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Particle Flux Measurements during TRACER (TRACER PFM) Field Campaign Report

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Acronyms and Abbreviations

AMF	ARM Mobile Facility
AOS	Aerosol Observing System
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
BNF	Bankhead National Forest
CPC	condensation particle counter
DOE	U.S. Department of Energy
ECOR	eddy correlation flux measurement system
HTDMA	humidified tandem differential mobility analyzer
NCSU	North Carolina State University
OPC	optical particle counter
POPS	portable optical particle spectrometer
rBC	refractory black carbon
RDMA	radial differential mobility analyzer
SGP	Southern Great Plains
SMPS	scanning mobility particle sizer
SONIC	sonic anemometer
SP2	single-particle soot photometer
TRACER	Tracking Aerosol Convection Interactions Experiment
UHSAS	ultra-high-sensitivity aerosol spectrometer

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1.0 Summary

The purpose of this campaign was to deploy an instrument payload collocated with a suite of in situ and remote-sensing aerosol instruments at the urban site at the La Porte, Texas Municipal Airport during the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Tracking Aerosol Convection Interactions Experiment (TRACER) in Houston, Texas. The payload was designed to characterize the physicochemical properties, mixing state, and size-resolved vertical fluxes of aerosols.

Deployed aerosol instruments associated with the flux measurements included three condensation particle counters (CPCs) with different lower cutoff diameters ($D > 2.5$ nm, $D > 10$ nm, and $D > 40$ nm), a single-particle soot photometer (SP2) measuring refractory black carbon, and a portable optical particle spectrometer (POPS) measuring the optical size distribution (~180 nm < D < ~5000 nm). A 10-m flux tower hosted a sonic anemometer (SONIC) for vertical velocity measurements. In addition, a humidified tandem differential mobility analyzer (HTDMA) was deployed to measure the growth factors and hygroscopicity parameter of 15-50 nm-sized particles in order to constrain the composition of compounds responsible for modal aerosol growth during new particle formation/growth events. Finally, a radial differential mobility analyzer (RDMA) was deployed to extend the size distribution measurement to 5 nm. Figure 1 shows the trailer and flux tower deployed during TRACER. The instruments were deployed between May25, 2022 and September 28, 2022.

Figure 1. The trailer and flux tower co-located with the aerosol mobile facility during the TRACER campaign. The flux tower is the tower with the black box mounted near the top at the right of the picture. Aerosol instruments were housed in the trailer situated to the left of the tower.

Data were collected from June 1, 2022 to September 26, 2022. Figure 2 provides an overview of the data coverage. One CPC (CPC3) failed early during the campaign and had to be sent for service to the manufacturer. The SP2 had a failed scattering detector, which required the return of the instrument to the principal investigator's laboratory for repair. Intermittent periods of incorrect data for CPC1 are due to the accumulation of water in the butanol reservoir. Other intermittent periods are due to power outages from storms and periods of regular instrument maintenance that require taking the instruments offline.

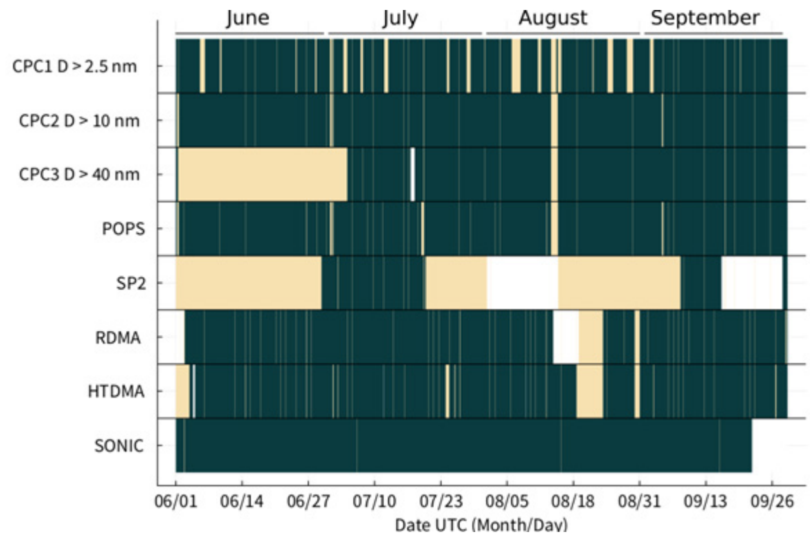

Figure 2. The colors show the performance of the instruments during the campaign: Dark green indicates normal operation, yellow indicates incorrect data, and white indicates questionable data.

