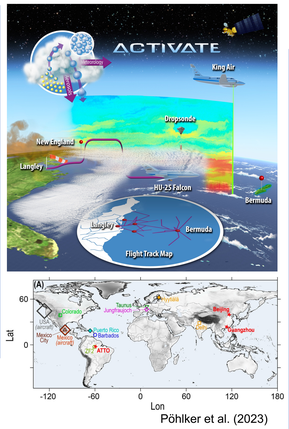
CCN Closure Analysis for the ACTIVATE Campaign

Cassidy Soloff[1], Ewan C. Crosbie[2,3], Simon Kirschler[4,5], Richard H. Moore[2], Michael A. Shook[2], Shuaiqi Tang[6,7], Kenneth L. Thornhill[2,3], Christiane Voigt[4,5], Hailong Wang[6], Edward L. Winstead[2,3], Luke D. Ziemba[2], Armin Sorooshian[1]

[1]University of Arizona, [2]NASA Langley Research Center, [3]Analytical Mechanics Associates, [4]Institute of Atmospheric Physics, German Aerospace Center, [5]Institute of Atmospheric Physics, University Mainz, Germany, [6] Pacific Northwest National Laboratory, [7]Nanjing University

Background and Objectives

- NASA's Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE): 162 joint research flights with two spatially coordinated aircraft over three years (2020 – 2022)
- Question: Can cloud condensation nuclei (CCN) concentrations be predicted using particle size spectra and chemical composition over the northwest Atlantic?
- Goal: Advance understanding of droplet activation in the northwest Atlantic, a complex region with diverse meteorology and influence from continental and marine aerosol sources
- The hygroscopicity parameter, κ, describes the ability of a particle to uptake water
- $\kappa_{org} = 0.12 \pm 0.02$ and $\kappa_{inorg} = 0.63 \pm 0.01$ perform well globally (Pöhlker et al., 2023)



Pöhlker et al. (2023)

Predicting CCN

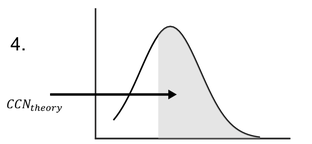
1. Merge data
   Size Distribution: Scanning Mobility Particle Sizer (SMPS) [10-100 nm] + Laser Aerosol Spectrometer (LAS) [100-3000 nm]
   Chemistry: Aerosol Mass Spectrometer (AMS)
   Cloud Screening: Fast Cloud Droplet Probe (FCDP)
   CCN Concentration: CCN spectrometer
2. Calculate bulk κ using κ_{org} and κ_{inorg} from Pöhlker et al. (2023)
3. Calculate critical activation diameter (D_c) for supersaturation (SS) of the CCN spectrometer using κ-Köhler theory
4. Integrate SMPS+LAS size distribution for D_p>D_c to get predicted CCN concentration

2. $$\kappa = \sum_i \varepsilon_i \kappa_i$$

3. $$S(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1-\kappa)} \exp\left(\frac{4\sigma_{s/a} M_w}{RT\rho_w D}\right)$$

Petters & Kreidenweis (2007)

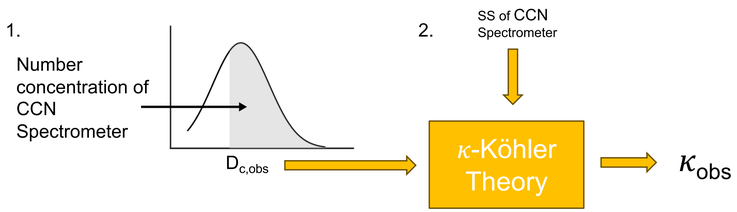
4.



Kulkarni et al. (2023)

Finding Observed κ

1. Back-integrating LAS+SMPS until the concentration equals the CCN concentration gives us observed D_c
2. Plugging into κ-Köhler, along with SS of CCN spectrometer, gives us κ_{obs}



Acknowledgments

NASA grant 80NSSC19K0442 in support of ACTIVATE, a NASA Earth Venture Suborbital-3 (EVS-3) investigation funded by NASA's Earth Science Division and managed through the Earth System Science Pathfinder Program Office.

