



Clarifying the controlling factors of the seasonal variation of droplet concentrations in marine low-level clouds

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Abstract. Marine boundary layer (MBL) clouds exert a strong cooling influence on Earth's climate, yet their response to aerosol perturbations remains one of the largest uncertainties in climate projections. A key challenge is identifying which aerosol and meteorological factors control cloud droplet number concentration (N_d), especially when boundary-layer structure alters the transport of surface-emitted aerosol to cloud base. Here, we combine aircraft and ground-based measurements from the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) campaign with the aerosol activation parameterization to quantify controlling factors on N_d during different seasons. We found that summer and winter boundary layer decoupling modulates the vertical transport of aerosols within the MBL and thereby influences cloud condensation nuclei number concentration (N_{CCN}) at cloud base. In both seasons, decoupling occurs for roughly 32% of the total time, during which ~60% of accumulation-mode and ~75% of Aitken-mode aerosols fail to reach the cloud layer. The out-of-cloud accumulation-mode particle number concentration (N_{acc}) is higher in summer than in winter, whereas the corresponding N_d is comparable between the two seasons. Sensitivity experiments show that N_{acc} is the dominant control on N_d in summer, whereas cloud droplet formation in winter is additionally sensitive to Aitken-mode aerosol number concentration (N_{ait}) and size. In contrast, seasonal variations in updraft velocity and aerosol hygroscopicity have weaker effects. These results help clarify how boundary-layer coupling and aerosol size distributions jointly regulate marine cloud droplet formation and provide observational constraints for representing aerosol-marine cloud interactions in climate models.



1 Introduction

Marine boundary layer (MBL) clouds exert a substantial net cooling effect on the global climate (Kang et al., 2022; Wu et al., 2017). This cooling effect is fundamentally governed by two key aerosol-cloud interactions (ACIs) (Grosvenor et al., 2014). The Twomey effect describes how an increase in aerosol concentrations increases cloud droplet number concentration (N_d), decreases mean droplet size, and thereby enhances cloud albedo with fixed cloud liquid water path (Twomey, 1977). The Albrecht effect suggests that the suppression of drizzle by these smaller droplets increases cloud lifetime (Albrecht, 1989). Consequently, the estimation of the cloud climate effect by climate models largely relies on the N_d .

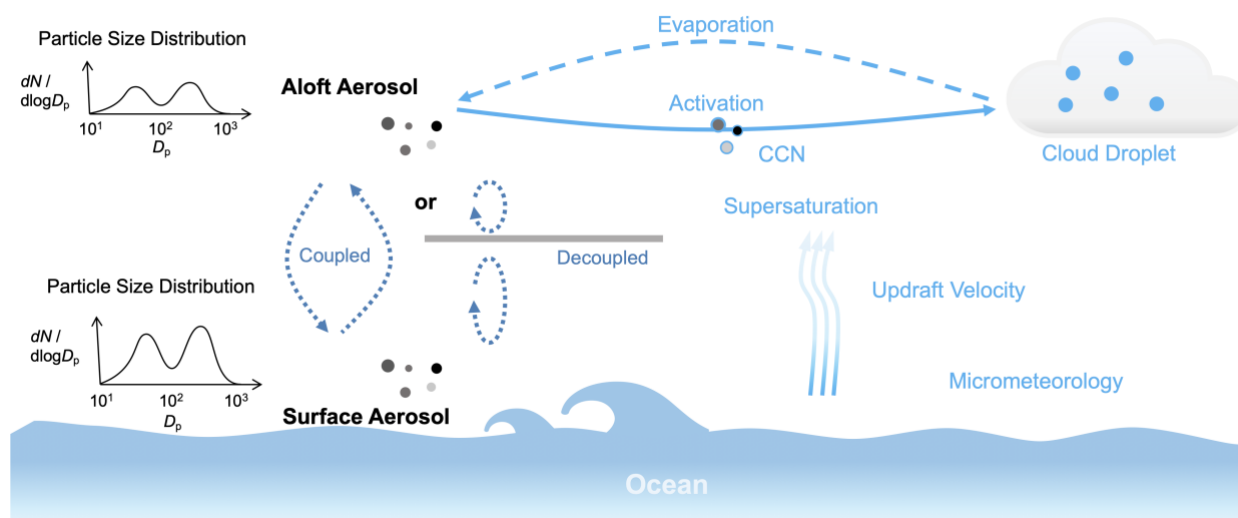
N_d is closely linked to the number concentration of aerosol particles available for activation near cloud. Aerosol particles that activate at a prescribed supersaturation S are called cloud condensation nuclei (CCN), and their number concentration is denoted N_{CCN} . Ideally, N_{CCN} would be measured in cloud-free air immediately below or adjacent to cloud base, where it most directly characterizes the aerosol population available for activation and minimizes the effects of MBL vertical distribution (Jia et al., 2026). Because of the difficulty in measuring out-of-cloud CCN at the global scale (Virtanen et al., 2025), studies commonly use aerosol proxies, such as aerosol optical depth (AOD) (Rosenfeld et al., 2023), surface-measured accumulation-mode aerosol number concentrations (N_{acc}) (Braga et al., 2022), and the number concentration of aerosol particles larger than the Hoppel minimum diameter (HM, typically ~40–150 nm) (Gong et al., 2023). The HM is a local minimum in the particle number size distribution (PNSD) between the Aitken (~30–80 nm) and accumulation modes (~80–1000 nm), which is often used as an empirical proxy for an effective activation diameter (Hoppel et al., 1990). However, the extent to which surface-based aerosol proxies represent cloud-base aerosol populations depends strongly on the MBL coupling state (Su et al., 2022). In a coupled MBL, vigorous mixing ensures ground aerosols represent those at the cloud base. Conversely, decoupled conditions create a vertical disconnect, where a sub-cloud stable layer restricts the upward transport of aerosols, causing surface and cloud-base aerosol populations to diverge. Thus, MBL coupling determines whether surface measurements characterize the aerosol reservoir at cloud base.

For aloft aerosol particles transported to cloud base, the fraction that activates is jointly controlled by meteorological conditions and aerosol properties (e.g., aerosol hygroscopicity (κ), particle size distribution (PNSD) of aerosols) (Braga et al., 2021; Efraim et al., 2024; Reutter et al., 2009). Among the meteorological factors, cloud-base updraft velocity (w) is a primary control on the maximum supersaturation (S_{max}), which further determines the minimum particle size that can be activated (Reutter et al., 2009; Gong et al., 2023). The sensitivity of N_d to N_{CCN} differs between CCN-limited and updraft-limited conditions. The former refers to the state where CCN are insufficient, in which N_d increases with the available aerosol number concentration. In contrast, the latter refers to the regime in which CCN supply is abundant, while the supersaturation generated by the updraft is insufficient to activate all available particles. N_d becomes more sensitive to w than to aerosol loading (Reutter et al., 2009; Noble and Hudson, 2014).

In addition to meteorological conditions, N_d is intricately sensitive to PNSD and κ of aerosols, and the relative importance of these properties remains debated. Recent evidence suggests that the effective aerosol population, i.e., aerosol particles that



exceed a critical activation diameter, is more decisive than the total number concentration (Hernández Pardo et al., 2019).
 Furthermore, in clean marine environments with low N_{acc} , Aitken-mode aerosols can serve as a vital CCN reservoir,
 particularly under stronger updrafts ($w \geq 0.6 \text{ m s}^{-1}$) that enable higher supersaturations and low N_{acc} (Pöhlker et al., 2021).
 Several studies indicate that κ significantly influences N_d under polluted conditions or at very low w , where supersaturation
 is limited (Reutter et al., 2009). Conversely, other observations suggest that variability in κ has a negligible effect on N_d
 compared to PNSD (Hernández Pardo et al., 2019; Karydis et al., 2012). Despite these advances, it remains unclear when
 surface-based aerosol proxies, particularly N_{acc} , adequately represent the cloud-base aerosol population. It is also unresolved
 how cloud-base w and aerosol PNSD jointly regulate the contribution of N_d .
 Both coupling state and aerosol population exhibit pronounced seasonal variability (Rémillard et al., 2012; Zheng et al.,
 2018). Whether the relative controls of meteorological parameter (i.e., w) and aerosol physicochemical properties on N_d
 persist across contrasting meteorological and aerosol regimes is unclear. To quantify the competing role of meteorological
 and aerosol drivers for N_d , we conducted comprehensive ground-based and aircraft observations during the Aerosol and
 Cloud Experiments in the Eastern North Atlantic (ACE-ENA) field campaign across two seasons. The Eastern North
 Atlantic (ENA) site serves as a natural laboratory with profound seasonal contrasts in synoptic weather patterns and aerosol
 loading (Wang et al., 2022; Wood et al., 2015). The mechanistic complexities associated with ACI within MBL are
 conceptually summarized in Fig. 1. Building on this conceptual framework, this study specifically aims to: (1) quantify how
 the atmospheric coupling and decoupling states modulate the vertical transport of aerosols, and (2) evaluate the relative
 impacts of meteorological conditions, specifically w , and aerosol physicochemical properties (e.g., κ , PNSD, aerosol number
 concentration) on N_d across summer and winter seasons. These insights are critical for refining the representation of marine-
 aerosol-cloud interactions in global climate models.





