



# **Organized Variations in MBL Cloud Microphysical Properties Observed by Aircraft and Satellite and Simulated by Model**

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## 22 Abstract

23 Marine boundary layer (MBL) clouds in subtropical regions strongly impact global energy  
24 balance, but complete understanding of the processes that control their microphysical properties  
25 remain elusive. We analyze aircraft in-situ measurements of MBL clouds for two selected cases  
26 from the ACE-ENA field campaign that contain mesoscale convective cells (MCCs) on the order  
27 of tens of kilometers embedded in the large-scale overcast cloud field. The aircraft flight tracks  
28 aligned with the MCC organization, such that vertically-stacked, horizontal flight legs alternated  
29 between sampling clouds along organized MCCs and sampling clouds between MCCs. This  
30 alignment is well-suited to study the distinctly different microphysical properties for the two cloud  
31 regimes. Clouds within organized MCCs had lower droplet concentrations, but larger droplet sizes  
32 and liquid water contents with enhanced drizzle relative to clouds between MCCs. While observed  
33 aerosol properties below these two cloud regimes are generally consistent with their corresponding  
34 cloud microphysical properties, preexisting organization of the aerosol field was probably not  
35 required in the development of the MCC organization. In contrast, the lower aerosol and CCN  
36 concentrations observed below the MCC cloud layer most likely developed from precipitation and  
37 coalescence scavenging. A cloud-resolving WRF model simulation with realistic large-scale  
38 forcing reproduces the MCC organization of the cloud field suggesting that updraft velocity is the  
39 key to explain the differences in cloud microphysics. Both observations and model simulations  
40 indicate that under moderate-heavy drizzling conditions, precipitation and coalescence scavenging  
41 dominates and drives spatial gradients of cloud droplets, aerosols and CCN concentrations rather  
42 than local sources.



## 43 1. Introduction

44 Owing to their substantial role in the Earth's radiation budget, and consequently, their effect on  
45 the Earth's climate, low-level stratiform clouds have been a topic of considerable interest since the  
46 publication of the classic paper describing their physics (Lilly, 1968). Marine boundary layer  
47 (MBL) clouds in subtropical regions strongly influence the regional and global climate system  
48 (e.g., Klein and Hartmann, 1993). Over the ocean, MBL clouds are common with a strong  
49 temperature inversion at the top of the MBL, which provides conditions favorable for MBL cloud  
50 formation (Lilly, 1968). These MBL clouds are maintained by vertical mixing, which is primarily  
51 due to the strong longwave radiative cooling at the cloud top generating turbulence to provide an  
52 upward moisture flux from the ocean surface (Albrecht et al., 1995; Rémillard et al., 2012; Wood,  
53 2012; Wood et al., 2015).

54 The climatic importance of the MBL cloud microphysical and macrophysical properties,  
55 particularly the cloud fraction (CF), cloud-droplet effective radius ( $r_e$ ), number concentration ( $N_c$ ),  
56 and liquid water content/path ( $LWC/LWP$ ), is widely recognized. Climate models disagree  
57 substantially in the magnitude of cloud feedback for the regimes of subtropical MBL clouds (Bony  
58 et al. 2005 and 2006, Lohmann et al. 2005), and suffer from the so-called 'too few, too bright'  
59 problem (Allan et al., 2007, Nam et al., 2012, Webb et al., 2013, IPCC 2013). The 'too few'  
60 problem, an underestimate in cloud amount, allows more solar radiation to reach the surface. The  
61 'too bright' problem, an overestimate in cloud albedo due to an overestimate in the amount of  
62 liquid water within the cloud, causes more sunlight to be reflected. It is therefore imperative to  
63 have more accurate MBL cloud properties through long-term ground-based observations, as well  
64 as aircraft in situ measurements, so that we can improve their representation in climate models.

65 Aerosol-cloud-precipitation interactions are a significant source of uncertainty for MBL clouds  
66 (e.g., Wood, 2012). Aerosol generation resulting from natural and anthropogenic activities is  
67 expected to have considerable, far-reaching effects on cloud development and the hydrological  
68 cycle. Though the aerosol direct effect can simply be thought of as a reduction of incoming solar  
69 radiation reaching the Earth surface, the aerosol indirect effect (AIE) involves a complex set of  
70 aerosol-cloud-precipitation interactions. AIEs include the alteration of cloud properties such as  
71 cloud lifetime, droplet size distribution, liquid water content and path ( $LWC$ ,  $LWP$ ), cloud optical  
72 depth (COD), and albedo (Penner et al., 2004; Dong et al., 2005, 2006, 2014; Ghan et al., 2016).



73 Several studies indicate that MBL clouds under the regions of relatively higher sub-cloud aerosol  
74 concentrations have reduced  $r_e$ , increased  $N_c$ , and enhanced  $LWC/LWP$  and COD than clouds under  
75 clean regions (Twohy et al., 2005; Lu et al., 2007; Dong et al., 2015). However, recent  
76 observational studies, e.g., Toll et al. (2017), indicate that the  $LWP$  response to increased aerosol  
77 concentrations is bidirectional and depends upon a host of different meteorological parameters.

78 The Eastern North Atlantic (ENA) is a region of persistent subtropical MBL clouds (Dong et al.,  
79 2014). The region is commonly covered by large swaths of stratocumulus cloud sheets. The  
80 stratocumuli commonly form to the east of the position of the subtropical ridge in a region of large-  
81 scale subsidence. Synoptic scale variability in low cloud cover has been associated with changes  
82 in the strength and position of the subtropical high (Wood, 2012). Even within synoptic regions  
83 with fully overcast low-level stratocumulus cloud decks, small scale organized mesoscale  
84 convective cells (MCCs) can develop (Miller et al. 1995). The Aerosol and Cloud Experiments in  
85 the Eastern North Atlantic (ACE-ENA) field campaign was conducted during two intensive  
86 observation periods (IOPs): early summer 2017 (June 21 to July 20) and winter 2018 (January 11  
87 to February 20) (Wang et al., 2019; Wu et al., 2020). The DOE Atmospheric Radiation  
88 Measurement (ARM) Aerial Facility (AAF) Gulfstream-159 (G-1) research aircraft flew from  
89 Terceira Island in the Azores during IOPs. There were 20 flights during the summer 2017 IOP and  
90 19 flights during the winter 2018 IOP. There are approximately 158 total hours with aircraft in-  
91 situ measurements during ACE-ENA IOP (Wang et al., 2019; Wu et al., 2020).

92 Two cases, July 18, 2017 and January 25, 2018, were selected during the ACE-ENA for this study.  
93 The aircraft in-situ measurements and satellite observations for the selected two cases reveal that  
94 there are significant, organized MCCs over an area of about 600 km<sup>2</sup>. The aircraft flight patterns  
95 were L shaped with one leg aligned with the cloud-level wind direction and the other leg across  
96 the cloud-level winds. In this study, we focus on these two cases in which aircraft sampled two  
97 distinct cloud regimes due to the alignment of the flight path with the mesoscale organization of  
98 the cloud field. In both cases, a portion of the flight path aligned with an organized band or MCC  
99 with enhanced drizzle, while the other portion of the flight path mostly crossed different organized  
100 bands or MCCs with light drizzle. The terms along-wind and cross-wind are used throughout this  
101 paper to distinguish the two different spatial regions sampled over the entire flight path. These



names are not meant to imply a causal link between the wind or flight direction and the observed differences in cloud microphysical properties.

In this study, we explore the large differences in cloud microphysical properties for two distinct cloud regimes embedded within large-scale overcast stratocumulus clouds. We further investigate the causes for the different MBL cloud microphysical properties for the July 18 case using a cloud-resolving WRF (CR-WRF) model simulation with realistic large-scale forcing. Section 2 specifies the in-situ aircraft data, satellite data, reanalysis meteorological data, and the CR-WRF model run used in this study. Results are shown in section 3 followed by a brief summary and discussion in section 4.

## 2. Data and Model

The aircraft data used in this study come from the ACE-ENA field campaign collected on July 18, 2017 and January 25, 2018. Cloud and drizzle microphysical properties are derived from measurements made by the Fast Cloud Droplet Probe (FCDP) and the 2-Dimensional Stereo (2DS-V) probe at 1 Hz. Microphysical parameters are computed each second using observations of droplet size distributions (DSDs) over a range of particle size bins. Aircraft position is provided by the Aircraft-Integrated Meteorological Measurement System (AIMMS-20).

