

1 **Understanding the Causes of Satellite–Model Discrepancies in**
2 **Aerosol–Cloud Interactions Using Near-LES Simulations of Marine**
3 **Boundary Layer Clouds**

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17 **Abstract.** Aerosol–cloud interactions (ACI) remain the largest source of uncertainty in model
18 estimates of anthropogenic radiative forcing, primarily because of deficiencies in representing
19 aerosol–cloud microphysical processes that lead to inconsistent cloud liquid water path (LWP)
20 responses to aerosol perturbations between observations and models. To investigate this
21 discrepancy, we conducted a series of large-eddy scale simulations driven by realistic
22 meteorology over the Eastern North Atlantic, and evaluated LWP susceptibility, precipitation
23 processes, and boundary layer thermodynamics using satellite and ground-based observations.

24 Simulated LWP responses show a strong dependence on cloud state. Non-precipitating thin
25 clouds exhibit a modest LWP decrease with increasing cloud droplet number concentration (N_d),
26 consistent in sign but weaker in magnitude than satellite estimates, reflecting enhanced turbulent
27 mixing and evaporation. The largest model-observation discrepancy occurs in non-precipitating
28 thick clouds, where simulated LWP susceptibilities are strongly positive (+0.32) while
29 observations indicate large negative values (−0.69). This discrepancy stems from excessive
30 precipitation driven by underestimated entrainment, overly active accretion, and overly broad
31 drop-size distributions in polluted conditions. While our high-resolution setup mitigates the
32 excessive drizzling common in coarser models and captures key regime transitions, these biases
33 persist—highlighting that improved parameterization of cloud-top processes, precipitation, and
34 aerosol effects are needed beyond simply increasing model resolution.

35 Additionally, misrepresented moisture inversions in reanalysis introduce a moist bias in cloud-
36 top relative humidity, further amplifying positive LWP susceptibility. Our results also suggest
37 that large negative N_d –LWP relationships in observations may reflect internal cloud processes
38 rather than true ACI effects.

42 **1. Introduction**

43 Marine boundary layer clouds exhibit substantial influence on Earth's radiation balance
44 due to their high albedo and extensive global coverage. Aerosols modulate cloud albedo through
45 changing cloud droplet number concentration (N_d), cloud liquid water path (LWP), and cloud
46 fraction. The estimated radiative cooling from aerosols partially offset the warming from
47 greenhouse gas emission (Slingo 1990). However, aerosol-cloud interaction (ACI) remains the
48 most uncertain component of anthropogenic radiative forcing (Foster et al., 2021). In particular,
49 liquid-phase cloud adjustments in LWP, cloud fraction, and cloud lifetime present the largest
50 uncertainties in determining the net radiative forcing of ACIs, especially under varying large-
51 scale conditions (Han et al., 2002; Small et al., 2009).

52 Among these uncertainties, the LWP response to aerosol perturbations has drawn
53 particular attention due to its large spread in both observations and numerical model simulations.
54 Theoretically, increasing aerosols would reduce droplet size and suppress precipitation, thereby
55 increasing LWP and cloud lifetime (Albrecht, 1989). However, smaller droplets might also
56 enhance evaporation and entrainment, leading to a reduced LWP in non-precipitating clouds
57 (Ackerman et al., 2004; Xue and Feingold, 2006; Bretherton et al., 2007). This competition
58 between processes leads to a bifurcated LWP response that varies with aerosol concentration,
59 cloud type, and background meteorology.

60 In recent years, numerous satellite studies have reported an overall decrease of LWP with
61 increasing N_d for non-precipitating clouds in polluted environments and an increase in LWP for
62 precipitating clouds (e.g., Gryspeerdt et al. 2019, 2021; Toll et al., 2019; Zhang et al., 2022,
63 2023; Qiu et al., 2024; Yuan et al., 2023; 2025). In contrast, current global climate models
64 (GCM) mostly simulate a positive LWP response to aerosol perturbation regardless the cloud
65 conditions, which leads to an over-estimation of the aerosol-induced radiative forcing that is
66 dominated by ACI (e.g., Ghan et al., 2016; Michibata et al., 2016; Mülmenstädt et al., 2024).
67 This discrepancy could stem from the poorly resolved cloud processes in GCM due to its coarse
68 horizontal resolution (~ 100 km). Recent development in computing have enable the global
69 convection-permitting models (GCPMs) with kilometer-scale grid spacing, serving as an
70 invaluable complement to the traditional climate models (e.g. Sato et al., 2019; Stevens et al.,
71 2019; Caldwell et al., 2021; Donahue et al., 2024). Notably, Sato et al. (2018) employed a
72 GCPM and simulated a negative LWP response, attributing it primarily to better resolved
73 evaporation and condensation processes from aerosol perturbations. Yet, other CPM studies with
74 finer resolution than Sato et al. (2018) mostly simulate an increase in LWP with aerosol
75 perturbations (e.g., Fons et al., 2024; Christensen et al, 2024), largely due to uncertainties in
76 microphysics schemes, particularly regarding the treatment of precipitation (White et al., 2017).

77 Since most current GCPMs and GCMs adopt two-moment microphysics schemes, it is
78 important to evaluate the precipitation parameterization in these schemes with observational
79 constraints, in addition to the influence of precipitation process on the simulated ACI.
80 Meanwhile, Terai et al. (2020) found that the lack of decrease in LWP in kilometer-scale models
81 could be due to the lack of resolving the sub-kilometer processes that are most relevant to ACI
82 processes. For example, they found that when increasing model resolution from 4 km to 250m,
83 the fraction of precipitating clouds largely decreases, especially for thick clouds, and the LWP
84 response becomes negative for non-precipitating clouds. Therefore, it is critical to assess the
85 benefit of increasing model resolution to near large-eddy simulation (LES) scale in representing
86 precipitation, as well as the evaporation-entrainment feedback responsible for LWP reduction

87 without altering the structure of the microphysics parameterization and ultimately reconcile the
88 LWP adjustment observed by satellite with those estimated by GCM and GCPM.

89 With model resolutions ranging from 25 m to 200 m, numerous LES studies have utilized
90 idealized meteorological conditions and have provided valuable process-level understanding on
91 the mechanisms governing cloud responses to aerosol perturbations (e.g., Xue and Feingold,
92 2006; Xue et al., 2008; Bretherton 2007; Seifert et al., 2015; Glassimire et al., 2019; Hoffman et
93 al., 2020; Chen et al., 2024; Zhang et al., 2024). However, ACI and cloud processes using
94 idealized simulations cannot be directly evaluated or constrained by observations, limiting their
95 ability to explain the divergent LWP response between the two. Additionally, many LES studies
96 are conducted with limited domain size, which cannot resolve mesoscale organization and
97 variability of cloud and precipitation, both of which have been shown to significantly affect
98 retrieved N_d -LWP relationships (e.g., Zhou and Feingold; 2023; Kokkola et al., 2025; Tian et al.,
99 2025). Finally, both aerosol and cloud fields are strongly modulated by synoptic conditions (e.g.,
100 Engström and Ekman, 2010, Zheng et al. 2011, Zheng et al. 2025). LES studies focused on a
101 small number of cases fail to capture the influence of cloud regimes and synoptic variabilities on
102 ACI, both of which determine the magnitude and sign of cloud responses to aerosol
103 perturbations.

104 The Eastern North Atlantic (ENA) region is uniquely suited to address this issue due to
105 its location at the transition between midlatitude and subtropical regimes, experiencing various
106 synoptic conditions and cloud regimes (e.g., Remillard & Tselioudis, 2015; Zheng et al., 2025).
107 In addition, the ENA region and the availability of long-term, high-quality ground-based
108 observations from the DOE Atmospheric Radiation Measurement (ARM) program make it
109 possible for process-level evaluation with the comprehensive observations. Marine boundary
110 layer (MBL) clouds in this region are frequently drizzling and sensitive to aerosol and
111 meteorological perturbations, making them ideal for studying aerosol-cloud-precipitation
112 interactions (Wood et al., 2015).

113 *The goal of this study is to evaluate key ACI processes, such as precipitation suppression
114 and evaporation-entrainment feedback, as well as precipitation treatment in a two-moment
115 scheme, through simulations approaching LES scales.* To address limitations in previous LES
116 studies, we perform a series of simulations using a nested-domain configuration to seamlessly
117 simulate the realistic circulations across different synoptic regimes, with the innermost domain
118 spanning $1^\circ \times 1^\circ$, consistent with typical GCM grid spacing and the spatial scale used in satellite
119 observations to quantify N_d -LWP relationships (introduced in Sect. 2). To investigate the
120 variation of ACI across different synoptic conditions, we simulated an ensemble of realistic
121 MBL cloud cases across three synoptic regimes, each characterized by northerly surface flow
122 over the ENA site. The classification of synoptic regimes is based on our previous study (Zheng
123 et al., 2025), in which seven major synoptic regimes were identified using both surface and mid-
124 level meteorological data. To enable a process-level evaluation of the parameterization of the
125 warm rain process, we leverage ground-based radar measurement from the DOE ARM ENA site
126 and apply a newly developed radar simulator for direct model-observation comparison.

128 **2. Data and Methodology**129 **2.1 Datasets**

130 This study adopts both satellite and ground-based observations to assess the simulated
 131 cloud, precipitation processes, and ACI processes. For satellite observations, we used cloud
 132 retrievals derived from the Spinning Enhanced Visible InfraRed Imager (SEVIRI) on the
 133 geostationary satellite Meteosat-10 and Meteosat-11 over the ENA region. The cloud retrievals
 134 are based on the methods developed by the Clouds and the Earth's Radiant Energy System
 135 (CERES) project using the Satellite ClOud and Radiation Property retrieval System (SatCORPS)
 136 algorithms (Minnis et al., 2011, 2021; Painemal et al., 2021). The SEVIRI Meteosat cloud
 137 retrieval products are pixel-level cloud retrievals produced by NASA LaRC SatCORPS group,
 138 specifically tailored to support the ARM program over the ARM ground-based observation sites.
 139 For Meteosat-10 and Meteosat-11 cloud retrievals, they have a spatial resolution of 4-km and 3-
 140 km at nadir and an hourly and half-hourly temporal resolution, respectively.

141 In this study, we used the cloud mask, cloud effective radius (r_e), cloud optical depth
 142 (τ), LWP, cloud phase, and cloud top height variables in the SEVIRI Meteosat cloud retrieval
 143 product (Minnis et al., 2011, 2021). We focus on warm boundary layer clouds with cloud top
 144 below 3km and a liquid cloud phase. The r_e and τ retrievals are based on the shortwave-infrared
 145 split window technique during the daytime. Cloud LWP is derived from r_e and τ using the
 146 equation: $LWP = \frac{4r_e\tau}{3Q_{ext}}$, where Q_{ext} represents the extinction efficiency and assumed constant of
 147 2.0. Cloud mask algorithm is consistent with the CERES Ed-4 algorithm, as described in Trepte
 148 et al. (2019), where cloudy and clear pixels are distinguished based on the calculated TOA
 149 clear-sky radiance. Cloud top height is derived from the retrieved cloud effective and top
 150 temperature, together with the boundary-layer temperature profiles and lapse rate, as described
 151 in Sun-Mack et al. (2014). Cloud N_d is retrieved based on the adiabatic assumptions for warm
 152 boundary layer clouds, based on the following equation:
 153

$$154 \quad N_d = \frac{\sqrt{5}}{2\pi k} \left(\frac{f_{ad}c_w\tau}{Q_{ext}\rho_w r_e^5} \right)^{1/2} \quad (1)$$

155 In Equation (1), k represents the ratio between the volume mean radius and r_e , and it is
 156 assumed to be constant of 0.8 for stratocumulus, f_{ad} is the adiabatic fraction, c_w is the
 157 condensation rate, Q_{ext} is the extinction coefficient, and ρ_w is the density of liquid water
 158 (Grosvenor et al., 2018).

159 To facilitate a consistent comparison, the satellite retrievals are adjusted to the same
 160 domain size as the simulation (e.g., $1^\circ \times 1^\circ$) and the pixel-level cloud retrievals are smoothed to
 161 25-km resolution to reduce impact from cloud heterogeneity and small-scale covariability on the
 162 estimated cloud susceptibility (e.g. Arola et al. 2022; Zhou and Feingold, 2023). In the context of
 163 ACI: cloud susceptibility quantifies how sensitive a cloud property responds to change in aerosol
 164 concentration or N_d . To constrain the spatial-temporal variation in meteorological conditions and
 165 cloud properties, cloud susceptibility is estimated as the regression slope between N_d and cloud
 166 properties within the $1^\circ \times 1^\circ$ domain at each time step of satellite observations. In this study, we
 167 quantify LWP and cloud fraction (CF) susceptibilities. Because of the non-linear relations
 168 between LWP and N_d , the LWP susceptibility is quantified in logarithm scale as
 169 $d\ln(LWP)/d\ln(N_d)$ (e.g., Gryspeerdt et al. 2019; Qiu et al., 2024), whereas CF susceptibility is
 170 quantified as $dCF/d\ln(N_d)$ (e.g., Kaufman et al. 2005; Chen et al., 2022; Qiu et al., 2024). Due

171 to the dependence of cloud responses on cloud regimes (e.g., Chen et al., 2014; Zhang et al.,
172 2022; Qiu et al., 2024), the estimated cloud susceptibilities are displayed in the N_d -LWP
173 parameter space as the classification of cloud states.

174 In addition to the satellite retrievals, we adopt the ground-based observation at the ARM
175 ENA site. Specifically, we use the ground-based cloud radar and lidar observations for process-
176 level evaluation of modeled precipitation processes. In this study, the radar reflectivity (Z_e) and
177 cloud boundaries are from the Active Remote Sensing of Clouds (ARSCL) value added product
178 (Clothiaux et al., 2001). To remove noise in the data, we smoothed the 4s reflectivity profiles
179 into 1-minute. Cloud top height is derived as the upper most range gate height with radar
180 reflectivity greater than the sensitivity threshold of the Ka-band zenith radar (-40 dBZ) combined
181 with the hydrometer layer top data in the ARSCL. Cloud base height is from the best-estimate
182 cloud base height variable in the ARSCL product. Thermodynamic profiles are derived from the
183 radiosonde data, which is launched at the ENA site twice daily at 0000 UTC and 1200 UTC.

184 The ground-based r_e and τ retrievals are based on the parameterization developed in
185 Dong et al. (1998), where r_e is retrieved from a radiative transfer model as described in Dong et
186 al. (1997) and parameterized as a function of cloud LWP, shortwave transmission ratio, and
187 cosine of solar zenith angle. Cloud LWP is retrieved from the brightness temperature measured
188 by the three-channel microwave radiometer (MWR3C) at 23.8, 30, and 90 GHz (Cadeddu et al.,
189 2013). The shortwave transmission ratio is calculated from the unshaded pyranometer from the
190 QCRAD product (Long and Shi, 2006), defined as the ratio between cloudy and clear-sky
191 shortwave irradiance.

192 Meteorological and thermodynamic variables are extracted from the European Center for
193 Medium-Range Weather Forecasts (ECMWF) ERA5 reanalysis data and used as the forcing for
194 the simulation. ERA5 is the fifth generation of the ECMWF reanalysis, replacing the ERA-
195 Interim reanalysis. ERA5 provides the best-estimate of the global atmosphere, land surface, and
196 ocean waves with a horizontal resolution of 31 km and an hourly output throughout (Hersbach et
197 al., 2020). Atmospheric variables are available on 137 vertical levels, ranging from 1000 hPa
198 (near surface) to 1 Pa (~80km).

