Observations and understanding of the role of giant CCN in influencing warm rain production from shallow liquid clouds in ACTIVATE

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Objectives

- Use observations from in-situ probes on the low flying aircraft (HU-25 Falcon) below cloud to quantify the coarse mode aerosol size distribution (1-50 μm ambient diameter) under clear sky conditions below cloud
 - Use the Cloud-Aerosol Spectrometer (CAS), the cloud droplet probes (CDP and FCDP), and 2D-S probe to collect aerosol size distributions at ambient RH, but then convert these to dry size distributions using RH measurements and constraints on coarse mode aerosol hygroscopicity
 - Compare the wind speed dependence of near-surface coarse mode distributions (giant CCN) with previous studies.
- Use in-cloud in situ cloud probe data from ACTIVATE to produce composite cloud droplet size distributions and apply the stochastic collection equation to calculate autoconversion and accretion rates following Wood (2005a,b).
 - Use these together with bulk information (e.g., cloud water, cloud droplet concentration, volume radius, and concentration of large droplets) derived from the in-situ measurements to evaluate and further develop a new autoconversion parameterization that incorporates GCCN.
 - The autoconversion scheme will be tested in large eddy simulations (UW-SAM).
- If time permits, work with the HSRL team at NASA Langley to attempt to constrain the hygroscopic growth of the coarse mode aerosol by combining in situ measurements with airborne high spectral resolution lidar data on the King Air aircraft in ACTIVATE.
 - The subcloud layer in the PBL is often well-mixed, so the RH vertical profile is known. Under such conditions, we will examine how lidar backscatter and aerosol extinction varies with height and attempt to predict this using the ambient in situ measurements and hygroscopicity.
 - Discrepancies between observed and predicted aerosol backscatter and extinction are likely indicative of uncertainty in the hygroscopic growth, and thus this method may provide an alternative method for constraining the coarse mode hygroscopicity, which may be difficult to otherwise estimate.

GCCN can increase drizzle in marine stratocumulus

- In Feingold et al. (1999), GCCN are droplets with r=20 micron added to clouds to explore the sensitivity.
- Impact of GCCN is sensitive to background CCN. Clean clouds are less sensitive because the low N_d leads to large cloud drops that can coalesce efficiently without GCCN.
- Significant impact on drizzle for GCCN concentrations higher than ~10⁻³ cm⁻³.
- Condensational growth of GCCN was bypassed but is critical for collisioncoalescence initiation



Feingold, G., Cotton, W. R., Kreidenweis, S. M., & Davis, J. T. (1999). The impact of giant cloud condensation nuclei on drizzle formation in stratocumulus: Implications for cloud radiative properties. Journal of the Atmospheric Sciences, 56(24), 4100–4117.

Precipitation nuclei



- Superdroplet LES permit the explicit treatment of condensational growth of the GCCN (left, from Dziekan et al., 2021)
- GCCN have dry diameters larger than 1.6 μ m and are taken from an observed size distribution from VOCALS (Jensen and Nugent, 2017). $N_{GCCN} = 0.28 \text{ cm}^{-3}$
- Results indicate significant impacts on precipitation when $N_{GCCN}/N_{CCN} > 10^{-3}$ (left).



Dziekan, P., Jensen, J. B., Grabowski, W. W., & Pawlowska, H. (2021). Impact of Giant Sea Salt Aerosol Particles on Precipitation in Marine Cumuli and Stratocumuli: Lagrangian Cloud Model Simulations. J. Atmos. Sci., **78**, 4127–4142.

Jensen, J. B., & Nugent, A. D. (2016). Condensational Growth of Drops Formed on Giant Sea-Salt Aerosol Particles. Journal of the Atmospheric Sciences, 74(3), 679–697.

