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Regime-based Aerosol-Cloud Interactions from CALIPSO-MODIS and the Energy Exascale Earth System Model version 2 (E3SMv2) over the Eastern North Atlantic

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Abstract. This study investigates aerosol-cloud interactions in marine boundary layer (MBL) clouds using a regime-based approach, combining satellite (CALIPSO-derived aerosol extinction coefficients and MODIS-derived cloud properties) with simulations from a 1° nudged Energy Exascale Earth System Model version 2 (E3SMv2), over a ~10°×10° domain in Eastern North Atlantic (ENA) from 2006 to 2014. The E3SMv2 captures observed seasonal variations in cloud droplet number concentrations (N_d) and liquid water path (LWP), though it systematically underestimates N_d. Using deep-learning-based clustering, ENA meteorology was partitioned into four distinct synoptic regimes, enabling regime-dependent aerosol-cloud interactions analyses. Both satellite and E3SMv2 reveal inverted-V relationships between LWP and N_d, though specific slopes vary across different regimes. In Pre-Trough regime, both datasets indicate rising LWP at low N_d, but model LWP peaks more rapidly, suggesting overly aggressive drizzle suppression. In Post-Trough regime and Ridge regime, satellite shows stronger negative LWP–N_d sensitivities while model predicts more exaggerated responses. While Trough regime exhibits a muted LWP response in satellite, and slightly negative response in model. Exaggerated model LWP sensitivities may stem from uncertainties in representing drizzle processes, entrainment, and turbulent mixing. As for cloud responses to aerosols, both datasets confirm that N_d increases with MBL aerosol extinction, although the simulated aerosol-cloud interactions appear overly sensitive to environmental conditions. Overall, E3SMv2 captures aerosol impacts on stratiform clouds effectively but performance deteriorates for deeper, dynamically complex clouds, highlighting the need for improved representations of cloud processes within climate models.

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1. Introduction

Marine boundary layer (MBL) clouds play a pivotal role in regulating the Earth's energy budget due to their extensive coverage over the oceans and high albedo. These low-level clouds reflect a substantial amount of incoming solar radiation back into space, thereby exerting a net cooling effect on the planet (Albrecht et al., 1995; Wood et al., 2015; Dong et al., 2023; Wall et al., 2023). Central to the quantification of the radiative impact of MBL cloud properties is to determine the sensitivity of clouds to the presence of aerosols. Aerosols can impact cloud microphysical properties, such as the cloud droplet number concentration (N_d) and droplet effective radius (r_e), and, consequently, alter cloud optical and macrophysical properties, including liquid water path (LWP) and cloud fraction (Twomey, 1977; Albrecht et al., 1989; McComiskey et al., 2009; Zheng et al., 2020). The interactions between aerosols and clouds, commonly termed aerosol-cloud interactions (ACI), contribute to one of the largest uncertainties in climate projections (IPCC, 2021). These uncertainties in the aerosol-induced cloud microphysical responses and the accompanying cloud adjustments stem from the inherent complexity of cloud microphysical processes (e.g., droplet activation, precipitation suppression, and entrainment-induced evaporation) and their interactions with dynamically evolving MBL, where aerosol perturbations can potentially contribute to either the brightening or darkening of clouds (Wall et al., 2022; Feingold et al., 2024).

Satellite remote sensing observations are essential in efforts to quantify the cloud adjustment to aerosol perturbations, by providing spatially extensive and temporally resolved datasets. Numerous studies using satellite data have demonstrated a significant relationship and progressively advanced our understanding of cloud adjustments to aerosols (e.g. Bellouin et al., 2020). Observational evidence frequently shows a negative correlation between N_d and LWP, particularly in high-aerosol environments, suggesting a reduction in LWP attributed to enhanced entrainment and evaporation driven by aerosol-induced smaller droplet sizes. However, distinguishing causality from correlations is a persistent challenge, as atmospheric variability, retrieval biases, and sampling limitations can obscure true aerosol-induced effects (Arola et al., 2022; Goren et al., 2023; Liu et al., 2024). For example, Zhang et al. (2022) demonstrate that the relationship between LWP and N_d is not only sensitive to aerosol loading but also modulated by the underlying meteorological conditions. Their work, along with other studies (McCoy et al., 2020; Gryspeerdt et al., 2022), highlight that distinct atmospheric regimes may lead to either enhancement or suppression of LWP with increased N_d.

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Parallel to observational advances, the modeling community has made significant progress in simulating aerosol-cloud interactions within global climate models (GCMs). Recent GCM versions incorporate more physically based cloud microphysics parameterizations, which enable the simulation of precipitation suppression and enhanced entrainment responding to aerosol changes. For instance, studies by Mülmenstädt et al. (2024a) show that some of the GCMs in the Coupled Model Intercomparison Project Phase 6 (CMIP6), namely the US Department of Energy (DOE) Exascale Earth System Model version 1 (E3SMv1; Golaz et al., 2019) and others, can capture the inverted V-shaped relationship between LWP and N_d, as often observed from satellite retrievals, although discrepancies persist regarding the causal interpretation of these relationships. Moreover, even though E3SMv1 simulates the general trend of increased N_d with higher aerosols, it overestimates N_d by more than two to three times relative to observations, indicating an overpredicted sensitivity of N_d to aerosols (Christensen et al. 2023; Varble et al., 2023). Tang et al. (2024) further highlight that even when E3SM version 2 (E3SMv2; Golaz et al., 2022) simulates the overall cloud macrostructure, the microphysical responses to aerosol perturbations are still subject to systematic uncertainties related to precipitation processes and turbulence–microphysics interactions. Collectively, these studies illustrate both the strides made and challenges remained in representing aerosol-cloud interactions in large-scale models. While some GCMs may successfully replicate observed negative LWP-N_d relationships under the present-day conditions, they struggle to accurately simulate the turbulence, entrainment, and precipitation feedback, that govern LWP adjustments in response to aerosol changes (Mülmenstädt et al., 2024b).

One of the major hurdles lies in isolating the direct influence of aerosols on cloud microphysics from the effects of meteorological variability. Inconsistent methodologies, such as different sampling strategies, aerosol proxies, and comparative metrics, further complicate the comparisons between observations and model outputs. For instance, studies leveraging vertical aerosol extinction profiles rather than column-integrated aerosol optical depth have achieved more robust correlations between aerosols and cloud microphysics, emphasizing the need for refined observational approaches (Painemal et al., 2019, 2020). Furthermore, retrieval errors in satellite measurements of N_d and LWP, compounded by issues such as vertical mismatch between aerosol and cloud layers, uncertainties in cloud retrievals, as well as updraft variability and precipitation effects, can bias the estimates of cloud sensitivity to aerosol perturbations (Quaas et al., 2020; Gryspeerdt et al., 2022; Jia et al., 2022; Alexandri et al., 2024). Similarly, analyses by Mülmenstädt et al. (2024b) and Zhang & Feingold (2023) have pointed out that uncertainties in the observed cloud adjustment processes, such as those related to the balance between precipitation suppression and enhanced entrainment-induced evaporation, remain a persistent source of discrepancy between observational inferences and model simulations. These challenges are compounded

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by the complex, multiscale nature of ACIs, where small-scale processes interact nonlinearly with larger-scale meteorological drivers. Nevertheless, assessing model performance in simulating cloud microphysical responses to aerosol perturbations using observation is a crucial step toward improving the process-level understanding.

The Eastern North Atlantic (ENA) region is uniquely advantageous for advancing our understanding of ACIs in MBL clouds (Wood et al., 2015; Tian et al., 2025). Located at the confluence of subtropical and midlatitude air masses, the ENA is characterized by diverse meteorological conditions and cloud regimes. This region frequently experiences well-organized stratocumulus cloud decks and other MBL cloud types, which are sensitive to both local and long-range transported aerosols (Wang et al., 2020; Wang et al., 2022). Observational studies have documented the distinct aerosol and cloud properties over the ENA, noting that the relatively pristine marine environment, combined with episodic aerosol perturbations, facilitates a clear separation of aerosol-induced cloud adjustments from meteorological confounders (Zheng et al., 2022b; Varble et al. 2023; Christensen et al. 2024; Qiu et al., 2024). Moreover, the ENA has been extensively sampled, with long-term observational data collected at multiple spatiotemporal resolutions from various platforms including the DOE's Atmospheric Radiation Measurement (ARM) research facility (Wood et al., 2015), and several satellite remote sensing products such as those from the Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO; Winker et al., 2009, 2010), and the MOderate Resolution Imaging Spectroradiometer (MODIS) on board Terra and Aqua (Barnes et al., 1998; Platnick et al., 2003; King et al., 2013). These comprehensive observational datasets make the ENA an ideal testbed for evaluating model parameterizations of aerosol-cloud interactions.

In this study, we employ an integrated approach that combines CALIPSO-derived aerosol extinction profiles and MODIS-derived cloud properties with simulations from the DOE E3SM version 2 (E3SMv2). By applying a clustered-meteorology-regime-based analysis over the ENA, we aim to isolate the impact of aerosol perturbations on cloud microphysical properties by regime and seek to provide a robust observational and modeling framework for quantitatively assessing aerosol-induced cloud adjustments. This approach is not only intended for reconciling discrepancies between satellite observations and model simulations but also for informing about potential model uncertainties in ACIs in connection with specific meteorological regimes. The data and methods used in this study are introduced in Section 2. The simulated aerosols, clouds, and their interactions are examined in Section 3, compared to the satellite observations. The meteorological-regime-based analysis of the aerosol-cloud interaction and cloud adjustments are discussed in Section 4. The conclusion and future work are presented in Section 5.

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2. Data and Method

2.1 Satellite retrievals of aerosols and clouds

In this study, aerosol extinction coefficient (σ_{EXT}) is a research product derived from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on the CALIPSO. These retrievals are produced at a 1 km along-track resolution using the Fernald–Klett iterative approach, constrained by an independent CALIOP-based aerosol optical depth (AOD), and are limited to cloud-free pixels. The retrieved profiles have been evaluated against airborne High Spectral Resolution Lidar (HSRL) measurements during the Caribbean 2010 field campaign and show good agreement. The detailed aerosol retrieval methodology and product evaluation are described in Painemal et al. (2019) and Li et al. (2022).

Cloud properties, including the liquid water path (LWP) and the cloud droplet number concentration (N_d), are obtained from MODIS Aqua at 1 km resolution using Clouds and Earth's Radiant Energy System (CERES) Edition 4.0 algorithms (Minnis et al., 2021). The MODIS-retrieved LWP is estimated to have uncertainties of approximately 10–15% when compared with ARM surface-based observations (Xi et al., 2014; Painemal et al., 2016). The N_d is retrieved using the adiabatic formulation (Painemal and Zuidema, 2011; Grosvenor et al., 2018):

$$N_d = \Gamma^{1/2} \frac{10}{4\pi \rho_W^{1/2} k} \frac{\tau^{1/2}}{r_e^{5/2}} ,$$

Where the cloud droplet effective radius (r_e) and cloud optical depth (τ) are estimated from MODIS 3.79 µm and 0.64 µm bands, respectively (Painemal et al., 2013, 2020). Γ denotes the adiabatic lapse rate of condensation (Albrecht et al., 1990), which is calculated from the cloud-top temperature and pressure derived from MODIS (Painemal et al., 2020). ρ_W is the water density and k represents the ratio between the cloud droplet volume mean radius and the effective radius, assumed to be constant at 0.8 (Martin et al., 1994).

To collocate aerosol and cloud retrievals from the two satellite datasets, the following matching method is employed. For each 1-km CALIOP aerosol pixel, five 1-km MODIS pixels are selected on each side of the CALIPSO track (thus 10 MODIS pixels in total). The CALIOP retrievals are first averaged to produce a 5 km along-track resolution product, and the MODIS cloud retrievals are aggregated into four 5 km × 5 km grids (two grids east and two west of the CALIPSO track). These datasets are then further averaged over approximately 25 km along-track segments, ensuring that the

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aerosol and cloud data are matched at similar spatial scales. Hence, the clear-sky aerosol extinction profiles can be collocated in the vicinity of the clouds, enabling the 'simultaneous' assessment of aerosol-cloud interaction from the satellite data around 1 p.m. local time over ENA region. For a detailed description of the data-matching strategy, please refer to Painemal et al. (2020) and Li et al. (2025).

Although the relative errors in N_d retrieval can be significant at the pixel scale (Grosvenor et al., 2018), previous studies have shown that the N_d compares well with measurements from 11 aircraft campaigns, demonstrating a decent correlation when sampling the marine stratocumulus clouds (Gryspeerdt et al., 2022). Moreover, the aggregated collocation method significantly reduces N_d bias (Painemal et al., 2020), resulting in a relationship between aerosol and cloud properties less affected by artifacts. In addition, the CloudSat Cloud Profiling Radar (CPR) is used to determine precipitation status from the satellite. The drizzle condition is defined as the ratio of pixels with maximum radar reflectivity between –15 and –7 dBZ to the total number of pixels (19) within the 25 km satellite collocated segment, while the thresholds for light rain and rain conditions are defined as maximum radar reflectivity greater than –7 dBZ but less than zero dBZ, and greater than 0 dBZ, respectively. Lastly, the collocated satellite datasets are analyzed within a ~10° × 10° domain over the ENA region (33.5–43.5°N, 23–33°W) with a local overpass time of approximately 1:30 p.m.. To simplify terminology, all aerosol and cloud properties retrieved from the different satellite products are hereafter referred to collectively as "satellite" data.