Table 1 provides a brief summary of data sets submitted to the ARM Data Center. The submitted files contain processed, calibrated, and quality-controlled data ready for use in science analysis.

Table 1. TRACER PFM data sets submitted to ARM Data Center.

Data Set	ARM Record ID	Content	Size
Tower Data	ARM 0773	Daily data including sonic anemometer wind vector and temperature, CPC1, CPC2, CPC3, POPS (16 bins), SP2 scattering, SP2 black carbon concentrations binned at 10 Hz frequency.	36 GB
SP2 Data	ARM 0777	Processed SP2 data containing 16 bin scattering and 16 bin rBC size distributions binned at 1 Hz.	2.6 GB
RDMA Data	ARM 0775	Inverted particle size distribution data from the RDMA.	28MB
HTDMA Data	ARM 0776	Inverted HTDMA data given hygroscopicity frequency distributions (see Figure 5 below).	961 MB

2.0 Results

Due to the extensive nature of the deployment, a complete conclusive analysis of the data set is not available yet. A few highlights of the ongoing preliminary analysis are provided here.

Sonic Anemometer/Heat fluxes: We intercompared the sensible heat flux derived from the 10-Hz sonic anemometer data from the deployed flux tower with those from the co-located DOE ARM eddy correlation flux measurement system (ECOR). An example of intercomparison is shown in Figure 3. The two time series show excellent agreement, suggesting that the deployed sonic anemometer acquired good data.

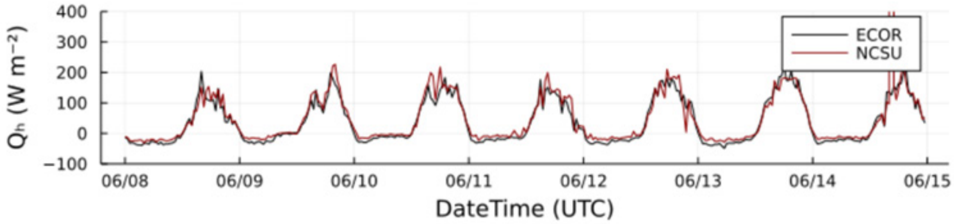

Figure 3. Comparison of ECOR-reported and NCSU tower-derived sensible heat fluxes.

Size Distributions: Figure 4 shows a comparison of the size distribution from the ARM Aerosol Observing System (AOS) instruments and the North Carolina State University (NCSU)-deployed SP2, POPS, and RDMA instruments. The mean spectral densities derived from the optical particle counters ultra-high-sensitivity aerosol spectrometer (UHSAS), optical particle counter (OPC), NCSU POPS, and NCSU SP2 are in excellent agreement. Note that channels 75-100 of the UHSAS are not shown due to an artifact in the data associated with that size range (gain stage). The spectral densities from the AOS UHSAS and AOS scanning mobility particle sizer (SMPS) and the AOS SMPS and NCSU RDMA disagree in the overlap region, with the AOS SMPS showing slightly larger values. However, the shapes of the spectra are similar and the temporal correlation of the integrated values in the overlap ranges and the correlations with the total particle counters are high.