Giant CCN measurements

- <u>Slide method</u>: Woodcock (1953), refined/automated with the giant nucleus impactor (GNI), an open stream impactor designed to collect giant haze particles with dry diameters from 1.6-18 μ m. Size distribution follows from microscopy analysis (Jensen et al., 2020)
- <u>Aircraft open stream wing probes</u> (scattering and shadow probes) have been used in clear sky (Colón-Robles et al., 2006; Lowenstein et al., 2010) to characterize the giant CCN size distribution
- General agreement between methods, confirming Woodcock (1953) dependence on wind

Jensen, J. B., et al. (2020). The Giant Nucleus Impactor (GNI)—A System for the Impaction and Automated Optical Sizing of Giant Aerosol Particles with Emphasis on Sea Salt. Part I: Basic Instrument and Algorithms. J. Atmos. Oceanic Technol., 37, 1551–1569.

Lowenstein, J. H., Blyth, A. M., & Lawson, R. P. (2010). Early evolution of the largest-sized droplets in maritime cumulus clouds. Quart. J. Roy. Meteorol. Soc., 136, 708–717.

► Colón-Robles, M., Rauber, R. M., & Jensen, J. B. (2006). Influence of low-level wind speed on droplet spectra near cloud base in trade wind cumulus. Geophys. Res. Lett., **33**(20).





▲ Woodcock, A. H. (1953). Salt nuclei in air as a function of altitude and wind force. J. Atmos. Sci., **10**, 362–371.



<u>Aerosol Cloud Meteorology Interactions</u> Over the Western <u>Atlantic Experiment</u>



Cloud-Aerosol Spectrometer (CAS)

- The CAS probe measures haze/cloud droplets using 30 bins from 0.5 um to 50 um diameter.
- Estimate coarse mode (GCCN) distributions below cloud base as a function of specific meteorological conditions (i.e., near surface wind speed).
- Aggressive screening to remove cloud and/or precipitation



Moore et al. 2022

Low flying HU-25 aircraft



How to predict the GCCN haze droplet DSD in clouds?

Ráoult

dominated

arow

Kelvin Raoult

 r_{drv} [µm]

100

- We have quite good observational constraints on Hydrated vs dry droplet phase space the dry GCCN distribution from sea spray 1000 Condensational growth of GCCN is necessary to Kelvin dominated serve as precipitation nuclei. evaporation • For $r_{dry} \gtrsim 1 \ \mu m$, we can simplify the droplet growth 100 equation: Growth for $\sigma = 0$ Kelvin term is negligible [mm] Raoult term tends to be substantially larger than the 10 typical supersaturation in PBL clouds • Analytical solution to droplet growth equation ($\sigma=0$, . II Y dry no Kelvin): hygroscopicity ("kappa") Raoult + Kelvin $r = (r_0^5 + 5G\kappa r_{drv}^3 t)^{1/5}$ terms [%] $r\frac{dr}{dt} = G\left(\sigma - \frac{A}{r} + \kappa \frac{r_{dry}^3}{r^3}\right)$ Initial radius droplet growth equation 0.1 0.01 10 0.1
 - Final radius insensitive to initial radius for GCCN (1-10 μ m), so: $r \approx (5G\kappa t)^{1/5} r_{dray}^{3/5}$

Precipitation nuclei effects: Parcel model setup

- Use Lagrangian parcel model with bin representation of the complete aerosol size distribution (Chen and Lamb 1994) to activate cloud droplets, and grow them with both diffusional growth and collision-coalescence
- Background aerosol includes a fine mode and a natural coarse mode that is wind speed dependent (taken from Woodcock 1953).
- Conduct simulations with different concentrations of fine mode particles (modal dry diameter 80 nm). Coarse mode particles (modal diameter 2.2 μm) are added so that the supermicron mass makes up 90% of the total injected aerosol mass.
- Lift parcel up to 300 m above cloud base with updraft of 0.4 m s⁻¹, then keep parcel at this level for 45 mins and evolve size distribution. Examine precipitation at end of simulation.

Chen, J.-P., & Lamb, D. (1994). Simulation of Cloud Microphysical and Chemical Processes Using a Multicomponent Framework. Part I: Description of the Microphysical Model. *J. Atmos. Sci.*, **51**, 2613–2630. Woodcock, A. H. (1953). Salt nuclei in air as a function of altitude and wind force. J. Atmos. Sci., **10**, 362–371.