199 2.2 WRF Model

200 We used the Weather Research and Forecasting (WRF) model version 4.4.2 (Skamarock
201 et al., 2021) for our simulations. In a companion study, Lee et al. (2025) used the WRF model at
202 near LES scale with interactive chemistry and aerosol schemes (WRF-Chem) and investigated
203 ACI and its feedback on both clouds and aerosols in the ENA region. As the WRF-Chem
204 simulations are 5-10 times more computationally expensive, the present study adopted the same
205 dynamical and physical configuration and conducted more experiments with prescribed aerosol
206 concentrations and realistic meteorology.

207 We employed four one-way nested domains in the model, with the domain size of
208 $27^\circ \times 27^\circ$, $9^\circ \times 9^\circ$, $3^\circ \times 3^\circ$, and $1^\circ \times 1^\circ$, and spatial resolution of 5km, 1.67 km, 0.56 km, and
209 190m, respectively, for d01, d02, d03, and d04 domain. The innermost domain (d04) exhibit a
210 domain size close to most GCM grid spacing and is consistent with the spatial scale for
211 quantification of cloud susceptibility in satellite study (e.g., Zhang et al., 2022, 2023; Qiu et al.,
212 2024). The spatial resolution of 190m is much higher than the CPMs and close to the LES scale.
213 All the analyses and evaluations in this study are based on output from the innermost domain
214 (d04). There are 75 vertical levels in the model with a model top of ~20 km, the grid spacing is
215 log-stretched with higher resolution of ~50 m near the surface and increases to ~150 m at the

height of ~1500m . As mentioned above, the initial and lateral boundary conditions for the outer domain are taken from the ERA5 reanalysis data.

The simulations are performed using the Rapid Radiative Transfer Model for Global Climate Models (RRTMG; Mlawer et al., 1997), and the Noah land surface model (Chen and Dudhia 2001). The Mellor–Yamada–Janjic (MYJ; Mellor and Yamada, 1982) planetary boundary layer (PBL) scheme and the shallow cumulus schemes (Hong and Jiang, 2018) are utilized for the outer domain (d01 and d02) only. Simulations in this study employ a two-moment Morrison microphysics scheme, which has been widely implemented in both CPMs and GCMs (Morrison et al., 2005; Morrison and Gettleman, 2008; Golaz et al., 2022). In the Morrison two-moment microphysics scheme, the DSD (ϕ) is defined as:

$$\phi(D) = N_0 D^\mu e^{-\lambda D}, \quad (2)$$

$$\eta = 0.0005714 N_d + 0.2714, \quad (3)$$

$$\mu = \frac{1}{\eta^2} - 1, \quad (4)$$

$$\lambda = \left[\frac{\pi \rho N_c \Gamma(\mu+4)}{6 q_c \Gamma(\mu+1)} \right]^{1/3}, \quad (5)$$

where D is the diameter, N_0 is the intercept parameter, μ is the shape parameter, λ is the slope parameter, η is the dispersion parameter which governs the width of the DSD (Morrison and Gettleman, 2008).

Instead of prescribing a constant cloud droplet number concentration, total aerosol number concentrations are prescribed as a constant throughout the domain with no explicit vertical variation or transport in all simulations. Aerosol activation follows the parameterization of Abdul-Razzak & Ghan (2000), with fixed assumptions for size distribution, chemical composition, aerosol type, and mixing state. The activated fraction mainly depends on the local supersaturation and updraft speed. The fixed aerosol field neglects spatial and temporal variability driven by emissions, long-range transport, wet scavenging, and CCN reactivation from evaporated raindrops. These missing processes can sustain higher CCN concentrations, suppress precipitation, and potentially exaggerate positive LWP responses.

Despite this simplification, our companion WRF-Chem study (Lee et al., 2025) shows that, even with full aerosol microphysics, wet scavenging, and aerosol reactivation, the simulated LWP responses remain broadly consistent with the results presented here, especially the positive susceptibility in precipitating clouds. This agreement suggests that the key findings of this work are robust, although the prescribed-aerosol assumption may still contribute to some of the quantitative discrepancies discussed in Section 3.

For each case, we run the model for 36 hours (except for the consecutive case on 21 July 2016, where the model was run for 60 hours), starting at 12:00 UTC of the previous day and the first 12 hours are used as model spin-up period. The time resolution of the model is 30 seconds in the outer domain for advection and physics calculation and is 1 second for the innermost domain. Model variables are output instantaneously for every 10 minutes for the innermost domain, similar as in satellite observation of snapshots.

To access the cloud responses to aerosol perturbations, we conduct three sets of simulations with different prescribed aerosol number concentration of $N=100$, 500 , and 1000 cm^{-3} for all 11 cases. Cloud susceptibility is quantified as the change in domain-mean cloud properties within the innermost domain at the same output time, comparing polluted and clean simulations (e.g. $N=1000$ vs. $N=100$, $N=500$ vs. $N=100$, and $N=1000$ vs. $N=500$). With constant and uniform aerosol concentration, the N_d -LWP relations resulting from internal cloud processes are able to be quantified within each experiment at the same output time. To minimize N_d -LWP

261 relations from cloud heterogeneity and small-scale covariability and to be consistent with the
262 quantification of cloud susceptibility in satellite observations, the pixel level model outputs are
263 smoothed to 25-km resolution and N_d -LWP relations are quantified as $d\ln(LWP)/d\ln(N_d)$
264 using the smoothed data.

265 To directly compare the WRF simulations with ground-based observations, we used the
266 Cloud Resolving Model Radar Simulator (CR-SIM; Oue et al. 2020). It is a forward-modeling
267 framework which uses consistent microphysics assumptions as in the atmospheric model (i.e.,
268 the two-moment Morrison scheme in this study) and emulates radar and lidar observables. Some
269 common radar and lidar variables include: the radar reflectivity factor at horizontal and vertical
270 polarization, depolarization ratio, Doppler velocity, spectrum width, lidar backscatter, attenuated
271 backscatter, lidar extinction coefficient, and so on. In this study, we analyzed the simulated radar
272 reflectivity factor to characterize cloud and precipitation properties.

273 To distinguish different precipitation modes and the microphysical growth processes that
274 transition clouds from non-precipitating to drizzling and raining, we investigate the vertical
275 transition from cloud to precipitation using the Contoured Frequency of Optical Depth Diagram
276 (CFODD) method (Suzuki et al., 2010) from both observations and model simulations. The
277 CFODD analysis calculates the frequency of radar reflectivity profiles as a function of in-cloud
278 optical depth (τ_d), where τ_d is calculated based on an adiabatic-condensation growth model and
279 it starts at zero at cloud top and increases downward. One benefit of the CFODD analysis is that
280 the slope of reflectivity directly relates to the droplet collection efficiency, where the slope of
281 reflectivity in the common geometric height depends on cloud water content (Suzuki et al.,
282 2010).

283 **2.3 Case Studies**

284 With the focus of MBL clouds in this study, cases are selected when both satellite and
285 ground-based observations define MBL clouds in the ENA region. For cloud type classification
286 in ground-based observations, we used the same method as in Zheng et al. (2025), where clouds
287 are classified into seven types based on the boundaries and duration of each cloud object. In this
288 study, we include both cumulus and stratocumulus clouds. Days are excluded when only shallow
289 cumulus clouds are detected to filter out clouds that are below the detectable resolution of the
290 Meteosat observations and to minimize uncertainties in the cloud microphysical retrievals from
291 the ground-based observations. We further exclude days with more than three layers of cloud in
292 the boundary layer to minimize uncertainty in cloud retrievals. Classification of cloud type in
293 Meteosat observations uses a similar method as the ground-based observations. Cloud objects are
294 defined as connected cloudy pixels, where low clouds are defined as clouds with 90th percentile
295 of cloud top height below 3km. Low clouds are further classified as stratiform clouds and
296 cumulus or broken stratiform clouds using an area threshold of 10,000 km² (Qiu and Williams,
297 2020).

298 We focus on summer months (June, July, August) in the ENA region, when this region is
299 often dominated by the Bermuda high-pressure systems and MBL clouds have the highest
300 occurrence frequency (e.g., Li et al., 2011; Mechem et al., 2018; Dong et al., 2014, 2023).
301 Previous studies found that the ARM measurements at the ENA site –located near the northern
302 shore of the Graciosa Island, the northernmost island in the Azores archipelago – can be
303 influenced by local emissions and island effects during southerly wind conditions. These impacts
304 include modification to the aerosol and CCN concentrations, boundary layer turbulence, and the
305 cloud field (e.g., Ghate et al., 2021, 2023). To minimize these influences, we focus on the three

306 synoptic regimes identified in Zheng et al. (2025) when the ENA site is influenced by northerly
307 surface wind: the high-ridge regime (characterized by a mid-tropospheric ridge and surface high-
308 pressure system), the post-trough regime, and the weak trough regime (Table S1, Figure S1).

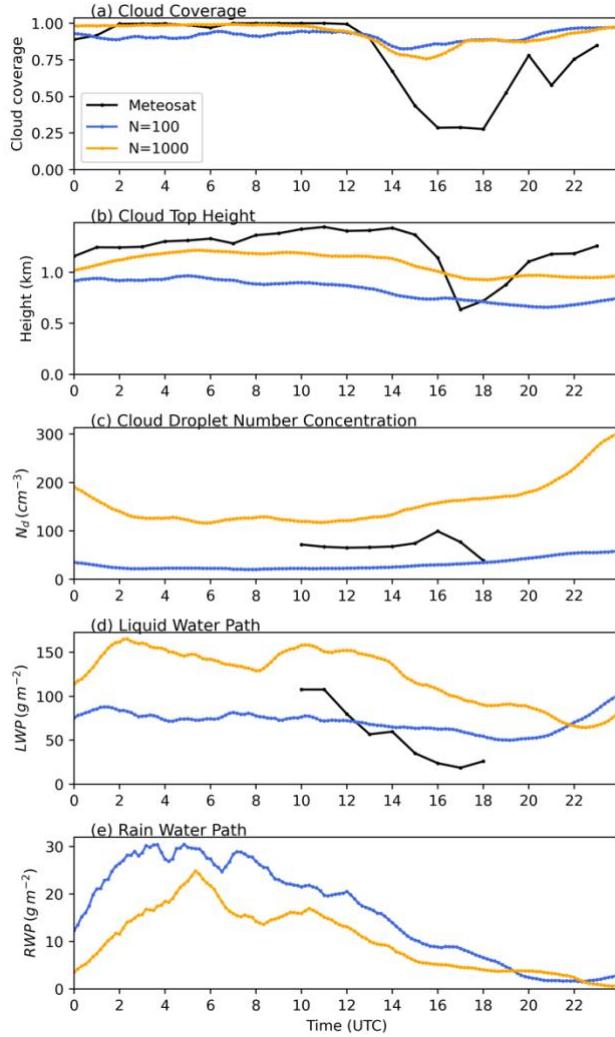
309 With the case selection criteria discussed above, there are a total 11 cases for the WRF
310 simulations, covering different cloud states and synoptic conditions. The general characteristics
311 of the 11 cases are listed in Table S1. The synoptic pattern for each case from ERA5 is shown in
312 Figure S1, the cloud fields observed from Meteosat are shown in Figure S2. WRF simulated
313 cloud fields in the N=100 and N=1000 experiments are shown in Figure S3, S4. To better
314 illustrate the large-scale cloud organization and compared with Meteosat observations, the
315 simulated LWP in domain 2 are shown. As seen in Figures S2-4, our WRF simulations well
316 capture the frontal systems and synoptic pattern of cloud fields across different cases.

317 **3 Results:**

318 **3.1 Case Study: Impacts of Aerosols on PBL Thermodynamics and Cloud Evolution**

319 Previous studies have demonstrated the distinct cloud responses to aerosol perturbations
320 between precipitating and non-precipitating regimes in both model simulations and observations
321 (e.g., Chen et al., 2014; Sato et al., 2018; Gryspeerdt et al., 2019; Fons et al., 2024; Qiu et al.,
322 2024). To explore these differences, we analyze two representative cases in our simulations: one
323 dominated by precipitating clouds and another by non-precipitating clouds, to highlight the
324 distinct interactions among aerosols, clouds, and PBL thermodynamics in the presence and
325 absence of precipitation.

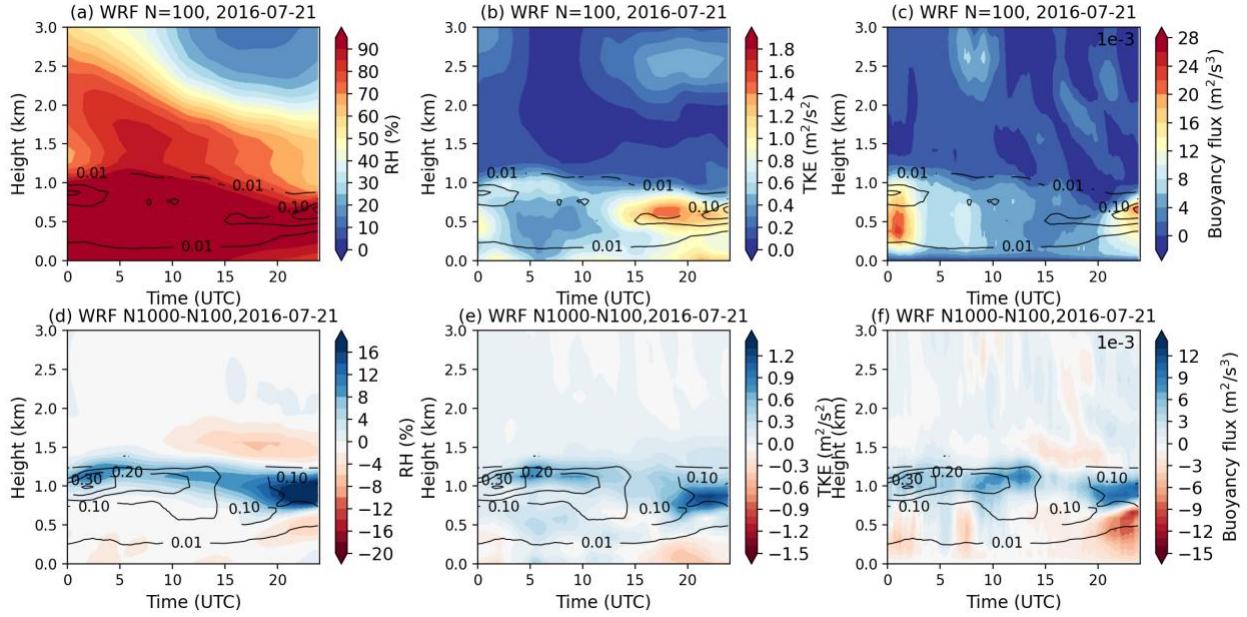
326 On 21 July 2016, the ENA site was presented by precipitating stratocumulus clouds from
327 00:00 UTC to 13:00 UTC, as seen from radar reflectivity profiles in Figure S5b. The clouds
328 dissipated from 12-18 UTC and redeveloped after 18 UTC (Figure 1a, black line). The sounding
329 observations show a moist and well-mixed boundary layer, with relative humidity (RH) near
330 saturation above cloud top (Figure S6). Our simulation captures the structure of the boundary
331 layer, with a moist layer above the cloud, and the cloud-top RH close to sounding observations
332 (99% and 96%, Figure S6c). Due to biases in the ERA5 reanalysis in representing the
333 temperature inversion, the boundary layer top in the model is ~500m lower than in sounding data
334 (Figure S6). Consequently, the simulated cloud tops are ~300–500 m lower than both satellite
335 and ground-based radar observations (Figure 1b, Figure S6).



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Figure 1. Time series of domain-averaged cloud properties from satellite observations and model simulation on 21 July 2016. (a) Cloud coverage, (b) cloud top height, (c) cloud liquid water path, and (d) rain-water path for N=100 (blue lines) and N=1000 (orange lines) experiments.

