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:

- This manuscript describes meta-analysis of a wide range of recently published INP characterizations of mineral dust samples analyzed by several different techniques. Data is reanalyzed to create comparable quantities, and discrepancies between the different datasets are discussed, with a focus on 1) different materials, 2) different measurement techniques, and 3) different sample ageing conditions. Particle coagulation and wet ageing processes are considered for the source of discrepancies between different measurement techniques. Parameterizations are constructed for aggregate data, divided by measurement technique and sample ageing conditions. There is a lot of interesting data and discussion within this paper, however there are significant weaknesses that prevent any strong conclusions from being made.

Foremost: aggregate equivalent diameters are constructed to examine possible impacts of aggregation on measured freezing temperatures. This is an interesting idea, with some interesting data. In the manuscript it is taken as proof of extensive aggregation, however, which does not logically follow from any of the data or discussion. One of the following needs to happen: 1) Compelling evidence needs to introduced that extensive aggregation is occurring in droplet freezing measurements or 2) much of the manuscript needs to be rewritten to remove the unwarranted conclusion.

We thank the reviewers for their remarks and realize that the message we are trying to relay has been confused and confounded with aggregation. The role of coagulation was meant as a potential suggestion causing the lower INA of dust particles at higher freezing temperatures.

The main idea behind the discussion aimed to say that an outcome of having large surface areas present within a single droplet promotes freezing at much warmer temperatures than the aerosol suspension experiments. As such if the surface area within the suspension measurements are considered as coming from one particle as is the case with the aerosol measurements, then that particle would have to be extremely large in the single aerosol experiments to induce freezing at the same temperature as the suspension measurements. That is really the main point we were trying to make.

Furthermore, to observe one freezing event in the atmosphere at such warm temperatures, a large surface area of dust particles would need to be present (assuming all have the same composition and evenly distributed active sites) to observe the freezing of one ice crystal. As such aggregation is of secondary importance here, the surface area argument holds even in the absence of aggregation. This means that for the suspension experiments to be relevant to the atmosphere, one must consider many small atmospherically suspended particles would have the same ice nucleation potential as one large 10 – 100 μm dust particle (Figure 4). i.e. since the concentration of large dust particles is very low, the requirement would be based on many

smaller particles fulfilling the same surface area magnitude to allow for freezing to be predicted by the n_s derived from the suspension experiments.

To address this concern, we have reduced discussions on the coagulation effect and illustrate the point above more clearly. The aggregation is not merely presented as potential side effect of the suspension experiments. Even in aggregates if water molecules can access the active site, this should not preclude ice nucleation and the accessible surface area by water molecules. Please refer to our response to comment 9 for further changes.

Second: Many strong conclusions and atmospheric implications are made without strong, or any, evidence, these need to be walked back or better supported. See comments on lines 500+ below.

We have revised the abstract, conclusion, and atmospheric implications in response to these comments. Please see our responses to the comments regarding lines 500+ below.

Despite these issues, I think there is a lot of interesting analysis and work that has gone into this paper, and I think it will be publishable and of broad interest with the right framing and revised discussion, but extensive revisions are required.

We thank the reviewer for this positive evaluation and have presented a major revision to the manuscript based on the comments from both Reviewer 1 and Reviewer 2. Please refer to our responses below and the revised manuscript.

Specific comments:

1. Section 2.2: In this section it was unclear to me if you are using equations 1-6 and applying them to literature datasets, or simply explaining how these quantities might be calculated and using literature values that have already been calculated.

For this section we are doing both, showing how the literature values are determined and for older studies or studies that did not derive n_s, we derive them here using these equations. In the revised manuscript we make it clear where we have derived the n_s values and where the n_s values have been already calculated in the existing literature.

I think it would be much more clear to move this information to the supplemental and expand the discussion to be more explicit about which quantities were used from each dataset to calculate each derived quantity. Right now it feels like there is enough information to make a few guesses but not enough information to be sure what you have actually done. Perhaps a very brief explanation should stay in the main manuscript and point to the supplemental.

In this work, we compiled ice nucleation data from previous studies. The active site surface density (n_s) and nucleation rate (J_{het}) of dust particles are either directly reported or derived in this work based on published ice nucleation data and particle surface area. All n_s and J_{het} (either reported or derived) are calculated based on Equations 1-6. The specific equations used in the calculation depend on the measurement methods applied in ice nucleation experiments, i.e., dry dispersion in chamber experiments (Eq. (1) – Eq. (2) and Eq. (5)) versus wet suspensions in droplet freezing measurements (Eq. (3) – Eq. (4) and Eq. (6)).

The derivation of n_s and J_{het} is a fundamental step in our analysis and forms the basis for comparing results across different experimental methods and dust species. Therefore, we would like to keep this information in the main text. To address the reviewer's concern, we revised the main text and added more descriptions to make this point clearer, and added a table in the SI based on the Reviewer recommendation as follows:

Lines 169-176: "In the present work, the n_s are either taken from previous studies or derived based on reported ice nucleation results and particle surface area. While all n_s are derived based on the following equations (Eq. (1) – Eq. (4)), the specific equations used differ depending on the measurement methods, i.e., chamber experiments with dry-dispersed aerosol (Eq. (1) – Eq. (2)) and DFTs with wet-suspended particles (Eq. (3) – Eq. (4)). Table S3 in the supporting information (SI) reports for which sources we calculated n_s based on the particle surface reported in the source of the study and for which ones the n_s was directly reported."

