Responses to Referees

The authors wish to thank both reviewers for their useful suggestions and thoughtful comments on means to improve and strengthen the paper. Below are our point-by-point responses to each of the reviewer's comments. Corresponding modifications are reflected in the manuscript and Supplementary Information (SI).

Referee #1

Reviewer's comment: *Consider subdividing and reorganizing the Introduction into further subsections, e.g. "Background," "Motivation," "Previous work," etc. to improve structural clarity.*

Authors' response: This is a good suggestion. We now add three subsection titles (i.e., 1.1. Background – Page 22048 Line 20; 1.2. State of the art of IN measurement techniques – Page 22049 Line 28; 1.3. Objectives – Page 22052 Line 27).

Reviewer's comment: *Page 22054, Line 5-13: The sentences before "In this study" should be part of the introduction, not in the Methods section.*

Authors' response: The following sentences now appear at the end of the first paragraph in the Introduction section (i.e., Page 22049 Line 2).

"In particular, yearly emission rates of soil dust are 1000 to 4000 teragrams, accounting for a major proportion of both the dust component and the total particle loading in the atmosphere (*Boucher et al*., 2013). The resulting radiative forcing directly exerted by mineral dust is estimated to range from -0.3 to +0.1 W m^{-2}. Therefore, dust slightly contributes to the direct cooling effect of aerosols. However, our understanding of the influence of the dust burden upon overall climate forcing, including its secondary effect on cloud albedo, remains highly uncertain, in part due to the absence of accurate INP representations in atmospheric models. Thus, the effective radiative forcing effect of airborne dust on current climate predictions remains unresolved."

Reviewer's comment: *Page 22055, Line 22-24: The sentence "The influence of dust washing..." seems to suggest that differences in IN propensity were measured for of washed and unwashed particles. However, it seems that in Welti et al., (2014), IC was used to confirm the presence of soluble material, but no experiments were actually performed to test for differences in IN propensity of washed and unwashed samples (rather, these tests are proposed as future work). Please verify that this sentence reflects the actual findings in the cited paper or include references that support this sentence.*

Authors' response: The reviewer is right in pointing out that *Welti et al*. (2014) 'proposed' (did not 'measure') the influence of soluble impurities from kaolinite rich minerals on ice nucleation (IN) propensity. As per the reviewer's suggestion, we have modified and updated the sentence in Page 22055 Lines 22-24.

Original: "The influence of dust washing and discharge of soluble materials on IN propensity has been previously reported (*Welti et al*., 2014)."
→
Modified: "The influence of dust washing and discharge of soluble materials on IN propensity has been previously proposed (*Welti et al*., 2014). More specifically, the authors postulated two different scenarios at different temperatures based on their observations. At temperatures below ~ -38 ºC, the washed dust component may have enhanced water condensation below water saturation, and a formed liquid layer presumably may have stabilized the subcritical ice embryo entrapped inside the liquid. The authors

proposed this capillary condensation process as a part of condensation freezing or homogeneous nucleation based on the previous observation (*Christenson*, 2013) and the theoretical framework (*Marcolli*, 2013). Above ~ -38 ºC, on the other hand, heterogeneous nucleation might have been suppressed because the liquid layer derived from the deliquescence of soluble impurities from individual particles may have diminished accessibility of water vapor to active sites (e.g., localized surface features such as cracks and edges), originally proposed by *Koehler et al*. (2010), preventing the ice embryo formation. In this study, suspended samples…"

Newly added reference:

Koehler, K. A., Kreidenweis, S. M., DeMott, P. J., Petters, M. D., Prenni, A. J., and Möhler, O.: Laboratory investigations of the impact of mineral dust aerosol on cold cloud formation, Atmos. Chem. Phys., 10, 11955–11968, doi:10.5194/acp-10-11955-2010, 2010.

Reviewer's comment: *Page 22061, Line 15-28: The explanation for why the DLS value for S_{total}/M_{total} is preferred over the TSI-OPS value needs to be clearer and more concise. It might make intuitive sense to use an SSA value that matches $n_{s,geo}$ for suspension and dry-dispersed measurements, but choosing one over the other need to be better justified. First of all, why is there such a large difference in the S_{total}/M_{total} values reported by DLS and TSI-OPS? Could there be a physical reason that suspended particles (through their interactions with and processing by water) actually have a significantly different S_{total}/M_{total} than dry particles?*

Authors' response: This difference in the $S_{\text{total}}/M_{\text{total}}$ values is presumably due to the fact that dry-dispersed particles are typically prone to agglomeration (i.e., Sect. 3.1) with more pronounced variation compared to suspended-particles.

As discussed in Page 22061 Lines 17-20, the 'representativeness' of our $S_{\text{total}}/M_{\text{total}}$ highly depends on the degree of agglomeration, and it could vary up to a factor of 13 based on our size distribution comparisons. Since we do not have size distribution measurements and associated $S_{\text{total}}/M_{\text{total}}$ for each suspension measurement, DLS-SSA is used for the data evaluation for the measurements with polydispersed suspended particles throughout this study. Nevertheless, the usage of DLS-SSA is reasonable because the presence of fewer agglomerates in suspended particles has been demonstrated with hematite particles and shown in Fig. 1 of *Hiranuma et al*. (2014b). We presume such a similarity might remain true for the illite NX particles. Furthermore, the use of DLS-SSA (= 6.54 m^2 g^{-1}) is reasonable because the conversion factor ranged for size-selected particle diameters from 200 to 1000 nm (as discussed in Page 22061 Lines 3-6) is similar to DLS-SSA (also similar to AIDA-SSA; i.e., Fig. 2b). We also note that the discussion of the potential effect of agglomerates is separately given in Sect. 4.4. The text in the manuscript is slightly changed (see modified Page 22061-22062 Lines 10-3; discussed below).

Reviewer's comment: *Shouldn't the text say 0.49 m^2 g^{-1} is 13 times smaller than 6.54 m^2 g^{-1}?*

Authors' response: Corrected (Page 22061 Line 18). Thank you.

Reviewer's comment: *It is unclear whether it is fair to say "$n_{s,BET}$ is especially representative of measurements with suspended samples because minimal corrections..." Yes, the resulting value is based on a relatively simple correction, but how does this make it "especially representative" of measurements if it is strongly dependent on the choice of S_{total}/M_{total}? Granted, the resulting value may indeed be representative of the measurements (though removing the "especially" qualifier would be more appropriate), but a stronger case needs to be made about the appropriate choice for S_{total}/M_{total}. Since using $n_{m,sus}$ requires an additional assumption than using $n_{m,geo}$, the latter does seem to be a better option,* *given a better explanation for the choice of S_{total}/M_{total}. Changing "where the latter..." to "so the latter..." would greatly improve the clarity.*

Authors' response: Discussed above (Page 2). For clarity, the authors have modified Pages 22061-22062 Lines 10-3. This section now reads:

"...in which, $n_{m,\mathrm{sus}}$ is the IN active mass for suspension measurements, α represents the ice activated fraction (= $N_{\mathrm{ice}}/N_{\mathrm{total}}$), which is the direct measurement of suspension experiments and some of the dry-dispersed particle methods. With an assumption of a uniform BET-SSA, the resulting $n_{s,\mathrm{BET}}$ may be representative of measurements with suspended samples because minimal corrections (only α and θ) are involved when compared to that with dry-dispersed particles. Owing to internal surface area and surface roughness, BET-SSA may be greater than DLS-SSA (*O'Sullivan et al*., 2014).

Alternatively, we can also convert ice-nucleating mass derived from suspension measurements, $n_{m,\mathrm{sus}}$, to $n_{s,\mathrm{geo}}$ using DLS-SSA to provide a reasonable comparison to dry-dispersed particle measurements. However, this process requires one more step than when using $n_{s,\mathrm{BET}}$ (with an additional assumption of constant size distribution for all suspensions) and two more steps than when using n_m. For our inter-comparison study, we used both $n_{s,\mathrm{BET}}$ and $n_{s,\mathrm{geo}}$. Because fewer conversion factors are involved, $n_{s,\mathrm{BET}}$ may be best suited for suspension measurements, and $n_{s,\mathrm{geo}}$ may be best suited for dry-dispersed particle measurements (Eqn. 3 to 4 or vice versa).

The usage of DLS-SSA for the calculation of $S_{\mathrm{total}}/M_{\mathrm{total}}$ of suspension measurements appears to be reasonable, as this leads to $n_{s,\mathrm{geo}}$ for suspension measurements nearly equivalent to $n_{s,\mathrm{geo}}$ for dry-dispersed particles. When $S_{\mathrm{total}}/M_{\mathrm{total}}$ is derived based on TSI-OPS measurements, a value of 0.49 $m^2\ g^{-1}$ is obtained, which is smaller by a factor of about thirteen compared to DLS-SSA. This difference may be mainly due to the fact that dry-dispersed particles are typically prone to agglomeration (discussed below, i.e., Sect. 3.1) compared to the measurements with suspended particles. The presence of fewer agglomerates in suspended particles is shown in Fig. 1 of *Hiranuma et al*. (2014b). Since the size distribution of a suspended sample for each experiment was not measured, DLS-SSA was used for the data evaluation for suspension measurements throughout this study."

In addition, we removed "especially" according to the reviewer's suggestion.

Reviewer's comment: *Page 22064, Line 4-15: Could there be aspects of the measurement techniques themselves (or differences in calibrations, corrections, etc.) that could contribute to the differences seen in the SA distributions in Figure 2? How might differences in optical, aerodynamic, and mobility sizing techniques contribute to the differences observed?*

Authors' response: Yes, the different types of dispersion methods, impactors and size segregating instruments used in the present work can contribute to the different degree of agglomeration and the differences in surface area distributions as discussed in Sect. 4.4 (Page 22084 Lines 1-3). See also the Supplement Table S1 for further details. However, we cannot quantitatively compare the effect of measurement techniques themselves on the observed differences in particle size distribution (though all particle sizing instruments have been calibrated well). This is beyond the scope of the current work. Hence, a continued investigation to obtain further insights into consistencies or discrepancies of particle dispersion and size distribution characterization as well as IN measurement techniques, perhaps by assembling and comparing them using identical test dust samples over similar thermodynamic conditions as demonstrated in ICIS-2007, is important (i.e., Page 22092 Lines 5-13).

