Impact of aerosol processing in the transition of a stratocumulus cloud system to open cells: A comparison of Lagrangian and bin microphysics schemes in LES

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Abstract

Stratocumulus clouds, a key component of global climate, are sensitive to aerosol properties. Aerosol-cloud-precipitation interactions in these clouds influence their closed-to-open cell dynamical transition and hence cloud cover and radiative forcing. This study uses large-eddy simulations with Lagrangian super-particle and bin microphysics schemes to investigate impacts of aerosol scavenging and physical processing by clouds on drizzle initiation and the cellular transition process. The simulation using Lagrangian microphysics with explicit representation of cloud-borne aerosol and scavenging shows significant aerosol processing that impacts precipitation generation and consequently the closed-to-open cell transition. Sensitivity simulations using the bin scheme and their comparison with the Lagrangian microphysics simulation suggest that reduced aerosol concentration due to scavenging is a primary microphysical catalyst for enhanced precipitation using the Lagrangian scheme. However, changes in the aerosol distribution shape through processing also contribute appreciably to the differences in precipitation rate. Thus, both aerosol scavenging and processing drive earlier rain formation and the transition to open cells in the simulation with Lagrangian microphysics. This study also highlights a shortcoming of Eulerian bin microphysics producing smaller mean drop radius and cloud water mixing ratios owing to numerical diffusion. Initially larger mean radius and cloud mixing ratios using the Lagrangian scheme induce faster rain development compared to the bin scheme. A positive feedback in turn accelerates aerosol removal and further rain production using the Lagrangian scheme and, consequently, reduced cloud droplet number, increased mean size, and increased droplet spectral width.



Impact of aerosol processing in the transition of a stratocumulus cloud system to open cells: A comparison of Lagrangian and bin microphysics schemes in LES

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Key Points:

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8	•	Substantial aerosol scavenging and physical processing by clouds, affecting driz-
9		zle formation, is simulated using Lagrangian microphysics.
10	•	Reduced aerosol concentration and size distribution changes impact aerosol removal
11		via precipitation and time to open cell formation.
12	•	A positive cloud-aerosol-rain feedback accelerates rain formation and aerosol re-
13		moval using the Lagrangian compared to bin scheme.

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Abstract 14

Stratocumulus clouds, a key component of global climate, are sensitive to aerosol prop-15 erties. Aerosol-cloud-precipitation interactions in these clouds influence their closed-to-16 open cell dynamical transition and hence cloud cover and radiative forcing. This study 17 uses large-eddy simulations with Lagrangian super-particle and bin microphysics schemes 18 to investigate impacts of aerosol scavenging and physical processing by clouds on driz-19 zle initiation and the cellular transition process. The simulation using Lagrangian mi-20 crophysics with explicit representation of cloud-borne aerosol and scavenging shows sig-21 nificant aerosol processing that impacts precipitation generation and consequently the 22 closed-to-open cell transition. Sensitivity simulations using the bin scheme and their com-23 parison with the Lagrangian microphysics simulation suggest that reduced aerosol con-24 centration due to scavenging is a primary microphysical catalyst for enhanced precip-25 itation using the Lagrangian scheme. However, changes in the aerosol distribution shape 26 through processing also contribute appreciably to the differences in precipitation rate. 27 Thus, both aerosol scavenging and processing drive earlier rain formation and the tran-28 sition to open cells in the simulation with Lagrangian microphysics. This study also high-29 lights a shortcoming of Eulerian bin microphysics producing smaller mean drop radius 30 and cloud water mixing ratios owing to numerical diffusion. Initially larger mean radius 31 and cloud mixing ratios using the Lagrangian scheme induce faster rain development com-32 pared to the bin scheme. A positive feedback in turn accelerates aerosol removal and fur-33 ther rain production using the Lagrangian scheme and, consequently, reduced cloud droplet 34 number, increased mean size, and increased droplet spectral width. 35

Plain Language Summary 36

Stratocumulus clouds reflect substantial solar radiation due to their extensive cov-37 erage and are therefore a critical component of regional and global climate. Aerosol par-38 ticles and their two-way interactions with these clouds impact drizzle formation and their 39 transition to a less reflective open-cellular cloud structure. In this study, we show that 40 both the reduction of aerosol concentration and changes in aerosol particle sizes due to 41 their interactions with cloud and drizzle drops impact precipitation formation and the 42 time required to transition to an open-cell structure. Such aerosol-cloud interactions are 43 highly simplified in commonly used cloud models due to the difficulty of tracking the so-44 lute mass of aerosols in individual cloud drops. A newer modeling approach called the 45 Lagrangian "super-doplet" method is a state-of-the-art tool that can better represent 46 aerosol-cloud interactions by explicitly tracking aerosol properties in cloud and drizzle 47 drops and reducing other numerical errors. We show that limitations of traditional cloud 48 models, particularly in how they represent aerosol scavenging and processing, affect the 49 predicted micro-scale cloud properties. This leads to delayed formation of rain and the 50 transition to an open cellular cloud structure compared to the simulation using the super-51 particle modeling approach. 52

1 Introduction 53

Stratocumulus clouds significantly affect the earth's energy balance (Slingo, 1990). 54 The mesoscale structure of boundary layer clouds, affecting cloud cover, is critical to their 55 radiative forcing and precipitation formation. Aerosol-cloud-precipitation interactions 56 in turn influence the mesoscale structure, dynamics, and lifetime of clouds. Aerosol im-57 pacts on marine low clouds are a key for global climate modeling (e.g., Zhang et al., 2016) 58 and underlie the potential for marine cloud brightening to offset global warming (Latham 59 et al., 2008; Diamond et al., 2022; Feingold et al., 2022). This study aims to elucidate 60 aerosol-cloud-precipitation interactions in stratocumulus clouds using state-of-the-art large-61 eddy simulations (LES) with two different approaches for size-resolved representation 62

of microphysics. 63

Aerosols induce changes to cloud microphysics (Twomey, 1974; Chandrakar et al., 64 2016), thereby influencing macroscopic cloud properties and processes (e.g., cloud thick-65 ness, precipitation formation, cloud cover, liquid water path, patterns of convection, ra-66 diative forcing, etc.) through various process interactions (Albrecht, 1989; Pincus & Baker, 67 1994; Ackerman et al., 2004; Wood, 2012; Glassmeier et al., 2021). Aerosol scavenging 68 and processing in stratocumulus cloud systems can substantially influence their dynam-69 ical states. For example, precipitation suppression from aerosol loading can alter pat-70 terns of latent heating and evaporative cooling, in turn affecting the dynamics, turbu-71 lent kinetic energy (TKE), and entrainment rate (Wood, 2012). Indeed, modeling stud-72 ies have suggested that the transformation of a cloud system from closed to open cells 73 can occur merely from a reduced aerosol concentration (Wang & Feingold, 2009). Aerosols 74 also influence cloud drop size distributions (DSDs), which could impact the entrainment 75 rate directly through the settling-entrainment and evaporation-entrainment feedbacks, 76 causing the liquid water path and cloud albedo to change (Ackerman et al., 2004; Brether-77 ton et al., 2007; Hill et al., 2008). In a clean marine environment, the response of clouds 78 to changes in aerosol concentration is expected to be more acute than in polluted con-79 ditions (Reutter et al., 2009; Chandrakar et al., 2017). A key question is, how do aerosol-80 cloud interactions with various associated feedbacks impact the stratocumulus transi-81 tion to a cleaner state? 82

