Anonymous Referee #2 (RC1)

Review of "Lagrangian particle-based simulation of aerosol-dependent vertical variation of cloud microphysics in a laboratory convection cloud chamber"

This paper presents a computational investigation of cloud droplet formation and growth in a 2-meter-tall convection chamber. A unique aspect of the work is the use of Lagrangian microphysics, which allows the time history of droplets to be investigated. Some of the results are intriguing and overall the work appears to be rigorous and technically correct (with one exception described below). The connections to atmospheric processes are somewhat tenuous, so it seems the paper would be more suitable for AMT than for ACP. To be published in ACP those connections should be strengthened.

Response: We thank the reviewer for this constructive comment. We strengthened the link to atmospheric cloud processes by revising Section 4 and the Conclusions to (i) frame our results in a timescale perspective using the ratio of a microphysical adjustment time (phase relaxation, τ_p) to a chamber-scale transport time (τ_m), summarized by Da (= τ_m/τ_p), (ii) relate the Da dependence of supersaturation variance and droplet growth to laboratory evidence from the Pi Chamber (Chandrakar et al., 2016), and (iii) connect the resulting Lagrangian "history sensitivity" to eddy-hopping and stochastic parcel views of condensational broadening in shallow cumulus and stratocumulus (e.g., Abade et al., 2018). This is further supported by recent adiabatic cloud simulations in which the main analysis is based on a 1D turbulent-parcel framework, supported by additional 3D LES results. These simulations show that eddy hopping effect is stronger under pristine conditions because a longer phase relaxation time permits larger supersaturation fluctuations for comparable vertical-velocity variability, and a larger peak cloud-base supersaturation activates a broader range of CCN (Grabowski et al., 2026). We also added a brief clarification that this Da differs from the entrainment-mixing Da commonly used at cloud edges, and that both mechanisms can coexist in atmospheric clouds (Lines 515-524, 577-604):

"From a Lagrangian perspective, we interpret our diagnostics as follows. At a given time, the super droplets within the lowest 0.2 m can be viewed as originating from a common near-bottom air mass. Turbulent motions then disperse these droplets along trajectories that differ in their net ascent and in their residence time within the near-bottom supersaturation source region. These differences produce distinct time-integrated supersaturation exposures, and therefore distinct condensational growth histories, even for droplets that start from the same near-bottom layer. In the clean (low-CCN) cases, supersaturation fluctuations are relatively large, so growth becomes strongly history dependent and the normalized radius increment, dr, varies substantially across trajectories. In the polluted (high-CCN) cases, supersaturation fluctuations are damped, so droplets experience more similar effective supersaturation histories and dr becomes more uniform. In our simulations, this transition is accompanied by a weakening of the vertical gradient in q_c, consistent with reduced contrasts between strongly ascending and weakly ascending trajectories as CCN increases."

"This behavior can be summarized by a timescale perspective in which Da (= τ_m/τ_p) compares a chamber-scale transport time, τ_m, to the phase relaxation time, τ_p. As CCN loading increases, the total droplet surface area increases, τ_p shortens, and supersaturation

perturbations are relaxed more rapidly toward quasi-equilibrium. The resulting reduction in supersaturation variance and narrowing of growth histories is consistent with laboratory evidence from the Pi Chamber (Chandrakar et al., 2016). Our simulations extend this picture by showing, in a fully three-dimensional Lagrangian framework, how the same mechanism links supersaturation damping to both the vertical structure of q_c and the sensitivity of condensational growth to vertical history for droplets originating from the same near-bottom air mass."

"In atmospheric clouds, this timescale view is closely related to eddy-hopping and stochastic parcel frameworks for condensational broadening in shallow cumulus and stratocumulus. When Da is unity, droplets adjust slowly enough that differences in Lagrangian residence time within regions of enhanced supersaturation are efficiently translated into differences in condensational growth, favoring spectral broadening. When Da is large, supersaturation is homogenized more rapidly, so droplets following distinct trajectories can nevertheless experience similar effective supersaturation histories and grow by comparable amounts. This suggests that higher CCN loading can weaken the imprint of turbulent history on condensational broadening in cloud interiors, consistent with stochastic entraining parcel studies (e.g., Abade et al., 2018). This interpretation is also consistent with recent adiabatic cloud simulations based primarily on 1D turbulent-parcel modeling, complemented by 3D LES cloud simulations, showing that eddy hopping effect and the associated turbulence-driven condensational broadening are stronger under pristine conditions because a longer phase relaxation time allows larger supersaturation fluctuations for the same turbulent vertical-velocity variability, and a larger peak cloud-base supersaturation activates a broader range of CCN (Grabowski et al., 2026).

We note that the Da used here differs from the Da commonly adopted in entrainment-mixing studies at cloud edges, where Da often compares a microphysical adjustment time (frequently evaporation in subsaturated entrained air) to a small-scale turbulent mixing time across cloudy and environmental filaments. In that framework, large Da implies microphysics can act faster than mixing, promoting inhomogeneous mixing signatures. By contrast, our Da compares phase relaxation to a chamber-scale transport time in a near-saturated, quasi-closed environment where condensational relaxation dominates. There, a shorter τ_p strengthens the negative feedback on supersaturation perturbations, so increasing CCN primarily damps supersaturation fluctuations and homogenizes condensational growth histories in the chamber interior. In atmospheric clouds, both behaviors can coexist, with CCN damping interior supersaturation variability while entrainment at cloud edges can still drive locally inhomogeneous mixing depending on humidity deficits and mixing scales."

The main criticism is regarding the way some of the results are discussed and interpreted. In multiple places in the manuscript the wording suggests that supersaturation builds up during ascent. This has the potential to be very confusing because some readers may associate this with the familiar increase of supersaturation with height in a convective cloud. For example, the explanation of increasing q_c with height being attributed to "active droplet activation and condensational growth during ascent" (lines 198-199) does not make sense. Symmetry suggests that the same argument should work for plumes descending from the top surface. A cold plume mixed into a warm background has nearly the same potential to generate supersaturation as a warm plume mixed into a cold background (see multiple previous papers

showing calculations and simulation results for convection-cloud chambers… usually the supersaturation profile is nearly constant with height, showing no significant bias toward the bottom or top surface). In contrast, arguments that appear later in the paper (such as on lines 380-382) make much more sense in light of the results shown. I would encourage the authors to revise the language in other parts of the paper that give the potentially misleading impression that droplet growth is tied to vertical ascent. The peak near normalized vertical displacement of negative 0.25, suggests that plumes that are efficiently mixed produce the strongest supersaturation (as opposed to a plume that simply travels upward without strong mixing). Supersaturation generation in a convection-cloud chamber is the result of mixing, not of ascent. That mixing may have a link to plume propagation distance and therefore to distance from the top or bottom boundaries, but it is not the ascent itself that is the source of supersaturation.

Response: We thank the reviewer for this helpful clarification. We have revised the sentence (Lines 231-233):

"indicating a relatively strong vertical increase in q_c that reflects efficient droplet activation and condensational growth in the lower part of the chamber where mixing between warm, moist air from the bottom boundary and cooler air aloft generates enhanced supersaturation"

Other important points to be addressed are:

What is the fundamental problem statement or hypothesis underlying the investigation? For example, on lines 58-60 the direction of this investigation is stated, but it is not clear why this topic would be of interest to explore.

Response: We thank the reviewer for pointing out the need to clarify the fundamental problem and hypothesis. In the revised manuscript, we have added a short paragraph (Lines 61-67):

"These previous chamber simulations raise a straightforward process level question. Why do q_c profiles tend to increase with height at low aerosol loading and become more vertically uniform as aerosol concentration increases, and which mechanisms control this behavior. In this study we test the hypothesis that changes in CCN concentration modify the balance between turbulent mixing and microphysical adjustment, and that this timescale balance controls both the vertical structure of q_c and the spread of condensational growth along Lagrangian trajectories. Our goal is to use trajectory resolved diagnostics in LES of a convection cloud chamber to quantify this aerosol dependent timescale regime and to provide a framework that can be used to interpret chamber experiments and link them to shallow boundary layer clouds."

A “free-slip” boundary condition at the top and bottom surfaces (line 106) is unphysical and may fundamentally alter the nature of the flow. Can this be justified as having negligible effect on the microphysical problem investigated in this study? If so, please provide clear evidence. If not, please clearly state this limitation in the abstract and provide a discussion of how the conclusions will change.

Response: We agree that a “free-slip” boundary condition at the top and bottom plates is an idealization relative to the physical chamber, because it does not explicitly represent viscous wall stresses at solid surfaces. In our simulations, the thin viscous layers adjacent to the plates are not resolved at the adopted grid spacing. Under such under-resolved near-wall conditions, enforcing a strict zero tangential velocity at the plates can produce an artificially strong grid-scale shear layer and lead to spurious numerical dissipation. We therefore use a stress-free tangential treatment as a pragmatic approximation at the resolved scale.

In principle, a more physical approach would be to represent the plate effects through parameterized near-wall fluxes, including momentum flux and potentially heat and moisture fluxes, applied within a thin layer adjacent to the boundaries. However, constraints on these fluxes in chamber conditions remain limited, and it is not straightforward to apply standard wall-function formulations (e.g., Monin–Obukhov similarity) in a closed laboratory geometry across different wall orientations. In particular, extending such similarity assumptions to top and side surfaces raises additional questions about physical consistency because the angle between gravity and the local wall-normal direction differs between the top/bottom plates and the sidewalls. We therefore treat the near-wall momentum exchange as a known limitation of the dynamical idealization and focus our interpretation on the chamber interior.