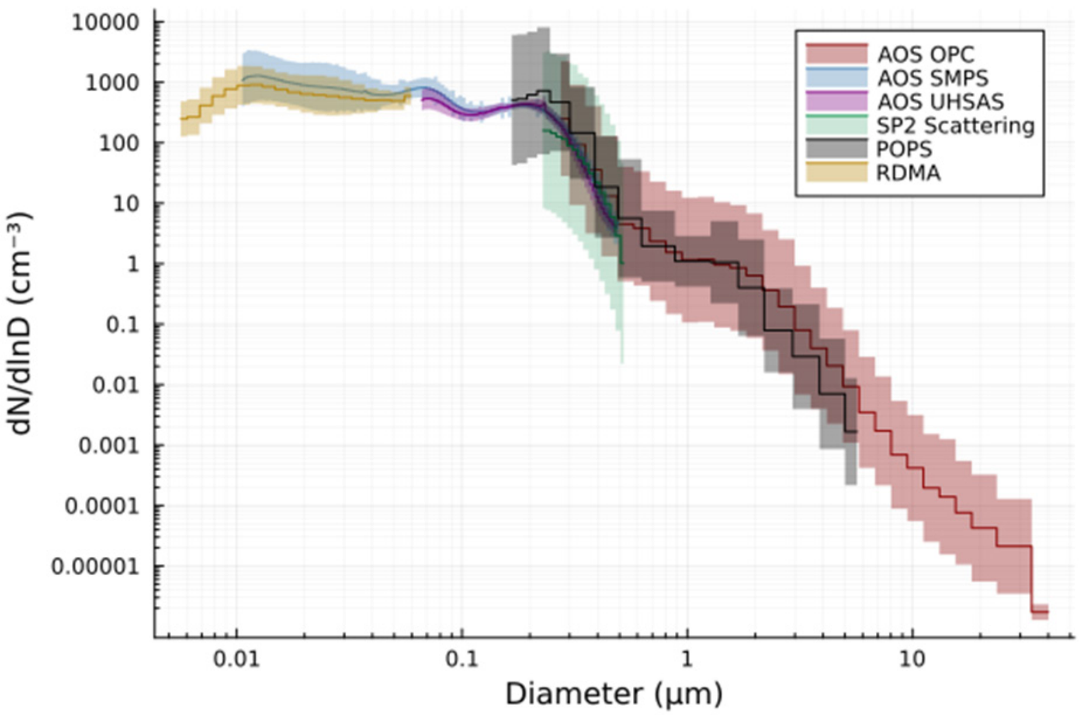

Figure 4. Comparison of size distributions for the AOS OPC, AOS SMPS, AOS UHSAS, NCSU POPS, NCSU SP2 Scattering, and NCSU RDMA. The solid lines correspond to the mean spectral density average for a single day. The shaded area corresponds to the mean multiplied by the geometric standard deviation and the mean divided by the geometric standard deviation.

Black carbon data: Refractory black carbon concentrations were measured by the SP2 and varied between 5 and 100 cm^{-3}. The mass mode of the black carbon distribution was typically at mass < 1 fg. The fraction of thinly and thickly coated black carbon particles varied throughout the day. The fractions of thinly and thickly coated particles showed a diurnal cycle and typically increased during the day, likely due to photochemical aging.

Hygroscopic growth factors: Hygroscopic growth factors were measured for dry particles between 15 and 50 nm. These data were inverted using the algorithms developed by Petters (2021). Figure 5 shows an example of this inversion. The HTDMA data generally showed bimodal distributions with less and more hygroscopic modes. The less hygroscopic mode generally had growth factors near 1 corresponding to a hygroscopicity parameter $\kappa = 0.09 \pm 0.11$. The hygroscopicity of the more hygroscopic mode was typically $\kappa = 0.29 \pm 0.18$. The relative fractions of the more and less hygroscopic modes were highly dynamic and varied between being dominated by non-hygroscopic and dominated by the hygroscopic population.

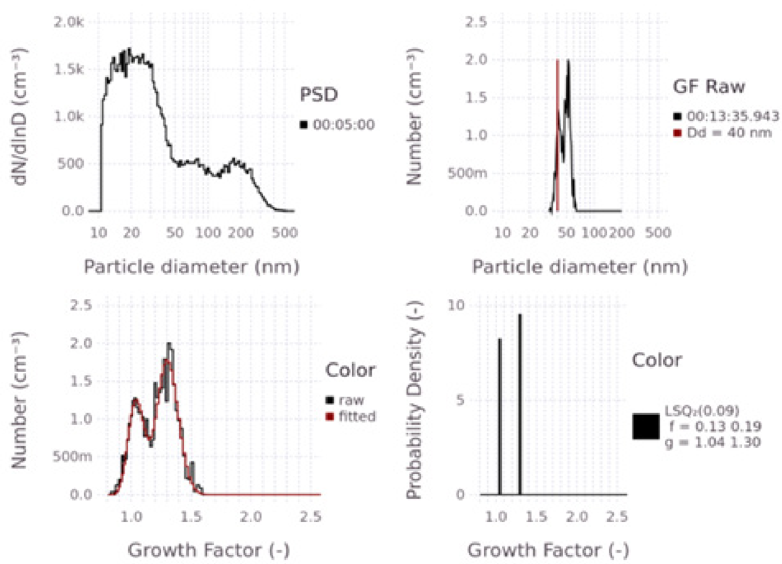

Figure 5. Example inversion of HTDMA data. Top left: measured aerosol size distribution. Top right: measured raw growth factor scan for 40 nm-size particles. Bottom left: raw growth factor data. Bottom right: inverted probability density distribution retrieving the growth factor and relative amounts of the less and more hygroscopic modes. See also Petters (2021) for additional details.

