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Aerosol Hygroscopic Growth, Mixing State, and Cloud Condensation Nuclei Activity during TRACER Field Campaign Report

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DISCLAIMER

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Acronyms and Abbreviations

ANC	ancillary site
ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
DMA	differential mobility analyzer
FIMS	fast integrated mobility spectrometer
HFIMS	relative humidity-controlled fast integrated mobility spectrometer
HTDMA	humidified tandem differential mobility analyzer
IOP	intensive operational period
RH	relative humidity
SCCN	size-resolved cloud condensation nuclei
TRACER	Tracking Aerosol Convection Interactions Experiment
UPS	uninterruptible power supply

Contents

Figures

1.0 Summary

Convective clouds play a critical role in the Earth's climate system. Recent research has shown that a realistic representation of convective processes is critical to constraining climate sensitivity in global climate models. Theoretical and modeling studies showed that aerosols could have strong dynamic feedback to convection in warm and humid environments through enhancing ice-related processes and condensational growth. A few observation-based studies also suggested the influence of aerosols on convective cloud and precipitation properties. However, robust observational quantification of an aerosol effect on convective clouds isolated from other factors remains elusive.

Understanding the impact of aerosol on convective clouds requires knowledge of the cloud condensation nuclei (CCN) spectrum, which represents the number of particles that uptake water and form cloud droplets as a function of supersaturation. The water uptake by aerosol is also of critical importance for the direct interaction of aerosol with radiation (i.e., aerosol direct effect) due to light scattering and absorption by aerosol. While both droplet activation under supersaturated conditions (i.e., relative humidity RH > 100%) and the hygroscopic growth under sub-saturated conditions (i.e., RH < 100%) are strongly influenced by particle hygroscopicity, the thermodynamic regimes and measurement methods are quite different. Aerosol particles, especially organic particles, can exhibit higher hygroscopicity for droplet activation than that for hygroscopic growth. In the subsaturated regime, the hygroscopicity of organic particles can also vary strongly with RH. However, global climate models usually treat organic species in aerosols with a constant hygroscopicity, potentially introducing substantial uncertainties in the quantification of aerosol radiative effects.

During the intensive operational period (IOP) of the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) use facility's Tracking Aerosol Convection Interactions Experiment (TRACER), two guest instruments, a size-resolved cloud condensation nuclei (SCCN) system and an RH-controlled fast integrated mobility spectrometer (HFIMS), were deployed at the ancillary site (ANC) from July 3rd to September 10th, 2022 (Figure 1).

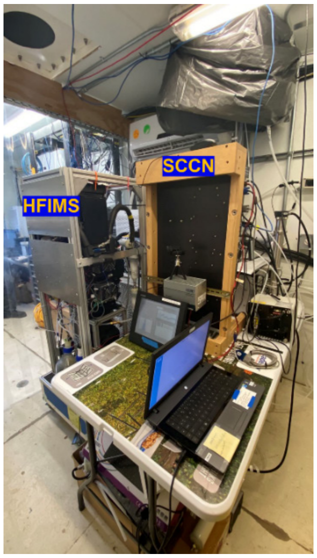

Figure 1. SCCN system and HFIMS deployed at the ANC site during TRACER IOP.

The SCCN system provides activation fraction spectra of size-selected particles and allows the derivation of particle hygroscopicity (κ_{CCN}) under supersaturated conditions (Frank et al. 2006, Moore et al. 2010). The spectrum of activation fraction was obtained for five dry particle sizes ranging from ~ 40 nm to ~ 165 nm every 60 to 90 minutes. The HFIMS is based on the FIMS technology invented by our group, and jointly developed by Aerosol Dynamics Inc. and our group with support from the U.S. Department of Energy Small Business Innovation Research program (Pinterich et al. 2017b, Wang et al. 2019, Zhang et al. 2022, Zhang et al. 2021). The key component of the HFIMS is a water-based FIMS, which measures the size distribution of humidified particles with high time resolution (Pinterich et al. 2017b, 2017a). By replacing the second DMA inside a traditional humidified tandem differential mobility analyzer (HTDMA) with the water-based FIMS, the measurements of the size distribution of humidified particles, and thus the hygroscopic growth factor, is significantly accelerated in the HFIMS. During the deployment, hygroscopic growth of particles with dry diameter of 35, 50, 75, 110, 165, and 265 nm at 75%, 85%, and 90% RH were measured every 22 minutes. Particle hygroscopicity under sub-saturated conditions (κ_{GF}) can be derived from the measured particle hygroscopic growth. The combination of the two instruments provided size-resolved aerosol hygroscopicity under both super- and sub-saturated conditions.

