

Dear Editor and Referees,

Please find below our point-by-point responses to the comments on our manuscript entitled “Immersion freezing of surface dust particles from Tengger Desert in China: effects of mineral composition and aging treatments on ice nucleating activity” by Xue et al.

We sincerely thank the Editor and Referees for your insightful comments and constructive suggestions, which have helped us improve the manuscript. We have carefully considered each comment and revised the manuscript accordingly. Our responses are provided below, and the corresponding changes are highlighted in the revised manuscript. We hope that the revisions satisfactorily address the concerns raised and that the manuscript is now suitable for publication.

During the revision process, Mr. Yingjie Zhou conducted additional experiments on temperature calibration and ice nucleation at different cooling rates, which were essential to addressing the reviewers' comments. With the agreements of all coauthors, we therefore added Yingjie Zhou as a coauthor which is reflected in the revised manuscript.

Thank you for your time and consideration.

On behalf of all co-authors,
Bingbing Wang

In the following point-by-point responses, reviewer's comments are in black font, our response is in light blue normal font, and the corresponding revision are in blue font. Line numbers in the responses refer to the revised manuscript. The line numbers in the referees' comments refer to the previous version of manuscript.

Referee #1

Xue et al. investigate immersion freezing of surface dust from the Tengger Desert and examine the effects of ammonium sulfate, NaCl, acids, and heating. The dataset is extensive. The combination of freezing measurements and single-particle analysis is useful.

Overall, I think the study is suitable for publication after minor revision. My main concerns are related to the interpretation of the mineralogical effects, the calculation of particle surface area, and several mechanistic claims that are stronger than supported by the measurements.

Major Comments:

1. The authors attribute the higher ice nucleation activity of Us-dust mainly to a ~2% increase in feldspar fraction. They further state that this increase “resulted in” a ~2 K increase in T₅₀ and an order-of-magnitude increase in N_s. However, it is difficult to find and establish such a causal relationship. The data are consistent with an important contribution from feldspar, but they do not quantitatively demonstrate that the 2% increase in feldspar alone caused the observed enhancement. The authors should use more cautious wording.

Response: Thank you for this important comment. Our single particle analysis showed an increase in the feldspar fraction between the two dust samples, indicating potentially important contribution from feldspar to ice nucleation. However, we agree that our data do not

quantitatively demonstrate that the 2% increase in feldspar content alone caused the observed enhancement in ice nucleation activity. We therefore revised the relevant statements throughout the manuscript to avoid implying a direct causal relationship and attributing the enhancement solely to the increased feldspar.

Revised text:

Line27 in Abstract: “A minor increase in feldspar content (~2% in surface area) quantified by single particle analysis may contribute to the enhanced ice nucleation active site density (N_s).”

Line330 in the main text: “Given that the ΔT_{50}^w of chlorite and illite are lower than Us-dust, the improved IMF ability of Us-dust relative to S-dust may be partly attributed to the increased feldspar content.”

Line635 in the main text: “The 2% increase in particle surface area of feldspar may contribute to the approximately 2 K higher T_{50} and one order of magnitude increase in N_s . ”

1. *N_s is central to the manuscript. However, the surface area is estimated from CCSEM/EDX measurements of aerosolized particles, mainly between approximately 0.2 and 2 μm , whereas the original dust was sieved to $<74 \mu\text{m}$. Nebulization, transport, and impaction can strongly bias the measured particle-size distribution, particularly for larger particles and aggregates. It is therefore not obvious that the CCSEM population accurately represents the particles present in the 1 μL freezing droplets. An independent measurement of the suspension particle-size distribution would be very helpful. Additionally, it's important to clearly state that the surface area used here is a physical/geometric surface area derived from CCSEM particle size, rather than a BET specific surface area. This distinction is important because many literature N_s datasets and parameterizations use BET-derived surface area. Direct comparison of N_s values may therefore not be strictly equivalent.*

Response:

We thank you for raising this important point. We agree that nebulization, transport, and impaction may introduce size biases in the CCSEM/EDX measurements. We have conducted additional dynamic light scattering (DLS) measurements to independently characterize the particle size distribution in the dust suspensions and estimate the corresponding surface area in a 1 μL droplet. For 0.1 wt% suspension, the estimated surface areas per droplet obtained from DLS and CCSEM were $1.0 \times 10^6 \mu\text{m}^2$ and $2.4 \times 10^6 \mu\text{m}^2$ for Us-dust, and $8.2 \times 10^5 \mu\text{m}^2$ and $2.5 \times 10^6 \mu\text{m}^2$ for S-dust, respectively. Although the estimates differ by about a factor of 2–3, they are within the same order of magnitude. This comparison provides an independent assessment of the approximate magnitude of the geometric surface area estimated from CCSEM/EDX, although it is not necessary quantitatively equivalent between the two methods.