One of the main motivations to deploy the “nano” HTDMA was to study the hygroscopicity of the material condensing onto particles during modal growth events. Figure 6 shows an example event from the campaign. The modal growth rate was very rapid (19 nm/hr). The hygroscopicity parameter increased from < 0.2 before the event to ~0.6 during the event, suggesting that hygroscopic inorganic materials, likely sulfates, are responsible for condensational aerosol growth during this specific event.

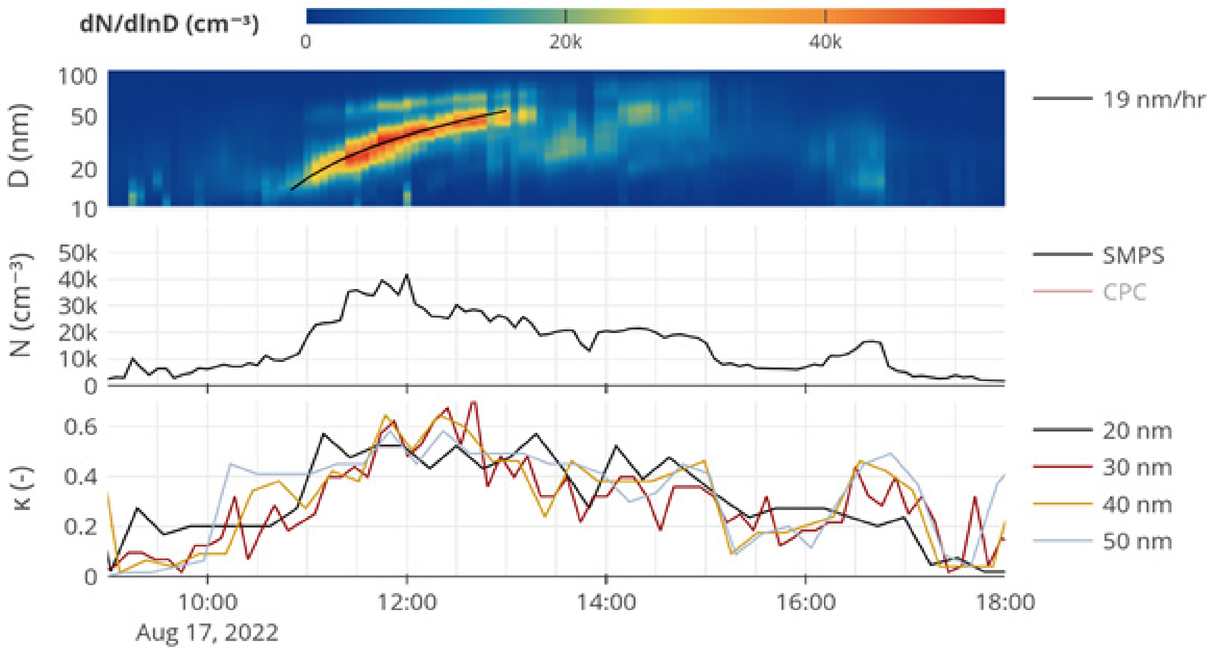

Figure 6. Top: size distribution from the AOS SMPS during a modal growth event. Middle: integrated number concentration from the SMPS. Bottom: evolution of the hygroscopicity (derived from most frequent growth factor after inversion) for 20-50 nm-sized particles.

