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Immersion freezing of surface dust particles from Tengger Desert in China: effects of mineral composition and aging treatments on ice nucleating activity

Jiao Xue[1], Shengkai Wang[1], Haoyu Qi[1], Yao Yao[1], Shulan Chen[1], Juying Lu[1], Tian Zhang[1], Qi Wang[2], Defeng Zhao[3], Youwei Hong[4], Yihua Cai[1], Bingbing Wang[1,*]

[1]State Key Laboratory of Marine Environmental Science, College of Ocean and Earth Sciences, Xiamen University, Xiamen, China

[2]China-ASEAN College of Marine Sciences, Xiamen University Malaysia, Selangor Darul Ehsan, Malaysia

[3]Shanghai Key Laboratory of Ocean-land-atmosphere Boundary Dynamics and Climate Change, Fudan University, Shanghai, China

[4]School of Resource and Environmental Science, Quanzhou Normal University, Quanzhou, China

* *Corresponding authors:* Bingbing Wang (Bingbing.Wang@xmu.edu.cn)

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Abstract

Mineral dust is a major source of ice nucleating particles (INPs) for mixed-phase clouds and thereby influences cloud properties, precipitation, and radiative forcing. However, the ice nucleation properties of natural dust and their response to atmospheric aging remain poorly constrained. Here, we investigated immersion freezing (IMF) of surface-collected dust from Tengger Desert, China, and examined the effects of internal mixing with ammonium sulfate (AS), NaCl, H_2SO_4, and HNO_3, as well as heat treatment. Median freezing temperature (T_{50}) of Tengger Desert dust increased from 251.6 K to 256.7 K as dust mass fraction increased from 1.4×10^{-4} wt% to 1.0×10^{-1} wt%. A minor increase in feldspar content (~2% in surface area) quantified by single particle analysis enhanced the ice nucleation active site density (N_s) by approximately one order of magnitude. Internal mixing with AS enhanced IMF activity, with the enhancements of T_{50} (0.4 K - 4.8 K), N_s, and heterogeneous ice nucleation rate coefficient (J_{het}) exhibiting sigmoidal response to AS concentration from 5×10^{-5} M to 5×10^{-2} M. This enhancement is attributed to NH_4^+ adsorption on dust surface. NaCl and acids (H_2SO_4 and HNO_3), as well as heat treatment, had no impact on the overall IMF activity in terms of T_{50}, although modest changes in N_s and J_{het} were observed above 256 K. We developed the experimentally constrained parameterizations of N_s and J_{het} for Tengger Desert dust with and without aging, which can be used in cloud modeling to improve our understanding of how atmospheric aging modifies the ice nucleation of natural dust.

Keywords: Ice nucleation, single particle analysis, elemental composition, atmospheric aging, heat treatment

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1. Introduction

Ice crystal formation is a fundamental atmospheric process that substantially modulate the microphysical properties and lifetime of cold clouds, precipitation, the Earth's radiation balance, and climate (Baker, 1997; Knopf and Alpert, 2023; Lohmann and Feichter, 2005; Rosenfeld et al., 2014; Storelvmo, 2017). Ice crystals may form via homogeneous or heterogeneous nucleation. Ice nucleating particles (INPs) reduce the free energy barrier for ice embryo formation and facilitate heterogeneous nucleation at lower relative humidity (RH) and higher temperatures than required for homogeneous nucleation (Pruppacher and Klett, 1997). Heterogeneous ice nucleation includes various pathways, including deposition nucleation, immersion freezing (IMF), condensation freezing, and contact freezing (Knopf et al., 2018; Ladino Moreno et al., 2013; Pruppacher and Klett, 1997; Vali et al., 2015). IMF is considered as the dominant heterogeneous nucleation mechanism in mixed-phase clouds (Kanji et al., 2017; Murray et al., 2012). Improved quantification of the IMF ability of diverse INP types is therefore critical for improving ice nucleation parameterizations and enhancing representations of aerosol-cloud interactions in cloud and climate models.

Mineral dust is a major INP class and contributes significantly to ice nucleation in mixed-phase clouds (Chen et al., 2024; DeMott et al., 2015; Hoose and Möhler, 2012; Kanji et al., 2017; Kawai et al., 2021; Ladino Moreno et al., 2013; Murray et al., 2012; Shi and Liu, 2019; Ullrich et al., 2017). Global model simulations indicate that the INP to dust mass ratio varies by an order of magnitude across dust source regions (Kawai and Matsui, 2025). Kawai et al. (2021) demonstrated that Asian dust contributes only 3.4% to global annual dust mass, yet accounts for more than 15% of global dust INPs. This disproportionate contribution is attributed to the long-range transport of Asian dust to the middle and upper troposphere for mixed-phase cloud formation. Despite this climatic implications, investigations on the ice nucleation properties of Asian dust are limited and often focus on particles collected during dust storms in urban environments. For instance, Zhao et al. (2024) found that dust from the 2021 Asian dust storms can

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trigger IMF between 230 K and 250 K under water subsaturated conditions. Ice residual analysis conducted by Iwata and Matsuki (2018) showed that clay minerals dominated the ice active fraction above 243 K, whereas atmospheric aging (e.g., internal mixing with sea salt or nitrate coatings) suppressed ice nucleation. Chen et al. (2021) reported differences in IMF ability of particles during East Asian dust storms above 248 K linked to different desert regions and transport pathways. Li et al (2022) showed that surface dust/salt mixed particles from Chinese deserts can trigger deposition nucleation at temperatures as low as 223 K. Detailed investigations of IMF characteristics of Asian dust from source regions and the controlling factors are therefore required to accurately quantify their impacts on cloud formation.

Several studies have characterized the ice nucleating properties of natural dust from various source regions (Boose et al., 2016; Kaufmann et al., 2016; Perkins et al., 2020). Kaufmann et al. (2016) conducted ice nucleation measurement on dust from source regions on four continents and compared results with reference minerals, finding that natural dust froze within a narrow temperature range and that mineralogical composition can largely explained the freezing behavior for most samples. Boose et al. (2016) reported that the IMF ability of dust from nine typical deserts correlates with K-feldspar content at 253 K and with the abundance of feldspar and quartz between 238 K and 245K, whereas high clay content suppress IMF ability. Feldspar, particularly K-rich feldspar, is consistently identified as a highly ice active component that plays an important role in the freezing behavior of natural dust at high temperatures (e.g., Atkinson et al., 2013; Harrison et al., 2016). Nevertheless, investigations on ice nucleation of natural dust from deserts remain limited relative to work on laboratory surrogate and monomineralic dust standards. Furthermore, the quantitative contributions by individual mineral components to ice nucleation activity of natural dust are not yet fully resolved.

Dust particles undergo atmospheric aging during long-range transport, including mixing with organics/inorganics and heterogeneous chemical reactions, which may

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alter their intrinsic ice nucleation activity (Cziczo et al., 2013; DeMott et al., 2003; Eastwood et al., 2009; Kanji et al., 2013; Kulkarni et al., 2015; Möhler et al., 2008; Sullivan et al., 2010a, b; Tobo et al., 2012). Addition of soluble species (e.g., AS and NaCl) lowers water activity of aqueous droplets and thereby depresses freezing temperatures (Koop et al., 2000; Koop and Zobrist, 2009; Rigg et al., 2013; Zobrist et al., 2008). Emerging evidence indicates that low concentrations of certain solutes can modify the freezing temperature of dust via chemical interactions with ice active mineral components (Kumar et al., 2018, 2019a, b; Reischel and Vali, 1975; Whale et al., 2018; Wilson and Haymet, 2009; Worthy et al., 2021; Yun et al., 2021). For example, Whale et al. (2018) observed that ammonium salts increase the freezing temperature of feldspar and quartz, whereas NaCl reduces it. Similarly, Worthy et al. (2021) found that AS increases the freezing temperature of K-rich feldspar, kaolinite, montmorillonite, and Arizona Test Dust (ATD), which is attributed to NH_4^+/K^+ exchange and NH_4^+ adsorption that change interfacial water structure facilitating ice nucleation. In contrast, acid treatments diminish the IMF ability of feldspar, which is likely associated with H_3O^+/K^+ exchange and incongruent dissolution (Yun et al., 2021). Different aging treatments (acidic and salt solutions, peroxide, heating) have distinct effects on ice nucleation activity of ATD, underscoring the complexity of atmospheric processes (Niedermeier et al., 2011; Perkins et al., 2020).

