

Interactive comment on “Measurements of ice nucleation by mineral dusts in the contact mode” **by K. W. Bunker et al.**

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General comments

We appreciate the reviewer's close reading of our experimental protocol and careful examination of Figure 3 in particular. The reviewer has pointed out some areas where we were not as clear as we had hoped to be. We welcome the chance for clarification. (See below for specific instances.)

That said, we also have some fundamental disagreements with the reviewer, primarily on the role of laboratory experiments in atmospheric science. As an example, the reviewer comments:

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There are major weaknesses in this study. For example, placing a water droplet on a cold plate is not the same as suspending a water droplet in air or let it fall freely. Therefore, results from cold plate studies cannot be extrapolated to contact nucleation in clouds readily.

While we agree that droplets on cold stages are not the same as droplets in clouds, we disagree that droplets on substrates cannot be used to gain physical insight into nucleation. Our aim in this study is to explore freezing when mineral dust is deposited at the air-water interface. Insofar as the curvature of the interface is neglected, the size of the water droplet is irrelevant. This assumption is valid unless there is reason to believe that, for example, the droplet's radius of curvature affects contact nucleation. We are aware of no such reason (unless the water droplet is on the order of nanometers in diameter), and therefore, a supercooled cloud droplet with a radius of ten microns and a supercooled three millimeter diameter droplet on a cold stage should behave in the same way in terms of whether they freeze when a mineral dust particle impinges upon them.

Our view is widespread in atmospheric science. As examples, consider two recent papers from this journal. Broadley et al. (2012) used a cold stage coupled with optical microscopy to examine freezing of water by illite and explicitly motivate their investigation by its relevance to processes in mixed phase clouds. Pinti et al. (2012) used emulsions of mineral dusts plus water in oil and lanolin to investigate the freezing behavior of water catalyzed by the dusts using differential scanning calorimetry. Again, their motivation was relevance to processes in mixed phase clouds. In those two cases, as in ours, the goal is to isolate the essential physics of the process under consideration.

We respectfully disagree with the reviewer. Experiments using droplets (even large ones) on substrates can be used to gain insight into nucleation.

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Specific comments

The reviewer's comments are italicized. Our responses are in roman text.

1. Laying a water droplet on a cold plate is not the same as suspending a water droplet in air or a free-falling water droplet in air. The difference in surface area can lead to significantly reduced collection efficiency of aerosol particles, as observed in this study. Therefore, the data obtained using this method require additional consideration and cannot be readily extrapolated to represent the real phenomena in the atmosphere.

2. The water droplet used here has a volume of 5 microliter. This is too big a water droplet and not representative of water droplets in clouds. This could be a cause to the low collection efficiency and low number of freezing events observed in the study.

Our response to the general issue of whether a droplet on a cold stage can tell us anything about processes in cold cloud is in the section above. The issue of whether a five microliter droplet can be used since most cloud droplets are smaller is similar.

The issue of collection efficiency is one where we failed to make ourselves understood. Collection efficiency is typically defined as the fraction of particles captured by a droplet given a volume that the droplet sweeps out (see e.g. equation 2 in Ladino et al (2011)). We have defined it as the fraction of the total number of particles through our system that are deposited to the droplet. (The geometry of the droplet does not enter our determination of this quantity.) To remove this source of confusion, we have revised the paper, replacing 'collection efficiency' with 'captured fraction'.

The reviewer asserts that the collection efficiency (now captured fraction, see preceding paragraph) is low. Low in comparison to what? In any case, we have designed our experiment such that aerosol particles are deposited to the surface of our test droplets; we measure the number deposited to the surface using scanning electron microscopy. It was not our intention to study collection efficiencies. Rather, our focus is on contact

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nucleation.

3. Temperature measurements in contact nucleation study are crucial and challenging. The authors performed experiments in a narrow window of temperatures (-15 °C, -17 °C and -18 °C). It is unclear that given the uncertainty in RTDs and thermocouples (± 0.5 °C and larger) how the authors are able to make distinct measurements between -17 °C and -18 °C and attribute data accordingly. It is necessary to have as wide a temperature range as possible to assess the importance of contact nucleation.

