Aerosol Intensive Operational Periods 2002 – 2003

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and Aerosol Working Group

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Outline

- Aerosols and ARM
- Aerosol-related areas requiring additional effort
    - Aerosol optical thickness and humidification
    - Aerosol absorption and diffuse irradiance closure
    - CCN
- In Situ Aerosol Absorption Measurement Characterization Experiment (2002)
    - Measurements and Instruments
- Aerosol IOP (2003)
    - Measurements and Instruments

ARM and Aerosols

Why does ARM need to know about aerosols?

- **Relate observed radiative fluxes and radiances in the atmosphere, spectrally resolved and as a function of position and time, to the temperature and composition of the atmosphere.**

Direct effects

• **How do aerosols affect the calculation of clear-sky radiation fields?**

Indirect Effects

• **What is influence of aerosols on cloud radiative and microphysical properties?**

ARM and Aerosols

What aerosol measurements and derived products are needed?

- **Assessing the direct impact of aerosols on radiation**
  (aerosol optical properties)
  - **aerosol optical thickness**
  - **single scattering albedo**
  - **phase function**
- **Modeling and developing parameterizations**
  (relating aerosol optical and physical properties)
  - **aerosol size distribution**
  - **aerosol composition**
- **Assessing and modeling cloud indirect effects**
  - **direct CCN measurements**
  - **aerosol size distribution and composition**

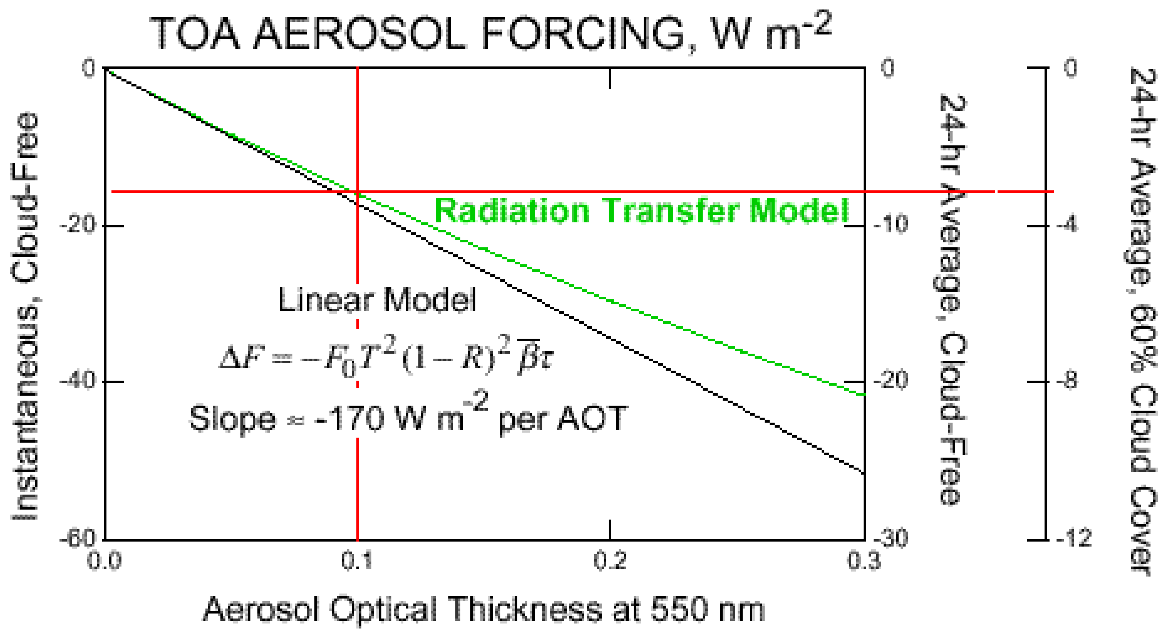
<u>Migration from longwave to shortwave requires a greater focus on aerosol-related issues</u>

DIRECT AEROSOL FORCING AT TOP OF ATMOSPHERE

Dependence on Aerosol Optical Thickness

Comparison of Linear Formula and Radiation Transfer Model

Particle radius r = 85 nm; surface reflectance R = 0.15; single scatter albedo $\omega_0 = 1$.



