Dear Editor,

Please find attached our revised manuscript and supporting information (SI). We have carefully addressed all the comments from the reviewers. The reviewers' comments are presented in black, and our responses are provided in red. We have also revised the main text accordingly and highlighted the changes in blue.

Reviewer 1:

I would like to thank the authors for their thoughtful and thorough response and revisions, the manuscript is much improved and many points have been clarified.

There are still several points which should be considered further, but this is an interesting and thought-provoking work. I believe the manuscript is publishable after minor revisions and does not need further review.

The equivalent diameter normalization is still somewhat unsatisfying to me in the methodology and reasoning, but because it is appropriately described and discussed I don't think we need to be in perfect agreement for this to be publishable. My comments regarding this:

1) The 50% and 10% freezing thresholds for solution and aerosol measurements are still largely arbitrary. I understand your point that a 50% threshold in aerosol studies isn't a good point of comparison to 50% freezing in solution studies, but that doesn't mean that 10/50% should compare well. If you have size distribution data, you could calculate a 10% active fraction (INP concentration/particle concentration) for solution measurements, and this would at least be a fair comparison. You can't do it with just a BET isotherm for suspension samples unfortunately, so it might remove a lot of viable points with the current method.

We thank the reviewer's comment. We agree that comparing the 10% freezing threshold for aerosol measurements with the 50% threshold for suspension measurements is not directly comparable. However, as the reviewer pointed out, the majority of published suspension studies report only BET-derived surface areas rather than total particle number concentrations or size distributions in suspensions, making it difficult to derive a 10% activated fraction from suspension measurements.

Adopting the 10% and 50% thresholds therefore represents a practical operational compromise that allows us to extract a characteristic freezing temperature from each measurement method while retaining maximum data coverage across the published datasets. We acknowledge that the choice of frozen or active fraction threshold may lead to a shift in the corresponding threshold freezing temperature. We added a statement to clarify this point in the manuscript in Line 375-376 "We note that the selection of different frozen or active fraction threshold may lead to a shift in the threshold freezing temperature."

2) A related issue: For solution studies, the 50% freezing threshold is not an intrinsic property of an ice nucleating material, but an extrinsic property that depends on the solution/suspension concentration and the size of droplets. Yes, it is used commonly in the literature and I take issue with it there too because meaningful comparisons between 50% freezing thresholds are only possible if concentration and droplet size are constant. Active fraction has this same issue to a

lesser extent because it only depends on aerosol size distribution (for a given ns, active fraction increases as size distribution shifts to larger sizes).

We completely agree with the reviewer that the 50% (median) freezing temperature is determined by droplet size and suspension concentration, specifically, the total particle surface area contained within each droplet. This is precisely the rationale behind Figure 3 showing a warmer freezing temperature of droplets with larger equivalent particle diameter (D_e) due to high particle concentration included within a droplet. The purpose of showing freezing temperature is not to compare the ice nucleation ability (INA) of different samples, but to point out the high concentration of particles used in suspension measurements would lead to a warmer freezing threshold of a droplet (Figure 4a). Therefore, there is a higher possibility of particle coagulation within suspension samples which may reduce the effective surface area for ice nucleation and lead to a lower suspension n_s values compared to those from aerosol measurements. When comparing INA of different samples, we derived ice active site surface density (n_s) that considers the droplet volume and particle size which allows for a fairer comparison.

3) With less importance placed on aggregation, I wonder if spherical equivalent diameter is the correct parameter to discuss. I can understand some of the appeal, but is there a physical reason why equivalent diameter with a linear fit is more meaningful than ns (IN active surface site density) with a parabolic fit? Because mathematically they produce the same curve shapes/fits.