Only the upper approximately 10 mL of the 20 mL dust suspension was used for both ice nucleation experiments and CCSEM/EDX analysis. Larger particles and aggregates may have settled toward the bottom of the vial before sampling, potentially explaining why the CCSEM/EDX measurements predominantly captured particles in the approximately 0.2–2 μm size range, despite the original dust being sieved to less than 74 μm . As shown in Fig. S4, the size range determined by CCSEM/EDX is in general consistent with DLS measurements, which show that the majority of particles are less than 3 μm (hydrodynamic diameter).

We explicitly stated in the revised manuscript that N_s was calculated using geometric surface area derived from CCSEM/EDX analysis, rather than BET specific surface area. Because these approaches quantify surface area with different underlying measurement principles, direct comparisons with literature N_s values or their parameterizations based on BET surface area should be made cautiously. Here, N_s was calculated using the geometric surface area estimated from CCSEM/EDX measurements. This approach provides a practical basis for estimating particle surface area from size resolved measurements and is relevant to atmospheric applications, where size distributions are more commonly available than BET measurements of collected aerosol samples. The corresponding clarifications and DLS results were added to the revised manuscript and Supplementary Information.

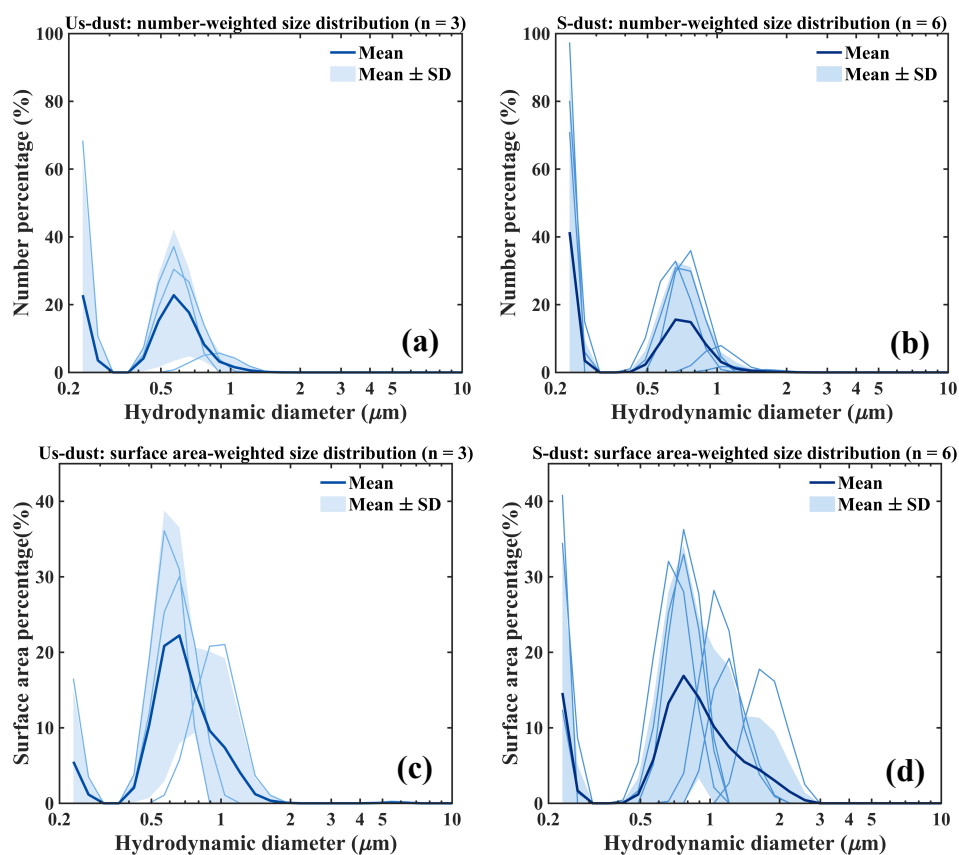


Figure S4. The normalized surface area-weighted (a, b) and number-weighted (c, d) particle size distribution determined by DLS for Us-dust (a, c) and S-dust (b, d) suspensions. Light blue lines show individual replicate measurements; n denotes the number of replicates. Dark blue lines indicate the mean, and shaded regions indicate ± 1 standard deviation (SD).

Revised text:

Line150 in the main text: “The upper portion (~10 mL) of each 20 mL dust suspension was collected and serially diluted with ultrapure for immediate use in the ice nucleation experiments.”

Line181 in the main text: “Briefly, the upper portion (~10 mL) of each 20 mL dust suspension was nebulized. The generated aerosols were carried by high-purity nitrogen through a diffusion dryer and collected onto TEM copper grids (01814-F, Ted Pella) using an impactor (SKC, Inc.).”

Line284 in the main text: “The surface area used to calculate N_s and J_{het} was estimated from the geometric particle diameters measured by CCSEM/EDX, rather than Brunauer-Emmett-Teller (BET) specific surface area. Because these methods quantify different surface area metrics, comparisons with literature N_s and J_{het} values or parameterizations based on BET surface area should be made cautiously. Dynamic light scattering (DLS) measurements were performed to independently assess the particle size distribution in the suspensions and estimate the geometric surface area per 1 μL droplet (Text S2 and Fig. S4). The DLS- and CCSEM-derived surface area estimates were within the same order of magnitude, although the latter were approximately 2-3 times higher.”