It should also be noted that all size distribution measurements with dry particles are converted and evaluated in volume equivalent diameter (as inferred in Page 22056 Line 16, Page 22057 Lines1, 13 and 23), and the consistency between DLS-based hydrodynamic diameter and AIDA-based volume equivalent diameter has been demonstrated in our previous study with hematite particles (Fig. 1 from *Hiranuma et al*., 2014b).

Reviewer's comment: *These (and perhaps other) possibilities should also be mentioned here as potential explanations for the observed differences in addition to possible agglomeration. A more detailed discussion should then appear in Section 4.*

Authors' response: We agreed and modified Page 22064 Lines 4-15. Now this part reads, "The surface area distribution of the DLS hydrodynamic diameter-based measurement (Fig. 2a) agreed well with *in situ* measurements from the AIDA chamber (Fig. 2b), suggesting the size distributions of dry illite NX particles during AIDA experiments were similar to those of suspension measurements. This observation is consistent with results presented in *Hiranuma et al*. (2014b). Briefly, the authors found agreement between the DLS-based hydrodynamic diameter and the AIDA-derived volume equivalent diameter of hematite particles. As opposed to the AIDA observation, the wider distributions and the shift in the mode diameters in the MRI-DCECC measurements towards a larger size (0.62 µm, Fig. 2c) when compared to Fig. 2a and b may indicate a higher degree of particle agglomeration as a result of different degrees of pulverization during the particle generation processes or particle coagulation at the high aerosol number concentration used for these measurements. A more pronounced agglomeration effect was observed by the TSI-OPS measurements (Fig. 2d), such that a surface area distribution of supermicron-sized particles was obtained. Thus, different types of dry particle dispersion methods can contribute to varying degrees of agglomeration and the observed differences in surface area distributions. Though all size segregating instruments used in the present study are well calibrated, we cannot rule out the effect of measurement techniques themselves on the observed differences in particle size distribution. In Sect. 4.4 we discuss whether agglomeration has an effect on the IN activity.".

Reviewer's comment: *Page 22068, Line 10-15: No results are discussed here for CU-RMCS. Include a brief summary here like for the other instruments.*

Authors' response: Page 22068 Lines 11-12 now reads, "The University of Colorado (CU)-RMCS examined the freezing abilities of droplets containing 1.0 wt% illite NX. CU-RMCS detected the warmest immersion freezing of illite NX particles at about -23 °C under the experimental conditions used in the present work (see the Supplementary Methods for further details).".

Reviewer's comment: *Page 22075-6, Line 25-1: Is the presence of agglomerates directly measured or just inferred from the results? If the latter is the case, it would be more appropriate for this sentence to say "...may have been carried out in the presence..."*

Authors' response: We thank the reviewer for this suggestion. Page 22075-6 Line 25-1 now reads, "We note that MRI-DCECC experiments may have been carried out in the presence of a high degree of agglomeration (Fig. 2c and d).".

Reviewer's comment: *Page 22083, Line 16-17: "agglomerated-fractions based on a relative comparison to D_{95}" implicitly assumes that differences in D_{95} are a result of agglomerations, rather than discussing the possibility of other contributing factors, such as differences in the hydrodynamic size-based, volume equivalent diameter-based, and optical size-based results.*

Authors' response: Discussed above (hydrodynamic vs. volume equivalent). The presence of larger D_{95} fraction is indicative of the presence of agglomerates.

Reviewer's comment: *Figure 10: In all other figures, $n_{s,geo}$ is the left column. Please change this figure to match the rest.*

Authors' response: No, the panel based on $n_{s,\mathrm{BET}}$ is the left column throughout (Figs. 6, 7 and 8). The figure caption is modified.

"Figure 10. Examination of mode dependency of heterogeneous ice nucleation of illite NX particles. A comparison of FRIDGE (default) and FRIDGE (imm.mode) in $n_{s,\mathrm{BET}}$ and $n_{s,\mathrm{geo}}$ are shown in (a) and (b), respectively. (c) and (d) show a comparison between EDB (contact), EDB (imm.), ZINC, IMCA-ZINC, and PNNL-CIC data in $n_{s,\mathrm{BET}}$ and $n_{s,\mathrm{geo}}$, respectively."

Reviewer's comment: *As a general technical comment, the authors are advised to check the consistency of past and present tenses used in the manuscript. Some specific examples are included below, but the flow of the text is sometimes interrupted by unexpected tense changes. Consider using the present tense whenever possible, especially when discussing work done for this study.*

Authors' response: Thank you. Corrected.

Reviewer's comment: *Another general technical comment, there are often missing spaces before and after mathematical expressions and symbols. Many are pointed out below, but the authors are advised to verify that all such cases are fixed.*

Authors' response: All fixed.

Reviewer's comment: *Page 22047, Line 27: Consider rewording "Only instruments making measurements with wet suspended samples were able to measure..."*

Authors' response: Reworded.

Reviewer's comment: *Page 22048, Line 3: Put a space between "to" and "n_s".*

Authors' response: Corrected.

Reviewer's comment: *Page 22048, Line 9: Remove comma after "spectra"*

Authors' response: Removed.

Reviewer's comment: *Page 22048, Line 16: Remove "an"*

Authors' response: Removed.

Reviewer's comment: *Page 22048, Line 17: Remove ",thereby,"*

Authors' response: Removed.

Reviewer's comment: *Page 22049, Line 23: Replace "towards immersion freezing properties" with "for immersion freezing".*

Authors' response: Replaced.

Reviewer's comment: *Page 22050, Line 3: remove ", which".*

Authors' response: Removed.

Reviewer's comment: *Page 22050, Line 6-9: For clarity, change to "Supersaturated conditions with respect to water and ice, as a function of temperature, were created in the simulation chamber vessel by a rapid pressure drop caused by mechanical expansion and subsequent cooling."*

Authors' response: Changed.

Reviewer's comment: *Page 22052, Line 28: Change "was" to "is"*

Authors' response: Changed.

Reviewer's comment: *Page 22053, Line 6-7: The meaning of "The dataset constitutes a function of..."is unclear. Consider rewording as "This dataset captures the functional dependence of... nucleation time on illite NX immersion freezing properties" or something similar.*

Authors' response: Thank you. For clarity, the sentence now reads, "The dataset captures the functional dependence of various experimental parameter variables, such as particle concentration, particle size, droplet size, temperature, cooling rate and nucleation time, on the immersion freezing properties of illite NX particles.".

Reviewer's comment: *Page 22053, Line 16: Is the hyphen between parameterization and approach necessary?*

Authors' response: Thank you. The hyphen is now removed.

Reviewer's comment: *Page 22055, Line 2: Consider using "irregular" rather than "deformed."*

Authors' response: Corrected.

Reviewer's comment: *Page 22057, Line 12: Replace "about 2" with "~2"*

Authors' response: Replaced.

Reviewer's comment: *Page 22057, Line 21: "is" is inconstant with the tense of the rest of the paragraph.*

Authors' response: Rephrased to "was".

Reviewer's comment: *Page 22058, Line 2: "in the table" should specify the table number.*

Authors' response: Rephrased to "As seen in Table 1,…".

Reviewer's comment: *Page 22059, Line 11-14: As this sentence is currently written, it seems to say that* $n_{s,geo}$ *represents the geometrically determined surface area (instead of the IN active surface-site density based on geometric size).*

Authors' response: We modified the sentence.

Original: "We now describe a method to parameterize surface area-scaled immersion freezing activities using the size equivalent ice nucleation active surface-site density (Connolly et al., 2009; Niemand et al., 2012; Hoose and Möhler, 2012), relating it to the geometrically determined surface area, $n_{\mathrm{s,geo}}$."

→

Modified: "We now describe a method to parameterize surface area-scaled immersion freezing activities using the size equivalent ice nucleation active surface-site density based on geometric size ($n_{s,geo}$; *Connolly et al*., 2009; *Niemand et al*., 2012; *Hoose and Möhler*, 2012)."

***Reviewer's comment:** Page 22060, Line 7: Consider replacing "under water suspended conditions" with "for experiments using suspended particles."*

Authors' response: No, $n_{s,BET}$ is applicable to both dry and suspension measurements. We now modify Page 22060 Lines 6-7.

"In addition, the IN efficiency can be related to the BET-SSA to estimate BET-inferred ice nucleation surface-site density, $n_{s,BET}$."

***Reviewer's comment:** Page 22060, Line 12: Replace ", therefore S_{total}" with "; therefore, S_{total}.*

Authors' response: Corrected.

***Reviewer's comment:** Page 22060, Line 15-17: Since you are not actually describing a list of steps, consider changing to "...$n_{s,BET}$, the geometric size-based ice nucleating mass, $n_{m,geo}$ (g^{-1}), was first calculated..."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22060, Line 17: Change S_{total}-to-M_{total} to S_{total}/M_{total} here and throughout for consistency with mathematical notation for the size-selected case.*

Authors' response: This is a good suggestion. Thank you. All corrected (Page 22061 Line 2; Page 22061 Line 17).

***Reviewer's comment:** Page 22061, Line 6: Changing "Lastly" to "Therefore" would provide consistency with the change on Page 22060, Line 15-17.*

Authors' response: Corrected.

***Reviewer's comment:** Page 22061, Line 25: Remove comma after "technique"*

Authors' response: Corrected.

***Reviewer's comment:** Page 22061, Line 27-28: Consider rewording "it is one step further when compared to $n_{s,BET}$ (with an additional assumption of constant size distribution for all suspensions) and two steps further compared to nm" as "this process requires one more step than when using $n_{s,BET}$ (with an additional assumption of constant size distribution for all suspensions) and two more steps than when using nm" for clarity.*

Authors' response: Thank you. Reworded.

***Reviewer's comment:** Page 22062, Line 2-3: Either specify "$n_{s,BET}$ is more representative for suspensions than... and $n_{s,geo}$ is better for for dry-dispersed particle measurements than..." or simply say something like "$n_{s,BET}$ is suited for suspensions, and $n_{s,geo}$ is suited for dry-dispersed particle measurements."*

Authors' response: Thank you. We modified the sentence.

"Because fewer conversion factors are involved, $n_{s,BET}$ may be best suited for suspension measurements, and $n_{s,geo}$ may be best suited for dry-dispersed particle measurements (Eqn. 3 to 4 or vice versa)."

