



Coarse-mode sea spray aerosols enhance warm rain formation in marine low clouds

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Abstract. Precipitation in low clouds affects cloud responses to aerosol perturbations, thereby impacting aerosol radiative forcing of climate, a significant but indeterminate contribution to uncertainty in future warming. The onset of drizzle formation in marine low clouds may be influenced by the presence of supermicron sea salt aerosols. These particles can act as giant CCN (GCCN) that benefit from rapid condensational growth to a size that initiates collision-coalescence. Aircraft-based in-situ observations of cloud microphysical properties and aerosol size distributions from the Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE) are analyzed from January–June 2022. Observations from a cloud aerosol spectrometer and a cloud droplet probe in clear air are used to quantify GCCN size distributions. Modest correlations are found between GCCN and near surface-level wind speeds when averaged together into wind speed bins, a result consistent with other studies. Observed GCCN distributions and cloud thicknesses are used to drive simulations within a 1-dimensional kinematic super droplet model, exploring the conditions under which the observed GCCN distributions induce a first-order impact on precipitation rate. The model demonstrates that even relatively small GCCN salt masses ($\sim 1 \mu\text{g m}^{-3}$) can accelerate the onset of drizzle in lightly precipitating marine low clouds. Similarly, a statistical analysis of in-cloud data shows the ratio of rainwater to total condensed water is higher when GCCN concentration/mass is above average. Together, the results indicate a significant influence of GCCN on precipitation formation in marine low clouds.

1 Introduction

Cloud-aerosol interactions have a significant impact on climate, particularly through their influence on marine stratocumulus clouds, which cover roughly 20 % of the planet's oceans and play a dominant role in modulating Earth's radiative energy balance (Hartmann and Short, 1980). These in-

teractions affect both cloud microphysics and macrophysics, including cloud albedo and lifetime, thereby altering the reflection of solar radiation and the overall radiative forcing of the climate system (Seinfeld et al., 2016). By regulating heat, moisture, and momentum exchanges between the ocean and atmosphere, the stratocumulus-topped boundary layer (STBL) profoundly influences large-scale climate sys-

tems (Wood, 2012). Marine stratocumuli reflect a large portion of the Earth's incoming solar radiation, while only minimally contributing to outgoing longwave radiation (Chen et al., 2000). This combination of radiative effects and expansive coverage makes stratocumulus clouds integral to the planet's overall radiative budget (Stephens and Greenwald, 1991; Hartmann et al., 1992; Wood, 2012; L'Ecuyer et al., 2019). Unfortunately, clouds are the largest uncertainty in estimates and interpretations of the Earth's rapidly changing energy budget, and much of the uncertainty revolves around how aerosols interact with stratocumulus clouds (Pachauri et al., 2014; Bellouin et al., 2020; von Salzen et al., 2025). The Twomey effect (Twomey, 1977) describes how increases in cloud droplet concentration resulting from aerosol-cloud interactions (ACI) reduce droplet size and increase total droplet surface area for a fixed amount of condensate. As a result, clouds reflect more solar radiation, even without changes in cloud liquid water content. However, changes in droplet size can also drive adjustments to cloud macrophysical properties by changing precipitation and cloud top entrainment rate (Ackerman et al., 2004; Wood, 2007; Bellouin et al., 2020). These precipitation and droplet sedimentation processes, which impact how cloud condensate and cover adjust in response to aerosol changes, remain challenging to quantify making them a persistent source of uncertainty in climate models (Stevens and Feingold, 2009). As these interactions occur on sub-kilometer scales, Earth system models poorly represent these cloud adjustments (e.g., Mülmenstädt and Feingold, 2018). Cloud condensation nuclei (CCN) play a crucial role in aerosol-cloud-precipitation processes, by increasing cloud droplet concentration, which reduces cloud droplet size for a given amount of condensate, thereby decreasing precipitation efficiency (Albrecht, 1989; Beard and Ochs, 1993; Pawlowska, 2003; Terai et al., 2012; Mann et al., 2014). The ability of a cloud system to create a few large droplets ($\sim 15\text{--}20\text{ }\mu\text{m}$ radius) greatly influences its capacity to generate precipitation through collision-coalescence (Hocking, 1959; Berry, 1967; Albrecht, 1989; Wood et al., 2009). There are two pathways to forming large droplets. First, reducing the overall number of CCN will result in each cloud droplet being bigger (for a given condensate amount), and then collision-coalescence may be able to grow a few lucky drops that end up accelerating the collision-coalescence. Second, if present in sufficient amounts, giant CCN can also grow efficiently by condensation to radii of $15\text{--}20\text{ }\mu\text{m}$.

Giant cloud condensation nuclei

Drizzle formation can be achieved through the existence of a tail of large “collector” droplets formed on giant cloud condensation nuclei (GCCN) that can initiate collision-coalescence (Ludlam, 1951; Johnson, 1982; Feingold et al., 1999; Jensen and Lee, 2008; Jensen and Nugent, 2017; Dziekan et al., 2021). In marine environments giant sea spray

particles with dry diameters larger than $\sim 2\text{ }\mu\text{m}$ enhances collision-coalescence, in contrast to smaller particles that suppress it. Based on this finding, we define GCCN as salt particles with dry diameters larger than $2\text{ }\mu\text{m}$. Condensational growth of GCCN from the ocean surface through the cloud is important for growing the particles to a size where they can initiate collision-coalescence (Ludlam, 1951; Jensen and Nugent, 2017). Despite a solid theoretical foundation, this effect has not yet been incorporated into physical parameterizations used in bulk microphysical models, which may lead to an underestimation of warm rain formation in some clouds (Jensen and Nugent, 2017; Dziekan et al., 2021). Cloud droplets grown on GCCN are highly concentrated solutions compared with the majority of cloud droplets, which are grown on much smaller accumulation mode aerosol particles (with typical dry diameters of $\sim 0.1\text{ }\mu\text{m}$). This accelerates, via the Raoult effect (Jensen and Nugent, 2017), the condensational growth rates of droplets grown on GCCN compared with those forming on accumulation mode CCN. In some cases, this may lead to a faster onset of warm rain (L'Ecuyer et al., 2009). The fundamental challenge to representing GCCN in microphysical models is that the condensational growth rate of cloud droplets depends not only on their size but also on the mass of salt they contain. GCCN-containing droplets must therefore be treated as a distinct category of cloud hydrometeor, increasing model complexity. Posselt and Lohmann (2008) attempted to incorporate GCCN effects into a global climate model by parameterizing GCCN by assuming they directly activate into raindrops; however, this approach neglects detailed condensational growth processes. It is computationally-challenging to accurately capture this aerosol-dependent condensational growth in microphysical models, but it can be achieved using super droplet methods (Shima et al., 2009, 2020; Li et al., 2022; Zmijewski et al., 2024), or by using two-dimensional bin microphysical schemes that independently keep track of hydrometeor wet sizes as well as the aerosol dry size (e.g., Flossman et al., 1985; Lebo and Morrison, 2013). Since droplets formed on GCCN remain highly concentrated salt solutions, they can more easily resist evaporation and the larger GCCN can even continue growing by condensation as they descend through (weakly) subsaturated downdrafts (Jensen and Nugent, 2017). This contrasts with smaller cloud droplets grown on accumulation mode aerosol, which typically lose mass and may completely evaporate in similar conditions.

Despite advances in understanding GCCN's role in collision-coalescence growth, significant uncertainties remain due to wide variations in the sizes and number concentrations reported in prior studies. For example, Jensen and Nugent (2017) found that introducing $1\text{ }\mu\text{m}$ dry radius GCCN at number concentrations of 0.3 cm^{-3} could influence drizzle in a microphysical parcel model, while Feingold et al. (1999) introduced $20\text{ }\mu\text{m}$ droplets, a proxy for condensationally-grown GCCN, into large eddy simulations at concentrations as low as 10^{-4} to 10^{-2} cm^{-3} and found im-

pacts on precipitation for the higher concentrations. Because condensational growth of GCCN was not treated explicitly in Feingold et al. (1999), it is difficult to compare this result with other studies, highlighting major differences in model assumptions and methods. Lowenstein et al. (2010) used aerosol and cloud probe observations of hydrated giant haze particles below marine trade cumulus to initialize a parcel model that produced a tail of large droplets ($r \sim 20\text{--}100\text{ }\mu\text{m}$) above cloud base that matched observations fairly on two flight days. However, model simulations were not conducted without GCCN, so the overall impact of the GCCN is unclear.

Dziekan et al. (2021) have performed the most sophisticated and realistic simulations of GCCN influences on low cloud precipitation to date, using a super droplet model embedded within a large eddy model. The model explicitly computes both the condensational and coalescence growth of GCCN and shows that GCCN from an observed flight over the southeastern Pacific is sufficient to increase cloud base precipitation rate in a marine stratocumulus cloud case by $\sim 50\%$ compared with the case with zero GCCN. A somewhat weaker influence ($\sim 10\%$) was found in a deeper trade cumulus case. The base LES case used in Dziekan et al. (2021) used observed GCCN measurements with near-surface GCCN dry salt mass of $\sim 6\text{ }\mu\text{g m}^{-3}$ and number concentration of $\sim 0.22\text{ cm}^{-3}$ (for dry diameters exceeding $2\text{ }\mu\text{m}$) with a near-surface wind speed of 5.4 m s^{-1} . Sensitivity tests were conducted for larger GCCN concentrations/masses to show that further increasing GCCN loading continued to increase precipitation rate, with a factor of 5 increase leading to precipitation rate increases of $\sim 300\%$ (stratocumulus) and $\sim 30\%$ (cumulus) above the rate without GCCN.

GCCN dry size distributions in previous studies are typically found to decrease approximately exponentially with increasing particle diameter (e.g., Woodcock, 1953; Lewis and Schwartz, 2004; Colón-Robles et al., 2006). Because the observed e -folding diameter for the decrease is relatively small (see Sect. 2.2 below), this means that reported GCCN number concentrations are particularly sensitive to the threshold dry diameter $D_{\min}$ used to define GCCN. Although several recent studies have adopted a threshold $D_{\min} \sim 2\text{ }\mu\text{m}$, studies do not always report what threshold diameter was used or use a hydrated size threshold rather than a dry one (e.g., Colón-Robles et al., 2006), making cross-study comparisons of observed GCCN and their influences on clouds challenging (Cooper et al., 2013).

This work aims to address critical gaps in understanding coarse-mode cloud-aerosol interactions in marine low clouds. We utilize a robust observational framework, which includes a very large number of flight legs across different seasons and meteorological conditions, established by the NASA Earth Venture Sub-Orbital (EVS-3) Aerosol Cloud Meteorology Interactions over the western Atlantic Experiment (ACTIVATE) field campaign. Building on previous work (e.g., Woodcock, 1953; Colón-Robles et al., 2006), we

first document coarse-mode aerosol size distributions under clear sky conditions below cloud for a wide variety of aircraft flights and examine how near-surface wind speed influences their number and mass concentration and size distribution. We then investigate the extent to which our observed variations in GCCN number concentrations can modulate precipitation in marine stratocumulus clouds using a super droplet model. The goal is to determine how each of the observed GCCN distributions alters the cloud droplet size distribution and accelerates precipitation formation under varying aerosol conditions and for a range of plausible macrophysical and microscale turbulence conditions. We also examine the influence of GCCN on observationally derived rain-water content (RWC), calculated from ACTIVATE observations, which provides a purely observational perspective on GCCN-precipitation relationships. Together, the modeling and observation analyses provide a comprehensive understanding of how GCCN impact warm rain formation. This analysis is necessary not only to improve our understanding of aerosol-cloud interactions, but also to contribute to better parameterizations that include the impact of GCCN.