Current studies have revealed diverse aging effects on the IMF ability of monomineralic particles and laboratory dust surrogates. Because of complex compositions of natural dust, results from simplified standards cannot be directly extrapolated to atmospheric dust. Consequently, the effects and mechanisms by which atmospheric aging alters the IMF of natural dust remain poorly constrained. Most aerosol-aware ice nucleation parameterizations generally distinguish broad particle types and relate freezing to temperature, humidity, particle abundance, or particle surface area (Burrows et al., 2022). However, they rarely represent atmospheric aging explicitly in regional and global models; when included, aging is treated with highly simplified assumptions.

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Improved process-level constraints are therefore required to develop and evaluate the aging dependent ice nucleation parameterizations.

This study investigates the intrinsic IMF ability of natural dust from Tengger Desert and its response to different aging processes. We investigated the impacts on the IMF activity of surface-collected dust after heat treatment and mixing with AS, NaCl, H_2SO_4, and HNO_3. Single particle analysis provided size-resolved elemental compositions and quantified the contribution of mineral components. We quantify changes in IMF ability of dust induced by each aging treatment, discuss plausible mechanisms, and propose corresponding ice nucleation parameterizations with potential atmospheric implication.

2 Experimental methods

2.1 Sample collection and preparation

Surface dust particles were collected from the Tengger Desert, China (37.57°N,104.93°E) (Fig. 1). The experimental procedure is summarized in Fig. 2. Dust particles was sieved (200 mesh, 74 μm, nylon) and used for suspension preparation. A weighed dust aliquot was mixed with 20 mL ultrapure water in a glass vial and manually shaken for 1-2 minutes to ensure thorough mixing. After standing for 5 min to allow coarse particles to settle, the upper 10 mL was collected as the first dust suspension (hereafter referred as S-dust). A second suspension was prepared identically but included 30 min of ultrasonic treatment to enhance dispersion of fine particles (hereafter referred as Us-dust). Both suspensions were serially diluted with ultrapure water and used immediately for ice nucleation experiments. To determine dust mass concentration, 6 mL of each suspension was filtered using 0.2 μm polycarbonate filter membrane (GTTP04700, Isopore^{TM}) and the insoluble part was weighed. Ultrasonic treatment increased the dust mass concentration in suspension by about 5 times. The following suspensions were used. For S-dust, the mass concentrations were 2.0×10^{-2} wt%, 3.0×10^{-3} wt%, and 7.0×10^{-4} wt%. The mass concentrations for Us-dust were 1.0×10^{-1} wt%, 1.87×10^{-2} wt%, 3.5×10^{-3} wt%, 7.0×10^{-4} wt%, and 1.4×10^{-4} wt%.

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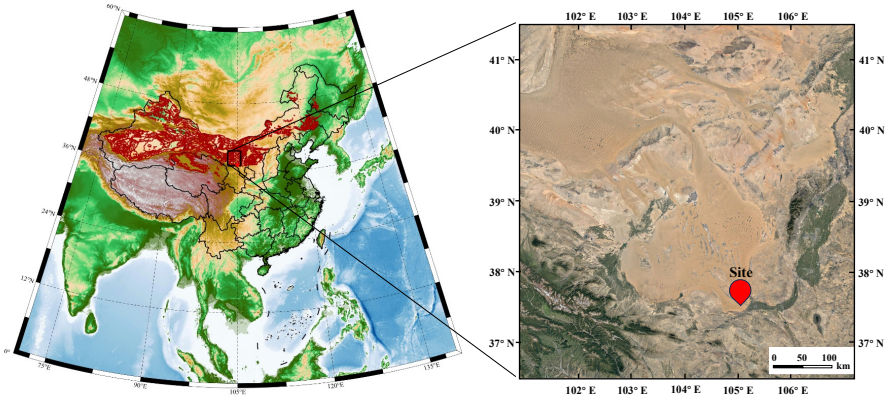

Figure 1. (a) Deserts in northern China (red shading) and the location of Tengger desert (black rectangle). (b) Sampling site (red dot) in Tengger Desert (Map data © 2026 Google).

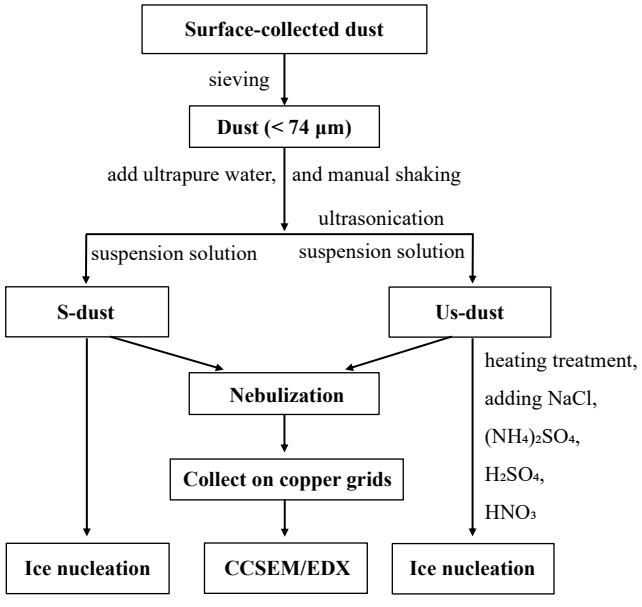

Figure 2. Schematic diagram of experimental procedures.

2.2 Aging treatments on Us-dust

All aging treatments used the Us-dust suspension with dust mass concentration of 1.87×10^{-2} wt%. We added AS, NaCl, HNO_3, and H_2SO_4 to the Us-dust suspension for

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12 hours prior to ice nucleation experiments. Final AS concentrations (99.95%, Sigma) were 5×10^{-5} M, 5×10^{-4} M, 5×10^{-3} M, 2×10^{-3} M, 1×10^{-3} M, and 5×10^{-2} M. Final NaCl concentrations (99.995%, Sigma) were 5×10^{-4} M, 5×10^{-3} M, 5×10^{-2} M. Final H_2SO_4 concentrations (99.7%, Hushi) were 2.5×10^{-4} M, 2.5×10^{-3} M, 2.5×10^{-2} M. Final HNO_3 concentrations (99.7%, Hushi) were 5×10^{-4} M, 5×10^{-3} M, 5×10^{-2} M. Heat treatment was conducted on the Us-dust suspension at 95 °C for 30 min.

2.3 Size-resolved composition of dust particles

Single particle analysis was conducted by the computer-controlled scanning electron microscopy equipped with an energy dispersive X-ray spectroscopy (CCSEM/EDX, Quanta 650, FEI Inc.; Genesis, EDAX Inc.) to determine particle size and elemental composition. Dust particles were collected from the suspensions following the method from our previous work (Xue et al., 2024; Zhao et al., 2024). Briefly, the suspensions were nebulized, carried by high-purity nitrogen through a diffusion dryer, and collected onto TEM copper grids (01814-F, Ted Pella) using an impactor (SKC, Inc.). CCSEM/EDX analysis at the working voltage of 20 kV followed the established method from our previous work (Knopf et al., 2014; Laskin et al., 2002; Wang et al., 2012; Xue et al., 2024; Zhao et al., 2024) and is described briefly here. The elemental composition and the area equipment dimeter (calculated by the 2-D projected particle area, hereafter referred as particle size) were quantified for about 1040 S-dust and 1180 Us-dust particles. We obtained atomic percentages for C, N, O, Na, Mg, Al, Si, P, Si, Cl, K, Ca, and Fe. C, N, and O determined by EDX were semiquantitative and excluded from classification. For each particle, the normalized atomic percentage of element X was calculated as $[X] = \frac{X}{Na+Mg+Al+Si+K+Ca+Fe+P+S+Cl}$. Based on the elemental composition, particles were grouped into five types, namely illite, kaolinite, chlorite, quartz, and feldspar, using a rule-based classification scheme (Fig. 3). This method has been widely used to identify different dust types based on the elemental fingerprints of monomineralic dust (Hu et al., 2022; Kandler et al., 2007; Wang et al., 2022; Zhao et al., 2024). Mean elemental compositions of each dust type were shown in Fig. S1.

