

1 **Distinctive aerosol-cloud-precipitation interactions in marine boundary layer clouds from the**
2 **ACE-ENA and SOCRATES aircraft field campaigns**

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12
13 **Abstract.** The aerosol-cloud-precipitation interactions within the cloud-topped Marine Boundary Layer
14 (MBL) are examined using aircraft in-situ measurements from Aerosol and Cloud Experiments in the
15 Eastern North Atlantic (ACE-ENA) and Southern Ocean Clouds Radiation Aerosol Transport
16 Experimental Study (SOCRATES) field campaigns. SOCRATES clouds exhibit a larger number
17 concentration and smaller cloud droplet effective radius (148.3 cm^{-3} and $8.0 \mu\text{m}$) compared to ACE-ENA
18 summertime (89.4 cm^{-3} and $9.0 \mu\text{m}$) and wintertime clouds (70.6 cm^{-3} and $9.8 \mu\text{m}$). The ACE-ENA clouds,
19 especially during the winter, feature stronger drizzle formation via droplet growth through enhanced
20 collision-coalescence, attributed to a relatively cleaner environment and deeper cloud layer. Furthermore,
21 the Aerosol-Cloud Interaction (ACI) indices from the two aircraft field campaigns exhibit distinct
22 sensitivities, indicating different cloud microphysical responses to aerosols. The ACE-ENA winter
23 season features relatively fewer aerosols, which are more likely activated into cloud droplets under the
24 conditions of sufficient water vapor availability and strong turbulence. The enriched aerosol loading
25 during ACE-ENA summer and SOCRATES generally leads to smaller cloud droplets competing for the

26 limited water vapor and exhibiting a stronger ACI. Notably, the precipitation susceptibilities are stronger
27 during the ACE-ENA than during the SOCRATES campaigns. The in-cloud drizzle behavior
28 significantly alters sub-cloud cloud condensation nuclei (CCN) budgets through the coalescence-
29 scavenging effect, and in turn, impact the ACI assessments. The results of this study can enhance the
30 understanding and aid in future model simulation and assessment of the aerosol-cloud interaction.

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32

33 **1. Introduction**

34 Marine boundary layer (MBL) clouds substantially impact the Earth's climate system (Dong and
35 Minnis, 2022). Sustained by large-scale subsidence and cloud-top longwave radiative cooling, MBL
36 clouds, typically located beneath the temperature inversion at the MBL top, persistently reflect the
37 incoming solar radiation and modulate the radiative balance (Lilly, 1968; Albrecht et al., 1995; Wood et
38 al., 2015; Dong et al., 2023). The climatic significance of MBL cloud radiative effects, which remains
39 largely uncertain (IPCC, 2022), is closely linked to cloud microphysical properties that are substantially
40 influenced by surrounding aerosol conditions (Chen et al., 2014; Feingold and McComiskey, 2016).
41 Observational evidence demonstrates that cloud microphysical responses to aerosols, defined as the
42 aerosol-cloud interaction (ACI), can be typically viewed as decreased cloud droplet effective radii (r_c)
43 and increased number concentrations (N_c) with more aerosol intrusion under conditions of comparable
44 cloud water content (Feingold and McComiskey, 2016). The ACIs have been extensively investigated
45 by different observational platforms, such as aircraft (Hill et al., 2009; Diamond et al., 2018; Gupta et
46 al., 2022), ground-based and satellite observations (Painemal et al., 2020; Zhang et al., 2022; Zheng et
47 al., 2022a), and model simulations (Wang et al., 2020; Christensen et al., 2023) over different maritime
48 regions like the southeast Pacific (Painemal and Zuidema, 2011), northeast Pacific (Braun et al., 2018),
49 southeast Atlantic (Gupta et al., 2022), and eastern North Atlantic (Zheng et al., 2022a).

50 Furthermore, a larger number of small cloud droplets can sometimes extend cloud longevity and
51 spatial coverage and modulate the precipitation processes in the MBL clouds, reflecting the cloud
52 adjustments to aerosol disturbances (Albrecht, 1989; Bellouin et al., 2020). Precipitation, particularly in
53 the form of drizzle, is common in MBL clouds (Wood et al., 2015; Wu et al., 2020), and the turbulence
54 forced by stratocumulus cloud-top radiative cooling can increase the cloud liquid water path and
55 contribute to drizzle production (Ghate et al., 2019, 2021). The drizzle formation and growth processes
56 are deeply entwined with the MBL aerosols and dynamics. Frequent aerosol intrusions in the MBL have
57 been found to have to lower the efficiency of collision-coalescence-induced which results in the
58 suppression of precipitation frequency and strength. Such phenomenon can be quantified and assessed
59 via the cloud precipitation susceptibility (Feingold and Seibert, 2009; Lu et al., 2009; Sorooshian et al.,
60 2009; Duong et al., 2011). The assessments of precipitation susceptibility are examined to be under the
61 influences of methodology (Terai et al., 2012), cloud morphology (Sorooshian et al., 2009; Jung et al.,
62 2016), ambient aerosol concentrations (Duong et al., 2011; Jung et al., 2016; Gupta et al., 2022), and
63 cloud thickness (Terai et al., 2012; Jung et al., 2016; Gupta et al., 2022). The in-cloud turbulence and
64 wind shear can effectively enhance collision-coalescence efficiency, stimulate drizzle formation and
65 growth, and consequently lead to enhanced precipitation rate and amount (Chen et al., 2011; Wu et al.,
66 2017). Cloud-top entrainment of dryer and warmer air can potentially deplete small cloud droplets and
67 shrink large droplets via evaporation, thereby impacting cloud-top microphysical processes depending
68 on the homogeneous or inhomogeneous mixing regimes (Lehmann et al., 2009; Jia et al., 2019).

69 Conversely, precipitation has been shown to exert a substantial influence on the MBL aerosol and
70 cloud condensation nuclei (CCN) budget through the coalescence-scavenging effect. The coalescence-
71 scavenging refers to the process in which cloud or drizzle droplets, containing aerosol particles inside,
72 merge with each other. Upon the collision-coalescence of cloud droplets, the dissolved aerosol masses
73 within the cloud droplets also collide and merge into a larger aerosol core, leading to larger aerosol
74 particles upon droplet evaporation. The sub-cloud aerosols are then replenished into the cloud layer,

75 experiencing growth within the cloud through cloud and drizzle droplet collision-coalescence and
76 subsequently falling and evaporating outside the cloud again. Eventually, the residual aerosols
77 undergoing this cloud-processing cycle will gradually decrease in number concentration and increase in
78 size (Flossmann et al., 1985; Feingold et al., 1996; Hudson and Noble, 2020; Hoffmann and Feingold,
79 2023). In addition, the drizzle drops, upon falling out of the cloud base, can result in net reductions in
80 sub-cloud aerosols and CCN budgets via precipitation scavenging processes (Wood, 2006; Zheng et al.,
81 2022b). Quantitative estimates of these effects remain ambiguous and inconclusive, which are subject to
82 multiple factors such as aerosol physicochemical characteristics, cloud morphology, and MBL dynamics
83 and thermodynamics conditions (Sorooshian et al., 2009; Duong et al., 2011; Diamond et al., 2018;
84 Brunke et al., 2022). Thus, more studies on the aforementioned processes regarding MBL aerosols and
85 clouds over different maritime regions are warranted to pursue an in-depth understanding of aerosol-
86 cloud-precipitation interactions (ACPIs).

87 The Eastern North Atlantic (ENA) stands as a desirable region for exploring MBL clouds in the
88 mid-latitude, with Graciosa Island in the Azores (39.09°N, 28.03°W) representing a focal point for
89 studies of ACPIs. Located between the mid-latitude and subtropical climate zones, Graciosa Island is
90 subject to the meteorological influence of both the Icelandic Low and the Azores High, and the influence
91 of aerosols ranging from pristine marine air masses to those heavily influenced by continental emissions
92 from North America and Northern Europe (Logan et al., 2014; Wood et al., 2015; Wang et al., 2020).
93 Addressing the need for sustained research into the MBL clouds, the recent Aerosol and Cloud
94 Experiments in the Eastern North Atlantic (ACE-ENA) aircraft campaign (J. Wang et al., 2022) was
95 conducted in the summer (June and July) 2017 (ACE-ENA Sum) and winter (January and February)
96 2018 (ACE-ENA Win). During these two intensive operation periods (IOPs) of ACE-ENA, the research
97 aircraft accrued abundant in-situ measurements of aerosols, clouds, and drizzle properties, providing
98 invaluable resources for studying the ACI and ACPI processes. During the summer, the Azores is located
99 at the eastern part of the high-pressure system, while during the winter, the center of the Azores high

100 shifts to the eastern Atlantic and is primarily located directly over the Azores (Mechem et al., 2018; J.
101 Wang et al., 2022). Furthermore, both ACE-ENA Sum and ACE-ENA Win IOPs of featured
102 anomalously strong high-pressure systems, compared to the 20-year climatology, as shown in Figure S1.
103 This meteorological pattern is favorable to the prevailing and persistent stratocumulus clouds observed
104 during ACE-ENA, especially during the winter IOP, where the enhanced large-scale subsidence can lead
105 to stronger and sharper temperature inversions above the stratocumulus-topped MBL (Rémillard and
106 Tselioudis, 2015; Jensen et al., 2021; Marcovecchio et al., 2022). The ACE-ENA Sum is characterized
107 by anomalously low MBL heights and substantial MBL decoupling (Miller et al., 2021; J. Wang et al.,
108 2022). The winter IOP was under the frequent impacts of the mid-latitude systems and prevalently
109 featured precipitation-generated cold pools, where evaporative cooling alters the thermodynamical
110 structure of the MBL, sustains and enhances turbulence mixing, hence contributes to dynamical
111 perturbations that can influence the behavior of the MBL (Terai and Wood, 2013; Zuidema et al., 2017;
112 Jensen et al., 2021; J. Wang et al., 2022; Smalley et al., 2024). In recent years, many observational studies
113 based on ACE-ENA data have focused on the seasonal contrasts of the aerosol distributions and sources
114 (Y. Wang et al., 2021b; Zawadowicz et al., 2021), the cloud and drizzle microphysics vertical
115 distributions (Wu et al., 2020a; Zheng et al., 2022b), and the impacts of MBL conditions on the cloud
116 structure and morphology (Jensen et al., 2021). However, they seldom analyze the comprehensive
117 interactions between aerosol, clouds, and precipitation.

118 Over the Southern Ocean (SO), the Southern Ocean Clouds Radiation Aerosol Transport
119 Experimental Study (SOCRATES) field campaign (McFarquhar et al., 2021) was conducted during the
120 austral summer (January and February 2018), which marks another valuable piece of the MBL cloud
121 research. The SO, being one of the cloudiest regions globally, is predominantly influenced by naturally
122 produced aerosols originating from oceanic sources due to its remoteness, where the anthropogenic and
123 biomass burning aerosols exert minimal influence over the region (McCoy et al., 2021; Sanchez et al.,
124 2021; Twohy et al., 2021; Zhang et al., 2023). The aerosol budget in this region is primarily shaped by

125 biological aerosols, which nucleate from the oxidation products of dimethyl sulfide (DMS) emissions, as
126 well as by sea spray aerosols. Hence, the SO provides an unparalleled natural laboratory for discerning
127 the influence of these natural aerosol emissions on the MBL clouds under a pre-industrial natural
128 environment. The summertime SO region, particularly near the SOCRATES focus area, is characterized
129 by more frequently closed-cell mesoscale cellular convection structures (Danker et al., 2022; Lang et al.,
130 2022). Furthermore, the MBL clouds over the SO predominantly consist of supercooled liquid water
131 droplets, which coexist with mixed- and ice-phase processes (Y. Wang et al., 2021a; Xi et al., 2022),
132 while the precipitation phases are examined to be primarily dominated by liquid hydrometeors (Tansey
133 et al., 2022; Kang et al., 2024). The in-situ measurements collected from SOCRATES have cultivated
134 many studies on aerosols, clouds, and precipitation over the SO using both in-situ measurements and
135 model simulations (McCoy et al., 2020; Altas et al., 2021; D'Alessandro et al., 2021), and provides an
136 opportunity to study the liquid cloud processes under a colder climate. As shown in Figure S1c, our
137 composite analysis of the synoptic pattern shows that the SOCRATES cloud cases used in this study are
138 located ahead of the anomalously strong thermal ridge and behind the thermal trough, providing an
139 environment favorable to closed cellular MBL cloud structures (McCoy et al., 2017; Lang et al., 2022).
140 Since the region of selected SOCRATES cloud cases crosses a larger latitudinal zone and is under more
141 consistent influence of mid-latitude cyclone systems than the ACE-ENA during the summer IOP, the
142 cloud sampling periods used in this study majority reside in the closed-cell MBL stratocumulus decks.

143 The cloud cases selected from the ACE-ENA and SOCRATES campaigns share similar cloud
144 morphology (stratocumulus) while experiencing different aerosol sources and meteorological conditions.
145 A synergistic approach that compares data from these different field campaigns can provide valuable
146 insights to the community regarding the dominant physical processes of the interactions between aerosols,
147 clouds, and precipitation under the influence of different MBL dynamic and thermodynamic conditions.
148 This study targets the similarities and differences in the MBL aerosol, cloud, and drizzle properties, their
149 distribution and evolution, and more appealingly, the ACIs and ACPIs between the two campaigns. The

150 data and methods used in this study are introduced in section 2. The aerosol and CCN properties in the
151 above- and sub-cloud regimes, as well as the vertical distributions of MBL cloud and drizzle properties,
152 are examined in section 3. The ACI, precipitation susceptibility and drizzle impacts on the sub-cloud
153 aerosols and CCN (ACPI) are discussed in section 4. Finally, the results are summarized, and the
154 importance of this study is discussed in section 5.

155

156 **2. Data and methods**

157 **2.1 Cloud and drizzle properties**

158 The in-situ measurements of MBL cloud properties are temporally synchronized to 1 Hz
159 resolution, corresponding to approximately 100 m (5 m) of horizontal (vertical) sampling. The sampling
160 locations of the selected cases are indicated by the white dots in Figure S1. The Fast Cloud Droplet Probe
161 (FCDP) onboard the aircraft during ACE-ENA can detect droplets with diameter (D_p) ranging from 1.5
162 μm to 50 μm , with the size bins of the probe between 1 and 3 μm (Glienke and Mei, 2020). SOCRATES
163 used a similar CDP to measure droplets from 2 μm to 50 μm at a 2 μm probe size bin width. Both ACE-
164 ENA and SOCRATES leverage the Two-Dimensional Stereo Particle Imaging Probe (2DS) to discern
165 droplets with diameters from 5 μm to 1280 μm (Lawson et al., 2006; Glienke and Mei, 2019). The 2DS
166 in-situ measurements are used as additional screening to eliminate the ice particles with diameters larger
167 than 200 μm . Moreover, the University of Washington Ice-Liquid Discriminator product, which is a
168 Machine-learning-based single-particle phase classification of the 2DS images (Atlas et al., 2021), is
169 used to identify small ice crystals when available. Through these three datasets, we can tease out the ice-
170 dominated period to the highest extent possible and focus on the liquid cloud processes and ACI during
171 SOCRATES (Wang et al., 2021).

