In Situ Measurements (from Aircraft) of Stratospheric Aerosol Size Distributions

Chuck Wilson

Duncan Axisa, Stephan Borrmann, Chuck Brock, Rushan Gao, Haf Jonsson, Aga Kupc, Mike Reeves, Christina Williamson (collaborators not responsible for errors)

DMT, MPIC, NOAA, NPS, NOAA, NCAR, NOAA (affiliations are approximate)

Photo from MACPEX

Requirements for Useful, Quantitative In Situ Size Distribution Measurements

- Quantitative sampling and transport
- Modifications to particles in sampling and transport are addressed
- Adequate size range covered
- Instruments are characterized so that instrument response is correctly converted to size distributions
- Sample flow rate large enough so that desired spatial features are resolved.
  - TAS around 200 m/s
  - Background concentrations as low as 5 particles/cm^3 (Borrmann averages over 30 s to get background characterization with modified UHSAS.)
- Data are available

Selected Previously Addressed Topics

- Satellite validation
- Heterogeneous chemistry
- Formation of aerosol and clouds in the stratosphere
- Stratospheric cloud and aerosol dynamics
- Impact of volcanic eruptions and pyrocumulus injections
- Emissions from aircraft and rockets
- Stratosphere-Troposphere exchange
- …

Up-Coming Topics

- Next Volcanic Eruption
  - New Particle formation, Aerosol Growth, Sedimentation
- Albedo Modification
  - Aerosol Characterization and Evolution
- ………

Platforms

- NASA U-2, ER-2, B-57, DC-8, Global Hawk
- Geophysica (M-55)
- GV, BAE-146 (at high latitudes)
- Maximum altitudes reached ~ 22 km depending on platform and payload