Supersaturation suppression from GCCN is not a major issue

- Address using a Lagrangian bin microphysics model
- Represent the aerosol size distribution as a two-mode lognormal distribution. Geometric mean diameter and standard deviation for the two modes:

 $\begin{array}{ll} \mathsf{GMD}_{\mathrm{fine.}} & : \ \mathsf{60-100} \ \mathsf{nm} \\ \mathsf{GMD}_{\mathrm{coarse}} & : \ \mathsf{2200} \ \mathsf{nm} \\ \sigma_{\mathrm{fine}} & : \ \mathsf{2.2} \\ \sigma_{\mathrm{coarse}} & : \ \mathsf{1.5} \end{array}$

- The ratio of the mass in the coarse mode to that in the fine mode can be varied from 0 (no coarse mode) to >100 to explore the sensitivity of the activated droplet concentration to increases in coarse mode particle mass.
- The droplet concentration is reduced only slightly, even with 80-90% of the salt mass in the coarse mode. The result is mostly insensitive to the size and concentration in the fine mode.



Precipitation Nuclei: Parcel model results

- Precipitation rate from drizzle drops is shown as a function of droplet concentration
- Precipitation suppression is clearly seen
- Increasing coarse mode (increasing *u* from 4 to 12 m s⁻¹) buffers precipitation suppression as N_d is increased
- Precipitation susceptibility (slope) in this case drops from ~1 for low GCCN to ~0.5 for u=12 m s⁻¹
- Need to include GCCN to fully represent precipitation susceptibility in warm clouds
- How to do this in simple models (GCM physics)?



Precipitation nuclei: scaling with ratio of GCCN to droplet concentration N_d

- For this cloud case, precipitation rate scales reasonably well with the ratio of the coarse mode concentration (D_{dry} >2 µm) to the cloud droplet concentration, i.e., $N_{a,giant}$ / N_d
- For N_{a,giant} /N_d ≥ 0.0005, precipitation rate exceeds a few tenths mm day⁻¹. Consistent with Dziekan et al. (2021)







New "GCCN-aware" autoconversion scheme

- In situ cloud DSDs used to derive autoconversion rate by integration of the stochastic collection equation (SCE)
- DSDs also provide bulk quantity inputs for autoconversion schemes (e.g., q_c and N_d for KK)
- SCE results are then compared with bulk formulations (see Wood 2005)
- New scheme uses the product of the size distribution close to the the autoconversion threshold ($r=20 \ \mu m$ in this case) and the cloud water q_c
 - $\left(\frac{dn}{dr}\right)_{r=20\mu m}$ can be estimated as the sum of the modeled "cloud" DSD and the GCCN DSD.



Khairoutdinov, M., & Kogan, Y. (2000). A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus. *Mon. Wea. Rev.*, **128**, 229–243. Wood, R. (2005). Drizzle in Stratiform Boundary Layer Clouds. Part II: Microphysical Aspects. *J. Atmos. Sci.*, **62**, 3034–3050.

AUTOCONVERSION PARAMETERIZATIONS vs OBSERVATIONS

Conclusions

- Started analysis of ACTIVATE data to characterize GCCN distributions in clear sky near the surface, and understand their dependence on wind speed (and perhaps temperature)
- Will analyze in-cloud data to evaluate/refine a new "GCCN aware" bulk parameterization of autoconversion that can be applied in low dimensional models (e.g., GCMs) but also in LES with bulk microphysics
- Lagrangian parcel model with explicit treatment of condensational growth and collisioncoalescence allows exploration of the effects of giant CCN on precipitation. Results suggest that:
 - •Supersaturation suppression due to competition for vapor from GCCN is unlikely to significantly reduce the activation of droplets
 - •Giant CCN (coarse mode particles, 1-10 micron dry diameter) from sea spray are likely to significantly impact precipitation formation.
- Parcel model will be used to test new analytic GCCN condensational growth scheme