Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

Relationships between supermicrometer particle concentrations and cloud water sea salt and dust concentrations: Analysis of MONARC and ACTIVATE data

Supplemental Information

S1. Minimum Diameter Definitions in Literature

Comparison between studies investigating the nature and effects of giant cloud condensation nuclei (GCCN) continues to be challenging due to the differences in how GCCN are quantified and defined across both models and field studies (Table S1). The selection of a minimum size threshold matters because past literature has shown that certain size ranges have specific GCCN effects.[1-4] Most studies in Table S1 relied on a minimum diameter threshold of 2 µm particle diameter followed by 1 µm. The range of minimum diameter thresholds extended from 0.5 µm to 5 µm, with the caveat that we omitted definitions applying to ultragiant CCN. Certain studies rely on measurements at ambient relative humidity, which complicates comparison to studies based on dry diameter. To address the latter limitation, parameterizations exist to convert from ambient to dry diameter (e.g.,[5, 6]).

Of relevance to GCCN size definitions is the more commonly used "coarse" designation for aerosol particles above a certain diameter. The United States Environmental Protection Agency (EPA) defines coarse particles as those with aerodynamic diameters between 2.5 and 10 µm.[7] Models define coarse aerosol types in various ways: dry diameters between 0.8 and 3.65 µm for sea salt and between 0.59 and 2.75 µm for dust (Community Atmosphere Model [CAM5.1]);[8] diameter above 2.5 µm (Goddard Earth Observing System model of atmospheric Chemistry [GEOS-Chem]);[9] dry radius between 0.005 µm and 60 µm for sea salt and dust (Global Aerosol Data Set [GADS] and Optical Properties of Aerosols and Clouds [OPAC]);[10-12] radius below 5 µm for dust (Navy Aerosol Analysis and Prediction System (NAAPS);[13, 14] diameter between 1 and 20 µm (Semi-Arid Climate and Environment Observation Station (SACOL);[15] diameter greater than 0.5 µm for sea salt and dust (Model for Ozone and Related Chemical Tracers, version 2 (MOZART-2).[16] The use of many different size-based definitions of coarse aerosol types in models contributes to the uncertainty of GCCN effects.

Table S1. Summary of selected studies examining GCCN in terms of study type, region, and how they defined the minimum size (D_p = diameter; r = radius, $r_{m,aero}$ = geometric mean radius) and whether the latter was at dry or ambient relative humidity (RH) conditions. Note that some studies used terminology other than GCCN such as "large/coarse particles". Furthermore, this table omits attention to ultragiant CCN, which is yet another class of large particles discussed in modeling and experimental studies. Blank cells indicate there was insufficient information with the caveat that modeling studies were assumed to have referred to dry diameters if they did not explicitly state so.

Reference	Instrument/Model	RH	Minimum size threshold	Region
Johnson et al.[17]	coated microscope glass sides	Ambient	$D_p > 1$ µm	Missouri
Ochs and Semonin[18]	numerical model of cloud microphysics	Dry	r > 1 µm	Missouri
Hindman and Radke[19]	coated microscope glass sides and Scanning Electron Microscope (SEM)	Ambient	$D_p \geq 2$ µm	Washington
Feingold et al.[20]	Forward Scattering Spectrometer Probe (FSSP) and numerical models	Ambient	r > 5 µm	Southern Ocean
Szumowski et al.[21]	Lagrangian drop-growth trajectory model	Dry	$D_p \geq 2$ µm	Hawaii
Yin et al.[22]	cloud model combined with dynamic and microphysical modes	Dry	$D_p > 2$ µm	
Lasher-Trapp et al.[23]	Three-dimensional cloud model	Dry	r > 1 µm	Florida
Blyth et al.[24]	Forward Scattering Spectrometer Probe (FSSP) and stochastic coalescence model		r > 1 µm	Florida
Levin et al.[25]	Scanning electron microscopy	Dry	$D_p > 0.5$ µm	Eastern Mediterranean
Colon-Robles et al.[26]	Forward Scattering Spectrometer Probe (FSSP)	Dry	r > 1 µm	Western Atlantic