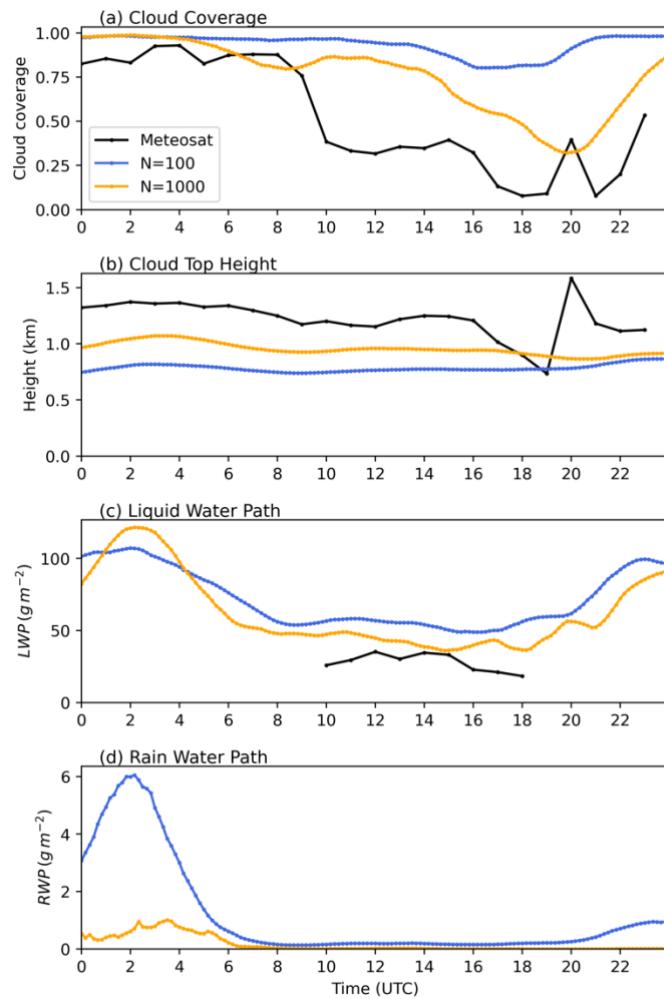
341 In the N=100 simulation, WRF model reproduces the overcast and precipitating
342 stratocumulus clouds, with a domain mean cloud cover varies between 0.90 to 0.94 from 00-13
343 UTC, which is slightly below that from Meteosat of 0.97 to 1.0 (Figure 1a, blue and black lines).
344 However, unlike observations, the simulated clouds do not dissipate after 14 UTC; both cloud
345 cover and LWP remain nearly constant throughout the day (Figures 1a, d, blue lines). With
346 increased aerosol concentration (N=1000), the simulated precipitation is suppressed (Figure 1e),
347 and the cloud layer remains overcast while deepening, accompanied by rising cloud tops and
348 increasing LWP (Figure 1b, c, orange lines). This cloud response arises from aerosol-induced
349 precipitation suppression and the corresponding changes in boundary layer processes, as
350 illustrated in Figure 2. The turbulent kinetic energy (TKE) is calculated as $\frac{1}{2}(\overline{u'^2} + \overline{v'^2} + \overline{w'^2})$,
351 with a unit of $m^2 s^{-2}$, and buoyancy flux is calculated as $g/\theta_0 \overline{w' \theta'_v}$, with a unit of $m^2 s^{-3}$.



352
353 Figure 2. Time series of domain-averaged thermodynamic profiles on 21 July 2016, for (a)
354 relative humidity, (b) turbulent kinetic energy (TKE) (unit: $m^2 s^{-2}$), (c) buoyancy flux (unit:
355 $m^2 s^{-3}$) in N=100 simulations, (d) changes in relative humidity profiles, (e) changes in TKE, (f)
356 changes in buoyancy flux between N=100 and N=1000 simulations. The black contours are
357 cloud water mixing ratio (unit: g/kg) in (a)-(c) N=100 and (d)-(f) N=1000 simulations.
358

359 In the simulations, increases in aerosol concentrations lead to higher N_d and smaller drop
360 size. As the two-moment Morrison scheme does not consider the cloud drop size in the
361 parameterization of evaporation, aerosol impacts on clouds and boundary layer occur through the
362 influence of precipitation on PBL structure. Specifically, aerosols suppress precipitation by
363 reducing autoconversion with increasing N_d , decreasing sedimentation rate and terminal velocity
364 from smaller droplets. The formation of drizzle release latent heat and reduce both entrainment
365 and the production of turbulent kinetic energy (TKE) by buoyancy; while the evaporation of
366 drizzle below cloud cool and moisten the sub-cloud layer that decrease buoyancy and TKE
367 (Stevens et al., 1998). As a result, the reduced precipitation increases both TKE and buoyancy
368 flux in the cloud layer and below cloud (Figure 2e, f). The enhanced turbulence and buoyancy
369 support vertical development of clouds, raising cloud tops and expanding the cloud layer upward
370 (Figures 1b and 2), while also increasing RH near the cloud top (Figure 2d).

371 On the second day (22 July 2016), the precipitating stratocumulus clouds transition into
372 non-precipitating thin stratus over the ENA site (Figure S7). The clouds were predominately
373 overcast from 00-09 UTC and dissipated after 10 UTC, with the domain-mean cloud coverage
374 decreasing from 0.8-0.9 to 0.1-0.2 (Figure 3a, black line). As shown in Figure S8, the boundary
375 layer was moist and well-mixed, capped by a sharp temperature inversion, and moisture
376 decreases rapidly above the inversion. WRF model reproduces the general thermodynamic
377 structure, including the inversion and moisture decline above the PBL. However, due to biases in
378 ERA5 thermodynamic profiles, the simulated PBL top is about 700m lower than observed
379 (Figure S8). Additionally, WRF model fails to capture the rapid decrease of moisture above
380 cloud top, resulting in a more humid layer above cloud with cloud-top RH of 87% in the model,
381 compared to 62% in sounding observation (Figure S8c).

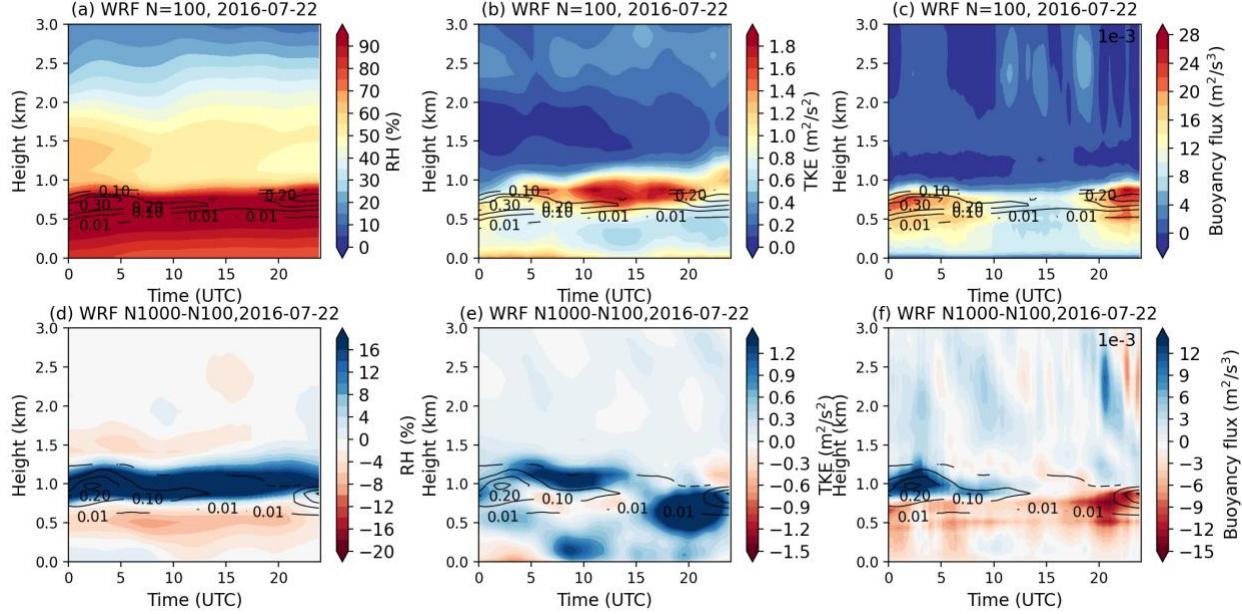


384 Figure 3. Time series of domain-averaged cloud properties from observations and model
 385 simulation on 22 July 2016. (a) Cloud coverage, (b) cloud top height, (c) cloud liquid water path,
 386 and (d) rain-water path for $N=100$ (blue lines) and $N=1000$ (orange lines) experiments.
 387

388 In the $N=100$ simulation, the simulated stratocumulus cloud generates light precipitation
 389 from 00-06 UTC, then it transitions to a non-precipitating thin cloud layer after 06 UTC (Figure
 390 3d, blue line). However, the cloud does not dissipate in the model. Domain-mean cloud cover
 391 remains between 0.85 to 0.95 throughout the day, and the simulated LWP is nearly twice that
 392 retrieved from Meteosat (Figure 3a and 3c, blue lines). When aerosol concentrations are
 393 increased to $N=1000$, clouds dissipate from 14-20 UTC, with a decreasing domain-mean cloud
 394 cover and becoming more consistent with observations (Figure 3a, orange line). Meanwhile,
 395 cloud tops rise slightly with increasing aerosol. The cloud dissipation reflects a net effect of
 396 aerosol induced changes in condensation, evaporation, turbulence, and buoyancy, as shown in
 397 Figure 4.

398 During the early phase (00–06 UTC), increased aerosol loading suppresses drizzle,
 399 leading to an increase in LWP and a decrease in RWP (Figure 3c, d). Similar as the first case, the
 400 suppressed precipitation enhances turbulence and increases TKE in and below cloud (Figure 4e),
 401 lift the cloud top, and lead to an increase in RH near cloud top (Figure 4d). Meanwhile, the free

402 tropospheric air above cloud top is relatively drier compared to the first case (Figure 4a). The
 403 increased turbulence and raised cloud top entrain dry air into the cloud and enhances
 404 evaporation. After 6 UTC, as clouds become non-precipitating in the N=100 experiment, the
 405 decrease of cloud water from evaporation starts to dominate the increase from precipitation
 406 suppression and lead to a net decrease in LWP. Reduced buoyancy weakens the upward transport
 407 of moisture and energy from the sub-cloud layer, further contributing to cloud dissipation. As a
 408 result, both cloud cover and LWP decrease with increasing aerosol. (Figure 3a, c).



409
 410 Figure 4. Time series of domain-averaged thermodynamic profiles on 22 July 2016, for (a)
 411 relative humidity, (b) turbulent kinetic energy (TKE) (unit: $m^2 s^{-2}$), (c) buoyancy flux (unit:
 412 $m^2 s^{-3}$) in N=100 simulations, (d) changes in relative humidity profiles, (e) changes in TKE,
 413 (f) changes in buoyancy flux between N=100 and N=1000 simulations. The black contours are
 414 cloud water mixing ratio (unit: g/kg) in (a)-(c) N=100 and (d)-(f) N=1000 simulations.
 415

416 The absence of afternoon cloud dissipation in WRF simulations are likely associated with
 417 model biases in the thermodynamic structure inherited from ERA5. For example, on 21 July 2016,
 418 ARM sounding observations show a pronounced decrease in specific humidity and relative
 419 humidity above the PBL between 14 and 20 UTC (figures not shown). This sharp drying leads to
 420 cloud erosion in the observations. However, WRF simulations or ERA5 reanalysis produces only a
 421 gradual reduction in moisture from 00 to 20 UTC (Figure 2a), maintaining a moist layer above cloud
 422 top and prevent cloud breakup. On 22 July 2016, the model reproduces the moisture gradient above
 423 PBL with a warm and dry layer above, the lifted cloud top in the N=1000 simulation entrain dry air
 424 into cloud system and dissipate clouds in the afternoon (Figure 3a). On days when ERA5 accurately
 425 capture the observed moisture decrease above PBL (e.g., 25 and 28 July 2016), the model
 426 reproduces both the dissipation and evening redevelopment of clouds seen in Meteosat data
 427 (figures not shown). This indicates that the diurnal evolution of MBL clouds is highly sensitive
 428 to the representation of diurnal variation in moisture as well as the moisture gradients near the
 429 inversion.

430 The prescribed, vertically uniform aerosol concentration further reinforces cloud
 431 persistence by maintaining elevated CCN levels and suppressing drizzle formation. The lack of

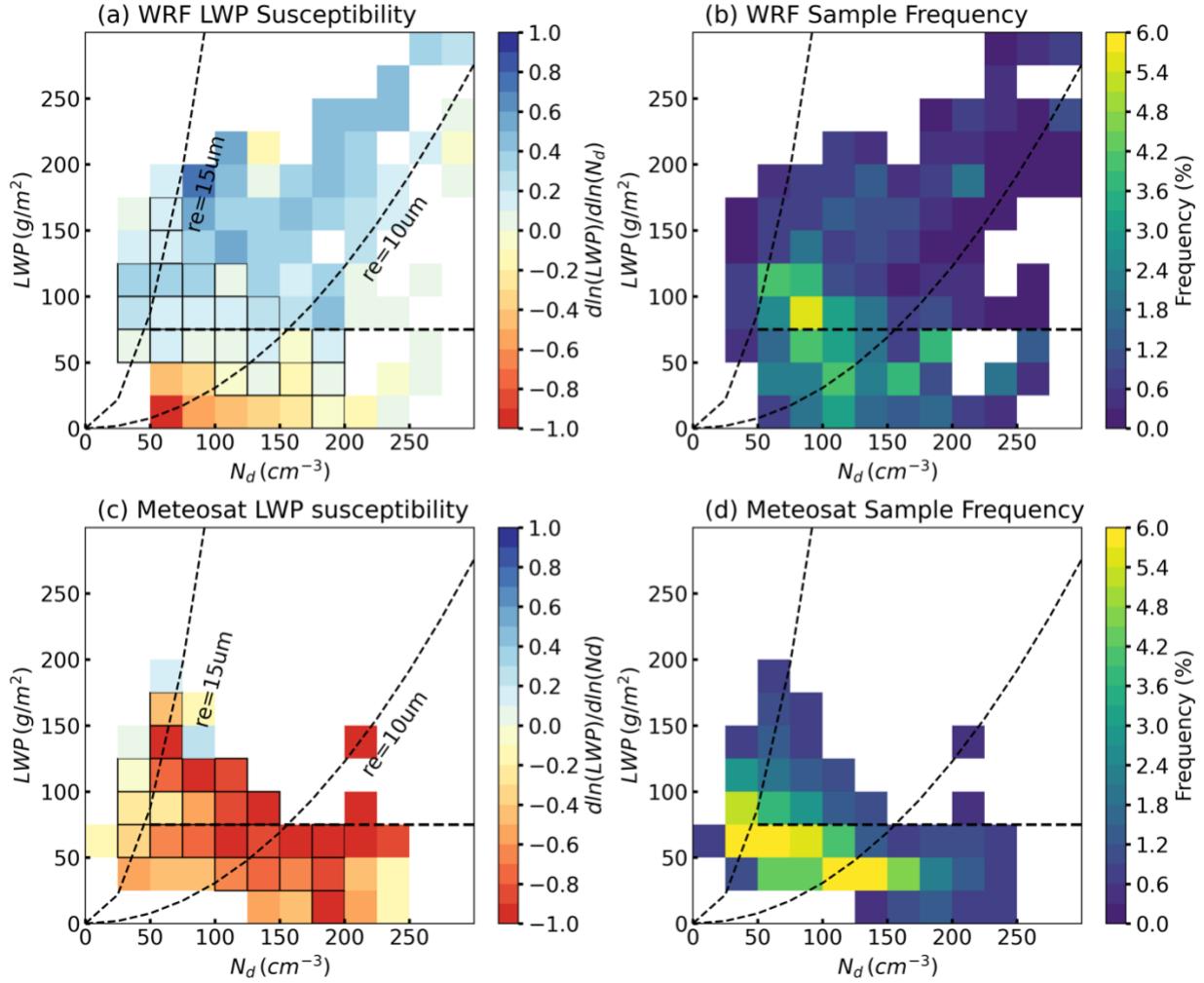
432 precipitation scavenging prevents cloud-base evaporative cooling and inhibits decoupling, both
433 of which would otherwise promote afternoon cloud breakup. The implications of thermodynamic
434 biases (e.g. the moist layer above cloud top and the underestimated PBL height) for the estimated
435 ACI are discussed in detail in Section 3.3.2

436 In a nutshell, precipitating and non-precipitating clouds react differently to aerosol
437 perturbations in our simulations. For precipitating clouds, aerosols increase LWP through
438 precipitation suppression and support vertical development of cloud through the impact of
439 precipitation on PBL dynamic and thermodynamics. For the non-precipitating case, PBL air is
440 drier compared to the first case, the enhanced turbulence and entrainment of dry air above leads
441 to evaporation and reduced buoyancy. The reduced buoyancy stabilizes PBL and decays the
442 cloud layer.