Global-average AOT 0.1 corresponds to global-average forcing -3.2 W m^{-2}.

Schwartz, BNL

Routine SGP Aerosol Measurements

Instrument	Primary Measurement	Derived
AOS (Surface)	- aerosol light scattering at 3 wavelengths (RH≤40%), (0.1, 10 μm size cuts) - aerosol absorption coefficient (PSAP) (RH≤40%), (0.1, 10 μm size cuts) - single scattering albedo - Angstrom exponents - total condensation particle concentration - ozone - aerosol number distribution (0.1 to 10 μm) - light scattering (green) as a function of relative humidity (f(RH))	- aerosol extinction coefficient - aerosol single scattering albedo - Angstrom coefficient - Hemsipheric backscatter fraction
CSPOT (Cimel) Sun and sky photometer	- AOT (6 wavelengths) - Angstrom exponents - Sky radiance in principal plane and almucantar	- aerosol size distribution - refractive index - single scatter albedo
MFRSR	- AOT (5 wavelengths) - Angstrom exponent	- ozone - NO_2
RSS	- direct spectral irradiance - diffuse spectral irradiance	- AOT
CART Raman Lidar	- aerosol backscatter profiles - aerosol extinction profiles - aerosol optical thickness profiles - water vapor mixing ratio profiles	- relative humidity
MPL	- relative aerosol backscatter	- aerosol backscatter profiles - aerosol extinction profiles
In Situ Aerosol Profiling (IAP)	- aerosol scattering (3 wavelengths) (dry) - aerosol absorption (1 wavelength) (dry) - hemispheric backscatter fraction (dry) - aerosol scattering (1 wavelength) (high RH) (future)	- single scatter albedo - AOT - Angstrom exponents
Aerosol Sample	- aerosol mass concentration - aerosol ionic composition	

Aerosol-related areas requiring additional effort

Aerosol humidification and AOT closure

Background: Parameterizations of aerosols in climate models requires relating aerosol optical characteristics (AOT, w_o) to physical characteristics (size, composition, mass)

Problem: Most in situ measurements of aerosol scattering, absorption and size distribution are made for dry and not ambient conditions

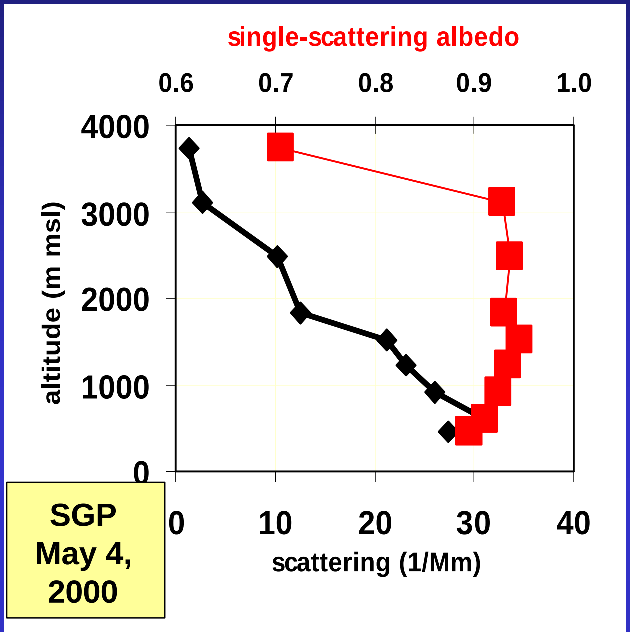
- **AOS, Gulfstream, IAP measure aerosols in dry state**
- **AOS measures f(RH) for scattering at surface only**
- **airborne in situ scattering and absorption underestimated AOT by 25-30% for high RH**
- **IAP measurements of aerosol extinction appear lower than lidar measurements, Sun photometer**

DOE ARM In-Situ Aerosol Profiling (IAP)