Clouds also affect aerosol properties and remove aerosol particles from the bound-83 ary layer by scavenging. Cloud condensation nuclei (CCN) activate cloud droplets, which 84 can grow to drizzle sizes mainly through collision-coalescence and are removed through 85 precipitation (a wet scavenging process). These in-cloud interactions may drive changes 86 in aerosol composition and size due to chemical (gas diffusion and aqueous chemistry) 87 as well as physical (diffusion/impaction of interstitial aerosol particles to droplets and 88 droplet coagulation) processing (Hoppel et al., 1990; Bower et al., 1999; Pierce et al., 2015). 89 Interstitial aerosols also grow through vapor deposition and coagulation (Seinfeld & Pan-90 dis, 1998). After evaporation of cloud and drizzle drops, the processed aerosols are again 91 available as a CCN source but with altered properties (Hoppel et al., 1990; Pierce et al., 92 2015; Chandrakar, Morrison, & Witte, 2022; Hoffmann & Feingold, 2023). This mod-93 ification of properties could make them more efficient CCN due to increased solute mass 94 through the physical and chemical processing (Hudson et al., 2015), thereby affecting 95 drizzle formation in stratocumulus clouds. Multiple cycles of aerosol processing are sug-96 gested to produce a bimodal aerosol size distribution shape as air masses are advected 97 from the coast to the remote ocean (Hoppel et al., 1990). The activation scavenging of aerosols to cloud droplets depends on the degree of competition for water vapor (Goren 99 & Rosenfeld, 2015; Chandrakar et al., 2017). Thus, as a cloud system gets cleaner through 100 precipitation formation and wet scavenging, the scavenging rate could accelerate in a pos-101 itive feedback loop, rapidly increasing the precipitation flux. Past studies (e.g., Rosen-102 feld et al., 2006; Wang & Feingold, 2009; Wood et al., 2011; Chandrakar, Morrison, & 103 Witte, 2022) have shown that drizzle formation can impact mesoscale cloud properties 104 and drive a transition in cloud cellular structure. Most earlier studies (e.g., Goren et al., 105 2019; Erfani et al., 2022) on this topic used bulk microphysics schemes, where the de-106 tails of aerosol processing are highly parameterized and therefore uncertain. A micro-107 physics scheme that can track aerosol properties inside and outside clouds/drizzle drops 108 is needed to capture aerosol processing in clouds with greater fidelity. 109

Representation of aerosol-cloud-precipitation interactions, even in detailed process
models, is a significant challenge. Aerosol scavenging and processing add substantial complexity to this problem. Eulerian microphysics schemes, both bulk and bin, have traditionally been used for past studies of the stratocumulus transition to open cells (e.g., Wang & Feingold, 2009; Mechem & Kogan, 2003; Berner et al., 2011; Duynkerke et al., 2004;
Glassmeier & Feingold, 2017; Goren et al., 2019). While relatively computationally efficient, several limitations (discussed below) could affect the ability of these schemes to
capture aerosol-cloud-precipitation interactions and their impact on macroscopic cloud

properties and mesoscale features. Lagrangian passive trajectory models with improved treatment of aerosols have also been used in the past for process-level studies of drizzle formation and aerosol-cloud interactions in stratocumulus (e.g., Feingold et al., 1999; Pinsky et al., 2008; Magaritz et al., 2009). However, these models neglected feedback between the dynamics and microphysics, used Eulerian bin schemes, and were mainly restricted to small two-dimensional domains.

With recent advances in computing power, LES using a mesoscale domain with de-124 tailed Lagrangian particle-based microphysics explicitly representing aerosol-cloud in-125 teractions is now feasible. This modeling setup can elucidate complex cloud-aerosol-precipitation-126 dynamics interactions with greater fidelity than models with traditional Eulerian bulk 127 or bin microphysics schemes. Lagrangian particle-based schemes track "super-particles" 128 in the modeled flow, each representing a multitude of real aerosol, cloud and rain par-129 ticles. In this work, we use the Super-Droplet Method (SDM) Lagrangian scheme (Shima 130 et al., 2009) in LES. Lagrangian particle-based schemes address key limitations in Eu-131 lerian bulk and bin schemes (Grabowski et al., 2019), particularly by their ability to track 132 cloud-borne aerosols explicitly as well as by eliminating numerical diffusion of cloud and 133 precipitation variables which is problematic for traditional Eulerian schemes (Morrison 134 et al., 2018; Lee et al., 2021; Chandrakar, Morrison, Grabowski, & Bryan, 2022). A few 135 recent studies (e.g., Andrejczuk et al., 2010; Dziekan et al., 2021; Chandrakar, Morri-136 son, & Witte, 2022; Hoffmann & Feingold, 2023) demonstrated the potential of Lagrangian 137 microphysics schemes in LES for simulating aerosol-cloud interactions in stratocumu-138 lus clouds. In this article, we investigate the role of aerosol scavenging and processing 139 in the cellular transition of a stratocumulus cloud field using a Lagrangian super-particle 140 scheme and compare results to those using an Eulerian bin scheme. Specifically, we in-141 vestigate the following science questions: 142

• How do the scavenging and processing of aerosols by physical cloud processes im-143 pact the evolution of cloud microphysical properties and drizzle formation? 144 • How do aerosol scavenging and processing influence the transition from closed to 145 open cells? What is the impact of aerosol processing on drizzle formation and the 146 cellular transition relative to that of aerosol scavenging alone? 147 • Considering the limitations of bin schemes, how do simulations with a bin scheme 148 evolve differently than those with SDM? How do cloud properties simulated us-149 ing SDM and the bin scheme compare after the transition to open cells? 150

The rest of the manuscript is outlined as follows. Section 2 discusses the model, simulation setup, and sensitivity cases. In the results section (Sec. 3), the evolution of cloud properties during the cellular transition from the control SDM simulation is presented first to illustrate results using the most detailed representation of aerosol and cloud physical processes. Aerosol scavenging and processing during the cellular transition are discussed in Sec. 3.2, and the SDM and bin simulations are compared in Sec. 3.3. Finally, we summarize the significant findings and provide conclusions in Sec. 4.

¹⁵⁸ 2 Model Description and Case Setup

The current study uses CM1 (Cloud Model 1; Bryan & Fritsch, 2002) non-hydrostatic dynamical core in a LES configuration to simulate the DYCOMS-II RF02 drizzling stratocumulus case from Ackerman et al. (2009). A prognostic subgrid turbulent kinetic energy scheme (Deardorff, 1980) is used for subgrid closure. The Lagrangian super-droplet method (SDM, Shima et al., 2020) and the Eulerian Tel Aviv University (TAU) bin scheme (Tzivion et al., 1987; Feingold et al., 1988; Stevens et al., 1996) are used as microphysical models in CM1.

