Life Cycle Evolution of Inhomogeneous Mixing in Shallow Cumulus Clouds

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Abstract

Understanding how entrainment and mixing shape the cloud droplet size distribution (DSD) is crucial for understanding the optical properties and precipitation efficiency of clouds. Different mixing scenarios, mainly homogeneous and inhomogeneous, shape the DSD in a distinct way and alter the cloud's impact on climate. However, the prevalence of these mixing scenarios and how they vary in space and time is still uncertain, as underlying processes are commonly unresolved by conventional numerical models. To overcome this challenge, we employ the \$L^3\$ model, which considers supersaturation fluctuations and turbulent mixing down to the finest relevant lengthscales, making it possible to represent different mixing scenarios realistically. We investigate the spatial and temporal evolution of mixing scenarios over the life cycle of shallow cumulus clouds for varying boundary layer humidities and aerosol concentrations. Our findings suggest homogeneous mixing is generally predominant in cumulus clouds, while different mixing scenarios occur concurrently in the same cloud. Notably, inhomogeneous mixing increases over the cloud life cycle across all analyzed cases. The mean and standard deviation of supersaturation are found to be the most capable indicators of this evolution, providing a comprehensive insight into the characteristics of mixing scenarios. Finally, we show inhomogeneous mixing is more prevalent in drier boundary layers and for higher aerosol concentrations, underscoring the need for a more comprehensive investigation of how these mixing dynamics evolve in a changing climate.











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Key Points: The evolution of predominant mixing scenarios during the shallow cumulus life cycle is investigated While homogeneous mixing is prevalent, inhomogeneous mixing gradually increases as the cloud ages Inhomogeneous mixing is more favored for higher aerosol concentrations and in drier boundary layers

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12 Abstract

Understanding how entrainment and mixing shape the cloud droplet size distribution (DSD) 13 is crucial for understanding the optical properties and precipitation efficiency of clouds. Dif-14 ferent mixing scenarios, mainly homogeneous and inhomogeneous, shape the DSD in a dis-15 tinct way and alter the cloud's impact on climate. However, the prevalence of these mixing 16 scenarios and how they vary in space and time is still uncertain, as underlying processes 17 are commonly unresolved by conventional numerical models. To overcome this challenge, we employ the L^3 model, which considers supersaturation fluctuations and turbulent mix-19 ing down to the finest relevant lengthscales, making it possible to represent different mixing 20 scenarios realistically. We investigate the spatial and temporal evolution of mixing scenar-21 ios over the life cycle of shallow cumulus clouds for varying boundary layer humidities and 22 aerosol concentrations. Our findings suggest homogeneous mixing is generally predominant 23 in cumulus clouds, while different mixing scenarios occur concurrently in the same cloud. 24 Notably, inhomogeneous mixing increases over the cloud life cycle across all analyzed cases. 25 The mean and standard deviation of supersaturation are found to be the most capable indi-26 cators of this evolution, providing a comprehensive insight into the characteristics of mixing 27 scenarios. Finally, we show inhomogeneous mixing is more prevalent in drier boundary lay-28 ers and for higher aerosol concentrations, underscoring the need for a more comprehensive 29 investigation of how these mixing dynamics evolve in a changing climate. 30

31 Plain Language Summary

Clouds play a crucial role in Earth's climate system by influencing the radiation bal-32 ance and moisture transfer. When clouds mix with their environment, a process known as 33 entrainment and mixing, the number and size of cloud droplets change, affecting cloud op-34 tical properties and their ability to precipitate. This mixing can happen in two major ways: 35 either the cloud and environmental air are well-mixed (homogeneous mixing) or unevenly 36 mixed (inhomogeneous mixing). However, accurately representing these effects, particularly 37 inhomogeneous mixing, is challenging in weather or climate models, as essential scales and 38 processes are unresolved. By applying a high-resolution model, we investigate how mixing scenarios evolve over the cloud life cycle. We show that while homogeneous mixing prevails, 40 older (mature or dissipating) clouds tend to mix more inhomogeneously. Moreover, clouds 41 in drier and more polluted environments are shown to favor inhomogeneous mixing. In these 42 environments, inhomogeneous mixing becomes the predominant mixing scenario for older 43 clouds, emphasizing the need for further investigations on the impact of different mixing sce-44 narios in a changing climate. 45

46 **1 Introduction**

The role of clouds in the climate system and their representation in climate models re-47 mains a significant uncertainty (Boucher et al., 2013; Forster et al., 2021). Entrainment and 48 mixing are among the most perplexing yet vital processes affecting the optical properties and precipitation efficiency of clouds by shaping the cloud droplet size distribution (DSD). 50 While the concept of extreme mixing scenarios, namely homogeneous and inhomogeneous, has been introduced a few decades ago (Baker et al., 1980a; Blyth, 1993), there are ongoing 52 discussions regarding the prevalence of specific mixing scenarios - whether each occurs pre-53 dominantly at the top, or lateral edges of a cloud, how it varies during the cloud life cycle, or 54 how environmental properties shape the mixing type (Burnet & Brenguier, 2007; Lehmann 55 et al., 2009; Jarecka et al., 2013; Schmeissner et al., 2015; Hoffmann et al., 2019; Yeom et 56 al., 2021; Desai et al., 2021; Gao et al., 2021; Hoffmann, 2023; Liu et al., 2023; Yeom et al., 57 2023). 58

In theory, the Damköhler number, defined as the ratio between the timescales for turbulent mixing (τ_{mix}) and the microphysical response (τ_{micro}), could be used to determine mixing scenarios (Baker & Latham, 1979; Baker et al., 1980b; Lehmann et al., 2009; Tölle ⁶² & Krueger, 2014). τ_{micro} quantifies the speed of droplet evaporation in the entrained air, and ⁶³ one example is the evaporation time scale (Lehmann et al., 2009; Jarecka et al., 2013; Tölle ⁶⁴ & Krueger, 2014), $\tau_{\text{evap}} = r_v^2 (-GS_e)^{-1}$, where $S_e < 0$ is the supersaturation of the entrained ⁶⁵ air and the growth parameter *G* summarizes the effects of vapor diffusion and heat conduc-⁶⁶ tion on droplet evaporation (Yau & Rogers, 1996). On the other hand, $\tau_{\text{mix}} = (l^2/\varepsilon)^{1/3}$ es-⁶⁷ timates how fast entrained air is homogenized with cloudy air by turbulent mixing, where *l* ⁶⁸ is the lengthscale of a scalar inhomogeneity, such as the entrained air inside the cloud, that ⁶⁹ breaks down to the Kolmogorov lengthscale through turbulent motions characterized by a ⁷⁰ turbulence kinetic energy dissipation rate ε .

