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Atmospheric Chemistry and Physics Discussions

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 Dr. Ladino's comments on our paper. He raises some important points, and we are glad to have the chance to clarify them.

We revisited the paper by Pitter and Pruppacher and realized that we can estimate a freezing efficiency from the information in the paper; the freezing efficiencies we calculate from their data are actually comparable to ours. We have incorporated that information into Section 3.1. We have also revised the Discussion (section 3.2) to include a discussion of the onset of freezing by contact nucleation, comparing our

results with Pitter and Pruppacher (1973), Svensson et al. (2009) and Ladino et al. (2011).

Furthermore, we have revised our description of the determination of the number of aerosol particles deposited to our test droplets. In particular, Dr. Ladino's questions concerning some of those details, taken together with points raised by Reviewer #1, have motivated us to revise that section of the paper to make it more explicit that our approach is empirical. Our general approach does not depend upon the fact that the air in the chamber is turbulent, and phoretic effects are accounted for because we are measuring the number of particles that are deposited to the droplet. Our approach would work even if the flow in the chamber were laminar and the droplet were slowly growing by condensation rather than slowly evaporating. We have revised section 2.4 in the paper to highlight this.

Specific comments

Dr. Ladino's comments are italicized. Our responses are in roman text.

1. Important literature studies are missing in the introduction (e.g., Pitter and Pruppahcer (1973), Rosinski and Nagamoto (1976), Levin and Yankofski (1983), Diehl and Mitra (1998), Diehl et al. (2002), Von Blohn et al. (20050, Durant and Shaw (2005), among others) and the role of contact freezing in cloud formation and its role in the hydrological cycle and the global radiative balance is not fully explained. Why is it important to study contact freezing and why is it believed to be the most efficient ice nucleation mode? Please provide some evidences from previous studies. Although the focus of the paper is mineral dust particles, it is worthwhile to discuss the very high onset freezing temperatures observed from bioaerosols (e.g. Levin and Yankofski (1983), Diehl et al. (2002)).

We have a different view of the purpose of the Introduction to a publication than does Dr. Ladino. Rather than writing a comprehensive survey, we were introducing contact nucleation by mineral dust. Many of the studies cited above are of contact nucleation, but due to some other type of material (as noted in the comment above). There are venues (e.g. *Reviews of Geophysics*) for comprehensive reviews. We feel that the Introduction to a journal article should do just that, introduce the topic at hand briefly and concisely.

We did provide a rationale for studying contact nucleation in the second paragraph of the Introduction. We also motivate study of contact nucleation in paragraphs 4 and 5 of the Introduction, where we cite specific studies showing that contact nucleation may be effective for higher temperatures and smaller particles than other modes.

We have added discussion of results from Rosinski and Nagamoto in section 3.3.

2. The obtained experimental results are compared with Svensson et.al (2009) and Ladino et.al (2011) but they are not compared with Pitter and Pruppacher (1973) who also used kaolinite particles in the contact freezing mode. Are the kaolinite samples used in Ladino et.al (2011), Svensson et.al (2009) and the present study the same? The purity of those samples is an issue and could explain the observed differences in the onset freezing temperatures as highlighted by Broadley et al. (2012).

We have revised the Discussion to include a discussion of Pitter and Pruppacher's results. Using their fraction of frozen droplets (their Figure 2), and the number of aerosol particles collected by the droplets, we estimate a freezing efficiency of 10^{-4} for kaolinite at -11 C, which is comparable to our freezing efficiency for kaolinite at -18 C. We note that their samples were not size selected and contained particles as large as 30 μm in diameter.

As we stated in section 2.1, the kaolinite powder was from Fluka, as was the kaolinite used by Ladino et al. (2011) and Svensson et al. (2009). As we stated in the paper,

a comparison of our freezing efficiencies with those from Svensson et al. and from Ladino et al. is difficult because of the discrepancy between our freezing efficiencies and what is reported in those two papers. Our freezing efficiencies are comparable to those we have calculated from the information in Pitter and Pruppacher.

However, motivated by this comment, we revised the paper to include a discussion of freezing onset in the contact mode. For that quantity, our results are comparable to all of the papers just cited.

3. As highlighted by Reviewer #1, the dynamical forces responsible for the collisions between the aerosol particles and the droplets in this study are not discussed. Do the collision rates agree with theoretical calculations? How important are the phoretic forces in your system? What is the relative humidity of the air mass carrying the aerosol particles? Is the droplet size controlled/measured and is droplet evaporation considered?

