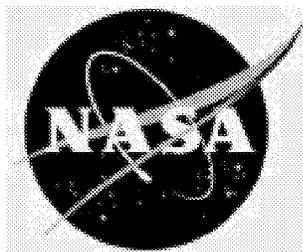


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NASA's Atmospheric Effects of Aviation Project

Results of the August 1999 Aerosol Measurement Intercomparison Workshop, Laboratory Phase

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June 2001

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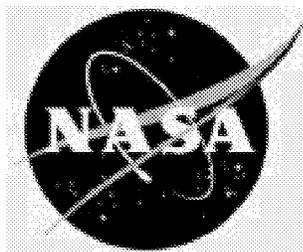
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Nomenclature

AEAP	Atmospheric Effects of Aviation Project
CNC	condensation nuclei counter
DMA	differential mobility analyzer
EAC	electrostatic aerosol classifiers
EI	emission index
GRC	John H. Glenn Research Center
HEPA	High-Efficiency Particulate Arresting
LaRC	Langley Research Center
LPC	laser particle counter
MASS	Mobile Aerosol Sampling System
NCAR	National Center for Atmospheric Research
N-MASS	Nucleation-Mass Aerosol Size Spectrometer
nASA	nanometer aerosol size analyzer
nDMA	nano-differential mobility analyzer
OSU	Oregon State University
PAGEMS	Particulate and Gaseous Emissions Measurement System
PSL	Particle Simulation Laboratory
SASS	Subsonic Assessment Program
SMPS	scanning mobility particle sizer
SUCCESS	Subsonic Assessment: Cloud and Control Effects Special Study
UD	University of Denver
UMN	University of Minnesota
UMR	University of Missouri-Rolla

1. Introduction

Because commercial aviation is anticipated to increase in both volume and extent substantially during the next two decades (Boeing 1996; Baughcum, Sutkus, and Henderson 1998; Mortlock and Van Alstyne 1998) and aircraft represent a unique source of anthropogenic emissions (inasmuch as they are routinely emitted above the boundary layer), the need to better understand their environmental impact is clear. NASA's Atmospheric Effects of Aviation Project (AEAP) addresses this need. A major component of the AEAP—the Subsonic Assessment Program (SASS)—was formed with the goal of characterizing the effects that the current fleet of commercial aircraft have on atmospheric chemical and radiative processes, and what effect they may have in the coming years, as air traffic increases (Friedl 1997). Aircraft are prolific sources of both soot and sulfate particles to the upper troposphere and lower stratosphere (Fahey, Keim, and Chan 1995; Anderson, Cofer, and Brunke 1998; Anderson, Cofer, and Hudgins 1998; Miake-Lye et al. 1998; Karcher and Fahey 1997). These particles may have an impact upon climate through direct absorption/reflectance of solar radiation (Pueschel et al. 1992); by altering cirrus cloud formation, reflectance, or duration; or by providing additional surface area upon which heterogeneous chemical processes, such as ozone destruction, can occur. Because of their potential to perturb these important atmospheric processes, the AEAP has placed a high priority upon gaining an increased understanding of particle formation and growth processes within aircraft plumes and upon characterizing the particulate emissions of turbine engines (Friedl 1997).

To accurately assess the atmospheric effects of aircraft-generated particulates as well as to develop and test predictive models for aircraft emissions, the number density, size, and composition of aerosols within engine exhaust and aging plumes must be understood and well characterized. Thus, the AEAP has funded several individual groups to make observations within a number of different measurement venues including behind aircraft in-flight, parked in run-up areas, as well as downstream of turbine engines mounted in test cells. Because the soot and sulfate particles within fresh plumes typically range from a few to a few tens of nanometers in diameter, measurement approaches are very limited. Butanol-based condensation nuclei counters (CNCs) have been used in a majority of aircraft emission studies (e.g., Hagen et al. 1998; Anderson, Cofer, and Brunke 1998; Anderson, Cofer, and Hudgins 1998; Fahey, Keim, and Chan 1995; Twohy, Gandrud, and Weinheimer 1998). Size distributions are obtained either using an electrostatic differential mobility analyzer coupled to a CNC (Hagen et al. 1998) or by employing a battery of CNCs with different lower size cut-offs (Anderson, Cofer, and Brunke 1998; Anderson, Cofer, and Hudgins 1998; Cofer, Anderson, and Bagwell 1998; Brock et al. 2000). For composition information, samples are typically delivered to a pair of identical CNCs, one with an unheated inlet to monitor total particulate emissions and the other with a heated inlet to evaporate sulfate/water particles, to determine the nonvolatile (or soot) particle fraction (e.g., Cofer, Anderson, and Bagwell 1998). Observations are reported in terms of emission indices (EIs) or the amount of pollutant generated per kilogram of fuel burned. This is most often determined by taking the ratio of the pollutant concentration to that of a conserved combustion tracer, such as CO₂. For aerosols, EIs are reported as the number of particles generated per kilogram of fuel burned. If size distribution measurements are also recorded, an inferred mass EI (grams per kilogram of fuel burned) can be estimated by multiplying the calculated aerosol volume times an assumed aerosol mass density. Gravimetric analysis of aircraft particulate emissions has not been successful because of the amount of exhaust gas volume required to obtain a measurable accumulation (weight) of particles on filtering mediums.

Aircraft aerosol EIs have been recorded by AEAP-sponsored investigators. For example, the University of Missouri-Rolla has reported measurements from behind aircraft operating in run-up areas (Whitefield and Hagen 1995), in-flight (Hagen, Trueblood, and Whitefield 1992), and in test cells (Whitefield and Trueblood 1993; Wey et al. 1998). The University of Denver has obtained airborne observations in aged aircraft plumes (Fahey, Keim, and Chan 1995) as well as very fresh exhaust

emissions (Brock et al. 2000). The Langley Group has made airborne, near-field measurements behind a variety of aircraft (Anderson, Cofer, and Brunke 1998; Anderson, Cofer, and Hudgins 1998; Anderson, Cofer, and McDougal 1999) as well as recent determinations of EIs in decaying plumes over the North Atlantic (Anderson et al., 1999).

Unfortunately, a wide variation in aerosol emissions indices and chemical and physical properties has been reported in the exhausts of subsonic commercial aircraft. Because a significant fraction of these aerosol measurements has been measured by various independent research teams involved in the SASS studies, it became necessary to determine if differences in aerosol measurement system components and/or techniques used by these groups were responsible for the wide variation in particle concentrations and properties observed in commercial aircraft exhaust. Because the data have, for the most part, been collected with similar aerosol counters, it appeared doubtful that the discrepancies would be explained by instrumental differences alone. Other sources of differences between the observations may include sampling issues, plume age, engine type and condition, environmental conditions, and fuel sulfur content.

Thus, to evaluate instrumental error as a source of the observed discrepancies an Aerosol Measurement Intercomparison Workshop was sponsored by NASA's AEAP/SASS Program. This workshop took place August 1–14, 1999, at the NASA Langley Research Center in Hampton, Virginia. The first week of the 2-week workshop was devoted to challenging the core detection elements of the aerosol measuring instruments with laboratory-generated aerosols. The second week was focused on testing the efficiency and sensitivity of the measurement systems by sampling the exhaust from an LaRC T-38 Talon aircraft. Invitations to the workshop were issued to all groups that participated as aerosol investigators in SASS-sponsored airborne and ground-based field experiments. This report is intended to summarize the results from the laboratory phase of the workshop.