Objective: Obtain a statistically-significant data set of vertical distribution of aerosol properties

Measurements: aerosol scattering and absorption above a similarly instrumented surface site

2-3 profiles/week for 1 year



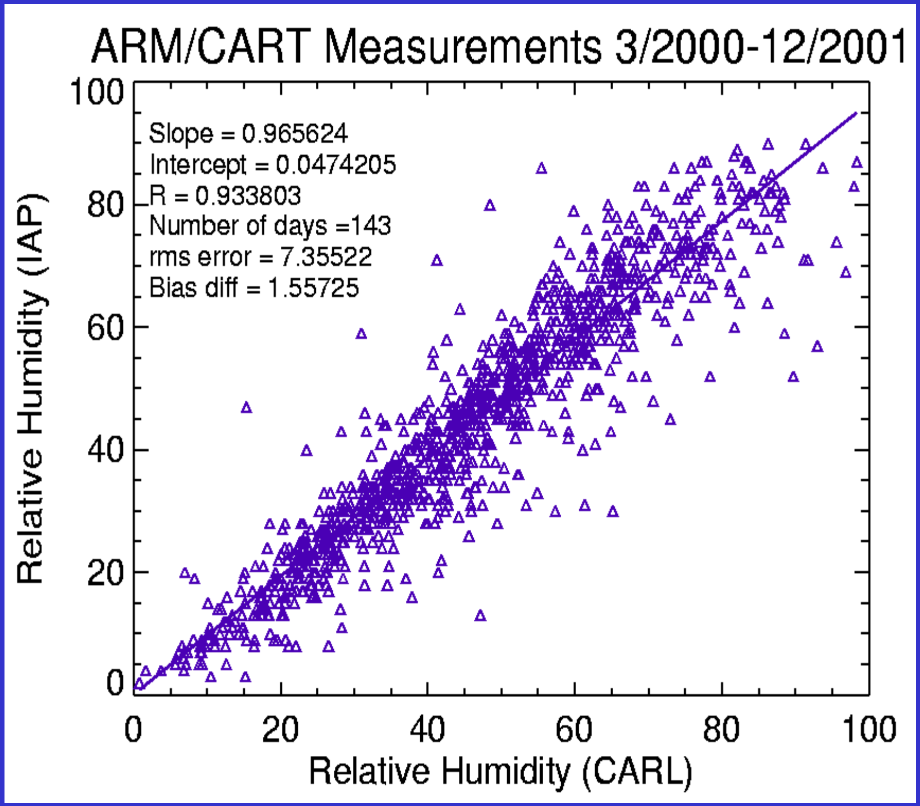
Aircraft

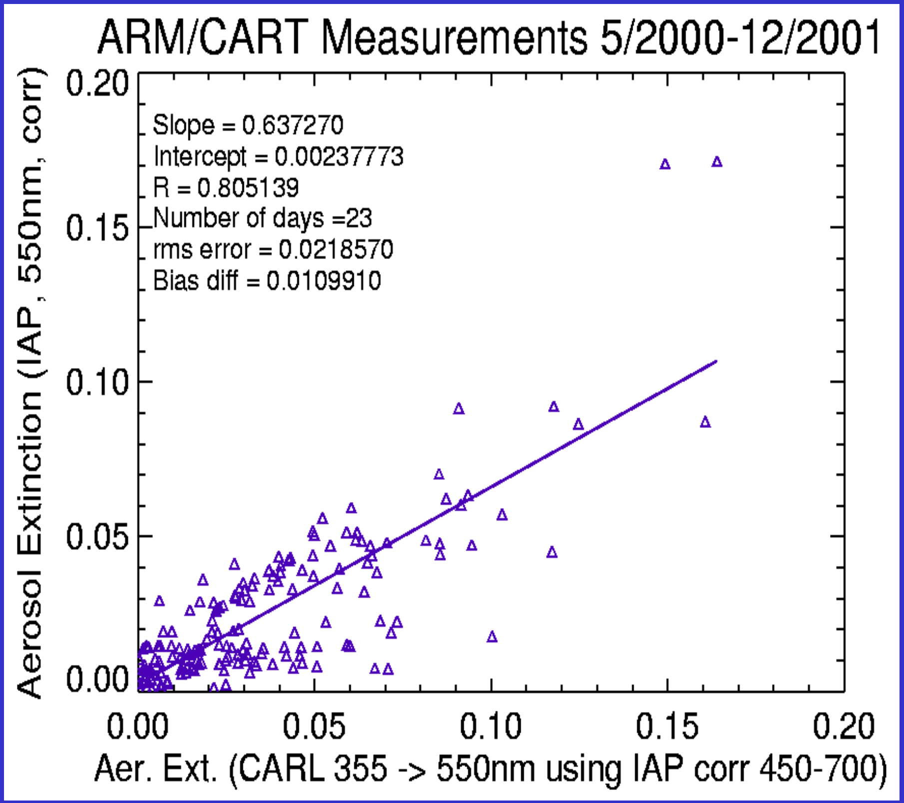
