4. page 16510 lines 7-8: "The results of the COSMO model for the vertical profiles of $N_{ice}$ are summarized in Figure 7. These results simulate…"

**Reviewer's comment:** A literature overview that shows the actual presence of hematite particles in cirrus clouds is missing. Only in section 3.2 the author mentions the present of hematite particles in the atmosphere but still not in cirrus clouds. It should be mentioned that *Matsuki et al*. (2010) did not mention that the dust particles measurements were taken from cirrus cloud, therefore we need to ask are we sure hematite particles can be found in cirrus clouds?

**Authors' response:** Discussed above.

**Reviewer's comment:** Additional information in the method part is needed. TSI mentioned on the SSPD 3433 web page that it can sufficient deagglomerate most dry particles in the range from 0.5 to 50 µm, in the paper the authors mention that they size selected 200nm, which is below the threshold of this equipment.

**Authors' response:** The previous AIDA study (Fig. 2 of *Skrotzki et al*., 2013) demonstrated the application of a SSPD for dry-dispersion of quasi-monodisperse (200 nm mode diameter) hematite into the AIDA vessel.

Page 16498 lines 16-17 now reads:

"A Small-Scale Powder Disperser (SSPD, TSI, Model 3433) was used to dry-disperse the quasi-monodispersed hematite particles into the AIDA vessel as demonstrated in *Skrotzki et al*. (2013)."

Reference:

Skrotzki, J., Connolly, P., Schnaiter, M., Saathoff, H., Möhler, O., Wagner, R., Niemand, M., Ebert, V., and Leisner, T.: The accommodation coefficient of water molecules on ice – cirrus cloud studies at the AIDA simulation chamber, Atmos. Chem. Phys., 13, 4451-4466, doi:10.5194/acp-13-4451-2013, 2013.

**Reviewer's comment:** In addition, did the author verify that the size that were selected by the DMA were similar to the size distribution that came out of the DMA?

**Authors' response:** DMA was not employed in our study. As described in lines 6-12 of the page 16498, we used laboratory-synthesized hematite particles that had been formed as a powder of equally sized hematite particles (~200 nm, 500 nm, 1000 nm area equivalent diameter; *Skrotzki et al*., 2013; *Hiranuma et al*., 2014). Hence, no additional size-segregation was necessary.

Reference:

Hiranuma, N., Hoffmann, N., Kiselev, A., Dreyer, A., Zhang, K., Kulkarni, G., Koop, T., and Möhler, O.: Influence of surface morphology on the immersion mode ice nucleation efficiency of hematite particles, Atmos. Chem. Phys., 14, 2315–2324, doi:10.5194/acp-14-2315-2014, 2014.

**Reviewer's comment:** Section 2.3 page 16500 line 27 to page 16501 line 3. Was a 1000nm surface area used for all three selected sizes, please clarify?

**Authors' response:** We chose only 1000 nm diameter as a simulated hematite size for our COSMO and SCAM5 modeling studies. Our choice of 1000 nm diameter and surface area is reasonable and representative for ice nucleation efficiency (inferred by $n_s$) of three different sizes of hematite particles studied at AIDA (page 16503 lines 18-25; i.e., different sizes of hematite particles were used to characterize ice nucleation efficiency, $n_s$, at a wide temperature range).

**Reviewer's comment:** The value of 1000nm that were taken from *Hiranuma et al*. (2014) does not represent size selected measurement but values from looking on the entire distribution, does the authors expect it to be the same value?

**Authors' response:** We use the term 'quasi-monodisperse' because of the presence of minor fraction of smaller agglomerates/fragments (Fig. 2 in *Skrotzki et al*., 2013; Fig. 1 in *Hiranuma et al*., 2014) within the sample, but this minor fraction have minimum influence on the surface-scaled $n_s$ parameterization presented in the current work.

The geometric standard deviation ($\sigma_g$) from *Hiranuma et al*. (2014) is correctly 1.05. What we have reported (0.097) is "width of the distribution" (= 2.303· 2 log $\sigma_g$.; *Huffman et al*., 2010). Our COSMO and SCAM5 modeling studies were performed based on either assuming monodisperse size distribution of 1000 nm or mode diameter of 1000 nm with $\sigma_g$ = 1.05. After all, the impact of introducing such distribution on the simulation results was small as presented in the manuscript (page 16509 line 28-page 16510 line 1).

For consistency, we have replaced "($\sigma$ = 0.097)" with "($\sigma_g$ = 1.05)" on page 16501 line 2.

Reference:

Huffman, J.A., Treutlein, B., and Pöschl, U.: Fluorescent biological aerosol particle concentrations and size distributions measured with an Ultraviolet Aerodynamic Particle Sizer (UV-APS) in Central Europe, Atmos. Chem. Phys., 10, 3215-3233, doi:10.5194/acp-10-3215-2010, 2010 (the supplemental Online Material is available at: http://www.atmos-chem-phys.net/10/3215/2010/acp-10-3215-2010-supplement.pdf).

**Reviewer's comment:** Page 16501 line 13 The author mentioned that for T>-36 the parameterization from M92 was used in the model, however it is known (and also mentioned on the page 16496 line 21) that M92 parameterization is relevant for -7>T>-23, therefore I am not sure if the model can represent well the creation of ice for temperature between -23<T<-36.

**Authors' response:** We agree with the reviewer that the M92 parameterization, which was developed based on the INP concentrations from CFDC observations in mid-latitudes, is strictly valid only in the temperature and supersaturation range of the measurements, i.e., between -7 and -20°C and 2 and 25% ice supersaturation. However, it has been extrapolated beyond this range in numerous previous model implementations (e.g., *Morrison et al*, 2003; *Seifert and Beheng*, 2006), because some information is needed in the whole temperature range above homogeneous freezing conditions. Here, we implemented the M92 parameterization, for $T$ > -36 °C, where hematite particles are not ice active, simply to ensure a reasonable spatio-temporal distribution of total water content (among water vapor, liquid and ice phase cloud particles) and to avoid unrealistic conditions (e.g., high supersaturations) in the lower troposphere in our model runs. Further, Figure 2 in *Seifert et al*. (2012) show the evidence that the deviation between the Phillips parameterization (*Phillips et al*., 2008) and the M92 parameterization can result in about an order of magnitude INP concentration mainly due to 'aerosol assumption' (i.e., number concentration). For these reasons, and given the inherent uncertainty of an aerosol-independent approach, we believe that the use of the M92 parameterization for $T$ > -36 °C in our conceptual modeling study is appropriate and reasonable.

References:

Morrison, H., Shupe, M. D., and Curry, J. A.: Modeling clouds observed at SHEBA using a bulk microphysics parameterization implemented into a single-column model, J. Geophys. Res., 108, doi:10.1029/2002JD002229, 2003.

Seifert, A., Köhler, C., and Beheng, K. D.: Aerosol-cloud-precipitation effects over Germany as simulated by a convective-scale numerical weather prediction model, Atmos. Chem. Phys., 12, 709–725, doi:10.5194/acp-12-709-2012, 2012.

Seifert, A. and Beheng, K.D.: A two-moment cloud microphysics parameterization for mixed-phase clouds. Part I: Model description, Meteorol. Atmos. Phys., 92, 45–66, doi:10.1007/s00703-005-0112-4, 2006.

Phillips, V.T.J., DeMott, P.J., and Andronache, C.: An empirical parameterization of heterogeneous ice nucleation for multiple chemical species of aerosol, J. Atmos. Sci., 65, 2757–2783, doi:http://dx.doi.org/10.1175/2007JAS2546.1, 2008.

**Reviewer's comment:** Page 16506 lines 4-8 I think that this part should also be mentioned in the introduction part.

**Authors' response:** The referee makes a good point. We have updated and moved the following sentence on the page 16497 line 21 after "…proxy for atmospheric dust particles":

"Hematite is used as an example of atmospheric mineral dust particles, which can also be found in the form of cloud-borne particles in shallow stratocumulus clouds (*Matsuki et al*., 2010). Natural hematite often exists in supermicron-sized silt particles and accounts for a few percent of the total dust particle mass (*Claquin et al*., 1999)."

**Reviewer's comment:** Page 16507 lines 14-22 Can the author say something about some of the points on figure 3 that does not match the $n_s$ isoline, spatially those measured by AIDA.

**Authors' response:** The reviewer is right. It may be indicative of isolines to be also as a function of composition. The authors, however, point out that the previous AIDA results for desert dust are in good agreement with the new $n_s$ isolines, at least within an order of magnitude, and their agreement also supports the general trend of the isolines. There is one outlier for SD2 at about -50°C. The reason for which is not known. Furthermore, the soot data show a somewhat less steep increase to lower temperatures. But, it confirms the general trend of isolines for deposition nucleation. These premises must be further examined in comparing to atmospherically relevant substrates and their ice nucleation activities in controlled laboratory settings.