2 Methods

2.1 Dataset

ACTIVATE is a NASA Sub-Orbital field campaign that flew aircraft and captured data from February 2020 through June 2022 with the intent of providing globally important data detailing changes in warming and cooling planetary feedbacks, atmospheric aerosols and sources, and marine boundary layer cloud systems (Sorooshian et al., 2023). The observations relevant to this study were collected during two deployments spanning January to June 2022 to capture different seasonal states and broaden the range of aerosol and meteorological conditions. Flights were conducted using a twin-aircraft design based out of NASA Langley Research Center in Virginia, with the HU-25 Falcon flying at lower altitudes (0.15 to 3 km) and the King Air flying above (8 to 10 km). In this study, only data from the low-flying Falcon are used. We utilize measurements taken from the Diode Laser Hygrometer (DLH) which measures relative humidity, Cloud Droplet Probe (CDP), Cloud-Aerosol Spectrometer (CAS), and the two-dimensional Stereo Probe (2D-S), all of which were externally mounted on the Falcon. The CAS measures from $0.5\text{ }\mu\text{m}$ diameter to $50\text{ }\mu\text{m}$ diameter and the CDP measures from 2 to $50\text{ }\mu\text{m}$ diameter. Both probes measure cloud droplet size distributions, from which aerosol and cloud droplet number concentration, liquid water content, effective radius, and effective variance are derived (Baumgardner et al., 2001; Lance, 2012). The 2D-S measures larger drop number size distribution for liquid and ice both separately and combined; liquid water content; effective diameter and median volume diameter (Kirschler et al., 2022). The 2D-S also measures a broader size range from 29 to $1465\text{ }\mu\text{m}$

diameter. While a Fast Cloud Droplet Probe (FCDP) was available during the campaign, it is not included in this analysis. The FCDP lower diameter threshold is 3 μm diameter, so it doesn't quite reach the 2 μm diameter threshold we were looking for. This study focuses on in-situ Falcon data collected between 150 m and 2.5 km altitude. A total of 75 joint flights on 43 d, logging upwards of 120 flight hours were conducted during the 2022 deployment, making it the most comprehensive year due to fewer pandemic disruptions compared to 2020–2021 (Sorooshian et al., 2023).

We focus on Falcon flight legs classified as BCB (below cloud base) to analyze coarse-mode aerosols (Sects. 2.2–3.2), and ACB (above cloud base) and BCT (below cloud top) when working in-cloud (Sect. 3.3). These legs are a part of “cloudy ensembles”, which are sequences of flight legs conducted in close succession that sample different vertical regions of the cloud and sub-cloud environment, where leg is about 3 min long (Sorooshian et al., 2023). The vast majority of available data come from cloudy ensembles, which account for over 90 % of the total flight legs in 2022. We did not utilize flight legs from designated “clear-sky” ensembles due to insufficient data (see Sorooshian et al., 2023). Less than 10 % of the total ensembles in 2022 are “clear-sky”. The systematic repetition of these flight ensembles has resulted in an extensive and structured dataset (Dadashazar et al., 2022; Sorooshian et al., 2023). However, we aim to use BCB legs without cloud contamination, so to filter for clear sky conditions, we apply a liquid water content threshold of 0.0025 g m^{-3} from the CAS and the CDP and remove data points above this threshold to remove contamination from cloud droplets. We further restrict the dataset by requiring that the precipitation number concentration, defined as the concentration of precipitation-sized particles measured by the 2D-S probe, remains below 100 m^{-3} . This additional criterion ensures that drizzle-sized drops are not present. We also remove data points with relative humidity (RH) above 95 %, given that there is a ~ 5 % error in RH measurements, to further ensure that the sampled air is subsaturated. Together, these thresholds ensure that the observed hydrated size distributions are indeed characterizing haze droplets in subsaturated conditions without cloud or drizzle droplet interference.

2.2 Constructing dry size distributions

To characterize aerosol number concentration and mass concentration above the Western Atlantic, we analyze size-resolved hydrated (ambient) aerosol distributions derived from CAS observations from only the below the cloud base flight legs. Above cloud base and below cloud top legs are used for analysis in Sect. 3.3. We use 1 Hz data for 75 flights on 43 different dates from January to June 2022. Each 1 Hz observation is taken from a typical 3 min below cloud base (BCB) flight leg and only samples passing our clear sky filtering criteria are retained. Though the CAS measures parti-

cles at 0.5 to 50 μm diameter, we select only measurements corresponding to haze particles (ambient hydrated particles) ranging from 2.5 to 50 μm diameter for both the CAS and CDP, which removes any accumulation mode particles before constructing our distributions. The CDP range is 2.5 to 50 μm diameter. It is important to note that this ambient size, for most RH values sampled, corresponds to a dry minimum diameter of less than 2 μm , which ensures that we capture all the sea spray particles with dry diameters greater than 2 μm , even as the RH varies along each flight leg. After the removal of flight legs exceeding clear-sky filtration process (see Sect. 2.1), the relative humidity analysis yielded 460 below cloud base legs with a mean altitude of 701 m. To contextualize our ambient (haze) size distributions, we compare our average ambient CAS and CDP distributions with Lewis and Schwartz (2004). We compare with their Fig. 22b, which shows a range of average size distributions across multiple studies at an average wind speed of $5\text{--}7 \text{ m s}^{-1}$, shown in Fig. 1. Although Lewis and Schwartz (2004) report size distributions across a broader range of wind speeds, our comparisons are limited to the $5\text{--}7 \text{ m s}^{-1}$ range, which is closest to the mean wind speed observed in our dataset (5.13 m s^{-1}) and contains the most robust sampling. Higher wind speed regimes reported in previous studies are not well represented in our dataset, limiting direct comparison. Their distributions are reported at 80 % RH (r_{80}), so we normalize our ambient measurements to this reference humidity using a hygroscopic growth factor derived from the mean relative humidity of each flight leg (see below). The resulting r_{80} values are used to rescale particle sizes while maintaining the measured number concentration distributions. All size distributions used for this comparison are in $\frac{dN}{d\ln D}$ to maintain consistency with Lewis and Schwartz (2004). We find that our CDP and CAS ambient distributions in this wind speed range, normalized to r_{80} , generally fall within the 3 standard deviations of the studies depicted in Lewis and Schwartz (2004) Fig. 22b.

We estimate dry salt aerosol size distributions from our hydrated measurements. To convert an ambient size distribution to a dry salt size distribution, we utilize the average flight leg's relative humidity with respect to liquid water, taken from the Diode Laser Hygrometer (DLH) aboard the Falcon (Diskin et al., 2002; Sorooshian et al., 2023), and the corresponding salt particle “growth factor” (Seinfeld and Pandis, 2006). The hygroscopic growth factor quantifies how aerosol particles absorb moisture from the atmosphere, leading to an increase in their size. This growth factor $g(\text{RH})$ is approximated with a simple fit based on the formulation from Tang et al. (1997) as:

$$g(\text{RH}) = \left(\frac{1.7}{1 - \text{RH}} \right)^{0.31}, \quad (1)$$

The growth factor $g(\text{RH})$ is a function of leg-mean relative humidity (RH) only (for coarse mode sea spray particles, assuming constant sea spray composition), not of diameter and

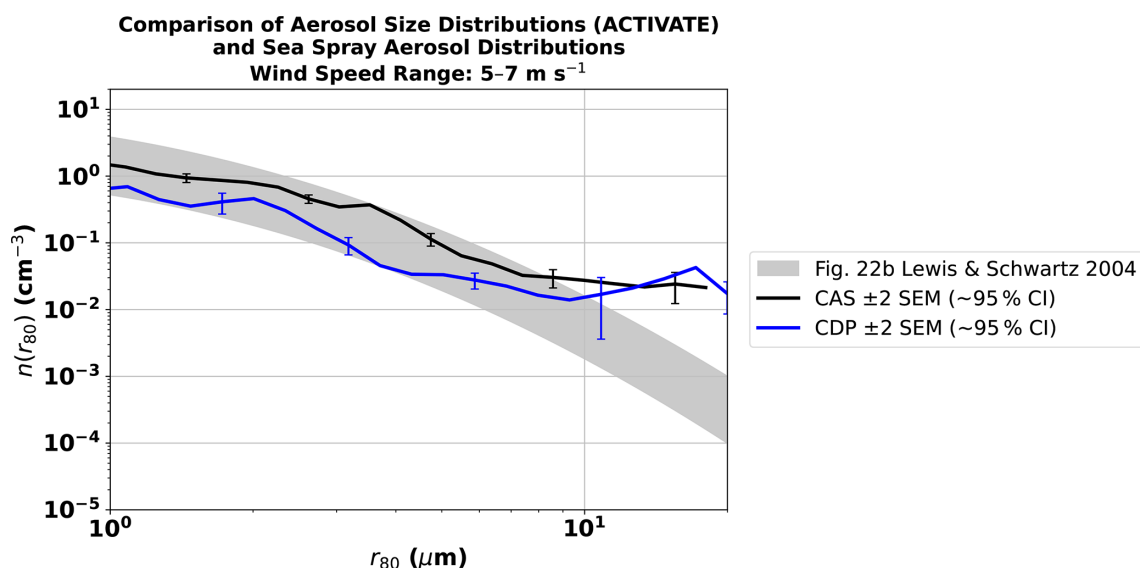


Figure 1. Normalized average ambient aerosol size distributions measured by the CAS (black) and CDP (blue) during ACTIVATE flights for wind speeds of 5–7 m s^{−1}. Error bars denote ±2 standard errors (95 % confidence interval) based on variability across contributing flight legs (see Sect. 2.3 for details). The grey shaded region represents the range of sea spray aerosol size distributions compiled from multiple studies by Lewis and Schwartz (2004).

therefore varies across all flight legs (Tang, 1996; Jensen and Lee, 2008). Ambient particle diameters are converted to dry diameters by dividing by $g(\text{RH})$, and the corresponding number size distributions, expressed as $\frac{dN}{dD}$, are transformed into dry-diameter space. Since RH differs across flight legs, the resulting dry diameter bins vary slightly; therefore, each dry size distribution is interpolated onto a common dry diameter grid spanning 2 to 25 μm before averaging. Average dry size distributions for each instrument are compared in Fig. 2. We calculate the mean total number concentration for the CAS to be 0.56 and 0.18 cm^{−3} for the CDP.

The flight legs in this study from ACTIVATE averaged a sample time of 180 s, which is relatively short compared with some other studies (Colón-Robles et al., 2006 used circle legs of ∼ 30 min duration). Low particle counts can result in “shot noise” and a flattening of the size distributions for the lower number concentrations of larger sized particles (Baumgardner et al., 2017). To quantify this effect, because low particle counts at larger diameters lead to elevated shot noise, we estimate the relative Poissonian counting error for each size bin using Eq. (2):

$$\text{relative error} = \frac{\sqrt{N_{\text{counts}}}}{N_{\text{counts}}}, \quad (2)$$

This allows us to identify the particle sizes for which the counting statistics are most reliable. Because of the quasi-exponential nature of the coarse mode size distributions and the limited probe sampling volume, the relative error exceeds 100 % for many bin diameters above ∼ 10 μm in many cases, so we choose to limit our exponential fitting (See Eq. 2) to the portion of each distribution smaller than 10 μm diame-

ter. The fitted parameters ($N_{0, \text{dry}}$ and $D_{e, \text{dry}}$) (See Eq. 2) and their covariance provides insight into how the abundance (concentration), characteristic size, and the overall mass of coarse-mode aerosol vary together for both instruments, illustrated in Fig. 3.