Cloud microphysical properties are computed from FCDP observations of droplet concentrations in 18 discrete size bins, which include droplets with diameters ranging from 1.5 to 46  $\mu\text{m}$ . Drizzle microphysical properties are computed from 2DS-V observations of droplet concentrations in 40 size bins, which includes drops with diameters ranging from 45 to 975  $\mu\text{m}$ . An observation is considered to contain cloud if the cloud droplet number concentration ( $N_c$ ) is greater than 5  $\text{cm}^{-3}$  and drizzle if the drizzle droplet number concentration ( $N_d$ ) is greater than 0.01  $\text{cm}^{-3}$ . Only observations verified to contain cloud or drizzle are included in the results presented. The cloud condensation nuclei (CCN) concentrations below cloud base and above cloud top measured by the Dual-Column CCN Counter were used to determine the number concentration of activated CCN at two supersaturation levels ( $S=0.15\%$  and  $S=0.35\%$ ) at 1 Hz. In this study, the CCN concentrations ( $N_{CCN}$ ) at  $S=0.15\%$  are used. The aerosol concentration below the cloud layer was observed by the Passive Cavity Aerosol Spectrometer (PCASP). The PCASP has 30 size bins, which include particle diameters ranging from 100 to 3200 nm. We used the PCASP observed aerosol concentrations rather than the Fast Integrated Mobility Spectrometer (FIMS) and the



132 Condensation Particle Counter (CPC) because the data have fewer spikes, its size range spanning  
 133 the accumulation size mode is a good proxy for condensation nuclei, and it is an external probe,  
 134 which alleviates concerns about processing time delay.

135 In addition to aircraft in-situ measurements, concurrent satellite observations and retrievals are  
 136 used in this study in order to investigate the spatiotemporal variations of MBL cloud properties.  
 137 The MBL cloud optical depth  $\tau$  and cloud-droplet effective radius  $r_e$  retrieved from the Meteosat-  
 138 9 satellite operated by the European Organization for the Exploitation of Meteorological Satellites  
 139 (EUMETSAT) were used to look for patterns in the large-scale horizontal MBL cloud structures  
 140 (Minnis et al., 2011). The large-scale meteorological conditions are obtained from the ERA5  
 141 reanalysis.

142 The Weather Research and Forecast (WRF) model version 3.6 is employed in this study to simulate  
 143 the case on July 18, 2017. Four nested domains are used with a horizontal resolution of 19.2 km,  
 144 4.8 km, 1.2 km, and 300 m, respectively. The innermost domain is configured as a quasi large-  
 145 eddy simulation with the 3D Smagorinsky first-order closure for eddy coefficient computation.  
 146 The 65 stretched sigma levels are used with a 40-m vertical resolution in PBL. The large-scale  
 147 forcing is adopted from ERA5 reanalysis (25 km resolution). To accurately depict cloud  
 148 microphysical processes in the MBL cloud, a spectral bin scheme is employed which utilizes 33  
 149 bins to represent cloud/rain drops and aerosols separately (Wang et al., 2013). The model integrates  
 150 from 12:00 UTC, July 17 to 00:00 UTC, July 19 and the first half day is considered as spin-up.  
 151 Shortwave and longwave radiation transfer calculations are accounted for by the Goddard and  
 152 RRTM schemes, respectively.

### 153 **3. Results and Discussion**

#### 154 **3.1 Cloud Results**

155 Figure 1 shows the horizontal and vertical flight paths on July 18, 2017 and January 25, 2018 over  
 156 the time that cloud data were collected for this study. In both cases, the aircraft flew at several  
 157 nearly constant altitude paths (horizontal legs) spanning from near cloud base to near cloud top.  
 158 The horizontal flight legs were vertically stacked, repeating L shaped patterns. In both cases, one  
 159 arm of the L was closely aligned with the cloud-level wind direction and motion, while the other  
 160 arm was across the cloud-level wind direction. Horizontal legs in the along-wind direction are



161 depicted in blue, while horizontal legs in the cross-wind direction are red. Each of these horizontal  
162 segments contains about 270 one-second observations of the DSD and is about 25 km in length  
163 using a representative aircraft speed of  $90 \text{ ms}^{-1}$ . The designations along-wind and cross-wind are  
164 simply used to separate the data collected along each direction. These names are not meant to  
165 imply a causal link between the wind or flight direction and the observed differences in cloud  
166 microphysical properties. However, what initially caught our attention was the distinctly different  
167 cloud microphysical properties observed in the along-wind flight legs compared with the cross-  
168 wind flight legs in the July 18 case. We later identified January 25 as a similar case observed  
169 during the winter IOP. Before detailing the distinctly different cloud microphysical properties  
170 along each flight leg direction, we will briefly look at the large-scale synoptic patterns from these  
171 two days.

172 The synoptic patterns at cloud altitude are shown in Figure 2. On July 18, the observation area was  
173 in between a high-pressure system well to the west-southwest and a low-pressure system to the  
174 northeast. The prevailing wind at 900 hPa was northwesterly. On January 25, the center of the 850  
175 hPa high was just a few hundred km southwest of the observation area, and the prevailing wind  
176 was north-northeasterly. The pressure gradient and hence cloud level winds are stronger in July.  
177 Mean wind speeds can be estimated from in-situ observations on the aircraft and the interpolated  
178 sounding at ENA site by averaging the wind speed over the cloud altitude range and observation  
179 time. The mean cloud-level wind speed on July 18 was  $7.7 \text{ ms}^{-1}$  from aircraft measurements and  
180  $7.5 \text{ ms}^{-1}$  from the interpolated sounding. The mean cloud-level wind speed on January 25 was  $2.9$   
181  $\text{ms}^{-1}$  from aircraft and  $0.84 \text{ ms}^{-1}$  from the interpolated sounding. The wind arrows in Fig. 2 are  
182 fixed size and do not indicate wind speed.

183 Satellite retrievals of COD provide a large-scale view of cloud horizontal structures. Figure 3  
184 shows the satellite-retrieved COD on July 18 at 10:00 and 10:30 UTC. Both images show bands  
185 with enhanced COD that roughly line up with the cloud-level wind direction (NW – SW). Between  
186 these bands, COD is reduced. Examination of the half-hourly satellite retrievals of COD over this  
187 region from 9:00 to 11:00 UTC indicates that the cloud field tended to organize into bands of  
188 higher COD that lined up along the prevailing winds. Between 10:00 and 10:30 UTC, as the aircraft  
189 was sampling the middle and upper portions of the clouds, a band of enhanced COD is aligned



190 with the along-wind flight track, while the cross-wind flight track flew through several bands  
 191 including their edges and centers with relatively large variations in COD.

192 Figure 4 shows the satellite retrieved COD on January 25 at 12:00 and 12:30 UTC. Again, there  
 193 are horizontal structures in the retrieved COD, but it is difficult to make out a dominant pattern of  
 194 banded structures as observed on July 18. Examination of the half-hourly satellite retrievals of  
 195 COD over this region from 11:00 to 1:00 UTC indicates that areas of enhanced COD tend to move  
 196 slowly along the prevailing northerly winds, though there is also dissipation and strengthening of  
 197 cloud elements happening. Between 12:00 and 12:30 UTC, as the aircraft was sampling the middle  
 198 and upper portions of the clouds, a region of enhanced COD covered much of the along-wind flight  
 199 track, while most of the cross-wind flight track was covered by a region of much lower COD. This  
 200 is consistent with the lower percentage of 1 Hz in-situ measurements in the cross-wind flight track  
 201 that met the cloud identification criteria in the middle of the clouds depicted in Fig. 1d.

202 The vertical thermodynamic structures obtained from the ARM interpolated sounding product are  
 203 shown in Figure 5. The atmosphere is well mixed below the cloud base with a strong temperature  
 204 inversion and drying immediately above the cloud top on both days, which is common for this  
 205 region. The lower atmosphere contains much more water vapor on July 18 with a lower cloud base  
 206 (~600 m) compared to January 25 (~1300 m). The mean cloud thickness is slightly greater on July  
 207 18 (~400 m) compared to January 25 (~300 m).

208 Figures 6 and 7 show that there are large systematic differences in the aircraft in-situ measured  
 209 cloud and drizzle microphysical properties between the along- and cross-wind legs on July 18 and  
 210 January 25, respectively. Cloud microphysical properties are computed from each measurement  
 211 of the FCDP droplet size distribution (DSD) from 1.5 to 46  $\mu\text{m}$ . Mean cloud properties are  
 212 computed by averaging all 1 Hz measurements that meet the cloud present criteria ( $N_c > 5 \text{ cm}^{-3}$ )  
 213 along each defined horizontal leg. Drizzle microphysical properties are computed from each  
 214 measurement of the 2DS-V drop size distribution from 45 to 975  $\mu\text{m}$ . Mean drizzle properties are  
 215 computed by averaging all 1 Hz measurements that meet the drizzle present criteria ( $N_d > 0.01 \text{ cm}^{-3}$ )  
 216 along each defined leg.

