https://doi.org/10.5194/egusphere-2023-2608
Preprint. Discussion started: 14 November 2023

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

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Abstract. The aerosol-cloud-precipitating interaction within the cloud-topped Marine Boundary Layer (MBL), are being examined using aircraft in-situ measurements from Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) and Southern Ocean Clouds Radiation Aerosol Transport Experimental Study (SOCRATES) field campaigns. SOCRATES clouds have a larger number of smaller cloud droplets compared to ACE-ENA summertime and wintertime clouds. The ACE-ENA clouds, especially in wintertime, exhibit pronounced drizzle formation and growth, attributed to the strong in-cloud turbulence that enhances the collision-coalescence process. Furthermore, the Aerosol-Cloud Interaction (ACI) indices from the two aircraft field campaigns suggest distinct sensitivities. The aerosols during ACE-ENA winter are more likely to be activated into cloud droplets due to more larger aerosols and strong vertical turbulence. The enriched aerosol loading during SOCRATES generally leads to smaller cloud droplets competing for available water vapor and exhibiting a stronger ACI. The ACI calculated near the cloud base was noticeably larger than the layer-mean and near-cloud-top, owing to the closer connection between the cloud layer and sub-cloud aerosols. Notably, the sensitivities of cloud base precipitating rates to cloud-droplet number concentrations are more pronounced during the ACE-

https://doi.org/10.5194/egusphere-2023-2608
Preprint. Discussion started: 14 November 2023

ENA than during the SOCRATES campaigns. The in-cloud drizzle evolutions significantly alter sub-cloud cloud condensation nuclei (CCN) budgets through the coalescence-scavenging effect, and in turn, impact the ACI assessments. The results of this study can enhance the understanding and aid in future model simulation and assessment of the aerosol-cloud interaction.

1. Introduction

Marine boundary layer (MBL) clouds substantially impact the Earth's climate system (Dong and Minnis, 2022). Sustained by large-scale subsidence and cloud-top longwave radiative cooling, MBL clouds, typically located beneath the temperature inversion at the MBL top, persistently reflect the incoming solar radiation and modulate the radiative balance (Lilly, 1968; Albrecht et al., 1995; Wood et al., 2015; Dong et al., 2023). The climatic significance of MBL cloud radiative effects, which remains largely uncertain (IPCC, 2022), is closely linked to cloud microphysical properties that are substantially influenced by surrounding aerosol conditions (Chen et al., 2014; Feingold and McComiskey, 2016). Observational evidence demonstrates that cloud microphysical responses to aerosols, defined as the aerosol-cloud interaction (ACI), can be typically viewed as decreased cloud droplet effective radii (r_c) and increased number concentrations (N_c) under an augmented aerosol intrusion. The ACIs have been extensively investigated by different observational platforms, such as aircraft, ground-based and satellite observations, and model simulations over different maritime regions (Hill et al., 2009; Diamond et al., 2018; Painemal et al., 2020; Wang et al., 2020; Zheng et al., 2022a).

Furthermore, more and smaller cloud droplets not only extend cloud longevity and spatial coverage but also modulate the precipitation processes, reflecting the cloud adjustments to aerosol disturbances (Albrecht, 1989; Bellouin et al., 2020). Since precipitation is common in MBL clouds, particularly in the form of drizzle (Wood et al., 2015; Wu et al., 2020), the formation and growth of drizzle drops are deeply entwined with the MBL aerosols and turbulence. On the one hand, aerosols have

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been found to suppress the precipitation frequency and strength by constantly buffering cloud droplet number concentrations via activation, hence increasing cloud precipitation susceptibility (Lu et al., 2009; Sorooshian et al., 2009; Duong et al., 2011). On the other hand, in-cloud turbulence and wind shear can effectively enhance collision-coalescence efficiency, stimulating drizzle formation and growth, and consequently leading to enhanced precipitation (Chen et al., 2011; Wu et al., 2017). Conversely, precipitation has been shown to induce a substantial influence on the MBL aerosol and cloud condensation nuclei (CCN) budget, via the coalescence-scavenging effect. As the drizzle drops descend, they are enlarged by collecting more cloud droplets within the cloud layer. However, the drizzle drops, once falling out of the cloud base, can result in net reductions in sub-cloud aerosols and CCN budgets via the coalescence-scavenging effect (Wood, 2006; Zheng et al., 2022b). Quantitative estimates of these effects remain ambiguous and inconclusive, which is subject to multiple factors such as aerosol physicochemical characteristics, cloud morphology, and MBL dynamics and thermodynamics conditions (Sorooshian et al., 2009; Duong et al., 2011; Diamond et al., 2018; Brunke et al., 2022). Thus, more studies on the aforementioned processes regarding MBL aerosols and clouds over different maritime regions are warranted to pursue an in-depth understanding of aerosol-cloud-precipitation interactions (ACPIs).

The Eastern North Atlantic (ENA) stands as a desirable region for exploring MBL clouds in the mid-latitude, with Graciosa Island in the Azores (39.09°N, 28.03°W) representing a focal point for such studies. Located between the mid-latitude and subtropical climate zones, Graciosa is subject to the meteorological influence of both the Icelandic Low and the Azores High, and the influence of aerosols ranging from pristine marine air masses to those heavily influenced by continental emissions from North America and Northern Europe (Logan et al., 2014; Wood et al., 2015; Wang et al., 2020). Addressing the need for sustained research into the MBL clouds, the recent Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) aircraft campaign (J. Wang et al., 2022) were conducted in the summer 2017 (ACEENA Sum) and winter 2018 (ACEENA Win). During these two intensive operation

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periods (IOPs) of ACE-ENA, the research aircraft accrued abundant in-situ measurements of aerosols, clouds, and drizzle properties, providing invaluable resources for studying the ACI and ACPI processes.

Over the Southern Ocean (SO), the Southern Ocean Clouds Radiation Aerosol Transport Experimental Study (SOCRATES) field campaign (McFarquhar et al., 2021) was conducted during the austral summer, which marks another valuable piece of the MBL cloud research. The SO, being one of the cloudiest regions globally, is predominantly influenced by naturally produced aerosols originating from oceanic sources due to its remoteness, where the anthropogenic and biomass burning aerosols exert minimal influence over the region (McCoy et al., 2021; Sanchez et al., 2021; Twohy et al., 2021; Zhang et al., 2023). The aerosol budget in this region is primarily shaped by biological aerosols, which nucleate from the oxidation products of dimethyl sulfide (DMS) emissions, as well as by sea spray aerosols. Hence, the SO provides an unparalleled natural laboratory for discerning the influence of these natural aerosol emissions on the MBL clouds. Furthermore, the MBL clouds over the SO predominantly consist of supercooled liquid water droplets, which coexist with mixed- and ice-phase processes (Y. Wang et al., 2021a; Xi et al., 2022), while the precipitation phases are examined to be primarily dominated by liquid hydrometeors (Tansey et al., 2022; Kang et al., 2023). The in-situ measurements collected from SOCRATES have cultivated numerous studies on aerosols, clouds, and precipitation over the SO using both in-situ measurements and model simulations (McCoy et al., 2020; Altas et al., 2021; D'Alessandro et al., 2021), and provides an opportunity to study the liquid cloud processes under a colder nature.

This study examines the similarities and differences in the MBL aerosol, cloud, drizzle properties, their distribution and evolution, and their ACIs and ACPIs from selected cloud cases during ACE-ENA and SOCRATES. The data and methods used in this study are introduced in section 2. The aerosol and CCN properties in the above- and sub-cloud regimes, and the vertical distributions of MBL cloud and drizzle properties are examined in section 3. The ACI, precipitation susceptibility and drizzle impacts on the sub-cloud aerosols and CCN (ACPI) are discussed in section 4. Finally, the findings are summarized along with the discussions of the importance of this study in section 5.

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2. Data and methods

2.1 Cloud and drizzle properties

The in-situ measurements of MBL cloud properties are temporally synchronized to 1 Hz resolution, corresponding to approximately 100 m (5 m) of horizontal (vertical) sampling. The Fast Cloud Droplet Probe (FCDP) onboard aircraft during ACE-ENA can detect droplets with diameter (D_p) ranging from 1.5 μm to 50 μm at resolutions between 1 and 3 μm (Glienke and Mei, 2020), while the SOCRATES used a similar CDP to measure droplets from 2 μm to 50 μm at a 2 μm resolution. Both ACE-ENA and SOCRATES leverage the Two-Dimensional Stereo Particle Imaging Probe (2DS) to discern droplets with diameters from 5 μm to 1280 μm (Lawson et al., 2006; Glienke and Mei, 2019). The 2DS in-situ measurements will be used as additional screening to eliminate large ice particles (D_p > 200 μm). Moreover, the University of Washington Ice–Liquid Discriminator product, which is a Machine-learning-based single-particle phase classification of the 2DS images, is used to identify small ice crystals when available. Through these three datasets, we can tease out the ice-dominated period to the utmost extent and focus on the liquid cloud processes and ACI during the SOCRATES (Atlas et al., 2021; Wang et al., 2021).

Although these in-situ measurements can provide "ground-truth" datasets, their uncertainties must be properly analyzed and data quality must be controlled before being applied to scientific studies. The uncertainties of FCDP in sizing and concentration are approximately 30% and 20%, respectively (Baumgardner et al., 2017). Considering the significant uncertainty in the concentration of smaller particles from a photodiode probe such as 2DS (Baumgardner & Korolev, 1997; Wang et al., 2021), a diameter of 40 μm is used as the demarcation line between cloud droplets and drizzle drops (Wood et al., 2005). Then droplet number concentrations in the overlapping size bin between FCDP and 2DS are redistributed assuming a gamma distribution, thereby a complete size spectrum of cloud and drizzle can

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be merged from FCDP and 2DS measurements. Hence, the cloud and drizzle microphysical properties can be calculated.

