On behalf of my coauthors, I would like to thank the reviewers for their careful reading and constructive comments. We appreciate their recognition of the need for contact-freezing measurements and of the value of our new C-ICE experimental approach. The reviewers did raise important questions regarding the validation of contact freezing and the possible contribution of immersion freezing. These comments motivated several additional experiments and analyses that have substantially strengthened the manuscript.

In particular, we have added C-ICE experiments using soluble ammonium sulfate particles as a mode-specific validation of impact-initiated contact freezing, direct post-melt observations of quartz particles to independently evaluate particle collection, and AgI immersion freezing experiments to assess the potential contribution of immersion freezing. We believe that these additions, together with the corresponding revisions to the experimental description and interpretation, have significantly improved the manuscript.

Below we address each reviewer comment point by point. The reviewer comments are shown in blue, our responses in black, and the revised manuscript text in green. Line numbers refer to the revised manuscript, and the corresponding changes are highlighted in the revised version.

Referee #2

The manuscript introduces a new experimental approach to quantify the temperature- dependent contact freezing probability for well defined aerosol particles in the laboratory. This is still an under researched field and therefore of interest for the cloud science community.

There are several critical points however, that should be resolved before publication.

Author's response: We appreciate your affirmation, comments, and suggestions on our work. We have expanded the Methods, added direct tests of freezing-mode ambiguity and particle collection, and revised the CE discussion accordingly. Each comment has been addressed point-by-point in the following contents, and the manuscript has been completely revised.

My main issues are:

The manuscript lacks a thorough description of the experimental setup and the measurement procedure and a detailed discussion of possible sources of errors. Questions that need to be answered:

1. How was the aerosol prepared (especially dry or wet dispersion?) Are particles allowed to collide with the droplet from the beginning (i.e. T=ambient) or when (or at which temperature) is the aerosol flow admitted?

   We appreciate this significant comment.

   The aerosols used in C-ICE are generated as dry solid particles using a custom-built solid aerosol generator with nitrogen as the carrier gas; that is, they are not introduced by nebulizing an aqueous suspension. Depending on the experiment, the dry aerosol is diluted and/or size selected before dehumidification and thermal conditioning. The aerosol stream is deliberately introduced only shortly before the expected freezing temperature range, which is established in preliminary experiments for each material or condition and then applied consistently within the corresponding series. This reduces both droplet evaporation and cumulative particle loading before the relevant temperature window.

"A custom-built solid aerosol generator is used to produce a stable particle-laden airflow. The generated aerosol is subsequently conditioned through dilution and/or size selection, allowing either polydisperse or size-selected particle populations to be delivered." (Lines 116-118)

"The dehumidifier setpoint is selected according to the aerosol material, its expected freezing temperature range, and the required drying conditions. For a given sample or experimental series, the selected setpoint is held constant. This configuration allows both the aerosol particles and the nitrogen carrier gas to approach a selected target conditioning temperature before collision, while simultaneously reducing the water vapor content of the aerosol stream." (Lines 124-127)

"Note that the aerosol stream is not applied continuously throughout the entire cooling ramp. Instead, aerosol delivery is initiated shortly before the expected freezing temperature range of the material under investigation. The approximate freezing range is first established from preliminary experiments, after which a consistent aerosol admission protocol is used for the corresponding experimental series. This strategy limits prolonged exposure of the droplet to the dry aerosol flow and minimizes excessive droplet evaporation and cumulative particle loading before the relevant freezing temperature window. Limiting particle accumulation is particularly important for reducing the potential contribution from delayed immersion freezing by particles collected prior to the observed freezing event." (Lines 139-145)

2. From this moment on, aerosol particles might be scavenged into the droplet and might cause immersion freezing at a later time. Please specify the rate of collisions between particles and the droplet, If I judge from Fig. 7b, ~10 particles collide with the droplet every second, so several hundred particles have been scavenged by the droplet before freezing is observed. How can contact freezing be disentangled from immersion freezing under these conditions?

We appreciate this constructive comment.