443 **3.2 Evaluation of LWP Susceptibility Across Cloud States and Synoptic Conditions**

444 The two cases in Section 3.1 demonstrate the impact of different cloud states and PBL
445 thermodynamics on cloud responses to aerosol perturbations. In order to evaluate the ACI
446 process across all simulated cloud states, we composite the cloud fields from all 11 cases and all
447 three aerosol concentrations (e.g. N=1000 vs. N=100, N=500 vs. N=100, and N=1000 vs.
448 N=500) to estimate the mean LWP response, and compare it with satellite retrievals, as shown in
449 Figure 5. More specifically, LWP susceptibility in WRF simulations is defined as the change in
450 domain mean cloud properties as $d\ln(LWP)/d\ln(N_d)$ between polluted and clean simulations
451 for each 10-minutely model output. To be consistent with satellite retrievals, we focus on
452 daytime with solar zenith angle less than 65°. Lastly, we use the LWP- N_d parameter space to
453 represent different cloud states. (Qiu et al., 2024).

454 Based on the relationships between r_e , LWP, and N_d in the satellite retrievals (e.g.,
455 $LWP = \frac{4r_e\tau}{3Q_{ext}}$, $N_d = \frac{\sqrt{5}}{2\pi k} \left(\frac{f_{ad}c_w\tau}{Q_{ext}\rho_w r_e^5} \right)^{1/2}$), $r_e = 15$ isolines is marked in the LWP- N_d parameter
456 space as an commonly used indicator of precipitation likelihood in the satellite retrieval (e.g.,
457 Gryspeerdt et al., 2019; Toll et al., 2019; Zhang et al., 2022; Qiu et al., 2024). Based on the
458 distinct LWP, cloud albedo and CF susceptibilities between cloud states, MBL clouds are
459 classified into three states: the precipitating clouds ($r_e > 15 \mu m$), the non-precipitating thick
460 clouds ($r_e < 15 \mu m$, $LWP > 75 \text{ gm}^{-2}$), and the non-precipitating thin clouds ($r_e < 15 \mu m$, $LWP <$
461 75 gm^{-2}) (Qiu et al., 2024). To be consistent with observational reference, the WRF simulated
462 cloud states are classified using the same definition. Similar to warm MBL clouds in
463 observations (e.g. Qiu et al., 2024), LWP responses to aerosol perturbation in model simulations
464 show clear dependence on cloud state (Figure 5a).



465
466 Figure 5. Mean liquid water path (LWP) susceptibility from (a) (b) WRF simulations and (c) (d)
467 Meteosat cloud retrievals during the daytime. (a) (c) cloud LWP susceptibility $d\ln(LWP)/$
468 $d\ln(N_d)$, (b) (d) frequency of occurrence of sample in each bin. The dashed lines indicate $r_e = 15$
469 μm , $r_e = 10 \mu m$, and $LWP = 75 \text{ gm}^{-2}$, as r_e thresholds for precipitation (precipitating clouds
470 located to the left of the line), and for thick clouds (with $LWP > 75 \text{ gm}^{-2}$), respectively. Black-
471 outlined bins denote cases where the WRF and Meteosat LWP susceptibilities differ significantly
472 ($p < 0.05$) based on a Welch's t-test.
473

474 For precipitating clouds ($r_e > 15 \mu m$), LWP slightly increases with N_d , with a mean
475 susceptibility of +0.15. The increase of LWP agrees with the precipitation suppression
476 mechanism. Meanwhile, there are only 4% of clouds in model simulations locate to the left of the
477 $r_e = 15 \mu m$ isotherm with small N_d , even with aerosol concentration set to 100 cm^{-3} (Figure 5b).
478 The non-precipitating thick clouds ($r_e < 15 \mu m$, $LWP > 75 \text{ gm}^{-2}$) is the dominant cloud state in
479 model simulation, with a total frequency of occurrence of 49%. Different from the evaporation-
480 entrainment feedback mechanism, LWP largely increases in the model with increasing aerosols,
481 with a mean susceptibility of +0.32. For non-precipitating thin clouds ($r_e < 15 \mu m$, $LWP < 75$
482 gm^{-2}), LWP decreases with aerosol perturbations with a mean of -0.14, which is consistent
483 with the second case shown in the previous section.

484 To evaluate model performance, we estimated LWP susceptibility from satellite retrievals
485 within the same domain and for the same 11 cases as the model simulations (Figures 5 c, d).
486 Specifically, LWP susceptibility was quantified as the regression slope between LWP and N_d
487 within the $1^\circ \times 1^\circ$ domain at each time step of satellite observations. For precipitating clouds,
488 LWP slightly decreases with increasing N_d in satellite data, consistent with the four-year
489 climatological mean feature in the ENA region reported in our previous study (Qiu et al., 2024).
490 This decrease of LWP with increasing N_d is likely associated with the depletion of LWP through
491 sedimentation–evaporation–entrainment feedbacks, which outweigh the increase of LWP from
492 precipitation suppression. In contrast, in model simulations, the lack of realistic evaporation–
493 entrainment feedback results in LWP increasing primarily through precipitation suppression. The
494 simulated LWP susceptibilities are significantly different with satellite observations at 95%
495 confidence level for most precipitating clouds (Figure 5a).

496 For non-precipitating thin clouds, the simulated decrease in LWP with increasing aerosol
497 concentration agrees in sign with satellite observations. However, the magnitude of this decrease
498 is weaker, and the simulated susceptibilities remain significantly different from satellite
499 estimates at 95% confidence level for most bins (Figure 5a, c). This model behavior contrast
500 with most GCM and coarse CPM studies, which often simulate an increase of LWP for non-
501 precipitating clouds (e.g., Fons et al., 2024; Christensen et al, 2024; Mülmenstädt et al., 2024;).
502 The improved representation in our high-resolution simulations arise from better-resolve PBL
503 turbulence and thermodynamics, which enhance the entrainment of dry air, accelerates
504 evaporation, reduces buoyancy, and promotes dissipation of the cloud system.

505 In contrast, for non-precipitating thick clouds, the model and observations diverge
506 substantially. In satellite observations, LWP decreases most strongly for this cloud state, with a
507 mean LWP susceptibility of -0.69 (Figure 5c). This observational estimate is consistent with the
508 climatological mean derived from four years of Meteosat data over the ENA region (Qiu et al.,
509 2024). In the model, however, LWP increases most strongly with increasing N_d for this cloud
510 state. Moreover, compared with satellite retrievals, model simulates substantially larger
511 population of polluted thick clouds characterized by high N_d and LWP. For example, non-
512 precipitating thick clouds are the dominant cloud state in the model, accounting for 49% of total
513 cloud occurrence (Figure 5b), whereas they are the least frequent in observations, at only 15.7%
514 (Figure 5d). Meanwhile, only 4% of simulated clouds fall into the precipitating cloud regime
515 with $N_d < 50$, compared to a 22.2% in the satellite observations

516 The overall overestimation of N_d likely arises from the prescribed aerosol concentration
517 in the model configuration, combined with the absence of precipitation scavenging. For
518 reference, the mean aerosol concentration over the ENA region during summer is approximately
519 400 cm^{-3} (e.g., Zhang et al., 2021; Wang et al., 2021; Zheng et al., 2024). The model's
520 overestimation of LWP may stem from its excessively positive LWP susceptibility in thick
521 clouds. As shown in Figure S9, simulated LWP in the N=100 experiment agrees reasonably well
522 with the Meteosat retrievals, with a mean value about 10% lower than observed. However, in the
523 N=500 and N=1000 simulations, the strong positive LWP susceptibility leads to increases in
524 LWP for clouds with $\text{LWP} > 75 \text{ gm}^{-2}$, resulting in mean values 30% and 40% higher than
525 Meteosat retrievals, respectively.

526 To further examine whether these discrepancies depend on large-scale meteorological
527 conditions, we assessed LWP susceptibility across different synoptic regimes. Because only one
528 case is available for the “weak-trough” regime (Table S1), our comparison focuses on the “high-
529 ridge” and the “post-trough” regimes (Figure S10). The “high-ridge” regime shows a higher

530 occurrence of non-precipitating thin clouds than the “post-trough” regime, with total frequencies
531 of 49% and 40%, respectively (Figures S10b, d). This more frequent non-precipitating thin cloud
532 in the model is consistent with our previous study based on six years of ground-based
533 observations at the ARM ENA site, which revealed that the “high-ridge” regime favors single-
534 layer stratocumulus clouds with shallower cloud depth and smaller LWP compared to the “post-
535 trough” regime (Zheng et al., 2025).

536 In addition, non-precipitating thin clouds in the “high-ridge” regime exhibit more
537 negative LWP susceptibilities than clouds with similar LWP and N_d in the “post-trough” regime.
538 This difference in LWP susceptibility is associated with the colder and drier air above clouds
539 under subsidence in the “high-ridge” regime, which facilitates cloud dissipation, as also
540 demonstrated in the case study. Furthermore, non-precipitating or lightly drizzling thick clouds
541 in both synoptic regimes manifest strong positive LWP susceptibilities, suggesting that the
542 model-observation discrepancy for this cloud state persist regardless of synoptic conditions and
543 therefore warrants further investigation. In summary, the mean LWP susceptibility from our
544 simulations were evaluated against satellite retrievals in the LWP- N_d parameter space across
545 different cloud states and synoptic conditions for a comprehensive comparison. The simulations
546 reproduce the observed decrease in LWP for non-precipitating thin clouds, although with weaker
547 magnitudes. For precipitating clouds, the model predicts a slight increase in LWP instead of the
548 weak decrease seen in satellite observations, reflecting the limited representation of evaporation-
549 entrainment feedback in the model. Large discrepancies remain for non-precipitating or lightly
550 drizzling thick clouds, where the model simulates too many polluted thick clouds and yields an
551 opposite (positive) LWP response compared to the strongly negative satellite signal.

552 In addition, the model-observation discrepancy persists across all synoptic regimes,
553 suggesting that they originate from the model’s representation of cloud microphysics,
554 precipitation, and aerosol-cloud coupling rather than from large-scale meteorological variability.
555 The consistency of these modeled LWP response, in agreement with previous LES studies of
556 similar cloud regimes (e.g., Wang et al., 2020; Lee et al., 2025), further motives the central focus
557 of the next section: diagnosing the physical mechanisms driving these biases. We show that three
558 leading factors dominate the discrepancy: excessive precipitation production in thick clouds, a
559 moist bias above cloud top, and satellite retrieved N_d -LWP relationships contaminated by
560 internal cloud processes.

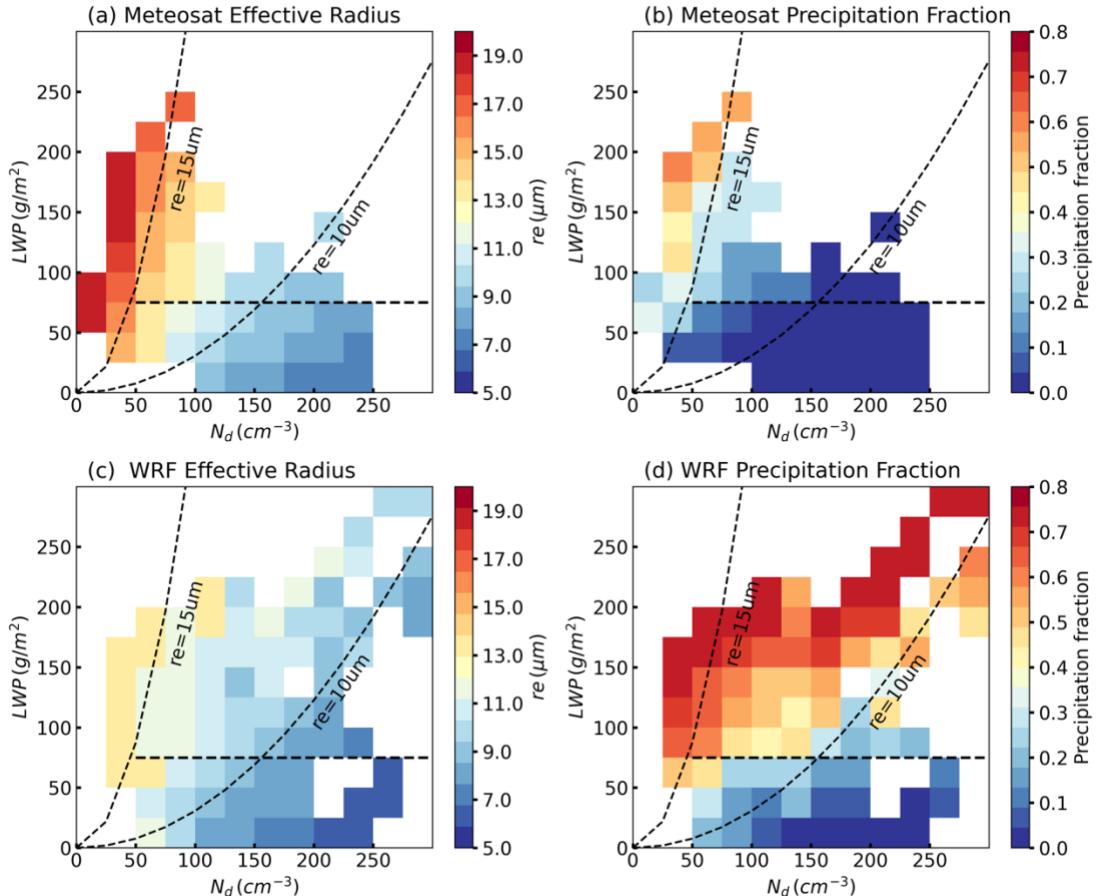
561 **3.3 Causes of Satellite–Model Discrepancies in LWP Susceptibility**

562 The satellite–model differences highlighted above point to systematic biases in how the
563 model represents cloud microphysics, precipitation processes, and entrainment pathways. In this
564 section, we diagnose the physical mechanisms driving these discrepancies, beginning with the
565 model’s precipitation efficiency.

