

## 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,  $\tau_p$ ) to a chamber-scale transport time ( $\tau_m$ ), summarized by  $Da (= \tau_m/\tau_p)$ , (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  $Da (= \tau_m/\tau_p)$  compares a chamber-scale transport time,  $\tau_m$ , to the phase relaxation time,  $\tau_p$ . As CCN loading increases, the total droplet surface area increases,  $\tau_p$  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  $\tau_p$  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  $3 \times 10^{-3} \text{ m}^2 \text{ s}^{-2}$  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  $\approx 5\%$ , SD  $\approx 0.9\%$ ) 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  $\sigma_S$  and the  $W$ – $S$  correlation (not shown)”

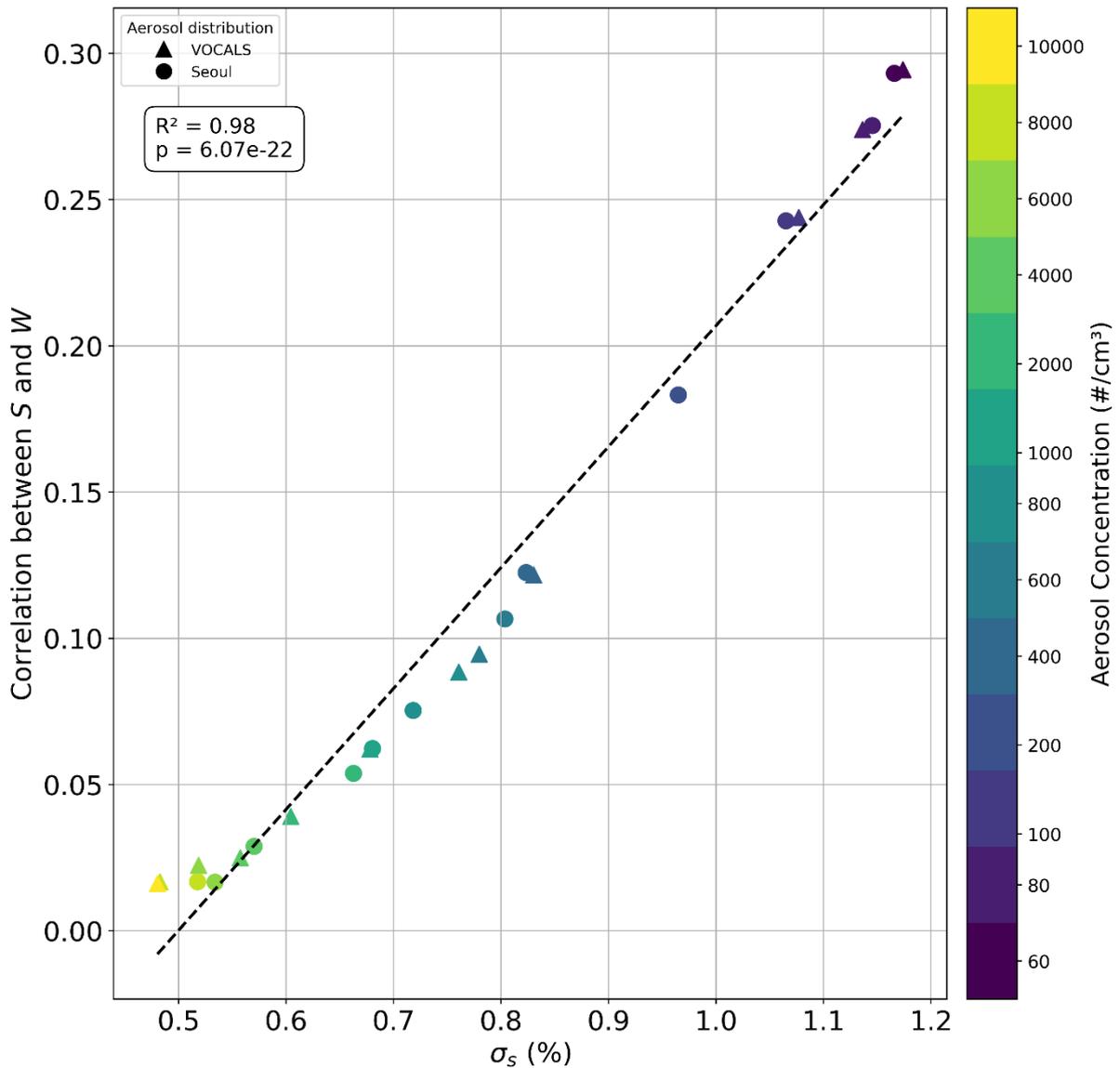


Figure R1. Scatter plot showing the relationship between the domain-averaged supersaturation standard deviation ( $\sigma_s$ ) and the correlation coefficient between supersaturation ( $S$ ) and vertical velocity ( $W$ ). Both  $\sigma_s$  and  $\text{corr}(S, W)$  are calculated using all grid points 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  $\tau_p$  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  $\tau_p$  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  $\text{cm}^{-3}$ , 