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Measurements of ice nucleation by mineral dusts in the contact mode

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**Atmospheric
Chemistry
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Measurements of ice nucleation by mineral dusts in the contact mode

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**Contact mode
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Abstract

Formation of ice in Earth's atmosphere at temperatures above approximately −20 °C is one of the outstanding problems in cloud physics. Contact nucleation has been suggested as a possible mechanism for freezing at relatively high temperatures; some
5   laboratory experiments have shown contact freezing activity at temperatures as high as −4 °C. We have investigated Arizona Test Dust and kaolinite as contact nuclei as a function of size and temperature and find that the fraction of submicron particles that are active as contact ice nuclei is less than 10^{-3} for −18 °C and greater. We also find that the different dusts are quite distinct in their effectiveness as contact nuclei; Arizona
10 Test Dust catalyzed freezing in the contact mode at all mobility diameters we tested at −18 °C whereas kaolinite triggered freezing only for mobility diameters of 1000 and 500 nm at that temperature.

1   Introduction

When freezing is initiated at temperatures greater than approximately −36 °C in Earth's
15 atmosphere, the phase transition must be catalyzed. Water will persist in a metastable, supercooled state at temperatures greater than −36 °C unless nucleation is triggered, usually by the presence of a foreign surface. For example, as most air travelers know, aircraft icing is a severe hazard. Buildup of ice on the wings of an airplane can cause it to lose lift, with catastrophic results (Jameson and Kostinski, 2000). That phenomenon
20 is an extreme example of heterogeneous freezing, as the water is liquid until it comes into contact with the aircraft. (In fact, if the plane were to fly through a cloud of ice crystals, the danger would be minimal, as the crystals would simply bounce off the wing).

In addition to the dangers it presents to aviation, freezing in the atmosphere is im-
25 portant because of the role that ice plays in Earth's radiative equilibrium and hydrologic cycle. Ice crystals interact with incoming solar radiation where the dominant effect is

scattering; they also interact with terrestrial radiation where they both absorb and scatter. The Bergeron-Wegener-Findeisen process, where ice grows at the expense of surrounding liquid water droplets in mixed phase clouds, is thought to be responsible for the majority of precipitation (Lau and Wu, 2003). Finally, freezing affects cloud dynamics through the latent heat of freezing and sublimation (Rosenfeld et al., 2008).

There are three pathways to heterogeneous nucleation – deposition, immersion/condensation, and contact freezing. (We group immersion and condensation nucleation because both occur with the foreign substance inside the droplet). Of the three, contact is the most mysterious. Though the details of the physical mechanisms underpinning deposition and immersion/condensation nucleation are not well understood, the nuclei can at least be measured in the atmosphere (Cantrell and Heysmfield, 2005). Contact nucleation is neither understood on a mechanistic level, nor are the particles responsible for it currently measured in the contact mode.

Contact freezing is distinct from other modes in that the surface of a supercooled droplet of water must come into contact with an aerosol particle. The interaction at the surface of the supercooled droplet initiates freezing. The possible importance of contact nucleation stems from the fact that it may be effective at higher temperatures than immersion/condensation nucleation and that smaller particles may initiate the phase transition. Recent work has shown that mineral dusts, the most common ice nuclei in the atmosphere, are not effective as nuclei in the immersion mode for temperatures greater than approximately $-20\,°C$ (Niedermeier et al., 2010; Lüönd et al., 2010; Murray et al., 2011). There are examples of freezing in the atmosphere at considerably higher temperatures, which opens the question as to what initiates nucleation in such warm conditions (see e.g. Rangno and Hobbs, 1991, 1994). Contact nucleation has been suggested as a candidate mechanism for such high temperature freezing and as a way to sustain mixed phase clouds in the Arctic (Morrison, 2005).

Some of the first experiments in contact nucleation of ice showed freezing at temperatures as high as $-4\,°C$ (Gokhale and Lewinter, 1971; Gokhale and Spengler, 1972). In those studies powders, usually with a diameter of a micrometer or greater, impacted

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upon droplets of water which were one to five millimeters in diameter. The freezing temperatures in that setup ranged from five to ten degrees greater than when the powder was mixed into the water droplet's volume (i.e. in the immersion mode). Subsequent work showed that aerosol particles as small as tens of nanometers in diameter catalyzed freezing in the contact mode at −10 °C (Sax and Goldsmith, 1972).