566 **3.3.1 Precipitation Efficiency**

567 A long-standing challenge in numerical models is the tendency to produce precipitation
568 too frequently and too lightly (Sun et al., 2006; Stephens et al., 2010). To assess the modeled
569 precipitation efficiency with observation, Figure 6 shows the mean cloud properties from
570 Meteosat observations, and from WRF simulations for the 11 cases combining all three aerosol
571 concentrations (N=100, 500, and 1000). As satellite retrieves r_e near cloud top, we used r_e at
572 ~100 m below cloud top in the simulation, which approximate $\tau = 2$ from cloud top for marine

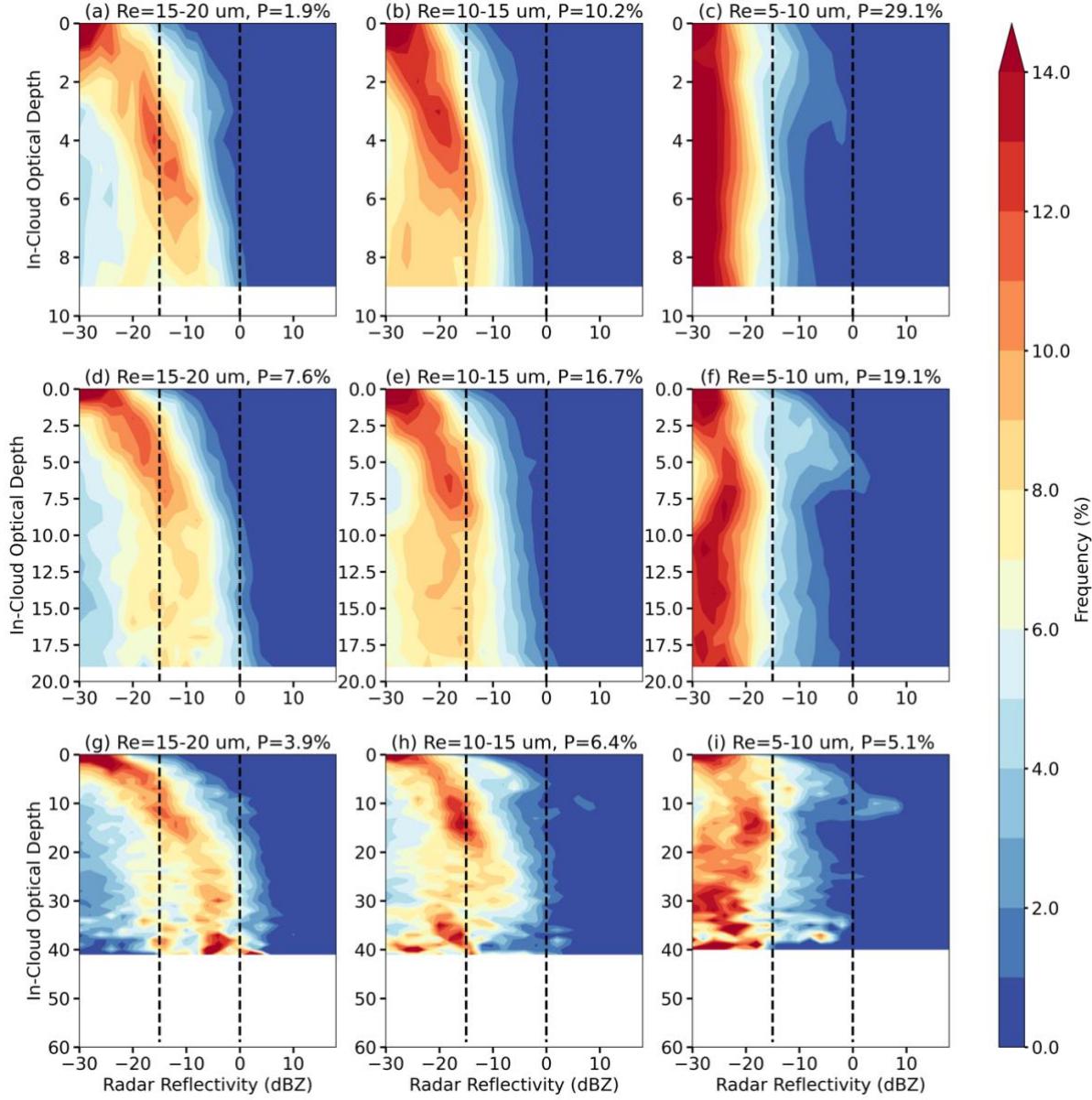
573 stratocumulus. The modeled pixel-level precipitation fraction is calculated as the area fraction of
 574 cloudy pixels with the column maximum radar reflectivity (Z_{max}) greater than -15 dBZ at
 575 each model output time (Haynes et al., 2009; Suzuki et al., 2015; Jing et al., 2017). Modeled
 576 radar reflectivity is from the radar simulator (CR-SIM), as discussed in the methodology. The
 577 precipitation fraction in Meteosat is calculated as the area fraction of clouds with $r_e > 15 \mu\text{m}$.
 578 Qiu et al. (2024) evaluated different effective radius thresholds and rain rate thresholds in
 579 satellite retrievals using precipitation masks derived from ground-based radar reflectivity at the
 580 ENA site, and concluded that the $r_e > 15 \mu\text{m}$ threshold showed the best agreement with
 581 observations.



582
 583 Figure 6. Mean cloud properties from (a), (b) Meteosat retrievals and (c), (d) WRF simulations
 584 during the daytime. (a), (c) effective radius, (b), (d) pixel-level precipitation fraction. The dashed
 585 lines indicate $r_e = 15 \mu\text{m}$, $r_e = 10 \mu\text{m}$, and $\text{LWP} = 75 \text{ gm}^{-2}$, as r_e thresholds for precipitation
 586 (precipitating clouds located to the left of the line), and for thick clouds (with $\text{LWP} > 75 \text{ gm}^{-2}$),
 587 respectively.

588 As shown in Figures 6a, c, the modeled r_e is $\sim 1\text{-}3 \mu\text{m}$ smaller than satellite retrievals for
 589 a similar cloud condition. Additionally, compared to observation, model generates precipitation
 590 too often at smaller drop size with $r_e > 10 \mu\text{m}$ and at higher N_d concentration (Figures 6b, d,
 591 $r_e = 10 \mu\text{m}$ dashed line). The large discrepancy in LWP susceptibility for thick clouds between
 592 the 10 and 15 μm isolines is likely linked to model bias in precipitation efficiency. To further
 593 investigate the model bias of excessive rain at smaller drop size and the positive LWP responses

594 to aerosol perturbations, we compared the modeled radar reflectivity profiles from the radar
 595 simulator with ARM observations using the CFODD framework. Based on the relationship
 596 between Z_e and the droplet collection efficiency (E_c), the vertical slope of Z_e as a function of in-
 597 cloud optical depth (τ_d) is directly linked to E_c , a steeper slope indicates a larger E_c (Suzuki et
 598 al., 2010).



599
 600 Figure 7. Frequency of radar reflectivity as a function of in-cloud optical depth (τ_d) for ARM
 601 ground-based observations during the daytime. Different rows are for different ranges of optical
 602 depth (τ): (a)-(c) clouds with $\tau < 10$, (d)-(f) clouds with $10 < \tau < 20$, (g)-(i) clouds with $\tau > 20$.
 603 Different columns are for different ranges of effective radius (r_e). The left, middle, and right
 604 columns are for $15 - 20 \mu\text{m}$, $10 - 15 \mu\text{m}$, and $5 - 10 \mu\text{m}$, respectively. The black dashed lines
 605 in each panel denote -15 dBZ and 0 dBZ , as thresholds of drizzle and rain, respectively. The
 606 percentage of sample (P) for each subgroup is denoted in the figure, with a total sample of
 607 91,737.

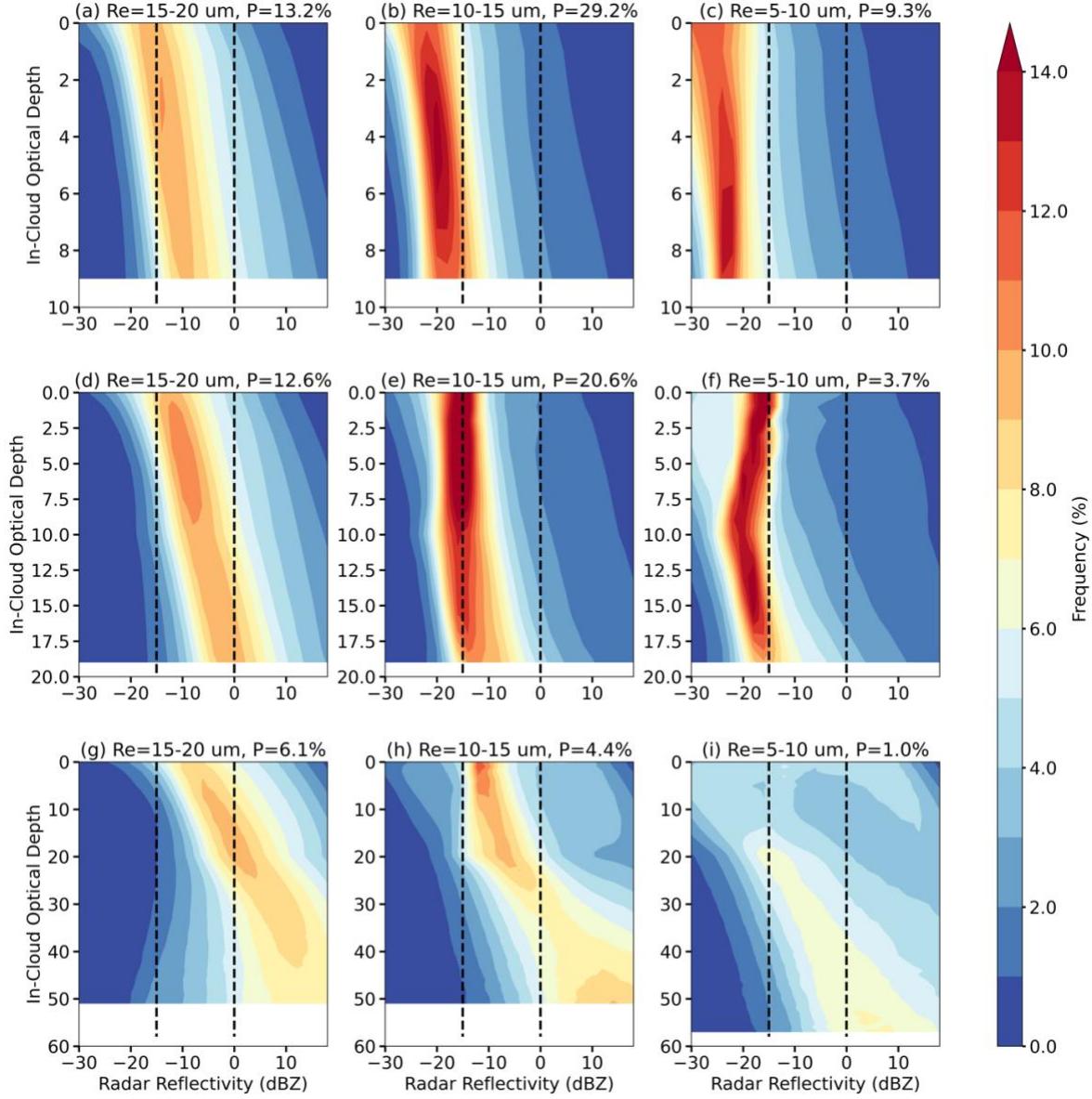
608 Ground-based radar reflectivity profiles and cloud retrievals at the ARM ENA site are
609 used as the ground truth. To reduce noise, radar reflectivity profiles and cloud boundary data are
610 smoothed to a 1-minute resolution. To increase the sample size, we analyzed the climate-mean
611 radar reflectivity profiles of stratocumulus and cumulus clouds observed during the summer
612 months (June to August) from 2016 to 2021, comprising a total of 91,737 profiles. Radar
613 reflectivity profiles derived from the selected 11 cases exhibit consistent characteristics (figure
614 not shown). To better distinguish microphysical processes such as autoconversion and accretion
615 from dynamical processes such as updraft, clouds are further categorized by both r_e and τ ranges.
616 MBL clouds are classified as non-precipitating clouds, drizzle, and rain using a reflectivity
617 threshold of $Z_e < -15$ dBZ, -15 dBZ $< Z_e < 0$ dBZ, and $Z_e > 0$ dBZ, respectively, as denoted
618 by black dashed lines in Figures 7 (Haynes et al., 2009; Suzuki et al., 2015; Jing et al., 2017).

619 Applying the same cloud state classification as in the satellite observations (e.g., $r_e >$
620 $15 \mu\text{m}$ for precipitating clouds and $\text{LWP} > 75 \text{ gm}^{-2}$ for thick clouds), the total frequency of
621 occurrence of precipitating, non-precipitating thin, and non-precipitating thick clouds are 30.7%,
622 46.3%, and 23.0%, based on six-year of ARM observations. These frequencies are consistent
623 with those derived from satellite data for the 11 cases (22.2%, 55.6%, and 22.2%, respectively;
624 Figure 5d). Therefore, the selected cases in this study are representative of the typical
625 distribution of MBL cloud types in the ENA region during summer.

626 As shown in the first column of Figure 7, in clean environment with $r_e > 15 \mu\text{m}$, the
627 observed MBL clouds start to drizzle with $Z_e > -15$ dBZ even in the thinnest category (Figure
628 7a), of which the cloud top is mostly non-precipitating ($Z_e < -25$ dBZ). Cloud drops rapidly
629 grow from cloud top downward and initiate drizzle at $\sim 4\text{-}6$ optical depth into the cloud.
630 However, most observed MBL clouds, even for the thickest category (Figure 7g), remain
631 drizzling rather than raining as most of the radar reflectivity is lower than 0 dBZ.

632 Figures 7b, e, h represent clouds with observed r_e of $10 - 15 \mu\text{m}$, indicating an increase
633 in N_d compared with clouds with similar τ and $r_e > 15 \mu\text{m}$ (Figures 7a, d, g). Precipitation in
634 these clouds is suppressed as the Z_e is mostly less than -15 dBZ in thin clouds ($\tau < 10$, Figures
635 7b). Thick clouds produce drizzle at $\sim \tau_d > 20$ and Z_e slightly decrease at cloud base, likely due
636 to mixing and evaporation (Figure 7h). When r_e decreases to below $10 \mu\text{m}$ (Figures 7c, f, i), Z_e
637 further reduces to around -20 to -30 dBZ throughout the cloud layer, indicating that precipitation
638 is further suppressed. The precipitation suppression effect is shown not only by the peak
639 frequency of Z_e , but also the slope of Z_e , which indicates the droplet collection efficiency as
640 discussed above. As seen in Figure 7, for clouds with similar thickness, the slope of Z_e decreases
641 with decreasing r_e , which reflects a weaker collision coalescence and accretion processes with
642 higher N_d and smaller cloud drops.

643 In thick clouds with $r_e < 10 \mu\text{m}$ (Figure 7i), most radar reflectivity remains below -25
644 dBZ in the lower cloud layer, while reflectivity slightly increases toward cloud top in the region
645 corresponding to $\sim 10\text{-}20$ optical depth into the cloud. Reflectivity then decreases again toward
646 cloud top. This vertical pattern is consistent with the structure of marine clouds reported in
647 Suzuki et al. (2010). The observed decrease of reflectivity near cloud top may be attributed to
648 entrainment and evaporation, or to the accretion process involving large droplets falling
649 downward, as indicated by localized reflectivity peaks exceeding -15 dBZ (Figure 7i).
650 Meanwhile, in clouds with small drop sizes, cloud deepening or dynamical processes have little
651 effect on precipitation based on observations.