172 Although these in-situ measurements can provide “ground-truth” datasets, their uncertainties
173 must be properly analyzed and data quality must be controlled before being applied to scientific studies.
174 The uncertainties of FCDP in sizing and concentration are approximately 30% and 20%, respectively

175 (Baumgardner et al., 2017). Considering the significant uncertainty in the concentration of smaller
 176 particles from a photodiode probe such as 2DS (Baumgardner & Korolev, 1997; Wang et al., 2021), a
 177 diameter of 40 μm is used as the demarcation line between cloud droplets and drizzle drops (Wood et al.,
 178 2005). Then droplet number concentrations in the overlapping size bin between FCDP and 2DS are
 179 redistributed assuming a gamma distribution, thereby a complete size spectrum of cloud and drizzle can
 180 be merged from FCDP and 2DS measurements. Hence, the cloud and drizzle microphysical properties
 181 can be calculated.

182 The cloud droplet number concentration (N_c) is given by:

$$183 \quad N_c = \int_2^{40} n(D_p) dD_p, \quad (1)$$

184 The cloud droplet effective radius (r_c , Hansen and Travis, 1974) is given by:

$$185 \quad r_c = \frac{\int_2^{40} r_p^3 n(D_p) dD_p}{\int_2^{40} r_p^2 n(D_p) dD_p}, \quad (2)$$

186 The cloud liquid water content (LWC_c) can be calculated by:

$$187 \quad LWC_c = \frac{4}{3} \pi \rho_w \int_2^{40} D^3 n(D_p) dD_p, \quad (3)$$

188 where ρ_w is water density.

189 Similarly, the drizzle drop number concentration (N_d) and liquid water content (LWC_d) can be calculated
 190 using the size distribution from 40 μm to 1280 μm . Particularly, the drizzle mean mass diameter (D_{mmd})
 191 is given by:

$$192 \quad D_{mmd} = \left(\frac{\int_{40}^{1280} D_p^3 n(D_p) dD_p}{\int_{40}^{1280} n(D_p) dD_p} \right)^{1/3}, \quad (4)$$

193 This quantity is chosen because the D_{mmd} denotes the diameter of average mass (the third-moment
 194 average) of the drizzle size distribution, which provides the link between the number concentration and
 195 the mass concentration of drizzle droplets in a sample (Hinds, 1999).

196 Adapting the method in Zheng et al. (2022b), the cloud base precipitation rate (R_{CB}) is given by:

$$197 \quad R_{CB}(mm/hr) = 6\pi * 10^{-4} \int_{40\mu m}^{1280\mu m} D_{p,mm}^3 n(D_{p,mm}) U_{\infty}(D_{p,mm}) dD_{p,mm}, \quad (5)$$

198 in order to match the unit conversion, the $D_{p,mm}$ is diameter in unit of mm, $n(D_{p,mm})$ is drizzle number
 199 concentration in every size bin with a unit of $\# m^3 mm^{-1}$, and $U_{\infty}(D_{p,mm})$ is terminal velocity in given
 200 size bin, which is calculated from the full Reynolds number theory as in Pruppacher and Klett (2010).

201 The combined threshold of $N_c > 5 cm^{-3}$ and $LWC_c > 0.01 g m^{-3}$ is used for determining the valid
 202 cloud samples and cloud boundaries (Wood, 2005; Zheng et al., 2022b). The complete cloud vertical
 203 profiles from sub-cloud to the above-cloud are selected during the ACE-ENA and SOCRATES IOPs, in
 204 which the flight strategy includes sawtooth and spiral cloud transects and ramping cloud sampling. The
 205 precipitation conditions are determined by whether samples of $N_d > 0.001 cm^{-3}$ exists below the cloud
 206 base height. In total, the selected numbers of cloud (precipitating cloud) profiles are 18 (13), 26 (13), and
 207 28 (24) for ACE-ENA Sum and ACE-ENA Win, and SOCRATES, respectively. The detailed selected
 208 cloud profiles, with their cloud-base heights (z_t), cloud-top heights (z_b) and cloud thicknesses ($H_c =$
 209 $z_t - z_b$) are listed in Table S1, along with the cloud profile macrophysics.

210 Furthermore, the assessments of ACI are significantly impacted by the MBL dynamic and
 211 thermodynamic conditions. Jones et al. (2011) suggested that the MBL would be in a well-mixed and
 212 coupled condition when the difference in liquid water potential temperature (θ_L) and total water mixing
 213 ratio (q_t) between the bottom of MBL and the inversion layer are less than 0.5 K and 0.5 g kg⁻¹,
 214 respectively. The cases selected for this study feature both coupled and decoupled MBL conditions,
 215 particularly during ACE-ENA Sum, which is characterized by anomalously low MBL heights and
 216 substantial MBL decoupling. Previous studies found that under decoupled conditions the aerosols, CCN,
 217 and moisture sources near the surface are disconnected from the cloud layer aloft, hence exerting a much
 218 less effective impact on cloud microphysics (Zheng et al., 2022a; Christensen et al., 2023; Su et al., 2024).
 219 Therefore, we adapt and modify the metric in Jones et al. (2011) to calculate the sub-cloud coupled layer,
 220 in order to quantify the degree to which aerosols and CCN measured sub-cloud are in a well-mixed state

221 and can represent the actual interaction (or contact) with the cloud layer. In this study, the q_t and θ_L at
222 the cloud base are calculated, and then their vertical variations are examined starting from the altitude of
223 cloud base (z_b) and looking downward. As such, the coupled point height (z_{cp}) is defined as the altitude
224 where the downward vertical changes in q_t and θ_L exceed 0.5 K and 0.5 g kg⁻¹, respectively. Hence, the
225 coupled layer thickness ($H_{cp} = z_t - z_{cp}$) is defined as the layer between the cloud top height (z_t) and
226 coupled point height (z_{cp}), hence the selection of the aerosols and CCN within the below-cloud part of
227 the coupled layer can be viewed as in contact with the cloud. An example of the coupled layer
228 identification is shown in Figure S2. Therefore, the degree of MBL decoupling (D_{cp}) can be quantified
229 as the ratio of the coupled point height (z_{cp}) to the cloud base height (z_b), where $D_{cp} = z_{cp}/z_b$. As
230 shown in Table S1, the ACE-ENA Sum feature with highest degree of decoupling (averaged $D_{cp} = 0.504$),
231 compared to the ACE-ENA Win ($D_{cp} = 0.370$) and SOCRATES ($D_{cp} = 0.277$).

232

233 **2.2 Aerosol properties**

234 The total aerosol number concentrations (N_a) from ACE-ENA and SOCRATES are measured by
235 the airborne Condensation Particle Counter (CPC) models 3772 and 3760A, which count the number of
236 aerosols with diameter (D_p) larger than 3 nm and 11 nm, respectively (Kuang and Mei, 2019;
237 SOCRATES Low Rate Data, 2022). Additionally, the Passive Cavity Aerosol Spectrometer (PCASP)
238 onboard the ACE-ENA aircraft is capable of sizing the aerosol with D_p ranging from 0.1 μm to 3.2 μm
239 (Goldberger, 2020). The ultra-high sensitivity aerosol spectrometer (UHSAS) measures the size-resolved
240 aerosol distribution from 0.06 μm to 1.0 μm during SOCRATES (Uin, 2016). Therefore, the number
241 concentrations of accumulation mode aerosols (N_{ACC} , 0.1 μm -1 μm) can be discerned from the PCASP
242 and UHSAS aerosol size distributions. Aitken mode aerosols (N_{Ait} , < 0.1 μm) from ACE-ENA are
243 retrieved by the fast integrated mobility spectrometer (FIMS), which can size the aerosol down to 9 nm
244 (Olfert et al., 2008), while the N_{Ait} from SOCRATES is limited to 0.06 μm – 0.1 μm due to the limitation

245 of UHSAS. As for the CCN measurements, the ACE-ENA utilized the Dual-Column CCN Counter at
246 two constant supersaturation levels of 0.15% and 0.35% (Uin and Mei, 2019), while the CCN number
247 concentration (N_{CCN}) during SOCRATES was measured under various supersaturation levels from 0.06%
248 to 0.87% using a scanning CCN counter (Roberts and Nenes, 2005). In this study, N_{CCN} at 0.35%
249 supersaturation ($N_{CCN0.35\%}$) is used to ensure a direct comparison between ACE-ENA and SOCRATES.
250 The aerosol measurements are in the temporal resolution of 1 Hz. Note that the aerosol and CCN data
251 are quality-controlled by removing the data point where the $N_c + N_d$ greater than 5 cm^{-3} or N_d greater
252 than 0.01 cm^{-3} , to filter out the contamination of the cloud droplets, and drizzle water splashing.

253 The sub-cloud aerosols and CCN are selected within the below cloud base part of the coupled
254 layer, which is described in the last section, in order to better assess the aerosol-cloud interactions. The
255 above-cloud aerosols and CCN are selected between the cloud top and 200 m above. Note that the
256 selection criteria of 200 m above the cloud top would inevitably induce uncertainty in the cloud top ACI
257 assessment, depending on the vertical trend of the individual aerosol profile. Over the Southeast Atlantic,
258 Gupta et al. (2021) conducted an analysis focusing particularly on the differing impacts when biomass
259 burning aerosols are in contact with marine stratocumulus cloud tops, using 100 m above as the
260 demarcation, versus when they are separated by various distances, and found that significant differences
261 were observed in cloud microphysics, owing to different droplet evaporation and nucleation, compared
262 to profiles which aerosols and cloud layer are separated. That result is in agreement with the modeling
263 sensitivity study over the Eastern North Atlantic by Wang et al. (2020), who found that aerosol plumes
264 can exert impacts on the cloud-top microphysics only when they are in close contact with the cloud layer.
265 During much of the ACE-ENA campaign, nearly constant (and sometimes decreasing) vertical
266 atmospheric profiles of aerosol concentration were observed within a few hundred meters above the
267 cloud top. Aerosol intrusions due to long-range transport, particularly during the summer season, were
268 observed to induce an elevated aerosol layer in higher altitudes that was not in contact with the cloud
269 layer. The frequent new particle formation events during SOCRATES significantly alter the free-

270 troposphere Aitken mode aerosol budget, but the aerosols would need to further subside to impact the
271 cloud (McCoy et al., 2021; Zhang et al., 2023). Note that from previous studies on ACE-ENA and
272 SOCRATES, the aerosol vertical profiles within ~200 m above the cloud layers are typically found to
273 have less variation (Wang et al., 2020; Wang et al., 2021; McCoy et al., 2021; Zhang et al., 2023), hence
274 representing the aerosol layers in contact with the cloud. Hence, the 200 m criterion used in this study
275 provides a sufficient sample size population for statistical analysis.

276

277 **3. Aerosol, cloud, and drizzle properties of selected cases**

278 **3.1 Aerosols and CCN in above- and sub-cloud regimes**

279 The probability density functions (PDFs) of aerosols, CCN, and cloud microphysical properties
280 from selected cases during the ACE-ENA and SOCRATES field campaigns are presented in Figure 1.
281 Notably, the N_a , N_{Acc} and $N_{CCN0.35\%}$ values from the SOCRATES are the highest among the three IOPs,
282 followed by the ACE-ENA Sum and ACE-ENA Win as illustrated in both above-cloud (Figs. 1a-1c) and
283 sub-cloud regimes (Figs. 1d-1f). Such variations can be linked to the different aerosol sources in the
284 ACE-ENA and SOCRATES regions, especially during the summer and winter seasons over the Azores.

285 In the SOCRATES region, according to the previous studies involving back-trajectory analyses,
286 dominant air masses within the MBL primarily originate from the south or from the west, skirting the
287 Antarctic coast (Zhang et al., 2023), while the air masses above the MBL follow a similar transport
288 pathway, they can also originate from the tip of southern Africa and be transported southeast along the
289 warm conveyor belt (McCoy et al., 2021). Above-cloud aerosol $N_{CCN0.35\%}$ values analyzed during
290 SOCRATES (674.6 cm^{-3}) are primarily constituted by the Aitken mode aerosols because the mean N_{Acc}
291 is only 62.5 cm^{-3} . Previously, McCoy et al. (2021) reported average $N_{CCN0.35\%}$ values of 680.69 cm^{-3} ,
292 546.28 cm^{-3} and 465.05 cm^{-3} for mid-troposphere, above and below cloud for the multiple SOCRATES
293 cases, respectively. For individual cases, the above cloud aerosols vary from a couple hundred to over a

294 thousand particles per cubic centimeter (McCoy et al., 2021; Zhang et al., 2023). These aerosols are
295 predominantly produced from the oxidation of biogenic gases, notably dimethyl sulfide (DMS) emitted
296 by marine biological productivity (Sanchez et al., 2018; McCoy et al., 2020). The rising air currents in
297 MBL transport these particles into the free troposphere with dominant aerosol population over the SO
298 (McCoy et al., 2021; Sanchez et al., 2021). Hence, it reinforces the notion that the SO represents a pre-
299 industrial marine environment where the influence of anthropogenic and biomass-burning aerosols is
300 mostly negligible (McCoy et al., 2020, 2021).

301 Conversely, the ENA region experiences aerosols of varied origins, spanning maritime air masses
302 to those heavily influenced by continental emissions from North America or Northern Europe, especially
303 during the summer season (Logan et al., 2014; Wang et al., 2020). The summer air mass back-trajectories
304 within the MBL strongly feature recirculating flow around the Azores high. During the wintertime,
305 however, the air masses predominantly originate in the FT, are transported above the MBL, and are then
306 further entrained to the MBL by large-scale subsidence, indicating less influence from continental
307 pollution (Y. Wang et al., 2021b). During the ACE-ENA Sum, the MBL is enriched by sulfate and
308 carbonaceous particles (Y. Wang et al., 2021b; Zawadowicz et al., 2021). This enhancement is attributed
309 both to local generation from DMS and to the long-range transport from the continental air masses,
310 resulting in the mean N_a of 312.6 cm^{-3} and 301.5 cm^{-3} for above- and sub-cloud regimes, respectively.
311 The ACE-ENA Win exhibits the lowest aerosol and CCN concentrations, predominantly sourced from
312 local maritime influences, and coupled with reduced continental air mass intrusions (Zheng et al., 2018;
313 Y. Wang et al., 2021b).