A renewed interest in ice nucleation has prompted more recent investigations into contact mode nucleation. Shaw et al. (2005) documented a clear shift in freezing catalyzed by volcanic ash in which the catalyst (i.e. the ash) did not have to impinge upon the surface of the water from the outside. The presence of the ash at the air-water interface was enough to initiate freezing at the higher temperature (see also Fornea et al., 2009). In one of the few studies to date to investigate the effect of size on contact nucleation, Ladino et al. (2011) found that kaolinite dust was more effective in the contact mode for 400 and 800 nm diameter particles for temperatures less than −15 °C. (Single particles of kaolinite are essentially inactive in the immersion mode for temperatures greater than −20 °C; Lüönd et al., 2010.)

We present results of contact nucleation by size selected mineral dusts showing that neither submicron Arizona Test Dust nor kaolinite is particularly effective as contact freezing nuclei. The fraction of dust active in the contact mode at −18 °C was less than 10^{-3}. Further, we demonstrate a dependence on particle size, temperature, and particle composition for contact nucleation. Before presenting these results, we briefly explain our experimental method.

2 Experiment

Contact nucleation experiments are challenging because the surface of a supercooled droplet of water must come into contact with an aerosol particle and freezing events initiated by that contact must be detected. Ideally, this would be done on an encounter-by-encounter basis. However, as we show below, such an approach is not feasible (yet) for the temperature range in our experiments. Instead, we use a supercooled

thermostatted droplet on a substrate which is exposed to a stream of aerosol laden air. Particles in the turbulent flow are deposited to the surface of the droplet. An LED laser focused through the droplet onto a photodiode monitors the phase change. Light is scattered upon freezing, resulting in a loss of signal at the diode. Figure 1 is a block schematic of the experimental setup. We describe the individual elements of the experiment in the subsections below.

2.1 Aerosol generation and size selection

We tested Arizona Test Dust (ATD, Powder Technology Inc.) and kaolinite (Fluka). Both were dispersed using a fluidized bed aerosol generator (TSI 3400A). The initial flow of filtered lab air (dew point of approximately −40 °C) is 15 l per minute. The mixing chamber, which has a volume of 11.4 l, served to stabilize the concentration of aerosol. Excess flow was vented to the room at that point. The sample mineral dust aerosol were then selected by their electrical mobility diameters with a differential mobility analyzer (DMA, TSI 3081). We selected sizes ranging from one micron to 62.5 nm. The sample flow through the DMA and sample chamber was one liter per minute for all of the sizes we used. Downstream from the nucleation chamber, the particle concentration was measured using a condensation particle counter (CPC, TSI 3772).

2.2 Temperature measurement and control

Temperature measurement and control were primarily in the nucleation chamber. To minimize temperature gradients, the sample flow is conditioned prior to entering the chamber by pulling it through 1.5 m of coiled copper tubing (0.25″ o.d.) that is submerged within the coolant used as a heat sink for the Peltier element (see below).

The nucleation chamber, shown in Fig. 2, is milled from two copper blocks for temperature stability and control and is surrounded with foam insulation (not shown). A Peltier element and PID control system (Ferrotec) are used to set the temperature of the nucleation chamber. Coolant from a Julabo CF40 circulating through a water block on the

hot side of the Peltier element serves as a heat sink. Four temperature sensors are used to monitor the chamber temperature. Two thin-film, $100\,\Omega$, platinum resistance thermal detectors (RTDs, Minco) are attached to the outside surface of the chamber; one RTD, which serves as the point of reference for the control of the temperature, is attached within a cavity 0.5 mm beneath the stage where the silanized glass slide is placed. The last temperature measurement is from a thermocouple that monitors the sample air temperature within the nucleation chamber. The RTDs and thermocouple were calibrated against each other before installation and have been checked against the melting point of water in the chamber.