217 On July 18 (Figure 6), the mean cloud-droplet radius ( $r_c$ ) and cloud liquid water content ( $LWC_c$ )  
 218 increase with altitude on both the along- and cross-wind sides as expected, but their means at each  
 219 leg altitude are quite different. Mean  $N_c$  is largest in the middle of the clouds and decreases toward





cloud top probably due impacts from cloud top entrainment and growth of cloud droplets to drizzle drops. The along-wind side has a larger mean  $r_c$  and  $LWC_c$ , but lower  $N_c$  than the cross-wind side at each leg altitude. The vertical distributions of drizzle drop radius,  $r_d$ , and number concentration,  $N_d$ , are opposite to their cloud counterparts with high concentrations of small drizzle drops near the cloud top and fewer, but larger drizzle drops near the cloud base. The highest  $N_d$  near the cloud top was converted from cloud droplets through the autoconversion process (Wood, 2005) and additional water vapor through the evaporation of smaller cloud droplets during cloud-top entrainment. These drizzle drops fall when the gravitational force exceeds the buoyancy force and grow further by collecting cloud droplets and small drizzle drops through the collision-coalescence process. As drizzle drops fall, accretion becomes increasingly important (Wood, 2005). Figure 6 also shows that more drizzle is being generated on the along-wind side as  $r_d$ ,  $N_d$ , and  $LWC_d$  are generally larger at the same leg altitudes throughout the clouds. The mean vertical profiles of the drizzle microphysical properties within the cloud vary generally as expected for both legs. Specifically,  $r_d$  decreases but  $N_d$  and  $LWC_d$  increase with altitude.

Figure 7 from January 25 is similar to Fig. 6 in that the profiles of the mean cloud and drizzle microphysical properties vary generally as expected with altitude. It is also similar in that the along-wind side has a lower  $N_c$ , but larger  $r_c$  and  $LWC_c$  with more drizzle than the cross-wind side. One difference is that the  $N_c$  is much smaller in the middle of the cloud on January 25. This is consistent with lower aerosol and CCN concentrations and less water vapor below the cloud layer on January 25 as described later.

Figures 8 and 9 show the differences between the cloud microphysical properties computed each second from the along- and cross-wind measurements of the DSD in the middle portion of the clouds on July 18 and January 25. Restricting in-situ measurements to the mid-cloud horizontal legs reduces the impact of cloud edge effects. Figures 8a and 9a show the mean DSDs along horizontal flight legs at approximately 850 m and 1380 m in altitude, respectively. The mean cloud DSDs based on FCDP observations extend from 1.5 to 45  $\mu\text{m}$  and the mean drizzle DSDs based on 2DS-V observations extend from 45 to 975  $\mu\text{m}$ . In both cases, there are higher concentrations of smaller cloud droplets with diameters less than 20 to 25  $\mu\text{m}$  observed on the cross-wind side, and higher concentrations of larger cloud droplets and drizzle drops on the along-wind side. Large



cloud droplets ( $r_c \sim 20 \mu\text{m}$ ) on the along-wind side in both cases provide embryonic drizzle droplets that grow to drizzle-sized drops through the collision-coalescence process near the cloud top.

The scatterplots in Fig. 8 for July 18 show almost bi-modal type differences between the along- and cross-wind observations. Generally, the along-wind observations have relatively tighter clusters with smaller  $N_c$ , but larger  $r_c$  and  $LWC_c$  than the cross-wind observations. The scatterplots in Fig. 9 for January 25 have much more overlap between the along- and cross-wind observations. But there is a cluster of along-wind observations with relatively lower  $N_c$ , larger  $r_c$  and higher  $LWC_c$  where no cross-wind observations fall. The larger  $r_c$  and higher  $LWC_c$  along-wind sides on both days most likely result from stronger updrafts and low-level moisture convergence, which is more likely to lead to drizzle formation. Further investigation this point is carried out by running a WRF model simulation, which is included section 3.3. The next section discusses cloud-aerosol interactions using observed aerosol data.

### 3.2 Aerosol Discussion

A plausible explanation for some of the differences in cloud properties might be different aerosol concentrations in along- and cross-wind sides. For example, fewer available CCN in the along-wind direction could result in lower  $N_c$  and large  $r_c$  (Platnick and Twomey, 1994; Painemal et al., 2015). The mean aerosol concentrations in the along- and cross-wind legs are similar to their corresponding  $N_c$  values in the middle of the cloud layer ( $80 \text{ cm}^{-3}$  and  $120 \text{ cm}^{-3}$ ) as shown in Figure 6b, while the mean CCN concentrations are greater and lower, respectively, than their  $N_c$  values near the cloud base ( $40 \text{ cm}^{-3}$  along and  $65 \text{ cm}^{-3}$  cross). This is possible since the CCN concentration was measured at 0.15% supersaturation. Using the 0.35% supersaturation observations (not shown), the mean CCN concentrations just below cloud base increase to just over  $70 \text{ cm}^{-3}$  in both the along- and cross-wind legs, which both are greater than  $N_c$  near the cloud base, but still lower than the  $N_c$  values in the middle of the cloud layer. Hudson and Noble (2013) report that effective cloud supersaturations often exceed 1% in stratus clouds that form in clean marine air, and thus both CCN measurements may be underestimates. We believe the aerosol concentrations measured by the PCASP instrument provide the best estimates of available CCN.

As shown in Figure 10, the mean observed aerosol and CCN concentrations are lower for the along-wind side relative to the cross-wind side for both (a) above ocean and (b) below cloud base legs. Specifically, the mean aerosol concentration measured just below cloud base on the along-



wind side is 28% lower than on the cross-wind side. The lower aerosol and CCN concentrations on the along-wind side may be largely due to drizzle coalescence scavenging. Following the method of Wood (2006), the loss rate of CCN over the entire cloud-containing mixed layer due to drizzle scavenging can be estimated from the cloud base precipitation rate, the depth of the MBL, and the cloud thickness. We computed precipitation rates at cloud base at 1 Hz using the observed DSDs for the horizontal legs near cloud base. Mean precipitation rates of  $6.4 \text{ mm day}^{-1}$  (moderate to heavy drizzle) and  $0.93 \text{ mm day}^{-1}$  (modest drizzle) were obtained by averaging over the cloud base legs for the along- and cross-wind measurements, respectively. The corresponding CCN loss rates are  $-463 \text{ cm}^{-3} \text{ day}^{-1}$  ( $-19 \text{ cm}^{-3} \text{ hr}^{-1}$ ) and  $-100 \text{ cm}^{-3} \text{ day}^{-1}$  ( $-4 \text{ cm}^{-3} \text{ hr}^{-1}$ ). The greater along-wind CCN loss rate is consistent with the lower aerosol concentrations observed on the along-wind side. While there are many factors that determine aerosol/CCN concentration, the estimated differential scavenging rate (along-wind minus cross-wind) by itself could produce the observed differences in below cloud aerosol in  $\sim 1.5 \text{ h}$ .

Figure 11 shows that the observed aerosol and CCN concentrations on January 25 are also lower on the along-wind leg, which has lower  $N_c$  and more drizzle, relative to the cross-wind leg, which higher has lower  $N_c$  and less drizzle. However, for this case the observed aerosol concentrations are lower than the middle cloud  $N_c$  and the observed CCN concentrations are lower than the cloud base  $N_c$  for both the along- and cross-wind legs. The observed aerosol concentration on the along-wind leg is 24% lower than on the cross-wind leg. Again, this difference may be largely due to drizzle coalescence scavenging. Mean precipitation rates at cloud base are  $11.2 \text{ mm day}^{-1}$  (moderate to heavy drizzle) and  $0.33 \text{ mm day}^{-1}$  (light drizzle) on the along- and cross-wind legs. The corresponding CCN loss rates are  $-277 \text{ cm}^{-3} \text{ day}^{-1}$  ( $-12 \text{ cm}^{-3} \text{ hr}^{-1}$ ) and  $-10 \text{ cm}^{-3} \text{ day}^{-1}$  ( $-0.4 \text{ cm}^{-3} \text{ hr}^{-1}$ ). Again, while there are many factors that determine aerosol/CCN concentration, the estimated differential scavenging rate by itself could produce the observed differences in above ocean aerosol in  $\sim 1 \text{ h}$ .