We agree that temperature measurements in any nucleation experiment are crucial and challenging. We chose -18 °C as a benchmark because it is in the range where immersion freezing is unlikely to be effective, and therefore ice initiation by contact freezing would be important. Immersion freezing becomes increasingly more likely for lower temperatures. (We are not claiming that contact freezing is unimportant for temperatures less than -18 °C, just that we chose to investigate a region where we believed that contact freezing would be the dominant freezing mechanism.) We, in fact, did try some experiments at temperatures higher than -15 °C, but did not observe freezing at those temperatures. We chose not to report that null result because we only did a few runs at the higher temperatures.

We state in section 2.2 (see line 10 on page 20296), "The temperatures we report are those from the RTD within the cavity under the slide (i.e. the RTD beneath the droplet stage)." The uncertainty of that RTD is much less than 0.5 K. We are confident that when we report those temperatures as different, they are, in fact, different. In the penultimate sentence of that paragraph, we state our uncertainty of the temperature of the droplet, ± 0.5 K. We recognize that there may be (probably are) temperature gradients within the droplet. It would be an error to ascribe a single temperature to the droplet. (See Gurganus et al. (2011) for a discussion on this point.) That said, we are confident that the droplet has a different average temperature when we set the temperature controller to -18 °C than when we set it to -17 °C. Finally, when we

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discuss the temperature dependence in section 3.2, we have not claimed a difference in the freezing efficiency for the two temperatures. We state just the opposite, "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."

We have revised the caption and legend to Figure 3 to re-emphasize the figure's main point. As stated in the Abstract, the fraction of particles active in the contact mode for any temperature ≥ -18 °C is below 10^{-3} .

4. Relative humidity is another key component in contact nucleation since evaporation of the water droplet can introduce thermophoresis and diffusiophoresis. The authors did not pay any attention to the issue of relative humidity in the experiments and did not correct for this in the analysis.

Thermo- and diffusiophoresis are certainly important considerations when calculating the flux of aerosol particles to the surface of the droplet. However, we took a pragmatic approach and circumvented those difficulties by implementing an experimental design that does not rely on a calculation to determine the flux of particles to the droplet. We expose the droplet to a stream of aerosol laden air, then measure the dust that is deposited using scanning electron microscopy. The effects of thermo- and diffusiophoresis are thus incorporated into our results. There is no need for a correction.

5. Due to the experimental design, the authors cannot easily measure or assess the number of aerosol particles deposited to the droplet. The authors "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." This is one of the largest sources of uncertainty in the measurement. The authors need to research other methods of assessing the number of aerosol particles that come into contact with the droplet.

6. Representation and quality of data is poor in the reviewer's opinion. On Figure 3, the
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error bars easily span several orders of magnitude for almost all data points, especially those for ATD at -17 °C. It is unclear why the uncertainty would be particularly large for -17 °C. It is also intriguing why the data for ATD at -17 °C and -18 °C are noticeably different by an order of magnitude.

The reviewer is correct in asserting that the determination of the number of particles deposited to the surface of the test droplet is one of the principal sources of uncertainty in our experiment. It is the primary source of uncertainty in the determination of the freezing efficiency (i.e. the ordinate in Figure 3), which we stated in the last sentence of section 2.4.

As for the magnitude of the uncertainty in the freezing efficiency, we note that the data are displayed on a logarithmic scale. Though the uncertainties may look large, they are modest. The largest uncertainty in the figure is $\pm 2.8 \times 10^{-4}$ for ATD at -17 °C. This is, in fact, smaller (in value) than the majority of the uncertainties reported by, for example, Svensson et al. (2009) (see their Figure 3). Other studies do not report uncertainties in the freezing efficiency at all (Ladino et al., 2011; Figure 8).

As we noted in the paper, the number of particles deposited to the droplet is the primary source of uncertainty in the determination of the fraction of particles passing through the system which are deposited to the droplet. That is, however, not the only source of uncertainty in the system. There is also an uncertainty in the number of particles passing through the system. As a conservative estimate, we use Poisson statistics and set the uncertainty in particle number equal to the square root of the total number. The variation in the total number of particles through the system is the reason for the variation in the magnitude of the uncertainties in Figure 3.

The reviewer also notes that the freezing efficiency for ATD at the two lowest temperatures do differ. They do not, however, disagree within the bounds of the uncertainty. We explicitly state in section 3.2, "Tests with ATD do not show a statistically significant difference between -18 and -17 °C."

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