Port view of rack

John Ogren
Betsy Andrews
NOAA/CMDL

Raman lidar and IAP Comparisons

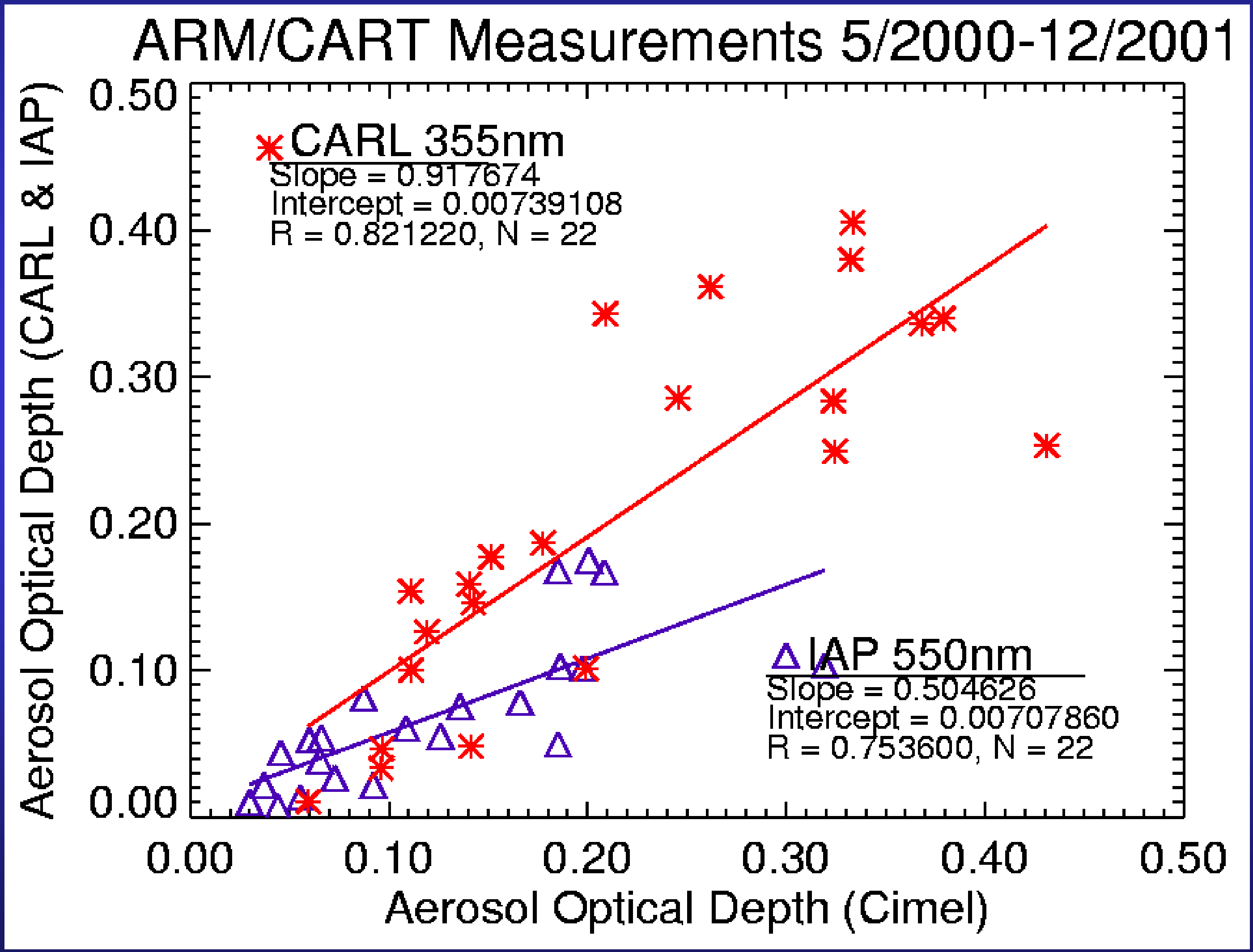
- Combine Raman lidar/IAP to study aerosol vertical variability
- Initial Comparisons
  - good agreement (within ~5-7%) in RH
  - differences in aerosol extinction