***Reviewer's comment:** Page 22062, Line 20-22: Change "wt '%" to "wt %'s" or "abundances" and "was measured" to "were measured."*

Authors' response: We thank the referee for this suggestion. We reworded "wt%" and "was measured" to "abundances" and "were measured", respectively.

***Reviewer's comment:** Page 22063, Line 2: Consider changing "published elsewhere" to "previously published."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22063, Line 17: Change "suggests" to 'suggest."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22063, Line 20-23: For clarity consider rewording, e.g. "Since illite NX particles have significant internal surface area, BET-derived surface areas can be expected to be larger than those derived from the laser diffraction technique. Supporting this notion, ..."*

Authors' response: Thank you for a good suggestion. We modified the sentences according to the reviewer's suggestion.

***Reviewer's comment:** Page 22063, Line 28: Change "These" to "this."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22064, Line 12: Change "discusses" to "discuss."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22065, Line 4: Would be clearer as "$n_s(T)$, (m_{-2} as a function of ºC)."*

Authors' response: Corrected.

***Reviewer's comment:** Page 22066, Line 14: Change "500 nm mobility diameter size" to "500 nm mobility diameter size-selected" for consistency.*

Authors' response: Corrected.

***Reviewer's comment:** Page 22066, Line 23-24: Consider changing "with droplets of volume from micro-liter to pico-liter" to "using droplets with volumes in the micro-liter to pico-liter range."*

Authors' response: Corrected to "using droplets with volumes in the microliter to picoliter range".

Reviewer's comment: *Page 22067, Line 1: Replace "; with the highest temperatures attained" with ". The highest temperatures are attained."*

Authors' response: Corrected.

Reviewer's comment: *Page 22067, Line 2: Add a comma before "which."*

Authors' response: Corrected.

Reviewer's comment: *Page 22067, Line 5: "$n_s(T)$" should be written in math mode.*

Authors' response: Corrected.

Reviewer's comment: *Page 22067, Line 23: Replace "to allow" with "that allows" for consistency.*

Authors' response: Corrected.

Reviewer's comment: *Page 22068, Line 20-21: Consider rewording "within previously reported uncertainties for immersion freezing experiments" as "for immersion freezing experiments, within previously reported uncertainties" for clarity.*

Authors' response: Corrected.

Reviewer's comment: *Page 22069, Line 4-6: Consider changing to "As demonstrated in DeMott et al. (2014), higher RH_w values were required for full expression of immersion freezing in the CFDC. The use of 105 % RH_w in CSU- CFDC does not capture INP activity for many natural dusts, up to a factor of three." for clarity.*

Authors' response: Changed.

Reviewer's comment: *Page 22069, Line 14: Remove "available" for clarity.*

Authors' response: Removed.

Reviewer's comment: *Page 22070, Line 7: Remove "one"*

Authors' response: Removed.

Reviewer's comment: *Page 22071, Line 5-6: It is unclear what is meant by "and, with a slightly better agreement, a time-dependent treatment." Please provide a clearer explanation.*

Authors' response: For clarity, the authors updated the text as, " The results from both instruments agreed well with each other from a data evaluation based on n_s, and this agreement was even improved when the different residence times in LACIS and the CSU-CFDC were accounted for (i.e., when nucleation rate coefficients were compared).".

Reviewer's comment: *Page 22071, Line 12: Change "from" to "than."*

Authors' response: Changed.

Reviewer's comment: *Page 22071, Line 14-15: Why is "(i.e., MRI-DCECC)" included? Also, replace ", which is N_{ice} of" with "of N_{ice} ="*

Authors' response: Thanks for pointing out this error. We deleted (i.e., MRI-DCECC) and replaced ", which is N_{ice} of" with "of N_{ice} = "as per the reviewer's suggestion.

Reviewer's comment: *Page 22071, Line 17: Change "their" to "the."*

Authors' response: Corrected.

Reviewer's comment: *Page 22071, Line 20: Replace "therefore" with "so" or use a semicolon to separate the clauses.*

Authors' response: Now the text reads, "…particles; therefore, an OPC threshold…".

Reviewer's comment: *Page 22071-2, Line 25-1: Replace "resulting in the data from PINC being in agreement with LACIS..." with "resulting in agreement between the data from PINC and data from LACIS..." for clarity.*

Authors' response: Corrected.

Reviewer's comment: *Page 22072, Line 9: Replace the comma with a semicolon to separate independent clauses.*

Authors' response: Corrected.

Reviewer's comment: *Page 22073, Line 21: Put a space between "in" and "n_s."*

Authors' response: Corrected.

Reviewer's comment: *Page 22074, Line 6-8: Consider changing ", whereas" to ". However," and placing a comma before "which" to avoid a run-on sentence.*

Authors' response: Corrected.

Reviewer's comment: *Page 22074, Line 15: Change "its" to "the."*

Authors' response: Corrected. This sentence is now moved to Page 22068 Lines 11-12, and it now reads, "CU-RMCS detected the warmest immersion freezing of illite NX particles at about -23 °C under the experimental conditions used in the present work (see the Supplementary Methods for further details).".

Reviewer's comment: *Page 22075, Line 22: Change "well agreed" to "agreed well."*

Authors' response: Corrected.

Reviewer's comment: *Page 22075, Line 24: Is "unique" necessary here?*

Authors' response: No. Deleted.

Reviewer's comment: *Page 22076, Line 2-4: A space is required before "n_s."*

Authors' response: Corrected.

Reviewer's comment: *Page 22077, Line 21: "axs" should be "axes."*

Authors' response: Corrected.

Reviewer's comment: *Page 22077, Line 24: Again, "especially" in this context is an unnecessary qualifier?*

Authors' response: The reviewer is correct. Deleted.

Reviewer's comment: *Page 22080, Line 21: Consider removing "to control of the conditions leading to" for clarity.*

Authors' response: We agree. It is not necessary and has been removed.

Reviewer's comment: *Page 22084, Line 13: Replace "be of" with "have."*

Authors' response: Corrected.

Reviewer's comment: *Page 22085, Line 1: Replace "shows" with "show"*

Authors' response: Corrected.

Reviewer's comment: *Page 22085, Line 16: Remove "of" for consistency.*

Authors' response: Corrected.

Reviewer's comment: *Page 22086, Line 11: Commas are unnecessary.*

Authors' response: Commas are now deleted.

Reviewer's comment: *Page 22086, Line 12-14: Consider changing "...PNNL-CIC and IMCA-ZINC both of which measured condensation/immersion and purely immersion mode freezing efficiency of particles, respectively, are in reasonable..." to "...PNNL-CIC and IMCA-ZINC measured condensation/immersion and purely immersion mode freezing efficiency of particles, respectively, and are in reasonable..." for clarity.*

Authors' response: Corrected.

Reviewer's comment: *Page 22087, Line 6-7: Change "K-feldspar and" and "orthoclase which" to "K-feldspar, and" and "orthoclase, which."*

Authors' response: Corrected.

Reviewer's comment: *Page 22088, Line 26: Change "the function" to "a function."*

Authors' response: Corrected.

Reviewer's comment: *Page 22089, Line 8: Change "the function" to "a function."*

Authors' response: Corrected.

Reviewer's comment: *Page 22089, Line 15: Change "(Garimella et al., 2014)" to "Garimella et al., (2014)."*

Authors' response: Corrected.

Additional revision: In addition to addressing the reviewers' comments, other editorial corrections (major ones) were made as below.

- Page 22047-22048 Lines 23-18: This paragraph now starts with a more general statement of how the different datasets compare and then discuss the possible difference between the dry-dispersed and suspended measurements further down in the paragraph. This paragraph now reads, "In general, the seventeen immersion freezing measurement techniques deviate, within a range of about 8 ˚C in terms of temperature, by three orders of magnitude with respect to n_s. In addition, we show evidence that the immersion freezing efficiency expressed in n_s of illite NX particles is relatively independent of droplet size, particle mass in suspension, particle size and cooling rate during freezing. A strong temperature dependence and weak time- and size dependence of the immersion freezing efficiency of illite rich clay mineral particles enabled the n_s parameterization solely as a function of temperature. We also characterized the $n_s(T)$ spectra and identified a section with a steep slope between -20 and -27 ˚C, where a large fraction of active sites of our test dust may trigger immersion freezing. This slope was followed by a region with a gentler slope at temperatures below -27 ˚C. While the agreement between different instruments was reasonable below ~ -27 ˚C, there seemed to be a different trend in the temperature-dependent ice nucleation activity from the suspension and dry-dispersed particle measurements for this mineral dust, in particular at higher temperatures. For instance, the ice nucleation activity expressed in n_s was smaller for the average of the wet suspended samples and higher for the average of the dry-dispersed aerosol samples between about -27 and -18 ˚C. Only instruments making measurements with wet suspended samples were able to measure ice nucleation above -18 ˚C. A possible explanation for the deviation between -27 and -18 ˚C is discussed. Multiple exponential distribution fits in both linear and log space for both specific surface area and geometric surface area are provided. These new fits, constrained by using identical reference samples, will help to compare IN measurement methods that are not included in the present study and IN data from future IN instruments.".

- The authors have realized that the averaging/fitting procedure in the linear space in Fig. 7 would bias the fit to higher n_s values. Therefore, we have added the fit in the log space in Fig. 7 and associated fit expressions in Table 3. We also present T-binned $n_{s,\mathrm{BET}}(T)$ and $n_{s,\mathrm{geo}}(T)$ spectra averaged in the log space in Fig. S3 (see the current version of SI Lines 724-741) in a similar way to Fig. 8. As can be seen in both Fig. S3 and Fig. 8, there seems a different trend between suspension and dry-dispersed particle measurements for this mineral dust. Thus, the choice of averaging procedure does not influence our data interpretation of this deviation (i.e., n_s from dry-dispersed methods > n_s from suspension methods) in this study.

  Accordingly, we have also modified the following texts to clarify the use of linear or log space:

  Page 22048 Lines 12-15 now reads, "Multiple exponential distribution fits in both linear and log space for both specific surface area and geometric surface area are provided.".

  Pages 22077-22078 Lines 27-1 now reads, "We also report the absolute values of $\Delta\log(n_s)/\Delta T$ for four T-segregated segments based on T-binned Lin. Avg. (multiple exponential distribution fit to the T-binned average data in the linear space), T-binned Max. (fit to the T-binned maxima in the linear space) and T-binned Min. (fit to the T-binned minima in the linear space) in Fig. 7 (i.e., T_1 to T_4).".