2.2 E3SM simulations

E3SMv2 is a fully coupled Earth System model (Golaz et al., 2022). Its atmospheric component, EAMv2, closely follows its predecessor EAMv1, as described in Rasch et al. (2019) and Xie et al. (2018), with only minor updates to its physical parameterizations. EAMv2 employs a spectral element dynamical core with approximately 110 km horizontal resolution and 72 vertical layers. The radiation and aerosol treatments in EAMv2 follow the Rapid Radiative Transfer Model (RRTM; Mlawer et al., 1997) and the four-mode version of the Modal Aerosol Module (MAM4; Liu et al., 2016; H. Wang et al. 2020), respectively. Turbulence, shallow convection, and cloud macrophysics are handled by the Cloud Layers Unified by Binormals (CLUBB) scheme (Larson, 2017; Golaz et al., 2022), while stratiform cloud microphysics is simulated by the Morrison-Gettelman (MG2) scheme (Gettelman and Morrison, 2015). Deep convection is represented by the Zhang and McFarlane Scheme (Zhang and McFarlane, 1995), as in EAMv1, but with a revised convective triggering function in EAMv2 that improves the simulation of precipitation and its diurnal cycle (Xie et al. 2019).

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In this study, EAMv2 was run at the standard resolution (~110 km) with the meteorology nudged to the ERA5 reanalysis. The nudged simulations reduce errors in simulated meteorological conditions, facilitating the examination of aerosol and clouds. Hourly outputs are available from 2006 to 2014 over the ~10°×10° ENA domain (33.5–43.5°N, 23–33°W), comprising 54 model columns. The aerosol extinction coefficient (σ_{EXT}) profile is directly outputted from the model. The MBL cloud samples are defined below 680 hPa to better match the satellite observations, and a cloud fraction threshold greater than 5% is used to determine a valid MBL cloud layer as in Kang et al. (2024). To further compare with the MODIS-retrieved cloud-top height (CTH), CTH is inferred by the diagnosed inversion height in E3SM. The inversion height is determined where $\left(\frac{\partial \theta_l}{\partial z}\right)\left(\frac{\partial RH}{\partial z}\right)$ is minimized, with the θ_l denoting liquid-water potential temperature and RH denoting relative humidity derived from the model outputs. Given the coarse vertical resolution of E3SM near the cloud top, this approach accounts for strong thermodynamic inversions and the effects of entraining dry air from the free troposphere (Erfani et al., 2022). The cloud-base height (CBH) is similarly identified using the 5% cloud fraction threshold. The in-cloud N_d is obtained from grid-box-averaged cloud liquid number, weighted by cloud fraction at each vertical level. Lastly, the cloud LWP is computed by integrating the in-cloud liquid water content (LWC) between cloud-top (CTH) and cloud-base (CBH) levels: $LWP = \int_{CBH}^{CTH} LWC\ dz$.

2.3 Clustering Model

Clustering meteorological patterns allows us to systematically characterize and categorize the diverse atmospheric conditions that modulate aerosol-cloud interactions in the ENA region. By identifying distinct meteorological regimes, we can, to some extent, isolate the aerosol-driven microphysical responses from the meteorological variability. In this study, we adapted an advanced deep learning-based clustering model proposed by Faruque et al. (2023), which features the architecture of the convolutional neural networks (CNN), and long short-term memory (LSTM) layers combined with a Deep Embedded Clustering (DEC) framework. This hybrid CNN-LSTM-DEC model was designed to capture complex spatiotemporal dependencies in meteorological data, overcoming limitations associated with conventional clustering methods that often treat spatial and temporal features separately (Zheng et al., 2025).

The clustering model was applied to ERA5 reanalysis data over a ~10°×10° domain (33.5–43.5°N, 23–33°W) in the ENA region for 2006–2014, with data at 1 pm local time daily to better match the time

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of satellite records. Input variables included 500 hPa geopotential height (Z500), mean sea level pressure (SLP), and the 10-meter u and v wind components. The CNN-LSTM-DEC architecture employs a sequence-to-sequence autoencoder with an encoder comprising four convolutional blocks (with filter sizes of 64, 128, 256, and 512), each followed by max-pooling to distill spatial features. An LSTM layer with 512 units captures temporal dependencies, and a dense layer with 256 units (using ReLU activation) defines the compressed latent space. A key aspect of our study was the fine-tuning of model hyperparameters through the grid search technique, which enabled us to systematically optimize clustering performance. The optimal configuration utilized a Stochastic Gradient Descent (SGD) optimizer with a learning rate of 0.01, momentum of 0.95, and a batch size of 32. Initially, latent features extracted by the encoder were clustered using K-means, with which clustering into four groups yielded the highest silhouette score of 0.267, compared to scores of 0.257, 0.178, and 0.167 for five, six, and seven clusters, respectively.

To further refine the clustering assignments, the DEC module was employed. By iteratively optimizing the combined clustering and reconstruction losses, the DEC module increased the silhouette score to 0.358, indicating enhanced cluster cohesion and separation. This two-step clustering process significantly reduced intra-cluster variability while accentuating differences between clusters, as suggested by Faruque et al. (2023). Their work also demonstrated that integrating both CNN and LSTM layers produces more robust latent representations, which are the reduced-dimensional encoding of the input that captures its most significant attributes, compared to CNN-only models or traditional approaches such as K-means and self-organizing maps. Furthermore, Zheng et al. (2025) showed that including additional meteorological variables, notably the 10-meter wind components, improved the model's ability to distinguish subtle synoptic regimes over the ENA region compared to studies that considered Z500 only (e.g., Mechem et al., 2018). Overall, the refined CNN-LSTM-DEC model demonstrates a marked improvement in clustering performance over traditional methods for analyzing large-scale meteorological phenomena.

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3. Aerosol and cloud properties from satellite and E3SMv2

3.1 Seasonal distribution of cloud properties

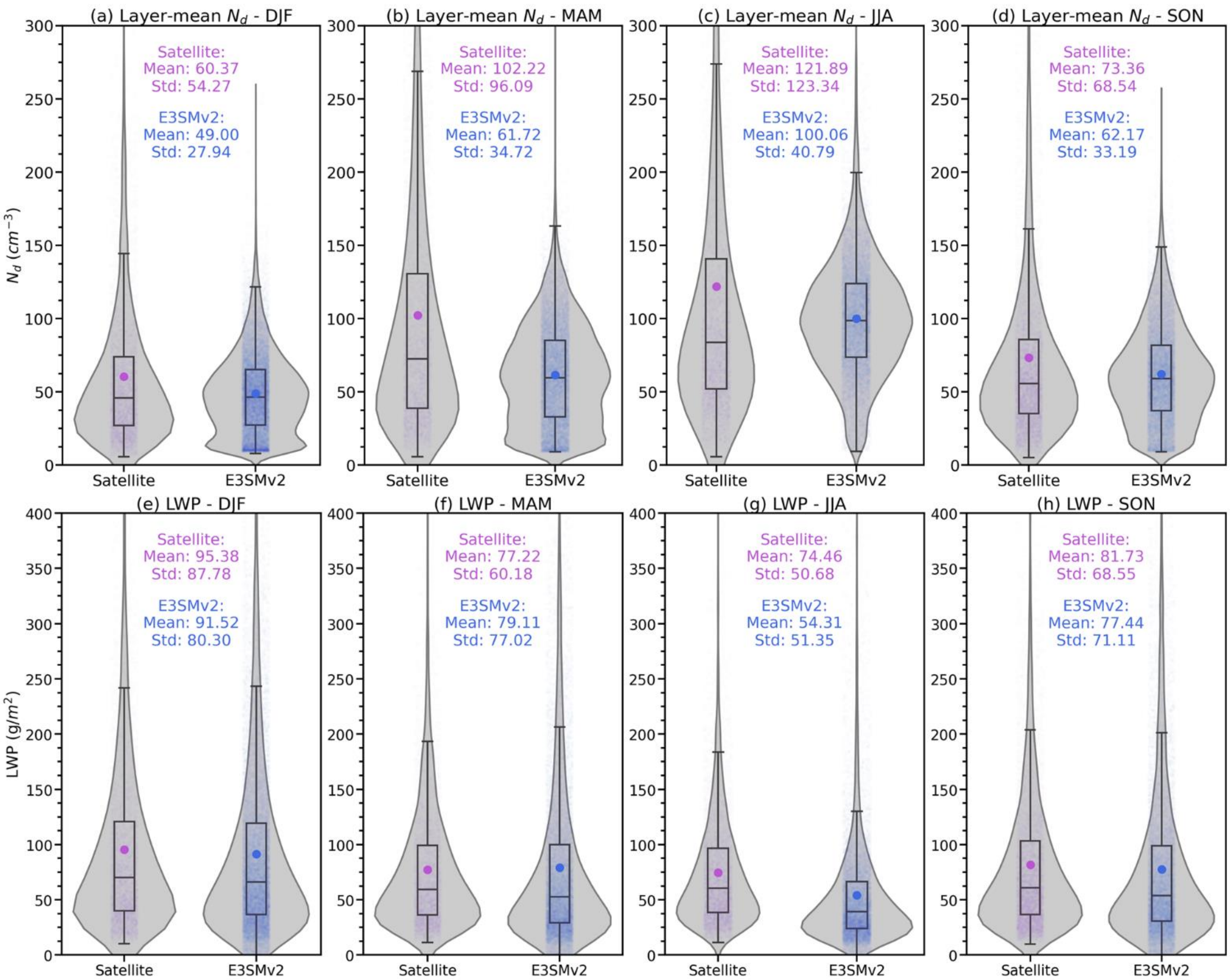

Figure 1. Violin plots of cloud droplet number concentration (N_d, top panels) and cloud liquid water path (LWP, bottom panels) from satellite retrievals (purple) and E3SMv2 simulations (blue), during winter, DJF (a, e), spring, MAM (b, f), summer, JJA (c, g), and fall, SON (d, h). The mean value is indicated by the color-coded dot. The smoothed shape of each violin shows the Gaussian kernel density estimate (KDE). From top to bottom within each violin, the box plot lines represent the third quartile (Q3, 75th percentile), median (Q2, 50th percentile), and first quartile (Q1, 25th percentile), respectively. The upper whisker extends to Q3 + 1.5 × IQR (interquartile range), and the lower whisker extends to Q1 − 1.5 × IQR.

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Figure 1 illustrates the seasonal variations in the layer-mean cloud droplet number concentration (N_d) for low-level clouds over the ENA, from satellite (MODIS) retrievals and E3SMv2 model. The annual mean statistics for N_d (LWP) are 88.34 ± 91.68 cm⁻³ (81.9 ± 66.46 g m⁻²) for MODIS and 66.27 ± 38.67 cm⁻³ (80.31 ± 73.38 g m⁻²) for E3SMv2. Satellite-derived N_d exhibits a pronounced annual cycle, with the highest mean values during summer (JJA, 121.89 cm⁻³) and spring (MAM, 102.22 cm⁻³), followed by (SON, 73.36 cm⁻³), and the lowest during (DJF, 60.37 cm⁻³). In comparison, the E3SMv2 simulates the seasonal variations, with N_d peaking in JJA (100.06 cm⁻³) and reaching a minimum in DJF (49.00 cm⁻³); however, the magnitude of these seasonal changes is smaller than that in the satellite data. Moreover, the model consistently underestimates N_d across all seasons, with particularly significant low biases during MAM and JJA, where the mean differences between the model and observations are 40.5 and 21.83 cm⁻³, respectively. Satellite observations display broader distributions with higher variability, manifested in larger standard deviations, especially during MAM (96.09 cm⁻³) and JJA (123.34 cm⁻³), indicating that the observed N_d encompasses a wider range of cloud conditions, likely influenced by diverse aerosol sources and variable meteorological conditions. In contrast, the E3SMv2 exhibits lower variability, with N_d distributions that are generally narrower, with distinct peaks at low concentrations in all seasons except JJA. The model bias of overproducing frequent low N_d scenarios in the MBL clouds in previous E3SM versions remains in E3SMv2 (Varble et al., 2023; Tang et al., 2023; Kang et al., 2024), possibly related to the model treatment of cloud droplet formation under low turbulence conditions, which is typical in the MBL stratus and stratocumulus (Shan et al., 2024; Wan et al., 2025).

Satellite-derived LWP in Fig. 1e–h displays modest seasonal variations, with the highest mean values observed during DJF (95.38 g m⁻²) and the lowest during JJA (74.46 g m⁻²). The spread of LWP distribution is the greatest in DJF, smallest in JJA, and intermediate in MAM and SON. E3SMv2 simulates the similar seasonal cycle of LWP, with the highest values in DJF (91.52 g m⁻²) and lowest values in JJA (54.31 g m⁻²). The model performs relatively well in reproducing the seasonal mean LWP during DJF and SON, compared to the observations. However, across all seasons, the model may be biased toward producing more frequent low LWP values, suggested by the more positively skewed distributions.