The temperatures we report are those from the RTD within the cavity under the slide (i.e. the RTD beneath the droplet stage). The vertical temperature gradient across the chamber is approximately one degree, though the temperature gradient across the droplet is less. The uncertainty in the temperature of the droplet is $\sim \pm 0.5\,°C$, arising principally from the temperature gradient across the chamber. The uncertainty of the droplet's temperature is estimated from melting point tests, which are sensitive to the entire volume of the droplet.

2.3 Test droplet

As noted above, contact nucleation requires that a supercooled droplet of water comes into contact with an aerosol particle. Our approach is to immobilize the water on a substrate and detect freezing events as aerosol particles deposit to it from the surrounding flow. The substrate, shown just above the RTD recess in Fig. 2, is a silanized glass slide, either prepared from clean slides by soaking them in Rain-X™ or purchased from Hampton Research and used as received. The silanized glass slide reduced the freezing temperature of the five microliter water droplets to less than $-20\,°C$, well outside the range of temperatures we investigated in this study. As discussed above, the droplets were exposed to aerosol by pulling sample air through the chamber.

As mentioned earlier, a red LED (1 mW, 650 nm), focused through the droplet onto a photodiode, was the means of detecting freezing events. Upon freezing, the

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transmission of light through the droplet was substantially reduced as the translucent, white ice in the droplet scattered the radiation. Coincident with the drop in signal from the photodiode, we observed a small spike in the temperature of the air as measured by the thermocouple shown in Fig. 2, a result of the droplet's latent heat of freezing. Freezing was visually confirmed in many experiments by immediately disassembling the chamber and inspecting the droplet visually. The droplet's phase was consistent with the state of the signal as measured by the photodiode in all cases.

2.4 Number of aerosol particles deposited to the droplet

The aim of our experiments is to determine the number of aerosol particles required to catalyze freezing as a function of temperature and type of aerosol. The crux of that endeavor is the number of aerosol particles deposited to the test droplet's surface before freezing. We cannot, as yet, detect an individual aerosol particle's interaction with the surface of our droplet. We are forced to use an empirical, statistical measure.

To determine the number of aerosol particles deposited to the surface of the droplet, we expose it to the turbulent sample flow for a specified period of time, then evaporate the droplet. We inspect the residue, using a scanning electron microscope (SEM) and energy dispersive X-Ray spectrometer. Because the dust particles agglomerate on the surface of the droplet, both as the air flows past during the experiment and as the droplet evaporates, we cannot simply count individual particles on the surface of the glass slide. To determine the number of particles deposited to the droplet, we measure the total, projected surface area of dust apparent within the area of the droplet's residue, then divide that by the average projected area of a single aerosol particle of the same mobility diameter. The average area of a particle of a given mobility diameter was determined in a separate experiment by collecting aerosol particles downstream of the DMA on a Nucleopore filter, then inspecting that using the SEM. The last step is necessary because mineral dust has a complicated morphology. The mobility diameter and the diameter as determined from a dust particle's projected area (i.e. from the SEM images) can be quite different.

It is not practical to perform this analysis for every experiment. We calculate a collection efficiency for a series of test droplets, then use the result to determine the number of aerosol particles deposited to the surface of the droplet for all further experiments. The collection efficiency is

$$CE = \frac{N_{\text{deposited}}}{Cq\Delta t} \tag{1}$$

where $N_{\text{deposited}}$ is the number of particles deposited to the surface of the droplet, as discussed above, C is the concentration of aerosol particles in the sample flow as measured by the CPC downstream of the chamber, q is the flow rate, and Δt is the duration of the experiment. In practice, the number of particles to which the droplet was exposed was calculated using TSI's AIM software.

We conducted four experiments using kaolinite with mobility diameters of 1000 and 500 nm (two experiments with each mobility diameter). The calculated average collection efficiency is $3.8 \times 10^{-3} \pm 2.6 \times 10^{-3}$. The uncertainty in the collection efficiency of the droplet arises primarily from the uncertainty in $N_{\text{deposited}}$, the number of aerosol particles collected by the droplets.