Page 22078 Lines 12-14 now reads, "In this figure, panels i, ii and iii show T-binned data averaged in the linear space of all seventeen instruments, all suspension type measurements, and all measurements that involved dry particles, respectively, while panel iv shows a comparison between suspension and dry-particle measurements.".

Table 3 legend now reads, "Table 3. List of the Gumbel cumulative distribution fit parameters to the $n_{s,\mathrm{BET}}$ and $n_{s,\mathrm{geo}}$ for T-binned ensemble dataset fitted in the linear space [All (lin)], ensemble dataset fitted in the log space [All (log)], ensemble maximum values ($\mathrm{All}_{\mathrm{max}}$), ensemble minimum values ($\mathrm{All}_{\mathrm{min}}$), suspension subset fitted in the linear space [Sus (lin)], suspension subset fitted in the log space [Sus (log)], dry-dispersed particle subset fitted in the linear space [Dry (lin)] and dry-dispersed particle subset fitted in the log space [Dry (log)]. Note that $\mathrm{All}_{\mathrm{max}}$ and $\mathrm{All}_{\mathrm{min}}$ are fitted in the linear space.".

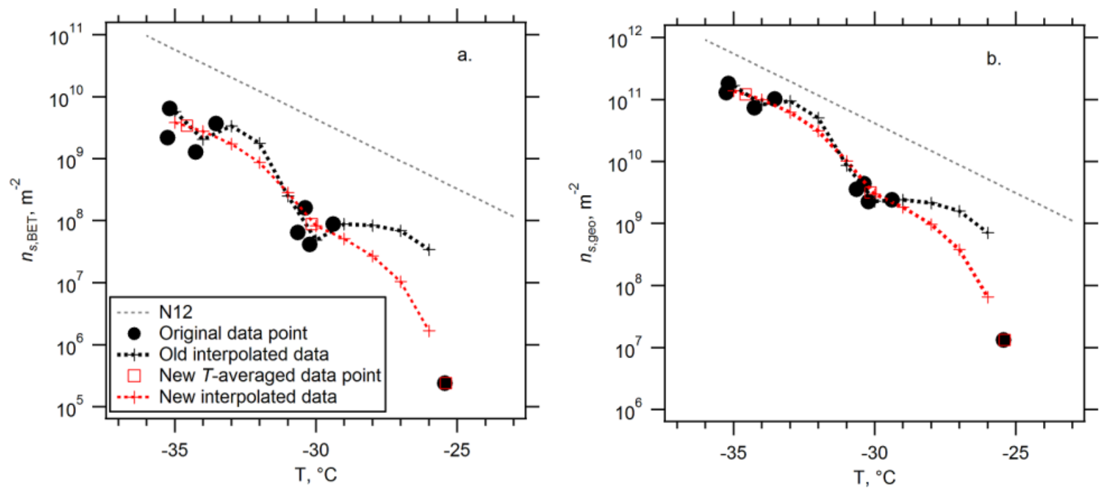
Figure 7 caption now reads, "The multiple exponential distribution fit in the linear space (T-binned Lin. Avg.) is expressed as $n_{s,\mathrm{BET}}(T) = \exp(23.82 \times \exp(-\exp(0.16 \times (T + 17.49))) + 1.39)$ or $n_{s,\mathrm{geo}}(T) = \exp(25.75 \times \exp(-\exp(0.13 \times (T + 17.17))) + 3.34)$. The same fit in the log space (T-binned Log. Avg.) is expressed as $n_{s,\mathrm{BET}}(T) = \exp(22.00 \times \exp(-\exp(0.16 \times (T + 20.07))) + 3.00)$ or $n_{s,\mathrm{geo}}(T) = \exp(22.93 \times \exp(-\exp(0.16 \times (T + 20.31))) + 5.72)$.".

Figure 8 caption now reads, "T-binned $n_{s,\mathrm{geo}}$ (a) and $n_{s,\mathrm{BET}}$ (b). T-binned data (i.e., average in the linear space with 1 ˚C bins for -37 ˚C $< T < -11$ ˚C) of $n_s(T)$ spectra are presented for…".

- We replaced MΩ with MΩ cm. MΩ was the wrong unit (Page 22055 Line 25; SI Line 66 and 190).

- The first sentence in 3.2.8 (Page 22069 Line 1) now reads, "This CFDC provided data for condensation/immersion freezing at around -21.2, -25.1 and -29.7 °C (a total of eight data points with two, two and four points at around each temperature, respectively), which extends to a warmer region than the AIDA measurements.". All data points are now presented in Figs. 4, 5 and 6.

- PINC provided data for immersion freezing at around -25.4, -30.2 and -34.6 °C (a total of nine data points with one, four and four points at around each temperature, respectively). This distribution of data points results in the black fit curve-shape (see black dotted line named 'old interpolated data' in the figure below). To obtain a more representative fit, we grouped/averaged those four data points at the averaged T and performed the same polynomial interpolation with only three data points (at -25.4, -30.2 and -34.6 °C) for -35 °C < T < -26 °C. New interpolated data fits the data better than the previous one and gives much better trace on the $\log(n_{s,\mathrm{ind.}})/\log(n_{s,\mathrm{fit}})$ data as shown in the Figs. S4-S8. Accordingly, the fit parameters (i.e., expressions in Table 3) as well as data representations (Figs. 4, 5, 7 and 8) have changed but only slightly.



Extra Figure. *T*-binned interpolated $n_s(T)$ data (black and red cross markers) for PINC based on the BET (a) and geometric (b) surface areas. Note that the interpolation is valid for -35 ˚C < *T* < -26 ˚C with 1 ˚C bins. Literature results (N12) are also shown.

Additionally, the authors added a new sentence in Page 22071 Line 17.

"PINC provided data for immersion freezing at around -25.4, -30.2 and -34.6 °C (a total of nine data points with one, four and four points at around each temperature, respectively).".

- The authors found the recent publication showing the IN activity of supermicron particles of mineral dust (i.e., *Wheeler et al*., 2014). We added this new reference in Page 22083 Lines 18-20.

  "Since dry aggregates can have large 'supermicron' sizes, they may have different IN propensities and efficiencies (*Wheeler et al*., 2014)…"

  Since we examined the size dependency by comparing only submicron range diameters vs. bulk throughout this study, we modified Page 22066 Lines 16- for clarity.

  "The results suggest size independence of n_s within the experimental uncertainties (a combination of binomial sampling error and the uncertainty of conversion of aerodynamic particle diameter to mass) for the range of examined size (500 nm vs. bulk) and mass concentrations…"

  We also modified another sentence in Page 22073 Lines 5-7.

  Original: "Specifically, a number of instruments (AIDA, LACIS, MRI-DCECC, PINC, PNNL-CIC and IMCA-ZINC) have shown size-independent *n*s values for dry-dispersed particles."
  →
  Modified: "Specifically, AIDA and MRI-DCECC have shown size-independent n_s values for submicron dry-dispersed particles."

  New Reference: Wheeler, M. J., Mason, R. H., Steunenberg, K., Wagstaff, M., Chou, C. and Bertram, A. K.: Immersion freezing of supermicron mineral dust particles: freezing results, testing different schemes for describing ice nucleation, and ice nucleation active site densities, J. Phys. Chem. A, Article ASAP, doi: 10.1021/jp507875q, 2014.

- For clarity, we added the following sentence in Page 22086 Line 20.

  "As described in the Supplementary Methods, immersion mode experiments were performed for the droplets, which were not activated via contact freezing."

- We modified the following sentence in Page 22087 Lines 26-27.

  Original: "…acid processing of K-feldspar which deactivated kaolinite samples."
  →
  Modified: "…acid processing of K-feldspar which deactivated Fluka-kaolinite."

  We also added another sentence in Page 22087 Line 28.

  "More quantitative investigations of the acid processing of both reference and atmospherically relevant materials and its influence on their immersion mode ice nucleation efficiencies are needed."

- Figure 2 now shows the surface area distributions normalized to 'the total surface area concentration'. Accordingly, the unit on the y-axis in Fig. 2 has been changed to $dS/d\log D_p$, arb.

- During the preparation of the revised version of the manuscript, we have recognized that the concentration of Na^+ cations measured with HPLC was biased by Na^+ leaching from the sodium borosilicate glass bottle used for sample storage. Therefore we have excluded this data from the Fig. 3. The measurements of other the cations (K^+, Ca^{2+} and Mg^{2+}) were not affected since these elements are not present in the sodium borosilicate glass chemical formula.

- The polydisperse and size-selected data from the MRI-DCECC measurements in Fig. 6 are 'combined' for the overall data visibility.

- Two new sentences have been added in Acknowledgement.

  "D. Niedermeier acknowledges financial support from the Alexander von Humboldt-foundation, Germany."

  "N. Hiranuma also thanks the AIDA technical team, including R. Buschbacher, T. Chudy, O. Dombrowski, E. Kranz, G. Scheurig and S. Vogt, for their professional support for the chamber maintenance and operation."

Referee #2 (Prof. Gabor Vali)

Reviewer's comment: *This paper is the result of a large effort in organization and in execution. It represents a significant step in clarifying the power and the limitations of laboratory ice nucleation studies. It also adds considerable new information about the ice nucleating capacity of the mineral illite NX. The authors are congratulated on conceiving and carrying out this work. The main accomplishment of this investigation is to show that many different measurement methods can be used to arrive at a quantitative evaluation of the ice nucleating ability of illite NX. Using the same sample of the mineral and performing measurements with the instruments located at their home bases is a useful alternative approach to the inter-comparison workshops with co-located instruments. Discrepancies among the various measurements in this intercomparison were about the same magnitude as those found for simultaneous measurements with a dust sample in the 2007 workshop (DeMott et al. 2011). Here a larger number of instruments were involved, with a greater diversity of operating principles, so the comparable result represents a success and perhaps even some advantage. It is worth noting that the results represents a substantial improvement over the long term; the scatter was much worse in the results of the 1975 workshop (Vali, 1976).*

However, the results also demonstrate fairly serious limitations. Discrepancies of about two orders of magnitude in the derived measures of ice nucleating ability indicate that comparisons of data obtained in different experiments - past and future - will have to be compared with that sort of variability in mind. Furthermore, measurements of the abundance of INPs in the atmosphere or in other systems have to be accepted with similar possible error ranges. The approach of using a sample powder distributed to different locations has its own difficulties, principally that of ensuring sample stability. It could be expected that a mineral powder is fairly stable but that is not absolutely certain. The effects of oxidation, humidity changes, radiation, aging, vapor adsorption, etc., cannot be separated from differences that arise due to variations in measurement techniques. Tests conducted with the suspensions to diagnose changes in composition (last paragraph on page 22055) is a step in the right direction and shows the possible importance of such tests.