Figure 12 also qualitatively supports the idea that the along- and cross-wind differences in below cloud aerosol are highly influenced by the differential rates of drizzle coalescence scavenging. In addition to having lower mean aerosol concentrations, the along-wind measurements have higher concentrations of large accumulation mode aerosols (diameter  $> 1.0 \mu\text{m}$ ) and lower concentrations of small accumulation mode aerosols (diameter  $< 0.3 \mu\text{m}$ ). The shift in aerosol size distribution is



309 expected as evaporated drizzle drops leave behind larger aerosols that combine the CCN collected  
310 during the coalescence process. On July 18, this effect is more pronounced in the leg just below  
311 the cloud base relative to the leg just above the ocean. The shift to larger aerosols on January 25  
312 observed just above the ocean is obvious, but even less pronounced. It would have been nice to  
313 have observations from a horizontal flight leg just below the cloud base on January 25.

### 314 3.3 WRF simulation

315 To further investigate different cloud microphysical properties in the along- and cross-wind legs,  
316 we employed a nested CR-WRF simulation with a spectral bin microphysics scheme to simulate  
317 the evolution of the cloud field on July 18. The model run starts from 12:00 UTC, July 17 to 00:00  
318 UTC, July 19 and the first half day is considered as model spin-up. The model begins with the  
319 large-scale observed background aerosol, which is uniform across the inner domain. The model  
320 shows that the specific meteorological conditions present on July 18 are conducive for the  
321 formation of organized bands of enhanced drizzle that align with the wind direction. Figure 13a  
322 shows that the WRF simulation does produce the basic pattern of COD captured by the satellite  
323 retrievals in Figure 3. Specifically, organized bands of higher COD line up with the wind direction  
324 without prescribing variation in the below cloud CCN.

325 Figure 13b shows the model generated vertical velocity at 848 m and 10:00 UTC. The banded  
326 regions of enhanced COD in the model are associated with narrow bands of enhanced upward  
327 vertical velocity. While the largest COD values coincide with the narrow band of upward vertical  
328 velocity, the band of enhanced COD extends laterally outward, indicating that drizzle formation  
329 extends laterally away from the updraft core. The organized structure of the mesoscale convective  
330 cells embedded in the overcast stratus cloud deck on July 18, 2017 was captured by both the  
331 satellite observations and model simulations. Substantial drizzle is often associated with cellular  
332 convection in stratocumulus layers (Albrecht et al., 1995; Miller et al., 1995). The stronger updraft  
333 core that coincided with the along-wind flight path enhances the activation of CCN (Hudson and  
334 Noble, 2013) which increases  $LWC_c$ . The enhanced updraft also lifts more large cloud droplets  
335 into the upper part of the cloud layer where cloud droplets start to convert into drizzle drops by the  
336 autoconversion process. Drizzle-size drops that form near the top of the cloud, then start to fall  
337 and grow by collecting cloud droplets and smaller drizzle drops through the accretion process,  
338 which further reduces  $N_c$  and  $N_d$ .



339 The drizzle growth processes of collision and coalescence effectively combine the CCN within the  
340 many collected droplets into a single, larger CCN that is left behind when drizzle drops evaporate  
341 after falling below the cloud base. In regions of active drizzle, this recycling of CCN through the  
342 cloud results in the reduction in the total CCN concentration (mainly loss of smaller CCN), but an  
343 increase in large CCN below cloud base. In both cases studied, clouds that were observed to be  
344 producing more drizzle (the along-wind flight segments) were associated with fewer, but larger  
345 CCN below the cloud base relative to the clouds that were producing less drizzle (the cross-wind  
346 flight segments).

#### 347 4. Summary and Conclusions

348 Aircraft in-situ measurements obtained on July 18, 2017 and January 25, 2018 during the ACE-  
349 ENA IOP reveal two distinct cloud regimes on each day within overcast MBL clouds. Specifically,  
350 the aircraft flight patterns consisted of horizontal legs at several altitudes below and through the  
351 cloud layers that alternated between along-wind flight paths sampling clouds within organized  
352 regions of enhanced convection and drizzle and cross-wind flight paths mostly sampling clouds  
353 through several MCCs or enhanced bands which include the edges and centers of MCCs. This  
354 alignment of the flight track and mesoscale cloud organization is well suited to study the distinctly  
355 different cloud microphysical properties for the two cloud regimes. The designations along-wind  
356 and cross-wind are simply used to distinguish the two spatial regions sampled. By chance the  
357 mesoscale cloud structures of enhanced convection fell within the along-track portions of the flight  
358 paths on these two days. We did not find evidence for a causal link between the wind or flight  
359 direction and the observed differences in cloud microphysical properties. The different cloud  
360 microphysical properties in the along- and cross-wind directions are primarily due to sampling  
361 different cloud regimes within the mesoscale pattern of cloud variation.

362 The aircraft in-situ measurements in the along-wind legs had smaller  $N_c$ , larger  $r_c$  and  $LWC_c$ , and  
363 more drizzle than those in the cross-wind legs. Based on the aircraft in situ measurements of cloud  
364 and aerosol properties, as well as satellite retrievals and a CR-WRF simulation, we conclude that  
365 the different MBL cloud microphysical properties in the along- and cross-wind flight observations  
366 are not due to variations in the background aerosol conditions. In other words, different aerosol  
367 and CCN concentrations below the cloud layer in the along- and cross-wind legs are not the causes  
368 of different cloud microphysical properties between two legs. In contrast, we did find that the



369 lower aerosol and CCN concentrations below the cloud layer in the along-wind legs were modified  
370 by drizzle coalescence scavenging. Specifically, the along-wind legs had enhanced drizzle and  
371 large CCN loss rates but higher concentrations of large accumulate mode aerosols, relative to the  
372 cross-wind observations where less drizzle production was observed.

373 The large-scale cloud field for these cases is best represented as bi-modal cloud structures with  
374 organized mesoscale structures of enhanced convection and drizzle that form within an overcast  
375 MBL cloud field. Combining the along- and cross-wind observations and treating the cloud field  
376 as a homogenous MBL cloud would be misleading in these cases. Mean vertical profiles produced  
377 by combining the along- and cross-wind observations shown in Figures 6 and 7 would not be  
378 representative of the cloud microphysical properties observed in either the organized regions of  
379 enhanced convection or the regions falling between them. Similarly, scatterplots and mean DSDs  
380 that do not distinguish the underlying cloud organization would miss the distinctly different  
381 microphysical properties of the clouds within each regime. This bi-modal behavior is crucial to be  
382 considered for both model simulations and ground-truth evaluation of remote sensing retrievals.

383 The results from this study have important implications on studying aerosol-cloud-precipitation  
384 interactions using aircraft, surface, or satellite measurements. Even with similar aerosol  
385 background conditions and within the same dynamic regime, clouds may organize into regions or  
386 bands with distinctly different microphysical properties. Caution should be taken when analyzing  
387 aerosol indirect effect with fixed  $LWP/LWC$ , since the differences in cloud microphysics might  
388 result from the cloud structure variations rather than from aerosol effects.

389



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406 (<https://cds.climate.copernicus.eu/cdsapp#!/home>).