We expect the chamber dynamics to be dominated by buoyancy-driven motions away from the plates, which are the flow features most relevant for the microphysical questions addressed here. This expectation is supported by previous Pi Chamber simulations using the same babyEULAG dynamical configuration. Grabowski et al. (2024) report simulated mean TKE of about 3×10^{-3} m^2 s^{-2} comparable to Pi-chamber estimates (Thomas et al., 2019, Fig. 6), and Grabowski (2020) shows that, without CCN, the simulated mean supersaturation and its standard deviation (mean ≈ 5%, SD ≈ 0.9%) agree with Pi-chamber observations (Thomas et al., 2019, Fig. 8). Finally, we conducted a sensitivity test by recomputing the key *W*–*S* correlations including the boundary-adjacent layers and over the full domain. While the absolute magnitude of the correlation is reduced, the aerosol-dependent trends are unchanged (Fig. R1). We have clarified these points and added an explicit limitation statement in the revised manuscript (Lines 113–130, 333-335).

“Although the free-slip condition at the lower and upper boundaries is idealized relative to the solid plates in an actual chamber, this dynamical configuration has been evaluated extensively in previous Pi Chamber simulations. Grabowski (2020) showed that, in the absence of CCN, the model reproduces the observed mean supersaturation and its standard deviation when averaged over the interior of the chamber, and Grabowski et al. (2024) demonstrated that the simulated mean TKE is close to values inferred from Pi Chamber observations. To represent the solid chamber walls within the horizontally periodic framework, we introduce internal sidewall regions at the 10th grid point in each horizontal direction. Around each of these planes, a three-grid-point-wide relaxation zone is applied in

which all three velocity components are relaxed toward zero and the thermodynamic variables are nudged toward the prescribed wall state (T = 289.5 K, RH = 100%) with a relaxation time scale of 0.1 s. Thus, the physical sidewalls of the chamber are mimicked by internal relaxation zones, while the outer lateral boundaries of the computational box remain strictly periodic. Because the thin viscous layers adjacent to the top and bottom plates are not resolved at our grid spacing, we do not attempt to represent viscous wall stresses at the resolved scale explicitly. A more complete treatment would require parameterized near-wall fluxes (momentum, and possibly heat and moisture) constrained by chamber conditions, but such constraints remain limited, and the applicability of similarity-based wall functions in a closed chamber, particularly across different wall orientations, is not well constrained. Unless stated otherwise, statistics are computed over an interior subdomain that excludes a few grid levels adjacent to the top and bottom plates to minimize sensitivity to under-resolved near-wall dynamics. The sensitivity of the key vertical velocity–supersaturation diagnostics to this choice is evaluated in Sect. 3.2."

"Repeating the analysis over the full domain, including the boundary-adjacent layers, reduces the absolute magnitude of the W–S correlation but does not change the aerosol-dependent trends or the tight relationship between σ_S and the W–S correlation (not shown)"

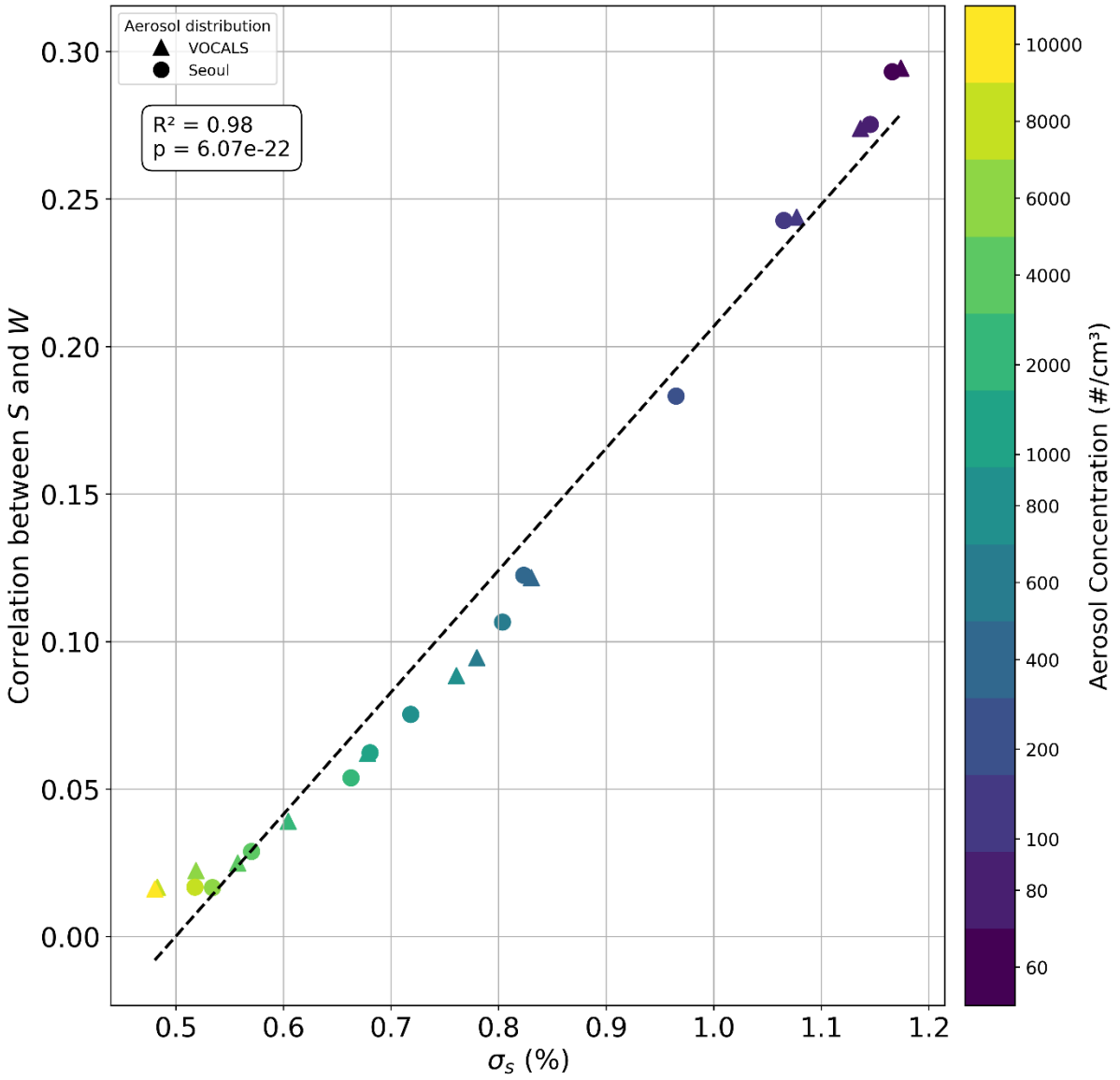

Figure R1. Scatter plot showing the relationship between the domain-averaged supersaturation standard deviation (σ_S) and the correlation coefficient between supersaturation (S) and vertical velocity (W). Both σ_S and corr(S, W) are calculated using all grid points in the domain, without excluding the uppermost and lowermost boundary layers for VOCALS (triangles) and Seoul (circles) aerosol cases. Each point represents the averaged value for the quasi-equilibrium period (15–30 minutes).

In multiple places in the manuscript it is pointed out that mean and fluctuations of supersaturation are reduced as cloud droplet concentration is increased. Suppression of supersaturation for shorter phase relaxation time has a clear theoretical interpretation; for example, see Equations 12 and 13 in Chandrakar et al. (Journal of the Atmospheric Sciences, 2018, "Influence of Turbulent Fluctuations on Cloud Droplet Size Dispersion and Aerosol Indirect Effects"; note that in that paper is the phase relaxation time). There are also multiple places in the manuscript that would be more clearly interpreted in terms of the quasi-steady

supersaturation. For example, on line 322, the statement that condensation causes "rapid vapor depletion… before S can develop" does not make sense. Without supersaturation, there is no vapor depletion. I suppose what is meant is the analog to quasi-steady supersaturation, which is the result of two rates: the rate at which supersaturation is produced versus the rate at which it is consumed. So the low supersaturation observed is simply a result of faster consumption by droplet growth (due to shorter phase relaxation time) for an unchanged rate of production. This is not a surprising result. Another example is lines 218-219: There the concept could be made clearer by drawing an analogy with the quasi-steady supersaturation in a convective cloud. For equivalent forcing, such as updraft strength, smaller phase relaxation time leads to lower quasi-steady supersaturation.