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2.2.2. Instrumentation

The aerosol measurement system (the DMA) consists of an electrostatic classifier that separates aerosols in a sample stream into size ranges and is connected to a TSI Model 3025 Ultrafine Condensation Particle Counter that counts the particles. A computer controls the classifier voltage sweep, and particle counts are directed into inversion software that results in the generation of 25 size bins. Flows are controlled by mass flow controllers. The system determines an aerosol size distribution from 5 to 250 nm.

The LaRC airborne instrument suite was designed for use aboard an aircraft to provide continuous measurements of total and nonvolatile aerosol number densities with crude particle sizing. The system is composed of two ultrafine condensation nuclei counters (TSI Model 3025 CNCs) and two standard condensation nuclei counters (TSI Model 3760 CNCs). Both types of CNCs have ≈ 1 Hz response times and are butanol based. Some evidence of sensitivity of butanol-based CNCs to particle composition has been reported (Ball, Hanson, and McMurry 1999). Extensive laboratory characterizations and calibrations indicate the ultrafine and standard CNCs have 50 percent size cutoffs at ≈ 5 and 13 to 15 nm, respectively, when operated in the flight configuration. To prevent saturating these instruments in the highly concentrated aircraft exhaust plumes, sample air is withdrawn from a sampling manifold through a critical flow orifice and immediately diluted by a factor of 10 to 50 with a concentric flow of filtered cabin air. This process provides the secondary benefit of allowing the CNCs to be operated at constant sample pressure and volumetric flow. The instruments are arrayed so that one each of the ultrafine and standard CNCs share a common inlet which can deliver sample either at cabin temperature ($\approx 20^\circ\text{C}$) or heated to 150° to 300°C by passage through a 15-cm heat-tape-wrapped tube. This arrangement allows quantification of total aerosols >5 and >15 nm along with the number of nonvolatile, presumably soot, particles and, by difference, volatile aerosols in the same size categories.

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2.3.2. Instrumentation

The NCAR system uses TSI Model 3760 butanol-based CNC counters to measure particle number concentration. In SUCCESS, the counters were used primarily to count evaporated cloud nuclei;

therefore, no dilution system was employed. Corrections for coincidence errors are applied, but become substantial above about $20\,000\text{ cm}^{-3}$. In SUCCESS, a manifold heated to 250°C was used ahead of one counter to measure particle volatility.

2.4. University of Denver Group

2.4.1. Participant

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2.4.2. Instrumentation

The Nucleation-Mode Aerosol Size Spectrometer (N-MASS) is composed of five CNCs operating in parallel. Each CNC senses the cumulative concentration of particles with diameters larger than a particular size. The 50 percent cut points are 5, 8, 15, 30, and 55 nm. The instrument operates with a constant internal pressure (60 mb for this intercomparison) and uses perfluorotributylamine as the working fluid. The data from the 5 CNCs are inverted by using a nonlinear iterative technique to give a size distribution from 5 to 55 nm. The N-MASS has operated at pressures from sea level to 60 mb, only size distribution information. A concentration range of up to $2 \times 10^5/\text{cm}^3$ can be measured; the operating pressures are sea level to 60 mb. The flow requirement is 1.2 volumetric L/min, controlled by a critical orifice.

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2.5.2. Instrumentation

The GRC Particulate and Gaseous Emissions Measurement System (PAGEMS) extractively samples combustion gases from aircraft gas turbine combustors and engines. Differential mobility analyzers and condensation nuclei counters are used in tandem to characterize the emissions in terms of particulate total concentration, size distribution, and volatility. A suite of gas analysis equipment measures CO₂, NO_x, and SO₂ levels in the combustion gases. Data can be collected over a wide range of operating conditions, pressures up to 250 psig, particulates 10 to 500 nm in size, 10⁴ to 10¹³ particles/cm³, and gas species down to parts per million levels.

2.6. University of Missouri-Rolla Group

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2.6.2. Instrumentation

The Mobile Aerosol Sampling System (MASS) uses extractive sampling from an aerosol source and delivers the sample to various aerosol characterizing devices: particle (CN) counters, an LPC, and one or more Electrostatic Aerosol Classifiers (EAC). The LPC is used for counting and sizing large particles by using optical techniques; the EAC is used to size particles at the small end of the size spectrum. A thermal discriminator can be used to remove volatile material from the particles before characterization. Our TSI Model 3025 Ultrafine CNC counts particles down to 3 nm in diameter. Holding tanks are used to capture airborne samples for quasi-real-time size distributions. Size distribution data can be acquired for particles in the diameter size range of 7 nm to 5 μm. Concentrations of total particles (CN), nonvolatile particles, and particles in specific size bands can be continuously measured. The system operation and data acquisition are computer supported.

Previously, the system has been deployed for airborne sampling missions on the NCAR Sabreliner, the DLR Dassault Falcon 20E, and the NASA DC-8 and for various ground-based field sampling projects, including the Glenn PSL and flametube facilities, USAF Phillips Laboratory, McDonnell Douglas, Air France, Pratt & Whitney, and Arnold Engineering and Development Center.

2.7. University of Minnesota Group

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2.7.2. Instrumentation

The UMN research instrumentation consisted of a nanometer aerosol size analyzer (nASA) that was capable of scanning 30 size channels between 3 and 100 nm in a total time of 3 sec. The analyzer includes a bipolar charger, an extended-length nanometer differential mobility analyzer, and an electrometer. This combination of components provides particle size spectra at a scan rate of 0.1 sec/channel. Particle concentrations were measured using a TSI Model 3025 Ultrafine Condensation Particle Counter. An additional TSI Scanning Mobility Particle Sizer (SMPS) was used during the laboratory phase of the polydispersed aerosol intercomparisons.

3. Calibration Aerosols

The laboratory phase of the intercomparison relied heavily on the University of Minnesota group, headed by David Pui and Da-ren Chen, supported by Po-Shin Lee and Hee-Siew Han. Their aerosol generation systems were used during the intercomparison workshop as calibrating sources for both monodispersed and polydispersed aerosols. Aerosol streams were generated by either a tube furnace (metallic silver particles) or an evaporation/condensation aerosol generator (sulfuric acid). The silver and sulfuric acid particles were surrogates for nonvolatile and volatile particles, respectively. When monodispersed aerosol was necessary to a test objective, a University of Minnesota nano-differential mobility analyzer (nDMA) was used to deliver monodispersed particles from the initially polydispersed aerosols (Chen, Pui, and Sem 1998). Output (particles/cm³) from the nDMA was constantly monitored by this group with a TSI Model 3025A Ultrafine CNC. The University of Minnesota characterization of these particles, therefore, served as the reference standard used to evaluate the other measurements. It should be noted that while a common sampling manifold with identical (length, diameter, and composition) tubing was used to feed samples to each system, the only attempt to determine particle losses incurring during transmission to the individual instruments was a diffusion loss calculation.

4. Data Presentation

Data presented in this report were selected to serve as summary representations of the intercomparison experiments from which general discussion and conclusions from these intercomparisons could be made. For these intercomparison experiments, we consider differences of 10–20 percent in individual measurements to be reasonably good agreement. Differences of 10–20 percent would certainly not explain some of the large differences observed in the field measurements conducted in the past. The complete data set for the laboratory phase is obtainable via CD-ROM.