**Reviewer's comment:** Page 16508 lines 3 I think that the part of supplement material 3 should be part of the paper, since all the measurements represent measurement below water saturation the reader should see in detail how that isonline above water saturation were calculated.

**Authors' response:** The authors prefer to keep it in the supplement. To address the reviewer's concern, we now rephrased the sentences in the page16508 lines 1-3 as:

Original: "The lower bound of $n_s$ value ($10^6$ $m^{-2}$) was set based on the minimum $n_s$ observed during AIDA expansions. The method used to constrain the $n_s$-isolines above 100% $RH_{ice}$ as discussed in the supplemental material (Fig. S3)."
→
Modified: "The lower bound of $n_s$ value ($10^6$ $m^{-2}$) was set based on the minimum $n_s$ observed during AIDA expansions. Since the certain regions of $n_s$-isolines (i.e., $n_s < 7.5$ x$10^{10}$ $m^{-2}$; blue lines in Fig. 2) can submerge below ice saturation, the correction was applied to shift them and maintain all isolines above 100% $RH_{ice}$. The procedure to constrain $n_s$ to >100% RHice is described in the supplement (Fig. S3)."

**Reviewer's comment:** Page 16508 lines 12-17 (regarding Figure 4b) Why using a third degree polynomial if it does not represent the experimental work?

**Authors' response:** In later sections (i.e., Simulation A vs. Simulation B in Figs. 5, 6 and 7 in Sect. 3.4), we demonstrate that the third degree polynomial fit parameterization yield similar simulation results (with respect to yielded ice crystal concentration) compared to the interpolated parameterization. We intended to introduce this fit owing to its model friendly aspect.

**Reviewer's comment:** Page 16509 first paragraph The whole section is not clear. It is not clear, which figure represent figure 4b? Some of the sentence needs clarification as lines 14-20. There is hardly reference to figure 6.

**Authors' response:** To clarify our study cases, we have modified and updated the texts in the following sections:

Page 16509 lines 9-13:

Original: "These include the pure homogeneous ice nucleation case (Simulation A), three cases with contributions from both the homogeneous and heterogeneous ice nucleation (hereafter combined case) described in Fig. 4 (Simulations B, C and D) and the simulation of the different lower boundaries of RHice ($RH^*_i$, Simulation D)."
→
Modified: "These include (Case 1) the pure homogeneous ice nucleation case, (Case 2-4) cases with contributions from both homogeneous and heterogeneous ice nucleation (hereinafter referred to as the combined case) described in Fig. 4a-c (corresponding to Simulations A, B and C) and (Case 5) the simulation of the different lower boundaries of $RH_{ice}$ ($RH^*_i$, Simulation D)."

Page 16509 lines 18-20:

Original: "The differences between the four parameterizations used in this study are small for both the combined cases and the pure heterogeneous cases."
→
Modified: "The differences between the three parameterizations derived from AIDA measurements, corresponding to Simulations A, B and D, are small for both the combined case and the pure heterogeneous ice nucleation case as presented in Fig. 6."

Page 16509 line 25:

Original: "The three parameterizations have largest differences…"
→
Modified: "The three parameterizations (i.e., Simulations A, B and D) have largest differences…"

**Reviewer's comment:** Discussions: The first paragraph is not so clear, the SCF was mentioned only briefly in secession 3.2. I recommend the authors to rewriting this paragraph again.

**Authors' response:** We now updated the first paragraph on the page 16511:

"As described in the previous section (Sect. 3.1), deposition mode freezing cannot solely explain the $n_s$-isoline observation below water saturation (-50 ˚C < T < -36 ˚C in Fig. 2). Although we presumed that SCF acts as a subset of immersion freezing and plays an important role in this region, further insight and evidence of SCF beyond cloud simulation chamber observations are required to correctly understand the contributions of both homogeneous and heterogeneous nucleation. High-resolution microscopic techniques with an integrated continuous cooling setup are needed to visualize the freezing process of a single particle and to fully understand the complex freezing processes involved in SCF on particle surfaces."

We feel the definition of SCF given in Sect. 3.1 (page 16505 lines 14-25) is sufficient and no repetitive discussion is necessary.

**Reviewer's comment:** Page 16511 lines 21-23 Not all the points on figure 3 match the hematite particles therefore I do not think it is correct to assume that all dust will behave in the same way is hematite particles.

**Authors' response:** To address the referee's concern, we now rephrased the related section.

Page 16511 lines 21-23:

Original: In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) provides indication on the validity of the assumption to treat all dust as hematite in deposition mode."
→
Modified: "In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) suggests that hematite has similar ice nucleation efficiency, inferred by $n_s$, as dust. The comparison between the observed profile of ice crystal number concentrations and the simulated ones (Figs. 5 and 6) also suggests the validity of the new parameterizations. These premises must be further examined in comparing to atmospherically relevant substrates (fresh and aged ones) and their ice nucleation activities in laboratory settings. In situ INP measurements, such as the number concentration and the types of INPs, at the upper troposphere can also help to constrain the parameterizations."

**Reviewer's comment:** Table 1:
Organize the table in a way that will be easier on the reader, for example based on *T* from warm to cold *T* or particle size.

**Authors' response:** Table 1 is now organized based on T(Evaluated $n_s$) - from low to high T(Evaluated $n_s$).

Table 1. Summary of aerosol measurements and AIDA ice nucleation experiments. All HALO experiments are from *Skrotzki et al.* (2013).

| | Aerosol Measurements | | | Ice Nucleation Measurements | | | | |
|---|---|---|---|---|---|---|---|---|
| Experiment ID | Hematite Diameter, nm | Total Number Conc., $cm^{-3}$ | Total Surface Conc., $\mu m^2\ cm^{-3}$ | Examined T Range, °C | Examined $RH_{ice}$ Range, % | Evaluated $n_s$, $m^{-2}$ | T(Evaluated $n_s$), °C | $RH_{ice}$(Evaluated $n_s$), % |
| HALO05_24 | 200 | 115.0 | 14.4 | -76.1 to -81.9 | 100.6 to 164.8 | $10^{11}$ | -78.2 | 136.4 |
| HALO04_09 | 500 | 112.5 | 26.9 | -75.8 to -80.1 | 100.3 to 149.8 | $10^{11}$ | -77.5 | 128.3 |
| HALO04_05 | 500 | 142.2 | 30.9 | -61.8 to -65.5 | 100.2 to 135.6 | $10^{11}$ | -62.6 | 111.1 |
| HALO05_18 | 200 | 161.9 | 21.8 | -60.3 to -65.2 | 100.1 to 124.5 | $10^{11}$ | -60.8 | 106.0 |
| HALO06_22 | 200 | 145.7 | 19.2 | -50.2 to -53.9 | 100.3 to 123.4 | $10^{11}$ | -50.7 | 106.7 |
| HALO06_21 | 200 | 245.0 | 32.9 | -50.3 to -53.8 | 100.4 to 115.8 | $10^{11}$ | -50.5 | 102.2 |
| INUIT01_26 | 1000 | 342.1 | 749.0 | -41.0 to -47.1 | 100.2 to 103.9 | $10^{10}$ | -41.2 | 102.2 |
| HALO06_20 | 200 | 168.7 | 22.4 | -39.8 to -44.4 | 100.4 to 128.8 | $10^{10}$ | -40.7 | 111.3 |
| HALO06_19 | 200 | 283.0 | 42.9 | -39.7 to -44.5 | 100.2 to 121.6 | $10^{10}$ | -40.6 | 109.2 |
| INUIT04_08 | 1000 | 193.0 | 647.0 | -39.3 to -45.4 | 100.0 to 113.2 | $10^{10}$ | -40.4 | 110.1 |
| INUIT04_10 | 1000 | 161.7 | 546.6 | -37.5 to -43.7 | 100.0 to 124.1 | $10^{10}$ | -40.1 | 123.3 |
| INUIT01_30 | 1000 | 414.5 | 889.7 | -34.6 to -42.0 | 100.2 to 127.1 | $2.5 \times 10^{8}$ | -37.0 | 122.8 |

**Reviewer's comment:** Please add to the table a column that will give information on which of experiments were taken from Skrotzki et al. (2013) and which one from Hiranuma et al. (2014) etc.

**Authors' response:** We have updated the table text as "Table 1. Summary of aerosol measurements and AIDA ice nucleation experiments. All HALO experiments are from *Skrotzki et al*. (2013).". Further information is given in text on the page 16503 lines 13-17.

**Reviewer's comment:** No information regarding how the evaluated $n_s$ values of $T$ and RH is mentioned in the text, why and how these value were chosen?