Response: We thank the reviewer for this helpful clarification. We have revised the relevant passages to interpret the reduction of mean supersaturation and its fluctuations at high CCN loading in terms of quasi-steady supersaturation and the phase relaxation time, following Chandrakar et al. (2018) (Lines 17-19, 253-258, 310-312, 347-351, 389-395, 547-558):

"Lagrangian trajectory analyses reveal that upward motion substantially influences droplet growth and activation under clean conditions, but this influence diminishes sharply in polluted environments where the shorter phase relaxation time keeps supersaturation close to a low quasi-steady value.",

"In the polluted cases, the enhanced condensational sink keeps supersaturation close to its low quasi-steady value during ascent, which limits additional condensational growth aloft and therefore reduces the vertical increase in q_c.",

"However, in polluted conditions, the short phase relaxation time causes supersaturation to relax rapidly toward its quasi-steady value, so supersaturation responds only weakly to fluctuations in vertical velocity W.",

"In polluted environments, the high number of activated droplets dramatically increases the total surface area available for condensation, which shortens τ_p and allows the condensational sink to balance the turbulent source of supersaturation over a short timescale. As a result, supersaturation remains low away from the production region near the lower boundary, and most condensational growth occurs there rather than being sustained throughout the full depth of the chamber.",

"This disparity can be expressed by the Damköhler number ($Da = \tau_m / \tau_p$), which quantifies the relative pace of condensation versus turbulent mixing. In polluted environments, $Da \gg 1$, which indicates that condensation adjusts much faster than vertical mixing, so the condensational sink rapidly balances the turbulent source and maintains a low quasi-steady supersaturation. As a result, supersaturation fluctuations and additional condensational growth aloft are strongly reduced, and q_c becomes nearly uniform with height. This behavior is consistent with the quasi-steady supersaturation framework of Chandrakar et al. (2018), in which both the mean supersaturation and its variance decrease as the system timescale $\tau_s = \tau_m \tau_p / (\tau_m + \tau_p)$ becomes smaller.",

"Conversely, at high aerosol concentrations, the short τ_p maintains a low quasi-steady supersaturation throughout the chamber, so condensational growth changes little with height and the q_c profile becomes nearly vertically uniform."

Figure 5 shows large oscillations in the mean supersaturation with height near the top and bottom boundaries. What is the source of this numerical instability? The concern is that the oscillations could strongly influence the correlation with vertical air speed, which is an important focus point in this investigation. This needs to be addressed and its influence on the w'-s' correlation needs to be checked.

Response: We assume that the reviewer refers to large supersaturation fluctuations near the horizontal chamber boundaries. Large values near the boundaries should not be surprising because resolved transport is inefficient there. This is similar to conditions near the surface in atmospheric convective boundary layer that lead to the superadiabatic temperature gradient near the surface. Please note that the dynamic model is run in the so-called implicit large eddy simulation (ILES) mode and there is no explicit representation of the turbulent mixing. This is especially relevant to the conditions near the boundaries. Supersaturation fluctuations that are evident when one moves away from the boundary are purely numerical. These are similar to the Gibbs phenomenon in numerical advection applying non-monotone advection scheme. Although the model applies monotone advection scheme, this is insufficient to maintain nonoscillatory supersaturation as it involves nonlinear coupling between the temperature and water vapor. Please read the discussion in Grabowski and Smolarkiewicz (Mon. Weather Rev. 1990). Such numerical problems are especially evident in ILES simulations.

We do not think these numerical aspects affect the W–*S* correlations (Figure 7), because the correlations reported in the manuscript are computed using interior grid points away from the upper and lower chamber boundaries. As a sensitivity test, we recomputed the same correlations, including grid points closer to the boundaries and also using the full domain. The impact is small, consistent with weak resolved vertical-velocity perturbations near rigid boundaries. This robustness is documented in Figure R1. Figure R1 uses the same x and y axes as Figure 7, but the statistics are computed over the full domain, including the boundary-adjacent layers. In contrast, Figure 7 uses only the interior (mid-level) region. Including the boundary layers reduces the absolute magnitude of the W-S correlation, but the main trends are unchanged. In particular, the W–S correlation decreases with increasing aerosol concentration in both figures, and the relationship between the x-axis diagnostic and the W–S correlation remains comparatively strong when the boundary-adjacent layers are included. We have revised the relevant passages in the revised manuscript (Lines 301-309):

"Before interpreting this relationship, we address a potential concern raised by the mean supersaturation profiles in Figure 5a,b, which exhibit oscillations near the lower and upper rigid boundaries. Two factors likely contribute to these near-wall oscillations. First, resolved transport and mixing are inefficient near rigid boundaries in ILES, which can allow sharp gradients to persist. Second, supersaturation is a diagnosed quantity, so small grid-scale perturbations in temperature and water vapor can translate into comparatively large fluctuations in S because of the nonlinear saturation relationship and advection–condensation coupling (Grabowski and Smolarkiewicz, 1990). Importantly, the W–S correlations reported

in this study are computed over the interior region away from the plates, and the main aerosol-dependent trends remain unchanged when the boundary-adjacent layers are included (not shown). With this in mind, we interpret the aerosol dependence of W–S coupling as follows."

Specific comments

Lines 102-105: In one sentence the "sidewall temperatures" are stated, but in the next line it is stated that the model is configured with "periodic lateral boundaries".

Response: We realize that the original wording was confusing. The babyEULAG domain is horizontally periodic, and the "sidewalls" are represented by internal relaxation zones that mimic solid walls inside this periodic box, following Grabowski et al. (2024). The prescribed sidewall temperature of 289.5 K is applied to these internal wall regions, while the outer lateral boundaries of the computational domain remain periodic. We have revised the model description in Section 2 to clarify this sidewall configuration (Line 118):

"To represent the solid chamber walls within the horizontally periodic framework, we introduce internal sidewall regions at the 10th grid point in horizontal direction at each vertical grid level."

Lines 112-114: How significant are droplet losses to the sidewalls compared to the bottom surface?

Response: We have now quantified the cumulative cloud water loss at each boundary for the period between 15 and 30 min for three VOCALS cases and added a short description in Section 2 (Lines 188-197):

"To further assess the role of the boundaries, we diagnosed the cumulative cloud water loss at each boundary for the period between 15 and 30 min in three VOCALS simulations spanning the aerosol range (60, 200, and 10K cm^{-3}, not shown). In the VOCALS (60) case, approximately 86 % of the cloud water loss during this period occurs at the lower boundary and about 14 % at the sidewalls, with almost negligible loss at the upper boundary. For VOCALS (200, CTRL) the corresponding fractions are about 70 % at the lower boundary, 30 % at the sidewalls, and less than 1 % at the upper boundary, whereas in VOCALS (10K) they are roughly 38, 59, and 3 %, respectively. These diagnostics show that both the bottom plate and the sidewalls act as important sinks for condensate, while the upper boundary remains a minor loss pathway. The decreasing share of condensate loss at the lower boundary from VOCALS (60) to VOCALS (10K) is consistent with reduced sedimentation losses at higher CCN concentrations and supports our interpretation that smaller mean droplet size for larger N_c favors condensate accumulation in the chamber interior."

Lines 157-158: What is the reason for the arbitrary use of 1 micrometer as a cutoff between haze and cloud droplets? For the broad aerosol size distributions used in this study, this can be very different than the true activation radii. An advantage of Lagrangian microphysics is that the aerosol properties are known for each super-droplet, so the boundary between haze and cloud can be determined for each super-droplet. Please explain whether this simplification has any significant consequences for the interpretation of results.

Response: We thank the reviewer for pointing this out. In the revised manuscript, we clarify that the 1 μm threshold is an operational definition that follows Yang et al. (2023) and is used only for statistical classification of "cloud droplets" and "haze particles". To test the sensitivity to this choice, we recomputed Δq_c using an activation criterion based on the critical radius of each super droplet (r > critical radius). The resulting dependence of Δq_c on aerosol concentration (Fig. R2) is nearly indistinguishable from that obtained with the original r > 1 μm definition (Fig. 3), with a mean relative difference of about 1.6 % between the Δq_c values computed using the r > 1 μm threshold and those computed using the r > critical radius criterion across all cases. Physically, even if some large but technically unactivated haze particles exceed 1 μm, they already contribute to bulk condensate similarly to small activated droplets, so treating r > 1 μm hydrometeors as "cloud droplets" in the statistics has no significant influence on our interpretation of the results.

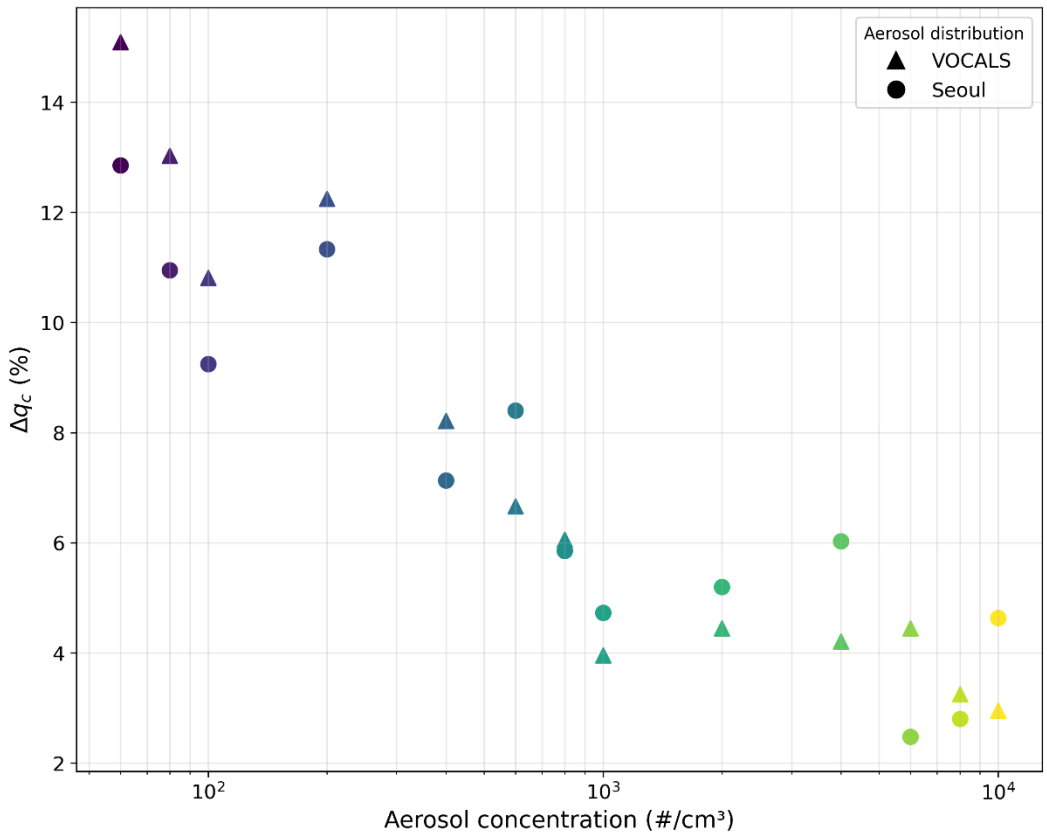

Figure R2. Δq_c as a function of aerosol concentration, where each point represents the averaged value for the quasi-equilibrium period (15–30 min) of each aerosol distribution. Here, Δq_c is computed using only droplets with a radius larger than their size-dependent critical radius (r > critical radius). Triangles denote VOCALS cases; circles denote Seoul cases.