85 **Figure 1.** Schematic illustrating the mechanistic complexities in quantifying aerosol-cloud interactions (ACIs) within the MBL in this study. The diagram highlights the intricate, competing factors governing aerosol activation and cloud droplet formation.

2 Methods and model simulation

2.1 Airborne measurements

90 During the ACE-ENA campaign, the Gulfstream-1 (G-1) research aircraft conducted 20 flights during the summer intensive observation period (IOP 1: 21 June–21 July 2017) and 19 flights during the winter IOP (IOP 2: 19 January–18 February 2018). These aircraft observations were available only during the daytime. The aircraft was based at Lajes Field on Terceira Island in the Azores. Flights typically featured horizontal legs at multiple levels, including near the ocean surface (~40 m above mean sea level, AMSL), below cloud base, outside clouds, within clouds, and at cloud top. Flight tracks, as shown in
 95 Fig. S1, were confined within a 50-km radius of the ENA observatory and spanned altitudes from ~40 to ~3000 m AMSL. We analyzed data from 16 research flights (6 in summer and 10 in winter) that contained at least one horizontal leg with both out-of-cloud aerosol and cloud droplet size distributions. Other flights were excluded because they lacked suitable cloud sampling or did not have stable out-of-cloud FIMS measurements. We also confirmed that the out-of-cloud aerosol size distributions along the horizontal leg remained relatively consistent so that the sampling was conducted within the same air
 100 parcel (Table S1).

The G-1 was equipped with a suite of instruments for characterizing meteorological parameters, aerosol particles, and cloud droplets (Wang et al., 2022). Particle number size distributions aloft ($PNSD_A$, $10 < D_p < 600$ nm) were measured at 1 Hz using a Fast Integrated Mobility Spectrometer (FIMS, Wang et al., 2018). We also calculated the aloft accumulation-mode (N_{acc-A}) and the aloft Aitken-mode particle number concentrations (N_{ait-A}) from $PNSD_A$ by using a two-mode log-normal
 105 fitting method (Gong et al., 2020). Total particle number concentrations on the flight (N_{10nm-A} ; $D_p > 10$ nm) were measured by a condensation particle counter (CPC; model 3772, TSI Inc., Shoreview, Minnesota). Cloud droplet size distributions (2–50 μ m) were measured at 1 Hz using a Fast Cloud Droplet Probe (FCDP, SPEC Inc., Boulder, Colorado), and cloud liquid water content (LWC) and measured N_d were derived by integrating the cloud droplet size distributions.

In this study, flight segments are used only when the out-of-cloud N_{10nm-A} was relatively stable, with the coefficient of
 110 variation within 20%. We also exclude flight segments influenced by new particle formation (NPF) or by pollution transported from the upper troposphere by removing cases where aerosol number concentration increases with altitude.

2.2 Surface measurements

Ground-based aerosol and meteorological measurements were collected at the ENA observatory on Graciosa Island, Azores (39°05'N, 28°01'W; 30.5 m AMSL). The dataset analyzed in this study encompasses the summer (June–August 2017) and
 115 winter (December 2017–February 2018) seasons and is restricted to daytime hours (06:00–21:00 local time) to match the



timing of aircraft observations. Surface meteorological conditions at the ENA site (Fig. S2) exhibit distinct seasonal patterns during the two IOPs. For example, summer temperatures are approximately 6 °C higher than winter (Fig. S2a). In contrast, ground relative humidity (RH) remains comparable across both seasons (Fig. S2b). Sea level pressure shows a slight difference, with median values of approximately 1021 hPa in summer and 1026 hPa in winter (Fig. S2c). Wind directions during summer and winter are comparable, mainly from the southwest and northeast (Fig. S2d). Winter is characterized by higher wind speeds, with distinctively more frequent occurrences exceeding 12 m s⁻¹.

Ground particle number size distributions (PNSD_G, 10 < D_p < 470 nm) were measured using a Scanning Mobility Particle Sizer (SMPS; model 3938, TSI Inc., Shoreview, Minnesota). The seasonal average PNSDs are shown in Fig. A1. Aerosol concentrations are higher in summer than in winter. The ground Aitken-mode (*N*_{ait-G}) and accumulation-mode aerosol number concentrations (*N*_{acc-G}) are calculated from PNSD_G by using the two-mode log-normal fitting method. *κ* was derived from the activation spectra of size-selected aerosols using a size-resolved cloud condensation nuclei (SCCN) system. The system couples a differential mobility analyzer (DMA 3081, TSI) with a condensation particle counter (CPC 3010, TSI) and a cloud condensation nuclei counter (CCNC). Aerosols were size-selected at 40, 50, 75, 100, and 150 nm. For each diameter, the CCNC supersaturation was scanned by varying the temperature gradient (Δ*T* = 4, 6.5, and 10 K) and/or the sampling flow rate (0.3 to 1.0 L min⁻¹). To represent the distinct hygroscopic properties of different aerosol populations, the Aitken-mode *κ* was defined as the median value across three selected diameters (40, 50, and 75 nm), while the accumulation-mode was represented by the *κ* measured at 150 nm (Gong et al., 2023; Reutter et al., 2009).

Updraft velocity at the cloud base height (CBH) is retrieved from the Doppler Lidar (Gong et al., 2023; Ghate et al., 2021), following the methodology of Borque et al. (2016). The lidar retrievals have a temporal resolution of 5 s and a vertical resolution of 30 m. During aircraft sampling periods, an additional CBH check was performed using LWC. Cloud bases are defined as the lowest altitude where LWC > 0.03 g m⁻³ persists for at least 10 s (10 consecutive data points). Thermodynamic profiles, specifically water vapor mixing ratio (*q_t*, g H₂O / kg wet air) and potential temperature (*θ*), were derived from balloon-borne radiosonde soundings (Holdridge, 2020). Table S2 lists the measurement parameters and instrumentation details.

To estimate the degree of coupling states below cloud base, we used layer-mean differences in *q_t* and *θ* between the near-ground level (represented at ~100 m, Zheng et al., 2021) and the layer immediately below cloud base (~50 m below cloud base height). The near-ground level was used to minimize direct surface conditions influences, while the below cloud base height was used to reduce potential retrieval uncertainty (Noble and Hudson, 2014; Jones et al., 2011). Specifically, the thermodynamic differences of *q_t* and *θ* were calculated as follows:

$$\Delta q_t = q_{t, \text{near-ground level}} - q_{t, \text{below cloud base}}, \quad (1)$$

$$\Delta \theta = \theta_{\text{below cloud base}} - \theta_{\text{near-ground level}}, \quad (2)$$

The Δ*q_t* and Δ*θ* values used here are leg-mean values from the two specified sampling levels. Consistent with previous studies of MBL coupling and decoupling (Zeider et al., 2025; Dong et al., 2015), our analysis was also restricted to cloudy



conditions. Clear-air periods without cloud occurrence were outside the scope of this work. We applied the criteria for the
 150 different coupling states proposed by Zeider et al. (2025). The vertical profiles are classified into four regimes based on the
 two-layer differences Δq_t and $\Delta\theta$ (Table 1).

Table 1. Criteria used to define the thermodynamic coupling states in the MBL.

Degree	Criteria
Strong coupled	$\Delta\theta \leq 1.0 \text{ K}, \Delta q_t \leq 0.8 \text{ g}\cdot\text{kg}^{-1}$
Moderate, coupled with high $\Delta\theta$	$\Delta\theta > 1.0 \text{ K}, \Delta q_t \leq 0.8 \text{ g}\cdot\text{kg}^{-1}$
Moderate, coupled with high Δq_t	$\Delta\theta \leq 1.0 \text{ K}, \Delta q_t > 0.8 \text{ g}\cdot\text{kg}^{-1}$
Decoupled	$\Delta\theta > 1.0 \text{ K}, \Delta q_t > 0.8 \text{ g}\cdot\text{kg}^{-1}$

2.2 Estimation of N_d using activation parameterization

N_d was also simulated using the physically based activation parameterization developed by Abdul-Razzak and Ghan (2000),
 155 hereinafter ARG. The ARG scheme provides an analytical expression in an ascending air parcel, based on the interplay
 between adiabatic cooling and water vapor depletion (particle growth).

The parameterization assumes a multi-modal lognormal PNSD. In this study, we considered two modes ($i = 1, 2$): the Aitken
 and accumulation modes. For each mode, the inputs include the aerosol number concentration (N), geometric mean diameter
 (μ), geometric standard deviation (σ), and the hygroscopicity parameter (κ).