Line58 in the Supplementary Information, Text S2: “Particle size distribution and surface area. To independently assess the particle surface area in the dust suspensions, additional measurements were performed using dynamic light scattering (DLS), and the corresponding geometric surface area per 1 μL droplet was estimated from the measured size distributions. The particle size distributions of suspended dust particles were measured using a DLS instrument (Zetasizer Lab, Malvern Panalytical, UK). The refractive index of the dust particles was set to 1.46, and water was used as the dispersant. The measurement temperature was maintained at 25 °C. For each sample, 1 mL of S-dust or Us-dust suspension was transferred into a quartz cuvette and analyzed. At least, three replicate measurements were performed for each sample. The scattered light intensity data were processed using the instrument software to obtain volume-weighted and number-weighted particle size distributions. The surface area was subsequently calculated from the volume-weighted particle size distribution.

For 0.1 wt% suspension, the estimated surface areas per droplet obtained from DLS and CCSEM were $1.0 \times 10^6 \mu\text{m}^2$ and $2.4 \times 10^6 \mu\text{m}^2$ for Us-dust, and $8.2 \times 10^5 \mu\text{m}^2$ and $2.5 \times 10^6 \mu\text{m}^2$ for S-dust, respectively. These two methods yielded estimates within the same order of magnitude, with the CCSEM-derived values approximately 2–3 times higher. These differences may reflect differences in particle size characterization and the underlying measurement principles. The original dust was sieved to $<74 \mu\text{m}$ and only the upper portion ($\sim 10 \text{ mL}$) of each 20 mL suspension was used for both ice nucleation experiments and CCSEM/EDX analysis. Larger particles and aggregates may have settled toward the bottom of the vial before sampling, potentially reducing their representation in the upper suspension and contributing to the smaller particle size range ($\sim 0.2\text{--}2 \mu\text{m}$) observed in the CCSEM/EDX measurements. As shown in Fig. S4, the size range determined by CCSEM/EDX is generally consistent with the DLS measurements, which indicate that the majority of particles have hydrodynamic diameter below $3 \mu\text{m}$. Nebulization, aerosol transport, and impaction may introduce size dependent biases in the particles collected for CCSEM/EDX analysis. Nevertheless, the agreement between the DLS- and CCSEM-derived surface area estimates suggests that these differences did not result in an order of magnitude discrepancy in the estimated surface area under the conditions examined.”

2. *The mechanism proposed for ammonium sulfate is too strong. The observed enhancement is consistent with mechanisms proposed in previous studies, but several mechanisms may contribute. Please change statements such as “directly support” to more cautious language unless additional surface characterization is provided.*

Response: Thank you for this important comment. We agree that our experimental results do not provide direct surface level evidence to establish a single mechanism responsible for the observed enhancement by AS. The observed enhancement is consistent with mechanisms proposed in previous studies, including NH_4^+ adsorption on mineral surfaces, but other mechanisms may also contribute. Further surface characterization would be needed to distinguish the relative contributions of these mechanisms. Therefore, we revised the relevant statements throughout the manuscript to avoid implying direct mechanistic evidence and to present NH_4^+ adsorption as a plausible and potentially important mechanism.

Revised text:

Line32 in the Abstract: “This sigmoidal response is consistent with a possible contribution from NH_4^+ adsorption on dust surface.”

Line387 in the main text: “The observed sigmoidal response of ΔT_{50} suggests a concentration dependent activation of ice nucleation sites on dust surface upon AS exposure.”

Line475 in the main text: “In addition, NH_4^+ adsorbed on dust surfaces may alter the arrangement and orientation of water molecules, potentially enhancing ice nucleation activity (Anim-Danso et al., 2016; Kumar et al., 2018; Whale et al., 2018; Worthy et al., 2021) (Anim-Danso et al., 2016; Kumar et al., 2018; Worthy et al., 2021).”

Line484 in the main text: “Our experimental data are consistent with NH_4^+ adsorption on mineral surface being an important mechanism contributing to the ice nucleation enhancement by AS. The ice nucleation enhancement on Us-dust by AS observed here may involve NH_4^+ adsorption on the dust surface, while other mechanisms, such as the participation of NH_4^+ in ion exchange (Worthy et al., 2021) and regulation of hydrogen-bond ordering on dust surfaces (Whale, 2022), may also contribute the observed enhancement.”

Line643 in the main text: “This supports the possibility that NH_4^+ adsorption on mineral surface and the associated activation of ice nucleation active sites may be an important mechanism for the ice nucleation enhancement by AS.”

