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 in the revised manuscript.

Reviewer 2:

- This manuscript compiles laboratory measurements of immersion freezing by mineral dust and seeks to derive revised parameterizations for dust ice nucleation under mixed-phase cloud conditions. The topic is important, and the effort involved in assembling a broad dataset is clear. The plots are interesting, and the compilation itself is potentially useful. The manuscript also raises a genuinely important issue, namely the difficulty of comparing measurements made using different experimental approaches.

However, I am afraid I am not persuaded by the central interpretation of the paper. In my view, the manuscript places far too much weight on the idea that there is a meaningful and general difference between 'aerosol' and 'suspension' methods, when the analysis presented does not convincingly isolate a method effect from the much more obvious and well-established effects of composition and material-specific behaviour. The paper's own compilation shows that dust ice nucleation activity varies enormously between mineral classes and within them, including among feldspars, quartz samples, ATD, and natural dusts. Yet these materials are then aggregated in ways that make the subsequent attribution to measurement method, in my view, untenable.

Indeed, I came away with rather a different impression from the one advanced by the authors. Looking at the figures, I am less struck by a clear demonstration of a fundamental dichotomy between aerosol and suspension approaches than by the extent to which the measurements are, in fact, as broadly consistent as they are, given the very large differences in mineralogy, sample origin, preparation, and experimental protocol represented in the compilation. The interesting conclusion here is not, to my mind, that one has established a robust and general method-dependent discrepancy, but rather that a wide and heterogeneous literature still shows a degree of coherence despite the fact that obviously unlike materials have been grouped together.

We appreciate the Reviewer 2's comments and would like to point out that we are not suggesting a method discrepancy. The methods used to analyze ice nucleation are robust for both the aerosol and the suspension methods. Instead, we are proposing that the suspension method results in the potential for water interactions with the mineral dust surface over longer time scales than the aerosol methods (few seconds water exposure before freezing). This interaction with water should not be considered inert as there is literature that shows mineral surfaces can undergo ion exchange and pH changes when suspended in water. As such we propose that the two methods are in their own right accurate, but this distinction may lead to measuring modified ice nucleation properties of dust. Indeed, some current work in our group measuring the ice nucleation abilities (INA) of two types of feldspars, montmorillonite and kaolinite shows that when generated from a wet suspension versus a dry dispersion using the same aerosol online ice nucleation technique, the INAs are indeed different across all 4 dust samples. We will

publish these results in a separate paper. We have modified the manuscript to make this point clearer, i.e. that there is not one method superior to the other, and that rather than a measurement discrepancy the results should consider how dust interactions with water can modify their INA (please refer to comment 2 and 3 below).

For that reason, I am not sure that this manuscript is revisable in any straightforward sense. To do this properly, it seems to me that one would need a rather different paper built around careful like-for-like comparison, explicit treatment of compositional heterogeneity, and a much more cautious interpretation of what can and cannot be inferred about method effects from compiled cross-study datasets. As it stands, I do not think the main conclusion is supported.

We acknowledge the Reviewer 2's concerns that chemical composition of dust particles is an important factor in determining the variability of INA of dust particles, which adds difficulty to comparing datasets crossed studies. To address this concern, we have added more quantitative descriptions of measurement methods for specific dust species (please refer to comment 1 below). We also revisited and modified our mineral categories for measurement comparison to better constrain the variability in INA of dust particles within the same mineralogical group (please refer to comment 1), enabling a comparison closer to "like-for-like comparison". For example, the results from K-feldspar and other feldspars have been separated, as well as three species belonging to clay particles. Differences between aerosol and suspension measurements are still evident within these refined mineral-specific categories. This suggests that despite constraining the heterogeneity in mineralogy, the water-particle interactions in wet and dry suspension techniques still lead to the observed differences.

We also acknowledge that there are limitations remaining in the binary classification of measurement methods. For example, compositional differences and experimental protocols within each mineralogical group may still introduce uncertainty in observed dust INA of aerosol and suspension measurements.

To address these concerns, we have rewritten the abstract and moderated our interpretation of method-dependent differences throughout the text and clearly highlighted these limitations (please refer to comments 1 and 3). We also revised the manuscript to avoid commenting on which measurement method is more correct or more atmospherically relevant, as both represent different experimental regimes (please refer to comment 2).

Major comments

1. The manuscript does not convincingly demonstrate a meaningful general difference between 'aerosol' and 'suspension' methods. The central claim of the paper is that measurement method introduces one of the largest sources of variability in dust ice nucleation activity, with dry-dispersed aerosol measurements yielding systematically higher ns than wet-suspension measurements, sometimes by several orders of magnitude. This claim appears in the abstract, is developed through Figs. 2–4, and is used to motivate the proposed method-specific parameterizations. I do not think the evidence presented is sufficient to support that conclusion.

The difficulty is that the comparison is heavily confounded by differences in the materials being compared. The manuscript itself repeatedly acknowledges that dust ice nucleation activity depends strongly on mineral composition and can vary substantially even within nominally the same mineral class. For example, the manuscript notes that K-feldspar-rich particles tend to be much more ice active than other feldspars; that non-potassium-rich feldspars can have much lower activity; that quartz samples vary; that ATD may be relatively active where K-feldspar is present; and that natural desert and volcanic dusts are diverse materials with differing nucleation behaviour.

We fully agree that variability in the INA of dust particles, can arise not only from measurement methods but also from other factors including compositional differences. To reduce the compositional effects, we have separated datasets by specific mineral types (illite, kaolinite, K-feldspar, non-potassium feldspar and ATD) in Figures 2 and S2. As shown in Figure S2 and listed below, differences in aerosol (red) and suspension (blue) n_s are observed for specific dust species. For example, the median n_s values between aerosol and suspension methods differ by 0–3 orders of magnitude at -31 < T < -18 ℃ for illite, by 3–5 orders of magnitude at -27 < T < -21 ℃ for ATD and by 0–1 orders of magnitude at -35 < T < -30 ℃ for kaolinite. The aerosol n_s of quartz, non-potassium feldspars and montmorillonite are not shown in Figure S2 due to the absence of or inadequate data from aerosol measurements in each temperature bin.

This issue is especially clear in the treatment of the different dust classes shown in Fig. 2. The manuscript aggregates feldspars, clays, ATDs, and natural dusts in ways that I do not think are well justified physically. For feldspars, the manuscript explicitly states that not all feldspars are equal, and that K-feldspar and non-potassium-rich feldspars can differ by orders of magnitude in ns. It therefore seems unreasonable to collect all feldspar data in the manner done in Fig. 2 and then interpret the resulting contrast as evidence for a difference between techniques. The feldspars used in the various studies may simply not be comparable substances. Even amongst relatively active k-feldspars there is plenty of evidence for a widespread in activity (Whale et al., 2017; Canet et al., 2025). If the authors think there is good reason to treat them as a common category for this purpose, that case needs to be made explicitly and convincingly. At present it is not.

