

The manuscript by Nagare et al. describes a comparison between immersion and contact freezing in the ETH Collision Ice Nucleation Chamber and the Immersion Mode Cooling Chamber/Zurich Ice Nucleation Chamber. Unlike previous studies, an enhancement of contact mode over the immersion mode was not seen.

I am supportive of publication. That said, there is some ambiguity in the way that the freezing efficiency is defined and/or used in the paper that should be clarified. In addition, I think that the authors should discuss the discrepancy between their measurements and those of other groups in more detail.

Freezing efficiency

I think I am not quite following how FE is defined and used. In the text FE is defined as

$$FE = \frac{FF}{N} \quad (1)$$

where FF is the fraction of droplets that have frozen and N is defined as the number of collisions between a droplet and aerosol particles. If I combine the definition of FF and equation 1, the result is

$$FE = \frac{\text{freezing events}}{\text{total droplet-aerosol collisions}} \quad (2)$$

which can be interpreted as the probability that a single collision between an unfrozen droplet and an aerosol particle results in a freezing event. This is the interpretation, used for example, in Hoffmann et al. [1] and Niehaus et al. [2].

My confusion is from the discussion around the case when the first collision between aerosol and droplet results in a freezing event. If that is the case, then intuitively, $FE = 1$. I don't understand the point of Equation 4 in the manuscript. The denominator is defined as the fraction of unfrozen droplets after N collisions, but $1 - e^{-N}$ is defined in Nagare et al. (2015) [3] as FF (see Equation 19). If the denominator is FF , then Equation 4 in the manuscript makes sense because it reduces to $FE = 1$.

I am also confused by the statement in lines 6 and 7 on page 6. "... FE should be independent of residence time when..." Shouldn't FE always be independent of residence time? The number of freezing events will depend on the residence time, since a longer residence time is a greater probability that the droplet will collide enough particles to catalyze freezing, but once you have normalized by the number of droplet-aerosol collisions, FE should be a probability, which is independent of time.

Figure 4 is also a source of confusion for me. How is $FE > 1$ possible, as is shown in panel b for $T < 245$. If I understand the definition of FE correctly, this implies that there are more freezing events than there are aerosol-droplet collisions. Under the assumption of panel b, shouldn't all these data points collapse to 1 for $T \leq 245$? I am also not understanding how FE is a function of the aerosol concentration. FF should be a function of the concentration, but the point of normalizing by the number of collisions is to remove that dependence.

Immersion vs. contact freezing

Based on a difference in the freezing efficiency with a residence time of 2 seconds vs. 4 seconds, the authors conclude that contact freezing is not enhanced over immersion freezing, at least not for the Arizona Test Dust, and perhaps not for silver iodide. This claim is also supported by the comparison to immersion freezing results using IMCA/ZINC. (See my discussion above concerning confusion about *FE* and a time dependence.)

This is a striking result, considering the previous work in the field. Gokhale and Goold [4] tested AgI for contact freezing, and showed that freezing was initiated at temperatures close to $-5\text{ }^{\circ}\text{C}$ in the contact mode while temperatures closer to $-15\text{ }^{\circ}\text{C}$ were necessary for immersion mode freezing. Sax and Goldsmith [5] also tested AgI, in a cold room with freely suspended droplets, and saw indications of a shift to lower temperatures for the onset of freezing for contact vs. immersion freezing. These results are directly relevant to the experiments described here. While the freezing efficiency is not reported in either of these papers, they do discuss a shift in the onset freezing temperature, which can be compared, for example, to Figure 1 in this paper, which unambiguously shows that droplets begin to freeze at about $-8\text{ }^{\circ}\text{C}$ in the immersion mode and only at $-13\text{ }^{\circ}\text{C}$ in the *FE(4s)* experiments in the contact mode. (See also Gokhale and Spengler [6] and Gokhale and Lewinter [7]. Tests with AgI were conducted in both of those studies as well.)

Similarly, Pitter and Pruppacher [8] tested kaolinite in a wind tunnel and found a clear shift to lower temperatures for the onset of freezing when changing from contact to immersion mode experiments. Niehaus et al. [9] report that they ran experiments in which a freezing event occurred, then melted the droplet and cooled it back to the original temperature. No freezing events occurred. Niehaus et al. concluded that these tests, in which the same aerosol were compared against themselves, that contact freezing was more probable than was immersion mode. (ATD was among the substances they tested.) Though AgI, kaolinite, nor ATD was tested in the study, in an even more convincing case of an enhancement of contact freezing over immersion mode, Durant and Shaw [10], visually confirmed a shift in the freezing temperature when a particle was at the air-water interface. (See their Figure 1.)

As noted above, I am in favor of publication because I believe that more information on these systems will help the community to unravel the complexities and, perhaps, make a determination as to just how important contact freezing is in Earth's atmosphere. That said, I believe that the authors should place their results more clearly in the context of previous work.

Other comments

Discussion of uncertainties:The authors discuss the uncertainty in the frozen fraction as stemming from the classification uncertainty of the IODE detector (see, e.g. the caption to Fig. 5.) Shouldn't the uncertainty in the number of

particles that have collided with the droplet be included when showing FE ? I realize that different values of N are used, depending on the value of CE , but even so, there is an uncertainty in CE and thus N . An indication of that uncertainty would help in interpreting these plots.

Discussion of contact freezing mechanisms: Pg. 8, line 29. “This mechanism was refuted by Fukuta (1975b).” “Refute” implies that he disproved Cooper’s mechanism. I think he rejected it, but did not disprove it.

Pg. 9, line 18. “This indicates that collision itself does not increase FE ...” There is evidence that the collision can increase FE . Davis et al. [11] recently showed that salt particles can initiate efflorescence upon contact with a supersaturated solution of a different salt. Niehaus and Cantrell [12] observed freezing events initiated by soluble salts at temperatures above the eutectic. Finally, Yang et al. [13] have observed freezing which may be in response to the movement of the triple line, as from a collision.

pg. 13, line3 28-29: “One reason for this may be that in CLINCH and IMCA/ZINC experiments the particles are free to realize the energetically most favorable position in or on the droplet.” This may be true, but it is also true for many of the other experiments noted above.

pg. 14, line 5: “Own observations...” I think you mean to start that sentence with “Our”.

Table A1: Include N .

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