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

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

The cloud droplet effective radius (r_c) is given by:

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

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

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

where ρ_w is water density.

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

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

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

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

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

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

The combined threshold of $N_c > 5$ cm^{-3} and $LWC_c > 0.01$ g m^{-3} is used for determining the valid cloud samples and cloud boundaries (Wood, 2005; Zheng et al., 2022b). The complete cloud vertical profiles from sub-cloud to the above-cloud are selected during the ACE-ENA and SOCRATES IOPs, in which the flight strategy includes sawtooth and spiral cloud transects and ramping cloud sampling. The precipitation conditions are determined by whether samples of $N_d > 0.001$ cm^{-3} exists below the cloud base height. In total, the selected numbers of cloud (precipitating cloud) profiles are 18 (13), 26 (13), and 28 (24) for ACE-ENA summer and winter IOPs along with SOCRATES, respectively. The detailed selected cloud profiles are listed in Table S1, along with the cloud profile macrophysics. Additionally, for the purposes of studying the sub-cloud aerosols that actually interact with the cloud, the sub-cloud mixed layer is determined following the threshold suggested by Jones et al. (2011). Starting from the cloud base and looking downward, the mixed layer altitude is defined as where the vertical changes in liquid water potential temperature (θ_L) and total water mixing ratio (q_t) exceed 0.5 K and 0.5 g/kg, respectively. Hence, the mixed layer thickness is defined as the difference in cloud top altitude and mixed layer altitude. An example of the mixed layer identification is shown in Figure S1. The sub-cloud aerosols are thus selected between the mixed layer altitude and cloud base in order to best represent the aerosol-cloud interactions, while the above-cloud aerosols are selected between the cloud top and 200 m above (Wang et al., 2020).

2.2 Aerosol properties

The total aerosol number concentrations (N_a) from ACE-ENA and SOCRATES are measured by the airborne Condensation Particle Counter (CPC) models 3772 and 3760A, which counts the number of aerosols with diameter (D_p) larger than 3 nm and 11 nm, respectively (Kuang and Mei, 2019; SOCRATES Low Rate Data, 2022). Additionally, the Passive Cavity Aerosol Spectrometer (PCASP) onboard the ACE-ENA aircraft is capable of sizing the aerosol with D_p ranging from 0.1 µm to 3.2 µm (Goldberger, 2020). While the ultra-high sensitivity aerosol spectrometer (UHSAS) measures the size-

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resolved aerosol distribution from 0.06 μm to 1.0 μm during SOCRATES (Uin, 2016). Therefore, the number concentrations of accumulation mode aerosols (N_{ACC}, 0.1 μm-1 μm) can be discerned from the PCASP and UHSAS aerosol size distributions. The Aitken mode aerosols (N_{Ait}, < 0.1 μm) from the ACE-ENA is given by the fast integrated mobility spectrometer (FIMS), which can size the aerosol down to 9 nm (Olfert et al., 2008), while the N_{Ait} from SOCRATES is limited to 0.06 μm – 0.1 μm due to the limitation of UHSAS. As for the CCN measurements, the ACE-ENA utilized the Dual-Column CCN Counter at two constant supersaturation levels of 0.15% and 0.35% (Uin and Mei, 2019), while the CCN number concentration (N_{CCN}) during SOCRATES was measured under various supersaturation levels from 0.06% to 0.87% using a scanning CCN counter (Roberts and Nenes, 2005). In this study, N_{CCN} at 0.35% supersaturation ($N_{CCN0.35\%}$) is used to ensure a direct comparison between ACE-ENA and SOCRATES. The aerosol measurements are in the temporal resolution of 1Hz. Note that the aerosol and CCN data are quality-controlled by removing the data point where the $N_c + N_d$ greater than 5 cm^{-3} or N_d greater than 0.01 cm^{-3}, to filter out the contamination of the cloud droplets, and drizzle water splashing.

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

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

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

In the SOCRATES region, dominant air masses primarily originate from the south or from the west, skirting the Antarctic coast (Zhang et al., 2023). The SOCRATES above-cloud aerosols (674.6 cm^{-}

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^{3}) are primarily constituted by the Aitken mode aerosols because the mean N_{Acc} is only 62.5 cm^{-3}. These aerosols are predominantly produced from the oxidation of biogenic gases, notably the dimethyl sulfide (DMS) emitted by marine biological productivity (Sanchez et al., 2018; McCoy et al., 2020). Conversely, the ENA region experiences aerosols of varied origins, spanning maritime air masses to those heavily influenced by continental emissions from North America or Northern Europe (Logan et al., 2014; Wang et al., 2020). During the summer ACE-ENA campaign, the MBL is enriched by sulfate and carbonaceous particles (Y. Wang et al., 2021b; Zawadowicz et al., 2021). This enhancement is attributed both to local generation from DMS and to the long-range transport from the continental air masses, resulting in the mean N_a of 312.6 cm^{-3} and 301.5 cm^{-3} for above- and sub-cloud regimes, respectively. The ACE-ENA winter exhibits the lowest aerosol and CCN concentrations, predominantly sourced from local maritime influences, and coupled with reduced continental air mass intrusions (Zheng et al., 2018; Y. Wang et al., 2021b).

Figure 1a reveals that there are more above-cloud N_a during the three IOPs than sub-cloud values, especially during the SOCRATES. The higher above-cloud N_a values from the three IOPs are primarily contributed by Aitken mode aerosols because their corresponding N_{Acc} values are much lower (Figs. 1a&b). It is interesting to note that the above-cloud $N_{CCN0.35\%}$ values exceed the N_{Acc} for all three IOPs (Figs. 1b&c), implying that a significant fraction of Aitken mode aerosols can be activated to become CCN, corroborating findings from earlier studies (McCoy et al., 2021; Zheng et al., 2021). For the sub-cloud regime, the N_a values during SOCRATES and ACE-ENA winter are ~70-80% of their corresponding above-cloud values, and the N_a during ACE-ENA summer is almost identical to its above-cloud value. Notice that the N_{Acc} values from three IOPs are more than doubled their above-cloud values, and most of them can be activated to become CCN at SS of 0.35%. It is interesting to note that the higher $N_{CCN0.35\%}$ at sub-cloud layer during SOCRATES may partially result from the cloud process on aerosols

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(Figs. 1e&f), which is suggested by previous studies (McCoy et al., 2021; Zhang et al., 2023), and will be further discussed.

To further investigate the above- and sub-cloud aerosol properties from three IOPs, the aerosol droplet size distributions are analyzed in Figure 2. It is evident that SOCRATES aerosols have the highest concentrations of Aitken mode particles (D_p = 0.06 – 0.1 μm, given that the < 0.06 μm is not available from UHSAS) for both the above- and sub-cloud regimes. McCoy et al. (2021) and Zheng et al. (2021) identified analogous origins and formations of the above-cloud Aitken mode aerosols over both the SO and ENA regions and concluded that these aerosols primarily originate from the nucleation of photo-oxidation products of DMS, notably H_2SO_4 and MSA, in the free troposphere (FT). The differential concentrations can be ascribed to the fact that sea-surface DMS concentrations in the SO are generally higher than those in the ENA region (Aumont et al., 2002; Zhang et al., 2023). Moreover, DMS emissions in the ENA during summer surpass those during winter (Zawadowicz et al., 2021). For the accumulation mode aerosols (0.1 – 1 μm), the N_{Acc} values for both above- and sub-cloud regimes during SOCRATES decrease monotonically with particle size. The results in Figure 2 further support the finding that Aitken mode aerosols are dominant over the SO. The N_{Acc} values during ACE-ENA show slight uplifts for the small accumulation mode aerosols (< 0.3 μm), particularly for summer, reflecting the signal of potential long-range transport of fine-mode aerosols (Wang et al., 2020; Y. Wang et al., 2021b). Consequently, such comparison reinforces the notion that the SO represents a largely pre-industrial marine environment, wherein the influence of anthropogenic and biomass-burning aerosols is minimal (McCoy et al., 2020, 2021; Zhang et al., 2023).

When contrasting the aerosol size distributions in the sub-cloud regime (Fig. 2b) with those in the above-cloud regime, the influence of cloud processing on aerosols is discernibly non-trivial, particularly under the cloud-topped MBL conditions examined in this study. While the FT aerosols can be further entrained down and contribute to the population of Aitken mode aerosols within the MBL, the sub-cloud

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aerosols can also be subject to the influence of new particle formation in the upper MBL, though arguably less effective than those within the FT (Zheng et al., 2021). Additionally, in-cloud Brownian capture can lead to a substantial reduction in Aitken mode aerosols (Hudson et al., 2015; Wyant et al., 2022), providing the rationale for the observed decrease in Aitken mode aerosols in the sub-cloud regime, especially for particles smaller than 0.07 µm. In addition, cloud chemical processing, such as the aqueous-phase condensation of sulfuric gas onto the aerosol cores inside the cloud droplets, is particularly pronounced during the transitioning of Aitken mode aerosols to accumulation mode aerosols (Hudson et al., 2015; Zhang et al., 2023).