One possible method to work with “shot noise” is to concatenate a set number of individual 3 min legs into “super legs” to create legs with longer sampling time. However, because the typical ACTIVATE sampling approach mostly involves sampling the different air masses on subsequent legs due to minimal backtracking, changing meteorological conditions would result in “super leg” averages that mix air from numerous locations. The number of legs logged per flight per day varies strongly (between 2–16 legs) and concatenating across multiple legs, flights, or days would risk introducing additional variability due to changing meteorological and flight conditions. Instead, to smooth shot noise, we fit an equation of exponential decay to each size distribution using Eq. (3):

$$n(D_{\text{dry}}) = N_{0, \text{dry}} e^{-\frac{D_{\text{dry}}}{D_{e, \text{dry}}}}, \quad (3)$$

where $N_{0, \text{dry}}$ represents the intercept and $D_{e, \text{dry}}$ represents the e -folding diameter, which controls the fall-off in concentration with size. This fit is performed using nonlinear least-squares regression, such that all included size bins contribute equally to the regression. As a result, bins with low particle counts, like those at larger diameters, may be subject to greater statistical uncertainty due to counting variability. To mitigate the influence of this “shot noise”, the fitting is restricted to dry diameters $\leq 10 \mu\text{m}$, where particle counts are

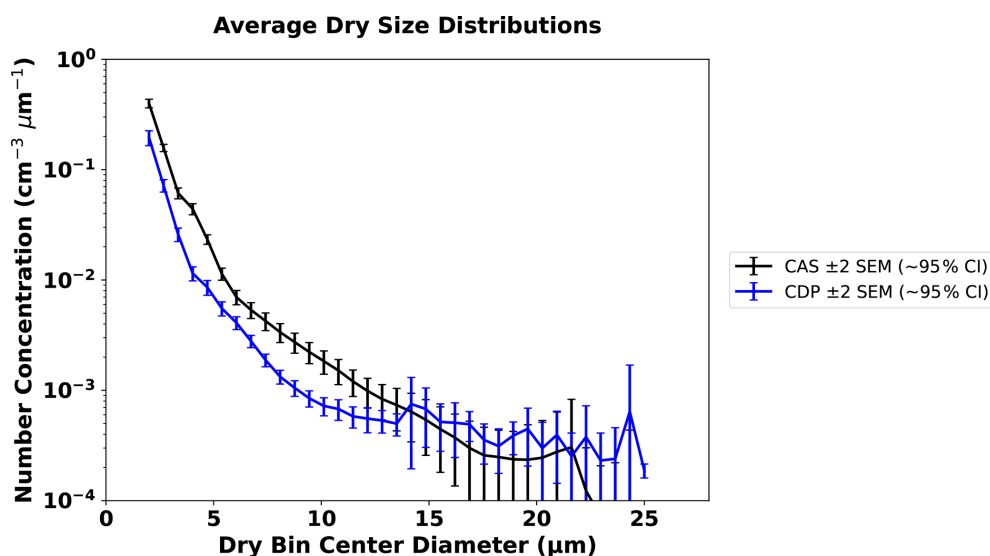


Figure 2. Average dry aerosol size distributions derived from CAS (black) and CDP (blue) measurements. Error bars represent ± 2 standard errors of the mean (95 % confidence interval) based on variability across contributing flight legs (see Sect. 2.3 for details) for January–June 2022 below cloud base flight legs.

higher and sampling uncertainty is reduced. The fitted parameters derived from these dry size distributions are further used for analysis in Sect. 3.2. The slope parameter from these fits, displayed in Fig. 3, provides a useful framework for looking at how variations in size distribution influence aerosol properties such as number concentration and mass concentration.

Larger values of $D_{e, \text{dry}}$ correspond to distributions with relatively more large particles. A strong positive relationship is observed for both instruments, with most points falling near a “slope of one” trend, indicating that increases in particle number are generally accompanied by increases in total mass. Although both instruments show a strong positive relationship between number concentration and mass concentration, notable differences are evident between the CAS and CDP observations. The CAS exhibits a slightly steeper relationship than the CDP. The total mass values from the two probes are generally similar, differing by roughly a factor of two, whereas the number concentrations differ more substantially, by up to a factor of five. In particular, lower concentrations, $\sim 10^{-2} \text{ cm}^{-3}$, are observed more frequently for the CDP than the CAS. The CDP also tends to have larger values of $D_{e, \text{dry}}$, which correspond to a slower decrease in number concentration with size. As a result, although the CDP measures fewer particles overall, these particles tend to be larger to compensate for the lower number concentration. This also yields total mass values that are more comparable to those observed by the CAS.

2.3 Wind speed binning

Near surface wind speed has been shown to be one of the leading controlling factors on bubble bursting and the production of sea spray aerosol (Woodcock, 1953; Monahan et al., 1983; Banner and Peregrine, 1993; Gemmrich and Farmer, 1999; Clarke et al., 2006). To explore the relationship between wind speed and coarse-mode aerosol number concentration for aerosol dry diameters larger than $2.0 \mu\text{m}$, which we define as $N_{d>2}$, we first apply a logarithmic wind profile correction (Stull, 1988) to all Falcon in-situ wind speed measurements, standardizing them to 10 m above sea level. Wind speeds were collected from the NASA Langley in situ aircraft state dataset which provides aircraft-derived wind measurements (Sorooshian et al., 2023). This correction accounts for variability in flight leg altitude and uses a roughness length of 0.02 m for open ocean conditions (Peña Díaz and Gryning, 2008). The mean 10 m wind speed across all valid below-cloud-base legs is 5.31 m s^{-1} with a standard deviation of 2.65 m s^{-1} . To assess uncertainty associated with the assumed surface roughness length, we repeated the correction using roughness lengths of 0.001 and 0.0001 m, representative of smoother ocean surfaces. These assumptions changed the mean estimated 10 m wind speed by approximately 0.8 and 1.2 m s^{-1} , respectively. The corrected wind speeds remained highly correlated with the original estimates ($r \geq 0.998$). These differences are smaller than the observed variability in wind speed and are generally less than the wind speed bin widths used in our analysis. Thus, the uncertainty in the roughness length assumption is unlikely to substantially affect the derived GCCN-wind speed relationships.

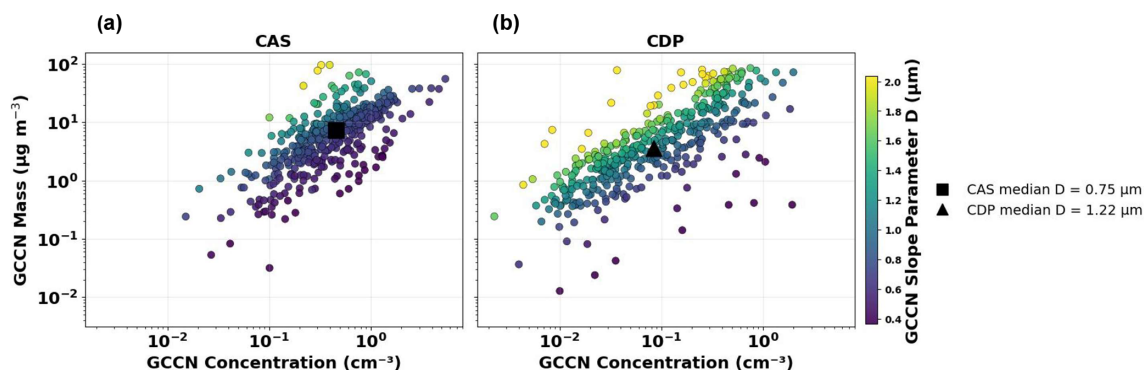


Figure 3. Figure 3 shows the relationship between GCCN dry mass concentration and number concentration for (a) the CAS and (b) CDP observations. Each point is colored by the fitted slope parameter $D_{e, \text{dry}}$, which characterizes the e -folding diameter of the dry size distribution.

Due to the relatively short flight legs in ACTIVATE, we aggregate flight legs into six wind speed bins that capture the full range of observed wind speeds (Fig. 4). Previous observational studies have shown a strong dependence on wind speed for aerosol number concentration, particularly in the coarse mode (Woodcock, 1953; Lewis and Schwartz, 2004; Colón-Robles et al., 2006). Given this established dependence, grouping legs by wind speed allows us to directly compare our results with prior findings (Sect. 4) while increasing the statistical robustness of our results, as the aggregation reduces variability from short sampling times. These bins are designed to represent the overall wind speed range in our dataset, 0 to 16 m s^{-1} . The selected wind speed ranges are 0–2.5, 2.5–3.5, 3.5–5, 5–7, 7–9, and $> 9 \text{ m s}^{-1}$, which we manually selected. The distribution of 10 m wind speeds in our dataset is not uniform. The chosen ranges (0–2.5, 2.5–3.5, 3.5–5, 5–7, 7–9, and $> 9 \text{ m s}^{-1}$) ensure that each of the 6 bins contains a sufficient number of flight legs while also isolating wind speed ranges commonly discussed in sea spray aerosol literature (Lewis and Schwartz, 2004, see Fig. 1). We repeated the analysis using smaller binning schemes, 4 and 5 wind speed bins (not shown), and found that the slope of the number concentration and wind speed relationship differed by less than 16 % compared to the 6-bin case, with all slopes overlapping within their respective uncertainties.

We examine the dependence of coarse-mode aerosol number concentration $N_{d>2}$, cm^{-3} , and mass, $M_{d>2}$, $\mu\text{g m}^{-3}$, on wind speed by analyzing binned number concentration and mass concentration as a function of wind speed for both the CAS and CDP (Fig. 4). Using both the CAS and the CDP enables us to evaluate whether the observed relationships hold consistently across different instruments. We take the mean dry size distribution, resulting in one average size distribution for each of the six wind speed bins, shown in Fig. 4 for GCCN number concentration. The standard error quantifies the uncertainty in the mean droplet number concentration at each particle size bin due to the finite number of legs within each wind speed range. Size distributions are in $\frac{dN}{dD}$ and stan-

dard error is calculated following Eq. (4):

$$\text{SE} = \frac{\sigma}{\sqrt{n}}, \quad (4)$$

where σ represents the standard deviation calculated across all flight legs within a given wind speed bin at each particle size bin, and n represents the number of flight legs included in that bin. In addition to computing standard error for our wind speed and number concentration and relationships, we also compute counting errors for total number concentration measured by each instrument. Counting uncertainties are assumed to follow Poisson statistics, where the uncertainty in particle counts is given by the square root of the number of counts. These uncertainties are propagated to number concentration by accounting for the instrument sample volume. The total counting error is obtained by adding the bin-level errors in quadrature, which reduces to:

$$\delta N = \sqrt{\sum_i C_i}, \quad (5)$$

where δN is the total counting error in the integrated concentration, N is the total number concentration obtained by summing over all size bins, and C_i is the concentration in bin i . The bin concentrations, C_i are calculated as the product of the mean size distribution and the bin width, δd_i .

While number concentration captures the abundance of coarse-mode aerosol particles, quantifying the mass concentration dependence on wind speed allows us to determine whether higher wind speeds produce larger particles. Following the same method of analysis from our wind speed versus number concentration relationships, we maintain the same 6 wind speed bins and mean size distributions in each bin. The total dry mass in each wind speed bin was calculated from the mean size distributions as:

$$M_{d>2} = \frac{\pi}{6} \rho_{\text{salt}} \sum_{i=D_{i>2\mu\text{m}}} C_i D_i^3, \quad (6)$$

where $M_{d>2}$ is the total dry mass in each wind speed bin, $\mu\text{g m}^{-3}$, C_i is the number concentration in bin i , D_i is the

particle diameter of bin i , and the density of salt (ρ_{salt}) is 2200 kg m^{-3} . Standard error is calculated in each bin following Eq. (4). Additionally, we compute instrument counting error for mass for both instruments following Eq. (7):

$$\delta M_{d>2} = \frac{\pi}{6} \rho_{\text{salt}} \left[\sum_{i=D_{i>2\mu\text{m}}} D_i^6 C_i \right]^{\frac{1}{2}}, \quad (7)$$

where $\delta M_{d>2}$ is the counting error in total mass. This formulation explicitly accounts for the influence of larger particles through the diameter term on both the mass and its associated uncertainty. Counting uncertainties in mass are small for both instruments, although they are larger than for number concentration, as mass is influenced by the largest particles, even if they are less numerous. However, these counting errors still remain much smaller than the leg-leg variability represented by the standard error, further showing that mass concentration is not dominated by limitations in particle counting.

We make sure to remove flight legs for which the average mass exceeded $100 \mu\text{g m}^{-3}$. These values represent extreme outliers relative to the typical range of observed GCCN mass and likely reflect measurement artifacts or anomalous sampling conditions. Excluding these cases prevents a small number of unusually large values from disproportionately influencing the statistical analysis. The resulting mean dry mass for the CAS is $10.3 \mu\text{g m}^{-3}$ (median = $7.4 \mu\text{g m}^{-3}$) and $12.2 \mu\text{g m}^{-3}$ (median = $3.6 \mu\text{g m}^{-3}$) for the CDP (Fig. 3).

2.4 Microphysics Model

To assess the potential role of giant cloud condensation nuclei (GCCN) in warm rain formation, we use a condensational and collisional super droplet model (Shima et al., 2009) to simulate cloud droplet evolution from the cloud base to cloud top. A column of air is lifted (details are provided below), resulting in saturation and condensational growth, followed by collision-coalescence and rainout. Haze particles grow, and a fraction activates to form cloud droplets as they are lofted above the cloud base, allowing us to determine their influence on cloud microphysics and precipitation initiation. This Lagrangian model tracks the growth of any specified dry aerosol size distribution, which may include a supermicron (GCCN) distribution. Multiple droplets are represented by a single computational particle with attributes including the multiplicity, the dry size, the wet size, and the location.