For feldspars, we have now separated K-feldspar and non-potassium rich feldspars and fitted parameterizations for the two datasets respectively, as shown in Figure 2 (the new figure is shown below). For K-feldspar, a method-dependent discrepancy remains evident. For non-potassium rich feldspars, comparison between suspension and aerosol measurements cannot be made due to the absence of aerosol n_s datasets. The large variability observed in n_s within non-potassium rich feldspars is attributed to the compositional difference among this dust group, for example, albite (Na-end member) exhibits the highest n_s compared to anorthite (the calcium-end member).

For clays, I do not find it obvious why illite, kaolinite, and montmorillonite should be aggregated in the way shown in Fig. 2. These are distinct minerals, and the manuscript provides no clear physical basis for expecting them to share a common ns behaviour such that method-dependent offsets can be meaningfully interpreted once pooled

For clay particles, we now separated the three clay species (illite, kaolinite and montmorillonite) into individual plots, as shown in Figure 2 and below. Nevertheless, differences in aerosol (hollow circles) and suspension (solid circles) n_s are still observed for all three species.

For ATD, the paper treats Arizona Test Dust as though it were effectively one material class. However, different ATD samples are well known to have different compositions e.g (Ehlers et al., 2026) and therefore could quite plausibly differ meaningfully in their ability to nucleate ice. I therefore do not think it makes sense to compare the ATD datasets in Fig. 2 as though they were directly interchangeable across techniques. Again, this is not like-with-like.

We agree that it would not make sense to compare ATD datasets as the reviewer states, especially since it is known that the composition of ATD changes with particle size (Vlasenko et al., 2005). However, we reviewed methodological details of the 10 collected ATD publications: 6 studies explicitly reported the INA of ATD A1 (ultrafine) (Sullivan et al., 2010a; Sullivan et al., 2010b; Niedermeier et al., 2011; Augustin-Bauditz et al., 2014; Kulkarni et al., 2014; Kulkarni et al., 2015), 2 used a nominal size cut (Chen et al., 2023; Kanji et al., 2013), 1 used ATD A2 (fine) (Perkins et al., 2019), and 1 did not specify the exact grade (Zolles et al., 2015). Since 60% of the dataset derives from the exact same standardized ATD grade (A1), we believe the general trend of ATD INA is representative. Moreover, given that we are trying to update a mineral dust parameterization that is as generalizable as possible – we think that comparing ATD in this way is not necessarily unmeaningful to come up with a parameterization that can be used in global climate models, that are not equipped to take mineralogical wise parametrizations to predict ice nucleation for dust in the cloud/climate models. We have explicitly added the specific ATD grades utilized in each study to Table S2 for the readers' clarity.

For natural dust, the problem is even more obvious. The natural dusts included here are clearly different substances from different desert and volcanic sources. It is not clear why one would expect them to nucleate ice similarly in the first place, let alone why they should be pooled in a way that supports a general conclusion about measurement method.

For natural dust, both desert dust (Boose et al., 2016; Kanji et al., 2019) and volcanic dust (Fahy et al., 2022) are included because they are most globally significant natural dust species. These two dust species are merged into the same group, as they share similarities in both mineralogical composition and INA, but differ from soil dust, another major natural dust type that contains a large fraction of biological compounds. As noted by Fahy et al. (2022): "Several primary minerals have been implicated as the ice-active phases in volcanic ash in previous studies, including alkali (Na/K-rich) feldspar, plagioclase (Na/Ca-rich) feldspar, and pyroxene. Similarly, K-feldspar (hereafter used to refer to K-rich alkali feldspar), Na/Ca-feldspar, and quartz are recognized as ice-active phases in mineral dust". The INA of volcanic ash is influenced by its mineralogical composition (Zolles et al., 2015), such as its feldspar content, which also aligns with desert dust. It is therefore valuable to include the n_s data from these two dust species to develop a generalized parameterization (Figure 3). We fully agree with the Reviewer that these two datasets should not be pooled together to support the conclusions about measurement methods, as the compositional difference between these two dust types cannot be overlooked. Consequently, these results have been removed from the discussion of

measurement methods and from Figure 2 and Figure S2. We also separated desert dust and volcanic dust when discussed the impact of aging treatments on INA of natural dust, as shown in Figure S5.

To address Reviewer's comment, we added more descriptions of differences between measurement methods for specific dust species and highlighted the uncertainty arising from compositional difference within each mineralogical group at Line 287-300: "Specifically, the median n_s values between aerosol and suspension methods differ by up to three orders of magnitude at -31 < T < -18 °C for illite, by 3–5 orders of magnitude at -27 < T < -21 °C for ATD and up to one order of magnitude at -35 < T < -30 °C for kaolinite. K-feldspar and kaolinite n_s show the closest agreement between the two methods, with differences generally within an order of magnitude. This comparison cannot be made for quartz and other non-potassium-rich feldspars because of the lack of aerosol n_s data for these dust species. We also acknowledge that these method-based comparisons within individual dust groups are still subject to uncertainties arising from compositional differences within individual dust categories. For example, the composition of ATD is known to vary with particle size (Vlasenko et al., 2005), which may influence its INA. Within the non–potassium-rich feldspar group, albite (Na-end member) exhibits a higher n_s compared to anorthite (the calcium-end member) (Harrison et al., 2016). Nevertheless, the consistent differences observed across multiple groups suggest that measurement method contributing to the variability in reported dust INA cannot be excluded but this is likely convoluted by changes in composition."

We have also rewritten the abstract to reduce emphasis on methodological differences:

"Dust aerosol plays a key role in cloud formation and evolution due to its high atmospheric abundance and efficient ice nucleation ability (INA). A generalized parameterization of dust-induced ice formation in climate models remains challenging, because dust INA varies substantially with mineral composition, aerosol generation methods, and atmospheric aging processes. In this study, we revisited the INA of dust particles under mixed-phase cloud conditions (MPC, -38 < T < 0 °C) compiled from previous laboratory studies. Our results indicate that INA of dust particles with different mineral compositions exhibit variability spanning up to six orders of magnitude in INA at -38 < T < -2 °C. When accounting for mineralogical differences, aerosol generation methods whether particles are dry-dispersed or wet-suspended also contribute to the reported variability. This sample preparation difference likely arises from variations in water-particle interactions time scales that are longer in the suspension than the aerosol measurements. Aging generally reduces dust INA, with chemical reactions inducing the strongest reduction, followed by thermal treatments and water/aqueous aging. Based on these findings, we developed a suite of n_s −based parameterizations to represent INA of dust particles with mixed and specific mineral composition. We also proposed parameterizations based on D_e (spherical equivalent particle diameter within a droplet), which predict droplet freezing across the full MPC temperature range using a single expression. These empirical parameterizations capture the general INA trends of dust particles and are expected to improve the accuracy of predictions of dust-induced cloud formation in climate models."