314 Figure 1a and 1d reveals that there are more above-cloud N_a during the three IOPs than sub-cloud
315 values, especially during the SOCRATES. The higher above-cloud N_a values from the three IOPs are
316 primarily contributed by Aitken mode aerosols because their corresponding N_{Acc} values are much lower
317 (Figs. 1a & 1b). It is interesting to note that the above-cloud $N_{CCN0.35\%}$ values exceed the N_{Acc} for all
318 three IOPs (Figs. 1b & 1c), implying that a significant fraction of Aitken mode aerosols can be activated

319 to become CCN, corroborating findings from earlier studies (McCoy et al., 2021; Zheng et al., 2021).
320 For the sub-cloud regime, the N_a values for SOCRATES and ACE-ENA Win are ~70-80% of their
321 corresponding above-cloud values, and the N_a during ACE-ENA Sum is almost identical to its above-
322 cloud value. Notice that the sub-cloud N_{Acc} values from the three IOPs are more than double the above-
323 cloud N_{Acc} values, and most of the sub-cloud accumulation mode aerosol can be activated to become
324 CCN at SS of 0.35%. It is interesting to note that the higher $N_{CCN0.35\%}$ at the sub-cloud layer during
325 SOCRATES may partially be a result of aerosols being positively impacted by cloud dynamic processes
326 (Figs. 1e & 1f), which is suggested by previous studies (McCoy et al., 2021; Zhang et al., 2023) and will
327 be further discussed in the following paragraphs.

328 To further investigate the above- and sub-cloud aerosol properties from the three IOPs, the aerosol
329 droplet size distributions are analyzed in Figure 2. It is evident that SOCRATES aerosols have the highest
330 concentrations of Aitken mode particles ($D_p = 0.06 - 0.1 \mu\text{m}$, given that the $< 0.06 \mu\text{m}$ is not available
331 from UHSAS) for the above- and sub-cloud regimes. McCoy et al. (2021) and Zheng et al. (2021)
332 identified analogous origins and formations of the above-cloud Aitken mode aerosols over the SO and
333 ENA regions and concluded that these aerosols primarily originate from the nucleation of photo-
334 oxidation products of DMS, notably H_2SO_4 and MSA, in the free troposphere. The differential
335 concentrations can be ascribed to the fact that sea-surface DMS concentrations in the SO are generally
336 higher than those in the ENA region (Aumont et al., 2002; Zhang et al., 2023). Moreover, DMS emissions
337 in the ENA during the summer season surpass those during winter (Zawadowicz et al., 2021). For the
338 accumulation mode aerosols ($0.1 - 1 \mu\text{m}$), the N_{Acc} values for both above- and sub-cloud regimes during
339 SOCRATES decrease monotonically with particle size. The results in Figure 2 further support the finding
340 that Aitken mode aerosols are dominant over the SO. The N_{Acc} values during ACE-ENA show slight
341 uplifts for the small accumulation mode aerosols ($< 0.3 \mu\text{m}$), particularly during the summer, reflecting
342 the signal of potential long-range transport of fine-mode aerosols (Wang et al., 2020; Y. Wang et al.,

343 2021b). Consequently, such comparison reinforces the notion that the SO represents a largely pre-
344 industrial marine environment, wherein the influence of anthropogenic and biomass-burning aerosols is
345 minimal (McCoy et al., 2020, 2021; Zhang et al., 2023).

346 When contrasting the aerosol size distributions in the sub-cloud regime (Fig. 2b) with those in the
347 above-cloud regime, the influence of cloud processing on aerosols is discernibly non-trivial, particularly
348 under the cloud-topped MBL conditions examined in this study. The FT aerosols can be entrained and
349 contribute to the population of Aitken mode aerosols within the MBL, and the sub-cloud aerosols can
350 also be subject to the influence of new particle formation in the upper MBL, though arguably less
351 effective than those within the FT (Zheng et al., 2021). Additionally, in-cloud Brownian capture can lead
352 to a substantial reduction in Aitken mode aerosols (Hudson et al., 2015; Wyant et al., 2022), providing
353 the rationale for the observed decrease in Aitken mode aerosols from above- to the sub-cloud regime,
354 especially for particles smaller than $0.07 \mu\text{m}$. In addition, cloud chemical processing, such as the
355 aqueous-phase condensation of sulfuric acid onto the aerosol cores inside the cloud droplets, is
356 particularly pronounced during the transitioning of Aitken mode aerosols to accumulation mode aerosols
357 (Hudson et al., 2015; Zhang et al., 2023).

358 The larger Aitken mode aerosols ($> 0.07 \mu\text{m}$) in the above- and sub-cloud regimes can effectively
359 grow to accumulation mode aerosols through coagulation and water vapor diffusional growth (Covert et
360 al., 1996), contributing to the elevated accumulation mode aerosol distribution and increased N_{Acc} in the
361 sub-cloud regime. These processes are evident by the decrease of critical supersaturations from above-
362 cloud (between 0.35% - 0.4%) to sub-cloud (between 0.3% - 0.35%) during SOCRATES (Fig. S3)
363 because the aerosol droplet sizes are enlarged and more readily become CCN. Furthermore, the collision-
364 coalescence combines mixtures of large and small cloud droplets, and results in the sub-cloud aerosol
365 residuals shifting towards the larger size upon the drizzle droplet evaporation below the cloud. This
366 partially elucidates the observed increase in the tail-end of the accumulation mode aerosol distribution
367 for all three IOPs. The elevation in sub-cloud coarse mode aerosols observed for both ACE-ENA IOPs

368 (as seen in Fig. 2) can be attributed to the evaporation of collision-coalescence-enlarged drizzle droplets
369 and the intrusion of sea spray aerosols (e.g., sea salt), as illustrated and analyzed based on a summertime
370 case study that exhibits the signal of cloud-processing aerosols (Zheng et al., 2022b), and the long-term
371 aerosol physicochemical properties over the ARM-ENA ground-based observatory (Zheng et al., 2018)
372 particularly during the winter season where the production of sea spray aerosol is prevalent.

373

374 **3.2 Distribution of bulk cloud microphysical properties**

375 The PDFs of MBL cloud microphysical properties (N_c , r_c , LWC_c) derived from aircraft in-situ
376 measurements from the three IOPs are shown in Figures 1g-1i. The mean microphysical properties for
377 the individual cloud profiles are listed in Table S2. The SOCRATES has the highest sub-cloud aerosols
378 and CCN, and subsequently feature a larger number of smaller cloud droplets, given the highest N_c
379 (148.3 cm^{-3}) and smallest r_c ($8 \text{ }\mu\text{m}$) among the three IOPs. These results have further confirmed and
380 reassured our understanding of the aerosol first indirect effect: a larger population of aerosols induce a
381 higher number concentration of small cloud droplets under constrained liquid water content conditions,
382 and thus the MBL clouds reflect more incoming solar radiation (Twomey, 1977). The ACE-ENA Win
383 clouds feature the fewest N_c (70.6 cm^{-3}) and largest r_c ($9.8 \text{ }\mu\text{m}$), while the N_c and r_c (89.4 cm^{-3} and $9 \text{ }\mu\text{m}$)
384 during ACE-ENA Sum fall between the SOCRATES and ACE-ENA Win values. Considering the
385 aerosol competing effect against the available water vapor, the relatively abundant aerosols in
386 SOCRATES might account for the observed narrower r_c distribution, which peaks between $6 - 10 \text{ }\mu\text{m}$.
387 SOCRATES has a lower cloud-layer water vapor mixing ratio (figure not shown) compared to ACE-
388 ENA because the SO region has been observed to contain less precipitable water vapor than the ENA
389 region due to the colder sea surface temperatures (Marcovecchio et al., 2023). Therefore, the aerosol and
390 cloud properties in Figure 1 promise further examination of different cloud microphysical responses to
391 aerosols via the ACI process. Note that the $N_{CCN0.35\%}$ are lower than N_c values during the ACE-ENA

392 Win, which is also confirmed in previous studies (J. Wang et al., 2022; Wang et al., 2023). This
393 interesting phenomenon can potentially be attributed to a combination of factors, including lower MBL
394 aerosol sources, stronger in-cloud coalescence-scavenging depletion of sub-cloud aerosols, and the
395 aircraft snapshots capturing the equilibrium states of aerosols and cloud due to enhanced aerosol
396 activations induced by stronger updrafts during the ACE-ENA Win (J. Wang et al., 2022). This thereby
397 compels further investigation into the potential impacts of precipitation on the MBL CCN budget which
398 is further discussed in Section 4.

399

400 **3.3 Vertical distributions of cloud and drizzle microphysics**

401 The vertical distributions of the cloud and drizzle microphysical properties within the cloud layer
402 from the three IOPs are shown in Figure 3. To ensure the representativeness of the vertical profiles, all
403 the in-cloud samples are vertically smoothed using a triangular moving average method, and are inverse-
404 distance weighted in every 50 m moving altitude windows. Furthermore, the altitude is then normalized
405 by $z_i = \frac{Z - Z_{base}}{Z_{top} - Z_{base}}$, where $z_i = 0$ denotes cloud base and $z_i = 1$ denotes cloud top. Consistent with
406 previous discussions on the bulk microphysics distribution, the mean N_c values from SOCRATES are
407 consistently higher than ACE-ENA Sum, and ACE-ENA Win for the entire cloud layer, with a slight
408 increase ranging from the cloud base to the upper-middle part ($z_i \approx 0.85$) and then decreasing toward
409 the cloud top (Fig. 3a). All r_c values from the three IOPs show a near-linear increase from cloud base to
410 top, with the smallest values observed during SOCRATES and the largest values observed during ACE-
411 ENA Win (Fig. 3b).

412 The warmer and drier air near the cloud top entrains into the cloud layer and further mixes
413 downward, often resulting in the evaporation of small cloud droplets and the shrinking of droplet sizes,
414 which oppose condensational growth (Desai et al., 2021). Decreases in both N_c and LWC_c , and the
415 reduced growth of r_c near the cloud top ($z_i > 0.85$) support signals of cloud-top entrainment mixing

416 during all three IOPs. It is interesting to note that the r_c values from SOCRATES increase monotonically
417 from cloud base to top, while the r_c values from both ACE-ENA Sum, and ACE-ENA Win increase until
418 $z_i \approx 0.8$ and then remain nearly constant, although all of their N_c values (at $z_i \approx 0.8$) decrease towards
419 the cloud top. When dry air entrainment occurs at the cloud top, some of the upper-level smaller cloud
420 droplets will evaporate, which leads to decreases in N_c (Fig. 3a). As a result, the nearly constant r_c values
421 (at $z_i > 0.8$) might represent the equilibrium balance between two competing processes: cloud droplet
422 condensational and collision-coalescence growths and the entrainment mixing evaporation effects.

423 Carrying the distinct discrepancies in the mean values for all layers, the N_c and r_c from ACE-
424 ENA Sum, and ACE-ENA Win clouds experienced similar vertical evolutions as SOCRATES. The
425 increases of r_c (Δr_c) from cloud base to cloud top are $4.03 \mu\text{m}$, $4.78 \mu\text{m}$ and $5.85 \mu\text{m}$, with percentage
426 increases of 66%, 68% and 79%, for SOCRATES, ACE-ENA Sum, and ACE-ENA Win, respectively.
427 Even though, theoretically, the condensational growth effect would be more pronounced on smaller cloud
428 droplets due to their smaller surface area (Wallace and Hobbs, 2006), SOCRATES exhibits the thickest
429 mean cloud thickness but experienced the least r_c increase among the three IOPs. This suggests that high
430 aerosol loading limits the overall growth of the cloud droplet size distribution (DSD) in SOCRATES
431 clouds, while the ACE-ENA Win clouds show the strongest r_c increase, in contrast. This comparison
432 indicates different cloud microphysical responses to aerosol perturbations in the three IOPs, which will
433 be further discussed in Section 4.1. The LWC_c values from the three IOPs are comparable to each other.
434 The vertical distributions of MBL cloud microphysical properties examined in this study are in good
435 agreement with the previous studies conducted on these two field campaigns (Wu et al., 2020a; Y. Wang
436 et al., 2021a; J. Wang et al., 2021; Wang et al., 2023). Cloud adiabaticity is a key parameter as it provides
437 insight into the degree of mixing and microphysical processes occurring within clouds. The sub-adiabatic
438 conditions indicate that the LWC_c is less than what would be expected in an adiabatic scenario, often due
439 to processes such as in-cloud collision-coalescence and entrainment mixing (Hill et al., 2009; Braun et

440 al., 2018; Gao et al., 2020; Wu et al., 2020b). In addition, the cloud adiabaticity is defined as $f_{ad} =$
 441 LWC_c/LWC_{ad} , where the LWC_{ad} denotes adiabatic LWC (Wu et al., 2020b). As shown in Figure S4,
 442 the clouds from all three IOPs feature certain levels of sub-adiabaticity above the cloud base. Considering
 443 the inter-cloud layer-mean f_{ad} , the campaign-mean f_{ad} values are 0.689 ± 0.229 , 0.542 ± 0.143 , and
 444 0.490 ± 0.207 for SOCRATES, ACE-ENA Sum, and ACE-ENA Win, respectively.

445 To quantitatively evaluate the impact of cloud-top entrainment mixing rate on cloud droplets, we
 446 adapt the method of Albrecht et al. (2016), where the cloud-top entrainment rate (w_e) can be expressed
 447 as

$$448 \quad w_e = A_\sigma * \sigma_w / R_{i\sigma}, \quad (6)$$

449 where the turbulence kinetic energy (TKE) dissipation coefficient A_σ is empirically taken as 26 as in
 450 Albrecht et al. (2016), and the $R_{i\sigma}$ is the buoyancy Richardson number calculated by $(g/\theta_0) *$
 451 $(\Delta\theta_v h / \sigma_w^2)$. σ_w denotes the standard deviation of vertical velocities taken near the cloud top ($z_i > 0.9$),
 452 and h is the MBL height. θ_0 is the reference potential temperature and $\Delta\theta_v$ is the virtual potential
 453 temperature difference across the temperature inversion layer above the cloud. Given the valid cloud-top
 454 virtual potential temperature and vertical velocity measurements for the selected cloud cases, the
 455 averaged w_e values are $0.570\pm 0.834 \text{ cm s}^{-1}$, $0.581\pm 0.560 \text{ cm s}^{-1}$, and $0.960\pm 1.127 \text{ cm s}^{-1}$ for SOCRATES,
 456 ACE-ENA Sum, and ACE-ENA Win, respectively. The stronger w_e during ACE-ENA Win might be
 457 induced by the generally weaker cloud-top inversions and stronger near-cloud top turbulence (Fig. 5a),
 458 compared to the summertime when the ENA is dominated by the large-scale high-pressure system (Ghate
 459 et al., 2021). Within the above-cloud inversion layer, the temperature (water vapor mixing ratio)
 460 differences ΔT (Δq) are 1.76 K (-1.75 g kg⁻¹), 1.54 K (-1.66 g kg⁻¹) and 1.48 K (-1.09 g kg⁻¹) for
 461 SOCRATES, ACE-ENA Sum, and ACE-ENA Win, respectively. The virtual potential temperature
 462 differences $\Delta\theta_v$ are 4.90 K, 5.16 K, and 3.82 K, for SOCRATES, ACE-ENA Sum, and ACE-ENA Win,
 463 respectively, indicating relatively dryer entrained airmasses during SOCRATES and ACE-ENA Sum.