Line 188: It's not incorrect, but it seems a little strange to use "altitude" instead of "height".

Response: Following the suggestion, we have replaced "altitude" with "height" throughout the manuscript and in the relevant figure captions.

Line 194: It isn't clear to me whether these refer to averages over horizontal planes at 0.3 m and 1.7 m, or to averages over layers (e.g., from 0 to 0.3 m and from 1.7 to 2.0 m). Also, is there any particular reason for choosing the value of 30 cm?

Response: In the revised manuscript, we clarify that the values are horizontal averages taken at the heights of z = 0.3 m and z = 1.7 m, not the averages over layers (0.0-0.3 m and 1.7-2.0 m). The level at 0.3 m was chosen to represent the lower source region while remaining several grid points away from the bottom plate, to avoid near-wall effects. The level at 1.7 m was selected to sample the upper part of the cloudy layer, where q_c peaks around 1.5 m, while still being well separated from the top boundary:

"where the numerator is the horizontal mean q_c at z=1.7 m minus the horizontal mean q_c at z=0.3 m."

Figure 3 caption: For the benefit of those reading the paper quickly, it would be helpful to provide a qualitative description of Δq_c within the caption.

Response: Thanks for this helpful suggestion. We have revised the Figure 3 caption:

"Large Δq_c indicates a strong increase of q_c with height, whereas small or negligible Δq_c would indicate a nearly uniform vertical q_c profile. "

Line 229: It is not clear what is meant by the "largest aerosol mode".

Response: In this sentence, “largest aerosol mode” was intended to refer to the largest-size aerosol particles in the size distribution, whose critical supersaturation is approximately 0.02%. We have revised the text to state this explicitly and replaced “largest aerosol mode” with clearer wording to avoid confusion (Lines 266-275):

“The minimum critical supersaturation (S_{crit}) in the prescribed aerosol spectrum is approximately 0.02%, corresponding to the largest aerosol particles in the size distribution. As aerosol loading increases, $\bar{S}$ steadily falls toward this value, often converging near 0.02% (see Fig. 6). This behavior is qualitatively consistent with Yang et al. (2025), who found that mean supersaturation approached the critical value for their monodisperse aerosol. In our polydisperse case, $\bar{S}$ does not collapse to a single number, but instead converges to a narrow range near the minimum S_{crit} set by the largest particles. When $\bar{S}$ is near this threshold, particles with S_{crit} close to $\bar{S}$ can repeatedly transition between weak activation and deactivation as turbulent S fluctuations move conditions slightly above or below their critical value, which effectively buffers $\bar{S}$. This buffering helps explain why, beyond a certain aerosol concentration, the domain-mean q_c no longer increases (Fig. 2): enhanced vapor competition limits additional net activation and condensational growth despite further increases in aerosol loading.”

Figure 5 caption: Clarify that the vertical profiles are for the horizontally-averaged supersaturation. Also, are the histograms for supersaturations within the full volume, or a subset of the chamber volume?

Response: We have revised the Figure 5 caption to clarify that the vertical profiles show horizontally averaged supersaturation and that the histograms are constructed from supersaturation sampled only over the interior region of the domain, excluding grid points within 0.08 m of the sidewalls and 0.1 m of the top and bottom boundaries:

“constructed from supersaturation values at interior grid points, excluding locations within 0.08 m of the sidewalls and 0.1 m of the top and bottom boundaries, during the same quasi-equilibrium period”

Line 265: Should be “high aerosol concentration”.

Response: We have corrected the phrase to “high aerosol concentration” in the revised manuscript. Thanks.

Line 267: Should be “The physical explanation…”.

Response: We have corrected the phrase. Thanks.

Lines 281-283: The “rapid equilibration” argument makes sense, if by that it means that the supersaturation is rapidly depleted. It is unclear what is meant by “droplets complete their growth before experiencing the full extent of vertical S variability”. Does it mean that the supersaturation is depleted after relatively short ascent, such that droplet growth ceases above some level? As written, this argument is too vague.

Response: We have clarified this argument in the revised manuscript as follows:

“In polluted environments, the high number of activated droplets dramatically increases the total surface area available for condensation, which shortens τ_p and allows the condensational sink to balance the turbulent source of supersaturation over a short timescale. As a result, supersaturation remains small away from the production region near the lower boundary, and most condensational growth occurs there rather than being sustained throughout the full depth of the chamber.”

Line 314: The definition “the timescale over which S environment evolves” is ambiguous. One could also say that the phase relaxation time is the characteristic time for the supersaturation to evolve (due to droplet growth). I assume what is meant is that it is the timescale on which the supersaturation varies due to turbulent mixing.

Response: We agree that our original wording was ambiguous. In the revised manuscript we now explicitly define τ_m as the turbulent mixing timescale (Lines 360-362); that is, the characteristic time over which supersaturation changes due to turbulent advection and mixing, and we distinguish this from the phase relaxation time τ_p, which describes changes due to condensational growth:

“that is, the characteristic time over which the supersaturation field changes due to turbulent advection and mixing, in contrast to the phase relaxation time (τ_p), which characterizes changes in supersaturation due to condensational growth.”

Line 316: The units are for a velocity standard deviation, not a velocity variance. Please correct either the wording or the units.

Response: Thanks for pointing this out. In the revised manuscript we replace “turbulent velocity variance” with “a characteristic rms turbulent velocity scale” (standard deviation).

Line 359: The statement that “sustained upward motion enhances droplet growth by exposing SDs to regions of higher s” is inconsistent with the results shown. For most of the simulated cases, a peak occurs at a negative value of normalized vertical displacement, which suggests the opposite of what is stated. Only in the two cleanest cases is the mean growth associated with positive vertical displacement.

Response: Thanks for this clarification. We have revised the discussion of Figure 12 (formerly Fig. 11) to clarify that enhanced growth is associated with trajectories that spend more time moving upward within the supersaturation source region near the chamber base, and we now explicitly explain why the maximum *dr* occurs near a negative normalized vertical displacement in polluted cases:

“This indicates that trajectories that spend more time moving upward within the supersaturation source region near the chamber base tend to experience enhanced condensational growth.”

Line 388: Should be “confinement of the S production zone”.

Response: Thanks. The phrase has been corrected to “confinement of the *S* production zone” in the revised manuscript.

Lines 403-406: The two statements that “activation fraction generally increases with normalized vertical displacement” and “it peaks around a normalized displacement of approximately -0.25” are inconsistent. There is an initial increase, but over most of the range of normalized vertical displacement the activation fraction decreases or is only weakly changing.

Response: We agree with the reviewer’s careful assessment and have revised the sentence in the revised manuscript:

"The activation fraction increases from the mostly descending trajectories toward moderately ascending ones and typically peaks near a normalized vertical displacement of about –0.25, then decreases or changes only weakly at higher normalized vertical displacements."

Lines 408-414: This discussion is not clear.

Response: We have rewritten the discussion at lines 480–487 to clarify the physical interpretation:

"In both the *dr* (Fig. 12) and activation fraction (Fig. 14) analyses, the key factor is that SDs whose normalized vertical displacement lies near −0.25 remain longer within the lower chamber layer ($z \lesssim 0.5$ m), where S is locally enhanced by mixing of air parcels with contrasting temperature and humidity from the upper and lower boundaries. As shown in Figure 5, horizontally averaged S tends to be the highest near the bottom boundary and decreases toward zero with height. Trajectories in this normalized vertical displacement range around −0.25 are repeatedly exposed to this supersaturation source region, which increases both their probability of activation and their condensational growth. In contrast, SDs with larger normalized vertical displacement ascend more quickly out of the lower layer and spend more time in regions where S has already been depleted, so both their activation fraction and *dr* are reduced."

Figure 14: Wouldn't it be more insightful to show the spatial locations of where activation occurred, rather than the locations of droplets that were activated within the last 20 seconds, which is sufficient time that they can be transported over the maximum dimension of the chamber? This seems like a missed opportunity to gain deeper understanding of what processes are controlling activation.

