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 #3

The manuscript describes a device/technique that is designed to measure impact initiated contact freezing. This is an interesting topic, and data is needed. I don't think we can currently assess just how important contact freezing may be in the atmosphere because we do not have the data needed for such an assessment.

Author's response: We appreciate your recognition of the need for quantitative contact freezing measurements. We agree that the current lack of experimental constraints limits our ability to assess the atmospheric importance of contact freezing. This is precisely the motivation for developing C-ICE: to provide controlled and quantitative measurements of particle-droplet contact freezing that can help build the experimental basis needed for future atmospheric assessments. The present manuscript therefore focuses on establishing and validating the experimental method rather than attempting to quantify the atmospheric contribution of contact freezing.

There are some points that need to be addressed in this manuscript though.

1. Contact freezing is so hard to measure because of the "contact" not the freezing. There are a lot of ways to determine whether a droplet freezes, and I think the method that the authors describe works well for this purpose. I am not convinced that the authors have adequately constrained the "contact" portion of contact freezing.

   We thank you for this important comment. We agree that constraining particle-droplet contact is the key challenge. Because individual submicron impacts cannot be directly resolved optically, we added complementary experimental constraints. Dry soluble ammonium sulfate particles provide a mode-specific test because they dissolve after entering the droplet and therefore do not sustain a conventional delayed immersion pathway. In addition, direct observations of particles retained in the target droplet independently confirm particle collection under the C-ICE geometry. Together, these additions strengthen the experimental constraint on both particle-droplet contact and freezing mode attribution.

   "Impact-initiated contact freezing involves ice nucleation associated with the arrival and physical contact of an aerosol particle with a supercooled droplet. Accurate identification of freezing onset is

therefore essential for relating the observed phase transition to the particle delivery history." (Lines 258-260)

"In this study, freezing-induced breath figures provide the primary operational criterion for identifying freezing onset at the single-droplet level, while grayscale analysis provides a complementary consistency check. This dual-criterion approach enables reproducible identification of the onset freezing temperature and provides the basis for subsequent frozen fraction analysis." (Lines 342-345)

"To our knowledge, this is the first laboratory report of impact-initiated contact freezing triggered by dry $(NH_4)_2SO_4$ particles, extending previous observations of contact freezing by other soluble salts (Niehaus and Cantrell, 2015). This finding also extends the extensive literature on ammonium sulfate deliquescence and efflorescence phase transitions (Onasch et al., 1999; Jordanov and Zellner, 2006), homogeneous freezing of aqueous ammonium sulfate droplets (Larson and Swanson, 2006; Knopf and Lopez, 2009), and heterogeneous ice nucleation involving solid or effloresced ammonium sulfate (Zuberi et al., 2001; Wise et al., 2009; Schill et al., 2014). A related collision-induced phase transition was demonstrated by Davis et al. (2015a; 2015b), who showed that contact with an external solid particle can trigger efflorescence of metastable aqueous ammonium sulfate droplets. More recently, Kirpes et al. (2022) observed solid organic-coated ammonium sulfate particles in the summertime Arctic at high RH, further demonstrating that ammonium sulfate can persist in unexpected solid phase states under atmospheric conditions.

Importantly, $(NH_4)_2SO_4$ provides a mode-specific validation of impact-initiated contact freezing in C-ICE. The dry crystalline particles dissolve after entering the pure water droplet, so no persistent solid ammonium sulfate surface remains to support conventional delayed immersion freezing. Once dissolved, ammonium sulfate lowers water activity and favors homogeneous freezing only at substantially lower temperatures. Consistent with this, Kohn et al. (2016) used $(NH_4)_2SO_4$ particles to form dilute cloud droplets and observed no heterogeneous freezing signal above 236 K (-37.15 °C), with freezing occurring only in the homogeneous regime. This is approximately 20 °C colder than the -17.7 to -14.4 °C freezing range observed here. Thus, neither intrinsic background freezing nor homogeneous or delayed immersion freezing following particle dissolution can reasonably account for the observed warm-temperature response. The $(NH_4)_2SO_4$ experiments therefore provide direct evidence that C-ICE can generate and detect impact-initiated contact freezing, while simultaneously demonstrating stable and reproducible system performance." (Lines 374-393)