5. Laboratory Phase

For CNCs, nucleation and subsequent growth to detectable particle sizes become much less efficient at very small particle diameters. The diameter at which 50 percent of the particles nucleate and grow to a measurable size (50 percent cutoff diameter) can vary significantly among different types of CNCs. Accordingly, one of the first goals of the workshop was for each group to determine the minimum particle diameter at which their instruments would count 50 percent of the supplied particles. These evaluations were done using both silver (nonvolatile) and sulfuric acid (volatile) particles. In addition, because many of the CNC systems are used as airborne systems, normally operated at reduced pressures, 50 percent cutoff evaluations were not only performed at ambient pressures but also at reduced pressure (≈ 200 torr). By means of these tests any differences in total particle counts due to different 50 percent cutoff diameters could be identified.

Another goal of the laboratory phase of the workshop was to determine the upper end of the dynamic concentration range. Particle counting problems often occur at high particle concentrations ($>10^4$ particles/cm³); this can be caused by particle coincidence in the detection beam, depletion of condensate, etc. To determine the effective dynamic concentration range, a 60-L vessel was filled with very concentrated Ag or sulfuric acid aerosol (initially $>10^6$ particles/cm³). The aerosol sample was then withdrawn and fed through a common manifold to the various instruments. As the sample was fed to the manifold, an equal volume of HEPA-filtered air was added to the vessel. This produced an exponential decay in particle concentration. When concentrations fell sufficiently, the instruments started counting properly, and in this manner the dynamic range of the various instruments could be evaluated.

Another objective of the laboratory phase of the workshop was to determine size-dependent efficiencies of the coupled CNC/DMA, and multichannel CNC systems as a function of particle type. The source aerosols were furnished in the manner described for the 50 percent cutoff diameter evaluations though all aerosols were polydispersed and at ambient pressure. These systems were evaluated for how well they sized the aerosol (geometric mean diameters) and the resulting total (integrated) particle counts.

6. Laboratory Test Results

The 50 percent cut points were assessed during the first several rounds of laboratory phase comparisons. Round 1 consisted of 10 runs of monodispersed Ag aerosol at ambient pressure, with diameters ranging from 3–15 nm, beginning and ending at 1322 and 1412 EDT, respectively, on August 3. The results for runs 1–10 are shown in figure 1 in terms of number concentrations measured by the individual instruments versus number concentrations supplied by UMN. The calibration aerosol diameters and concentrations were characterized before introduction into a manifold designed to split the flows for

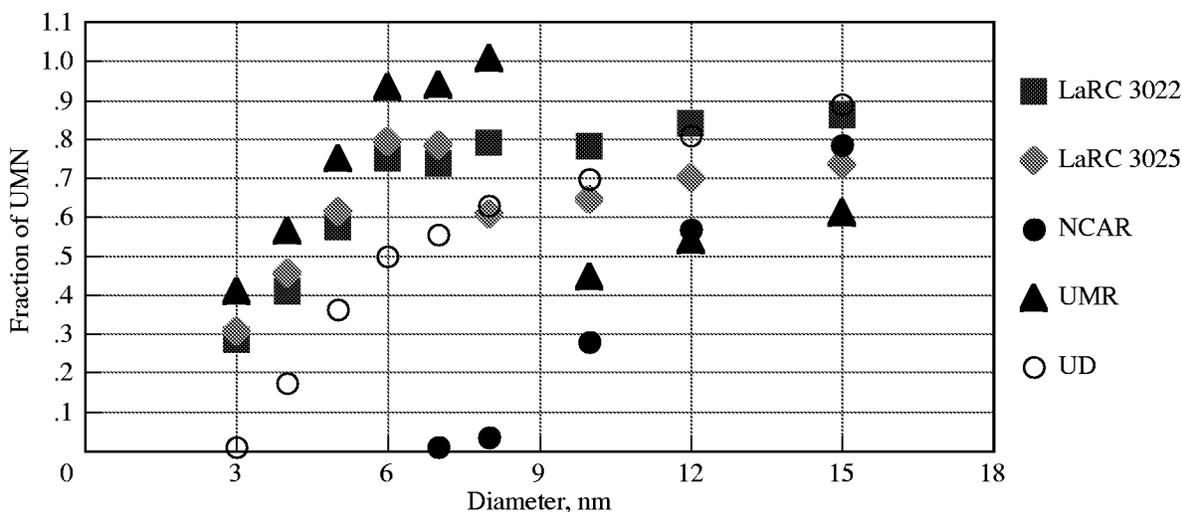


Figure 1. Round 1, Ag aerosol.

delivery to the individual instruments. No diffusion corrections have been applied to these data. All counters, except the OSU/NCAR 3760, responded to diameters approaching as small as 3 nm. The reason for the somewhat erratic drop in counting efficiency appearing at about 7–10 nm, and observed by many of the instruments, is not known.

In this report, the 50 percent cut points are defined as when the instruments are counting 50 percent of the particles UMN reports as being delivered to the distribution manifold. We realize that this neglects any losses in the delivery system. Using this method, the UMR counts in figure 1 reveal a 50 percent cut point between 3 and 4 nm. The LaRC 3025 and 3022 CNCs had cut points at about 4–5 nm, and the UD N-MASS at about 6 nm. The OSU/NCAR 3760 CNC can be seen to have a 50 percent cut point at about 12 nm. These cut points are typical of what would be anticipated.

Round 2 consisted of runs 11–30, also of monodispersed Ag particles at ambient pressure. In this case, the aerosol size ranged from 3–80 nm in diameter. These runs were conducted between 1416 and 1638 on August 3. Again, figure 2 is presented in terms of number concentrations counted versus number concentrations supplied by UMN. Reasonably good agreement (10–20 percent) is shown in figure 2 among the various participants, and greater than 80 percent of the particles were being measured by all instruments at diameters >35 nm. The 50 percent cut points can be determined easier by viewing the bottom plot in figure 2. The UMR cut point was about 4 nm, LaRC about 5 nm, UD slightly less than 8 nm, and the OSU/NCAR 3760 cut point at about 13 nm.

Round 3 consisted of runs 31–54 using monodispersed H_2SO_4 aerosol at ambient pressure over the range of 4–95 nm. Runs 31–44 were conducted between 1806 and 1859 on August 3, while runs 45–54 were conducted between 0941 and 1017 on August 4. Results from runs 31–44 are presented in figure 3(a). Figure 3(a) shows that relative agreement with the UMN-supplied concentrations at diameters ≥ 40 nm was achieved by all participants except UD and GRC. We can offer no explanation for the large differences at smaller diameters but assume that the disagreement existed due to problems in aerosol generation. Because these were the first H_2SO_4 runs conducted through the tubing and manifold, it is certainly possible that the smaller sizes of aerosol were fully or partially neutralized by reactions with adsorbed or desorbed ammonia. Results from runs 45–54, conducted on the following day, are presented in figure 3(b). These results are much more consistent with prior results. It can be deduced from this figure that the 50 percent cut points determined previously with Ag aerosol do not change in

