



1 Coarse-mode sea spray aerosols enhance warm rain formation in 2 in marine low clouds.

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

32 1 Introduction

33 Cloud-aerosol interactions have a significant impact on climate, particularly through their influence on marine
34 stratocumulus clouds, which cover roughly 20% of the planet's oceans and play a dominant role in modulating Earth's
35 radiative energy balance (Hartmann and Short 1980). These interactions affect both cloud microphysics and
36 macrophysics, including cloud albedo and lifetime, thereby altering the reflection of solar radiation and the overall
37 radiative forcing of the climate system (Seinfeld et al., 2016). By regulating heat, moisture, and momentum exchanges
38 between the ocean and atmosphere, the stratocumulus-topped boundary layer (STBL) profoundly influences large-
39 scale climate systems (Wood 2012). Marine stratocumuli reflect a large portion of the Earth's incoming solar radiation,



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

63 **1.1 Giant cloud condensation nuclei**

64 Drizzle formation can be achieved through the existence of a tail of large "collector" droplets formed on giant cloud
65 condensation nuclei (GCCN) that can initiate collision-coalescence (Ludlam 1951; Johnson 1982; Feingold et al.,
66 1999; Jensen and Lee 2008, Jensen and Nugent 2017; Dziekan et al., 2021). In marine environments giant sea spray
67 particles with dry diameters larger than $\sim 2\ \mu\text{m}$ enhances collision-coalescence, in contrast to smaller particles that
68 suppress it. Based on this finding, we define GCCN as salt particles with dry diameters larger than $2\ \mu\text{m}$.
69 Condensational growth of GCCN from the ocean surface through the cloud is important for growing the particles to a
70 size where they can initiate collision-coalescence (Ludlam 1951; Jensen and Nugent 2017). Despite a solid theoretical
71 foundation, this effect has not yet been incorporated into physical parameterizations used in bulk microphysical
72 models, which may lead to an underestimation of warm rain formation in some clouds (Jensen and Nugent 2017;
73 Dziekan et al., 2021). Cloud droplets grown on GCCN are highly concentrated solutions compared with the majority
74 of cloud droplets, which are grown on much smaller accumulation mode aerosol particles (with typical dry diameters
75 of $\sim 0.1\ \mu\text{m}$). This accelerates, via the Raoult effect (Jensen and Nugent 2017), the condensational growth rates of



76 droplets grown on GCCN compared with those forming on accumulation mode CCN. In some cases, this may lead to
77 a faster onset of warm rain (L'Ecuyer et al., 2009). The fundamental challenge to representing GCCN in microphysical
78 models is that the condensational growth rate of cloud droplets depends not only on their size but also on the mass of
79 salt they contain. GCCN-containing droplets must therefore be treated as a distinct category of cloud hydrometeor,
80 increasing model complexity. Posselt and Lohmann (2008) attempted to incorporate GCCN effects into a global
81 climate model by parameterizing GCCN by assuming they directly activate into raindrops; however, this approach
82 neglects detailed condensational growth processes. It is computationally-challenging to accurately capture this
83 aerosol-dependent condensational growth in microphysical models, but it can be achieved using super droplet methods
84 (Shima et al., 2009; Shima et al., 2020; Li et al., 2022; Zmijewski et al., 2024), or by using two-dimensional bin
85 microphysical schemes that independently keep track of hydrometeor wet sizes as well as the aerosol dry size (e.g.,
86 Flossman 1985; Lebo and Morrison 2013). Since droplets formed on GCCN remain highly concentrated salt solutions,
87 they can more easily resist evaporation and the larger GCCN can even continue growing by condensation as they
88 descend through (weakly) subsaturated downdrafts (Jensen and Nugent 2017). This contrasts with smaller cloud
89 droplets grown on accumulation mode aerosol, which typically lose mass and may completely evaporate in similar
90 conditions.

91
92 Despite advances in understanding GCCN's role in collision-coalescence growth, significant uncertainties remain due
93 to wide variations in the sizes and concentrations reported in prior studies. For example, Jensen and Nugent (2017)
94 found that introducing 1 μm dry radius GCCN at concentrations of 0.3 cm^{-3} could influence drizzle in a microphysical
95 parcel model, while Feingold et al. (1999) introduced 20 μm droplets, a proxy for condensationally-grown GCCN,
96 into large eddy simulations at concentrations as low as 10^{-4} to 10^{-2} cm^{-3} and found impacts on precipitation for the
97 higher concentrations. Because condensational growth of GCCN was not treated explicitly in Feingold et al. (1999),
98 it is difficult to compare this result with other studies, highlighting major differences in model assumptions and
99 methods. Lowenstein et al. (2010) used aerosol and cloud probe observations of hydrated giant haze particles below
100 marine trade cumulus to initialize a parcel model that produced a tail of large droplets ($r \sim 20\text{-}100 \mu\text{m}$) above cloud
101 base that matched observations fairly on two flight days. However, model simulations were not conducted without
102 GCCN, so the overall impact of the GCCN is unclear.

103
104 Dziekan et al. (2021) have performed the most sophisticated and realistic simulations of GCCN influences on low
105 cloud precipitation to date, using a super droplet model embedded within a large eddy model. The model explicitly
106 computes both the condensational and coalescence growth of GCCN, and shows that GCCN from an observed flight
107 over the southeastern Pacific is sufficient to increase cloud base precipitation rate in a marine stratocumulus cloud
108 case by $\sim 50\%$ compared with the case with zero GCCN. A somewhat weaker influence ($\sim 10\%$) was found in a deeper
109 trade cumulus case. The base LES case used in Dziekan et al. (2021) used observed GCCN measurements with near-
110 surface GCCN dry salt mass of $\sim 6 \mu\text{g m}^{-3}$ and number concentration of $\sim 0.22 \text{ cm}^{-3}$ (for dry diameters exceeding $2 \mu\text{m}$)
111 with a near-surface wind speed of 5.4 m s^{-1} . Sensitivity tests were conducted for larger GCCN concentrations/masses



112 to show that further increasing GCCN loading continued to increase precipitation rate, with a factor of 5 increase
113 leading to precipitation rate increases of ~300% (stratocumulus) and ~30% (cumulus) above the rate without GCCN.
114
115 GCCN dry size distributions in previous studies are typically found to decrease approximately exponentially with
116 increasing particle diameter (e.g., Woodcock, 1953; Lewis and Schwartz, 2004; Colón-Robles et al., 2006). Because
117 the observed e-folding diameter for the decrease is relatively small (see section 2.2 below), this means that reported
118 GCCN number concentrations are particularly sensitive to the threshold dry diameter D_{min} used to define GCCN.
119 Although several recent studies have adopted a threshold $D_{min} \sim 2 \mu\text{m}$, studies do not always report what threshold
120 diameter was used or use a hydrated size threshold rather than a dry one (e.g., Colón-Robles et al., 2006), making
121 cross-study comparisons of observed GCCN and their influences on clouds challenging (Cooper et al., 2013).

122

123 This work aims to address critical gaps in understanding coarse-mode cloud-aerosol interactions in marine low clouds.
124 We utilize a robust observational framework, which includes a very large number of flight legs across different seasons
125 and meteorological conditions, established by the NASA Earth Venture Sub-Orbital (EVS-3) Aerosol Cloud
126 meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE) field campaign. Building on previous
127 work (e.g., Woodcock, 1953; Colón-Robles et al., 2006), we first document coarse-mode aerosol size distributions
128 under clear sky conditions below cloud for a wide variety of aircraft flights and examine how near-surface wind speed
129 influences their number and mass concentration and size distribution. We then investigate the extent to which our
130 observed variations in GCCN concentrations can modulate precipitation in marine stratocumulus clouds using a super
131 droplet model. The goal is to determine how each of the observed GCCN distributions alters the cloud droplet size
132 distribution and accelerates precipitation formation under varying aerosol conditions and for a range of plausible
133 macrophysical and microscale turbulence conditions. We also examine the influence of GCCN on observationally
134 derived rainwater content (RWC), calculated from ACTIVATE observations, which provides a purely observational
135 perspective on GCCN-precipitation relationships. Together, the modeling and observation analyses provide a
136 comprehensive understanding of how GCCN impact warm rain formation. This analysis is necessary not only to
137 improve our understanding of aerosol-cloud interactions, but also to contribute to better parameterizations that include
138 the impact of GCCN.