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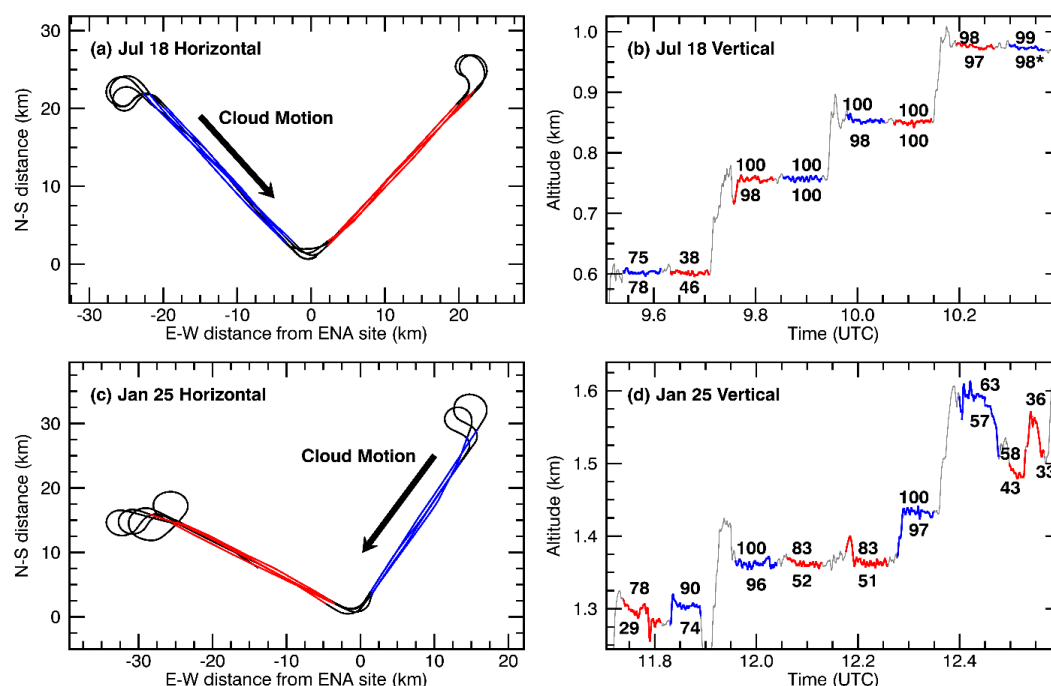
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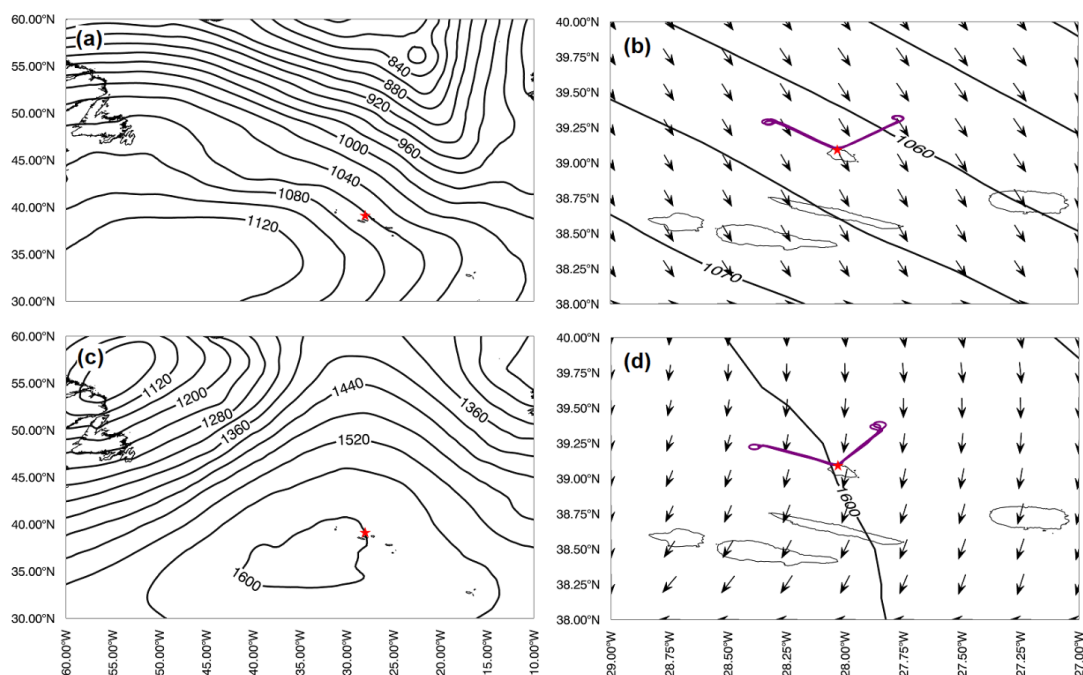
532

533 **Figure 1.** Horizontal and vertical flight paths for the periods of data collection on July 18 (a, b)  
 534 and January 25 (c, d). Along-wind legs are blue and cross-wind legs are red. Horizontal distances  
 535 are relative to the ARM radar facility at (39.09°N, 28.03°W). The numbers above and below the  
 536 flight legs in the vertical plots are the percentage of observations along the leg that meet the criteria  
 537 for cloud and drizzle, respectively (\* denotes that over 80% of the 2DS-V observations were  
 538 missing).

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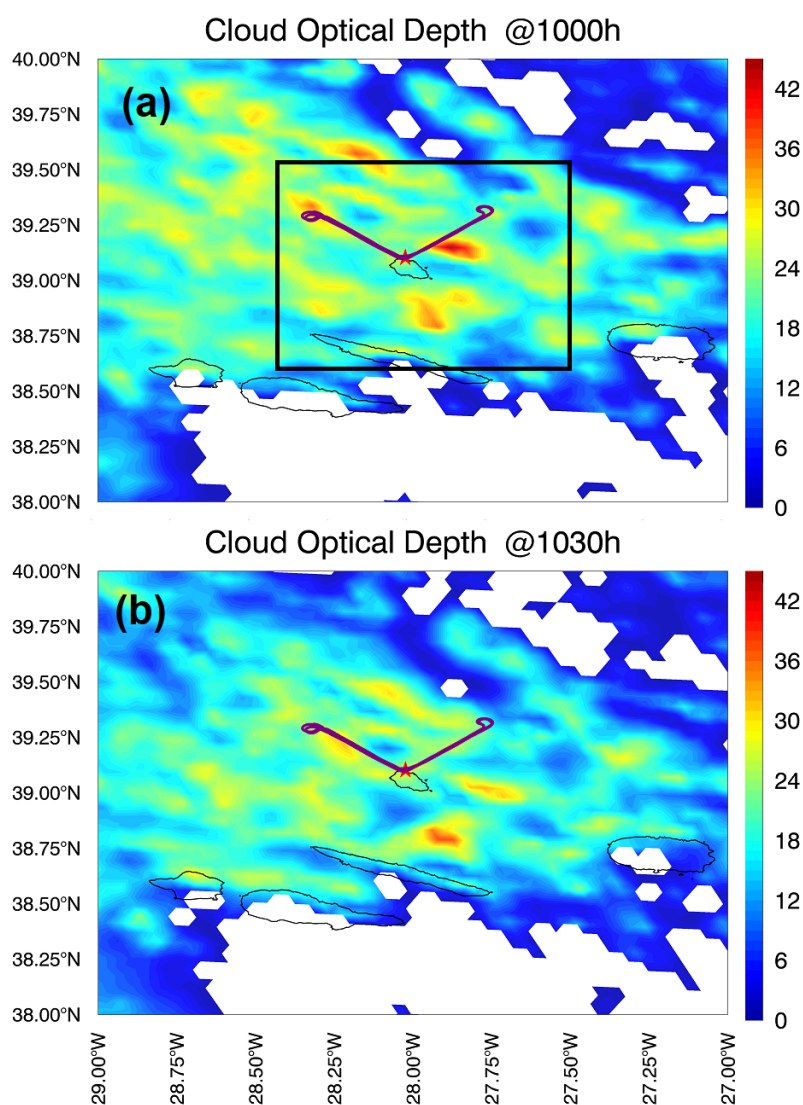
542

543 **Figure 2.** 900 hPa geopotential height from ERA5 on July 18, 2017 (a, b) and 850 hPa geopotential  
 544 height on January 25, 2018 (c, d). The red star indicates the position ARM radar facility. Arrows  
 545 in (b) and (d) represent the wind direction only within a grid box of  $2^\circ \times 2^\circ$ , not the wind speed.  
 546 Purple lines show the horizontal flight track.