When the turbulent mixing is faster than the microphysical response ($\tau_{mix} \ll \tau_{micro}$), 71 homogeneous mixing dominates, and all droplets experience the same subsaturation and par-72 tially evaporate without reducing the cloud droplet number concentration (N_c) . When the 73 turbulent mixing is slower than the microphysical response ($\tau_{micro} \ll \tau_{mix}$), inhomogeneous 74 mixing is expected, and only those droplets exposed to the entrained air are assumed to evap-75 orate completely, resulting in a decrease in N_c while the mean droplet size remains constant. 76 A third mixing scenario occurs when the DSD contains numerous small droplets or droplets 77 ascent during mixing. In this case, the mean droplet radius increases by the complete evap-78 oration of the smallest droplets, which narrows the DSD (Krueger et al., 2008; Luo et al., 79 2022; Lim & Hoffmann, 2023). Thus, each mixing scenario shapes the DSD in a distinct 80 way. 81

However, conventional models commonly fail to represent these distinct ways and as-82 sume homogeneous mixing on all unresolved scales, although it has been argued that ho-83 mogeneous mixing only dominates on the lengthscale below a few decimeters (Baker et al., 84 1980b; Lehmann et al., 2009). Such assumption can misrepresent DSD shape changes during 85 mixing, with commensurate effects on cloud optical properties (Chosson et al., 2007; Slawinska et al., 2008; Hill et al., 2009; Xu et al., 2022), as well as the generation of precipitation 87 (Hoffmann et al., 2019). While early studies suggested inhomogeneous mixing had a negligible effect on cloud optical properties (Hill et al., 2009), recent research indicates it can 89 reduce the cloud optical depth by up to 5% in stratocumulus clouds (Xu et al., 2022). This 90 indicates the need for mixing parameterizations that adjust cloud microphysics based on the 91 prevailing mixing scenario. 92

Early studies attempted to parameterize mixing scenarios using a predetermined pa-93 rameter, adjusting N_c based on an assumed mixing scenario (Morrison & Grabowski, 2008; 94 Hill et al., 2009). This approach, however, assumes a single mixing scenario for all clouds, 95 ignoring the possibility of multiple mixing scenarios occurring concurrently in the same 96 cloud. In recent years, studies focused on investigating the relationship between mixing sce-97 narios and various cloud properties (Tölle & Krueger, 2014; Pinsky et al., 2016; Luo et al., 98 2020, 2021, 2022; Lim & Hoffmann, 2023), leading to parameterizations that adapt to cloud 99 properties interactively (Xu et al., 2022). Thus, it is essential to gain comprehensive insights 100 into how mixing scenarios vary spatiotemporally and to identify the cloud properties and 101 environmental factors that influence this variability. 102

Still, examining mixing scenario characteristics is challenging due to the lack of mod-103 els representing all relevant scales of entrainment and mixing. In this study, we employ the 104 L^3 model, a large eddy simulation (LES) model coupled with a Lagrangian cloud model 105 (LCM) and the linear eddy model (LEM), which allows to explicitly represent LES sub-grid 106 scale mixing on scales as small as a few centimeters (Hoffmann & Feingold, 2019; Hoffmann 107 et al., 2019). This approach was found capable of considering and classifying different mix-108 ing scenarios in a single cumulus congestus cloud (Lim & Hoffmann, 2023). This study aims 109 to extend our understanding of the aforementioned three mixing scenarios by investigating 110 how these change in the life cycle of shallow cumulus clouds, developing naturally from a 111 maritime boundary layer. 112

Cumulus clouds exhibit distinct life cycles, which include developing, mature, and dis-113 sipating stages, each with distinct dynamical and microphysical characteristics (Grinnell et 114 al., 1996; Zhao & Austin, 2005; Katzwinkel et al., 2014). Although inhomogeneous mix-115 ing is less prevalent in cumulus clouds compared to weakly turbulent stratocumulus clouds 116 (Jarecka et al., 2013; Hoffmann et al., 2019), observational evidence suggests that the pre-117 dominant mixing type might change over the cloud life cycle (Lehmann et al., 2009; Schmeiss-118 ner et al., 2015). Therefore, investigating the life cycle dependency of mixing scenarios pro-119 vides ample opportunities to elucidate the relationship between mixing scenarios, environ-120 mental conditions, and microphysical parameters that may be applicable beyond cumulus 121 clouds. In addition to investigating the effect of the internal variability over the cloud life cy-122 cle on mixing scenarios, we investigate how varying aerosol concentrations and boundary 123 layer humidities affect these dynamics. 124

- Therefore, this study aims at deepening our understanding of mixing scenario dynamics, including:
 - Classifying various mixing scenarios within a single cumulus cloud.
 - Investigating how mixing scenarios evolve over the cloud life cycle.
 - Identifying which microphysical property or environmental condition affects these changes most.

This paper is structured as follows. Section 2 presents the L^3 model framework, simulation settings, and the method used to determine the cloud life stages. Section 3 shows the changes in the predominant mixing scenario type and the concurrent cloud properties over the cloud life cycle. Section 4 compares the vertical distribution of these properties under different aerosol concentrations and boundary layer humidities. Finally, we conclude our paper in Sec. 5.

137 **2** Model and Simulation

138 **2.1** The L^3 model

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This study uses the L^3 model, the LES-LCM-LEM coupled model, representing supersaturation fluctuations and turbulent mixing on the LES sub-grid scale (SGS) (Hoffmann et al., 2019). The dynamical core of our simulations is the System for Atmospheric Modeling (SAM) by Khairoutdinov and Randall (2003), a nonhydrostatic, anelastic LES model.

The LCM models cloud microphysical processes employing individually simulated 143 computational particles, LCM particles or "superdroplets" as in Shima et al. (2009), where each particle represents a group of identical hydrometeors with the same properties, such 145 as liquid radius and aerosol mass. In the L^3 model, the absolute supersaturation fluctuation 146 (δ') of a virtual air volume around each LCM particle that deviates from the LES-resolved 147 absolute supersaturation, $\bar{\delta} = \bar{q_v} - q_s(\bar{T}, p)$, is stored and updated throughout the particle growth history. Here, $\bar{q_v}$ is the LES water vapor mixing ratio, and q_s is the saturation vapor 149 mixing ratio calculated from the LES absolute temperature \overline{T} and hydrostatic pressure p. 150 Thus, the diffusional growth of cloud droplets in L^3 is not only determined by $\bar{\delta}$ but also by 151 δ' , which is primarily determined by the LEM. 152

 δ' is redistributed among the LCM particles located in the same LES grid box by the 153 LEM, an explicit turbulence and mixing model developed by Kerstein (1988). In the LEM, 154 the LCM particles in the same LES grid box are arranged in a virtual one-dimensional array, 155 with each LCM particle representing one LEM grid box. The LEM grid size is determined 156 by dividing the LES vertical grid size by the number of LCM particles in the LES grid box. 157 Segments are randomly chosen from this one-dimensional array and internally rearranged 158 to mimic turbulent compression and folding using the so-called triplet map (Kerstein, 1988; 159 Krueger et al., 1997), which is applied with a frequency determined by the LES SGS tur-160

¹⁶¹ bulence intensity. Interested readers are referred to Hoffmann et al. (2019), Hoffmann and ¹⁶² Feingold (2019), and Lim and Hoffmann (2023) for details on the L^3 model.