Since the model is stochastic, we use an ensemble size of 16 for each case, with 20 200 super droplets across the whole cloud. The cloud thickness is 400 m, representing a typical precipitating warm cloud. The submicron aerosol is represented with a log-normal distribution with a fixed median radius of $0.1 \mu\text{m}$ and a fixed geometric standard deviation of 1.835, consistent with the 7th modal aerosol module (MAM7) from the 5th Community Atmosphere Model (CAM5) (Liu et al., 2012), except to avoid complicating the activation process, we only simulate particles larger than

$0.1 \mu\text{m}$ in radius. The updraft velocity, w , as a function of time is given by Eq. (8), which linearly decreases to zero by $t = 800 \text{ s}$:

$$w(t) = w_0 \left(1 - \frac{t}{800 \text{ s}} \right) \text{ if } t < 800 \text{ s else } 0, \quad (8)$$

There is no downdraft in this simulation. w_0 represents the peak updraft speed (1 m s^{-1}). The resulting vertically integrated liquid water path (LWP) after lifting is about 385 g m^{-2} , which is representative of the thicker parts of marine boundary layer clouds that are likely to be precipitating (Rémillard et al., 2012). The simulation assumes an initially uniform RH of 99 %. The column has a uniform pressure of 1000 hPa and a uniform temperature of 300 K. With such a thin cloud, this is a decent approximation. Pressure changes and latent heating are neglected. These conditions produce a LWP of 385 g m^{-2} which is deemed the “base” LWP for these simulations, but we also incorporate sensitivity tests with a low LWP of 100 g m^{-2} for simulations to explore the dependence of precipitation on GCCN in thinner clouds (Sect. 3.2).

The submicron aerosol number concentration for the base case is set to 50 cm^{-3} , which is typical of relatively clean marine conditions (Heintzenberg et al., 2000), but we also vary this from 10 to 100 cm^{-3} to explore sensitivity to background submicron aerosol number concentrations. Particles in the accumulation mode and coarse mode are assumed to be hygroscopic (hygroscopicity parameter, “kappa”, of 0.7, Petters and Kreidenweis, 2007). For any desired kappa, we use the transformation $r^3 = 0.7r^3/\text{kappa}'$ to find the corresponding effective dry radius. Using the parametric collision kernel given by Ayala et al. (2008) and the ordinary Hall collision efficiencies, we choose two representative levels of turbulence defined by the eddy diffusivity: $0 \text{ cm}^2 \text{ s}^{-3}$ (no microscale turbulence) and $50 \text{ cm}^2 \text{ s}^{-3}$. For turbulent cases, the Taylor-microscale Reynolds number is fixed at 75.

As the model runs, particles remain within the column, where the updraft initiates their growth via condensation upon reaching supersaturation. They continue growing for 800 s, after which the updraft ceases and drops sediment out. Both condensation and collision coalescence can take place throughout and after the ascent. The total simulation length is 1 h, selected as a typical cloud lifetime, and the vertical resolution is 25 m. Particles are initially uniformly distributed in height. Precipitation is measured as the mass of drops that reach below the cloud base with radii greater than $30 \mu\text{m}$. This threshold is meant to estimate in a real cloud which particles can reach the surface before evaporating.

The goal of these simulations is to determine the effect of GCCN on precipitation, both in terms of mass concentration of GCCN and total concentration of GCCN. To ensure computational tractability, we use GCCN size distributions from only one instrument, the CAS. The mass concentrations from the CAS and CDP, and their relation to number concentrations are quite similar (Fig. 3). So, running the simulations

using data from both instruments introduces considerable redundancy and would yield similar results. We use all 460 flight leg dry size distributions to serve as model input. We extract the slope and intercept parameters from exponentially fitted dry size distributions (Sect. 2.2), and compute the total coarse-mode dry mass of sea salt ($\mu\text{g m}^{-3}$) for each GCCN size distribution using Eq. (8). Coarse-mode dry GCCN mass M_{dry} is determined as:

$$M_{\text{dry}} = \frac{\pi}{6} \rho_s N_{0,\text{dry}} \int_{(D_{\text{dry}})_{\min}=2\mu\text{m}}^{\infty} (D_{\text{dry}})^3 e^{-\frac{D_{\text{dry}}}{D_{e,\text{dry}}}} dD_{\text{dry}}, \quad (9)$$

The density of sea salt, ρ_s , is 2200 kg m^{-3} , M_{dry} is the total dry mass concentration per flight leg (expressed in units of $\mu\text{g m}^{-3}$), $N_{0,\text{dry}}$ is the dry intercept, $(D_{\text{dry}})_{\min}$ is the minimum dry diameter, D_{dry} is the dry diameter (μm), and $D_{e,\text{dry}}$ is the e -folding diameter previously extracted from the exponential fit to the dry size distributions.

3 Results

3.1 Wind speed influence on coarse-mode aerosol number concentration and mass concentration

Both number and mass concentrations increase with wind speed, indicating enhanced production of coarse-mode particles under stronger surface winds (Fig. 4). However, substantial variability exists within each wind speed bin, indicating that wind speed alone does not fully constrain the observed aerosol properties. A similar positive relationship is observed between coarse-mode mass concentration and wind speed for both instruments. The relationship between mass concentration and wind speed also shows a statistically significant relationship for both instruments, with fitted slopes of 0.63 ± 0.51 and $0.31 \pm 0.29\text{ }\mu\text{g m}^{-3}$ for the CAS and CDP, respectively, and coefficients of determination of $R^2 = 0.61$ and $R^2 = 0.54$. While both mass concentration and number concentration increase with wind speed, the large error bars show that substantial variability remains between flight legs.

The slopes for the number concentration–wind speed fits (Fig. 4 for the CAS and CDP respectively (Table 1)), uncertainties represent ± 2 standard errors of the mean (SEM; 95 % CI). The slopes agree within statistical uncertainty. Furthermore, the overall relationship between $N_{d>2}$, of individual flight legs and wind speed for both instruments show statistically significant (Table 1) consistent positive trends, with increasing number concentrations at higher wind speeds. Coefficients of determination are $R^2 = 0.84$ and $R^2 = 0.64$ for the CAS and CDP, respectively. However, there is substantial variability between flight legs, reflected in the large error bars, demonstrating consistence with prior studies (e.g. Colón-Robles et al., 2006) that wind speed alone does not fully explain the observed variability in GCCN number concentrations.

3.2 Modeling GCCN influence on precipitation

To evaluate how variability in observed GCCN properties influences precipitation, we apply the model across the full set of flight-leg size distributions. The resulting relationships between accumulated rainfall and GCCN number concentration and mass concentration are shown in Fig. 5.

The simulations show a clear and consistent enhancement of accumulated rainfall with increasing GCCN mass concentration (Fig. 5a) and number concentration (Fig. 5b). Across all model ensembles, representing different background aerosol number concentrations N_a , median accumulated rainfall increases systematically with GCCN concentration or mass, showing that higher GCCN levels are associated with more efficient warm-rain production over the one-hour simulation. This behavior is seen in the median curves, as well as, in the distribution of individual flight-leg simulations, represented by the scatter. The scatter from the full set of flight legs closely follows the log-binned median response in both panels. This agreement is important because it shows that the relationships in Fig. 5 reflect a consistent response across the full population of observed GCCN size distributions. For the low-LWP simulations (100 g m^{-2}), modeled rainfall magnitudes are also broadly consistent with those reported by Jensen and Lee (2008) under comparable cloud droplet concentrations. Across the overlapping $M_{d>2}$ range, the two modeling approaches generally agree within approximately a factor of three, providing additional support for the magnitude of the GCCN-related precipitation response simulated here.

The low N_a (10 cm^{-3}) ensembles of simulations consistently produce the most rainfall over the simulation, for a given GCCN concentration ($N_{d>2}$, Fig. 5a) or mass ($M_{d>2}$, Fig. 5b). In contrast, the high N_a simulations produce considerably less rainfall, with the “base” ensemble lying below these two limits. The sensitivity of the rain to $N_{d>2}$ and $M_{d>2}$ is much stronger, in a relative sense, for higher aerosol background (larger N_a). This demonstrates that the background aerosol environment of the simulations clearly influences not only the precipitation (as is expected from numerous previous observational and modeling studies), but also helps set the effectiveness of GCCN in influencing warm rain. Under low N_a conditions, the cloud contains fewer competing smaller droplets, which leads to more effective collision-coalescence of droplets, leading to enhanced rainfall. The high N_a ensemble conditions distribute available water across a larger number of droplets, slowing the rate of accumulated rainfall. The shape of these curves is also important. For the low N_a ensembles, rainfall increases at a slower rate with $M_{d>2}$ and $N_{d>2}$, and tends to weaken somewhat at the highest values, particularly for $N_{d>2}$ (Fig. 5a). This implies that continuing to add more GCCN can yield smaller additional increases in accumulated rain at a certain point, (or possibly even drop off). In contrast, accumulated rainfall for the high N_a and “base” ensembles appear to in-

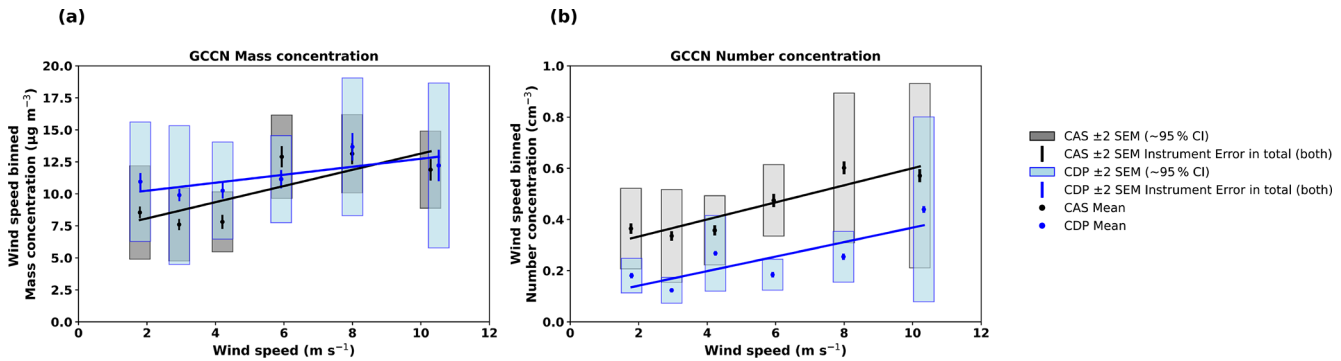


Figure 4. Relationship between wind speed and number concentration, (a) $N_{d>2}$, and (b) GCCN dry mass concentration, $M_{d>2}$ (right). Data are grouped into six wind speed bins, and each point represents the mean value derived from the average dry size distribution within each bin. Shaded boxes denote ± 2 standard errors of the mean (95 % confidence interval) across the flight legs contributing to each bin, while vertical bars represent $\pm 2\sigma$ instrument counting error propagated through the integrated mass concentration or number concentration calculation. Black symbols correspond to CAS measurements and blue symbols correspond to CDP measurements. Solid lines indicate linear least-squares fits to the binned means and are reported in Table 1.

Table 1. Linear regression statistics describing the relationship between wind speed and wind-speed-binned GCCN dry mass and number concentration for CAS and CDP measurements. Slopes (m) in units of $\mu\text{g m}^{-3}$ per m s^{-1} , intercepts (b), and coefficients of determination (R^2) are derived from least-squares fits of the form $y = mx + b$ applied to the mean values within each wind speed bin. The slopes for both GCCN mass concentration and number concentration are positive for both instruments. While all slopes are statistically significant at the 95 % CI, the large uncertainties associated with the mass relationships, particularly for CDP, indicate a more variable dependence on wind speed.