3. *Line 208: Please justify the use of a cooling rate of 10 K/min, which is substantially faster than the ~1 K/min commonly used in droplet-freezing studies. Such a rapid cooling rate may increase thermal lag and temperature uncertainty, and can also affect the derived J_{het} . Please explain why 10 K/min was chosen and clarify how the stated temperature uncertainty of ± 0.5 K was validated under this dynamic cooling condition.*

Response: Thank you for raising this important concern. A cooling rate of 10 K min^{-1} was selected based on previous studies (Knopf and Lopez, 2009; Koop et al., 1998) and to reduce the time available for water vapor transfer between supercooled droplets and ice crystals (frozen droplets), thereby limiting potential interference from the Bergeron–Wegener–Findeisen process. We conducted additional experiments to evaluate the thermal lags and the influence on freezing temperature at different cooling rates. **The freezing temperature and the related analysis have been updated after calibration including the figures and tables, which are all highlighted in the revised manuscript. The main conclusions remain after this temperature calibration.**

First, we calibrated the temperature measurements using the melting points of decane (243.45 K) and dodecane (263.55 K) (Knopf and Lopez, 2009) at heating rates of $0.5\text{--}10 \text{ K min}^{-1}$, following Budke and Koop (2015). At 10 K min^{-1} , the measured melting temperatures differed from the literature values by approximately 2.1 K for decane and 1.6 K for dodecane (Fig. S2).

The absolute difference between the calibrated temperatures and the literature values was determined for individual calibration. We reported a conservative temperature uncertainty as the standard deviations of these absolute differences, as shown as error bars in Fig. S2, which is smaller than 0.5 K at 243 K and 0.3 K at 263 K. We applied the resulting calibration fit (Fig. S2) to all the raw freezing temperatures. We have clarified this calibration procedure and conservative temperature uncertainty in Text S1. We also clarify that applying the heating mode calibration to the cooling mode freezing measurements assumes comparable thermal lag during heating and cooling at the same temperature change rate.

Second, we conducted additional freezing experiments using water droplets and Tengger Desert dust suspensions at different cooling rates. The median freezing temperature (T_{50}) showed little variation at cooling rate between 0.5 and 2 K min⁻¹, whereas the difference in T_{50} between 10 and 1 K min⁻¹ was +1.1 K at 255 K and -0.9 K at 245 K (Fig. S3). Previous studies have demonstrated that immersion freezing can exhibit cooling rate dependence due to the time dependent nature of nucleation, although the magnitude of this effect varies among ice nucleating materials and with particle surface area (Broadley et al., 2012; Wright et al., 2013). Studies of kaolinite containing droplets reported an ~2 K decrease in freezing temperature when the cooling rate increased from 1 to 10 K min⁻¹, whereas smaller shifts have been reported for pure water droplets (Murray et al., 2011, 2012). Taken together, these studies indicate that the approximately 1 K difference observed in our study is consistent with previously reported cooling rate effects, although it cannot be attributed exclusively to time dependent nucleation, and potential contributions from temperature uncertainties cannot be excluded. We have therefore clarified that our measurements and derived J_{het} values correspond to a cooling rate of 10 K min⁻¹. Comparisons of J_{het} values with studies using different cooling rates should be made cautiously. The corresponding discussion has been added to the revised manuscript and Supplementary Information.

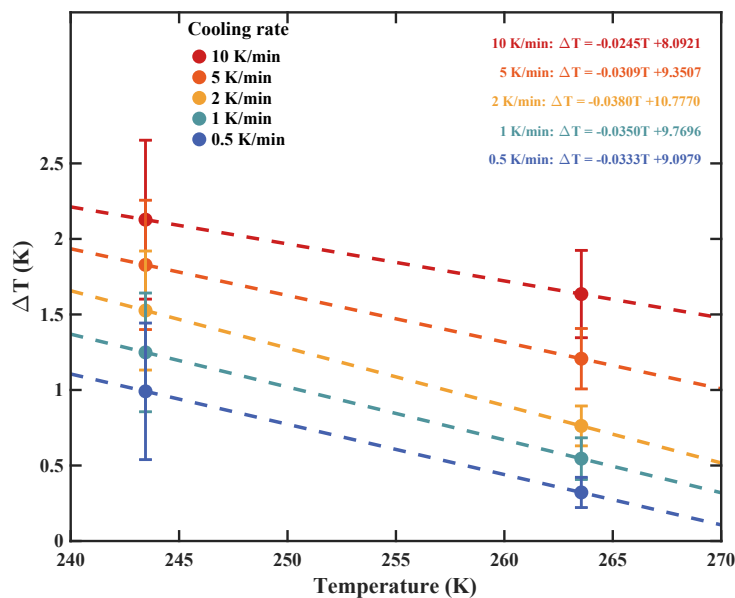


Figure S2. Temperature calibration based on the differences between the experimentally measured melting temperatures of decane and dodecane droplets and their respective literature

values as heating rate of 0.5-10 K min⁻¹. Error bars indicate the standard deviations of the absolute deviations from the reference values.