464 Considering the near cloud-top proportion of cloud where the LWC_c experienced decrease, the difference
465 in LWC_c (between the cloud-top value and the upper-middle cloud maximum for the mean profiles) for
466 the ACE-ENA Sum (-0.032 g m^{-3}) is higher than the reductions in winter (-0.018 g m^{-3}) and SOCRATES
467 (-0.009 g m^{-3}), albeit that the w_e for ACE-ENA Sum is comparable to SOCRATES, and much lower than
468 ACE-ENA Win values. The warmer and dryer entrained air can partially contribute to the greater LWC_c
469 reduction and the lower f_{ad} (0.39) during the ACE-ENA Sum than those during the ACE-ENA Win
470 ($f_{ad} = 0.45$) and SOCRATES ($f_{ad} = 0.66$) near the cloud top (Fig. S4). For the three IOPs, the N_c and
471 LWC_c exhibit stable trends from the cloud base, followed by noticeable decreases near the cloud top
472 mixing zone, while the changes in r_c trends near the cloud top were not as dramatic as the others. Such
473 characteristics of the cloud microphysics vertical profiles indicate the signal of inhomogeneous mixing,
474 which occurs when dry and warm air mixes unevenly and slowly with the cloud air, hence partially
475 evaporating the cloud droplets (Lehmann et al., 2009; Lu et al., 2011). The results are consistent with
476 previous research results regarding stratocumulus clouds over multiple field campaigns (Brenguier et al.,
477 2011; Jia et al., 2019) and with the findings for selected cases during ACE-ENA (Yeom et al., 2021) and
478 SOCRATES (Sanchez et al., 2020). The near-cloud top r_c profiles ($z_i > 0.8$) for the ACE-ENA cases
479 exhibit fewer increases compared to SOCRATES, which could be possibly attributed to more effective
480 mixing due to the stronger entrainment rate, particularly during ACE-ENA Win, eventually reaching a
481 smaller equilibrium in terms of mean sizes.

482 Figures 3d-3f illustrate the normalized profiles of MBL drizzle microphysical properties. The N_d
483 values from the three IOPs mimic each other, which all maximize at the cloud top and then monotonically
484 decrease toward the cloud base (Fig. 3d), while their LWC_d values follow a similar trend, albeit with
485 relatively large differences (Fig. 3f). In contrast to the N_d and LWC_d trends, the D_{mmd} gradually increase
486 from cloud top to cloud base (Fig. 3e), making physical sense since the drizzle droplets are typically
487 formed near the cloud top and continuously grow via collision-coalescence process while falling. The

488 ACE-ENA Win drizzle D_{mmd} and LWC_d values are distinctively larger than those in ACE-ENA Sum
489 and SOCRATES. It is interesting to note that near the cloud top ($z_i > 0.9$), ACE-ENA Win has
490 comparable N_d but much larger D_{mmd} than the other two IOPs, suggesting that there were more large
491 drizzle embryos formed from large cloud droplets (Fig. 3b) during ACE-ENA Win. It is noteworthy that
492 the D_{mmd} in the lower-half region of the ACE-ENA Win clouds experienced rapid growth from $\sim 80 \mu\text{m}$
493 to $\sim 105 \mu\text{m}$ (Fig. 3e), and this increment of $\sim 25 \mu\text{m}$ contributed to most of the D_{mmd} growth from cloud
494 top to cloud base ($33.5 \mu\text{m}$), indicating a stronger warm-rain process during the winter.

495 In order to further analyze the cloud-to-drizzle conversion processes, the cloud and drizzle DSDs
496 are categorized into four segments based on their relative position within the cloud layer (Fig. 4): upper
497 cloud ($z_i > 0.8$, Fig. 4a), upper-middle cloud ($0.5 \leq z_i < 0.8$, Fig. 4b), lower-middle cloud ($0.2 \leq z_i <$
498 0.5 , Fig. 4c) and lower cloud ($z_i < 0.2$, Fig. 4d). The cloud DSDs ($D_p < 40 \mu\text{m}$) from the three IOPs
499 gradually shift towards larger sizes, moving from the lower to the upper cloud regions. This is
500 accompanied by the narrowing of the cloud DSD ranges, as evidenced by the decline in the relative
501 dispersion of cloud droplets (ε), which is defined as the ratio between the standard deviation and the
502 mean radius of the distribution. At the lower portion of the cloud (Fig. 4d), the relatively greater value
503 of ε represents the co-existence of the newly formed small cloud droplets from recently activated CCN
504 and the sedimentation of larger droplets from the upper sections of the cloud. In addition, the
505 discrepancies in ε between the three IOPs may be attributed to the sub-cloud aerosol differences, which
506 essentially resided in different microphysical regimes. Y. Wang et al. (2021a) stated that higher aerosol
507 loading would lead to increased ε due to the water vapor competition effect, supporting the discrepancy
508 between SOCRATES and ACE-ENA Sum, which can be categorized as a water vapor-limited regime.
509 Meanwhile, the ACE-ENA Win exhibits characteristics of an aerosol-limited regime, in which the cloud
510 DSDs tend to be narrower than in the water-limited regime, due to enhanced droplet growth, and the ε
511 values further decrease with height via the condensational narrowing effect (J. Chen et al., 2018).

512 Notably, for the four cloud portions from cloud base to cloud top, the skewness of summer (winter)
513 cloud DSDs are 0.627 (0.271), 0.358 (0.175), 0.098 (-0.063), and -0.362 (-0.554), respectively. The cloud
514 DSDs during ACE-ENA Win exhibit a more pronounced negative skew (to the left) than those during
515 ACE-ENA Sum, which can be partially attributed to the activation of more sub-cloud coarse mode
516 aerosols becoming larger cloud embryos, as demonstrated in Fig. 2. These coarse mode aerosols, whether
517 from primary production of sea spray or the residuals of evaporated drizzle drops, are more easily
518 activated (or re-activated) into larger cloud droplets when they intrude (or recirculate) into the cloud
519 layer (Hudson and Noble, 2020; Hoffmann and Feingold, 2023). Nevertheless, it is challenging to
520 pinpoint the actual origins of coarse mode aerosols from the perspective of aircraft observational
521 snapshots, thus requiring further numerical modeling work. Ascending within the cloud, the process of
522 water vapor condensation perpetually pushes the DSD towards larger sizes, culminating in a more
523 negatively skewed DSD. Concurrently, the cloud-top entrainment mixing plays a pivotal role in
524 minimizing ϵ in the upper cloud region, as elaborated by Lu et al. (2023).

525 In the upper region of the cloud (Fig. 4a), the ACE-ENA Win clouds contain more cloud droplets
526 close to 40 μm , albeit the mean N_c is lower. This scenario is conducive to the formation of larger drizzle
527 embryos compared to summertime clouds, as depicted in Fig. 3e. In comparison, the SOCRATES clouds
528 feature a pronounced log-normal DSD than during ACE-ENA, as the DSDs peak at $D_p \sim 15 \mu\text{m}$
529 throughout the cloud, and subsequently, the lack of larger cloud droplets resulted in the smaller drizzle
530 embryos near the cloud top. As the newly formed drizzle drops descend and continuously grow through
531 the collision-coalescence process, the drizzle DSDs ($D_p > 40 \mu\text{m}$) are noticeably broadened. From upper
532 to lower cloud regions, the longer tails of the drizzle DSDs expand at the cost of smaller drizzle drops
533 and cloud droplets via the collision-coalescence process. The clouds observed during ACE-ENA,
534 especially in wintertime, contain more large drizzle drops ($D_p > 200 \mu\text{m}$) than SOCRATES, which is
535 reflected in the distinct differences in the vertical D_{mmd} as shown in Fig. 3e.

536 It has been intensively studied that in-cloud turbulence can stimulate collision-coalescence and
537 consequently enhance the drizzle evolution processes (Pinsky et al., 2007; Grabowski and Wang, 2013;
538 Wu et al., 2017; S. Chen et al., 2018). The turbulence strength is characterized by the turbulence kinetic
539 energy (TKE), which is calculated as:

$$540 \quad TKE = \frac{1}{2}(\overline{u'^2} + \overline{v'^2} + \overline{w'^2}), \quad (7)$$

541 where the turbulent perturbations of vertical ($\overline{w'^2}$) and horizontal ($\overline{u'^2}$ and $\overline{v'^2}$) components are
542 calculated as the simple moving variance in a 10 s window centered at the measurement time, without
543 window weighting function, using 1 Hz data for all three IOPs. The w data is confined to an absolute
544 aircraft roll angle of less than 5° (Cooper et al., 2016). Given the average aircraft ground speed of ~ 140
545 m s^{-1} and vertical speed of $\sim 5 \text{ m s}^{-1}$ (Atlas et al., 2020), the smallest resolved wavelength is 140 m. Hence,
546 within the 10s moving window, the ~ 50 m in the integral vertical range is able to resolve the eddies up
547 to ~ 1400 m in size, and preserve the potential of capturing the inertial subrange.

548 As shown in Figure 5, the vertical wind variances (Fig. 5b) in ACE-ENA Win (layer-mean of
549 $0.244 \text{ m}^2 \text{ s}^{-2}$) are generally higher than those in ACE-ENA Sum ($0.153 \text{ m}^2 \text{ s}^{-2}$) and SOCRATES (0.147
550 $\text{m}^2 \text{ s}^{-2}$), while the horizontal wind variances (Figs. 5c & 5d) are comparable between ACE-ENA Sum
551 and ACE-ENA Win but much higher than the SOCRATES, resulting in higher TKE during ACE-ENA.
552 Note that the higher w'^2 near cloud top corresponds to the stronger entrainment rate in ACE-ENA Win.
553 Near the cloud top, turbulence effectively enhances coalescence between the larger cloud droplets,
554 primarily by increasing the relative velocities between droplets (Magaritz-Ronen et al., 2016; Ghate and
555 Cadeddu, 2019), and this is especially true for the vertical component w'^2 of TKE. The horizontal
556 turbulence components, the u'^2 and v'^2 can also play a role in mixing the ambient air masses and
557 contribute to the broadening of DSD (Wu et al., 2017). The use of TKE provides an illustration that in-
558 cloud turbulence during ACE-ENA is stronger than that observed during SOCRATES. That being said,
559 the quantitative evaluation of the turbulent enhancement of collision-coalescence requires access to the

560 eddy dissipation rate, as typically used in model parameterizations (Grabowski and Wang, 2013; Witte
561 et al., 2019). The smallest scales resolvable with the 1 Hz measurement used in this study are on the
562 order of 140 meters, thus capturing only the larger-scale end of the inertial subrange and larger turbulent
563 motions. Consequently, the ability to resolve smaller eddies and turbulent structures, crucial for
564 understanding the energy cascade within the inertial subrange, is limited by coarse spatial and temporal
565 resolutions and aliasing issues (Siebert et al., 2010; Muñoz-Esparza et al., 2018; Kim et al., 2022).
566 Therefore, to fully resolve the spectrum of turbulence and quantitatively examine energy dissipation and
567 mixing processes, access to higher-frequency measurements is required to capture smaller eddies within
568 the inertial subrange (Siebert et al., 2010; Lu et al., 2011; Waclawczyk et al., 2017). Additionally, further
569 quantifying the entrainment-mixing mechanisms also requires high-frequency eddy dissipation and
570 accurate examination of the mixing time scale (Lehmann et al., 2009; Lu et al., 2011) for individual
571 profiles. Though currently beyond the scope of this study, utilizing the high-rate measurements of
572 velocities available from SOCRATES (at 25 Hz) and ACE-ENA (at 20 Hz) to explore those mechanisms
573 further will be of interest to future investigations.

574 Drizzle formation and evolution in the ACE-ENA Win clouds are noticeably stronger than in the
575 other two IOPs, which could be attributed to multiple factors. First, the ambient aerosols and CCN during
576 winter are substantially fewer, featuring clean environments that promote the formation of generally
577 larger cloud droplets due to the availability of more water content per droplet. Larger cloud droplets are
578 more likely to collide and coalesce into drizzle drops, leading to relatively heavier precipitation (Chen et
579 al., 2011; Duong et al., 2011; Mann et al., 2014). Furthermore, deeper cloud layers with mean thickness
580 of (392.4 m) during ACE-ENA Win were observed when compared to the ACE-ENA Sum clouds (336.3
581 m). In a thicker cloud layer with sufficient turbulence, the residence times of large cloud droplets and
582 drizzle drops would become longer, and the chance of collision-coalescence growth could be effectively
583 increased by recirculating the drizzle drops (Brost et al., 1982; Feingold et al., 1996; Magaritz et al.,
584 2009; Ghate et al., 2021). Additionally, the prevalence of winter season precipitation-evaporation-

585 induced MBL cold pools disturbs the MBL thermodynamics and contribute to turbulent mixing (Zuidema
 586 et al., 2017) can provide a strong dynamical forcing perturbation to the warm-rain process (Jenson et al.,
 587 2021; J. Wang et al., 2022; Smalley et al., 2024). The physical hypotheses from previous studies could
 588 potentially serve as the explanation for the phenomena that the ACE-ENA Win drizzle DSD is
 589 sufficiently broadened, and the D_{mmd} is enlarged toward the cloud base. In comparison, although the
 590 SOCRATES exhibits even thicker clouds (487.4 m), the drizzle processes are seemingly suppressed by
 591 the much higher ambient aerosol and CCN concentrations.