Kogan and Mechem[27]	Coupled Ocean / Atmosphere Mesoscale Prediction System (COAMPS)	Ambient	$r > 1$ µm	Western Atlantic
Teller and Levin[28]	Tel-Aviv University Cloud Model (TAU-CM)	Dry	$D_p > 0.5$ µm	Eastern Mediterranean
Zhang et al.[1]	One-dimensional cloud microphysics model	Dry	$r > 1$ µm	
Mechem and Kogan[29]	Bulk parametrization model	Dry	$r > 3$ µm	Western Atlantic
Jensen and Lee[2]	stochastic Monte Carlo cloud model	Dry	$r > 0.5$ µm	
Rosenfeld et al.[30]	conceptual model	Dry	$D_p > 1$ µm	
Cheng et al.[31]	Regional Atmospheric Modeling System (RAMS)	Dry	$D_p \geq 5$ µm	Western Atlantic
Kogan et al.[32]	Systems for Atmospheric Modeling - Explicit Microphysics (SAMEX)	Dry	$r > 1$ µm	Western Atlantic
Dagan et al.[3]	Tel-Aviv University Cloud Model (TAU-CM)	Dry	$D_p > 1$ µm	Idealized tropical moist environment
Jung et al.[33]	Cloud and Aerosol Spectrometer (CAS)	Ambient	$D_p \geq 1$ µm	Northeastern Pacific Ocean
Sorooshian et al.[34]	Cloud and Aerosol Spectrometer (CAS)	Ambient	$D_p > 2$ µm	Northeastern Pacific Ocean
Dadashazar et al.[35]	Cloud and Aerosol Spectrometer (CAS)	Ambient	$D_p > 5$ µm	Northeastern Pacific Ocean
Jensen et al.[4]	Drop growth model	Dry	$r > 0.5$ µm	Idealized marine environment
Dror et al.[36]	Optical particle counter (OPC) and Scanning Mobility Particle Sizer (SMPS)	Dry	$D_p > 5$ µm	Atlantic Ocean, Caribbean Sea, and Pacific Ocean

Schlosser et al.[37]	Cloud and Aerosol Spectrometer (CAS)	Dry	$D_p \geq 1$ μm	Northeastern Pacific Ocean
Hoffmann and Feingold[38]	Models of varying complexity	Dry	$r_{m,aero} = 0.9$ μm	

Table S2. Linear regression results between MINALT/ACT GCCN particle volume concentrations (μm^3 cm^{-3}) above different minimum dry diameters versus cloud water tracer species for MONARC, ACTIVATE, ACTIVATE winter, and ACTIVATE summer. The values in the table represent R^2 with number of points used in the calculations in parenthesis. The bolded values are statistically significant with p values below 0.05.