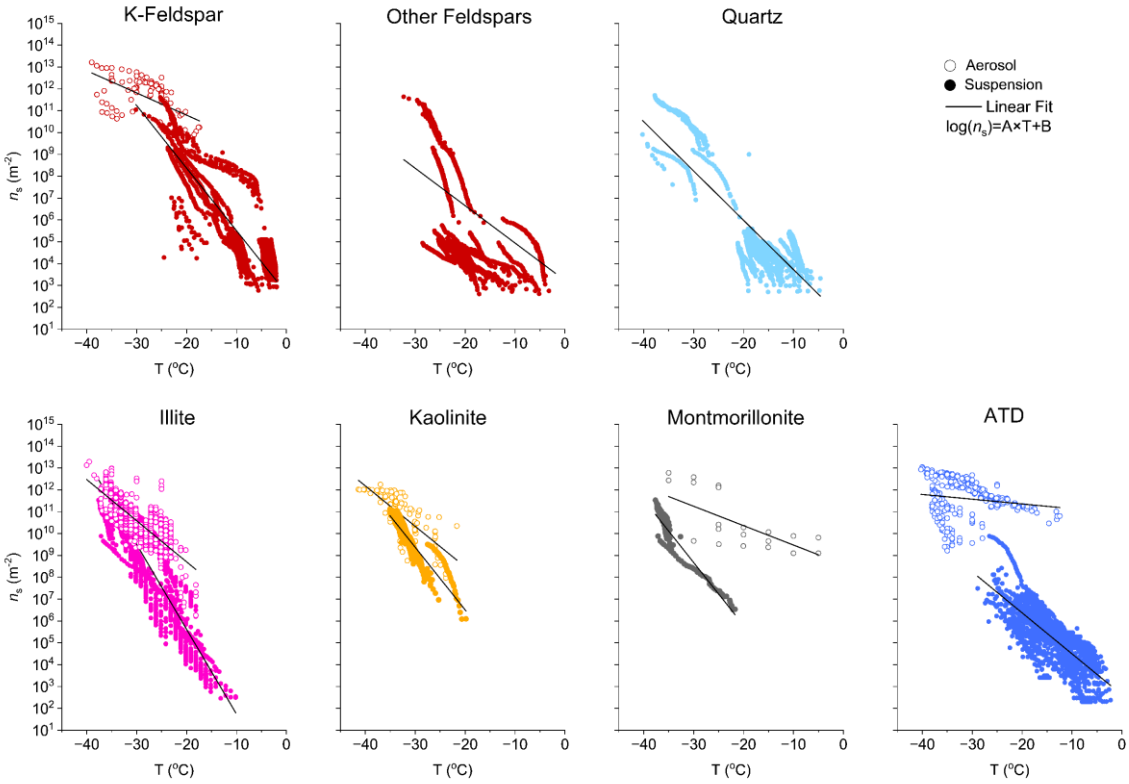


Figure 2. n_s of dust particles with specific mineral compositions. Empty and filled data point denote measurements using dry-dispersed aerosol and wet suspended particles, respectively. K-feldspar and non-potassium-rich feldspars are shown separately due to their distinct INA. For quartz and non-potassium-rich 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.

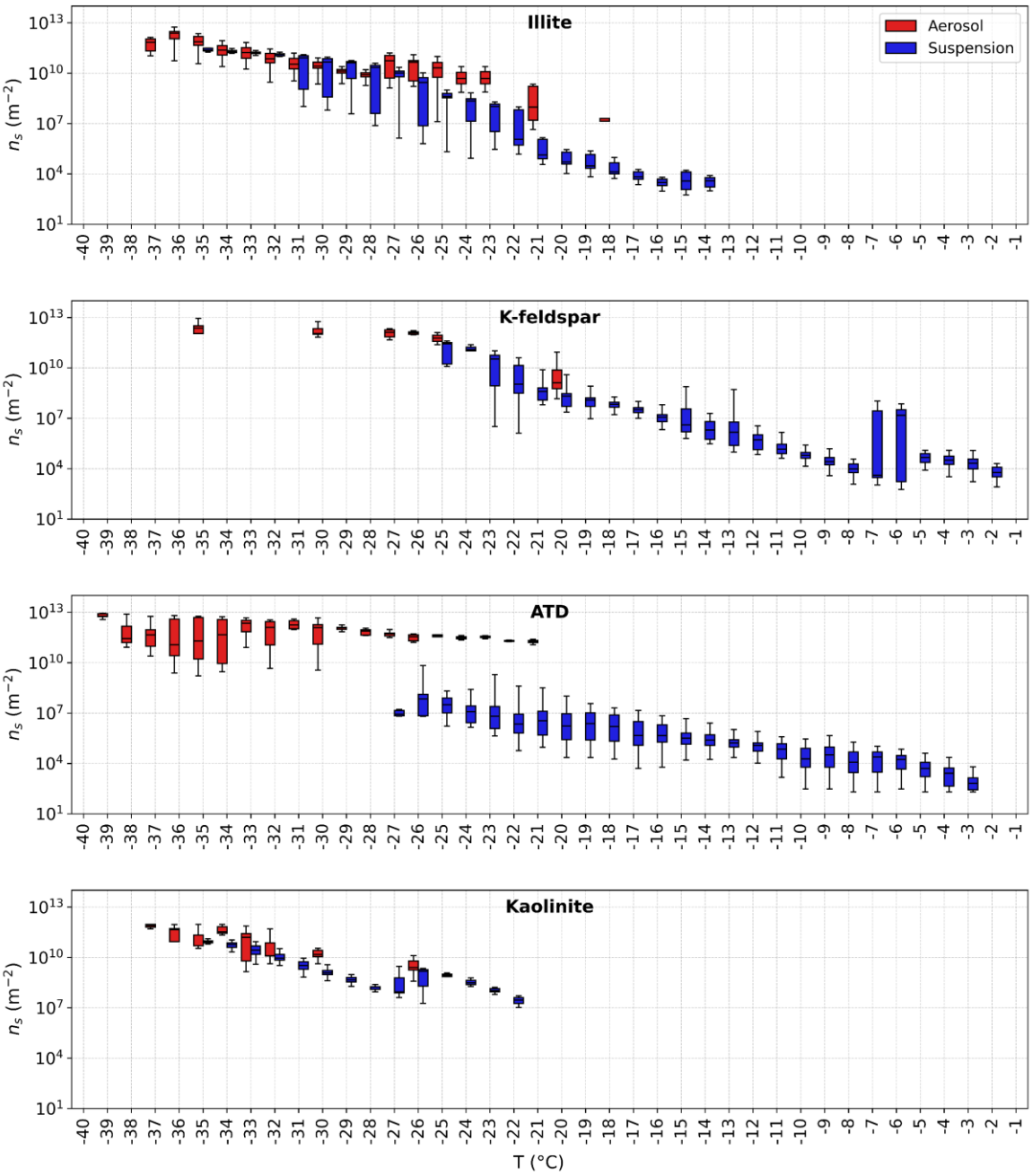


Figure S2. Boxplots of n_s dust particles with different mineral compositions measured from dry-dispersed particles (aerosol, red) and wet-suspended particles (suspension, blue). The n_s were binned into 1 °C intervals at -40 < T <-3 °C. n_s of quartz, non-potassium feldspars and montmorillonite are not shown due to the absence of or inadequte data from aerosol measurements for each temperature bin.