References

Pöhlker, M.L., Pöhlker, C., Quaas, J., Mülmenstädt, J., Pozzer, A., Andreae, M.O., Artaxo, P., Block, K., Coe, H., Ervens, B., Gallimore, P., Gaston, C.J., Gunthe, S.S., Henning, S., Herrmann, H., Krüger, O.O., McFiggans, G., Poulain, L., Raj, S.S., Reyes-Villegas, E., Royer, H.M., Walter, D., Wang, Y., Pöschl, U., 2023. Global organic and inorganic aerosol hygroscopicity and its effect on radiative forcing. Nat Commun 14, 6139. 10.1038/s41467-023-41695-8.
Petters, M.D., Kreidenweis, S.M., 2007. A single parameter representation of hygroscopic growth and cloud condensation nucleus activity. Atmos. Chem. Phys. 7, 1961-1971. https://doi.org/10.5194/acp-7-1961-2007.
Kulkarni, G., Mei, F., Shilling, J.E., Wang, J., Reveggino, R.P., Flynn, C., Zelenyuk, A., Fast, J., 2023. Cloud Condensation Nuclei Closure Study Using Airborne Measurements Over the Southern Great Plains. J Geophys Res-Atmos 128, e2022JD037964. https://doi.org/10.1029/2022JD037964.

Results

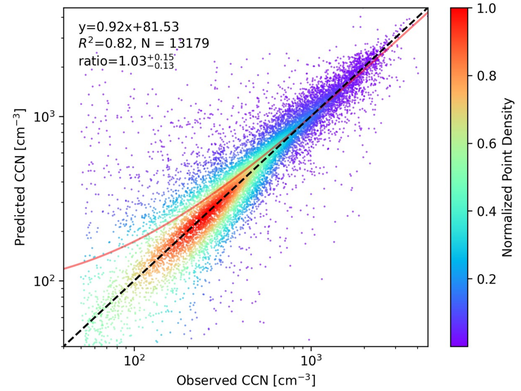


Figure 1. Predicted CCN concentrations versus the measured CCN concentration across all ACTIVATE flights. Points are color-coded based on a Gaussian kernel density normalized by the peak density. The equation of the best-fit line, R^2, number of points (N), and the median ratio of predicted over observed CCN (plus or minus the difference between the median and the 75th/25th percentiles) are shown in the top left corner.

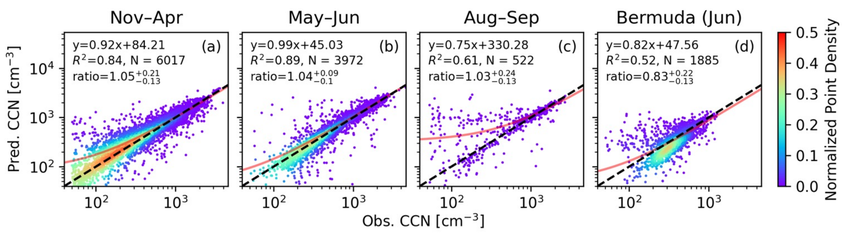


Figure 2. Same as Fig. 1 but categorized by season.

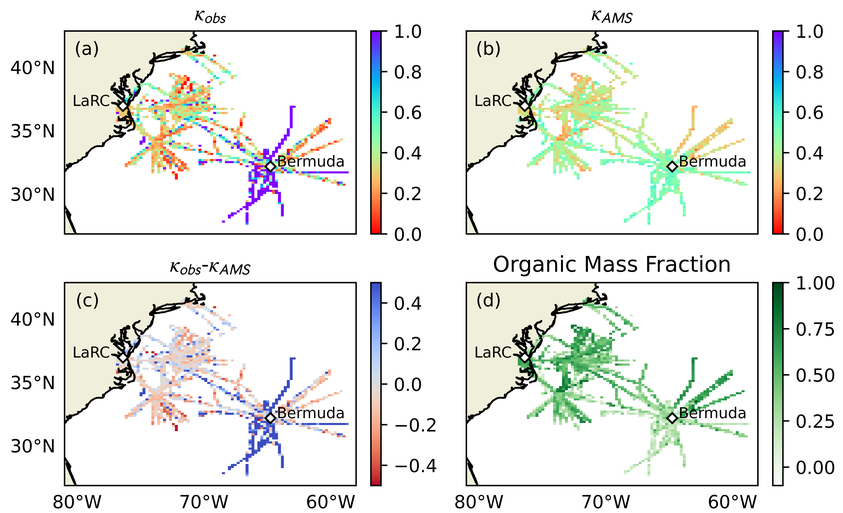


Figure 3. Spatial distribution of median (a) κ_{obs}, (b) κ_{AMS}, (c) their difference, and (d) OMF across all ACTIVATE flights. The two major bases of operations are shown with markers: NASA Langley Research Center (LaRC) in Hampton, Virginia and Bermuda. Each pixel has latitude by longitude dimensions of 0.16° by 0.23°.