We first clarify that the ~10 s^{-1} rate applies to the polydisperse aerosol over part of its size range; it does not apply to the DMA-selected 200 and 800 nm experiments. The median effective collision rate for both size-selected AgI populations is approximately 0.17 s^{-1}. We now state this explicitly.

More importantly, the revised manuscript no longer relies on AgI alone to identify freezing mode. We added ammonium sulfate as a soluble, mode-specific test: after entry into the pure water droplet, the dry crystalline particle dissolves, eliminating a persistent solid ammonium-sulfate surface that could drive conventional delayed immersion freezing. We also performed AgI immersion freezing measurements across five concentrations and restrict aerosol exposure to the relevant temperature window. In conclusion, AgI is used as a benchmark material now to evaluate the freezing-mode sensitivity and size-resolved performance of C-ICE.

"Silver iodide (AgI) was used as a benchmark material to further evaluate the freezing mode sensitivity and size-resolved performance of C-ICE. In addition to the contact freezing measurements, immersion freezing experiments were performed using the same cold stage and hydrophobic glass substrate. Following the droplet array approach used in Chen et al. (2021), 70 droplets of 1 µL AgI suspension were analyzed per experiment at the same cooling rate of 1 °C min^{-1} used in C-ICE. Frozen fractions and 95% confidence intervals were calculated using the Wilson score method in Eq. (2). A series of

AgI suspensions from 0.0001 to 1 wt% was prepared to span and exceed the particle loading relevant to the polydisperse AgI contact freezing measurements. In C-ICE experiments, the upper-bound AgI mass concentration in the 1 µL target droplet was estimated from the OPC-measured particle size distributions assuming CE_{upper}=1, i.e., that all nozzle-delivered particles were collected. Assuming spherical particles with an AgI density of 5.68 g cm^{-3}, the resulting upper-bound concentration has a median of 0.0051 wt% and an interquartile range of 0.0037-0.0076 wt% (Table S3). The actual concentrations of the prepared immersion freezing suspensions are summarized in Table S4." (Lines 544-554)

"Figure 8 compares the frozen fraction behavior of AgI in immersion and impact-initiated contact freezing. For immersion freezing, the temperature of the first observed freezing event ranges from approximately -4.6 to -7.2 °C across the concentration series, warmer than the polydisperse AgI C-ICE response. However, the immersion freezing curves show a pronounced concentration dependence, shifting systematically toward lower temperatures as AgI concentration decreases. At 0.0001 wt%, the immersion T_{50} approaches that of the polydisperse C-ICE experiment. The corresponding pure-water/background curves are comparable, indicating that the differences among the AgI datasets arise primarily from particle exposure rather than from differences in the cold-stage baseline. A similar mode dependence was reported by Nagare et al. (2016), who observed that heterogeneous freezing of AgI in the CLINCH contact freezing experiments occurred at lower temperatures than in the corresponding IMCA-ZINC immersion experiments.

These results illustrate an important point for freezing mode comparison. When the immersion mode activity of a material is substantially weaker than its contact freezing response at comparable particle loading, freezing observed following aerosol-droplet interaction can be attributed more confidently to impact-initiated contact freezing. In contrast, for highly immersion active materials such as AgI, particles collected before the observed freezing event may contribute to the measured response, particularly as particle loading increases. Therefore, a rigorous quantitative comparison of intrinsic contact and immersion freezing efficiencies requires careful matching of particle loading, surface area, and particle-water interaction history. The AgI experiments presented here are intended primarily as a benchmark of C-ICE performance and as a direct assessment of the potential immersion contribution, rather than as a complete quantitative ranking of the two freezing modes. The ammonium sulfate experiments in Sect. 3.2 provide the more direct mode-specific validation of impact-initiated contact freezing." (Lines 562-579)