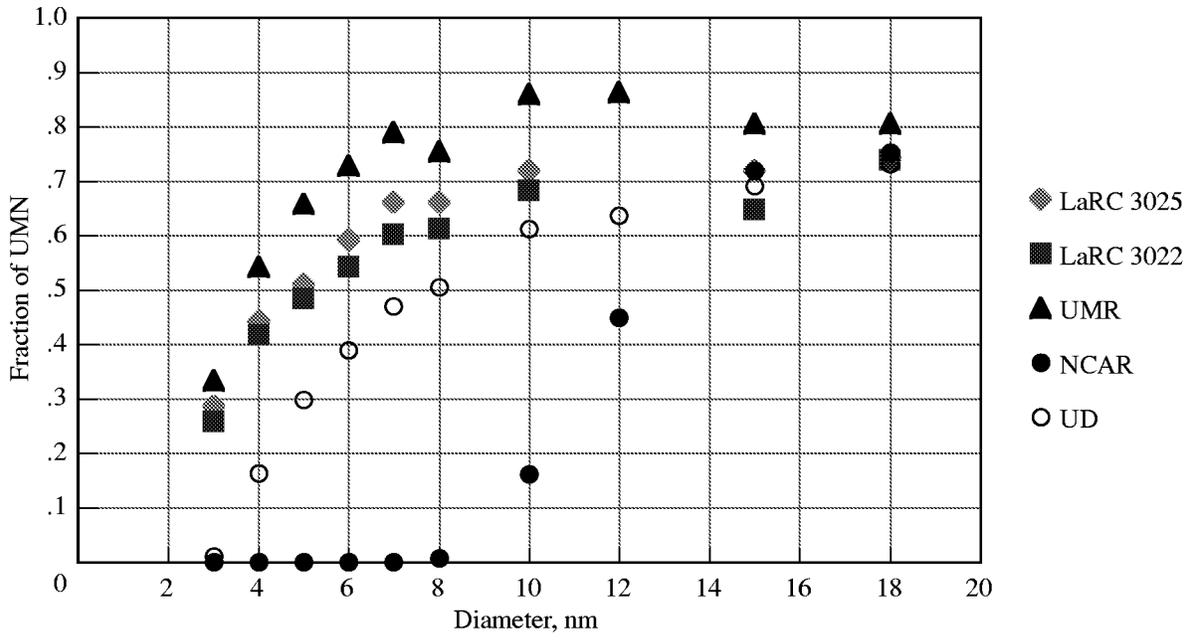
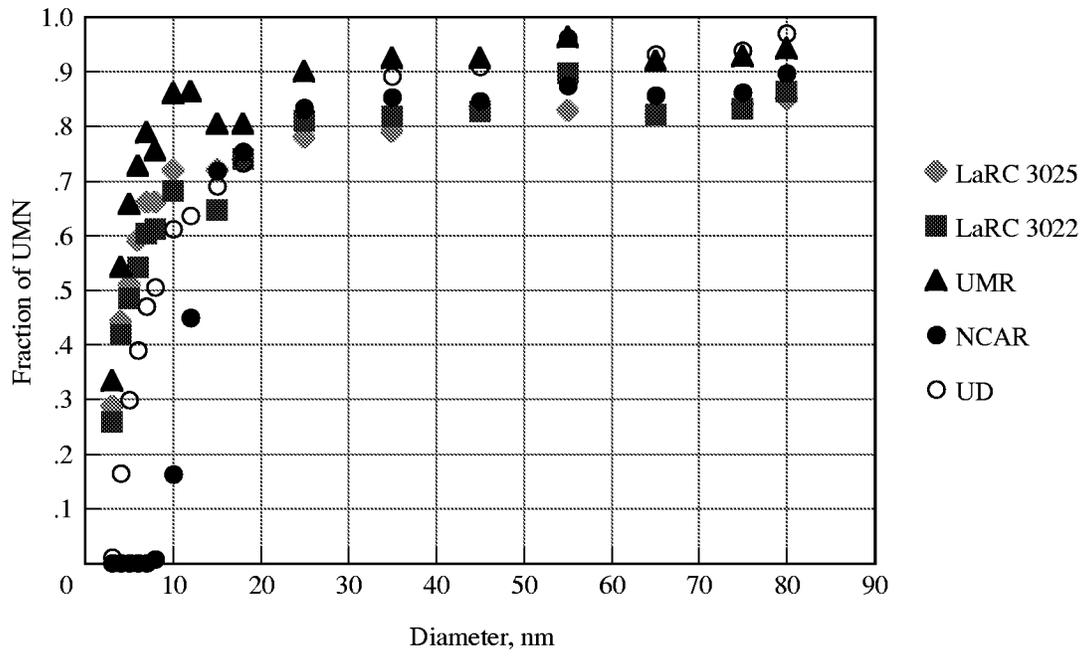
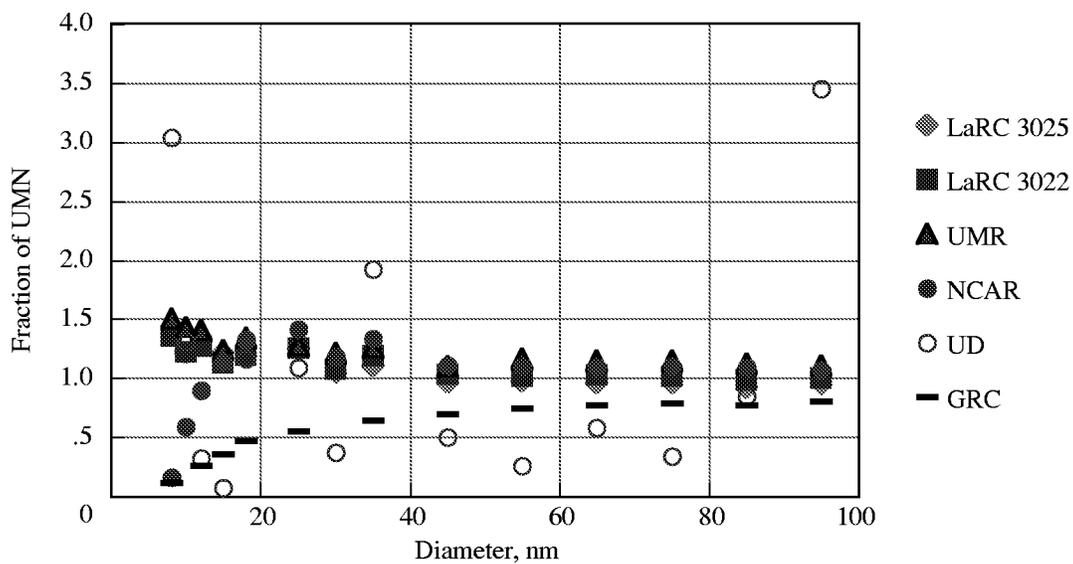
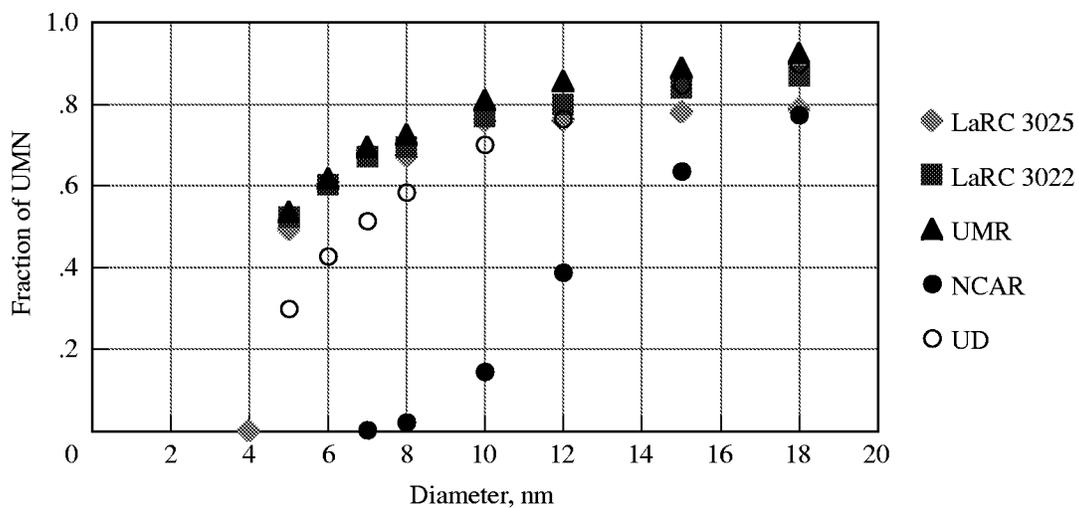


Figure 2. Round 2, Ag aerosol.