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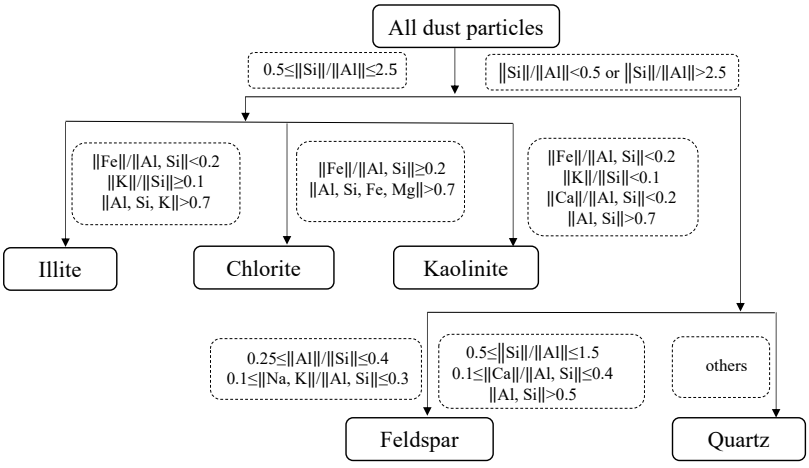

Figure 3. Particle classification scheme based on the relative atomic percentages of elements obtained from CCSEM/EDX analysis of individual particles.

2.4 Immersion freezing experiments

IMF experiments were performed using a temperature-controlled cooling stage coupled to an optical microscope, following the method described by Knopf and Forrester (2011) and Yao et al. (2022). About 20-25 of 1 µL droplets (dust suspensions or ultrapure water) were pipetted onto a hydrophobic glass slide, covered with a second hydrophobic glass slide and a custom copper spacer, and sealed with vacuum grease to isolate the droplets from ambient environment. The sample cell was placed on the cooling stage for subsequent freezing experiment. Sample temperature was regulated by circulating liquid nitrogen through the cooling stage and a heating foil under feedback from a temperature controller. Droplet temperature was calibrated against the melting points of decane, dodecane, and ice. A conservative temperature uncertainty was estimated to be less than ± 0.5 K. Freezing experiment was conducted at 10 $K \cdot min^{-1}$ while a digital camera recorded the freezing process at one picture per second. At least three independent samples were measured, each with more than three repeated runs, yielding about 200 – 270 freezing events per droplet type. Freezing temperature of each droplet was determined by analyzing the recorded images. Ultrapure water droplets (resistivity > 18.2 MΩ cm) served as background. The freezing temperatures were corrected for

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freezing point depression induced by solutes (ΔT_f), which was calculated as follows:

$$\Delta T_f = i \times K_f \times m_{solute}, \quad (1)$$

where i is van't Hoff factor, K_f is cryoscopic constant (1.86 K kg mol^{-1}), and m_{solute} is solute molality (Atkins and de Paula, 2014; Worthy et al., 2021).

2.5 Frozen fraction and ice nucleating active site density

Frozen fraction ($f_{ice}(T)$) is defined as the ratio of the number of frozen droplets (N_{ice}) at a certain temperature (T) to the total number of droplets (N_{total}), i.e., $f_{ice}(T) = N_{ice}/N_{total}$. Frozen fraction reflects the distribution of ice formation event at different temperatures. Ice nucleation active site density (N_s) is the number of ice active site per unit particle surface area and is calculated using Eq. (2) according to Vali (1971):

$$N_s = \frac{-\ln\left(1 - f_{ice}(T)\right)}{A} \quad [m^{-2}] \quad (2)$$

where A is the total particle surface area immersed in a single droplet. A was calculated by the particle mass in the droplet and the average surface area per unit particle mass (A_m). Particle mass in each droplet equals the dust mass concentration in suspension times the droplet volume (1 µL). A_m was estimated from CCSEM/EDX particle data by

$$A_m = \frac{\sum S_i}{\sum S_i \times \rho_i} \quad (3)$$

where S_i is the surface area of the individual particle i (assumed spherical, $S_i = \pi {d_i}^2$); d_i is the diameter of the particle i; ρ_i is the density of different particle types, specifically, ρ_{quartz} = 2.62 g/cm^{-3}, $\rho_{chlorite}$ = 2.42 g/cm^{-3}, ρ_{illite} = 2.6 g/cm^{-3}, $\rho_{kaolinite}$ = 2.6 g/cm^{-3}, and $\rho_{feldspar}$ = 2.6 g/cm^{-3} (Menut et al., 2020). Uncertainty in N_s (95% confidence intervals) was calculated using Poisson nucleation statistics (Koop et al., 1997).

2.6 Heterogeneous ice nucleation rate coefficient (J_{het})

Experimental data were used to derive J_{het}, providing a time-dependent description of heterogeneous nucleation. J_{het} at temperature T^i for a given temperature interval (ΔT) was calculated based on the method from previous studies (Alpert et al., 2011; Koop et al., 1997; Zobrist et al., 2007):

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$$J_{\text{het}}(T^i) = \frac{N^i_{\text{ice}}}{t^i_{\text{tot}} \times A^i} \quad (4)$$

where N^i_{ice} is the total number of ice nucleation events in the i^{th} temperature interval, and $t^i_{\text{tot}} \times A^i$ is product of observation time and dust surface area available for nucleation in that interval. $t^i_{\text{tot}} \times A^i$ is given by

$$t^i_{\text{tot}} \times A^i = \sum_{j=1}^{N^i_{\text{ice}}} \frac{\left(T^i_{\text{st}} - T^i_{\text{ice},j}\right)}{c_{\text{r}}} A^i_{liq,j} + \frac{\Delta T}{c_{\text{r}}} A^i_{liq} \quad (5)$$

where T^i_{st} is the start temperature of i^{th} temperature interval; $T^i_{\text{ice},j}$ and $A^i_{liq,j}$ are the freezing temperature and dust surface area of the j^{th} droplet that froze within the interval; A^i_{liq} is the total dust surface area in the droplets remaining liquid at the end of the interval; c_{r} is the cooling rate (10 K min^{-1}); ΔT of 0.5 K is used.

The experimentally derived J_{het} was parameterized using the water activity based framework which relates the droplet freezing temperature uniquely to droplet water activity (Alpert et al., 2022; Knopf and Alpert, 2013; Xue et al., 2024; Zhao et al., 2024). It has been applied to both homogeneous and heterogeneous ice nucleation (China et al., 2017; Knopf and Alpert, 2013; Koop and Zobrist, 2009). In this framework, Δa_{w} is used to describe the freezing ability of droplets, which represents the deviation of the water activity of droplets at freezing temperature from the water activity at the ice-melting equilibrium state at the same temperature (Koop et al., 2000). Δa_{w} was calculated for each droplet at its freezing temperature. The corresponding J_{het} was parameterized by a liner regression of log (J_{het}) against Δa_{w} (or temperature).

3 Results and discussion

3.1 Characteristics of Tengger Desert dust at single particle level

Figure 4 shows the particle classification for the S-dust and Us-dust samples. Particles were mainly in the size range of 0.2-2 μm. S-dust and Us-dust exhibited a peak size of about 0.35 μm and 0.7 μm, respectively. Size distributions of the same particle type between S-dust and Us-dust were slightly different (Fig. 4b, f). Particle number

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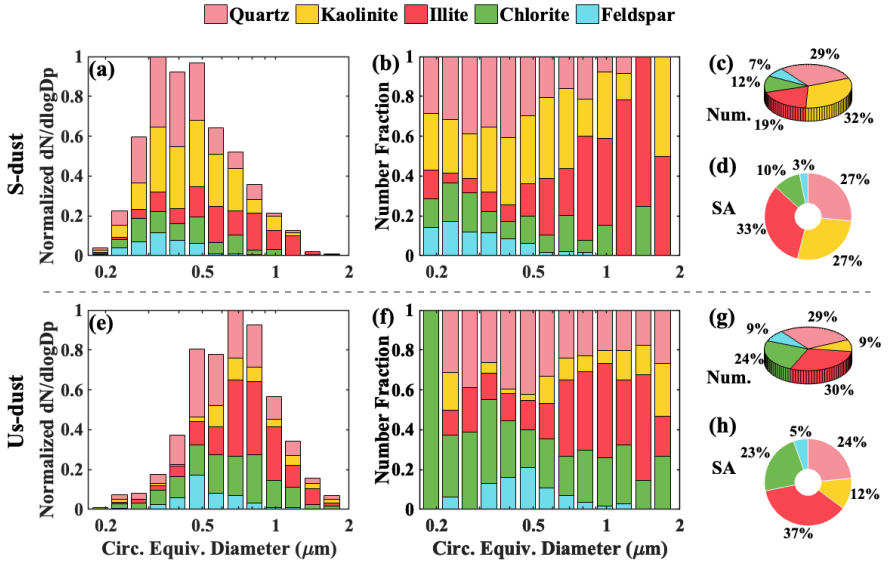
268 contribution of Us-dust by kaolinite, quartz, illite, chlorite, and feldspar was 32%, 29%,
269 19%, 12%, and 7%, respectively. Compared with S-dust, chlorite, illite, and feldspar in
270 Us-dust is about 12%, 11%, and 2% higher, respectively, whereas kaolinite decreased
271 by 23% and quartz remained unchanged (Fig. 4c, g). The contribution to particle surface
272 area of Us-dust by illite, quartz, chlorite, kaolinite, and feldspar was 37%, 24%, 23%,
273 12%, and 5%, respectively. Compared with S-dust, Us-dust showed higher
274 contributions from chlorite (13%), illite (4%), and feldspar (2%), and lower
275 contributions from quartz (3%) and kaolinite (15%) (Fig. 4d, h).