2. The paper appears to assume that 'aerosol' approaches are, in some sense, the correct ones. The manuscript seems to carry an implicit assumption that dry-dispersed 'aerosol' methods give the more correct or atmospherically representative measure of dust ice nucleation activity, whereas suspension methods are treated as being depressed by water exposure and aggregation. This perspective/bias runs through the discussion and is especially clear in the atmospheric implications section, where aerosol-derived ns is presented as representative of fresh near-source dust, while suspension-derived ns is treated more as something affected by artefacts or

processing. I do not think this is adequately thought through or justified. It may indeed be the case that some droplet-freezing experiments are influenced by aggregation or prolonged water contact. But the converse is also true: many aerosol instruments have short residence times compared with the timescales over which particles may be immersed in cloud droplets in the atmosphere. If there are rapid aging effects, or if immersion time itself matters, then short-residence-time aerosol measurements are also only probing a particular experimental regime, not some obviously privileged 'truth.' The manuscript acknowledges different water-contact times and briefly notes time dependence via Jhet but the discussion still treats aerosol methods as more representative in a way that seems asserted rather than demonstrated.

If the authors wish to argue that one class of method is more atmospherically relevant than the other, then they need to define the atmospheric scenario and provide evidence for that claim. As things stand, I do not think the asymmetry in how the two method classes are treated is warranted.

We agree with Reviewer 2 that we have in our discussion of the different methods inadvertently implied one technique is superior to the other, which was not our initial intention. Instead, the discussion should remain neutral and emphasize that different methods probe different experimental regimes. To address this comment, along with similar concerns raised by Reviewer 1, we have thoroughly revised the relevant sections of the manuscript:

We have revised the text from "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." (Lines 567-571 in the original manuscript) 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.

We also revised "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" (Lines 617 to 621 in original manuscript) to "Because aerosol instruments typically have short water-particle contact times, they capture the INA of dust particles under conditions of rapid water activation. 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." (Lines 714-720 in revised manuscript).

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 moderated our discussion of the coagulation effect throughout the text (please refer to comment 4), clarifying that it does not occur in all suspension measurements, but may arise under conditions of high particle concentrations.

3. The 'aerosol' / 'suspension' binary is itself too coarse. The manuscript tends to treat 'aerosol' methods and 'suspension' methods as though they were two coherent and internally homogeneous methodological classes. I do not think that is really tenable. Each category contains a range of techniques with different sample preparation routes, concentrations, residence times, droplet sizes, cooling rates, activation conditions, and thermal histories. The manuscript itself notes variation in chamber residence times and in droplet-freezing conditions across studies.

We agree with Reviewer 2 that treating aerosol and suspension methods as two distinct homogeneous categories may introduce uncertainty in measuring INA of dust particles, as each category encompasses a range of experimental techniques and conditions. However, the primary goal of our study is to develop parameterizations that capture the general INA of dust particles and can be used in models. Therefore, we focus on the major sources of variability in INA of dust particles, such as measurement method, chemical composition, and aging processes, rather than providing a detailed evaluation of every individual experiment.

To address Reviewer 2's concerns, we explain in the Atmospheric Implications section the limitations of our binary measurement categories and the uncertainties arising from methodological differences within each measurement category in Lines 748-756 (revised manuscript): "In the present work, a binary classification of measurement methods (i.e., aerosol

versus suspension) was adopted. We note that studies using both methods encompass a wide range of experimental techniques and conditions, including differences in sample preparation, particle concentration, residence time and droplet size (as listed in Table S2). These variations can introduce uncertainty in dust INA even within the same measurement category, which is not explicitly accounted for in the present work. We therefore encourage future studies to carefully control and document sample preparation and experimental conditions, which will help minimize differences arising from methodological variability and enable a direct comparison across studies."

4. The manuscript puts too much weight on the aggregation/coagulation hypothesis. The paper repeatedly advances the idea that the lower ns values obtained in suspension measurements, particularly at warmer temperatures, may be explained by particle coagulation in highly concentrated suspensions, which reduces effective surface area available for nucleation. This is discussed in Section 3.2 and is then used to motivate the De based treatment.

This may be a plausible hypothesis, but I do not think the paper provides actual evidence sufficient to carry the weight placed upon it. Nor, in truth, did I think the prior literature cited established this in a definitive physical sense either. The reasoning here seems entirely inferential: there is an apparent discrepancy between compiled aerosol and suspension datasets, one can imagine aggregation occurring in concentrated suspensions, and therefore aggregation is proposed as an important explanation for the discrepancy. One can easily perform calculation showing such a mechanism to be plausible, given a set of assumptions. But that is not the same thing as demonstrating the mechanism.

Moreover, impairment of ice nucleation activity by aggregation would presumably require that the aggregation in some way reduces access of water to the active nucleating surfaces or otherwise alters the availability of those sites. I do not see physical evidence for that process being provided here. The manuscript's De analysis shows that suspension-derived equivalent particle sizes are often large and interprets this as evidence of unrealistic multi-particle loading and coagulation, but this does not in itself establish the causal mechanism by which freezing efficiency is impaired.

I therefore think the language throughout the paper should be much more cautious. Aggregation/coagulation may be one possible contributor to the posited discrepancies (if these do indeed exist), but it is certainly not shown here to be the dominant explanation.

We thank Reviewer 2 for the comments. The potential role of coagulation effect is indirectly suggested by the D_e results, where unexpected high D_e values lead to distinct differences in droplet freezing temperatures observed for aerosol and suspension measurements, while comparable D_e values result in similar droplet freezing temperatures between two methods (Figure 4). It was also suggested by previous studies of illite by Emersic et al. (2015). However, we fully agree with Reviewer 2 that we did not provide direct observational evidence for particle coagulation in this work and this effect should be interpreted with caution.

To address the Reviewer's concerns, we have revised and tempered the discussion of the coagulation effect and its role in the observed methodological difference throughout the text,

clarifying that it is not expected to occur in all suspension measurements, but could arise under conditions of high particle concentrations. It is therefore considered as one possible factor that may influence some suspension measurements under specific experimental conditions, rather than a general explanation for differences between all suspension and aerosol measurements.