Overall, the observed higher (lower) N_d and lower (higher) LWP during the warm (cold) seasons are consistent with previous studies based on ground-based and satellite remote sensing as well as aircraft in situ measurements (Wu et al., 2020; Varble et al., 2023; Zheng et al., 2024). While the E3SMv2 successfully simulates the broad seasonal variations in both N_d and LWP, its consistent underestimation

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of N_d and limited representation of variability, especially during the warm seasons, indicate that further refinements are needed to better capture the full spectrum of aerosol-cloud interactions and cloud microphysical processes.

3.2 Responses of LWP to N_d

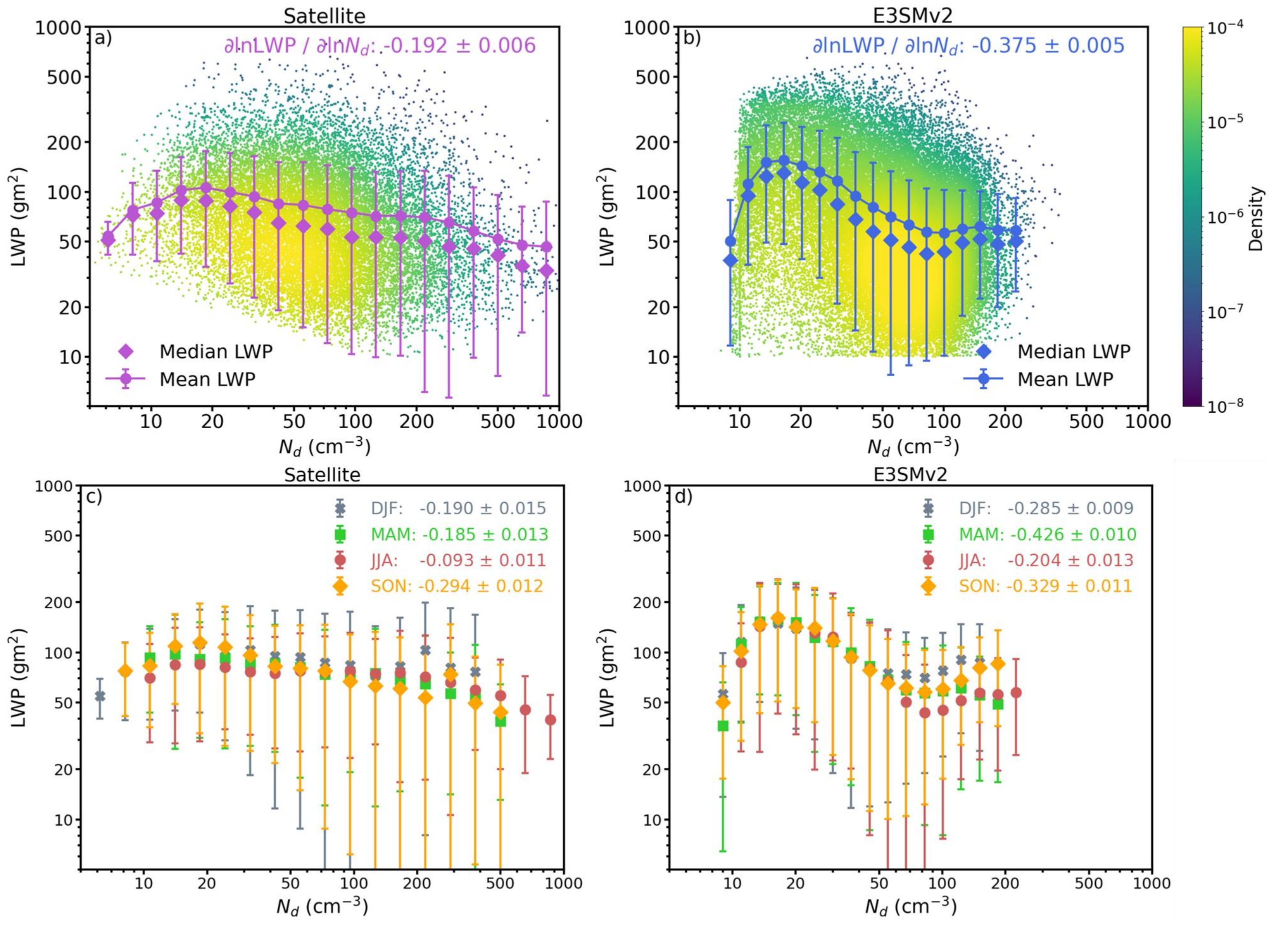

Figure 2. Top Panel: Bulk LWP binned as a function of N_d from a) satellite and b) E3SMv2, the Gaussian kernel density estimate (KDE) is shown as color-shaded scatter points area; Bottom panel: seasonality of the LWP dependences from c) satellite and d) E3SMv2.

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The dependence of LWP on N_d for both satellite data and E3SMv2 simulations, is presented in Figure 2. We quantify the response of LWP to changes in N_d using a LWP adjustment index defined as

$$\mathcal{L}_0 = \frac{\partial \ln(LWP)}{\partial \ln(N_d)}.$$

The bulk LWP adjustment, derived from satellite observations and model simulations, is -0.192 ± 0.006 and -0.375 ± 0.005, respectively, consistent with previous satellite studies over the eastern Atlantic region (Gryspeerdt et al., 2019; Christensen et al., 2023; Zhang et al., 2025). Notably, LWP exhibits a distinct pattern in observations and E3SMv2: it initially increases with N_d and then reverses to a decreasing trend, forming an inverted-V shape with a reversal point near N_d ~20 cm^{-3}. However, while the negative LWP response at high N_d in observations is primarily attributed to entrainment and evaporative processes, the model simulations appear to generate a similar, yet exaggerated, shape predominantly through parameterized precipitation suppression (Mülmenstädt and Feingold et al., 2018; Mülmenstädt et al., 2024b).

In the satellite observations (Fig. 2a), at lower cloud droplet concentrations, with the increased aerosol concentration, cloud droplets become more numerous in smaller sizes. It leads to decrease in efficiency in colliding and coalescing into raindrops and suppress precipitation (Albrecht, 1989). This suppression results in less cloud water being lost through rainfall and, consequently, an increase in LWP. Furthermore, the combined effects of entrainment-sedimentation feedback and precipitation-stabilization may lead to weaker entrainment drying in relatively clean clouds, further enhancing the LWP under lower N_d conditions (Bretherton et al., 2007; Wood, 2012; Possner et al., 2018). In contrast, at higher N_d levels, the increased abundance of small droplets expands the surface area available for evaporation at the cloud top (Ackerman et al., 2004; Gupta et al., 2021; Zhang et al., 2022; Zheng et al., 2023). This enhancement in evaporation promotes cooling and intensifies localized turbulent mixing, which, in turn, facilitates the entrainment of dry air from above the cloud. The subsequent mixing further accelerates the evaporation of cloud droplets, reducing the overall liquid water content and decreasing LWP.

Global climate models such as E3SMv2 typically lack the resolution to explicitly simulate small-scale turbulent mixing and entrainment, instead, relying on bulk parameterizations that tend to overestimate precipitation suppression (Varble et al., 2023; Mülmenstädt et al., 2024a; Y. Zhang et al., 2024). Noticeably, E3SMv2 (Fig. 2b) captures the relationship between LWP versus N_d qualitatively. However, the model systematically produces higher LWP at low N_d, and lower LWP at high N_d than observation, suggesting that E3SMv2 overestimate the sensitivity of LWP to $N_{d,}$. This discrepancy may reflect uncertainties in the parameterization of cloud adjustments, particularly those involving entrainment and aerosol-cloud microphysical interactions in E3SMv2, as discussed further in Section 4.

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The inverted-V shape in the LWP–N_d relationship persists across seasons in both satellite observations and E3SMv2 simulations (Figs. 2c and 2d), although its intensity and turning point vary. Satellite observations (Fig. 2c) generally exhibit a weaker negative slope of LWP with N_d during winter (DJF) and autumn (SON) than in summer (JJA), with transitional behavior in spring (MAM). In contrast, E3SMv2 simulations (Fig. 2d) reveal more pronounced LWP changes with N_d in every season, consistent with the model tendency shown in the bulk relationship. Despite the overall similarity in seasonal patterns, the discrepancies between the model and observations become more apparent when the data are stratified by season.

3.3 Vertical distribution of aerosol extinction coefficient

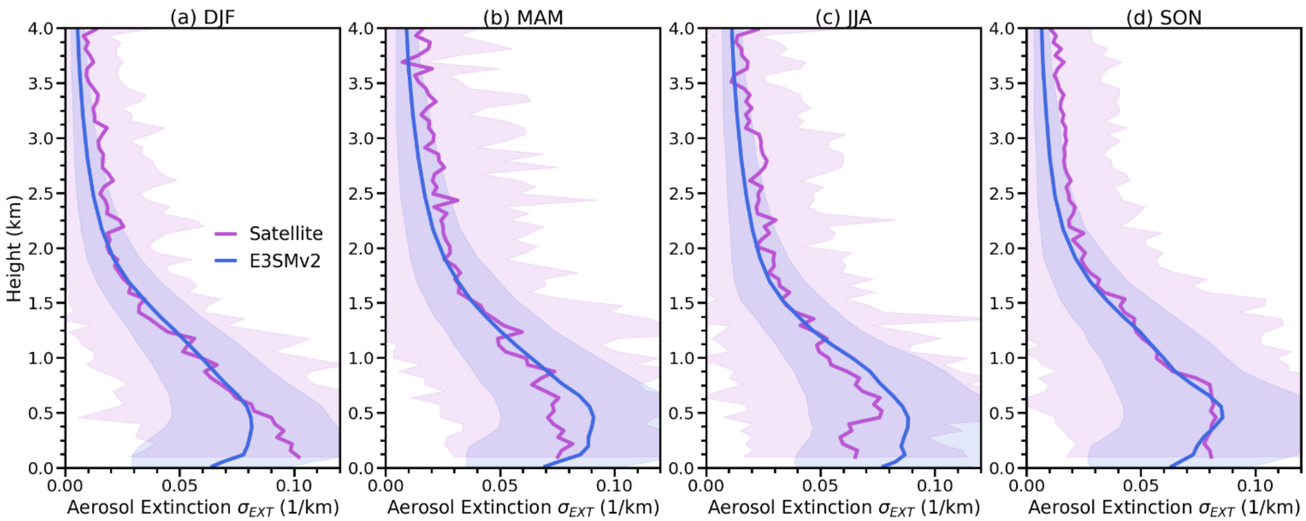

Figure 3. Domain averaged vertical distribution of aerosol extinction coefficients (σ_{EXT}) in the presence of clouds from Satellite (purple) and E3SMv2 (blue) during a) winter, DJF; b) spring, MAM; c) summer, JJA, and d) fall, SON.

The observed seasonal variations in vertically resolved aerosol extinction profiles over the ENA are presented in Figure 3. Both satellite observations and the E3SMv2 model display a characteristic decrease in aerosol extinction coefficient with altitude, with maximum values near the surface (i.e., below approximately 1 km) and a rapid decline above around 2 km. This steep gradient suggests that the

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aerosols are more concentrated within the MBL, consistent with surface aerosol sources such as the oxidation of dimethyl sulfide (DMS) and sea spray aerosols (Zheng et al., 2018; Wang et al., 2021; Ghate et al., 2023). Also, the relatively higher relative humidity within the MBL might also impacts the optical properties of aerosols (Baynard et al., 2006; Feng et al., 2016).

In winter (DJF) and fall (SON), the model tends to underestimate the aerosol extinction coefficient (σ_{EXT}) below 1 km compared to satellite observations, while overestimating the near-surface extinction during spring and summer. Recent studies have identified several factors that may explain these seasonal biases. For example, Logan et al. (2014) and Zheng et al., (2018) demonstrated that MBL aerosol properties are highly sensitive to local meteorological conditions and long-range transport events. During the cold seasons (DJF and SON), the ENA region experiences high wind speeds in the MBL due to an intensified pressure gradient between the Icelandic Low and the Azores High (Logan et al., 2014; Ghate et al., 2021). The observed high σ_{EXT} reflects the contribution from coarse-mode aerosols, including enhanced sea salt emissions under high wind conditions and long-range transported dust aerosols associated with increased extratropical cyclone activity (Logan et al., 2014; Gläser et al., 2015; Wood et al., 2015). In this context, the underestimation of σ_{EXT} by E3SMv2 may result from underpredicted sea spray aerosols and long-range transported dust (Burrows et al., 2020; Wang et al., 2020; Feng et al., 2022; Qin et al., 2024).