276
277 **Figure 4.** Particle size distribution and contribution of dust types for S-dust (upper panel, a-d)
278 and Us-dust (lower panel, e-h). Normalized particle size distribution (a, e); relative number
279 fraction of each particle type as a function of size (b, f); the number (c, g) and surface area (d,
280 h) contribution of each particle type.

281

282 **3.2 Ice nucleation ability of Tengger Desert dust**

283 Figure 5 summarizes IMF experimental results for ultrapure water droplets and dust-
284 containing droplets. Both S-dust and Us-dust at the investigated concentrations induced
285 IMF at higher temperatures than water droplets (Fig. 5a). T_{50} changed from 250.4 K to
286 252.7 K for S-dust as concentration increased from 7.0×10^{-4} wt% to 2.0×10^{-2} wt%. T_{50}

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changed from 251.6 K to 256.7 K for Us-dust as concentration rose from 1.4×10^{-4} wt% to 1.0×10^{-1} wt%. T_{50} shows that Us-dust nucleated ice more efficient than S-dust (Fig. 5b). At comparable dust surface area concentration, T_{50} of Us-dust was about 2 K higher than S-dust. T_{50} increased by about 1.5 K and 1.7 K per decade increase in dust surface area concentration for S-dust and Us-dust, respectively. We defined $\Delta T_{50}{}^{w}$, as the T_{50} difference between dust-containing droplets and water droplets. Figure 5c compares the $\Delta T_{50}{}^{w}$ for S-dust, Us-dust, and other monomineralic dust. $\Delta T_{50}{}^{w}$ of monomineralic dust ranks as follows: K-feldspar > Na/Ca-feldspar > quartz > illite > kaolinite > chlorite. $T_{50}{}^{w}$ for Us-dust was consistent with Na/Ca-feldspar but low than K-feldspar, while $\Delta T_{50}{}^{w}$ for S-dust was lower than Na/Ca-feldspar, but higher than chlorite and illite. We have shown that the contributions of chlorite, illite, and feldspar in both number and surface area were higher in Us-dust compared to S-dust (Fig. 4). Given that the $\Delta T_{50}{}^{w}$ of chlorite and illite are lower than U-dust, the improved IMF ability of Us-dust relative to S-dust can be primarily attributed to the increased feldspar content.

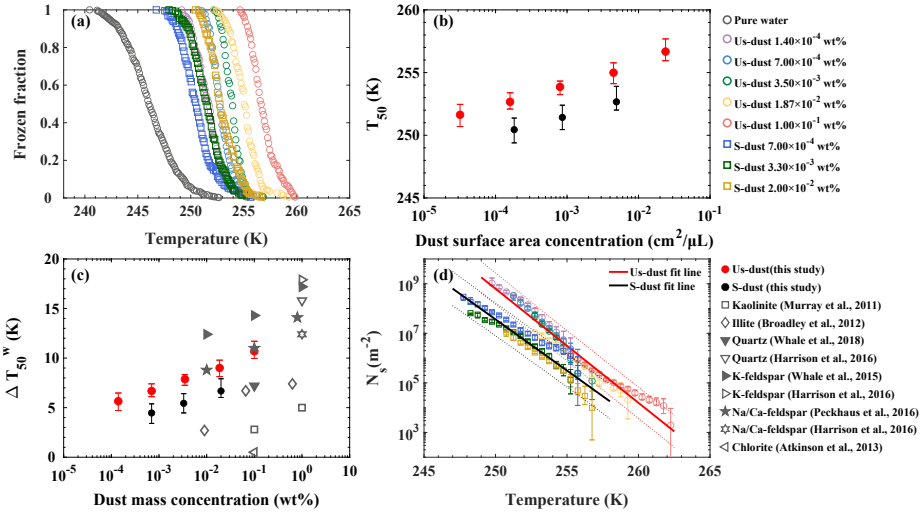

Figure 5. (a) Frozen fraction of water droplets, Us-dust and S-dust containing droplets. (b) T_{50} as a function of dust surface area concentration within a droplet. (c) $\Delta T_{50}{}^{w}$ as a function of dust mass concentration. Error bars (b, c) in temperature correspond to the 25th and 75th percentiles of freezing temperature. Gray symbols (c) represent monomineralic dust from previous studies (Atkinson et al., 2013; Broadley et al., 2012; Harrison et al., 2016; Murray et al., 2011;

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Peckhaus et al., 2016; Whale et al., 2015, 2018). (d) N_s as a function of temperature. Error bars for N_s correspond to the 95% confidence intervals. Black and red lines were fitted lines for Us-dust and S-dust, respectively. Dotted lines indicate the 95% prediction intervals.

Figure 5d shows the N_s for S-dust and Us-dust with parameterizations using the function of $log_{10}(N_s) = m_1 \times T + n_1$. The values of m_1 and n_1 are shown in Table 1. Overall, N_s for Us-dust was about one order of magnitude higher than S-dust. As shown in Fig. 6, N_s for Us-dust was comparable to feldspar, falling in between Na/Ca-feldspar and K-feldspar. S-dust was between Na/Ca-feldspar and quartz, and was distinctly higher than illite and chlorite. Previous studies has suggested that K-feldspar may enhance the ice nucleation activity (Boose et al., 2019; Harrison et al., 2016). Notably, both Us-dust and S-dust showed stronger temperature dependences of N_s than previously reported for ATD, natural dust from deserts, and airborne particles during dust storms (Chen et al., 2021; Hader et al., 2014; Niemand et al., 2012; Reicher et al., 2019; Ullrich et al., 2017). Compared to these referenced particles, Us-dust and S-dust shared similar N_s at temperatures below 255 K, but had significantly lower N_s above 255 K. N_s of Us-dust and S-dust dominated by submicron particles, especially for S-dust, consistent with submicron airborne particles collected during dust storms originated from same deserts (Chen et al., 2021) (Fig. 6b). However, N_s for Tengger desert dust was up to 3 orders of magnitude lower than the N_s parameterizations reported by Niemand et al. (2012) and Ullrich et al. (2017) at high temperatures. This observed discrepancy in N_s is likely attributed to the differences in particle size and composition of these surface-collected or airborne dust particles. Additionally, atmospheric aging on dust during long-range transport may also contribute to the discrepancy.