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 from "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" (Lines 325-327 in the original manuscript) to "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." (Lines 376-379 in the revised manuscript).

We modified from "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." (Lines 340-348 in the original manuscript) to "In this work, we did not 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 may occur under conditions of high particle concentrations." (Lines 392-407 in the revised manuscript).

We modified from "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" (Lines 353-354 in the original manuscript) to "These findings suggest that the 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 412-415 in the revised manuscript).

We modified from "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." (Lines 567-571 in the original manuscript) 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

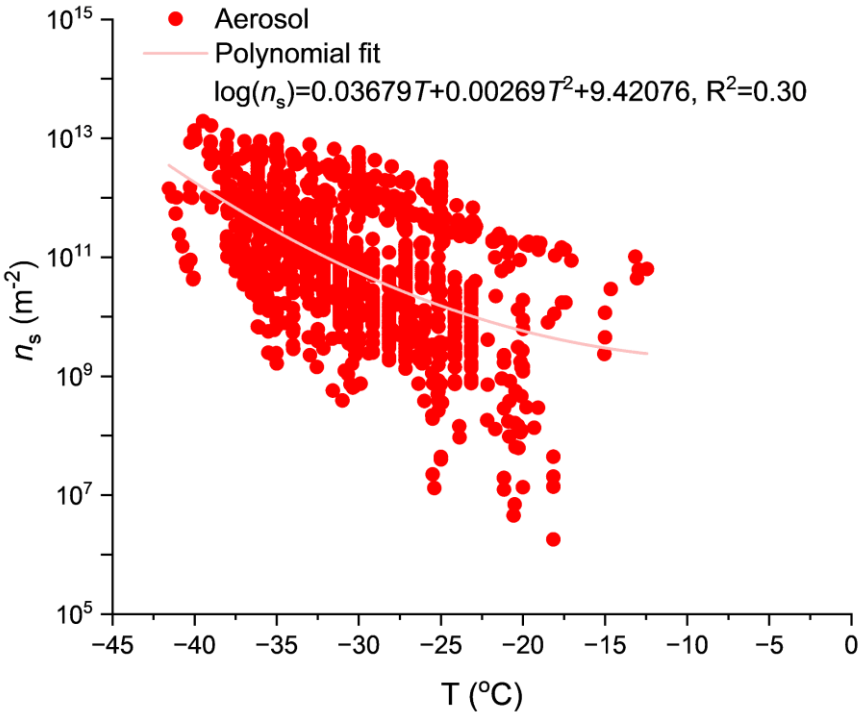
freezing if the water molecules can access the aggregated surfaces of the particles in the droplet." (Lines 660-664 in the revised manuscript).

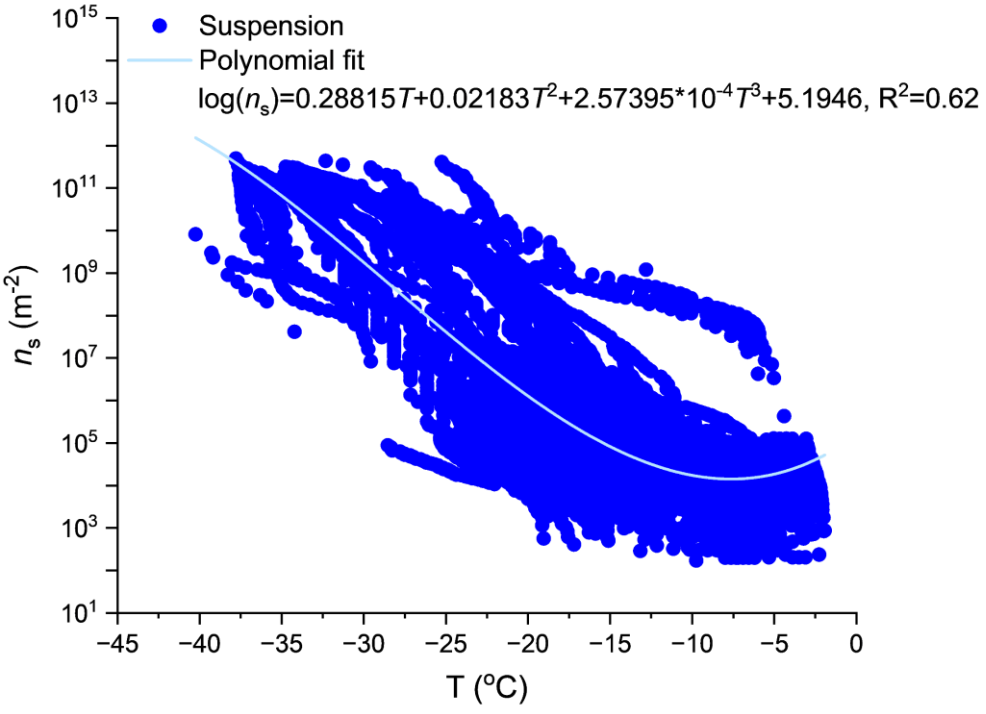
5. I am not at all convinced by the manuscript's implicit assumption that the fitted relationships should be linear in the chosen coordinates. I know it is pretty commonly observed/assumed, but what physical reason is there for this? The paper repeatedly adopts forms such as log(ns)=AT+B, but no real justification is given for why such a relationship should be expected for a compilation that mixes different minerals, different samples, different methods, and different aging pathways. In that setting, a straight line in log-space may simply be a convenient summary of a broad and heterogeneous cloud of data, rather than evidence of a common physical law. This concern is reinforced by the fact that the authors themselves introduce two separate temperature regimes for the suspension data, which rather undermines the idea that a simple linear form is appropriate in the first place. At minimum, I think the manuscript should acknowledge that the chosen fits are empirical conveniences and should avoid implying that the linearity itself has some deeper 'physically grounded' significance.

We acknowledge that the log-linear fits adopted in this work are empirical parametrizations intended as convenient summaries of the compiled datasets rather than evidence of a physical law. Nevertheless, these fits capture the general temperature trend of dust INA. For example, aerosol n_s increases with decreasing temperature and suspension n_s exhibits two-step temperature dependences (Figure 3 and Figure S3). Furthermore, the loglinear fits of n_s aligns with the majority studies that reported an exponential temperature dependence for dust INA (Niemand et al., 2012; Ullrich et al., 2017). Other fitting approaches were also tested, including polynomial and exponential forms, but they did not provide improved representations of the compiled data compared to log-linear fits. For example, the results of the polynomial fittings for aerosol and suspension n_s are shown in figures below. For aerosol n_s, the second-order polynomial fit shows no improvement in n_s representation compared to log-linear fits. A third-order polynomial fit leads to a higher correlation for suspension n_s (R^2=0.62 versus R^2=0.50 for log-linear fit). However, it fails to capture the expected physical behavior of n_s, instead of monotonically increasing as temperature decreases, the fitted curve exhibits a minima at temperatures above -10 °C. We also attempted a constrained polynomial fit by enforcing a strict monotonic increase of n_s with decreasing temperature, as shown in the second plot below. However, this constrained fit fails to capture the two-step temperature dependence of n_s (Figure S3), shifting the transition temperature from -18 °C to ≈-12 °C. The results of the exponential fittings for aerosol and suspension n_s are shown below (the second plot) as well. The data clearly demonstrates that the exponential function cannot successfully capture the temperature dependence of n_s in either method (R^2=0.03 for aerosol measurements versus R^2=0.04 for suspension measurements). Moreover, linear fits provide a simple and practical way to implement these parameterizations in models.