**Authors' response:** We thank the reviewer for pointing out this error. We have added information regarding "evaluated $n_s$, $T$ and $RH_{ice}$", which were used to verify size-independency of the $n_s$ values (page 16503 lines 18-25), in the following sections:

Page 16503 line 21

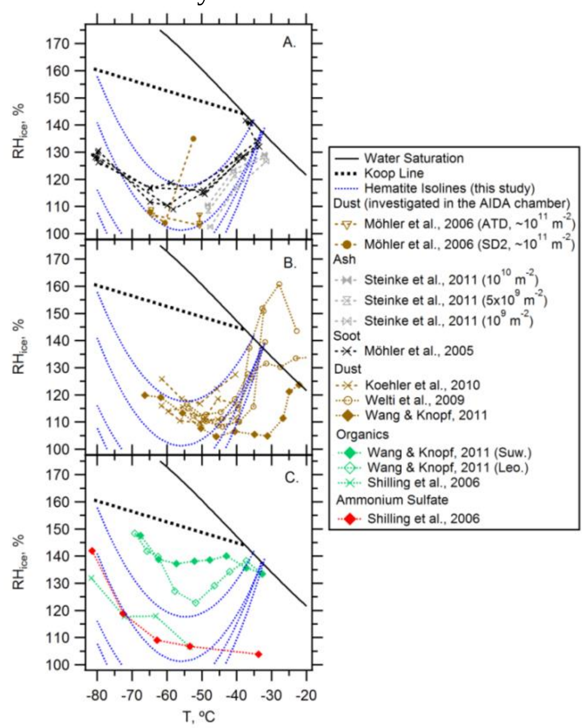
Original: "the $n_s$ values"
→
Modified: "the evaluated $n_s$ values"

Page 16503 lines 22-23

Original: "(i.e., INUIT04_08, 1000 nm, HALO06_19,200 nm, and HALO06_20, 200 nm)"
→
Modified: "(see corresponding $RH_{ice}$ and $T$ at Evaluated $n_s$ in Table 1 for INUIT04_08, 1000 nm, HALO06_19, 200 nm, and HALO06_20, 200 nm)"

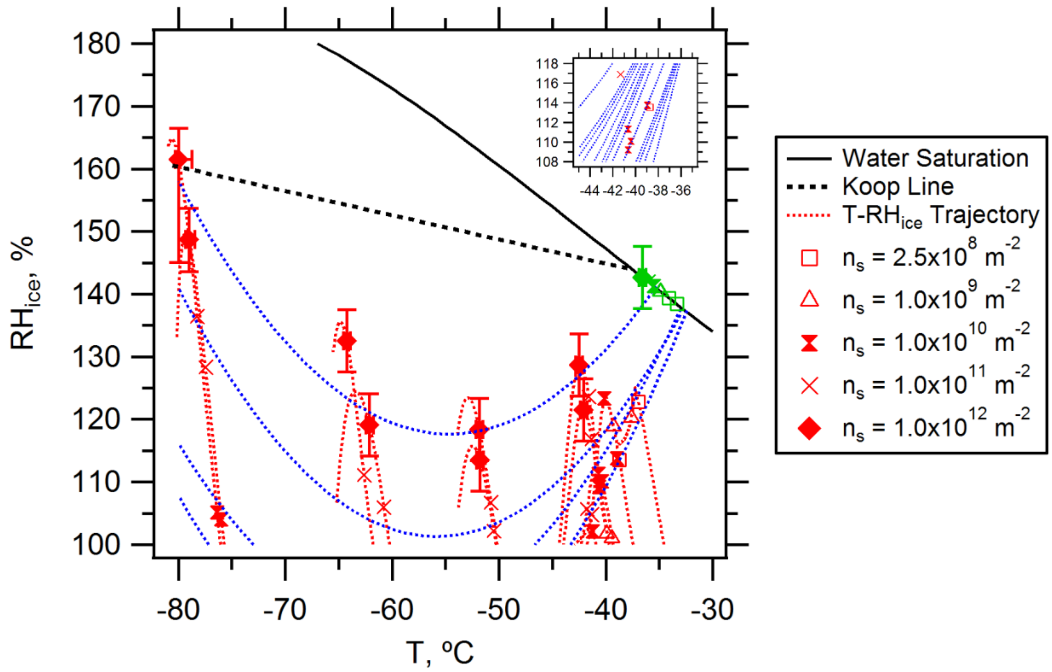
**Reviewer's comment:** Figure 2 and 3
It will be better to increase the figures size on the expend of the legend which take too much space of the figure, which does not allow to see all the values.

**Authors' response:** Modified with smaller symbols.



**Reviewer's comment:** Figure 2
It is very hard to distinguish between the different values; it might be the red color, especially around -40°C where the points are overlapping one another. Please mark them in a different marker or color. Please also mark in a different color the immersion freezing measurement so it will be easy to identify them from the one from this work (mentioned also on the page 16504 lines 23).

**Authors' response:** We modified Figure 2 with smaller symbols as well as its caption as:




Original caption: "…The data on the water saturation line represents the previously reported results of immersion freezing (*Hiranuma et al*., 2014)."
→
Modified: "…The data indicated by green color on the water saturation line represent the previously reported results of immersion freezing (*Hiranuma et al*., 2014)."

**Reviewer's comment:** Figure 5 and 6
It is very hard to distinguish between some of the lines. Perhaps using another color indicator or showing only pressure of 0-600 will highlight the differences. An addition line which will represent observation will be a nice addition for this plot, something that will represent the observation to show how the new parameterization is compared to it.

**Authors' response:** This is a good suggestion. We added the observed profiles of ice crystal number concentrations in the figure. The observational data were collected over the SGP site on eight days of April 2010 during the Small PARTicles In CirrUS (SPARTICUS) campaign (*Zhang et al*., 2013). See the figure 5&6 captions for details. Please note that for the aircraft measurements, we only consider samples when ice crystal number is above the detection limit (*Zhang et al*., 2013). Accordingly, for modeled profiles we replaced the grid-box mean values with the in-cloud values.

Reference:


Zhang, K., Liu, X., Wang, M., Comstock, J. M., Mitchell, D. L., Mishra, S., and Mace, G. G.: Evaluating and constraining ice cloud parameterizations in CAM5 using aircraft measurements from the SPARTICUS campaign, Atmos. Chem. Phys., 13, 4963-4982, doi:10.5194/acp-13-4963-2013, 2013.

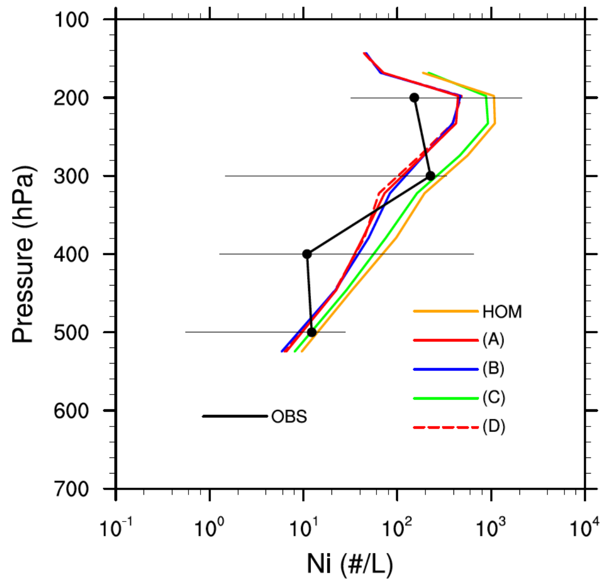



Figure 5. Monthly mean profiles of the simulated in-cloud ice crystal number concentrations (Ni) over the ARM SGP site. The four cases shown in the figure include the pure homogeneous ice nucleation case (HOM) and four combined (heterogeneous + homogeneous) ice nucleation cases: (A) AIDA Interp. + Homogeneous; (B) AIDA Fit + Homogeneous; (C) P13 + Homogeneous; and (D) AIDA Interp. ($RH^*_i$=105%) + Homogeneous. Black dots show the observed mean profile of Ni. Left and right ends of the horizontal bars indicate the $10^{th}$ and $90^{th}$ percentiles of the observed Ni values at each pressure level.

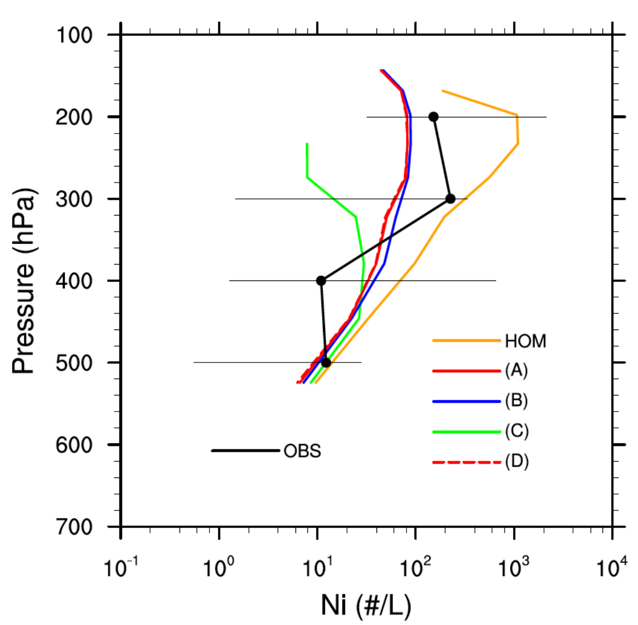



Figure 6. Monthly mean profiles of the simulated in-cloud ice crystal number concentrations over the ARM SGP site. The four cases shown in the figure include the pure homogeneous ice nucleation case (HOM) and four pure heterogeneous ice nucleation cases: (A) AIDA Interp.; (B) AIDA Fit; (C) P13; and (D) AIDA Interp. ($RH^*_i$=105%). Black dots show the observed mean profile of Ni. Left and right ends of the horizontal bars indicate the $10^{th}$ and $90^{th}$ percentiles of the observed Ni values at each pressure level.