Raman lidar/IAP/Cimel AOT comparison

IAP measurements of aerosol optical thickness appear lower than lidar measurements, Sun photometer



Aerosol-related areas requiring additional effort

Aerosol absorption and diffuse flux closure

• Models overestimate diffuse downward irradiance so that attempts to reconcile the differences between measurements and models require large aerosol absorption and/or changes in surface albedo

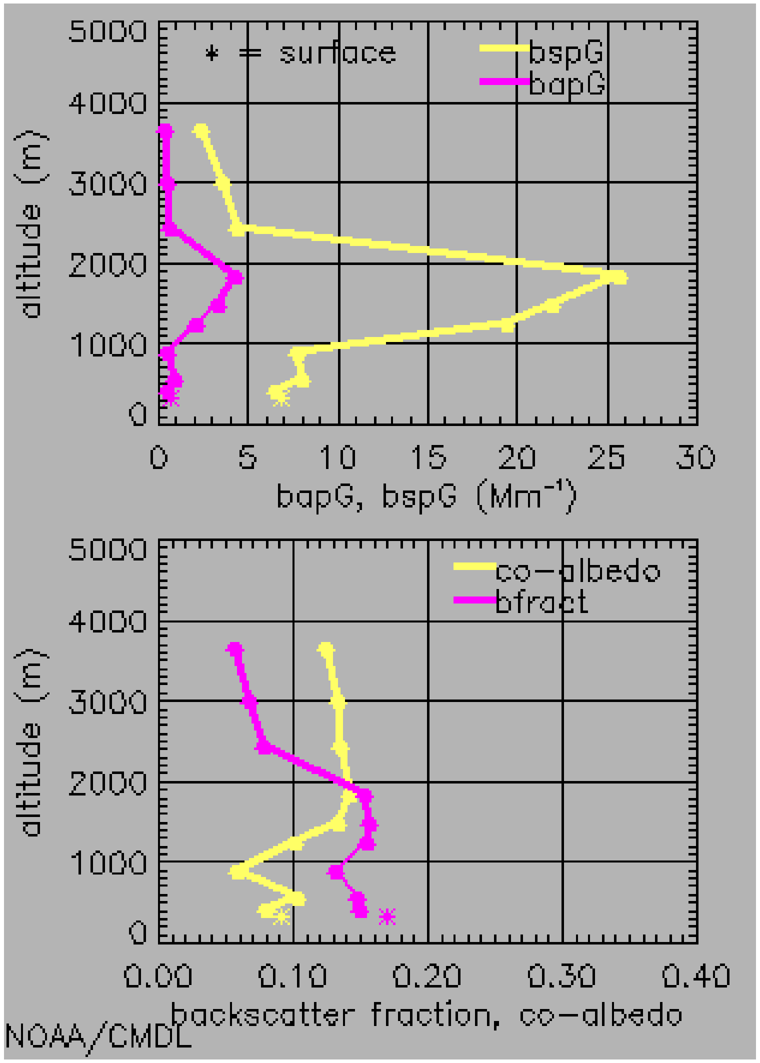
• Using best estimates of both aerosol absorption and surface albedo reduces but does not eliminate differences between measurements and models of diffuse irradiance (see poster by Powell, Kato, et al.)