2.2 Simulation

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A maritime shallow-cumulus-topped boundary layer is simulated based on the Barbados Oceanographic and Meteorological Experiment (BOMEX) LES intercomparison case (Siebesma et al., 2003). The simulations are initialized with constant heat and moisture fluxes of 8×10^{-3} K m s⁻¹ and 5.2×10^{-5} m s⁻¹, respectively. The total model domain is 5.12 km $\times 5.12$ km $\times 2.56$ km in *x*, *y*, and *z* directions with isotropic 40 m grid spacing. The model time step Δt is 1 s, and the total model integration time is 9 h. The results are analyzed only for the last 6 h of each simulation.

In each grid box, 100 computational particles are initialized as sea salt (NaCl) aerosol 171 particles. Each particle represents the same number of real aerosol particles or droplets. This 172 number of computational particles ensures 40 cm LEM resolution, corresponding to the typ-173 ical transition length scale from inhomogeneous to homogeneous mixing (Lehmann et al., 174 2009), as noted in Lim and Hoffmann (2023). In other words, all relevant scales of inhomo-175 geneous mixing are resolved. Aerosols are initialized with two log-normal distributions, hav-176 ing number concentrations of 118 and 11 cm⁻³ (in total 129 cm⁻³), geometric mean radii 177 of $r_{\rm m}$ = 19 and 56 nm and geometric standard deviations of $\sigma_{\rm r}$ = 3.3 and 1.6, respectively (Derksen et al., 2009). To investigate the effect of different aerosol number concentra-179 tions, we simulate three additional aerosol concentrations by halving (64.5 cm^{-3}) , doubling 180 (258 cm⁻³), and quadrupling (516 cm⁻³) the number concentrations of each mode. For sim-181 plicity, halved aerosol concentration cases are referred to as the 64 cm⁻³ cases. The initial dry aerosol radii are randomly generated by following the log-normal distribution parameters 183 given above, and the corresponding equilibrium wet radii of each aerosol are determined by 184 considering the initial ambient humidity. Sedimentation and collision-coalescence processes 185 are not considered as the simulated clouds are assessed to barely precipitate, especially when 186 the initial aerosol concentration exceeds 64.5 cm^{-3} . 187

In addition to the standard BOMEX configuration (i.e., control boundary layer humidity), two different initial vapor mixing ratio profiles are used: one with a drier and another with a moister boundary layer to investigate the effects of boundary layer humidity. For this purpose, we employ two additional moisture profiles, one with a higher and the other with a lower water vapor mixing ratio q_v (±0.9 g kg⁻¹) within the cloud layer (Fig. 1a), as in Drueke et al. (2020). In this study, the simulation without modifying the initial q_v profile and with the aerosol concentration of 129 cm⁻³ is referred to as the "control case".

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2.3 Cloud Clock for Shallow Cumulus Cloud Life Cycle Classification

Individual clouds are detected as three-dimensional volumes from snapshots of the cloud field. Every 60 s, cloudy grid boxes are identified if their liquid water mixing ratio $q_c \ge 0.01$ g kg⁻¹. Then, all adjacent cloudy grid boxes are considered to belong to the same cloud. This study considers only clouds consisting of ten or more grid boxes for analysis, and approximately 20 - 30 individual clouds matching these conditions are detected in each snapshot (cf. Fig. 1b).

We apply the 'cloud clock' by Witte et al. (2014) to classify the cloud life stage. The 'cloud clock' uses two variables. First, the cloud-volume-averaged total water mixing ratio (q_t) normalized by the corresponding sub-cloud mixed layer value,

$$q_t^* = \frac{q_t}{q_{t,\text{ml}}},\tag{1}$$

with the prescribed mixed layer $q_{t,ml} = 17.0 \text{ g kg}^{-1}$. Second, the cloud-volume-averaged potential virtual temperature perturbation,

$$\Delta \theta_{\rm v} = \theta_{\rm v,c} - \overline{\theta_{\rm v,clr}},\tag{2}$$

Figure 1. a) Initial potential temperature (red solid line) and vapor mixing ratio profiles for three environmental conditions used in the simulations (black solid line: the control case, dash-dotted line: moister case, and dotted line: drier case). b) An overview of clouds from the control case, color-coded according to their classified life cycle stages at t = 529 min. c) Distributions of the 'cloud clock' variables q_t^* and $\Delta \theta_v$ from each single cloud, for the control case. Different colors in b) and c) indicate different life stages (blue: developing, green: mature, and blue: dissipating)

207	is used. Here, $\Delta \theta_v$ is calculated from the difference between the in-cloud virtual potential
208	temperature ($\theta_{v,c}$) and the horizontal mean non-cloudy or clear sky value of θ_v , at the same
209	level ($\overline{\theta_{v,clr}}$). Witte et al. (2014) suggested that in shallow cumulus cloud fields, a single-
210	pulsed cloud with $q_t^* > 0.9$ and $\Delta \theta_v > 0$ can be classified as "developing", $q_t^* < 0.85$ and
211	$\Delta \theta_{\rm v} < 0$ as "dissipating stage", and remaining cases as an intermediate or mature stage.

In this study, shallow cumulus clouds in moister and drier boundary layers are analyzed, potentially changing $q_{t,ml}$. To find appropriate limits to identify life stages, we use K-means clustering by training q_t^* and $\Delta \theta_v$ instead of following the aforementioned criteria by Witte et al. (2014). Interestingly, the K-means clustering classification aligns well with their criteria but is more sensitive to $\Delta \theta_v$ (Fig. 1c).

K-means clustering criteria defines developing clouds by $\Delta \theta_{\rm v} > 0$ with $q_{\rm t}^* > 0.85$, dis-217 sipating clouds by $\Delta \theta_v < -0.25$ with $q_t^* < 0.85$, and mature clouds for other cases (Fig. 1c). 218 For drier and moister cases, q_1^* values are changed while its differences in each stage and 219 $\Delta \theta_{\rm v}$ criteria are consistent. Note that this study does not separate potentially multi-pulsed 220 clouds affecting both q_t^* and $\Delta \theta_v$ (Heus et al., 2009; Witte et al., 2014). For the control case, 221 approximately 4,000 clouds are classified to be in the developing stage, 3,000 in the mature 222 stage, and 1,500 in the dissipating stage from 360 snapshots (example for at t = 529 min is 223 shown in Fig. 1b). 224

3 Evolution of Cloud and Mixing Scenarios

Figure 2 shows the general characteristics of the different simulated cases. The im-226 posed changes in the initial cloud layer q_v (> 500 m) result in a drier or moister sub-cloud 227 layer due to the mixing between two layers (Fig. 2a), while the potential temperature θ barely 228 varies between the simulated cases (Fig. 2b). ε is higher when the boundary layer is moister, 229 especially at the cloud top, due to the higher cloud fraction (f_c) stimulating more turbulence 230 production in this region (Fig. 2c and d). Overall, more clouds are formed in the moister 231 boundary layer and reach higher depths (Fig. 2d and e). Different initial aerosol concentra-232 tions primarily affect N_c (Fig. 2f). In drier boundary layers, the lower q_c for almost the same 233 $f_{\rm c}$ (especially at 1000 m) indicates more dilution by entrainment and mixing (Fig. 2e). 234

Figure 2. Vertical profiles of a) vapor mixing ratio (q_v) , b) potential temperature (θ) , c) dissipation rate (ε) , d) cloud fraction (f_c) , e) cloud water mixing ratio (q_c) , f) cloud droplet number concentration (N_c) . The line colors indicate different initial aerosol concentrations (black: 64 cm⁻³, blue: 129 cm⁻³, green: 258 cm⁻³, and red: 516 cm⁻³). Line style depicts the cases with different boundary layer humidity (solid: control boundary layer, dashed: drier boundary layer, and dotted: moister boundary layer). Results are averaged over the last six hours of the simulation.