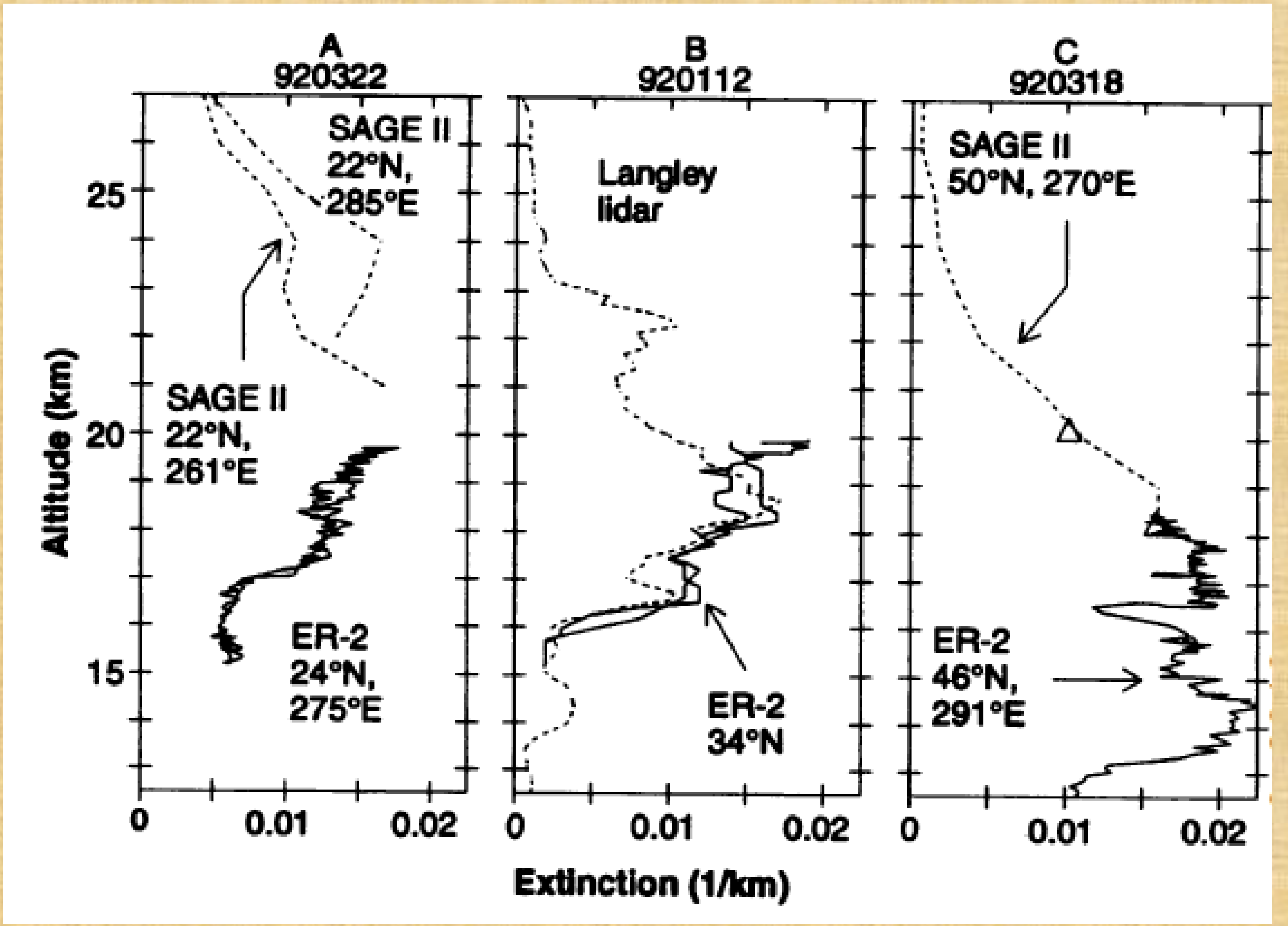
Sampling and Transport

- Submicron particles can be accurately sampled with inlets having solid diffusers and operating under nearly isokinetic conditions
  - Actively controlled inlets may require heavy pumps
  - Instrumented, passive inlets pumped by aircraft motion can provide quantitative sampling when anisokinetic sampling efficiencies are accounted for
- Loses in sampling and transport must be evaluated quantitatively

Modification in Sampling and Transport

- Subsonic aircraft in the UTLS generate ~20 C in compressive heating in sampling
  - The water on a sulfuric acid-water particle evaporates in about 0.25 s.
  - Many sensors operate at warmer temperatures. (OPC around 30 C in sensing volume. Particles accommodate in hundredths of a second.)
- Modified diameters are sensed with CPC and OPC sensors inside aircraft
- Reported data should state the conditions associated with the diameter.
- Less is known about volatility of PSC or organic components.

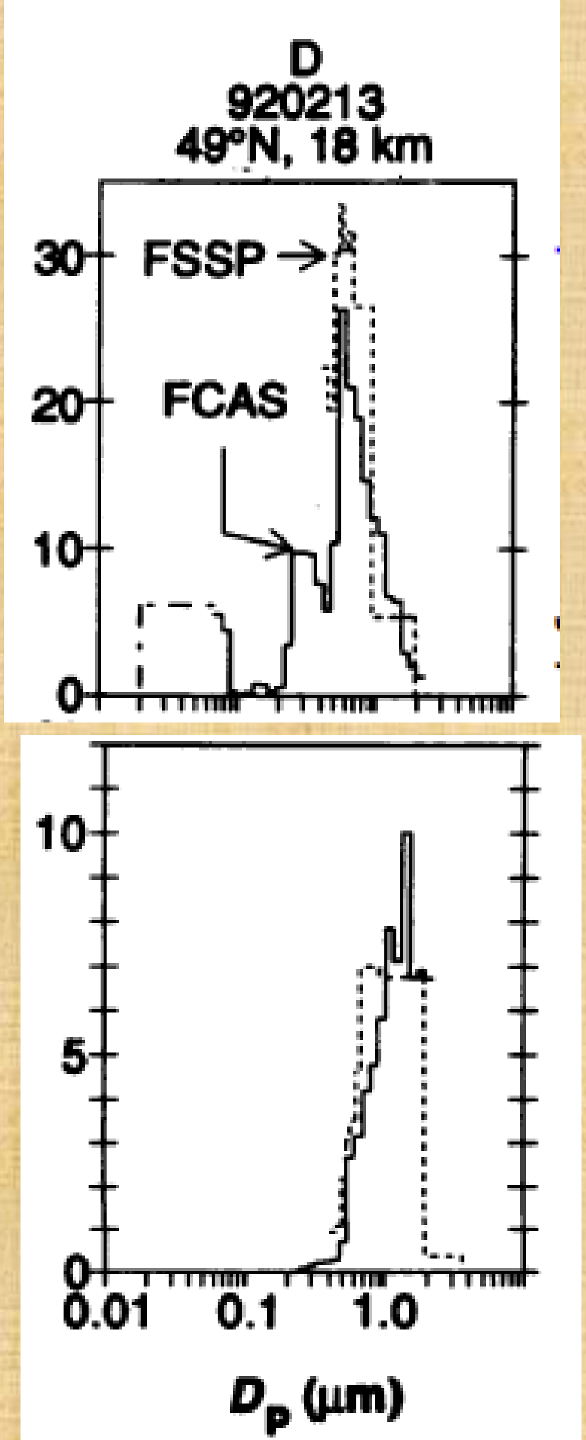
In Situ Observations of Aerosol…..
Wilson et al., Science, 261, 1140, 1993.