During spring and summer (MAM and JJA), the ENA is characterized by enhanced formation of secondary organic aerosols (SOA) and DMS-derived sulfate (Zheng et al., 2018; Sanchez et al., 2018), dominated by fine-mode aerosols. Sea salt concentrations, which are usually higher in winter, contribute less during the warm months owing to lower wind speeds. Additionally, as the development of the Azores High in summer inhibits large-scale dust transport to the ENA, thereby reducing contributions from coarse-mode aerosols (Wang et al., 2021). Conversely, the overestimation of σ_{EXT} by E3SMv2 may be partially attributed to the overproduced sulfate and organic matter at the surface (Hassan et al., 2024; Huang et al., 2024). On the other hand, the aerosol retrievals from CALIPSO have certain limitations at very low aerosol conditions (Painemal et al., 2019), which might contribute to the model-data differences in the free troposphere. In general, the E3SMv2 well captures the observed seasonal variations in aerosol vertical profile and mean extinctions in the MBL compared to the satellite observations, which allows us to further examine the simulated cloud responses to aerosol changes

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3.3 N_d susceptibility to aerosol extinction coefficient

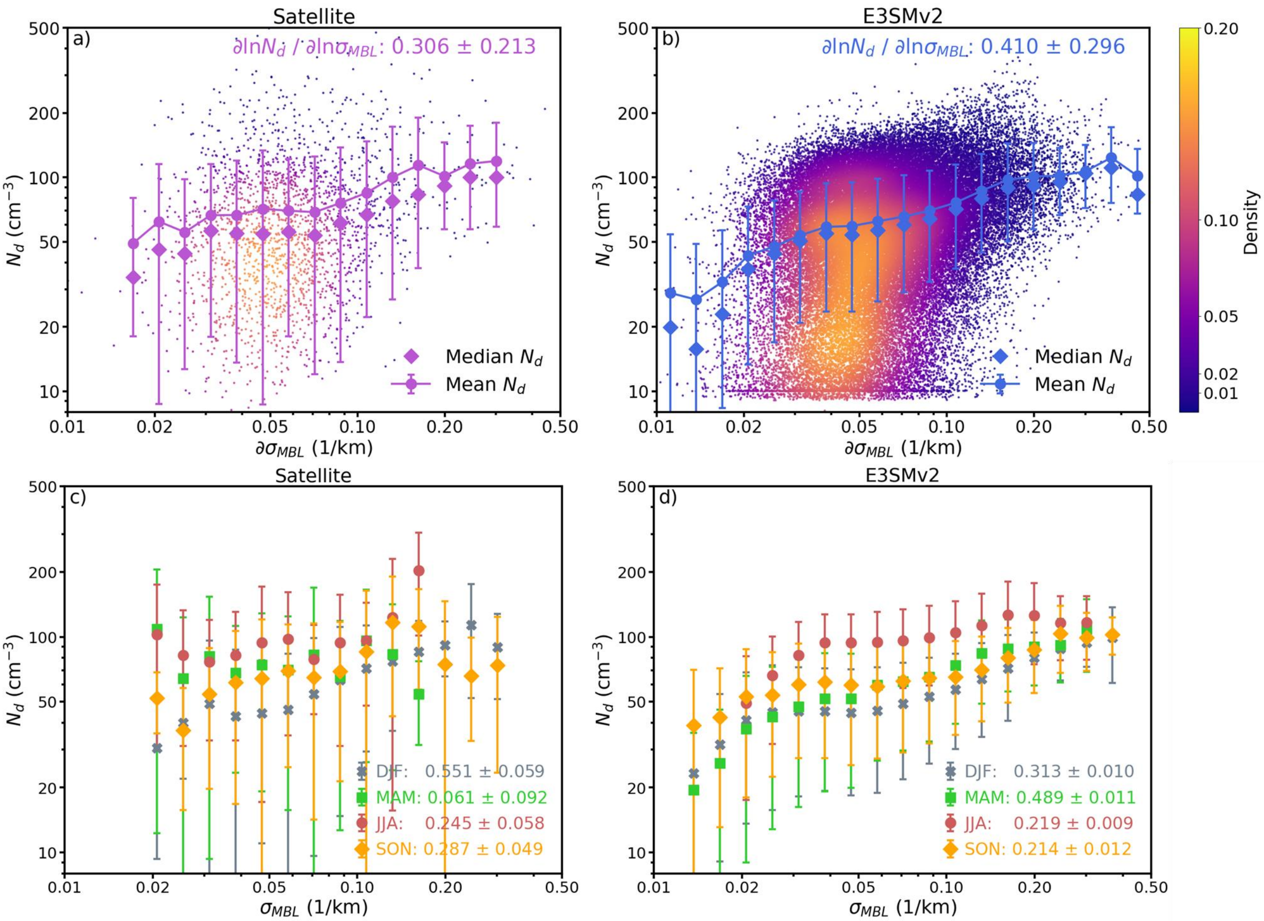

Figure 4. Top Panel: N_d binned as a function of σ_{MBL} from a) satellites and b) E3SMv2. The Gaussian kernel density estimate (KDE) is shown as color-shaded scatter points area, the mean N_d values in σ_{MBL} bins are shown as solid-dotted line, and the median N_d is denoted by the diamond. Bottom panel: seasonality of the N_d dependences from c) satellite and d) E3SMv2, colored symbols denote mean N_d in σ_{MBL} bins, and whiskers denote standard deviations.

To quantify the aerosol impact on cloud microphysics, we define an aerosol-cloud interaction (ACI) index as:

$$ACI_N = \frac{\partial \ln(N_d)}{\partial \ln(\sigma_{MBL})},$$

where the σ_{MBL} denotes the mean value of the below-cloud-top σ_{EXT}. This parameter represents the sensitivity of N_d to changes in aerosols within the marine boundary layer (MBL). Although a robust

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ACI assessment can be performed using aerosol information near the cloud base such as from in situ aircraft measurements (Gupta et al., 2021; Zheng et al., 2022a), limitations in satellite retrievals of accurate cloud base height and the challenge for models to pinpoint the cloud base from coarse vertical resolution. Moreover, the coarse vertical resolution of the model makes it difficult to vertically collocate CALIOP aerosol layer and the model output. Hence, those reasons necessitate the use of the mean aerosol properties within the below-cloud-top MBL in the present study. This approach facilitates a more consistent comparative assessment of aerosols between satellite observations and model simulations.

The top row of Figure 4 compares the satellite-derived and E3SMv2-simulated relationships between N_d and σ_{MBL}. Satellite observations (Fig. 4a) show that N_d generally increases with σ_{MBL}; however, the sensitivity is moderate with considerable scatter ($ACI_N = 0.306 \pm 0.213$). This susceptibility falls within the range reported in previous satellite studies (Quaas et al., 2020; Jia et al., 2022), although it is relatively moderate compared to those analyses based on alternative observational platforms such as ground-based observations and in-situ measurements (McComiskey et al., 2009; Zheng et al., 2024). Nonetheless, the positive ACI_N indicates that, on average, higher aerosol concentrations are associated with increased cloud droplet numbers, consistent with the well-known Twomey effect.

In contrast, the E3SMv2 model (Fig. 4b) exhibits a stronger dependence of N_d on σ_{MBL}, with a noticeably higher slope. Although the model reproduces the qualitative relationship, its steeper slope suggests that the microphysical or aerosol activation schemes in E3SMv2 may be overly sensitive, as also indicated by previous studies (Christensen et al., 2023; Varble et al., 2023). In other words, for the same fractional change in σ_{MBL}, E3SMv2 predicts a larger fractional change in N_d compared to satellite data. This model–observation discrepancy may reflect uncertainties in how E3SMv2 parameterizes aerosol activation, updraft velocities at the cloud base, or boundary-layer processes such as entrainment and mixing (Tang et al., 2024; Wan et al., 2025), as discussed further in Section 4.

In terms of seasonal variations in N_d susceptibility, the ACI_N derived from satellite data (Fig. 4c) reveals large differences across seasons: winter (DJF) shows a markedly stronger response (~0.55) compared to spring (MAM) at (~0.06). Summer (JJA) and fall (SON) display intermediate ACI_N values. In contrast, the E3SMv2 results (Fig. 4d) also exhibit a clear positive relationship in every season, but associated with a narrower range of ACI_N values. Notably, the simulated springtime (MAM) ACI_N (~0.49) is considerably stronger than that observed from satellites, whereas a weaker slope is simulated in winter. In summer (JJA) and fall (SON), the slopes are closer to satellite-derived values, yet E3SMv2 tends to exhibit a higher and more consistent responsiveness of N_d to aerosol increases.

Overall, the seasonal assessments suggest that E3SMv2 consistently yields both stronger LWP responses to N_d and enhanced N_d responses to aerosols compared to satellite-derived relationships. A

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notable limitation of this seasonal grouping is that it fails to disentangle the complexities of aerosol-cloud interactions from characteristic meteorological variations. Aggregating data into seasons makes it challenging to unambiguously attribute changes in cloud properties to aerosol variations rather than to shifts in large-scale dynamics or thermodynamic conditions (Zheng et al., 2022b; Zhang et al., 2023). This limitation necessitates the regime-based analysis in Section 4, in which samples are clustered according to dominant meteorological regimes; an approach that can more effectively isolate microphysical processes from the confounding effects of synoptic-scale and seasonal variability (Mülmenstädt et al., 2012; Mechem et al., 2018; Zheng et al., 2025).

4 Regime-based Analysis of Aerosol, cloud properties, and their interactions

4.1 Distinctive Meteorological Regimes over ENA

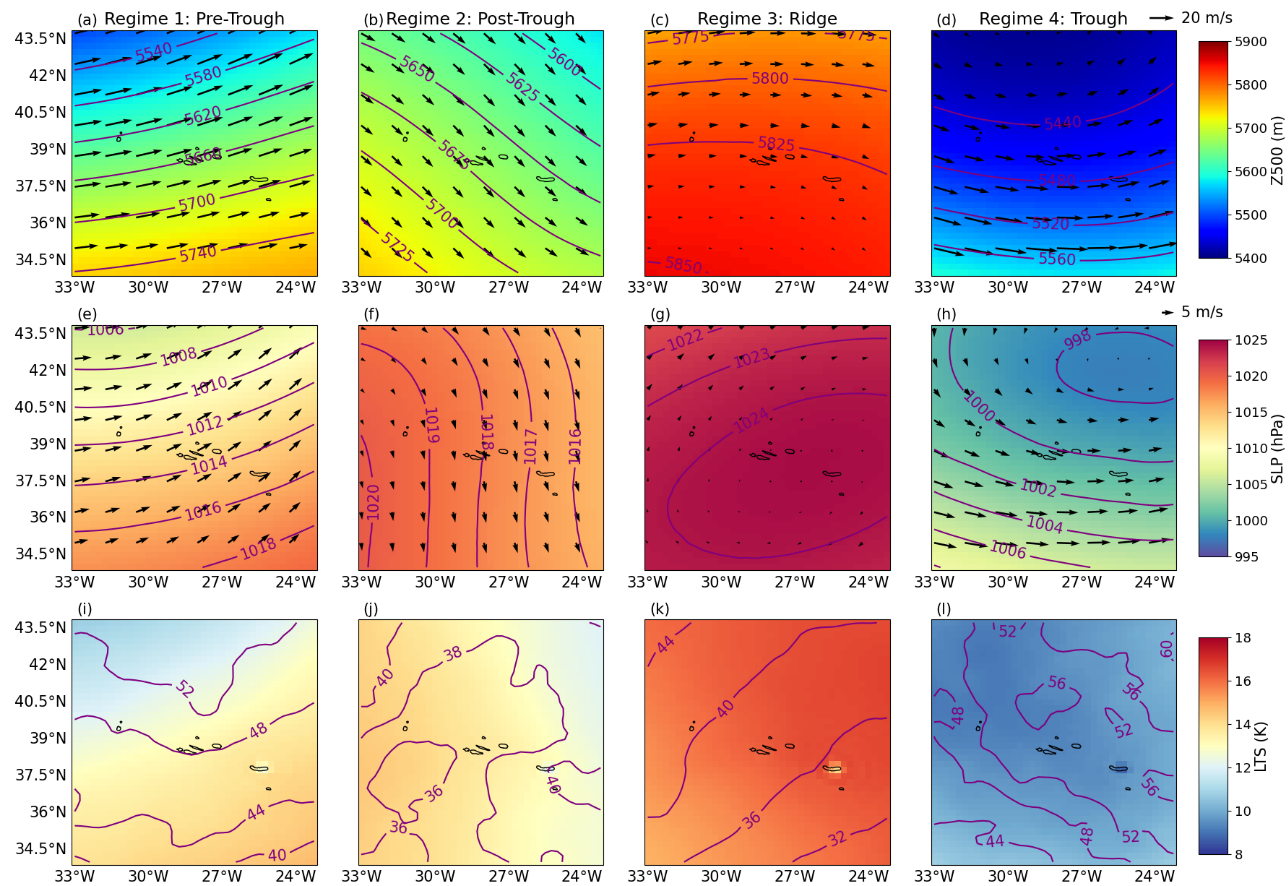

Figure 5. Meteorological composites for each synoptic regime classified by the clustering model: Regime 1 (a, e, i), Regime 2 (b, f, j), Regime 3 (c, g, k), and Regime 4 (d, h, l). The first row shows 500 hPa geopotential height (Z500) shaded and contoured, with 500 hPa wind vectors overlaid. The

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second row presents sea level pressure (SLP) in both shaded and contoured formats, together with 10 m surface wind vectors. The third row displays lower tropospheric stability (LTS) at 700 hPa as the shaded field and 700 hPa relative humidity (RH700) in contours.