4. Since the authors claim that there is turbulence inside the chamber, how does it affect the collision rates as compared to a laminar flow (e.g., Vohl et al. (1999))? Are there any particle losses due to turbulence?

As we discussed in our reply to Reviewer #1, phoretic forces are certainly important considerations when calculating the flux of particles to the surface of the droplet. However, we deliberately implemented an approach that bypasses those difficulties. 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.

We are confident that the turbulence in the chamber enhances particle deposition to the surface of the test droplets. We have not attempted to quantify that because our approach to the determination of the number of particles deposited to the droplet is empirical. Our focus is on the nucleation, not on the details of how the particles are

deposited to the surface of the droplet.

There are undoubtedly particle losses within our system due to the turbulence. But again, those losses are automatically accounted for in our approach because we measure the particles deposited to the droplet for given number of particles measured by the CPC (see Figure 1 in the paper).

As stated in the paper (1st paragraph, section 2.1), the dew point of the air we use is $\sim$ -40 C. We use 5 microliter droplets as dispensed from a Hampton microliter syringe. Droplet evaporation is considered insofar as our method of determining the number of particles deposited to the droplet is empirical. The droplet was slowly evaporating during the tests we did to determine the number of particles that hit the surface of the droplets so we believe that the results should be applicable to the contact nucleation tests, where the droplet was also slowly evaporating.

5. Since the collision efficiency strongly depends on the aerosol particle size, I am concerned about the "monodispersity" of the size selected particles (especially for large particles). How accurate are the IN particle sizes and how narrow are the monodisperse size distributions?

We were careful to specify that the diameters specified in Figure 3 are the electrical mobility diameters. The DMA is selecting for the correct electrical mobility. (We have verified this using polystyrene latex spheres and NaCl aerosol.) However, as we discuss in the paper (see the final two paragraphs of section 3.3), electrical mobility may not have a one-to-one correspondence to other measures of particle size (e.g. area equivalent diameter, as measured from SEM images) for irregularly shaped particles like mineral dust. We are preparing a separate paper on just this topic. Just what exactly the mobility diameter of aspherical, rough particles represents in terms of other measures of the particles' sizes is beyond the scope of this paper.

6. It was mentioned in the text that the number of deposited particles was calculated

only for 500 and 1000 nm particles but not for the smaller ATD particles presented in figure 3. Why not? How is it possible to present an error bar for those small particles? What is the utility to calculate CE and how was it used to interpret your results? How do you define the number of freezing events if only one droplet is used?

Each freezing event represents a separate droplet. Once a droplet froze, we disassembled the chamber and replaced the droplet for further tests. We have added a paragraph in section 2.3 clarifying this.

As noted above and in the response to Reviewer #1, the collision efficiency is not calculated, but determined empirically.

We did tests to determine the number of particles deposited to our test droplets as described in section 2.4. Because the air flow within the chamber is turbulent, we do not expect the fraction of the total number of aerosol particles within the airstream which are deposited to the droplet to depend strongly on the particle size. For a laminar flow, it would certainly depend on size, since the diffusion coefficient is inversely proportional to the particle size. In contrast, the turbulent diffusion coefficient is independent of size. We did not find a size dependence with the tests we conducted.

SEM scans of the smallest particles (62.5 nm mobility diameter) reveal that they are the most aspherical and have the largest variability in the area equivalent diameter given a specified mobility diameter. This would bias our procedure for determining the number of aerosol particles deposited to the droplets. As mentioned above, we are preparing a separate paper addressing this issue.

We have added a sentence to the last paragraph of section 2.4, clarifying the fact that the captured fraction determined for mobility diameters of 500 and 1000 nm is used for all contact nucleation tests.

7. Ladino et al. (2011) did not determine any experimental collection efficiencies. Those values were determined in Ladino et al. (2011a), however the methodology used

to calculated CE in Ladino et al. (2011a) is wrongly described at present. Aerosol particles and cloud droplets were injected into CLINCH to allow collisions between them. At the bottom section of the chamber, the total injected droplets and the aerosol particles captured by those droplets were collected in a plastic bottle in order to determine the total aerosol mass (by ICP-MS) to calculate the collection efficiencies.

We apologize for the error. We have added a reference to "Experimental Study of Collection Efficiencies between Submicron Aerosols and Cloud Droplets" in *J. Atmos. Sci.* and revised the paper to correctly describe the procedure used in that paper to determine the number of aerosol particles collected per droplet and thus the collection efficiency of the droplets.

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