What do the results say about the success of this endeavor? First, the greater degree of agreement among the measurements with suspensions shows that those methods have greater control and fewer uncertainties than the tests with dry aerosols. The downside to the drop-freezing tests is that the background noise level is relatively high, restricting measurements to temperatures above -20 ºC or -25 ºC at best. Second, the scatter in the results for dry aerosol methods is due to diverse operating principles on which the measurements rely. These uncertainties are difficult to surmount. Third, the results support the notion that the frequency of nucleating sites per particle is proportional to the surface are of the particle for illite NX and similar materials.

Authors' response: The authors highly appreciate Prof. Gabor Vali for his comments above, giving a good overview and summary of our study along with previous achievements made by the ice nucleation research community. As mentioned above, there are indeed some important limitations, emerging from instrumental and analytical perspectives, which must be overcome in working towards a complete understanding of the deviation in ice-nucleating ability of examined (and future) techniques.

Here is our response to Prof. Vali's comments.

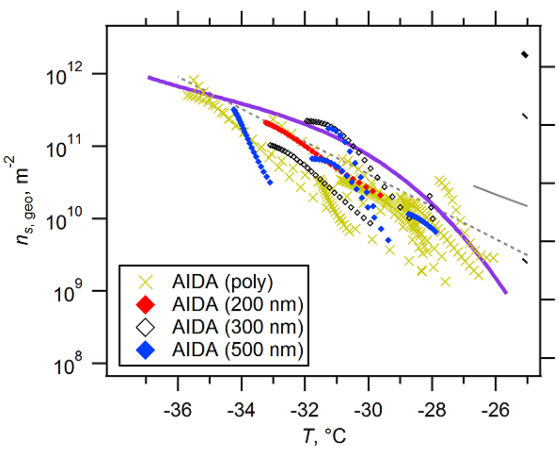
Reviewer's comment: *One could argue that the scatter of measurements are a combination of the instrumental variations and of incomplete fulfillment of the assumptions of the analysis.*

Authors' response: As mentioned in Page 22091 Lines 7-13, the individual uncertainties of each instrument cannot be greater than the discrepancy among the results from the different instruments (~8 ˚C in terms of temperature and up to three orders of magnitude with respect to n_s), suggesting that all instruments may be reasonably precise but it is still difficult to find overall agreement between current IN measurement techniques, at least using illite NX as the standard and allowing partners to analyze it independently. For instance, it is still difficult to compare ice nucleation results because sample preparation techniques and measurement methods (e.g., particle dispersion and size distribution characterization) differ from group to group, which also can contribute to the scatter of data (i.e., n_s diversity). Hence, a continued investigation to obtain further insights into consistencies or diversity of IN measurement techniques, perhaps by assembling and comparing them using identically processed test dust samples over similar thermodynamic conditions as demonstrated in ICIS-2007, is important (i.e., Page 22092 Lines 5-13).

Reviewer's comment: *Can the authors state what they consider the proof of adequacy of the n_s analysis? The size-sorted results? Also, could they explain what is meant (22090/15) by "uniform distribution of active sites for available S_{total}"? Independence of site density from particle size? How well is that proven?*

Authors' response: *Hoose and Möhler* (2012) compiled ice nucleation efficiencies of atmospheric aerosol by evaluating aerosol-specific 'singular' freezing onsets when or after specific ambient conditions were met. Such time-independent and surface area-scaled n_s formulations, originally developed by *Connolly et al*. (2009) and *Niemand et al*. (2012) on the basis of earlier suggestions by *DeMott et al*. (1995), have been recently adapted to assess the nucleation in a wide range of atmospherically relevant *T*-RH_{ice} conditions (i.e., T > -78 °C; *Hiranuma et al*., 2014a).

In the present work, we examined two premises of the n_s analysis, namely time independence and size independence. For the former, strong temperature dependence and weak time dependence of immersion freezing using illite NX particles are presented in Sect. 4.3 of the current manuscript. For the latter, we previously demonstrated the size independence of the n_s value using two different sizes of submicron hematite particles (200 and 1000 nm volume equivalent diameter; *Hiranuma et al*., 2014a). This was based on the AIDA deposition mode nucleation experiments. We have evidence that this size independence of the n_s value remains true for submicron illite NX particles based on the AIDA and CSU-IS, in which the n_s values derived from polydisperse and quasi-monodisperse populations overlap (See Figs. 4b and 5b, Figs. 4g and 5g). We also present the magnified version of Fig. 5g below [Note temperature and n_s uncertainty for the AIDA immersion experiment is ± 0.3 ˚C and ± 35%, respectively (*Möhler et al*., 2003; *Steinke et al*., 2011)]. Additionally, a size independence of the freezing behavior for particles with different sizes was reported in *Wex et al*. (2014) and *Augustin-Bauditz et al.* (2014). Nevertheless, more experiments with size-selected particles, in particular those larger than 0.5 µm, are needed to further investigate the size dependence of n_s (*Wheeler et al*., 2014).



Extra Figure. Magnified section (*T* < -25 ºC and $n_s > 10^8$ m^{-2}) of Fig. 5G with three size subcategorizations. The number in brackets represents the DMA set point size in mobility diameter. This figure is not shown in the manuscript since the data used to generate this figure (i.e., *T*, n_s and DMA size setpoint) are summarized in publically accessible data base available at http://imk-aaf-s1.imk-aaf.kit.edu/inuit/ as already mentioned in Page 22059 Lines 6-9.

For clarity, we modified Page 22068 Lines 19-. The text now reads, "Ice-nucleating efficiencies of both polydisperse and quasi-monodisperse illite NX particles were investigated in this study. n_s of DMA size-selected illite NX particles (200, 300 and 500 nm mobility diameter) agreed well with that of the polydisperse population for immersion freezing experiments, within previously reported uncertainties (T ± 0.3 ˚C and n_s ± 35%; *Steinke et al*., 2011).".

We added the following sentence in Page 22068 Line 23.

Added text: "Previously, *Hiranuma et al* (2014a) demonstrated the size independence of the n_s value using two different sizes of submicron hematite particles (200 and 1000 nm volume equivalent diameter) based on AIDA deposition mode nucleation experiments. Such a similarity might remain true for the immersion mode freezing of mineral dust particles that are smaller than 1 µm diameter."

We also added the following sentences in Page 22071 Line 6.

Added text: "...were compared). Furthermore, a size independence of the immersion mode freezing was seen for Fluka-kaolinite particles with mobility diameters of 300 and 700 nm in *Wex et al*. (2014), and for illite NX particles when comparing particles with mobility diameters of 500 nm to bulk material (*Augustin-Bauditz et al*., 2014)."

In addition, Page 22053 Lines 13-15 now reads, "Results of freezing efficiencies at specific temperatures are presented using the ice nucleation active surface-site density (n_s) parameterization (e.g., *Connolly et al*., 2009; *Niemand et al*., 2012; *Hoose and Möhler*, 2012) developed on the basis of suggestions by *DeMott et al*. (1995).".

New Reference:

Augustin-Bauditz, S., Wex, H., Kanter, S., Ebert, M., Stolz, F., Prager, A., Niedermeier, D. and Stratmann, F.: The immersion mode ice nucleation behavior of mineral dusts: A comparison of different pure and surface modifed dusts, Geophys. Res. Lett., 41, doi:10.1002/2014GL061317, 2014.

DeMott, P. J.: Quantitative descriptions of ice formation mechanisms of silver iodide-type aerosols, Atmos. Res., 38, 63–99, doi:10.1016/0169-8095(94)00088-U, 1995.

Reviewer's comment: *The overview of the results in Fig. 6 is not as informative as should be. This graph is valuable in demonstrating the overall trend of the results. However, the author might consider also displaying the results in terms of the ratios of the individual measurements to the geometric mean of all the data across the temperature range covered. That type of display would provide a clearer depiction of the data for evaluating trends with respect to each measurement technique. Also, it would be useful to see results presented separately for the suspension measurements and for the dry aerosol measurements. The influence of sample size is neglected in the analysis. Weighting data points by error ranges resulting from sample sizes would have been useful.*

Authors' response: This is a good suggestion. We added new figures (Fig. S4-S8) in SI Lines 742-787. The authors would like to present these figures after introducing the T-binned figures (i.e., Fig. 8) because the ratios are in part calculated based on the T-binned interpolated data. Accordingly, we also add the following paragraph in the manuscript Page 22079 Line 2.

"In addition, T-binned $n_{s,\mathrm{BET}}(T)$ and $n_{s,\mathrm{geo}}(T)$ spectra averaged in the log space are presented in Fig. S3. Similarly, we also present T-binned ratios of the individual measurements to the log fit of the data [All (log), Sus (log) or Dry (log) from Table 3] across the temperature range covered for all the measurement techniques (-37 ˚C < T < -11 ˚C) in the Supplement Figs. S4-S8. These figures provide inter-comparisons of the n_s deviations across the various techniques employed in this study."

The text added in the updated version of SI Lines 742-754 reads, "Figures S4 depicts the n_s diversity in $\log(n_{s,\mathrm{ind.}})/\log(n_{s,\mathrm{fit}})$, which represents the ratio of the individual measurements ($n_{s,\mathrm{ind.}}$) to the log fit line to either all data [All (log)], the suspension data [Sus (log)] or the dry-dispersed particle data [Dry (log)] as $n_{s,\mathrm{fit}}$. The interpolated T-binned data (i.e., interpolated data points in Figs. 4 and 5) are used for $n_{s,\mathrm{ind.}}$. The fit in the log space, which is derived from the parameters summarized in Table 3, is used as a denominator to avoid a bias of sudden jump of the reference value at certain temperatures where the number of available data changes. As shown in the figure, data deviation (i.e., scatter from the Avg. $\log(n_{s,\mathrm{ind.}})/\log(n_{s,\mathrm{fit}}) = 1$ line) can be seen in both suspension measurements and dry aerosol measurements. This deviation is observed with all the $n_{s,\mathrm{fit}}$ cases [All (log), Sus (log) and Dry (log)]. Additionally, the scatter of individual non-T-binned data and the validity of interpolations are presented in Figs. S5-S8. In specific, these four figures (Figs. S5-S8) complement panels a.ii and a.iii, panels b.ii and b.iii, panels a.iv anda.v and panels b.iv and b.v. from Fig. S4, respectively, in greater detail."