Lines 191-197: "To account for the stochastic nature (time dependence) of ice nucleation, the **time dependent metrics**—heterogeneous ice nucleation rate (J_{het}, m^{-2} s^{-1}), can be calculated based on following equations (Eq. (5) and Eq. (6)). J_{het} describes the number of nucleation events per unit particle surface and unit time. Similar to n_s, J_{het} values are either taken from previous studies or derived from reported ice nucleation results and residence time of aerosol particles or droplets for aerosol (Eq. (5)) and suspension measurements (Eq. (6)), respectively. Table S3 indicates for which study J_{het} was calculated and for which ones the J_{het} was directly reported."

Furthermore, we have added Table S3 to the SI to say which studies the n_s and J_{het} are derived by us and from which studies we mined the data directly, as also shown below.

Table S3. Summary of ice nucleation metrics derived for each study, indicating whether values were calculated in this work, extracted directly from literature, or not available.

References	Ice active site surface density (n_s)	Source (derived or reported from literature)	Ice nucleation rates (J_{het})	Source (derived or reported from literature)
Salam et al. (2007)	Yes	Calculated	Yes	Calculated
Sullivan et al. (2010a)	Yes	Calculated	Yes	Calculated
Sullivan et al. (2010b)	Yes	Calculated	Yes	Calculated
Murray et al. (2011)	Yes	From the literature	Yes	From the literature
Niedermeier et al. (2011)	Yes	Calculated	Yes	Calculated
Tobo et al. (2012)	Yes	Calculated	Yes	Calculated
Kanji et al. (2013a)	Yes	From the literature	Yes	Calculated
Atkinson et al. (2013)	Yes	From the literature	No	
Wex et al. (2014)	Yes	From the literature	Yes	From the literature
Augustin-Bauditz et al. (2014)	Yes	Calculated	Yes	Calculated
Sihvonen et al. (2014)	No	-	No	-
Kulkarni et al. (2014)	Yes	Calculated	Yes	Calculated
Kulkarni et al. (2015)	Yes	Calculated	Yes	Calculated
Emersic et al. (2015)	Yes	From the literature	No	
Hiranuma et al. (2015)	Yes	From the literature	Yes	From the literature
Zolles et al. (2015)	Yes	From the literature	No	
Peckhaus et al. (2016)	Yes	From the literature	Yes	Calculated
Harrison et al. (2016)	Yes	From the literature	Yes	Calculated
Boose et al. (2016)	Yes	From the literature	Yes	Calculated
Demott et al. (2018)	Yes	From the literature	No	
Perkins et al. (2019)	Yes	Calculated	No	
Harrison et al. (2019)	Yes	From the literature	Yes	Calculated
Kanji et al. (2019)	Yes	From the literature	Yes	Calculated
Fahy et al. (2022)	Yes	From the literature	Yes	Calculated
Chen et al. (2023)	Yes	From the literature	Yes	Calculated

2. Line 232 or thereabout: There is a lot of discussion of feldspar ice nucleation, and the variability in mineral dust ice nucleation properties, and I feel that inclusion of Whale 2017 would greatly benefit this section. In that work, for feldspar samples measured using the same technique, there are >5 orders of magnitude difference in ns, in a similar manner to the combined data within your manuscript. This certainly seems relevant for understanding the origin of the variability and should be discussed.

We agree with the Reviewer, it is a good idea to include Whale et al. (2017). We added this citation in Lines 258-263 as follows: "Whale et al. (2017) examined different types of feldspars and showed that the exceptional INA of alkali feldspars arises from specific topographical features: the perthitic microtexture resulting from phase separation into Na- and K- rich regions. This would explain the variability in ice nucleation across the feldspar group, with the Na/K feldspars showing superior INA than other feldspars which lack such microtextural features."

3. Line 253: I don't think it is fair to say "1-8 orders of magnitude higher" when some of the aerosol and suspension points overlap, maybe 0-8?

We agree with the Reviewer and changed it to "aerosol n_s of K-feldspar is up to 2 orders of magnitude higher than suspension n_s at T =-20 °C", now in Lines 268-269.

4. Line 270: similar to above, maybe "0-6 orders"… or "up to 6 orders" might sound better. Also shows up line 428 and 565.

We changed the statements according to Reviewer 1's comments:

Lines 303-Line 305: "The median values of aerosol n_s are up to 6 orders of magnitude higher than suspension n_s at -38 < T < -18 °C".

Lines 494-496: "For example, the difference between n_s from dry-dispersion and wet-suspension can reach up to 6 orders of magnitude in the same temperature range (Figure 3 and Figure S3)."

Lines 656-658: "Dry-dispersed particles generally demonstrate higher n_s than those produced from wet suspensions, with discrepancies of up to 6 orders of magnitude at -38 < T < -18 °C."

5. Line 276: My understanding based on the equations included, is that the way you are estimating Jhet will shift ns values in an identical way based on experimental cooling rate or residence time, is that correct?

Yes, your understanding is entirely correct. Both n_s and J_{het} are metrics used to quantify the ice nucleation ability (INA) of dust particles. n_s is based on the singular description of ice nucleation, which assumes that ice nucleation is only temperature-dependent and time-independent.

Such that any relative change between the relative ordering of experiments in ns and jhet is due to cooling rate changes? Some discussion of this would be helpful I think. I also think that more

explicit discussion (related to my comments on section 2.2) surrounding what assumptions go into this estimation of jhet and their validity would be helpful.

J_{het} was obtained by scaling n_s with the residence time of an aerosol particle exposed to ice formation conditions (Eq. (5)). For DFT measurements with wet-suspended particles, J_{het} represents the differential nucleation rate over a specific time interval (Δt), which can be calculated based on the reported cooling rate in each freezing experiment (Eq. (6)). Compared to n_s, J_{het} accounts for time dependence of an ice formation event. Therefore, if the residence time and cooling rate are the same, the relative ordering of J_{het} values should remain identical to that in n_s.