Figure 3. Illustration on the IHMD estimation. Properties of LCM particles before mixing (state i) are estimated by backtracking the particles in the same grid box in state f. The IHMD is estimated from the particles in state f by comparing them to their properties in state i.

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3.1 Inhomogeneous Mixing Degree Relative Frequency

Within clouds, different mixing scenarios take place in different locations at the same time. Therefore, it is essential to have a method to quantify the predominant mixing scenario. To classify the mixing scenario in an LES grid box, we use the inhomogeneous mixing degree,

IHMD =
$$\frac{\ln (N_{c,f}/N_{c,i})}{\ln (q_{c,f}/q_{c,i})}$$
, (3)

defined in Lim and Hoffmann (2023), a parameter based on the conceptual cloud microphys-240 ical changes for idealized mixing scenarios (Andrejczuk et al., 2006; Morrison & Grabowski, 241 2008). The IHMD is estimated based on the changes in N_c and q_c before and after mixing, 242 denoted by the subscript i and f, respectively. In extreme homogeneous cases (IHMD = 0), 243 $N_{\rm c}$ remains constant while $q_{\rm c}$ decreases, implying the partial evaporation of droplets. In ex-244 treme inhomogeneous mixing scenarios (IHMD = 1), N_c changes proportionally to q_c , implying the complete evaporation of droplets. When IHMD is over 1, a decrease in N_c over-246 whelms the decrease in $q_{\rm c}$, resulting in an increase in the droplet mean volume radius due to 247 the complete evaporation of small droplets. 248

The IHMD is determined by tracking LCM particle properties over a 30 s timelag 249 (t_{lag}) between the before (i) and after (f) mixing state. The IHMD is estimated in regions 250 where mixing occurs defined by $q_{c,f}/q_{c,i} < 0.9$, while both $q_{c,i}$ and $q_{c,f}$ are required to 251 be larger than 0.01 g kg⁻¹. In addition, the LCM mean supersaturation before mixing $\overline{S_i}$ = 252 $\sum_{n=1}^{N_p} S_{i,n} / N_p < -0.05$ to ensure that subsequent changes are driven by entrained air. Here, 253 $S_{i,n}^{n-1} = (\bar{\delta}_i + \delta'_{i,n})/q_s$, where the subscript *n* denotes the index of individual LCM particle 254 and N_p represents the total number of LCM particles within the same LES grid box at state f. 255 Properties before mixing, such as $q_{c,i}$ and S are estimated by backtracking all particles in the 256

same grid box at the state f. Thus, the particles in state i are not necessarily in the same grid box, and droplets not residing in the grid box at state f are not included in state i property estimation (cf. the neglected droplet in Fig. 3). Note, however, that the particles tend to have similar growth histories for $t_{\text{lag}} = 30$ s (Lim & Hoffmann, 2023).

Both N_c and q_c typically decrease during entrainment and mixing by dilution and 261 evaporation. However, it is possible that entrained aerosol particles activate during mixing 262 (so-called secondary activation), leading to an increase in $N_{\rm c,f}$ and potentially $q_{\rm c,f}$ (Krueger 263 et al., 2008; Chen et al., 2020; Chandrakar et al., 2021). This study incorporates this effect 264 by considering the increased $N_{c,f}$ and $q_{c,f}$ (cf. the yellow droplet in Fig. 3). Incorporat-265 ing secondary activation makes the IHMD smaller (Fig. S1a), primarily by increasing $N_{c,f}$, 266 counterbalancing a possible decrease in droplet number by complete evaporation. Never-267 theless, the overall change in IHMD is negligible (Fig. S1a), as secondary activation is rare 268 (Fig. S1b), potentially due to the aforementioned supersaturation restriction to under -0.05269 before mixing. More detailed explanations on IHMD estimation can be found in Sec. 3 of 270 Lim and Hoffmann (2023). 271

To determine the predominant mixing type of a cloud, IHMDs are classified as one of three major mixing types. Homogeneous mixing is associated where $0 \le IHMD < 0.5$ and inhomogeneous mixing where $0.5 \le IHMD \le 1$ considering all intermediate mixing scenarios. Narrowing mixing is associated with $1 < IHMD \le 5$. In extreme cases, IHMD can exceed 5, but approximately 98 % of the IHMDs are smaller than this threshold and not included in the estimation. Subsequently, the relative frequency of each mixing scenario, the mixing scenario relative frequency,

$$MSRF_{X} = n_{X}/n, \tag{4}$$

is determined, where *n* is the number of all mixing events in the cloud and n_X is the number 279 of all classified homogeneous, inhomogeneous, or narrowing mixing events depending on 280 subscript X. For instance, out of ten mixing events in the entire cloud, six are classified ho-281 mogeneous, three inhomogeneous, and one narrowing. These values correspond to $MSRF_X$ 282 of 0.6, 0.3, and 0.1, respectively. Thus, MSRF_X is beneficial to determine the predominant 283 mixing type in a cloud (e.g., homogeneous in this example), as other mixing scenarios also 284 occur in the same cloud at the same time, while the average of the IHMDs could be biased by 285 potentially large IHMDs during narrowing mixing. 286

Figure 4a shows the MSRF_X of each scenario in each cloud as a violin plot for the control case. Each data point represents the value from an individual cloud in the indicated life stage. The homogeneous mixing scenario is the most dominant, and the narrowing mixing scenario is rare throughout the cloud life cycle. Most importantly, the inhomogeneous mixing scenario becomes more frequent as the cloud ages while the dominance of homogeneous mixing decreases commensurately, confirming observational evidence (e.g., Lehmann et al., 2009; Schmeissner et al., 2015).

Results from all simulated cases consistently indicate a trend towards increased inhomogeneous mixing as the cloud ages regardless of the initial conditions (Fig. 4b, c and d). The increase in MSRF_I from the developing to dissipating stage remains approximately the same for all simulated cases, suggesting a universality in the evolution of inhomogeneous mixing throughout the cumulus cloud life cycle.