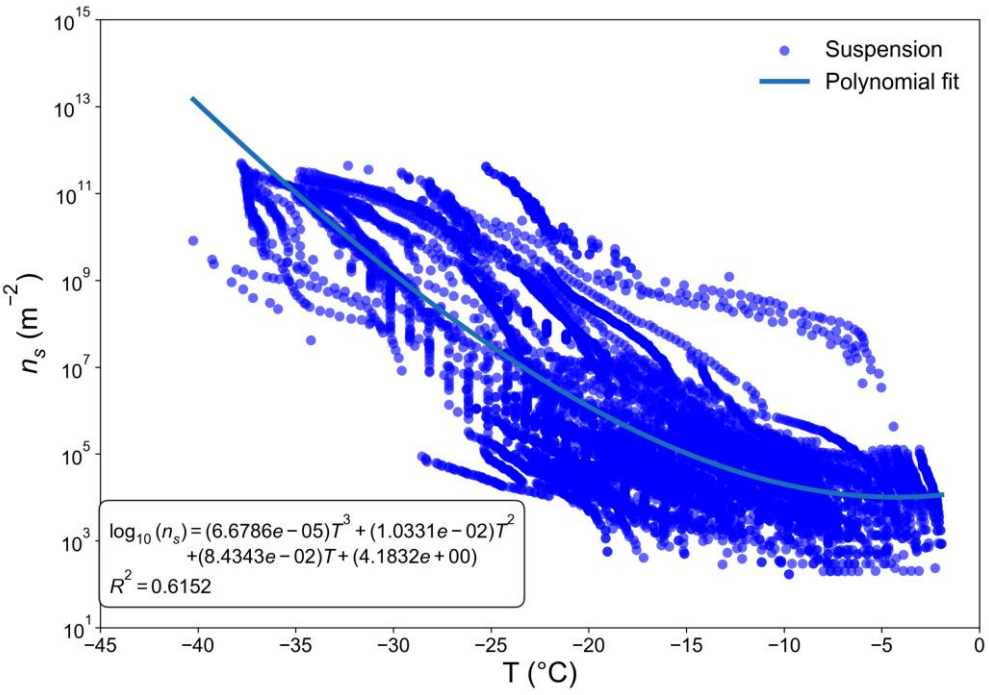
To address Reviewer's concerns and to clarify why log-linear form was selected, we have added the following statements in Lines 433-436: "This log-linear form is an empirical parameterization chosen to summarize the compiled datasets, capturing the general trend of temperature dependence of n_s and J_{het}. Alternative fitting approaches, including polynomial and exponential forms, were also tested but did not improve the representation of the data.".

Polynomial fits:

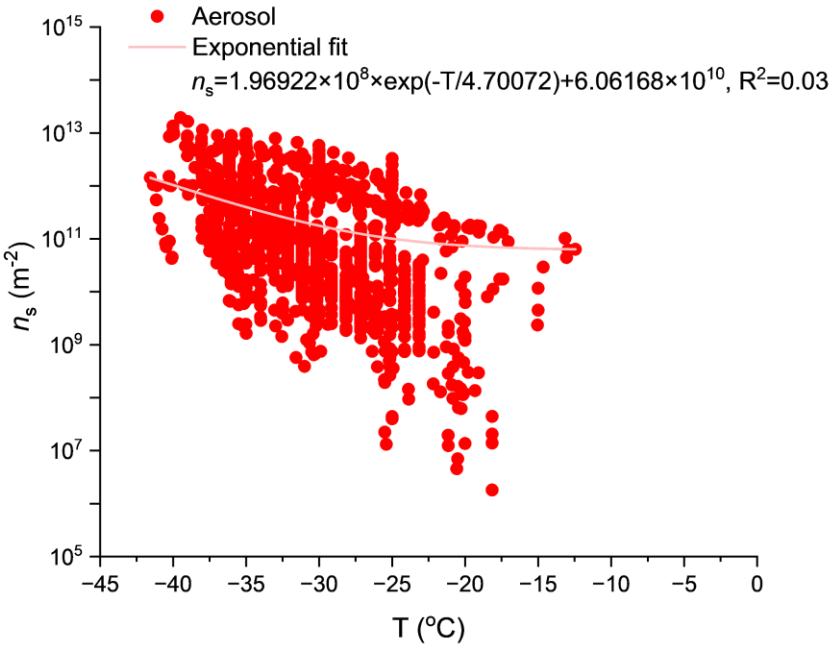


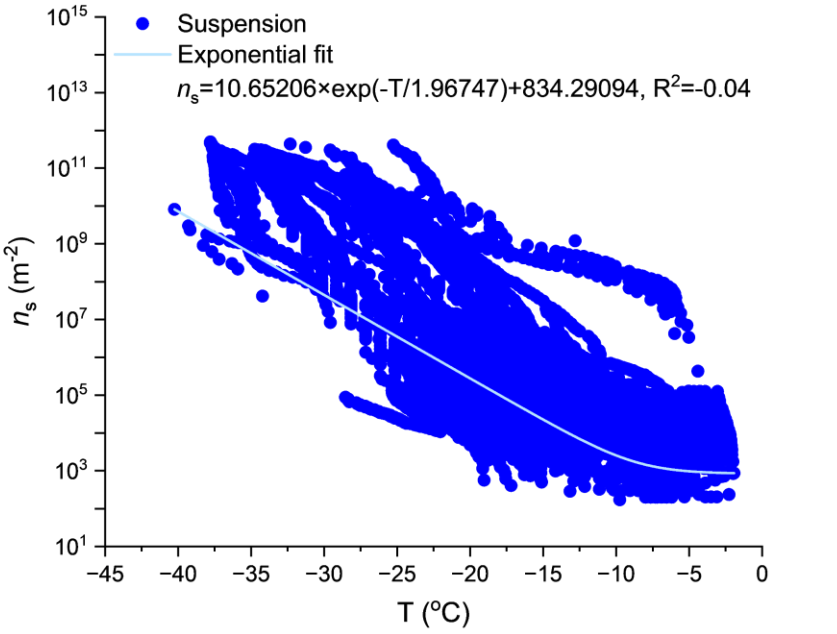


Polynomial fits (a strict monotonic increase of n_s):



Exponential fits:





6. I also take issue with much of what is said here about aging and water exposure. The discussion is presented rather too neatly, whereas the literature on these processes is more extensive and more nuanced than is reflected in the manuscript, see e.g (Reischel and Vali, 1975; Klumpp et al., 2022; Whale, 2022; Ren et al., 2022) amongst many others. Different dusts behave differently, the timescales matter, and I do not think the present paper supports the level of confidence with which some of these statements are made. I would suggest either engaging more carefully with the wider literature or substantially toning down the claims.