We had few statements in section 2.2 to reflect the singular and stochastic nature of ice formation, for example "n_s is based on the singular description of ice nucleation, which assumes a strong temperature dependence and time is neglected because of the secondary importance of time compared to temperature for dust particles" in Line 158-162 and "To account for the stochastic nature (time dependence) of ice nucleation, the time dependent metrics—heterogeneous ice nucleation rate (J_{het}, m^{-2} s^{-1}), can be calculated based on following equations (Eq. (5) and Eq. (6))" in Line 191-193.

Following Reviewer 1's suggestions, we have revised or added descriptions in section 2.2 about the temperature-dependent and stochastic nature of ice nucleation and explicitly state the assumptions underlying the calculations of J_{het} and n_s:

Lines 158-163: "n_s indicates the number of ice nucleation events occurring per unit of particle surface. n_s is based on the singular description of ice nucleation, which assumes a strong temperature dependence and time is neglected because of the secondary importance of time compared to temperature for dust particles (Welti et al., 2012; Kanji et al., 2013b; Budke and Koop, 2015; Wright and Petters, 2013; Vali and Snider, 2015).".

Lines 170-174: "In the present work, the n_s are either taken from previous studies or derived based on reported ice nucleation results and particle surface area. While all n_s are derived based on the following equations (Eq. (1) – Eq. (4)), the specific equations used differ depending on the measurement methods, i.e., chamber experiments with dry-dispersed aerosol (Eq. (1) – Eq. (2)) and DFTs with wet-suspended particles (Eq. (3) – Eq. (4)).

Lines 191-197: "To account for the stochastic nature (time dependence) of ice nucleation, the **time dependent metrics**—heterogeneous ice nucleation rate (J_{het}, m^{-2} s^{-1}), can be calculated based on following equations (Eq. (5) and Eq. (6)). J_{het} describes the number of nucleation events per unit particle surface and unit time. Similar to n_s, J_{het} values are either taken from previous studies or derived from reported ice nucleation results and residence time of aerosol particles or droplets for aerosol (Eq. (5)) and suspension measurements (Eq. (6)), respectively. Table S3 indicates for which study J_{het} was calculated and for which ones the J_{het} was directly reported".

Lines 206-213: "We note that the practice of using cooling rate or chamber residence times can underestimate J_{het}, because the real time of nucleation could be less than the residence time or the time interval at a given temperature (cooling rate). Since the true ice nucleation time is not

possible to observe as we only detect freezing once a crystal has nucleated and grown to detectable sizes, the nucleation rates presented are a lower limit. We further note that compared to n_s, J_{het} scales the INA of dust particles by the time of an ice formation event. Therefore, if different datasets are measured with identical cooling rates or residence times, the relative ordering of their J_{het} values will remain consistent with that of n_s."

6. Line 283: The "two step temperature dependence" is weakest in natural dust samples, and doesn't appear for the aerosol samples. I'm not convinced that this statement is well-supported by the data presented thus far.

We agree with the reviewer when considering very few individual dust samples in the suspension category, but the majority of the suspension data shows a week temperature dependence above -20 C. As such our discussion about the two-step temperature dependence of J_{het} being less pronounced is specifically for the datasets encompassing all dust species and specifically for suspension measurements, as shown in Figure 3. We did not discuss this temperature dependence for individual dust species. The parameterizations developed from temperature dependent datasets are intended to represent the average ice nucleation behavior of dust types with mixed mineral compositions, as noted in the main text Line 462-465: ", the parameterizations developed from the dataset of all dust types (Figure 3) represent the average freezing behavior of dust particles with mixed mineral compositions, which are suitable for predicting INPs in an atmosphere with compositionally diverse dust or for global-average applications ".

We also emphasize that this two-step dependence is primarily validated by the suspension samples rather than aerosol samples. As indicated in Lines 312-313, "Similar to n_s, aerosol J_{het} values are systematically higher than suspension J_{het} values (Figure 3b)". For individual dust species, only a one-step temperature dependence is observed, as shown in Figure 2.

7. Line 310: referencing eq. 1 and 3 here seems worse than just redefining the terms – Sp is the surface area of a single aerosol and Sd is the total surface area of all suspended aerosol in a droplet.

We revised the text to "The diameter of the spherical equivalent particle is calculated based on $D_e = \sqrt{\frac{S_p \text{ or } S_d}{\pi}}$, where the S_p is the surface area of an individual aerosol particle in aerosol measurement, and S_d is the total surface area of suspended particles in a droplet in suspension measurement.", now in Lines 345-348. We also give more information about the D_e method "The D_e method treats the combined surface area of multiple particles as that of a single INP. The underlying assumption is that a multi-particle droplet (those measured by DFT) can be conceptually dispersed into many smaller, single-particle droplets. Given that the freezing of a droplet is triggered by the most active ice-nucleating site on particle surface and the probability of encountering such an active site scales with the total particle surface area, the D_e method normalizes a muti-particle droplet into an equivalent single INP that shares the same ice-nucleating probability. This approach enables a direct comparison among different ice nucleation measurements" in Line 348-355.

8. Line 312: Why are you choosing different definitions for "freezing temperature" for suspension and aerosol measurements? That needs to be justified.

We thank the reviewer for raising this important point. The selected freezing temperature must represent the ice formation behavior of the majority of the droplets or aerosol particle population. For suspension measurements, the median freezing temperature where 50% of the total droplets are frozen (frozen fraction = 50%) is a widely used and robust metric that effectively captures the average freezing temperature of the droplet population. In contrast, aerosol measurements evaluate the ice formation of individual particles and quantify the activated fraction (AF) of INPs in the total aerosol populations. Not all samples can reach an AF of 50% due to the inherent rarity of single INPs (i.e. the samples are not aggregated) in the aerosol population, the poor ice nucleation abilities of some aerosol species, and instrument limitations (e.g., small sampling air volume). Therefore, an AF of 10% is widely used in aerosol ice nucleation studies as a statistically significant threshold that represents an ice active state of the aerosol population.