Table 1 The m_1 and n_1 values for the N_s parameterizations of Tengger Desert dust as a function of temperature (T), $\log(N_s) = m_1 \times T + n_1$

Samples	m_1	LCL_{m1}	UCL_{m1}	n_1	LCL_{n1}	UCL_{n1}	RMSE	R^2
Us-dust	-0.46	-0.49	-0.43	123.93	117.00	130.85	0.30	0.95
S-dust	-0.41	-0.45	-0.37	110.94	100.68	121.19	0.31	0.91

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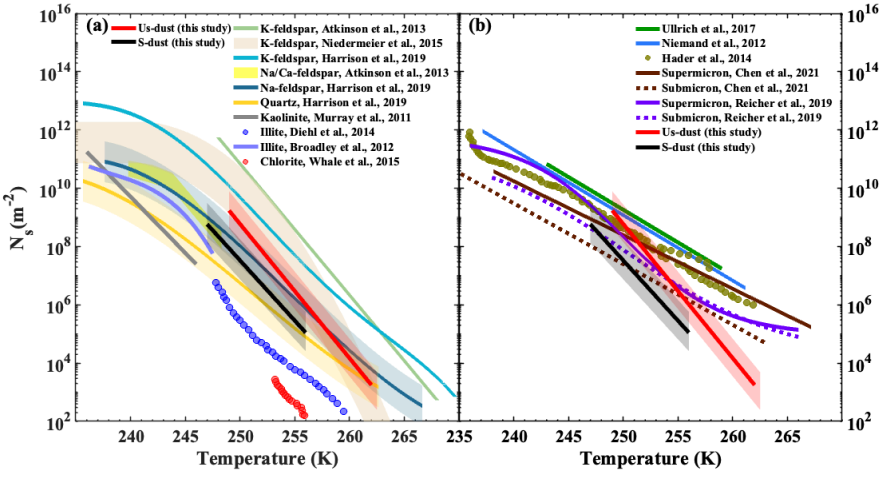

Figure 6. (a) Comparison of N_s parameterizations for Us-dust, S-dust, and monomineralic dusts from previous studies (Atkinson et al., 2013; Broadley et al., 2012; Diehl et al., 2014; Harrison et al., 2016; Murray et al., 2011; Niedermeier et al., 2011; Whale et al., 2015). Shade areas of N_s for Us-dust and S-dust are the 95% prediction intervals. (b) Comparison of N_s parameterizations for Us-dust, S-dust, ATD (Hader et al., 2014), soil samples, desert dust, and airborne dust (Chen et al., 2021; Niemand et al., 2012; Reicher et al., 2019; Ullrich et al., 2017). The detail sample information for each parameterization is shown in Table S1.

3.3 Ice nucleation ability of dust mixed with ammonium sulfate

Figure 7 presents the IMF results for Us-dust at 1.87×10^{-2} wt% after mixing with AS at various concentrations. AS enhanced the IMF ability of Us-dust, leading to higher freezing temperature and N_s as AS concentration increases (Fig. 7a, b). At AS concentration above 5×10^{-3} M, N_s was approximately two orders of magnitude larger than untreated Us-dust. We define $\Delta T_{50}{}^{u}$ as the T_{50} difference between AS treated and untreated Us-dust, using T_{50} of 254.9 K for the untreated Us-dust at 1.87×10^{-2} wt%. $\Delta T_{50}{}^{u}$ increased from 0.4 K to 4.8 K as AS concentration increased from 5×10^{-5} M to 5×10^{-2} M (Fig. 7c), following a sigmoidal response reproduced via a Boltzmann function as:

$$\Delta T_{50}^{u} = 0.4819 + 4.496/\left(1 + exp\left((\log_{10}[AS] + 3.062)/-0.3119\right)\right) \quad (6).$$

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Previous studies have proposed that the interaction between dust and low concentration AS enhances the IMF ability of dust (Whale et al., 2018; Worthy et al., 2021). The observed sigmoidal response reflects progressive activation of ice nucleation sites on dust surface upon AS exposure. Minor ice nucleation sites were already activated at low AS concentration down to 5×10^{-4} M, resulting a ΔT_{50}^{u} of 0.4 K. Increase in AS concentration to 5×10^{-3} M substantially activated the ice nucleation sites, leading to a rapid increase of ΔT_{50}^{u} to 4.8 K. Further increase AS to 5×10^{-2} M led to surface saturation of Us-dust by AS, whereby no additional nucleation sites were activated and thus ΔT_{50}^{u} remained constant. The underlying mechanism is further discussed in Section 3.5.1.

ΔT_{50}^{u} for Us-dust mixed with AS fell within the upper range reported for other minerals (Fig.7c). At AS concentrations of 10^{-4} M – 10^{-3} M, Us-dust exhibited comparable ΔT_{50}^{u} to K-feldspar, but was greater at higher AS concentrations than those reported by Whale et al. (2018) and Worthy et al. (2021). To eliminate biases originating from inconsistent dust concentrations in these studies, AS content was normalized by dust concentration and expressed as the mass ratio of AS to dust (AS/Dust) for intercomparison (Fig. 7d). ΔT_{50}^{u} increased with AS/Dust ratio with a more pronounced sigmoidal shape, which is fitted by a Boltzmann function as:

$$\Delta T_{50}^{u} = 0.4104 + 4.37/\left(1 + exp\left((\log_{10}(AS/Dust) + 0.1844)/-0.2838\right)\right) \quad (7).$$

When plotted against AS/Dust, ΔT_{50}^{u} for Us-dust generally converges to the range of ΔT_{50}^{u} for K-feldspar (Whale et al., 2018; Worthy et al., 2021). However, ΔT_{50}^{u} for K-feldspar tended to be a linear response against either log values of AS concentration or AS/Dust ratios. ΔT_{50}^{u} for Us-dust was greater than quartz but lower than kaolinite at the investigated AS concentrations. ΔT_{50}^{u} for Us-dust was slightly lower than ATD reported by Worthy et al. (2021), but higher than ATD from Whale et al. (2018). These differences in ΔT_{50}^{u} or its trend was likely controlled by divergent interfacial interactions between AS and various minerals of the investigated dust. In addition, Worthy et al. (2021) suggested that those inconsistencies may be associated with the different contact time between AS and dust.

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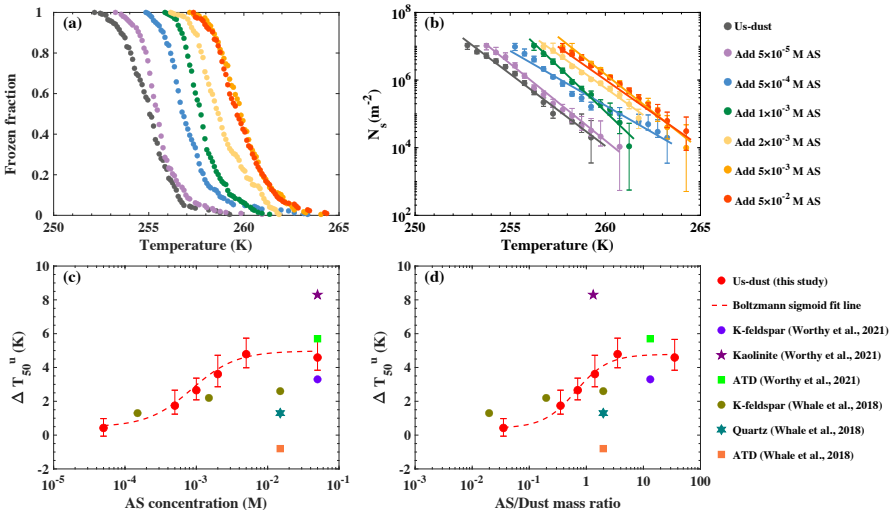

Figure 7. (a) Frozen fraction for Us-dust (1.87×10^{-2} wt%) mixed with ammonium sulfate (AS) at different concentrations. (b) N_s with error bars corresponding to the 95% confidence intervals. Solid lines are parameterizations of N_s. (c) $\Delta T_{50}{}^{u}$ as a function of AS concentration. (d) $\Delta T_{50}{}^{u}$ as a function of the mass ratio of AS to dust (AS/Dust). Dashed lines (c, d) represent the Boltzmann sigmoid fitting. Error bars (c, d) represent the 25% and 75% percentiles. The literature data in panels (c) and (d) are from Whale et al. (2018) and Worthy et al. (2021).

3.4 Ice nucleation ability of dust mixed with NaCl or acids and after heating

Figure 8 shows that $\Delta T_{50}{}^{u}$ for Us-dust were close to zero after heat treatment, mixed with NaCl, or acid (H_2SO_4 or HNO_3) treatment. This indicates that these treatments had no impacts on IMF ability of Us-dust when comparing the T_{50} values. However, these treatments had certain effects on the IMF ability at freezing temperature above 256 K as evidenced by the deviations in N_s (Fig. 8d-f) and frozen fraction (Fig. S2). Specifically, heat and H_2SO_4 treatments, as well as higher concentrations of NaCl (5×10^{-2} M) and HNO_3 (2.5×10^{-2} M) treatments imposed a reduction in N_s, whereas lower concentrations of NaCl (5×10^{-4} M) and HNO_3 (5×10^{-4} M and 5×10^{-3} M) treatments led to an increase in N_s. The impact of heat treatment on Us-dust was consistent with the results from Zolles et al (2015) showing no significant change in the IMF ability of K-feldspar, Na/Ca-feldspar, Kaolinite, and ATD after heating at 523 K

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for 4-5 h, as they stated that the $\Delta T_{50}{}^{u}$ were within the uncertainty.