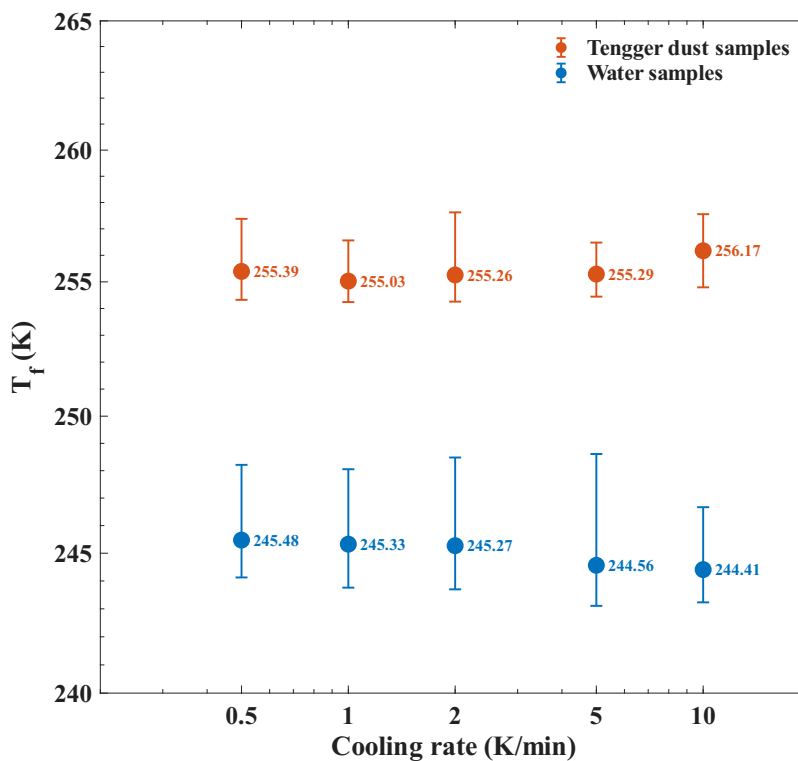


Figure S3. Median freezing temperatures of water droplets and Tengger Desert dust suspensions measured at different cooling rates. Error bars indicate the 25th and 75th percentiles.

Revised text:

Line213 in the main text: “Droplet temperature was calibrated against the melting points of decane and dodecane and a conservative temperature uncertainty was estimated to be ± 0.5 K (Text S1 and Fig. S2).”

Line216 in the main text: “The cooling rate of 10 K min⁻¹ was selected following previous studies (Knopf and Lopez, 2009; Koop et al., 1998) to reduce the time available for water vapor transfer between supercooled droplets and ice crystals and minimize potential interference from the Bergeron–Wegener–Findeisen process.”

Line229 in the main text: “Additional freezing experiments were conducted using water droplets and Tengger Desert dust suspensions at different cooling rates (Text S1 and Fig. S3). T₅₀ showed little variation at cooling rate between 0.5 and 2 K min⁻¹, whereas the difference in T₅₀ between 10 and 1 K min⁻¹ were +1.1 K at 255 K and -0.9 K at 245 K. These differences may partly reflect the cooling rate effect due to the time dependent nature of nucleation, although this effect varies among ice nucleating materials and with particle surface area (Broadley et al., 2012; Murray et al., 2011, 2012; Wright et al., 2013). Potential contributions to these differences from temperature uncertainties cannot be excluded.”

Line280 in the main text: “Note that the derived J_{het} values reported in this study correspond to a cooling rate of 10 K min⁻¹; therefore, comparisons with studies using different cooling rates should be made with caution.”

Line19 in the Supplementary Information, Text S1: “Temperature calibration and freezing temperature at different heating/cooling rates.

Following Budke and Koop (2015), temperature calibration was performed using the melting points of decane (243.45 K) and dodecane (263.55 K) (Knopf and Lopez, 2009) at heating rates of 0.5–10 K min⁻¹. These reference temperatures span much of the freezing temperature range investigated in this study (245–265 K). Calibration was conducted in heating mode because the thermal lag of an organic crystal above its melting temperature is usually negligible (Budke and Koop, 2015). In contrast, using the temperature of liquid to solid phase transition (e.g, freezing of water droplets) in cooling mode for calibration will lead to supercooling of a liquid. In addition to the variability of freezing temperature due to the stochastic nature of crystallization, even minor supercooling and the stochastic nature would bias a calibration in cooling mode. The reference substances were deposited on hydrophobic glass slides prepared in the same manner as those used for the droplet freezing experiments. The measured melting temperatures were compared with literature values to establish the calibration, which was then applied to the raw freezing temperatures. Applying the heating mode calibration to the cooling mode freezing measurements assumes comparable thermal lag during heating and cooling at the same temperature change rate. At a heating rate of 10 K min⁻¹, the deviations from the literature melting points were approximately 2.1 K for decane and 1.6 K for dodecane (Fig. S2). The absolute difference between the calibrated temperatures and the literature values was determined for individual calibration. We reported a conservative temperature uncertainty of ± 0.5 K as the higher values of standard deviations of these absolute differences at 243 and 263 K, as shown as error bars in Fig. S2 which is smaller than 0.5 K at 243 K and 0.3 K at 263 K.