	Na$^+$ MINALT	**Na$^+$ ACT**	**Cl$^-$ MINALT**	**Cl$^-$ ACT**	**nss Ca^{2+} MINALT**	**nss Ca^{2+} ACT**
MONARC						
$V_{>1}$	**0.41 (30)**	**0.23 (30)**	**0.34 (30)**	0.11 (30)	**0.25 (27)**	0.03 (27)
$V_{>2}$	**0.38 (27)**	**0.17 (22)**	**0.28 (27)**	0.06 (22)	0.14 (24)	0.01 (20)
$V_{>3}$	0.12 (24)	0.07 (13)	0.12 (24)	0.01 (13)	0.09 (21)	0.00 (12)
$V_{>5}$	0.02 (12)	0.01 (8)	0.01 (12)	0.01 (8)	0.00 (11)	0.04 (7)
$V_{>10}$	0.00 (0)	0.02 (1)	0.00 (0)	0.02 (1)	0.00 (0)	0.05 (1)
ACTIVATE: All						
$V_{>1}$	**0.46 (53)**	0.04 (51)	**0.44 (53)**	0.05 (51)	0.13 (15)	0.00 (15)
$V_{>2}$	**0.46 (53)**	0.04 (51)	**0.44 (53)**	0.05 (51)	0.12 (15)	0.00 (15)
$V_{>3}$	**0.44 (53)**	0.03 (51)	**0.42 (53)**	0.03 (51)	0.12 (15)	0.03 (15)
$V_{>5}$	**0.51 (53)**	0.04 (46)	**0.49 (53)**	0.05 (46)	0.04 (15)	0.03 (13)
$V_{>10}$	**0.58 (44)**	0.00 (24)	**0.57 (44)**	0.00 (24)	0.09 (14)	0.01 (4)
ACTIVATE: Winter						
$V_{>1}$	**0.14 (32)**	0.01 (32)	**0.14 (32)**	0.01 (32)	0.70 (5)	0.21 (5)
$V_{>2}$	**0.15 (32)**	0.01 (32)	**0.14 (32)**	0.01 (32)	0.70 (5)	0.21 (5)
$V_{>3}$	**0.15 (32)**	0.00 (32)	**0.15 (32)**	0.00 (32)	0.70 (5)	0.17 (5)
$V_{>5}$	**0.15 (32)**	0.00 (31)	**0.15 (32)**	0.00 (31)	0.26 (5)	0.14 (5)
$V_{>10}$	**0.19 (28)**	0.06 (20)	**0.20 (28)**	0.05 (20)	0.59 (5)	0.13 (2)
ACTIVATE: Summer						
$V_{>1}$	**0.53 (21)**	0.11 (19)	**0.51 (21)**	0.12 (19)	0.46 (10)	0.02 (10)
$V_{>2}$	**0.54 (21)**	0.10 (19)	**0.51 (21)**	0.11 (19)	0.48 (10)	0.02 (10)
$V_{>3}$	**0.56 (21)**	0.04 (19)	**0.54 (21)**	0.04 (19)	0.36 (10)	0.04 (10)
$V_{>5}$	**0.63 (21)**	0.12 (15)	**0.62 (21)**	0.13 (15)	0.61 (10)	0.02 (8)
$V_{>10}$	**0.74 (16)**	0.00 (4)	**0.73 (16)**	0.00 (4)	0.44 (9)	0.03 (2)

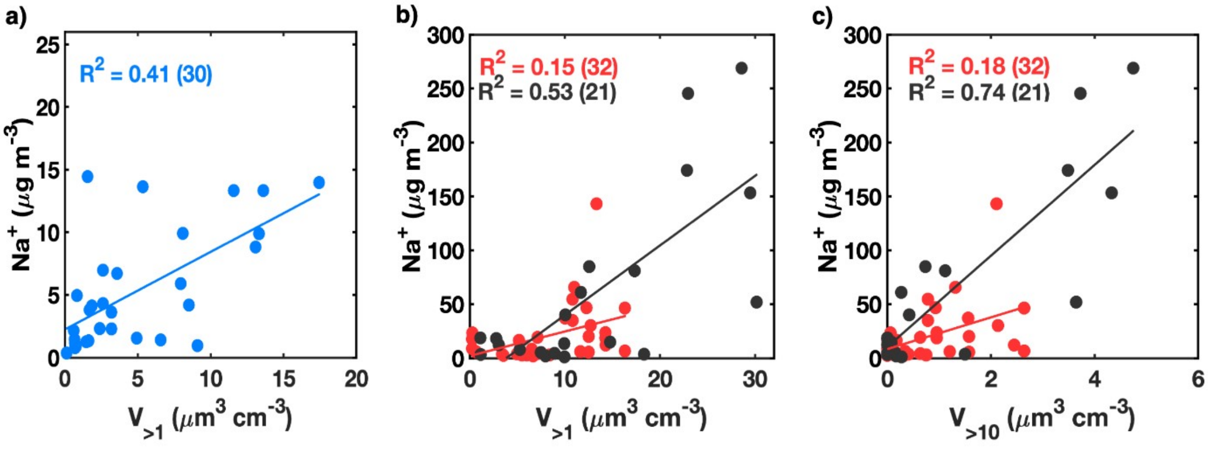

Fig. S1. Cloud water sodium versus MINALT particle volume concentration above a dry diameter of (a) 1 µm for MONARC, (b) 1 µm for ACTIVATE separated by winter (red) and summer (black), and (c) 10 µm for ACTIVATE separated by winter (red) and summer (black). Number of data points are in parenthesis. The R^2 value of the cumulative data points for panels b and c are 0.46 and 0.58, respectively.