592

593 **4 Aerosol-cloud-precipitation interactions (ACPIs)**

594 **4.1 Cloud microphysical responses on aerosols**

595 The impacts of aerosol loading on cloud microphysical properties can be assessed by the aerosol-
 596 cloud interaction (ACI) index, which can be quantified as both:

$$597 \quad ACI_N = \frac{\partial \ln(N_c)}{\partial \ln(N_{CCN,0.35\%})}, \quad (8)$$

598 and

$$599 \quad ACI_r = -\frac{\partial \ln(r_c)}{\partial \ln(N_{CCN,0.35\%})}, \quad (9)$$

600 which emphasizes the cloud microphysical responses to CCN via the relative logarithmic change of N_c
 601 and r_c to the change in $N_{CCN,0.35\%}$ (Feingold et al., 2003; McComiskey et al., 2009). Physically, the ACI
 602 process involves aerosols intruding into the cloud layer, activating as cloud droplets, and subsequently
 603 altering cloud DSD and dispersion (Zheng et al., 2022a&b) under varying water vapor conditions.
 604 Therefore, the cloud microphysical responses within the lower region of the cloud are assessed, which is
 605 the first stage in which the sub-cloud CCN can directly interact with the cloud droplets. Furthermore, the
 606 similarity in the vertical integral of LWC_c (as shown in Fig. 3c) provides comparable liquid water
 607 between three IOPs for the assessment of newly generated cloud embryos from activated CCN because
 608 the ACI_r is normally assessed under a fixed liquid water (Zheng et al., 2020).

609 Considering all the cases from three IOPs with available CCN measurements (some cases without
610 CCN measurements during SOCRATES), the N_c and r_c at the lower cloud ($z_i < 0.2$) are plotted against
611 the sub-cloud $N_{CCN,0.35\%}$ in Figures 6a and 6b, and the ACI indices are calculated as $ACI_{N,CB}$ and $ACI_{r,CB}$
612 (CB denoting the assessment near the cloud base). The ACI indices from three IOPs are in the ACI range
613 of the previous studies in MBL clouds (Twohy et al., 2005; Lu et al., 2009; Diamond et al., 2018) using
614 aircraft in-situ measurements. Note that the availability of valid sub-cloud measurements inevitably
615 limits the sample size, especially for SOCRATES, as shown in Table S2. As shown in Figure 6a, the
616 $ACI_{N,CB}$ for ACE-ENA Win (0.748) is higher than ACE-ENA Sum (0.617), indicating that N_c is more
617 sensitive to the sub-cloud $N_{CCN,0.35\%}$ during the winter. In other words, aerosols intruding into the cloud
618 layer are easily activated to become cloud droplets. The N_c sensitivity for the SOCRATES cloud (0.692)
619 lies between the two ACE-ENA IOPs. The $ACI_{N,CB}$ values from three IOPs are generally higher than the
620 ACI_N values from the layer-mean N_c against the sub-cloud $N_{CCN,0.35\%}$ (not shown). Previous studies have
621 shown that the enhanced vertical turbulence (updraft velocity) can effectively facilitate CCN
622 replenishment into the cloud layer (Hu et al., 2021; Zheng et al., 2022a&b) and increase the actual in-
623 cloud supersaturation (Brunke et al., 2022), thus leading to a more efficient cloud droplet formation,
624 enhancing the $ACI_{N,CB}$. By correlating the mean TKE values with the CCN activation ratio
625 ($N_c/N_{CCN,0.35\%}$) for all individual cloud cases, the three IOPs show moderate but statistically significant
626 correlation coefficients of 0.36, 0.55, and 0.51 for ACE-ENA Sum, ACE-ENA Win, and SOCRATES,
627 respectively. This result reinforces the notion that the CCN activation fractions, particularly during the
628 ACE-ENA Win, are significantly correlated with in-cloud turbulence intensities. Furthermore, more
629 coarse mode aerosols during ACE-ENA Win are also favorable to the activation efficiency (Dusek et al.,
630 2006).

631 As for the r_c responses to CCN (Fig. 6b), the typical Twomey effect, where more CCN compete
632 against available water vapor and result in smaller cloud droplets, is evident by different cloud

633 susceptibility between the three IOPs. SOCRATES features a higher $ACI_{r, CB}$ (0.311), suggesting that an
 634 increase in $N_{CCN, 0.35\%}$ can result in a significant decrease in r_c , compared to ACE-ENA Sum (0.206) and
 635 ACE-ENA Win (0.263). Although the absolute range of variation for r_c during SOCRATES is smaller,
 636 the slope is much deeper (Fig. 6b). Recall that the sub-cloud $N_{CCN, 0.35\%}$ during SOCRATES is generally
 637 higher than ACE-ENA and contains more small-sized aerosols (as indicated in Fig. 2b). Consequently,
 638 after activation, the lower part of the cloud exhibits a higher number of smaller cloud droplets for
 639 SOCRATES, as shown in Fig. 4d. Therefore, as more CCN intrudes into the cloud, the competition for
 640 water vapor among newly-activated cloud droplets becomes more pronounced, given similar water
 641 availability. In contrast, the presence of larger cloud droplets near the cloud base, whether activated from
 642 coarse-mode aerosols or remaining as residuals from collision-coalescence, would elevate the r_c
 643 especially under the relatively less CCN condition, hence inevitably dampening the $ACI_{r, CB}$ during ACE-
 644 ENA. However, a more comprehensive investigation into the cloud microphysical responses to CCN
 645 intrusions under a larger range of various water supply conditions, and further untangling the ACI from
 646 the meteorological influences, will require additional aircraft cases from more field campaigns. Examples
 647 include, the VAMOS Ocean-Cloud-Atmosphere-Land Study (VOCALS; Wood et al., 2011), the Cloud
 648 System Evolution over the Trades (CSET; Albrecht et al., 2019), the ObseRvations of CLouds above
 649 Aerosols and their intEractionS (ORACLES; Redemann et al., 2021), and the Aerosol Cloud
 650 meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE; Sorooshian et al., 2019).
 651 Note that the $ACI_{r, CB}$ values in Figure 6b are also larger than the results from the layer-mean r_c against
 652 sub-cloud $N_{CCN, 0.35\%}$, since the layer-mean microphysics is more subject to the cloud droplet evolution
 653 processes such as condensational growth and collision-coalescence.

654 To investigate the ACI indices at the upper level of the cloud, the N_c and r_c at the upper cloud
 655 ($z_i > 0.8$) are plotted against the above-cloud $N_{CCN, 0.35\%}$ in Figures 6c and 6d, and the ACI indices are
 656 calculated as $ACI_{N, CT}$ and $ACI_{r, CT}$ (denoting the assessments near the cloud top). Compared to the

657 $ACI_{N,CB}$ and $ACI_{r,CB}$, the $ACI_{N,CT}$ and $ACI_{r,CT}$ are much weaker, especially for $ACI_{r,CT}$, as the near cloud
658 top droplets are too large for above-cloud aerosols to exert a significant influence on r_c (Diamond et al.,
659 2018; Gupta et al., 2022). The weaker cloud top N_c dependence on the $N_{CCN,0.35\%}$ could be due to the
660 legacy of the sub-cloud CCN impacts on N_c being conveyed to the cloud top. This occurs because FT
661 aerosols and CCN can be entrained into the MBL before and during the cloud process, as observed in the
662 assessment of inter-cloud cases. Note that the LWC_c near the cloud top for the three IOPs are not
663 comparable to each other, which might also induce uncertainty in the near-cloud-top ACI assessment.
664 These weaker relationships support the notion that although the aerosols entrained into the upper-cloud
665 region can affect the cloud microphysics to a certain degree, the effects are less pronounced than those
666 from the sub-cloud aerosols (Diamond et al., 2018, Wang et al., 2020) because the MBL cloud N_c and r_c
667 variations are dominated by the condensational growth, collision-coalescence, and entrainment mixing
668 processes near the cloud top.

669

670 **4.2 Precipitation susceptibility**

671 The precipitation susceptibility relies on the assessment of relative responses in the precipitation
672 rate to the change in N_c (Feingold and Seibert, 2009; Sorooshian et al., 2009), which is defined as:

$$673 \quad S_o = -\frac{\partial \ln(R_{CB})}{\partial \ln(N_c)}, \quad (10)$$

674 where the R_{CB} is the cloud base precipitation rate calculated in section 2 (equation 5). By incorporating
675 all the cloud cases, including both precipitating and non-precipitating clouds (the R_{CB} can also be
676 calculated based on the drizzle DSD near the cloud base), the S_o accounts for the impact of cloud droplets
677 on the potential precipitation ability of the cloud (Terai et al., 2012).

678 As shown in Figure 7a, the R_{CB} values generally have a negative correlation with increased layer-
679 mean N_c for all three IOPs. The S_o values are 0.979, 1.229, and 1.638, with the absolute values of
680 correlation coefficients being 0.33, 0.29, and 0.45 for SOCRATES, ACE-ENA Sum, and ACE-ENA Win,

681 respectively. The regression relationships are statistically significant with $p < 0.05$ for all three IOPs.
682 These correlation coefficient values fall within the reasonable range found in previous studies on
683 precipitation susceptibility in MBL stratus and stratocumulus clouds (Jung et al., 2016; Gupta et al.,
684 2022), and indicate statistically significant dependences of R_{CB} on N_c . Previous study by Terai et al.
685 (2012) found that the S_o values decrease with the increasing cloud thickness over the southeast Pacific,
686 and Jung et al. (2016) found that the S_o is more pronounced within the medium-deep clouds with
687 thickness ~ 300 - 400 m in the MBL stratocumulus over the eastern Pacific. Gupta et al. (2022) found that
688 the S_o values are generally higher under low ambient N_a condition in the southeastern Atlantic MBL. In
689 this study, R_{CB} for the ACE-ENA Win is more susceptible to the layer-mean N_c than the ACE-ENA Sum
690 and SOCRATES, which can be partially attributed to the existence of more large drizzle drops (as shown
691 in Fig. 4d) near the cloud base in ACE-ENA Win. As previously discussed, the ACE-ENA Win featured
692 enhanced collision-coalescence suggested by the stronger in-cloud turbulence, and a possible drizzle-
693 recirculating process as indicated by the previous study. And such mechanisms might explain the low N_c
694 conditions with more large drizzle drops, leading to the increase of S_o values during ACE-ENA Win. In
695 comparison, the aerosol of SOCRATES is largely composed of fine Aitken mode aerosol, which results
696 in smaller cloud droplets. Thus, collision-coalescence is ineffective during SOCRATES, which leads to
697 the relatively narrower drizzle DSDs, where the warm-rain processes are suppressed, and in turn,
698 diminishing the sensitivity of R_{CB} to N_c (Stevens and Feingold, 2009; Fan et al., 2020; Gupta et al., 2022).

699 It is well known that the R_{CB} can be parameterized or predicted by assuming an approximate
700 relation with N_c and cloud thickness (H_c), which is usually parameterized in the form of $R_{CB} \propto c H_c^3 N_c^{-1}$
701 (Lu et al., 2009; Kang et al., 2024). Following the same method, we derive the relationships from three
702 IOPs in Figure 7b, where the R_{CB} are positively (negatively) proportional to the H_c (N_c), with the
703 exponential parameters in the range of the typical values in the MBL clouds (Comstock et al., 2004; van
704 Zanten et al., 2005; Lu et al., 2009). The statistical coefficient of determination (R^2) values of R_{CB} against

705 H_c (N_c) are 0.696 (0.177), 0.419 (0.212) and 0.165 (0.295), for the ACE-ENA Sum, winter and
706 SOCRATES, respectively, suggesting that the R_{CB} in ACE-ENA clouds may be more determined by H_c ,
707 while the R_{CB} in SOCRATES clouds could be less dependent on both H_c and N_c . Note that the
708 relationship for SOCRATES in this study reveals a similar R_{CB} dependence on N_c but a smaller
709 dependence on the cloud thickness than the study by Kang et al. (2024), who concluded a relationship of
710 $R_{CB} = 1.41 \times 10^{-9} H_c^{3.1} N_a^{-0.8}$, based on the rain rate retrieved from radar and lidar measurements and
711 the aerosol concentration also from the SOCRATES. The discrepancies are possibly due to the different
712 sample selections and different methods in the R_{CB} calculation. Note that the mean cloud thicknesses of
713 ACE-ENA Sum (336.3 m), ACE-ENA Win (392.4 m) and SOCRATES (487.4 m), are within the
714 thickness range found to exhibit stronger S_o (Terai et al., 2012; Jung et al., 2016; Gupta et al., 2022).

715

716

717 **4.3 Drizzle impacts on sub-cloud CCN and implication to ACI**

718 Multiple studies on the MBL clouds have concluded that the in-cloud drizzle formation and
719 evolution processes can effectively impact the sub-cloud CCN budgets via the coalescence-scavenging
720 effect (Wood, 2006; Wood et al., 2012; Diamond et al., 2018; Zheng et al., 2022b; Zhang et al., 2023).
721 Drizzle drops are formed and grow via the collision-coalescence process by collecting cloud droplets and
722 small drizzle drops, resulting in the consumption of CCN (the precursor of cloud droplet), but in the
723 meantime, the in-cloud N_c can be continuously buffered by the sub-cloud CCN replenishment. Although
724 the sub-cloud aerosols (especially in large size) would be added if the drizzle fell and evaporated outside
725 the cloud, the increment cannot compensate for the loss. Therefore, the net result of the whole process is
726 usually presented as the depletion of sub-cloud CCN residuals, and such drizzle modulation on the CCN
727 budget could be substantial in moderate-to-light drizzles or even non-precipitating clouds, depending on
728 the collision-coalescence efficiency (Feingold et al., 1996; Wood, 2006; Kang et al., 2022).