We conducted additional freezing experiments using water droplets and Tengger Desert dust suspensions at different cooling rates. T_{50} showed little variation at cooling rate between 0.5 and 2 K min⁻¹, whereas the difference in T_{50} between 10 and 1 K min⁻¹ was +1.1 K at 255 K and -0.9 K at 245 K (Fig. S3). Previous studies have demonstrated that immersion freezing can exhibit cooling rate dependence due to the time dependent nature of nucleation, although the magnitude of this effect varies among ice nucleating materials and with particle surface area (Broadley et al., 2012; Wright et al., 2013). Studies of kaolinite containing droplets reported an approximately 2 K decrease in freezing temperature when the cooling rate increased from 1 to 10 K min⁻¹, whereas smaller shifts have been reported for pure water droplets (Murray et al., 2011, 2012). Taken together, these studies indicate that the approximately 1 K difference observed here is consistent with previously reported cooling rate effects, although it cannot be attributed exclusively to time dependent nucleation, and potential contributions from temperature uncertainties cannot be excluded.”

4. *Section 4: The proposed AS-dependent parameterization is potentially useful, but it is derived from one surface dust sample and one aqueous treatment protocol. The atmospheric example is illustrative, but it gives the impression that the parameterization can already be directly implemented for internally mixed atmospheric dust. I suggest describing this as an experimentally constrained sensitivity parameterization rather than a generally applicable atmospheric aging parameterization.*

Response: Thank you for this important comment. We agree that the proposed parameterizations are constrained by experiments using a single dust sample from the Tengger Desert, which should be interpreted as experimentally constrained parameterizations. We have

revised the Abstract, Section 4 title, relevant texts, and Table 2 caption to clarify this limitation. We emphasize that our study provides an illustration of potential type of formulation for ice nucleation parameterization, rather than a generally applicable parameterization for internally mixed atmospheric dust with AS. We also emphasize that further validation using dust from different sources and additional aging conditions is needed before broader implementation in cloud models.

Revised text:

Line35 in the Abstract: “We developed experimentally constrained sensitivity parameterizations of N_s and J_{het} for Tengger Desert dust with and without aging, which may be used in cloud modeling to study how atmospheric aging modifies the ice nucleation of natural dust.”.

Line576 in the main text: We changed the title for Section 4, “Atmospheric implication” to “Atmospheric implication: impact of AS on IMF by Tengger Desert dust”.

Line593 in the main text: “We applied the proposed parameterizations for Tengger Desert dust internally mixed with AS to estimate their N_s and J_{het} under representative atmospheric conditions.”.

Line 618 in the main text: We changed the caption for Table 2 to “The k , a , b , c , and d values for parameterizations of $N_s(T, r)$, $J_{het}(T, r)$, and $J_{het}(\Delta a_w, r)$ presented in Eqs.8-10 for Tengger Desert dust mixed with AS.”.

Line666 in the main text: “These parameterizations illustrate how experimentally derived changes in dust ice nucleation associated with AS aging can be represented in a model framework. However, they are constrained by experiments using a single dust source and aging protocol, and this study primarily investigated the effects of inorganic aging on the IMF activity. Future studies should investigate the effects of organic compounds and other atmospheric aging processes on dust ice nucleation. Further experiments and ice nucleation parameterizations involving dust from different sources and a wider range of atmospheric aging conditions are needed to evaluate their broader applicability in cloud models.”

Specific comments:

1. *Methods state 95 °C for 30 min, while it states 90 °C for 30 min in Line 496. Please correct this.*

Response: Thank you for pointing this out. It was 95 °C. We corrected the statement.

2. *Section 3.5.4: The present heat treatment is very different from 523 K for 4–5 h (Zolles et al., 2015). These treatments may affect different components. Therefore, the comparison with Zolles et al. needs care.*

Response: We agree with you that different heating treatments may impact on the ice nucleation ability of different components and that a direct comparison should be made with caution. We revised the statement to clarify that the comparison is intended only to indicate a qualitative consistency in the response of mineral dust to heat treatment. We retained the comparison with Zolles et al (2015) because they provide relevant data on the immersion freezing activity of several relevant mineral dust components after heat treatments. In addition, we removed the previous comparison with Zolles et al (2015) in Section 3.5.4 to avoid overextending the comparison.

Revised text:

Line439 in the main text: “The impact of heat treatment on Us-dust was qualitatively consistent with the results from Zolles et al (2015), who reported marginal change in the IMF ability of K-feldspar, Na/Ca-feldspar, Kaolinite, and ATD after heating at 523 K for 4-5 h, with the ΔT_{50}^u values remaining within their measurement uncertainty.”