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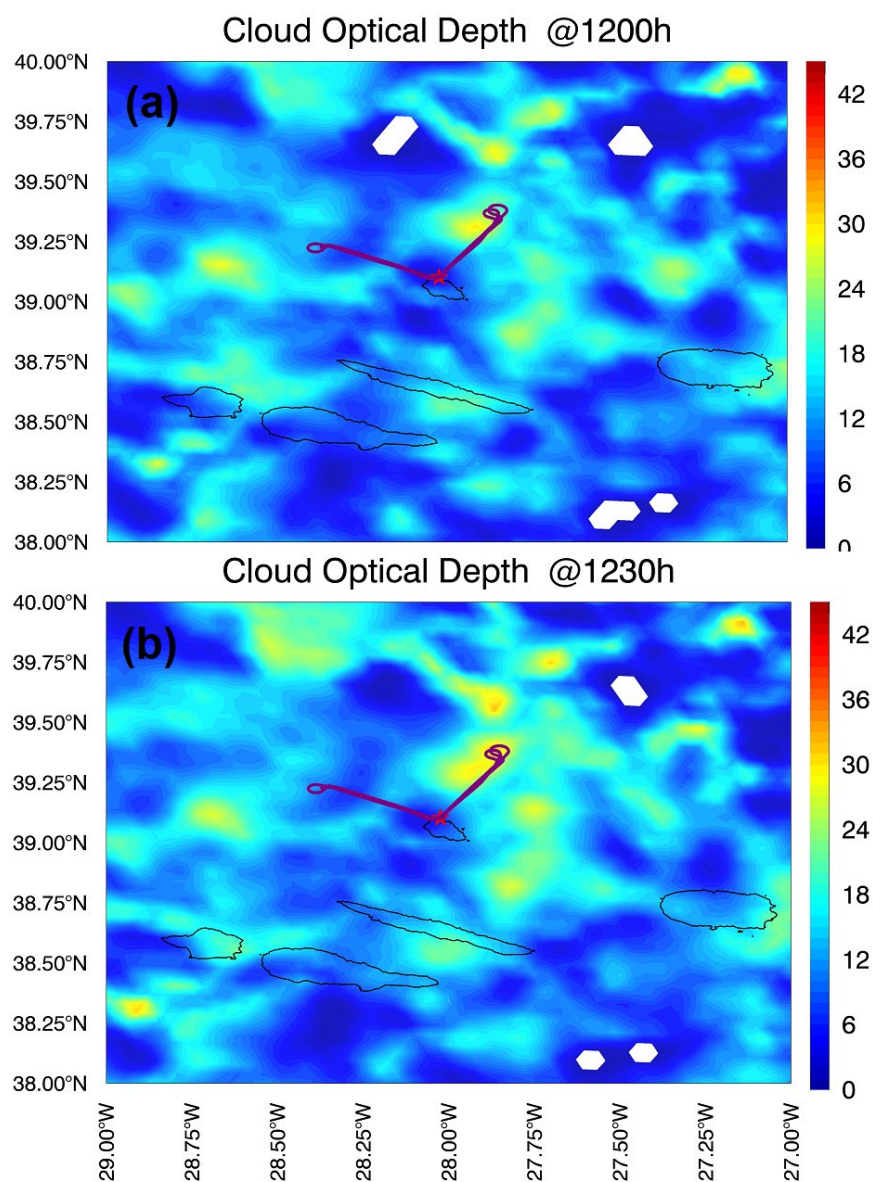
551 **Figure 3.** Meteosat images (4-km resolution) for cloud optical depth on July 18, 2017, at (a) 10  
 552 and (b) 10:30 UTC over a 2° by 2° box centered on the ARM radar facility marked with a star.  
 553 Horizontal flight track shown in purple. Region inside black box in (a) shows corresponding WRF  
 554 simulation inner domain shown in Fig. 13.

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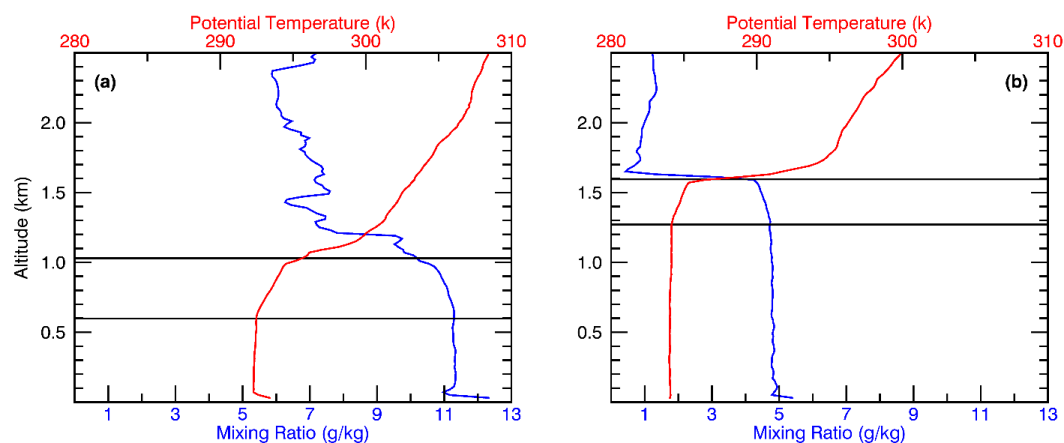
558

559 **Figure 4.** Meteosat images (4-km resolution) for cloud optical depth on January 25, 2018, at (a)  
 560 12:00 and (b) 12:30 UTC over a 2° by 2° box centered on the ARM radar facility marked with a  
 561 star. Horizontal flight track shown in purple.

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566 **Figure 5.** Profiles of potential temperature and mixing ratio on (a) July 18 and (b) January 25.

567 Values obtained from the ARM interpolated sounding product. Horizontal black lines indicate the

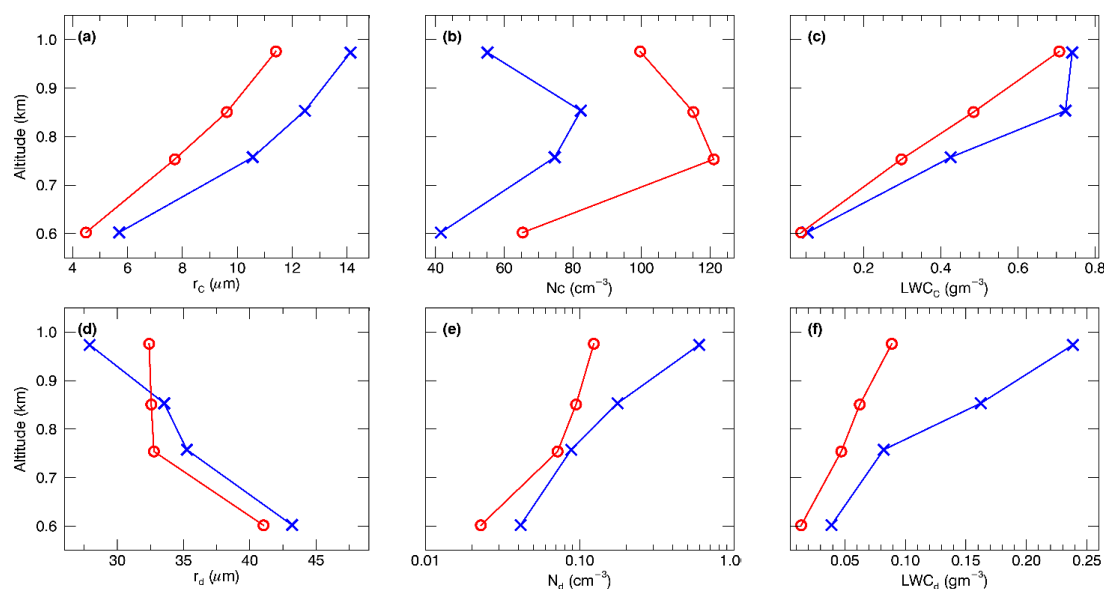
568 approximate altitudes of the cloud top and cloud base.

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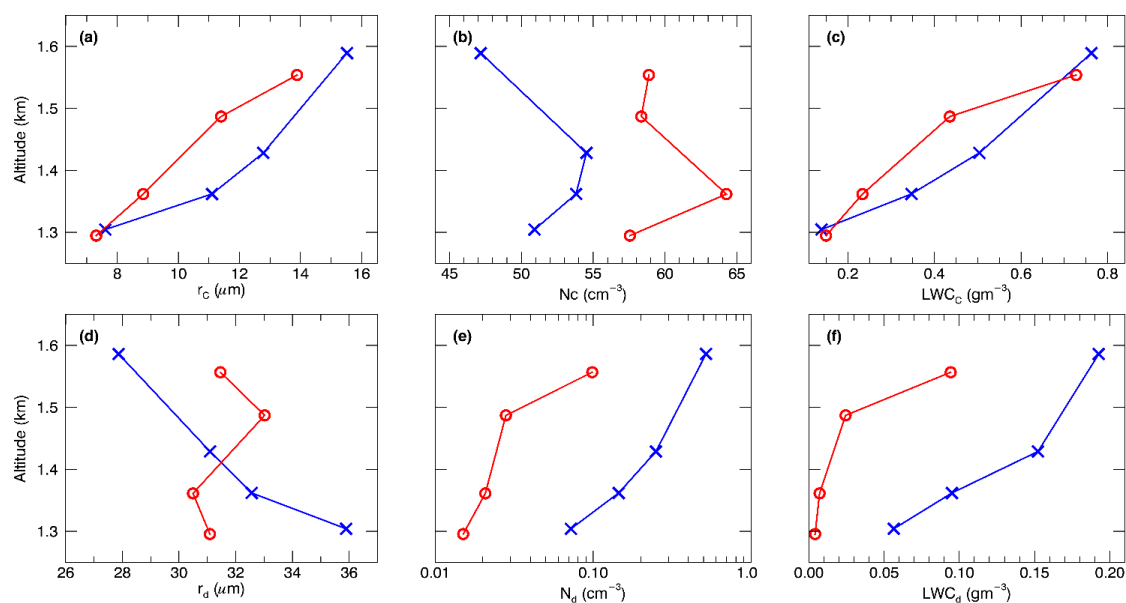
572

573 **Figure 6.** Mean values of various cloud and drizzle microphysical properties computed from the  
 574 1 Hz aircraft observations along the horizontal legs shown in Fig. 1b for July 18: (a) cloud droplet  
 575 radius, (b) cloud droplet number concentration, (c) cloud droplet liquid water content, (d) drizzle  
 576 v radius, (e) drizzle droplet number concentration, and (f) drizzle droplet liquid water content.