3   Results and discussion

Figure 3 shows the efficiency of contact nucleation for Arizona Test Dust and kaolinite as a function of mobility diameter and temperature. We define contact nucleation efficiency as the number of the freezing events per number of particles deposited to the droplet. By adopting that definition, we assume that the freezing transition is initiated immediately upon an aerosol particle's contact with the surface. In essence, we assume that the last aerosol particle to come into contact with the surface is the one that initiates freezing. Though we cannot rule out the possibility that an aerosol particle might sit on the surface for some time, then initiate freezing, in experiments where we

halted the deposition of particles to the surface after some time but kept the droplet at the subzero temperature, we observed no freezing events.

3.1 Effectiveness of dust in the contact mode

The most striking result, apparent from Fig. 3, is that both ATD and kaolinite are ineffective as contact nuclei at $-18\,°C$ and higher. On average, more than 1000 aerosol particles are required to initiate freezing in the contact mode at the lowest temperatures we tested; for most sizes and temperatures, the fraction of aerosol particles active as contact nuclei is less than 10^{-4}.

Though the fraction of aerosol particles active as freezing nuclei in the contact mode is low, it is higher than the active fraction in the immersion mode for comparable temperatures. For example, Jones et al. (2011) report a freezing fraction of 2×10^{-3} at water saturation for ATD at $-24.9\,°C$, the highest temperature tested in their experiments for ATD. The ATD in that study was not size selected. Connolly et al. (2009) found that ATD did initiate freezing in the immersion mode at $-18\,°C$ in the AIDA chamber. However, no freezing was reported at $-12\,°C$. (Connolly et al., 2009, did not report the active fraction.) Freezing at temperatures greater than $-20\,°C$ has not been reported for submicron clay mineral dusts (Fig. 3b, Hoose and Möhler, 2012).

The freezing efficiencies shown in Fig. 3 are quite different than those found in two recent studies of contact nucleation by submicron mineral dust. Both Svensson et al. (2009) and Ladino et al. (2011) found freezing efficiencies greater than unity for some of the ranges they explored. Quantitative evaluation of their results is hampered by ambiguities in the number of aerosol particles collected by their test droplets before freezing.

Svensson et al. (2009) used an electrodynamic trap to suspend droplets with a starting diameter of 60 μm, then observed freezing events as a flow of air with suspended kaolinite particles flowed past the droplet. The kaolinite was not size selected. The number of particle-droplet collisions was inferred by assuming that droplets froze upon the first collision at the lowest temperature tested. Svensson et al. (2009) report

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a freezing efficiency of approximately 0.93 for the kaolinite dust at −10 °C for a high flow of humidified air. We observed no freezing for contact nucleation by kaolinite at any temperature greater than −18 °C, though we did not employ humidified air.

Ladino et al. (2011) also report freezing efficiencies greater than unity, specifically cit-
ing the number of aerosol particles collected by their test droplets as the critical factor. The number of aerosol particles deposited to a falling drop's surface was determined by calculating the collection efficiency of a droplet falling through a reservoir of aerosol particles, then multiplying that efficiency by the volume swept out by the falling droplet. Between one and nine of every thousand droplets captured an aerosol particle on its
traversal of the chamber, whereas, for example, 20 % of the droplets froze at −25 °C in tests of 800 nm diameter particles as contact nuclei.

As noted above, the fraction of dust particles active in the contact mode in our exper-
iments is much lower than those derived from two recent experiments. However, our freezing efficiencies are much closer to those derived from the only field measurement
of contact nuclei of which we know that is recorded in the peer-reviewed literature. Deshler and Vali (1992) used small droplets suspended from thermocouples, detect-
ing crystallization by the increase in temperature from the latent heat of freezing. They calculated the number of aerosol particles deposited to the test droplets using the tem-
perature and relative humidity in the test chamber as well as the aerosol concentration.
To simplify the calculations, they based the calculation on a single sized aerosol par-
ticle. In experiments over the course of two years in Laramie, Wyoming, USA, they observed an average contact nucleus concentration of $3.1 \, \mathrm{l^{-1}}$ active at a temperature of −18 °C for an assumed particle radius of 0.01 μm. If we assume an average aerosol concentration of $100 \, \mathrm{cm^{-3}}$ for the conditions prevalent in Laramie, their efficiency of
3.1×10^{-5} is comparable to our reported efficiencies.