Previous studies show that NaCl and acids can reduce the IMF ability of K-feldspar, kaolinite, and quartz (Reischel and Vali, 1975; Whale, 2022; Whale et al., 2018; Yun et al., 2021) (Fig. 8a, b). For K-feldspar and kaolinite, higher NaCl concentration led to lower $\Delta T_{50}{}^{u}$ (negative values). Higher HCl and HNO_3 concentrations led to a stronger reduction in $\Delta T_{50}{}^{u}$ for K-feldspar (Reischel and Vali, 1975; Whale et al., 2018). However, NaCl and HCl did not affect the IMF ability of ATD (Link et al., 2020; Perkins et al., 2020), which was similar to Us-dust. Although, NaCl, heat, and acid treatments did not substantially change the T_{50} of Us-dust, they altered the frozen fraction and N_s, leading to the enhancement or reduction in the IMF ability of Us-dust at warmer freezing temperature. A similar phenomenon was also observed for ATD when mixed with inorganic acids (Perkins et al., 2020).

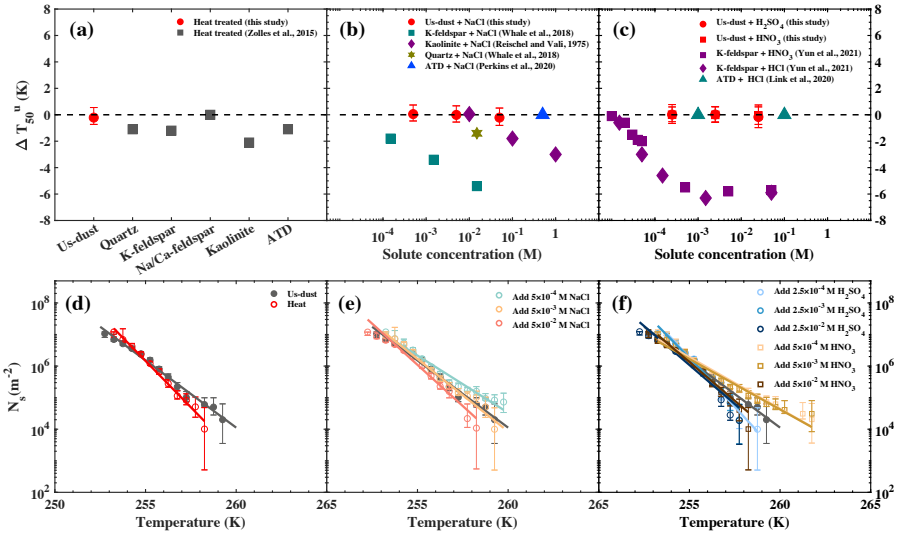

Figure 8. Comparison of $\Delta T_{50}{}^{u}$ (a-c) and N_s (d-f) for Us-dust treated by heating, adding NaCl, H_2SO_4 and HNO_3. Error bars for $\Delta T_{50}{}^{u}$ represent the 25% and 75% percentiles. Error bars for N_s correspond to the 95% confidence intervals. Solid lines are parameterizations of N_s. The comparison data in panels (a, b, c) are from previous studies (Link et al., 2020; Perkins et al., 2020; Reischel and Vali, 1975; Whale et al., 2018; Yun et al., 2021; Zolles et al., 2015).

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3.5 Potential mechanisms for the change in ice nucleation ability

3.5.1 $(NH_4)_2SO_4$ addition

The contribution to particle surface area of Us-dust by illite, quartz, chlorite, kaolinite, and feldspar was 37%, 24%, 23%, 12%, and 5%, respectively. In Us-dust, only about 13% of felspar type particles have K element (i.e., K-feldspar-like) and the rest was Na/Ca-feldspar (Fig. S3). Previous studies have shown that the K-feldspar exhibits higher IMF activity than Na/Ca-feldspar (Atkinson et al., 2013; Harrison et al., 2016; Peckhaus et al., 2016; Whale et al., 2017; Zolles et al., 2015). Although K-feldspar-like particles accounted for only a small fraction of Us-dust surface area, the $\Delta T_{50}{}^{u}$ values were comparable to those reported for K-feldspar, suggesting that feldspar may provide a disproportionate contribution to the highly active sites responsible for the observed enhancement as compared with S-dust. However, K-feldspar alone could not fully account the enhancement at AS/Dust mass ratio greater than 1 (Fig. 7c, d), implying contributions from other minerals (e.g., kaolinite) in Us-dust.

Ammonium ions (NH_4^+) in solution can replace the K^+ on the surface of K-feldspar, forming interfacial sites that promote ice nucleation (Worthy et al., 2021). In addition, NH_4^+ adsorbed on dust surfaces potential can alter the arrangement and orientation of water molecules, thereby enhancing ice nucleation activity (Anim-Danso et al., 2016; Kumar et al., 2018; Worthy et al., 2021). At the pH (~5.5–6.6) of droplets with AS, the surfaces of feldspar (point of zero charge (PZC) < 2), quartz (PZC ≈ 2), and kaolinite (PZC ≈ 3–6) were all negatively charged and can adsorb NH_4^+ due to the lack of charge-balancing cations (Kumar et al., 2018, 2019a; Worthy et al., 2021). $\Delta T_{50}{}^{u}$ of Us-dust exhibited a Boltzmann-type (sigmoidal) dependence on AS concentration, suggesting that the enhancement is consistent a threshold-saturation interfacial process from low to high AS coverage on dust surface (Fig.7c, d). Our experimental data directly support that NH_4^+ adsorption on mineral surface is the primary mechanism for the ice nucleation enhancement by AS. The ice nucleation enhancement on Us-dust by AS observed here was initiated by NH_4^+ adsorption on the dust surface, while the participation of NH_4^+ in ion exchange on feldspar (Worthy et al., 2021) and regulation of hydrogen-bond

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ordering at dust surface (Whale, 2022) may further amplify the enhancement.

3.5.2 NaCl addition

Previous studies have shown that the effect of near-neutral alkali salt solutions on the IMF of mineral dust depends on the particle type and their surface properties (Kumar et al., 2018, 2019a, b; Whale et al., 2018). Suppression of the IMF on feldspar by NaCl may be attributed additional surface process beyond colligative behavior, in which NaCl can increase the interfacial free energy to form ice cluster (Whale et al., 2018). Kumar et al. (2018) suggested that alkali cations may adsorb onto negatively charged mineral surface and form an interfacial double layer on feldspar that block ice nucleation sites. In contrast, Perkins et al. (2020) showed that NaCl solution has no effect on the IMF activity of ATD (Fig.8b). Similarly, no significant change in T_{50} was observed for Us-dust in this study at NaCl concentration from 5×10^{-4} M to 5×10^{-2} M (Fig.8b). However, N_s of Us-dust at warmer temperature (256 K–260 K) gradually decrease with increasing NaCl concentration, indicating selective suppression of the most ice active sites (Fig.8e). Because feldspar tends to respond more strongly to NaCl suppression (Whale et al., 2018) and is highly active at warm temperatures (Atkinson et al., 2013; Boose et al., 2019), the observed high sensitivity of Us-dust to NaCl above 256 K are likely associated with feldspar. At lower temperatures, non-feldspar minerals, which is less sensitive to NaCl than feldspar (Fig.8b), control the freezing of Us-dust.

3.5.3 Acid treatment

Previous studies have found that the effect of acid treatments on IMF of mineral dust is sample dependent. Suppression on the IMF activity of feldspar and ATD by H_2SO_4 treatment was linked to acid-driven surface reactions, loss of surface K^+, and formation of sulfate-containing products on particle surface (Augustin-Bauditz et al., 2014; Kumar et al., 2018; Niedermeier et al., 2011). For HNO_3, HCl, and organic acid treatment, Yun et al. (2021) proposed that the reduced IMF ability of K-rich feldspar was associated with H_3O^+/K^+ exchange and incongruent dissolution of Al and Si. Sullivan et al. (Sullivan et al., 2010b) found that HNO_3 did not significantly suppress the freezing by ATD above water saturation. However, Perkins et al. (2020) showed that acid treatment on ATD affected the nucleation sites at warm temperatures, while

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ice active sites at low temperatures remain unaffected. For Us-dust, no significant shift in $\Delta T_{50}{}^{u}$ was observed after treatment with H_2SO_4 (2.5×10^{-4} M to 2.5×10^{-2} M) or HNO_3 (5×10^{-4} M to 5×10^{-2} M), indicating that limited impact on IMF activity in terms of T_{50}. However, above 256 K, N_s of Us-dust was reduced by HNO_3 at 5×10^{-2} M and H_2SO_4 at all tested concentrations, but was slightly enhanced by HNO_3 at 5×10^{-4} M and 5×10^{-3} M. These imply that acids have effects on nucleation sites of Us-dust at warmer temperatures, but direction and magnitude are dependent on acid type and concentration.