The MSRF_H in developing clouds are over 0.5 for all simulated cases and generally decrease by about 0.1 toward the dissipating stages (Fig. 4b), while the MSRF_I increases by 0.1 between the developing and dissipating stages. Notably, for the dry boundary layer with the highest aerosol number concentration ($N_a = 516 \text{ cm}^{-3}$), inhomogeneous mixing is more frequent than homogeneous mixing in the dissipating stage (Fig. 4b and c). Meanwhile, MSRF_N slightly increases as the cloud ages, but this change is relatively small, with less than 0.02 (see Fig.4d).

Figure 4. a) Distribution of $MSRF_X$ (homogeneous, inhomogeneous, and narrowing) from individual clouds over different cloud life cycles (developing, mature, and dissipating) for the control case. Mixing scenario medians are connected with red dotted lines to highlight the life cycle changes. The median $MSRF_X$ for b) homogeneous, c) inhomogeneous, and d) narrowing mixing in individual clouds as a function of the cloud life stage (developing, mature, and dissipating) are shown for all simulated cases. The solid line depicts the control case, the dashed line depicts the drier case, and the dotted line indicates the moister case. The colors of the line indicate different initial aerosol number concentrations (black: 64 cm⁻³, blue: 129 cm⁻³, green: 258 cm⁻³ and red: 516 cm⁻³).

Figure 5. Distributions of cloud volume-averaged median a) vertical velocity (W), b) typical cloud size, c) liquid water path (LWP), d) cloud droplet number concentration (N_c) , e) adiabatic fraction (AF), f) mean supersaturation (\overline{S}) , g) standard deviation of supersaturation (σ_S) , h) droplet mean radius (r_m) , and i) droplet radius relative dispersion (d_r) for different cloud life stages. The solid line and circle marker indicate the results obtained from cases with control boundary layer humidity. The dashed line and triangle marker indicate the result obtained from a case with a drier boundary layer. The dotted line and square marker denote the results of a case with a moister boundary layer. The colors distinguish cases with different initial aerosol number concentrations. In each panel, the correlation coefficients (ρ_X) with respect to MSRF_X (homogeneous, inhomogeneous, and narrowing) are indicated.

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3.2 The Evolution of Cloud-Volume-Averaged Properties over the Life Cycle

To investigate how inhomogeneous mixing evolves, we illustrate the properties related to entrainment and mixing averaged across the entire volume of individual clouds, as represented by:

$$\overline{\phi}_{\text{cloud}} = \frac{1}{V_{\text{cloud}}} \int_{V_{\text{cloud}}} \phi \, dV, \tag{5}$$

where V_{cloud} is the total cloud volume and ϕ is cloud property of interest, such as ε or N_{c} .

Figure 5 shows the median of $\overline{\phi}_{cloud}$ from individual clouds in different life stages. For clarity, we omit the overbar and subscript in the following.

Over the cloud life cycle, clouds become less buoyant, and their mean vertical velocity (*W*) decreases similarly in all cases (Fig. 5a). The typical cloud size, determined from the

square root of vertically projected cloudy area (e.g., Neggers et al., 2003), decreases over the 315 life cycle (Fig. 5b). The liquid water path (LWP) also decreases toward the mature stage and 316 remains almost constant afterward (Fig. 5c). In Fig. S2, we show that when considering only 317 clouds larger than 300 m in cloud size, the cloud size increases from the developing to the 318 mature stage and decreases in the dissipating stage. Concurrently, the LWP increases toward 319 the dissipating stage due to the expected large number of deeper clouds toward the end of the 320 cloud life cycle. Other variables barely change if analyzed clouds are constrained to larger 321 cloud sizes. 322

Figure 5e shows the adiabatic fraction (AF), a ratio between the LWC and the adiabatic LWC at the corresponding level, indicating the degree of cloud dilution (Eytan et al., 2021). As clouds age, clouds are diluted by mixing with environmental air, which decreases AF. Generally, AF decreases similarly over the cloud life cycle in all cases. However, AFs are smaller in more polluted cases, where entrainment is typically stronger (Wang et al., 2003). We should note that values shown in Fig. 5 are averaged over the entire cloud volume, affected by extremely low AFs at the cloud edge (McFarlane & Grabowski, 2007).

The cloud dilution causes a decrease in the mean supersaturation \overline{S} experienced by 330 the LCM particles and an increase in its standard deviation ($\sigma_{\rm S}$) during the cloud life cycle 331 (Fig. 5f and g). More dilution and a lower environmental \overline{S} in the driver boundary layer cases 332 leads to a more substantial decrease in \overline{S} . Additionally, the lower \overline{S} in the cases with higher 333 aerosol concentration supports the aforementioned stronger entrainment. Thus, more dilution 334 in the drier and polluted boundary layer cases leads to a higher $\sigma_{\rm S}$ (Fig. 5g). This indicates a 335 more inhomogeneous mixture of S in the cloud, a prerequisite for increasing inhomogeneous 336 mixing on the cloud edges. 337

Dilution also decreases N_c over the cloud life cycle, while the initial aerosol number 338 concentration strongly determines N_c (Fig. 5d). The droplet mean radius increases over the 339 cloud life cycle (Fig. 5h), while droplet size variance increases more due to continuous en-340 trainment and mixing, allowing the droplet size relative dispersion d_r , the ratio between 341 the mean and standard deviation of the droplet radius, to increase over the cloud life cycle 342 (Fig. 5i). The increase in $d_{\rm r}$ supports the slight increase in narrowing mixing over the cloud 343 life cycle (Fig. 4d). Note that the increase in narrowing mixing is balanced by the decreasing 344 W (Fig. 5a), which prevents further narrowing by lifting (Lim & Hoffmann, 2023). 345

Overall, cloud-averaged properties and MSRF_X exhibit consistent changes over the cloud life cycle, regardless of the aerosol concentration and boundary layer humidity (Figs. 4, and 5). We now briefly discuss the correlation of median values ρ_X , shown in Fig. 5. ρ_X is estimated with 36 sets of median values (12 simulated cases and three mixing stages) for MSRF_X (Fig. 4b, c, and d) and the properties shown in Fig. 5. A complete correlation matrix can be found in Fig. S3.

For MSRF_I, \overline{S} shows the strongest negative correlation (-0.9), alongside the cloud clock variables (q_t^* and $\Delta \theta_v$, as indicated in Fig. S3). On the other hand, σ_S shows the strongest positive correlation with MSRF_I (0.9), while other properties such as *W*, cloud size, and AF show a negative correlation around -0.8.

For MSRF_H, correlations are generally lower. The same variables show an inverse relationship compared to MSRF_I as expected. Maximum correlation is 0.87 for $\Delta \theta_{\rm v}$, while others range from 0.8 to 0.87. For MSRF_H and MSRF_I, other properties (LWP, $d_{\rm r}$, $r_{\rm m}$ and $N_{\rm c}$) show weak correlations under ±0.6.

The correlations are even weaker for MSRF_N. Here, AF shows the strongest correlation of -0.66, followed by W, q_t^* , cloud size, $\Delta \theta_v$ and \overline{S} with correlations ranging from -0.5 to -0.6. Interestingly, the DSD shape-related parameters, such as d_r and r_m show higher positive (0.38) and negative (-0.42) correlation than σ_S (0.33), suggesting a distinct underlying process for narrowing mixing, in contrast to other two scenarios.