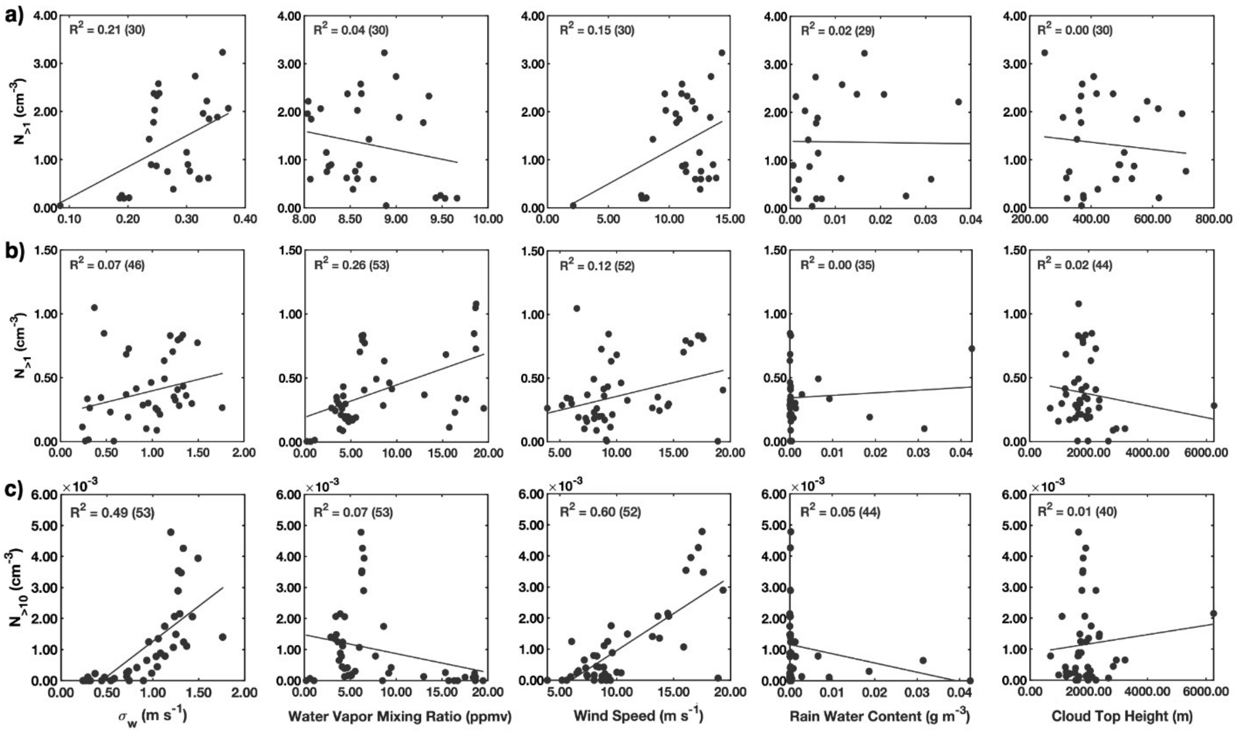

Fig. S2. Scatterplots of $N_{>1}$ versus five environmental parameters for both (a) MONARC and (b) ACTIVATE, in addition to (c) $N_{>10}$ versus the same parameters for ACTIVATE. Number of data points are in parenthesis.