We added the following statements in Lines 359-367 to make it clearer:
"The freezing temperatures at FF = 50% and AF = 10% are the representative freezing thresholds for suspension and aerosol measurements, respectively. While a 50% FF captures the median freezing behavior of a droplet population where aerosol particles are aggregated in individual droplets, achieving a 50% AF in aerosol measurements is often impractical due to the measurement on single sub-micron aerosol particles in droplets and therefore the inherent rarity of INPs. The lower INA of some aerosol species in a single droplet leads to lower detected INA. Consequently, a 10% AF is adopted as a widely reported, statistically significant threshold for evaluating the ice-active state of aerosols in chamber experiments."

9. Lines 315-330: So you have constructed an equivalent diameter for all particles in solution drops and compared to actual aerosol diameters, after briefly discussing coagulation. I don't understand quite how you think these comparisons are meaningful, however. It certainly doesn't seem to follow that droplet freezing measurements are only relevant to >10 micron single particles within the atmosphere, when you have only constructed a "what if everything coagulates" product with no evidence that coagulation is actually occurring in these measurements… Related to this, droplet freezing measurements often use serial dilutions to access different INP concentration ranges, with the most dilute samples differing by >1000 fold. In most cases there is overlap in INP concentration between dilutions, which would not be the case if the degree of aggregation were changing with concentrations. Some discussion of this is relevant and warranted in this section.

The 1000-fold dilution series is only possible when droplet freezing assays have small enough droplets where the pure water samples freeze at T < -35 °C. Diluting by too much can yield freezing in the background of the instrument. The purpose of constructing the equivalent diameter of particles (D_e) in aerosol and suspension measurements is to highlight the high particle concentrations used in some of the suspension measurements is what yields higher freezing temperatures. These high particle concentrations (large total particle surface) are commonly used to enhance the freezing signal above water background (i.e., detection limits).

To avoid any confusion, we revised these statements to clarify this point more neutrally in Lines 370-379: "For example, suspension D_e values (≈ 10–7000 μm) are 1–3 orders of magnitude larger than those from aerosol measurements at T = -25 °C. Particles with different mineral compositions indicate a similar trend (Figure 4b), with larger suspension D_e values. This explains why suspension droplet experiments freeze at warmer temperatures than droplets activated from individual aerosol particles (Figure 4), as a greater particle surface area increases the probability of ice formation. At high particle concentrations in suspension measurements, coagulation may occur which cannot be fully verified in this study. Should it occur, coagulation could reduce the effective surface area available for ice nucleation, but would only be relevant if water were unable to access the aggregated regions of the dust particles."

We agree that we did not provide direct observational evidence for particle coagulation in this work and it may not be relevant in every suspension measurement. Our dataset also includes suspension measurements with small D_e values (≈ 1 μm) at -38 < T < -22 °C, which are comparable to aerosol measurements (Figure 4). These D_e data obtained from picoliter-size droplets with lower particle concentrations (D_e≈ 1 μm), which indirectly suggest that particle coagulation is not expected in all suspension experiments, but may occur under conditions of high particle concentrations. This observation is consistent with the idea that many DFT measurements employ serial dilution to probe suspensions with different particle concentration ranges and overlapping INP concentrations are often observed across the dilution series.

To address reviewer's concern about the observational evidence and dilution series, we added the following statements:
Lines 392-396: "In this work, it is not possible to provide direct observational evidence for particle coagulation or its impact on INA of dust particles. Nevertheless, we noted that there are few suspension measurements with D_e (≈ 1 μm) at -38 < T < -22 °C that also occur for aerosol measurements (Figure 4), suggesting that agreement between the two methods is observed at sufficiently low particle suspension concentrations."

Lines 403-407: "Many DFT measurements employ serial dilution to probe suspensions with different INP concentration ranges and overlapping INP concentrations are often observed across the dilution series. These dilution results combined with our D_e data (Figure 4), suggest that particle coagulation is not expected in all suspension experiments, but could not be excluded as a possibility under conditions of high particle concentrations.".

We have also tempered the discussion of the coagulation effect and its role in the observed methodological difference throughout the text in response to Reviewer 2's comment (Comment 4):

We deleted lines (Lines 355-364 in original manuscript): "However, such coagulation-induced effects are primarily artifacts of the high particle concentrations used in laboratory suspensions and are unlikely to occur under typical atmospheric conditions, where individual cloud droplets generally contain only a single dust particle. As a result, n_s values obtained from aerosol measurements with individual particles are more representative of dust particles, which remain largely unaggregated and have minimal interactions with water. While suspension n_s, especially that corresponds to high particle concentrations, are unlikely to be relevant for dust

particle freezing in the atmosphere regarding coagulation, they may reflect the INA of particles subjected to water-aging processes, which will be discussed in section 3.3. If ice formation parameterizations are developed based on suspension n_s, they will likely underestimate the INA of dry and fresh dust aerosol particles."

We deleted the statements (Lines 401 to 403 in original manuscript): "As mentioned above, suspension-derived n_s may involve measurements using unrealistically high particle concentrations that promote artificial coagulation, which is unlikely to occur in real-world cloud droplets containing only one particle.".

We deleted lines (Lines 543-549 in the original manuscript): " It should also be noted that water aging only reduce dust n_s by 2 orders of magnitude at -26 < T < -2 °C, which is smaller than the n_s discrepancy observed between aerosol and suspension measurements within the same temperature range (up to 6 orders of magnitude at -26< T < -20 °C). This suggests that water aging alone cannot fully explain the n_s discrepancy caused by measurement method, the artificial coagulation effect (section 3.2) may be an important contributing factor".