• Large uncertainties in w_o and surface albedo

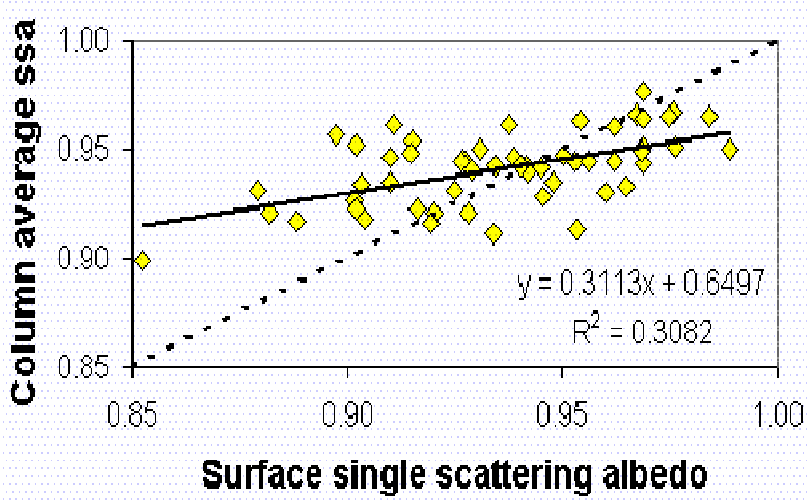
• surface AOS - (PSAP w_o ~ 0.9-1.0)
- **photoacoustic method suggests AOS PSAP *under*estimates w_o**
- **AOS measures dry aerosols (not ambient conditions)**

Aerosol Single Scattering Albedo

IAP Measurements April 4, 2000



- IAP flights often show large vertical variability in ω_o
- Surface measurements often differ from column averages



- Resolution requires concerted effort to measure aerosol absorption

John Ogren, Elisabeth Andrews NOAA/CMDL

Aerosol Absorption

In Situ Aerosol Absorption Measurement Characterization Experiment

- **Objectives**
  - **Characterize uncertainties associated with existing ARM SGP surface and airborne measurements of aerosol absorption and extinction**
  - **Develop methodology to employ new instruments to more accurately measure profiles of aerosol absorption, scattering, and extinction under low AOT conditions during Aerosol IOP**
- **Desert Research Institute**
- **June, 2002**
- **"Closure" experiment for aerosol absorption, extinction under controlled conditions**

In Situ Aerosol Absorption Measurement Characterization Experiment

- Aerosol absorption – PSAP, aethalometer, photoacoustic, absorption photometer
- Aerosol scattering – TSI, integrating sphere nephelometers
- Aerosol extinction – Cavity ringdown instruments

Table 1. Instruments for aerosol optics measurements.

Instrument	Group Responsible	Measurement	Wavelength (nm)
Cavity Ringdown	DRI	σ_{ep}	532
Cavity Ringdown	NASA Ames	σ_{ep}	690, 1550
TSI 3 Wavelength Nephelometer	NOAA/CMDL	σ_{sp}, σ_{bsp}	450, 550, 700
Integrating Sphere Nephelometer	DRI	σ_{sp}	532
Particle Soot Absorption Photometer (PSAP)	NOAA/CMDL	σ_{ap} (filter-based)	565[a]
Aethalometer	DRI or NOAA/CMDL	σ_{ap} (filter-based)	370, 450, 571, 615, 660, 880 950
Photoacoustic Instrument	DRI	σ_{ap}	496.5, 514.5, 532[b], 632, 685, 1047
EMS Andersen Absorption Photometer	DLR	σ_{ap} (filter-based)	670

[a]adjusted through calibration to 550 nm.
[b]primary wavelength for this study.