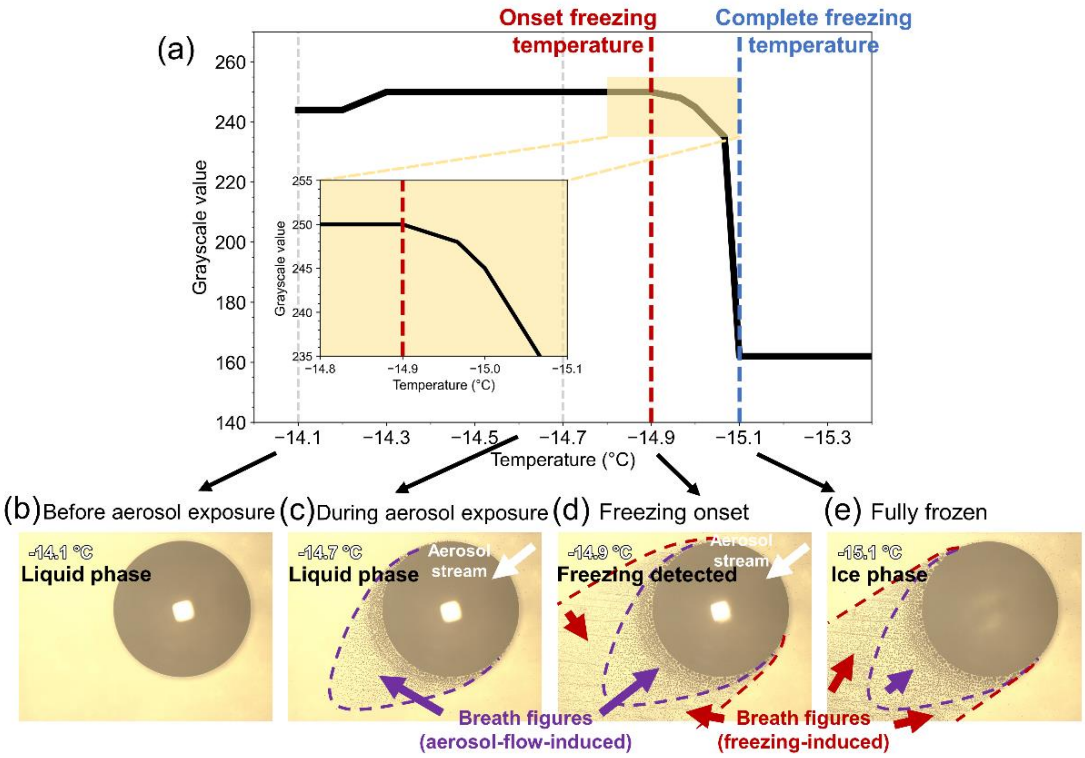


Figure R3.1. Optical identification of freezing onset in the C-ICE system. (a) Evolution of the grayscale value of a single supercooled pure-water droplet as a function of temperature during a representative C-ICE experiment with polydisperse ammonium sulfate ($(NH_4)_2SO_4$) particles. The onset freezing temperature is indicated by the red dashed line, while the complete freezing temperature is marked by the blue dashed line. The shaded inset highlights the sensitivity of the grayscale signal to the onset freezing event. Representative optical images illustrating the evolution of the droplet during the experiment are shown in panels (b-e), with the corresponding temperatures indicated in the upper-left corner of each image and linked to panel (a) by black arrows. (b) Supercooled liquid droplet before introduction of the aerosol stream (-14.1 °C). (c) Liquid droplet during aerosol exposure, showing localized breath figures in the wake region of the droplet (-14.7 °C). (d) Freezing onset, marked by the abrupt appearance and expansion of secondary breath figures (-14.9 °C). (e) Fully frozen droplet (ice phase; -15.1 °C). ... (Fig. 2)