We added the following sentence on the page 16509 line 13 after "… Simulation D).":

"In addition, the observed profile of ice crystal number concentrations is also shown in comparison to the simulations in Fig. 5. The observational data were collected over the SGP site on eight days of April 2010 during the Small PARTicles In CirrUS (SPARTICUS) campaign (*Zhang et al*., 2013)."

We added the following sentence on the page 16509 line 18 after "… heterogeneous nucleation.":

"The observed mean profile of in-cloud ice crystal number concentrations is in agreement with the simulated ones."

We also modified the sentences on the page 16511 lines 20-23:

Original: In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) provides indication on the validity of the assumption to treat all dust as hematite in deposition mode."
→
Modified: "In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) suggests that hematite has similar ice nucleation efficiency, inferred by $n_s$, as dust. The comparison between the observed profile of ice crystal number concentrations and the simulated ones (Figs. 5 and 6) also suggests the validity of the new parameterizations. These premises must be further examined in comparing to atmospherically relevant substrates (fresh and aged ones) and their ice nucleation activities in laboratory settings. In situ INP measurements, such as the number concentration and the types of INPs, at the upper troposphere can also help to constrain the parameterizations."

**Reviewer's comment:** Consider changing figure 5 and 6 to one figure as 5a and b, it will be easier for the reader to understand the comparison of the two.

**Authors' response:** We use another alphabetical sub-category (Simulations A-D), and we feel introducing another one will be confusing. We now modified the text and introduced Figure 6 explicitly at the reviewer's prior suggestion. So we would like to keep figure numbers as is.

**Additional revision:** In addition to addressing the reviewer's comments, other editorial corrections (major and non-miscellaneous ones) were made as below.

Since a temperature is a numerical measure, we would like to replace "warm or cold" with "high or low": page 16494 line 14, page 16494 line 16, page 16494 line 21, page 16500 line 21, page 16504 line 16, page 16511 line 13.

Abstract

Page 16494 lines 4-5: "critical in order to" → "important to"
Page 16494 line 8: "water saturated conditions that were realized" → "water subsaturated conditions. These conditions were achieved"

Introduction

Page 16495 line 2: "constitute" → now rephrased to "represent a"
Page 16495 lines15-19: now reads, "Briefly, deposition mode induces ice formation when water vapor is directly deposited onto the INP, immersion and condensation freezing can induce ice formation when

freezing is initiated by the INP immersed within the supercooled droplet or solution droplet, and contact freezing can initiate at the moment when an INP comes into contact with a supercooled droplet."
Page 16495 line 21: "is accounted for by" → now reads, "results from"
Page 16495 lines 22-24: "However, representativeness of freezing mechanisms in cirrus clouds is still ambiguous (e.g., *Sassen and Khvorostyanov*, 2008)." → now reads, "However, freezing mechanisms in cirrus clouds are still uncertain (e.g., *Sassen and Khvorostyanov*, 2008)."
Page 16496 line 19: "descriptions such as *Meyers et al*. (1992, henceforth M92)" → now reads, "descriptions given in *Meyers et al*. (1992, hereinafter referred to as M92)"
Page 16497 lines 4-6: now reads, "They showed that the heterogeneous INP number concentration obtained from a CNT-based parameterization is typically higher by several factors than that of Phillips's parameterization under identical test conditions"
Page 16497 lines 19-20: "Herein, as part of the Ice Nucleation research UnIT (so called INUIT), we conducted a comprehensive investigations on examining the" → now reads, "Within the framework of Ice Nucleation research UnIT (INUIT), we comprehensively investigated"
Page 16497 lines 26-28: "Applications of the fitted $n_s$ parameterization derived from these measurements to atmospheric modeling simulations were also performed." → now reads, "In addition to developing new dust parameterizations from these AIDA measurements (Sects. 3.1-3.3), the fitted $n_s$ parameterization was also applied to atmospheric modeling simulations (Sect. 3.4)."

Method

Page 16498 lines 7-10: "well defined surface area, and are therefore good to investigate and relate $T$-$RH_{ice}$ dependent ice nucleation efficiency to surface area (*Hiranuma et al*., 2014)." → now reads, "well-defined surface area. Hence, they are suited well for investigating $T$-$RH_{ice}$-dependent ice nucleation efficiency and relating it to the surface area (*Hiranuma et al*., 2014)."
Page 16500 line 19: "complementally covering" → now reads, "that complement each other and cover"
Page 16501 lines 12-14: "was used to simulate ice formation of background particles at $T$ > -36°C. In specific, the aerosol-independent M92 scheme was used in this study. A combination of these parameterizations was advantageous…" → now reads, "was used to simulate ice formation of background particles at $T$ > -36°C, namely, the aerosol-independent M92 scheme. These parameterizations were combined…"
Page 16501 line 18: "To better understand how the AIDA $n_s$-based parameterization compares to" → now reads, "To better understand to what an extent the AIDA $n_s$-based parameterization differs from"
Page 16502 lines 4-8: now reads, "The single column model resembles a single column of a GCM and can be based on either observational data or suitable model output data. The complex feedbacks between the simulated column and other columns due to large-scale dynamics, except cloud detrainment from shallow and deep convection, are not considered. Consequently, the single column model is an ideal tool for testing ice cloud parameterizations."
Page 16502 lines 20-21: "More specifically, COSMO is the high resolution limited-area model, which allows an assessment of clouds" → now reads, "COSMO is the high-resolution limited-area model to assess clouds"

Results

Page 16503 lines 13-14: "In the present study we also used the AIDA results reported in *Skrotzki et al*. (2013) and reconciled to $n_s$ values" → now reads, "In addition, we used the AIDA results reported by *Skrotzki et al*. (2013) and reconciled them with the $n_s$ values"
Page 16503 lines 25-26: "Further, an advantage of using 1000 nm diameter hematite particles was that, being of , comparatively larger surface area," → now reads, "The advantage of using 1000 nm diameter hematite particles was that, due to their comparatively larger surface area,"
Page 16505 line 16: "Supportively," → now reads, "To support this,"

Page 16505 line 21: "SCF may have plausible relevance" → now reads, "SCF may be of relevance"
Page 16507 lines 14-16: now reads, "As seen in Figure 3, the results from previous studies suggest the necessity of increasing $RH_{ice}$ to maintain a constant $n_s$ value below $T$ ~-55°C. They also indicate that nucleation may be triggered by SCF…"
Page 16508 lines 7-8: "are necessary in order to" → now reads, "are required to"
Page 16508 lines 17-20: now reads, "The third approach (Fig. 4c) consisted in applying the equivalent $n_s$ for deposition nucleation of hematite particles parameterized using the method introduced by *Phillips et al*. (2008 and 2013). In detail, we characterized the nucleation activity…"
Page 16509 lines 13-14: "We observe that ice crystal formation from" → now reads, "It is found that ice crystal formation due to"
Page 16509 line 24: "balances" → now rephrased to "compensates"
Page 16510 line 21: "This enables us to estimate" → "This segregation allows for an estimation of"

Discussion

Page 16511 line 14: "and deserves more attention." → now reads, ", and this highlights the need for further investigation."
Page 16511 lines 14-21: "Substantial differences between the empirical approach of P13 and our parameterization developed in this study are presumably attributed to the difference in lab- or field data, highlighting the need for further characterizations of atmospherically relevant substrates and their ice nucleation activities in laboratory settings. Nevertheless, *Niemand et al*. (2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation. → now reads, "Nevertheless, the previous study (e.g., *Niemand et al*., 2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation."
Page 16511 line 24 – Page 16512 line 2: now reads, "Finally, to further develop more atmospherically relevant parameterizations other than fit-based parameterizations with artificial test aerosol, the relationship between $1/T$ and ln $S_{ice}$ for a constant nucleation rate or $n_s$ based on the CNT can be analyzed (i.e., Eqns. A10-A11 in *Hoose and Möhler*, 2012). In this way, the composition specific $n_s(T\text{-}S_{ice})$ values, where the transition from SCF to deposition nucleation (or vice versa) occurs, may be better defined and can be then be used as an inexpensive model friendly parameterization."