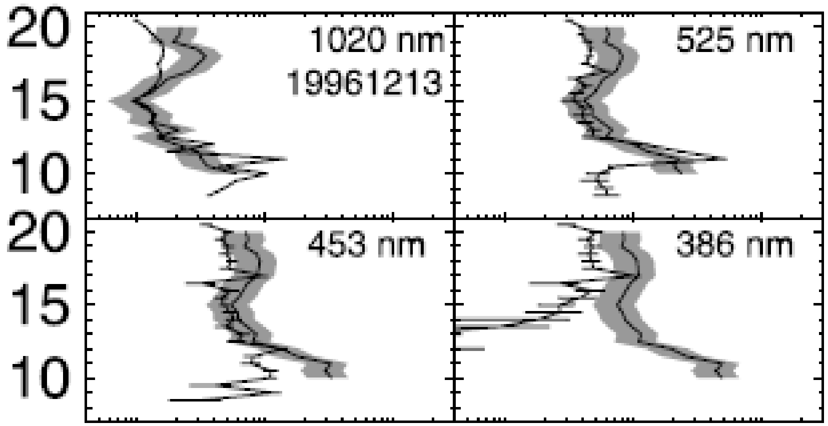
- Pinatubo Aerosol
- Focused Cavity Aerosol Spectrometer (FCAS) and Forward Scattering Spectrometer Probe (FSSP-300)
- Super-micron particles present
- Ambient Diameters assuming H_2SO_4-H_2O

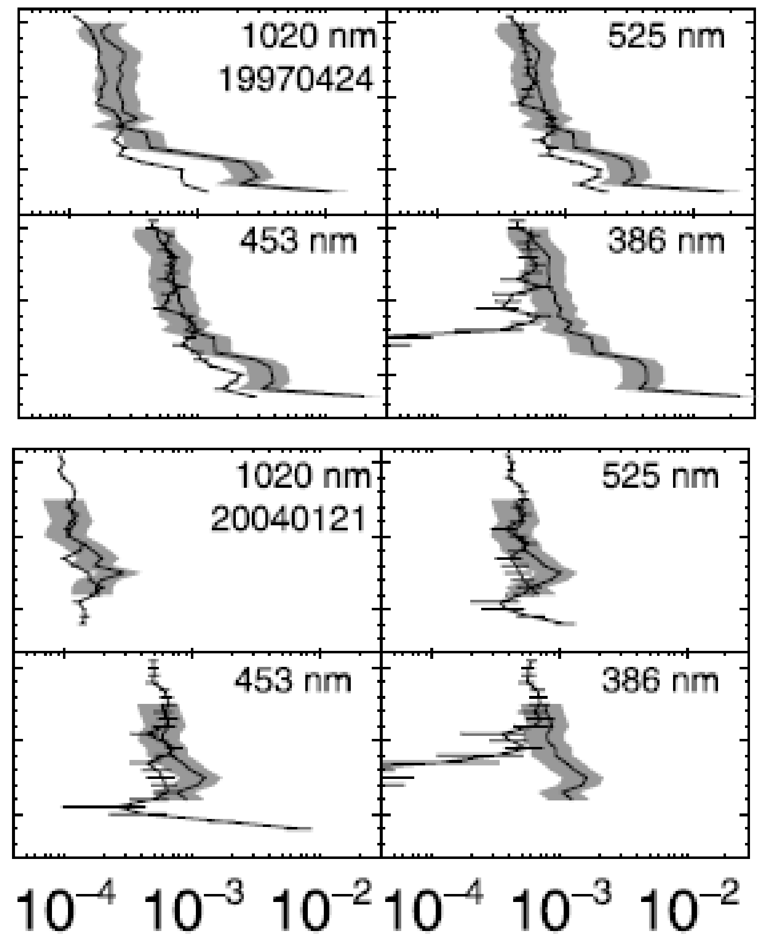
dN/dLogDp

dV/dLogDp



Reeves, J. M., et al. (2008), Comparison of aerosol extinction coefficients, surface area density, and volume density from SAGE II and in situ aircraft measurements, J. Geophys. Res., 113, D10202, doi:10.1029/2007JD009357.





Extinction Profiles. (km^{-1}). Shaded is 1 sigma FCAS uncertainty of 35%. SAGE II profiles show published error bars.

Surface area profiles overlap less often than do the extinction profiles. FCAS surface uncertainty is 28%

Reeves, J. M., et al. (2008), Comparison of aerosol extinction coefficients, surface area density, and volume density from SAGE II and in situ aircraft measurements, J. Geophys. Res., 113, D10202, doi:10.1029/2007JD009357.