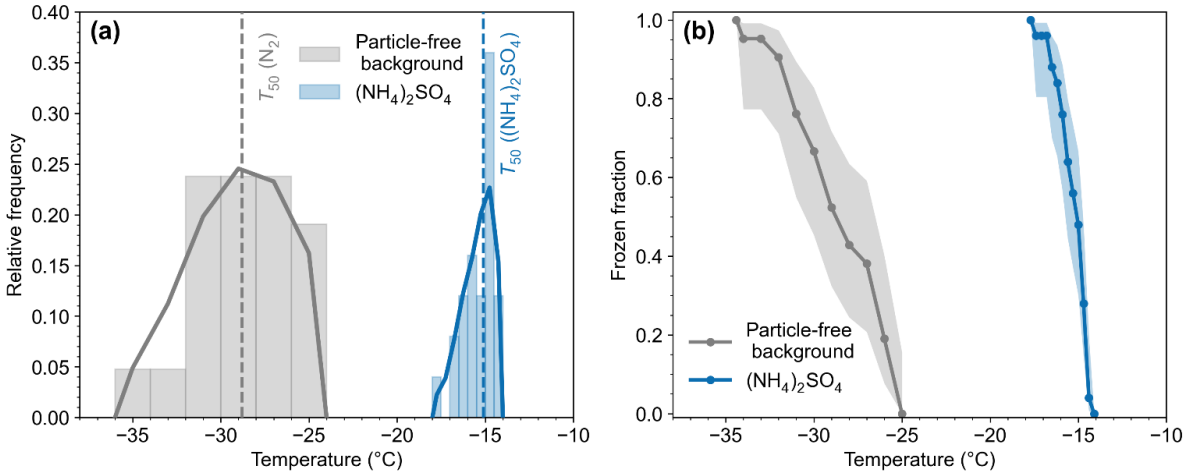


Figure R3.2. Mode-specific validation and background freezing behavior of the C-ICE system. (a) Distributions of freezing temperatures obtained from repeated experiments conducted under identical conditions for the particle-free background (i.e., no aerosol particles present; grey; n = 21) and for polydisperse ammonium sulfate ($(NH_4)_2SO_4$) particles... (Fig. 3)

2. The assumption is that AgI aerosol will cause freezing of the droplet upon contact, but that remains an assumption. The collision rate can be calculated, as the authors have done, but it isn't experimentally verified. The particles may be deposited onto the surface of the droplet, then catalyze freezing at some later time. Is that still contact freezing? The stream of air that is used for the aerosol flow is undoubtedly setting up some circulation within the droplet, which could entrain particles from the surface into the interior, which means that you now have a convolution of contact freezing and immersion freezing. Has there been an attempt to show that this is not happening?

In the case of AgI, we agree that particles may be collected by the droplet and subsequently contribute to immersion freezing. For this reason, the revised manuscript no longer uses AgI alone as evidence for contact freezing. Instead, we address this ambiguity in two complementary ways: (i) dry ammonium sulfate is used as a mode-specific validation material; and (ii) AgI immersion freezing is measured directly over 0.0001-1 wt% to evaluate how strongly the immersion response depends on particle loading.

Separately, particle collection is independently evaluated using quartz. For both DMA selected 800 nm quartz and polydisperse quartz, retained particle features were directly observed within the target droplet after freezing and melting. These observations provide an experimental check on particle collection that does not rely on the freezing event itself.

"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)

"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)