160 First, the maximum supersaturation ($S_{\max}$) is determined by Eq. (3):

$$S_{\max} = \left\{ \sum_{i=1}^I \frac{1}{S_{mi}^2} \left[f_i \left(\frac{\zeta}{\eta_i} \right)^{\frac{3}{2}} + g_i \left(\frac{S_{mi}^2}{\eta_i + 3\zeta} \right)^{\frac{3}{4}} \right] \right\}^{-\frac{1}{2}}, \quad (3)$$

where $I=2$ is the total number of aerosol modes considered in this study. S_{mi} is the critical supersaturation for a particle with a
 diameter equivalent to the μ of mode i . The terms η_i and ζ are dimensionless parameters, which are both dependent on the w ,
 surface tension, growth coefficient that relates to the diffusivity of water vapor in air and air thermal conductivity
 165 parameters, and temperature-dependent thermodynamic parameters that link updraft-driven cooling to the condensational
 sink of water vapor. And η_i is also related to aerosol number concentration in mode i . The geometric coefficients f_i and g_i ,
 which depend on the σ of mode i , are defined as Eqs. (4) and (5):

$$f_i = 0.5 \exp(2.5 \ln^2 \sigma_i), \quad (4)$$

$$g_i = 1 + 0.25 \ln \sigma_i, \quad (5)$$

170 Once $S_{\max}$ is determined, the total activated N_d is obtained by integrating the PNSD for all particles with a critical
 supersaturation less than $S_{\max}$. This is expressed using the complementary error function as Eq. (6):



$$N_d = \sum_{i=1}^I N_i \frac{1}{2} [1 - \operatorname{erf}(u_i)], \quad (6)$$

where N_i is the number concentration of aerosol mode i . $I=2$ is the total number of aerosol modes considered in this study. u_i is a dimensionless parameter relating the critical supersaturation of the mode to the $S_{\max}$ as Eq. (7):

$$u_i = \frac{2 \ln\left(\frac{S_{mi}}{S_{\max}}\right)}{3\sqrt{2} \ln \sigma_i}, \quad (7)$$

This analytical approach allows for the efficient calculation of calculated N_d under varying w and aerosol conditions without the need for explicit time-dependent numerical integration.

3 Results and Discussion

3.1 Difference between measured N_d and $N_{\text{acc-A}}$ in different seasons

The relationship between measured N_d in low-level clouds ($< \sim 2200$ m in this study) and the out-of-clouds $N_{\text{acc-A}}$ measured in adjacent cloud-free air exhibits distinct seasonal contrasts (Fig. 2). Each point represents the paired median values of measured N_d and $N_{\text{acc-A}}$ within a given flight altitude bin. The median values of $N_{\text{acc-A}}$ span ~ 21 – 361 cm^{-3} in summer and ~ 10 – 131 cm^{-3} in winter. The seasonal median $N_{\text{acc-A}}$ values were 220.87 cm^{-3} in summer and 33.48 cm^{-3} in winter, with corresponding mean $N_{\text{acc-A}}$ of 219.54 ± 95.79 cm^{-3} in summer and 48.74 ± 34.33 cm^{-3} in winter. The summer-to-winter median ratio of $N_{\text{acc-A}}$ was 6.60. In comparison, the seasonal median of measured N_d values were 135.78 cm^{-3} in summer and 87.09 cm^{-3} in winter, with corresponding mean measured N_d of 133.35 ± 57.73 cm^{-3} in summer and 88.74 ± 42.82 cm^{-3} in winter. Measured N_d corresponds to a smaller summer-to-winter median ratio of 1.56. Although both summertime measured N_d and $N_{\text{acc-A}}$ were higher on average, the seasonal contrast in measured N_d was much weaker than that in $N_{\text{acc-A}}$, consistent with the results from Wang et al. (2022). A zero-intercept linear fit $N_d = aN_{\text{acc-A}}$ was used to quantify the difference between measured N_d and $N_{\text{acc-A}}$. The fitted slope a was 0.57 in summer, which was lower than a in winter ($a = 1.46$). The wintertime slope greater than unity suggests that accumulation-mode aerosols alone are insufficient to account for the observed droplet population, implying a possible contribution from Aitken-mode particles to measured N_d in winter. In contrast, the lower summertime slope suggests that accumulation-mode aerosols are sufficient to activate into cloud droplets during summer.

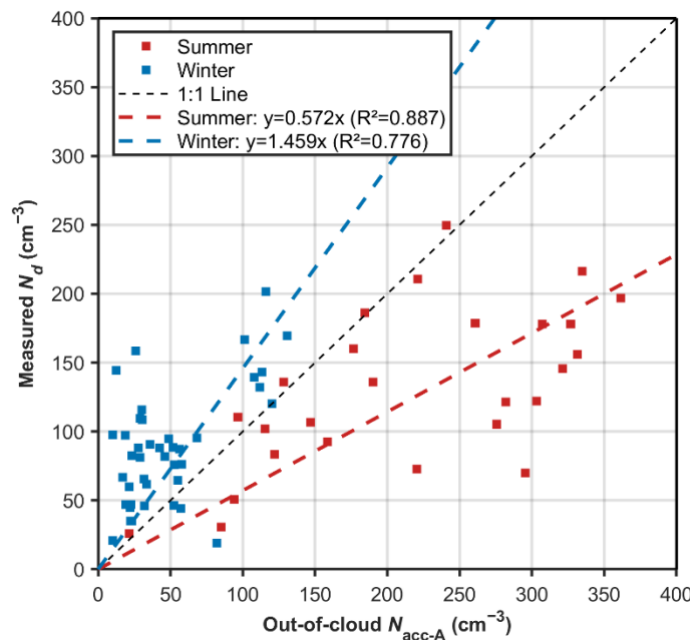


Figure 2. The measured N_d versus N_{acc-A} in adjacent cloud-free air. Red and blue squares represent data from the summer (Jun–Aug 2017) and winter (Dec 2017–Feb 2018) periods, respectively. The red and blue dashed lines indicate the corresponding zero-intercept linear fits. The black dashed line indicates the 1:1 relationship.

Potential mechanisms driving the seasonal cycle of N_d and the enhanced winter activation efficiency include: (1) differences in the degree of MBL coupling between winter and summer regulate aerosol vertical gradients. Specifically, decoupled states restrict the upward transport of surface aerosols, resulting in lower aerosol concentrations aloft. (2) seasonal differences in cloud-base updraft velocity may affect cloud activation. (3) seasonal differences in aerosol physicochemical properties, such as Aitken and accumulation mode geometric mean diameters (μ_{ait} and μ_{acc}), concentrations (N_{ait} and N_{acc}), aerosol geometric standard deviation (σ_{ait} and σ_{acc}), and hygroscopicity in both Aitken and accumulation modes (κ_{ait} and κ_{acc}). In the following section, we will quantify the controlling factors of the seasonal variation of marine low-level N_d .

3.2 Mixing states of MBL affect aerosol vertical transport

3.2.1 Cases study

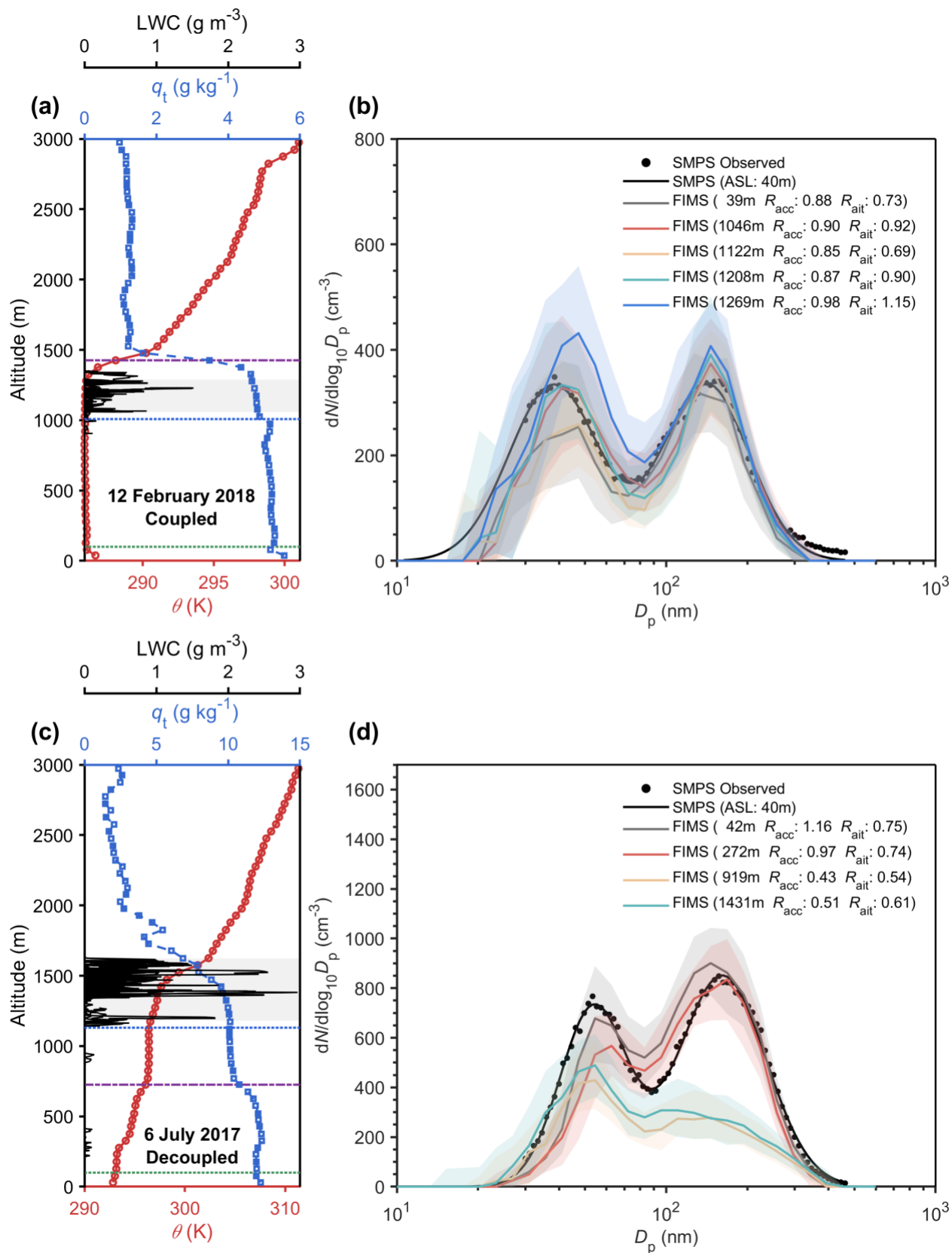
Figures 3a–3d illustrate representative cases of vertical profiles of thermodynamic properties (q_t and θ) and PNSD during a coupled (12 February 2018) and a decoupled MBL (6 July 2017), respectively. Figures 3a and 3c show the profiles of q_t and θ , while Figures 3b and 3d show the corresponding PNSD profiles for the coupled and decoupled cases, respectively. The grey shading denotes the cloudy layer between the cloud base and cloud top, while the purple dash-dotted line marks the maximum height directly influenced by the surface fluxes. The green and blue dotted lines mark the near-ground level and



cloud base height, respectively. The vertical differences in q_t and θ between these two levels were used to assess the coupling states.