Table 3. Mean Fractional Differences of All Available Points for Aerosol Extinctions and of All Surface Area and Volume Points From 12-km Altitude and Higher[a]

Wavelength, nm	Mean Fractional Difference, %	Number of Points
1020	14 ± 5	142
525	19 ± 4	139
452	24 ± 4	138
386	79 ± 6	113
Surface area	55 ± 2	110
Volume	35 ± 2	110

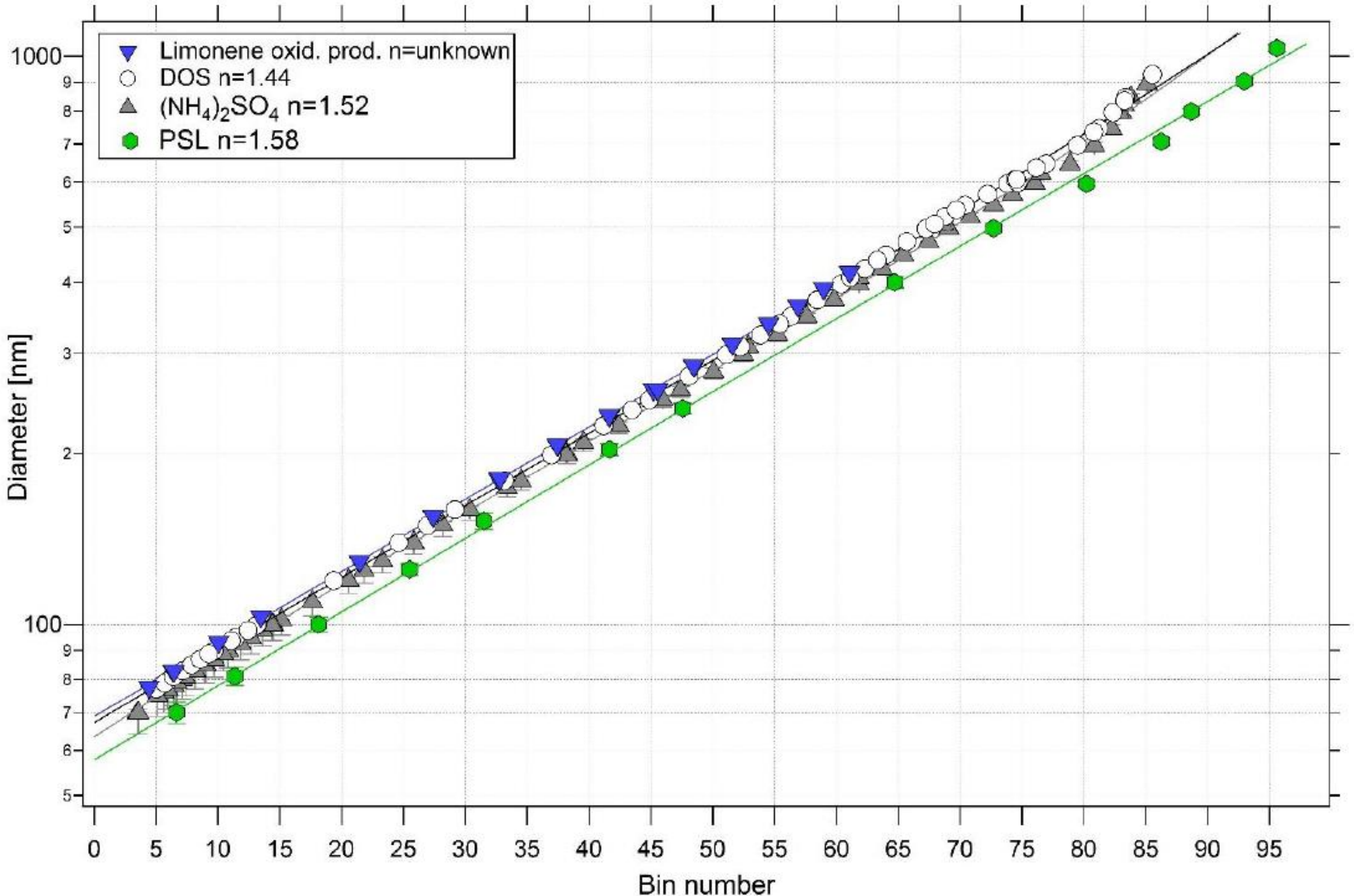
[a]The fractional differences for extinction are calculated with respect to the average: (FCAS-SAGE)/Avg, but for surface and volume are calculated as (FCAS-SAGE)/FCAS. Uncertainties are the standard error ($\sigma/\sqrt{N}$) of the means.