"The median effective particle collision rate is 0.17 s^{-1} for both 200 nm and 800 nm populations, with corresponding mean rates of 0.20 s^{-1} and 0.29 s^{-1}, respectively (Table S5). Thus, the warmer freezing of the 800 nm particles occurs under comparable particle-delivery rates, further supporting a particle-size dependence of the AgI freezing response. In contrast, the polydisperse aerosol produces substantially higher collision rates over much of the size range. Submicron particles alone contribute on the order of ~10 collisions per second, with additional contributions from coarse-mode particles (~1 collision per second). The greater frequency and broader size distribution of particle-droplet collisions likely contribute to the warmer and broader freezing response of the polydisperse particles. The frozen fraction behavior therefore reflects both particle ice-nucleating properties and the size-dependent frequency of effective particle-droplet collisions. Particle number concentrations and size distributions for the AgI particles are shown in Figs. S10 and S11 and Table S6." (Lines 615-624)

"Overall, the AgI results demonstrate that C-ICE can resolve both freezing-mode effects and particle-size-dependent C-ICE freezing response under controlled collision conditions. AgI serves as a useful benchmark for evaluating C-ICE performance, while also illustrating why quantitative contact-immersion comparisons must explicitly account for particle loading, surface area, and interaction history." (Lines 625-628)

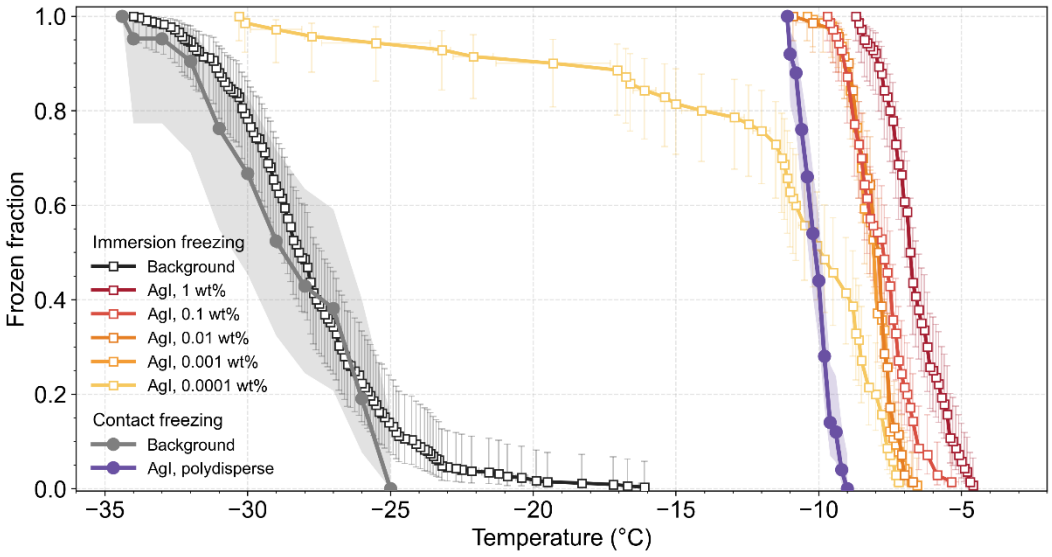


Figure R2.1. Comparison of AgI immersion and impact-initiated contact freezing. Frozen fraction as a function of temperature is shown for immersion freezing experiments using AgI suspensions spanning 0.0001–1 wt% (open symbols), polydisperse AgI impact-initiated contact freezing experiments in C-ICE (purple filled symbols), and the corresponding particle-free/background measurements. Immersion freezing experiments used 70 droplets of 1 µL per condition on the same cold-stage platform and at the same cooling rate as the C-ICE experiments. Error bars and shaded regions represent 95% confidence intervals calculated using the Wilson score method. Actual AgI suspension concentrations are listed in Table S4. (Fig. 8)

3. Can deposition ice formation on the particles in the precooler or in the high humidity environment of the experiment chamber be excluded? The precooler might include regions with a temperature gradient normal to the flow direction and thereby may create regions of supersaturation similar to a diffusion chamber.

We are grateful for this comment.