Statistics for wind speed binned GCCN mass and number concentration				
Variable	Instrument	Slope ± 2 SE (95 % CI)	Int. ± 2 SE (95 % CI)	R^2
Mass concentration	CAS	0.63 ± 0.51	7 ± 3	0.61
Mass concentration	CDP	0.31 ± 0.29	10 ± 2	0.54
Number Concentration	CAS	0.033 ± 0.015	0.3 ± 0.1	0.84
Number Concentration	CDP	0.029 ± 0.022	0.1 ± 0.1	0.64

crease more rapidly with $N_{d>2}$ and $M_{d>2}$ when GCCN levels are low. This is consistent with how the low-LWP ensembles also increase rapidly with GCCN levels, but the rain remains very small ($< 10^{-2}$ mm in an hour) until GCCN loadings increase significantly. The limited liquid water available in these conditions initially suppresses droplet growth and hinders collision-coalescence between the droplets formed on submicron aerosol and also the tail of growing GCCN particles accreting cloud droplets. The presence of GCCN alone is not enough to generate rainfall if the available liquid water is limited.

Furthermore, in both panels (Fig. 5), increasing GCCN is associated with more accumulated rainfall, but the correlation with ($M_{d>2}$) is significantly tighter than for number concentration $N_{d>2}$, as seen with the individual simulations for flight leg averages (circles in Fig. 5). This is particularly evident in the low LWP ensemble, where higher GCCN mass produces greater accumulated rainfall than comparable increases in number concentration. These larger particles accelerate the growth of precipitation sized drops, leading to

greater accumulated rainfall. Finally, the influence of microscale turbulence on the collection kernel rate is also important, although more subtle than the effect of background conditions and GCCN. For all ensembles, the median rain accumulation curves for both turbulence levels remain closely clustered together, indicating that the overall trend of the GCCN rainfall relationships is robust across the range of dissipation rates given. However, it is clearly seen that increased level of turbulence increases rain rates across all ensembles. Microscale turbulence (as represented here by turbulent dissipation rate) impacts the efficiency of droplet growth and collision-coalescence, by increasing the likelihood of collisions between droplets. The influence of microscale turbulence is secondary to the effects of background aerosol concentration and LWP. Across most ensembles, the simulations with enhanced turbulence generally produce slightly greater accumulated rainfall, although the corresponding curves remain closely clustered. Overall, the behavior of the ensembles shows that the magnitude of rainfall is dependent not

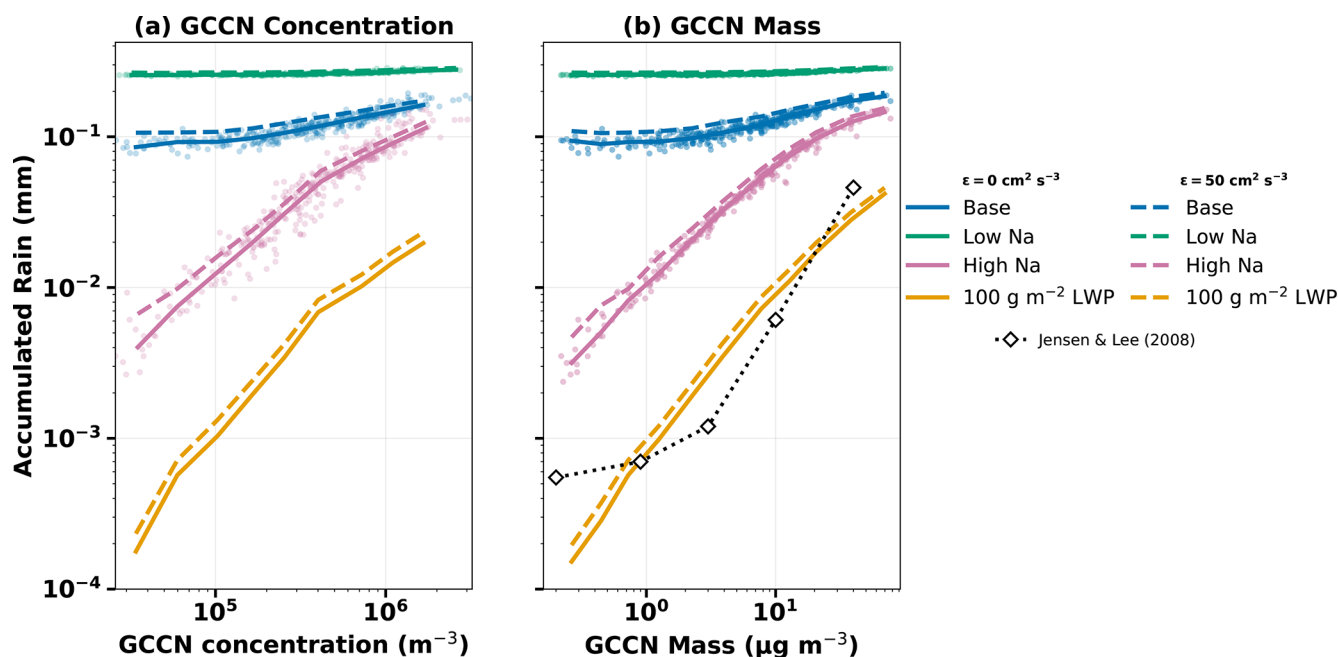


Figure 5. 1-D kinematic super droplet microphysics model simulations showing accumulated rainfall over the 1 h simulation, as a function of GCCN number concentration $N_{d>2}$ (a) and GCCN mass concentration $M_{d>2}$ (b). Model simulations were performed for all flight legs. For the “base” simulation ensemble (blue), the liquid water path is fixed at 385 g m^{-2} and the submicron aerosol concentration N_a is 50 cm^{-3} . Also shown are low LWP (100 g m^{-2}) simulations with the same N_a (yellow). Two levels of turbulent dissipation rate ($\epsilon = 0$ and $50 \text{ cm}^2 \text{ s}^{-3}$) are shown in each ensemble to represent increasing levels of turbulence. High N_a ensembles use a number concentration of 100 cm^{-3} and low N_a ensembles use a number concentration of cm^{-3} . Colored curves show the binned median responses for each ensemble, while the scatter points represent model estimates using individual flight legs. The median response provides a representative description of the ensemble behavior. The Jensen and Lee (2008) cases correspond to surface wind speeds of 1, 2, 4, 8, and 16 m s^{-1} , converted to mass concentrations for particles with $M_{d>2}$ of 0.2, 0.9, 3, 10, and $40 \mu\text{g m}^{-3}$, respectively. The Jensen and Lee (2008) cases have an LWP of approximately 100 g m^{-2} , making them most directly comparable to our low-LWP simulation ensemble and are shown by the dotted black line with open diamonds. Jensen and Lee (2008) report rain rate mm h^{-1} , while our simulations show accumulated rainfall (mm) over a 1 h simulation.

only on the size and number of particles, but on the amount of available cloud water.

3.3 Observations of GCCN influence on precipitation

While the model results in Sect. 3.2 illustrate theoretically how giant CCN can accelerate precipitation rates, it is difficult to provide a direct comparison with the model due to sampling challenges with the dataset. ACTIVATE lacks a radar measurement collocated with the GCCN observations, and the majority of the flight legs are undertaken in a survey-type mode rather than providing repeated sampling of an air mass at different levels. These limitations make the dataset less amenable to closure studies that focus specifically on relating precipitation to cloud and aerosol variability. Instead, we can use aircraft in situ sampling of rain water content (RWC), together with cloud water content (CWC), and cloud droplet concentration (N_d) and develop a composite analysis that is used to assess whether flights with high GCCN concentrations (or mass) contain more or less precipitation than those with low GCCN levels. Rainwater content (RWC) is a direct measure of the condensate converted into

precipitation-sized drops, making it a key proxy for evaluating the role of GCCN in affecting the cloud to rain conversion. To isolate the role of GCCN, we analyze RWC within set bins of LWC and N_d , which constrain the cloud macrophysical and microphysical state and hold it constant. This allows us to evaluate the effect of $N_{d>2}$ and ($M_{d>2}$) on RWC while minimizing the potentially confounding influence of background cloud variability. So, in addition to using the modeling described in Sect. 3.2, we use the in situ measured cloud and precipitation size distribution to derive observational estimates of rainwater content and total liquid water content, which is defined as:

$$\text{LWC} = \text{RWC} + \text{CWC} \quad (10)$$

We set a threshold diameter of $50 \mu\text{m}$ to separate cloud from rain drops (Glienke et al., 2017; Ziemba et al., 2023). As we saw in the ensemble simulations (Fig. 5), GCCN can accelerate collision-coalescence, which manifests in an increased rate of conversion of CWC to RWC (Posselt and Lohmann, 2008). It is widely appreciated that the rate of conversion of cloud to rain in a cloud is strongly dependent upon the

amount of condensate produced in an updraft by condensation (LWC), and is sensitive to the cloud droplet concentration, N_d , as higher droplet numbers suppress precipitation formation by limiting droplet growth (Berry and Reinhardt, 1974; Khairoutdinov and Kogan, 2000; Wood, 2005). ACTIVATE does not have aircraft remote sensors that provide liquid water path (LWP) measurements. Instead, we can use the total LWC, provided in Eq. (10) as the sum of the CAS CWC and the 2D-S RWC (precipitation water), where the 2D-S RWC is calculated using only bins with diameters greater than $50\text{ }\mu\text{m}$ to avoid double counting. We determined in-cloud conditions (N_d , CWC) using the CAS for the results shown here, but an equivalent analysis using the results determining cloud using the CDP (not shown) yielded similar conclusions in terms of the correlation with GCCN. Equivalent sensitivity analyses using CDP defined in-cloud conditions and combined CAS and CDP in-cloud conditions yielded the same overall conclusions; therefore, only the CAS defined analysis is shown.

Our analysis here is carried out using 1 Hz in-cloud data from the CAS and 2D-S because coalescence tends to be strongly spatially variable, so the use of leg means is inappropriate. To gather in-cloud data, we take measurements from above cloud base legs (ACB) and below cloud top legs (BCT), both of which focus on cloud sampling. The ACB and BCT legs are combined into a unified dataset. We compute a total in-cloud number concentration N_d from the CAS, along with CWC and RWC estimates from the CAS and 2D-S respectively for every second of cloudy data. Here, N_c is the droplet number concentration measured by the CAS, for diameters smaller than $50\text{ }\mu\text{m}$, while N_r is the raindrop number concentration measured by the 2D-S from 62.70 to $1464.90\text{ }\mu\text{m}$ diameter. The total number concentration used in this analysis is N_d , which is $N_r + N_c$. In-cloud conditions from the CAS are identified using a CWC threshold $> 0.01\text{ g m}^{-3}$, ensuring that only data points within cloud are considered. Raindrop number concentration is also estimated but are 2–3 orders of magnitude smaller than N_c and therefore do not contribute substantially to total droplet number concentration. RWC estimates use bins with diameters of 62.70 to $1464.90\text{ }\mu\text{m}$ captured from the 2D-S via the following Eq. (10):

$$\text{RWC} = \frac{\pi}{6} \times \rho_w \sum_{i=6}^{128} n_i \times D_i^3 \Delta D_i \quad (11)$$

The density of water, $\rho_w = 1000\text{ g m}^{-3}$, n_i is the observed, binned size distribution (dn/dD) in bin i , D_i is the bin central diameter, and ΔD_i is the bin width. The 2D-S has 128 bins, but the first 5 are highly uncertain due to poorly-defined sizing and sample volume for the smallest rain drops, and these are not used. The smallest 2D-S bins also overlap with the largest CAS bins, so our approach also avoids the potential for double counting. The CWC and RWC are summed to compute the total liquid water content (g m^{-3}) per second, Eq. (11).

To analyze the fractional contribution of rainwater to the total liquid within the cloud, we compute the percentage ratio of rainwater to total condensate:

$$\xi = \frac{\text{RWC}}{\text{LWC}} \times 100 \quad (12)$$

All the flights and legs used for the previous analysis are included. Based on our understanding of precipitation formation, ξ will increase as the total condensate level increases and is expected to also be higher for low N_d . By controlling for these variables, we can then investigate the potential sensitivity to GCCN mass concentration and number concentration (as is suggested by the microphysical modeling, Sect. 3.2). To control for these variables, we place ξ into discrete bins of LWC and N_d . These bins are logarithmically spaced (4 in each dimension to give 16 total bins) to capture observed variability. We then take the mean RWC and LWC in each of the bins and compute ξ as the ratio of these means, i.e., $\xi(\text{LWC}, N_d) = \text{RWC}/\text{LWC} \times 100$. The mean N_d over all cloudy data is 125 cm^{-3} (10th–90th percentile: 15 – 235 cm^{-3}), and the mean LWC is 0.17 g m^{-3} (10th–90th percentile: 0.047 – 0.32 g m^{-3}).