Line533 in the main text: “Niedermeier et al. (2011) reported that the IMF activity of ATD remained unchanged after heating.”

3. *Line 141: Please clarify what is meant by “ultrapure water” here. It would be helpful to provide the water source/system and relevant water-quality information.*

Response: Thank you for pointing this out. We add this information in the text.

Revised text: Line141 in the main text: “A weighed dust aliquot was mixed with 20 mL ultrapure water (Millipore, resistivity > 18.2 M Ω cm) in a glass vial and manually shaken for 1-2 minutes to ensure thorough mixing.”

4. *Line 425: Typo “felspar”.*

Response: We corrected the typo.

Referee #2

Authors have investigated immersion freezing (IMF) of surface-collected dust from Tengger Desert, China, and examined the effects of internal mixing with ammonium sulfate (AS), NaCl, H₂SO₄, and HNO₃, as well as heat treatment. The effects of increase in dust mass fraction on Median freezing temperature (T₅₀) of Tengger Desert dust have been shown while no effect of NaCl and acids (H₂SO₄ and HNO₃), as well as heat treatment in terms of T₅₀, on the overall IMF activity, was noted.

Limited understanding on impacts of chemical aging on ice nucleation for airborne dust is available. The experimentally constrained parameterizations of N_s and J_{het} for Tengger Desert dust with and without aging presented in this study may be used in cloud modeling to further understand that how atmospheric aging modifies the ice nucleation of natural dust. Overall, the work is extensive and may be accepted after minor revision. Following are the specific comments:

1. The whole manuscript revolves around comparison of IMF characteristics in normal dust and sonicated dust. Specific details of sonication treatment (temperature at which ultrasonication was done, make and model of ultrasonicator used for the purpose etc) may be provided. Further authors may explain that why 30 min sonication is preferred.

Response: We thank the reviewer for this helpful suggestion. We added details of the sonication treatment, including the ultrasonicator model and manufacturer, as well as the treatment temperature. The 30 min sonication duration was selected following the approach described by Knopf and Alpert (2013) to facilitate the dispersion of fine dust particles in the 20 mL suspension. The corresponding details were added to the revised manuscript.

Revised text: Line144 in the main text: “A second suspension was prepared using the same procedure but subjected to ultrasonic treatment at 40 kHz for 30 min at 25 °C in an ultrasonic bath (KQ2200DA, Kunshan Ultrasonic Instrument Ltd.). This sample is hereafter referred to as ultrasonicated dust (Us-dust). The sonication duration was selected based on Knopf and Alpert (2013) to reduce particle aggregation and promote the dispersion of fine dust particles in the 20 mL suspension.”

2. Why cooling rate of $10 \text{ K} \cdot \text{min}^{-1}$ has been used? Appropriate reference for this may be provided. What is the advantage of doing this instead of usual $2 \text{ K} \cdot \text{min}^{-1}$ or $1 \text{ K} \cdot \text{min}^{-1}$

Response: A cooling rate of $10 \text{ K} \cdot \text{min}^{-1}$ was selected based on previous studies (Knopf and Lopez, 2009; Koop et al., 1998) and to reduce the time available for water vapor transfer between supercooled droplets and ice crystals (frozen droplets), thereby limiting potential interference from the Bergeron–Wegener–Findeisen process. We conducted additional experiments to evaluate the thermal lags and the influence on freezing temperature at different cooling rates. Please see the details in the response to the similar concern, Comment #3 raised by Reviewer #1. We revised the manuscript accordingly.

3. L359: Increase in AS concentration and activation of the ice nucleation sites needs more explanation.

Response: We thank you for this helpful comment. We agree that this statement requires further explanation. We had briefly mentioned that the underlying mechanism will be discussed in Section 3.5.1. As discussed in Section 3.5.1, previous studies have suggested that interactions between ammonium ions (NH_4^+) and dust surfaces may enhance the IMF ability of dust. Based on our results, we propose that increasing AS concentrations progressively promote the activation of ice nucleation sites on the dust surface, resulting in the observed sigmoidal response of ΔT_{50}^u . At relatively low AS concentrations, only a small fraction of susceptible sites may be activated. As the AS concentration increases, additional sites may become active, leading to a more pronounced enhancement in ice nucleation activity. At higher AS concentrations, the enhancement approaches a plateau, potentially because most of the susceptible sites have been activated or because the relevant surface interactions approach saturation. We have revised the discussion in the main text to clarify this proposed interpretation and its potential underlying mechanism.