Response: Thank you for this helpful suggestion. We have revised Figure 15 (formerly Fig. 14) to show the three-dimensional spatial locations of first activation events (and their vertical distribution) rather than the locations of droplets after 20 seconds, which more directly illustrates where activation occurs in the chamber (Lines 493-501):

"Figures 15a and 15c show the three-dimensional locations of the first activation events for 50,000 SDs that were randomly selected from the population of unactivated SDs located between 0 and 0.2 m at 1600 s for VOCALS (200, CTRL) and VOCALS (10K) cases, respectively. Figures 15b and 15d present the corresponding vertical histograms using 0.02 m height bins. For VOCALS (200, CTRL), 24,423 of the tracked SDs activate between 1600 and 1620 s, and a substantial fraction of these activation events occur well above the source layer near the lower boundary, with activation locations extending through much of the chamber depth. For VOCALS (10K), only 6,512 SDs activate, and these activation events are almost entirely confined to heights below about 0.3 m, with a weaker secondary maximum

near the chamber top where *S* is also elevated, consistent with the vertical structure of *S*, as shown in Figure 5a."

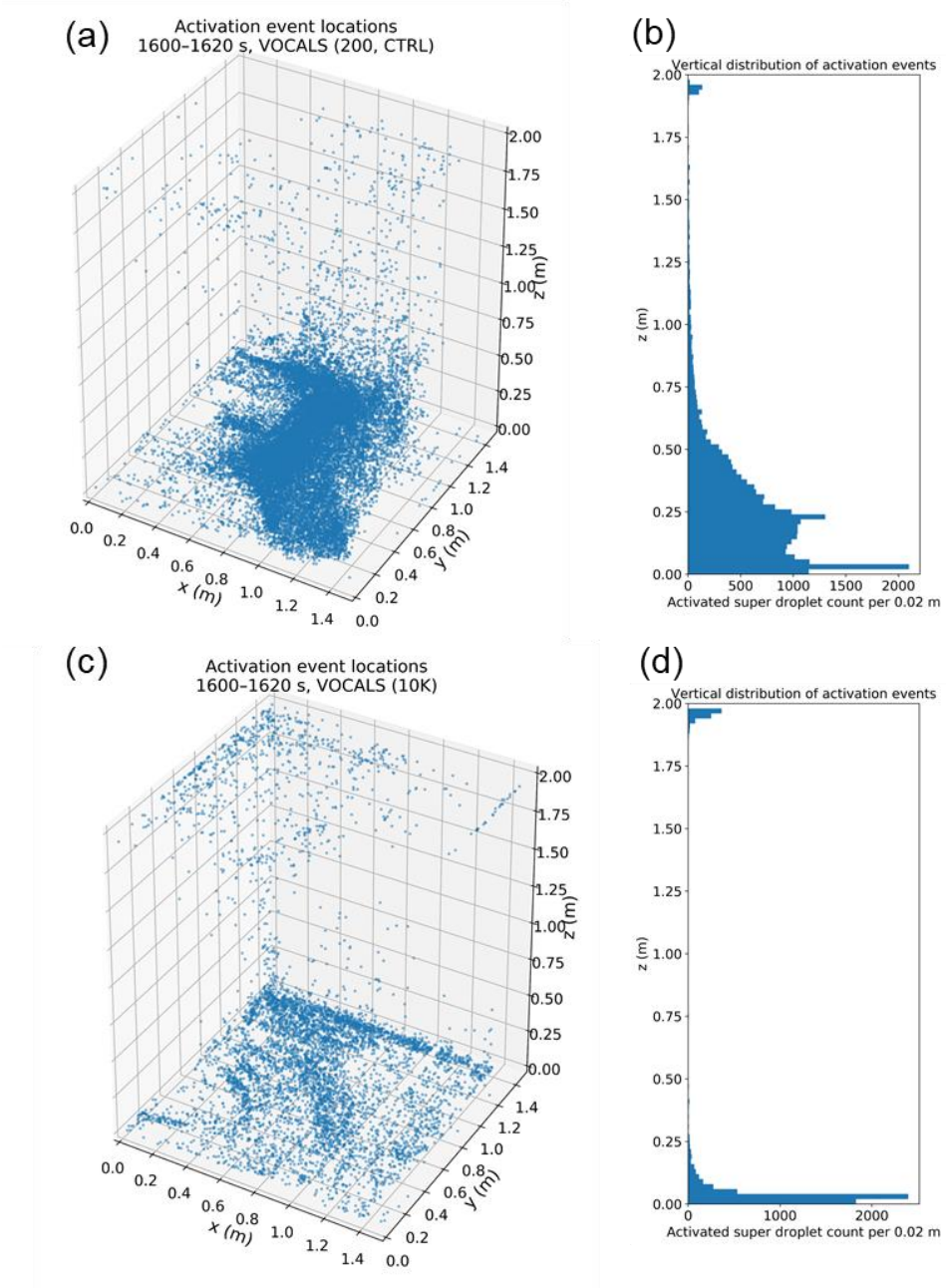

Figure 15. Spatial distribution and vertical frequency of activation events between 1600 and 1620 s. Panels (a) and (c) show three-dimensional locations of the first activation events for 50,000 SDs that were randomly selected from the population of unactivated SDs located between 0 and 0.2 m at 1600 s for VOCALS (200, CTRL) and VOCALS (10K) cases, respectively. Panels (b) and (d) show the corresponding vertical distributions of activation events, expressed as the number of activated super droplets per 0.02 m height bin.

Lines 456 and 463: After reading the paper and looking at the evidence, I disagree with the statements that “droplets experienced sustained condensational growth during ascent” and “droplet growth (dr) and activation strongly correlated with upward displacement”.

Response: We thank the reviewer for this insightful and critical comment. We have revised the corresponding sentences in the Conclusions to more accurately reflect the presented evidence (Lines 547-558):

"Conversely, at high aerosol concentrations, the short phase relaxation time induces a low quasi-steady supersaturation throughout the chamber, so condensational growth changes little with height and the cloud water mixing ratio profile becomes nearly vertically uniform. The Damköhler number ($Da = \tau_m/\tau_p$), defined as the ratio of turbulent mixing timescales (τ_m) to microphysical timescales (τ_p), provides a useful framework for interpreting this transition: under clean conditions, Da is of order unity, so condensation and vertical mixing act on comparable timescales, allowing vertical transport to imprint supersaturation variability on droplet growth. In contrast, polluted conditions correspond to large Da ($\gg$ 1), indicating that condensation depletes supersaturation more rapidly than mixing can regenerate it. Lagrangian trajectory analyses further show that in clean cases, droplet growth and activation are maximized for trajectories with moderate upward displacement (normalized vertical displacement) that remain long within the supersaturation source region near the chamber base. In contrast, in polluted cases, the dependence of growth and activation on normalized vertical displacement is much weaker, consistent with the more uniform and strongly damped supersaturation field."

References

Chandrakar, K. K., Cantrell, W., Krueger, S., Shaw, R. A., and Wunsch, S.: Supersaturation fluctuations in moist turbulent Rayleigh–Bénard convection: a two-scalar transport problem, J. Fluid Mech., 884, A19, https://doi.org/10.1017/jfm.2019.895, 2020.

Grabowski, W. W., Chandrakar, K. K., and Morrison, H.: Broadening of adiabatic droplet spectra through eddy hopping: Polluted versus pristine environments, J. Atmos. Sci., in review, 2026.

Anonymous Referee #3 (RC2)

The manuscript examines how aerosol loading and vertical transport jointly shape cloud microphysical structure in a turbulent convection chamber. The simulations are conducted using large-eddy simulations coupled with a Lagrangian super-droplet model, and the main findings focus on mean vertical profiles of liquid water content within the chamber. The results indicate that cloud water generally increases with height due to continued droplet activation and growth during ascent. However, this vertical gradient weakens substantially under polluted conditions, where enhanced competition for water vapor suppresses supersaturation variability and leads to more vertically uniform cloud water profiles. These findings underscore the role of aerosol loading in controlling vertical cloud microphysics and emphasize the coupled effects of vapor competition and turbulent vertical dynamics, with potential implications for aerosol–cloud interaction parameterizations in both laboratory and atmospheric contexts. While the topic of the manuscript is highly interesting and detailed modeling is indeed required to understand the processes governing droplet formation in cloud chambers, I have significant reservations regarding both the content and the conclusions drawn. Consequently, I cannot recommend acceptance of the manuscript in its current form. The analysis is highly technical and limited to a single experimental configuration intended to mimic a specific chamber setup. However, I do not find sufficient scientific novelty in the results when compared to previous studies using the same modeling framework or to the established understanding of how aerosol concentration controls supersaturation. In this respect, the manuscript may be more suitable for a journal with an instrumental or methodological focus, such as Atmospheric Measurement Techniques or a similar outlet.

Response: We thank the referee for the careful reading and for the clear summary of the manuscript. We also appreciate the referee's legitimate concern that the current results may appear to have insufficient scientific novelty relative to established understanding of aerosol control on supersaturation.

We agree that the qualitative damping of supersaturation (S) variability at higher CCN is well known. Our novelty is therefore not the existence of this damping, but the advances in three specific aspects, when compared to established understanding from prior chamber simulations: we show how the damping reorganizes the vertical cloud-water structure $q_c(z)$ in a turbulent, wall-forced chamber (i.e., the transition from strong increase with height to nearly uniform profiles); using the Lagrangian framework, we quantify the corresponding collapse of growth-history sensitivity across droplets originating from the same near-bottom air mass; and to address the concern that the study uses a single chamber configuration, we interpret these behaviors through a nondimensional timescale framework ($Da = \tau_m/\tau_p$) that identifies a transition from a history-sensitive to a history-insensitive regime, providing a geometry-independent process explanation.