Both the D_e and the n_s based parameterizations are developed based on their empirical relationships with temperature. The D_e based parameterizations describe how the freezing temperature of a droplet changes with the total particle surface contained within that droplet, which highlights the issue with describing freezing thresholds by median freezing temperature, (see comment above) whereas the n_s based parameterizations describe the surface-normalized INA of dust particles as a function of temperature. We therefore do not consider either parameterization to be intrinsically more meaningful or physically superior to the other, as they emphasize different ways of representing droplet freezing and are useful under different application conditions:

We pointed out in Line 675-Line 677 for D_e based parameterizations: "Unlike the n_s parameterizations, it does not require distinguishing between measurement methods and temperature regimes, allowing for representing droplet freezing over the entire MPC temperature range with a single expression." and in Line 693-695 for n_s based parameterizations: "All n_s-based parameterizations can be applied to estimate the contribution of dust aerosol to INP concentrations and primary ice formation in different atmospheric scenarios, if the corresponding surface area of dust particles is known.".

Ultimately for #2 and 3 above, it could be that given a relatively small range of experimental conditions used for experiments (that impact suspension concentrations, drop volumes, and aerosol distributions) the impact is only some added spread to the data. The fact that freezing temperature changes approximately logarithmically with some of these variables also helps you, and could be discussed. But I think these are still important issues that more careful attention should be paid to.

We agree that variations in suspension concentrations, droplet volumes and aerosol properties can affect the freezing temperature of samples. The spread in freezing temperature observed at similar D_e values in Figure 4 also suggests the influence of additional experimental factors that influence droplet freezing. Despite this variability, a clear log-linear relationship between freezing temperature and D_e remain across the compiled datasets, indicating that the developed D_e parameterizations capture the general trend of droplet freezing. To address reviewer's concern, we added the following statements at Line 478-Line 481: "We also note the variability in freezing temperature observed at similar D_e values (Figure 4), indicating that other factors other than D_e, including droplet volume, dust mineral composition and other experimental conditions, may also influence droplet freezing."

Reviewer 2:

Chen and Kanji present a literature synthesis of the ice nucleation ability of mineral dust aerosol. Many published studies from the past two decades are aggregated. The data are analyzed in the ice nucleation active site framework. Two main threads in the literature discussion, dry vs. wet suspension methods and sample again in water are revisited. Active site based parameterizations are developed and the authors' point of view on the observations is presented.

This manuscript has received two primary referee reports to which the authors have responded in great detail. The discussion is valuable reading for researchers that are interested in the topic. In this referee's opinion, the collation of the datasets provides a valuable summary of the current state of knowledge. The stated hypotheses in the abstract that aerosol generation methods contribute to variability and water may act as a physical or chemical medium of aging are plausible. However, I am not convinced that the authors found compelling new evidence that supports these hypotheses beyond what has been known and discussed previously. Nevertheless, the compilation is valuable as it provides empirical bounds for ns of mineral dust that can be used to evaluate sensitivity in field studies and model simulations. Since the discussion is published alongside the manuscript, and the compilation is intrinsically valuable, I support publication of this manuscript in ACP.

Toward the last point, evaluation of model and earth system sensitivity, I want to encourage the authors to translate the ns-band parameterization to INP number concentration vs. temperature using typical mineral dust surface areas. Those values should be compared with atmospheric compilations of INP number concentration. I believe (though I could be wrong) that this will show the atmospheric irrelevance of dust at T > −15 °C, which in this referee's view reflects some general consensus in the field.

We thank the reviewer for the comments. We absolutely agree with the reviewer that translating the n_s-band parameterization to INP concentration and comparing to observations would be an exciting addition to the paper. However, to do this well, in different areas, at different temperatures (altitudes) and for different dust species (scaled pure components or mixtures) is beyond the scope of this paper. We are in fact in an ongoing project doing exactly that, where we estimate atmospheric INP concentrations based on our developed n_s parameterizations (shown in Figure 3) and dust concentration outputs from a climate model. From the ongoing