139 **2 Methods**

140 **2.1 Dataset**

141 ACTIVATE is a NASA Sub-Orbital field campaign that flew aircraft and captured data from February 2020 through
142 June 2022 with the intent of providing globally important data detailing changes in warming and cooling planetary
143 feedbacks, atmospheric aerosols and sources, and marine boundary layer cloud systems (Sorooshian et al., 2023). The
144 observations relevant to this study were collected during two deployments spanning January to June 2022 to capture
145 different seasonal states and broaden the range of aerosol and meteorological conditions. Flights were conducted using
146 a twin-aircraft design based out of NASA Langley Research Center in Virginia, with the HU-25 Falcon flying at lower



147 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
148 are used. We utilize measurements taken from the Diode Laser Hygrometer (DLH) which measures relative humidity,
149 Cloud Droplet Probe (CDP), Cloud-Aerosol Spectrometer (CAS), and the two-dimensional Stereo Probe (2D-S), all
150 of which were externally mounted on the Falcon. The CAS measures from 0.5 μm diameter to 50 μm diameter and
151 the CDP measures from 2 μm to 50 μm diameter. Both probes measure cloud droplet size distributions, from which
152 aerosol and cloud droplet number concentration, liquid water content, effective radius, and effective variance are
153 derived (Baumgardner 2001; Lance 2012). The 2D-S measures larger drop number size distribution for liquid and ice
154 both separately and combined; liquid water content; effective diameter and median volume diameter (Kirschler et al.,
155 2023). The 2D-S also measures a broader size range from 29 to 1465 μm diameter. While a Fast Cloud Droplet Probe
156 (FCDP) was available during the campaign, it is not included in this analysis. The FCDP lower diameter threshold is
157 3 μm diameter, so it doesn't quite reach the 2 μm diameter threshold we were looking for. This study focuses on in-
158 situ Falcon data collected between 150 m and 2.5 km altitude. A total of 75 joint flights on 43 days, logging upwards
159 of 120 flight hours were conducted during the 2022 deployment, making it the most comprehensive year due to fewer
160 pandemic disruptions compared to 2020–2021 (Sorooshian et al., 2023).

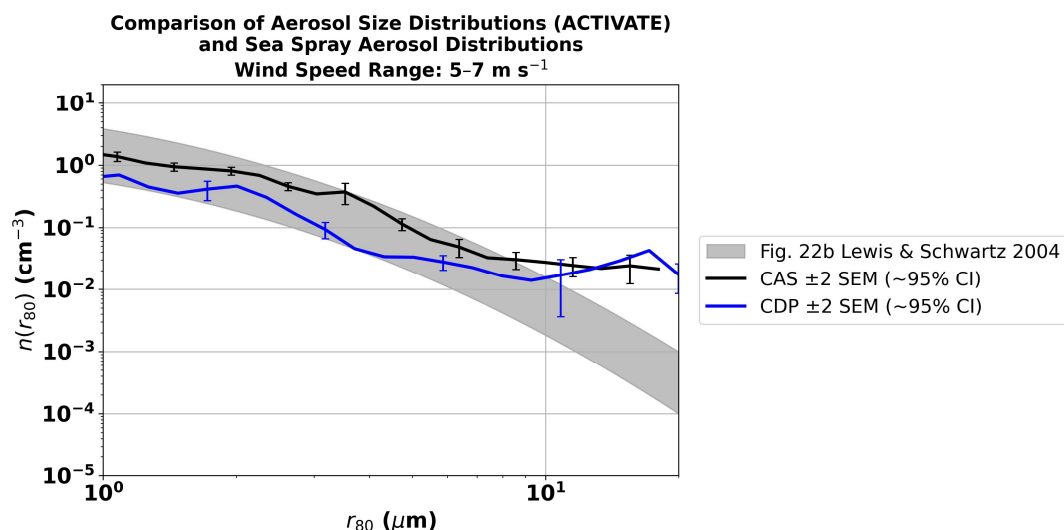
161 We focus on Falcon flight legs classified as BCB (below cloud base) to analyze coarse-mode aerosols (section 2.2-
162 3.2), and ACB (above cloud base) and BCT (below cloud top) when working in-cloud (section 3.3). These legs are a
163 part of “cloudy ensembles”, which are sequences of flight legs conducted in close succession that sample different
164 vertical regions of the cloud and sub-cloud environment, where leg is about 3 minutes long (Sorooshian et al., 2023).
165 The vast majority of available data come from cloudy ensembles, which account for over 90% of the total flight legs
166 in 2022. We did not utilize flight legs from designated “clear-sky” ensembles due to insufficient data (see Sorooshian
167 et al., 2023). Less than 10% of the total ensembles in 2022 are “clear-sky.” The systematic repetition of these flight
168 ensembles has resulted in an extensive and structured dataset (Dadashazar et al., 2022; Sorooshian et al., 2023).
169 However, we aim to use BCB legs without cloud contamination, so to filter for clear sky conditions, we apply a liquid
170 water content threshold of 0.0025 g m^{-3} from the CAS and the CDP and remove data points above this threshold to
171 remove contamination from cloud droplets. We further restrict the dataset by requiring that the precipitation number
172 concentration, defined as the concentration of precipitation-sized particles measured by the 2D-S probe, remains below
173 100 m^{-3} . This additional criterion ensures that drizzle-sized drops are not present. We also remove data points with
174 relative humidity (RH) above 95%, given that there is a ~5% error in RH measurements, to further ensure that the
175 sampled air is subsaturated. Together, these thresholds ensure that the observed hydrated size distributions are indeed
176 characterizing haze droplets in subsaturated conditions without cloud or drizzle droplet interference.

177 **2.2 Constructing dry size distributions**

178 To characterize aerosol concentration above the Western Atlantic, we analyze size-resolved hydrated (ambient)
179 aerosol distributions derived from CAS observations from only the below the cloud base flight legs. Above cloud base
180 and below cloud top legs are used for analysis in section 3.3. We use 1 Hz data for 75 flights on 43 different dates
181 from January to June 2022. Each 1 Hz observation is taken from a typical 3 minute below cloud base (BCB) flight leg
182 and only samples passing our clear sky filtering criteria are retained. Though the CAS measures particles at 0.5 μm to



183 50 μm diameter, we select only measurements corresponding to haze particles (ambient hydrated particles) ranging
 184 from 2.5 μm to 50 μm diameter for both the CAS and CDP, which removes any accumulation mode particles before
 185 constructing our distributions. The CDP range is 2.5 μm to 50 μm diameter. It is important to note that this ambient
 186 size, for most RH values sampled, corresponds to a dry minimum diameter of less than 2 μm , which ensures that we
 187 capture all the sea spray particles with dry diameters greater than 2 μm , even as the RH varies along each flight leg.
 188 After the removal of flight legs exceeding clear-sky filtration process (see section 2.1), the relative humidity analysis
 189 yielded 460 below cloud base legs with a mean altitude of 701 m. To contextualize our ambient (haze) size
 190 distributions, we compare our average ambient CAS and CDP distributions with Lewis and Schwartz (2004). We
 191 compare with their Fig. 22b, which shows a range of average size distributions across multiple studies at an average
 192 wind speed of 5-7 m s^{-1} , shown in Figure 1. Although Lewis and Schwartz (2004) report size distributions across a
 193 broader range of wind speeds, our comparisons are limited to the 5-7 m s^{-1} range, which is closest to the mean wind
 194 speed observed in our dataset (5.13 m s^{-1}) and contains the most robust sampling. Higher wind speed regimes reported
 195 in previous studies are not well represented in our dataset, limiting direct comparison. Their distributions are reported
 196 at 80% RH (r_{80}), so we normalize our ambient measurements to this reference humidity using a hygroscopic growth
 197 factor derived from the mean relative humidity of each flight leg (see below). The resulting r_{80} values are used to
 198 rescale particle sizes while maintaining the measured number concentration distributions. All size distributions used
 199 for this comparison are in $\frac{dN}{d \ln D}$ to maintain consistency with Lewis and Schwartz (2004). We find that our CDP and
 200 CAS ambient distributions in this wind speed range, normalized to r_{80} , generally fall within the 3 standard deviations
 201 of the studies depicted in Lewis and Schwartz Fig. 22b.
 202



203
 204 Figure 1 Normalized average ambient aerosol size distributions measured by the CAS (black) and CDP (blue) during
 205 ACTIVATE flights for wind speeds of 5-7 m s^{-1} . Error bars denote ± 2 standard errors (95% confidence interval)



206 based on variability across contributing flight legs (see section 2.3 for details). The grey shaded region represents the
 207 range of sea spray aerosol size distributions compiled from multiple studies by Lewis and Schwartz.

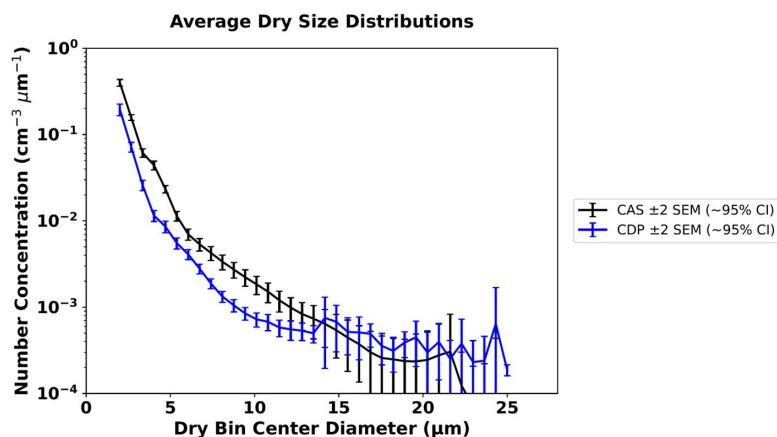
208

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

$$215 \quad g(RH) = \left(\frac{1.7}{1-RH} \right)^{0.31}, \quad (1)$$

216 The growth factor $g(RH)$ is a function of leg-mean relative humidity (RH) only (for coarse mode sea spray particles,
 217 assuming constant sea spray composition), not of diameter and therefore varies across all flight legs (Tang 1996;
 218 Jensen and Lee 2008). Ambient particle diameters are converted to dry diameters by dividing by $g(RH)$, and the
 219 corresponding number size distributions, expressed as $\frac{dN}{dD}$, are transformed into dry-diameter space. Since RH differs
 220 across flight legs, the resulting dry diameter bins vary slightly; therefore, each dry size distribution is interpolated onto
 221 a common dry diameter grid spanning 2 to 25 μm before averaging. Average dry size distributions for each instrument
 222 are compared in Figure 2. We calculate the mean total concentration for the CAS to be 0.56 cm^{-3} and 0.18 cm^{-3} for
 223 the CDP.