2.0 Results

2.1 Preliminary Results

We classified the airmasses arriving at the ANC site during the IOP into two clusters. The first cluster represents marine airmasses from the Gulf of Mexico with minimum influences from continental emissions. The second cluster includes the continental airmasses that often were strongly influenced by urban emissions (Figure 2).

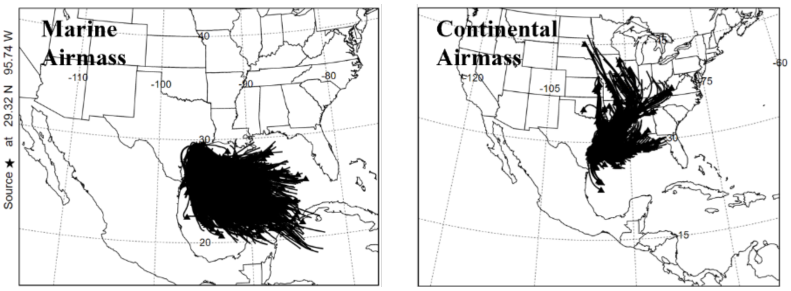

Figure 2. Clustered backward trajectories of airmass arriving at the ANC site during the TRACER IOP.

Figure 3 shows the particle size distribution, chemical composition, average κ_{GF} under RH=90%, and κ_{CCN} on August 16th (marine airmass) and August 13th (continental airmass). The marine airmass exhibits a bimodal size distribution while the aerosols in the continental airmass on August 13th display a unimodal size distribution (Figure 3a). For the marine airmass, sulfate is the dominant chemical species throughout the day, followed by organics and ammonium, and the contribution from nitrate to the submicron aerosol mass is minor. In comparison, for the continental airmass, organics are the most dominant, followed by nitrate, sulfate, and ammonium (Figure 3b). Both κ_{GF} and κ_{CCN} show a general increasing trend with particle diameter (Figure 3c,d), suggesting higher fractions of more hygroscopic species (e.g., sulfate) in larger particles. Aerosols in the marine airmass show higher hygroscopicity than those in the continental airmass (Figure 3c,d), consistent with the fact that the composition of aerosols in marine airmasses is more dominated by sulfate.