New particle formation and flux measurements: New particle formation could not be directly detected by our equipment or equipment deployed by ARM or NCSU. We therefore adopt the language "small-particle event" that indicates the appearance of small particles followed by modal growth. In general, small-particle events were relatively uncommon during the TRACER intensive operational period at the main site. A total of 10 small-particle/growth events were identified through manual analysis. Figure 7 shows an example of a small-particle/growth event on August 17, 2022 (same as in Figure 6). This particular example can be classified as a "Class B" event. During Class B events, the first detected particle-mode diameter does not occur at the smallest diameter measured by the instrument. New particle formation aloft followed by downward mixing – which has been the driving hypothesis for this work – has been invoked previously to explain the Class B pattern in surface observations. Note the gradual increase in 3-10 nm concentration, which is consistent with the downward vertical mixing hypothesis. However, the preliminary fluxes shown in Figure 7 do not appear to show this. An important obscuring factor for flux measurements at the site is the high temporal variability of the number concentration due to nearby sources (non-stationarity of the timeseries). How to properly derive fluxes in this non-stationary environment is currently under investigation.

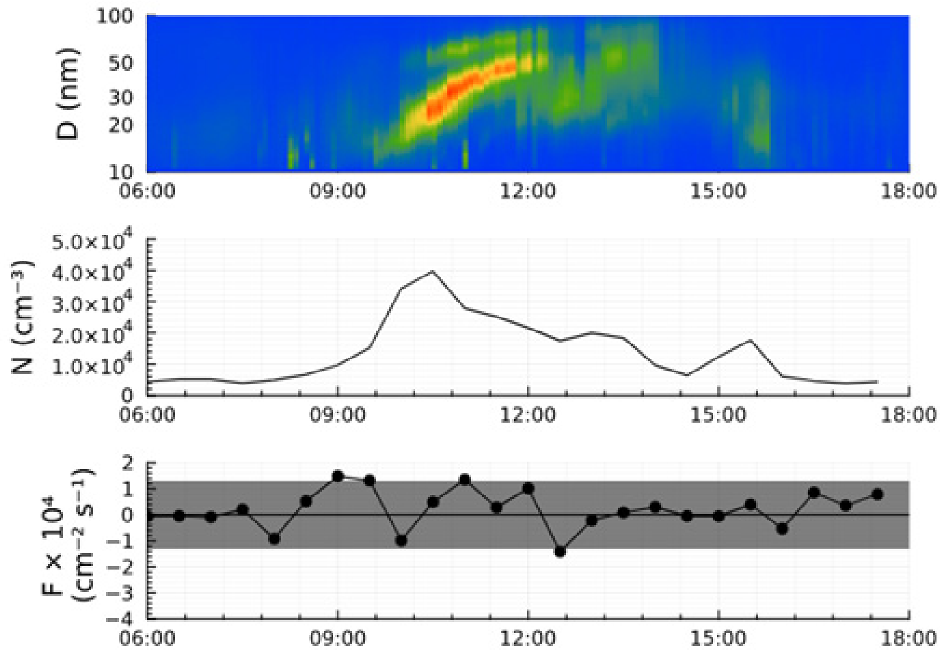

Figure 7. (a) Time series of aerosol size distribution against local time on 8/17 during TRACER. Color is dN/dlnD. (b) Concentration of 3-10-nm particles measured by the difference in CPCs from our flux tower, (c) Flux derived using the method given in Islam et al. (2022). Grey shading is the limit of detection.

3.0 Publications and References

3.1 Publications

Islam, MM, N Meskhidze, A Rasheeda Satheesh, and MD Petters. 2022. "Turbulent flux measurements of the near-surface and residual-layer small particle events." *Journal of Geophysical Research − Atmospheres* 127(17): e2021JD036289, https://doi.org/10.1029/2021JD036289

Kasparoglu, S, MM Islam, N Meskhidze, and MD Petters. 2022. "Characterization of a modified printed optical particle spectrometer for high-frequency and high-precision laboratory and field measurements." *Atmospheric Measurement Techniques* 15(17): 5007–5018, https://doi.org/10.5194/amt-15-5007-2022

Petters, MD. 2021. "Revisiting matrix-based inversion of scanning mobility particle sizer (SMPS) and humidified tandem differential mobility analyzer (HTDMA) data." *Atmospheric Measurement Techniques* 14(12): 7909–7928, https://doi.org/10.5194/amt-14-7909-2021

Petters, MD, T Pujiastuti, A Rasheeda Satheesh, S Kasparoglu, B Sutherland, and NM Meskhidze. "Wind-Driven Emissions of Coarse-Mode Particles in an Urban Environment." To be submitted to *Atmospheric Chemistry and Physics*, 2023.