(a) Runs 31–44.



(b) Runs 45–54.

Figure 3. Round 3, H_2SO_4 aerosol.

any significant manner with volatile sulfuric acid aerosol. Essentially, the 50 percent cut points for all ambient pressure assessments were as follows: LaRC 3025, ≈ 5 nm; LaRC 3022, ≈ 5 nm; UMR, ≈ 5 nm; UD, ≈ 7 nm; OSU/NCAR, ≈ 12 – 13 nm.

Round 4 consisted of runs 55–71 with monodispersed H_2SO_4 furnished to the distribution manifold at reduced pressure (≈ 200 torr). These runs were conducted on August 4 between 1353 and 1458 EDT. These results are shown in figure 4. No diameters were generated that were less than 12 nm in this series. Thus, except for the OSU/NCAR and LaRC 3760 CNCs, cut points could not be determined for the other instrumentation.

It is clear in the bottom plot that the 3760 CNCs had suffered a slight loss in cut point efficiency, yielding cut point diameters of about 15 nm. These data indicate changes in counting efficiencies at reduced pressure. Measured number concentrations for particles >20 nm in diameter seem to indicate a loss in sensitivity for all instruments at reduced pressure relative to the UMN-supplied aerosols. Results from round 6, shown in figure 5, also suggest a loss of sensitivity at 200 torr. At 200 torr the grouping of 3760 CNCs versus 3025 CNC becomes much more apparent (indicating a greater differential loss in counting efficiency for the 3760 CNCs at 200 torr) than at ambient. This same behavior had been observed in our laboratory at LaRC with our 3760 and 3025 CNCs (Cofer, Anderson, and Bagwell 1998).

Round 5 consisted of runs 72–81, conducted between 0920 and 1212 on August 5. These runs were conducted with polydispersed Ag aerosol, initially furnished at very high concentrations from a 60-L vessel. In figure 6, Tank run 79 is presented and is representative of the typical results from all these high-concentration runs. In figure 6, the LaRC 3022 CNC can be seen to perform very well at high concentrations. The 3022 model CNC is known for having a large dynamic range (relative to the 3025 and 3760 series CNCs). With the exception of the UD N-MASS instrument, all CNCs began to develop counting problems between 10^4 and 10^5 particles/cm³. All CNCs can be seen to count well at concentrations between 10^4 and 10^2 particles/cm³. The N-MASS counted almost as well at high concentrations as the 3022. As stated earlier in this report, these results, in general, were not unexpected.

Round 6 consisted of runs 82–99 conducted on August 5 between 1335 and 1442. The aerosol was monodispersed Ag over the range of 5–85 nm at a pressure of 200 torr. Careful examination of top plot in figure 5, and comparison with top plot of figure 4, suggests a less efficient particle counting efficiency (unlike at ambient pressure) with the Ag aerosol than with the H_2SO_4 aerosol for all the butanol-based CNCs. The N-MASS (halocarbon condensate), however, does not indicate any significant particle counting efficiency change between H_2SO_4 and Ag aerosol. Another interesting aspect of figure 5 is that the 3760 CNCs sporadically reach 50 percent of the UMN-supplied aerosol, and thus, do not have a 50 percent cut point as defined in this report. The 3025 CNCs appear (bottom plot in fig. 5) to have 50 percent cut points at around 10 nm in diameter (rather than at 4–5 nm diameters) at 200 torr.

A calculated diffusion-loss coefficient (based on tube length, diameter, pressure, and flow rate) for loss during laminar pipe flow, was applied to the data presented in bottom plot in figure 5 and is shown in figure 7. Interestingly, the 50 percent cut points for all instruments compare much more favorably with those determined during the ambient aerosol sampling. Diffusional loss would be expected to increase at reduced pressure and with decreasing particle diameter. In fact, a 50 percent cut point can now be determined for the 3760 CNCs after diffusion correction. However, it should be equally apparent that the reduced counting efficiency at the larger particle diameters at 200 torr has changed little. The slight but progressive tendency for the counting efficiency to drop as particle diameters increase (>15 nm) suggest that diffusional losses (a much larger correction factor at small diameters) may have

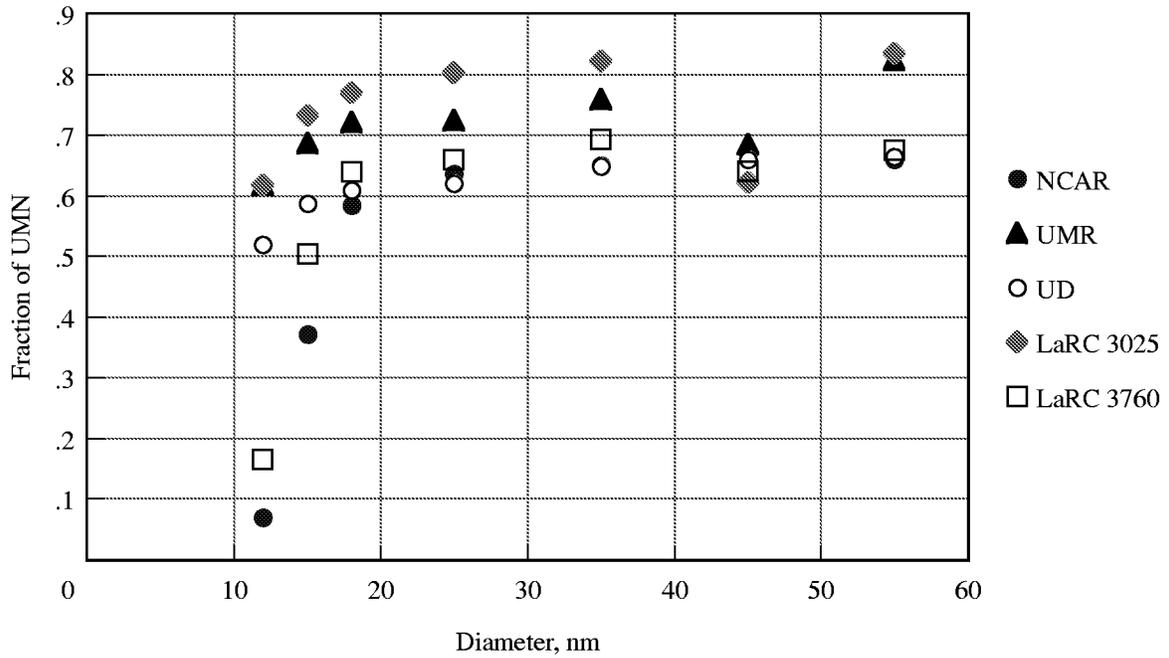
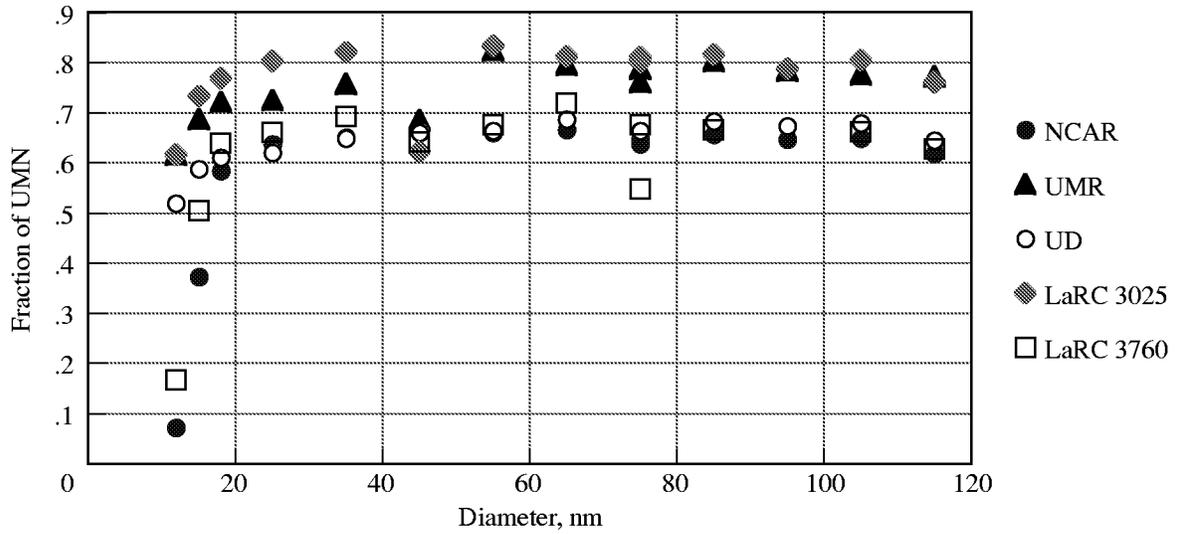