As expected, high LWC and lower N_d are two primary controls on rainwater content (Fig. 6), so we can use this as a framework to evaluate the potential impact of GCCN on RWC. However, this analysis does not currently isolate the impacts of GCCN on this specific measure of precipitation efficiency, defined here by the ratio of RWC to LWC. The model results (Sect. 3.2), showed that, while both GCCN number concentration, $N_{d>2}$ and mass concentration, $M_{d>2}$ impact precipitation, $M_{d>2}$ seems to have a better correlation than number concentration. We perform our observational rainwater content analysis for both GCCN variables. To do this, we subdivide the dataset into cases with high and low $M_{d>2}$ (and a separate split for $N_{d>2}$). For both GCCN variables, we combine the CAS and CDP measurements to provide a single flight-mean estimate of GCCN number concentration and mass. We use our previously calculated leg-mean $M_{d>2}$ (Sect. 2.3) for each instrument and then combined across the CAS and CDP for each flight. This approach incorporates information from both probes while allowing the analysis to center on the distinction between the effects of $M_{d>2}$ and $N_{d>2}$ rather than the differences between the instruments. Because the cloudy data cannot be precisely collocated with the out-of-cloud GCCN measurements, which are also made using different flight legs, and flights are relatively short, we decided to use the flight-mean values (across all legs used to determine means for each flight) and then assign all the cloud data from the cloudy legs from that flight as high or low GCCN clouds on the basis of the flight mean GCCN mass or number concentration, depending on the analysis. This use of flight-mean GCCN value introduces uncertainty because substantial leg-to-leg variability occurs within individual flights, resulting in overlap between the leg-mean values associated with the “high” and “low” GCCN flight

categories. For the combined CAS and CDP measurements used in this analysis, the difference between the mean high and low GCCN populations is approximately 2.0 times the median in-flight standard deviation for $M_{d>2}$ and 2.4 times the median in-flight standard deviation for $N_{d>2}$. Thus, the flight-mean categories separate relatively higher and lower GCCN environments on average, but the overlap among individual legs introduces considerable uncertainty, particularly for the mass-based classification. We produce an average GCCN mass for each of our 75 flights. To categorize each flight, we take the top 50 % of flight mean $M_{d>2}$ (or $N_{d>2}$) (37 flights) as “high” GCCN and the bottom 50 % (38 flights) as “low”. To assess the robustness of our results using 50 % split categories, we repeated the same analysis with a “high” category of the top 20 % of flights and a “low” category of the bottom 20 % of flights. We also tested a “high” category of the top 40 % of flights and a “low” category of the bottom 40 % of flights (not shown). We then separate the calculated RWC data into high and low GCCN flight categories (Fig. 7).

RWC is evaluated as a function of total LWC and N_d for both the “high” and “low” GCCN categories and higher RWC/LWC is generally observed in the high GCCN category relative to the low GCCN category. Ratios were calculated only in bins where the combined number of available cloudy 1 Hz data points across the high and low GCCN subsets is greater than or equal to 100, depicted in Fig. 7; bins that did not meet these criteria are masked. This visualization provides a normalized metric for comparing the relative impact of GCCN under varying cloud conditions.

To assess the statistical significance of RWC/LWC differences between high and low GCCN conditions (either using $M_{d>2}$ or $N_{d>2}$ to characterize GCCN) we applied a bootstrap resampling approach within each joint N_d and LWC bin. Bootstrapping is well suited for this analysis because the flight-mean data are unevenly distributed across bins and may not follow a normal distribution, violating assumptions of traditional parametric tests. For each bin, we generated 10 000 bootstrap realizations by resampling flights with replacement separately for the high and low subsets. For each realization we computed the ratio of mean RWC/LWC between the two groups, producing a distribution of possible outcomes for each bin. This resulted in a distribution of possible outcomes per bin, from which we computed 90 % confidence intervals, mean values, and the percentage of bootstrap samples indicating an enhancement in RWC/LWC in the presence of GCCN.

Figure 7 shows that higher GCCN conditions are generally associated with enhanced RWC/LWC for the vast majority of the LWC– N_d bins for both $M_{d>2}$ and $N_{d>2}$. This enhancement is especially robust (high confidence) in the central, more heavily populated bins of the distribution, located at moderate levels of LWC and N_d , where the percentage of bootstrap realizations greater than 1 is near 100 % and present 90 % confidence intervals entirely above unity. In contrast, a small number of less populated bins (gray boxes

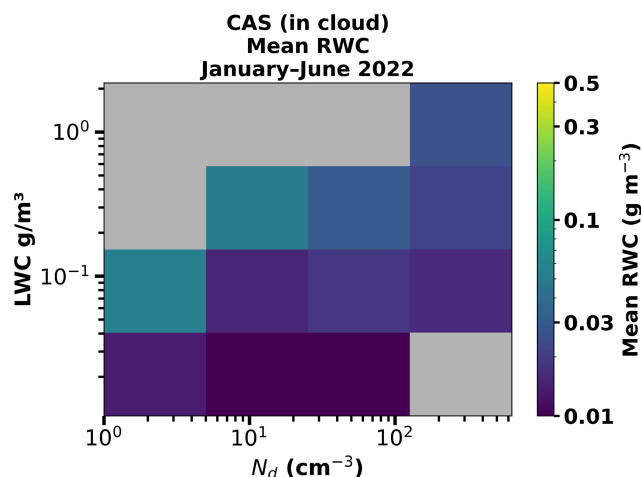


Figure 6. Mean RWC as a function of LWC and N_d for the CAS. Dark purple indicates lower mean RWC and light blue to yellow indicates higher mean RWC. Gray bins indicate bins where fewer than 100 valid data points are available across the compared subsets and are excluded from analysis. Highest levels of mean RWC are observed for moderate LWC– N_d . We found our analysis not to be sensitive to the choice of percentile threshold used.

in Fig. 7) show weaker or even negative association with GCCN. These areas indicate that the GCCN-associated enhancement is not uniform across all cloud conditions. It is dependent on the background cloud microphysical properties and is stronger under a subset of the observed cloud conditions.

The overall enhancement response is largely statistically similar for both $M_{d>2}$ and $N_{d>2}$. Although the magnitude of the High/Low ratio differs between the two variables for individual bins, neither metric produces a consistently larger response and the 90 % confidence intervals for most corresponding bins overlap. This similarity may partly reflect the positive covariance between flight-mean $M_{d>2}$ and $N_{d>2}$ ($R^2 = 0.22$; Fig. S2), which limits our ability to independently distinguish their respective influences in the observational dataset. Thus, while the model results from Sect. 3.2 show that GCCN mass concentration may be a better predictor of precipitation response than GCCN number concentration, the observations do not clearly distinguish this enhanced precipitation response to mass concentration compared with number concentration. Nevertheless, the observational results are consistent with the modeling results in showing enhanced precipitation under higher GCCN conditions. In several of the most densely sampled LWC– N_d bins, RWC/LWC is enhanced by approximately a factor of 2 or more under both higher GCCN mass concentration and higher GCCN number concentration. The observations therefore support an overall association between GCCN and enhanced precipitation response.

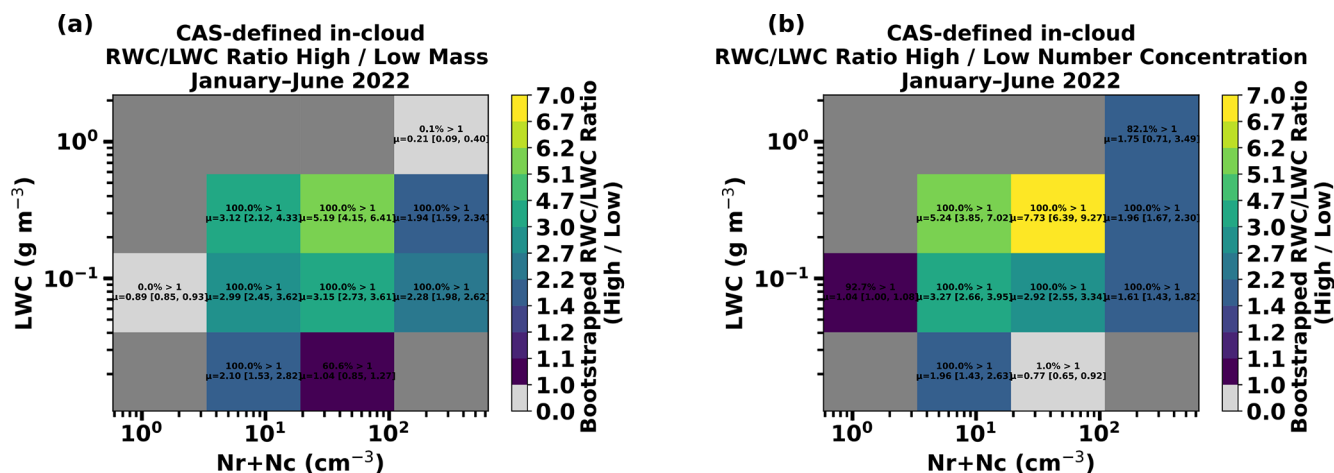


Figure 7. Observationally derived precipitation efficiency responses, defined as RWC/LWC, across bins (boxes) of LWC and $N_r + N_c$ for cases with high versus low $M_{d>2}$ (a) and high versus low $N_{d>2}$ (b). GCCN mass and number concentration are derived from the combined CAS and CDP measurements, while the CAS is used to define in-cloud conditions. Colors indicate the bootstrapped ratio of mean RWC/LWC in high GCCN cases relative to low GCCN cases within each bin (High/Low), while light gray bins indicate High/Low ratios with less than unity. Text within each box shows the percentage of bootstrap realizations producing ratios greater than 1, the mean ratio, and the 90 % confidence interval of the bootstrap distribution. Light gray bins indicate bins where fewer than 100 valid data points are available across the compared subsets and are excluded from analysis. Dark gray bins met the minimum sampling threshold but yielded bootstrap distributions whose mean ratio did not exceed 1, indicating no statistically robust reduction of RWC/LWC under higher GCCN conditions in those bins.

4 Discussion

Together the ACTIVATE observational analysis in addition to results from the microphysics model provide a preponderance of evidence that GCCN are able to exert a significant influence on warm-rain formation. Both the in-situ measurements and model simulations show that increasing GCCN is associated with enhanced precipitation production. The model simulations show that $M_{d>2}$ exhibits a stronger and more consistent relationship than number concentration; however, the observational analysis shows broadly similar RWC/LWC responses to both GCCN metrics and does not clearly distinguish their individual influences. Some of this observational ambiguity may reflect greater uncertainty in $M_{d>2}$ than in $N_{d>2}$. $M_{d>2}$ is more sensitive to the sampling and sizing of relatively rare large particles, and the uncertainty in particle diameter can therefore disproportionately affect the derived mass concentration. This additional uncertainty could reduce the separation between the High and Low $M_{d>2}$ flight groups (Sect. 3.3) and weaken the observed RWC/LWC response to mass, particularly for flights near the High/Low threshold. Due to the exponential nature of the GCCN size distribution for $D_{\text{dry}} > 2 \mu\text{m}$, GCCN number concentration tends to be dominated by salt particles with sizes close to the $2 \mu\text{m}$ threshold, but these particles are less effective at promoting precipitation than slightly larger particles with dry diameters in the range $4\text{--}10 \mu\text{m}$. This influence of GCCN appears to be captured well using the overall dry mass of salt particles, which suggests a possible way forward to parameterizing the GCCN effects on warm rain

production in marine clouds. Our analyses also provide statistical observational evidence from a large number of clouds that LWC and N_d play an important role in staging the primary conditions for precipitation formation, and that the presence of GCCN can impact how efficiently rain develops. The modeling provides additional support. Varying the submicron aerosol concentrations (N_a) and LWP have leading order influences on the production of warm rain (Terai et al., 2012; Pawlowska, 2003; vanZanten et al., 2005; Wood 2005; Mann et al., 2014). Accumulated rainfall is greatest for low N_a conditions and weakest for low LWP conditions. Low N_a conditions promote collision-coalescence, while low LWP limits droplet growth and suppresses rainfall. Furthermore, RWC/LWC ratios were higher and more consistent for moderate LWC– N_d bins, thus showing strong agreement with model simulations. These comparisons are made within the same LWC– N_d regimes and therefore reduce the influence of differences in these primary cloud properties. The resulting enhancement in precipitation efficiency under higher GCCN conditions supports an association between GCCN and precipitation beyond that attributable to differences in LWC and N_d alone.