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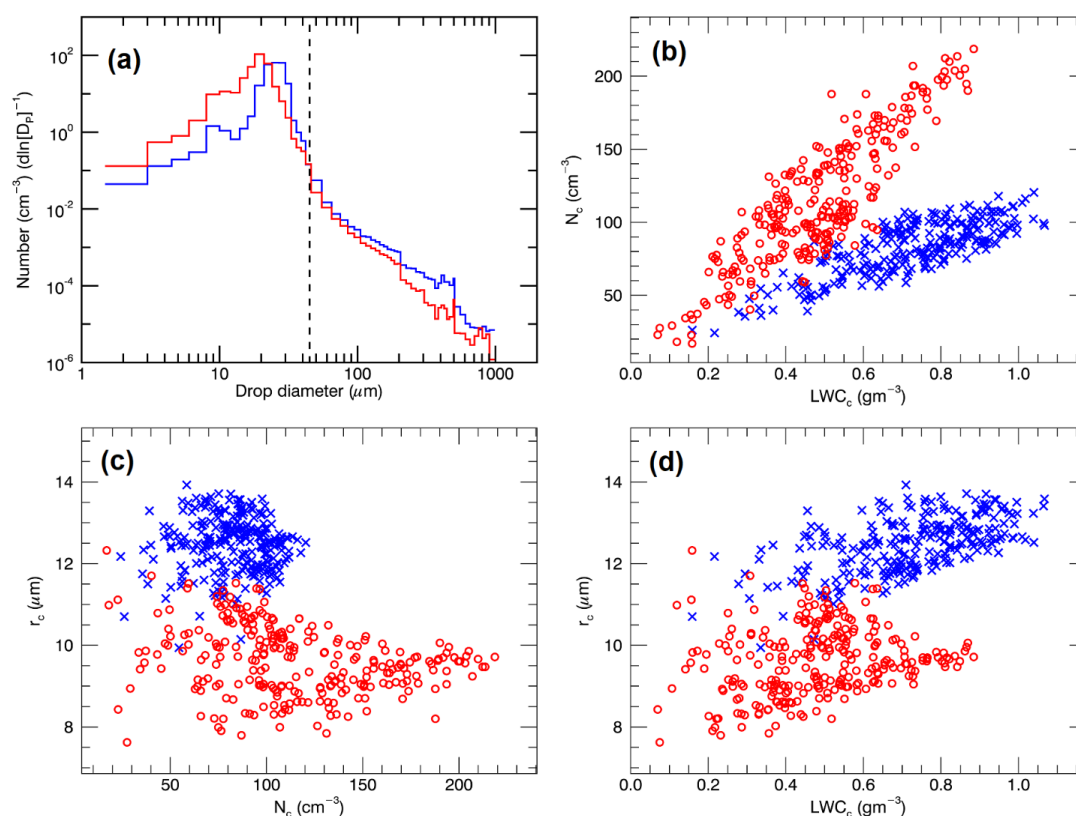
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581 **Figure 7.** Same as Fig. 6, except for the mean values calculated from the 1 Hz aircraft in situ  
 582 measurements along the horizontal legs shown in Fig. 1d on January 25.

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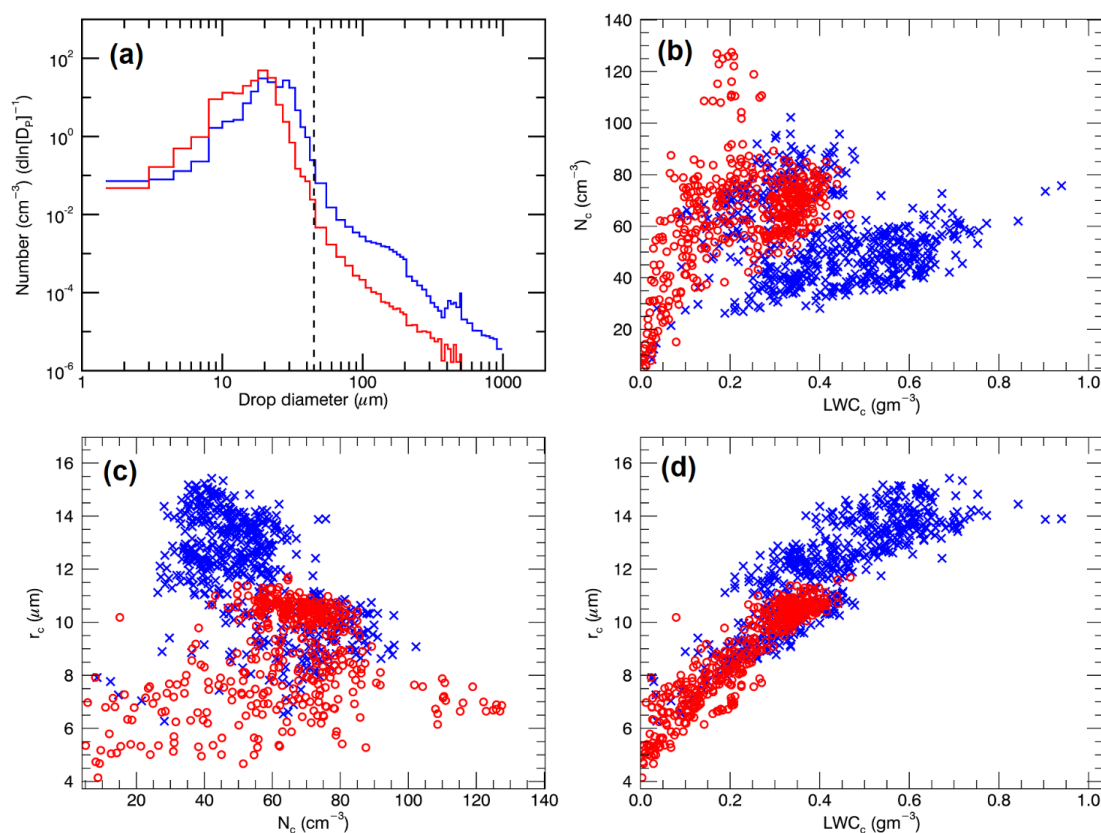
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587 **Figure 8.** (a) Mean droplet size distribution (DSD) computed by averaging all 1 Hz in-situ  
 588 measurements made for the horizontal legs at approximately 850 m altitude in the along-wind  
 589 (blue) and cross-wind (red) directions on July 18. Approximate time range 10.0 to 10.15 UTC  
 590 (refer to Fig. 1b). Vertical dashed line at drop diameter of 45  $\mu\text{m}$  indicates the division between  
 591 cloud-sized and drizzle-sized drops. The remaining panels are scatterplots of (b) cloud-droplet  
 592 effective radius ( $r_c$ ) vs. cloud-droplet number concentration ( $N_c$ ), (c)  $N_c$  vs. cloud liquid water  
 593 content ( $LWC_c$ ) and (d)  $r_c$  vs.  $LWC_c$  derived from the 1 Hz in situ measurements taken along the  
 594 850 m horizontal legs.

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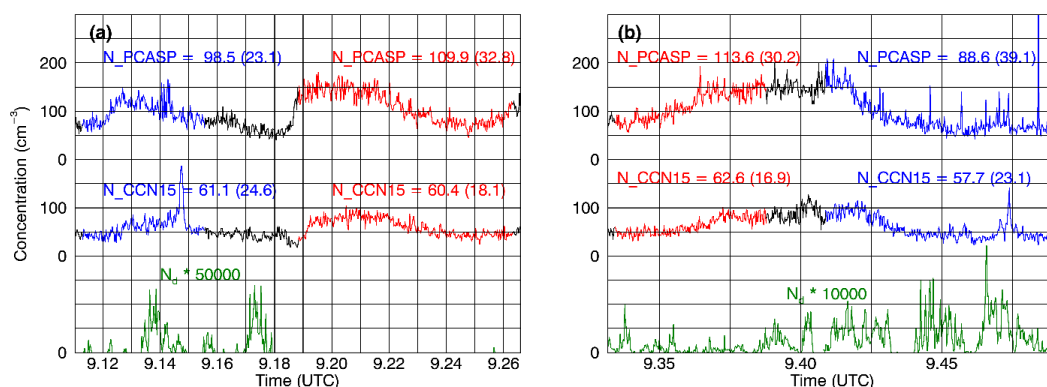
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599 **Figure 9.** Same as Fig. 8, except for the in-situ measurements taken on January 25 when sampling  
 600 the four horizontal legs in the middle altitude. Approximate time range is 11.95 to 12.35 UTC  
 601 (refer to Fig. 1d).