To strengthen the atmospheric relevance (and ACP suitability), we revised Section 4 and the Concluding Remarks to explicitly connect this Da-based regime view to Lagrangian interpretations of condensational growth in turbulent shallow clouds (e.g., eddy-hopping and condensational broadening) and to clarify the distinction between chamber circulation and atmospheric updraft terminology. This interpretation is consistent with recent adiabatic cloud simulations in which the main analysis is based on a 1D turbulent-parcel framework,

supported by additional 3D LES results. These simulations show that eddy hopping effect is stronger in pristine conditions because a longer phase relaxation time allows larger supersaturation fluctuations for comparable vertical-velocity variability, and a larger peak cloud-base supersaturation activates a broader range of CCN (Grabowski et al., 2026). This framing emphasizes a nondimensional process interpretation that can be transferred beyond a specific chamber geometry (Lines 515-524, 577-604):

"From a Lagrangian perspective, we interpret our diagnostics as follows. At a given time, the super droplets within the lowest 0.2 m can be viewed as originating from a common near-bottom air mass. Turbulent motions then disperse these droplets along trajectories that differ in their net ascent and in their residence time within the near-bottom supersaturation source region. These differences produce distinct time-integrated supersaturation exposures, and therefore distinct condensational growth histories, even for droplets that start from the same near-bottom layer. In the clean (low-CCN) cases, supersaturation fluctuations are relatively large, so growth becomes strongly history dependent and the normalized radius increment, *dr*, varies substantially across trajectories. In the polluted (high-CCN) cases, supersaturation fluctuations are damped, so droplets experience more similar effective supersaturation histories and *dr* becomes more uniform. In our simulations, this transition is accompanied by a weakening of the vertical gradient in q_c, consistent with reduced contrasts between strongly ascending and weakly ascending trajectories as CCN increases."

"This behavior can be summarized by a timescale perspective in which *Da* ($= \tau_m/\tau_p$) compares a chamber-scale transport time, τ_m, to the phase relaxation time, τ_p. As CCN loading increases, the total droplet surface area increases, τ_p shortens, and supersaturation perturbations are relaxed more rapidly toward quasi-equilibrium. The resulting reduction in supersaturation variance and narrowing of growth histories is consistent with laboratory evidence from the Pi Chamber (Chandrakar et al., 2016). Our simulations extend this picture by showing, in a fully three-dimensional Lagrangian framework, how the same mechanism links supersaturation damping to both the vertical structure of q_c and the sensitivity of condensational growth to vertical history for droplets originating from the same near-bottom air mass."

"In atmospheric clouds, this timescale view is closely related to eddy-hopping and stochastic parcel frameworks for condensational broadening in shallow cumulus and stratocumulus. When Da is unity, droplets adjust slowly enough that differences in Lagrangian residence time within regions of enhanced supersaturation are efficiently translated into differences in condensational growth, favoring spectral broadening. When Da is large, supersaturation is homogenized more rapidly, so droplets following distinct trajectories can nevertheless experience similar effective supersaturation histories and grow by comparable amounts. This suggests that higher CCN loading can weaken the imprint of turbulent history on condensational broadening in cloud interiors, consistent with stochastic entraining parcel studies (e.g., Abade et al., 2018). This interpretation is also consistent with recent adiabatic cloud simulations based primarily on 1D turbulent-parcel modeling, complemented by 3D LES cloud simulations, showing that eddy hopping effect and the associated turbulence-driven condensational broadening are stronger under pristine conditions because a longer phase relaxation time allows larger supersaturation fluctuations for the same turbulent vertical-velocity variability, and a larger peak cloud-base supersaturation activates a broader range of CCN (Grabowski et al., 2026).

We note that the Da used here differs from the Da commonly adopted in entrainment-mixing studies at cloud edges, where Da often compares a microphysical adjustment time (frequently evaporation in subsaturated entrained air) to a small-scale turbulent mixing time across cloudy and environmental filaments. In that framework, large Da implies microphysics can act faster than mixing, promoting inhomogeneous mixing signatures. By contrast, our Da compares phase relaxation to a chamber-scale transport time in a near-saturated, quasi-closed environment where condensational relaxation dominates. There, a shorter τ_p strengthens the negative feedback on supersaturation perturbations, so increasing CCN primarily damps supersaturation fluctuations and homogenizes condensational growth histories in the chamber interior. In atmospheric clouds, both behaviors can coexist, with CCN damping interior supersaturation variability while entrainment at cloud edges can still drive locally inhomogeneous mixing depending on humidity deficits and mixing scales."

I also agree with the first reviewer regarding issues with terminology. Although I am not an expert in cloud chamber experiments, I find the discussion of updrafts to be potentially misleading. The manuscript explicitly states that "the positive correlation between supersaturation (S) and vertical velocity (W) in the chamber arises not from adiabatic cooling driven by the updraft itself, but rather from the mixing of air volumes with different thermodynamic properties originating from the lower and upper boundaries." Given that the dynamical processes in the chamber differ substantially from those in real atmospheric clouds, the interpretation of droplet activation in terms of updrafts at different altitudes appears questionable. If activation is primarily driven by mixing, one would expect similar effects to occur in downdrafts as well, which is not addressed in the discussion

Response: We thank the referee for this important clarification regarding terminology and the interpretation of vertical motion in the chamber. We agree that supersaturation in a convection cloud chamber is generated primarily by turbulent mixing of air masses with different thermodynamic properties, rather than by adiabatic cooling in an atmospheric-updraft sense. To avoid confusion, we revised the text to de-emphasize "updraft" terminology and to describe the flow in terms of the ascending and descending branches of the chamber circulation and their associated mixing pathways (Lines 231–233):

"indicating a relatively strong vertical increase in q_c that reflects efficient droplet activation and condensational growth in the lower part of the chamber where mixing between warm, moist air from the bottom boundary and cooler air aloft generates enhanced supersaturation"

Regarding the referee's point about downdrafts, our simulations show that descending motions in the chamber interior are predominantly associated with subsaturation and therefore with low q_c. This is illustrated in Fig. 8 (computed from randomly sampled interior grid points away from chamber boundaries), where negative S occurs preferentially for $W<0$. Physically, this arises because the descending branch carries air that has been cooled aloft and typically depleted in water vapor by prior condensation; when this air mixes with its surroundings, the resulting water-vapor mixing ratio is often insufficient to maintain saturation at the local temperature, yielding $S<0$ and favoring evaporation rather than continued activation and growth. Similar behavior has also been reported in other chamber simulations (e.g., Wang et al., 2024, Fig. 7), which show reduced q_c (and smaller droplets) in descending regions. We therefore interpret activation and sustained condensational growth as occurring where mixing

produces supersaturation, whereas descending motions more often sample mixing states that yield subsaturation and reduced q_c. We have added a brief description of this point in the main text to more directly illustrate the W–S coupling using the S–W scatterplots (Lines 314–318):

"To illustrate this *W–S* coupling more directly, Figure 8 shows the scatterplot of supersaturation and vertical velocity. The scatterplots show that positive *S* values are preferentially associated with updrafts (*W*>0), whereas negative *S* occurs mainly in downdrafts (*W*<0), consistent with condensation being most effective during ascent. As aerosol concentration increases, both the positive and negative *S* tails contract toward zero, making the *W–S* relationship more symmetric and thereby weakening the *W–S* coupling. "

Some specific comments:

Lines 194–195: "The denominator is the mean of qc at both levels, where active cloud formation occurs during the quasi-equilibrium period." Is droplet activation assumed to occur only near the top and bottom of the chamber? If so, what is the physical justification for this assumption? Given the turbulent nature of the chamber, mixing should occur throughout the domain, and activation would therefore not be limited to only two vertical levels.

Response: We thank the referee for pointing out that the wording in Lines 194–195 was potentially misleading. We did not intend to imply that droplet activation is confined only to two discrete heights. The levels z=0.3 m and z=1.7 m are used only as reference heights to define a normalized metric of the vertical contrast in q_c during the quasi-equilibrium period. We have revised the text accordingly by removing the phrase "where active cloud formation occurs" and clarifying that the denominator serves only as a normalization factor. The two reference heights are chosen to be symmetrically offset from the lower and upper plates (0.3 m from each boundary) to reduce sensitivity to boundary-adjacent layers (Lines 226-229):

"where the numerator is the horizontal mean q_c at z=1.7 m minus the horizontal mean q_c at z=0.3 m, and the denominator is the mean q_c at these two reference heights during the quasi-equilibrium period, which is used to normalize the vertical-contrast metric. These heights are chosen to be symmetrically offset from the lower and upper plates (0.3 m from each boundary) to reduce sensitivity to boundary-adjacent layers."

Lines 216–218: "…the number of droplets increases, leading to stronger competition for available water vapor. As a result, each droplet grows more quickly, consuming vapor rapidly… This rapid vapor depletion suppresses the buildup of S during ascent, thereby limiting the vertical increase in qc." This description appears inconsistent. Individual droplets should grow more slowly when supersaturation is reduced, even though the total condensational growth rate (and thus qc) may increase due to a higher number of droplets. Please rephrase to clearly distinguish between individual droplet growth and bulk condensational growth.