Conclusion

Page 16512 lines 21-23: "Elaborating observed suppression of SCF near water saturation and enlightening the physical processes on observed transitions in nucleation modes for various types of atmospheric particles are important as future works." → now reads, "The observed active SCF near water saturation and physical processes at the transitions of nucleation modes still remain to be studied in detail for various types of atmospheric particles."
Page 16513 lines 1-5: now reads, "Our new parameterization revealed a minimum deviation of $N_{ice}$ values estimated by SCAM5 at minimum $RH_{ice}$ values for ice formation (100% or 105%) compared to COSMO. This deviation suggests different sensitivities of the model to the lower bound of the $RH_{ice}$ value owing to the presence of the model-resolved supersaturation…"
Page 16513 line 7: "suppression in ice nucleation" →now rephrased to "more ice nucleation"

References

Pages 16514-16519: All available doi numbers are now added in references.
Page 16517: Mishra et al., which is a study on optical properties, has been excluded.

**Referee #2**

**Reviewer's comment:** 1) Justification for using hematite to represent "atmospheric dust" at the start of the paper, or stating this as a point of evaluation in this manuscript.

**Authors' response:** We have updated the last paragraph of the introduction section with some more clarification on why we selected hematite particles as a test substrate as well as a proxy of atmospheric dust in this study.

We chose hematite because it is a good model substance to infer $n_s$ values over a wide $T$ and $RH_{ice}$ range (i.e., page 16500 lines 9-12 and lines 16-21 and Sect. 3.3). The atmospheric relevance was demonstrated by comparing the model hematite results with more relevant desert dust aerosol results (page 16506 lines 20-25).

In this work, we demonstrated that the formulated hematite $n_s$-isolines are reasonably comparable to that of desert dust samples, previously studied in AIDA (i.e., ATD and SD2; *Möhler et al*., 2006), at $n_s \sim 10^{-11}$ $m^{-2}$ (Fig. 3A and associated text on the page 16506 lines 20-25; page 16507 lines 20-22). Nonetheless, we feel it is important to point out and re-emphasize that this work is a conceptual study with laboratory-synthesized hematite particles as a model aerosol for deposition ice nucleation over a wide range of $T$ and $RH_{ice}$. In this study, we did not attempt to conclude how much hematite content contributes to ice formation in cirrus clouds. For clarity, we have added the following sentence on the page 16498 line 3 after "…existing parameterizations.":

"It is important to note that the purpose of the current study is to perform a conceptual study with laboratory-synthesized hematite particles as a model aerosol for deposition ice nucleation, over a wide range of $T$ and $RH_{ice}$, but not to quantify how much hematite content contributes to ice formation in cirrus clouds."

**Reviewer's comment:** 2) Recognizing the fact that P13 is at least inclusive of some atmospheric INP data, so the mystery of it not agreeing with lab data below -40 ˚C is neither a new discovery, nor is it explained by new data presented herein. As an explanation of lower temperature discrepancies with laboratory dust data, P13 offered that coatings are common and may limit deposition nucleation. This may not be the full reason, but this paper needs to better explain or speculate as to why the modeling community should use the new parameterization in preference to P13.

**Authors' response:** The referee makes a good point in stating the fact that we are not working on the premise that there is one true dust-derived $n_s(T, S_i)$ and thus we cannot conclusively say which parameterization does better job for predicting/estimating dust-derived INPs. To address referee's concern, we modified the relevant texts and also soften the tense of P13 vs. our new parameterization in the following sections:

Page 16510 lines 24-27:

Original: "Our result shows less ice crystal formation with P13 compared to the AIDA $n_s$-isoline-based parameterization. The observed discrepancy between the new parameterization and P13 may largely reflect the difference in parameterization based on lab- or field data. Furthermore, strong supersaturation dependence of $n_s$ at cold $T$ was not well constrained by P13, presumably due to a limited amount of data." →
Modified: "Our results show diversity between P13 and the AIDA $n_s$-isoline-based parameterization. Ice crystal formation was less for P13 and more for the new parameterization. A possible explanation for the observed deviation may be due to the difference in parameterization based on lab- or field data. For

instance, atmospheric aging and processing (i.e., surface coating and associated heterogeneous surface reactions) may have altered ice-nucleating propensity and limited deposition nucleation of dust-derived INPs in the P13 parameterization for the field data-derived parameterization as discussed in *Phillips et al*. (2008)."

Page 16511 lines 14-23:

Original: "Substantial differences between the empirical approach of P13 and our parameterization developed in this study are presumably attributed to the difference in lab- or field data, highlighting the need for further characterizations of atmospherically relevant substrates and their ice nucleation activities in laboratory settings. Nevertheless, *Niemand et al*. (2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation. In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) provides indication on the validity of the assumption to treat all dust as hematite in deposition mode."
→
Modified: "Nevertheless, the previous study (e.g., *Niemand et al*., 2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation. In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) suggests that hematite has similar ice nucleation efficiency, inferred by $n_s$, as dust. The comparison between the observed profile of ice crystal number concentrations and the simulated ones (Figs. 5 and 6) also suggests the validity of the new parameterizations. These premises must be further examined in comparing to atmospherically relevant substrates (fresh and aged ones) and their ice nucleation activities in laboratory settings. In situ INP measurements, such as the number concentration and the types of INPs, at the upper troposphere can also help to constrain the parameterizations."

Page 16509 lines 3-6:

Original: "To conclude, the discrepancy between a new parameterization and P13 is substantially large, and the consequence of this discrepancy towards cloud properties is demonstrated in the following section."
→
Modified: "AIDA $n_s$-isoline-based parameterization suggests strong supersaturation dependence of $n_s$ at low *T*. Observed diversity between a new parameterization (Figs. 4a and 4b) and P13 (Fig. 4c) may result in different ice crystal forming propensities and may predict different cloud properties. The potential consequence of observed diversity is demonstrated using conceptual models and discussed in the following section."

In addition, we also moved the following discussion of upper and lower *T* boundary (originally page 16508 line 24- page 16509 line 3) and inserted to the page 16508 line 17, after "… shown in Fig. 4b." since these boundaries are more relevant to $n_s$ fit presented in Fig. 4b:

"Note that the upper temperature boundary of -36 ˚C was assigned as the interface between immersion mode- and deposition mode ice nucleation (*Hiranuma et al*., 2014), and the lower boundary of −78 ˚C is the limit introduced by interpolating the hematite-isoline curves."

**Reviewer's comment:** 3) Relatedly, the parameterization seems to drastically limit ice supersaturation at low temperatures in atmospheric situations, while still producing ample ice crystals. Some discussion of the realism of that result should be added.

**Authors' response:** The main reason for the higher ice number concentrations despite lower supersaturations is that our laboratory-based parameterization yields higher values of $n_s$ at e.g. at ~110% than the P13 parameterization gives at e.g. ~130%. In addition, also with the new parameterization, supersaturations up to 120% are reached occasionally. One possible explanation for this simulated high ice formation is the contribution of updrafts, where supersaturation conditions are maintained. Nevertheless, since atmospheric measurements of supersaturations in cirrus regions seem still uncertain, and high atmospheric supersaturations are still under debate (e.g., *Peter et al*., 2006; *Krämer et al*., 2009), we cannot say for certain about the realism of this feature and, therefore, prefer no discussion in the current manuscript.

References:

Peter, T., Marcolli, C., Spichtinger, P., Corti, T., Baker, M. B. and Koop, T.: When dry air is too humid, Science, 314, 1399-1402, doi:10.1126/science.1135199, 2006.

Krämer, M., Schiller, C., Afchine, A., Bauer, R., Gensch, I., Mangold, A., Schlicht, S., Spelten, N., Sitnikov, N., Borrmann, S., de Reus, M., and Spichtinger, P.: Ice supersaturations and cirrus cloud crystal numbers, Atmos. Chem. Phys., 9, 3505-3522, doi:10.5194/acp-9-3505-2009, 2009.

**Reviewer's comment:** Lines 21-24, but also a general comment: The major question this begs is why we should deem that hematite is representative for cirrus, and why we should deem that the lab studies are relevant for atmospheric particles? This seems like a topic in itself.

**Authors' response:** We did not select hematite because it is a major atmospheric dust component in cirrus clouds, but we chose hematite because it is a good model substance to infer $n_s$ values over a wide $T$ and $RH_{ice}$ range (discussed above). The compatibility between hematite particles and desert dust particles has already been demonstrated in the first version of our manuscript (i.e., Fig. 3A and associated text on the page 16506 lines 20-25; page 16507 lines 20-22).

To further clarify, we have updated the abstract. Modifications are performed to the old text as follows:

Page 16494 lines 5-7:

Original: "The surface-scaled ice nucleation efficiencies of hematite particles, inferred by $n_s$, were derived from AIDA…"
→
Modified: "The ice nucleation active surface-site density ($n_s$) of hematite particles, used as a proxy for atmospheric dust particles, were derived from AIDA…"

Page 16494 lines 16-19:

Original: "We implemented new $n_s$ parameterizations into two cloud models to investigate its sensitivity and compare with the existing ice nucleation schemes towards simulating cirrus cloud properties."
→
Modified: "We implemented new hematite-derived $n_s$ parameterizations, which is comparable to the previous AIDA measurements of desert dust, into two conceptual cloud models to investigate their sensitivity in comparison to the existing ice nucleation schemes for simulating cirrus cloud properties."