We deleted lines (Lines 602-605 in the original manuscript): "However, the reduction in n_s caused by water/aqueous aging alone cannot account for the discrepancy in n_s observed for different measurement methods (up to 6 orders of magnitude at -26 < T < -20 °C), indicating that the artificial coagulation effect has a greater effect on the determined dust INA."

We moderated the statements in the abstract: from "This discrepancy likely arises from different water-particle interactions between the two methods, including particle coagulation at artificially high particle concentration and surface modification by water" to "This sample preparation difference likely arises from variations in water-particle interactions time scales that are longer in the suspension than the aerosol measurements.".

We modified Lines 325-327 in the original manuscript "However, such elevated particle concentrations in suspension measurements are also expected to enhance particle coagulation, which reduces the effective surface area available for ice nucleation and thereby decreases the freezing efficiency of suspension droplets" to Lines 376-379 in the revised manuscript: "At high particle concentrations in suspension measurements, coagulation may occur which cannot be fully verified in this study. Should it occur, coagulation could reduce the effective surface area available for ice nucleation, but would only be relevant if water were unable to access the aggregated regions of the dust particles.".

We modified Lines 340-348 in the original manuscript: "We also noted that there are few suspension D_e (≈ 1 μm) at 38 < T < -22 °C similar to that of aerosol measurements (Figure 4). These data correspond to suspension measurements with picoliter-size droplets, rather than from microliter-droplet measurements at warmer temperatures and therefore result in dust equivalent particle sizes (≈ 1 μm) that are more atmospherically relevant. Simulations by Emersic et al. (2015) suggest that picoliter-size droplets are unlikely to contain multiple particles to result in significant coagulation effects compared to microliter-sized droplets. Consequently, the freezing behavior of picoliter-size droplets is more comparable to droplets activated from single particles in aerosol measurements." to Lines 392-407 in the revised manuscript: "In this work, it is not possible to provide direct observational evidence for particle

coagulation or its impact on INA of dust particles. Nevertheless, we noted that there are few suspension measurements with D_e (≈ 1 μm) at -38 < T < -22 °C that also occur for aerosol measurements (Figure 4), suggesting that agreement between the two methods is observed at sufficiently low particle suspension concentrations. These data correspond to suspension measurements with picoliter-size droplets, (compared to microliter-droplet measurements) at warmer temperatures and therefore result in dust equivalent particle sizes (≈ 1 μm) that are more atmospherically relevant. Simulations by Emersic et al. (2015) suggest that picoliter-size droplets are unlikely to contain multiple particles, reducing the likelihood of significant coagulation effects compared to microliter-sized droplets. Consequently, the freezing behavior of picoliter-size droplets is more comparable to droplets activated from single particles in aerosol measurements. Many DFT measurements employ serial dilution to probe suspensions with different INP concentration ranges and overlapping INP concentrations are often observed across the dilution series. These dilution results combined with our D_e data (Figure 4), suggest that particle coagulation is not expected in all suspension experiments, but could not be excluded as a possibility under conditions of high particle concentrations.".

We modified Lines 353-354 in the original manuscript: "These findings suggest that coagulation effect may partly explain the different n_s values obtained between two methods, especially that observed for relatively warm temperatures" to Lines 412-415 in the revised manuscript: "These findings suggest that coagulation effect may be one possible factor contributing to the different n_s values obtained between two methods, especially that observed for relatively warm temperatures".

Lines 567-571 in the original manuscript: "Compared with dry-dispersed particles, suspension droplets with artificially high particle concentration undergo stronger particle coagulation, which reduces the effective surface area available for ice nucleation. In addition, particles experience longer water-contact time in suspension measurements, which may modify their surfaces and thereby reducing their INA" to Lines 660-664 in the revised manuscript: "Compared with dry-dispersed aerosol measurements, suspension experiments, particularly under conditions of higher particle concentrations may lead to particle coagulation within droplets, which can reduce the effective surface area available for ice nucleation. However, this should not influence the results of freezing if the water molecules can access the aggregated surfaces of the particles in the droplet.".

10. Lines 367: The transition to parameterizations seems abrupt, especially immediately following a teaser for discussing relevant "|water-ageing" processes. Maybe this belongs elsewhere in the manuscript, or perhaps it just needs to be tied together better.

The placement of the parameterizations in this section was intentional, as our logical flow is to first introduce the roles of chemical composition (section 3.1) and measurement methods (this section), and then build the ice formation parameterizations upon these variables.

To improve the flow, we have revised the transitional paragraph to "The findings presented in section 3.1 and the current section highlight that both chemical composition and measurement methods are important factors causing the observed variability in the INA of dust particles. Building upon these findings, we develop specific parameterizations in the following sections

to systematically represent the INA of dust particles. We fitted the n_s and J_{het} temperature dependent parameterizations using a log-linear form:" in Lines 427 to 431.

11. Figure 5: The aged and fresh data for aerosol/suspension look so similar, can you do significance testing to see if there are population level differences? Maybe a simple thing would be to see how well the best fit lines for fresh populations fit the aged data (calculating R^2 values). There are lots of fancier ways to check for significance between the two populations at a given temperature, or across all temperatures (which should probably be restricted to where you have data for both, i.e. >-27C for suspensions).

We thank Reviewer 1 for this insightful comment. Visually, fresh and aged data may appear similar because they are plotted on a log scale, and a large variability is observed for each dataset at individual temperature bins. To address the reviewer's concern, we performed statistical analysis to quantify the differences in the INA of fresh and aged samples for both aerosol and suspension measurements, respectively. Specifically, we conducted two types of significance analysis, including Analysis of Covariance (ANCOVA) to assess the overall difference in INA between fresh and age samples across temperatures and non-parametric Mann-Whitney U-test to assess the difference within individual temperature bin of 1 ℃.