work we can already see that the modeled INP concentrations with our new parameterizations being compared with field observations conducted over the Southern Great Plains (SGP) site are approximately 0.01 – 0.1 L^{-1} at $-5 \leq T \leq -15$ °C, which are lower than the total INP concentrations of 1 – 2 L^{-1} measured by PINE at $T = -15$ °C. We also modeled INP concentration and compared them with the observed INP concentrations during the Aerosol-Ice Formation Closure Pilot Study. The modeled dust INP concentrations are 0.001 – 0.02 L^{-1} at T $-5 \leq T \leq -15$ °C, compared with 0.001 ~100 L^{-1} from the observations by Ice Spectrometer (IS) and Portable Ice Nucleation Experiment (PINE) measurements at identical temperatures.

For both simulations, the modeled INPs using our proposed n_s parameterization in this work show closer agreement with the field observations than those predicted using the parameterizations of Demott et al. (2015) and Niemand et al. (2012). The detailed results are being further investigated and discussed in a separate study. Nevertheless, the model simulations suggest our dust n_s parameterizations do not overestimate the atmospheric INP concentration under the investigated conditions.

An exception may be the anomalously large ns values reported for a subset of ATD and montmorillonite observations. It may well be that those larger ns would result in significant and perhaps unrealistically large INP concentration at T = −10 °C. For example, ns = 10^11 m−2, for ATD at −10 °C, would result in INP = 1000 L−1 assuming a moderate dust surface area of 10^−5 m2 m−3. I am unaware that such high INP concentrations have been measured in the atmosphere at this temperature. If ns is indeed this large, there should be multiple lines of evidence supporting it. Importantly, such concentrations would be readily observable with "online" instruments. Residuals analyzed from such an event would easily prove that mineral dust active at −10 °C is present and dominant in the atmosphere. Whether my personal reading of the preponderance of evidence is correct or not, this is a testable hypothesis that a paper like this should advance to truly advance the lively debate between the different camps.

We agree that the high n_s values observed for ATD and montmorillonite from aerosol measurements at $T > -10$ °C are close to the limit of detection the instrument used. When developing the general n_s parameterizations for aerosol and suspension measurements (Figure 3), these potentially uncertain data points at $T > -10$ °C were excluded. To clarify this in the manuscript, we added the following statements to the caption of Figures 2 and 3, respectively: "The aerosol n_s data points measured at $T > -10$ °C for ATD and montmorillonite are excluded from parameterization development, as these measurements are likely to reach the lower limit of detection for aerosol instruments." and "The aerosol n_s data points measured at $T > -10$ °C are shown for completeness but are excluded from parameterization development, as these measurements are likely to reach the lower limit of detection for aerosol instruments.".

In addition, when composition-based n_s parameterizations are used to estimate INP concentration for a specific mineral group, the mineral-specific surface area concentration should be used rather than the total dust surface-area concentration. For example, the effective illite surface-area concentration can be calculated by multiplying the total dust surface-area concentration by the surface-area fraction of illite. Because the mineralogy at the surface is not available, the mass fraction is often used. To clarify this point, we added the following statements in Line 700-704: "When composition-based n_s parameterizations are used to

estimate INP concentrations for a specific mineral group, the effective surface area should ideally be calculated by scaling the total dust surface area by the surface area fraction of that mineral. However, since the surface area fraction of individual minerals is generally unknown, the mass fraction is commonly used as a proxy (Price et al., 2018).".