**Reviewer's comment:** The parameterization you choose to compare to is constrained by measurements of atmospheric IN, and it notes already that the results are inconsistent with cloud chamber data for pure mineral dusts. Hence, it should be understood that this is not a new story, but a remaining mystery of sorts.

This paper may shed some additional light on it due to the fact that it is hard to imagine a different situation for any pure dust aerosol, so perhaps the atmospheric measurements had some unresolved issues. Nevertheless, this paper needs to state more clearly what its goals are besides the new dust parameterization.

**Authors' response:** For clarity, we have modified the following sentence:

Page 16497 lines 26-28: "Applications of the fitted $n_s$ parameterization derived from these measurements to atmospheric modeling simulations were also performed." → now reads, "In addition to developing new dust parameterizations from these AIDA measurements (Sects. 3.1-3.3), the fitted $n_s$ parameterization was also applied to atmospheric modeling simulations (Sect. 3.4)."

In addition, we have added the following sentence on the page 16498 line 3 after "…existing parameterizations.":

"It is important to note that the purpose of the current study is to perform a conceptual study with laboratory-synthesized hematite particles as a model aerosol for deposition ice nucleation, over a wide range of $T$ and $RH_{ice}$, but not to quantify how much hematite content contributes to ice formation in cirrus clouds."

**Reviewer's comment:** Page 16496, lines 8-10: Regarding this statement, I do not understand what is meant by Welti et al. introducing this idea. The concept of a freezing mechanism of solutions on particles at below water saturation has a history in going back to Zuberi et al. (2002), Archuleta et al. (2005) and so forth. Welti et al. referenced this earlier work (exploring it in the context of CNT), as does the present paper later in the manuscript.

**Authors' response:** We thank the reviewer's suggestion for these useful literatures (*Zuberi et al*., 2002; *Hung et al*., 2003; *Archuleta et al*., 2005) and are now added on the page 16496 lines 8-10:

"Recently, *Welti et al*. (2014) introduced the relevance of soluble components of mineral dust (i.e., Fluka kaolinite) to condensation freezing below water saturation."
→
"Previous laboratory studies introduced the concept of a freezing mechanism of solutions on particles at below water saturation (*Zuberi et al*., 2002; *Hung et al*., 2003; *Archuleta et al*., 2005). More recently, *Welti et al*. (2014) explored the relevance of soluble components of mineral dust (i.e., Fluka kaolinite) to condensation freezing below water saturation in the context of classical nucleation theory (CNT)."

The CNT abbreviation is now used in the page 16496 line 24:

"Besides, classical nucleation theory (CNT)-based" → "Besides, CNT-based"

Added references:

Archuleta, C.M., DeMott, P.J., and Kreidenweis, S.M.: Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures, Atmos. Chem. Phys., 5, 2617-2634, doi:10.5194/acp-5-2617-2005, 2005.

Zuberi, B., Bertram, A.K., Cassa, C.A., Molina, L.T., and Molina, M.J.: Heterogeneous nucleation of ice in $(NH_4)_2SO_4-H_2O$ particles with mineral dust immersions, Geophys. Res. Lett., 29, 1504, doi:10.1029/2001GL014289, 2002.

Hung H.-M., Malinowski, A., and Martin, S. T.: Kinetics of heterogeneous ice nucleation on the surfaces of mineral dust cores inserted into aqueous ammonium sulfate particles, J. Phys. Chem. A., 107, 1296–1306, doi: 10.1021/jp021593y, 2003.

**Reviewer's comment:** Page 16497, lines 7-9: There is no doubt that there is a need for better parameterizations for cloud models, but an issue not mentioned is if one can be certain that laboratory measurements are representative for the atmosphere. So actually, what this seems to argue for are more and better in situ measurements. Apparently, these may not be as straightforward as setting conditions in instruments and measuring INP. At least, I think the authors should be open to all possibilities.

**Authors' response:** The reviewer makes a good point. The efforts to constrain model uncertainties from both in situ measurements and lab experiments are important and might be complementary. We updated the text in the following section to address the referee's comment and to clarify our point.

Page 16497 lines 7-9:
Original: "Therefore, systematic laboratory measurements to develop water subsaturated ice nucleation parameterizations for the range of atmospherically relevant $T$ -$RH_{ice}$ conditions are needed to better represent ice nucleation processes in cloud models."
→
Modified: "To gain insight on what triggers such deviation and to constrain model uncertainties, more and better in situ measurements are necessary (*Cziczo and Froyd*, 2014). In specific, identifying and quantifying sources, global spatio-temporal distribution and mixing-state of INPs might support to reduce model assumptions. In parallel, systematic laboratory measurements are indeed needed to develop water subsaturated ice nucleation parameterizations for the range of atmospherically relevant $T$-$RH_{ice}$ conditions for a better representation of ice nucleation processes in cloud models and to support in situ measurements."

Added reference

Cziczo, D.J. and Froyd, K.D.: Sampling the composition of cirrus ice residuals, Atmospheric Research, 142, 15−31, doi: 10.1016/j.atmosres.2013.06.012, 2014.

**Reviewer's comment:** Page 16498, line 6: What is the basis for selecting hematite particles as a proxy in this case?

**Authors' response:** Their uniform chemico-physical properties, such as shape (cubic), chemical composition ($Fe_2O_3$) and size (~200, ~500 and ~1000 nm diameter), which makes them as suitable test substrates to characterize ice nucleation efficiency ($n_s$) at a wide temperature range (i.e., page 16503 lines 18-25).

We have made the following minor modification on text (page 16498 line 6):

Original: "Laboratory-generated cubic hematite particles…"
→
Modified: "Laboratory-generated cubic hematite particles that have homogeneous chemico-physical properties…"

**Reviewer's comment:** Page 16501, lines 13-14: I am a bit surprised at the selection of use of the aerosolindependent M92 scheme to represent heterogeneous nucleation when applying a dust parameterization for cirrus levels. Can you please state if the values predicted by M92 are capped at some low temperature, or is it extended far beyond its usual valid (where data were represented in the original paper) temperature range? Is there a reason that the immersion freezing parameterization for hematite was not joined to the one for deposition?

**Authors' response:** We used the M92 parameterization only for $T$ >-36 ˚C (discussed above; see the authors' response to the comment from reviewer 1 on the page 4). Immersion freezing is considered as a part of SCF (details discussed below) and incorporated in our $n_s$-isoline-based parameterization. Use of the M92 parameterization for T>-36°C is necessary in the model applications because hematite is a rather inefficient ice nucleus in this temperature range, where other INP (e.g., biological ice nuclei, feldspars, etc.) are likely to contribute significantly to ice formation. If the M92 parameterization was not used in this range, too much supersaturated air and supercooled water would be transported to cirrus levels, and the impact of INP in cirrus clouds would be overestimated.

**Reviewer's comment:** Page 16501, lines 21-24: Further clarification is also needed regarding the use of P13. First, does the parameterization allow initialization otherwise in accord with the present parameterization in terms of the numbers of dust particles? Second, even if ice nucleation below water saturation is used for mineral dust particles only, does not this create some likely overlap with the M92 scheme? That is, I would expect the P13 formulation to make ice prior to homogeneous freezing temperatures, where the M92 formulation is already generating ice.

**Authors' response:** Since we re-formulated the P13 parameterization in terms of $n_s$ (Fig. 2c), P13 was implemented in the same way (i.e., same $T$ and $RH_{ice}$ ranges) as the AIDA-derived parameterization by assuming 200 dust particles per L with the identical aerosol surface area. With our approach, also no ice formation was allowed in the regime covered by M92. This makes the comparison of two parameterizations consistent, using M92 at $T$ > -36°C.

**Reviewer's comment:** Page 16502, line 22: Is a time step of 20s reasonable or sufficient for cirrus simulations?

**Authors' response:** In contrast to a parcel model that actively resolves the depletion of water vapor through heterogeneous ice formation, we use the parameterization-scheme of *Kärcher et a*l. (2006) to parameterize the processes on small time scales, such as depletion of water vapor. *Kärcher et al*. (2006) demonstrated the implementation of their scheme into a global model with much longer time steps (~30 min).

For clarity, we now added the following sentence at the end of section 2.3.2 (page 16503 line 8):

"The latter was used to parameterize the competition of water vapor between homogeneous and heterogeneous freezing"

Reference:

Kärcher, B., Hendricks, J., and Lohmann, U.: Physically based parameterization of cirrus cloud formation for use in global atmospheric models, J. Geophys. Res., 111, D01205, doi:10.1029/2005JD006219, 2006.