From both above- to sub-cloud regimes, the larger Aitken mode aerosols (> 0.07 µm) can be effectively enlarged to accumulation mode aerosols through coagulation and water vapor diffusional growth (Covert et al., 1996), contributing to the elevated accumulation mode aerosol distribution and increased N_{Acc} in the sub-cloud regime. These processes are particularly evidenced by the decrease of critical supersaturations from above-cloud (between 0.35% - 0.4%) to sub-cloud (between 0.3% - 0.35%) during SOCRATES (Fig. S2) because the aerosol droplet sizes are enlarged and more readily become CCN. Furthermore, the in-cloud coalescence process combines mixtures of large and small cloud droplets, and results in the sub-cloud aerosol residuals shifting towards the larger size, upon the droplet evaporation below the cloud (often manifested as drizzle). This partially elucidates the observed increase in the tail-end of the accumulation mode aerosol distribution for all three IOPs. The elevation in sub-cloud coarse mode aerosols observed for both ACE-ENA IOPs (as seen in Fig. S3) can be attributed both to the coalescence-enlargement process as well as the intrusion of marine aerosols (e.g., sea salt).

3.2 Bulk cloud microphysical properties distribution

The PDFs of MBL cloud microphysical properties (N_c, r_c, LWC_c) derived from aircraft in-situ measurements from the three IOPs are shown in Figures 1g-1i. The mean microphysical properties for the individual cloud profiles are listed in Table S2. The results in Figure 1 have demonstrated that

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aerosol/CCN sources and concentrations, especially from the sub-cloud regime, play an important role in cloud droplet formation and evolution. For example, the SOCRATES has the highest sub-cloud aerosols and CCN, and subsequently feature a larger number of smaller cloud droplets, given the highest N_c (148.3 cm^{-3}) and smallest r_c (8 µm) among the three IOPs. These results have further improved the understanding of the aerosol first indirect effect: more aerosols induce more smaller cloud droplets (higher N_c and smaller r_c) under the constrained liquid water content conditions, thus the MBL clouds reflect more incoming solar radiation (Twomey, 1977). The ACE-ENA wintertime clouds feature the fewest N_c (70.6 cm^{-3}) and largest r_c (9.8 µm), while the N_c and r_c (89.4 cm^{-3} and 9 µm) during ACE-ENA summer fall between the SOCRATES and ACE-ENA winter values. Considering the aerosol competing effect against the available water vapor, the relatively abundant aerosols in SOCRATES might account for the narrower r_c distribution, which peaks between 6 – 10 µm. SOCRATES has a lower cloud-layer water vapor mixing ratio (figure not shown) compared to ACE-ENA because the SO region has been observed to contain less precipitable water vapor than the ENA region due to the colder sea surface temperatures (Marcovecchio et al., 2023). Therefore, the aerosol and cloud properties in Figure 1 promise further examination of different cloud microphysical responses to aerosols via the ACI process. Note that the $N_{CCN0.35\%}$ values are less than N_c values during the ACE-ENA winter IOP, thereby offering compelling further investigation on potential impacts of precipitation on the MBL CCN budget. These aerosol-cloud-precipitation interactions (ACPIs) will be discussed in Section 4.

3.3 Vertical distributions of cloud and drizzle microphysics

The vertical distributions of the cloud and drizzle microphysical properties within the cloud layer from the three IOPs are shown in Figure 3. To ensure the representative vertical profiles, all the in-cloud samples are vertically smoothed by using a triangular moving average method, and are inverse distance weighted in every 50 m moving altitude windows. Furthermore, the altitude is then normalized 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 previous discussions on the bulk microphysics distribution, the mean N_c values from SOCRATES are consistently higher than ACE-ENA summer and winter for the entire cloud layer, with a slight increase ranging from the cloud base to the upper-middle part ($z_i \approx 0.85$) and then decreasing toward the cloud top due to cloud-top entrainment (Fig. 3a). All r_c values from the three IOPs show a near-linear increase from cloud base to top, with the smallest values observed during SOCRATES and the largest values observed during ACE-ENA winter (Fig. 3b).

The warmer and drier air near the cloud top entrained into the cloud layer and further mixing downward often results in the evaporation of small cloud droplets and the shrinking of droplet sizes which oppose condensational growth (Desai et al., 2021). Decreases in both N_c and LWC_c, and the reduced growth of r_c near the cloud top ($z_i > 0.85$) support signals of cloud-top entrainment mixing during all three IOPs. It is interesting to note that the r_c values from SOCRATES increase monotonically from cloud base to top, while the r_c values from both ACE-ENA summer and winter increase until $z_i \approx 0.8$ and then remain nearly constant, although all of their N_c values (at $z_i \approx 0.8$) decrease towards the cloud top. When dry air entrainment occurs at the cloud top, some of the upper-level smaller cloud droplets will evaporate, which provides an extra water vapor source and leads to decreases in N_c (Fig. 3a). This extra water vapor can then be re-condensed on the cloud droplets and potentially enlarge the droplet size. This effect is more pronounced on smaller cloud droplets since there is a smaller surface area (Wallace and Hobbs, 2006), as shown in the continuous growth of r_c during SOCRATES. On the other hand, the cloud-top entrainment mixing can shrink the large cloud droplets via evaporation. The nearly constant r_c values (at $z_i > 0.8$) represents the balance of these two competing processes. The impact of these two processes on cloud droplets depends on the cloud-top entrainment mixing rate.

While carrying the distinct discrepancies in the mean values for all layers, the N_c and r_c from ACE-ENA summer and winter clouds experienced similar vertical evolutions as the SOCRATES. The

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increases of r_c (Δr_c) from cloud base to cloud top are 4.03 μm, 4.78 μm and 5.85 μm for SOCRATES, ACE-ENA summer and winter, respectively. Consequently, the LWC_c values from the three IOPs are comparable to each other. The vertical distributions of MBL cloud microphysical properties examined in this study are in good agreement with the previous studies conducted on these two field campaigns (Wu et al., 2020a; Y. Wang et al., 2021a; J. Wang et al., 2021; Wang et al., 2023). In addition, the cloud adiabaticity is defined as $f_{ad} = LWC_c/LWC_{ad}$, where the LWC_{ad} denotes adiabatic LWC (Wu et al., 2020b). As shown in Figure S4, the clouds from all three IOPs feature certain levels of sub-adiabaticity above the cloud base. The layer-mean f_{ad} values are 0.681±0.083, 0.476±0.106, and 0.447±0.100 for SOCRATES, ACE-ENA summer and winter, respectively. It has been well known that the cloud sub-adiabaticity is primarily induced by the in-cloud collision-coalescence and the entrainment mixing processes (Hill et al., 2009; Braun et al., 2018; Gao et al., 2020; Wu et al., 2020b).

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

$$w_e = A_\sigma * \sigma_w / R_{i\sigma} , \tag{6}$$

where the turbulence kinetic energy (TKE) dissipation coefficient A_σ is empirically taken as 26 as in Albrecht et al. (2016), and the $R_{i\sigma}$ is the buoyancy Richardson number calculated by $(g/\theta_0) * (\Delta\theta_v h/\sigma_w^2)$. σ_w denotes the standard deviation of vertical velocities taken near the cloud top (z_i > 0.9), and h is the MBL height. θ_0 is the reference potential temperature and $\Delta\theta_v$ is the virtual potential temperature difference across the temperature inversion layer above the cloud. Given the valid vertical velocity measurements for the selected cloud cases, the averaged w_e values are 0.570±0.834 cm s^{-1}, 0.581±0.560 cm s^{-1}, and 0.960±1.127 cm s^{-1} for SOCRATES, ACE-ENA summer and winter, respectively. The stronger w_e during ACE-ENA winter might be induced by the generally weaker cloud-top inversions and stronger near-cloud top turbulence, compared to the summertime when the ENA is

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dominated by the large-scale high-pressure system (Ghate et al., 2021). Considering the near cloud-top proportion of cloud where the LWC_c experienced decrease, the reduction of LWC_c for the ACE-ENA summer (-0.032 g m^{-3}) is higher than the reductions in winter (-0.018 g m^{-3}) and SOCRATES (-0.009 g m^{-3}), albeit that the w_e for ACE-ENA summer is comparable to SOCRATES, and much lower than ACE-ENA winter values. Within the above-cloud inversion layer, the temperature (water vapor mixing ratio) 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 SOCRATES, ACE-ENA summer and winter, respectively. Therefore, the warmer and dryer entrained air can partially contribute to the greater LWC_c reduction and the lower f_{ad} (0.39) during the ACE-ENA summer than those during the ACE-ENA winter (f_{ad} = 0.45) and SOCRATES (f_{ad} = 0.66) near the cloud top (Fig. S4).

Figures 3d-3f illustrate the normalized profiles of MBL drizzle microphysical properties. The N_d values from the three IOPs mimic each other, which all maximize at the cloud top and then monotonically decrease toward the cloud base (Fig. 3d), while their LWC_d values follow a similar trend, albeit with relatively large differences (Fig. 3f). In contrast to the N_d and LWC_d trends, the D_{mmd} gradually increase from cloud top to cloud base (Fig. 3e), making physical sense since the drizzle droplets are typically formed near cloud top and continuously grow via collision-coalescence process while falling. The ACE-ENA wintertime drizzle D_{mmd} and LWC_d are distinctively larger than those in summertime and SOCRATES. It is interesting to note that near the cloud top ($z_i > 0.9$), the ACE-ENA winter has comparable N_d but much larger D_{mmd} than the other two IOPs, suggesting that there were more large drizzle embryos formed from large cloud droplets (Fig. 3b) during ACE-ENA winter.