The SDM simulation has 128 super-particles on average per grid box representing 166 aerosol, cloud, and rain particles. The Lagrangian transport equation of SDM uses a ve-167 locity field linearly interpolated to the location of each particle from the CM1 Eulerian 168 wind field and a Lagrangian subgrid velocity associated with each particle driven by LES 169 subgrid turbulence statistics (Chandrakar et al., 2021). SDM tracks solute mass in haze, 170 cloud and rain drops. It explicitly represents the activation and wet growth of aerosols 171 by accounting for curvature and solute effects in the condensation growth equation for 172 all particles. SDM uses a stochastic collision-coalescence scheme (Shima et al., 2009) with 173 the Hall coalescence kernel for drop coalescence. Impaction aerosol scavenging is repre-174 sented using the Brownian coagulation kernel (Seinfeld & Pandis, 2016). There is no ex-175 ternal aerosol source in the boundary layer (only internal sources through regeneration 176 from drop evaporation and entrainment from the free troposphere) to simplify the in-177 vestigation of the transition. See Chandrakar et al. (2021); Chandrakar, Morrison, Grabowski, 178 and Bryan (2022) for more details of the CM1-SDM model. 179

Contrary to SDM, the version of TAU used here does not track the solute mass in 180 cloud and rain drops. It tracks the water mass of haze aerosols but assumes a quasi-equilibrium 181 wet radius following the Köhler curve. When droplets activate, they are placed in ap-182 propriate bins based on their critical radius. During evaporation, deactivated cloud droplets 183 are placed in aerosol bins, which are re-filled up to the background (initial) aerosol con-184 centration. The bins are re-filled starting with the smallest aerosol bin that is depleted 185 relative to the background aerosol, and moving to progressively larger bins until all de-186 activated drops are transferred to aerosol during the time step. The aerosol size distri-187 bution in each LES grid cell is tracked using 20 logarithmically spaced bins. Cloud/rain 188 drop size distributions are represented using 35 mass-doubling bins for number and mass 189 mixing ratios (two-moment) with a minimum radius of $1.56 \ \mu m$. Drop and aerosol bins 190 are advected using the scalar advection scheme in CM1. TAU solves the stochastic col-191 lection equation as in (Tzivion et al., 1987) using the Hall kernel. Further details of CM1-192 TAU and CM1-SDM are given in (Chandrakar, Morrison, Grabowski, & Bryan, 2022), 193 where they are compared in LES of a cumulus congestus cloud. 194

The case setup is the same as presented in Chandrakar, Morrison, and Witte (2022). 195 The boundary layer is driven by constant surface latent and sensible heat fluxes (Ackerman 196 et al., 2009) and a bulk longwave radiative cooling approach based Stevens et al. (2005). 197 A uniform large-scale horizontal divergence is applied to represent subsidence. The ini-198 tial wind shear is set to zero to simplify the setup and avoid the influence of shear on 199 the mesoscale organization and transition to open cells. The simulation domain size is 200 $50 \times 25 \text{ km}^2$ horizontally and 1.5 km vertically with uniform 100 m horizontal and 5 m 201 vertical grid spacings. A relatively fine vertical grid spacing is used to represent entrain-202 ment and cloud processes better (Mellado et al., 2018), and coarser horizontal grid spac-203 ing is necessary for the feasibility of using a mesoscale domain. These grid spacings are 204 finer than some previous studies of the cellular transition (e.g., Goren et al., 2019) but 205 the same as our recent work in Chandrakar, Morrison, and Witte (2022). The CM1 gov-206 erning equations of the Eulerian fields and the TAU bin scheme are integrated using 0.6 207 s time steps. The Lagrangian SDM scheme requires smaller substeps due to condensa-208 tion/evaporation calculation with curvature and solute terms and the stochastic coales-209 cence scheme (0.15 s for the current case). The SDM and TAU simulations are initial-210 ized with a uniform bi-modal lognormal distribution of ammonium sulfate aerosols from 211 Ackerman et al. (2009) but with a factor of five reduced concentration (34 mg^{-1}) to ac-212 celerate the cellular transition process. The maximum radius of the initial (dry) aerosol 213 distribution is set to 1 μ m. 214

Various sensitivity tests are designed to investigate the impacts of aerosol scavenging and processing and to target the science questions posed in the introduction; these tests are listed in Tab. 1. In some of the tests using TAU, enhanced scavenging is considered by adding only a fraction of deactivated drops back to the aerosol population when

Case	Microphysics scheme	Aerosol input	Aerosol impaction scaveng- ing
SDM	SDM	Aerosols from DYCOMS-II	Explicitly using a diffusion kernel
TAUSC90	TAU	Aerosols from DYCOMS-II	Implicit scavenging (replen- ish only 90% of deactivated drops to aerosols)
TAUSC80	TAU	Aerosols from DYCOMS-II	Implicit scavenging (replen- ish only 80% of deactivated drops to aerosols)
TAUSC70	TAU	Aerosols from DYCOMS-II	Implicit scavenging (replen- ish only 70% of deactivated drops to aerosols)
TAU-AERO-HI- PROC	TAU	Processed distribution shape from SDM but with the ini- tial concentration	NO
TAU-AERO-LO- PROC	TAU	Processed distribution shape and number from SDM (mixing ratio: $16.5 mq^{-1}$)	NO
TAU-AERO-LO	TAU	Aerosol number from SDM (mixing ratio: 16.5 mg^{-1}) but with the initial distribu- tion shape	NO

 Table 1. Microphysical sensitivity tests presented in this article.

cloud and rain drops evaporate (which we refer to as "implicit" scavenging) to compen-219 sate for neglecting impaction scavenging. For example, in TAUSC90, TAUSC80, and TAUSC80, 220 90%, 80%, and 70% of deactivated drops are added back to the smallest unfilled aerosol 221 bin relative to the initial background concentration, respectively. These sensitivity tests 222 also help evaluate the impact of aerosol scavenging on cloud fields and precipitation evo-223 lution. The sensitivity test TAU-AERO-LO-PROC uses the processed aerosol distribu-224 tion from SDM at 5 hours input into TAU at a simulation time of 3.5 hours. This test 225 investigates the impact of the processed aerosol size distribution shape on the subsequent 226 evolution of precipitation and the transition to open cells. TAU-AERO-HI-PROC is the 227 same as TAU-AERO-LO-PROC except it scales the input processed aerosol distribution 228 to give the same total aerosol concentration as at the beginning of the baseline TAU and 229 SDM runs. TAU-AERO-LO also modifies the input aerosol at 3.5 hours and uses the same 230 aerosol concentration as TAU-AERO-LO-PROC but with the initial aerosol distribution 231 shape. Thus, comparing TAU-AERO-LO-PROC, TAU-AERO-HI-PROC, and TAU-AERO-232 LO helps to isolate the impact of reduced concentration via scavenging versus changes 233 to the shape of the aerosol distribution via processing. 234

235 3 Results

236 237

3.1 Cloud field evolution in the SDM simulation with explicit aerosol physics

Figure 1(left) displays the cloud water path (CWP) and transient macroscopic cloud structure as the cloud system evolves with aerosol-cloud microphysical interactions in the SDM run. The cloud structure grows with time to larger organized closed cells with "cloud holes" at the edges. At 422 min of simulation time, CWP shows a clear break-



Figure 1. Comparison of cloud water path at different times during the cellular transition in SDM and TAUSC90 runs. The colorbars show magnitude in kg m^{-2} .



