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TRACER-Carbonaceous Aerosol Thrust (CAT)-Los Alamos National Laboratory (LANL) Field Campaign Report

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March 2023



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Acronyms and Abbreviations

AMF	ARM Mobile Facility
AOS	Aerosol Observing System
ARM	Atmospheric Radiation Measurement
BC	black carbon
CAT	Carbonaceous Aerosol Thrust
H-CAPS	humidified cavity attenuated phase shift-single scattering albedo particulate matter monitor
IOP	intensive operational period
LANL	Los Alamos National Laboratory
PASS-3	photoacoustic soot spectrometer
PI	principal investigator
ppm	parts per million
RH	relative humidity
SP2	single-particle soot photometer
SP-AMS	soot particle aerosol mass spectrometer
TRACER	Tracking Aerosol Convection Interactions Experiment
UC-Davis	University of California-Davis

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1.0 Summary

The TRacking Aerosol Convection interactions ExpeRiment Carbonaceous Aerosols Thrust by Los Alamos National Laboratory (TRACER-CAT-LANL) was designed to complement the larger U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility TRACER science goals to understand convective cloud life cycles and aerosol-convection interactions. TRACER-CAT-LANL focused on understanding the relationship between particle composition and light absorption, and the influence of water uptake on this relationship. Our suite of measurements was designed to provide insight into how different internally mixed particles can act as efficient cloud seeds and contribute to warm-cloud invigoration. For example, light absorption by absorbing aerosols such as black carbon (BC) can increase localized warming, and in turn influence cloud formation and convective systems. However, it remains unclear to what extent coatings on BC further enhance absorption in the atmosphere as well as the specific role that water contributes to this enhancement. For this reason, TRACER-CAT-LANL characterized and quantified dry and humidified optical properties and measured the chemical composition of carbonaceous aerosols, including organic carbon and BC, over a period of one month in the summer during the TRACER intensive operational period (IOP).

We deployed the LANL-Guest-Aerosol Observing System (AOS) that was designed with a shared inlet, drying, and monitoring system at LANL to house a suite of aerosol and trace gas instrumentation based on the design of the new ARM AOS (Uin et al. 2019). We deployed the container and LANL-Guest instruments to Houston, Texas in July 2022 (<u>https://www.arm.gov/research/campaigns/amf2022tracer-cat-lanl</u>) for a one-month IOP. Our measurements were made at the T0 site in La Porte, Texas, to co-locate with the First ARM Mobile Facility (AMF1) Aerosol Observing System (AOS; see Figure 1). Unique in this deployment were the first field measurements using the humidified cavity attenuated phase shift-single scattering albedo particulate matter monitor (H-CAPS; Carrico et al. 2021).



Figure 1. (a) The LANL-TRACER-CAT science team (from left to right: Kyle Gorkowski, Allison Aiken, Manvendra Dubey, Spencer Jordan, Aaron Meyer, James Lee, Katherine Benedict, not pictured: Abu Sayeed Md Shawon) on the roof of the LANL-Guest AOS, (b) the LANL-Guest-AOS deployed at T0 during TRACER pictured on the far left, adjacent to the ARM AMF1 AOS.

The LANL-Guest-AOS was designed to house LANL plus additional guest instruments. For TRACER-CAT-LANL we hosted the instrumentation for TRACER-CAT-University of California-Davis

(UC-Davis)(https://www.arm.gov/research/campaigns/osc2022tracer-cat-ucdavis) led by Chris Cappa (principal investigator [PI]) and Qi Zhang (Co-Investigator) in addition to the LANL suite. Combining complementary measurements in one container enabled enhanced measurements for scientific discovery. For example, both the LANL and UC-Davis teams deployed a version of the H-CAPS on the shared inlet system to collectively determine wavelength-dependent properties of the humidified and non-humidified aerosol. Bulk and size-resolved aerosol chemical speciation was also measured with two separate soot particle aerosol mass spectrometers (SP-AMS). One alternately measured bulk non-refractory and BC plus non-refractory particle composition (LANL) while the other focused on the composition of the BC-containing particles in bulk and single-particle modes (UC-Davis).

On average throughout the July IOP, there were low urban and primary aerosol mass concentrations, with short periods (5-30 minutes) of direct plume intercepts. These periods were characterized by elevated refractory BC, non-refractory organic carbon, greenhouse gas, and carbon monoxide (CO) concentrations (see Figure 2). During the IOP, a number of particle growth events in our measured size distributions were also observed that included particles from 20 nm to 20 μ m. These events occurred earlier in the month, whereas larger particle size distributions were observed later in the month (see Figure 3). Regional models indicated that these larger particles were likely due to the long-range transport of Saharan dust.