224



225

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

229

230 The flight legs in this study from ACTIVATE averaged a sample time of 180 seconds, which is relatively short
 231 compared with some other studies (Cólón-Robles et al., 2006 used circle legs of ~ 30 -minute duration). Low particle



232 counts can result in “shot noise” and a flattening of the size distributions for the lower concentrations of larger sized
233 particles (Baumgardner et al., 2017). To quantify this effect, because low particle counts at larger diameters lead to
234 elevated shot noise, we estimate the relative Poissonian counting error for each size bin using Eq. (2):

$$235 \quad \text{relative error} = \frac{\sqrt{N_{\text{counts}}}}{N_{\text{counts}}}, \quad (2)$$

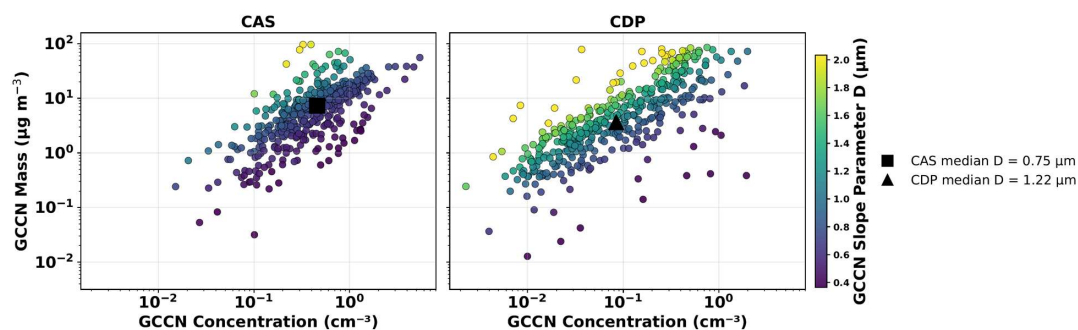
236 This allows us to identify the particle sizes for which the counting statistics are most reliable. Because of the quasi-
237 exponential nature of the coarse mode size distributions and the limited probe sampling volume, the relative error
238 exceeds 100% for many bin diameters above $\sim 10 \mu\text{m}$ in many cases, so we choose to limit our exponential fitting (See
239 equation 2) to the portion of each distribution smaller than $10 \mu\text{m}$ diameter. The fitted parameters ($N_{0, \text{dry}}$ and $D_{e, \text{dry}}$)
240 (See equation 2) and their covariance provides insight into how the abundance (concentration), characteristic size, and
241 the overall mass of coarse-mode aerosol vary together for both instruments, illustrated in Fig. 3.

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

$$249 \quad n(D_{\text{dry}}) = N_{0, \text{dry}} e^{-\frac{D_{\text{dry}}}{D_{e, \text{dry}}}}, \quad (3)$$

250 where $N_{0, \text{dry}}$ represents the intercept and $D_{e, \text{dry}}$ represents the e-folding diameter, which controls the fall-off in
251 concentration with size. This fit is performed using nonlinear least-squares regression, such that all included size bins
252 contribute equally to the regression. As a result, bins with low particle counts, like those at larger diameters, may be
253 subject to greater statistical uncertainty due to counting variability. To mitigate the influence of this “shot noise”, the
254 fitting is restricted to dry diameters $\leq 10 \mu\text{m}$, where particle counts are higher and sampling uncertainty is reduced.
255 The fitted parameters derived from these dry size distributions are further used for analysis in section 3.2. The slope
256 parameter from these fits, displayed in Figure 3, provides a useful framework for looking at how variations in size
257 distribution influence aerosol properties such as number concentration and mass.

258



259

260 Figure 3 shows the relationship between GCCN dry mass and number concentration CAS and CDP observations. Each
 261 point is colored by the fitted slope parameter $D_{e,dry}$, which characterizes the e-folding diameter of the dry size
 262 distribution.

263

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

275 2.3 Wind speed binning

276 Near surface wind speed has been shown to be one of the leading controlling factors on bubble bursting and the
 277 production of sea spray aerosol (Woodcock 1953; Monahan et al. 1986; Banner and Peregrine, 1993; Gemmrich and
 278 Farmer, 1999; Clarke et al., 2006). To explore the relationship between wind speed and coarse-mode aerosol
 279 concentration for aerosol dry diameters larger than $2.0 \mu\text{m}$, which we define as $N_{d>2}$, we first apply a logarithmic
 280 wind profile correction (Stull 1988) to all Falcon in-situ wind speed measurements, standardizing them to 10 m above
 281 sea level. Wind speeds were collected from the NASA Langley in situ aircraft state dataset which provides aircraft-
 282 derived wind measurements (Sorooshian et al., 2023). This correction accounts for variability in flight leg altitude and
 283 uses a roughness length of 0.02 m for open ocean conditions (Peña and Gryning 2008). The mean 10 m wind speed
 284 across all valid below-cloud-base legs is 5.13 m s^{-1} with a standard deviation of 2.65 m s^{-1} .

285



286 Due to the relatively short flight legs in ACTIVATE, we aggregate flight legs into six wind speed bins that capture
287 the full range of observed wind speeds (Fig. 4). Previous observational studies have shown a strong dependence on
288 wind speed for aerosol concentration, particularly in the coarse mode (Woodcock 1953; Lewis and Schwartz 2004;
289 Colón-Robles et al. 2006). Given this established dependence, grouping legs by wind speed allows us to directly
290 compare our results with prior findings (section 4) while increasing the statistical robustness of our results, as the
291 aggregation reduces variability from short sampling times. These bins are designed to represent the overall wind speed
292 range in our dataset, 0 m s⁻¹ 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 m
293 s⁻¹, which we manually selected. The distribution of 10 m wind speeds in our dataset is not uniform. The chosen ranges
294 (0–2.5, 2.5–3.5, 3.5–5, 5–7, 7–9, and >9 m s⁻¹) ensure that each of the 6 bins contains a sufficient number of flight
295 legs while also isolating wind speed ranges commonly discussed in sea spray aerosol literature (Lewis and Schwartz
296 2004, see Fig. 1). We repeated the analysis using smaller binning schemes, 4 and 5 wind speed bins (not shown), and
297 found that the slope of the concentration and wind speed relationship differed by less than 16% compared to the 6-bin
298 case, with all slopes overlapping within their respective uncertainties.

299

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

$$307 \quad SE = \frac{\sigma}{\sqrt{n}}, \quad (4)$$

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

$$315 \quad \delta N = \sqrt{\sum_i C_i}, \quad (5)$$

316 where δN is the total counting error in the integrated concentration, N is the total concentration obtained by summing
317 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
318 mean size distribution and the bin width, δd_i .

319 While number concentration captures the abundance of coarse-mode aerosol particles, quantifying the mass
320 dependence on wind speed allows us to determine whether higher wind speeds produce larger particles. Following the



321 same method of analysis from our wind speed versus concentration relationships, we maintain the same 6 wind speed
322 bins and mean size distributions in each bin. The total dry mass in each wind speed bin was calculated from the mean
323 size distributions as:

$$324 \quad M_{d>2} = \frac{\pi}{6} \rho_{salt} \sum_{i=D_i>2 \mu m} C_i D_i^3, \quad (6)$$

325 where $M_{d>2}$ is the total dry mass in each wind speed bin, $\mu\text{g m}^{-3}$, C_i is the concentration in bin i , D_i is the particle
326 diameter of bin i , and the density of salt (ρ_{salt}) is $2200, \text{kg m}^{-3}$. Standard error is calculated in each bin

327 following Eq. (4). Additionally, we compute instrument counting error for mass for both instruments following Eq.
328 (7):

$$329 \quad \delta M_{d>2} = \frac{\pi}{6} \rho_{salt} \left[\sum_{i=D_i>2 \mu m} D_i^6 C_i \right]^{\frac{1}{2}}, \quad (7)$$

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

336 We make sure to remove flight legs for which the average mass exceeded $100 \mu\text{g m}^{-3}$. These values represent extreme
337 outliers relative to the typical range of observed GCCN mass and likely reflect measurement artifacts or anomalous
338 sampling conditions. Excluding these cases prevents a small number of unusually large values from disproportionately
339 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
340 $12.2 \mu\text{g m}^{-3}$ (median = $3.6 \mu\text{g m}^{-3}$) for the CDP (Fig. 3).