We acknowledge that different dust species may behave differently to aging processes. The INA of fresh and aged dust particles with specific species has been shown in Figure S5 (also shown

below). We have already presented detailed discussions about the effect of aging on specific dust species (Lines 521-550 in the revised manuscript):

"To account for this, Figure S5 provides elaborated n_s boxplots for individual dust species, including K-feldspar, non-potassium-rich feldspars, quartz, ATD and natural dust (separated desert and volcanic dust). Clay particles (illite, kaolinite, and montmorillonite) are not shown due to limited data from two methods for comparison between fresh and aged particles. Given the significant contribution of clay particles to dust mass (Atkinson et al., 2013), additional measurements are needed to quantify the aging effect on their INA. Figure S5 indicates that aging may have different effects on INA of different dust species. Specifically, ATD and natural dust (both desert and volcanic dust) show a decrease of 1–2 orders of magnitude in n_s at -39 < T < -3 °C after aging, with the effect becoming more pronounced at lower temperatures. In contrast, the n_s changes of K-feldspar and quartz particles are generally within one order of magnitude, indicating that these dust species are relatively insensitive to aging. This observation is consistent with Harrison et al. (2016), who found most K-feldspars exhibited no significant change in INA after water aging. While Harrison et al. (2019) noted that certain quartz can be highly sensitive to water aging, only one of the three samples exhibited a pronounced decrease in n_s after 10 months of water aging, whereas n_s decrease of other quartz samples were within one order of magnitude. Their findings therefore align with our results showing insensitivity of quartz to aging treatments. The response of non-potassium-rich feldspar ("other feldspars" in Figure S5) (Harrison et al., 2019; Peckhaus et al., 2016) to aging process, including albite and anorthite, is more variable. Depending on temperature, aging can increase, decrease or leave their n_s unchanged (Figure S4). For most temperatures, the n_s changes of other feldspars remain within one order of magnitude. An exception occurs at -25 < T < -23 °C, where the n_s of aged particles increased by 2–5 orders of magnitude compared to fresh particles. This occurs because most fresh non-potassium-rich feldspars exhibit relatively low INA. However, one aged sample from Harrison et al. (2016) displayed markedly higher INA, which strongly influenced the median n_s values. This variability reflects both the compositional diversity of feldspars and the limited number of available datasets for aged non-potassium-rich feldspars, as indicated in Figure S5. Overall, our results suggest that feldspars (both K-feldspar and other feldspars) and quartz are generally insensitive to aging process compared to the natural dust and ATD samples presented here.".

We acknowledge that our discussion of aqueous aging is relatively simplified, as we intentionally report the impact of aging pathways on the general INA of dust particles. To address this concern, we have researched the literature and added the following discussions on aqueous aging in Lines 607-621 (revised manuscript): "The findings regarding water/aqueous aging are consistent with previous studies, which report that immersion in solutions generally lowers the INA of dust particles. For example, Reischel and Vali (1975) reported freezing suppression of freezing in kaolinite-containing particles by NaI, KI, NaCl, CsCl and K_2SO_4. Abdelmonem et al. (2017) observed a reduction in the INA of α-alumina immersed in solutions of various pHs. Exceptions occur for ammonia or ammonium salts, where increases in INA have been observed for quartz (Whale et al., 2018), kaolinite (Reischel and Vali, 1975; Kumar et al., 2019) and feldspars (Klumpp et al., 2022; Whale, 2022; Whale et al., 2018). Whale (2022) attributed this enhancement to the disruption of thermodynamically unfavorable hydrogen

ordering in ice clusters induced by water-orienting nucleating surfaces (i.e., dust surfaces) in NH_4^+-containing solutions. In contrast, Kumar et al. (2019) hypothesized that the adsorption of NH_3 and NH_4^+ on the feldspar and kaolinite promotes the arrangement of water molecules into an ice-like pattern through hydrogen bonding, and thereby enhancing the ice nucleation of dust particles. These exceptions highlight the variability in the freezing behavior of different dust species in response to diverse aging processes (Figure 6).”.

The conclusion that aging leads to a reduction in the INA of dust particles (Figure 5) should thus be interpreted as a general or averaged trend across dust particles with diverse mineralogical compositions and subjected to various aging processes. Moreover, the reduced INA after aging has also been observed for natural dust samples (Figure S5), indicating that our conclusion of “the reduced INA after aging” is relevant of atmospheric dust particles. This has been now clarified in Lines 621-626: “Therefore, the fitting curves in Figure 6 represent the average INA of aged dust particles with mixed mineral compositions and interact with various solutes. We note that the actual dust INA may deviate from these average trends in specific atmospheric environments where certain reactive solutes (e.g., ammonium salts) dominate the aging pathways. A reduction in INA after aging has also been observed in natural dust samples (Figure S5), indicating that this general aging effect is representative of atmospheric dust behavior.”

We also acknowledge that the time effect on the INA of aged dust particles is not explicitly discussed in this work due to limited available datasets investigating aging time effect on similar dust species. We now point out this limitation of our parameterizations in Lines 767-769: “In addition, these aging parameterizations do not account for the time scale of aging. Future studies should systematically investigate time-dependent aging to improve INP predictions across different stages of dust plume evolution.”.