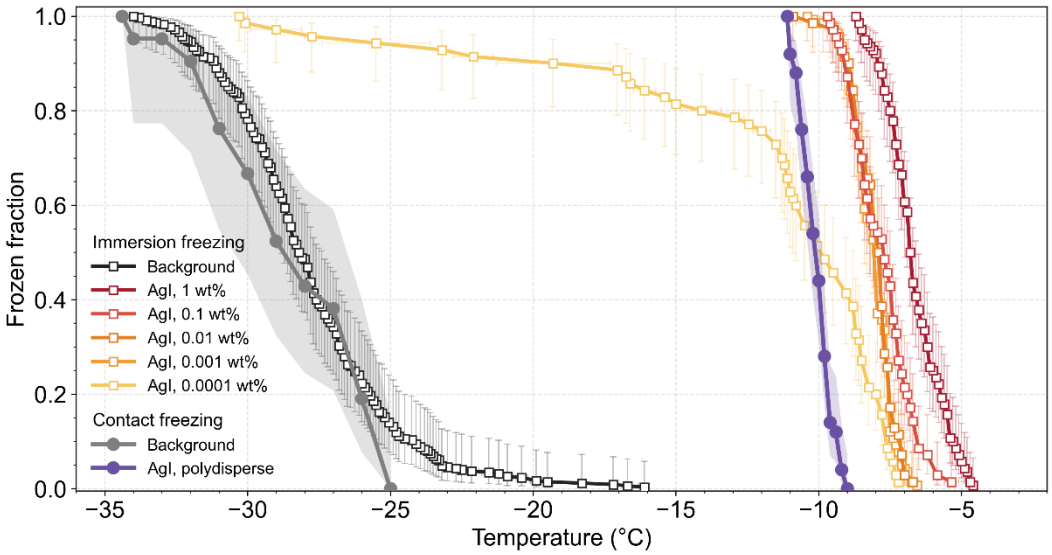


Figure R3.3. Comparison of AgI immersion and impact-initiated contact freezing. Frozen fraction as a function of temperature is shown for immersion freezing experiments using AgI suspensions spanning 0.0001–1 wt% (open symbols), polydisperse AgI impact-initiated contact freezing experiments in C-ICE (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. This issue is one that almost every attempt to measure contact freezing must confront. Davis et al used the direct approach for contact efflorescence, which isn't always possible for freezing and submicron particles. Hoffmann et al. studied contact freezing in an electrodynamic trap, and they were not able to "see" the particles colliding with the suspended droplets, but they were able to draw some conclusions about contact vs immersion freezing by examining the functional form of the frozen fraction vs time curves (see their Fig. 2). (They also verified collision rates by looking at particle residuals in droplets with an SEM.) Niehaus et al studied contact freezing, using droplets on a cold stage. They attempted to constrain contact vs immersion freezing by recording the first freezing event as "contact", then melting the droplet and subsequently cooling to the original temperature. Presumably the particle at the surface would have been subsumed into the bulk, where it would catalyze freezing in the immersion mode.

They did not see such events, leading to the conclusion that they were measuring contact freezing.

We greatly appreciate these examples. They motivated us to add independent experimental checks to the C-ICE study. Analogous to the residual particle approach noted by the referee, we directly observed retained quartz particle features within the target droplet after freezing and melting, providing an independent evaluation of particle collection. We also performed AgI immersion freezing experiments across five concentrations to directly assess the potential contribution of delayed immersion freezing.

In addition, we introduced dry ammonium sulfate as a mode-specific validation material. Because ammonium sulfate dissolves after entering the water droplet, it does not provide a persistent solid surface for immersion freezing. Together, these experiments provide independent constraints on particle collection and freezing-mode attribution.

"In this section, we combine the physically based collision model described in Sect. 2.3 with experimental constraints from size-selected AgI measurements and direct observations of quartz particle features retained in the target droplet after freezing and melting to derive an observation-constrained collision efficiency for C-ICE." (Lines 438-440)

"To independently evaluate particle collection without relying on the freezing response itself, additional C-ICE experiments were conducted using quartz particles. For DMA selected 800 nm quartz particles, numerous optically resolved quartz features were observed within the droplet after freezing and subsequent melting (Fig. S8), directly confirming substantial particle collection under the actual nozzle-droplet geometry. A separate experiment with polydisperse quartz particles similarly revealed a large population of retained particles. Automated image analysis identified 3046 particle features within the post-melt droplet (Fig. S9).