As shown in these figures, data deviation (i.e., scatter from the Avg. $\log(n_{s,\mathrm{ind.}})/\log(n_{s,\mathrm{fit}}) = 1$ line) can be seen in both suspension measurements and dry aerosol measurements. This deviation is observed with all the $n_{s,\mathrm{fit}}$ cases [All (log), Sus (log) and Dry (log)].

Page 22074 Lines 27-28 now reads, "Similarly, dry-dispersed particle measurements also exhibit scattered data for their measured temperature ranges.".

Page 22073 Lines 7-9 reads:

Original: "Overall, compared to suspension measurements, dry-dispersed particle measurements showed more pronounced diversity between measurements."
→
Modified: "Overall, compared to suspension measurements, dry-dispersed particle measurements showed higher n_s values."

Page 22073 Lines 14-15 deleted.

Deleted: "In-depth discussions of potential reasons for diversity specific to dry-dispersed particle measurements are given below (Sect. 4.)."

Reviewer's comment: *22059/Eq. (1) Since analysis of the data is being conducted with the time-independent singular approximation, it is somewhat misleading and needless to introduce Jimm in Eq. (1). This rate is not used in subsequent steps and there is no definition of what values of t are used for the different experimental methods. I recommend deleting the middle part of Eq. (1).*

Authors' response: The reviewer is correct in pointing out that we do not use J_{imm} in the rest of the manuscript. We simplified the equation as:

$$n_{s,\mathrm{geo}}(T) = -\ln\left(1 - \frac{N_{\mathrm{ice}}(T)}{N_{\mathrm{total}}}\right)\left(\frac{1}{S_{\mathrm{ve}}}\right), \quad (1)$$

Reviewer's comment: *There seems to be another problem with Eq. (1) in that it is unclear whether the logarithms is taken over both bracketed terms or only the first one. Is the equation dimensionally correct?*

Authors' response: Eqn. 1 is dimensionally correct. Logarithm is taken over the first bracketed term. The units on both sides match.

Reviewer's comment: *The value of writing Eqs. (1) - (3) in terms of size bins isn't really useful for this paper, since no size-resolved data are presented and neither were the measurements performed in a size-resolved manner.*

Authors' response: Prof. Vali is correct. None of the measurement PI produced n_s as a function of size; therefore, size summation symbols ($\sum_{i=1}^{n}$) from Eqns. 1 to 3 are omitted.

Reviewer's comment: *Could the authors address what uncertainties arise due to shape assumption, conversion to BET and DLS surface area?*

Authors' response: Shape assumption: For SMPS and APS measurements, a dynamic shape factor of 1.49 (± 0.12) was accounted for and used to estimate the volume equivalent diameter as mentioned in Page 22057 Lines 1-5. As an OPC measures optical scattering intensities from the particles which are converted to actual particle sizes by the Mie theory assuming spherical particles of known refractive index, an OPC cannot accurately measure sizes of non-spherical or irregularly structured particles. This typically results in overestimations of their actual sizes compared to the optical particle sizes by a factor of about two for non-spherical particles as discussed in Page 22057 Lines 11-14. This correction was also included to estimate the volume equivalent diameter.

BET and DLS surface area: For BET surface area, our experimental uncertainty is 124.4 ± 1.5 $m^2 g^{-1}$. The manufacturer report for the reproducibility of the DLS measurement is <5% (1% for 100 nm Polystyrene). We note that, as discussed in Page 22061 Lines 17-20, the 'representativeness' of our DLS surface area highly depends on the degree of agglomeration, and it could vary up to a factor of 13 based on our size distribution comparisons (i.e., potential effect of agglomeration, Sect. 4.4). For instance, agglomeration can reduce the surface area exposed to air or available to water as well as the S_{total}/M_{total} value (mentioned in Page 22061 Lines 14-15). Since we do not have size distribution measurements and associated S_{total}/M_{total} for each suspension measurement, DLS-SSA is presumably used for the data evaluation for the measurements with polydsispersed suspended particles throughout this study. Nevertheless, the usage of DLS-SSA is reasonable since the presence of fewer agglomerates in suspended particles has been demonstrated with hematite particles as shown in Fig. 1 of *Hiranuma et al*. (2014b). We presume such a similarity might remain true for the illite NX particles. Furthermore, the use of DLS-SSA (= 6.54 $m^2 g^{-1}$) is reasonable because the conversion factor ranged for size-selected particle diameters from 200 nm to 1000 nm (as discussed in Page 22061 Lines 3-6) is similar to DLS-SSA (also similar to AIDA-SSA; i.e., Fig. 2b). We also note that the discussion of potential effect of agglomerates is separately given in Sect. 4.4.

For clarity, we modified Pages 22061-22062 Lines 10-3, and it now reads:

"...in which, $n_{m,\text{sus}}$ is the IN active mass for suspension measurements, α represents the ice activated fraction (= N_{ice}/N_{total}), which is the direct measurement of suspension experiments and some of the dry-dispersed particle methods. With an assumption of a uniform BET-SSA, the resulting $n_{s,\text{BET}}$ may be representative of measurements with suspended samples because minimal corrections (only α and θ) are involved when compared to that with dry-dispersed particles. Owing to internal surface area and surface roughness, BET-SSA may be greater than DLS-SSA (*O'Sullivan et al*., 2014).

Alternatively, we can also convert ice-nucleating mass derived from suspension measurements, $n_{m,\mathrm{sus}}$, to $n_{s,\mathrm{geo}}$ using DLS-SSA to provide a reasonable comparison to dry-dispersed particle measurements. However, this process requires one more step than when using $n_{s,\mathrm{BET}}$ (with an additional assumption of constant size distribution for all suspensions) and two more steps than when using n_m. For our inter-comparison study, we used both $n_{s,\mathrm{BET}}$ and $n_{s,\mathrm{geo}}$. Because fewer conversion factors are involved, $n_{s,\mathrm{BET}}$ may be best suited for suspension measurements, and $n_{s,\mathrm{geo}}$ may be best suited for dry-dispersed particle measurements (Eqn. 3 to 4 or vice versa).

The usage of DLS-SSA for the calculation of $S_{\mathrm{total}}/M_{\mathrm{total}}$ of suspension measurements appears to be reasonable, as this leads to $n_{s,\mathrm{geo}}$ for suspension measurements nearly equivalent to $n_{s,\mathrm{geo}}$ for dry-dispersed particles. When $S_{\mathrm{total}}/M_{\mathrm{total}}$ is derived based on TSI-OPS measurements, a value of 0.49 $m^2\ g^{-1}$ is obtained, which is smaller by a factor of about thirteen compared to DLS-SSA. This difference may be mainly due to the fact that dry-dispersed particles are typically prone to agglomeration (discussed below, i.e., Sect. 3.1) compared to the measurements with suspended particles. The presence of fewer agglomerates in suspended particles is shown in Fig. 1 of *Hiranuma et al*. (2014b). Since the size distribution of a suspended sample for each experiment was not measured, DLS-SSA was used for the data evaluation for suspension measurements throughout this study."

Reviewer's comment: *22063/7-10 The authors state that the ". . . effects of impurities upon ice nucleation activity cannot be evaluated . . ." and that the impurities may be responsible for variations in ice nucleating efficiency at various temperatures. The underlying assumption here is that there is a specific temperature of activity associated with each component or impurity. If that is what the authors mean evidence need to be presented. Since that claim is made in the literature only for illite NX, the generalization here made is questionable.*

Authors' response: The authors agree that the generalization is not appropriate here. We modified the sentence in Page 22063 Lines 7-8:

Original: "Therefore, the possible effect of impurities upon the ice nucleation activity cannot be evaluated on the basis of its bulk analysis of the chemical composition."
→
Modified: "Therefore, the possible effect of these observed impurities in illite NX upon the ice nucleation activity cannot be evaluated on the basis of its bulk analysis of the chemical composition."

Reviewer's comment: *22063/13 It is unclear what special advantage illite NX has as a reference material over other minerals or other materials. The scatter in measured ice nucleating ability by different methods counters this statement.*

Authors' response: As stated in Page 22052 Line 27-, the objective of the INUIT project is to investigate the immersion freezing behavior of 'reference' particles. The INUIT group finds this commercially available illite NX is a quantitatively ideal reference of illite rich 'clay' material that can be shared among a large number of PIs. Moreover, illite NX samples contain relatively fewer impurities (e.g., quartz) when compared to other test dusts (e.g., IMt-1 illite contains 10 – 15% quartz based on manufacture report of clay mineral society; Arizona test dust contains ~17.1 % quartz as reported in Broadley et al. (2012)).

Besides illite NX, the INUIT group has comprehensively investigated the immersion freezing activities of Snomax (*Wex et al*., 2014b) and hematite (*Hiranuma et al*., 2014a and b) over the last three years. We will continue investigating the ice-nucleating ability of the other reference materials (e.g., K-feldspar) and more atmospherically relevant materials (e.g., soil dust, proteinaceous- and non-proteinaceous biological particles) in the next few years.

We now shorten and simplify the sentences for clarity:

Original: "Nonetheless, detection of non-illite mineral components implies the possibility of a wide range of ice nucleation efficiencies by the test sample at various temperatures. Hence, the illite NX sample may reflect the complexities of natural dust particles, which typically contain multiple sites with differing nucleation abilities, and can therefore be used as a reference material to mimic ice nucleation activity of physically and chemically complex natural dusts."
→
Modified: "Nonetheless, detection of non-illite mineral components may reflect the complexities of natural dust particles, which typically contain multiple sites with differing nucleation abilities. Thus, illite rich clay mineral can be used as a reference material to mimic the ice nucleation activity of physically and chemically complex natural dusts (*Murray et al*., 2012)."