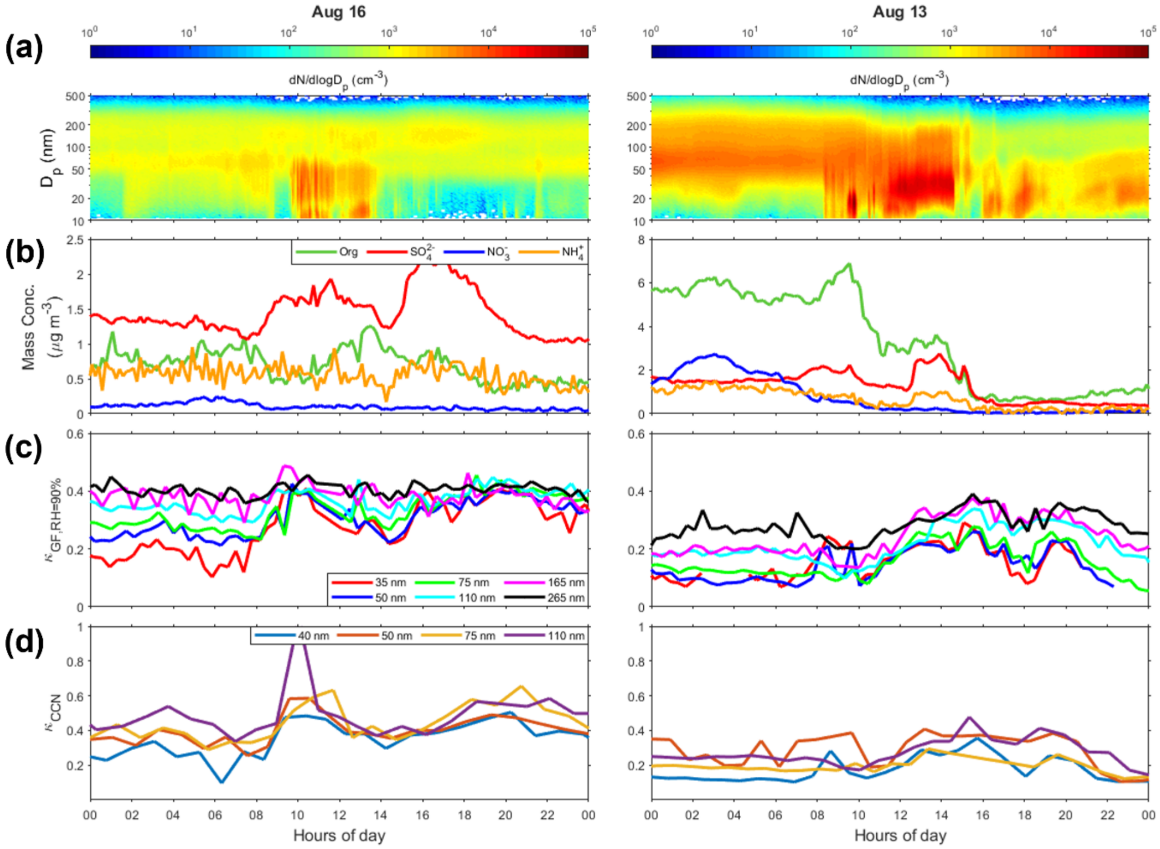

Figure 3. (a) Contour plot of particle number size distributions. (b) Mass concentrations of organics, sulfate, nitrate, and ammonium. (c) Average κ_{GF} for particles with diameters of 35, 50, 75, 110, 165, and 265 nm at 90% RH. (d) κ_{CCN} for particles with diameters of 40, 50, 75, and 110 nm. The left and right panels show data collected on August 16th (marine airmass) and August 13th (continental airmass), respectively.

2.2 Further Research Opportunities

Data from these two instruments will allow high-level data quality checks through closure studies, and the development of a CCN spectrum with high time resolution and value-added aerosol microphysics data sets that include size spectrum, mixing state, hygroscopicity growth, and CCN activity. These value-added data sets will allow the scientific community to examine both the impact of aerosols on physical processes and properties of deep convective systems and the impact of convective systems on the transport and cloud-processing of aerosols.

Data from the HFIMS and SCCN system can be combined with other ARM Aerosol Observing System measurements to quantify the variations of aerosol hygroscopicity with saturation regime and RH, and elucidate the key controlling factors and underlying mechanisms for the variations. The measurements during the two-month deployment will allow us to statistically examine the values of κ_{CCN} and κ_{GF} for ambient aerosols. We will compare the values of κ_{CCN} and κ_{GF}, and examine the variation of κ_{GF} with

RH for representative aerosol types, including relatively clean background aerosols from the Gulf of Mexico (i.e., marine airmass), and those strongly influenced by urban pollution and/or regional biogenic emissions (continental airmass). We can gain important insights into the underlying mechanisms by examining the trend of κ_{GF} with RH and the dependence of κ_{CCN} with particle size and composition.

3.0 Publications and References

3.1 Presentations

Li, J, J Zhang, X Gong, S Spielman, A Singh, C Kuang, M Zawadowicz, and J Wang. 2022. "Size-resolved Aerosol Hygroscopicities under both Supersaturated and Subsaturated Conditions at a Rural Site during TRACER." ARM/ASR Principal Investigators Meeting, Washington, D.C.

3.2 References

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Pinterich, T, SR Spielman, S Hering, and J Wang. 2017a. "A water-based fast integrated mobility spectrometer (WFIMS) with enhanced dynamic size range." *Aerosol Science and Technology* 51(10): 1212−1222, https://doi.org/10.1080/02786826.2017.1338664

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Zhang, JS, S Spielman, Y Wang, GJ Zheng, XD Gong, S Hering, and J Wang. 2021. "Rapid measurement of RH-dependent aerosol hygroscopic growth using a humidity-controlled fast integrated mobility spectrometer (HFIMS)." *Atmospheric Measurement Techniques* 14(8): 5625−5635, https://doi.org/10.5194/amt-14-5625-2021

4.0 Lessons Learned

Measurements were interrupted near the end of August due to a power outage and failure of UPS. A more robust UPS unit is highly desirable.

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