The post-melt evolution of the polydisperse quartz experiment is shown in Figs. 7a-d. After melting, retained quartz particles are distributed throughout the droplet. As evaporation proceeds and the droplet contracts, the particles are progressively redistributed and concentrated, eventually forming aggregates near the shrinking droplet footprint. This behavior makes particle counting after complete evaporation less representative of the initially collected population. Therefore, particle identification was performed shortly after complete melting to avoid extensive aggregation (Figs. 7a and S9a). Figure 7e compares the nozzle-delivered particle size distribution measured by an OPC, the corresponding collision-weighted distribution calculated using CE_{constr}, and the optically detected retained quartz features. The optical measurements independently support substantial particle collection and are broadly consistent with the magnitude predicted using CE_{constr} over the optically resolvable size range. The optically derived diameters are apparent equivalent diameters (Fig. S9b) rather than direct physical particle diameters because imaging through the curved droplet introduces refraction and magnification. Each detected feature was assigned an apparent equivalent circular diameter and grouped using the OPC size bin boundaries. In addition, the practical optical detection limit is approximately 0.8 µm, compared with approximately 0.3 µm for the OPC. Therefore, Fig. 7e is not intended as an exact bin-by-bin size closure; rather, it provides an independent experimental evaluation of the particle collection using the actual C-ICE geometry.

In summary, the physically based CE_{model}, the size-resolved AgI constraints, and the independent quartz collection measurements provide a consistent experimental basis for CE_{constr}. This observation-constrained efficiency is subsequently used to convert measured aerosol number concentrations and

size distributions into effective particle collision rates onto the target droplet." (Lines 519-542)

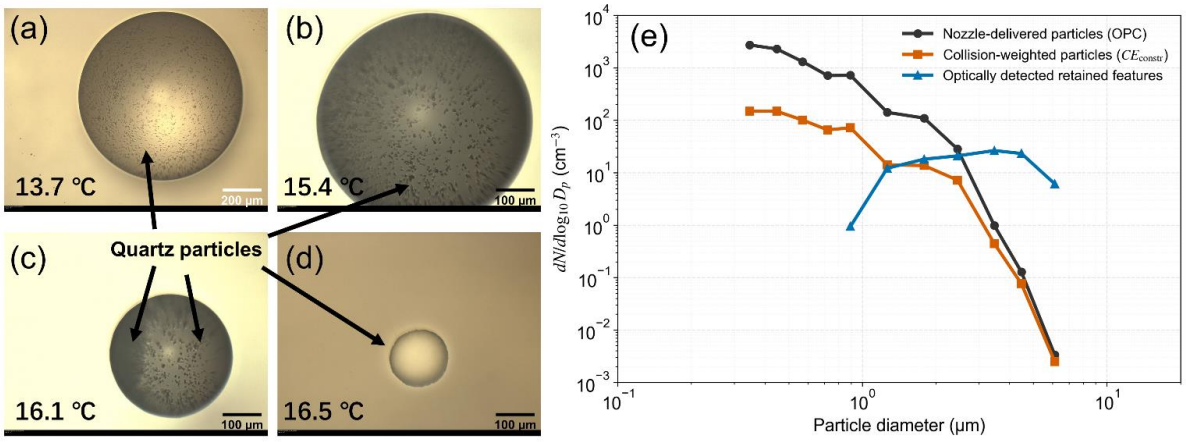


Figure R3.4. Independent experimental evaluation of particle collection in the C-ICE system using polydisperse quartz particles. (a-d) Optical images showing retained quartz particles after melting, as the liquid droplet progressively evaporated and contracted during warming from 13.7 to 16.5 °C. Particles are initially distributed throughout the liquid droplet and become progressively redistributed and concentrated as the droplet contracts. (e) Comparison of the nozzle-delivered particle number size distribution measured by OPC (black), the corresponding collision-weighted distribution calculated using CE_{constr} (orange), and the optically detected retained particle features (blue). A total of 3046 particle features were identified and assigned apparent equivalent circular diameters, grouped according to the OPC size bin boundaries, and converted to an equivalent number concentration using the total aerosol volume delivered during the pre-freezing aerosol exposure period. The resulting distribution was normalized by the corresponding logarithmic bin widths. Diameters for the OPC-derived distributions represent measured particle sizes, whereas the optical distribution represents apparent equivalent circular diameters of retained features. Because the curved droplet acts as an optical lens and the practical optical detection threshold is approximately 0.8 µm, the optical and OPC diameters are not directly equivalent. The comparison provides an independent experimental evaluation of particle collection rather than an exact size-resolved closure. (Fig. 7)