3.2 Temperature dependence of contact nucleation

As expected from classical nucleation theory and from previous experiments (see e.g. Ladino et al., 2011; Svensson et al., 2009), our results show that Arizona Test Dust

and kaolinite are more likely to catalyze freezing in the contact mode with decreasing temperature. Tests with ATD do not show a statistically significant difference between −18 and −17 °C. All of the sizes we tested froze, and the trend was similar for both temperatures. Only the larger particles of kaolinite froze at −18 °C (see Sect. 3.3).

5   We observed no freezing events for kaolinite at −15 °C. Particles of ATD with mobility diameters less than 250 nm did not freeze at −15 °C either. Clearly, there is a temperature dependence for contact nucleation, though it is entangled with size and composition effects in our experiments.

3.3   Size dependence of contact nucleation

10   At −18 and −17 °C, the mean freezing efficiencies for ATD show a decrease with decreasing mobility diameter for diameters of 250 nm and larger, though we cannot rule out the absence of a trend with certainty because of the large uncertainties in our data. The increase in the mean freezing efficiency between 125 and 62.5 nm mobility diameter for ATD at −18 °C appears to be statistically significant, though we are hesitant
15   to state that unequivocally because of our temperature uncertainty of ±0.5 °C and the large uncertainties in the efficiencies at −17 °C.

       However, for ATD, the size dependence at −15 °C is clear. We observed freezing events for mobility diameters of 1000, 500 and 250 nm, but not for 125 and 62.5 nm. The smaller particles did not freeze at the higher temperature despite the fact that the
20   number of particles deposited to the droplets at −15 °C was comparable to the number deposited at the lower temperatures. Similarly, kaolinite triggered nucleation at −18 °C for mobility diameters of 1000 and 500 nm, but not for any smaller than that. Though the size dependence on contact freezing seen in our data is also convolved with the effects of temperature and composition, it is clearly there. Ladino et al. (2011) also found that
25   larger particles were more effective as contact nuclei than were smaller ones.

       Though we observe a dependence of the freezing efficiency on the mobility diameter, we note that interpretation of the mobility diameter is problematic for irregularly shaped particles such as mineral dust. Figure 4 is two scanning electron microscope (SEM)

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images of Arizona Test Dust, taken using a tilted stage, which provides at least an indication of the dust's three dimensional shape. It is apparent from the figure that characterizing the size of the dust by a single number (e.g. the mobility diameter) may be misleading in some cases. For example, the dust particle in the upper panel has an aspect ratio of approximately three to one in the dimensions that are clearly visible in the figure. The particle in the lower image is more compact; a single diameter for it would be more appropriate. Complementary studies of atmospheric aerosol with SEM and atomic force microscopy have shown that many particles are best characterized as cylinders, rather than spheres (Barkay et al., 2005).

The size, aspect ratio, and even surface roughness (see Sect. 3.4) of the dust may play a critical role in the particles' efficacy as freezing nuclei in the contact mode, but exploration of those topics is beyond the scope of this paper. We note, however, that the mobility diameters reported in Fig. 3 do not capture the full range of the particles' morphology.

3.4 Arizona Test Dust vs. kaolinite as contact nuclei

The final aspect of Fig. 3 that we address is the difference in the efficiency of ATD versus kaolinite. ATD catalyzed freezing at −17 and −18 °C at every mobility diameter we tested whereas we observed freezing with kaolinite only at −18 °C and only for the two largest mobility diameters. At −15 °C, ATD initiated freezing for the largest diameters; kaolinite did not. We conclude that Arizona Test Dust is a better contact nucleus than kaolinite.

The reason (or reasons) for ATD's enhanced activity over kaolinite in the contact mode is not known. ATD is more effective than clay minerals in the immersion/condensation mode (Fig. 3a, b, Hoose and Möhler, 2012). One hypothesis is that ATD is rougher than clay minerals as a result of the milling process it undergoes and that the greater surface roughness is more conducive to nucleation. Our examination of SEM images of size selected mineral dust particles also indicates that the smaller particles are more elongated (i.e. they have a higher aspect ratio). These points are

speculation at present, since no predictive theory for or mechanistic explanation of contact nucleation exists.