Figure 4. Round 4, H_2SO_4 aerosol at 200 torr.

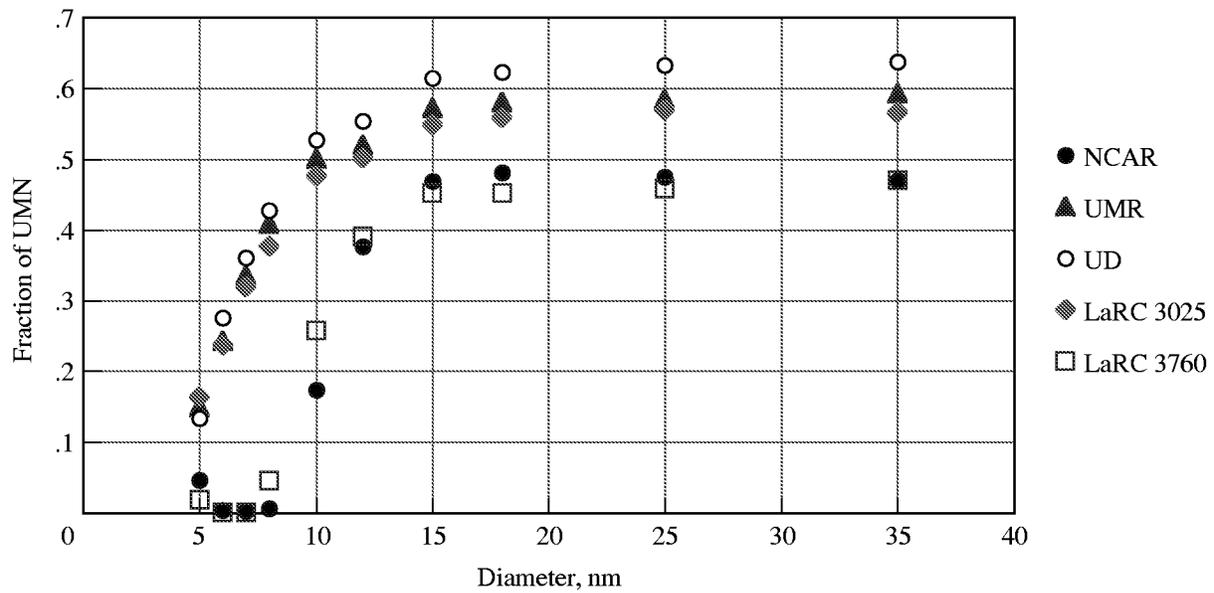
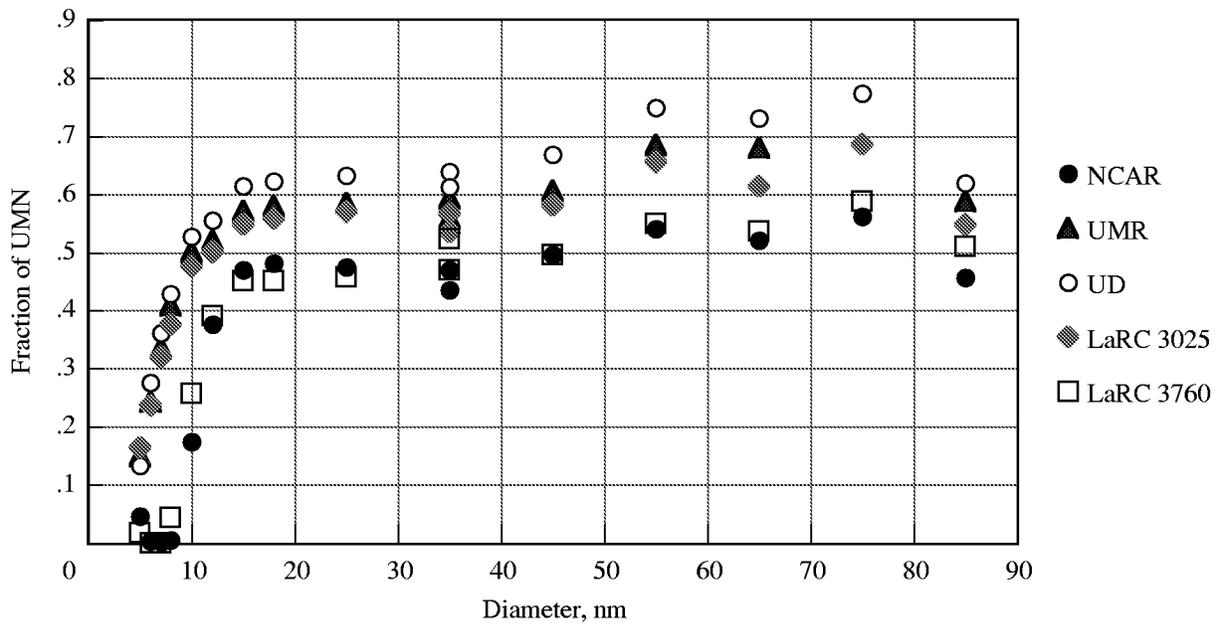


Figure 5. Round 6, Ag aerosol at 200 torr.