In order to further analyze the cloud-to-drizzle conversion processes, the cloud and drizzle droplet size distributions (DSD) are categorized into four segments based on their relative position within the cloud layer (Fig. 4): upper cloud ($z_i > 0.8$, Fig. 4a), upper-middle cloud ($0.5 \le z_i < 0.8$, Fig. 4b), lower-middle cloud ($0.2 \le z_i < 0.5$, Fig. 4c) and lower cloud ($z_i < 0.2$, Fig. 4d). The cloud DSDs ($D_p < 40$

μm) from the three IOPs gradually shift towards larger sizes, moving from the lower to the upper cloud regions. This is accompanied by the narrowing of the DSD ranges, as evidenced by the decline in the cloud relative dispersion (ε). The relative dispersion of cloud droplets (ε) is a parameter that represents the DSD and is defined as the ratio between the standard deviation and the mean radius of the distribution. At the lower portion of the cloud (Fig. 4d), the relatively greater value of ε clearly represents the co-existence of the newly formed small cloud droplets from recently activated CCNs and the sedimentation of larger droplets from the upper sections of the cloud. In addition, the discrepancies in ε between the three IOPs may be attributed to the sub-cloud aerosol differences, as Y. Wang et al. (2021a) stated that the higher aerosol loading leads to increased ε due to the water vapor competition effect.

Notably, the cloud DSDs during ACE-ENA winter exhibit a more pronounced negative skew (to the left) than those during ACE-ENA summer, possibly due to the activation of more sub-cloud coarse mode aerosols to become larger cloud embryos, as demonstrated in Fig. S3. Ascending within the cloud, the process of water vapor condensation perpetually pushes the DSD towards larger sizes, culminating in a more negatively skewed DSD. Concurrently, the cloud-top entrainment mixing plays a pivotal role in minimizing ε in the upper cloud region, as elaborated by Lu et al. (2023). Note that in the upper region of the cloud (Fig. 4a), the ACE-ENA winter clouds contain more cloud droplets close to 40 μm, albeit the mean N_c is lower. This scenario is conducive to the formation of larger drizzle embryos compared to summertime clouds, as depicted in Fig. 3e. In comparison, the SOCRATES clouds feature a pronounced log-normal DSD than the ACE-ENA, as the DSDs peak at D_p~15 μm throughout the cloud, and subsequently, the lack of larger cloud droplets resulted in the smaller drizzle embryos near the cloud top. As the newly formed drizzle drops descend and continuously grow through the collision-coalescence process, the drizzle DSDs ($D_p > 40$ μm) are noticeably broadened. From upper to lower cloud regions, the longer tails of the drizzle DSDs expand at the cost of smaller drizzle drops and cloud droplets via the collision-coalescence process. The clouds observed during ACE-ENA, especially in wintertime, contain

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more large drizzle drops ($D_p > 200$ μm) than SOCRATES, which is reflected in the distinct differences in the vertical D_{mmd} as shown in Fig. 3e.

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

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

where the turbulent perturbations of vertical ($\overline{w'^2}$) and horizontal ($\overline{u'^2}$ and $\overline{v'^2}$) components are calculated as the moving variance in a 10s window centered at the measurement time. The w data is confined to an absolute aircraft roll angle of less than 5° (Cooper et al., 2016).

As shown in Figure 5, the vertical wind variances (Fig. 5b) in ACE-ENA winter (layer-mean of 0.244 m^2 s^{-2}) are generally higher than those in summer (0.153 m^2 s^{-2}) and SOCRATES (0.147 m^2 s^{-2}), while the horizontal wind variances are comparable between ACE-ENA winter and summer but much higher than the SOCRATES, resulting in higher TKE during ACE-ENA. Near the cloud top, turbulence effectively enhances coalescence between the larger cloud droplets, primarily by increasing the relative velocities between droplets (Magaritz-Ronen et al., 2016; Ghate and Cadeddu, 2019), and this is especially true for the vertical component of TKE. Note that the values of the vertical turbulence are generally within the ranges of previous studies (Atlas et al., 2020; Ghate et al., 2021). Hence, the stronger drizzle formation in the ACE-ENA winter clouds can be attributed to the higher w'^2 (Fig. 5b) near cloud top (which also explains the stronger entrainment in winter), while the similar w'^2 correspond to the comparable N_d between the ACE-ENA summer and SOCRATES. The presence of turbulence throughout the cloud layer induces differential motions among the cloud droplets, increasing the likelihood of drizzle collecting smaller droplets and the droplets coalescing (Brost et al., 1982; Feingold

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et al., 1996). As a result, the drizzle DSD is sufficiently broadened, and the D_{mmd} is enlarged toward the cloud base.

It is noteworthy that the D_{mmd} in the lower-half region of the ACE-ENA winter clouds experienced rapid growth from ~80 µm to ~105 µm (Fig. 3e), and this increment of ~25 µm contributed to most of the D_{mmd} growth from cloud top to cloud base (33.5 µm). Such phenomena may potentially owe to the much stronger w'^2 in the low half cloud (Fig. 5b), where the sufficient updraft can slow down the descent of drizzle drops, thus recirculating them and elongating the drizzle residence time in the lower part of the cloud, effectively enhancing the collision-coalescence growth (Feingold et al., 1996; Magaritz et al., 2009). In terms of the horizontal turbulence components, the u'^2 and v'^2 can play a role in mixing the ambient air masses and contribute to the broadening of DSD (Wu et al., 2017). However, the vertical turbulence is generally more directly influential to the drizzle formation and evolution (Brost et al., 1982; Nicholls, 1984; Pinsky and Khain, 1997; Ghate et al., 2021).

4 Aerosol-cloud-precipitation interactions (ACPIs)

4.1 Cloud microphysical responses on aerosols

The impacts of different aerosol loadings onto the cloud microphysical properties can be assessed by the aerosol-cloud interaction (ACI) indices, which can be quantified as:

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

and

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

which emphasizes the cloud microphysical responses to CCN via the relative logarithmic change of N_c and r_c to the change in $N_{CCN,0.35\%}$ (Feingold et al., 2003; McComiskey et al., 2009). Physically, the ACI process involves aerosols intruding into the cloud layer, activating as cloud droplets, and subsequently altering cloud DSD and dispersion (Zheng et al., 2022a&b) under various water vapor availabilities.

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Therefore, the cloud microphysical responses within the lower region of the cloud are assessed, which is the first stage in which the sub-cloud CCN can directly interact with the cloud droplets. Furthermore, the similarity in the mean LWC_c (as shown in Fig. 3c) provides comparable liquid water between three IOPs for the assessment of newly generated cloud embryos from activated CCN because the ACI_r is normally assessed under a fixed liquid water (Zheng et al., 2020).

Considering all the cases from three IOPs with available CCN measurements (some cases without CCN measurements during SOCRATES), the N_c and r_c at the lower cloud (z_i < 0.2) are plotted against the sub-cloud $N_{CCN,0.35\%}$ in Figure 6, and the ACI indices are calculated as $ACI_{N,CB}$ and $ACI_{r,CB}$ (CB denoting the assessment near the cloud base). Note that the availability of valid sub-cloud measurements inevitably limit the sample size, especially for SOCRATES, as shown in Table S2. As shown in Figure 6a, the $ACI_{N,CB}$ for the ACE-ENA wintertime (0.748) is higher than the summertime (0.617), indicating that N_c is more sensitive to the sub-cloud $N_{CCN,0.35\%}$. In other words, aerosols intruding into the cloud layer are easily activated to become cloud droplets, which agrees with the previous assessment by J. Wang et al. (2022) on the seasonal dependency of the relationship between N_c and aerosols. The N_c sensitivity for the SOCRATES cloud (0.692) lies between the two ACE-ENA IOPs. The $ACI_{N,CB}$ values from three IOPs are generally higher than the ACI_N values from the layer-mean N_c against the sub-cloud $N_{CCN0.35\%}$ (not shown). Previous studies have shown that the enhanced vertical turbulence (updraft velocity) can effectively facilitate CCN replenishment into the cloud layer (Zheng et al., 2022a&b) and increase the actual in-cloud supersaturation (Brunke et al., 2022), thus leading to a more efficient cloud droplet formation, enhancing the $ACI_{N,CB}$. Furthermore, more coarse mode aerosols during ACE-ENA winter are also favorable to the activation efficiency (Dusek et al., 2006).

As for the r_c responses to CCN (Fig. 6b), the typical Twomey effect, where more CCN compete against available water vapor and result in smaller cloud droplets, is evidenced by different cloud susceptibility between the three IOPs. The SOCRATES features a higher $ACI_{r,CB}$ (0.311), suggesting

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that an increase in $N_{CCN,0.35\%}$ can result in a significant decrease in r_c, compared to ACE-ENA summer (0.206) and winter (0.263). Although the absolute range of variation for r_c during SOCRATES is smaller, the slope is much deeper (Fig. 6b). Recall that the sub-cloud $N_{CCN,0.35\%}$ during SOCRATES is generally higher and is constituted by more small-sized aerosols (as indicated in Fig. 2b). Consequently, after activation, the lower part of the cloud exhibits a higher number of smaller cloud droplets, as shown in Fig. 4d, even under the relatively less $N_{CCN,0.35\%}$ condition for SOCRATES. Therefore, as more CCN intrudes into the cloud, the competition for water vapor among newly-activated cloud droplets becomes more pronounced, given similar water availability. In contrast, the presence of larger cloud droplets near the cloud base, whether activated from coarse-mode aerosols or remaining as residuals from cloud coalescence, would elevate the r_c especially under the relatively more CCN condition, hence inevitably dampening the $ACI_{r,CB}$ during ACE-ENA. However, a more comprehensive investigation into the cloud microphysical responses to CCN intrusions under various water supply conditions will require additional aircraft cases from more field campaigns. Note that the $ACI_{r,CB}$ values in Figure 6b are also larger than the results from the layer-mean r_c against sub-cloud $N_{CCN,0.35\%}$, since the layer-mean microphysics is more subject to the cloud droplet evolution processes such as condensational growth and collision-coalescence. The ACI indices from three IOPs are in the ACI range of the previous studies in MBL clouds (Twohy et al., 2005; Lu et al., 2009; Diamond et al., 2018) using aircraft in-situ measurements.