Figure 6. Distributions of mixing region volume-averaged a) mean supersaturation of droplets in supersaturated air, b) mean supersaturation of droplets in subsaturated air, c) droplet radius relative dispersion during mixing. The blue violin plot on the left side of each panel depicts values before mixing, and the red violin plot on the right side depicts values after mixing. All data are obtained from the control case.

This correlation analysis shows that homogeneous and inhomogeneous mixing evolution is highly affected by the \overline{S} and σ_S changes over the cloud life cycle apart from the expected strong correlation with cloud clock variables. It is noteworthy that correlation among the values mentioned above (q_t^* , $\Delta \theta_v$, \overline{S} , σ_S , W, cloud size, and AF) are also strong (Fig. S3), indicating the complex interactions of these variables over the cloud life cycle.

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3.3 Mixing Region Properties Change During Cloud Life Cycle

The properties averaged over the entire cloud volume provide a valuable overview of how cloud properties change over the cloud life cycle. However, entrainment and mixing occur at the cloud edge, and the properties in this region may differ from those averaged over the entire cloud volume. Thus, it is also important to discuss the properties before and after mixing in the regions where entrainment and mixing occur (Fig. 6).

To distinguish the entrained and cloudy air properties during a mixing event, we assume that subsaturated particles are affected by the entrained air, whereas air unaffected by entrainment remains supersaturated. By using the supersaturation around each LCM particle, we define the average *S* of supersaturated droplets as $\overline{S_{\text{pos.}}}$ and the average *S* of subsaturated droplets as $\overline{S_{\text{neg.}}}$. As expected, $\overline{S_{\text{pos.}}}$ decreases and $\overline{S_{\text{neg.}}}$ increase during mixing at any cloud life stage (Fig. 6a and b).

Over the cloud life cycle, $\overline{S_{\text{pos.}}}$ decreases slightly, and $\overline{S_{\text{neg.}}}$ decreases substantially 382 (Fig. 6b). Moreover, $\overline{S_{\text{neg.}}}$ after mixing decreases as the cloud ages. This indicates that clouds 383 in the mature and dissipating stages have lower S, primarily due to the remnants of the en-384 trained air without full restoration of S inside the cloud (Fig. 5e). Additionally, the lower S 385 inside mature and dissipating clouds may be caused by the lower \overline{S} of the entrained air in 386 these stages. We will discuss this further in Sec. 4.2. Regardless of its origin, stronger sub-387 saturations decrease τ_{micro} , which creates more favorable conditions for inhomogeneous mix-388 ing scenarios as the cloud ages. 389

In the mixing region, d_r generally increases over the cloud life cycle (Fig. 6c). Lim and Hoffmann (2023) showed that in the homogeneous mixing scenarios, d_r increases after mixing, whereas d_r barely changes or even decreases in the inhomogeneous and narrowing mixing scenarios, respectively. While homogeneous mixing is the predominant scenario, leading to a general increase in d_r after mixing, this increase becomes weaker as the cloud matures due to the increased inhomogeneous mixing frequency (Fig. 6c).

Figure 7. Distributions of a) cloud base height (z_b) , b) cloud top height (z_t) and c) number of mixing events in normalized cloud depth (z^*) . The colors of the bar indicate different cloud life stages (blue: developing, green: mature, and red: dissipating). All data are obtained from the control case.

396 4 Vertical Profiles

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Shallow cumulus clouds exhibit a range of different cloud base and top heights. A developing cloud does not always extend across the entire cloud layer. Similarly, dissipating clouds dissociate from their base (Zhao & Austin, 2005). Figure 7a and b depict that the cloud base (z_b) and top height (z_t) increase over the cloud life cycle. This indicates that the actual vertical location where the cloud interacts with its surrounding environment also changes over the cloud life cycle. Thus, the vertical distribution of MSRF_X and other properties in normalized cloud depth,

$$z^* = \frac{z - z_b}{z_t - z_b},\tag{6}$$

is used instead of a vertical profile in the following section. By using z_t and z_b of each individual cloud, z^* is binned into intervals of 0.1 ranging from $z^* = 0$ to 1.

4.1 Mixing Scenario Relative Frequency Profile

Figure 8 shows the vertical distribution of $MSRF_X$ for the control case (Fig. 8e) and the deviations from it in all other simulated cases (denoted as $\Delta MSRF_X$), which are obtained by subtracting the control case $MSRF_X$ profile from the $MSRF_X$ profile of the considered cases. The $MSRF_X$ profiles are determined by estimating the relative frequency of each mixing scenario within each z^* bin. In the control case profile, homogeneous mixing is the predominant scenario in the developing stage (Fig. 8e), in agreement with the volume-averaged results (Fig. 4), especially at $z^* > 0.3$ where mixing is most frequent (Fig. 7c).

In the mature and dissipating stage, homogeneous mixing gradually decreases, especially near the cloud top, while it becomes more frequent near the cloud base. This decrease in the MSRF_H at the cloud top is associated with an increase in MSRF_I. In contrast, the increase in MSRF_H at the cloud base is related to a decrease in the MSRF_N (Fig. 8e).

To relate $MSRF_X$ profile changes to mixing relevant properties, we show profiles of 418 these properties averaged among individual clouds for different life stages, in-cloud, and the 419 environmental shell around the cloud (Fig. 9). Environmental shell values are obtained from 420 the first horizontally adjacent grid boxes outside the cloud, constituting a potential source 421 of entrained air. Each profile is cloud area-weight averaged, considering the vertical distri-422 bution of cloud area across normalized height. In addition, mixing region profiles averaged 423 over the regions where IHMDs are estimated are also shown. For the mixing region profiles, 424 values before and after the mixing state are averaged, while other profiles show instantaneous 425 values. We note that values at $z^* > 0.9$ in Fig. 9 tend to be affected by the cloud top over-426

Figure 8. The central panel (e) shows the vertical distribution of mixing scenario relative frequency (MSRF_X) for $N_a = 129 \text{ cm}^{-3}$ under control boundary layer moisture conditions. Surrounding panels illustrate Δ MSRF_X, derived by subtracting the control case MSRF_X profile from the MSRF_X profiles of each specific case. Columns are arranged by aerosol concentration: $N_a = 64 \text{ cm}^{-3}$ (first), $N_a = 129 \text{ cm}^{-3}$ (second), and $N_a = 258 \text{ cm}^{-3}$ (third). Rows indicate the initial boundary layer conditions: moist (top), control (middle), and dry (bottom). Varying line styles represent distinct mixing scenarios—solid for homogeneous (HM), dashed for inhomogeneous (IM), and dotted for narrowing mixing (NM) scenarios. Different cloud life stages are indicated by color: blue (developing), green (mature), and red (dissipating).

shooting the inversion height (~ 1500 m in the simulated BOMEX case), and should be inter preted with care.