Modification, Calibration, and Performance of the Ultra-High Sensitivity Aerosol Spectrometer for Particle Size Distribution and Volatility Measurements During the Atmospheric Tomography (ATom) Airborne Campaign

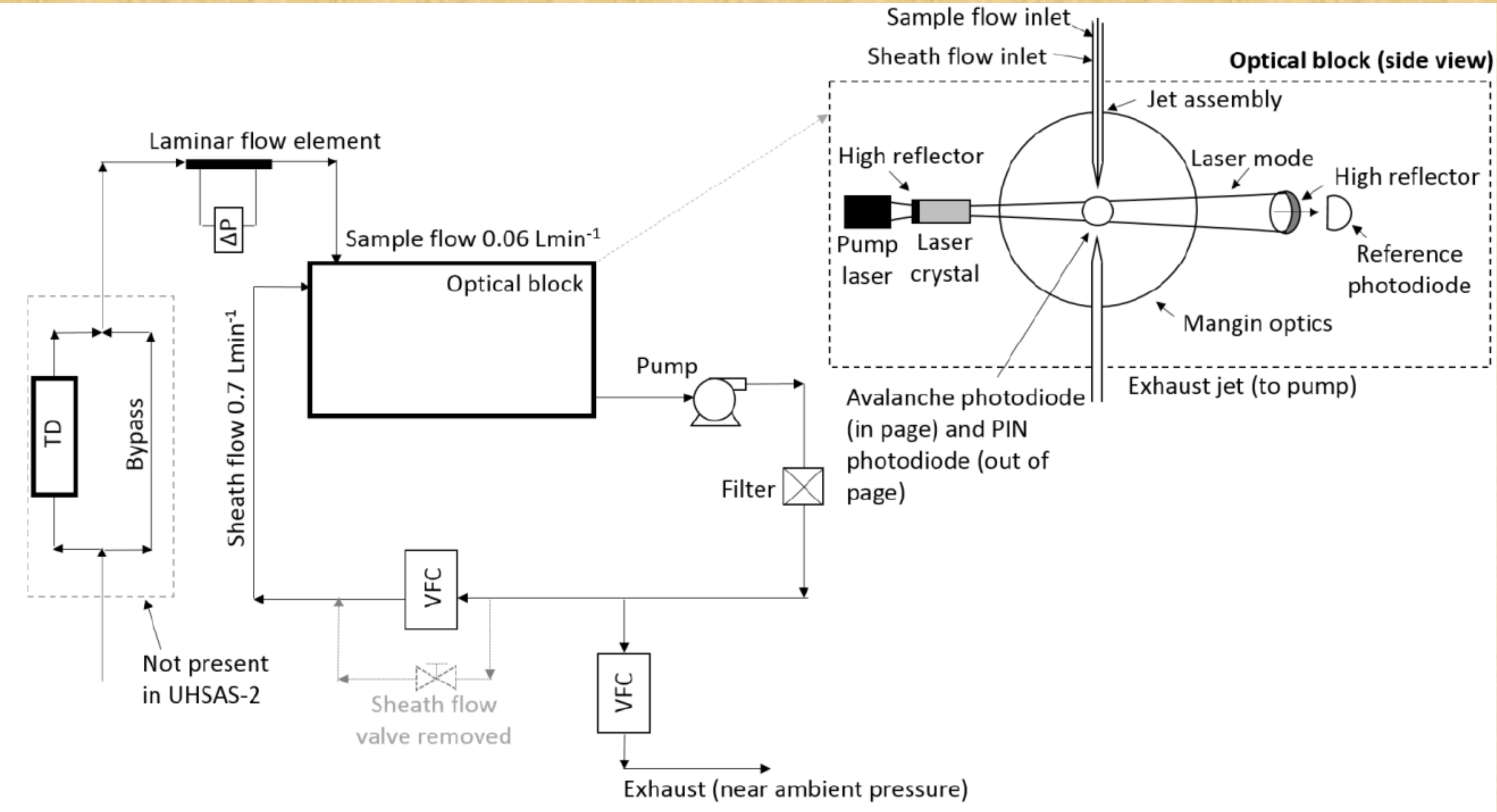
Draft – do not cite.

Agnieszka Kupc[1,2], Christina Williamson[1,2], Nicholas L. Wagner[1,2], Mathews Richardson[1,2], Charles A. Brock[1]

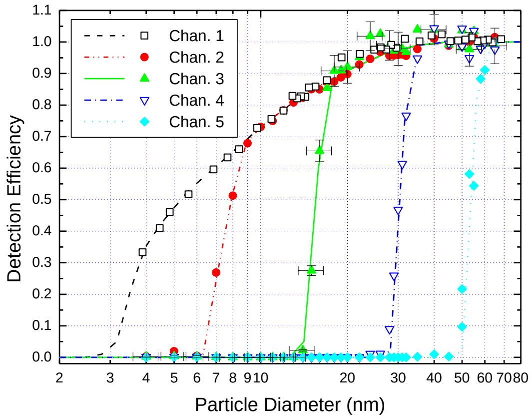
Ground-type UHSAS modified for use on NASA DC-8.
Modified flow system.
Response characterization with 4 types of aerosol.
Uncertainties due to various factors analyzed.
Detection efficiency as a function of pressure not analyzed.