729 The CCN loss rate due to the coalescence-scavenging effect can be calculated as:

$$730 \quad L_{CCN} = -\frac{K H_c}{H_{cp}} * N_c * R_{CB}, \quad (11)$$

731 where the constant K ($2.25 \text{ m}^2 \text{ kg}^{-1}$) denotes the drizzle collection efficiency (Wood et al., 2006; Diamond
732 et al., 2018). H_c is cloud thickness, and H_{cp} is the coupled layer thickness to ensure the change in the
733 cloud layer can be sufficiently conveyed throughout the layer. The calculated CCN loss rate for individual
734 cases is listed in Table S2. Considering all cloud (precipitating cloud) scenarios, the mean CCN loss rates
735 are $-7.69 \pm 13.96 \text{ cm}^{-3} \text{ h}^{-1}$ ($-10.45 \pm 15.56 \text{ cm}^{-3} \text{ h}^{-1}$), $-6.29 \pm 11.65 \text{ cm}^{-3} \text{ h}^{-1}$ ($-12.11 \pm 14.64 \text{ cm}^{-3} \text{ h}^{-1}$), and
736 $-4.94 \pm 7.96 \text{ cm}^{-3} \text{ h}^{-1}$ ($-5.58 \pm 8.43 \text{ cm}^{-3} \text{ h}^{-1}$) for ACE-ENA Sum, ACE-ENA Win and SOCRATES,
737 respectively. As the results indicate, the ACE-ENA clouds experience a more substantial sub-cloud CCN
738 loss than SOCRATES, especially in wintertime precipitating clouds. Recall that the assessment of
739 $ACI_{r,CB}$ relies on the relative changes of r_c and N_{CCN} , while the different L_{CCN} for individual cases can
740 result in the shrinking of the N_{CCN} variation ranges (imagine the abundant CCN are depleted by the
741 coalescence-scavenging). In other words, the given change in r_c corresponds to a narrowed change in
742 N_{CCN} . Mathematically speaking, the assessment of $ACI_{r,CB}$ depends on the ratio of the numerator (change
743 in r_c) and the denominator (change in N_{CCN}). Under the circumstances of substantial cloud-processing to
744 the aerosols, the altered sub-cloud CCN budgets are reflected as a smaller denominator, versus the less
745 altered numerator, hence mathematically presented as an enlarged $ACI_{r,CB}$. Therefore, the coalescence-
746 scavenging effect can not only deplete the sub-cloud CCN, but also quantitatively amplify the assessment
747 of cloud microphysics susceptibilities (Feingold et al., 1999; Duong et al., 2011; Jung et al., 2016; Zheng
748 et al., 2022b). In order to examine the potential impact of the aforementioned processes on the ACI
749 assessment, a sensitivity analysis is conducted by simply retrospectively the sub-cloud $N_{CCN0.35\%}$
750 according to their L_{CCN} . For each retrospective time step ΔT , the r_c values are held unchanged, and the
751 retrospective $N_{CCN0.35\%}$ values for individual cloud cases are given by $N_{CCN0.35\%} - L_{CCN} * \Delta T$, and then
752 the $ACI_{r,CB}$ can be recalculated. Note that assuming a constant r_c value over time inevitably induces

753 uncertainty and biases, as it does not consider the microphysical processes affecting the cloud droplet
754 mean size. However, previous numerical experiments show that the noticeable impact on the cloud mean
755 radius through collision-coalescence necessitates a high degree of CCN depletion, and the quantified
756 percentage changes in droplet mean sizes are several times less than the changes in CCN depletion
757 (Feingold et al., 1996). Hence, the retrospective method, from an observational snapshot point of view,
758 provides a direction that enables the assessment of $ACI_{r,CB}$ as if before the sub-cloud aerosols and CCN
759 are scavenged by in-cloud coalescence-scavenging and precipitation scavenging processes.

760 As shown in Figure 8, the $ACI_{r,CB}$ values tend to decrease with the retrospective time, which
761 indicates the retrospective CCN variation range is enlarged and counteracting the coalescence-
762 scavenging amplification. The detailed illustration of the different $ACI_{r,CB}$ calculated from the scattered
763 r_c and sub-cloud $N_{CCN0.35\%}$ is shown in Figure S5. Note that the $ACI_{r,CB}$ decreasing rates for the
764 precipitating clouds (Fig. 8b) are not as strong as for all clouds because the non-precipitating clouds have
765 smaller L_{CCN} largely due to weaker collision-coalescence. Hence, the retrospective period used here
766 might quickly exceed the actual time of cloud-processing to become effective on aerosol and CCN. In
767 other words, the actual time needed to trace back to the sub-cloud CCN concentration before they were
768 cloud-processed, is shorter than the retrospective time tested here in Figure 8. This results in the faster
769 decrease of $ACI_{r,CB}$ in the non-precipitating cloud. The retrospective of the sub-cloud CCN budget will
770 yield an alternative assessment of ACI, assuming that the drizzle processes have not yet significantly
771 impacted the sub-cloud CCN budget, especially for the assessment under the precipitating clouds.
772 However, examining the exact precipitating timing is challenging since the aircraft provides a snapshot
773 of the cloud and aerosol information. Thus, this retrospective study only provides a possible direction,
774 and the result should be interpreted with caution.

775

776

777 5. Summary and Conclusions

778 Based on the aircraft in-situ measurements during ACE-ENA and SOCRATES, the vertical
779 distributions and the evolutions of the aerosol, cloud, and drizzle properties are investigated under cloud-
780 topped MBL environments. The aerosols and CCN from SOCRATES are the highest among the three
781 IOPs, followed by ACE-ENA Sum, and ACE-ENA Win in descending order in both above- and sub-
782 cloud regimes. The differences can be attributed to the differences in aerosol size distributions between
783 ACE-ENA and SOCRATES, which are largely due to the aerosol sources in those regions. The
784 SOCRATES features the pre-industrial natural environment enriched by aerosols from marine biological
785 productivity and without the contamination of anthropogenic aerosols, while the ACE-ENA features the
786 aerosols from varied sources, including maritime and continental emissions, with distinct seasonal
787 variations. Examining the aerosol size distributions in sub-cloud versus above-cloud regimes manifests
788 the significant influence of cloud processing on aerosols. According to previous studies, physical
789 processing like in-cloud Brownian capture can reduce Aitken mode aerosols, while the chemical
790 processes transform Aitken mode aerosols to larger sizes, moving them toward the accumulation mode.
791 In addition, the in-cloud coalescence processes could also shift sub-cloud aerosol residuals to larger sizes,
792 as multiple aerosols combine into a single aerosol core inside the cloud droplet during collision-
793 coalescence. Those physical mechanisms could potentially explain the observed increase in the tail of
794 the aerosol size distribution for all IOPs, and it will be of interest for future research to prove such
795 hypotheses.

796 As for the cloud and drizzle properties, the SOCRATES clouds feature a larger number of smaller
797 cloud droplets than the ACE-ENA Sum and ACE-ENA Win clouds, with the r_c growth (and percent
798 increases), from cloud base to top, being $4.03 \mu\text{m}$ (0.66%), $4.78 \mu\text{m}$ (0.68%), and $5.85 \mu\text{m}$ (0.79%) for
799 SOCRATES, ACE-ENA Sum, and ACE-ENA Win, respectively. The cloud-top entrainment mixing is
800 evident in the observed decline of both N_c and LWC_c near the cloud top. The mean cloud-top entrainment
801 rates (w_e) are $0.570 \pm 0.834 \text{ cm s}^{-1}$, $0.581 \pm 0.560 \text{ cm s}^{-1}$, and $0.960 \pm 1.127 \text{ cm s}^{-1}$ for SOCRATES, ACE-

802 ENA Sum, and ACE-ENA Win, respectively. The strongest w_e during ACE-ENA Win is a result of
803 weaker cloud-top inversions and stronger near-cloud-top turbulence. The values of the TKE for three
804 IOPs are generally within the ranges of previous studies (Atlas et al., 2020; Ghate et al., 2021). For drizzle
805 vertical distribution, N_d from the three IOPs all exhibit decreases from cloud top to cloud base, while
806 D_{mmd} are in opposite directions with a maximum at the cloud base. The ACE-ENA Win clouds feature
807 more prominent drizzle formation and evolution owing to the combined effects of relatively cleaner
808 environment, deeper cloud layer, and slightly stronger in-cloud vertical turbulence, which is speculated
809 to substantially enhance the collision-coalescence and the drizzle re-circulating processes, compared to
810 the other two IOPs. Satellite retrievals of droplet number concentration heavily rely on the adiabatic
811 cloud assumption and are usually given as a constant of $f_{ad} = 0.8$, the in-situ observational evidence
812 found in this study further confirms the unrealistic nature of this assumption. It will be of interest to
813 utilize multiple aircraft measurements (campaigns) to explore the variability of MBL cloud and drizzle
814 microphysical properties over different marine regions. This can help examine potential predictors for
815 f_{ad} , which will aid in satellite-based retrievals and aerosol-cloud interaction assessments (Painemal and
816 Zuidema, 2011; Grosvenor et al., 2018; Painemal et al., 2021).

817 Comparing the seasonality of cloud-base precipitation rate (R_{CB}) during ACE-ENA, more cases
818 with large observed R_{CB} during the winter season, which is consistent with J. Wang et al. (2022). Notably,
819 the sensitivity of R_{CB} to N_c is more pronounced for the ACE-ENA during both winter (with precipitation
820 susceptibility $S_o = 1.638$) and summer ($S_o = 1.229$) compared to the SOCRATES ($S_o = 0.979$). This
821 could be possibly hypothesized as the result of turbulence-driven in-cloud droplet interactions, which
822 could result in much higher R_{CB} induced by larger drizzle drops near the cloud base for ACE-ENA,
823 especially under low N_c conditions. Furthermore, R_{CB} can be approximated by a relationship involving
824 N_c and H_c , as suggested in prior research. The relationships established in this study indicate that the S_o
825 in ACE-ENA clouds can be partially determined by H_c , while in SOCRATES clouds the S_o is less

826 influenced by H_c and N_c . Based on the physical mechanisms found in the previous study, a possible
827 hypothesis can be leveraged to explain the observed results. That is, the combination of a deeper cloud
828 layer and relatively lower ambient aerosol concentration, eventually leading to stronger drizzle
829 production and evolution during ACE-ENA, especially during the winter season, results in more robust
830 precipitation susceptibility. And further numerical simulations and experiments are warranted to prove
831 this hypothesis. Note that considering the combined factors of aerosol loadings, cloud morphology and
832 thicknesses, and the assessment methodology, the derived S_o values in this study are generally higher (or
833 close to the upper end) compared to previous studies (Lu et al., 2009; Duong et al., 2011; Terai et al.,
834 2012; Jung et al., 2016; Gupta et al., 2022).

835 The investigations of the ACI via the $ACI_{N,CB}$ and $ACI_{r,CB}$ indices reveal that during the ACE-
836 ENA Win, N_c is more sensitive to changes in $N_{CCN0.35\%}$, indicating aerosols more readily activate to
837 become cloud droplets compared to those in the ACE-ENA Sum, which is consistent with the previous
838 assessment by J. Wang et al. (2022) on the seasonal dependency of the relationship between N_c and
839 aerosols. One influencing factor is the strong dynamic mechanism that speeds up the infusion of CCN
840 into the cloud layer, thus aiding droplet formation. The moderate but statistically significant correlation
841 coefficients between the CCN activation fractions and the TKE agree with a previous study that found
842 the local activation fraction of CCN to be strongly associated with increased updrafts (Hu et al., 2021).
843 Furthermore, the presence of larger aerosols during ACE-ENA Win enhances the droplet activation
844 process. The SOCRATES IOP highlights a higher $ACI_{r,CB}$, indicating a pronounced decrease in r_c with
845 increasing $N_{CCN0.35\%}$. The $ACI_{r,CB}$ in ACE-ENA is dampened by the presence of more large cloud
846 droplets near the cloud base, particularly under relatively higher $N_{CCN0.35\%}$. However, the combined
847 effect of the relatively cleaner environment and sufficient water vapor results in stronger cloud
848 microphysical responses during the ACE-ENA wintertime than in the summertime. Note that the ACI
849 indices from this study lie in the higher end of the ACI ranges estimated via remote sensing (McComiskey

850 et al., 2009; Dong et al., 2015; Zheng et al., 2022a) possibly because the aircraft assessment of ACI is
851 based on measurements where the aerosols are in direct contact with the cloud layer. Arguably, the
852 assessment of N_c responses to $N_{CCN0.35\%}$ would inevitably be affected by the collision-coalescence
853 process near the cloud base, where simultaneously, the CCN replenishment buffers the N_c and the
854 collision-coalescence process depletes N_c . Hence, finding a layer where these two effects maintain a
855 dynamic balance in N_c might aid in a more accurate assessment and more fundamental understanding of
856 the ACI, which might be revealed by the LES or parcel model simulations.

857 Additionally, the in-cloud drizzle formation and evolution processes significantly influence the
858 sub-cloud CCN budgets via the coalescence-scavenging effect, which can potentially exaggerate the
859 assessment of cloud microphysics susceptibilities. Based on the CCN loss rate (L_{CCN}) from ACE-ENA
860 and SOCRATES, a sensitivity analysis is performed focusing on retrospectively adjusting the sub-cloud
861 CCN according to their L_{CCN} . Results showed that this adjustment led to a decreased $ACI_{r,CB}$,
862 highlighting the significance of the coalescence-scavenging process on the ACI assessment. However,
863 due to the fact that aircraft only provide a snapshot of the clouds and aerosol information, determining
864 the precise drizzle timing for the individual cloud is challenging. Hence, findings from this retrospective
865 approach provide only a direction or theory, and should be taken cautiously. Nevertheless, pursuing
866 further modeling experiments on this matter may be worthwhile. For example, the exact drizzling time
867 could be pinpointed within a model using an Eulerian framework or traced using a Lagrangian framework.
868 Nevertheless, the CCN adjustment could more accurately reflect the true characteristics of the cloud and
869 the MBL CCN budget, potentially aiding in a more precise assessment of ACI. Therefore, future research
870 would focus on model simulations of MBL clouds from ACE-ENA and SOCRATES and further assess
871 the modeled ACI under the observational constraints, as well as the continuous development of the warm
872 rain microphysical parameterizations, in order to aid in the better represent the MBL clouds in multiple
873 regions.

874

875 *Data availability.* The ACE-ENA field campaign data can be accessed from the Department of Energy
876 Atmospheric Radiation Measurement data archive ([https://iop.archive.arm.gov/arm-iop-](https://iop.archive.arm.gov/arm-iop-file/2017/ena/aceena/)
877 [file/2017/ena/aceena/](https://iop.archive.arm.gov/arm-iop-file/2017/ena/aceena/)). The SOCRATES field campaign data are publicly archived on the National
878 Center for Atmospheric Research (NCAR) Earth Observing Laboratory
879 (https://data.eol.ucar.edu/master_lists/generated/socrates/).