ATD is widely used as a proxy for atmospheric mineral dust. However, it is a commercial test dust with a higher K-feldspar fraction than typical atmospheric dust (Murray et al., 2012) and the surface area/mass fraction of ATD-like dust particles in the atmosphere is unknown. We performed an illustrative calculation using illite. The illite n_s from our aerosol parameterization is $10^{7.76346}$ m^{-2} at T = -15 °C (Figure 2). We assumed that illite accounts for 70% of the total dust surface area in the atmosphere, representing a relatively high illite abundance scenario (2 – 70% mass fraction of illite in transported airborne dust reported by Murray et al. (2012)). The assumed total dust surface area concentration in the atmosphere is 50 μm^2 cm^{-3} taken from Harrison et al. (2019). When considering that illite can account for 2 – 70 % mass fraction of transported airborne dust, with an average of ≈45% (Murray et al., 2012), this estimate leads to an INP concentration of ≈1.3 L^{-1}. In comparison, the dust INP concentration reported in Kanji et al. (2017) is 0.1–10 L^{-1} at T = −15 °C, where the higher range is expected to be present in dust events. If the suspension n_s parameterization is used or if a lower fraction of illite is assumed, the estimated dust INP concentration would decrease. In summary, both our preliminary model simulations and the illustrative illite calculation suggest that our n_s parameterizations do not lead to unrealistically high estimates of atmospheric INP concentrations, supporting their atmospheric relevance.

We also added the atmospheric relevance calculation at Line 700-718: "When composition-based n_s parameterizations are used to estimate INP concentrations for a specific mineral group, the effective surface area should ideally be calculated by scaling the total dust surface area by the surface area fraction of that mineral. However, since the surface area fraction of individual minerals is generally unknown, the mass fraction is commonly used as a proxy (Price et al., 2018). To illustrate the application of a composition-specific parameterization, we used the illite n_s from our aerosol parameterization, which is $10^{7.76346}$ m^{-2} at T = −15 °C (Figure 2 and Table S4). The assumed total dust surface area concentration in the atmosphere is 50 μm^2 cm^{-3} taken from Harrison et al. (2019). When considering that illite can account for 2 – 70 % mass fraction of transported airborne dust, with an average of ≈45% (Murray et al., 2012), this estimate leads to an INP concentration of ≈1.3 L^{-1}. In comparison, the dust INP concentration reported in Kanji et al. (2017) is 0.1–10 L^{-1} at T = −15 °C, where the higher range is expected to be present in dust events. When the parameterization from Figure 3 (Table S4) is used (relevant for global average applications), the total dust surface area concentration can be used directly, without the need to scale based on a single mineral. The aerosol n_s is $10^{8.99443}$ m^{-2} at T = −15 °C (Figure 3 and Table S4). Using the same total dust surface area concentration, this gives an estimated dust INP concentration of ≈50 L^{-1} at T = −15 °C, higher than the aforementioned range of 0.1–10 L^{-1}. Using the suspension n_s parameterization or assuming a lower dust surface area concentration would yield a lower estimated dust INP concentration.".

Other comments

The aggregation of data in Figure 1 lacks any direct notion of measurement uncertainty and judgment whether all of the data are, and should be, considered equally valid. Simply throwing all data into the average will not reduce system uncertainty. Eventually some data will need to be marked as suspect until there is independent verification.

We agree, Figure 1 did not explicitly identify potentially uncertain data points from individual study, as its primary purpose is to illustrate the overall ice nucleation ability (INA) across different mineral compositions. To address this concern, we added 95% prediction bands for the n_s datasets in Figure 2 (also shown below), while accounting for systematic differences between measurement techniques. Data points falling outside these prediction bounds represent deviations and are treated as outliers when deriving n_s based parameterizations. When comparing the INA of dust particles across different mineral compositions (Figure S2), measurement methods (Figure S4), and aging processes (Figure 6), we calculated median $n_s$values for each temperature bin. Points falling outside 1.5 × IQR (Interquartile Range) in $\log(n_s)$ were identified and excluded as statistical outliers. The conclusions made from these comparisons thus capture the major trends after filtering out these outliers. We also excluded aerosol n_s data points measured at $T > -10$ °C for parameterization development (Figure 3), as these measurements are likely to reach the lower limit of detection for aerosol instruments.