**Reviewer's comment:** Page 16504, lines 23-25: Could you say a little more about how immersion freezing data is used to "constrain" fitting curves? Because immersion freezing is requisite at the warmer temperatures? Or because you consider SCF as a subset of immersion freezing? Yet, you have not yet introduced the concept of SCF in the paper to this point.

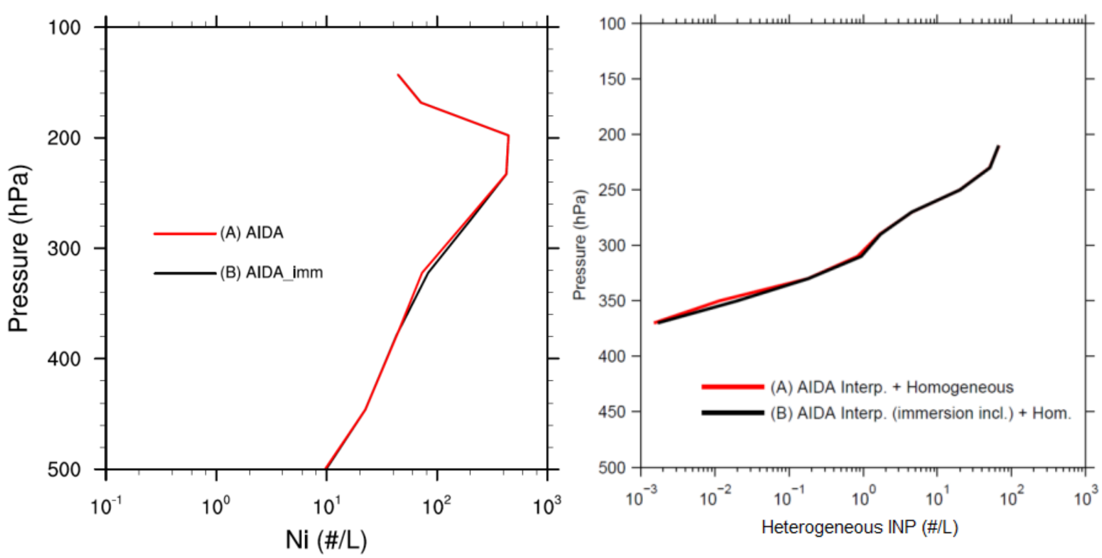
**Authors' response:** We thank the referee to raise this important point. We now describe how immersion freezing data are used on the page 16504 lines 23-25:

Original: "Previous AIDA measurements of immersion freezing (i.e., INUIT04_13 and INUIT01_28 from *Hiranuma et al*., 2014) are also shown and used to constrain the fitted curves."
→
Modified: "Previous AIDA results of two immersion freezing experiments (i.e., INUIT04_13 and INUIT01_28 from *Hiranuma et al*., 2014) are also shown on the water saturation line and used to constrain the fitted curves because immersion freezing is considered part of isolines. Since the $n_s$ values presented in Fig. 3 of *Hiranuma et al*. (2014) only extends up to ~$10^9$ $m^{-2}$, the data points of higher $n_s$ values were extrapolated based on the observed values from two measurements."

The review is correct in pointing out that SCF is considered as a subset of immersion freezing. We now updated the first paragraph on the page 16511:

"As described in the previous section (Sect. 3.1), deposition mode freezing cannot solely explain the $n_s$-isoline observation below water saturation (-50 ˚C < T < -36 ˚C in Fig. 2). Although we presumed that SCF acts as a subset of immersion freezing and plays an important role in this region, further insight and evidence of SCF beyond cloud simulation chamber observations are required to correctly understand the contributions of both homogeneous and heterogeneous nucleation. High-resolution microscopic techniques with an integrated continuous cooling setup are needed to visualize the freezing process of a single particle and to fully understand the complex freezing processes involved in SCF on particle surfaces."

We have evidence that both including immersion freezing data and not including them (but extrapolating the $n_s$-isoline fit to water saturation line) can reproduce similar (almost identical) simulation results in the simulations with SCAM5 and COSMO (please see figures below).




**Extra Figure.** Comparison of two parameterizations (i.e., including immersion data vs. extrapolating without immersion data) in monthly mean profiles of the simulated in-cloud ice crystal number concentrations with SCAM5 (left) and the heterogeneous INP number concentration averaged over two days with COSMO (right).

We feel that we provide the definition of SCF given in right place (Sect. 3.1 on the page 16505 lines 14-25) and no earlier discussion is necessary.

**Reviewer's comment:** Page 16505, line 9: I suggest replacing "while concurrent" with "during"

**Authors' response:** Corrected.

**Reviewer's comment:** Page 16505, line 14: Considering this sentence ending, ": : :pointed to a new freezing process in this study" I believe that "point" would be more appropriate. However, there seems a basic problem in saying it is a new freezing process, when what is imagined is something much like what has been discussed explicitly in recent papers and has been alluded to in some previous ones. Should it say that these results appear to support the existence of a pore or surface freezing process, as discussed in recent literature (e.g., Marcolli , 2014). Then move on to define it as SCF, rather than PCF.

**Authors' response:** We agree. We now rephrased the sentence on the page 16505 lines 12-14 as:

"Therefore, other microphysical processes at the particle surface and/or perhaps even within the bulk phase may be responsible for this *T* -dependent behavior and these results appear to support the existence of a pore or surface freezing process, as discussed in recent literature (e.g., *Marcolli*, 2014)."

**Reviewer's comment:** Page 16506: I must question the semantics of what can be called "atmospheric dust particles" because I am not terribly convinced that the hematite results encapsulated by $n_s$ fits show that they are comparable to actual atmospheric dust, but rather to soil dust samples brought into the laboratory. Are any of the referenced studies, for any temperature regime, actual measurements of atmospheric dust, rather than surrogates for such?

**Authors' response:** *Matsuki et al*. (2010) studied atmospheric dust particles. More specifically, the authors deduced that airborne dust residuals collected through the counter virtual impactor (*Ogren et al*., 1985) during flights in stratocumulus clouds over Niger contained hematite.

Reference:

Ogren, J. A., Heintzenberg J., and Charlson R. J.: In-situ sampling of clouds with a droplet to aerosol converter, Geophys. Res. Lett. 12, 121–124, doi: 10.1029/GL012i003p00121, 1985.

**Reviewer's comment:** Page 16506, line 9: "are" compared.

**Authors' response:** Corrected.

**Reviewer's comment:** Page 16506, line 16: Is "mobile" the right word for such ice nuclei counters? Mobile implies that they move, rather than the fact that they are movable or able to be installed on aircraft. Perhaps "portable"?

**Authors' response:** Thank you. We agree and corrected.

**Reviewer's comment:** Page 16508, lines 3-5: The lower bound was explained, but not the upper. What controls the upper bound, why physically does $n_s$ remain constant up to the water saturation line, and why presumably? It is explained rather obtusely at present. Does it mean simply that experimental data does not exist in this regime?

**Authors' response:** The answer is yes. Within our experimental conditions, the highest estimated $n_s$ was ~$10^{12}$ $m^{-2}$ before the peak $RH_{ice}$ (i.e., Fig. 2), even accounting for continuous increase in $n_s$ after depletion of supersaturation (i.e., Fig. S2), and we simply did not want to extrapolate the isoline fit to 'unexplored' region.

We have now rephrased the sentence (page 16508 lines 3-5) to:

"Above the upper bound of $10^{12}$ $m^{-2}$, $n_s$ presumably remains constant up to the water saturation line in $T$ - $RH_{ice}$ space (no experimental data is available in this range)."

**Reviewer's comment:** Page 16509: Variously here, the "three" and "four" parameterizations are mentioned. Please clarify.

**Authors' response:** To clarify our study cases, we have modified and updated the texts in the following sections:

Page 16509 lines 9-13:

Original: "These include the pure homogeneous ice nucleation case (Simulation A), three cases with contributions from both the homogeneous and heterogeneous ice nucleation (hereafter combined case) described in Fig. 4 (Simulations B, C and D) and the simulation of the different lower boundaries of RHice ($RH^*_i$ , Simulation D)."
→
Modified: "These include (Case 1) the pure homogeneous ice nucleation case, (Case 2-4) cases with contributions from both homogeneous and heterogeneous ice nucleation (hereinafter referred to as the combined case) described in Fig. 4a-c (corresponding to Simulations A, B and C) and (Case 5) the simulation of the different lower boundaries of $RH_{ice}$ ($RH^*_i$ , Simulation D)."

Page 16509 lines 18-20:

Original: "The differences between the four parameterizations used in this study are small for both the combined cases and the pure heterogeneous cases."
→
Modified: "The differences between the three parameterizations derived from AIDA measurements, corresponding to Simulations A, B and D, are small for both the combined case and the pure heterogeneous ice nucleation case as presented in Fig. 6."