REFERENCES

1. L. Zhang, D. V. Michelangeli and P. A. Taylor, Influence of aerosol concentration on precipitation formation in low-level, warm stratiform clouds, *Journal of Aerosol Science*, 2006, **37**, 203-217.
2. J. B. Jensen and S. Lee, Giant sea-salt aerosols and warm rain formation in marine stratocumulus, *Journal of the Atmospheric Sciences*, 2008, **65**, 3678-3694.
3. G. Dagan, I. Koren and O. Altaratz, Aerosol effects on the timing of warm rain processes, *Geophysical Research Letters*, 2015, **42**, 4590-4598.
4. J. B. Jensen and A. D. Nugent, Condensational growth of drops formed on giant sea-salt aerosol particles, *Journal of the Atmospheric Sciences*, 2017, **74**, 679-697.
5. H. E. Gerber, *Relative-humidity parameterization of the Navy Aerosol Model (NAM)*, N. R. Laboratory Report 8956, Washington, D.C., 1985.
6. J. S. Reid, B. Brooks, K. K. Crahan, D. A. Hegg, T. F. Eck, N. O'Neill, G. de Leeuw, E. A. Reid and K. D. Anderson, Reconciliation of coarse mode sea-salt aerosol particle size measurements and parameterizations at a subtropical ocean receptor site, *Journal of Geophysical Research: Atmospheres*, 2006, **111**.
7. W. E. Wilson and H. H. Suh, Fine particles and coarse particles: concentration relationships relevant to epidemiologic studies, *Journal of the Air and Waste Management Association*, 1997, **47**, 1238-1249.
8. X. Liu, R. C. Easter, S. J. Ghan, R. Zaveri, P. Rasch, X. Shi, J.-F. Lamarque, A. Gettelman, H. Morrison and F. Vitt, Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5, *Geoscientific Model Development*, 2012, **5**, 709.
9. R. J. Park, Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *Journal of Geophysical Research*, 2004, **109**.
10. L. Jaeglé, P. K. Quinn, T. S. Bates, B. Alexander and J. T. Lin, Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations, *Atmospheric Chemistry and Physics* 2011, **11**, 3137-3157.
11. M. Hess, P. Koepke and I. Schult, Optical properties of aerosols and clouds: The software package OPAC, *Bulletin of the American Meteorological Society*, 1998, **79**, 831-844.
12. P. Köpke, M. Hess, I. Schult and E. Shettle, *Global Aerosol Data Set*, Max-Planck-Inst. für Meteorologie, 1997.
13. T. F. Hogan and T. E. Rosmond, The description of the Navy Operational Global Atmospheric Prediction System's spectral forecast model, *Monthly Weather Review*, 1991, **119**, 1786-1815.
14. T. F. Hogan and L. R. Brody, Sensitivity studies of the Navy's global forecast model parameterizations and evaluation of improvements to NOGAPS, *Monthly Weather Review*, 1993, **121**, 2373-2395.
15. J. Huang, W. Zhang, J. Zuo, J. Bi, J. Shi, X. Wang, Z. Chang, Z. Huang, S. Yang and B. Zhang, An overview of the semi-arid climate and environment research observatory over the Loess Plateau, *Advances in Atmospheric Sciences*, 2008, **25**, 906.