Large-scale meteorological fields, specifically, the 500 hPa geopotential height, 10 m horizontal winds, and sea level pressure at 13:00 LT (1 p.m. local time) from the ERA5 reanalysis, are used as inputs to a deep-learning-based clustering model. The determination of cluster numbers is based on a combination of silhouette score analysis (i.e., measures of cluster cohesion and separation for different cluster numbers) and the sensitivity of aerosol and cloud property distinctions to the chosen number of clusters. A total of 3,286 daily samples spanning 2004–2016, representing the afternoon (13:00 LT) meteorology over the ENA, were used in the clustering. This approach results in the identification of four distinct synoptic-scale regimes (Figure 5).

Regime 1 (Figs. 5a, 5e, and 5i) represents the Pre-Trough phase, characterized by a developing trough approaching the Azores. This regime features strong southwesterly winds at both 500 hPa and near the surface, moderate mid-level moisture, and low lower tropospheric stability (LTS). Such conditions typically precede frontal development and passage and are associated with the early stages of midlatitude cyclone progression, therefore resulting in a wet free troposphere and low LTS (Mechem et al., 2018; Zheng et al., 2025).

Regime 2 denotes the typical Post-Trough condition that follows the passage of a trough, characterized by prevailing northwesterly winds (Fig. 5b) and transitional stability. The SLP field reveals a relatively weak pressure gradient, corresponding to a post-frontal environment in which drier and colder air is advected into the region (Figs. 5f and 5j). Although LTS remains moderate, it is slightly higher than in Regime 1, reflecting the gradual stabilization behind the frontal system. Taken together, Regimes 1 and 2 depict the typical meteorological evolution associated with mid-latitude cyclones traversing the ENA, which occur regularly throughout the year (Table S1), particularly during colder seasons when mid-latitude cyclone activity is more frequent (Wood et al., 2015; Mechem et al., 2018).

Regime 3 corresponds to the Ridge phase, in which a pronounced ridge dominates the region. Both the SLP and 500 hPa geopotential height (Z500) fields constitute a broad anticyclonic pattern, with relatively weak and variable winds from the surface up to 500 hPa. This pattern coincides with the driest free troposphere and the most stable lower troposphere observed among all regimes, which generally favor a more coupled and shallower (MBL) (Carrillo et al., 2015; Zheng et al., 2025). As indicated in Table S1, Regime 3 is the most frequently occurring regime (63.8%) in the region, peaking during the summer months when the center of the Azores High predominantly lies to the southwest of the Azores

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Islands. This finding is consistent with previous studies (Mechem et al., 2018; Wang et al., 2022), and the synoptic categorization is similar to that in Painemal et al. (2023) for the Western North Atlantic.

Regime 4 represents a typical Trough phase, characterized by a canonical 500 hPa trough with stronger cyclonic flow at the surface. The lower troposphere exhibits reduced static stability, while the contours of relative humidity at 700 hPa (RH700, Fig. 4l) indicate a more humid troposphere. These conditions imply enhanced ascent and moist processes. Among the four regimes, Regime 4 is the least frequent (3.4%) and is largely confined to the colder seasons (winter and spring), confirming the findings from previous studies (Wood et al., 2015; Mechem et al., 2018; Wang et al., 2022).

4.2 Cloud properties under different regimes

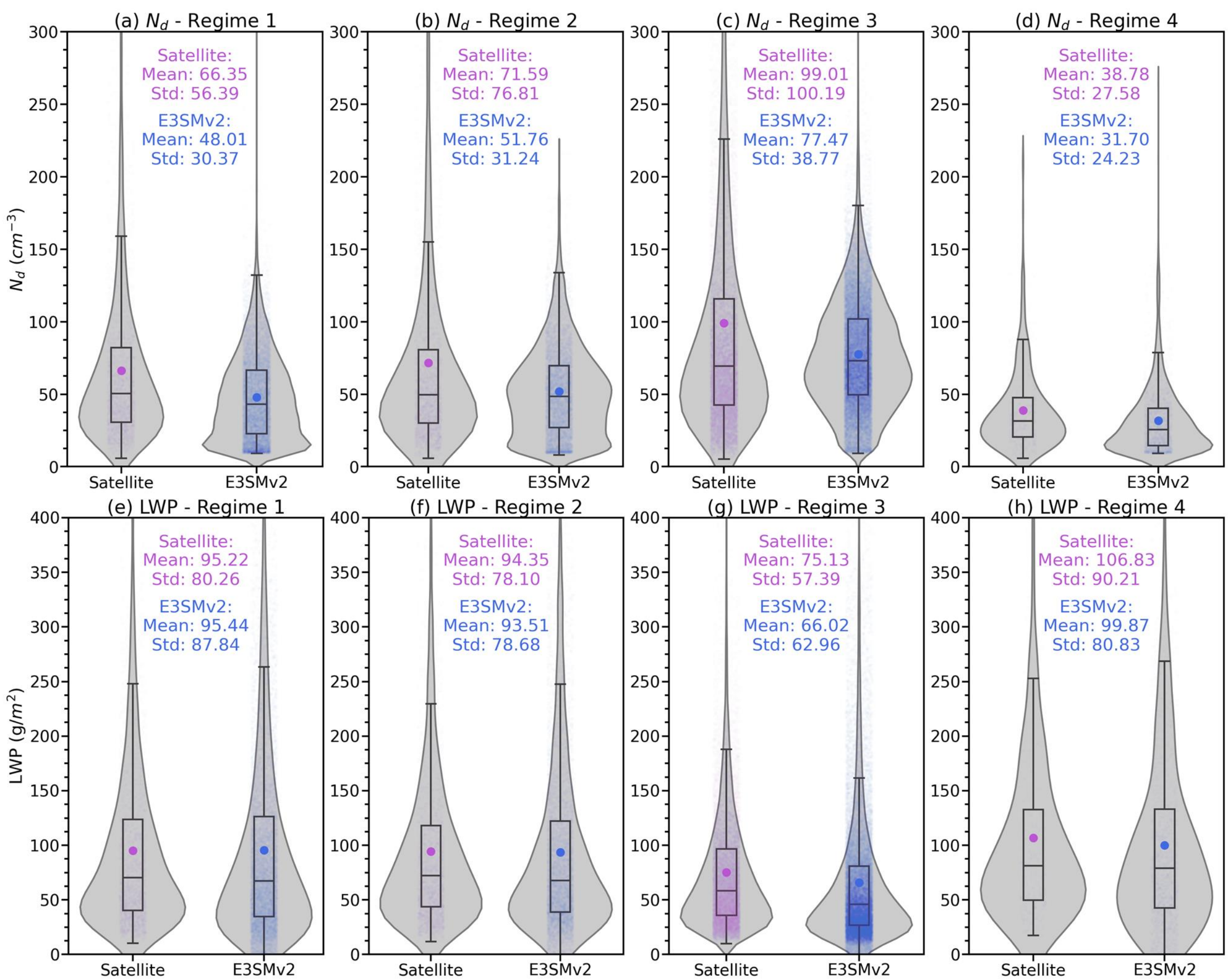

Figure 6. Violin plots of cloud droplet number concentration (N_d, top panels) and cloud liquid water path (LWP, bottom panels) from satellite retrievals (purple) and E3SMv2 simulations (blue), grouped by

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Regime 1 (a, e), Regime 2 (b, f), Regime 3 (c, g), and Regime 4 (d, h). The mean value is indicated by the color-coded dot. The smoothed shape of each violin shows the Gaussian kernel density estimate (KDE). From top to bottom within each violin, the box plot lines represent the third quartile (Q3, 75th percentile), median (Q2, 50th percentile), and first quartile (Q1, 25th percentile), respectively. The upper whisker extends to Q3 + 1.5 × IQR (IQR: interquartile range), and the lower whisker extends to Q1 − 1.5 × IQR.

In order to provide synergy on the meteorology and cloud for reference in this study, a brief and qualitative summary of meteorology patterns and cloud and precipitation status are listed in Table 1. Furthermore, based on the four distinct meteorological regimes identified through clustering, we stratify cloud properties from both satellite retrievals and E3SMv2 simulations. Both N_d and LWP exhibit systematic regime-dependent behavior and model biases (Fig. 6). And the detailed quantities listed in Table 2.

Across all regimes, E3SMv2 tends to underestimate N_d while representing LWP more accurately. A rigorous comparison of surface precipitation rates between satellite data and E3SMv2 is limited in this study due to lack of collocated precipitation rate measurements from the satellite. Therefore, we choose to compare the in-cloud fractional occurrences of rain and rain LWP from E3SMv2 with the fractional occurrences of drizzle, light rain, and rain from CloudSat (Table S2). In addition, CTH statistics, stratified by regime from both satellite and E3SMv2 datasets, are presented in Table 2, with composite maps shown in Figures S1 and S2. To quantify the variability of CTH under different regimes, Moran's I indices, a measure of spatial autocorrelation, were computed and then normalized for each regime to account for the differences in spatial resolution between the satellite and E3SMv2 datasets. A normalized Moran's I index of 0 means the least autocorrelated, and 1 means the most autocorrelated. The normalized Moran's I indices for the satellite (E3SMv2) datasets are 0.61 (0.0), 0.64 (0.89), 1.0 (1.0), and 0.0 (0.13) for Regimes 1, 2, 3, and 4, respectively.

Regime 1, characterized by an approaching trough with southwesterly flow, moderate moisture, and relatively weak subsidence, exhibits a mean N_d of 66.35 cm^{-3} in satellite observations, whereas E3SMv2 simulates a lower mean N_d of 48.01 cm^{-3} with reduced variability. The mean LWP in E3SMv2 (95.44 g m^{-2}) is similar to that of the satellite retrievals (95.22 g m^{-2}), with a slightly broader distribution. These findings suggest that while E3SMv2 captures the overall liquid water content in cloud layers under pre-frontal conditions, it systematically underestimates N_d. Moreover, the CTH in Regime 1 remains low (Table 2), reflecting a shallower MBL in the transitional environment of the approaching front (Jeong et

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al., 2022). Precipitation statistics indicate moderate fractions of drizzle and light rain (Table S2), consistent with the notion that although pre-trough instability favors drizzle production, the boundary layer does not fully deepen to support frequent or intense rainfall (Wood, 2005; Wu et al., 2020; Zheng et al., 2022). The partial uplift and moderate moisture convergence can occasionally enhance cloud thickness, as indicated by a moderately high normalized Moran's I index (0.61), yet the overall shallower structure typically limits heavier precipitation.

In the post-trough environment of Regime 2, the northwesterly flow advects drier and cooler air behind the frontal system. Such conditions are typically associated with increased subsidence and a deeper MBL conducive to the development of stratocumulus clouds (Wu et al., 2020; Jensen et al., 2021; Jeong et al., 2022). Consequently, satellite-derived CTH values (Table 2 and Fig. S1b) are higher than those in Regime 1, indicating a deeper MBL. Furthermore, clouds in Regime 2 are also associated with the highest fractional occurrences of drizzle among the four regimes (Table S2). Drizzle formation and turbulent mixing can lead to heterogeneous cloud structures with less cloud adiabaticity (Wu et al., 2020), as reflected by a normalized Moran's I index of 0.64 and in Figure S1b, in contrast to the more uniform CTH variation in Regime 3. E3SMv2 captures the relative CTH variation and precipitation frequency reasonably well under this regime. As for Regime 1, E3SMv2 still systematically underestimates N_d (51.76 cm^{-3} vs. 71.59 cm^{-3}) while simulating the observed mean LWP well (93.51 vs. 94.35 g m^{-2}).

Regime 3 features with a pronounced ridge, reduced convective activity, and generally lower moisture in the boundary layer. Satellite retrievals show the highest mean N_d (99.01 cm^{-3}, Fig. 6c), which could be attributed to the less active drizzle processes and droplet evaporation in shallower cloud deck within a more stable MBL (Wood et al., 2012; Zheng et al., 2023). In contrast, E3SMv2 simulates a lower mean N_d (77.47 cm^{-3}) and fails to capture the broad observed distribution. Moreover, LWP in this ridge regime is the lowest among all regimes, with E3SMv2 underestimating satellite observations (66.02 vs. 75.13 g m^{-2}; Fig. 6g). From both satellite and model perspectives, the cloud field is characterized by shallower, more homogeneous decks that span large horizontal areas yet produce only light or sporadic drizzle, as reflected by lower CTH (Figs. S1c and S2c) and the lowest precipitation fractions among the four regimes (Table S2). Overall, the results in Regime 3 align with the signature of shallow stratus and stratocumulus clouds (Rémillard and Tselioudis, 2015; Mechem et al., 2018; Wu et al., 2020; Jensen et al., 2021).

Under a well-developed trough in Regime 4, satellite observations record the lowest N_d (38.78 cm^{-3}) but the highest LWP (106.83 g m^{-2}) among four regimes, reflecting the prevalence of deep and warm-rain-active cloud systems formed by strong uplift and abundant moisture. These conditions yield the highest and most variable CTH among the four regimes (Table 2), along with frequent

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precipitation in the form of drizzle or rain. Stronger vertical motion promotes the development of deeper clouds with higher rainfall efficiency, contributing to spatially heterogeneous precipitating cloud fields observed in both the satellite data and model simulations (Figs. S1d and S2d; Table S2). Notably, E3SMv2 simulates the high LWP (99.87 g m^{-2}) and elevated liquid water content as in the observations, but underestimates variability. Similarly, the model underestimates N_d (31.70 cm^{-3}) relative to satellite observations as in the other regimes.