To ensure a robust comparison, ANCOVA was strictly restricted to the shared overlapping temperature ranges where data for both fresh and aged samples exist (-42 < T < -20 ℃ for aerosol measurements and -28 < T < -2 ℃ for suspension measurements). The ANCOVA results yielded a highly significant difference (p<0.05) for both aerosol measurements and suspension measurements. This indicates that the aging process significantly alters the INA temperature dependence of dust particles.

The results of U-test within individual temperature bins (1 °C) are shown in the tables below for aerosol (Table S5) and suspension measurements (Table S6), respectively. As shown in the tables, a statistically significant difference (p<0.05) was obtained for 45% and 76% of temperature bins in the aerosol and suspension measurements, respectively. This indicates that while the INA of fresh and aged samples may show similarities in certain specific temperature bins, both ANCOVA across temperatures and individual temperature bin statistical analyses provide strong quantitative evidence that the aging process leads to a change in the INA of the dust particles.

We added the following statements in Lines 496-502: "To statistically evaluate the impact of aging on the INA of dust particles, significance analyses were performed for both aerosol (-41 < T < -20 ℃) and suspension measurements (-27 < T < -3 ℃). The results yielded statistically significant differences (p < 0.05) between fresh and aged samples in both measurement methods, indicating that the aging process alters the INA of dust particles. The details about the significance analyses are provided in Table S5 and S6 and Text S1.".

We also added the following descriptions and tables in Text S1: "We conducted two types of significance analysis, including Analysis of Covariance (ANCOVA) to assess the overall difference in INA between unaged and aged samples across temperatures and non-parametric Mann-Whitney U-test to assess the difference within individual temperature bin of 1 ℃.

To ensure a robust comparison, ANCOVA was strictly restricted to the shared overlapping temperature ranges where data for both unaged and aged samples exist (-42 < T < -20 °C for aerosol measurements and -28 < T < -2 °C for suspension measurements). The ANCOVA results yielded a highly significant difference (p<0.05) between aerosol and suspension measurements. This indicates that the aging process significantly alters the INA temperature dependence of dust particles.

The results of U-test within individual temperature bins (1 °C) are shown in the tables below for aerosol (Table S5) and suspension measurements (Table S6), respectively. As shown in the tables, a statistically significant difference (p<0.05) was obtained for 45% and 76% of temperature bins in the aerosol and suspension measurements, respectively. This indicates that while the INA of unaged and aged samples may show similarities in certain specific temperature bins, both ANCOVA across temperatures and individual temperature bin statistical analyses provide strong quantitative evidence that the aging process leads to a change in the INA of the dust particles. For temperature bins where aging did not lead to significant differences, the underlying reason may be that aging effects vary depending on the aging treatments (Figure 6) or that specific mineral species are insensitive to aging (Figure S5). A detailed discussion on how different aging treatments influence the INA of different mineral dust is provided in section 3.3."

Table S5. Results of non-parametric Mann-Whitney U-tests comparing the ice nucleation activity (INA) between unaged and aged dust samples in aerosol measurements.

Measurement	Temperature bins (°C)	P value	Significant
Aerosol	-42		Not enough data
Aerosol	-41		Not enough data
Aerosol	-40	>0.05	No
Aerosol	-39	>0.05	No
Aerosol	-38	<0.05	Yes
Aerosol	-37	<0.05	Yes
Aerosol	-36	<0.05	Yes
Aerosol	-35	>0.05	No
Aerosol	-34	>0.05	No
Aerosol	-33	>0.05	No
Aerosol	-32	<0.05	Yes
Aerosol	-31	>0.05	No
Aerosol	-30	<0.05	Yes
Aerosol	-29	<0.05	Yes
Aerosol	-28	>0.05	No
Aerosol	-27	>0.05	No
Aerosol	-26	>0.05	No
Aerosol	-25	<0.05	Yes
Aerosol	-24	<0.05	Yes
Aerosol	-23		Not enough data
Aerosol	-22	<0.05	Yes
Aerosol	-21	>0.05	No
Aerosol	-20	>0.05	No

Table S6. Results of non-parametric Mann-Whitney U-tests comparing the ice nucleation activity (INA) between unaged and aged dust samples in suspension measurements.

System	Temperature bins (°C)	P value	Significant
Suspension	-38		Not enough data
Suspension	-37		Not enough data
Suspension	-36		Not enough data
Suspension	-35		Not enough data
Suspension	-34		Not enough data
Suspension	-33		Not enough data
Suspension	-32		Not enough data
Suspension	-31		Not enough data
Suspension	-30		Not enough data
Suspension	-29		Not enough data
Suspension	-28		Not enough data
Suspension	-27	<0.05	Yes
Suspension	-26	>0.05	No
Suspension	-25	>0.05	No
Suspension	-24	<0.05	Yes
Suspension	-23	<0.05	Yes
Suspension	-22	<0.05	Yes
Suspension	-21	<0.05	Yes
Suspension	-20	>0.05	No
Suspension	-19	<0.05	Yes
Suspension	-18	<0.05	Yes
Suspension	-17	<0.05	Yes
Suspension	-16	<0.05	Yes
Suspension	-15	<0.05	Yes
Suspension	-14	<0.05	Yes
Suspension	-13	<0.05	Yes
Suspension	-12	<0.05	Yes
Suspension	-11	<0.05	Yes
Suspension	-10	<0.05	Yes
Suspension	-9	<0.05	Yes
Suspension	-8	<0.05	Yes
Suspension	-7	<0.05	Yes
Suspension	-6	<0.05	Yes
Suspension	-5	>0.05	No
Suspension	-4	>0.05	No
Suspension	-3	>0.05	No