2.0 Results

TRACER-CAT-LANL collected data continuously from July 1 to July 31, 2022. A heterogeneous mixture of fresh and aged aerosols was observed from local urban, shipping, and industrial emissions, regional fires, and long-range transport of Saharan dust. Figure 2 shows the carbon dioxide (CO₂), methane (CH₄), and carbon monoxide (CO) trace gas concentrations in parts per million (ppm). Distinct short plumes were observed frequently at night, presumably from a local source yet to be identified.

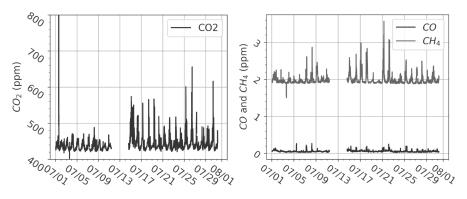


Figure 2. Trace gas measurements of distinct and recurrent short plumes using 10 min-averaged data.

Figure 3 shows the calculated total aerosol mass loadings for the PM_1 and PM_{10} size fractions to highlight the supermicron dust periods that impacted the site. An urban fire period was also observed. In future analysis we will investigate the speciation differences between what appear to be new particle formation events during the first half of the study and the dust periods that dominated the second half of the IOP.

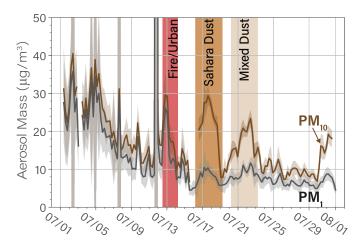


Figure 3. The time series of PM₁ and PM₁₀ aerosol mass concentrations showed distinct submicron and supermicron particle events. Data are plotted as 4-hour averages with standard deviation as shading.

The optical properties measured by the H-CAPS and photoacoustic soot spectrometer (PASS-3) are shown in Figure 4. The truncation corrections for the humidified optical properties of ambient aerosol is an area of active development and will be the focus of a future publication. An accurate truncation correction will give us a humidity-dependent aerosol absorption enhancement, which is a critical parameterization for more humid climate scenarios.

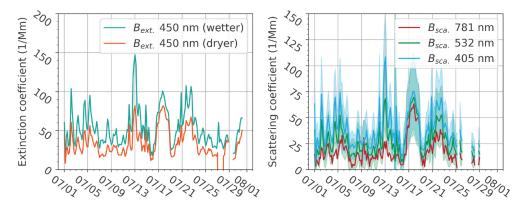


Figure 4. Humidity dependence of aerosol optical properties measured with the custom humidified cavity attenuated phase shift-single scattering albedo particulate matter monitor (H-CAPS) built by LANL. The right plot is the scattering coefficient as measured by the photoacoustic soot spectrometer (PASS-3). Data are shown as 4-hour averages for plotting purposes.

Complementing the physical and optical aerosol measurements were the chemically speciated measurements from the SP-AMS and single-particle soot photometer (SP2) shown in Figure 5. Aerosol chemical composition is be used to determine chemical processing and source identification for the local, regional, and long-range transported plumes. This data set, combined with co-located trace gas measurements (Figure 2), will be used to investigate the sources and chemical processing of the different plume events observed during the deployment, e.g., the nighttime local emissions.

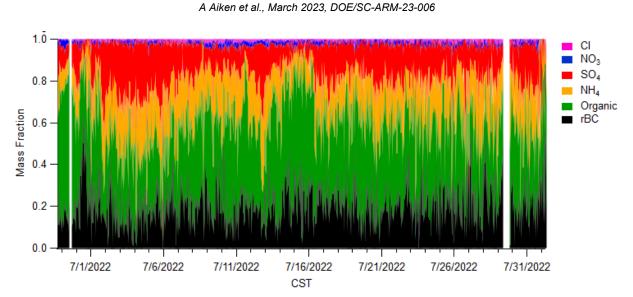


Figure 5. Aerosol speciated non-refractory organic and inorganic mass fractions as measured by the soot particle aerosol mass spectrometer (SP-AMS) and the single-particle soot photometer (SP2).