Overall, these results demonstrate the clear meteorological impact on both cloud microphysical (N_d) and macrophysical (LWP) properties. Pre- and post-trough conditions (Regimes 1 and 2) favor moderate N_d and LWP, while ridge-dominated conditions (Regime 3) promote stable, stratiform-dominated environments with high mean N_d but relatively low LWP. In contrast, developed troughs (Regime 4) yield lower N_d yet substantially higher LWP in more vertically developed cloud systems. Although E3SMv2 captures the LWP mean and distributions across these synoptic regimes, it systematically underestimates N_d. This discrepancy suggests that the challenges in simulating cloud droplets are irrespective of the meteorological influences.

Table 1. Summary of meteorological and cloud categories in different regimes.

Regimes	Meteorological Patterns	Cloud Status
R1 Pre-Trough	Approaching trough; strong SW winds at 500 hPa & surface; moderate mid-level moisture; low LTS	Lower CTH; moderate drizzle / light rain
R2 Post-Trough	Post-frontal NW winds; moderate LTS; weak pressure gradient; drier, free troposphere	Higher CTH; high drizzle fraction but low overall precipitation
R3 Ridge	Broad anticyclonic ridge; high-pressure-dominated surface; driest free troposphere; highest LTS	Shallow MBL; Lowest CTH; minimal precipitation
R4 Trough	Canonical trough; strong cyclonic surface flow; lowest LTS; moist free troposphere	Highest & most variable CTH; most frequent drizzle / rain

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Table 2. Regime-based aerosol and cloud variables from Satellite and E3SMv2

	Regime 1	**Regime 2**	**Regime 3**	**Regime 4**
Satellite				
$N_d\ (cm^{-3})$	66.35 ± 56.39	71.59 ± 76.81	99.01 ± 100.19	38.78 ± 27.58
$LWP\ (gm^2)$	95.22 ± 80.26	94.35 ± 78.10	75.13 ± 57.39	106.83 ± 90.21
$CTH\ (km)$	1.59 ± 0.46	1.76 ± 0.42	1.46 ± 0.50	1.70 ± 0.45
$\sigma_{MBL}\ (1/km)$	0.075 ± 0.047	0.070 ± 0.046	0.063 ± 0.039	0.067 ± 0.048
E3SMv2				
$N_d\ (cm^{-3})$	48.01 ± 30.37	51.76 ± 31.24	77.47 ± 38.77	31.70 ± 24.23
$LWP\ (gm^2)$	95.44 ± 87.84	93.51 ± 78.68	66.02 ± 62.96	99.87 ± 80.83
$CTH\ (km)$	1.62 ± 0.84	1.76 ± 0.68	1.41 ± 0.59	1.81 ± 0.88
$\sigma_{MBL}\ (1/km)$	0.070 ± 0.038	0.061 ± 0.033	0.071 ± 0.041	0.063 ± 0.032

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4.3 LWP-N_d relationships under different regimes

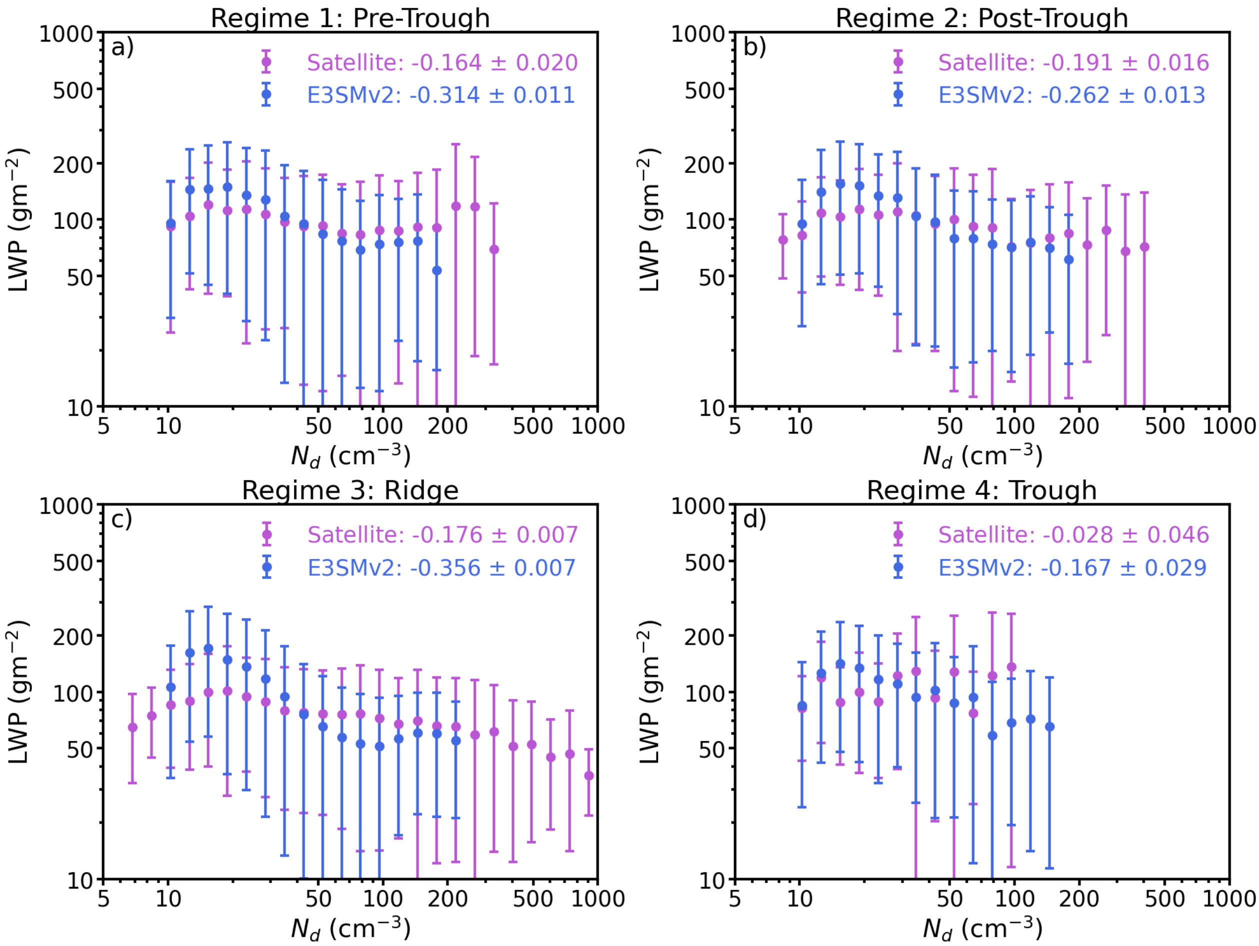

Figure 7. LWP responses on N_d from satellite (purple) and E3SMv2 (blue) for a) Regime 1, b) Regime 2, c) Regime 3, and d) Regime 4. Colored dots denote mean LWP in N_d bins, and whiskers denote standard deviations. The quantitative LWP adjustment index ($\mathcal{L}_0$) is denoted in the legend.

The relationship between LWP and N_d across the four synoptic regimes in both satellite retrievals and the E3SMv2 model exhibits the characteristic inverted-V shape (Fig. 7), as shown in the seasonal assessment in Section 3. Furthermore, regime-specific meteorological differences, particularly variations in stability and moisture transport, strongly influence the shape and peak of the LWP–N_d relationship. The quantitative LWP adjustment ($\mathcal{L}_0$) values are listed in Table 3.

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Noticeably, satellite retrievals in the Ridge regime (R3) display a more pronounced inverted-V shape compared to themselves in other regimes, with LWP consistently declining as N_d increases at high values (Fig. 7c). Under Ridge conditions, strong subsidence limits vertical growth, reduces moisture convergence, and produces a shallower, more stable cloud layer. Although the mixing rate from entraining drier free-tropospheric air might not be as intense as in thicker and more turbulent clouds, the drying effect propagates more efficiently through the thinner cloud layer, exerting a significant impact (Sanchez et al., 2020; Chun et al., 2023). Furthermore, since the Ridge regime features more high-N_d conditions, the resulting smaller cloud droplets are more susceptible to entrainment-evaporation, leading to a more efficient removal of cloud water (Possner et al., 2020). In contrast, the E3SMv2 model significantly overestimates the maximum LWP, and yields a significantly greater LWP decline with N_d ($\mathcal{L}_0$ values of -0.356 for model vs. -0.176 for satellite).

Under Regime 1 (Pre-Trough, Fig. 7a), the satellite and model data show a rise in LWP as N_d increases from very low values, while the characteristic inverted-V shape is partially masked by increased scatter in the satellite-retrieved LWP at high N_d, likely due to the complexity of drizzle, entrainment, and mixing processes in the pre-frontal environment. The enhanced drizzle formation, indicated by the relatively higher drizzle and rain fractions in clouds, can reduce the temperature gradient near cloud-top via condensation warming, stabilizing the cloud layer and partially offsetting the entrainment cooling and retaining the LWP. Similar to those in Regime 3, the model also exhibits a nearly double $\mathcal{L}_0$ (-0.314) compared to satellite (-0.164). One possible explanation is that the model microphysics suppresses drizzle too aggressively when N_d increases occur at lower values (Varble et al., 2023; Mülmenstädt et al., 2024b), thereby promoting the accumulation of cloud liquid water prior to the onset of entrainment-drying or precipitation. Furthermore, the MG2 microphysics scheme may trigger the entrainment–evaporation feedback too rapidly in the model, resulting in an LWP adjustment timescale much shorter than observed (Zhou et al., 2025). As a result, the model depletes cloud liquid water too quickly (Xie et al., 2018; Rasch et al., 2019). These combined model uncertainties lead to an excessively steeper $\mathcal{L}_0$ compared to satellite observations.

In the Post-Trough regime (Regime 2; Fig. 7b), the residual dynamic forcing from the trough coupled with the onset of entrainment-induced evaporation, produces an intermediate LWP–N_d sensitivity, particularly under the high N_d condition, between the Pre-Trough and Ridge regimes. On the one hand, in the Post-Trough environment, the cloud field is more variable and relatively thicker, which is found to be favorable for stronger entrainment rates (Wood, 2007; Wu et al., 2020; Chun et al., 2023), hence depleting LWP. On the other hand, this effect might be closely intermingled with patches of high LWP sustained by the precipitation-stabilization effect (Possner et al., 2020; Wu et al., 2020). These

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competing effects result in an averaged LWP decline with increasing N_d that is stronger than in the Pre-Trough state but remains less pronounced than the sharp decrease observed in the Ridge regime. Compared to the satellite observations, E3SMv2 underestimates LWP at high N_d, coupled with the slightly steeper LWP–N_d slope. The model bias likely results from parameterized entrainment and in-cloud mixing (Y. Zhang et al., 2023), which may intensify cloud-top evaporation cooling and promote LWP loss; in contrast, real clouds exhibit more heterogeneous mixing that better preserves liquid water at high N_d.

In Regime 4 (Fig. 7d), characterized by a well-developed trough with enhanced ascent and high moisture availability, clouds deepen, and LWP peaks at relatively high N_d, as observed by satellites. The muted LWP decrease seen in satellite data may be largely a consequence of combined factors. Since the satellite N_d is derived assuming an adiabatic vertical profile and a constant N_d throughout the cloud, the expected increase in subadiabaticity for Regime 4, as suggested by the enhanced precipitation, might induce retrieval bias that dampens the apparent sensitivity of LWP to N_d (Grosvenor et al., 2018). At the same time, retrievals tend to average over heterogeneous cloud fields in the Trough regime, where cloud properties vary on small scales, further smoothing out the true microphysical sensitivity (Gryspeerdt et al., 2022). Excluding the retrieval-induced bias, strong updrafts in this regime can promote moisture convergence, which in turn maintain or even enhance LWP despite higher N_d (Goren et al., 2018; Zhang et al., 2022; Painemal et al., 2023). In some cases, heavy precipitation may also drive locally precipitation-generated cold pooling that enhances the cloud base updraft that helps maintain LWP following precipitation. Such muted LWP response to N_d, has been reported for the stratocumulus clouds over the southeast Pacific, particularly in thickening stratocumulus clouds with a higher likelihood of producing more intense precipitation (Smalley et al., 2024), suggesting the precipitation-suppressing overwhelms the entrainment-drying effect on LWP. In contrast, E3SMv2 maintains an evident inverted-V shape of LWP–N_d curve as in other regimes, albeit with a smaller $\mathcal{L}_0$ (-0.167), resulting from the explicitly parameterized microphysical processes.

Overall, the model simulations exhibit an excessively sharp rise-and-fall pattern in LWP, producing an exaggerated inverted-V in the LWP–N_d relationship, compared to the more subtle shapes in satellite results. This suggests that microphysical feedback to cloud water may be triggered too early and too rapidly in the model. Such behavior may originate from the MG2 microphysics scheme's nonlinear autoconversion rate (Gettelman and Morrison, 2015), which acts to suppress drizzle too aggressively at low N_d, thereby retaining excess LWP, but truncates liquid water accumulation as N_d increases (Wang et al., 2023; Ovchinnikov et al., 2024). These limitations hinder E3SMv2's ability to capture observed cloud feedback, particularly in regimes with shallower clouds (e.g., Pre-Trough and

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581 Ridge). Addressing these issues may require recalibrating autoconversion rates using observational constraints (e.g., ARM data) and improving scale-aware entrainment schemes that better differentiate mixing regimes.