3.2 Presentations

Petters, MD, S Kasparoglu, A Rasheeda Satheesh, T Pujiastuti, B Sutherland, and N Meskhidze. 2023. “Aerosol Mixing State and Eddy-Covariance Particle Flux Measurements during the TRACER Campaign.” Presented at the TRacking Aerosol Convection interactions ExpeRiment (TRACER) Workshop. Texas Southern University, Houston, Texas.

Kasparoglu, S, A Rasheeda Satheesh, T Pujiastuti, B Sutherland, N Meskhidze, and MD Petters. 2023. “Aerosol Mixing State and Aerosol Flux Measurements during the TRACER campaign: Implications for new particle formation in a polluted urban environment.” Submitted to 41st annual conference of the American Association for Aerosol Research (AAAR). Portland, Oregon.

Petters, MD, T Pujiastuti, A Rasheeda Satheesh, S Kasparoglu, B Sutherland, and NM Meskhidze. 2023. “Wind-driven emissions of course-mode particles in an urban environment.” Abstracts submitted to the Atmospheric System Research Science Team Meeting, 2023, and the AAAR Meeting, 2023.

4.0 Lessons Learned

Experience using the ARM facility: Working with the ARM team at the TRACER M1 site was flawless.

Lessons Learned: A detailed account of lessons learned will be included in the final report to DOE with the associated Atmospheric System Research (ASR) proposal. Briefly, despite various short-term instrument failures (Figure 2), we are proud of the degree of operational readiness for the duration of the campaign. However, maintaining our aerosol trailer and flux tower with a single postdoctoral student over a 4.5-month period proved more challenging than anticipated. A shorter intensive operational period or increased staffing will be needed to conduct future experiments.

The in situ flux measurements proved more difficult than anticipated for several reasons. For technical reasons, including accessibility for instrument maintenance, availability of air conditioning, and availability of space, it was necessary to house the equipment inside the trailer. This resulted in the need to partially dry the aerosol to prevent condensation (which was one of the reasons for the SP2 and CPC instrument failures). Transporting the aerosol from the tower to the trailer reduced the instrument response, which led to a diminution of the measured fluxes. We selected the TRACER main site to be co-located with the Doppler lidar and the full suite of AOS measurements. Indeed, as part of our ASR project, we were able to retrieve excellent flux measurements from the Doppler lidar (not further reported here; see Petters et al. 2023), which were originally envisioned as an important comparison point with ground-level eddy covariance/POPS flux measurements. However, the surface aerosol time series at the site were highly non-stationary for all size ranges. Here, non-stationarity refers to advective components unrelated to turbulent vertical transport. These also increase the limit of detection and thus make it difficult to discern fluxes against statistical noise. Furthermore, even though the POPS performed excellently (Figure 4), and even though we have shown that the instrument response time of the used POPS is sufficiently fast for flux measurements (Kasparoglu et al. 2022), low counting statistics in the POPS was found to reduce the response significantly. Again, this makes it more difficult to discern fluxes against statistical noise.

Thus, the lessons learned can be summarized as follows:

- A site with less heterogeneous aerosol conditions (as was found during our preliminary campaign at the ARM Southern Great Plains [SGP] site) would be more conducive to flux measurements.
- A (much) taller flux tower would be desirable to (a) increase the flux footprint, (b) make it less critical to have instruments with fast response time due to more coherent turbulence, and (c) smooth out some of the aerosol heterogeneity experienced at the surface.
- Carefully reconsider the deployed aerosol counters to optimize response time considering the raw time response, counting statistics, the need for climate control, and the need for aerosol drying.
- Locate the aerosol measurements closely to the sonic anemometer to prevent transport through the tubing. This was logistically impossible with our setup due to space and power constraints, and/or the need to climate control and frequently service the detector. In general, turbulent flow is preferred to avoid tube damping. However, turbulent flow also strongly increases sample losses of ultrafine particles during transit, thus necessitating laminar flow. It is thus best to eliminate the need for tubing as much as possible. A walkup tower with ample space for instrumentation near the sonic will be needed for this.

These lessons have also been shared with ARM personnel during a teleconference for the third ARM Mobile Facility (AMF3) Bankhead National Forest (BNF) Tower CPC (Aerosol Flux) Design Review.

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