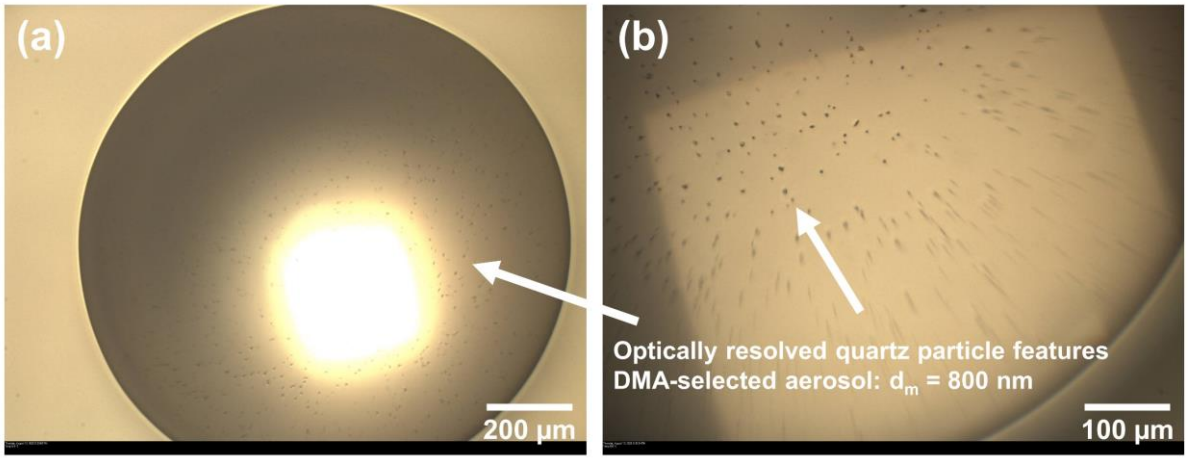


Figure R3.5. Optical observation of retained quartz particle features following exposure to DMA selected 800 nm quartz particles. (a) Post-melt image of the target droplet following exposure to DMA

selected quartz particles with a nominal mobility diameter (dm) of 800 nm. Numerous dark particle features are visible within the droplet. (b) Enlarged view of the droplet interior showing optically resolved quartz particle features, highlighted by the white arrow. Scale bars are 200 µm in (a) and 100 µm in (b). (Fig. S8)

4. Considering the emphasis that the authors place on the "impact" in this manuscript, I think some discussion of the context and theory of contact freezing is warranted. I'm not advocating for a review paper, but outlining why impact might matter is important for this discussion. The two papers I would compare and contrast are Cooper and Fukuta.

We agree and have clarified the theoretical context and, importantly, the operational meaning of "impact-initiated". The term is used to describe the experiment, in which dynamically delivered aerosol contact a droplet surface in C-ICE. This differs from experimental setups in which a particle is statically placed at or brought to the interface. However, while impaction occurs in the C-ICE experimental procedure, we did not intend to imply that mechanical momentum or pressure alone is the microscopic trigger. In response to the Reviewer's comment, we also add a brief discussion of the classical Cooper (1974) and Fukuta (1975) concepts: Cooper emphasized the thermodynamic importance of an INP intersecting the air-water interface, whereas Fukuta emphasized transient enhancement associated with wetting/contact-line motion during contact. Both motivate attention to the moment of contact without requiring a unique mechanical-impact mechanism.