652
653 Figure 8. Frequency of radar reflectivity as a function of in-cloud optical depth (τ_d) for WRF
654 N=100 simulation. Different rows are for different ranges of optical depth (τ): (a)-(c) clouds with
655 $\tau < 10$, (d)-(f) clouds with $10 < \tau < 20$, (g)-(i) clouds with $\tau > 20$. Different columns are for
656 different ranges of effective radius (r_e). The left, middle, and right columns are for $15 - 20 \mu\text{m}$,
657 $10 - 15 \mu\text{m}$, and $5 - 10 \mu\text{m}$, respectively. The black dashed lines in each panel denote -15
658 dBZ and 0 dBZ, as thresholds of drizzle and rain, respectively. The percentage of sample (P) for
659 each subgroup is denoted in the figure.

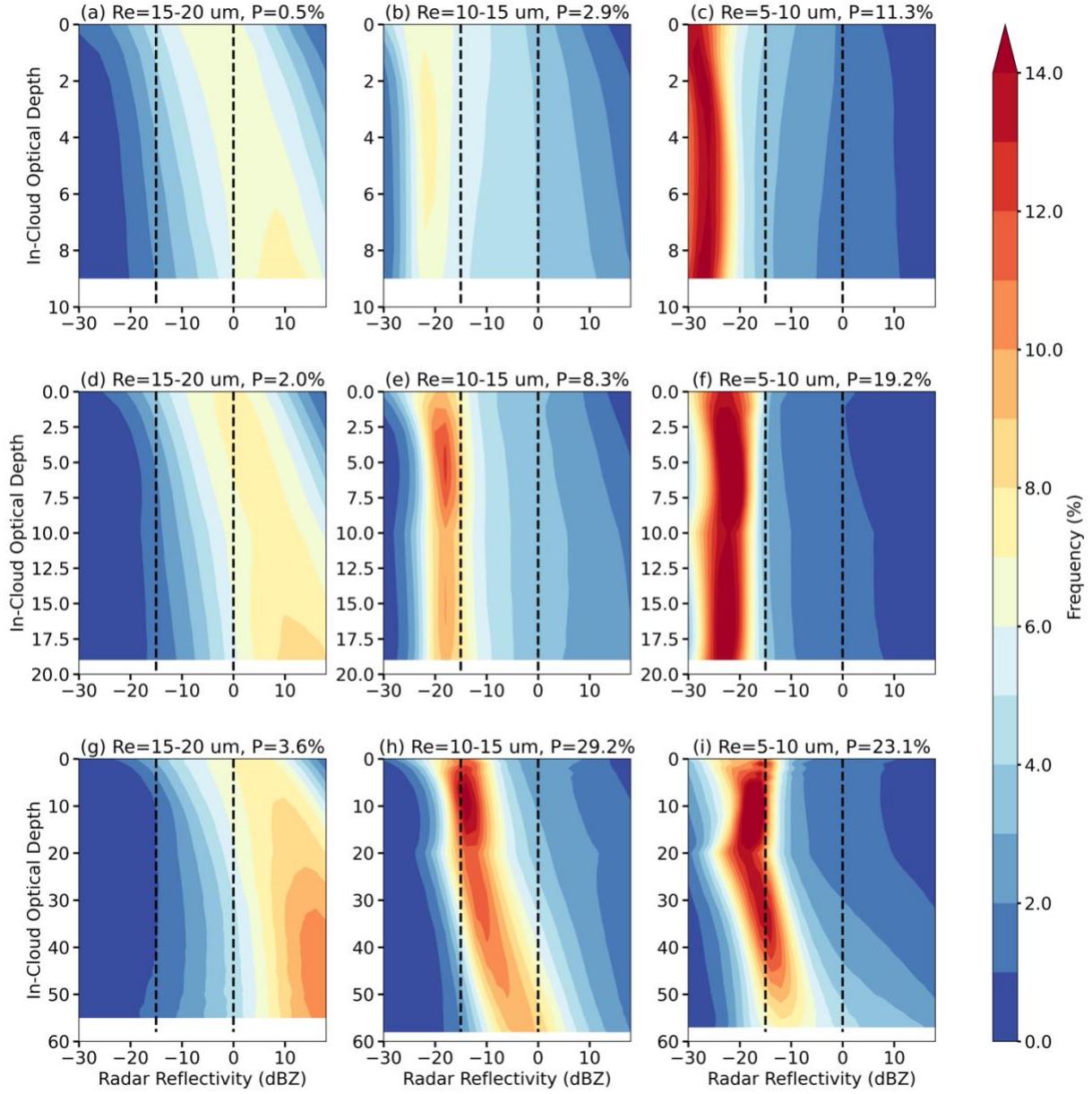
660
661 Compared to the “ground truth”, our model simulations reasonably identify the non-
662 precipitating regime in clouds with $r_e < 10 \mu\text{m}$ and $\tau < 20$, when cloud drops are too small for
663 efficient collision coalescence (Figures 8c, f). Additionally, drizzle initiates at the same r_e and τ
664 ranges as in observations: for example, the maximum frequency of Z_e exceeds -15 dBZ in thin
665 clouds with $r_e > 15 \mu\text{m}$ and $\tau < 10$ (Figure 8a) or in thick clouds with $r_e = 10 - 15 \mu\text{m}$ and $\tau =$

666 10 – 20 (Figure 8e). This result is different from GCM or GCPM where models hardly simulate
667 any non-precipitating clouds, or drizzle initiate too early in the cloud (e.g. 5-10 optical depth;
668 Jing et al. 2017, 2019; Michibata and Suzuki, 2020). The better resolved non-precipitating
669 regime as well as the transition from non-precipitating cloud to drizzle process in our simulations
670 reveal the importance of model resolution to better simulate precipitation.

671 On the other hand, model overestimates precipitation in both intensity and the frequency
672 of occurrence in optical thick with $\tau > 20$, the simulations produce rain with peak Z_e exceeding 0
673 dBZ in all size ranges, even in clouds with $r_e < 10 \mu\text{m}$ (Figure 8 g-i). Furthermore, precipitation
674 initiates too early near cloud top: all precipitating clouds in the model start to drizzle or even rain
675 at cloud top (Figures 8a, d, e, g, h, i). Based on the features shown in the CFODD analysis, the
676 overestimation of precipitation could be attributed to the following four aspects in the
677 parameterization.

678 First, the overestimation of reflectivity at cloud top indicates that autoconversion is
679 activated too early in clouds near the top. With the same aerosol concentration, clouds with less
680 activated N_d exhibit larger r_e (Figure 6c). As the autoconversion rate scaled non-linearly with N_d
681 (e.g. $\frac{\partial q_c}{\partial t} = 1350q_c^{2.47}N_d^{-1.79}$), clouds with larger drop size (e.g. $\sim 15\text{--}20 \mu\text{m}$) have smaller N_d ,
682 and therefore exhibit larger autoconversion rate. Second, the overestimation of reflectivity near
683 cloud top could be due to underestimation of entrainment rate or evaporation rate from the moist
684 layer above. As seen in Figure 8, the simulated Z_e does not decrease towards cloud top or cloud
685 base as in the observations, which indicates an underestimation of entrainment and evaporation.
686 Third, the overproduction of rain in the model indicates an overestimation of the accretion
687 process. In the Morrison scheme, accretion is parameterized as a function of cloud water and
688 rainwater content; thus, when autoconversion is triggered too early, accretion also initiates too
689 early. This bias is amplified in thick clouds, which have greater liquid water content and longer
690 path for droplet collection (Figures 8 g-i). For thick clouds with small drop size (Figure 8i), they
691 remain non-precipitating at the cloud top, indicating that autoconversion is appropriately
692 suppressed by small drop size. However, these clouds still produce rain, suggesting an
693 overestimation of accretion. Lastly, the excessive rain production in thick clouds also point to an
694 overly broad parameterized drop size distribution (DSD), which lead to an early initiation of
695 autoconversion at cloud top and rain formation in clouds with large r_e .

696 Overall, in $N=100$ simulation (Figure 8), most modeled MBL clouds are optically thin (τ
697 < 20) and exhibit medium ($r_e = 10\text{--}15 \mu\text{m}$, 49.8%) or large droplet sizes ($r_e = 15\text{--}20 \mu\text{m}$,
698 25.8%). Compared to observations, model produces more clouds with larger drop size, while
699 observations show a majority with $r_e < 10 \mu\text{m}$ (53.3%; Figure 7, third column). Meanwhile,
700 although the aerosol concentration is prescribed, the model predicts N_d through aerosol
701 activation and microphysical processes, resulting in variabilities in N_d . For clouds with given
702 optical depth, a decrease in r_e indicates an increase in N_d . This increase in N_d is associated with
703 both lower peak of Z_e and a reduced vertical Z_e gradients in the CFODD, suggesting aerosol-
704 induced precipitation suppression. Lastly, cloud dynamics plays a stronger role in the simulation
705 than in observations. For example, thicker clouds in the model show higher peak Z_e values and
706 broader Z_e distribution than thinner clouds with same r_e , whereas this enhancement is less
707 evident in ARM observations.



708
709 Figure 9. Frequency of radar reflectivity as a function of in-cloud optical depth (τ_d) for WRF
710 N=500 simulation. Different rows are for different ranges of optical depth (τ): (a)-(c) clouds with
711 $\tau < 10$, (d)-(f) clouds with $10 < \tau < 20$, (g)-(i) clouds with $\tau > 20$. Different columns are for
712 different ranges of effective radius (r_e). The left, middle, and right columns are for $15 - 20 \mu m$,
713 $10 - 15 \mu m$, and $5 - 10 \mu m$, respectively. The black dashed lines in each panel denote -15
714 dBZ and 0 dBZ, as thresholds of drizzle and rain, respectively. The percentage of sample (P) for
715 each subgroup is denoted in the figure.
716

717 Comparing simulations with different prescribed aerosol concentrations, we observe that
718 with increasing aerosols and decreasing drop size, precipitation is suppressed. This is evidenced
719 by the shift of frequency of occurrence of precipitating clouds, along with reduced peak Z_e and
720 shallower gradient of Z_e . For example, the most common cloud type shifts from thin clouds with

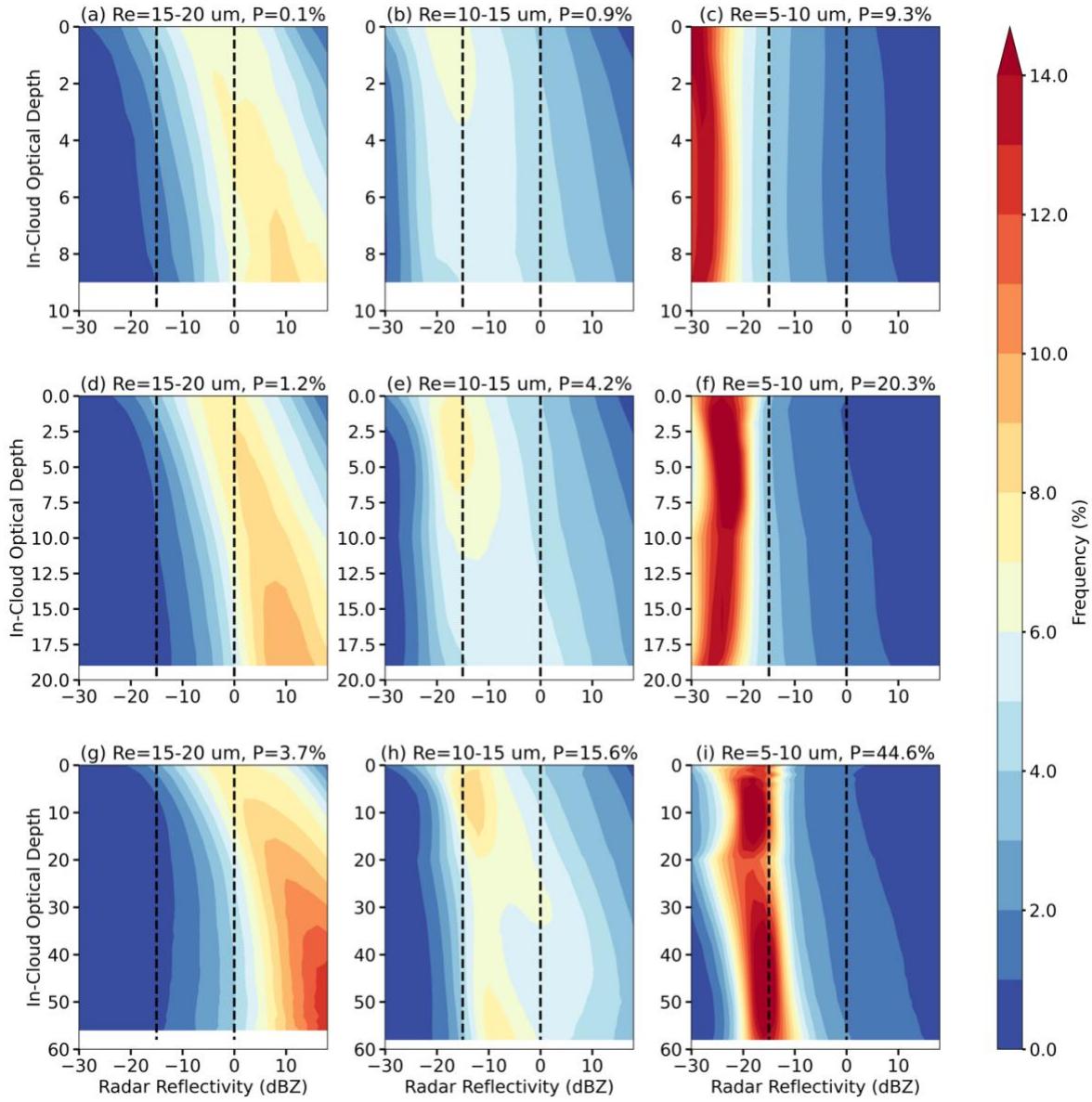
721 moderate r_e in the N=100 simulation (Figures 8b, e) to thicker clouds with smaller r_e in the
722 N=500 run (Figures 9h, i), revealing a typical cloud response to precipitation suppression.
723 Meanwhile, the percentage of clouds with $r_e = 15 - 20 \mu\text{m}$ decreases significantly from 31.9%
724 in N=100 to 6.1% in N=500 simulations. As a result, the droplet size distribution in N=500
725 simulation aligns better with ARM observations, although clouds are still thicker in model. For
726 clouds with similar r_e and τ , both the peak Z_e and its vertical gradient decrease with increasing
727 aerosol concentrations due to the reduced autoconversion with higher N_d . In particular, thick
728 clouds with medium r_e ($r_e = 10 - 15 \mu\text{m}$, $\tau > 20$, Figure 9h) transition from raining to drizzling
729 in the N=500 simulation, aligning more closely with observations.

730 For clouds with $r_e > 15 \mu\text{m}$, rain becomes stronger compared to the N=100 simulation,
731 even in the thinnest cloud (Figures 9 a, d, g vs. Figures 8 a, d, g). While the enhancement of
732 precipitation with increasing aerosol concentration may initially seem counter-intuitive, it can be
733 explained by the parameterization of DSD in the model. For clouds with similar τ , increasing r_e
734 is associated with higher LWP and q_c , but lower N_d . Based on Equation (5), the slope parameter
735 λ decreases with increasing r_e , resulting in a broader DSD with a flatter slope. Additionally, the
736 dispersion parameter η is proportional to N_d so that polluted clouds in N=500 simulation also
737 exhibit broader DSDs. As a result, even under suppressed autoconversion due to higher N_d , the
738 extended tail of the broader DSD initiates autoconversion, enhances accretion from higher fall
739 speed, and ultimately enhances precipitation in the N=500 simulation. Note that this type of
740 cloud occurs much less frequently in the N=500 simulation (6.1%) than in the N=100 simulation
741 (31.9%).