4 Conclusions

Tests of size selected Arizona Test Dust (ATD) and kaolinite at -15, -17 and $-18\,^\circ$C show that they are relatively ineffective as contact nuclei. For most sizes, less than one in ten thousand particles of ATD catalyzed freezing at $-18\,^\circ$C. At higher temperatures, the efficiency (defined as number of freezing events per number of particles deposited to the supercooled test droplet) was even lower. The experiments show that there is a temperature, size, and composition dependence to contact freezing by these two mineral dusts.

On average, the freezing efficiency for both ATD and kaolinite was higher for the lower temperatures. At $-15\,^\circ$C, ATD with mobility diameters of 125 and 62.5 nm did not catalyze freezing; larger diameters did initiate freezing at that temperature. Kaolinite exhibited contact nucleation activity only at $-18\,^\circ$C and only for mobility diameters of 500 and 1000 nm.

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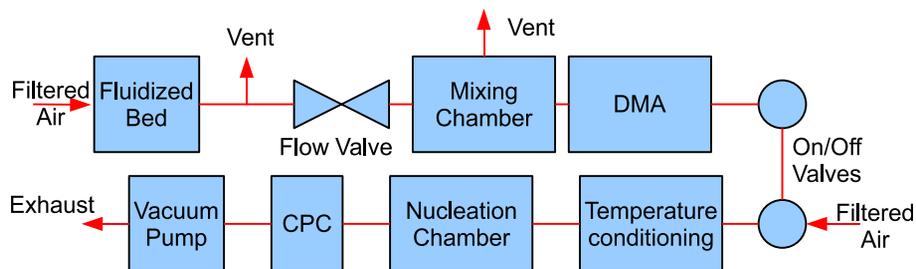
Fig. 1. Block diagram of the experiment. Aerosol particles are dispersed using a fluidized bed, then size selected with a differential mobility analyzer. The aerosol laden air stream is then cooled to a temperature close to the one selected for the experiment and pulled past the test droplet. The concentration is monitored with a condensation particle counter.