16. X. Tie, S. Madronich, S. Walters, D. P. Edwards, P. Ginoux, N. Mahowald, R. Zhang, C. Lou and G. Brasseur, Assessment of the global impact of aerosols on tropospheric oxidants, *Journal of Geophysical Research: Atmospheres*, 2005, **110**.
17. D. B. Johnson, Ultragiant urban aerosol particles, *Science*, 1976, **194**, 941-942.
18. H. T. Ochs, III and R. G. Semonin, Sensitivity of a Cloud Microphysical Model to an Urban Environment, *Journal of Applied Meteorology*, 1979, **18**, 1118-1129.
19. E. E. Hindman, P. V. Hobbs and L. F. Radke, Cloud condensation nucleus size distributions and their effects on cloud droplet size distributions, *Journal of the Atmospheric Sciences*, 1977, **34**, 951-956.
20. G. Feingold, W. R. Cotton, S. M. Kreidenweis and J. T. Davis, The impact of giant cloud condensation nuclei on drizzle formation in stratocumulus: Implications for cloud radiative properties, *Journal of the Atmospheric Sciences*, 1999, **56**, 4100-4117.
21. M. J. Szumowski, R. M. Rauber and H. T. Ochs, III, The Microphysical Structure and Evolution of Hawaiian Rainband Clouds. Part III: A Test of the Ultragiant Nuclei Hypothesis, *Journal of the Atmospheric Sciences*, 1999, **56**, 1980-2003.
22. Y. Yin, Z. Levin, T. G. Reisin and S. Tzivion, The effects of giant cloud condensation nuclei on the development of precipitation in convective clouds—A numerical study, *Atmospheric Research*, 2000, **53**, 91-116.
23. S. G. Lasher-Trapp, C. A. Knight and J. M. Straka, Early Radar Echoes from Ultragiant Aerosol in a Cumulus Congestus: Modeling and Observations, *Journal of the Atmospheric Sciences*, 2001, **58**, 3545-3562.
24. A. M. Blyth, S. G. Lasher-Trapp, W. A. Cooper, C. A. Knight and J. Latham, The role of giant and ultragiant nuclei in the formation of early radar echoes in warm cumulus clouds, *Journal of the Atmospheric Sciences*, 2003, **60**, 2557-2572.
25. Z. Levin, A. Teller, E. Ganor and Y. Yin, On the interactions of mineral dust, sea-salt particles, and clouds: A measurement and modeling study from the Mediterranean Israeli Dust Experiment campaign, *Journal of Geophysical Research: Atmospheres*, 2005, **110**.
26. M. Colón-Robles, R. M. Rauber and J. B. Jensen, Influence of low-level wind speed on droplet spectra near cloud base in trade wind cumulus, *Geophysical Research Letters*, 2006, **33**.
27. Y. L. Kogan and D. B. Mechem, *Improvement of the Cloud Physics Formulation in the US Navy Coupled Ocean-Atmosphere Mesoscale Prediction System (COAMPS)*, Oklahoma Univ Norman, 2006.
28. A. Teller and Z. Levin, The effects of aerosols on precipitation and dimensions of subtropical clouds: a sensitivity study using a numerical cloud model, *Atmospheric Chemistry and Physics,* 2006, **6**, 67-80.
29. D. B. Mechem and Y. L. Kogan, A bulk parameterization of giant CCN, *Journal of the Atmospheric Sciences*, 2008, **65**, 2458-2466.
30. D. Rosenfeld, U. Lohmann, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell and M. O. Andreae, Flood or drought: How do aerosols affect precipitation?, *Science*, 2008, **321**, 1309-1313.
31. W. Y. Cheng, G. G. Carrió, W. R. Cotton and S. M. Saleeby, Influence of cloud condensation and giant cloud condensation nuclei on the development of precipitating trade wind cumuli in a large eddy simulation, *Journal of Geophysical Research: Atmospheres*, 2009, **114**.

32. Y. L. Kogan, D. B. Mechem and K. Choi, Effects of sea-salt aerosols on precipitation in simulations of shallow cumulus, *Journal of the Atmospheric Sciences*, 2012, **69**, 463-483.
33. E. Jung, B. A. Albrecht, H. H. Jonsson, Y.-C. Chen, J. H. Seinfeld, A. Sorooshian, A. R. Metcalf, S. Song, M. Fang and L. M. Russell, Precipitation effects of giant cloud condensation nuclei artificially introduced into stratocumulus clouds, *Atmospheric Chemistry and Physics*, 2015, **15**, 5645-5658.
34. A. Sorooshian, G. Prabhakar, H. Jonsson, R. K. Woods, R. C. Flagan and J. H. Seinfeld, On the presence of giant particles downwind of ships in the marine boundary layer, *Geophysical Research Letters*, 2015, **42**, 2024-2030.
35. H. Dadashazar, Z. Wang, E. Crosbie, M. Brunke, X. Zeng, H. Jonsson, R. K. Woods, R. C. Flagan, J. H. Seinfeld and A. Sorooshian, Relationships between giant sea salt particles and clouds inferred from aircraft physicochemical data, *Journal of Geophysical Research: Atmospheres*, 2017, **122**, 3421-3434.
36. T. Dror, J. M. Flores, O. Altaratz, G. Dagan, Z. Levin, A. Vardi and I. Koren, Sensitivity of warm clouds to large particles in measured marine aerosol size distributions–a theoretical study, *Atmospheric Chemistry and Physics*, 2020, **20**, 15297-15306.
37. J. S. Schlosser, H. Dadashazar, E. L. Edwards, A. Hossein Mardi, G. Prabhakar, C. Stahl, H. H. Jonsson and A. Sorooshian, Relationships between supermicrometer sea salt aerosol and marine boundary layer conditions: insights from repeated identical flight patterns, *Journal of Geophysical Research: Atmospheres*, 2020, **125**, e2019JD032346.
38. F. Hoffmann and G. Feingold, Cloud Microphysical Implications for Marine Cloud Brightening: The Importance of the Seeded Particle Size Distribution, *Journal of the Atmospheric Sciences*, 2021, **78**, 3247-3262.