3.5.4 Heat treatment

Previous studies have shown that heat treatment either suppress the IMF ability of ATD or exerts no effect. Zolles et al. (2015) and Niedermeier et al. (2011) both reported that the IMF activity of ATD remined unchanged after heating. Perkins et al. (2020) found stronger heating caused larger changes in freezing behaver of ATD. They suggested that ice active sites at warmer temperatures are more sensitive to heating, which may be associated with defects, heat sensitive components, or other labile sites on the ATD surface. In addition, Pereira et al. (2022) observed reduced the IMF ability of agricultural soil dust after heating which was attributed to heat sensitive components, like organic matter. In our study, heat treatment on Us-dust at 90°C for 30 min led to the loss of ice active sites at warm temperatures while ice active sites at lower temperatures retained intact. This result is consistent with Perkin et al. (2022) suggesting that ice active sites in complex dust may have different heat sensitivities.

3.6 Heterogeneous ice nucleation rate coefficient

The experimentally derived J_{het} and its parameterizations as functions of Δa_w and temperature are presented in Fig.9 and Fig. S4, respectively. Us-dust nucleated ice starting from relatively low Δa_w (~0.1) and high temperature (i. e., 262 K) at the investigated concentrations. The temperature and Δa_w dependence of J_{het} for Us-dust is substantially steeper than for S-dust. This is associated with the differences in particle size and composition before and after ultrasonic treatment (Section 3.1). AS enhanced the J_{het} of Us-dust more progressively with AS concentration. Only minor variations in J_{het} for Us-dust (i.e., less than one order of magnitude) were observed above 256 K after heat, acid (H_2SO_4 and HNO_3), and NaCl treatments (Fig. S4). J_{het} of Us-dust is about

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100 times higher than kaolinite and illite, but lower than Aluminum oxide and Iron oxide at the investigated Δa_w (Fig. 9c). Compared with non-mineral or biological substances, in terms of J_{het} (Fig. 9d), Us-dust is less efficient than fungal spores (Iannone et al., 2011), comparable to 1-nonadecanol (Knopf and Forrester, 2011), but more efficient than marine microbes (*Thalassiosira Pseudonana* and *Nannochloris Atomus*) (Alpert et al., 2011; Knopf et al., 2011), and humic like substances (leonardite and Pahokee Peat) (Rigg et al., 2013). After mixed with AS, J_{het} for Us-dust shifted toward but still lower than aluminum oxide, iron oxide, and fungal spores.

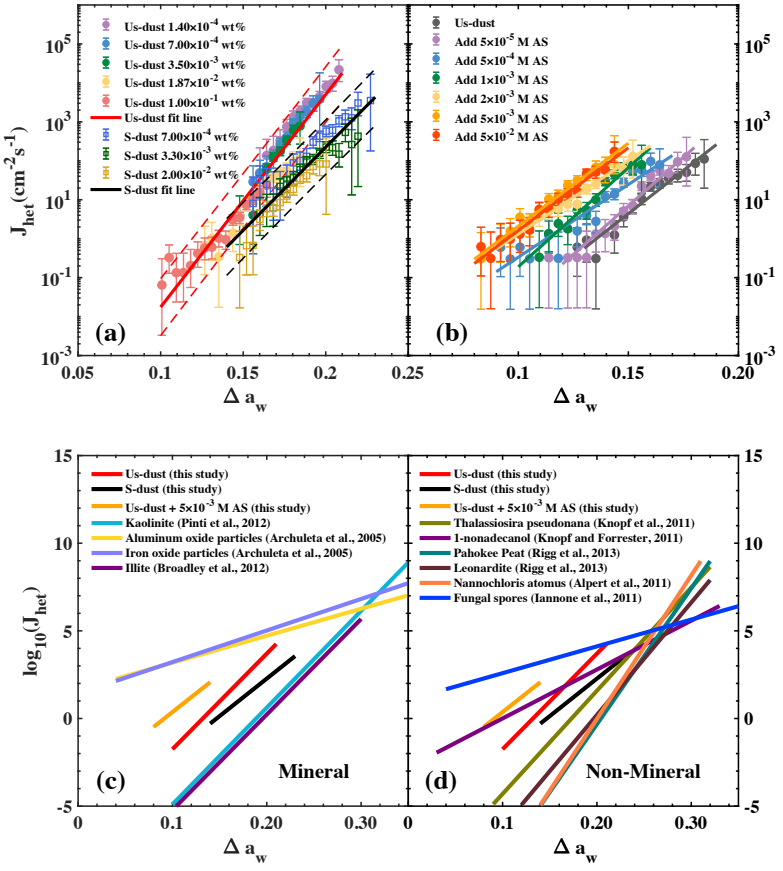

Figure 9. J_{het} as a function of Δa_w for Us-dust and S-dust (a), and Us-dust (1.87×10^{-2} wt%) mixed with AS (b). Comparison J_{het} of Us-dust, S-dust, and Us-dust mixed with 0.005 M AS with minerals (c) and non-minerals (d) from previous studies (Alpert et al., 2011; Archuleta et al., 2005; Broadley et al., 2012; Iannone et al., 2011; Knopf et al., 2002, 2011; Knopf and Alpert,

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2013; Knopf and Forrester, 2011; Murray et al., 2011; Pinti et al., 2012; Rigg et al., 2013). Dashed lines (a) are the 95% prediction intervals. Solid lines (a, b) are parameterizations according to $\log(J_{het}) = m_2 \times \Delta a_w + n_2$ and the values of m_2 and n_2 are listed in Table S2.

4. Atmospheric implication

As desert dust is emitted into the atmosphere and transported over long distances to different environments or high altitude, it may be aged and mixed with AS. The effect on the IMF activity by AS should be represented in parameterizations and considered in cloud modeling. Including AS concentration as another variable, we proposed parameterizations of N_s and J_{het} to consider the impact of AS on IMF of Us-dust. Parameterizations as a function of AS to dust mass ratio (AS/Dust, r) and temperature (T) or Δa_w were developed as follows:

$$N_s = 10^{(k_1 \times T + B_1)}, \quad (8)$$

$$J_{het} = 10^{(k_2 \times T + B_2)}, \quad (9)$$

$$J_{het} = 10^{(k_3 \times \Delta a_w + B_3)}, \quad (10)$$

where the parameters B (B_1, B_2, and B_3) are fitted by a Boltzmann sigmoid function, $B = a + \frac{b}{\left(1+e^{\frac{\log_{10}(r)+c}{d}}\right)}$. The fitting parameters are listed in Table 2. Evaluation of N_s (T, r), J_{het} (T, r), and J_{het} (Δa_w, r) parameterizations against the experimental observations is shown in Figs. S5-S7. The proposed parameterizations reproduce the observed impacts of AS on the IMF ability of Us-dust at the investigated AS and dust concentrations (AS < 0.1M, AS/Dust = 0.03~30).

We applied the proposed parameterizations to estimate N_s and J_{het} for Tengger Desert dust internally mixed with AS under representative atmospheric conditions. As shown in Fig. 10, both N_s and J_{het} increase markedly with decreasing temperature (increasing Δa_w) and with increasing AS/Dust mass ratio, consistent with the observed AS enhancement on IMF activity. As an illustrative case, an AS particle with an initial diameter of 0.5 μm takes up water and grows into a 10 μm droplet, corresponding to an AS concentration of about 0.002 M. When a Tengger Desert dust particle (0.5 μm and 10^{-13} g) is incorporated into this 10 μm AS droplet (AS/Dust of 1.16), the parameterizations predict N_s of 4.88×10^5 cm^{-2} and J_{het} of 6.5 cm^{-2} s^{-1} at 260 K. Under

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these conditions, Δa_w is 0.12. J_{het} value determined from J_{het} (Δa_w, r) parameterization is 6.2 cm^{-2} s^{-1}, consistent with the value calculated by J_{het} (T, r) parameterization. In contrast, the corresponding N_s, J_{het} (Δa_w), and J_{het} (T) for untreated Tengger Desert dust is 2.14×10^4 cm^{-2}, 0.22 cm^{-2} s^{-1}, and 0.35 cm^{-2} s^{-1} at 260 K, respectively, about 20-30 times lower than dust mixed with AS. This example suggests that internal mixing with AS can substantially increase the contribution of Tengger Desert dust to IMF under atmospherically relevant conditions.