Reference:

Wex, H., Augustin-Bauditz, S., Boose, Y., Budke, C., Curtius, J., Diehl, K., Dreyer, A., Frank, F., Hartmann, S., Hiranuma, N., Jantsch, E., Kanji, Z. A., Kiselev, A., Koop, T., Möhler, O., Niedermeier, D., Nillius, B., Rösch, M., Rose, D., Schmidt, C., Steinke, I., and Stratmann, F.: Intercomparing different devices for the investigation of ice-nucleating particles using Snomax® as test substance, Atmos. Chem. Phys. Discuss., 14, 22321–22384, doi:10.5194/acpd-14-22321-2014, 2014b. (Accepted in Atmos. Chem. Phys. On Dec. 20, 2014).

Reviewer's comment: *22066/8-10 What would it have meant if the results showed different $n_s(T)$ spectra for different mass concentrations? Dilution of samples with clean water is not normally expected to change the derived spectra. The statement here is a confirmation of that expectation not a new result.*

Authors' response: Yes, this observation is indeed expected when the experiments work properly. Therefore such observation is an important consistency check for this type of experiment. We have added two sentences for explanation.

We modified Page 22066 Lines 8-10, and the text now reads,

"Immersion freezing efficiency of illite NX particles collapsed into a single $n_s(T)$ spectrum, i.e. IN efficiency does not depend on suspended particle mass for the concentration range studied here. This observation is a check for consistency and it implies that ice nucleation is indeed triggered by suspended illite NX particles, and neither by impurities contained in the water used for dilution nor at the glass surface supporting the droplets. If IN efficiency did depend on suspended particle mass, different $n_s(T)$ spectra would result from the various illite NX concentrations, which are shifted by the respective dilution factor."

Reviewer's comment: *22066/14-17 Do the values given represent a cut-off size or the center of a narrow band in sizes?*

Authors' response: The latter is correct. The size is a narrow band of sizes centered at the mobility size selected by a DMA, as discussed in detail in the supplemental material (and as done by other PIs in this work).

Reviewer's comment: *22067/5 Typo in $n_s(T)$*

Authors' response: Corrected.

Reviewer's comment: *22067 What is meant by 'effective' surface?*

Authors' response: The word 'effective' does not add any values in the text. For clarity, we removed "effective":

Original: "…, implying that the absence of an effective surface in contact with a substrate has a negligible effect on immersion freezing for our experimental conditions."
→
Modified: "…,implying that the surface making contact with the substrate has a negligible effect on immersion freezing for our experimental conditions."

Reviewer's comment: *22067/15 abbreviate pL and nL as in previous paragraphs*

Authors' response: Page 22068 Line 2 now reads, "~400 picoliter to 150 nanoliter".

Reviewer's comment: *22068/22 Size-independence is a significant finding and deserves more detailed description (limits if validity, degree of agreement . . .)*

Authors' response: Page 22068 Lines 19- now reads, "Ice-nucleating efficiencies of both polydisperse and quasi-monodisperse illite NX particles were investigated in this study. n_s of DMA size-selected illite NX particles (200, 300 and 500 nm mobility diameter) agreed well with that of the polydisperse population for immersion freezing experiments, within previously reported uncertainties ($T \pm 0.3$ ˚C and $n_s \pm 35\%$; *Steinke et al*., 2011).".

We now add the following sentence in Page 22068 Line 23.

Added text: "Previously, *Hiranuma et al* (2014a) demonstrated the size independence of the n_s value using two different sizes of submicron hematite particles (200 and 1000 nm volume equivalent diameter) based on AIDA deposition mode nucleation experiments. Such a similarity might remain true for the immersion mode freezing of mineral dust particles that are smaller than 1 µm diameter."

We also added the following sentences in Page 22071 Line 6.

Added text: "... were compared). Furthermore, a size independence of the immersion mode freezing was seen for Fluka-kaolinite particles with mobility diameters of 300 and 700 nm in *Wex et al*. (2014), and for illite NX particles when comparing particles with mobility diameters of 500 nm to bulk material (*Augustin-Bauditz et al*., 2014)."

Reviewer's comment: *In Fig. 4 what does "AIDA size selected" refer to?*

Authors' response: As discussed above, a DMA was used to generate quasi-monodisperse particles (200, 300 and 500 nm mobility diameter) in the AIDA study.

Reviewer's comment: *22069/21 What discrepancy is being referred to?*

Authors' response: For clarity, we replaced "the discrepancy" with "high n_s values when compared to the other measurements".

Reviewer's comment: *22073/16 The title of Section 3.3 is not a good reflection of what is actually described.*

Authors' response: We agree, and the title of the Sect. 3.3 now reads, "Inter-comparisons based on the slope parameter of $n_s(T)$ spectra".

Reviewer's comment: *22073/17 I would have found it useful to have Figure 6 ahead of the detailed presentation of the results from each instrument. Discussions refer to differences from the overall trend, etc. which are not readily perceived from Figs. 4 and 5.*

Authors' response: Considering the large number of instruments involved in this inter-comparison paper, we found (after internal discussion) that it is best to discuss the individual instrument results prior to the compiled results.

Reviewer's comment: *22073/21 Typo: inns*

Authors' response: Thank you. Corrected.

Reviewer's comment: *22073/22-27 It is unclear to me whether these statements refer to the overall trend or some group of data sets.*

Authors' response: Within this T range (i.e., -27 ˚C $\leq T \leq$ -18 ˚C), the immersion results from all suspension measurements and a majority of dry measurements coexist (see the investigated T range for each technique in Table 1). Exceptions include LACIS, EDB and IMCA-ZINC.

Accordingly, we rephrased the sentence as:

Original: "Diversity is especially pronounced (for several orders of magnitude inn_s) at -27 ˚C < T < -18 ˚C, where the results from suspension measurements and a majority of dry measurements coexist."
→
Modified: "Diversity is especially pronounced for several orders of magnitude in n_s at -27 ˚C $\leq T \leq$ -18 ˚C, where the results from suspension measurements and a majority of dry measurements coexist (see the investigated T range for each technique in Table 1)."

Reviewer's comment: *22074/5 Aren't the numerical values of the slopes negative?*

Authors' response: Prof. Vali is right. For consistency and clarity, we added the definition of the slope parameter in Page 22073 Line 24.

"…slope in the spectrum (i.e., the absolute value of $\Delta\log(n_s)/\Delta T$ in log m^{-2} ˚C^{-1}, hereafter denoted as $\Delta\log(n_s)/\Delta T$)…".

Reviewer's comment: *22074/10 Since the fraction of active sites is reflected by the absolute values of n_s, it is unclear what the authors want to express here.*

Authors' response: We have modified this part of sentence to read:

Original: "…, suggesting that a large fraction of active sites of our test dust may trigger immersion freezing at…"
→
Modified: "…,suggesting that a dominant fraction of INP contained in our test dust becomes ice active in immersion freezing at …"

Reviewer's comment: *22074/14-20 There appears to be some repetition here.*

Authors' response: The authors thank Prof. Vali for pointing out this error. We have rephrased Page 22074 Lines 13-21 as:

"Similar observations are made by most of the other suspension measurement techniques. In short, most suspension methods capture the … containing 1.0 wt% illite NX (see the Supplementary Methods)."

Reviewer's comment: *22074/27 A possibly significant point is being made here - the amount of scatter in suspension measurements versus dry aerosol measurements - but this is masked by the larger number of the latter type of data. The authors could examine this difference in a rather simple way and it would be very useful to have that analysis presented in the paper.*

Authors' response: Discussed above (i.e., Figs. S4-S8; SI Lines 742-787).

Reviewer's comment: *22076/11-14 A resounding conclusion is stated here only to be qualified in lines 14-17, with more analysis promised. This is confusing. The reference to uniform distribution is not supported by any specific result.*

Authors' response: We agree with you. We do not have evidence to support the premise that active sites are uniformly distributed. For this reason, we delete Page 22076 Lines 11-14.

Reviewer's comment: *22076/19 Grammar issue: the past tense in this sentence conflicts with the reference to the section to follow and the next sentence which uses the present tense.*

Authors' response: "was elucidated" → "is further discussed"

Reviewer's comment: *22077/2 Typo: space missing between in and n_s.*

Authors' response: Corrected.

Reviewer's comment: *22077/2 What does shifting of activation temperatures mean?*

Authors' response: Matching the n_s values by shifting Ts horizontally rather than doing that for n_s vertically.

Page 22077 Lines 2-3 now reads:

"…whereas others may shift activation temperatures horizontally to match the n_s values from other instruments, perhaps biasing the overall accuracy and precision of instruments."

Reviewer's comment: *22077/5 So-called T-binned data presentation does hardly deserves to be used as section heading. It is a fairly standard procedure.*

Authors' response: Heading changed to "4.1 Dry vs. suspension $n_s(T)$ data".

Reviewer's comment: *22077/13 Typo: space between 'r' and 'for'*

Authors' response: Corrected.

Reviewer's comment: *22077/20 Grammar: past tene used here is out of sync with the rest of the writing*

Authors' response: Corrected.

Reviewer's comment: *22077/21 Don't the values of Hor(max − min) and Ver(max − min) depend on where those are taken? Are the values indicated in the graphs picked for particular reason? Are these the maxima within the gray bands for each value?*

Authors' response: Correct, $Hor_{Max\text{-}Min}$ and $Ver_{Max\text{-}Min}$ depend on temperature. The values shown on the Figure 7 are the "maximum" deviation we can find across all the measurements.

Fig. 7A Hor (-36.8 °C < T < -29.0 °C at n_s ~ 5.2e+09); Ver ($\log(n_{s,\max}/n_{s,\min})$ = 3.0 at -21 °C)
Fig. 7B Hor (-36.7 °C < T < -29.2 °C at n_s ~ 1.5e+11); Ver ($\log(n_{s,\max}/n_{s,\min})$ = 3.0 at -20 °C)

For clarity, we modified the Fig. 7 legend as:

"…The maximum deviation between maxima and minima in horizontal axis (in T ºC) and vertical axis [in $\log(n_{s,\max}/n_{s,\min})$] corresponds to $Hor_{Max\text{-}Min}$ and $Ver_{Max\text{-}Min}$, respectively."

We also modified Page 22077 Lines 20-22:

"It is observed that the largest deviation between the maxima and minima in the horizontal and vertical axes, corresponding to $Hor_{Max\text{-}Min}$ and $Ver_{Max\text{-}Min}$, respectively, shown in Fig. 7, is similar for both $n_{s,\mathrm{BET}}$ (Fig. 7a) and $n_{s,\mathrm{geo}}$ (Fig. 7b)."