215 This coupled case exhibits strong agreement between airborne FIMS-measured PNSD_A and ground-based SMPS-measured PNSD_G at approximately 40 m altitude (Fig. 3b). PNSD from ground to higher altitudes shows distinct Aitken and accumulation modes. The airborne-to-ground concentration ratios for the accumulation mode ($R_{\text{acc}} = N_{\text{acc-A}} / N_{\text{acc-G}}$) and the Aitken mode ($R_{\text{ait}} = N_{\text{ait-A}} / N_{\text{ait-G}}$) are about 0.85–0.98 and 0.69–1.15, respectively. At the top-flight level (~1269 m), the Aitken and accumulation-mode concentrations remain comparable to the ground-level concentrations ($R_{\text{ait}} = 1.15$ and $R_{\text{acc}} =$
 220 0.98). These good agreements between ground-level and flight measurements again indicate a well-mixed state of aerosol particles along MBL.

In the decoupled case (Fig. 3c and d), both Δq_t and $\Delta \theta$ exceeded the thresholds for the coupled regimes from the near-ground level to the cloud base height. The vertical thermodynamic structure shows a well-mixed surface layer (~725 m) separated from the upper MBL below cloud base height. Figure 3c shows q_t and θ are relatively homogeneous ($\Delta \theta \leq 1.0$ K, $\Delta q_t \leq 0.8$
 225 $\text{g} \cdot \text{kg}^{-1}$) below approximately 725 m (shown as the purple dash-dotted line). The primary cloud base height was located near 1181 m, with intermittent cloud fragments below. Airborne PNSD_A measurements at 42, 272, 919, and 1431 m AMSL show that the two lowest levels (42 and 272 m) were within the surface mixed layer. As shown in Fig. 3d, the PNSD_A at 42 m closely matches the ground-based PNSD_G , with R_{acc} and R_{ait} values of 1.16 and 0.75, respectively. At the two sampling levels above the mixed-layer top (919 and 1431 m), N_{acc} decreases sharply, with R_{acc} dropping to 0.43–0.51, while the R_{ait} shows a
 230 relatively moderate decrease to 0.54–0.61. This decrease suggests that decoupling depletes the transportation of surface aerosol to the upper MBL. The stronger depletion of accumulation-mode aerosols than of Aitken-mode aerosols indicates other aerosol-removal processes, such as cloud processing or scavenging of aerosol particles within fragmented cloud layers below the primary cloud base.





235 **Figure 3.** Comparison of aerosol and meteorological properties for a coupled case on 12 February 2018 (a, b) and a
 decoupled case on 6 July 2017 (c, d). (a, c) Vertical profiles of potential temperature (θ ; red circle) and water vapor mixing
 ratio (q ; blue square) from balloon-borne radiosondes, alongside aircraft-measured liquid water content (LWC; black). Gray
 shaded area marks the cloudy layer between the cloud-top and cloud-base levels. The purple dash-dotted line denotes the
 decoupling interface within the marine boundary layer (MBL), and the green and blue dotted lines mark the near-ground
 240 level and below the cloud base, respectively. (b, d) Particle number size distributions (PNSDs) over the diameter range 10-
 1000 nm measured at different altitudes within the MBL. Black dots represent ground-based SMPS measurements (PNSD_G),
 while the black solid lines represent the fitted PNSD_G. Colored lines represent airborne FIMS measurements at different
 altitudes (PNSD_A). The ratios of aloft-to-ground concentrations for the Aitken mode ($R_{ait} = N_{ait-A} / N_{ait-G}$) and the
 accumulation mode ($R_{acc} = N_{acc-A} / N_{acc-G}$) are indicated in the legend.

245 3.2.2 Statistical analysis of coupled states in two seasons

To verify the impact of coupled states on aerosol vertical transport, we conducted statistical analysis of R_{ait} (N_{ait-A} / N_{ait-G}) and
 R_{acc} (N_{acc-A} / N_{acc-G}) for 36 and 56 flight cases during the summer (IOP1) and the winter campaigns (IOP2), respectively. As
 shown in Fig. 4a, the majority of the data was collected under coupled conditions (22/36 cases in summer and 42/56 cases in
 winter), with 14 decoupled cases observed during the summer period and 14 in winter. The low frequency of decoupled
 250 conditions in the aircraft dataset is likely attributed to the timing of flight operations, which were concentrated around local
 noon, when the MBL is typically well mixed.

For coupled cases, the R_{acc} (the dark yellow boxplots) remains highly consistent (median ~0.84–0.94) from the ground up to
 1000 m, whereas the R_{ait} (the dark purple boxplots) exhibits a pronounced decrease with height, with a median of
 approximately 0.37–0.54 from the ground up to 1000 m. The decreasing trends of Aitken-mode aerosol concentrations were
 255 also observed over the Southern Great Plains by Wang et al. (2023), and they attributed this reduction to interstitial
 coagulation with cloud droplets. Moreover, the significant discrepancy between ground-level and aloft Aitken-mode
 concentrations may stem from enhanced ground-level Aitken aerosol production during summer at the ENA site (Gallo et
 al., 2023). The oxidation products from marine dimethyl sulfide (DMS) and secondary organic aerosol (SOA) formed via the
 photo-oxidation of oxygenated volatile organic compounds (OVOCs) likely drive the growth of ground Aitken-mode
 260 aerosols (Mungall et al., 2017; Zheng et al., 2018). The strong decrease of accumulation-mode and Aitken-mode aerosols
 may be attributed to the scavenging of aerosol particles within fragmented cloud layers below the primary cloud base.

For decoupled cases, R_{acc} (the light yellow boxplots) and R_{ait} (the light purple boxplots) show a sharp decrease from the
 ground up to 1000 m, with median values of $R_{acc} = 0.39$ and $R_{ait} = 0.24$, corresponding to reductions of 60.5% and 75.1%,
 respectively. This decrease confirms that thermodynamic decoupling acts as a physical barrier that restricts the upward
 265 transport of near-surface aerosols. A modest increase is observed at higher altitudes, approximately 1000–1500 m, where R_{ait}
 and R_{acc} increase to 0.66 and 0.57, respectively. Since cases with evident pollution and free-tropospheric entrainment have
 been removed, these increasing states likely reflect residual aerosol from previous MBL mixing or weak intermittent mixing

near the upper boundary layer rather than direct surface-to-cloud-base transport. Nevertheless, the reductions in aerosol concentration at high altitude compared to ground level reinforce the need to consider MBL coupling when using surface
 270 aerosol measurements as proxies for cloud-base CCN.

Figure 4b presents the statistical proportion of MBL coupling states during summer and winter derived from $\Delta\theta$ and Δq_t measured by high-resolution balloon-borne soundings across 368 cases. Coupled regimes dominate in both summer and winter, whereas decoupled cases account for approximately one-third of cases, 32.6% in summer and 32.0% in winter. The two moderately coupled categories together account for 22–25% across all cases. Following Zeider et al. (2025), the
 275 moderately coupled category with high $\Delta\theta$ is more influenced by processes above the MBL, whereas the moderate category with high Δq_t reflects stronger surface influences. The much larger occurrence of moderately coupled with high $\Delta\theta$ cases in summer than in winter (9.1% versus 1.7%) suggests a stronger influence of above-MBL entrainment during summer.