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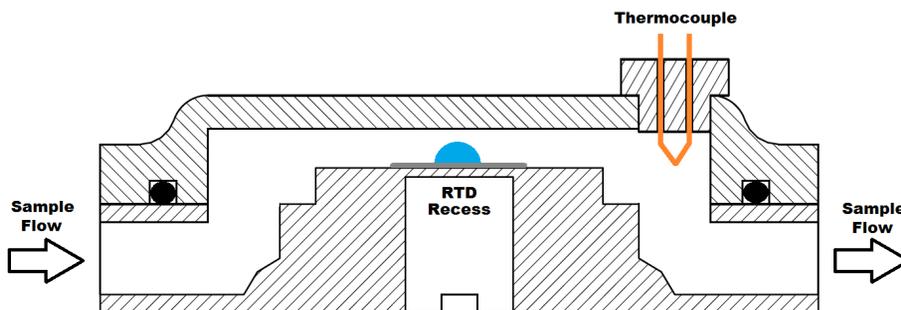
Fig. 2. Cross section of the nucleation chamber. The aerosol sample flows through the block as shown, past the droplet in the center of the chamber. The temperature control loop is via the rtd sitting under the droplet. Temperature is measured at three other places, including the thermocouple, which protrudes into the airstream. (The other two rtds are not shown in the Figure). The droplet sits on a silanized glass slide.
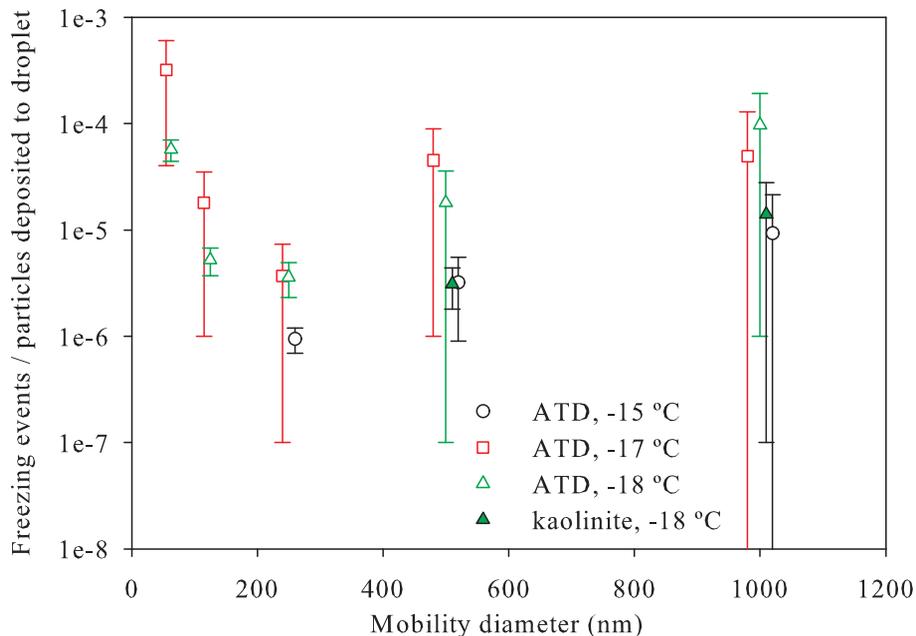
Fig. 3. Freezing efficiencies of Arizona Test Dust and kaolinite as a function of temperature and mobility diameter which show that neither dust is particularly effective as a contact nucleus for the temperatures tested. We tested mobility diameters of 1000, 500, 250, 125, and 62.5 nm. The mobility diameters are slightly offset in the plot to facilitate comparison of the data. Note that ATD produced no freezing events at $-15\,^\circ$C for dust of mobility diameters 125 and 62.5 nm while kaolinite did not catalyze freezing at any temperature warmer than $-18\,^\circ$C and then only for diameters of 1000 and 500 nm. The error bars are calculated from the uncertainty in the number of particles to which the droplet was exposed and the fraction of those that were deposited to the droplet (see above). In cases where the lower limit of the uncertainty was less than zero, we set the lower bound to 10^{-8} to enable plotting on the logarithmic scale.

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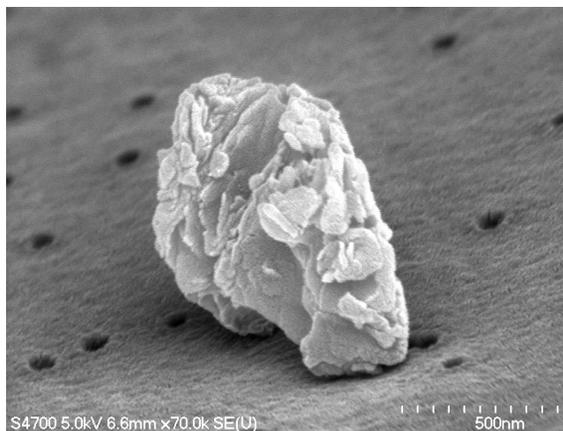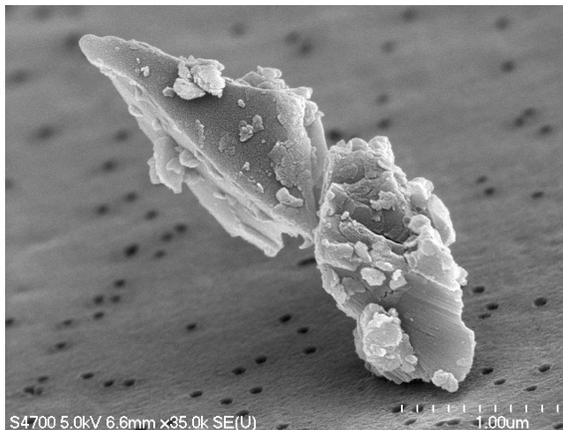
Fig. 4. Two SEM images of Arizona Test Dust on Nucleopore filters, illustrating the complexity of characterizing dust with a single diameter. The images were acquired by tilting the SEM stage. Neither of the two particles shown are spherical, though the particle in the lower panel is more compact.