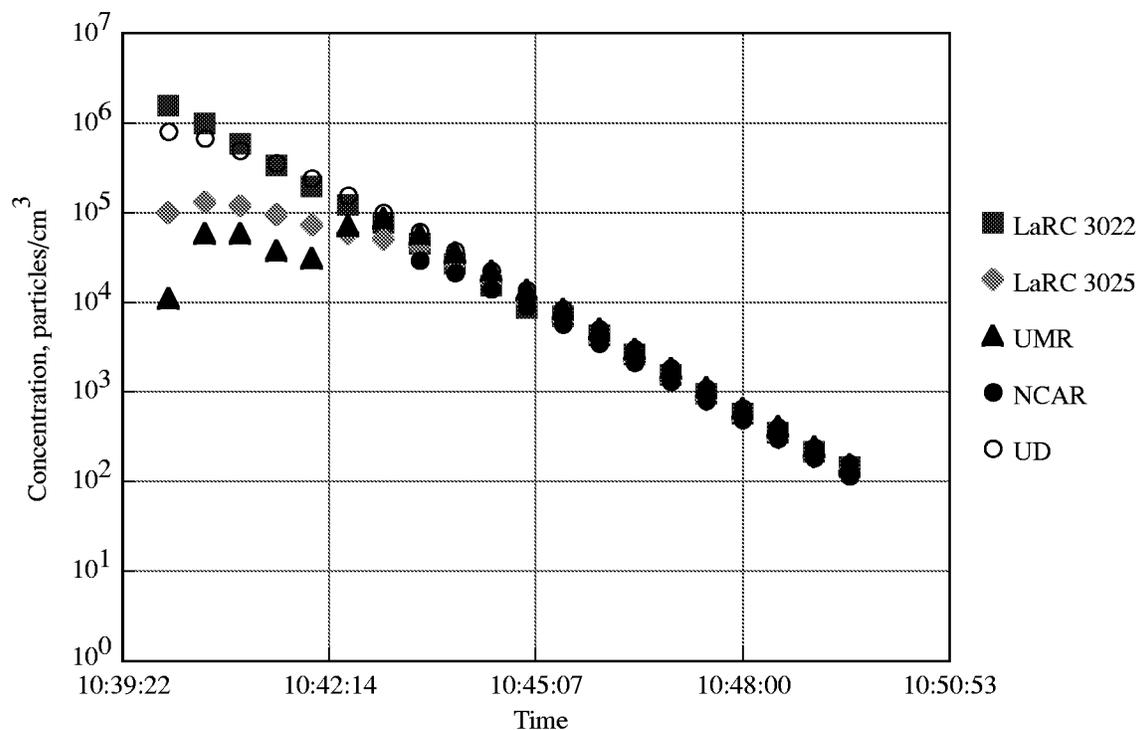


Figure 6. High concentration for tank run 79.

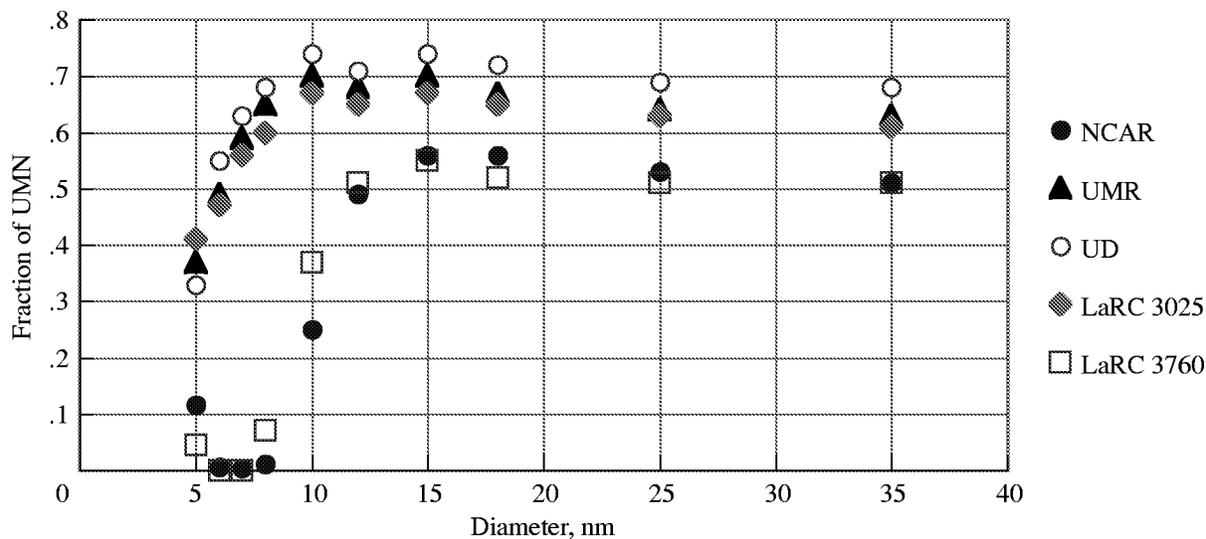


Figure 7. Diffusion corrected, Ag aerosol at 200 torr.

been slightly overestimated. Note that diffusion corrections were not applied to the reference concentrations reported by the UMR group, which might also cause the apparent bias.

Round 7 consisted of runs 100–102 conducted between 1527 and 1638 on August 5. The results for these runs were highly inconsistent with reported counts both exceeding and being well below UMN-supplied aerosol concentrations. These runs were considered flawed and are not graphically shown in

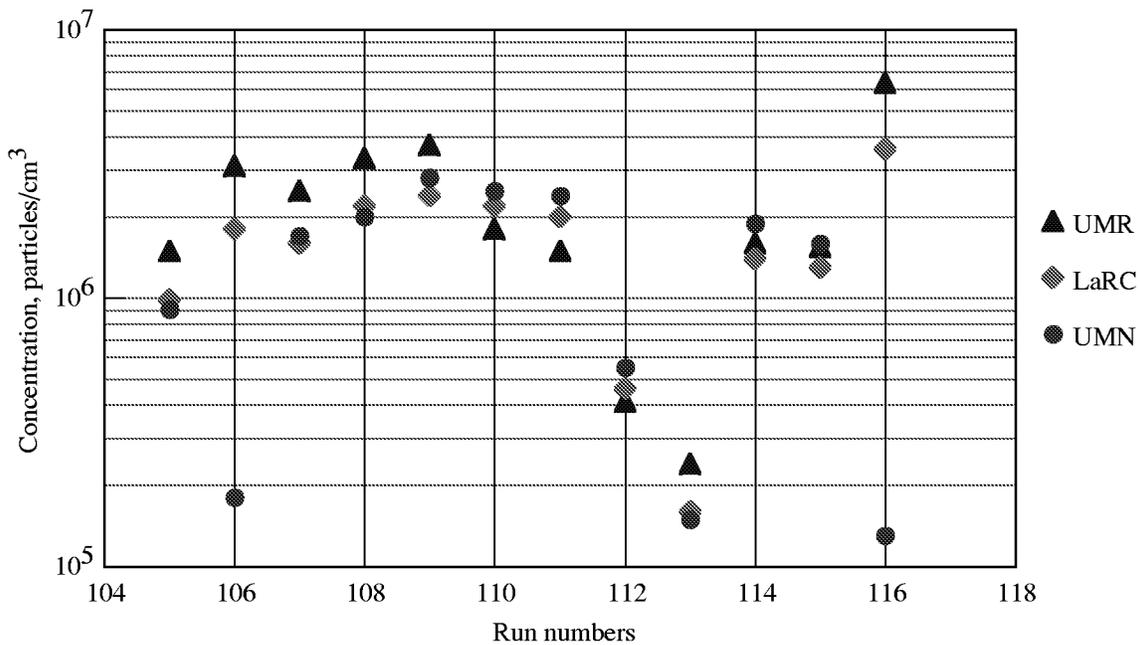
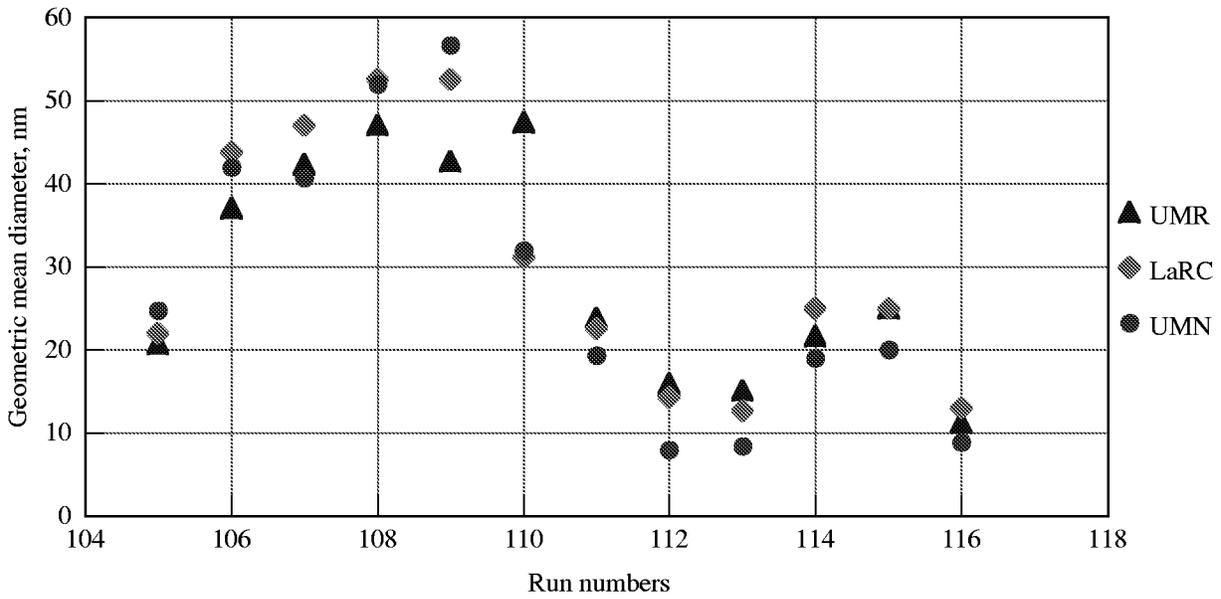


