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.

We thank the reviewer for the careful reading of the manuscript and the suggestions for improvement. We address the comments below point by point (in italic):

Freezing efficiency

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

$$FE = \frac{FF}{N} \tag{1}$$

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

$$FE = \frac{freezing\ events}{total\ droplet-aerosol\ collisions} \tag{2}$$

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 dont 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.

There is an ambiguity in the freezing efficiency determined by the CLINCH experiment when the droplet collides with more than 1 particle while it passes through the chamber, because in this case, it is not clear which collision is responsible for freezing. A second or a third collision might occur although they are not needed to freeze the droplet when it has frozen already on the first collision. To convert from FF to FE, only collisions of particles with liquid droplets should be counted. The reviewer is right that Eq. 4 should only be used when FE = 1. This is the case for AgI at temperatures for which the plateau condition applied in Nagare et al. (2015) is valid. Using Eq. (1), FE at the highest AgI concentration is lowest. A result that is not easily explained. Therefore, we introduced Eq. (4) for this case.

From this it can be concluded that experiments should be run such that the average number of collisions is close to but below one. In this case, Eq. (1) of the manuscript can always be used. We have improved the discussion of Eqs. (1) and (4) in the revised manuscript.

I am also confused by the statement in lines 6 and 7 on page 6. "...FE should be independent of residence time when..." Shouldnt 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.

In the case of collisional contact freezing, FE should be indeed independent of time. However, in the case of immersion freezing and adhesion freezing, FE depends on time. This is why we use FE(4s) > FE(2s) as criterion against collisional contact freezing. We have improved the text in the revised manuscript to make this clearer.

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, shouldnt all these data points collapse to 1 for $T \le 245$? 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.

FE > 1 is due to homogeneous freezing and measurement uncertainties. We add this statement to the revised manuscript. Indeed, FE should not be a function of concentration.

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 °C in the contact mode while temperatures closer to -15 °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 °C in the immersion mode and only at -13 °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.)

We expected to observe a higher freezing efficiency for AgI in CLINCH than in IMCA/ZINC. We were very astonished that we observed the opposite. We went therefore back to literature and studied the previous work. Because so many studies have been performed with AgI as ice nucleus, we decided that a profound discussion of all this literature would make the paper too long. Therefore we wrote a companion paper, which reviews ice nucleation studies with AgI. This paper is now also published in ACPD: Marcolli C., Nagare, B., Welti, A., and Lohmann U.: Ice nucleation efficiency of AgI: review and new insights, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-142, 2016.

In this paper, we refer to the literature mentioned by the reviewer. On page 4, we write:

Gokhale and Goold (1968) performed contact nucleation experiments by sprinkling AgI particles on supercooled droplets on a hydrophobic plate. They observed that the particles (5 – 400 µm in diameter) remained on the surface of the drops and initiated freezing at the initial stage temperature of 268 K. However, they did not quantify the number of particles present, which precludes an evaluation in terms of surface area. They performed similar experiments for an AgI smoke produced from an AgI string generator with particle diameters from 50 – 100 nm. These particles initiated freezing of 50 % of droplets at 263 K when the stage was cooled at a rate of 1.3 K/min. Gokhale and Goold (1968) concluded that these freezing temperatures are 5 – 10 K higher than the ones observed by Hoffer (1961) for droplets embedded in an oil with immersed AgI particles and attributed it to an enhanced freezing probability for dry particles on a surface compared with particles immersed in the droplet. However, a strict comparison is not possible because in both studies, information is lacking to quantify the surface area present per droplet. In a follow-up study, Gokhale and Lewinter (1971) monitored the freezing process of 2 mm water droplets with a movie camera and observed that nucleation was initiated at the point of particle contact and continued from there over the entire surface of the drop. The interior of the drop froze at a much slower rate.

On page 5, we write:

Sax and Goldsmith (1972) performed contact and immersion freezing experiments in a cloud chamber. Freely falling droplets with diameters of 40 – 160 µm (average: 100 µm) intercepted a horizontal aerosol stream of $5\cdot10^6$ cm^{-3} AgI particles with 30 nm diameter (size range from 10 – 40 nm) for 0.04 s (1 cm in vertical extent). The aerosol was produced by heating an AgI-coated resistance wire to T = 700°C in a nitrogen stream. For contact freezing experiments the droplets were brought in thermal equilibrium before intercepting the aerosol stream. After coagulation with the AgI particles, the droplets proceeded into an observation chamber where frozen droplets were *distinguished visually from liquid ones. Coagulation of 100 µm droplets with 30 nm particles were dominated by Brownian motion. Assuming a collision efficiency of ca. 0.3, around 100 particles would be captured by the droplet (note that this number is higher than the collection of only 1 particle estimated by Sax and Goldsmith, 1972). For immersion freezing experiments, the droplets passed the aerosol stream at T > 273 K, before they were cooled to the target temperature. Residence time in the chamber was around 4 s. Immersion freezing occurred at 2 K lower temperature than contact freezing.*

In Marcolli et al. (2016) we give possible explanations why FE in IMCA/ZINC was higher than in CLINCH. To avoid telling the same in two papers, we do not want to extend the discussion in the present manuscript but prefer to refer to the companion paper.

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.)

Thank you for pointing out the study of Pitter and Pruppacher. We now refer to it in the revised manuscript. We referred to the Niehaus et al. (2014) paper in the introduction and discussed it in Section 5.5. We add in the revised manuscript that Niehaus et al. concluded that contact freezing was more probable than immersion freezing for the ice nuclei that they investigated.

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.). Shouldnt 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.

We agree that the main uncertainty for the comparison of freezing mechanisms results from uncertainties in CE. We think that it is more transparent to show results with different assumptions of CE rather than to draw huge error bars. To make it clearer that the uncertainty of FE is due to the uncertainty of CE, we added the following sentence to the Section 4.3 in the revised manuscript: "The difference in FE between panels (c) and (d) must be considered as an uncertainty in FE due to the lack of reliable theoretical values of CE in the investigated temperature and particle size range." In Section 4.4 we add: "For panel (a) the theoretical formulations were used, while panels (b) and (c) give the upper and lower limit of FE, respectively."

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.
Thank you for pointing this out, we change it as suggested.

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.
Thank you for pointing out these papers. Niehaus and Cantrell indeed made plausible that the collision triggered freezing. To have an effect, the particles that collided with the droplets needed to be large so that the impact led to a mechanical disturbance. For 10 µm NaCl particles no effect was observed. Therefore, this process is likely not active in our experiment. However, we mention this study in the revised manuscript to illustrate that the collision may induce freezing when the impact is large enough. Davis et al. investigated contact efflorescence by bringing soluble salts in contact with supersaturated solution droplets. In this case, nucleation has to occur immediately after contact before the salt particle dissolves in the solution droplet. There is no immersion mode setup conceivable to compare the freezing efficiencies. Therefore it cannot be concluded whether the collision itself is responsible for freezing. In the experiment by Yang et al., no collision was involved in ice nucleation but a movement of the three-phase contact line when the droplet adjusted to the changing electric field.

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.
Indeed, the experimental conditions have to be considered in detail. In case of AgI, a detailed discussion is given in the companion paper.

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

Thank you for pointing this out.

Table A1: Include N.

We add N in the revised manuscript.

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