Response: We thank the referee for pointing out this inconsistency. Our wording was incorrect: under high CCN loading, individual droplets do not grow faster because supersaturation is

reduced. Instead, the larger total droplet surface area strengthens the bulk condensational sink, so supersaturation relaxes more rapidly toward a low quasi-steady value (shorter τ_p). In this sense, droplet growth equilibrates sooner and additional growth aloft is limited, which weakens the vertical increase in q_c. We have revised the text to clearly distinguish individual droplet growth from the bulk condensational uptake (Lines 253-256):

"As a result, supersaturation is driven rapidly toward a low quasi-steady value, so individual droplets experience weaker supersaturation and grow more slowly, but the population equilibrates sooner and exhibits limited additional growth aloft, thereby weakening the vertical increase in q_c."

Figure 3: How do the mean droplet size and droplet number concentration vary with height in the chamber? Is it possible that the more vertically uniform qc profiles arise from changes in supersaturation as a function of height and its interaction with the aerosol distribution? Because the chamber top is colder, condensation rates should be reduced, potentially facilitating more efficient activation and higher droplet number concentrations. As aerosol concentration increases, critical supersaturation increases and small variations in become less important, which may naturally lead to weaker vertical gradients in qc.

Response: To answer the referee's question, we draw the diagnostic figure (Figure R3) that show the vertical contrasts ΔN_c and ΔR_m defined analogously to Δq_c and averaged over the quasi-equilibrium period (15–30 min). As shown in Fig. R3, ΔN_c decreases systematically with increasing aerosol concentration, approaching small (near-zero) values at high CCN loading, consistent with the reduced vertical contrast in q_c under polluted conditions. In contrast, ΔR_m exhibits no clear monotonic dependence on aerosol concentration. These results indicate that the aerosol-dependent weakening of the vertical q_c gradient is primarily associated with a reduced vertical contrast in droplet number (i.e., diminished additional activation aloft), rather than a systematic increase of droplet size aloft.

Finally, the scatterplot of *S*–*W* (Fig. 8) show that descending motions in the chamber interior are predominantly subsaturated, and downdraft regions are associated with lower q_c and smaller droplets, consistent with other chamber simulations (e.g., Wang et al., 2024, Fig. 7). Together, these diagnostics support our interpretation that higher CCN loading damps supersaturation variability and reduces the sensitivity of activation and condensational growth to vertical history, leading to more vertically uniform q_c profiles.

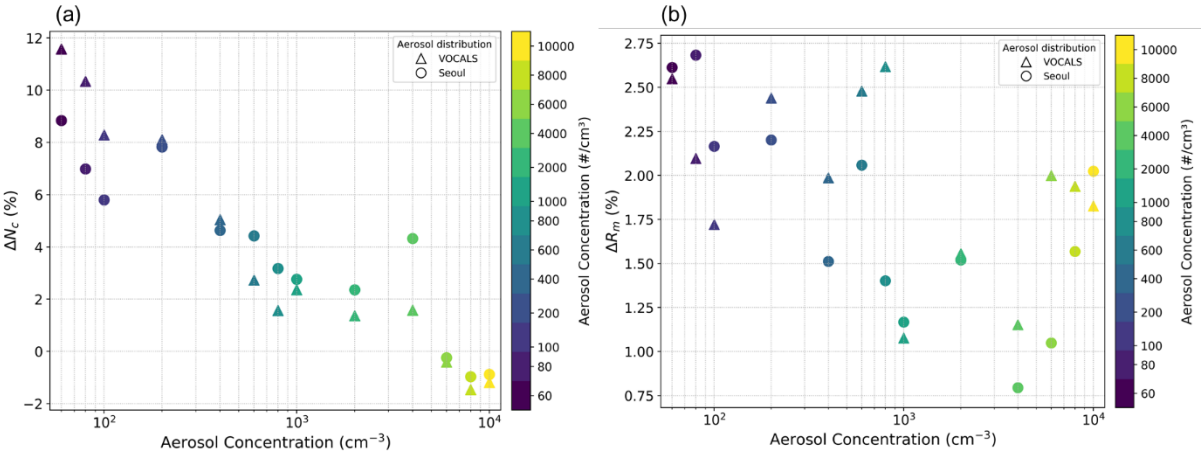

Figure R3. Vertical contrasts in droplet number concentration (N_c) and mean droplet size (R_m) as a function of aerosol concentration. Panel (a) shows ΔN_c (%) and panel (b) shows ΔR_m (%), where ΔX is defined analogously to Δqc as $\Delta X=(X_{1.7\,\mathrm{m}}-X_{0.3\,\mathrm{m}})/[0.5\,(X_{1.7\,\mathrm{m}}+X_{0.3\,\mathrm{m}})]\times100\%$, with X denoting the horizontal-mean value at each height. All quantities are averaged over the quasi-equilibrium period (15–30 min). Triangles denote VOCALS cases and circles denote Seoul cases; marker colors indicate aerosol concentration (cm^{-3}).

Figure 5: The manuscript repeatedly states that supersaturation fluctuations are maintained by mixing of air parcels. However, from basic thermodynamics, mixing two supersaturated air parcels at different temperatures should still yield supersaturated air. Since all boundaries in the model domain are supersaturated with respect to water, what mechanism produces negative supersaturation values in the simulation? Additionally, what causes the strong fluctuations in mean S near the top and bottom boundaries?

Response: That is basically because of condenesational loss of water vapor. Negative supersaturation (S<0) occurs when descending air that has been cooled aloft and typically depleted in water vapor by prior condensation mixes with its surroundings; the resulting water-vapor mixing ratio can be insufficient to maintain saturation at the local temperature, yielding S<0 and favoring evaporation rather than continued activation and growth. Similar behavior has also been reported in other chamber simulations (e.g., Wang et al., 2024, Fig. 7), which show reduced q_c (and smaller droplets) in downdraft regions.

The strong oscillations in the mean S profile near the top and bottom plates are the same issue raised by Anonymous Referee #2 and are discussed in the revised manuscript (Lines 301–309):

"Before interpreting this relationship, we address a potential concern raised by the mean supersaturation profiles in Figure 5a,b, which exhibit oscillations near the lower and upper rigid boundaries. Two factors likely contribute to these near-wall oscillations. First, resolved transport and mixing are inefficient near rigid boundaries in ILES, which can allow sharp gradients to persist. Second, supersaturation is a diagnosed quantity, so small grid-scale perturbations in temperature and water vapor can translate into comparatively large fluctuations in S because of the nonlinear saturation relationship and advection–condensation coupling (Grabowski and Smolarkiewicz, 1990). Importantly, the W–S correlations reported

in Figure 7 are computed over the interior region away from the plates, and the aerosol-dependent trends in Figure 7 remain unchanged when the boundary-adjacent layers are included (not shown). With this in mind, we interpret the aerosol dependence of W–S coupling as follows."

We attribute these near-wall oscillations primarily to the ILES configuration, where resolved transport and mixing are inefficient near rigid boundaries, and to the fact that supersaturation is a diagnosed nonlinear quantity. So small grid-scale perturbations in temperature and water vapor can translate into comparatively large fluctuations in S via advection–condensation coupling (Grabowski and Smolarkiewicz, 1990). Importantly, the W–S correlations in the main analysis are computed over the interior region excluding boundary-adjacent layers, and a sensitivity test including the boundary layers (and the full domain) shows that while the absolute correlation magnitude is reduced, the aerosol-dependent trends remain unchanged (Fig. R1). While Fig. 5 shows intermittent oscillations with both positive and negative S near the boundaries, the tendency for predominantly positive supersaturation in the near-bottom boundary layer is a robust feature also reported in one-dimensional turbulence model (Chandrakar et al., 2020). This supports our interpretation of the lower layer as a potential activation region. We have added this clarification and the supporting citations in the discussion of Fig. 5 in the revised manuscript (Lines 259-263):
"Notably, the near-bottom layer exhibits predominantly positive mean supersaturation, although both positive and negative values occur intermittently. This behavior is consistent with moist Rayleigh–Bénard convection results from a one-dimensional turbulence (ODT) framework (Chandrakar et al., 2020), supporting the role of the lower boundary layer as a plausible activation region"

Lines 231–232: "In contrast, because our size distribution spans multiple modes, $\bar{s}$ does not collapse exactly to one number but instead asymptotes to a narrow range just above the largest-mode critical S (∼0.02%)." This explanation is difficult to justify physically. The close correspondence between $\bar{s}$ and the largest-mode critical supersaturation may be coincidental rather than mechanistically constrained.

Response: We realize that "the largest aerosol mode" was not clearly understood. What we intended to say is that the minimum critical supersaturation is designated to the largest particles in the aerosol distribution. We have revised the text to state this explicitly and removed "largest aerosol mode." We also clarify that as aerosol loading increases, $\bar{S}$ approaches this minimum S_{crit}, so particles near this threshold can alternate between weak activation and deactivation as S fluctuates around the threshold (Lines 270-279):

"The minimum critical supersaturation (S_{crit}) in the prescribed aerosol spectrum is approximately 0.02%, designated to the largest aerosol particles in the size distribution. As aerosol loading increases, $\bar{S}$ steadily falls toward this value, often converging near 0.02% (see Fig. 6). This behavior is qualitatively consistent with Yang et al. (2025), who found that mean supersaturation approached the critical value for their monodisperse aerosol. In our polydisperse case, $\bar{S}$ does not collapse to a single value, but instead converges to a narrow range near the minimum S_{crit} set by the largest particles. When $\bar{S}$ is near this threshold, particles with S_{crit} close to $\bar{S}$ can repeatedly transition between weak activation and

deactivation as turbulent S fluctuations move conditions slightly above or below their critical value, which effectively buffers $\bar{S}$. This buffering helps explain why, beyond a certain aerosol concentration, the domain-mean q_c no longer increases (Fig. 2): enhanced vapor competition limits additional net activation and condensational growth despite further increases in aerosol loading."