We have clarified the intended thermodynamic role of the dehumidifier. In the representative one-dimensional calculation, the gas can reach ice saturation within the cooled section, after which excess water vapor is removed at the cooled inner wall. The outlet stream is therefore dried rather than maintained in a sustained supersaturated state as in a diffusion chamber. In addition, the cold-stage chamber is continuously purged with dry N_2 and remains globally subsaturated with respect to liquid water. We do not claim that the one-dimensional model excludes every possible microscopic or transient local supersaturation within the conditioning tube, but sustained deposition nucleation conditions in the aerosol stream are not expected under the operating concept. The manuscript now explicitly identifies

Fig. 4 as a representative model calculation rather than a direct measurement of local humidity fields.

"At the outlet, the aerosol particles and nitrogen carrier gas have been both dried and thermally conditioned toward the selected setpoint before entering the cold stage. Because the dehumidifier temperature is fixed during an experimental series while the cold-stage temperature continues to decrease, the conditioned aerosol stream is not necessarily at the same instantaneous temperature as the target droplet. The dehumidifier provides a reproducible and sufficiently dry thermodynamic state prior to particle delivery. Together with the continuous dry nitrogen purge of the cold-stage chamber, this conditioning suppresses widespread background condensation while allowing localized aerosol-flow-induced and freezing-induced breath figures to remain readily distinguishable." (Lines 404-410)

"The -15 °C condition illustrates one typical operating scenario and does not represent a universal setpoint for all C-ICE experiments. The dehumidifier temperature is selected according to the aerosol material, expected freezing-temperature range, and drying requirements and is held constant within a given experimental series." (Lines 419-421)

4. On collision efficiency (CE): impact CE should depend on the distance between nozzle and droplet (like in an impactor). What was the distance between nozzle and droplet? Why is this not considered when calculating impact CE? What was assumed for the charges on droplet and aerosol particle? Even tiny charges on the droplet may play a role here. Were polarization (image charge) effects taken into account?

We are grateful for this comment.

The nozzle-to-droplet distance is 5 mm, and 4.0 m s^{-1} is the nozzle-exit gas velocity rather than the particle velocity at the droplet surface. We agree that nozzle spacing and the resulting impinging-jet flow can affect particle trajectories. The analytical CE_{model} is therefore treated as a physical baseline; the actual C-ICE geometry is constrained empirically through CE_{exp} at 200 and 800 nm and independently evaluated using post-melt quartz collection.

For the size-selected experiments, the aerosol charge distribution was conditioned using a neutralizer prior to DMA classification, although the downstream particle charge distribution was not measured directly. The droplet charge was neither measured nor actively controlled. For the broader dry-generated aerosols, additional charging may also occur during generation and transport. The electrical term in the CE_{model} is evaluated using a representative empirical charging parameter to assess its possible relative contribution, not as a direct measurement. Polarization/image-charge effects are not explicitly resolved. We now state this limitation and identify dedicated charge-controlled experiments as a future requirement.

"For size-selected experiments, aerosol particles were charge equilibrated using a neutralizer and subsequently classified with a differential mobility analyzer (DMA, model 3080, TSI Inc., USA) at nominal electrical mobility diameters up to 1.0 μm. Contributions from multiply charged larger particles cannot be completely excluded." (Lines 119-121)

"In this work, a nozzle orifice diameter of 0.8 mm, an aerosol flow rate of 120 cm^3 min^{-1}, and a nozzle-to-droplet distance of 5 mm were employed, corresponding to a mean gas velocity of 4.0 m s^{-1} at the nozzle exit. This configuration provides directed particle delivery to the target droplet, while the

impinging flow may generate surface shear and internal circulation within the droplet. The resulting particle-droplet collision efficiency is quantified and discussed in Sects. 2.3 and 3.3." (Lines 135-139)

"Particle charge distributions were not measured directly in this study. Although particles generated and transported under dry conditions may acquire charge through particle-particle and particle-surface interactions, the detailed influence of particle charging and image-charge effects is not explicitly resolved here. Dedicated charge-controlled experiments would be required to quantify these effects." (Lines 244-247)

"Note that the temperature, RH, and electric force values are modeling assumptions rather than direct measurements of the local thermodynamic or electrical conditions near the droplet and are used primarily to illustrate the relative magnitude of the individual mechanisms." (Lines 457-459)