To investigate the ACI indices at the upper level of the cloud, the N_c and r_c at the upper cloud (z_i > 0.8) are plotted against the above-cloud $N_{CCN,0.35\%}$ in Figure S5, and the ACI indices are calculated as $ACI_{N,CT}$ and $ACI_{r,CT}$ (denoting the assessments near the cloud top). Compared to the $ACI_{N,CB}$ and $ACI_{r,CB}$, the $ACI_{N,CT}$ and $ACI_{r,CT}$ are much weaker, especially for $ACI_{r,CT}$. These weaker relationships support the notion that though the aerosols entrained into the upper-cloud region can affect the cloud microphysics to a certain degree, the effects are less pronounced than those from the sub-cloud aerosols

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(Diamond et al., 2018, Wang et al., 2020) because the MBL cloud N_c and r_c variations are dominated by the condensational growth process and the collision-coalescence process near the cloud top.

4.2 Precipitation susceptibility

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

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

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

As shown in Figure 7a, the R_{CB} values generally have a negative correlation with increased layer-mean N_c with the S_0 values of 0.979, 1.229 and 1.638, respectively, for SOCRATES, ACE-ENA summer and winter. Note that the R_{CB} for the ACE-ENA winter is more susceptible to the layer-mean N_c than the ACE-ENA summer and SOCRATES, which can be partially attributed to the existence of more large drizzle drops (as shown in Fig. 4d) near the cloud base. As previously discussed, these large drizzle drops are induced by the turbulence-enhanced in-cloud collision-coalescence and the drizzle-recirculating processes, especially under low N_c conditions, hence increasing the S_o values. Comparing the seasonality during ACE-ENA, more cases with large observed R_{CB} during the winter season are consistent with J. Wang et al. (2022). The relatively narrower drizzle DSD in SOCRATES may further diminish the sensitivity of R_{CB} to N_c. Note that the derived S_o values in this study are generally higher (or close to the upper end) compared to previous studies (Lu et al., 2009; Duong et al., 2011; Terai et al., 2012; Jung et al., 2016), which is possibly due to decreasing S_o within the thicker cloud (Terai et al.,

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2012). The mean cloud thicknesses of the ACE-ENA summer (368.7 m), winter (400.8 m) and SOCRATES (487.4 m) are inversely proportional to their S_o values.

In addition, it is well known that the R_{CB} can be parameterized or predicted via an approximate relation with N_c and cloud thickness (H_c), which is usually parameterized in the form of $R_{CB} \propto c\ H_c^3\ N_c^{-1}$ (Lu et al., 2009). Following the same method, we derive the relationships from three IOPs in Figure 7b, where the R_{CB} are positively (negatively) proportional to the H_c (N_c), with the exponential parameters in the range of the typical values in the MBL clouds (Comstock et al., 2004; vanZanten et al., 2005; Lu et al., 2009). The statistical R^2 values of R_{CB} against H_c (N_c) are 0.696 (0.177), 0.419 (0.212) and 0.165 (0.295), for the ACE-ENA summer, winter and SOCRATES, respectively, suggesting that the R_{CB} in ACE-ENA clouds may be more determined by H_c, while the R_{CB} in SOCRATES cloud are more related to N_c. Note that the relationship for SOCRATES in this study reveals a similar R_{CB} dependence on N_c but a smaller dependence on the cloud thickness than the study by Kang et al. (2023), who concluded a relationship of $R_{CB} = 1.73e - 10\ H_c^{3.6} N_a^{-1}$, based on the rain rate retrieved from radar and lidar measurements and the aerosol concentration also from the SOCRATES. The discrepancies are possibly due to the different sample selections and different methods in the R_{CB} calculation.

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

Multiple studies on the MBL clouds have concluded that the in-cloud drizzle formation and evolution processes can effectively impact the sub-cloud CCN budgets via the coalescence-scavenging effect (Wood, 2006; Wood et al., 2012; Diamond et al., 2018; Zheng et al., 2022b; Zhang et al., 2023). Drizzle drops are formed and grow via the collision-coalescence process by collecting cloud droplets and small drizzle drops, resulting in the consumption of CCN (the precursor of cloud droplet), but in the meantime, the in-cloud N_c can be continuously buffered by the sub-cloud CCN replenishment. Although the sub-cloud aerosols (especially in large size) would be added if the drizzle fell and evaporated outside

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the cloud, the increment cannot compensate for the loss. Therefore, the net result of the whole process is usually presented as the depletion of sub-cloud CCN residuals, and such drizzle modulation on the CCN budget could be substantial in moderate-to-light drizzles or even non-precipitating clouds, depending on the collision-coalescence efficiency (Feingold et al., 1996; Wood, 2006; Kang et al., 2022).

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

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

where the constant K (2.25 m^2 kg^{-1}) denotes the drizzle collection efficiency (Wood et al., 2006; Diamond et al., 2018). H_c is cloud thickness, and H_{mix} is the thickness of the mixed layer to ensure the change in the cloud layer can be sufficiently conveyed throughout the layer. The calculated CCN loss rate for individual cases is listed in Table S2. Considering all cloud (precipitating cloud) scenarios, the mean CCN loss rates are -7.69±13.96 cm^{-3} (-10.45±15.56 cm^{-3}), -6.29±11.65 cm^{-3} (12.11±14.64cm^{-3}), and -4.94±7.96 cm^{-3} (-5.58±8.43cm^{-3}) for ACE-ENA summer, winter and SOCRATES, respectively. Indicating the ACE-ENA clouds suffer more substantial sub-cloud CCN loss than the SOCRATES, especially for the wintertime precipitating clouds. Recall that the assessment of $ACI_{r,CB}$ relies on the relative changes of r_c and N_{CCN}, while the different L_{CCN} for individual cases can result in the shrinking of the N_{CCN} variation ranges (imagine the abundant CCN are depleted by the coalescence scavenging). In other words, the given change in r_c corresponds to a narrowed change in N_{CCN}, hence mathematically presented as an enlarged $ACI_{r,CB}$. Hence, the coalescence-scavenging effect can not only deplete the sub-cloud CCN, but also quantitatively amplify the assessment of cloud microphysics susceptibilities (Feingold et al., 1999; Duong et al., 2011; Jung et al., 2016; Zheng et al., 2022b). In order to examine the potential impact of the aforementioned processes on the ACI assessment, a sensitivity analysis is conducted by simply retrospecting the sub-cloud $N_{CCN0.35\%}$ according to their L_{CCN} and recalculate the $ACI_{r,CB}$.

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As shown in Figure 8, the $ACI_{r,CB}$ values tend to decrease with the retrospective time, which indicates the retrospective CCN variation range is enlarged and counteracting the coalescence-scavenging amplification. The detailed illustration of the different $ACI_{r,CB}$ calculated from the scattered r_c and sub-cloud $N_{CCN0.35\%}$ is shown in Figure S6. Note that the $ACI_{r,CB}$ decreasing rates for the precipitating clouds (Fig. 8b) are not as strong as for all clouds because the non-precipitating clouds have smaller L_{CCN} largely due to weaker collision-coalescence. Hence, the time retrospected might quickly exceed the actual time scale of processing of cloud droplets. In other words, the time needed to store the sub-cloud CCN to the budget before cloud existence is shorter. Therefore, the retrospective of the sub-cloud CCN budget will yield an alternative assessment of ACI, assuming that the drizzle processes have not yet significantly impacted the sub-cloud CCN budget, especially for the assessment under the precipitating clouds. However, examining the exact precipitating timing is challenging since the aircraft provides a snapshot of the cloud and aerosol information. Thus, this retrospective study only provides a possible direction, and the result should be interpreted with caution.

5. Summary and Conclusions

Based on the aircraft in-situ measurements during ACE-ENA and SOCRATES, the vertical distributions and the evolutions of the aerosol, cloud, and drizzle properties are investigated under the cloud-topped MBL environments. The aerosols and CCN from SOCRATES are the highest among the three IOPs, followed by ACE-ENA summer and winter in descending order in both above- and sub-cloud regimes. The differences can be attributed to the aerosol sources in both regions, where the SOCRATES represents the pristine natural environment enriched by aerosols from marine biological productivity and without the contamination of anthropogenic aerosols, while the ACE-ENA features the aerosols from varied sources, including maritime and continental emissions, with distinct seasonal variations. Examining the aerosol size distributions in sub-cloud versus above-cloud regimes manifests the

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significant influence of cloud processing on aerosols. Physical processing like in-cloud Brownian capture can reduce Aitken mode aerosols, while the chemical processes transform Aitken mode aerosols to larger sizes, moving them toward the accumulation mode. In addition, the in-cloud coalescence processes shift sub-cloud aerosol residuals to larger sizes, explaining the observed increase in the tail-end of the aerosol distribution for all IOPs.

As for the cloud and drizzle properties, the SOCRATES clouds feature more and smaller cloud droplets than the ACE-ENA summertime and wintertime clouds, with the r_c growths, from cloud base to top, being 4.03 μm, 4.78 μm, and 5.85 μm for SOCRATES, ACE-ENA summer, and winter, respectively. The cloud-top entrainment mixing is evidenced in the observed decline of both N_c and LWC_c near the cloud top. The mean cloud-top entrainment rates (w_e) as a function of cloud top virtual potential temperature and vertical velocity are 0.570±0.834 cm s^{-1}, 0.581±0.560 cm s^{-1}, and 0.960±1.127 cm s^{-1} for SOCRATES, ACE-ENA summer and winter, respectively. The strongest w_e during ACE-ENA winter is owing to weaker cloud-top inversions and stronger near-cloud-top turbulence. For drizzle vertical distribution, N_d from the three IOPs all exhibit decreases from cloud top to cloud base, while D_{mmd} are in opposite directions with a maximum at the cloud base. The ACE-ENA wintertime clouds feature more prominent drizzle formation and evolution owing to the strong in-cloud TKE, especially the strongest vertical turbulence in the lower half of the cloud layer, which substantially enhances the collision-coalescence process, compared to the other two IOPs.