341 **2.4 Microphysics Model**

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

350 Since the model is stochastic, we use an ensemble size of 16 for each case, with 20,200 super droplets across the whole
351 cloud. The cloud thickness is 400 m, representing a typical precipitating warm cloud. The submicron aerosol is
352 represented with a log-normal distribution with a fixed median radius of 0.1 microns and a fixed geometric standard
353 deviation of 1.835, consistent with the 7th modal aerosol module (MAM7) from the 5th Community Atmosphere



354 Model (CAM5) (Liu et al. 2012), except to avoid complicating the activation process, we only simulate particles larger
355 than 0.1 micron in radius. The updraft velocity, w , as a function of time is given by Eq. (8), which linearly decreases
356 to zero by $t = 800$ s:

$$357 \quad w(t) = w_0 \left(1 - \frac{t}{800 \text{ s}}\right) \text{ if } t < 800 \text{ s else } 0, \quad (8)$$

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

366 The submicron aerosol number concentration for the base case is set to 50 cm^{-3} , which is typical of relatively clean
367 marine conditions (Heintzenberg et al., 2000), but we also vary this from 10 cm^{-3} to 100 cm^{-3} to explore sensitivity to
368 background submicron aerosol concentrations. Particles in the accumulation mode *and* coarse mode are assumed to
369 be hygroscopic (hygroscopicity parameter, “kappa”, of 0.7, Petters and Kreidenweis 2008). For any desired kappa, we
370 use the transformation $r^3 = 0.7r^3/\text{kappa}$ to find the corresponding effective dry radius. Using the parametric collision
371 kernel given by Ayala et al. (2008) and the ordinary Hall collision efficiencies, we choose two representative levels
372 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,
373 the Taylor-microscale Reynolds number is fixed at 75.

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

381

382 The goal of these simulations is to determine the effect of GCCN on precipitation, both in terms of mass of GCCN
383 and total concentration of GCCN. For computational efficiency, we use size distributions from only one instrument,
384 the CAS. The masses for the CAS and CDP, and how mass and number depend on each other are quite similar (Fig.
385 3). So, running the simulations on both instruments would likely yield similar results. We use all 460 flight leg dry
386 size distributions to serve as model input. We extract the slope and intercept parameters from exponentially fitted dry
387 size distributions (section 2.2), and compute the total coarse-mode dry mass of sea salt ($\mu\text{g m}^{-3}$) for each GCCN size
388 distribution using Eq. (8). Coarse-mode dry GCCN mass M_{dry} is determined as:



389
$$M_{dry} = \frac{\pi}{6} \rho_s N_{0,dry} \int_{(D_{dry})_{min}=2 \mu m}^{\infty} (D_{dry})^3 e^{-\frac{D_{dry}}{D_{e,dry}}} dD_{dry}, \quad (9)$$

390 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
 391 of $\mu\text{g m}^{-3}$), $N_{0,dry}$ is the dry intercept, $(D_{dry})_{min}$ is the minimum dry diameter, D_{dry} is the dry diameter (μm), and
 392 $D_{e,dry}$ is the e-folding diameter previously extracted from the exponential fit to the dry size distributions.

393

394

395 **3 Results**

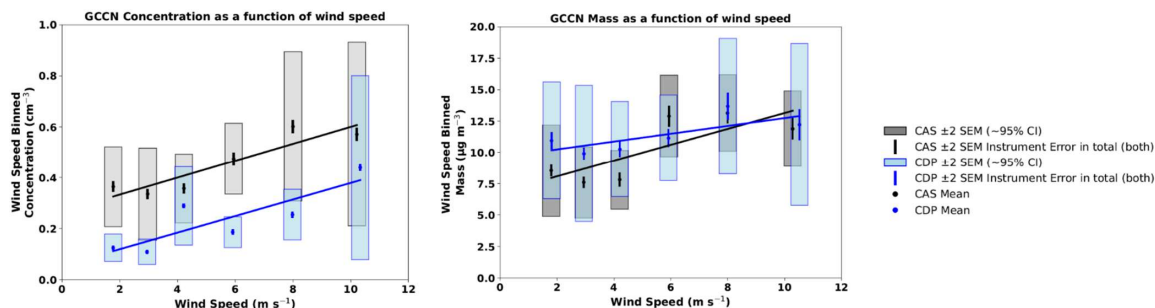
396 **3.1 Wind speed influence on coarse-mode aerosol concentration and mass**

397 Both concentration and mass increase with wind speed, indicating enhanced production of coarse-mode particles under
 398 stronger surface winds (Fig.4). However, substantial variability exists within each wind speed bin, suggesting that
 399 wind speed alone does not fully constrain the observed aerosol properties. A similar positive relationship observed
 400 between coarse-mode mass and wind speed for both instruments. The relationship between mass and wind speed also
 401 shows a statistically significant relationship for both instruments, with fitted slopes of 0.63 ± 0.51 and $0.31 \pm$
 402 $0.29 \mu\text{g m}^{-3}$ for the CAS and CDP, respectively, and coefficients of determination of $R^2 = 0.61$ and $R^2 = 0.54$.
 403 While both mass and concentration increase with wind speed, the large error bars show that substantial variability
 404 remains between flight legs.

405

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408

409 Figure 4 Relationship between wind speed and number concentration, $N_{d>2}$, (left) and GCCN dry mass, $M_{d>2}$, (right).
 410 Data are grouped into six wind speed bins, and each point represents the mean value derived from the average dry size
 411 distribution within each bin. Shaded boxes denote ± 2 standard errors of the mean (95% confidence interval) across
 412 the flight legs contributing to each bin, while vertical bars represent $\pm 2\sigma$ instrument counting error propagated
 413 through the integrated mass or concentration calculation. Black symbols correspond to CAS measurements and blue



414 symbols correspond to CDP measurements. Solid lines indicate linear least-squares fits to the binned means and are
 415 reported in Table 1.

416

Statistics for wind speed binned GCCN mass and concentration

Variable	Instrument	Slope \pm 2 SE (95% CI)	Int. \pm 2 SE (95% CI)	R ²
Mass	CAS	0.63 \pm 0.51	7 \pm 3	0.61
Mass	CDP	0.31 \pm 0.29	10 \pm 2	0.54
Concentration	CAS	0.033 \pm 0.015	0.3 \pm 0.1	0.84
Concentration	CDP	0.029 \pm 0.022	0.1 \pm 0.1	0.64

417

418 Table 1 Linear regression statistics describing the relationship between wind speed and wind-speed-binned GCCN
 419 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
 420 (b), and coefficients of determination (R^2) are derived from least-squares fits of the form $y = mx + b$ applied to the
 421 mean values within each wind speed bin. The slopes for both GCCN mass and concentration are positive for both
 422 instruments. While all slopes are statistically significant at the 95% CI, the large uncertainties associated with the mass
 423 relationships, particularly for CDP, indicate a more variable dependence on wind speed.

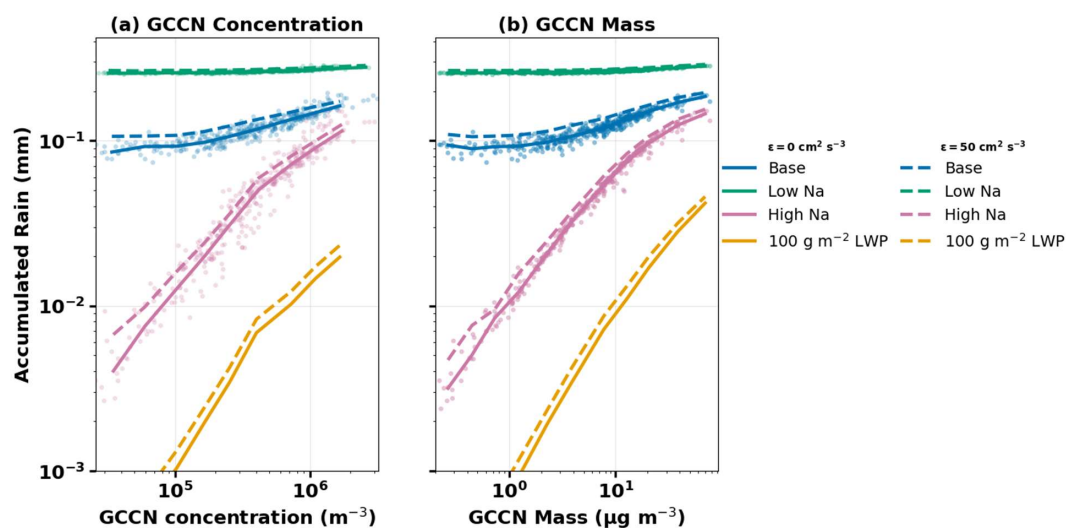
424

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

433 3.2 Modeling GCCN influence on precipitation

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

437



438

439

440 Figure 5 1-D kinematic super droplet microphysics model simulations showing accumulated rainfall over the 1 hr
 441 simulation, as a function of GCCN concentration ($N_{d>2}$) (a) and GCCN mass ($M_{d>2}$) (b). Model simulations were
 442 performed for all flight legs. For the “base” simulation ensemble (blue), the liquid water path is fixed at 385 g m^{-2} and
 443 the submicron aerosol concentration N_a is 50 cm^{-3} . Also shown are low LWP (100 g m^{-2}) simulations with the same
 444 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
 445 increasing levels of turbulence. High N_a ensembles use a concentration of 100 cm^{-3} and low N_a ensembles use a
 446 concentration of 10 cm^{-3} . Colored curves show the binned median responses for each ensemble, while the scatter
 447 points represent model estimates using individual flight legs. The median response provides a representative
 448 description of the ensemble behavior.