742 When continuously increasing aerosol concentration from N=500 to N=1000 (Figure 9
743 vs. Figure 10), the CFODD of reflectivity changes little, indicating a saturation of the
744 precipitation suppression effect and the broadening of DSD. More clouds shift to the non-
745 precipitating thick clouds subgroup with $r_e < 10 \mu\text{m}$ and $\tau > 20$ (44.6%, Figure 10i).

746 In summary, we evaluated the vertical development of precipitation in the model using
747 ARM radar reflectivity profiles. Our simulations realistically reproduce the non-precipitating
748 regime and the transition to drizzling clouds at similar r_e and τ ranges as ARM observations.
749 Meanwhile, model overestimates precipitation for optically thick clouds and clouds with $r_e >$
750 $15 \mu\text{m}$. This overestimation could be attributed to the early initiation of the autoconversion
751 process, which leads to an early onset of rain near the cloud top. The excessive accretion rates,
752 along with underestimation of entrainment and evaporation, lead to an overproduction of rain in
753 the model, especially in thick clouds with larger water content and longer droplet collection path.
754 Additionally, the parameterized DSD is too broad in the model, especially for polluted clouds
755 with large N_d and large r_e .

756 As the model reasonably captures the properties of non-precipitating thin clouds in
757 agreement with ARM observations, the simulated LWP susceptibility aligns well with satellite-
758 based estimates. In contrast, the overestimation of precipitation in thick clouds leads to a
759 predominantly positive LWP susceptibility in the model due to the precipitation suppression
760 effect. However, satellite observations indicate that these clouds are typically non-precipitating,
761 where entrainment drying dominates, resulting in a negative LWP susceptibility. This highlights
762 the need to improve the parameterization of precipitation processes: particularly autoconversion,
763 accretion, and DSD representation, in order to better simulate ACI across all cloud regimes.



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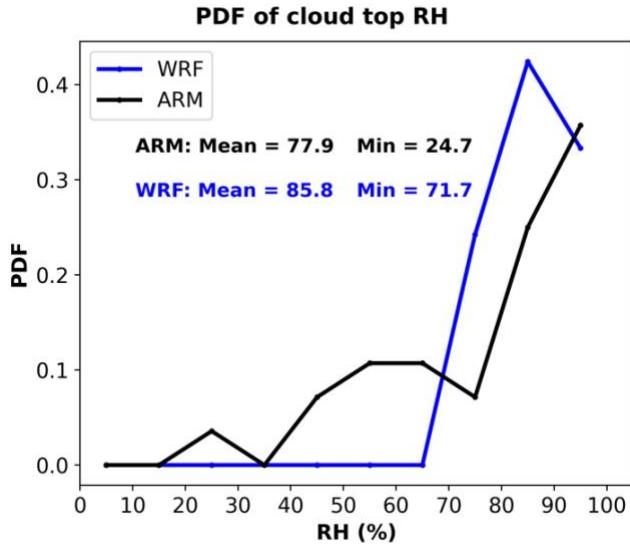
Figure 10. Frequency of radar reflectivity as a function of in-cloud optical depth (τ_d) for WRF N=1000 simulation. Different rows are for different ranges of optical depth (τ): (a)-(c) clouds with $\tau < 10$, (d)-(f) clouds with $10 < \tau < 20$, (g)-(i) clouds with $\tau > 20$. Different columns are for different ranges of effective radius (r_e). The left, middle, and right columns are for $15 - 20 \mu\text{m}$, $10 - 15 \mu\text{m}$, and $5 - 10 \mu\text{m}$, respectively. The black dashed lines in each panel denote -15 dBZ and 0 dBZ, as thresholds of drizzle and rain, respectively. The percentage of sample (P) for each subgroup is denoted in the figure.

While our analysis focuses on the two-moment Morrison scheme, Christensen et al. (2024) found that the choice of microphysics and PBL schemes accounts for only about 30 % of the variability in simulated ACI, much smaller than the variability across meteorological conditions and cloud states. Since this study encompasses 11 cases spanning diverse synoptic regimes and cloud types, the overall conclusions are unlikely to change substantially with

778 alternative two-moment bulk microphysics schemes. Nonetheless, future investigations using
779 multiple microphysics schemes would be valuable for quantifying the robustness of the
780 precipitation parameterization and its role in ACI uncertainty.

781 **3.3.2 Model Bias in Capturing Inversions**

782 As discussed in case study in Section 3.1, ERA5 profiles fail to accurately represent the
783 location and strength of inversions over the ENA region. These biases lead to an underestimated
784 boundary layer height and an overestimated RH above cloud top in the simulations. Figure 11
785 compares the probability density function (PDF) of cloud-top RH between ARM sounding
786 observations and WRF simulations across all 11 cases for N=1000 simulation. Different aerosol
787 concentrations (e.g., N=100, N=500) show consistent results (not shown). In ARM observations,
788 cloud-top height is derived from the radar reflectivity profile, as described in the method section;
789 while in WRF simulations, cloud top is defined as the highest model level where the cloud water
790 mixing ratio exceeds 0.001 g/kg. The RH is sampled at ~100m above cloud top in both data. We
791 further compare the cloud-top heights in WRF simulations defined using cloud water mixing
792 ratio and radar reflectivity profiles with $Z_e > -40$ dBZ from the radar simulator. The two
793 approaches yield nearly identical results, with a mean difference of less than 40m (figure not
794 shown).

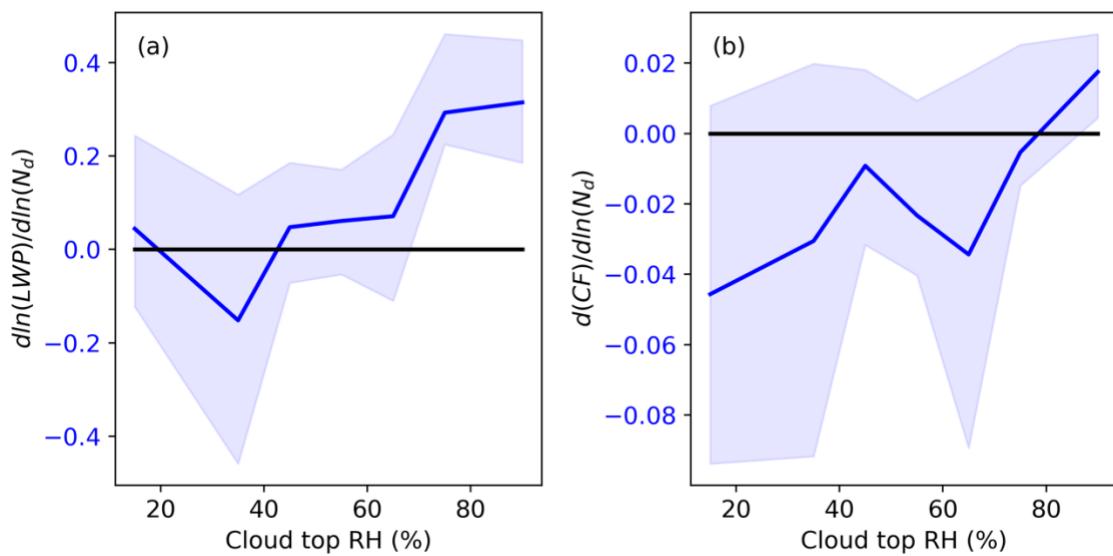


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796 Figure 11. PDF of cloud top relative humidity (RH) for WRF simulations (blue line) and ARM
797 sounding observations (black line).

798
799 To ensure a meaningful comparison between WRF output and ground-based
800 observations, cloud-top RH from WRF is averaged over a 10km \times 10km grid box centered at the
801 ARM ENA site for each sounding time, given the ~1.2-1.4 km mean cloud-top height for MBL
802 clouds and ~7 m/s prevailing wind speed at ENA during summer (Wood et al., 2015; Wu et al.,
803 2020). As seen in Figure 11, WRF simulations exhibit a systematic wet bias in cloud-top RH,
804 with the mean values 7.9% higher than those from observations and with no RH values below
805 71%.

806 Figure 12 shows the mean relationship between cloud top RH and cloud susceptibilities
807 calculated based on domain mean values for all three simulations (e.g. N=1000 vs. N=100,

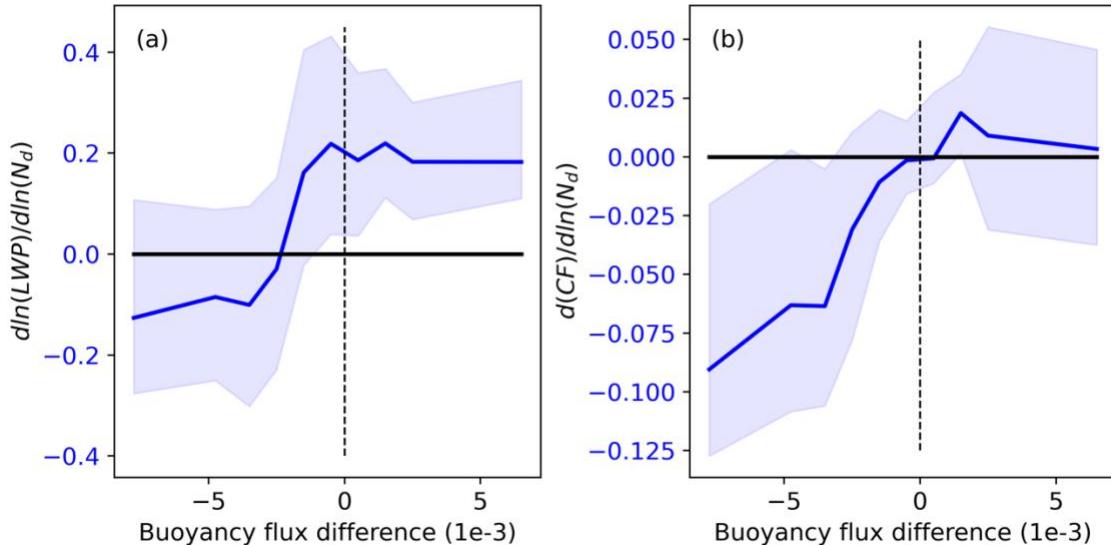
808 N=500 vs. N=100, and N=1000 vs. N=500). The cloud top RH is the domain mean RH value at
 809 ~100m above cloud top for all simulations. As seen in Figure 12a, we find a positive correlation
 810 between cloud-top RH and LWP susceptibility in the simulations, which is consistent with cloud
 811 responses shown in case study where a dry layer above cloud promotes evaporation and decrease
 812 LWP. Additionally, these positive relationships are consistent among different aerosol
 813 concentrations (e.g., N=1000 vs. N=100 or N=500 vs. N=100; figures not shown). Meanwhile, as
 814 seen in Figure 12, cloud top moisture has a more evident impact on cloud LWP than cloud cover.
 815 Relations between cloud top moisture and cloud susceptibilities found in our simulations are
 816 consistent with that in satellite observations around the globe (e.g. Toll et al., 2019; Yuan et al.,
 817 2023), except that LWP susceptibility is mostly negative while CF susceptibility is mostly
 818 positive in satellite data.



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 820 Figure 12. Dependence of (a) LWP susceptibility and (b) CF susceptibility on cloud top relative
 821 humidity in WRF simulations during the daytime. The solid blue line shows the median value of
 822 each RH bins and the shaded area shows the lower and upper 25th percentiles.
 823

824 Based on the relationship between cloud susceptibility and cloud-top RH, the over-
 825 estimated cloud-top RH of 8% may lead to an overestimation of 0.04 and 0.005 in LWP and CF
 826 susceptibility, respectively. Meanwhile, the under-estimated cloud-top height of 480m could
 827 result in an under-estimation of LWP and CF susceptibility of 0.18 and 0.02, respectively
 828 (figures not shown). Future modeling studies over the ENA region need to improve the initial
 829 and boundary conditions, e.g., through data assimilations.
 830

831 To further illustrate the influence of cloud-top evaporation on LWP and CF adjustment
 832 rate, we analyzed the relationship between cloud susceptibilities and change in the cloud-layer
 833 buoyancy flux. As shown in the case study, buoyancy flux increases with aerosol perturbation in
 834 precipitating clouds due to precipitation suppression, whereas it decreases in non-precipitating
 835 clouds due to enhanced entrainment driven evaporation. Thus, changes in buoyancy flux serves
 836 as a proxy for both cloud-top evaporation and precipitation suppression effects.



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Figure 13. Dependence of (a) LWP susceptibility and (b) CF susceptibility on changes in buoyancy flux in the cloud layer in WRF simulations during the daytime. The solid blue line shows the median value of each buoyancy flux bins and the shaded area shows the lower and upper 25th percentiles.

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In Figure 13, changes in cloud-layer buoyancy flux is calculated as the difference in domain-mean values between polluted and clean experiments (e.g., $N=1000$ vs. $N=100$, $N=500$ vs. $N=100$, and $N=1000$ vs. $N=500$), averaged in the cloud layer defined by the domain-mean cloud water mixing ratio. As shown in Figure 13, two distinct regimes emerge: when cloud-layer buoyancy flux substantially decrease with increasing aerosols, both LWP and CF decrease; when changes in buoyancy flux is small negative or positive, LWP and CF susceptibilities are generally positive or near zero. These results, together with those in Figure 12, support the conclusion that the reduction in LWP and CF in the model is primarily driven by cloud-top evaporation associated with enhanced entrainment. The absence of negative LWP responses in earlier modeling studies may be attributed to inadequate resolution of the interactions among boundary layer turbulence, entrainment, and cloud-top evaporation.

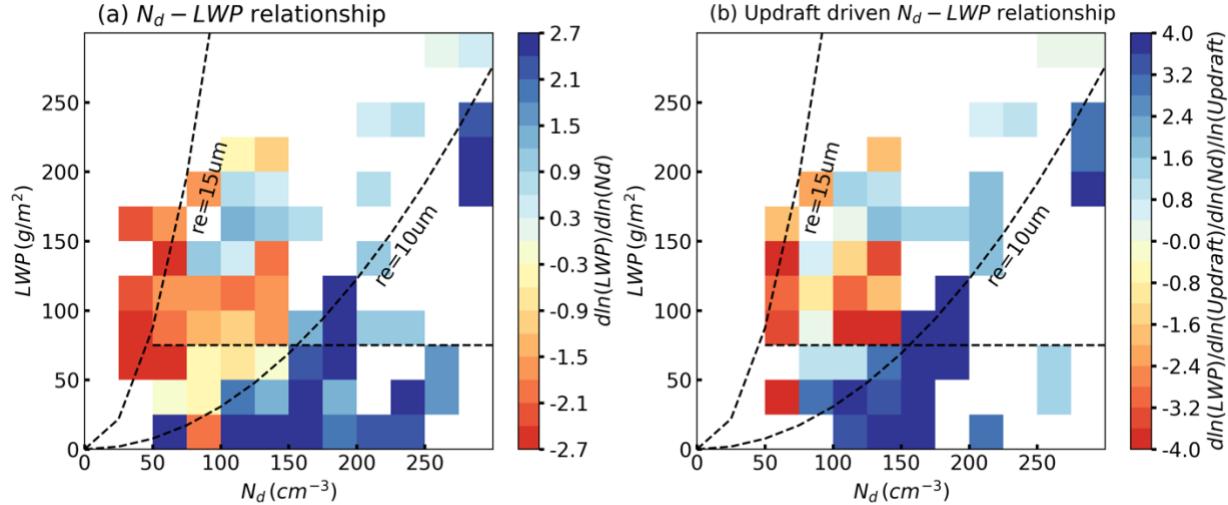
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3.3.3 LWP Adjustment from Internal Cloud Processes and Precipitation Heterogeneity

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In addition to model biases in representing precipitation processes and PBL thermodynamic profiles, one leading factor contributing to the discrepancy in ACI estimates lies in how ACI is diagnosed in numerical studies versus observations. In model simulations, ACI can be isolated using controlled experiments by varying aerosol concentrations while holding meteorology constant. In satellite-based analysis, however, the retrieved ACI signal inevitably includes not only aerosol-induced cloud responses but also N_d –LWP covariability arising from internal cloud processes, even under strict spatial and temporal sampling constraints. Diagnosing these internal cloud processes in satellite observations is difficult because key governing variables, such as cloud-base updraft speed, TKE, entrainment rate are not directly measured or retrieved. In contrast, model simulations allow us to quantify the N_d –LWP relationships driven by internal cloud processes by examining their spatial covariation under homogeneous aerosol conditions.