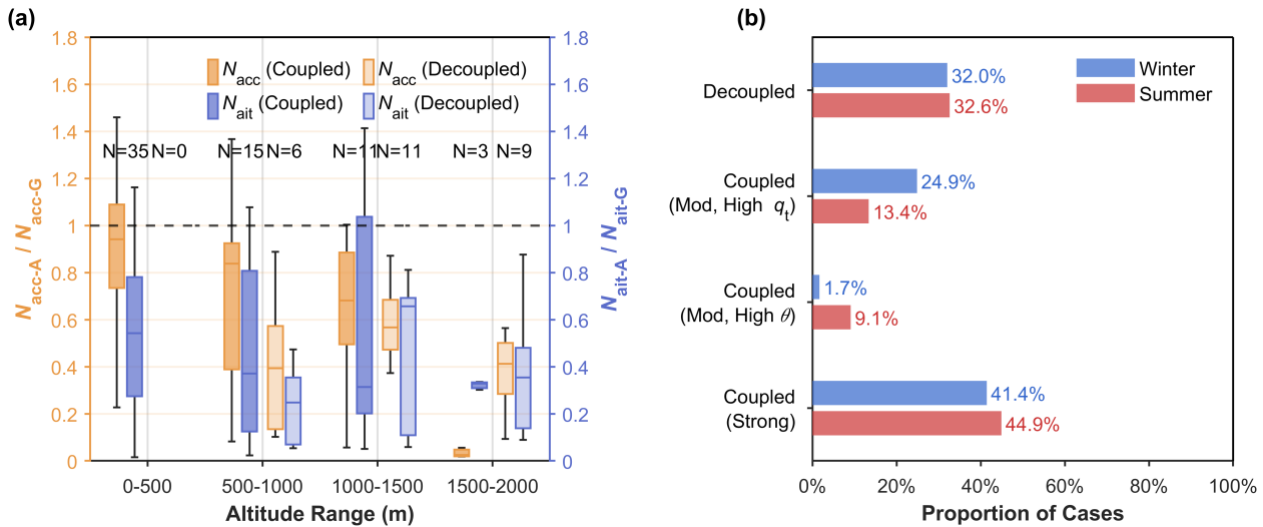


Figure 4. Statistical analysis of aerosol transport and MBL coupling states. (a) Vertical profiles of the aloft-to-ground
 280 aerosol concentration ratios for the accumulation mode (N_{acc-A} / N_{acc-G} , blue) and Aitken mode (N_{ait-A} / N_{ait-G} , red), derived from aircraft and ENA site measurements. (b) Seasonal proportion of coupling states (summer vs. winter) derived from balloon-borne radiosonde soundings.

3.3 Quantify the impacts of updraft velocity and aerosol properties on N_d

3.3.1 Difference between the ARG-simulated and the measured N_d

285 We analyzed the probability density functions (PDFs) of key meteorological and aerosol parameters governing ACI in summer and winter (Fig. A2). These include the meteorological parameter (updraft, w) and aerosol physicochemical properties, specifically the size (N_{ait} , N_{acc} , μ_{ait} , μ_{acc} , σ_{ait} , and σ_{acc} of the PNSD) and hygroscopicity (κ_{ait} and κ_{acc}) of aerosol. The PDF analysis reveals a seasonal contrast in both updraft conditions and aerosol properties. Regarding w , summer is



associated with relatively weak updrafts, with a median updraft velocity of 0.16 m s^{-1} , whereas winter exhibits comparatively stronger updraft conditions (median $w = 0.24 \text{ m s}^{-1}$).

In addition to updraft differences, the PDF analysis shows seasonal shifts in the physical characteristics of aloft aerosols. Summer is characterized by higher aerosol loading, with N_{ait} (204.81 cm^{-3} , median value) and N_{acc} (240.92 cm^{-3} , median value) more than four- and sevenfold higher than their winter median values (49.54 and 33.48 cm^{-3}), respectively. The Aitken-mode diameters of aerosol in summer (median of $\mu_{\text{ait}} = 57.18 \text{ nm}$) are significantly larger than those in winter (median of $\mu_{\text{ait}} = 37.08 \text{ nm}$), but the accumulation-mode diameters in both seasons are comparable. The σ_{ait} and σ_{acc} remain stable with inter-seasonal differences of $\sigma \leq 0.05$. These results indicate that the seasonal variation of aerosols is featured by changes in aerosol abundance and particle mode size, while the aerosol geometric standard deviation remains relatively constant. Meanwhile, κ values of ground-level aerosol are comparable between summer and winter, with differences of κ_{ait} and κ_{acc} being 0.01 and 0.07 , respectively, suggesting similar aerosol chemical compositions between summer and winter.

We applied the ARG scheme using w , κ_{ait} , κ_{acc} , N_{ait} , N_{acc} , μ_{ait} , μ_{acc} , σ_{ait} , and σ_{acc} of the PNSD to ARG-simulated N_d and quantify their relative contributions to ARG-simulated N_d variability. We note that the κ_{ait} , κ_{acc} values are measured at ground level, but N_{ait} , N_{acc} , μ_{ait} , μ_{acc} , σ_{ait} , and σ_{acc} are measured at cloud level. To the best of our knowledge, this is the first time that the measured out-of-cloud PNSD parameters have been integrated into the ARG model to estimate their contribution to ARG-simulated N_d . Figure 5 illustrates the comparison between in-situ measured N_d and ARG-simulated N_d . The simulation achieves a strong correlation in both seasons ($R^2 > 0.81$). The observed-to-simulated N_d ratio is approximately 0.5 in summer and slightly below 1.0 in winter, indicating that the dominant controls on ARG-simulated N_d differ between seasons, as discussed in the following section.

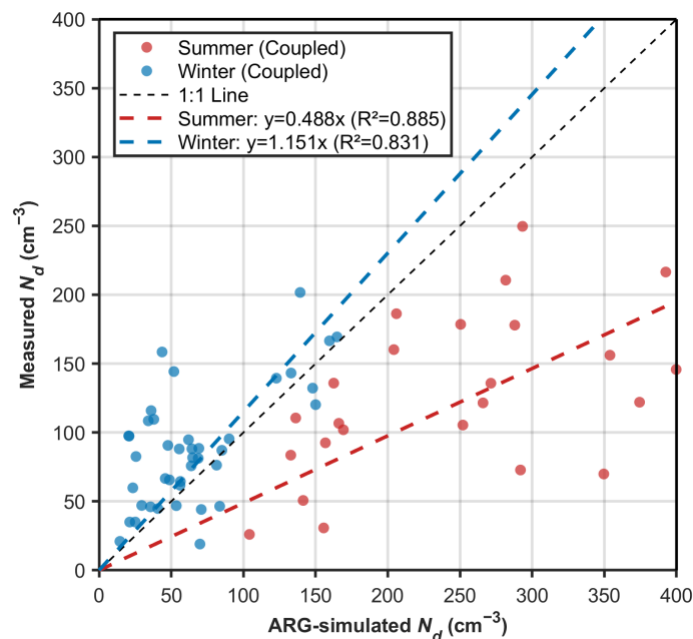




Figure 5. Comparison of ARG-simulated cloud droplet number concentrations (N_d) simulated by the ARG scheme versus those measured by FCDP on the G-1 aircraft in cloud. Red and blue dots represent data from the summer (June–August 2017) and winter (December 2017 – February 2018) campaigns, respectively. The black dashed line indicates the 1:1 relationship. Colored dashed lines represent the corresponding zero-intercept linear fits.

3.3.2 Controlling factors of cloud droplet formation

To evaluate the drivers of the contrasting seasonal biases in N_d , we performed a cross-substitution sensitivity analysis between summer and winter (Figs. 6 and 7) based on the 9 primary factors controlling aerosol CCN activation. Specifically, in each experiment, one input variable in the ARG parameterization was replaced at a time with its corresponding median value (Table S3) from the opposite season, while the remaining eight input parameters were kept at the original values of the season. Specifically, one summer input parameter was replaced by its winter median value for the summer simulations, while one winter input parameter was replaced by its summer median value for the winter simulations. The ARG parameterization was then rerun using the modified set of inputs. The sensitivity of aerosol activation to each factor is quantified as b / a , where a is the original regression slope between measured and ARG-simulated N_d and b is the slope after replacing one input parameter with its corresponding opposite-season median value. Figures 6 and 7 illustrate how changes in individual input variables affect the performance of the ARG-simulated N_d , and help identify which variables contribute to its contrasting seasonal biases.

We first test whether the seasonal difference in w can explain the N_d discrepancy. Summer updrafts are slightly weaker than winter updrafts, with summer median $w = 0.16 \text{ m s}^{-1}$, while winter $w = 0.24 \text{ m s}^{-1}$. Replacing summer w with the winter season median value increases ARG-simulated N_d by $\sim 7\%$ (Fig. 6a). This response can be explained by the fact that summer clouds are aerosol-rich but updraft-limited, increasing w enhances the peak supersaturation and allows additional aerosol particles to activate. Since the original simulation already overestimates N_d , the additional increase in w would further increase simulated N_d and therefore slightly degrade the agreement with observations.

The sensitivity tests with aerosol physical properties indicate N_{acc} plays a governing role in cloud droplet formation. Substituting summer N_{acc} with the lower winter values (33.48 cm^{-3} , $\sim 1/8$ of the summer original median value) substantially reduces the ARG-simulated N_d (Fig. 6c). This perturbation produces the largest response among all tested parameters, with a slope ratio of 1.79 (Fig. 6c), indicating that summer N_d is primarily controlled by the abundance of accumulation-mode particles. Physically, a weak summer w limits the production of supersaturation, while abundant accumulation-mode particles provide a large population of potential CCN and a large total particle surface area for water-vapor condensation. The condensation growth consumes water vapor, suppressing the maximum supersaturation. Under weak w conditions, particles compete for the limited supersaturation, and only a fraction of the available particles can activate into cloud droplets.

Aitken-mode properties have weaker effects than accumulation-mode aerosols. Replacing summer μ_{ait} (with a median value of 57.18 nm) by the winter smaller value (37.08 nm) produces a modest response, with a slope ratio of 1.17 (Fig. 6f).