584 **4.4 N_d susceptibility to aerosols under different regimes**

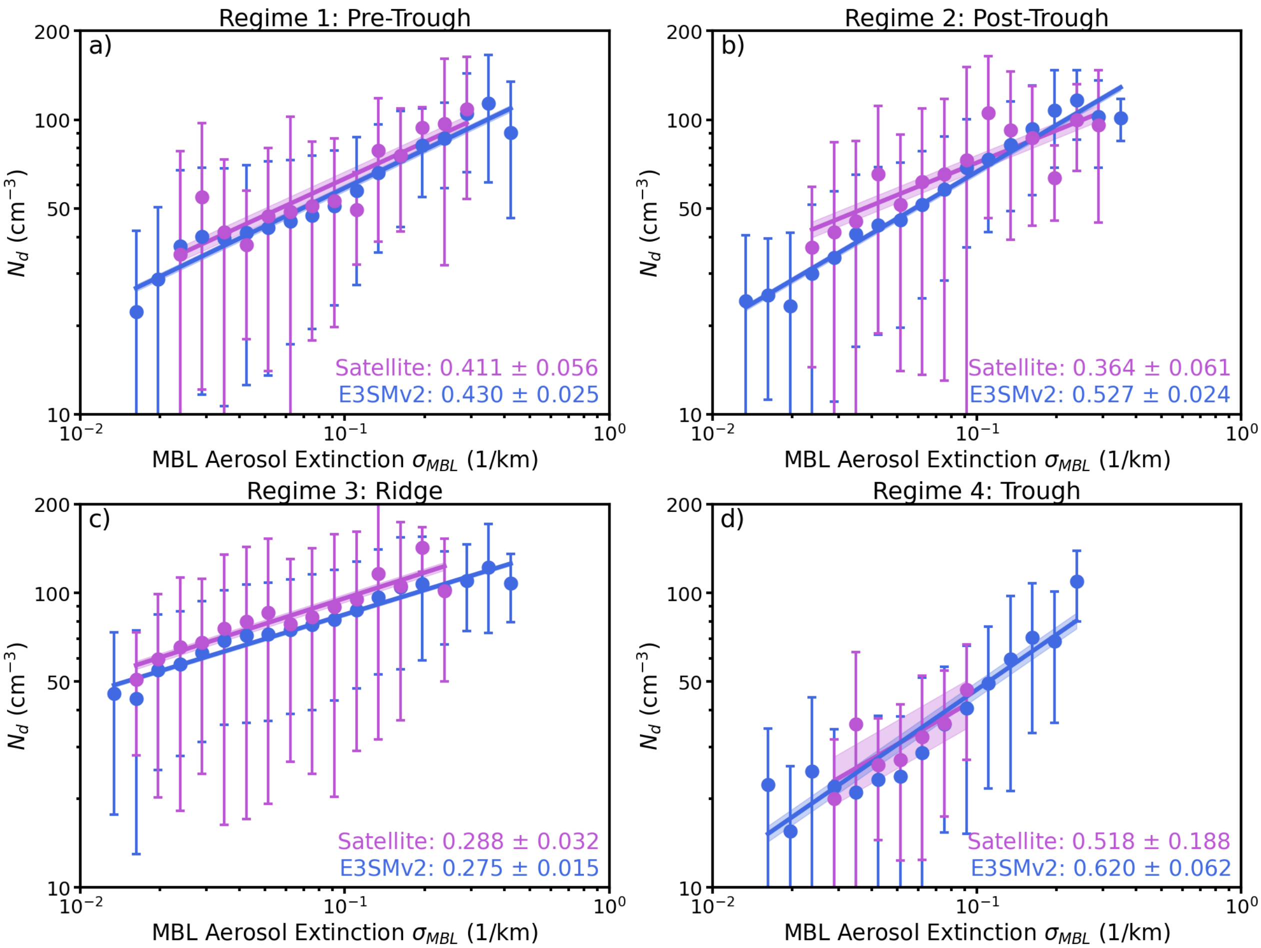

Figure 8. Cloud droplet number concentrations (N_d) dependence on the mean MBL aerosol extinction coefficient (σ_{MBL}) from satellite retrievals (purple) and E3SMv2 simulation (blue), under a) Regime 1; b) Regime 2; c) Regime 3 and d) Regime 4. The solid line indicates the regression line, and the N_d susceptibility ($ACI_N = \partial \ln(N_d)/\ln(\partial\sigma_{MBL})$) is listed in the legend.

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Figure 8 illustrates how N_d responds to changes in MBL aerosol extinction (σ_{MBL}) across each meteorological regime, as quantified by the aerosol–cloud interaction index (ACI_N). Under all regimes, both observations and E3SMv2 simulations show that N_d generally increases with increasing σ_{MBL}, reflecting the typical ACI in which cloud droplet concentration rises with increased aerosol loading. And it is worth noting that the model is able to simulate the quantitative relationship between N_d and σ_{MBL}, with exception of Regime 2. The ACI_N values are listed in Table 3. Since each regime is characterized by distinct large-scale meteorological conditions, these environmental factors influence the ACI remarkably, as the aerosol activation is highly influenced by updraft strength, moisture availability, and in-cloud supersaturation (Chen et al., 2011; Kirschler et al., 2022; Zheng et al., 2024).

Both satellite retrievals and E3SMv2 yield the lowest N_d susceptibility under the Ridge scenario (Regime 3, Fig. 8c), consistent with the stabilizing effect of subsidence that suppresses updraft variability and limits cloud depth. Furthermore, solar heating on cloud top, especially prominent in the local afternoon, can offset the longwave radiative cooling, thereby stabilize the cloud layer and reduce in-cloud supersaturation (Wood, 2012; Zheng et al., 2018), hence dampening the N_d susceptibility to aerosols. Such mechanism can exert a larger influence on Regime 3, as it is dominated by the warm season (summer and fall) occurrences. The good agreement between the model and satellite observations under these steady conditions suggests that, when cloud-top mixing and convective vigor are limited, E3SMv2 aerosol activation and microphysics perform reasonably well.

Conversely, under trough conditions (Regime 4), retrievals and E3SMv2 produce the highest N_d susceptibility among all regimes (Fig. 8d). In these conditions, stronger updraft and abundant moisture produce deeper MBL clouds, and effectively increase the in-cloud supersaturation (Gong et al., 2023). Therefore, the environmental conditions and the relatively less N_d provide aerosols a greater opportunity to modulate droplet numbers (Hudson and Noble, 2014; Zheng et al., 2024). The E3SMv2 exhibits a steeper slope than that observed, suggesting that the model may overestimate the efficiency with which additional aerosol activates into new droplets, though the discrepancy is less pronounced than those in Regime 2.

Interestingly, the largest overestimation of ACI_N by E3SMv2 occurs under Post-Trough conditions (Regime 2, Fig. 8b). In this regime, the model exhibits a much higher ACI_N than observations, suggesting that the model either overestimates aerosol activation or underrepresents processes that limit droplet concentration, such as drizzle formation and entrainment mixing. In thicker MBL clouds, the greater vertical extent may allow for enhanced droplet recirculation and collision–coalescence, resulting in a reduction of N_d that dampens aerosol effects (O et al., 2018; Zheng et al., 2024). Under Pre-Trough

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conditions (Regime 1, Fig. 8a), the model slightly overestimates N_d susceptibility yet agrees better with observations compared to Post-Trough. Previous studies have shown that E3SMv1 exhibits greater sensitivity of N_d to aerosols than do ground-based and satellite observations (Christensen et al., 2023; Varble et al., 2023), and this issue appears to persist in E3SMv2 over the ENA, consistent with Huang et al. (2024). It is possible that the model does not accurately represent the postfrontal boundary layer, possibly due to the unresolved subgrid turbulence (Ma et al., 2022), which may lead to an exaggerated N_d response to aerosol extinction.

Note that under low MBL aerosol (σ_{MBL}) conditions, the model yields more occasions of low N_d compared to the satellite, though it can be due to the limited sample sizes in the satellite, yet the N_d increases with σ_{MBL} are also more subtle, especially in the Post-Trough and Ridge regimes. Previous studies suggest that when environmental conditions limit activation and updraft, the resulting droplet numbers are systematically low (Tang et al., 2023; Varble et al., 2023). In other words, the model parameterizations lead to under-activation of aerosols in low-aerosol or weak-turbulence regimes. In contrast, sensitivity experiments by Wan et al. (2025) show that while stronger updrafts can boost N_d, doing so would undesirably increase the effective radiative forcing. Therefore, the model aerosol activation scheme might be oversensitive to the environmental factors, as shown in the present study. It is also noteworthy that E3SMv2 simulates a greater extent of σ_{MBL} than is observed by satellites under all scenarios, particularly in Regime 4, where precipitation is more prevalent. This discrepancy may arise from insufficient drizzle scavenging at moderate to high aerosol loads (Shan et al., 2024), whereby the model fails to effectively remove aerosols, rendering it overly sensitive to incremental changes in σ_{MBL}.

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4.5 Aerosol-cloud interactions under different regimes

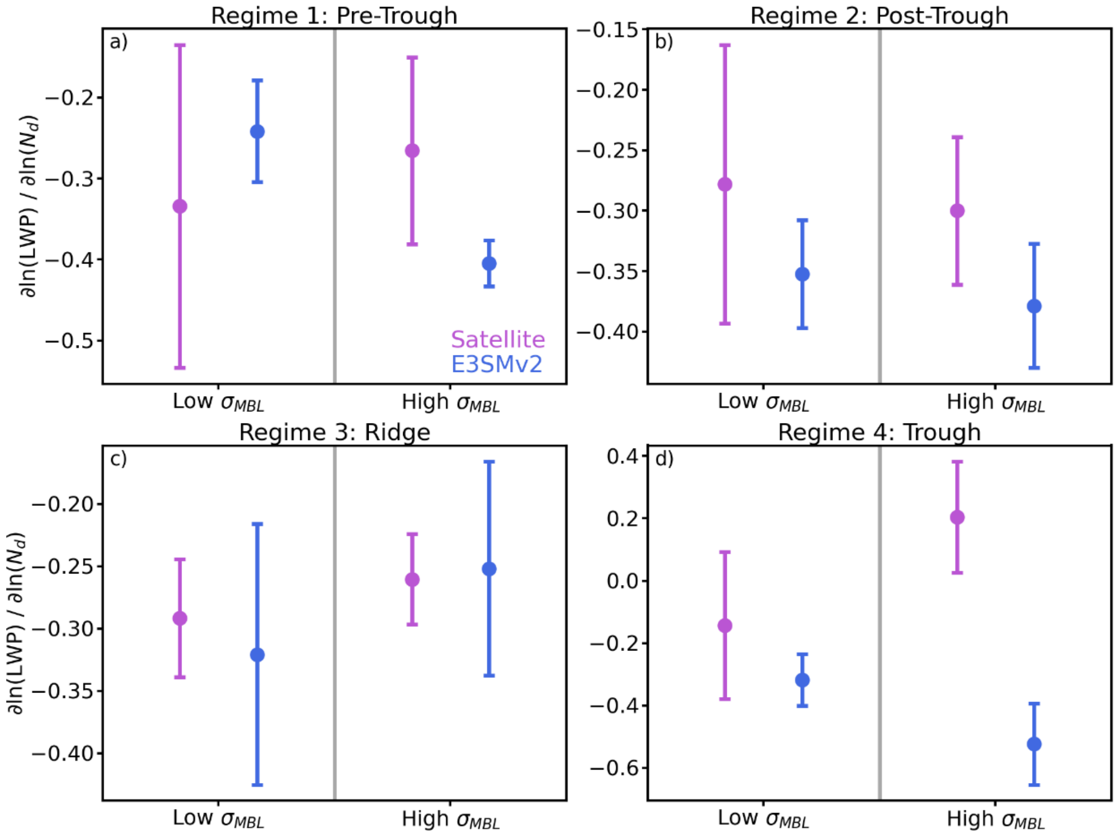

Figure 9. LWP adjustment due to N_d under low and high σ_{MBL} categories, separated by the median σ_{MBL} values (gray line) from the aggregate satellite (purple) and E3SMv2 (blue) dataset. For a) Regime 1; b) Regime 2; c) Regime 3; d) Regime 4.

In order to further illustrate the impact of aerosols on the behavior of the LWP–N_d relationship, both satellite and E3SMv2 data are grouped into lower and higher half σ_{MBL} categories, with the median σ_{MBL} value (0.594) derived from the aggregate of the datasets (Fig. 9).