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  $\mu\text{m}$  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  $\Delta q_c$  using an activation criterion based on the critical radius of each super droplet ( $r >$  critical radius). The resulting dependence of  $\Delta q_c$  on aerosol concentration (Fig. R2) is nearly indistinguishable from that obtained with the original  $r >$  1  $\mu\text{m}$  definition (Fig. 3), with a mean relative difference of about 1.6 % between the  $\Delta q_c$  values computed using the  $r >$  1  $\mu\text{m}$  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  $\mu\text{m}$ , they already contribute to bulk condensate similarly to small activated droplets, so treating  $r >$  1  $\mu\text{m}$  hydrometeors as “cloud droplets” in the statistics has no significant influence on our interpretation of the results.

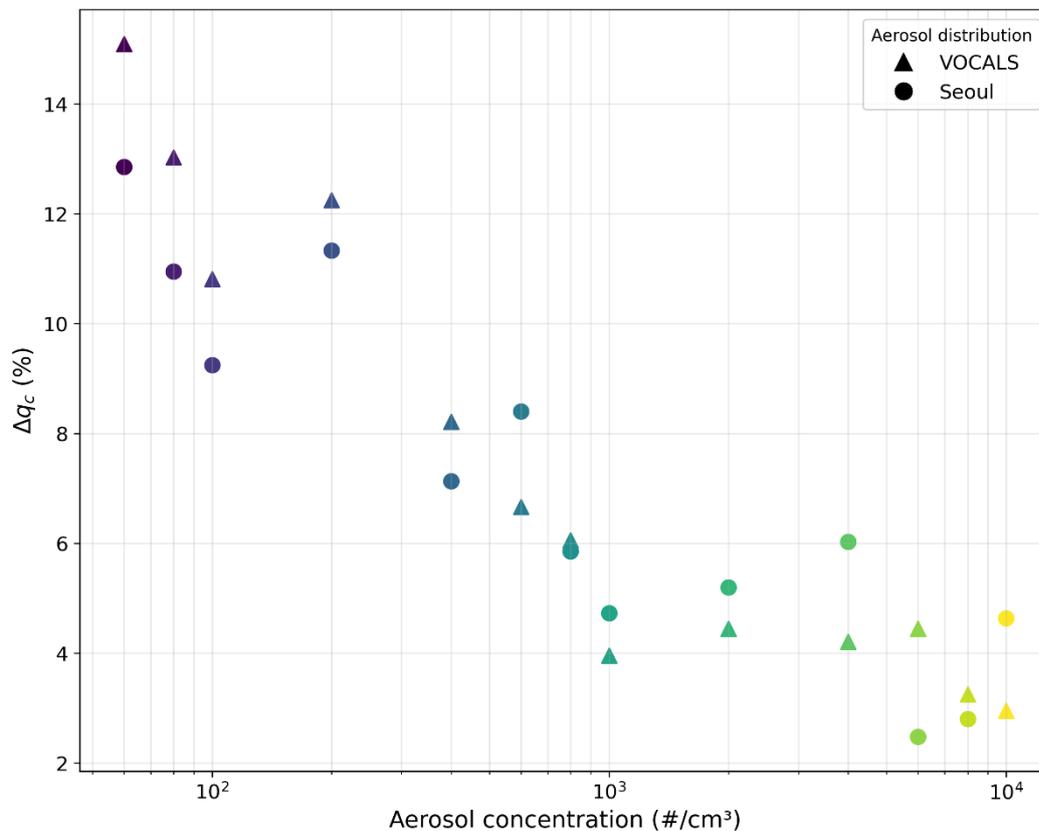


Figure R2.  $\Delta q_c$  as a function of aerosol concentration, where each point represents the averaged value for the quasi-equilibrium period (15–30 min) of each aerosol distribution. Here,  $\Delta q_c$  is computed using only droplets with a radius larger than their size-dependent critical radius ( $r > \text{critical radius}$ ). 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  $\Delta q_c$  within the caption.