Revised text: Line385 in the main text: “Previous studies have suggested that the interaction between dust and ammonium ions (NH_4^+) can enhance the IMF ability of dust (Anim-Danso et al., 2016; Kumar et al., 2018; Whale et al., 2018; Worthy et al., 2021). The observed sigmoidal response of ΔT_{50}^u suggests a concentration dependent activation of ice nucleation sites on dust surface upon AS exposure. At relatively low AS concentration down to $5 \times 10^{-5} \text{ M}$, the enhancement was modest, with a ΔT_{50}^u of 0.4 K , suggesting that only a small fraction of susceptible ice nucleation sites was activated. As the AS concentration increased to $5 \times 10^{-3} \text{ M}$, ΔT_{50}^u increased markedly to 4.8 K , consistent with a substantial activation of additional ice nucleation sites. At a higher AS concentration of $5 \times 10^{-2} \text{ M}$, ΔT_{50}^u remained nearly constant, suggesting that the enhancement had approached a plateau. One possible explanation is that most of the susceptible ice nucleation sites had already been activated, potentially reflecting saturation of the relevant surface interactions involving NH_4^+ . This interpretation is consistent with the proposed role of ammonium–dust interactions in enhancing ice nucleation, although the underlying molecular mechanism remains to be confirmed. Further discussion is provided in Section 3.5.1.”

4. L437: Pl see usage of potential in sentence- NH_4^+ adsorbed on dust surfaces potential can alter the arrangement.

Response: Thank you for your correction. We revised the sentence which also response to the comment #2 from Reviewer #1.

Revised text: Line475 in the main text: “In addition, NH_4^+ adsorbed on dust surfaces may alter the arrangement and orientation of water molecules, potentially enhancing ice nucleation activity (Anim-Danso et al., 2016; Kumar et al., 2018; Worthy et al., 2021).”

5. Why most effects of acid treatments have been reported on Us-dust only and not on S-dust?

Response: We thank you for this comment. The experimental design of this study primarily focused on investigating the potential mechanisms by which aging treatments affect the ice nucleation activity of dust. Therefore, we initially selected a single dust sample, Us-dust, for the aging experiments rather than conducting parallel experiments on both S-dust and Us-dust. This choice was based on our initial expectation that, because both samples originated from the same source, their responses to aging treatments might be broadly similar. However, we acknowledge that this assumption has not been experimentally verified. We clarified this limitation in the revised manuscript and acknowledge that further experiments are needed to determine the responses to aging for dust from different sources.

Revised text: Line671 in the main text: “Further experiments and ice nucleation parameterizations involving dust from different sources and a wider range of atmospheric aging conditions are needed to evaluate their broader applicability in cloud models.”

6. Heat treatment is the most common step for IMF study. Authors may elaborate upon the section pertaining to the effects of heat treatment. Heat treatment effect on S-dust is missing. Further, for Us-dust, loss of ice active sites at warm temperatures and change in T50 is reported while no discussion on change in profile has been included.

Response: Thank you for this valuable suggestion. Heat treatment is commonly used to investigate thermal sensitive ice nucleating components, including potential biological and organic materials. In this study, heating Us-dust reduced N_s at warmer temperatures, suggesting the deactivation of thermally labile ice active sites. We expanded the discussion to address this thermal response and the potential roles of intrinsic dust properties and heat-sensitive components. Heat treatment experiments were not conducted on S-dust because both samples originated from the same dust material, and similar responses were initially expected. However, this assumption remains unverified, and further experiments are needed to determine whether sonication affects the thermal sensitivity of dust INPs. This limitation is now acknowledged in the revised manuscript.

Revised text: Line528 in the main text: “In this study, heat treatment was used to examine changes in IMF activity and the potential thermal deactivation of labile ice active sites.”

Revised text: Line537 in the main text: “In our study, heat treatment on Us-dust at 95°C for 30 min reduced ice nucleation activity at warm temperatures, whereas activity at lower temperatures was largely retained. This response is consistent with Perkins et al. (2020), suggesting that ice active sites in complex dust samples differ in thermal sensitivity. The preferential loss of activity at warmer temperatures suggests the deactivation of thermally labile ice active sites, which may be intrinsic to the mineral dust itself or associated with heat sensitive components, including biological materials or organic matter. However, the specific origins of these labile ice active sites were not determined here and require further investigation. Heat treatment experiments were not conducted on S-dust; therefore, whether sonication influences the thermal sensitivity of dust INPs remains uncertain.”

7. Aging is a very wide process which includes internal mixing /coating with organic as well as inorganic components. However, the focus of this manuscript is on inorganic processes only but authors may add a section stating limitation and may be counter effect of aging where organics are involved.

Response: Thank you for this important comment. We agree that atmospheric aging encompasses a wide range of processes. Organic coatings may also modify the ice nucleating properties of dust, including potentially suppressing IMF activity. To clarify the scope and limitations of our study, we have added a discussion highlighting the need to investigate the effects of aging by organics under atmospherically relevant conditions.

Revised text: Line668 in the main text: “However, they are constrained by experiments using a single dust source and aging protocol, and this study primarily investigated the effects of inorganic aging on the IMF activity. Future studies should investigate the effects of organic compounds and other atmospheric aging processes on dust ice nucleation. Further experiments and ice nucleation parameterizations involving dust from different sources and a wider range of atmospheric aging conditions are needed to evaluate their broader applicability in cloud models.”.

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