The ε profiles (Fig. 9a) show higher values inside the cloud than the environment, and 429 its maximum is near the cloud top in every life stage, as observed by Gerber et al. (2008). 430 High in-cloud ε at the cloud top indicates a shorter τ_{mix} in this region, which explains the 431 high MSRF_H near the cloud top (Fig. 8e). In the mixing region, ε also increases with height, 432 but the values are lower than in the in-cloud region. This is because the mixture of less tur-433 bulent environmental air and potentially more turbulent cloudy air results in an intermediate 434 ε in the mixing region. As the cloud ages, ε decreases at the cloud top, and the MSRF_H in 435 this region decreases accordingly (Fig. 8e). Below the cloud top, on the other hand, ε and 436 accordingly MSRF_H slightly increase over the cloud life cycle (Fig. 9a). 437

⁴³⁸ While in-cloud N_c decreases with height and over the cloud life cycle as expected, N_c ⁴³⁹ in the mixing region at $z^* > 0.5$ increases over time, (Fig. 9b). This indicates that the mixing ⁴⁴⁰ and in-cloud regions of the cloud become less distinct as the cloud ages, i.e., mixing poten-⁴⁴¹ tially affects a larger fraction of the cloud. Similarly, r_m and d_r in the mixing region approach ⁴⁴² their in-cloud values as the cloud ages, supporting this idea (Fig. 9c and d).

It is expected that $r_{\rm m}$ increases with height in all life stages (Fig. 9c). Over the cloud life cycle, $r_{\rm m}$ increases at the cloud base and decreases near the cloud top, while the overall changes are insubstantial. This is because the cloud base elevates over the cloud life cycle (cf. Fig. 7a). The correspondingly larger droplets enable higher MSRF_H and less MSRF_N at the cloud base as the cloud ages (cf. Fig. 8e).

⁴⁴⁸ While *S* slightly decreases with height and over the cloud life cycle (Fig. 9e), σ_S sub-⁴⁴⁹ stantially increases (Fig. 9f). When σ_S is high enough, even high ε at the cloud top might ⁴⁵⁰ not be able to homogenize the supersaturation field. However, when σ_S is rather low (e.g., ⁴⁵¹ $\sigma_S < 0.01$), and \overline{S} and ε are sufficiently large (e.g., at the cloud top in the developing stage) ⁴⁵² homogeneous mixing dominates (Fig. 8e).

Except for \overline{S} , environmental properties change negligible with cloud life stages compared to in-cloud properties (see dashed lines in Fig. 9e). Changes in environmental \overline{S} are mainly due to the aforementioned elevated position of the cloud (cf. Fig. 4), where \overline{S} is lower (Fig. S4e). This implies that drier entrained air also stimulates the lower σ_S as the cloud ages. Thus, considering changes in the vertical location of clouds over time is an important factor in evaluating the life cycle evolution of shallow cumulus cloud mixing characteristics.

The complex interplay between vertical profiles of variables shown in Fig. 9 affect-459 ing MSRF_X underscores the need for a variable that contains more comprehensive meaning. 460 Thus, we estimate the transition length scale $l^* = \varepsilon^{\frac{1}{2}} \tau_{evp}^{\frac{3}{2}}$ (Baker et al., 1980b; Lehmann et 461 al., 2009), which indicates the length scale where Damköhler number becomes 1, separating 462 homogeneous and inhomogeneous mixing. Accordingly, smaller l^* indicate more potential 463 for inhomogeneous mixing. To estimate l^* , we first estimate τ_{evp} (Fig. 9g) using profiles of 464 $r_{\rm m}$ (Fig. 9c) and $\overline{S_{\rm neg.}}$ (not shown) to predict the entrained S for the in-cloud and mixing re-465 gions. 466

⁴⁶⁷ τ_{evp} generally decreases over the cloud life cycle due to the strong decrease in \overline{S} which ⁴⁶⁸ overruns the increase in r_m . As ε increases less than τ_{evp} decreases (Fig. 9a and g), l^* de-⁴⁶⁹ creases over the cloud life cycle, indicating more potential for inhomogeneous mixing (Fig. 9h). ⁴⁷⁰ This implies that the life cycle mixing scenario evolution is driven by *S* affecting τ_{evp} .

471 We should note that the small l^* at the cloud top does not necessarily indicate more in-472 homogeneous mixing in this region. The vertical distribution of MSRF_X does not consider 473 the absolute number of mixing events, and vertical profiles of variables do not capture the 474 variance in each mixing event. Nonetheless, the general pattern in MSRF_X changes, espe-475 cially near the cloud top where the mixing events are dominant (Fig. 7c), aligns well with life 476 cycle changes in MSRF_X in the total cloud volume.

Figure 9. Vertical profiles of a) dissipation rate (ε) b) cloud droplet number concentration (N_c), c) mean droplet radius (r_m), d) droplet radius relative dispersion (d_r), e) mean $S(\overline{S})$, and f) standard deviation of $S(\sigma_S)$. The line style depicts the region where each variable is averaged (solid: in-cloud, dashed: environment, and dotted: mixing region). Vertical profiles of in-cloud g) evaporation time scale (τ_{evp}) and h) transition length scale (l^*) are also shown. The line colors indicate different cloud life stages (blue: developing, green: mature, and red: dissipating)

Figure 10. Vertical distribution of the difference between the moist-pristine case and the control case (a, b, and c) and between dry-polluted case and the control case (d, e, and f) for the dissipation rate (1st column), mean supersaturation (2nd column) and supersaturation standard deviation (3rd column). The line colors indicate different cloud life stages (blue: developing, green: mature, and red: dissipating), and the line style depicts the region where each variable is averaged (solid: in-cloud, dashed: environment, and dotted: mixing region). The gray solid line denotes no difference, $\Delta = 0$.

4.2 Microphysical versus Environmental Factors Determining Mixing Scenarios

We have shown that changes in σ_S , \overline{S} , ε , and r_m play crucial roles in determining how mixing scenarios change over the cloud life cycle. The following section will discuss these scenarios under varied environmental conditions.

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⁴⁸¹ Overall, Δ MSRF_X profiles are uniform, suggesting a consistent change in vertical ⁴⁸² MSRF_X profiles while values differ (Fig. 8), confirming results shown in Fig. 4. The ho-⁴⁸³ mogeneous mixing scenario occurs more frequently in moister boundary layers (Fig. 8b) ⁴⁸⁴ and pristine environments (Fig. 8d) than in the control case. On the other hand, the inhomo-⁴⁸⁵ geneous mixing scenario is more frequent in drier boundary layers (Fig. 8h) and for higher ⁴⁸⁶ aerosol concentrations (Fig. 8f).

To compare the changes in the properties affecting entrainment and mixing, we pick a more homogeneous case ($N_a = 64 \text{ cm}^{-3}$ and moister boundary layer, referred to as "moist pristine" in Fig. 10a, b and c), and a more inhomogeneous case ($N_a = 258 \text{ cm}^{-3}$ and drier boundary layer, referred to as "dry polluted" in Fig. 10d, e and f).