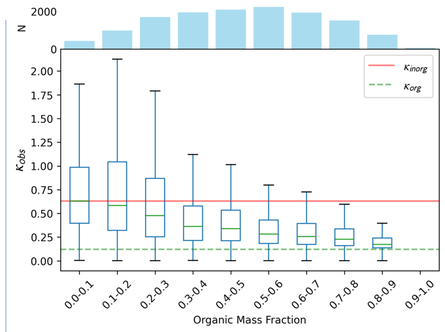


Figure 4. Box plots of observed κ segmented by AMS OMF. The number of points (N) is shown at the top of the plot for each OMF bin. Boxplots are shown only when there were > 75 observations. κ_{org} of 0.12 and κ_{inorg} of 0.63 (adopted from Pöhlker et al. (2023)) plotted in red and green horizontal lines, respectively.

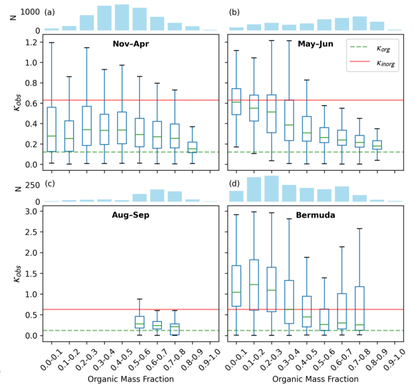


Figure 5. Same as Fig. 4 but separated by season.

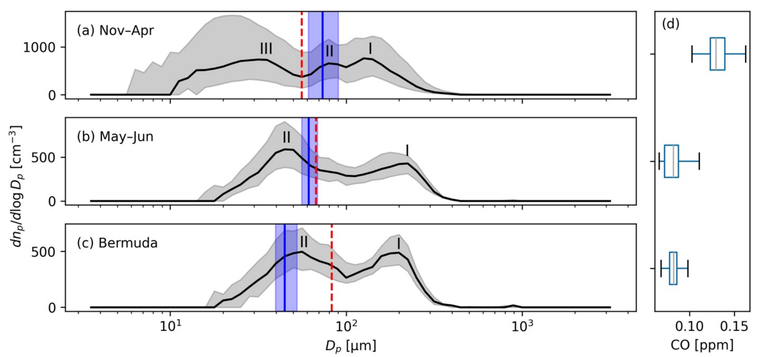


Figure 6. This figure uses data when OMF < 0.3. Panels a–c display median size distributions with the shaded regions representing the 75th and 25th percentiles. The blue line is the median $D_{p,c}$ with the shaded region representing the 75th and 25th percentiles. The red dashed line is the effective diameter. The Roman numerals mark the locations of the peaks from largest to smallest. Panel d displays boxplots of CO concentrations corresponding to the adjacent seasons in panels a–c.

Conclusions

- Assuming internally mixed particles and κ values from Pöhlker et al. (2023) produced to a CCN_{pred}/CCN_{obs} near unity (1.03) when using the entire dataset . The May–Jun season performed best (CCN_{pred}/CCN_{obs} = 1.04) and Bermuda measurements performed worst (CCN_{pred}/CCN_{obs} = 0.83).
- When averaged across the entire campaign, κ_{obs} aligned with κ_{AMS} across the full spectrum of OMF. Spatially, the median difference between κ_{obs} and κ_{AMS} was minimal near the coast, but near Bermuda, particles were much more hygroscopic than predicted by the AMS.
- κ_{obs} had the highest variability at low OMF fractions, with κ_{obs} >> κ_{inorg} in Bermuda and κ_{obs} < κ_{inorg} for Nov–Apr when OMF < 0.3. These variations in κ could have arisen from differences in continental emissions and mixing states between seasons.