Figure 2. Cloud droplet activation locations in X-Y plane during (a) closed and (b) open cellular phase of the stratocumulus evolution from the SDM run. It is based on individual super-particle outputs from 30 sec of simulation time. The activation radius threshold based on the Köhler theory is used to determine activated super-particles.



Figure 3. Average in-cloud vertical profiles of (a) cloud water mixing ratio (b) rain water mixing ratio (c) cloud droplet number mixing ratio (d) rain drop number mixing ratio (e) mean radius, and (d) radius standard deviation at different simulation times from the SDM run. These profiles are obtained from spatiotemporal averaging (3 samples within 15 min time windows) over cloudy grid cells ($qc > 10^{-5}$ kg kg⁻¹).

down from closed to open cell structures with large cloud water path in narrow updrafts 242 at cell boundaries and wider cloud-free regions in downdrafts. Figure 2 illustrates the 243 pattern of drop activation at two different simulation times with closed versus open cells 244 from the SDM simulation, demonstrating distinctly different droplet activation variabil-245 ity associated with mesoscale cellular structures. During the initial closed cell phase, droplet 246 activation occurs in small-scale updrafts scattered throughout the cloud field, and after 247 transitioning to the open cell state, it mainly occurs in stronger cloud updrafts located 248 near cell boundaries. For the same setup, Chandrakar, Morrison, and Witte (2022) showed 249 that this cellular transition occurs when the drop coalescence time becomes shorter than 250 the large eddy turnover timescale, leading to greater drop coalescence and increased pre-251 cipitation flux. While the eddy turnover timescale is fairly steady, the coalescence timescale 252 is linked to the mean drop radius and DSD width and evolves over time. This evolution 253 of the DSD is closely tied to the aerosol concentration in the cloud system, driving droplet 254 activation and controlling the phase relaxation timescale (a measure of the competition 255 for the water vapor field). This suggests a critical role of aerosol removal (scavenging) 256 in the transition process for a given meteorological condition and begs further investi-257 gation. The role of aerosol scavenging in the transition has been suggested in past stud-258 ies as well (e.g., Mechem et al., 2006; Kazil et al., 2011; Berner et al., 2013; Erfani et al., 259 2022), but using models that did not explicitly represent cloud-borne aerosols and cloud 260 processing and scavenging of aerosols. 261

Figure 3 shows changes in the mean vertical profiles of cloud properties (40 μ m cloud-262 rain threshold radius) as the cloud field evolves. The peak cloud water mixing ratio in-263 creases with time up to four hours $(4.08 \times 10^{-4} \text{ to } 5.35 \times 10^{-4} \text{ kg kg}^{-1})$. Then it slightly 264 decreases when transitioning to open cells. This trend of cloud water is opposite to the 265 rain water evolution; the mean rain mixing ratio increases slowly $(4.55 \times 10^{-6} \text{ to } 3.92 \times 10^{-6} \text{ to }$ 266 10^{-5} kg kg⁻¹) during the first four hours but increases at a faster rate thereafter $(1.06 \times$ 267 10^{-4} kg kg⁻¹ at 7 hours). However, below ~ 500 m, the mean cloud water mixing ra-268 tio has a larger magnitude during the open cellular phase (6-7 hours) than at earlier times 269 (e.g., $\sim 5.67 \times 10^{-5}$ kg kg⁻¹ increase between 4-7 hours at 400 m). This is likely from 270 drizzle drops that evaporate to smaller sizes (crossing the 40 μ m cloud-rain threshold 271 radius) in downdrafts. This causes fluctuations in the cloud base height (and affects droplet 272 activation as well, not shown), as evident by the droplet number concentration profiles. 273 A sharp vertical gradient in cloud droplet number concentration near the cloud base be-274 comes smoother over time, and in the open cellular phase, the drop concentration is higher 275 below ~ 370 m than in the closed cell phase (e.g., ~ $3.6 \times 10^6 \text{ kg}^{-1}$ increase between 276 4-7 hours at 350 m). But overall, the droplet concentration decreases with time at all 277 other altitudes due to aerosol scavenging, i.e., droplet loss from collision-coalescence and 278 deactivation that overwhelms the activation flux. Could this also hint at cloud process-279 ing of aerosols that produce larger CCN after evaporation in the lower part of the bound-280 ary layer? We explore this possibility in detail in later subsections. Droplet concentra-281 tion profiles in the upper part of the cloud layer show a peak near the cloud top, indi-282 cating activation of entrained aerosols. The rain drop concentration peaks around 700 283 m and decreases above due to transport/sedimentation, such that the height of the peak 284 rain drop concentration is about 150 m below cloud top (see Fig. 1 in Chandrakar, Mor-285 rison, & Witte, 2022). 286

The mean cloud droplet radius R_m increases above 400 m, as expected for rising cloud parcels. Near cloud top, R_m decreases due to entrainment and evaporation as well as activation of new (small) droplets. Below 400 m, R_m is larger because of evaporating drizzle drops crossing the cloud-rain size threshold. The increase in mean radius with decreasing height is less sharp after transitioning to open cells (the difference in R_m between 200 and 410 m changes from ~29 to 13 μ m between 1 to 7 hours). This is due to differences in droplet activation, as discussed above.

Vertical profiles of the standard deviation of droplet radius within each grid cell 294 σ_R , averaged over all cloudy points, exhibit some interesting features as the transition 295 progresses. During the first two hours, σ_R increases with height by about 1.5-2.5 μ m be-296 tween ~ 400 and 600 m (note it slightly decreases between ~ 600 m to just below cloud 297 top at 2 hours). This increase of σ_R with altitude is from the mixing of cloud parcels with 298 different growth histories in closed cell updrafts as well as some mixing at the updraft-299 downdraft interface (Chandrakar, Morrison, & Witte, 2022). A sharper increase of σ_R 300 near cloud top ($\Delta \sigma_R \sim 0.5, 2$, and 1.7 μ m at t = 1, 4, and 7 hours, respectively) is from 301 entertainment-mixing and associated drop activation that changes with time due to the 302 change in turbulence intensity and boundary layer aerosol concentration. Interestingly, 303 profiles after 2 hours show a decrease of σ_R with altitude above 450 m (decrease between 304 450 and 800 m of \sim 1.8 μ m at 3 hours and 3.7 μ m at 7 hours), although the magnitude 305 of the average σ_R increases with time after 4 hours. The rate of decrease with altitude 306 also increases with time. More evaporation of drizzle drops in downdrafts, their mixing 307 with updraft regions, and higher variability in drop age due to the overturning motion 308 of large eddies lead to a larger DSD width at lower altitudes at later times when there 309 is more drizzle in clouds (Chandrakar, Morrison, & Witte, 2022). The magnitude of the 310 mean σ_R increases with time due to greater supersaturation variability as the cloud gets 311 cleaner and as drop collision-coalescence increases. Mean σ_R below 400 m also increases 312 sharply with decreasing altitude (difference in σ_R between 200 and 400 m increasing from 313 $\sim 4-7.6 \ \mu m$ between 1 and 5 hr) because of evaporating drizzle drops. After the tran-314 sition to open cells, activation of new droplets between 200 and 400 m dominates the dis-315 tribution and reduces the magnitude of σ_R . 316