The cloud sub-adiabaticity f_{ad} values are 0.681±0.083, 0.476±0.106, and 0.447±0.100 for SOCRATES, ACE-ENA summer and winter, respectively. While the satellite retrievals of droplet number concentration heavily rely on the adiabatic cloud assumption and are usually given as a constant of f_{ad} = 0.8. Hence, the in-situ observational evidence of the variability of MBL cloud and drizzle microphysical properties over different regions shed light on the further understanding of the satellite

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retrievals, particularly the satellite-based aerosol-cloud interaction assessment (Painemal and Zuidema, 2011; Grosvenor et al., 2018; Painemal et al., 2021).

Notably, the sensitivity of cloud base precipitation rate (R_{CB}) to N_c is more pronounced for the ACE-ENA during both winter (with S_o = 1.638) and summer (1.229) compared to the SOCRATES (S_o = 0.979). This is partly due to the much higher R_{CB} induced by larger drizzle drops near the cloud base for ACE-ENA, a result of turbulence-driven in-cloud droplet interactions, especially under low N_c condition. Furthermore, R_{CB} can be approximated by a relationship involving N_c and H_c, as suggested in prior research. The relationships established in this study indicate that ACE-ENA clouds, are largely determined by H_c, while SOCRATES clouds are more influenced by the N_c.

The investigations of the ACI via the $ACI_{N,CB}$ and $ACI_{r,CB}$ indices reveal that during the ACE-ENA wintertime, N_c is more sensitive to changes in $N_{CCN0.35\%}$, indicating aerosols more readily activate to become cloud droplets. One influencing factor is the strong vertical turbulence, which speeds up the infusion of CCN into the cloud layer, thus aiding droplet formation. Furthermore, the presence of larger aerosols during ACE-ENA winter enhances the droplet activation process. The SOCRATES IOP highlights a higher $ACI_{r,CB}$, indicating a pronounced decrease in r_c with increasing $N_{CCN0.35\%}$. While the $ACI_{r,CB}$ in ACE-ENA is dampened by the presence of more larger cloud droplets near the cloud base, particularly under relatively higher $N_{CCN0.35\%}$. Note that the ACI indices from this study lie in the higher end of the ACI ranges estimated via remote sensing (McComiskey et al., 2009; Dong et al., 2015; Zheng et al., 2022a) because the aircraft assessment provides more connected circumstances between the aerosols and cloud layer. Arguably, the assessment of N_c responses to $N_{CCN0.35\%}$ would inevitably be affected by the collision-coalescence process near the cloud base, where simultaneously, the CCN replenishment buffers the N_c and the collision-coalescence process depletes N_c. Hence, finding a layer where these two effects maintain a dynamic balance in N_c might aid in a more accurate assessment and

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more fundamental understanding of the ACI, which might be revealed by the LES or parcel model simulations.

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

Data availability. The ACE-ENA field campaign data can be accessed from the Department of Energy Atmospheric Radiation Measurement data archive (https://iop.archive.arm.gov/arm-iop-file/2017/ena/aceena/). The SOCRATES field campaign data are publicly archived on the National

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Center for Atmospheric Research (NCAR) Earth Observing Laboratory (https://data.eol.ucar.edu/master_lists/generated/socrates/).

Author contributions. The original idea of this study is discussed by XZ, XD, and BX. XZ performed the analyses and wrote the manuscript. XZ, XD, BX, TL, and YW participated in further scientific discussions and provided substantial comments and edits on the paper.

Competing interests. At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

Acknowledgments. This work was supported by the NSF grants AGS-2031750/2031751/20211752 at the University of Arizona, Texas A&M University and Stanford University, respectively. The authors sincerely thank the investigators and mentors from the ACE-ENA and SOCRATES field campaigns for making the data publicly available.

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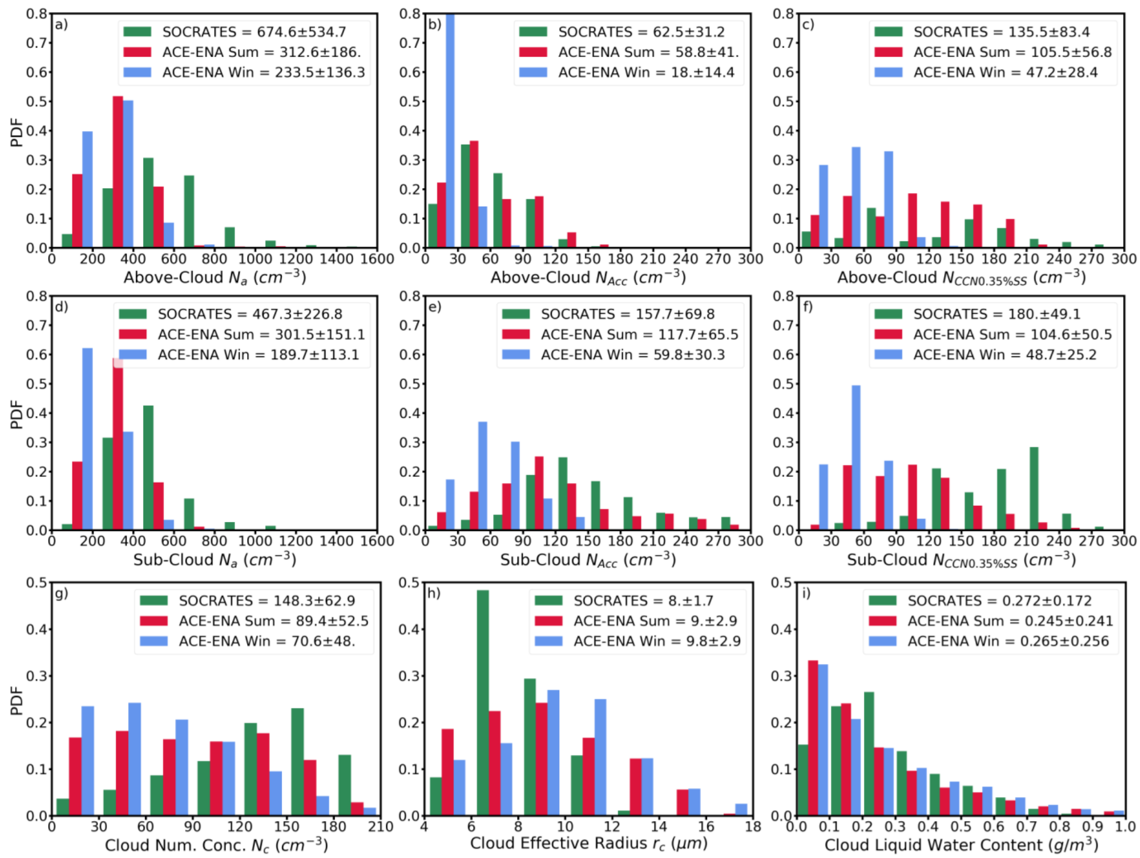

Figure 1. Probability Density Functions (PDFs) of N_a, N_{ACC} and $N_{CCN0.35\%}$ in the above-cloud (a, b, c) and sub-cloud (d, e, f) regimes; and the cloud microphysical properties of N_c (g), r_c (h), and LWC_c (f) within cloud layer. ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

https://doi.org/10.5194/egusphere-2023-2608
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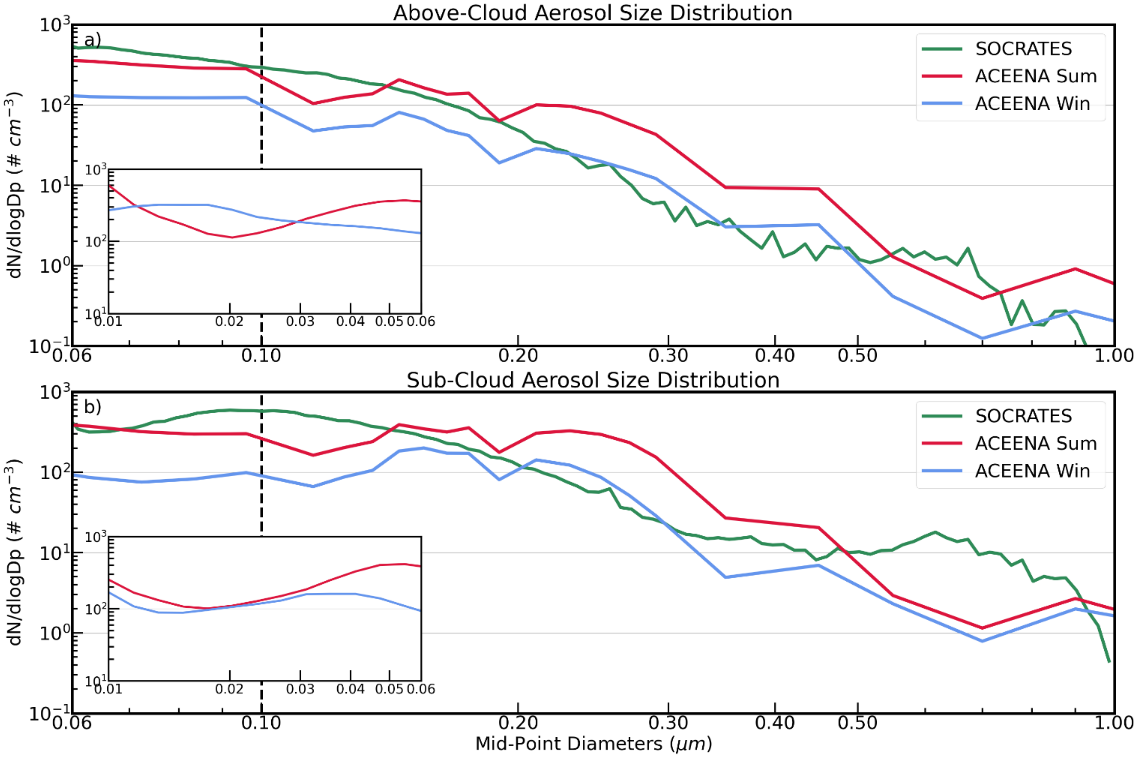