880

881 *Author contributions.* The original idea of this study is discussed by XZ, XD, and BX. XZ performed the
882 analyses and wrote the manuscript. XZ, XD, BX, TL, and YW participated in further scientific
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884

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892

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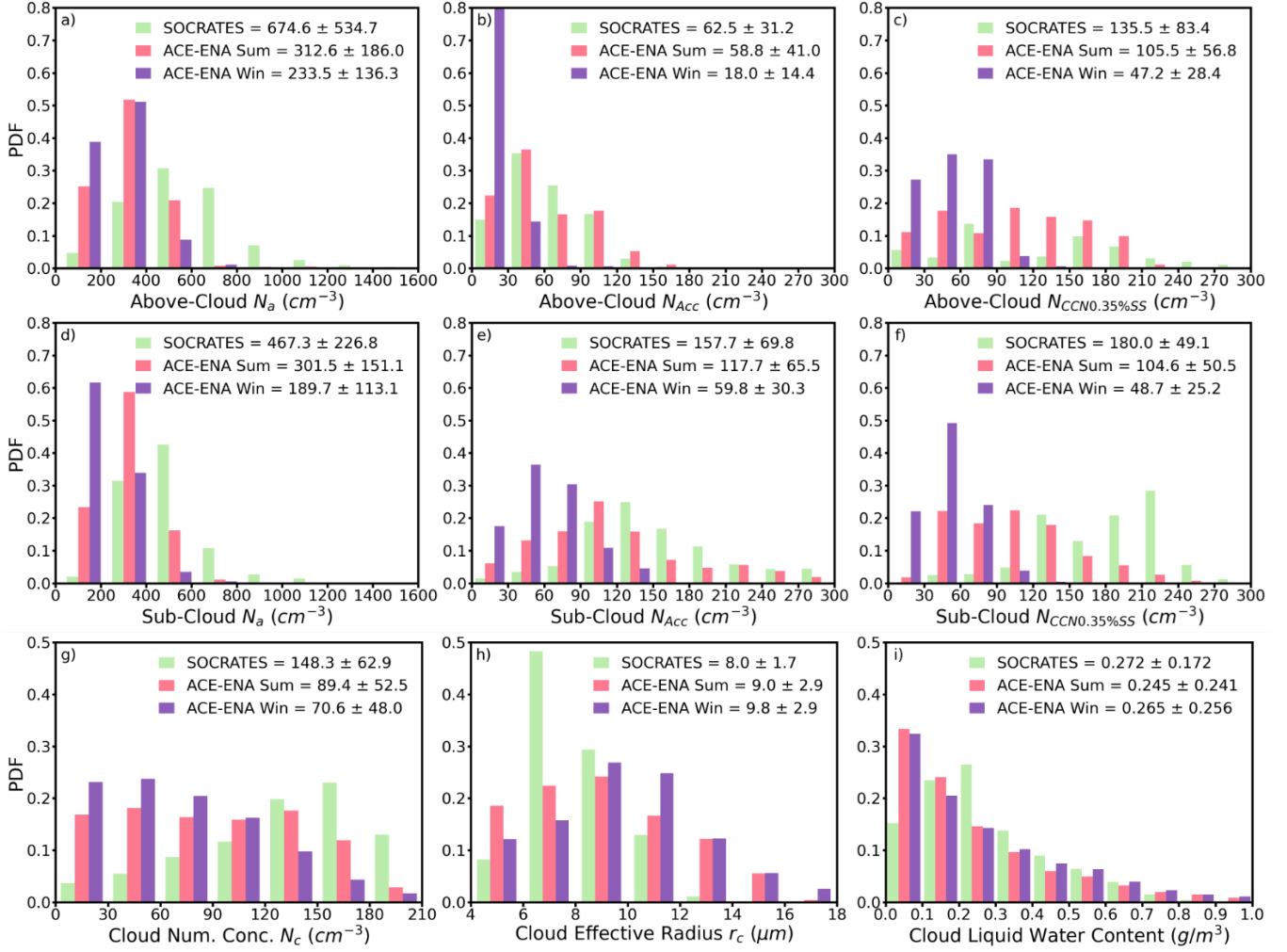


Figure 1. Probability Density Functions (PDFs) of N_a , N_{ACC} and $N_{CCN0.35\%}$ in the above-cloud (a, b, c) and sub-cloud (d, e, f) regimes; and the cloud microphysical properties of N_c (g), r_c (h), and LWC_c (f) within cloud layer. The statistical metrics in the legends denote the mean and standard deviation values for all samples in three IOPs. The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

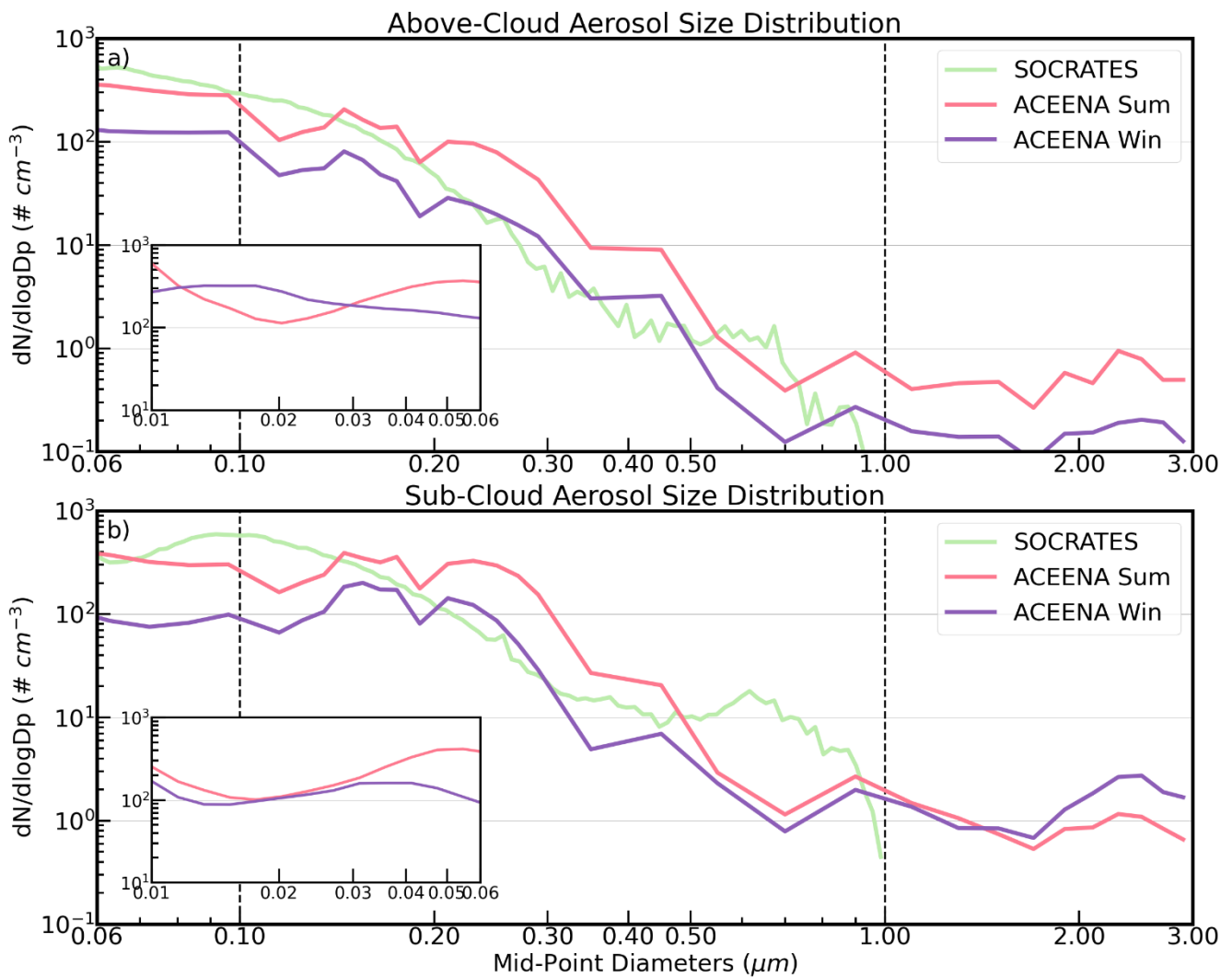


Figure 2. Aerosol size distributions ($D_p = 0.06 - 3 \mu\text{m}$) for above-cloud (a) and sub-cloud (b) regimes. The vertical dashed line at $D_p = 0.1 \mu\text{m}$ and at $D_p = 1 \mu\text{m}$ denotes the demarcations between Accumulation mode, Aitken mode and Coarse mode aerosols. The inner plots denote a smaller range of Aitken mode size distribution ($D_p = 0.01 - 0.06 \mu\text{m}$) available from ACE-ENA. The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

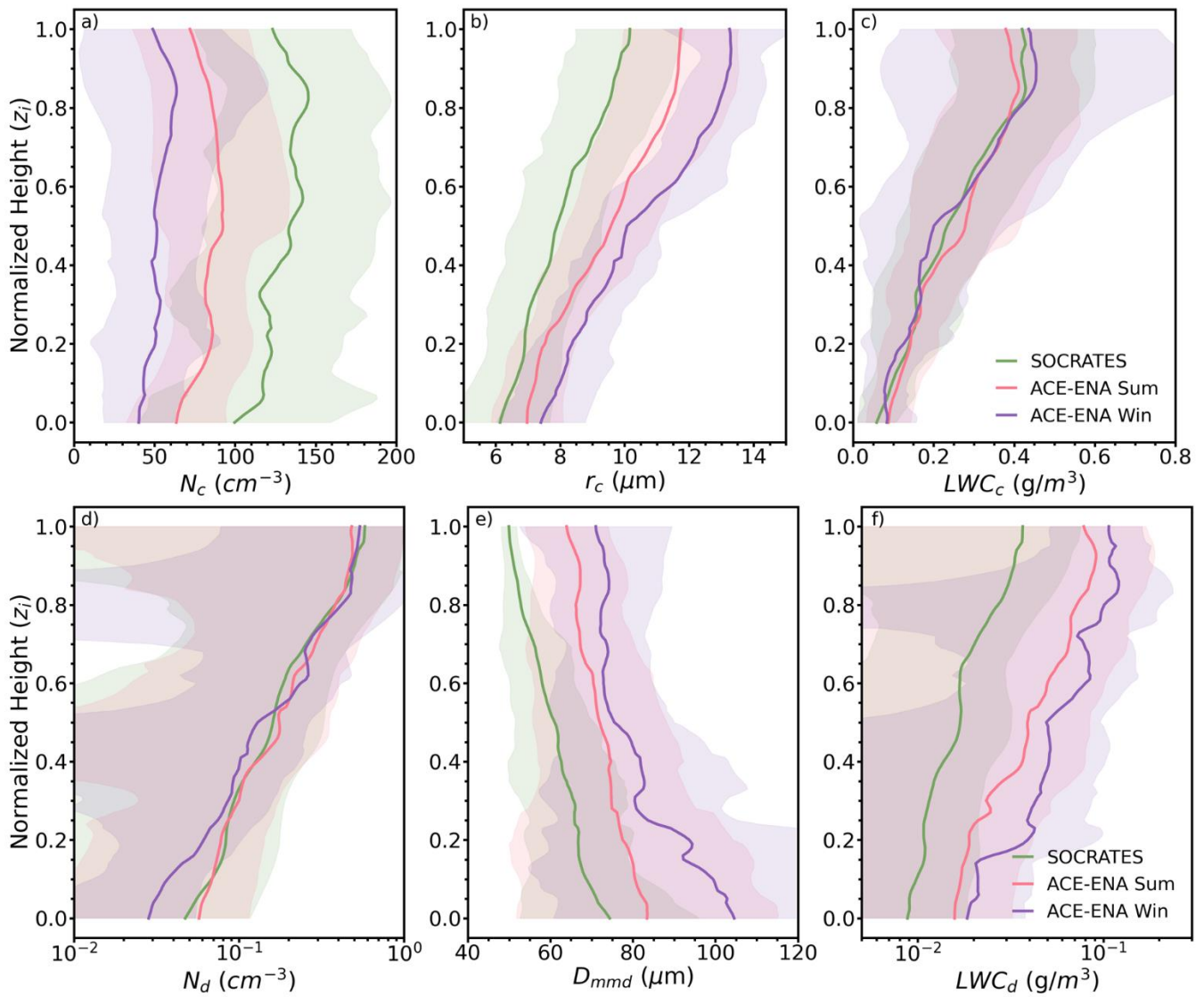
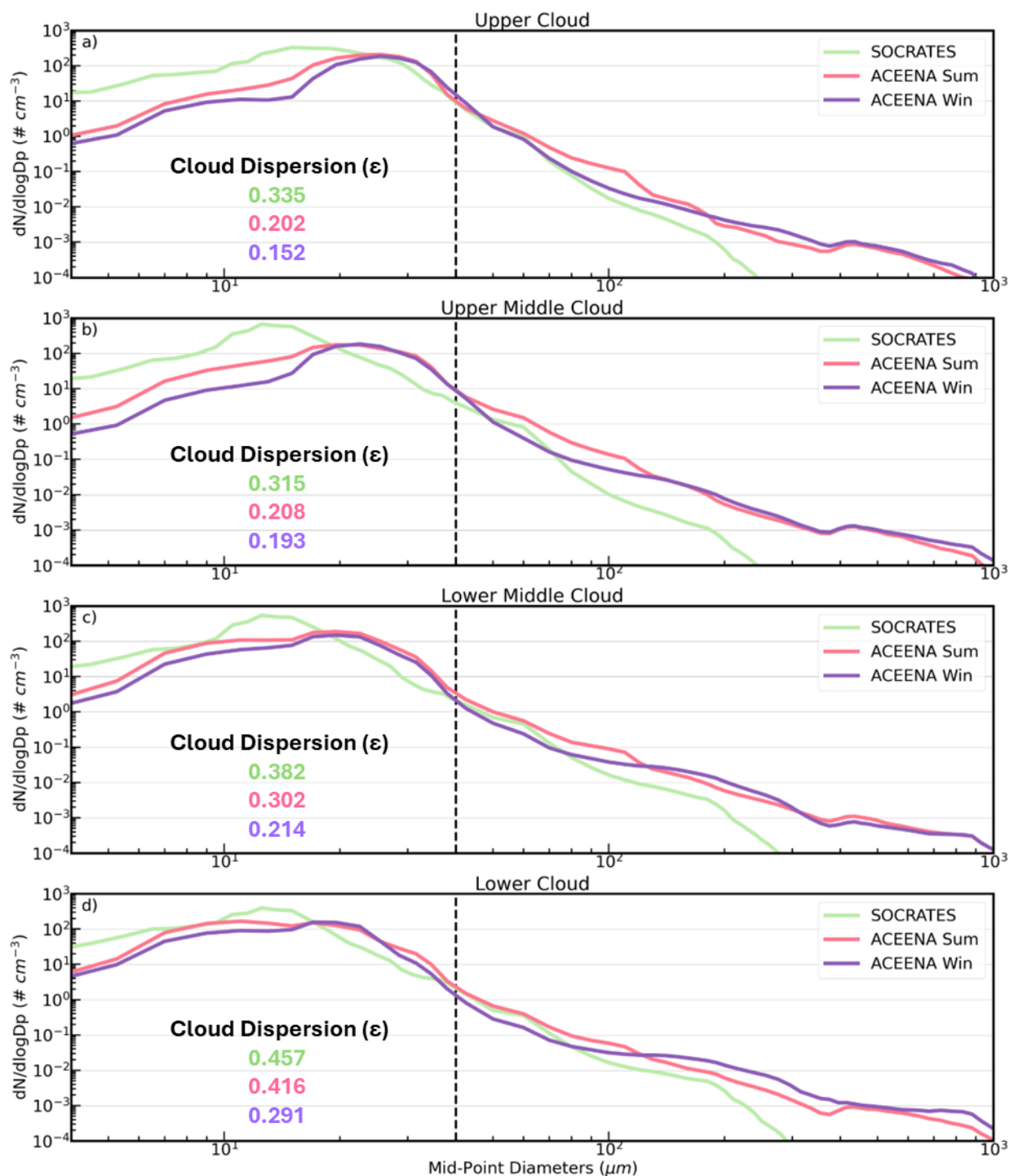


Figure 3. Vertical distributions of N_c (a), r_c (b), LWC_c (c), N_d (d), D_{mmd} (e), and LWC_d (f). Here the $z_i = 0$ denotes cloud base and $z_i = 1$ denotes cloud top. Shaded areas denote the inter-cloud-case standard deviations. The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.