Results from the *more homogeneous case* show larger \overline{S} and smaller σ_S (Fig. 10b and c), although ε is lower (Fig. 10a). Likewise, a moister boundary layer with the same aerosol concentration shows more homogeneous mixing (Fig. 8b) due to a higher \overline{S} and lower σ_S (Fig. S5e and f), while ε is lower (Fig. S5a).

The more inhomogeneous case shows the opposite trend to the more homogeneous case showing smaller \overline{S} and larger σ_S , regardless of ε (Fig. 10d, e, and f). This is consistent in a drier boundary layer with the same aerosol concentration, while other variables are largely unchanged (Fig. S6e and f), implying that the changes in \overline{S} and σ_S are a stronger indicator in determining the predominant mixing type than ε .

Interestingly, homogeneous and inhomogeneous Δ MSRF_X profiles in the case with a drier boundary layer (Fig. 8h) closely resemble profiles from a doubling of the aerosol concentrations (Fig. 8f). Similar pairs are shown between the moister boundary layer and halved aerosol concentration cases (Fig. 8b and d) and counteracting cases (Fig. 8c and g). This suggests that the microphysical and environmental properties can have equivalent impacts on mixing scenarios.

However, inhomogeneous mixing is always more frequent in a drier boundary layer (Fig. 8g, h and i). This is because in the drier boundary layers, \overline{S} is lower, and σ_S are higher (Fig. S6), whereas, in the more polluted boundary layer, changes in \overline{S} and σ_S are negligible compared to r_m and N_c change (Fig. S7). Therefore, while the complex interplay between \overline{S} , σ_S , r_m and N_c all affects the mixing scenario, \overline{S} and σ_S play the most important role.

The narrowing mixing scenario is more related to the aerosol number concentration. A 511 higher aerosol concentration (258 cm⁻³), which implies a higher N_c and a smaller r_m , con-512 sistently results in a slightly higher MSRF_Ns than in the control case. This remains true even in moist boundary layers where homogeneous mixing is more prevalent (Fig. 8c, f, and i). 514 On the other hand, for lower aerosol concentrations (64 cm^{-3}), narrowing mixing is always 515 less frequent regardless of the boundary layer humidity due to the correspondingly larger 516 droplets (Fig. 8a, d, and g). Thus, the narrowing mixing scenario is less dependent on the en-517 vironmental properties than homogeneous and inhomogeneous mixing scenarios and is pri-518 marily related to the prevalence of small droplets in DSD, as outlined in Lim and Hoffmann 519 (2023).520

521 **5 Discussions and Conclusions**

⁵²² Unraveling the intricate yet crucial process of entrainment and mixing is essential for ⁵²³ understanding clouds and their influence on the climate due to their significant influence on ⁵²⁴ the radiation budget and precipitation formation. However, key questions remain regarding the prevalence and variability of distinct mixing scenarios in space and time. Therefore, it is important to understand the relationship between different mixing scenarios and the environmental and microphysical factors determining them. In this study, we use a large eddy simulation (LES) model coupled with a Lagrangian cloud model (LCM) and a linear eddy model (LEM), the L^3 model, to simulate maritime shallow cumulus clouds, classifying them into different life stages and analyzing the evolution of mixing scenarios under varying environmental conditions.

- ⁵³² The key insights of this paper are:
- While different mixing scenarios occur concurrently, we confirm the evolution from a homogeneous mixing-dominant phase to a more inhomogeneous mixing phase over the cloud life cycle, irrespective of initial boundary layer humidity and aerosol concentration.
- In most cases, homogeneous mixing is dominant, but inhomogeneous mixing can prevail in the dissipating stages of shallow cumuli in drier and more polluted boundary layers.
- The mean (\overline{S}) and standard deviation (σ_S) of in-cloud supersaturation S encapsulate the evolution from homogeneous to inhomogeneous mixing and the vertical distribution of mixing scenarios. The values of the mean and standard deviation of S are changed either due to the accumulated effects of entrainment and mixing events or by the clouds shifting upward in the dissipating stage.

In summary, this study expands the understanding from previously observed increas-545 ingly inhomogeneous mixing in diluted clouds (e.g., Lehmann et al., 2009; Schmeissner et 546 al., 2015) by suggesting S as a useful parameter to estimate mixing scenarios without more 547 complex measures, based on the high correlation between S and σ_S . Less diluted developing 548 clouds with higher dissipation rates indicate faster homogenization and lower σ_S . Higher S 549 in these clouds imply slower droplet evaporation, making homogeneous mixing more favor-550 able. Over the cloud life cycle, dilution causes entrained air to accumulate inside the cloud 551 without completely saturating. Thus, in the dissipating stage, clouds have higher $\sigma_{\rm S}$ and lower S, making the homogenization take longer, enabling more inhomogeneous mixing. 553

The narrowing mixing scenario (Lim & Hoffmann, 2023) is more dependent on the abundance of small droplets than \overline{S} and σ_S . This confirms that the narrowing mixing scenario is not a part of homogeneous or inhomogeneous mixing but only depends on the availability of small droplets, providing ample opportunities for complete evaporation (Lim & Hoffmann, 2023).

In conclusion, our findings highlight the need for in-depth consideration of supersatu-559 ration fluctuations and mixing scenarios, as cumulus clouds cannot be characterized by one 560 single mixing scenario. As inhomogeneous mixing becomes more dominant in drier and 561 more polluted environments, its impact can alter in a changing climate. Further studies could 562 determine these relationships across different cloud types, such as in stratocumulus, where 563 recent observation showed that homogeneous mixing is more frequent in the moister environ-564 ment (Yeom et al., 2023), aligning with the results shown in this paper. Moreover, investigat-565 ing how inhomogeneous mixing changes the cloud life cycle will deepen our understanding 566 of the effects of different mixing scenarios on cloud dynamics and their climate implications.

568 **Open Research**

Simulation results for this research are available in Lim (2023). The System for Atmo spheric Modeling (SAM) code is available under the link http://rossby.msrc.sunysb.edu/SAM.html,
 with permission from its developer, Dr. M. Khairoutdinov (Khairoutdinov & Randall, 2003).

⁵⁷² Figures are made with Matplotlib version 3.3.4, available under the link https://matplotlib.org/.

- 573 K-means clustering and multiple-linear regression analysis are done with Scikit-learn: Ma-
- ⁵⁷⁴ chine Learning in Python (Pedregosa et al., 2011).

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Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.

Figure 7.

Figure 8.

Figure 9.

Control case b) d) a) C) 1.0 -I Region I In-cloud 10 0.8 Environment Mixing region 0.6 * N Life stage Developing 0.4 Mature I Dissipating 0.2 E in the second se 11 Ĩ E 0.0 50 75 20 40 10 0.0 0.2 0.4 25 5 0 0 ε (cm² s⁻³) *r*_m (μm) $N_{\rm c}~({\rm cm}^{-3})$ d_r h) e) **f)** g) A CONTRACT OF A 1.0 0.8 0.6 11 11 11 * N Ш 0.4 1 0.2 0.0 -0.20.0 0.00 0.01 10⁰ 10^{1} 10⁰ 10^{1} -0.4 au_{evp} (s) *I** (m) σ_S

Figure 10.