Figure 2. Aerosol size distributions (D_p = 0.06 – 1 μm) for above-cloud (a) and sub-cloud (b) regimes. The vertical dashed line at D_p = 0.1 μm denotes the demarcation between Accumulation mode and Aitken mode. The inner plots denote the Aitken mode size distribution (D_p = 0.06 – 1 μm) from ACE-ENA. ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

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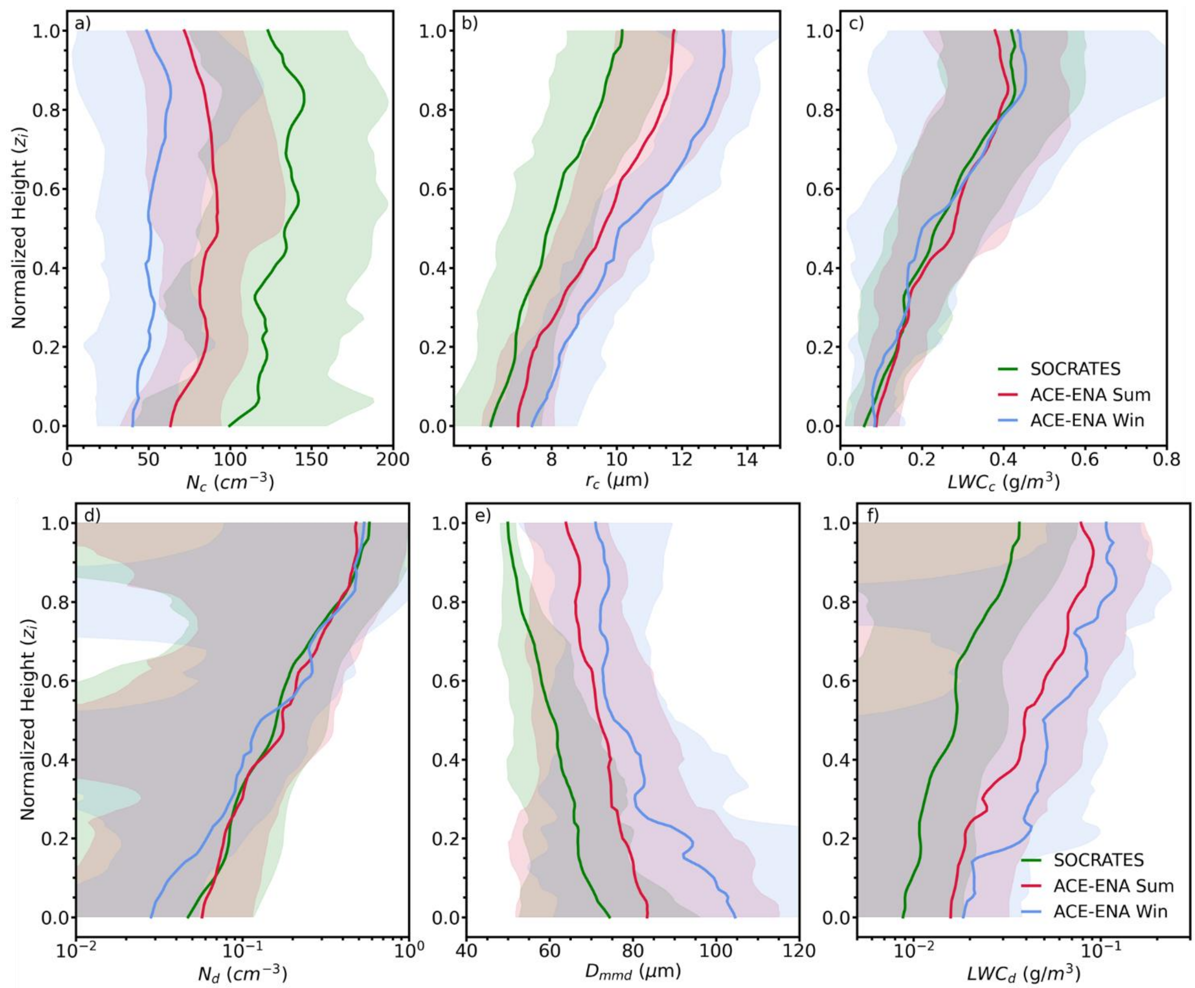

Figure 3. Vertical distributions of N_c (a), r_c (b), LWC_c (c), N_d (d), D_{mmd} (e), and LWC_d (f). Here the $z_i = 0$ denotes cloud base and $z_i = 1$ denotes cloud top.

https://doi.org/10.5194/egusphere-2023-2608
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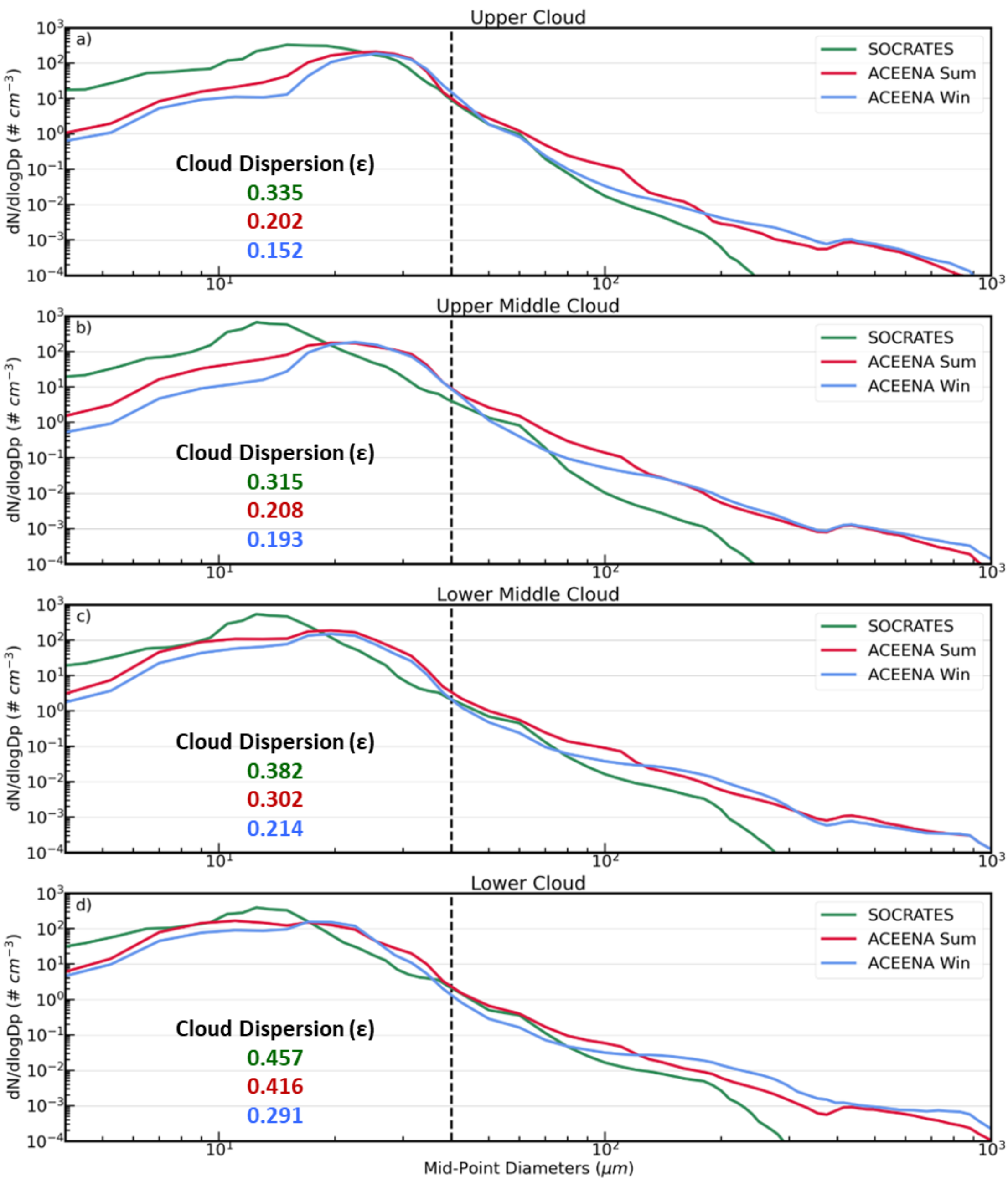