Replacing summer N_{ait} with the winter median value of 49.54 cm^{-3} ($\sim 1/4$ of the summer original median value) yields a slightly weaker response than μ_{ait} , with a slope ratio of 1.14 (Fig. 6b). This indicates that μ_{ait} , which is also the proximity of Aitken-mode particles to the critical activation diameter, is slightly more important than their total number. The smaller
345 winter μ_{ait} shifts the Aitken-mode population farther from the activation threshold, whereas changes in N_{ait} mainly affect the number of particles that are too small to activate.

Other aerosol physical shape parameters (μ_{acc} , σ_{ait} , and σ_{acc}) and chemical properties (such as κ_{ait} and κ_{acc}) exert only a minor influence, with narrow sensitivity ratios of 0.96–1.01 (Figs. 6d–e and 6g–i). This weak sensitivity likely reflects that moderate changes in aerosol geometric standard deviation or hygroscopicity parameters do not substantially alter the ARG-
350 simulated N_d .

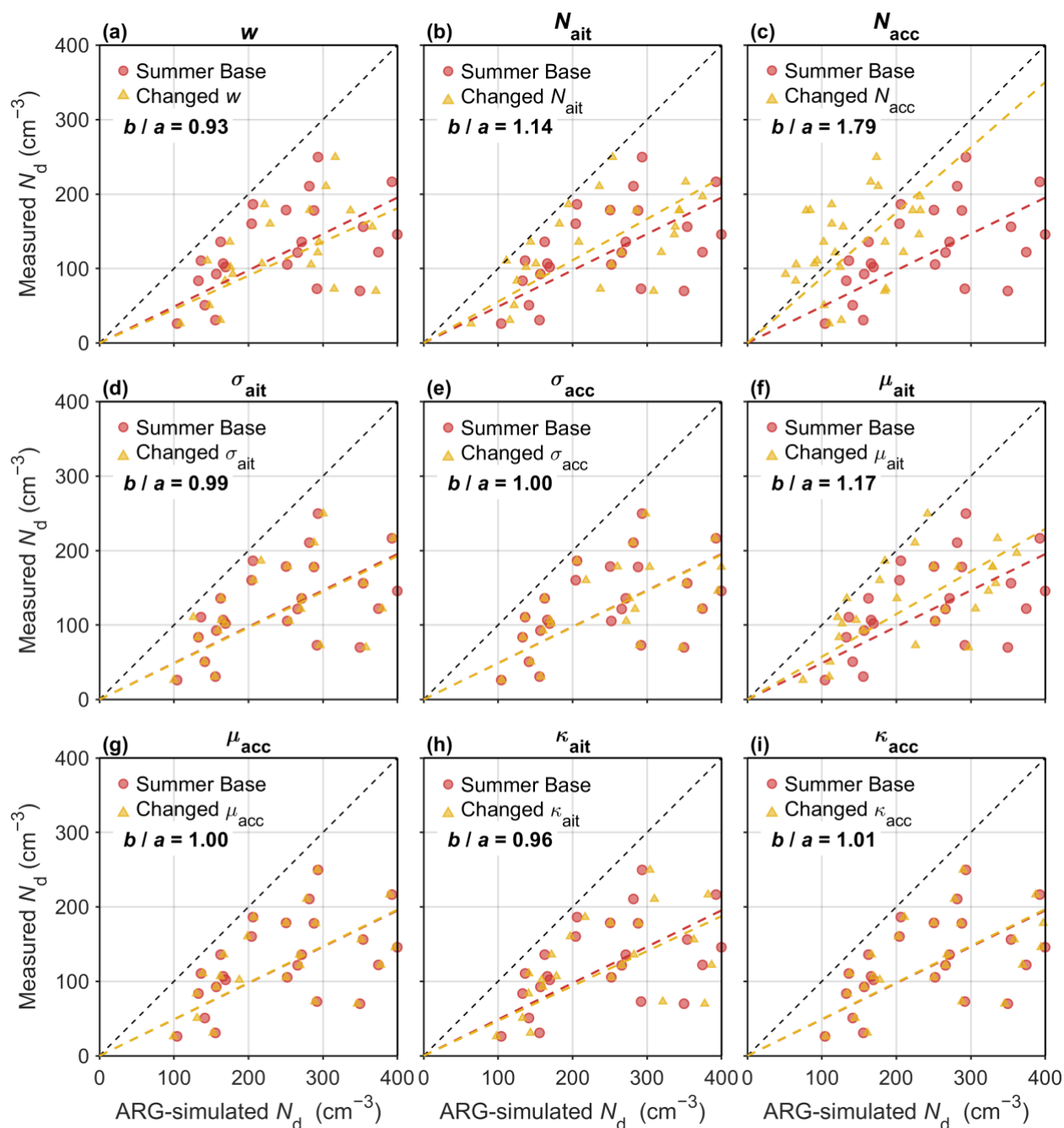


Figure 6. Quantifying the impact of individual meteorological and aerosol parameters on the closure between measured and ARG-simulated cloud droplet number concentration in the summer period. In each experiment, the summer simulations were repeated by replacing a single input parameter with its corresponding winter median value, while all other input parameters were kept at their original summer values: (a) updraft velocity (w); (b-c) number concentrations (N_{ait} and N_{acc}) for Aitken and accumulation modes, respectively; (d-e) geometric standard deviations (σ_{ait} and σ_{acc}); (f-g) geometric mean diameter (μ_{ait} and



μ_{acc}); and (h-i) hygroscopicity parameters (κ_{ait} and κ_{acc}). Here, a denotes the original regression slope between measured and ARG-simulated N_d , and b denotes the regression slope after replacing the indicated input parameter with its winter median value. b / a represents the relative change in the regression slope caused by the single-parameter substitution.

360 Winter is characterized by a lower aerosol loading ($N_{acc} = 33.48 \text{ cm}^{-3}$, $\sim 1/8$ of the summer original median value) and a relatively stronger updraft (with a median $w = 0.24 \text{ m s}^{-1}$, compared with $w = 0.16 \text{ m s}^{-1}$ in summer). Consistent with the summer sensitivity results, ARG-simulated winter N_d remains a moderate response to the w with a sensitivity ratio of 1.08 (Fig. 7a). The responses to σ_{ait} , σ_{acc} , κ_{ait} , and κ_{acc} are also weak, with narrow sensitivity ratios of 0.97–1.04 (Figs. 7d–e and 7g–i). These results indicate that updraft, hygroscopicity, and aerosol geometric standard deviation stay relatively consistent
 365 and exert secondary influences on N_d in both seasons.

Among the aerosol physical parameters, N_{acc} continues to exert a dominant control. Replacing the low winter N_{acc} with high summer values introduces a significant change of ARG-simulated winter N_d with a sensitivity ratio of 0.32 (Fig. 7c). This strong response highlights the primary role of accumulation-mode particles as readily activatable CCN in both seasons. Winter N_d also exhibits similar sensitivity to both N_{ait} and μ_{ait} . Substituting winter N_{ait} with the larger summer value yields a
 370 ratio of 0.75 (Fig. 7b), and replacing the smaller winter μ_{ait} with the larger summer value gives a ratio of 0.76 (Fig. 7f). These comparable responses suggest that both the abundance of Aitken-mode particles and their diameters regulate winter cloud droplet activation. Figure A3 shows that the activation fraction of N_d / N_{acc} frequently exceeds 2.0 in winter, indicating that accumulation-mode particles alone cannot explain the ARG-simulated N_d . This is because accumulation-mode aerosol has a low concentration in winter, they are insufficient to consume the generated water vapor. The resulting high S_{max} enables the
 375 activation of Aitken-mode particles. These findings confirm that in the remote winter MBL, the system lacks sufficient accumulation-mode aerosol to contribute to the CCN population, leaving Aitken-mode aerosol particles to act as critical drivers for cloud droplet formation.

Overall, the sensitivity experiments show that N_d is mainly controlled by aerosol availability rather than by changes in background w . More specifically, aerosol-rich conditions in summer intensify the competition for water vapor among
 380 particles, thereby suppressing the generation of high supersaturation. In winter, the limited availability of accumulation-mode aerosol weakens this competition, producing high supersaturation conditions that favor the activation of smaller particles, thereby enhancing the sensitivity of N_d to Aitken-mode properties (N_{ait} and μ_{ait}).