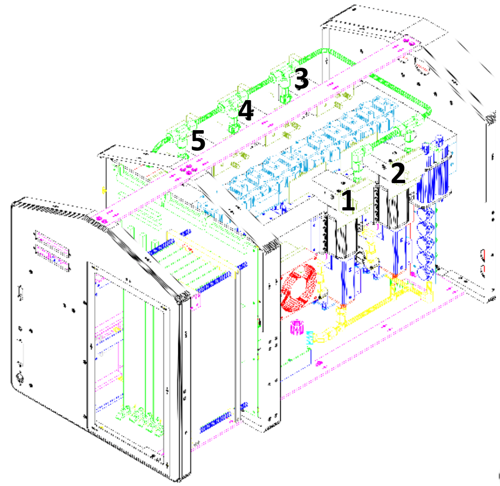


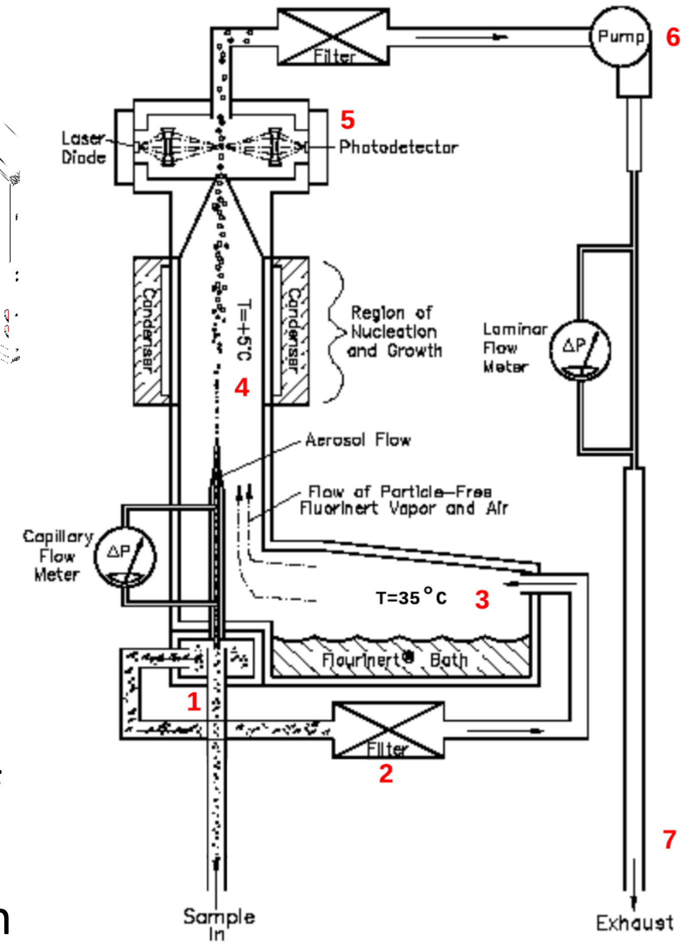
UHSAS Function and Modifications



Nuclei Mode Aerosol Size Spectrometer (NMASS)







- There are 5 CPC modules in parallel. The working principal of the CPC consists of three processes: (1) creation of a supersaturated vapor from a working fluid, (2) rapid growth of aerosol particles by condensation of the supersaturated vapor, and (3) optical detection of the particles that did grow.
- 50% cut points are determined by calibration
- All five CPCs operate at 60 mb in stratospheric application.

An instrument for fast-response measurement of particle size distributions in the 3-60nm size range

Christina Williamson, Agnieszka Kupc, James Wilson, David W. Gesler, J. Michael Reeves, Frank Erdesz, Richard Mcglaughlin, Charles A. Brock