The parameterizations developed here link dust IMF properties to AS loading and to temperature or Δa_w, providing a practical means to predict AS driven changes in IMF activity. N_s (T, r) can be combined with dust particle surface area to estimate INP concentrations under the singular hypothesis, while J_{het} (T, r) and J_{het} (Δa_w, r) can be used in time-dependent freezing frameworks to predict INP production rates. These parameterizations based on measurements of the Tengger Desert dust are constrained by AS concentrations below 0.1 M.

Table 2 The k, a, b, c, and d values for parameterizations of N_s (T, r), J_{het} (T, r), and J_{het} (Δa_w, r) along with Eqs.8-10.

Parametric formula	N_s **(T, r)**	J_{het} **(T, r)**	J_{het} **(Δa_w,r)**
k	-0.38	-0.34	42.09
a	103.13	89.65	-3.81
LCL_a	102.05	89.23	-4.41
UCL_a	104.2	90.08	-3.22
b	1.85	-1.58	-1.6
LCL_b	0.35	-2.35	-2.63
UCL_b	3.34	-0.81	-0.56
c	-0.33	-0.26	-0.23
LCL_c	-1.02	-0.65	-0.75
UCL_c	0.36	0.14	0.29
d	-0.38	0.33	0.31
LCL_d	-1.12	-0.08	-0.21
UCL_d	0.37	0.75	0.84
$RMSE$	0.2	0.22	0.23
R^2	0.94	0.93	0.92

Note: the parameter k was determined through a grid search that minimizes the sum of absolute errors.

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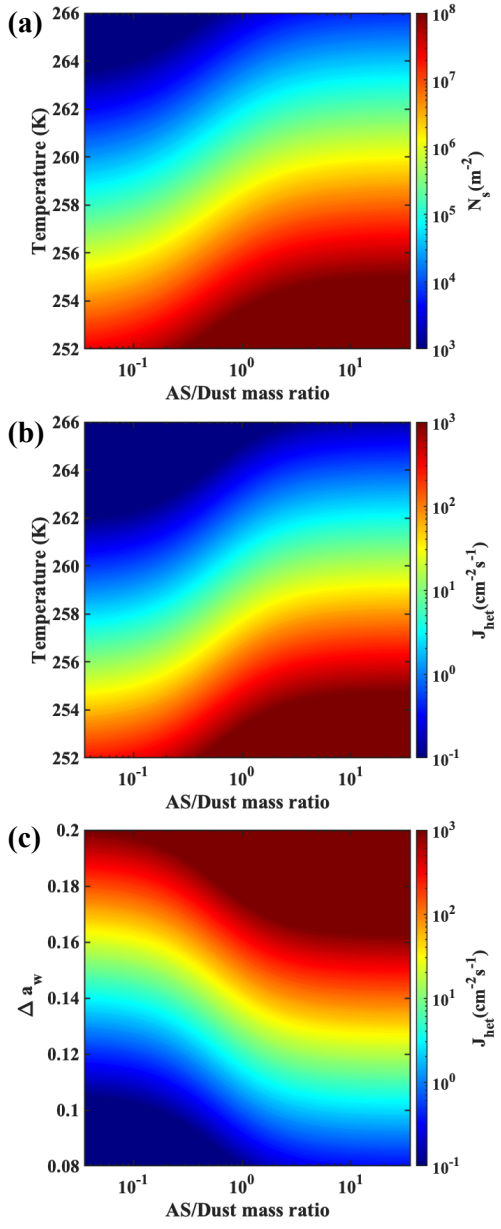

Figure 10. Prediction of N_s and J_{het} based on the parameterization of Us-dust mixed with AS. (a) N_s and (b) J_{het} as a function of temperature and AS/Dust mass ratio, (c) J_{het} as a function of Δa_w and AS/Dust mass ratio.

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5. Conclusion

We investigated the IMF of surface-collected dust from Tengger Desert and its response to various aging treatments, including mixing with NaCl and AS, heating, and acid treatment with H_2SO_4 and HNO_3. Tengger Desert dust initiated the IMF at temperatures as high as 262.3 K, with T_{50} increased from 251.6 K to 256.7 K as the dust mass fraction rose from 1.4×10^{-4} wt% to 1.0×10^{-1} wt%. Single particle analysis showed that ultrasonic treatment of dust in water substantially modified the suspended dust population. The ultrasonic treatment shifted the modal particle diameter from approximately 0.35 to 0.7 μm and altered mineralogical composition with increased contributions from chlorite, illite, and feldspar, reduced kaolinite, and minor change in quartz. The 2% increase in particle number and surface area of feldspar resulted in about 2 K higher T_{50} and one order of magnitude increase in N_s. This result suggests that mineralogical composition of complex natural dust can strongly influence ice nucleation properties, especially when it contains components with high ice nucleation activity.

Among the investigated aging treatments, AS enhanced the ice nucleation activity of Us-dust. When AS changed from 5×10^{-5} M to 5×10^{-2} M, the increase in T_{50} for Us-dust at 1.87×10^{-2} wt% changed from 0.4 K to 4.8 K. The corresponding increments in T_{50}, as well as N_s and J_{het}, followed a Boltzmann sigmoidal response. This directly support that NH_4^+ adsorption on mineral surface activating ice active sites is the primary mechanism for the ice nucleation enhancement by AS. Other treatments on Us-dust by NaCl, acid (H_2SO_4 and HNO_3), and heating have minor impacts on the IMF behavior in terms of T_{50}. However, heat and H_2SO_4 treatments, as well as higher concentrations of NaCl and HNO_3 (>2.5×10^{-2} M), led to a reduction in N_s and J_{het} above 256 K, whereas lower concentrations of NaCl (5×10^{-4} M) and HNO_3 (< 5×10^{-3} M) produced modest increases. These results indicate the presence of labile ice active sites at warmer temperatures with variably sensitive to chemical and thermal processing. Ice nucleation modeling studies should consider these effects by NaCl, H_2SO_4 and HNO_3.

This study provided experimental constraints on the IMF properties of Tengger Desert dust for the modeling assessments on the impacts of East Asian dust to mixed-phase

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cloud formation. Parameterizations of the experimentally derived $N_s(T)$ and $J_{het}(T$ or $\Delta a_w)$ of desert dust were proposed. N_s of Us-dust was comparable to those reported for feldspar and showed stronger temperature dependence than previously reported desert dust. Although N_s were broadly comparable to literature values below 255 K, Tengger Desert dust exhibited up to 2-3 orders of magnitude lower than commonly used parameterizations for desert dust of Niemand et al. (2012) and Ullrich et al. (2017) above 255 K. J_{het} of Us-dust was higher than those reported for kaolinite, illite, some marine microorganisms, and humic-like substances, but was lower than aluminum oxide, iron oxide, and fungal spores. Considering the impact of AS, we proposed parameterizations of N_s (T, r), J_{het} (T, r), and J_{het} (a_w, r) as a function of the mass ratio of AS/Dust. These parameterizations can be implemented in models to investigate the impact of internally mixed dust-AS on cloud formation.

The parameterizations are constrained to surface dust from Tengger Desert and the investigated AS concentrations. Particle size distribution, mineralogical composition, and surface properties of the investigated Tengger Desert dust may differ from airborne dust, which may introduce uncertainty when the proposed parameterizations are applied. The impacts of chemical aging with wider ranges of atmospheric substances on ice nucleation for authentic airborne dust should be further explored. Investigations on ice nucleation of dust from other source regions with atmospheric aging are required to provide better constrains on the effects of mineral composition and aging for broader applicability of ice nucleation parameterizations.

Data availability. All data are given in the main text or in the Supplement.

Supplement. The supplement related to this article is available online.

Author contributions. JX wrote the first draft of the manuscript. SKW, HYQ, SLC, and QW performed the experiments. YY set up the ice nucleation experimental system. JX, SKW, JYL, YWH, and TZ performed the data analysis. YHC collected the samples. BBW, DFZ, and YHC initiated the study. BBW supervised the project. BBW and JX led the discussion and all authors contributed to the data interpretation and the manuscript writing.

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Competing interests. At least one of the (co-)authors is a member of the editorial board of *Atmospheric Chemistry and Physics.*

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