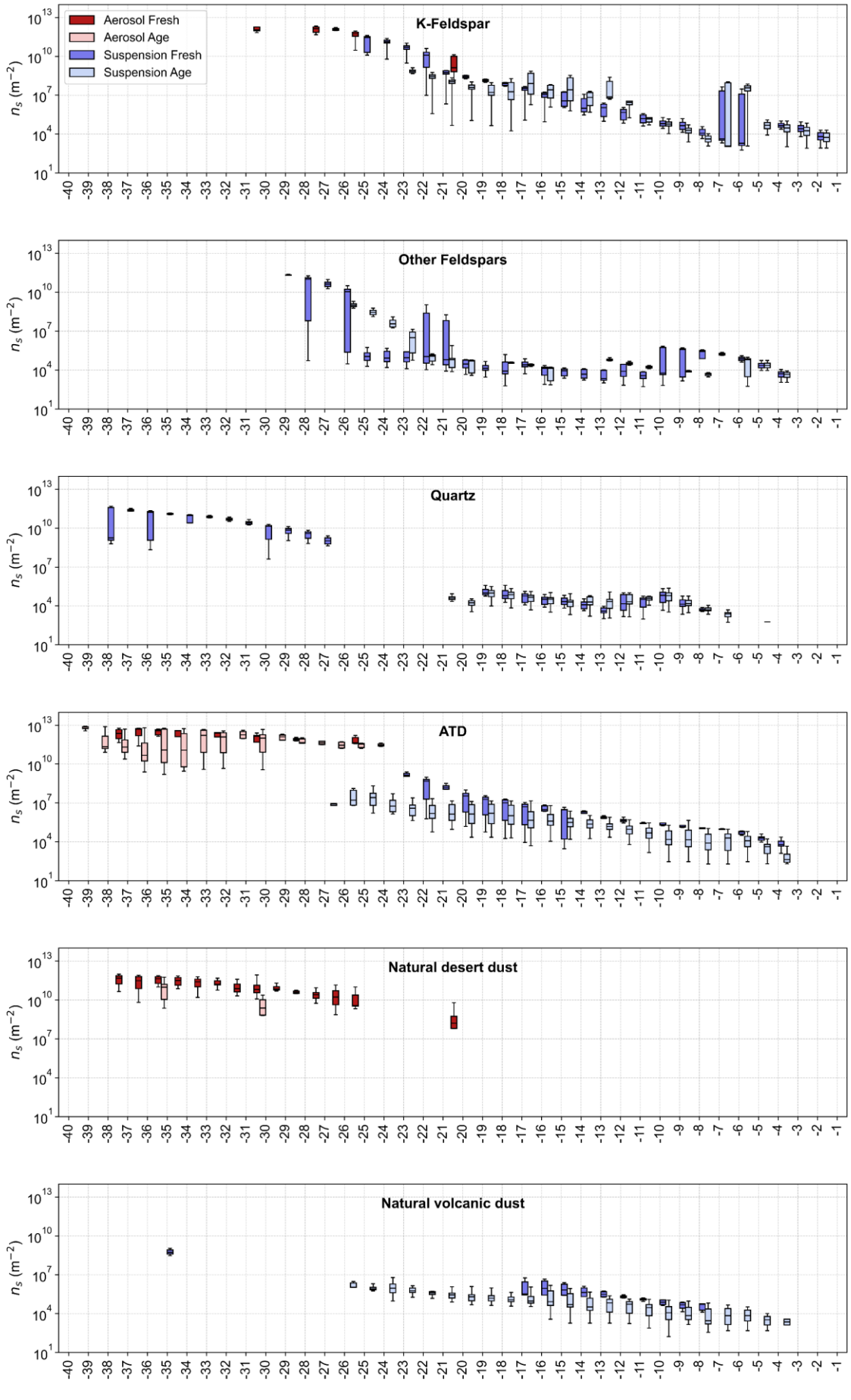

Aerosol Fresh
Aerosol Age
Suspension Fresh
Suspension Age
K-Feldspar
Other Feldspars
Quartz
ATD
Natural desert dust
Natural volcanic dust
n_s (m^{-2})
T (°C)

Figure S5. Boxplots of n_s for dust particles with different compositions, including K-feldspar, non-potassium-rich feldspar (other feldspars), quartz, ATD and natural dust. The n_s data were binned into 1 °C intervals at -40 < T <-1 °C.

7. I want to stress that I do think the plots are interesting. There is value in assembling this literature, and there is value in showing the broad spread of measurements and the practical challenges this presents for parameterization. But, to my mind, a quite different conclusion should be drawn. The most striking thing about the compilation is not that it proves a fundamental and general discrepancy between aerosol and suspension methods. Rather, it is that a body of literature spanning many minerals, sample origins, and experimental methods still shows some consistency as it does. That suggests to me that the interesting scientific question is not 'which method is right, and how do we correct the other one?' but 'under what specific circumstances, and for which materials, do different experimental approaches diverge meaningfully?' The present manuscript is not really set up to answer that question, because it aggregates unlike things too readily and then draws a conclusion that rests on that aggregation. For that reason, I think that if this work were to be developed properly, it would need to become a rather different paper.

We completely agree with Reviewer 2 that the more fundamental scientific question is not which method is "correct," as both methods represent a regime of experimental conditions. To address this concern, we have revised our manuscript about the two methods accordingly (please refer to comment 2 and 3). We now also discuss that despite the complexity in mineral dust composition where the similarities still yield overlapping results among the two methods and discuss which level of experimental complexity can be accounted for in parameterizations developed for use in cloud and climate models.

For example, we pointed out in Line 393-396 comparable D_e values are found between two methods at low particle suspension concentrations: "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.". Comparable n_s and J_{het} was also observed at low temperatures between aerosol and suspension measurements: "The median values of aerosol n_s are up to 6 orders of magnitude higher than suspension n_s at -38 < T < -18 °C, with the n_s gap narrowing with decreasing temperatures" in Line 303-305 and "The gap between aerosol and suspension J_{het} narrows with decreasing temperature and becomes comparable (within one order of magnitude) at T < -27 °C." in Line 314-315. The similarity among two methods at lower temperature is likely because: "DFT measurements at relatively lower temperatures (-38 < T < -19 °C) were conducted using picoliter-size droplets (Table S2) (Atkinson et al., 2013; Hiranuma et al., 2015; Zolles et al., 2015) with low particle concentration and minimal potential for particle coagulation." (Line 410-412 in revised manuscript).

We also pointed out the magnitude of methodological divergence is influenced by dust mineralogy, where K-feldspar and kaolinite show a smaller difference compared to other species in Line 287-291: "Specifically, the median n_s values between aerosol and suspension methods differ by up to three orders of magnitude at -31 < T < -18 °C for illite, by 3–5 orders

of magnitude at -27 < T < -21 °C for ATD and up to one order of magnitude at -35 < T < -30 °C for kaolinite. K-feldspar and kaolinite n_s show the closest agreement between the two methods, with differences generally within an order of magnitude." and moderated the corresponding conclusion of methodological divergence related to dust compositions in Line 298-300: "Nevertheless, the consistent differences observed across multiple groups suggest that measurement method contributing to the variability in reported dust INA cannot be excluded but this is likely convoluted by changes in composition".

We discussed the atmospheric applications of n_s parameterizations in models in Line 708-712: "The parameterizations that represent the average freezing ability of dust particles are appropriate for atmospheres with compositionally diverse dust or for global-average applications. The composition-specific parameterizations are tailored to conditions where the INP population is largely governed by a single mineral type, making them more suitable for regional or local applications" and in Line 714-723: "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. 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". For D_e -based parameterizations, we stated "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." in Line 723-727.

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