"By contrast, impact-initiated contact freezing involves the directed collision of an externally generated aerosol particle with a supercooled droplet. Classical contact nucleation theories provide different microscopic explanations for why this moment of contact may promote ice nucleation. Cooper (1974) proposed that a subcritical ice embryo formed on a particle in the vapor phase may become supercritical upon contact with supercooled liquid water, whereas Fukuta (1975) emphasized transient nucleation enhancement during wetting as the water front advances along the particle surface. These hypotheses emphasize the importance of the contact event but do not require mechanical momentum itself to be the sole triggering mechanism. Here, we use the term "impact-initiated contact freezing" operationally to describe freezing associated with directed particle-droplet collision, without implying a unique microscopic nucleation mechanism." (Lines 55-62)

5. I would not stress "impact" in this manuscript. I don't think "contact" is meaningfully measured, and impact even less so. What kind of impact will a half micron diameter particle have as it settles through the viscous boundary layer next to the droplet? The carrier gas may be moving at 4 m/s, but the particles are relaxing to a much, much lower speed as they approach the surface.

We agree. As discussed above, contact nucleation is clearly occurring, but the role of the force of impact, if any, is not determined in the experiment. The 4.0 m s^{-1} nozzle-exit gas velocity should not be interpreted as the particle impact velocity at the droplet surface. The revised manuscript now states this distinction and avoids using "impact" as a claim about a unique mechanical-pressure mechanism. We retain "impact-initiated" as an operational descriptor of the directed aerosol-delivery configuration, which distinguishes it from surface-initiated contact-freezing methods. The actual collection behavior is constrained experimentally through CE_{exp} and independently evaluated with quartz particles.

"Here, we use the term "impact-initiated contact freezing" operationally to describe freezing associated with directed particle-droplet collision, without implying a unique microscopic nucleation mechanism." (Lines 60-62)

"The performance and interpretation of the system are evaluated through four complementary analyses. First, the optical response is used to resolve freezing onset from subsequent bulk solidification. Second, dry ammonium sulfate particles, together with particle-free background measurements and aerosol-conditioning simulations, provide mode-specific validation of impact-initiated contact freezing and system reproducibility. Third, a physically based collision model is combined with freezing-inferred collision efficiencies and independent post-melt quartz particle observations to constrain particle collection in the actual C-ICE geometry. Finally, silver iodide is used as a benchmark material to evaluate freezing mode sensitivity and particle size dependent contact freezing. Together, these analyses establish C-ICE as a quantitative experimental system for investigating impact-initiated contact freezing and for developing physically constrained representations of particle-droplet interactions." (Lines 90-98)

"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." (Lines 135-137)

Other points:

1. lines 135-136: the assumption that contact freezing is promoted by a finite number of active sites is questionable. Contact freezing has been shown for soluble salts, where the existence of active sites is almost certainly not applicable. See Niehaus and Cantrell for the contact freezing results, but see the Davis paper already cited, where the phase transition is efflorescence, not freezing.

   We appreciate this constructive comment and have removed the singular active site assumption from the frozen fraction analysis. The revised method defines $f_{ice}(T)$ empirically from repeated independent droplets and does not require a singular or time dependent stochastic nucleation model. The new ammonium sulfate experiments also directly demonstrate why the analysis should not depend on a persistent active site picture.

   "Each experiment in C-ICE employs a single freshly pipetted pure water droplet, and experiments are repeated multiple times under identical conditions. Accordingly, the freezing of each droplet and its associated freezing temperature are treated as independent realizations. The total number of droplets (N_{tot}) corresponds to the number of repeated experiments conducted for a given set of conditions. The frozen fraction at temperature T, f_{ice} (T), is therefore defined empirically as the fraction of droplets that have frozen at temperatures warmer than or equal to T, which is given by (Vali, 1971)" (Lines 162-166)

   "This definition does not require an assumption regarding whether the underlying nucleation process follows a strictly singular or time dependent stochastic description. Because all experiments are conducted using the same prescribed cooling rate, temperature and experimental time remain consistently linked within each dataset." (Lines 168-171)

2. I think the data in Figure 3 also cast some doubt on the singular hypothesis. The range of freezing temperatures for the background is pretty broad. If the freezing were singular, wouldn't that be a lot narrower?

Admittedly, the observed range of temperatures for background is rather broad. In C-ICE, the particle-free background experiments represent the intrinsic freezing behavior of identical UHPLC-water droplets under the same cooling protocol and nitrogen flow perturbation as in the contact experiments. It is possible that agitation from the nitrogen perturbation enhances freezing or from trace impurities in the water could promote heterogeneous/immersion-type freezing.