As in previous observational studies, our results demonstrate that, on average, higher wind speeds increase both the size and number of GCCN, though the impact is stronger for concentration. This positive relationship between increasing wind speed and GCCN production is consistent with other studies (Woodcock, 1953; Ortiz-Suslow, et al., 2016; Monahan et al., 1983; Banner and Peregrine, 1993; Gemmrich and Farmer, 1999; Lewis and Schwartz, 2004;

Ortiz-Suslow, et al., 2016; Clarke et al., 2006; Colón-Robles et al., 2006; Jensen and Lee, 2008; Jensen and Nugent, 2017). In particular, Colón-Robles et al., 2006, which analyzed coarse-mode aerosols from a similar cloud probe (FSSP) to those used in this study, found a strong positive relationship between $N_{d>2}$ and wind speed ($R = 0.87$, $R^2 = 0.76$) over a wind speed range of approximately $5\text{--}14\text{ m s}^{-1}$, with a slope of 0.023 cm^{-3} (their Fig. 1a). Their wind speed range is very similar to ours, though they are looking at hydrated particles, whereas ours are dry. However, this is still comparable to our results, which show slopes of $0.033 \pm 0.015\text{ }\mu\text{g m}^{-3} (\text{m s}^{-1})^{-1}$ and $0.029 \pm 0.022\text{ }\mu\text{g m}^{-3} (\text{m s}^{-1})^{-1}$, for the CAS and CDP respectively (Table 1). Their correlation with wind speed for the FSSP is stronger than the CDP ($R^2 = 0.64$), but weaker than the CAS ($R^2 = 0.84$). The absolute concentrations reported by Colón-Robles et al. (2006) are generally lower than those observed here, with their Fig. 1a showing values ranging from about 0.05 to 0.3 cm^{-3} compared to about 0.1 to 0.6 cm^{-3} in this study (Fig. 4). Furthermore, their size distribution analysis (their Fig. 2) shows that stronger wind speeds increase particle concentrations across the coarse-mode spectrum, particularly for diameters larger than $10\text{ }\mu\text{m}$ (their Fig. 2a–b), with dry particle measurements from a Giant Nucleus Impactor (GNI) extending to $30\text{--}40\text{ }\mu\text{m}$ (their Fig. 2c). Our dry size distributions also follow exponential decay, with a tail extending to about $20\text{--}25\text{ }\mu\text{m}$ (Fig. 2), a slightly smaller range than in their GNI analysis. However, while both studies show the presence of large dry particles, the number concentrations of these particles in Colón-Robles et al. (2006) decrease rapidly with increasing diameter and remain lower than ours beyond $\sim 15\text{ }\mu\text{m}$ (their Fig. 2c). Our dry size distributions exhibit relatively higher number concentrations across the coarse-mode tail, particularly in the $15\text{--}25\text{ }\mu\text{m}$ range. Though their study was based on 30 min flight legs during the Rain in Shallow Cumulus over the Ocean (RICO) campaign (2004–2005) over the trade winds in the western Atlantic, the similarity in slope, correlation strength, and overlapping wind speed ranges indicates a consistent relationship between wind speed and coarse-mode aerosol number concentration with this study. Although a positive relationship between wind speed and $N_{d>2}$ is observed, the strength of this dependence is weaker than would be expected from sea spray aerosol flux parameterizations, which typically show a much stronger increase in particle production with wind speed (Woodcock, 1953; Monahan et al., 1983; Lewis and Schwartz, 2004).

Furthermore, though our relationship between mass and wind speed is weaker than for number concentration, it is still moderate and positive, consistent with other observational studies (Woodcock, 1953; Liu et al., 2021). For example, Woodcock (1953) reports GCCN mass values of approximately $\sim 8\text{ }\mu\text{g m}^{-3}$ at wind speeds of $5.5\text{--}7.9\text{ m s}^{-1}$ (their Fig. 2). In our study, the mean $M_{d>2}$ is $10.3\text{ }\mu\text{g m}^{-3}$ for the CAS and $12.2\text{ }\mu\text{g m}^{-3}$ for the CDP for a similar mean wind

speed of 5.13 m s^{-1} , which falls within the Woodcock (1953) wind regime, further allowing us to directly compare between studies. Both our study and Woodcock (1953) show considerable amounts of scatter, reflected in the large spread of values within individual wind speed bins (Fig. 4) and the moderate coefficients of determination ($R^2 = 0.61$ and 0.54 for the CAS and CDP, respectively). Despite the agreement in magnitude, the relatively modest coefficients of determination and substantial scatter indicate that the dependence of GCCN mass concentration on wind speed is weaker than would be expected from wind speed driven sea spray production alone. This highlights that variability in $M_{d>2}$ and $N_{d>2}$ relationships cannot be explained by wind speed alone. Wind speed represents only one component of the processes controlling GCCN variability in the midlatitude marine environment.

Potential contributors to the remaining variability in GCCN not explained by wind speed are seasonal dependence, which may reflect changes in meteorological conditions such as rapidly changing wind regimes and sea surface temperature that can lead to changes in production rates or in the size of GCCN that are produced (Chen et al., 1973; Lewis and Schwartz, 2004; Jaeglé et al., 2011; Painemal et al., 2021). There is an expected seasonal dependence on GCCN production because higher sea surface temperatures reduce water viscosity and impact bubble rise speeds and size distributions (Jaeglé et al., 2011). The ACTIVATE measurements used here span January–June, and so we examine the seasonal trend of GCCN mass, slope parameter, and number concentration over the Western Atlantic (Fig. 8). These variables are shown as flight-leg mean values (Sect. 2), organized chronologically by flight date, and grouped by month to highlight seasonal variability. Monthly summary statistics and inter-instrument agreement are also provided in Tables 2 and 3 to further quantify these trends.

In general, months with higher $M_{d>2}$ are associated with larger slope parameters (Table 3), indicating a shift toward relatively larger particles. However, GCCN number concentration does not exhibit a consistent relationship with slope parameter as shown by the lack of a clear connection between monthly number concentration and slope values (Table 3). Agreement between the CAS and CDP varies by variable and by month (Table 2). Across all months, correlations are strongest for number concentration ($R^2 = 0.83$) and slope parameter ($R^2 = 0.70$), while substantially weaker for mass concentration ($R^2 = 0.31$). However, this agreement is not uniform across individual months for any of the variables. For example, correlations are very weak in March for mass ($R^2 = 0.065$), slope parameter ($R^2 = 0.008$), and number concentration ($R^2 = 0.001$), while much stronger in June ($R^2 = 0.805$, 0.832 , and 0.969 , respectively). These results indicate that while the two instruments capture similar overall trends, their agreement depends on both the variable considered and the temporal scale of analysis. For most months, the CAS exhibits a higher monthly median for $M_{d>2}$ and

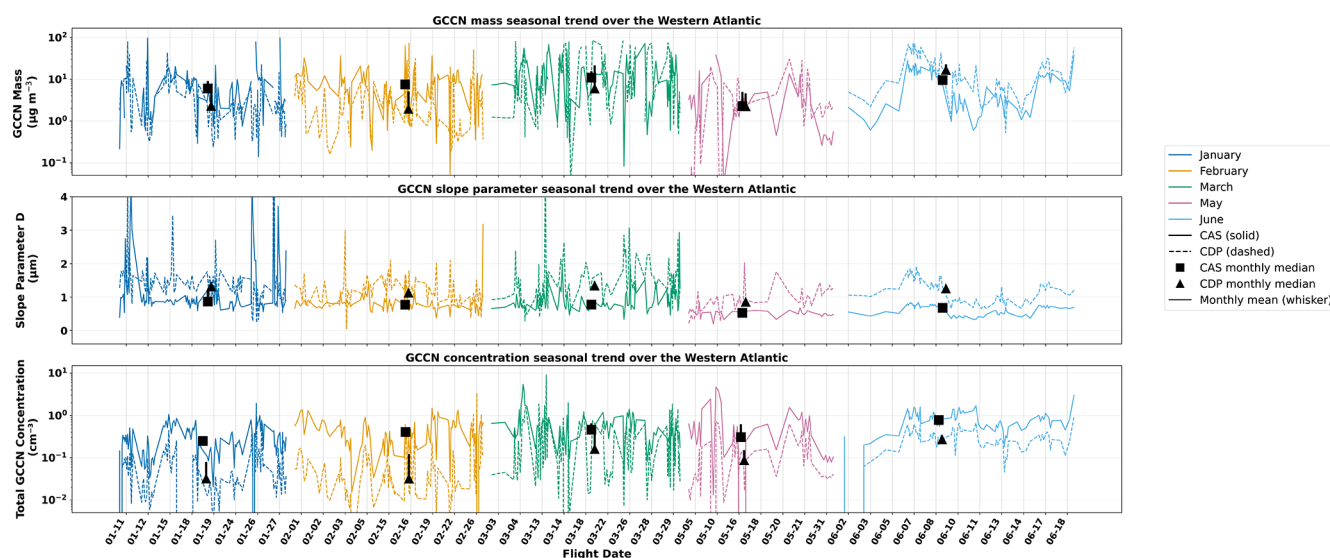


Figure 8. Monthly trends (January–June 2022) of GCCN mass $M_{d>2}$ (top), slope parameter $D_{e, \text{dry}}$ (middle), and total GCCN number concentration $N_{d>2}$ (bottom). Values are shown as flight-leg means plotted by flight date and grouped by month. Solid lines (and black squares for monthly medians) represent measurements from the CAS, while dashed lines (and black triangles) represent measurements from the CDP. Whiskers on the black median symbols show the corresponding monthly means.

Table 2. Monthly correlations between the CAS and the CDP across all months, as well as, within each month. GCCN mass concentration $M_{d>2}$, slope parameter $D_{e, \text{dry}}$, and number concentration $N_{d>2}$ shown as Pearson correlation coefficients r with corresponding coefficients of determination R^2 in parentheses. The full trends between the two instruments across all months are lowest and relatively weak for mass ($R^2 = 0.3$) and highest for the slope parameter ($R^2 = 0.7$) and number concentration ($R^2 = 0.8$). For all three variables, intra-month CAS and the CDP estimates are mostly positively correlated, and are highest during the most in the month of June. The weaker correlation between the two instruments for GCCN mass measurements likely reflects a greater sensitivity of mass to more poorly sampled larger particles, which are less numerous and more variable in space and time, leading to increased measurement differences between instruments.

Monthly Correlations between CAS and CDP ($r(R)^2$)			
Month	Mass ($\mu\text{g m}^{-3}$)	Slope Parameter D (μm)	N_d (cm^{-3})
January	0.69 (0.48)	0.77 (0.59)	0.75 (0.57)
February	0.18 (0.033)	0.61 (0.37)	0.33 (0.11)
March	−0.26 (0.065)	0.089 (0.008)	0.026 (0.001)
May	0.54 (0.29)	−0.66 (0.43)	0.58 (0.34)
June	0.90 (0.81)	0.91 (0.83)	0.98 (0.97)
All months	0.56 (0.31)	0.84 (0.70)	0.91 (0.83)

$N_{d>2}$, and a lower monthly median for slope compared to the CDP (Table 3). Across January through June 2022, there is no clear seasonal trend in $M_{d>2}$, $N_{d>2}$, or slope parameter, as monthly means and medians do not show a consistent increase or decrease over time (Table 3). Instead, values fluctuate between months, including peaks and dips across individual flights within the same month (Fig. 8). GCCN properties across this time period appear to be driven more by individual flight conditions than by a coherent seasonal cycle, though this could be due to the conditions that were selectively targeted with each flight. The absence of a clear sea-

sonal trend suggests that variations in $M_{d>2}$, size, and $N_{d>2}$ are not largely driven by large-scale seasonal changes.