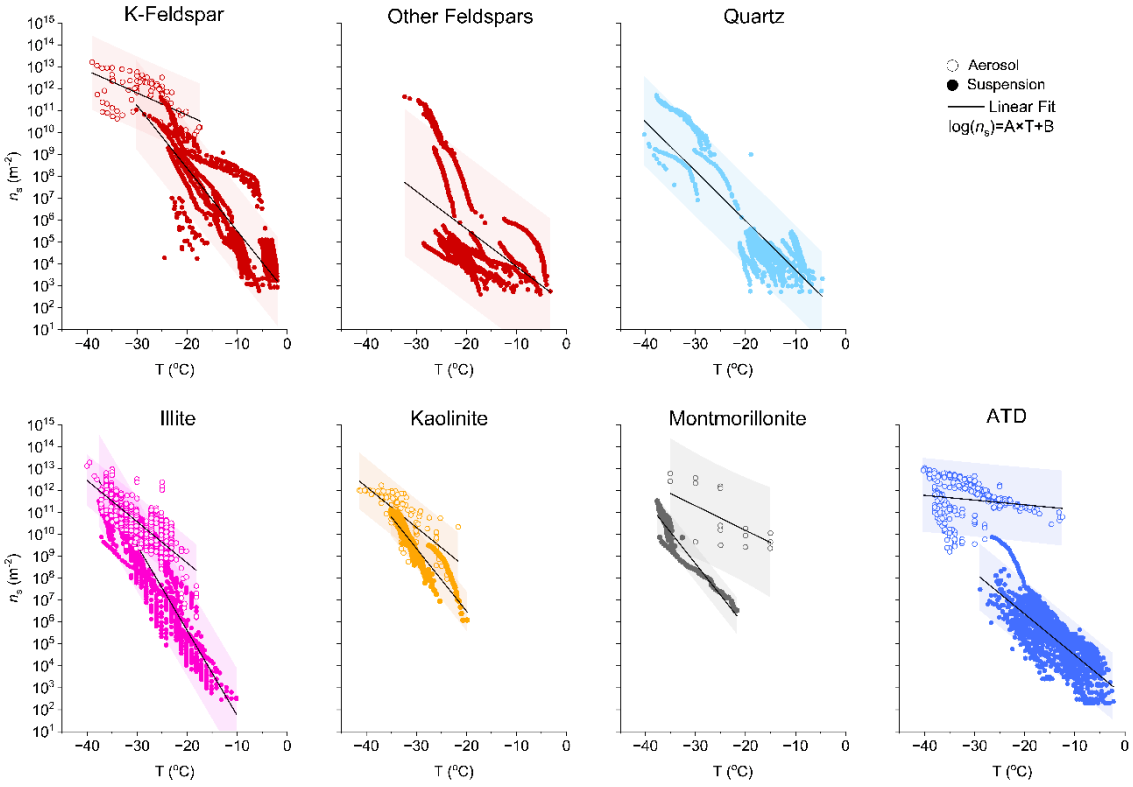


Figure 2. n_s of dust particles with specific mineral compositions. Empty and filled data point denote measurements using dry-dispersed aerosol and wet suspended particles, respectively. K-feldspar and non-potassium-rich feldspars are shown separately due to their distinct INA. For quartz and non-potassium-rich feldspars, only data from suspension measurements are available. Solid lines indicate linear fits of $\log(n_s)$ versus temperature, plotted separately for each measurement method. The values for fitting parameters (A and B) for each plot are listed in

Table S4. The data used in this figure were obtained from the studies listed in Table S3. The shaded area indicates 95% prediction band of each fitting curve. The aerosol n_s data points measured at T > −10 °C for ATD and montmorillonite are excluded from parameterization development, as these measurements are likely to reach the lower limit of detection for aerosol instruments.

Demott => DeMott throughout the manuscript

We have changed Demott to DeMott throughout the manuscript.

Please use proper minus signs (−) throughout the manuscript

We have changed minus signs (−) throughout the manuscript.

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