Lines 234–236: Can this hypothesis be tested by explicitly accounting for water uptake in the aerosol phase? What role does water availability from the chamber walls play, and could it contribute to the apparent stagnation behavior?

Response: We tested this hypothesis by separating liquid water into "cloud" water (our operational definition, r>1μm) and total liquid water (all particles), and defining a haze-water reservoir $q_{c,haze} \equiv q_{c,total} - q_{c,cloud}$. We find that $q_{c,haze}$ increases systematically with aerosol concentration (Fig. R4), indicating that under polluted conditions an increasing fraction of condensate resides in sub-micron, near-threshold particles rather than in fully activated cloud droplets. This supports our interpretation that near-threshold activation–deactivation (haze–droplet interaction) might contribute to the apparent stagnation of $q_{c,cloud}$ at high aerosol loading. We also note that, at a given aerosol number concentration, VOCALS cases tend to exhibit larger $q_{c,haze}$ than Seoul cases, consistent with its size spectrum containing a larger fraction of large dry aerosol particles (Fig. 1) that can hold more water below the r>1μm cloud threshold.

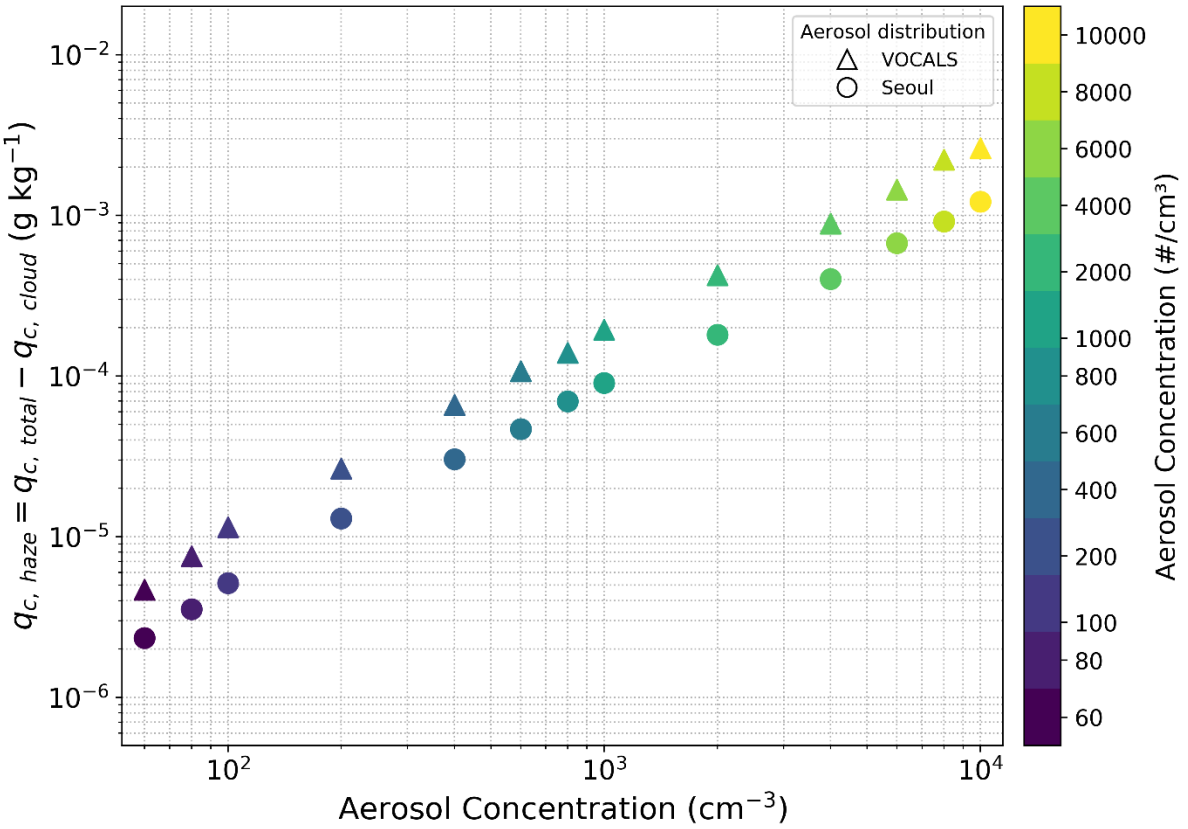

Figure R4. Aerosol-loading dependence of the "haze water" proxy $q_{c,haze} \equiv q_{c,total} - q_{c,cloud}$. Here,

$q_{c,cloud}$ is defined using the paper's cloud criterion (liquid water from particles with r>1 μm), while $q_{c,total}$ includes all liquid water (r>0 μm). Markers denote aerosol size distributions (VOCALS: triangles; Seoul: circles), and colors indicate the prescribed aerosol concentration (cm^{-3}); both axes are shown on logarithmic scales. Each point represents the averaged value for the quasi-equilibrium period (15–30 minutes).

Lines 264–265: "Downward motions near the upper boundary carry cooler and drier air down into the chamber, generating negative S perturbations." According to the model setup, the upper boundary is at 100% relative humidity. If mixing is the dominant process affecting temperature and moisture, why do negative supersaturation values form?

Response: Although the upper boundary is prescribed at RH = 100%, that air is saturated at a colder temperature and therefore has a low absolute water-vapor mixing ratio. When this cold, low-q_v air is transported downward and mixed into the warmer chamber interior, the local saturation mixing ratio increases rapidly with temperature, while q_v remains almost the same. As a result, the mixed air is often unable to maintain saturation at the local temperature and becomes subsaturated (S < 0). This is consistent with our diagnostics (Fig. 8), which show that negative S occurs preferentially in descending motions (W < 0), and with prior chamber studies reporting negatively skewed S and the most negative S in downdrafts (e.g., Anderson et al., 2021). We revised the text to clarify that negative S is associated with the warming and mixing of cold saturated air from aloft into a warmer environment, rather than implying that the upper boundary itself is "dry." (Lines 322-325):

"Conversely, downward motions transport air that is saturated at the cold upper boundary (low absolute q_v) into the warmer chamber interior; as this air warms and mixes, saturation vapor mixing ratio increases faster than q_v, so the local relative humidity decreases and subsaturation ($S < 0$) can occur. This behavior is consistent with observations from laboratory chamber experiments, which report the most negative S in downdrafts (Anderson et al., 2021)."

Figure 10 and related analysis: The main message of this figure is unclear. Please explain more explicitly how this analysis supports the manuscript's conclusions. Why is the analysis restricted to particles initially located near the bottom of the chamber rather than applied to all particles?

Response: The goal of Figure 11 (formerly Fig. 10) and related analysis is to define a simple Lagrangian metric that quantifies each droplet's recent vertical-history (predominantly ascending vs. mixed ascent–descent) and to show that this metric clearly separates distinct transport pathways in the chamber. This supports our main conclusion that vertical-history can strongly differentiate growth in clean cases, but that this sensitivity diminishes at high aerosol loading when supersaturation rapidly relaxes toward a quasi-steady value ($\tau_p \ll \tau_m$).

We restrict the analysis to SDs sampled from the same near-base source layer (z = 0–0.2 m) at the start of each 20 s tracking window so that all trajectories begin from a comparable origin region. This reduces a major confounding factor: if SDs from the full depth were included, differences in growth and height would largely reflect where droplets happened to be at the

start (e.g., proximity to the lower vs. upper boundary and the associated S environment) rather than differences produced by turbulent transport and supersaturation history along the trajectories.

Lines 363–364: “Suggesting that condensational growth becomes increasingly decoupled from vertical motion due to limited S variability under polluted conditions.” Or simply because there is more particles to use the same amount of water that is limited by the transport rate from the walls, and that’s why the relative growth is less.

Response: We agree with the referee that moisture availability in the chamber is ultimately controlled by the rate at which water vapor is supplied from the boundaries and transported into the interior, so that increasing the number of particles can reduce the per-particle growth simply by partitioning a finite vapor supply among more droplets. In our framework, this effect is expressed through the same source–sink balance that determines the quasi-steady supersaturation: higher aerosol loading increases the total droplet surface area and strengthens the condensational sink (shorter τ_p), so supersaturation is rapidly buffered near a low quasi-steady value with reduced variability. As a result, condensational growth becomes less sensitive to vertical-motion-induced supersaturation fluctuations, which is what we referred to as “decoupling.” We have revised the sentence to make this source–sink (transport-limited source and sink-enhanced relaxation) interpretation explicit (Lines 434-437):

“Suggesting that condensational growth becomes less sensitive to vertical motion because S is rapidly buffered near a low quasi-steady value set by the balance between the boundary-forced vapor supply (transport) and the enhanced condensational sink at high aerosol loading (short τ_p).”

References

Chandrakar, K. K., Cantrell, W., Krueger, S., Shaw, R. A., and Wunsch, S.: Supersaturation fluctuations in moist turbulent Rayleigh–Bénard convection: a two-scalar transport problem, J. Fluid Mech., 884, A19, https://doi.org/10.1017/jfm.2019.895, 2020.

Grabowski, W. W., Chandrakar, K. K., and Morrison, H.: Broadening of adiabatic droplet spectra through eddy hopping: Polluted versus pristine environments, J. Atmos. Sci., in review, 2026.