449

450 The simulations show a clear and consistent enhancement of accumulated rainfall with increasing GCCN mass (Fig.
 451 5a) and concentration (Fig. 5b). Across all model ensembles, median accumulated rainfall increases systematically
 452 with GCCN concentration or mass, showing that higher GCCN levels are associated with more efficient warm-rain
 453 production over the one-hour simulation. This behavior is seen in the median curves, as well as, in the distribution of
 454 individual flight-leg simulations, represented by the scatter. The scatter from the full set of flight legs closely follows
 455 the log-binned median response in both panels. This agreement is important because it shows that the relationships in
 456 Fig. 5 reflect a consistent response across the full population of observed GCCN size distributions.

457

458 The low N_a (10 cm^{-3}) ensembles of simulations consistently produce the most rainfall over the simulation, for a given
 459 GCCN concentration ($N_{d>2}$, Fig 5a) or mass ($M_{d>2}$, Fig 5b). In contrast, the high N_a simulations produce considerably
 460 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
 461 much stronger, in a relative sense, for higher aerosol background (larger N_a). This demonstrates that the background
 462 aerosol environment of the simulations clearly influences not only the precipitation (as is expected from numerous



463 previous observational and modeling studies), but also helps set the effectiveness of GCCN in influencing warm rain.
464 Under low N_a conditions, the cloud contains fewer competing smaller droplets, which leads to more effective collision-
465 coalescence of droplets, leading to enhanced rainfall. The high N_a ensemble conditions distribute available water
466 across a larger number of droplets, slowing the rate of accumulated rainfall. The shape of these curves is also
467 important. For the low N_a ensembles, rainfall increases at a slower rate with GCCN mass and concentration and tends
468 to weaken somewhat at the highest values, particularly for concentration (Fig. 5a). This implies that continuing to add
469 more GCCN can yield smaller additional increases in accumulated rain at a certain point, (or possibly even drop off).
470 In contrast, accumulated rainfall for the high N_a and “base” ensembles appear to increase more rapidly with $N_{d>2}$ and
471 $M_{d>2}$ when GCCN levels are low. This is consistent with how the low-LWP ensembles also increase rapidly with
472 GCCN levels, but the rain remains very small ($<10^{-2}$ mm in an hour) until GCCN loadings increase significantly. The
473 limited liquid water available in these conditions initially suppresses droplet growth and hinders collision-coalescence
474 between the droplets formed on submicron aerosol and also the tail of growing GCCN particles accreting cloud
475 droplets. The presence of GCCN alone is not enough to generate rainfall if the available liquid water is limited.

476

477 Furthermore, in both panels (Fig 5), increasing GCCN is associated with more accumulated rainfall, but the correlation
478 with GCCN mass ($M_{d>2}$) is significantly tighter than for number concentration $N_{d>2}$, as seen with the individual
479 simulations for flight leg averages (circles in Fig. 5). This is particularly evident in the low LWP ensemble, where
480 higher GCCN mass produces greater accumulated rainfall than comparable increases in number concentration. These
481 larger particles accelerate the growth of precipitation sized drops, leading to greater accumulated rainfall. Finally, the
482 influence of microscale turbulence on the collection kernel rate is also important, although more subtle than the effect
483 of background conditions and GCCN. For all ensembles, the median rain accumulation curves for both turbulence
484 levels remain closely clustered together, indicating that the overall trend of the GCCN rainfall relationships is robust
485 across the range of dissipation rates given. However, it is clearly seen that increased level of turbulence increases rain
486 rates across all ensembles. Microscale turbulence (as represented here by turbulent dissipation rate) impacts the
487 efficiency of droplet growth and collision-coalescence, by increasing the likelihood of collisions between droplets.
488 For reasons that are not entirely clear, the relative impact of microscale turbulence is greatest for the base N_a ensemble,
489 and is also most important at the lower precipitation rates, where it can help overcome the barrier to precipitation
490 formation caused by cloud droplets that are too small to effectively collide (high N_a). Overall, the behavior of the
491 ensembles show that the magnitude of rainfall is dependent not only on the size and number of particles, but on the
492 amount of available cloud water.

493 3.3 Observations of GCCN influence on precipitation

494 While the model results in section 3.2 illustrate theoretically how giant CCN can accelerate precipitation rates, it is
495 difficult to provide a direct comparison with the model due to sampling challenges with the dataset. ACTIVATE lacks
496 a radar measurement collocated with the GCCN observations, and the majority of the flight legs are undertaken in a
497 survey-type mode rather than providing repeated sampling of an air mass at different levels. These limitation makes
498 the dataset less amenable to closure studies that focus specifically on relating precipitation to cloud and aerosol



499 variability. Instead, we can use aircraft in situ sampling of rain water content (RWC), together with cloud water content
500 (CWC), and cloud droplet concentration (N_d) and develop a composite analysis that is used to assess whether flights
501 with high GCCN concentrations (or mass) contain more or less precipitation than those with low GCCN levels.
502 Rainwater content (RWC) is a direct measure of the condensate converted into precipitation-sized drops, making it a
503 key proxy for evaluating the role of GCCN in affecting the cloud to rain conversion. To isolate the role of GCCN, we
504 analyze RWC within set bins of LWC and N_d , which constrain the cloud macrophysical and microphysical state and
505 hold it constant. This allows us to evaluate the effect of GCCN mass and concentration on RWC while minimizing
506 the potentially confounding influence of background cloud variability. So, in addition to using the modeling described
507 in section 3.2, we use the in situ measured cloud and precipitation size distribution to derive observational estimates
508 of rainwater content and total liquid water content, which is defined as:

$$509 \quad LWC = RWC + CWC \quad (9)$$

510 We set a threshold diameter of $50 \mu\text{m}$ to separate cloud from rain drops (Glienke et al., 2017; Sanchez et al., 2023). As
511 we saw in the ensemble simulations (Fig. 5), GCCN can accelerate collision-coalescence, which manifests in an
512 increased rate of conversion of CWC to RWC (Posselt and Lohmann 2008). It is widely appreciated that the rate of
513 conversion of cloud to rain in a cloud is strongly dependent upon the amount of condensate produced in an updraft by
514 condensation (LWC), and is sensitive to the cloud droplet concentration, N_d , as higher droplet numbers suppress
515 precipitation formation by limiting droplet growth (Berry and Reinhardt 1974; Khairoutdinov and Kogan 2000; Wood
516 2005b). ACTIVATE does not have aircraft remote sensors that provide liquid water path (LWP) measurements.
517 Instead, we can use the total LWC, provided in Eq. (10) as the sum of the CAS or CDP CWC and the 2D-S RWC
518 (precipitation water), where the 2D-S RWC is calculated using only bins with diameters greater than $50 \mu\text{m}$ to avoid
519 double counting.

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

$$529 \quad RWC = \frac{\pi}{6} \times \rho_w \sum_{i=6}^{128} n_i \times D_i^3 \Delta D_i \quad (10)$$

530 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
531 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
532 sizing and sample volume for the smallest rain drops, and these are not used. The smallest 2D-S bins also overlap with
533 the largest CAS and CDP bins, so our approach also avoids the potential for double counting. The CWC and RWC
534 are summed to compute the total liquid water content (g m^{-3}) per second, Eq. (11).



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

$$537 \quad \xi = \frac{RWC}{LWC} \times 100 \quad (11)$$

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

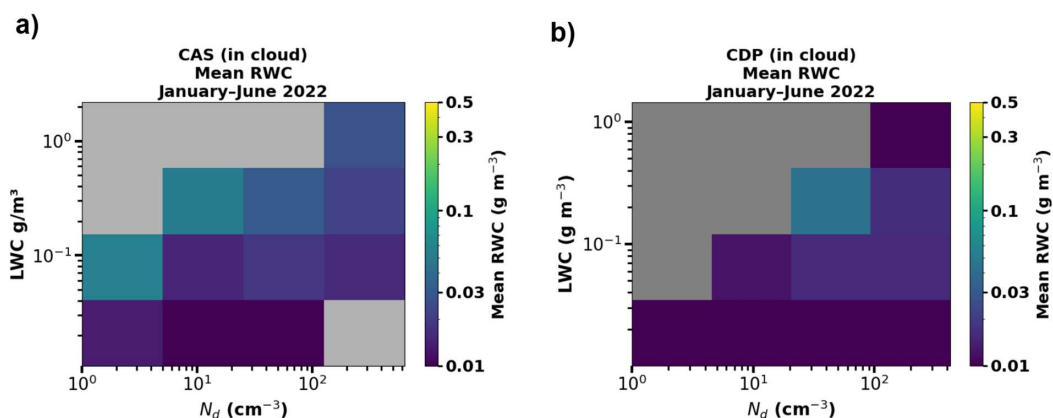
546 As expected, high LWC and lower N_d are two primary controls on rainwater content (Fig. 6), so we can use this as a
547 framework to evaluate the potential impact of GCCN on RWC. However, this analysis does not currently isolate the
548 impacts of GCCN on this specific measure of precipitation efficiency, defined here by the ratio of RWC to LWC. The
549 model results (section 3.2), showed that, while both GCCN number concentration and mass impact precipitation, mass
550 seems to have a better correlation than number concentration. We perform our observational rainwater content analysis
551 for both GCCN variables. To do this, we subdivide the dataset into cases with high and low GCCN mass $M_{d>2}$ (and
552 a separate split for GCCN number concentration $N_{d>2}$). We use our previously calculated leg-mean GCCN $M_{d>2}$
553 (section 2.3). Because the cloudy data cannot be precisely collocated with the out-of-cloud GCCN measurements,
554 which are also made using different flight legs, and flights are relatively short, we decided to use the flight-mean
555 GCCN mass (across all legs used to determine means for each flight) and then assign all the cloud data from the cloudy
556 legs from that flight as high or low GCCN clouds on the basis of the flight mean GCCN concentration, depending on
557 the analysis. We produce an average GCCN mass for each of our 75 flights. To categorize each flight, we take the
558 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
559 assess the robustness of our results using 50% split categories, we repeated the same analysis with a "high" category
560 of the top 20% of flights and a "low" category of the bottom 20% of flights. We also tested a "high" category of the
561 top 40% of flights and a "low" category of the bottom 40% of flights (not shown). We then separate the calculated
562 RWC data into high and low GCCN flight categories (Fig. 7).