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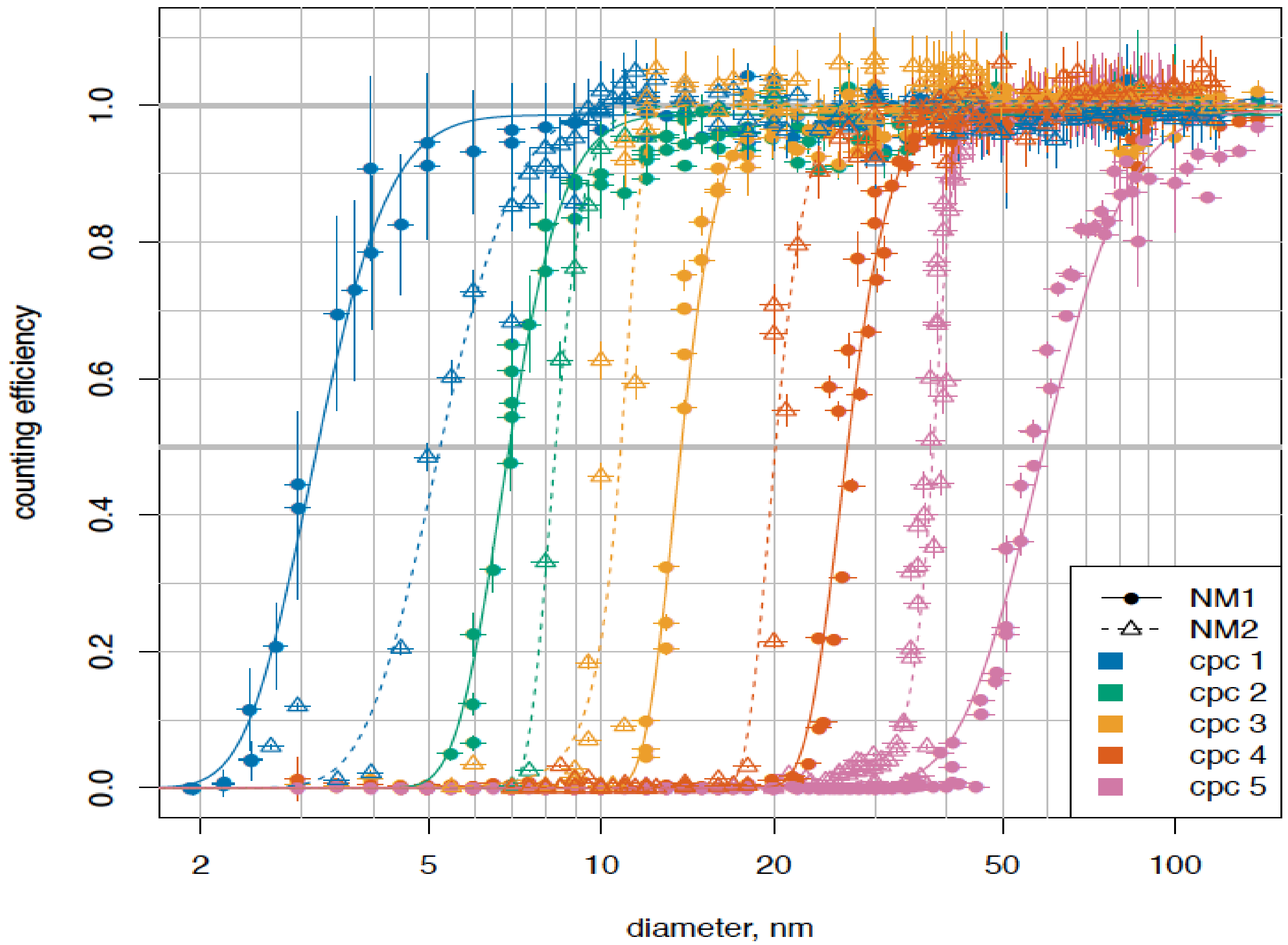

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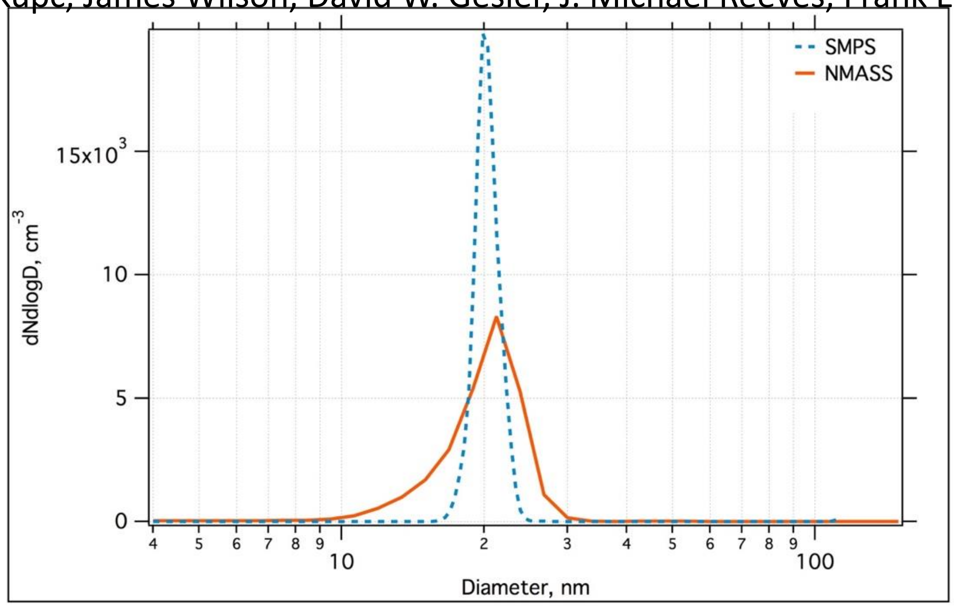
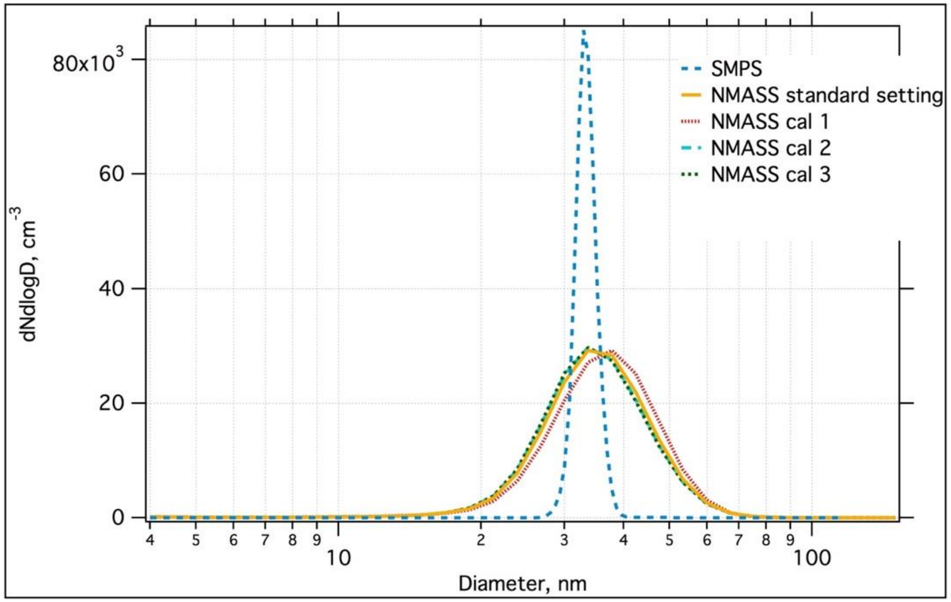
20 nm and 32 nm DMA selected Ammonium Sulfate aerosol