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3.2 Role of aerosol scavenging on the transition to open cells

When a stratocumulus system evolves with time, it progressively gets cleaner with-318 out significant external aerosol sources. The wet scavenging of aerosols through cloud 319 formation and coalescence causes the concentrations of both cloud droplets and aerosols 320 to decrease with time. The coalescence of cloud and drizzle drops and diffusion of aerosols 321 to drops increase the solute mass of aerosols in drops. Once they evaporate in the lower 322 part of the boundary layer, they are available again for cloud formation but with a greater 323 potential for activation and growth due to their larger sizes. Figure 4 demonstrates this 324 physical aerosol processing by clouds and resultant dry aerosol distribution after suffi-325 cient cloud cycling (time = 7 hours) at different altitudes. At all altitudes, the right tail 326 of dry aerosol distributions is broader than the initial background aerosol distribution. 327 This enhanced concentration of large aerosols and larger maximum sizes than the orig-328 inal aerosol distribution (2.5 μ m versus 1 μ m) suggests significant processing of aerosols. 329 With a decrease in altitude, the relative concentration of large aerosol particles in the 330 distribution tail and maximum particle size increase. At 200-250 m, an additional large 331 aerosol mode with modal radius of about 300 nm is apparent. This large aerosol mode 332 at lower levels is explained by the higher fall speeds of large drizzle drops, which are more 333 likely to contain large solute mass compared to smaller cloud drops. These faster-falling 334 drizzle drops are able to survive well below cloud base prior to evaporating, thereby re-335 generating large aerosol particles at low levels. 336

Figure 5 shows the decay of in-cloud and boundary layer averaged aerosol concen-337 trations in the SDM and TAU runs listed in Tab. 1. A sharp reduction of in-cloud aerosols 338 (Fig. 5a) within the first hour ($\sim 45\%$ in 30 min) in all of the TAU runs suggests a higher 339 activation rate, but this is balanced by a higher deactivation rate to give a similar de-340 cay rate of boundary layer-averaged aerosol concentration as SDM (Fig. 5b). After the 341 first hour, aerosol concentration is depleted significantly faster in SDM compared to TAUSC90 342 and TAUSC80 (e-folding timescale ~ 8.3 and 6.9 hours, respectively, versus 5.6 hours 343 for SDM in the cloud layer) in both the cloud layer and through the entire boundary layer. 344 A faster decay rate in SDM is due to impaction scavenging of aerosols by cloud and driz-345 zle drops (neglected in TAU) and faster removal through precipitation. Larger differences 346



Figure 4. Distributions of solute radius in activated droplets (solid lines) at different altitudes from t = 7 hours from the SDM run and their comparison with the initial aerosol distribution (black dashed-line).



Figure 5. Time series of aerosol mixing ratio averaged over (a) cloudy grid cells $(qc > 10^{-5} \text{ kg} \text{ kg}^{-1})$ and (b) the entire boundary layer up to the average cloud top. Different lines are from the SDM and TAU runs listed in Table 1. Squares mark the transition time to open cells.

in average boundary layer aerosol concentration between SDM and TAU begin around
 the time when surface precipitation deviates significantly between the simulations (dis cussed in the following subsection).

As shown in Fig. 5, the implicit scavenging in TAUSC90 and TAUSC80 is insuf-350 ficient to match the in-cloud aerosol decay in the SDM run. With an increase in implicit 351 scavenging compared to TAUSC90 and TAUSC80, the TAUSC70 run is similar to the 352 SDM aerosol decay rate within the cloud layer (~ 5.9 hours e-folding timescale for TAUSC70). 353 However, TAUSC70 still produces insufficient depletion of average boundary layer aerosol 354 concentration, with a mean aerosol number mixing ratio about $3-4 \text{ mg}^{-1}$ higher in TAUSC70 355 than SDM (e-folding timescale 8.1 hours versus 6.6 hours). Reduced loss of boundary 356 layer aerosol suggests that the precipitation loss is lower for TAU runs; that is, there is 357 less drizzle formation and more evaporation in the boundary layer. It is important to 358 note that aerosol processing is not considered in any of these TAU runs (TAUSC90, TAUSC80. 359 TAUSC70) while it is considered in SDM. This also impacts how aerosols participate in 360 cloud cycling and, thus, affects the aerosol removal in a positive feedback loop. 361

The impact of aerosol processing is further investigated by using TAU but with a 362 processed aerosol distribution from SDM (at 5 hours, about 1.5 hours before the tran-363 sition to open cells in SDM). The processed aerosol distribution is included in TAU at a simulation time of 3.5 hours (TAU-AERO-LO-PROC), and the aerosol concentration 365 thereafter is rapidly depleted (a factor ~ 1.5 decrease of the in-cloud aerosol concentra-366 tion within an hour) as aerosols activate as cloud droplets. This rapid depletion through 367 droplet activation in TAU could be due to the quasi-equilibrium assumption and neglect 368 of growth kinetics during the activation process (Chuang et al., 1997). A greater acti-369 370 vation loss of aerosols in TAU compared to SDM is also consistent with larger droplet activation rates near cloud base in cumulus congestus simulations (Chandrakar, Mor-371 rison, Grabowski, & Bryan, 2022). After this initial adjustment, the aerosol concentra-372 tion is significantly lower in TAU-AERO-LO-PROC. However, the decrease in aerosol 373 concentration is much slower in the cloud layer after this initial adjustment since no ad-374 ditional impaction scavenging exists. To contrast the impact of this clean and processed 375 aerosol environment, we performed two additional sensitivity simulations (restarting from 376 the same time at 3.5 hours): (a) TAU-AERO-LO, with the same reduced aerosol con-377 centration as TAU-AERO-LO-PROC but with the initial (unprocessed) aerosol distri-378 bution shape; and (b) TAU-AERO-HI-PROC, with the processed distribution shape from 379 SDM but with the initial aerosol concentration. For TAU-AERO-LO, in-cloud aerosols 380 decrease at a similar rate as TAU-AERO-LO-PROC during the rapid adjustment period 381 from 3.5 to 4.5 hours. However, the loss of aerosol in the boundary layer in TAU-AERO-382 LO is somewhat slower than TAU-AERO-LO-PROC, consistent with an enhanced precipitation-383 induced loss when the processed aerosol distribution shape is used in TAU-AERO-LO-384 PROC. Since TAU-AERO-HI-PROC is restarted at 3.5 hours with the initial aerosol con-385 centration, it has a significantly higher aerosol concentration than in all other runs. TAU-386 AERO-HI-PROC shows a similar rate of decrease in both cloud layer and boundary layer aerosol concentration as TAUSC90, even without any implicit scavenging in TAU-AERO-388 HI-PROC. This implies that the processed aerosol distribution shape leads to enhanced 389 scavenging in TAU-AERO-HI-PROC, which compensates for the lack of implicit scav-390 enging. 391