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



569 To assess the statistical significance of RWC/LWC differences between high and low GCCN conditions (either using
 570 $M_{d>2}$ or $N_{d>2}$ to characterize GCCN) we applied a bootstrap resampling approach within each joint N_d and LWC bin.
 571 Bootstrapping is well suited for this analysis because the flight-mean data are unevenly distributed across bins and
 572 may not follow a normal distribution, violating assumptions of traditional parametric tests. For each bin, we generated
 573 10,000 bootstrap realizations by resampling flight legs with replacement separately for the high and low subsets. For
 574 each realization we computed the ratio of mean RWC/LWC between the two groups, producing a distribution of
 575 possible outcomes for each bin. This resulted in a distribution of possible outcomes per bin, from which we computed
 576 90% confidence intervals, mean and median values, and the percentage of bootstrap samples indicating an
 577 enhancement in RWC/LWC in the presence of GCCN.

578



579

580 Figure 6 Mean RWC as a function of LWC and N_d for the CAS (a) and the CDP (b). Dark purple indicates lower mean
 581 RWC and light blue to yellow indicates higher mean RWC. Gray bins indicate bins where fewer than 100 valid data
 582 points are available across the compared subsets and are excluded from analysis. For both instruments, highest levels
 583 of mean RWC are observed for moderate LWC- N_d . We found our analysis not to be sensitive to the choice of percentile
 584 threshold used.

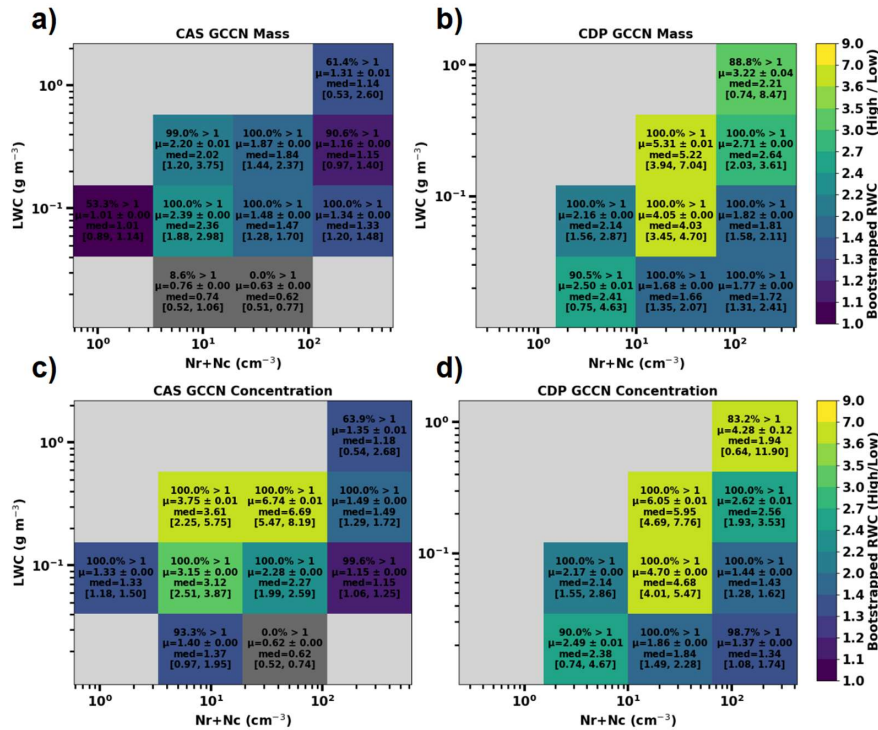
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591 Figure 7 Observationally derived precipitation efficiency responses, defined as RWC/LWC, across bins (boxes) of
 592 LWC and N_d for cases with high versus low GCCN mass (top row) and high versus low GCCN concentration (bottom
 593 row), calculated separately for CAS (left column) and CDP (right column). Colors indicate the bootstrapped ratio of
 594 mean RWC/LWC in high GCCN cases relative to low GCCN cases within each bin (High/Low). Values greater than
 595 unity, which pertains to the colored boxes without gray shading, indicate a larger ratio under higher GCCN conditions.
 596 Text within each box shows the percentage of bootstrap realizations producing ratios greater than 1, the mean ratio
 597 +/- standard error of the mean, and the 90% confidence interval of the bootstrap distribution. Light gray bins indicate
 598 bins where fewer than 100 valid data points are available across the compared subsets and are excluded from analysis.
 599 Dark gray bins met the minimum sampling threshold but yielded bootstrap distributions whose mean ratio did not
 600 exceed 1, indicating no statistically robust enhancement of RWC/LWC under higher GCCN conditions in those bins.

601 Figure 7 shows that higher GCCN conditions are generally associated with enhanced RWC/LWC for the vast majority
 602 of the LWC- N_d bins. The observed differences in RWC/LWC also remained consistent across these thresholds,
 603 indicating that our results are robust with the 50% split. In all three split cases, the same pattern was observed; flights
 604 in the high GCCN category always exhibited larger RWC/LWC than flights in the low GCCN category across the
 605 same regions of LWC- N_d space. In all CDP bins with sufficient sampling, the ratio is >1 (Fig. 7b, d), while the CAS
 606 has 3 bins (two for GCCN mass and one for number concentration) where the mean ratio is <1 (Fig. 7a,c). For the
 607 CAS, all of these bins are located at very low LWC and moderate N_d . Overall, the majority of the bins with sufficient
 608 sampling for both instruments, contain data points where the ratio is exceeded by more than 90% of the data points in



609 that bin. This indicates that higher GCCN mass or concentration is typically associated with larger RWC/LWC values
610 relative to lower GCCN conditions. This enhancement is especially robust (high confidence) in the central bins of the
611 distribution, located at moderate levels of LWC and N_d , where the percentage of bootstrap realizations greater than 1
612 is near 100%.

613 Between the two instruments, the strongest enhancements are found with the CDP dataset, particularly for GCCN
614 mass. In the CDP mass panel (7b) several bins exhibit mean values of 1.5-6. By contrast, the CAS mass panel (7a)
615 does not show bins where mean RWC/LWC exceeds 2.4. The CDP mass analysis therefore shows stronger
616 enhancement of RWC/LWC on average than the concentration analysis, while the CAS results show relatively
617 stronger enhancement for GCCN concentration than for mass. The especially strong enhancements in the CDP mass
618 panel support the model results from section 3.2 that GCCN mass may be a better predictor of precipitation response
619 than GCCN concentration alone. The presence of sufficiently large particles versus the presence of more but smaller
620 coarse mode appears to play a more important role in initiating collision-coalescence than number alone. Nevertheless,
621 both probes suggest that rainwater contents are likely enhanced by a factor of 2 or more for GCCN mass in the upper
622 half of the observed distribution compared with the lower half. This finding is broadly consistent with the modeling
623 results (Fig. 5) that suggest precipitation accumulation may be approximately doubled for the upper half of the GCCN
624 masses.

625 Furthermore, the highest ratios are not found uniformly across all bins. Instead, the largest RWC/LWC enhancements
626 in Fig. 7 are in regions where Fig. 6 shows the highest mean RWC, corresponding to moderate-to-high LWC and
627 relatively low-to-moderate N_d . For the CAS, bins with high LWC and high N_d yield lower ratios. Similarly, bins with
628 lower LWC and low N_d show weaker enhancement, likely a result of insufficient condensate. The CDP shows a
629 relatively similar pattern to the CAS in low LWC- N_d space; however, for both mass and concentration, the highest bin
630 of LWC and N_d yields relatively high mean ratios (>3) than bins with the same N_d but lower LWC. This suggests that
631 sufficiently high LWC might offset the suppressing effect of high N_d promoting rapid droplet growth and increased
632 collision-coalescence, thus allowing GCCN to enhance our metric of precipitation efficiency even in high N_d
633 conditions.