Four different inversions





Some Estimated Parameters

Instrument	Sample Flowrate
NMASS	~4 cc/s at 60 mb
UHSAS	~1 cc/s at stagnation pressure
FCAS	5 to 30 cc/s at stagnation pressure
POPS	Few cc/s

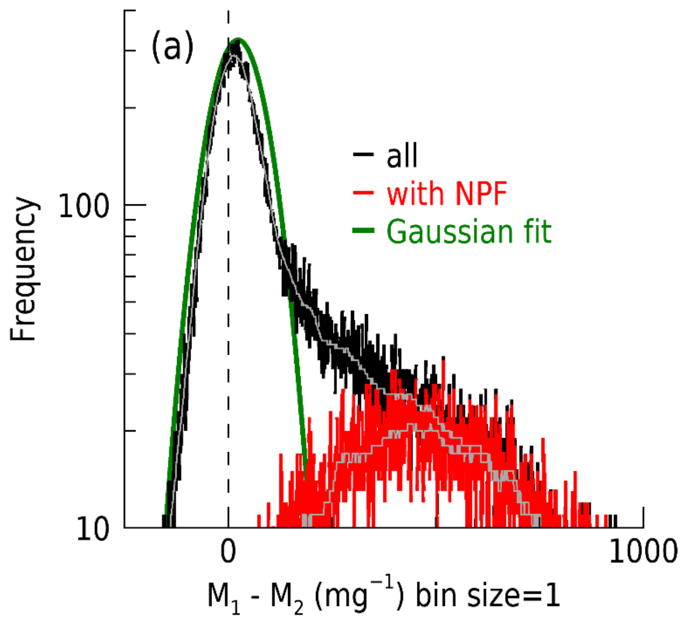
Instrument	Diameter Uncertainty
NMASS	~7%*
UHSAS	Several Percent
FCAS	<2.5%*
POPS	Several Percent

Inlet	Figure of Merit
Passive, Near Isokinetic Inlet	Typically within 20% of isokinetic. Corrections Made

* Requires sufficient sample size

Albedo modification experimental design will need to accommodate sample flowrates

Detection of aerosol new particle formation



$$\text{Relative Difference}=\frac{M_1 - M_2}{\sqrt{(b_1^2 C_1 + b_2^2 C_2)}}$$

$M_i = b_i C_i$
M_i are mixing ratios from CPC_i
C_i are counts from CPC_i
b_i is dependent on sample flow

- M_1-M_2 equals the mixing ratio (particles/mg air) of particles smaller than the lower detection limit of CPC_2 (~8 nm) and larger than the lower detection limit of CPC_1 (~4 nm).
- If relative difference > 3, the chance that M_1 exceeds M_2 only as a result of statistical fluctuations is less than 0.13% (certainty of 99.87%).
- Criteria is to assign NPF when relative difference > 3. Of all the out-of-cloud data points, 22% contained NPF.
- In this case, NPF occurs when nucleated particles grow to a detectable size (> 4 nm).