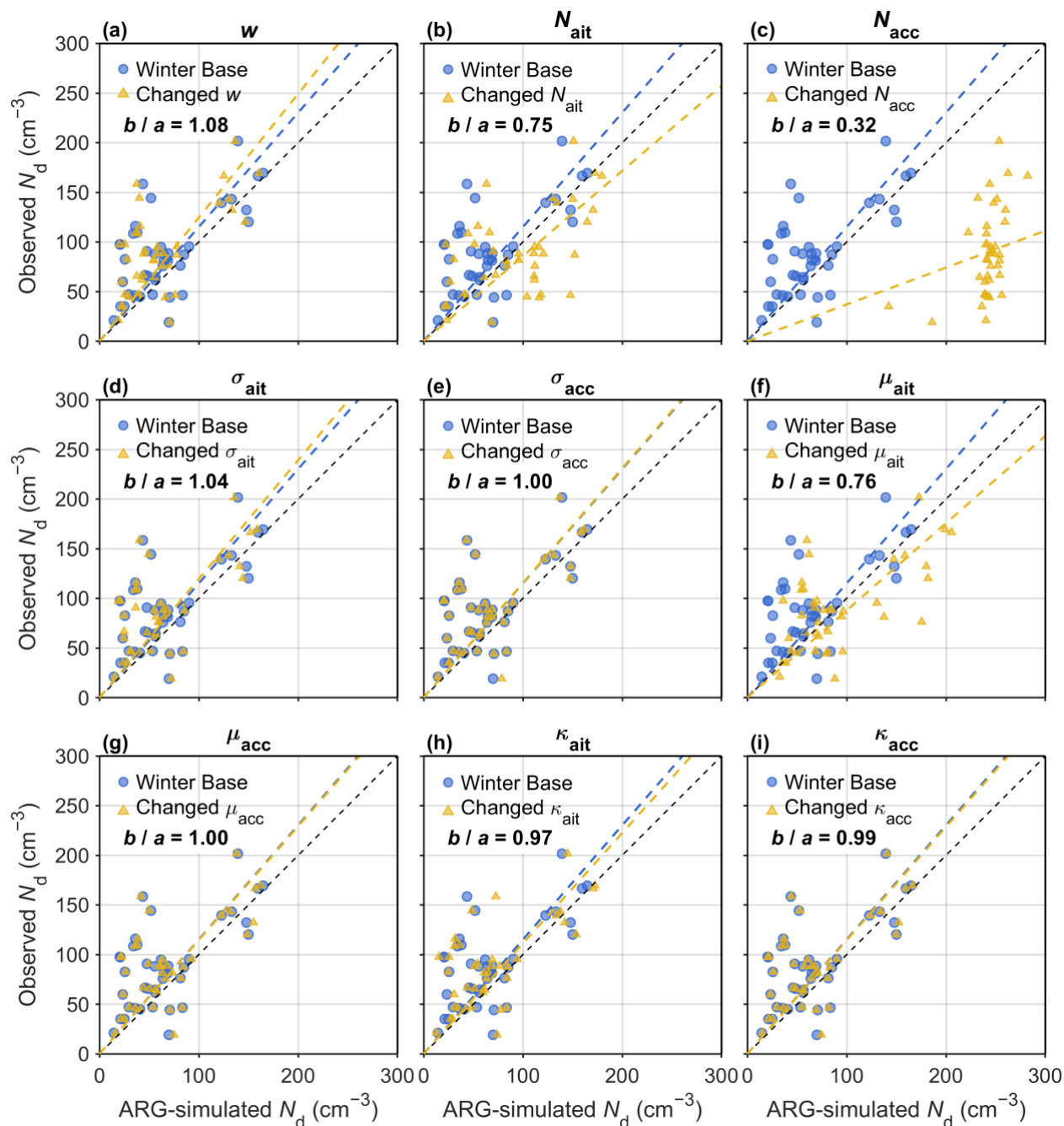


Figure 7. Quantifying the impact of individual meteorological and aerosol parameters on the closure between measured and ARG-simulated winter cloud droplet number concentration. In each experiment, the winter simulations were repeated by replacing a single input parameter with its corresponding summer median value, while all other input parameters were kept at their original winter values: (a) updraft velocity (w); (b-c) number concentrations (N_{ait} and N_{acc}) for Aitken and accumulation modes, respectively; (d-e) geometric standard deviations (σ_{ait} and σ_{acc}); (f-g) geometric mean diameter (μ_{ait} and



390 μ_{acc}); and (h-i) hygroscopicity parameters (κ_{ait} and κ_{acc}). Here, a denotes the original regression slope between measured and ARG-simulated N_d , and b denotes the regression slope after replacing the indicated input parameter with its summer median value. b / a represents the relative change in the regression slope caused by the single-parameter substitution.

4 Conclusions

This study examines how seasonal differences in marine boundary layer (MBL) structure regulate the vertical transport of surface aerosols and the activation of aloft aerosols into cloud droplets, and hence cloud droplet number concentration (N_d) over the Eastern North Atlantic (ENA). By integrating aircraft measurements from the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) campaign with synchronous ground-based observations, we show that seasonal changes in boundary-layer coupling states and aerosol availability jointly regulate cloud droplet activation.

The vertical structure of the MBL, whether it is well-mixed (coupled) or not (decoupled), modulates the seasonally available near-surface aerosol for cloud activation. Our analysis does not explicitly account for additional sources of MBL aerosols, such as new particle formation in the upper MBL and aerosol entrainment from the free troposphere, which may also be important sources of the MBL aerosol population (Wang et al., 2021; Zheng et al., 2018). These processes were not considered as independent factors because their individual contributions cannot be explicitly quantified within the present parameterization. Instead, our analysis focuses on how the MBL coupling states affect the vertical transport efficiency of near-surface aerosols and the representativeness of ground-based aerosol measurements for cloud-level activated aerosols. Thermodynamic decoupling is highly prevalent (occurring in $\sim 1/3$ of both summer and winter), and the two seasons show quite comparable frequency (summer about 32.6% and winter about 32.0%). This decoupling limits the vertical transport of near-surface aerosols into the cloud layer and reduces the representativeness of ground-based aerosol measurements for cloud-level cloud condensation nuclei (CCN). Consistently, our observations indicate that only 60% of the near-surface accumulation-mode aerosol concentration (used as a proxy for CCN) is aloft under the decoupled case.

410 Sensitivity analyses using the ARG parameterization and out-of-cloud particle parameters further quantify the major physical drivers of N_d in each season. In summer, simulated N_d is highly sensitive to accumulation-mode aerosol concentrations (N_{acc} , with a sensitivity ratio of 1.79) but only weakly sensitive to seasonal changes in w (sensitivity ratio of 0.93).

The winter MBL is characterized by lower accumulation-mode aerosol concentration and a more CCN-limited activation regime. The lower N_{acc} reduces competition for water vapor among aerosol particles, enabling high supersaturations that favor the activation of Aitken-mode particles. This is evidenced by the high N_d / N_{acc} ratio (> 2) observed in winter, indicating that accumulation-mode particles alone cannot explain cloud droplet numbers and that Aitken-mode particles also contribute. Winter N_d is also sensitive to the Aitken-mode number concentration (N_{ait}) and geometric mean diameter (μ_{ait}), with the sensitivity ratios of 0.75 and 0.76, respectively.

420 Across both seasons, aerosol hygroscopicity parameter (κ) and geometric standard deviation parameter (σ) exert only a secondary influence (sensitivity ratios of 0.96–1.04). These results show that seasonal N_d variability is controlled primarily



by aerosol abundance and size-related physical properties, with N_{acc} predominantly contributing to CCN in both seasons, and Aitken-mode particles becoming important under winter CCN-limited conditions. Because the sensitivity analysis used out-of-cloud aerosol properties, the influence of MBL coupling states is implicitly reflected in the aerosol inputs rather than treated as a separate sensitivity factor.

425 In summary, this study provides two main observational constraints for representing marine aerosol-cloud interactions in models. First, the representativeness of surface aerosol measurements for cloud-base CCN depends strongly on the MBL vertical mixing states and has large uncertainty under decoupled MBL conditions. Models should therefore account for the coupled or decoupled structure of the MBL when linking surface aerosol properties to cloud droplet activation, rather than assuming that surface aerosol concentrations are directly representative of the cloud layer. Second, commonly used aerosol
430 activation parameterizations (e.g., Abdul-Razzak and Ghan, 2000) may exhibit biased under high N_{acc} conditions. Our sensitivity analysis shows that N_{acc} dominates simulated N_d in summer, under abundant accumulation-mode aerosol conditions, whereas N_{ait} and μ_{ait} become additionally important under winter accumulation-mode-limited conditions. In contrast, aerosol size-distribution parameters (σ), κ , and updraft velocity (w) had comparatively weaker effects over the observed ranges.

435 These results also have implications for marine cloud brightening (MCB), which proposes spraying fine sea-salt particles into marine clouds to make them brighter and more reflective. Based on the results in this study, we think MCB has diminishing effectiveness under aerosol-rich, weak-updraft summer conditions, because additional seed particles could intensify competition for water vapor, reduce S_{max} , and limit further increases in N_d and cloud albedo. Consequently, MCB would likely fail to increase N_d or cloud albedo during these high aerosol loading regimes.

440 **Appendix A**

This appendix provides 3 additional figures that are discussed in the main manuscript. Figure A1 presents ground particle size distributions (PNSD_G) measured by SMPS during the summer and winter seasons. Figure A2 presents probability density functions (PDFs) of key parameters controlling ACI. Figure A3 presents the sensitivity of the aerosol activation fraction (N_d / N_{acc}) to w , calculated using the Abdul-Razzak and Ghan (2000) parameterization.



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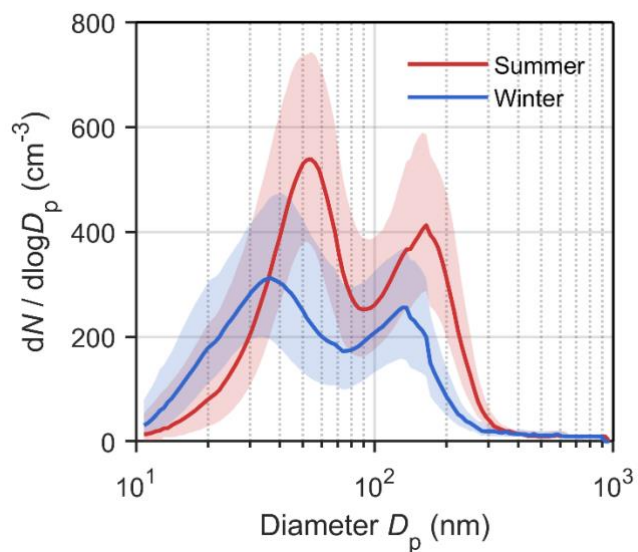


Figure A1. Ground particle size distributions (PNSD_G) measured by SMPS during the summer and winter seasons. Solid lines represent the median distributions, and the shaded regions denote the interquartile range (25th–75th percentile).