Impacts of aerosol scavenging and processing on the transition to open cells are demon-392 strated in Fig. 1. Cloud water path for the SDM run, which includes both physical pro-393 cessing of aerosols and impaction scavenging, evolves to an open cell structure signifi-394 cantly faster than TAUSC90 (in about one-half of the simulation time) and the other 395 TAU runs. The sizes of the closed cells are also larger in the TAU runs than in SDM. 396 Squares in Fig. 5a also mark the transition point from closed to open cells (defined by 397 cloud fraction $\langle \approx 56\% \rangle$ for all runs. These results show that increased implicit scaveng-398 ing in TAU (from TAUSC90 to TAUSC80 and TAUSC70) leads to a systematic decrease 399

in the time of transition to open cells. Similarly, reduced aerosol concentration in TAU-400 AERO-LO compared to the other TAU simulations leads to a faster transition. With the 401 processed aerosol distribution input at 3.5 hours in TAU-AERO-LO-PROC, the tran-402 sition happens earlier than in TAU-AERO-LO by nearly 2 hours. Moreover, the tran-403 sition is about 45 min faster with the processed aerosol distribution in TAU-AERO-HI-404 PROC than TAUSC90 despite the lower in-cloud and boundary layer aerosol concentra-405 tions and the presence of implicit scavenging in the latter. Overall, these results indi-406 cate that both the loss of aerosol concentration from scavenging as well as the change 407 in aerosol size distribution shape from aerosol processing are important in determining 408 the timing of the transition to open cells. 409

410 411

3.3 Comparison of cloud, precipitation, and dynamics evolution in Lagrangian SDM and Eulerian TAU bin simulations

Figure 6 compares the average properties of cloud and rain (cloud and rain water 412 mass and number mixing ratios, mean drop radius, and spectral width), vertically in-413 tegrated quantities (cloud fraction and liquid water path), dynamics (TKE and verti-414 cal velocity skewness), and the surface precipitation rate. The mean and standard de-415 viation of the drop radius are based on drop size distributions in each grid cell. The cloud 416 and rain statistics are averaged over all cloudy grid cells ($qc > 10^{-5}$ kg kg⁻¹). The peak 417 cloud water mixing ratio $(2.8 \times 10^{-4} \text{ kg kg}^{-1})$ is the second largest for SDM (after 2.9× 418 10^{-4} kg kg⁻¹ for TAU-AERO-HI-PROC), and it reaches a peak magnitude earlier than all TAU runs (4 versus 8 hours). A lower magnitude of cloud water in most TAU runs 420 is consistent with greater evaporation from numerical diffusion along cloud boundaries 421 (compared to the Lagrangian treatment in SDM), similar to differences between TAU 422 and SDM in cumulus congestus simulations (e.g., Chandrakar, Morrison, Grabowski, & 423 Bryan, 2022). The rain water mixing ratio also increases significantly faster to a larger 424 magnitude in SDM (8.25×10^{-5} kg kg⁻¹ at 7.45 hours) than in all TAU runs (e.g., 7.42× 425 10^{-5} kg kg⁻¹ at 13.08 hours for TAUSC70). The rain water mixing ratio also increases 426 faster with an increase in the implicit scavenging rate in TAU (from TAUSC90 to TAUSC80 427 and TAUSC70). Changes in rain water mixing ratio are generally consistent with changes 428 in cloud mixing ratio, with faster increases in rain mixing ratio associated with decreases 429 in cloud mixing ratio. For example, cloud water mixing ratio decreases faster after ~ 7 430 hours with an increase in the implicit scavenging for TAU. With the processed aerosol 431 distribution input into TAU-AERO-LO-PROC at 3.5 hours, rain water mixing ratio in-432 creases faster than in TAU-AERO-LO with its unprocessed aerosol distribution. Thus, 433 both increased scavenging as well as aerosol processing increase the formation of rain us-434 ing TAU. After initial adjustment, the cloud water evolution for TAU-AERO-LO-PROC 435 nearly matches SDM. The rate of rain water increase over time is also similar between 436 SDM and TAU-AERO-LO-PROC. The cloud droplet number mixing ratio is similar be-437 tween the SDM and TAU runs before ~ 3 hours, but the rain drop number mixing ra-438 tio is higher in SDM. After 3 hours, differences in the cloud and rain drop number mix-439 ing ratios among the simulations are similar to the mass mixing ratios. 440

The mean drop size increases over time faster in SDM than all of the TAU runs, 441 which is consistent with the cloud water mixing ratio plot. A larger mean radius in SDM 442 (for a similar drop number concentration during the first three hours) is also consistent 443 with spurious evaporation in the bin scheme from numerical diffusion of liquid water in 444 physical space. The numerical representation of the Eulerian advection of the bin vari-445 ables in the physical space might cause a spurious spread of cloud water to sub-saturated 446 downdrafts and cloud holes, leading to the evaporation of cloud droplets. This numer-117 ical artifact seems to produce more homogeneous mixing-like behavior in TAU, i.e., liq-448 uid mass loss with a nearly similar number relative to SDM. Due to the Lagrangian frame-449 work, such numerical artifacts are absent in SDM. Reduction of cloud water with a sim-450 ilar droplet number as SDM (and thus smaller mean radius) also occurred in the upper 451 part of the simulated cumulus congestus cloud using the TAU bin scheme in (Chandrakar, 452



Figure 6. Time evolution of average in-cloud statistics $(qc > 10^{-5} \text{ kg kg}^{-1})$ from the SDM and TAU runs listed in Table 1: (a) cloud water mixing ratio, (b) rain water mixing ratio, (c) cloud droplet number mixing ratio, (d) rain droplet number mixing ratio, (e) mean droplet radius, (f) standard deviation of droplet radius, (g) cloud fraction, and (h) liquid water path. Also shown are mean turbulence statistics in the cloud layer: (i) resolved turbulent kinetic energy and (j) vertical velocity skewness, and the (k) domain-mean surface precipitation rate.



Figure 7. Domain-averaged vertical profiles of (a) cloud water, (b) rain water, (c) cloud droplet number, and (d) rain drop number mixing ratios from SDM and TAU runs during the open cellular phase (approximately the same cloud fraction 56%). Note that the simulation time used for the comparison varies since the transition time differs among the runs.



Figure 8. Comparison of in-cloud average $(qc > 10^{-5} \text{ kg kg}^{-1})$ vertical profiles of (a) cloud water, (b) rain water, (c) cloud droplet number, (d) rain drop number mixing ratios, (e) mean droplet radius, and (f) standard deviation of droplet radius from the SDM and TAU runs during the open cellular phase (same as Fig. 7 except in-cloud versus domain-averaged quantities).