12. Line 575: you aren't really accounting for the variability in composition and measurement methods, are you? Just fitting the aggregate data separately for the two techniques. You haven't synthesized them together in any way though…

We agree with Reviewer 1 that we did not synthesize the variability in the INA of dust particles caused by composition and measurement methods. To account for the variability in the composition, one would need to normalize to a reference composition and require that the dust

compositions for all studied be known. However, this is not possible but a recommendation for composition to be reported is now included in the manuscript. Instead, we developed composition-specific and measurement methods-specific ice formation parameterizations. Therefore, we have revised our text (Line 575 in original manuscript) from "By accounting for the variability in dust INA caused by mineral composition and measurement methods" to: "To capture the variability in the dust INA caused by mineral composition and measurement methods", Lines 670-671. We also added the following statements in the Atmospheric implications section to encourage the report of mineralogical composition of dust particles, Lines 758-764: "In addition, given that mineralogical compositions strongly influence the INA of dust particles and that compositional variability exists across particle sizes even within the same dust category, we recommend that future research present mineralogical analyses alongside ice nucleation measurements. If the composition of natural desert dust and its corresponding mass or surface area fractions are known, a compositionally weighted parameterization can be developed based on the existing composition-specific parameterizations to describe the INA of natural dust."

13. Line 616-621: Do you actually know that dry-dispersed measurements are more representative of freshly emitted aerosol? I don't think there is strong evidence in your analysis that this is the case, and I also don't think it is well-supported by your citations. I think more realistically, we still have two different techniques with discrepancies in what they measure but we don't know which is more representative of ambient concentrations or aerosol-cloud interactions.

We agree with the Reviewer about this point. In fact in discussing this point more internally – we have also considered that the dry dispersed may not be representative since the sample sourcing for the dry dispersed experiment varies a lot, even if it is dry dispersed, how the sample was produced, either sampled from the surface of a desert, commercially available sample, ground samples. Just the dry dispersion does not qualify it for atmospheric relevance. We have now removed an explicit recommendation of one or the other being more atmospherically relevant but instead discuss the reasons and cases where the different approaches are relevant.

To address the similar concern from Reviewer 2 (the comment 2 and 7) We have also revised our statements about different measurement methods throughout the main text and to avoid commenting which method is correct or more atmospherically as both measurement methods represent certain regime of experimental conditions.

We deleted the statement of (Lines 374 to 376 in original manuscript): "This aerosol n_s parameterization represents the INA of dry and near-source dust aerosol that have a negligible effect from water exposure:".

We deleted previous statements that suggested aerosol measurements are more representative of atmospheric dust (Lines 355 to 364 in original manuscript): "However, such coagulation-induced effects are primarily artifacts of the high particle concentrations used in laboratory suspensions and are unlikely to occur under typical atmospheric conditions, where individual cloud droplets generally contain only a single dust particle. As a result, n_s values obtained from aerosol measurements with individual particles are more representative of dust particles, which remain largely unaggregated and have minimal interactions with water. While suspension n_s,

especially that corresponds to high particle concentrations, are unlikely to be relevant for dust particles freezing in the atmosphere regarding coagulation, they may reflect the INA of particles subjected to water-aging processes, which will be discussed in section 3.3. If ice formation parameterizations are developed based on suspension n_s, they will likely underestimate the INA of dry and fresh dust aerosol particles.".

We also deleted the statements of (Lines 401 to 403 in original manuscript) "As mentioned above, suspension-derived n_s may involve measurements using unrealistically high particle concentrations that promote artificial coagulation, which is unlikely to occur in real-world cloud droplets containing only one particle.".

We also revised (Lines 617 to 621 in original manuscript) "Therefore, n_s parameterizations derived from dry dispersed aerosol are most representative of freshly emitted, dry dust near source regions, while those derived from suspension measurements are more relevant to particles that have undergone water-induced changes like being transported in cloudy conditions where the particles are subjected to CCN activated trajectories" to Lines 714-720 (in revised manuscript): "Because aerosol instruments typically have short water-particle contact times, they capture the INA of dust particles under conditions of rapid water activation followed by ice nucleation. Therefore, n_s parameterizations derived from dry-dispersed aerosol would be representative of dust particles undergoing limited water-mediated processing. In contrast, suspension methods evaluate the INA of particles with longer water interaction time, therefore n_s parameterizations derived from suspension measurements are representative of particles that have undergone water-induced changes. ".

We have revised the text (Lines 567-571 in the original manuscript): "Compared with dry-dispersed particles, suspension droplets with artificially high particle concentration undergo stronger particle coagulation, which reduces the effective surface area available for ice nucleation. In addition, particles experience longer water-contact time in suspension measurements, which may modify their surfaces and thereby reducing their INA." to: "Compared with dry-dispersed aerosol measurements, suspension experiments, particularly under conditions of higher particle concentrations may lead to particle coagulation within droplets, which can reduce the effective surface area available for ice nucleation. However, this should not influence the results of freezing if the water molecules can access the aggregated surfaces of the particles in the droplet. Moreover, particles experience different water-contact times in aerosol and suspension measurements, which may contribute to differences in their INA." in Lines 660-666 in the revised manuscript.

14. Line 626: I don't think you have shown that De based parameters are atmospherically relevant in any ways, and this discussion seems poorly supported…

The D_e parameter, as shown in Figure 4 and listed below, represents the total particle surface concentration in droplets and its influence on droplet freezing temperature. D_e values span a broad range, reflecting both atmospherically relevant particle concentrations (e.g., D_e<10 μm) and extremely high particle concentrations used in some suspension measurements (D_e >100 μm). While these large sizes are unlikely to occur in the atmosphere, they highlight the reason

why the suspension experiments freeze at much warmer temperatures. This exercise is merely to show that for atmospherically relevant cases freezing would be expected at much lower temperatures for droplets with smaller dust surface areas or smaller particles, unless really large particles or very large surface area equivalents are present.