Figure 8. Polydispersed DMA analyses.

this report. Similarly, round 8 consisted of a single run conducted on August 5, run 103, a polydispersed H₂SO₄ aerosol at 760 torr; this was a size distribution run, for which no group other than the UMN team reported any results. This run is also not presented graphically.

Round 9 consisted of runs 104–116 and was conducted on August 6, between 1000 and 1428 EDT. Runs 104–109 were with H₂SO₄ aerosol, and 110–116 used Ag aerosol; all were polydispersed. Only UMN, UMR, and LaRC participated in this phase. In top plot in figure 8, results of the size distribution measurements are summarized in terms of run number versus geometric mean diameter. The degree of scatter in these data, and what might have contributed to it, is addressed subsequently in the section

“Discussion”. However, the degree of scatter appears comparable among the Ag and H₂SO₄ aerosols, indicating no systematic change due to aerosol composition. In bottom plot in figure 8, results from these runs are displayed as a function of total particle concentrations per cubic centimeter of volume, determined by integrating counts over all sampled bins. Most of these runs indicated good counting agreement among systems. The large differences observed between both the UMR and LaRC analyses with concentrations reported/supplied by UMN in runs 106 and 116 are unexplained.

Runs 118–127 (completing the laboratory phase) were sampling probe evaluations and will be discussed in a later publication, which will address the two probes (a UMR water-cooled probe and a new LaRC high-flow probe), and discuss results obtained from sampling aircraft exhaust from the port engine (J85-GE-5H) of a LaRC T-38 Talon aircraft at various power levels.

7. Discussion

The first set of experiments conducted during the laboratory phase of the intercomparison workshop were designed to determine the 50 percent cut points for the various particle counters. In general, the instruments behaved as expected. The TSI Model 3025 Condensation Particle Counters all had 50 percent cut points at about 5 nm or less at ambient pressures. The UMR 3025 achieved the best cut point at about 4 nm. The TSI Model 3760 Condensation Particle Counters operated by different groups gave similar results at ambient pressures, yielding 50 percent cut points at diameters around 11–13 nm. The UD N-MASS consistently produced cut points at diameters of about 6–8 nm, but this instrument was always operated at 60 mb of pressure internally, regardless of the calibration source pressure. All counters were found to count within 80–95 percent of the UMN-supplied concentrations for particles 30 nm or greater in diameter at ambient pressures. These cut point determinations (at atmospheric pressure) revealed slightly higher diameters than those reported in Wiedensohler et al. 1997 for the same types of condensation particle counters. Our definition of the 50 percent cut points, however, was based totally on the UMN-reported source inputs and reflected no transmission losses during sample delivery to the individual instruments.

The 50 percent cut point determinations at reduced pressure (≈ 200 torr) revealed a loss in sensitivity; that is, the butanol-based CNCs reached 50 percent counting efficiencies at larger particle diameters. Not only did the 50 percent cut point efficiencies drop, but steady state counting efficiencies (reached at the larger particle diameters where initial particle size is not significantly impacting nucleation/growth processes) were also observed to drop from the typical 80–95 percent observed at ambient pressure to as low as 48–65 percent for Ag aerosol at 200 torr. These results appear in conflict with those reported by Hermann and Wiedensohler (1996) and Wiedensohler et al. (1997) but agree with the theoretical calculations of Zhang and Lui (1991). In addition, this loss of efficiency at reduced pressure was more notable with the 3760 counters. (See figs. 4 and 5.) Although large particle counting efficiencies were observed to be worse for Ag aerosol than for H₂SO₄ aerosol with the TSI butanol-based instruments, this result was not reflected in the fluorocarbon-based N-MASS instrument.

All condensation particle counters (3025 and 3760) were found to saturate at particle concentrations between 10^4 and 10^5 particles/cm³. The N-MASS instrument was found to accurately determine particles in concentrations as high as 3×10^5 particles/cm³. The TSI Model 3022 was found to accurately measure up to 10^6 particles/cm³ at ambient pressure. These results were also largely consistent with our expectations.

The particle size distribution measurements based on differential mobility analyses (DMA) deserve some discussion. The UMN scanning mobility particle sizer (SMPS) utilized voltage sweeps (scans) to produce a progressively changing “monodispersed” aerosol that is fed directly into a 3025 CNC.

The resulting counts are processed with inversion software to produce artificial bins, ranging from 5–100 nm. The LaRC system also utilized voltage sweeps and required inversion software that produced 25 bins, ranging from 5 to 250 nm in diameter. The principal difference between the UMR system and the UMN and LaRC systems was that UMR employed holding tanks to rapidly accumulate aerosol samples that were quickly transferred to their DMA system, producing quasi-real-time size distributions. Small differences in the inversion software used by the different groups, particularly with regard to curve fitting routines, could have produced the minor differences observed in about 80 percent of the DMA intercomparison runs. When runs 104–109 (H₂SO₄ aerosols) are contrasted with runs 110–116 (Ag aerosols), the amount of scatter appears comparable, indicating no strong sensitivity to aerosol composition. However, all these runs were made at approximately ambient pressure, where the butanol-based CNCs had not indicated any sensitivity to composition, as was suggested by the 200 torr runs.

In closing, small counting differences (10–20 percent) were often observed among the CNC and the DMA size distributions. Occasionally large differences were found, but this usually involved differences between the instruments under evaluation with the given UMN source concentrations. These differences, nevertheless, do not appear large or frequent enough to explain some of the major differences (factors of 5–10) obtained by the different groups behind aircraft operating in run-up areas, in actual flight, and in jet engine test cells. The second part of the tests (the sampling probe evaluations and actual data obtained on the tarmac behind the LaRC T-38 aircraft) will further address these issues in a future publication.

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