1275 **Figure 4.** Cloud and drizzle size distributions for a) upper cloud ($z_i > 0.8$), b) upper-middle cloud ($0.5 \leq$
 1276 $z_i < 0.8$), c) lower-middle cloud ($0.2 \leq z_i < 0.5$) and d) lower cloud ($z_i < 0.2$). The vertical dashed
 1277 line at $D_p = 40 \mu\text{m}$ denotes the demarcation between cloud droplets and drizzle drops. The ACE-ENA
 1278 Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

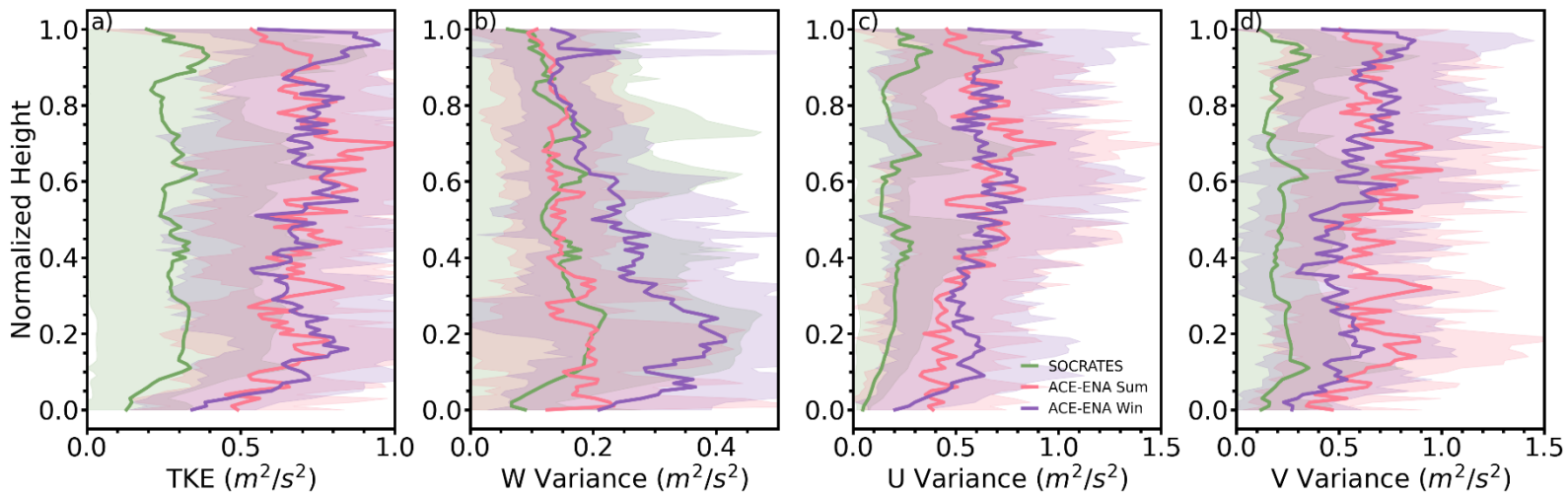


Figure 5. Vertical distributions of in-cloud TKE (a), w'^2 (b), u'^2 (c), and v'^2 (d). Shaded areas denote the inter-cloud-case standard deviations. The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

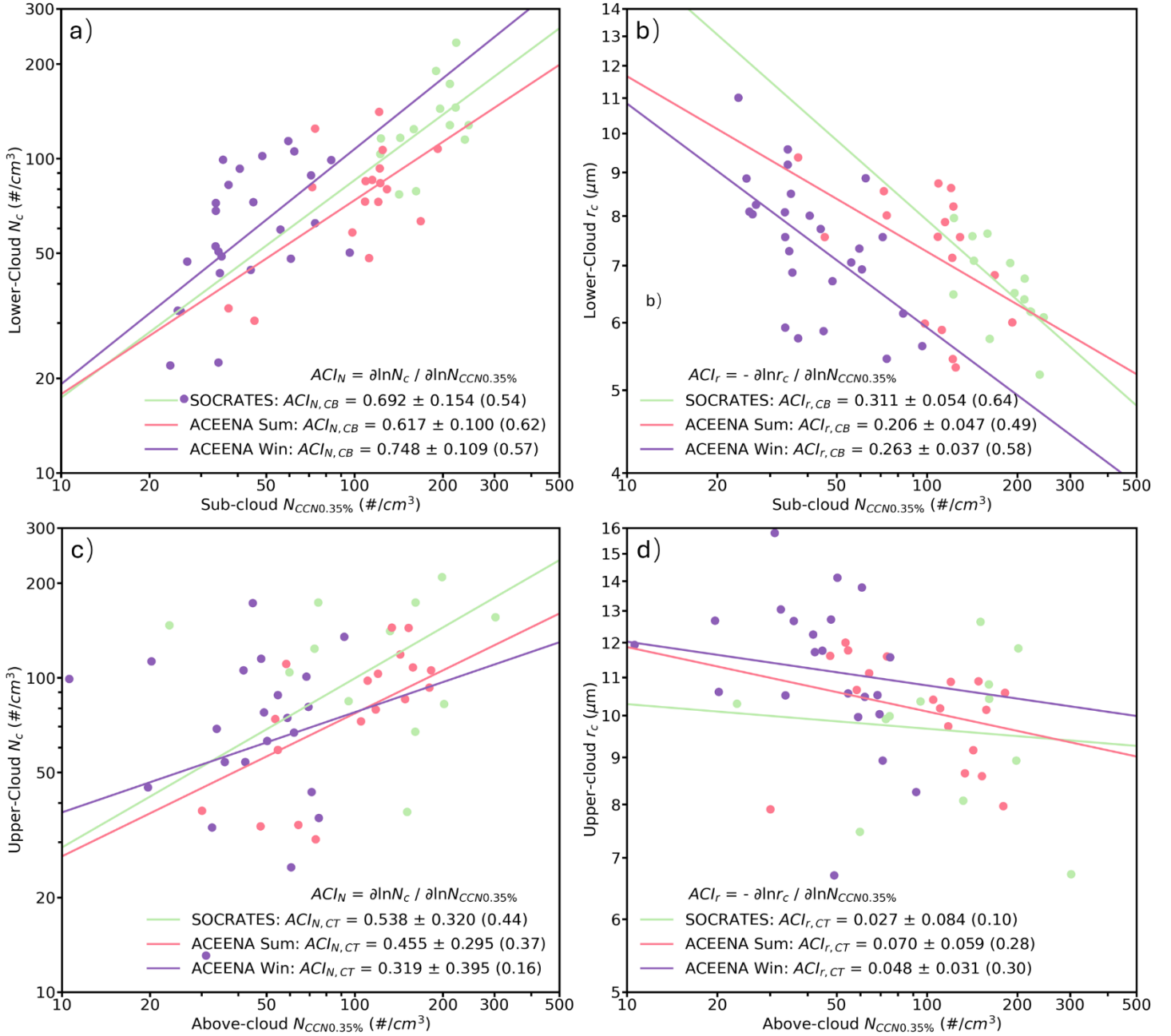


Figure 6. Scatterplots of the a) N_c and b) r_c at the lower-cloud ($z_i < 0.2$) against the sub-cloud $N_{CCN0.35\%}$, and the c) N_c and d) r_c at the upper-cloud ($z_i > 0.8$) against the above-cloud $N_{CCN0.35\%}$. The statistical metrics in the legends denote the ACI values and standard errors, and the absolute values of correlation coefficients (in parentheses). The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

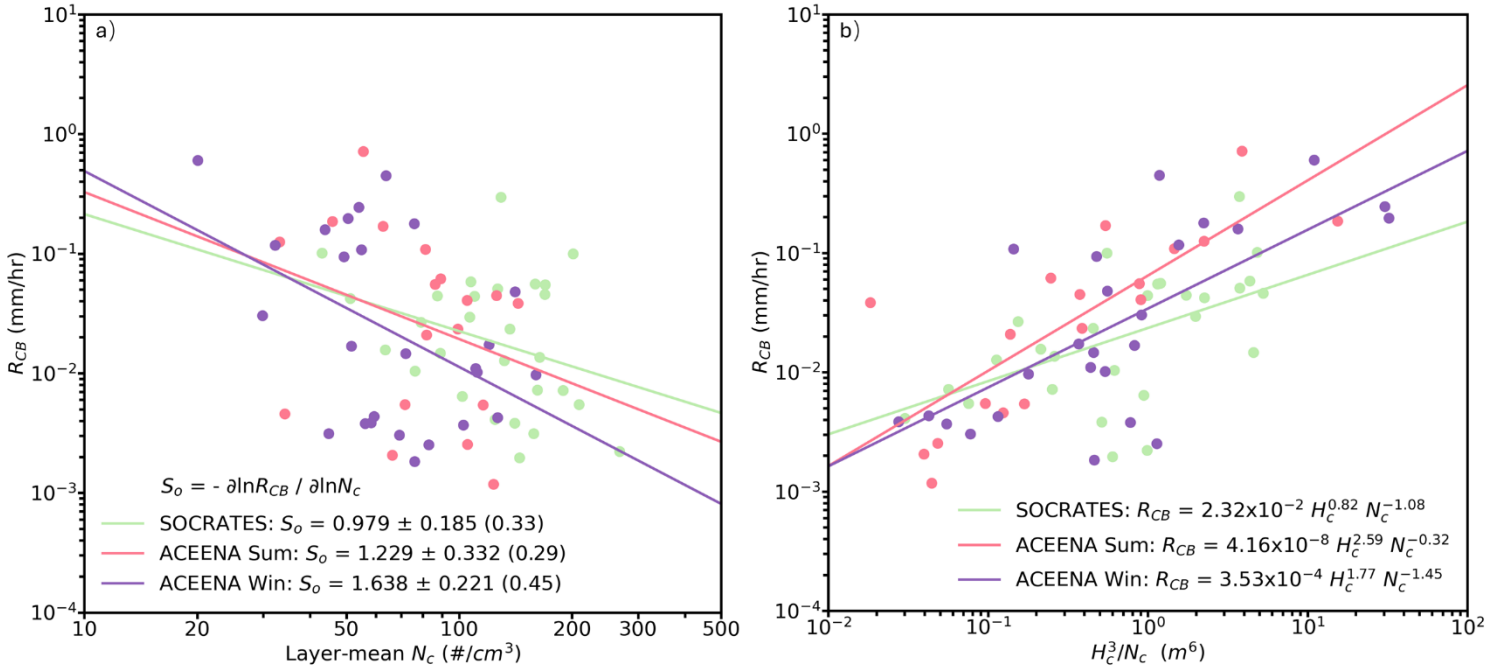


Figure 7. Scatterplots of the cloud base precipitation rate R_{CB} against the a) layer-mean N_c and b) H_c^3/N_c . The ACE-ENA Sum, ACE-ENA Win, and SOCRATES are color-coded with pink, purple, and green, respectively.

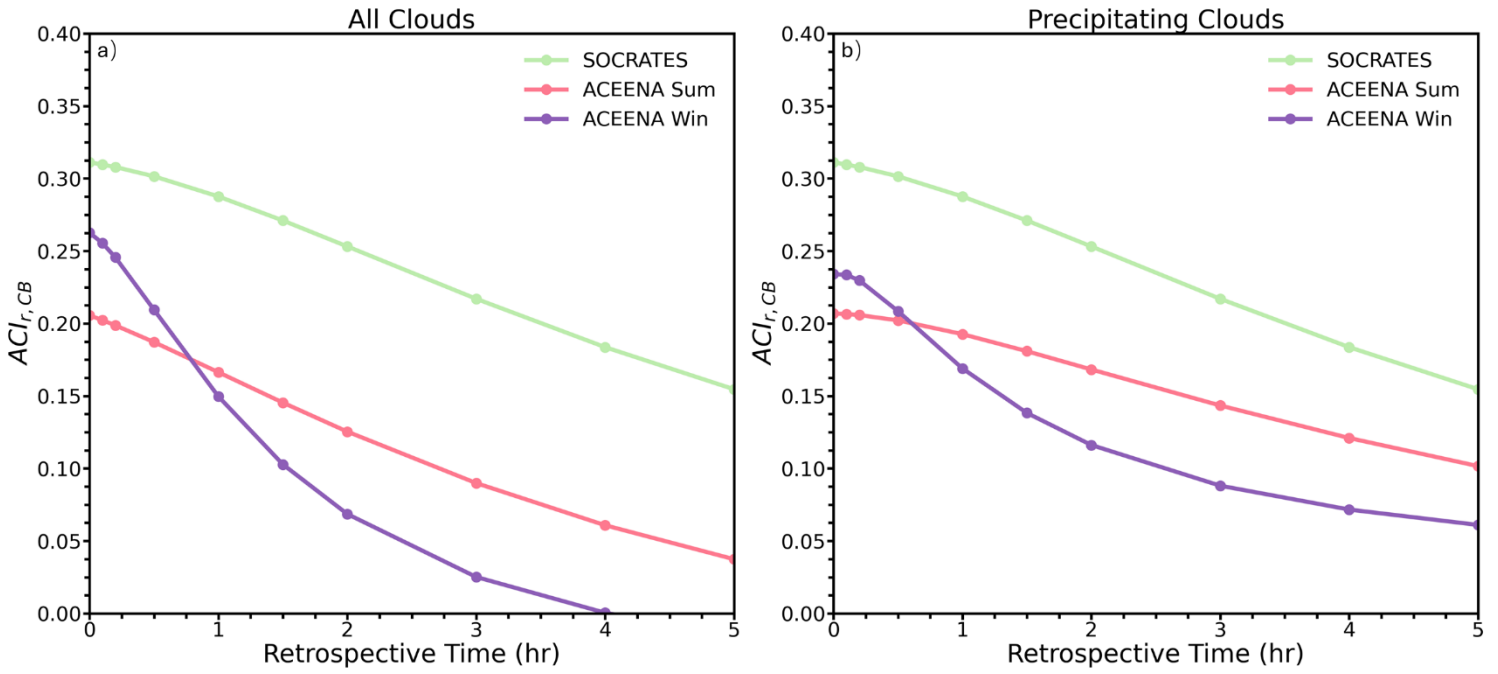


Figure 8. $ACI_{r,CB}$ as a function of the sub-cloud $N_{CCN0.35\%}$ retrospective time for a) all clouds and b) precipitating clouds.