In addition, the new AgI immersion measurements also show that, for materials with strong immersion activity, the freezing response depends on particle loading and therefore requires careful control when comparing nucleation modes. Because the empirical frozen fraction analysis used in this study does not require a singular active site assumption, we have removed this assumption from Sect. 2.2 and now describe the background distribution empirically.

"This definition does not require an assumption regarding whether the underlying nucleation process follows a strictly singular or time dependent stochastic description. Because all experiments are conducted using the same prescribed cooling rate, temperature and experimental time remain consistently linked within each dataset." (Lines 168-171)

"No specific microscopic nucleation pathway is assigned to these background events, and contributions from trace impurities or the substrate cannot be completely excluded." (Lines 356-357)

3. Section 3.1, breath figures: The authors present a compelling case that the breath figures are indicative of freezing. The case is not compelling that this is the direct result of an impact. The freezing event might have been catalyzed in the immersion mode as outlined above.

We appreciate this constructive comment.

Section 3.1 now uses breath figures only as an operational indicator of freezing onset; it does not use the breath figure morphology itself as proof of contact mode. Freezing mode attribution is handled separately in Sect. 3.2 using the ammonium sulfate mode-specific experiments and in Sect. 3.4 through the AgI immersion comparison.

"The performance and interpretation of the system are evaluated through four complementary analyses. First, the optical response is used to resolve freezing onset from subsequent bulk solidification. Second, dry ammonium sulfate particles, together with particle-free background measurements and aerosol-conditioning simulations, provide mode-specific validation of impact-initiated contact freezing and system reproducibility. Third, a physically based collision model is combined with freezing-inferred collision efficiencies and independent post-melt quartz particle observations to constrain particle collection in the actual C-ICE geometry. Finally, silver iodide is used as a benchmark material to evaluate freezing mode sensitivity and particle size dependent contact freezing." (Lines 90-96)

"3.1 Optical identification of freezing onset in C-ICE" (Line 257)

4. Line 488: "...DMA-selected aerosols…" Is there an effort to correct for the fact that there will be doubly charged aerosol, which are approximately twice as big?

We thank you for this comment.

A neutralizer is used upstream of the DMA so that the size-selected aerosol approaches an equilibrium bipolar charge distribution before mobility classification. Nevertheless, multiply charged larger particles cannot be completely excluded, so the revised manuscript refers to 200 and 800 nm as nominal mobility diameters rather than exact physical diameters. We have added this limitation explicitly.

"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)

5. Lines 499-502: The authors assert that the broader range of freezing T seen when they use polydisperse aerosol is a result of the range of particle sizes. The shift to higher T is apparent, but is the slope of the curve appreciably different?

This is an excellent point. The two size-selected populations show steeper frozen fraction transitions than the polydisperse case, while the polydisperse aerosol also has substantially higher collision rates over much of the measured size range. We interpret the warmer and broader polydisperse response as reflecting the combined effects of a broader particle size population and more frequent particle-droplet collisions. In the revised text, we do not claim that particle size spread alone determines the slope.

"The two size-selected populations also exhibit steeper frozen fraction slopes than the polydisperse case. These systematic differences under the same C-ICE geometry and thermodynamic conditions support a particle-size dependence of the AgI freezing response. The warmer freezing observed for 800 nm relative to 200 nm AgI is consistent with previous evidence that contact freezing efficiency depends strongly on particle size. Hoffmann et al. (2013a) reported a pronounced size dependence for size-selected illite particles, with larger particles exhibiting higher single-contact freezing probabilities. More broadly, AgI ice-nucleating activity is known to depend on available surface area and interfacial configuration (Marcolli et al., 2016). Larger particles may promote freezing through a combination of increased contact area, differences in collision/contact and wetting dynamics, and a higher probability of containing active nucleation sites. The present measurements do not isolate these mechanisms individually, but demonstrate that C-ICE can resolve particle size-dependent freezing behavior under controlled collision conditions." (Lines 583-592)

"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 616-624)