3.0 Publications and References

3.1 Presentations

Dubey, MK, K Gorkowski, KB Benedict, AC Aiken, C Carrico, T Capek, C Mazzoleni, R Farley, Q Zhang, and C Cappa. 2022. "Predicting Mixing and Humidity Effects on Absorbing Aerosol Optics: Laboratory, Wildfire and TRACER-CAT Results." ARM/ASR Principal Investigator Meeting. Rockville, Maryland.

Dal Porto, R, CD Cappa, RN Farley, JE Lee, K Gorkowski, K Benedict, Q Zhang, AC Aiken, and MK Dubey. 2022. "Impact of humidification on ambient aerosol optical properties; Field observations from TRACER-CAT in Houston, Texas." American Geophysical Union 2022 Fall Meeting. Chicago, Illinois.

Dubey, MK, CD Cappa, K Gorkowski, RN Farley, K Benedict, ASM Shawon, S Jordan, Q Zhang, JE Lee, R Dal Porto, AC Aiken, and A Meyer. 2022. "Humidity Effects on Mixed Carbon and Dust Aerosols in Houston Measured to Evaluate Radiative Forcing and Deep Convection Modules in Climate Models." American Geophysical Union 2022 Fall Meeting. Chicago, Illinois.

Gorkowski, K, R Dal Porto, RN Farley, S Jordan, A Meyer, JE Lee, MK Dubey, CD Cappa, Q Zhang, ASM Shawon, AC Aiken, and K Benedict. 2022. "Laboratory and Ambient Comparison of Hygroscopicity and Optical Properties of Aerosols." American Geophysical Union 2022 Fall Meeting. Chicago, Illinois.

Lee, JE, K Gorkowski, K Benedict, A Meyer, AC Aiken, Q Zhang, RN Farley, R Dal Porto, CD Cappa, and MK Dubey. 2022. "Mixed carbon aerosol properties in Houston, Texas (TRACER-CAT): Role of Chemistry in Absorption, Scattering, and Humidification." American Geophysical Union 2022 Fall Meeting. Chicago, Illinois.

3.2 Planned Submissions

Gorkowski et al. 2023. "Humidity-Dependent Absorption Enhancements of Long-Range Transported Dust."

Lee et al. 2023. "Midnight Plumes: Hyperlocal Nighttime Emissions in Houston."

Farley et al. 2023. "Composition of internally mixed black carbon in Houston during TRACER-CAT."

Dal Porto et al. 2023. "Optical properties of mixed absorbing aerosols in Houston during TRACER-CAT."

3.3 Collaborations

Co-deployed TRACER-CAT measurements with UC Davis (PI Cappa and Co-PI Zhang).

Coordinated science with other TRACER teams at Baylor University, University of Houston, and Brookhaven National Laboratory.

3.4 References

Carrico, CM, TJ Capek, KJ Gorkowski, JT Lam, S Gulick, J Karacaoglu, JE Lee, C Dungan, AC Aiken, TB Onasch, A Freedman, C Mazzoleni, and MK Dubey. 2021. "Humidified single-scattering albedometer (H-CAPS-PM_{SSA}): Design, data analysis, and validation." *Aerosol Science and Technology* 55(7): 749–768, <u>https://doi.org/10.1080/02786826.2021.1895430</u>

Uin, J, AC Aiken, MK Dubey, C Kuang, M Pekour, C Salwen, AJ Sedlacek, G Senum, S Smith, J Wang, TB Watson and SR Springston. 2019. "Atmospheric Radiation Measurement (ARM) Aerosol Observing Systems (AOS) for Surface-Based In Situ Atmospheric Aerosol and Trace Gas Measurements." *Journal of Atmospheric and Oceanic Technology* 36(12): 2429–2447, <u>https://doi.org/10.1175/JTECH-D-19-0077.1</u>

4.0 Lessons Learned

- 1. **Humidity:** In humid environments (like Houston) it is best to over-engineer the drying of the aerosol samples. We achieved an aerosol sample humidity of 50% relative humidity for most of the campaign with our current setup. The instruments would have performed better (needing less maintenance and providing more stable measurements) if the inlet RH was lower (30% RH).
- 2. **Battery Backup:** The power distribution inside the LANL-Guest-AOS was operated using one large, centralized battery backup system. It would have been better to use individual battery backups for each instrument rack because the points of failure would have been distributed. As was the case for our central battery backup, when its backup battery failed we did not have an easy way to fix it. This was exacerbated by the supply chain constraints of acquiring a large replacement battery during the IOP.





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