However, to further investigate potential drivers of this variability, we also examined the seasonal behavior of wind speed, which is a primary control on sea-spray aerosol production. We show the seasonal evolution of GCCN mass and mean wind speed across the Western Atlantic (Fig. 9). This analysis focuses only on mass concentration, as we found it to be more influential in enhancing precipitation formation during our model simulations. As in Fig. 8, variables are shown as flight-leg means organized chronologically by

Table 3. Monthly summary statistics for $M_{d>2}$, slope parameter D , and $N_{d>2}$ measured by the CAS and CDP. Values are reported as monthly means, with corresponding medians in parentheses. Across all months, the CAS exhibits higher mean $N_{d>2}$, while the CDP generally shows larger slope parameters, indicating a greater relative contribution from larger particles. For January, February, and May, the CAS shows higher mass measurements, but the CDP records higher mass in March and June. No clear or consistent seasonal trend is observed across the sampled months.

Monthly Summary Statistics (mean(median))						
Month	CAS N_d (cm^{-3})	CDP N_d (cm^{-3})	CAS Slope Parameter D (μm)	CDP Slope Parameter D (μm)	CAS Mass ($\mu\text{g m}^{-3}$)	CDP Mass ($\mu\text{g m}^{-3}$)
January	0.31 (0.25)	0.08 (0.032)	1.2 (0.87)	1.4 (1.3)	9 (6)	5.6 (2.3)
February	0.47 (0.4)	0.12 (0.032)	0.82 (0.77)	1.2 (1.1)	9 (7.5)	5.1 (1.9)
March	0.65 (0.46)	0.47 (0.16)	0.91 (0.78)	1.5 (1.3)	15 (11)	21 (6)
May	0.62 (0.31)	0.15 (0.087)	0.53 (0.53)	0.9 (0.86)	5 (2.3)	4.6 (2.2)
June	0.79 (0.78)	0.29 (0.27)	0.63 (0.68)	1.2 (1.3)	9.7 (9.6)	23 (17)

flight date and grouped by month to highlight seasonal variability.

Wind speed exhibits substantial variability between individual flight legs but does not show a systematic increase or decrease from January through early June (Fig. 9). A corresponding scatterplot with individual leg means, colored by month, for mass concentration and wind speed is also exhibited in Fig. S1. While a modest increase in both wind speed and $M_{d>2}$ is observed toward the final flight dates in late June, this is not sustained throughout all months. For example, the short periods of elevated mass for particular flights (e.g. 13 and 14 March) occurs without a corresponding substantial increase in wind speed. Short-term decreases in both variables, such as those observed in early June, suggest some co-variability, but this relationship is not consistent temporally. These results are consistent with the binned analysis presented in Sect. 3.1, where $M_{d>2}$ showed only a moderate dependence on wind speed. The wind speeds recorded in ACTIVATE are relatively low; ($< 16 \text{ m s}^{-1}$). It is possible that this relationship between $M_{d>2}$ and wind speed might be stronger, given more extreme wind speeds. Overall, these results indicate that while wind speed likely contributes to GCCN production on shorter timescales, it does not appear to be the dominant driver of changes in GCCN mass concentration.

The potential influence of sea surface temperature (SST) was not explicitly tested in this study. A more targeted analysis using SST measurements, after accounting for the dependence of GCCN on wind speed, would help determine whether SST explains additional variability in GCCN production. Despite the consistency of the results presented here, several limitations of the analysis should be considered. First, while the ACTIVATE campaign contains an unprecedented amount of data due to its numerous flights, the aircraft did not contain a radar, which limits our ability to directly measure precipitation. Instead, we rely on in-situ measurements to derive rainwater content, which does not capture the precipitation falling out of the clouds and reaching the ground. Addi-

tionally, the relatively short duration of individual flight legs limits the sampled air volume, $\sim 6 \times 10^{-3} \text{ m}^3$ for the CAS and $\sim 7.7 \times 10^{-3} \text{ m}^3$, thus the need to bin flight legs for wind speed analysis. While counting uncertainties in total $N_{d>2}$ are small ($\sim 0.01 \text{ cm}^{-3}$) and much smaller than the observed leg-to-leg variability, uncertainties in mass are slightly larger because mass is strongly influenced by the largest and least frequent particles. As reflected in Eq. (7), the mass uncertainty scales with D_i^6 , making it particularly sensitive to incomplete sampling of the larger size range. As a result, individual flight legs may under or over represent large particles, leading to increased variability in mass estimates. Grouping flight legs into wind speed bins increases the effective sample volume and reduces this sampling noise.

As for the modeling setup, we must address the immaturity of our implementation of turbulence. We naively assumed that the collision efficiency is unaffected by changes in the geometric collision rates, but the total effect of how turbulence should change the collision rates of cloud droplets is considered an unsolved problem due to the extremely high computational cost required to compute such rates. In this study, we found that GCCN generally have a higher potential impact on precipitation than turbulence in each case (Fig. 5). Should any future increases in collision efficiency caused by turbulence be found to occur, it could ultimately change this finding. Regardless, we are much more confident in the robustness of the sensitivity of each non-turbulent case to GCCN.

Possible future work could build on this analysis by incorporating additional years of ACTIVATE observations, including data from 2020 and 2021, to better assess interannual variability. A pivotal remaining challenge is to more accurately distinguish between the influence of GCCN mass and number concentration on precipitation. Addressing this will require observations spanning a broader range of cloud microphysical conditions and aerosol populations, particularly cases in which GCCN mass and number concentration vary more independently. This approach could help identify other

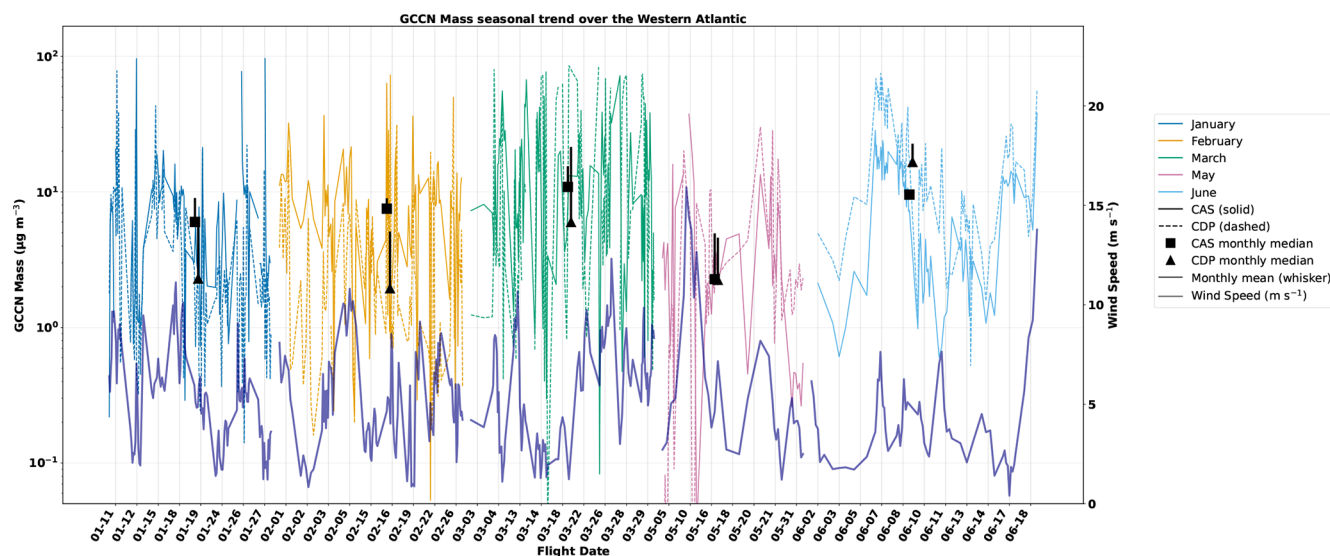


Figure 9. Monthly trends (January–June 2022) of mass are shown as flight leg means plotted by date and grouped by month. Solid and dashed lines represent measurements from the CAS and CDP, respectively. Black squares (CAS) and triangles (CDP) indicate monthly medians, with whiskers showing corresponding monthly means. The purple line represents the mean flight leg wind speed (Sect. 3.1).

cloud environments, in which precipitation is more sensitive to GCCN mass versus number concentration. Furthermore, the use of instruments specifically designed to measure giant nuclei, such as a giant nucleus impactor, would provide improved characterization of the largest particles that dominate mass and are most relevant for collision-coalescence processes. These improvements would help further constrain the role of GCCN in precipitation formation and refine their representation in both observational and modeling frameworks. A step forward in this direction is demonstrated by some studies using cloud water samples to infer the amount of sea salt in clouds based on the mass concentration of its dominant species, sodium and chloride (Dadashazar et al., 2017; MacDonald et al., 2020; Gonzalez et al., 2022; Zeider et al., 2025; Ajayi et al., 2026).

5 Concluding Remarks

This study combined in-situ aircraft observations from the ACTIVATE campaign with super-droplet simulations to investigate the role of GCCN in warm-rain formation in marine low clouds over the western North Atlantic. Both observations and model simulations consistently indicate that increasing GCCN abundance enhances warm-rain formation, particularly in clouds with sufficient LWC and moderate N_d . While both GCCN number concentration and mass concentration increase with wind speed, the observed dependence is weaker than would be expected from sea-spray aerosol flux parameterizations, indicating that additional environmental factors contribute to GCCN variability in the marine atmosphere.

Together, the observational and modeling results provide evidence that GCCN influence precipitation development through enhanced collision-coalescence. $M_{d>2}$ was found to be a better predictor of precipitation enhancement than $N_{d>2}$ alone during model simulations. Furthermore, our results are broadly consistent with previous studies that have identified positive relationships between wind speed and coarse-mode sea-salt aerosol production. However, the variability in our wind speed results demonstrates that wind speed alone is not enough to explain GCCN variability in mass and number concentration. Additional factors, including meteorological conditions, may contribute to the observed scatter.

Several limitations should be considered when interpreting these results. The ACTIVATE aircraft observations do not directly measure surface precipitation and rely instead on in-situ cloud and rainwater measurements. Additionally, estimates of GCCN mass concentration remain sensitive to sampling uncertainties associated with the largest and least frequent particles. The representation of turbulence within the microphysical model also introduces uncertainty in the simulated precipitation response. Despite these limitations, the consistency between the observational and modeling analyses provides strong evidence that GCCN can substantially influence warm-rain formation in marine low clouds. Overall, these findings improve our understanding of aerosol-cloud-precipitation interactions by demonstrating that coarse-mode sea spray aerosols can enhance precipitation efficiency even at relatively low concentrations. The stronger relationship between precipitation and GCCN mass concentration in the modeling results may provide a useful framework for representing GCCN effects in future observational and modeling studies, but further analyses incorporating additional years

of observations, a broader range of cloud conditions, and targeted measurements of coarse-mode sea salt particles are needed to better distinguish the relative roles of GCCN mass and number concentration and constrain their representation in microphysics models.

Code availability. The code used for data processing, analysis, and figure generation in this study is publicly available on GitHub. Scripts related to the CAS instrument are available at <https://github.com/katherineemifsud/ACTIVATE-2024-2025/tree/master/CAS> (last access: 20 October 2026) and scripts related to the CDP instrument are available at: <https://github.com/katherineemifsud/ACTIVATE-2024-2025/tree/master/CDP> (last access: 20 October 2026). These repositories include routines for observational size distribution processing, statistical analysis, model comparison, and rainwater content analysis. The code used for the model simulations is available at https://github.com/jbarr444/SDM_GCCN (Barr, 2026).

Data availability. Data from the ACTIVATE campaign are publicly available through the NASA Atmospheric Science Data Center (ASDC) at <https://doi.org/10.5067/SUBORBITAL/ACTIVATE/DATA001> (NASA Langley Research Center, 2020) and were accessed through the Sub-Orbital Order Tool (SOOT) at <https://asdc.larc.nasa.gov/soot/browse/ACTIVATE/2022> (last access: 1 July 2026). The datasets are organized by year, aircraft platform, and instrument, with variables provided within each instrument category.

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Author contributions. KM and RW conceived the study. JB performed the model experiments. KM conducted the analysis and prepared the manuscript with contributions from co-authors. 2D-S and FCDP datasets were created by CV. GD, YC, and JD produced DLH datasets. RM, LZ, EC, CR, MS contributed the CAS and CDP datasets. AS served as the principal investigator of ACTIVATE and provided the scientific leadership and campaign coordination that made this study possible.

Competing interests. At least one of the (co-)authors is a member of the editorial board of *Atmospheric Chemistry and Physics*. The peer-review process was guided by an independent editor, and the authors also have no other competing interests to declare.

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