Morrison, Grabowski, & Bryan, 2022). A larger mean drop radius is likely the trigger 453 for faster development of rain within the first 3 hours in SDM. A positive feedback through 454 aerosol scavenging and processing (i.e., more aerosol scavenging with processing due to 455 faster rain growth that, in turn, enhances the acceleration of rain formation) is respon-456 sible for further enhancement of rain growth and aerosol removal in SDM, and conse-457 quently, changes in cloud droplet number and mean size. Thus, this three-component 458 feedback, coupling cloud, rain, and aerosols, initially triggered by a larger mean cloud 459 radius in SDM than TAU (due to the numerical diffusion in physical space in TAU), drives 460 a faster transition to open cells in the SDM run. As discussed earlier, TAU does not ex-461 plicitly consider aerosol processing and recirculation of the processed aerosols into clouds, 462 which contributes to slower rain formation and transition to open cells. A faster increase 463 of the mean drop size in TAU-AERO-LO-PROC compared to TAU-AERO-LO is shown 464 in Fig. 6, providing clear evidence of the impact of the processed aerosol size distribu-465 tion. The mean drop size response for different implicit scavenging rates in (TAUSC90, 466 TAUSC80, TAUSC70) is consistent with the droplet mixing ratio differences. 467

The standard deviation of drop radius is similar between the SDM and TAU runs 468 up to three hours of simulation time. After ~ 3.5 hours, the radius standard deviation 469 is significantly higher for SDM than all TAU runs (\sim 7.14 versus 5.68 μ m). Interestingly, 470 this result contrasts with previous findings of broader droplet spectra using bin micro-471 physics (relative to SDM) in simulations of cumulus congestus clouds (Chandrakar, Morrison, Grabowski, & Brvan, 2022). A larger spectral width in cumulus congestus sim-473 ulations is caused by significant numerical diffusion in droplet size space arising from ver-474 tical transport in the strong updraft environment with 50-100 m vertical grid spacing. 475 However, offline tests in Morrison et al. (2018) showed that the vertical grid spacing is a key factor in numerical broadening of DSDs, and the vertical grid spacing is a factor 477 of ten smaller in the current stratocumulus case (5 m). Thus, the numerical broaden-478 ing in the current stratocumulus TAU runs appears to have less impact than in the con-479 gestus case of Chandrakar, Morrison, Grabowski, and Bryan (2022). A smaller spectral 480 width in TAU runs than SDM is also likely from reduced collision-coalescence and rain 481 formation, as shown by the rain drop number and mass ratio time series. Similar to the 482 mean drop radius, the mean radius standard deviation in the TAU runs also has a faster 483 rate of increase after five hours with an increase in implicit scavenging. The mean ra-484 dius standard deviation also responds similarly to the mean radius when the processed 485 aerosol distribution from SDM is input into TAU (TAU-AERO-LO-PROC and TAU-AERO-486 HI-PROC). However, interestingly, over time the mean radius standard deviation in all 487 of the TAU runs saturates to approximately the same value ($\sim 5.5 \ \mu m$) after the tran-488 sition from closed to open cells. 489

The integral cloud properties like cloud fraction and liquid water path follow time evolution consistent with the cloud water mixing ratio for each case. However, the cloud fraction is the lowest for SDM but it has a higher mean cloud water mixing ratio than the TAU runs, which is again consistent with numerical diffusion in TAU. The mean liquid water path (LWP) also increases faster over time in the SDM run, following the pattern of the mean cloud water mixing ratio.

Dynamical quantities also show a wide range of diversity among the simulations. 496 497 The mean turbulent kinetic energy (TKE) follows the LWP response. Mean TKE valuses from the SDM and TAU simulations are similar prior to ~ 4.5 hours, but it decreases 498 thereafter in SDM with the reduced LWP and increased precipitation. The mean ver-499 tical velocity skewness in the cloud layer increases (i.e., updrafts get stronger) up to \sim 500 2 hours and then decreases for SDM. However, for TAU runs, the vertical velocity skew-501 ness peak occurs later (~ 3.3 hours), likely due to the delay in rain formation affecting 502 the sedimentation flux and, consequently, the stability of the cloud layer. The surface 503 precipitation rate, a key quantity for the cellular transition process and aerosol removal 504 (Stevens et al., 2005; Savic-Jovcic & Stevens, 2008; Wood et al., 2008; Goren & Rosen-505

feld, 2012), also increases significantly faster in SDM than the TAU runs consistent with 506 the other quantities discussed above. In SDM, the mean precipitation rate sharply in-507 creases beginning at ~ 4 hours, coincident with the decrease in cloud water mixing ra-508 tio, liquid water path, and TKE. For TAU runs, the surface precipitation rate increases 509 earlier as the implicit aerosol scavenging is increased, but surface precipitation is sub-510 stantially delayed relative to SDM in all cases. When the aerosol distribution from SDM 511 is used in TAU (TAU-AERO-LO-PROC), the rate of increase in precipitation is simi-512 lar to SDM although delayed about 1.5 hours owing to spinup since the processed aerosol 513 is not input in TAU-AERO-LO-PROC until 3.5 hours. Moreover, the increase in pre-514 cipitation is delayed ~ 45 min in TAU-AERO-LO compared to TAU-AERO-LO-PROC. 515 This illustrates the impact of changes in aerosol size distribution shape from aerosol pro-516 cessing since both runs input the same reduced aerosol concentration from SDM. 517

Figure 7 compares the domain-averaged vertical profiles (without any threshold for 518 cloud water) of cloud and rain water number and mass mixing ratios after the transi-519 tion to open cells. These profiles are similar between the SDM and TAU runs. However, 520 the cloud top is slightly lower, and the cloud water mixing ratio is slightly larger for SDM 521 compared to all TAU runs between 350-700 m. When comparing the average profiles of 522 cloud and rain quantities based on a threshold for cloudy grid cells $qc > 10^{-5}$ kg kg⁻¹ 523 (Fig. 8), differences between SDM and TAU runs are more evident. Between 450-800 m, 524 SDM has a significantly higher mean cloud water mixing ratio (SDM versus TAU peaks: 525 $4.8 \times 10^{-4} \text{ kg kg}^{-1}$ versus $3.5 \times 10^{-4} \text{ kg kg}^{-1}$). This suggests the cloud field is more 526 diffused in TAU even after the transition to open cells, again likely from the numerical 527 diffusion. The in-cloud rain water peak is also slightly stronger in SDM $(9.3 \times 10^{-5} \text{ kg})$ 528 kg^{-1}) than in TAU (max ~ 8.1×10⁻⁵ kg kg⁻¹), and the rain drop number mixing ratio is larger in SDM (peak: $1.3 \times 10^5 \text{ kg}^{-1}$) than TAU (peak max ~ $1.0 \times 10^5 \text{ kg}^{-1}$) at 530 all levels below 750 m. A smaller drop number but similar rain water below the peak rain-531 water implies that the mean rain drop size is slightly larger in TAU runs. The cloud droplet 532 number mixing ratio profiles (averaged over cloudy grid cells) differ drastically between 533 SDM and TAU runs. SDM has a nearly uniform number mixing ratio in the cloud layer 534 that sharply decreases near the top. Similarly, below 350 m, it gradually drops to zero. 535 However, for the TAU runs, there is a nearly uniform number mixing ratio above 500 536 m, but it increases below this level to a significantly larger value than in SDM. This re-537 sult suggests more activation of cloud droplets at lower levels from moistening of the bound-538 ary layer in TAU. A more diffuse liquid water field in the Eulerian TAU simulations could 539 explain boundary layer moistening via increased evaporation in downdrafts. Moreover, 540 as discussed earlier, the quasi-equilibrium assumption and the neglect of growth kinet-541 ics during the activation process (Chuang et al., 1997) may also cause a higher droplet 542 activation rate in TAU. The presence of a large concentration of in-cloud drizzle drops 543 (likely near the cloud-drizzle threshold) and small cloud droplet concentration in SDM causes the mean and standard deviation of drop radius to deviate from TAU runs be-545 low 450 m (a larger mean and standard deviation in SDM). Otherwise, they are nearly 546 the same for SDM and TAU runs above 450 m. 547