Page 16509 line 25:

Original: "The three parameterizations have largest differences…"
→
Modified: "The three parameterizations (i.e., Simulations A, B and D) have largest differences…"

**Reviewer's comment:** Page 16510, COSMO simulations: I find a lot missing in the discussion here. First, the averaging used to obtain the results is not totally clear. What defines a cloudy area for the results shown in Figures 7 and 8? Does it imply that areas with very low ice content and low ice concentrations are averaged along with others of higher optical depth or higher ice water content across the domain? The lower ice concentrations in P13, despite the inclusion of homogeneous freezing are striking and somewhat surprising unless the small regions where stronger ice formation occurs (e.g., homogeneous freezing regions) are averaged out, while the higher values in the AIDA-based parameterizations surprise me if they are averaged over all cloudy parcels, even at low ice supersaturations.

**Authors' response:**

We thank the reviewer for pointing out that this discussion was too condensed and partly misleading. Regarding the averaging conditions, we have applied the following procedure in order to obtain consistent results for the different simulations:

The INP concentrations as well as $T$ and $RH_{ice}$ conditions allowing for hematite to nucleate through deposition mode, i.e. T<-36°C and $RH_{ice}$ >100%, are extracted and binned in pressure intervals for subsequent averaging, which yields the data in Figure 7. This averaging procedure was implemented for both the AIDA-based parameterization and P13. In case of P13, this leads to averaging also over zeros, when temperatures are below -36°C, but the supersaturation is low enough that the parameterization returns $n_s = 0$ (white areas of Figure 4C).

Note that Fig. 7 shows only the heterogeneous INP concentration. Although contained in the simulation and calculated right afterwards heterogeneous ice formation, the homogeneously formed ice is not contained in the mean values of Figure 7. Here we compare only ice that is formed by deposition nucleation, rather than "total" ice occurring in the model. In this way, the effects of sedimentation, advection and turbulent diffusion of ice crystals (that had been formed earlier in the model and may be present in T<-36°C and $RH_{ice}$>100% regions due to transport) are disregarded in order to extract the heterogeneous ice formation.

We have modified the test on the page 16510 lines 9-13:

Original: "$N_{ice}$ was spatially averaged over all cloudy areas of the model domain for freezing conditions of cubic hematite particles. As shown in Fig. 7, the mean $N_{ice}$ resulting from the parameterization based on P13 is smaller than that from the AIDA $n_s$-isoline-based parameterization by more than two orders of magnitude."

→

Modified: "$N_{ice}$ was spatially averaged over all areas of the model domain which in principle allow for deposition nucleation in our simulations, i.e. conditions below -36°C and above 100% $RH_{ice}$. Because not always $n_s$ is larger than zero (white areas of Fig. 2), also areas without ice formation are contained. It is also noteworthy that only purely heterogeneous ice formation is presented rather than the total ice occurring in the model. As shown in Fig. 7, the mean $N_{ice}$ resulting from the parameterization based on P13 is smaller than that obtained from the AIDA $n_s$-isoline-based parameterization by more than two orders of magnitude. This large difference results predominantly from the inactivity of P13 at low $RH_{ice}$."

The x-axis in Fig. 7 and caption have been modified:

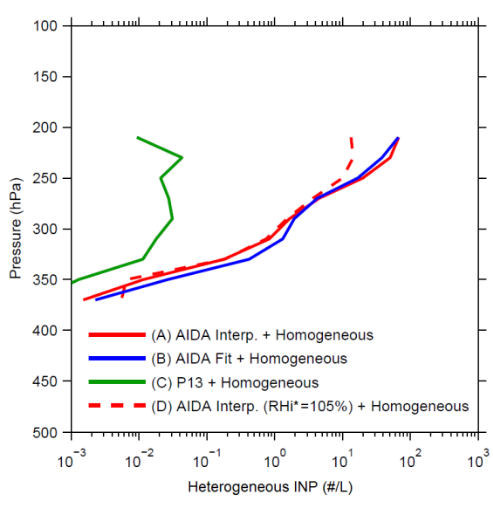



**Figure 7.** The mean heterogeneous INP number concentrations simulated in COSMO.

**Reviewer's comment:** Next, the results not only show a vast difference between the domain averaged ice crystal concentrations, but the relative picture of maximum supersaturation evident in Figure 8 is drastically different. If I follow this correctly, it would be highly unlikely to find an area supersaturated with respect to ice by more than 20

**Authors' response:** Our presentation is correct. Most of ice formation, in case of using AIDA-derived parameterization, is triggered below $RH_{ice}$ 120% (Fig. 8A) owing to our $n_s$ parameterization which shows non-negligible ice formation even at low $RH_{ice}$ and simulated atmospheric conditions in COSMO.

**Reviewer's comment:** Page 16510, last sentence: The data in P13 are clearly limited at lower temperatures. That paper acknowledged this issue and also discussed discrepancies between field and laboratory data at temperatures below -35 ˚C, offering some speculation about this. Can you say that your results confirm these things and can you speak to whether or not the modeling provides any new insights?

**Authors' response**: What we see from the results of both models is that the modeled ice concentrations are very sensitive to deposition nucleation at low temperatures when heterogeneous INPs (hematite particles in this study) compete with homogeneous freezing. Because the low supersaturations have a high frequency of occurrence in the atmosphere, it is highly desirable that atmopherically relevant IN (mineral dust and others) are described more precisely with respect to their deposition nucleation ability in the cirrus regime.

**Reviewer's comment:** Page 16511, lines 14-18: I wonder if the discrepancies between the parameterizations noted here do not actually highlight most the need for more high quality atmospheric measurements of INP's in the upper troposphere. This is what stands out to me at least. The laboratory measurements seem consistent.

**Authors' response:** The authors believe that in situ measurements of INPs, such as the number concentration and the types of INPs, can constrain lab based parameterizations. Though direct observation of the aerosol freezing mechanism in the upper troposphere where coincides with the low water vapor pressure (*Cziczo and Froyd*, 2014), investigating chemical characterization of cirrus residual particles can give a crucial hint and promote the investigation of ice formation mechanism by using laboratory instruments (e.g., AIDA).

We have rephrased the sentences on the page 16511 lines 14-23:

Original: "Substantial differences between the empirical approach of P13 and our parameterization developed in this study are presumably attributed to the difference in lab- or field data, highlighting the need for further characterizations of atmospherically relevant substrates and their ice nucleation activities in laboratory settings. Nevertheless, *Niemand et al*. (2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation. In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) provides indication on the validity of the assumption to treat all dust as hematite in deposition mode."

→

Modified"Nevertheless, the previous study (e.g., *Niemand et al*., 2012) demonstrated that different dusts exhibit similar $n_s$ in immersion mode freezing and perhaps such a similarity remains true for deposition mode ice nucleation. In fact, comparison between our AIDA $n_s$-based parameterization with hematite particles and *Möhler et al*. (2006) with ATD and SD2 (Fig. 3a) suggests that hematite has similar ice nucleation efficiency, inferred by $n_s$, as dust. The comparison between the observed profile of ice crystal number concentrations and the simulated ones (Figs. 5 and 6) also suggests the validity of the new parameterizations. These premises must be further examined in comparing to atmospherically relevant substrates (fresh and aged ones) and their ice nucleation activities in laboratory settings. In situ INP measurements, such as the number concentration and the types of INPs, at the upper troposphere can also help to constrain the parameterizations."

**Reviewer's comment:** Page 16512, lines 14-16: I did not note anywhere in the paper where a hypothesis for the appearance of an RH-dependent ice nucleation below -60 ˚C was discussed. Can you suggest anything?

**Authors' response:** Discussed on the page 16504 lines 25-27:

For clarity, we have added "(i.e., $RH_{ice}$-dependent ice nucleation regime)" in the end of this sentence (page 16504 lines 25-27):

"Figure 2 shows several important features of $n_s$-isoline curves. First, below −60 ˚C, $n_s$-isolines showed an increase in $RH_{ice}$ required to maintain a constant *ns* (i.e., $n_s > 2.5\times10^8$ m$^{-2}$) with decreasing $T$ (i.e., $RH_{ice}$-dependent ice nucleation regime)."

**Reviewer's comment:** Last sentence: Can you comment on whether or not you believe that the stronger ice nucleation at limited ice supersaturations on the basis of the new parameterization is realistic for the atmosphere? How might this be examined?

**Authors' response:** How strong ice nucleation at lower $RH_{ice}$ can be examined and quantified by conducting 'direct' measurements of INP concentration as a function of T and $RH_{ice}$. This could ultimately verify how realistic the lab results are for the atmosphere. Nonetheless, both experimental and model results (Fig. 2 and 8A) suggest the new parameterization to induce substantial ice formation even at low $RH_{ice}$. Hence, the new parameterization strongly suggests the role of $T$ and more ice nucleation when compared to the existing empirical parameterization, presumably allowing more ice activation under water subsaturated conditions.

**Additional revision:** In addition to addressing the reviewer's comments, other editorial corrections (major and non-miscellaneous ones) were made and listed above on the pages 11-13 of authors' responses.