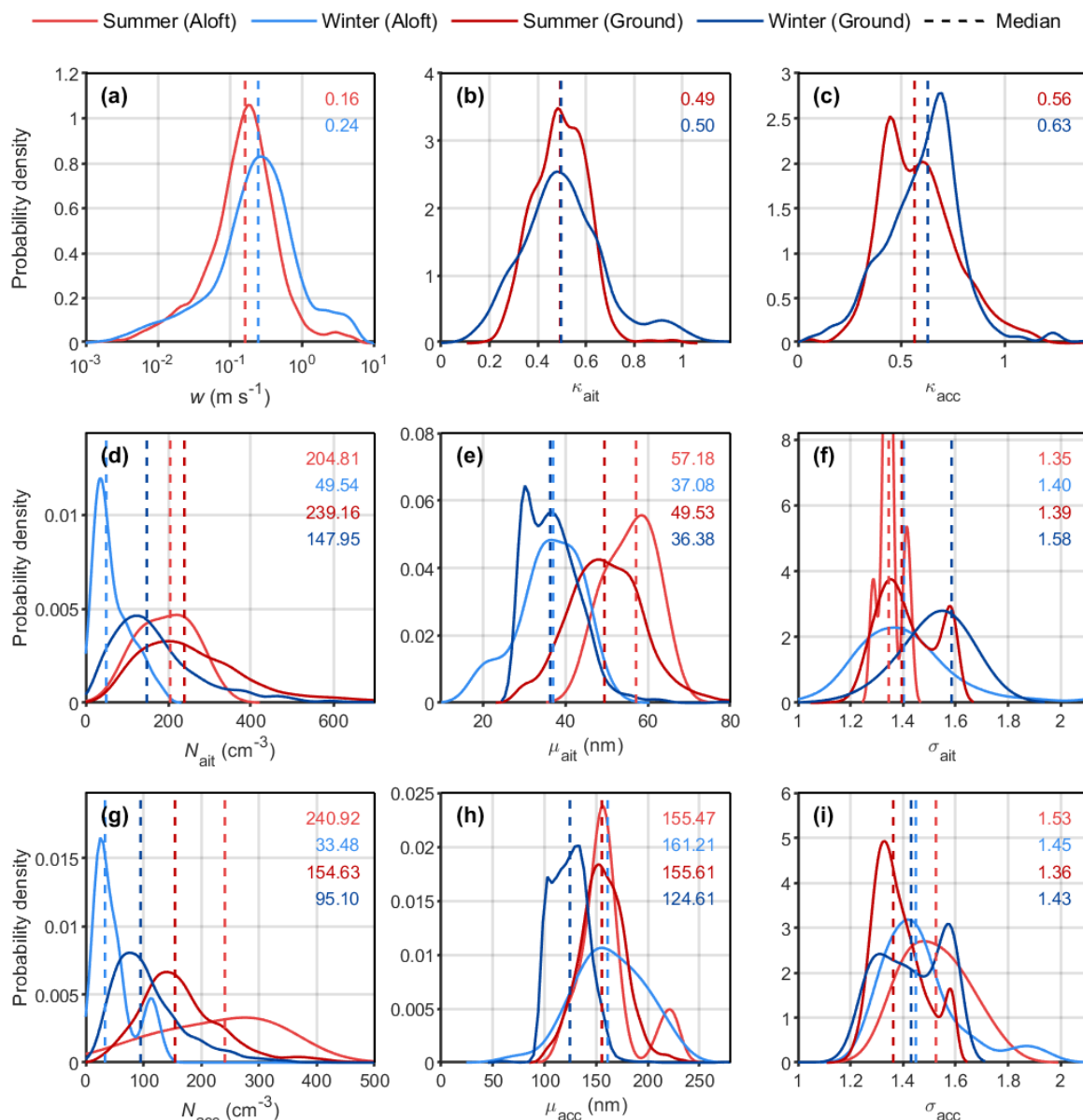


Figure A2. Probability density functions (PDFs) of key parameters controlling aerosol-cloud interactions (ACI): (a) updraft velocity (w); (b–c) aerosol hygroscopicity (κ) for the Aitken and accumulation modes, respectively; (d–f) Aitken-mode properties, including (d) number concentration (N_{ait}), (e) geometric mean diameter (μ_{ait}), and (f) geometric standard deviation (σ_{ait}); and (g–i) accumulation-mode properties, including (g) number concentration (N_{acc}), (h) geometric mean diameter (μ_{acc}), and (i) geometric standard deviation (σ_{acc}).

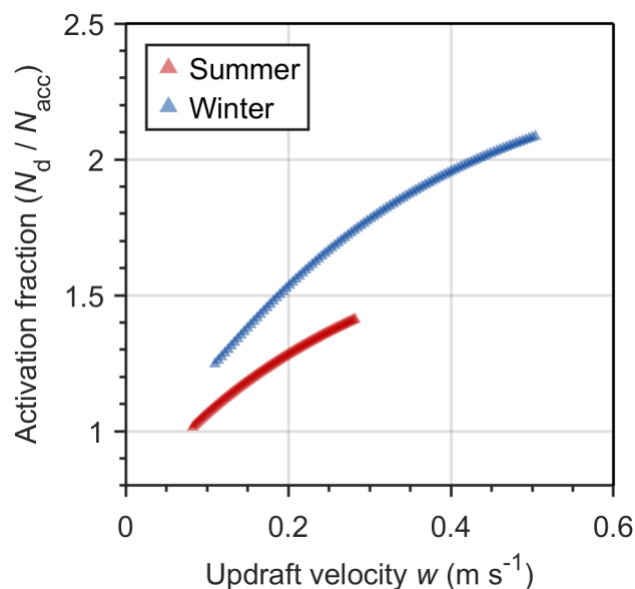


Figure A3. Sensitivity of the aerosol activation fraction (N_d / N_{acc}) to updraft velocity (w), calculated using the Abdul-
 455 Razzak and Ghan (2000) parameterization. The plotted x-axis range corresponds to the interquartile range (25th–75th
 percentiles) of the observed updraft velocity for summer and winter seasons.

Appendix B: Summarize the Abbreviations of Variables

ACE-ENA: Aerosol and Cloud Experiments in the Eastern North Atlantic

ACI: Aerosol-cloud interaction

460 AMSL: Above mean sea level

ARG: Abdul-Razzak and Ghan Scheme

CCN: Cloud condensation nuclei

ENA: Eastern North Atlantic

HM: Hoppel minimum

465 LWC: Liquid water content

MBL: Marine boundary layer

N_d : Cloud droplet number concentration

N_{CCN} : Cloud condensation nuclei number concentration

N_{10nm-A} : Total particle number concentration aloft for $D_p > 10$ nm

470 N_{ait} : Aitken-mode aerosol number concentration

N_{acc} : Accumulation-mode aerosol number concentration

N_{ait-G} : Ground Aitken-mode aerosol number concentration



$N_{\text{acc-G}}$: Ground accumulation-mode aerosol number concentration

$N_{\text{ait-A}}$: Aloft Aitken-mode aerosol number concentration

475 $N_{\text{acc-A}}$: Aloft accumulation-mode aerosol number concentration

PNSD: Particle number size distribution

PNSD_G : Ground particle number size distribution

PNSD_A : Aloft particle number size distribution

SCCN: Size-resolved cloud condensation nuclei measurement

480 w : Updraft velocity

q_i : Water vapor mixing ratio

Δq_i : Difference in water vapor mixing ratio between the near-ground level and below cloud base

θ : Potential temperature

$\Delta\theta$: Difference in potential temperature between the near-ground level and below cloud base

485 κ : Aerosol hygroscopicity parameter

μ_{ait} : Geometric mean diameter of the Aitken-mode aerosol size distribution

μ_{acc} : Geometric mean diameter of the accumulation-mode aerosol size distribution

σ_{ait} : Geometric standard deviation of the Aitken-mode aerosol size distribution

σ_{acc} : Geometric standard deviation of the accumulation-mode aerosol size distribution

490 **Data availability**

The ACE-ENA data used in this study are available at the ARM Climate Research Facility data archive (www.arm.gov/research/campaigns/aaf2017ace-ena).

Author contributions

L.W. and X.G. contributed to Conceptualization, Data curation, and Formal analysis. L.W. performed Investigation, Methodology, Visualization, and Writing – original draft. X.G. contributed to Data curation and assisted with formal analysis. J.Z., J.C., X.X., Y.C., S.C., Y.Z., Z.Z., Y.W., and X.G. provided help with the Methodology, as well as the Validation. X.G. served as the lead supervisor for the study. All authors contributed to Writing – review & editing, read and approved the final manuscript.

Competing interests

500 The authors declare that there are no conflicts of interest for this manuscript.



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