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

"Large  $\Delta q_c$  indicates a strong increase of  $q_c$  with height, whereas small or negligible  $\Delta q_c$  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 ( $S_{\text{crit}}$ ) 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**  $S_{\text{crit}}$  set by the largest particles. When  $\bar{S}$  is near this threshold, particles with  $S_{\text{crit}}$  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  $\tau_p$  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  $\tau_m$  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  $\tau_p$ , 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 ( $\tau_p$ ), 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.”

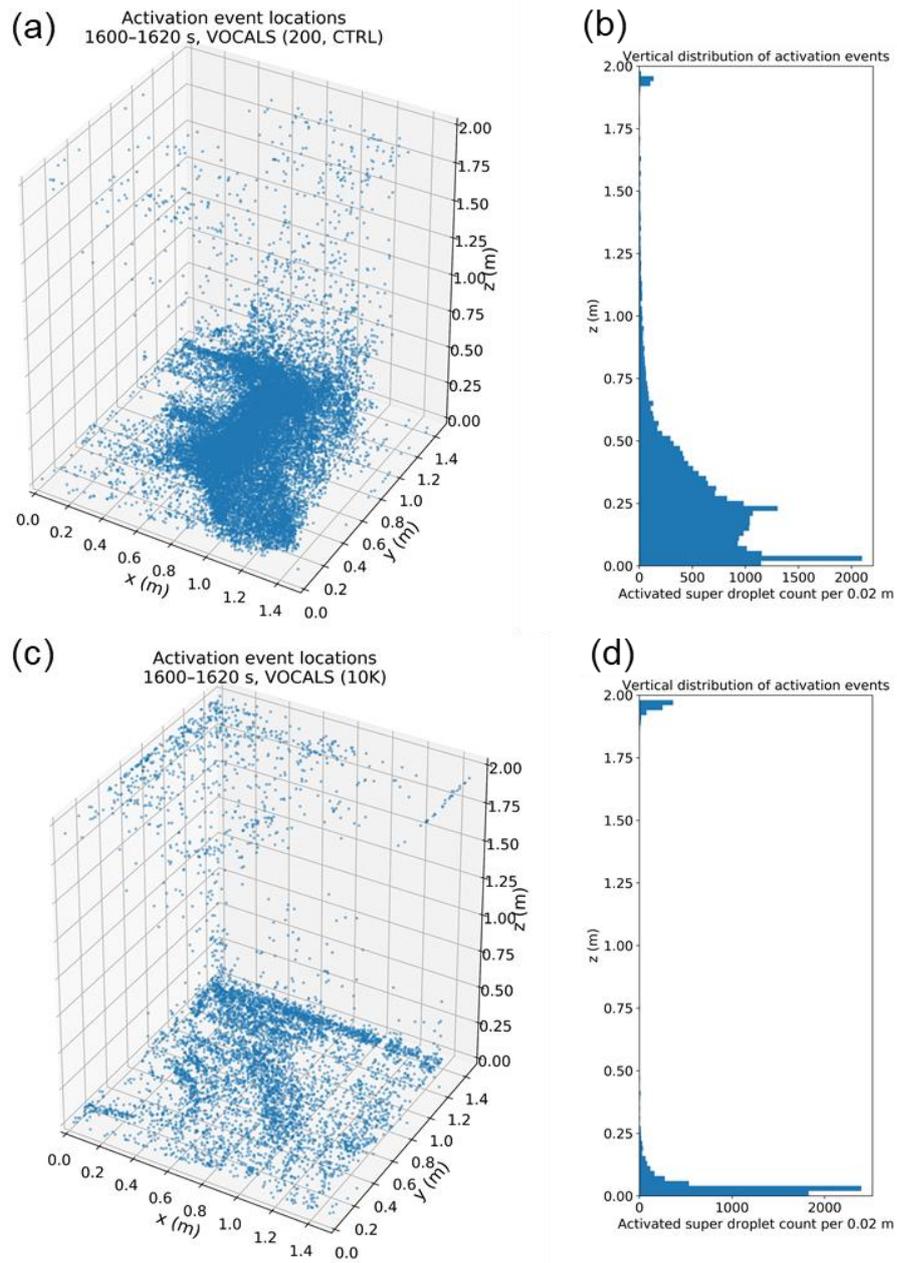


Figure 15. Spatial distribution and vertical frequency of activation events between 1600 and 1620 s. Panels (a) and (c) show three-dimensional locations of the first activation events for 50,000 SDs that were randomly selected from the population of unactivated SDs located between 0 and 0.2 m at 1600 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 ( $\tau_m$ ) to microphysical timescales ( $\tau_p$ ), 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

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