Due to the observed linear relationship between D_e and droplet freezing temperatures, D_e-based parameterizations can serve as a simplified empirical tool for predicting droplet freezing temperature if the particle size is known. Compared to n_s parameterizations which are developed for different measurement methods (aerosol versus suspension), the implementation of D_e-based parameterizations is simpler and does not require detailed information regarding water-particle interactions (which would be needed for the n_s parameterizations to capture the different slope dependencies). Despite including extreme experimental values, the D_e-based parameterizations fully cover particle sizes relevant for atmospheric dust (typically D_e<10 μm), making it suitable for applications under atmospheric conditions.

To clarify how D_e was derived and the underlying assumptions, we added Line 344-355: "We calculated the spherical equivalent diameter (D_e) of dust particles contained in a droplet for both measurement methods. The diameter of the spherical equivalent particle is calculated based on $D_e = \sqrt{\frac{S_p \text{ or } S_d}{\pi}}$, where the S_p is the surface area of an individual aerosol particle in aerosol measurement, and S_d is the total surface area of suspended particles in a droplet in suspension measurement. The D_e method treats the combined surface area of multiple particles as that of a single INP. The underlying assumption is that a multi-particle droplet (those measured by DFT) can be conceptually dispersed into many smaller, single-particle droplets. Given that the freezing of a droplet is triggered by the most active ice-nucleating site on particle surface and the probability of encountering such an active site scales with the total particle surface area, the D_e method normalizes a muti-particle droplet into an equivalent single INP that shares the same ice-nucleating probability. This approach enables a direct comparison among different ice nucleation measurements".

The statements about extremely high and atmospherically relevant D_e values are in Lines 370-375: "For example, suspension D_e values (≈ 10–7000 μm) are 1–3 orders of magnitude larger than those from aerosol measurements at T = -25 °C. Particles with different mineral compositions indicate a similar trend (Figure 4b), with larger suspension D_e values. This explains why suspension droplet experiments freeze at warmer temperatures than droplets activated from individual aerosol particles (Figure 4), as a greater particle surface area increases the probability of ice formation." and in Lines 393-399: "Nevertheless, we noted that there are few suspension measurements with D_e (≈ 1 μm) at -38 < T < -22 °C that also occur for aerosol measurements (Figure 4) suggesting that agreement between the two methods is observed at sufficiently low particle suspension concentrations. These data correspond to suspension measurements with picoliter-size droplets, (compared to microliter-droplet measurements) at warmer temperatures and therefore result in dust equivalent particle sizes (≈ 1 μm) that are more atmospherically relevant."

The atmospheric implication of D_e-based parameterizations was stated in Line 720-727: "Applying aerosol and suspension n_s-based parameterizations to atmospheric conditions

requires additional information on water-particle interactions (e.g., particle concentration, solutes and interaction time) and further physicochemical characterizations of dust particles. D_e-based parameterizations provide a simplified empirical alternative. It links immersion freezing temperatures of dust particles directly to particle size even when details of particle-water interactions are unknown. This approach is independent of measurement methods and allows prediction of dust freezing without the need to consider complex aging trajectories in models".

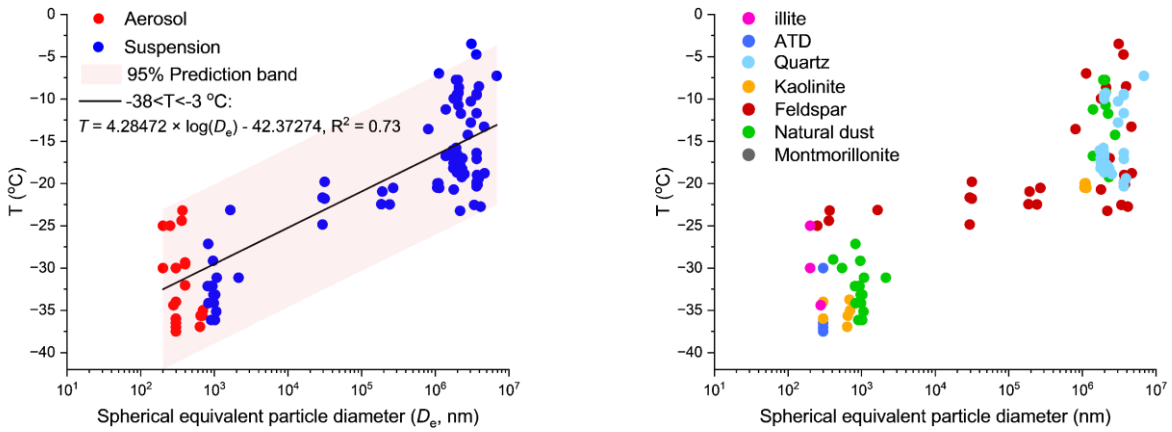

Figure 4. Spherical equivalent diameter (D_e) of dust particles in droplets derived from (a) aerosol (red) and suspension (blue) measurements, and (b) dust particles with different mineral compositions. The temperature shown in this figure represents the point at which 50% of droplets (FF=50%) are frozen in the suspension measurements, and at which 10% of particles (AF=10%) are ice activated in the aerosol measurements. D_e is calculated from studies in which S_p and S_d were provided or could be derived. The solid line is the fitting curve of freezing temperature versus log (D_e), which are in the form: $T = A \times \log(D_e) + B$ (values for parameter A and B are listed in this figure).

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