548 4 Summary and Conclusions

Shallow boundary layer clouds are a critical component of global climate and are 549 sensitive to anthropogenic pollution. Complex interactions between aerosol, cloud, and 550 precipitation at various scales make these clouds interesting but also challenging to rep-551 resent in climate models. Even process-level models struggle to represent aerosol-cloud-552 precipitation interactions explicitly due complications from cloud-borne aerosol and aerosol 553 processing. Lagrangian particle-based microphysics schemes are particularly well-suited 554 to account for these interactions with fewer simplifications and assumptions compared 555 to traditional Eulerian schemes. We utilized this tool (the Super-Droplet Method, or SDM) 556 to investigate the evolution of cloud micro- and macro-physical properties and physical 557

aerosol scavenging and processing during the mesoscale transition to open cells, extend ing the analysis presented in Chandrakar, Morrison, and Witte (2022). By contrasting
 results from SDM with simulations using the TAU bin scheme, we determined the impact of aerosol scavenging and processing on precipitation generation and the transition
 process. The impacts of simplifying aerosol-cloud interactions and numerical diffusion
 in the Eulerian bin scheme (TAU) on cloud field evolution, the mesoscale transition, and
 cloud properties in the final open-cell state were further investigated by comparing SDM
 and TAU results.

The simulation using Lagrangian microphysics with explicit aerosol activation and 566 impaction scavenging showed significant physical processing of aerosols that impacted 567 precipitation generation and evolution of the cloud field. Both changes in aerosol dis-568 tribution shape from processing and reduced concentration from scavenging were shown 569 to be important for the aerosol removal rate through precipitation. Past observations 570 (e.g., Sharon et al., 2006) indicated that the presence of extremely clean conditions co-571 incides with the cellular regions. In this study, we showed that physical processing of aerosol 572 and changes in aerosol size distribution shape also impact the time for transition to open 573 cells; an earlier transition occurred due to the processed aerosol distribution shape. 574

This study provided detailed insights into the evolution of the vertical structure 575 of cloud and rain quantities during the closed-to-open-cell transition. During the open 576 cell stage, moistening of the boundary layer caused droplet activation to occur at lower 577 levels than the initial cloud base, resulting in a smaller DSD mean and standard devi-578 ation at lower levels using SDM. Although the DSD mean and standard deviation increased overall, DSD width had a progressively steeper decreasing profile with height dur-580 581 ing the transition to open cells, contrasting with an increasing profile during the earlier closed cell phase. This was explained by more drizzle formation, the evaporation of driz-582 zle drops in downdrafts, and mixing of drops of different growth history producing broader 583 DSDs at lower compared to upper levels after the transition to open cells. 584

The simulation with SDM was compared to TAU simulations that applied an im-585 plicit representation of aerosol scavenging without aerosol processing. The TAU runs had 586 a significant delay in the transition to open cells compared to SDM, even when the im-587 plicit aerosol scavenging was increased to match the in-cloud aerosol concentration evo-588 lution in SDM. TAU runs could not capture the rapid decay of boundary layer aerosol 589 concentration that occurred with SDM. Sensitivity tests suggested that the representa-590 tion of aerosol processing is important for removing aerosols through precipitation. Spu-591 rious evaporation in TAU from numerical diffusion along cloud boundaries also reduced 592 cloud water and mean droplet size compared to SDM. A larger mean drop radius and 593 greater cloud water mixing ratios helped to initiate faster rain development in SDM. Af-594 ter the initial phase, a positive feedback associated with enhanced aerosol scavenging due 595 to faster rain growth in turn further enhanced rain formation in SDM. This feedback was responsible for accelerated aerosol removal and, consequently, reduced cloud droplet num-597 ber, increased mean droplet size, and increased spectral width. Thus, a positive feed-598 back via aerosol-cloud-precipitation interactions combined with initially larger mean ra-599 dius and cloud water mixing ratios helped to drive a faster transition to open cells in SDM 600 compared to TAU. 601

The absence of aerosol processing and recirculation of the processed aerosols into 602 clouds in TAU was another key factor for slower rain formation and transition to open 603 cells. Interestingly, this result contrasts with previous findings of broader droplet spec-604 tra using bin microphysics (relative to SDM), which led to faster precipitation genera-605 tion in simulations of cumulus congestus clouds (Chandrakar, Morrison, Grabowski, & 606 Bryan, 2022). This suggests that aerosol-cloud-precipitation interactions in SDM dom-607 inated the effects of numerical broadening in TAU for this stratocumulus case (a much 608 smaller vertical grid spacing, which impacts the degree of numerical DSD broadening, 609 in the current study compared to the earlier cumulus congestus simulations may have 610

also contributed to a reduced impact of numerical DSD broadening here). After the tran sition to open cells, some differences in cloud and rain quantities between SDM and TAU
 persisted. More droplet activation below 400 m in TAU runs caused a deviation in the
 mean and standard deviation of drop radius compared to SDM.

This study highlights the importance of aerosol scavenging and physical aerosol pro-615 cessing on drizzle formation and the open cell transition using a state-of-the-art mod-616 eling tool (LES with a super-particle microphysics scheme) with an explicit, detailed rep-617 resentation of aerosol-cloud interactions. Recent advances in computing power have made 618 LES with Lagrangian particle-based microphysics feasible over a mesoscale domain. This 619 work built upon previous studies on aerosol processing and the mesoscale transition us-620 ing models with simpler microphysics schemes (e.g., Feingold & Kreidenweis, 2002; Kazil 621 et al., 2011; Goren et al., 2019; Erfani et al., 2022). Further studies on several related 622 topics, including detailed quantification of aerosol scavenging and processing in differ-623 ent cloud regions, statistics of the recycling of processed aerosols, impacts of initially clean 624 versus polluted aerosol conditions, and the role of chemical aerosol processing, are pos-625 sible using LES with the Lagrangian super-particle scheme. This would further improve 626 understanding of aerosol-cloud interactions in stratocumulus clouds and help to inform 627 bulk microphysics parameterizations. 628

⁶²⁹ Open Research

The current study used the CM1 model release 19.8 for the simulations presented here. CM1 code with detail documentation and DYCOMS-II RF02 case setup is available at https://www2.mmm.ucar.edu/people/bryan/cm1/. SDM code provided by Shinichiro Shima is available at https://github.com/Shima-Lab/SCALE-SDM_BOMEX_Sato2018.

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Figure 1.



Figure 2.



(a) t = 1.3 hr

Figure 3.





Figure 4.



Figure 5.



Figure 6 Part-I.



Figure 6 Part-II.



Figure 7.



Figure 8.