634 **4 Discussion**

635 Together the ACTIVATE observational analysis in addition to results from the microphysics model provide a
636 preponderance of evidence that GCCN are able to exert a significant influence on warm-rain formation. Both the in-
637 situ measurements and model simulations show that increasing GCCN is associated with enhanced precipitation
638 production, with GCCN mass exhibiting a stronger and more consistent relationship than number concentration. Due
639 to the exponential nature of the GCCN size distribution for $D_{dry} > 2 \mu\text{m}$, GCCN number concentration tends to be
640 dominated by salt particles with sizes close to the $2 \mu\text{m}$ threshold, but these particles are less effective at promoting
641 precipitation than slightly particles with dry diameters in the range $4\text{-}10 \mu\text{m}$ (Barr and Wood 2026). This influence of
642 GCCN appears to be captured well using the overall dry mass of salt particles, which suggests a possible way forward
643 to parameterizing the GCCN effects on warm rain production in marine clouds. Our analyses also provide statistical
644 observational evidence from a large number of clouds that LWC and N_d play an important role in staging the primary



645 conditions for precipitation formation, and that the presence of GCCN can impact how efficiently rain develops. The
646 modeling provides additional support. Varying the submicron aerosol concentrations (N_a) and LWP have leading order
647 influences on the production of warm rain (Terai et al. 2012; Pawlowska 2003, Van Zanten 2005; Wood 2005a, b;
648 Mann 2014). Accumulated rainfall is greatest for low N_a conditions and weakest for low LWP conditions. Low N_a
649 conditions promote collision-coalescence, while low LWP limits droplet growth and suppresses rainfall. Furthermore,
650 RWC/LWC ratios were higher and more consistent for moderate LWC- N_d bins, thus showing strong agreement with
651 model simulations. These comparisons are made within the same LWC- N_d regimes and therefore isolate the influence
652 of GCCN. This demonstrates that enhanced precipitation efficiency is directly associated with the presence of GCCN
653 rather than differences in cloud properties.

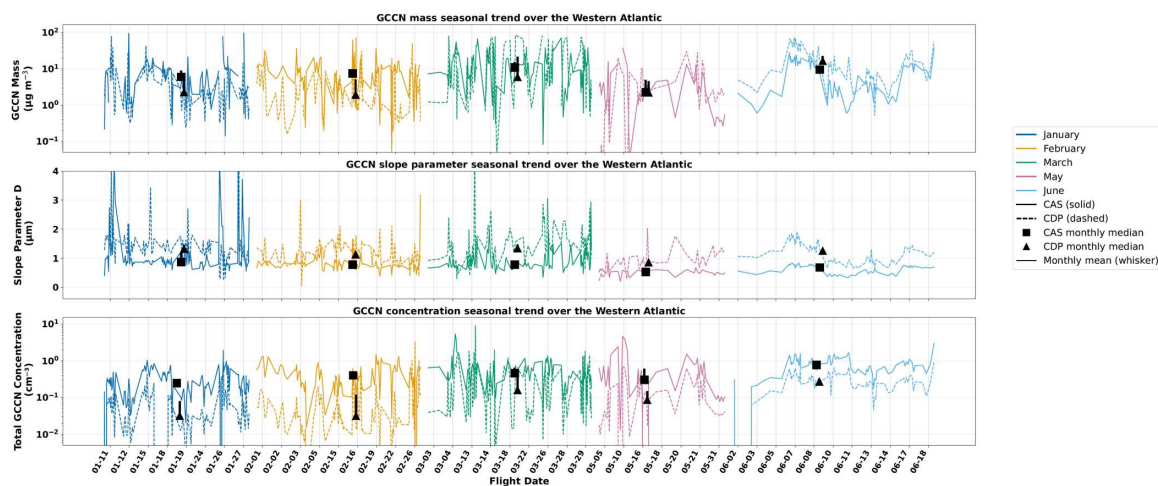
654 As in previous observational studies, our results demonstrate that, on average, higher wind speeds increase both the
655 size and number of GCCN, though the impact is stronger for concentration. This positive relationship between
656 increasing wind speed and GCCN production is consistent with other studies (Woodcock 1953; Monahan et al.
657 1986; Banner and Peregrine, 1993; Gemmrich and Farmer, 1999; Lewis and Schwartz 2004; Clarke et al. 2006;
658 Colón-Robles et al., 2006; Jensen and Lee 2008; Jensen and Nugent 2017). In particular, Colón-Robles et al., 2006,
659 which analyzed coarse-mode aerosols from a similar cloud probe (FSSP) to those used in this study, found a strong
660 positive relationship between concentration and wind speed ($R = 0.87$, $R^2 = 0.76$) over a wind speed range of
661 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,
662 though they are looking at hydrated particles, whereas ours are dry. However, this is still comparable to our results,
663 which show slopes of $0.033 \pm 0.015\ \mu\text{g m}^{-3}(\text{m s}^{-1})^{-1}$ and $0.029 \pm 0.022\ \mu\text{g m}^{-3}(\text{m s}^{-1})^{-1}$, for the CAS and CDP
664 respectively (Table 1). Their correlation with wind speed for the FSSP is stronger than the CDP ($R^2 = 0.64$), but weaker
665 than the CAS ($R^2 = 0.84$). The absolute concentrations reported by Colón-Robles et al. (2006) are generally lower
666 than those observed here, with their Fig. 1a showing values ranging from about 0.05 to 0.3 cm^{-3} compared to about
667 0.1 to 0.6 cm^{-3} in this study (Fig. 4). Furthermore, their size distribution analysis (their Fig. 2) shows that stronger
668 wind speeds increase particle concentrations across the coarse-mode spectrum, particularly for diameters larger than
669 $10\ \mu\text{m}$ (their Fig. 2a-b), with dry particle measurements from a Giant Nucleus Impactor (GNI) extending to $30\text{-}40$
670 μm (their Fig. 2c). Our dry size distributions also follow exponential decay, with a tail extending to about $20\text{-}25\ \mu\text{m}$
671 (Fig. 2), a slightly smaller range than in their GNI analysis. However, while both studies show the presence of large
672 dry particles, the concentrations of these particles in Colón-Robles et al. (2006) decrease rapidly with increasing
673 diameter and remain lower than ours beyond $\sim 15\ \mu\text{m}$ (their Fig. 2c). Our dry size distributions exhibit relatively
674 higher concentrations across the coarse-mode tail, particularly in the $15\text{-}25\ \mu\text{m}$ range. Though this study was based
675 on 30-minute flight legs during the Rain in Shallow Cumulus over the Ocean (RICO) campaign (2004-2005) over the
676 trade winds in the western Atlantic, the similarity in slope, correlation strength, and overlapping wind speed ranges
677 indicates a consistent relationship between wind speed and coarse-mode aerosol concentration with this study.
678 Although a positive relationship between wind speed and GCCN concentration is observed, the strength of this
679 dependence is weaker than would be expected from sea spray aerosol flux parameterizations, which typically show a
680 much stronger increase in particle production with wind speed (Woodcock 1953; Monahan et al., 1986; Lewis and
681 Schwartz, 2004).



682 Furthermore, though our relationship between mass and wind speed is weaker than for concentration, it is still
 683 moderate and positive, consistent with other observational studies (Woodcock 1953; Liu et al., 2021). For example,
 684 Woodcock (1953) reports GCCN mass values of approximately $\sim 8 \mu\text{g m}^{-3}$ at wind speeds of $5.5\text{-}7.9 \text{ m s}^{-1}$ (their Fig.
 685 2). In our study, the mean $M_{d>2}$ is $10.3 \mu\text{g m}^{-3}$ for the CAS and $12.2 \mu\text{g m}^{-3}$ for the CDP for a similar mean wind speed
 686 of 5.13 m s^{-1} , which falls within the Woodcock (1953) wind regime, further allowing us to directly compare between
 687 studies. Both our study and Woodcock (1953) show considerable amounts of scatter, reflected in the large spread of
 688 values within individual wind speed bins (Fig. 4) and the moderate coefficients of determination ($R^2 = 0.61$ and 0.54
 689 for the CAS and CDP, respectively). Despite the agreement in magnitude, the relatively modest coefficients of
 690 determination and substantial scatter indicate that the dependence of GCCN mass on wind speed is weaker than would
 691 be expected from wind speed driven sea spray production alone. This highlights that variability in GCCN
 692 concentration and mass relationships cannot be explained by wind speed alone. Wind speed represents only one
 693 component of the processes controlling GCCN variability in the midlatitude marine environment.