Figure 4. Cloud and drizzle size distributions for a) upper cloud ($z_i > 0.8$), b) upper-middle cloud ($0.5 \leq z_i < 0.8$), c) lower-middle cloud ($0.2 \leq z_i < 0.5$) and d) lower cloud ($z_i < 0.2$). The vertical dashed line at D_p = 40 µm denotes the demarcation between cloud droplets and drizzle drops. ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

https://doi.org/10.5194/egusphere-2023-2608
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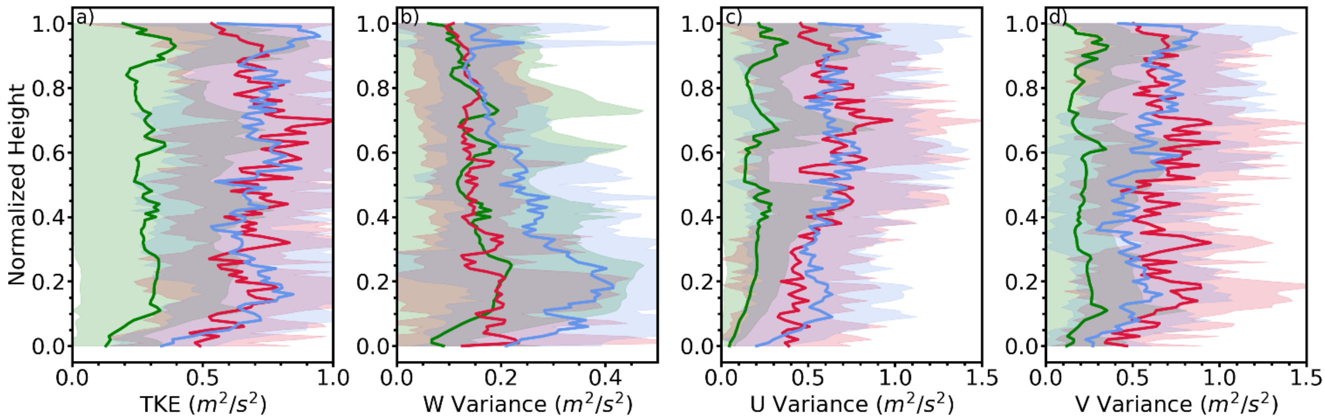

Figure 5. Vertical distributions of in-cloud TKE (a), w'^2 (b), u'^2 (c) and v (d). ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

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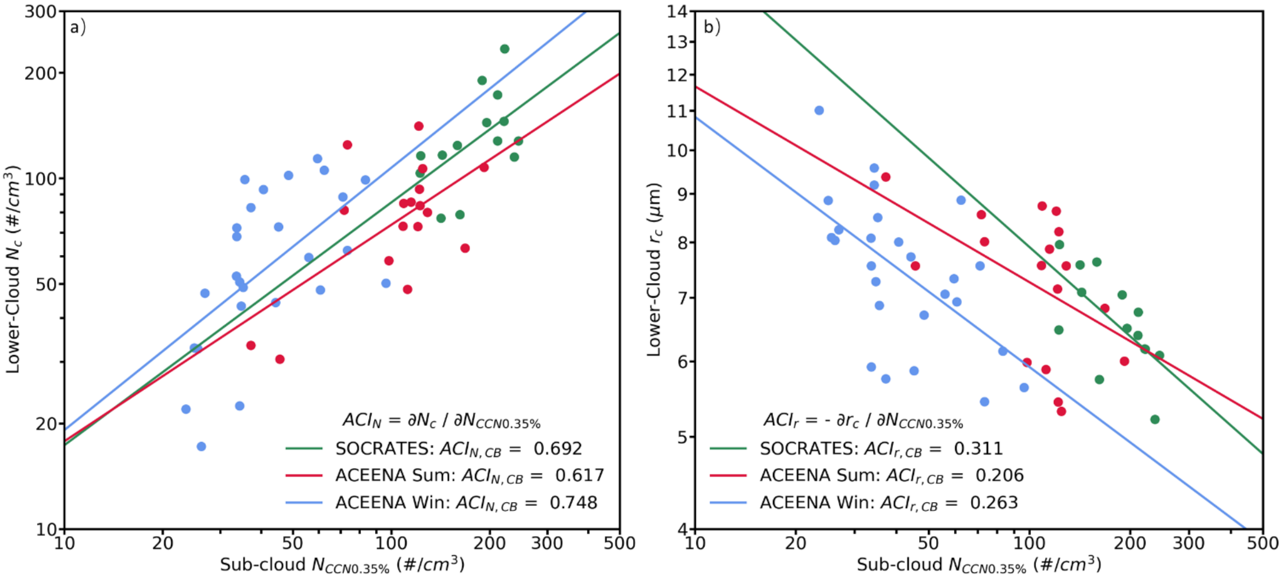

Figure 6. Scatterplots of the a) N_c and b) r_c at the lower-cloud (z_i < 0.2) against the sub-cloud $N_{CCN0.35\%}$. ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

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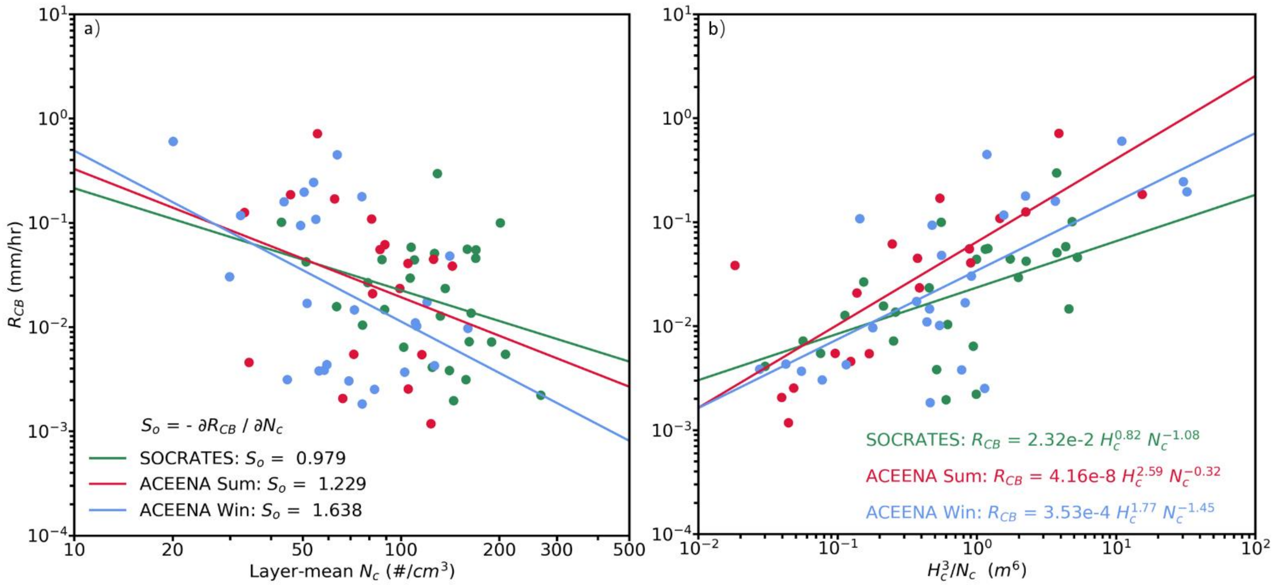

Figure 7. Scatterplots of the cloud base precipitation rate R_{CB} against the a) layer-mean N_c and b) H_c^3/N_c. ACE-ENA summer, winter and SOCRATES are color-coded with red, blue and green, respectively.

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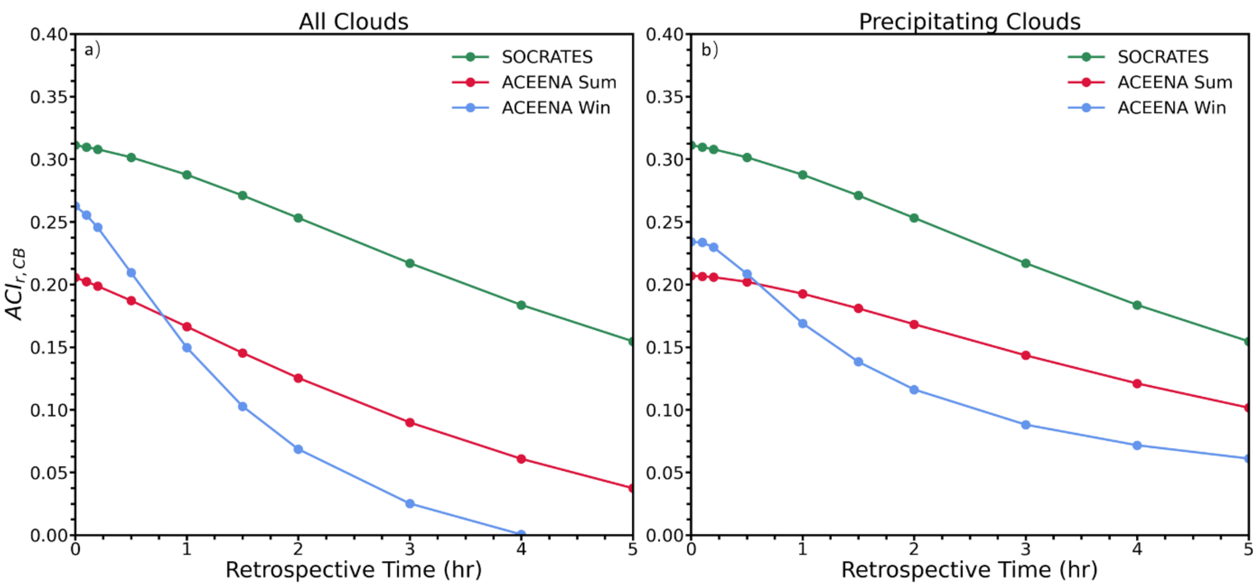

Figure 8. $ACI_{r,CB}$ as a function of the sub-cloud $N_{CCN0.35\%}$ retrospective time for a) all clouds and b) precipitating clouds.