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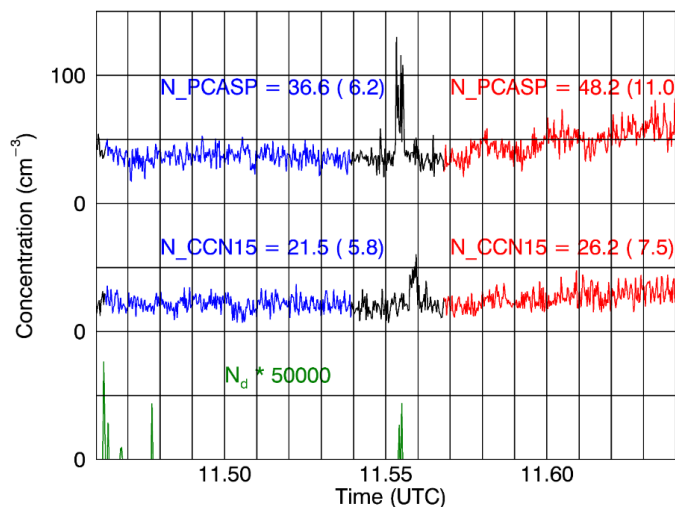
605

606 **Figure 10.** Observations of aerosol number concentration from the PCASP instrument and CCN  
 607 at 0.15% supersaturation from the CCN counter on July 18 for horizontal legs (a) just above the  
 608 ocean surface at 50 m altitude and (b) just below cloud base at 440 m altitude. Blue indicates the  
 609 along-wind leg portion and red the cross-wind leg portion. Numbers show the mean and standard  
 610 deviation for all 1 Hz in-situ measurements along each leg. Lower green lines are the observed  
 611 drizzle drop concentrations scaled by factors of 50000 and 10000, respectively.

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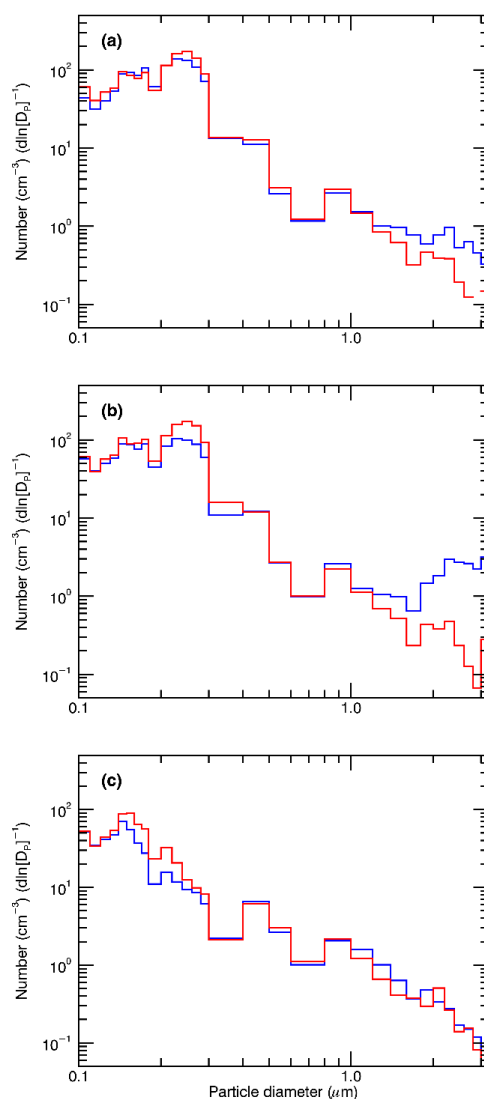
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616 **Figure 11.** Same as Fig. 10, except for the in-situ measurements made at approximately 30 m  
 617 above the ocean surface on January 25.

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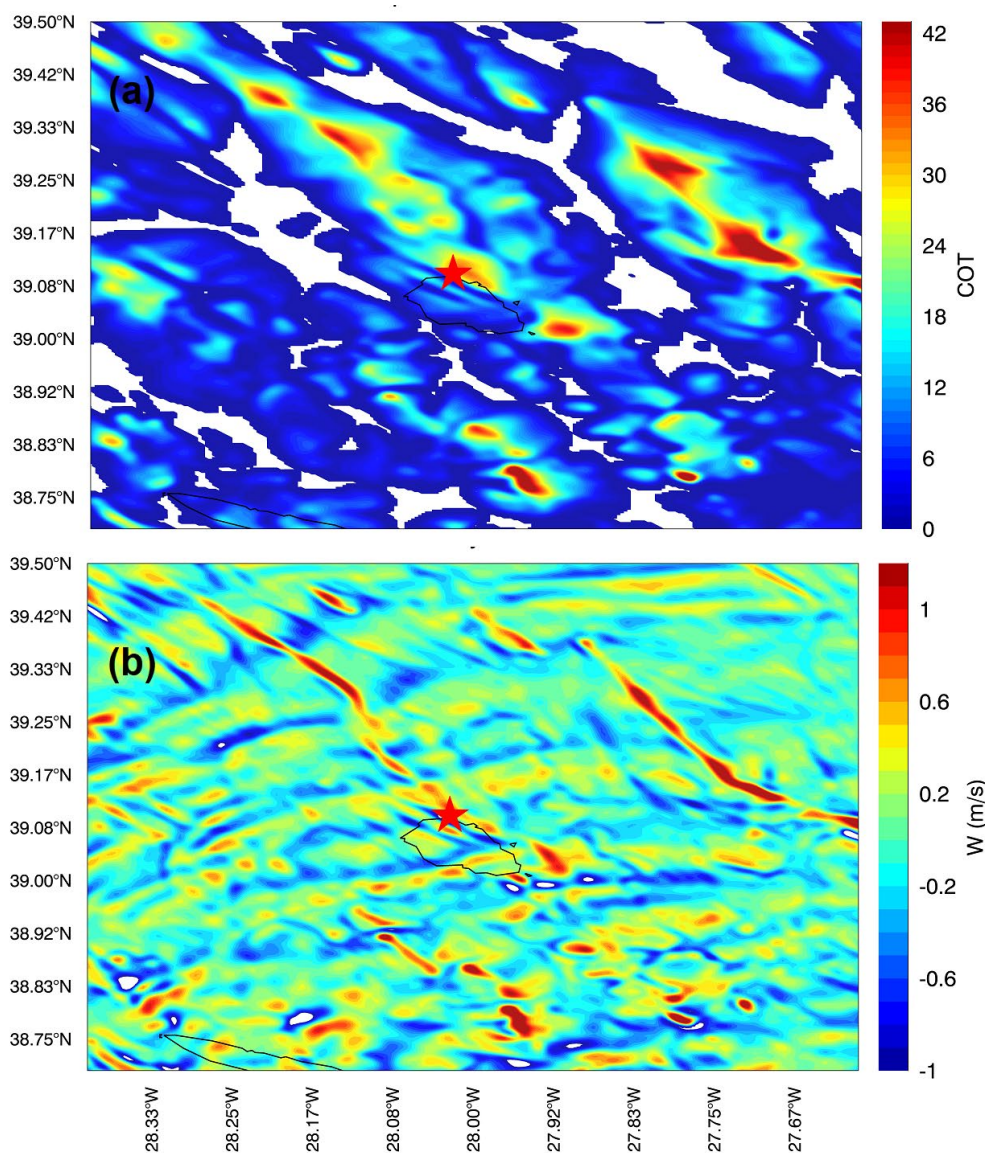
621

622 **Figure 12.** Mean aerosol size distribution computed by averaging all 1 Hz in-situ measurements  
 623 of the size distribution from the PCASP instrument made along horizontal legs (a) on July 18 just  
 624 above the ocean surface, (b) on July 18 just below cloud base, and (c) on January 25 just above  
 625 the ocean surface. Blue indicates the along-wind leg portion and red the cross-wind leg portion.

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630 **Figure 13.** WRF simulation of (a) cloud optical thickness (COT) and (b) vertical velocity ( $w$ ) at

631 848 m at 10:00 UTC over the inner domain of the model run. Red star indicates ARM radar facility.

632 The black box in Fig. 3a outlines the WRF inner domain relative to the satellite imagery.

633