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

704



705



706 Figure 8 Monthly trends (January - June 2022) of GCCN mass $M_{d>2}$ (top), slope parameter $D_{e,dry}$ (middle), and total
 707 GCCN concentration $N_{d>2}$ (bottom). Values are shown as flight-leg means plotted by flight date and grouped by
 708 month. Solid lines (and black squares for monthly medians) represent measurements from the CAS, while dashed
 709 lines (and black triangles) represent measurements from the CDP. Whiskers on the black median symbols show the
 710 corresponding monthly means.
 711

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)

712
 713 Table 2 Monthly correlations between the CAS and the CDP across all months, as well as, within each month. GCCN
 714 mass $M_{d>2}$, slope parameter $D_{e,dry}$, and concentration $N_{d>2}$ shown as Pearson correlation coefficients r with
 715 corresponding coefficients of determination R^2 in parentheses. The full trends between the two instruments across all
 716 months are lowest and relatively weak for mass ($R^2=0.3$) and highest for the slope parameter ($R^2=0.7$) and number
 717 concentration ($R^2=0.8$). For all three variables, intra-month CAS and the CDP estimates are mostly positively
 718 correlated, and are highest during the most in the month of June. The weaker correlation between the two instruments
 719 for GCCN mass measurements likely reflects a greater sensitivity of mass to more poorly sampled larger particles,
 720 which are less numerous and more variable in space and time, leading to increased measurement differences between
 721 instruments.



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)

722

723 Table 3 Monthly summary statistics for GCCN mass, slope parameter D , and concentration (N_d) measured by the CAS
 724 and CDP. Values are reported as monthly means, with corresponding medians in parentheses. Across all months, the
 725 CAS exhibits higher mean concentration, while the CDP generally shows larger slope parameters, indicating a greater
 726 relative contribution from larger particles. For January, February, and May, the CAS shows higher mass
 727 measurements, but the CDP records higher mass in March and June. No clear or consistent seasonal trend is observed
 728 across the sampled months.

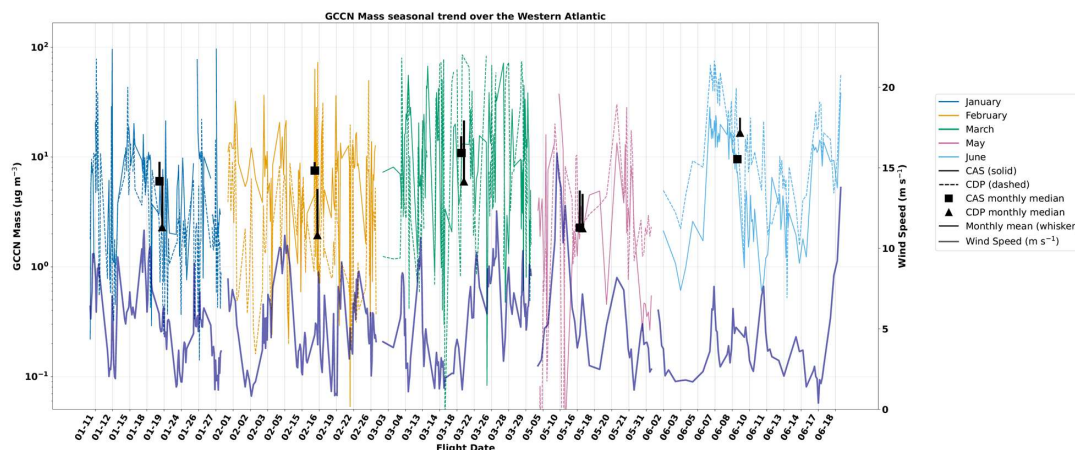
729 In general, months with higher GCCN mass are associated with larger slope parameters (Table 3), indicating a shift
 730 toward relatively larger particles. However, GCCN concentration does not exhibit a consistent relationship with slope
 731 parameter as shown by the lack of a clear connection between monthly concentration and slope values (Table 3).
 732 Agreement between the CAS and CDP varies by variable and by month (Table 2). Across all months, correlations are
 733 strongest for concentration ($R^2 = 0.83$) and slope parameter ($R^2 = 0.70$), while substantially weaker for mass ($R^2 =$
 734 0.31). However, this agreement is not uniform across individual months for any of the variables. For example,
 735 correlations are very weak in March for mass ($R^2 = 0.065$), slope parameter ($R^2 = 0.008$), and concentration ($R^2 =$
 736 0.001), while much stronger in June ($R^2 = 0.805$, 0.832 , and 0.969 , respectively). These results indicate that while the
 737 two instruments capture similar overall trends, their agreement depends on both the variable considered and the
 738 temporal scale of analysis. For most months, the CAS exhibits a higher monthly median for mass and concentration,
 739 and a lower monthly median for slope compared to the CDP (Table 3). Across January through June 2022, there is no
 740 clear seasonal trend in GCCN mass, concentration, or slope parameter, as monthly means and medians do not show a
 741 consistent increase or decrease over time (Table 3). Instead, values fluctuate between months, including peaks and
 742 dips across individual flights within the same month (Fig. 8). GCCN properties across this time period appear to be
 743 driven more by individual flight conditions than by a coherent seasonal cycle, though this could be due to the
 744 conditions that were selectively targeted with each flight. The absence of a clear seasonal trend suggests that variations
 745 in GCCN mass, size, and concentration are not largely driven by large-scale seasonal changes.

746 However, to further investigate potential drivers of this variability, we also examined the seasonal behavior of wind
 747 speed, which is a primary control on sea-spray aerosol production. We show the seasonal evolution of GCCN mass
 748 and mean wind speed across the Western Atlantic (Fig. 9). This analysis focuses only on mass, as we found it to be



749 more influential in enhancing precipitation formation. As in Fig. 8, variables are shown as flight-leg means organized
 750 chronologically by flight date and grouped by month to highlight seasonal variability.

751



752

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

757

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

769 The potential influence of sea surface temperature (SST) was not explicitly tested in this study. A more targeted
 770 analysis using SST measurements, after accounting for the dependence of GCCN on wind speed, would help determine
 771 whether SST explains additional variability in GCCN production. Despite the consistency of the results presented



772 here, several limitations of the analysis should be considered. First, while the ACTIVATE campaign contains an
773 unprecedented amount of data due to its numerous flights, the aircraft did not contain a radar, which limits our ability
774 to directly measure precipitation. Instead, we rely on in-situ measurements to derive rainwater content, which does
775 not capture the precipitation falling out of the clouds and reaching the ground. Additionally, the relatively short
776 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
777 need to bin flight legs for wind speed analysis. While counting uncertainties in total concentration are small (~ 0.01
778 cm^{-3}) and much smaller than the observed leg-to-leg variability, uncertainties in mass are slightly larger because mass
779 is strongly influenced by the largest and least frequent particles. As reflected in Eq. (7), the mass uncertainty scales
780 with D_i^6 , making it particularly sensitive to incomplete sampling of the larger size range. As a result, individual flight
781 legs may under or over represent large particles, leading to increased variability in mass estimates. Grouping flight
782 legs into wind speed bins increases the effective sample volume and reduces this sampling noise.

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

790 Possible future work could build on this analysis by incorporating additional years of ACTIVATE observations,
791 including data from 2020 and 2021, to better assess interannual variability. Furthermore, the use of instruments
792 specifically designed to measure giant nuclei, such as a giant nucleus impactor, would provide improved
793 characterization of the largest particles that dominate mass and are most relevant for collision-coalescence processes.
794 These improvements would help further constrain the role of GCCN in precipitation formation and refine their
795 representation in both observational and modeling frameworks. A step forward in this direction is demonstrated by
796 some studies using cloud water samples to infer the amount of sea salt in clouds based on the mass concentration of
797 its dominant species, sodium and chloride (Dadashazar et al., 2017; MacDonald et al., 2020; Gonzalez et al., 2022;
798 Zeider et al., 2025; Ajayi et al., 2026).

799 5 Concluding Remarks

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



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

814 Several limitations should be considered when interpreting these results. The ACTIVATE aircraft observations do not
815 directly measure surface precipitation and rely instead on in-situ cloud and rainwater measurements.

816 Additionally, estimates of GCCN mass remain sensitive to sampling uncertainties associated with the largest and least
817 frequent particles. The representation of turbulence within the microphysical model also introduces uncertainty in the
818 simulated precipitation response. Despite these limitations, the consistency between the observational and modeling
819 analyses provides strong evidence that GCCN can substantially influence warm-rain formation in marine low clouds.
820 Overall, these findings improve our understanding of aerosol-cloud-precipitation interactions by demonstrating that
821 coarse-mode sea spray aerosols can enhance precipitation efficiency even at relatively low concentrations. The
822 stronger relationship between precipitation and GCCN mass than number concentration suggests that mass-based
823 metrics may provide a useful framework for representing GCCN effects in future observational and modeling studies.
824 Further analyses incorporating additional years of observations and targeted measurements of coarse mode seas salt
825 particles would help better constrain the role of GCCN in marine clouds and their representation in microphysics
826 models.

827 **Code availability**

828 The code used for data processing, analysis, and figure generation in this study is publicly available on GitHub. Scripts
829 related to the CAS instrument are available at <https://github.com/katherineemifsud/ACTIVATE-2024-2025/tree/master/CAS> and scripts related to the CDP instrument are available at:
830 <https://github.com/katherineemifsud/ACTIVATE-2024-2025/tree/master/CDP>. These repositories include routines for
831 observational size distribution processing, statistical analysis, model comparison, and rainwater content analysis.
832

833 **Data Availability**

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

838 **Author Contributions**

839 KM and RW conceived the study. JB performed the model experiments. KM conducted the analysis and prepared
840 the manuscript with contributions from co-authors.



841 **Competing Interests**

842 **Disclaimer**

843

844 **Acknowledgments**

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