Since $Ver_{Max\text{-}Min}$ and $Hor_{Max\text{-}Min}$ are similar for n_s BET and for n_s geo in this definition, we also modified Page 22077 Lines 22-26.

"Nevertheless, $n_{s,\mathrm{BET}}$ is representative of measurements with suspended samples because fewer corrections are involved for its estimation when compared to that with dry-dispersed particles."

As discussed and shown above, we cannot say for certain that $n_{s,\mathrm{BET}}$ is a better proxy for inter-comparison of the IN measurements. Therefore, Page 22091 Lines 13-15 now reads, "In addition, two different n_s metrics, $n_{s,\mathrm{geo}}$ and $n_{s,\mathrm{BET}}$, were compared, and we found that $n_{s,\mathrm{BET}}$ is a better proxy for suspension-based IN measurements, while $n_{s,\mathrm{geo}}$ is better for dry-dispersed particle measurements.".

Reviewer's comment: *22078/1-3 What is the reason for expecting the results here given?*

Authors' response: We mean the results are consistent with the results described in Sect. 3.3 (i.e., Inter-comparisons based on the slope parameter of $n_s(T)$ spectra). We modified:

Original: "As expected, the slope is comparable to A13 in the T_1 to T_3 segment (-11 to -27 ˚C), while the slope in the T_4 segment is similar to N12. The largest deviations in $Ver_{Max\text{-}Min}$, corresponding to two to three orders of magnitude of n_s, were…"
→
Modified: "The slopes are comparable to the slope of the A13 parameterization in the T_1 to T_3 segments (-11 to -27 ˚C), while the slope in the T_4 segment is similar to those of the N12 parameterizations. These results are consistent with the results described in Sect. 3.3. Further, $Ver_{Max\text{-}Min}$ for roughly three orders of magnitude with respect to n_s is …"

Reviewer's comment: *22078/3-7 The valuation of Ver(max − min) is too limited. Only the point of its highest value is commented on. It would be useful to provide more information about its numerical value across the entire temperature range. 22078/7-10 What meaning do the authors attach to Hor(max −*

min)? Clearly, the numerical value of Hor(max − min) is much larger than any temperature measurement error. Is the authors' interpretation related to variations in the activity of sites between one or other measurement method? If so, what reasons can be given for such changes? If Hor(max−min) is just a reflection of the spread along the abscissa, it does not merit the introduction of a new parameter.

Authors' response: The Ver $_{Max\text{-}Min}$ value provides the maximum deviation in $\log(n_{s,max}/n_{s,min})$, and we would like to keep it as is (discussed above). As now stated in Page 22078 Lines 3-4, $Ver_{Max\text{-}Min}$ values varied up to three orders of magnitude (or $\log(n_{s,max}/n_{s,min})$ ~ 3.0). The max Ver value was observed ~ -20 ºC (now reads, "…$Ver_{Max\text{-}Min}$ for roughly three orders of magnitude with respect to n_s is observed in a temperature region around ~ -20 ˚C for both $n_{s,BET}(T)$ and $n_{s,geo}(T)$ spectra.").

Likewise, the $Hor_{Max\text{-}Min}$ value provides the maximum deviation of the seventeen immersion freezing measurement techniques (about 8 ˚C in terms of temperature). We would also like to keep $Hor_{Max\text{-}Min}$ discussions. In the paper $Hor_{Max\text{-}Min}$ deviation is discussed, see Page 22078 lines 7-10 [now reads, "…our $Hor_{Max\text{-}Min}$ shows that the seventeen measurements are in reasonable agreement within 7.8 ˚C (-36.8 ˚C, -33.0 ˚C, -29.0 ˚C (*min, log fit, max*)) at $n_{s,BET}$ of 5.2 x 10^9 m^{-2} and 7.5 ˚C (-36.7 ˚C, -32.8 ˚C, -29.2 ˚C (*min, log fit, max*)) at $n_{s,geo}$ of 1.5 x 10^{11} m^{-2}"].

Accordingly, we also modified the following sentences in Abstract and Conclusion.

Abstract: Page 22048 Lines 1-3: "the seventeen immersion freezing measurement techniques deviate, within the range of about 8 ˚C in terms of temperature, by three orders of magnitude with respect to n_s.".

Conclusion: Page 22091 Lines 7-9: "the seventeen immersion freezing measurement techniques deviate, within a range of about 8 ˚C in terms of temperature, by three orders of magnitude with respect to n_s."

***Reviewer's comment:** 22078/17 Please clarify what you mean by pronounced freezing and differences. In fact, the intention behind this whole sentence is a bit vague.*

Authors' response: We modified the sentence to clarify this. We changed words: "pronounced freezing and differences" → "abrupt increase in $\Delta\log(n_s)/\Delta T$ and n_s deviations"

***Reviewer's comment:** 22078/19 This paragraph mixes past and present tense wording.*

Authors' response: We modified the sentence to clarify this. We changed words to, "…over a wide range is of great advantage…".

***Reviewer's comment:** 22078/23 The distinction drawn for experimental methods using dry aerosol inputs as 'working on a particle by particle basis' is vague. Doesn't the evaluation of suspension measurements also assume that each nucleating site is located on a different particle? The authors are hinting at a subtle point which is not explored in detail and is poorly expressed by what is said. The main difference, in my view, is that suspension methods run into background problems at cold temperatures and that dry aerosol methods lack sensitivity (sample volume) at warmer temperatures.*

Authors' response: We have changed the text according to the reviewer's suggestion.

Original: "In turn, dry-dispersed particle measurements were advantageous for their capacity to work on a particle by particle basis and can readily explore particle size dependencies. Further, these measurements…"
→

Modified: "suspension experiments with small picoliter or nanoliter droplets allow measurements right down to the homogeneous freezing limit (~ -37 ºC; *Koop et al*., 2000). In turn, suspension methods with microliter droplets may run into 'background problems' at temperatures below about -20 °C to -25 °C for samples that do not contain many IN active at these temperatures, because then impurities contained in the water may trigger freezing. Conversely, dry aerosol methods lack sensitivity for detecting rare IN at high temperatures because of their low sample volume. These dry particle measurements are in general good…"

We also add the text regarding background freezing in FRIDGE (SI Lines 304-307):

"Background freezing induced by impurities in the water was observed at T < -23 °C. This background freezing contributed to less than 15 % of the overall freezing in the range of -25 °C < T < -23 °C and was accounted for the n_s estimation."

Reviewer's comment: *22078/29 Freezing efficiency is not defined.*

Authors' response: Corrected. The authors meant n_s.

Reviewer's comment: *22079/4-11 While it is easy to agree with the general point being made here, the meaning of many parts of this paragraph is quite vague. What is meant by systematic uncertainty, absolute standard technique, . . .? I think that what is said in this paragraph would be better placed in the Introduction.*

Authors' response: We agree with Prof. Vali. This paragraph is not engaged with the IN discussion; therefore, the 1st paragraph discussion does not add any scientific merit to the manuscript. It is now removed.

Reviewer's comment: *22082/23 Was the SBM fit obtained using the LACIS data points or to the straight line shown in Fig. 9?*

Authors' response: A contact angle distribution was fitted to the frozen fractions measured with LACIS. When then SBM calculations are done and the resulting frozen fractions are converted to n_s, this results in the straight lines shown in Fig. 9.

The text in Page 22082 Lines 23-29 was modified.

"Specifically, a contact angle distribution was fitted to the LACIS measurements and was used, together with the soccer ball model (SBM; *Niedermeier et al*., 2011 and 2014), to simulate frozen fractions for different residence times varying over four orders of magnitude (i.e., 1, 10, 100 and 1000 s residence time). These frozen fractions were then used to calculate n_s, shown as lines in Fig. 9. More specifically, frozen fractions for 500 nm diameter illite NX particles were calculated based on SBM to obtain $n_s(T)$ spectra."

Reviewer's comment: *22085/5 Are particles removed from the filter with full efficiency in the washing process? If that is not sure, it should be mentioned as a potential explanation of the observed discrepancy.*

Authors' response: High efficiency particle removal has been demonstrated by the authors. SI Lines 286-288 now reads, "It is noteworthy that the application of the ultrasonic bath and its high efficiency in the washing process for particle removal were demonstrated with a similar experimental setup employed by *Ardon-Dryer and Levin* (2014).".

Reviewer's comment: *22085/7 Description of this method for FRIDGE is missing in the Supplementary Methods.*

Authors' response: We thank you for pointing out this error. We now add the texts for the FRIDGE immersion mode operation in SI lines 275-307.

Reviewer's comment: *22090/23 This paragraph is rather confusing, specially the first sentence.*

Authors' response: We rewrote the paragraph based on the modifications discussed earlier (i.e., Fig S4-S8; SI Lines 740-787):

"Furthermore, comparisons of the suspension subsets against the dry-dispersed particle techniques were performed. Dry samples alone showed higher n_s values compared to the pre-suspended samples above -27 ºC. A possible explanation for this deviation (i.e., n_s from dry-dispersed methods > n_s from suspension methods) may be the surface modification of the illite NX particles (e.g., due to ion dissolution effects in the aqueous suspension)."

Reviewer's comment: *22091/28 Could you clarify what is meant by 'temperature change is the major driver of immersion freezing"?*

Authors' response: We wanted to point out that our observations show that immersion freezing efficiency of illite NX particles is temperature-dependent and increases as the temperature decreases. We revised the text to clarify this point.

"…our observations show that temperature is the major variable influencing the immersion freezing of illite NX particles, as the n_s values in general increase while temperature decreases."

Reviewer's comment: *22092/1 What is the connection of this sentence to the previous one?*

Authors' response: There is no connection. For clarity, we modified the sentence. Page 22091/22092 Lines 28-4 now reads, "In addition, our results of n_s and absolute values of $\Delta\log(n_s)/\Delta T$ distributions across a wide range of temperatures imply that clay minerals may contain various freezing activation energies, and the immersion freezing nature of clay minerals (e.g., illite NX) in a wide range of temperatures cannot be fitted by simple exponential functions but are governed by a hybrid of multi-exponential functions (a combination of scaled A13 and N12 parameterizations).".

Additional revision: In addition to addressing the reviewers' comments, other editorial corrections (major ones) are made as seen in the last four pages (Pages 12-15) of our response to the reviewer #1.