866 To ensure consistency with satellite methodology and suppress small-scale cloud heterogeneity,
 867 pixel-level model outputs are aggregated to a $25\text{ km} \times 25\text{ km}$ grid.

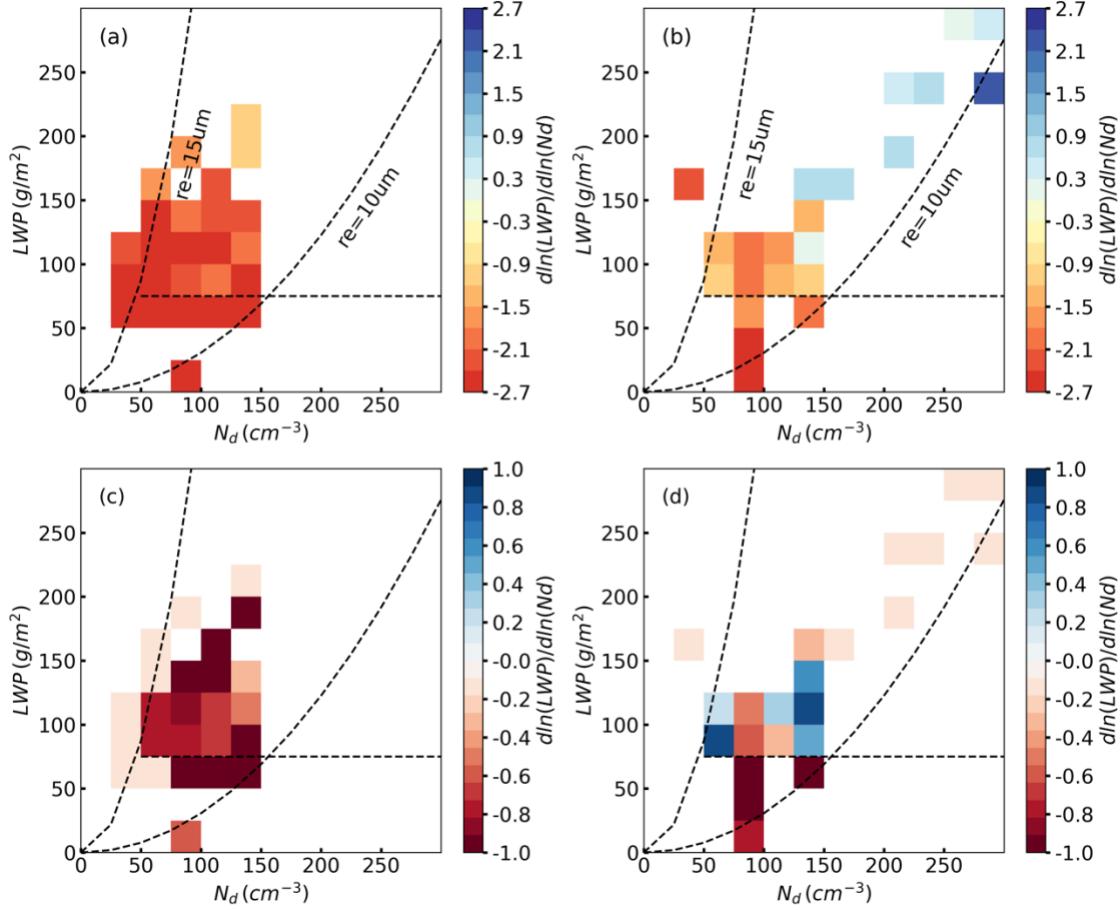


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 869 Figure 14. (a) LWP- N_d relations stem from internal cloud processes (b) LWP- N_d relations driven
 870 by cloud base updraft speed in WRF simulations during the daytime.

871
 872 Figure 14a shows the resulting N_d -LWP relationships across all cases and all aerosol
 873 concentrations, revealing opposing signs between different cloud regimes: a strong positive
 874 correlation for non-precipitating clouds and a strong negative correlation for precipitating clouds.
 875 To understand this contrast, we examine whether both N_d and LWP co-vary with a third
 876 parameter indicative of internal dynamics. Cloud-base updraft speed emerges as a physical
 877 meaningful driver: the ratio of $\frac{d\ln(LWP)}{d\ln(\text{Updraft})}$ to $\frac{d\ln(N_d)}{d\ln(\text{Updraft})}$ in Figure 14b closely mirrors the N_d -
 878 LWP relations in Figure 14a. This indicates that cloud base updraft speed largely governs the
 879 opposing responses. In non-precipitating clouds, stronger updrafts enhance supersaturation,
 880 activation, and condensation, increasing both N_d and LWP, and resulting in a positive N_d -LWP
 881 relationship. In precipitating clouds, stronger updrafts increase LWP and rain rate, but
 882 precipitation formation reduces N_d via coalescence and collection, leading to a negative relation.

883 Furthermore, mesoscale variability in precipitation structure can further modulate the
 884 N_d -LWP relationship in precipitating clouds. To test this hypothesis, precipitating cases
 885 (domain-mean precipitation fraction > 0.1) are further divided into heterogeneous and
 886 homogeneous categories based on the spatial standard deviation of precipitation fraction using
 887 the upper and lower 50th percentile, respectively (Figure 15). Precipitation fraction is defined as
 888 the areal fraction of cloud pixels with the column maximum reflectivity greater than -15 dBZ
 889 (Figure 6).

890 In heterogeneous convective precipitation (Figure 15a), strong and spatially variable
 891 latent heating release enhances buoyancy within clouds, while rain evaporation and downdrafts
 892 generate cold pools. Both processes act to intensify updrafts, which in turn promote rapid droplet
 893 growth and increase the cloud's capacity to retain liquid water, leading to higher LWP and
 894 precipitation. Meanwhile, stronger coalescence and precipitation scavenging reduce N_d . Such
 895 opposite changes in LWP and N_d amplify the negative N_d -LWP relationship (Figure 15c). In
 896 homogeneous stratiform precipitation, latent heating is more spatially uniform and stratification
 897 inhibits localized buoyancy-driven updrafts. Weaker coalescence and less efficient scavenging
 898 lead to a less negative N_d -LWP relationship (Figure 15d).



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Figure 15. Same as Figure 14, but for scenes with (a) heterogeneous and (b) homogeneous
precipitation fraction. (c) and (d) show the difference between (a) and (b) with Figure 14a.903
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In summary, even though clouds with $\text{LWP} > 75 \text{ g/m}^2$ and $r_e < 15 \mu\text{m}$ are typically
classified as non-precipitating thick clouds in observational ACI studies, pixel-level data real that
20–35% of these clouds produce precipitation (Figure 6a). The strongly negative LWP
susceptibilities inferred from satellite data for non-precipitating thick clouds may partly arise
from internal cloud processes driven by updraft speed and mesoscale precipitation structure,
rather than from aerosol–cloud interactions alone. providing a plausible explanation for the
model–observation discrepancy. Meanwhile, non-precipitating thin clouds with $\text{LWP} < 75 \text{ g/m}^2$
and $r_e < 15 \mu\text{m}$ exhibit low pixel-level precipitation fractions (typically < 0.1 , Figure 6a), and the
positive N_d –LWP relationships arising from internal cloud processes may bias satellite-derived
LWP susceptibility toward more positive values, further expanding the model–observation gap.
The opposing signs of N_d –LWP relationships in Figures 14a and 5c for non-precipitating thin
clouds highlight the need for additional process-level analysis in future study.915
4. Conclusions and Discussions916
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Previous studies found that model simulations and observations often reveal opposing
results in LWP responses to aerosol perturbations for MBL clouds. For example, satellite-based

918 assessments indicate a decrease of cloud LWP with aerosol perturbations, especially in polluted
919 conditions for non-precipitating clouds (e.g., Gryspeerdt et al. 2019; Toll et al., 2019; Zhang et
920 al., 2022, 2023; Qiu et al., 2024; Yuan et al., 2023; 2025). On the other hand, most GCMs and
921 CPMs simulate an increase of LWP with increasing aerosols (e.g., Ghan et al., 2016; Michibata
922 et al., 2016; Mülmenstädt et al., 2024; Fons et al., 2024; Christensen et al, 2024). Previous
923 studies found that increasing model resolution to sub-kilometer can improve the representation of
924 precipitation process and model performance in ACI by resolving the small-scale process most
925 relevant to ACI (e.g., Terai et al., 2020). It remains unclear how well models perform at close to
926 LES scale in representing the ACI feedback when using realistic meteorological conditions and
927 large case ensembles across various cloud states and synoptic regimes.

928 To address these gaps, our study makes three key advances: (1) we conduct a series of
929 realistic near-LES-scale case studies that enable direct comparison with ground-based and
930 satellite observations to reconcile observed–modeled discrepancies; (2) we examine a large
931 ensemble of MBL cloud cases spanning a range of cloud states and synoptic conditions to
932 capture the diversity of ACI responses; and (3) we use the same two-moment microphysics
933 scheme implemented in several GCMs and CPMs, making our findings directly relevant for
934 improving microphysical parameterizations in climate models.

935 The simulated MBL clouds generally match the satellite observation in domain mean
936 cloud coverage and mesoscale organization (Figures 1, 3, S2-S4), while the model may struggle
937 to capture the diurnal evolution of clouds, especially the dissipation of clouds in the afternoon.
938 Model overestimate cloud LWP, especially in the polluted runs and underestimated cloud top
939 height compared to satellite retrievals. To show the dependence of cloud responses on cloud
940 state, LWP susceptibilities are displayed in the N_d -LWP parameter space (Figure 5). For non-
941 precipitating thin clouds, our simulations show a consistently negative but weaker LWP
942 susceptibility compared to satellite observations, with a mean of -0.13 . The negative LWP
943 susceptibility likely result from the better resolved turbulence, condensation/evaporation
944 processes and their feedback on PBL thermodynamics. More specifically, increases in aerosols
945 enhance turbulence and TKE in the cloud layer. With the dry air above, the entrained dry air
946 intensifies evaporation, reduces buoyancy flux in the cloud layer and leads to dissipations of
947 clouds (Figure 4, 13).

948 For precipitating clouds, our model predicts a slight increase in LWP with the mean
949 susceptibility of $+0.15$, which is consistent with the precipitation suppression hypothesis and the
950 climatological mean cloud response for heavily precipitating clouds (e.g., Qiu et al., 2024). For
951 non-precipitating thick clouds, model simulations and satellite observations show the largest
952 disagreement with opposite LWP susceptibilities of $+0.32$ vs. -0.69 , respectively. Meanwhile,
953 the non-precipitating thick clouds are the dominant cloud state in the model, with a total
954 frequency of 49%, compared to a 15.7% frequency of occurrence in satellite observations. The
955 overestimation of N_d arise from the overestimated aerosol concentration in the configuration,
956 combined with the absence of precipitation scavenging in the model. The overestimation of LWP
957 is due to the positive LWP susceptibility in thick clouds where LWP in N=100 simulation show
958 good agreement with satellite retrievals (Figure S9)

959 Our analyses indicate that such discrepancy could mainly result from the overestimation
960 of precipitation for thick clouds: where MBL clouds in simulations produce precipitation at
961 much smaller cloud drop size (e.g., $r_e > 10 \mu m$) and in more polluted conditions compared to
962 satellite observations (Figure 6). Based on ARM radar observations, our simulations reasonably
963 capture the non-precipitating regime and the transition from non-precipitating to drizzling clouds

964 within the same r_e and τ range as observed (Figures 7, 8). Our simulation result appears to better
965 represent marine clouds than GCM or GCPM, which often initiate drizzle or rain at cloud top
966 and rarely simulate non-precipitating clouds (e.g., Jing et al. 2017, 2019; Michibata and Suzuki,
967 2020). However, several biases remain. In non-precipitating clouds, the model shows near-
968 constant Z_e profile with height, whereas observations show a decrease near cloud top, suggesting
969 an underestimation of entrainment and evaporation (Figure 8). In thicker clouds ($\tau > 20$), drizzle
970 often initiates too early at cloud top ($Z_e > -15$ dBZ), indicating excessive autoconversion. This
971 early onset allows raindrops to grow too large through prolonged collection in deeper clouds,
972 resulting in overestimated rain rates ($Z_e > 0$ dBZ), whereas observations show only drizzle
973 (Figure 8). Additionally, stronger rain in polluted cases with large r_e points to an overly broad
974 DSD, as the dispersion parameter η in the Morrison scheme increases with N_d , and the DSD
975 slope flattens with larger r_e (Figures 9, 10). The overestimation of precipitation for thick clouds
976 results in an increase in LWP from precipitation suppression in the simulation.

977 The overestimation of LWP susceptibility may also stem from biases in ERA5 and WRF
978 profiles in representing the location and strength of moisture inversions (Figures S6, S8), leading
979 to shallower PBL and a moist bias above the clouds in the simulations (Figure 11). Consistent
980 with observations, model simulations show a positive correlation between LWP susceptibility
981 and cloud-top RH, suggesting that the wet bias in cloud-top RH contributes to the positive bias in
982 LWP susceptibility (Figure 12).

983 Lastly, we find that part of the discrepancy in quantified ACI may stem from N_d -LWP
984 relationships driven by internal cloud processes that are mixed with the ACI signals in satellite
985 observations. Using model simulations with homogenous aerosol concentrations, we isolate these
986 internally driven N_d -LWP relationships. Our results reveal large opposing signals between
987 precipitating clouds (large negative relationships) and non-precipitating clouds (large positive
988 relationships), primarily governed by cloud base updraft speed (Figures 14) and modulated by
989 mesoscale cloud and precipitation organization (Figure 15). Therefore, the strongly negative
990 LWP susceptibility observed in thick clouds in satellite data could reflect internal cloud
991 dynamics rather than true ACI.

992 This study shows that while the discrepancy in ACI assessments between observations
993 and models can be reduced by increasing model resolution for precipitating and non-precipitation
994 thin clouds, the positive bias in the LWP susceptibility for non-precipitating thick clouds
995 persists. This bias is attributed to parameterization deficiencies in the microphysics scheme and
996 model biases in lower tropospheric thermodynamics over the ENA region. These findings may
997 motivate improvements in precipitation parameterizations and encourage their process-level
998 evaluation against observations.

999

1000 **Data availability:**

1001 The WRF model used, version 4.2.2, is freely available from the developers' website (<https://github.com/wrf-model/WRF/releases>, WRF, 2022). SEVIRI Meteosat cloud retrieval products, produced by NASA LaRC
1002 SatCORPS group, are available from the Atmospheric Radiation Measurement (ARM) Data Discovery website at
1003 <https://adc.arm.gov/discovery/>, Minnis Cloud Products Using Visst Algorithm. The ARM ground-based radar and
1004 lidar observations (KAZRARSCL), LWP retrievals, and balloon sounding observations are available from ARM
1005 Data Discovery.

1007

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1018

1019 **Author contribution:**

1020 SQ carried out the data analysis and wrote the manuscript. XZ and HL ran the simulations. PW provided the ground-
1021 based cloud microphysics retrievals. All authors contributed to the design of the study, interpretation of the results,
1022 and edit the manuscript.

1023

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1026

1027 **Competing interests:**

1028 The authors declare that they have no conflict of interest.

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