In the Pre-Trough regime (Fig. 9a), clouds exhibit low tops and moderate precipitation, featuring a regime where precipitation suppression competes with entrainment drying. Satellite retrievals show a weaker negative LWP–N_d sensitivity under high aerosol loading (i.e., LWP declines less steeply with N_d), whereas E3SMv2 simulates a steeper negative slope. The satellites may observe a weaker decline in LWP with increasing N_d because moderate precipitation in this regime allows sub-cloud drizzle evaporation to moisten the boundary layer, which weakens the in-cloud humidity gradient and reduces

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the entrainment efficiency, thereby buffering LWP losses (Wang et al., 2010; Chen et al., 2011; Jia et al., 2022). In the Ridge regime (Regime 3; Fig. 9c), characterized by shallow clouds and minimal precipitation, both satellite retrievals and E3SMv2 simulations yield a relatively weak (less negative) LWP–N_d sensitivity under high aerosol conditions. At higher aerosol loadings, increased N_d suppresses precipitation more effectively in these shallow clouds, stabilizing LWP by limiting drizzle loss. This mechanistic alignment between observations and E3SMv2 may explain their convergence toward a weaker negative slope (Quaas et al., 2020; Jia et al., 2022).

Post-Trough clouds (Regime 2; Fig. 9b), with greater cloud-top heights and moderate precipitation, exhibit a steeper negative LWP–N_d sensitivity in both observations and E3SMv2. The greater depth of Post-Trough clouds amplifies the vertical moisture gradient between the cloud layer and the overlying dry free troposphere. In this scenario, the primary mechanism reducing LWP is enhanced entrainment-driven evaporation. Once droplets are smaller (high N_d), they evaporate faster at cloud top, enhancing evaporative cooling. This strengthens entrainment through buoyancy reversal, creating positive feedback that accelerates LWP loss (Gryspeerdt et al., 2019). Although precipitation suppression can still play a role, the net result in a deeper cloud is often dominated by entrainment drying rather than by the retention of liquid water. Consequently, as aerosol loading increases, the slope of LWP–N_d becomes more negative, aligning with findings that deeper clouds, with stronger inversions, exhibit steeper negative slopes in LWP–N_d (Zhang and Feingold, 2023).

The Trough regime (Fig. 9d), marked by deep, precipitating clouds and unstable conditions, highlights a key model-observation discrepancy: satellites detect a weak or even positive LWP–N_d relationship at high aerosol loadings, while E3SMv2 simulates a steeply negative slope. It could be possible that, under the moist and unstable environment, the increasing N_d provides more surface areas for water vapor condensation and hence offsets the entrainment drying loss (Gryspeerdt et al., 2019). Also, under this regime with relatively low N_d, the increasing evaporation is favorable for more latent heat release, allowing the cloud to be invigorated and expand in vertical extent, hence increasing the LWP (Altaratz et al., 2014). Moreover, satellite retrievals in this regime may be also biased by vertical cloud inhomogeneity and drizzle contamination, which could artificially inflate LWP estimates in high-N_d conditions (Zhang et al., 2022). Meanwhile, E3SMv2 relies on parameterizations for turbulence and entrainment that are calibrated for shallow stratocumulus and may not fully capture the intermittency of entrainment in thicker, more precipitating cloud regimes (Gettelman and Morrison, 2015; Wang et al., 2023; Tang et al., 2023). This may cause the model to amplify entrainment drying relative to what might be observed, thereby producing a more strongly negative LWP–N_d slope than indicated by satellites.

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Ultimately, these contrasting signals reflect both retrieval limitations in complex cloud systems and the model's sensitivity to microphysical closure assumptions (Christensen et al., 2023).

Such model-satellite discrepancies are further confirmed in the bulk indirect susceptibility, which quantifies the integrated response of LWP to changes in marine boundary layer aerosol extinction. As shown in Table 3, satellite-derived susceptibilities range from –0.070 to –0.015, while E3SMv2 simulations consistently yield larger negative values (from –0.138 to –0.098), especially for Regime 2 and 3, which feature more stratiform-like clouds. That is, for a given increase in aerosol loading, E3SMv2 predicts a stronger LWP reduction than satellite observations. This systematic overestimation by E3SMv2 indicates that the model may be overly sensitive to aerosol perturbations, translating into an exaggerated indirect effect on LWP. Potential causes for this discrepancy include overly aggressive drizzle suppression in the MG2 microphysics scheme, which may retain excess LWP at low N_d but prematurely truncate water accumulation as N_d increases. In addition, limitations in representing subgrid-scale turbulent mixing and entrainment could further contribute to the observed biases. While E3SMv2 captures the qualitative trends of aerosol–cloud interactions, the quantitative discrepancies highlight the need for further refinement in aerosol activation and cloud microphysical parameterizations, as well as improved process-level representations of drizzle processes and entrainment mixing. Addressing these issues is essential for enhancing the model ability to simulate the indirect effects of aerosols on cloud properties in the ENA region.

Table 3. Indirect susceptibility of LWP to Aerosol for Satellite and E3SMv2

	Regime 1	**Regime 2**	**Regime 3**	**Regime 4**
		$\frac{\partial ln(LWP)}{\partial ln(N_d)}$		
Satellite	**-0.164 ± 0.019**	**-0.191 ± 0.016**	**-0.176 ± 0.007**	**-0.028 ± 0.046**
E3SMv2	**-0.314 ± 0.011**	**-0.262 ± 0.013**	**-0.356 ± 0.007**	**-0.167 ± 0.029**
		$\frac{\partial ln(N_d)}{\partial ln(\sigma_{MBL})}$		
Satellite	**0.411 ± 0.056**	**0.364 ± 0.061**	**0.288 ± 0.032**	**0.518 ± 0.188**
E3SMv2	**0.430 ± 0.025**	**0.527 ± 0.024**	**0.275 ± 0.015**	**0.616 ± 0.062**

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	$\frac{\partial ln(LWP)}{\partial ln(N_d)} * \frac{\partial ln(N_d)}{\partial ln(\sigma_{MBL})}$			
Satellite*	**-0.067 ± 0.012**	**-0.070 ± 0.013**	**-0.051 ± 0.006**	**-0.015 ± 0.024**
E3SMv2*	**-0.135 ± 0.009**	**-0.138 ± 0.009**	**-0.098 ± 0.006**	**-0.104 ± 0.021**

*The errors are given by propagating uncertainties of the $\partial \ln(LWP)/\partial \ln(N_d)$ and $\partial ln(N_d)/\partial \ln(\sigma_{MBL})$, based on the multiplicative error propagation method.

5. Summary and Conclusions

This study investigated aerosol-cloud interaction processes over the ENA by assessing satellite retrievals and simulations from E3SMv2. Using CALIPSO-derived aerosol extinction and MODIS cloud properties, along with model output from a 1° nudged E3SMv2 simulation over a ~10°×10° domain from 2006 to 2014, we investigated the seasonal and the meteorological-regime-based variations of marine low-cloud properties and examined how LWP responds to changes in N_d and to variations in MBL aerosol extinction.

Our analysis reveals distinct seasonal variations in cloud and aerosol properties, with both satellite and model data displaying higher N_d and lower LWP in warm seasons and lower N_d and higher LWP in cold seasons. E3SMv2 systematically underestimates the N_d but produces a more comparable LWP simulation, confirming previous studies. In general, satellite retrievals and E3SMv2 simulations capture the qualitative trends where higher N_d under increased aerosol loading and a characteristic inverted-V relationship between LWP and N_d. However, while the LWP response to N_d in the satellite dataset is primarily attributed to precipitation suppression and entrainment and evaporative processes, the model simulations appear to generate a similar but more dramatic shape due to deficiencies in representing these processes within the model parameterizations. Furthermore, it is possible that satellite retrieval biases and sampling strategies may contribute to the observed inverted-V behavior in the LWP–N_d relationship (Grosvenor et al., 2018; Arola et al., 2022; Gryspeerdt et al., 2022). Although we addressed retrieval uncertainties to some extent by applying several data screenings documented in Painemal et al. (2020), in-situ observations are still needed to confirm the LWP–N_d relationship. Another possibility is that the inverted-V shape reflects natural spatial variability that leads to both increases and decreases of N_d with LWP (Goren et al., 2025).

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Next, we employ a deep-learning-based clustering method to partition the ENA meteorology into four distinct synoptic regimes. This clustering approach more effectively captures the complex spatial and temporal variability of large-scale meteorological fields, compared to the traditional methods. The detailed regime-based analysis further demonstrated that meteorological conditions play a critical role in modulating aerosol-cloud interactions. The characteristic inverted-V shape in the LWP–N_d relationships persist across different synoptic regimes, with pronounced regime-dependent discrepancies between satellite retrievals and E3SMv2 simulations.

Regime 3 (Ridge) shows the strongest simulated inverted-V response among all four regimes, where LWP consistently decreases at high N_d, driven by the stronger contribution of the entrainment-drying effect in a thinner, more stable cloud layer. In the pre-trough regime, the LWP decline with high N_d is partially masked by the interplay between precipitation-stabilization and entrainment-mixing processes. In the Post-Trough regime, deeper clouds with intermediate precipitation display a more negative LWP–N_d sensitivity due to enhanced entrainment-drying effect, a process the model appears to overestimate compared to observations. Conversely, the Trough regime, characterized by deep, precipitating clouds, features complex behavior where satellite retrievals yield a muted LWP response, likely because of precipitation-suppressing overwhelming the entrainment-drying effect on LWP. The model overpredicts the LWP responses across all regimes with different magnitudes. While both datasets show an increasing LWP with rising N_d at low N_d concentrations, E3SMv2 reaches its peak LWP at lower N_d values, suggesting that its microphysics parameterization may act to suppress drizzle too aggressively and also inadequately represent subgrid moisture variability. Moreover, the E3SMv2 simulates steeper declines of LWP with N_d at higher values, indicating that the model parameterization induces more rapid entrainment-drying effects contributing to excessive LWP loss.

Both satellite retrievals and E3SMv2 simulations consistently show that N_d increases with increasing MBL aerosol extinction across all synoptic regimes. However, the model-derived sensitivity varies considerably with meteorological conditions compared to satellite. In the regimes with relatively shallow clouds, both data and model exhibit the lower N_d susceptibility to aerosol. In contrast, E3SMv2 shows a markedly steeper N_d response than observed in regimes with deeper and more precipitating clouds, likely due to an overestimation of aerosol emission and activation or underrepresentation of limiting processes such as insufficient drizzle scavenging. Such discrepancies suggest that the parameterizations in model aerosol activation schemes might be overly sensitive to environmentally controlled factors, which leads to a larger range of ACI_N.

The regime-wise LWP responses on N_d are analyzed under low and high MBL aerosol extinction conditions. In shallower and less precipitating cloud conditions, such as the Ridge regime, both satellite

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retrievals and E3SMv2 simulations converge to a weaker (less negative) sensitivity under high aerosol loading. By contrast, in more precipitating and vertically extended regimes, E3SMv2 exhibits significantly stronger LWP depletions on N_d with increasing aerosols. The E3SMv2 operates at relatively coarse horizontal and vertical resolutions, and its microphysics parameterizations, particularly the MG2 scheme, tend to overemphasize the entrainment-driven drying and droplet evaporation as it was calibrated for shallow stratus and stratocumulus conditions (Tang et al., 2024). As a result, the transition from shallow to deep cloud regimes, where natural processes evolve continuously, may not be adequately captured, leading to an exaggerated drying signal in the model, hence amplifying the negative LWP–N_d slope.

In summary, our findings report the satellite-observed and model-simulated range of aerosol-cloud interaction indices from both seasonal and regime-based perspectives over the ENA. Moreover, the regime-based analysis demonstrates that the interplay between aerosol loading and cloud microphysics is highly sensitive to the prevailing meteorological conditions. While E3SMv2 reliably reproduces the overall trends in aerosol effects on stratiform clouds, its performance degrades in deeper, more dynamically complex regimes. Given uncertainties in the satellite observations, it is critical for future studies to integrate datasets from airborne, ground-based, and satellite platforms. This strategy would enable the quantification of errors as well as corroborating the results presented here.

On the modeling side, increasing horizontal and vertical resolution may help improve representations of entrainment and drizzle processes. Furthermore, recalibrating autoconversion rates and developing scale-aware entrainment schemes, ideally constrained by high-resolution observational data from field campaigns such as ARM, may reduce the persistent uncertainties in simulating aerosol-cloud interactions, particularly under the dynamic meteorological transitions typical of the ENA region. However, bridging the gap between shallow and deep cloud regimes remains particularly challenging, as current model schemes treat them separately despite the gradual transitions that may be observed in nature.

Future research will focus on exploring the transferability of this regime-based analysis to other global marine regions, assessing the scaling effects and exploring the process-level understanding of aerosol-cloud interactions within models, and extending the investigation to include aerosol-cloud-radiation interactions, thereby providing better constraints on effective radiative forcing. Such efforts are essential to refine microphysical parameterizations and enhance the overall fidelity of climate models in representing these critical processes.

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Code and Data availability

The E3SMv2 nudged simulation output is available at: https://zenodo.org/records/15670340. The ERA5 reanalysis is available at: https://cds.climate.copernicus.eu/datasets/reanalysis-era5-pressure-levels?tab=overview. The original clustering model is available at: https://zenodo.org/records/14720991. The hyperparameter-tuned model and the collocated CALIPSO-MODIS dataset are available upon request.

Author contributions

The idea of this study was developed by XZ, YF, and DP. XZ performed the analyses and wrote the manuscript under the supervision of YF. MZ performed the nudged E3SM simulation. DP and ZL constructed the collocated CALIPSO-MODIS dataset. XZ, YF, DP, MZ, SX, ZL, RJ and BL participated in further scientific discussions and provided substantial comments and edits on the paper.

Competing interests

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

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