In Situ Aerosol Absorption Measurement Characterization Experiment

- Dry aerosols produced in laboratory
- Aerosol generator and mixing chamber from NOAA/CMDL
- Aerosols collected and analyzed for elemental and organic carbon

Table 3. Aerosols and preparation.

Aerosol or Preparation	Group Responsible	Comment
Aerosol Generator for externally mixed black and white particles	NOAA/CMDL	Will allow for external mixtures of the various aerosols listed below.
Differential Mobility Analyzer (DMA)	NOAA/CMDL	Monodisperse aerosol generation between 10 – 500 nm diam.
Mixing vessel	NOAA/CMDL	Multiple inlets and outlets for aerosols with controls for creating aerosols of varying single scattering albedos
Kerosene soot	DRI	A sooting kerosene lamp is diluted with filtered air. The optical and physical properties of this soot have been extensively characterized by DRI.
Diesel soot	NOAA/CMDL	A small diesel engine will be used generate diesel soot particulates.
Graphitic carbon	NOAA/CMDL	A small carbon vane pump will be used to mechanically generate fine graphite particles.
NaCl aerosols	NOAA/CMDL	Typical white aerosols generated using an atomizer.
$(NH_4)_2SO_4$ aerosols	NOAA/CMDL	Typical white aerosols generated using an atomizer.
'White Smoke Candles'	DRI	Cole Palmer white smoke candles are used as an aid for observing flow patterns. This smoke can be externally mixed with soot, or used by itself.
Ambient Aerosol. Primarily an urban aerosol source, though with some smoke from local fires likely.	DRI	An inlet will be prepared to ingest ambient aerosol from the outside.

• Aerosols collected and analyzed for elemental and organic carbon

Table 2. Instruments for aerosol characterization.

Instrument	Group Responsible	Measurement
Quartz Filter	**DRI**	**Elemental and organic carbon analysis**
Nuclepore filter	**DRI**	**Electron microscopy for particle size/morphology studies.**
Tapered Element Oscillating Microbalance (TEOM)	DRI	Total Aerosol Mass as a function of time.
TSI Dusttrak	DRI	Optical scattering surrogate for total aerosol mass
Teflon Filter	DRI	Total Filter aerosol mass, optical densitometer measurement aerosol absorption with white light.
Scanning Mobility Particle Sizer (SMPS)	DRI	Particle size distribution from 10 to 500 nm diameter.
GRIMM Optical Particle Sizer	DRI	Particle size distribution from 0.5 to 20 μm diameter.

Aerosol IOP

Located over SGP during May, 2003

1. Aerosol absorption and diffuse flux closure
   - Concerted effort to measure aerosol scattering, absorption
     - Surface
       - photoacoustic instrument to calibrate surface and airborne PSAP
       - multi-wavelength measurements of absorption and optical thickness
     - Aircraft
       - calibrated PSAP
       - Sun photometer (provide closure on AOT)
       - nephelometers (standard+ambient)
       - measure hygroscopic humidification factor
       - accurate broadband and spectrally resolved radiation measurements
2. Aerosol humidification and AOT closure
   - Airborne measurements
     - Sun photometer (provide closure on AOT)
     - nephelometers (scattering)
     - additional PSAP (absorption)
     - cavity ringdown instruments (extinction)
     - measure aerosol hygroscopic humidification factor
3. CCN and clouds
   - CCN
   - cloud liquid water path

Aerosol IOP: Aerosol Optical Thickness (AOT) closure

Hypotheses:

• How well do the routine CART Raman lidar and In Situ Aerosol Profiling measure of aerosol scattering and extinction profiles and AOT?
• How well can the surface measurements of aerosol scattering humidification factor be used for aerosols aloft?

Background:

• Aerosol extinction, optical thickness from periodic IAP flights differ from lidar, Sun photometer measurements
• Desire to identify reasons for random differences in AOT from lidar and Sun photometer
• Potentially large error in assumptions of aerosol humidification factor

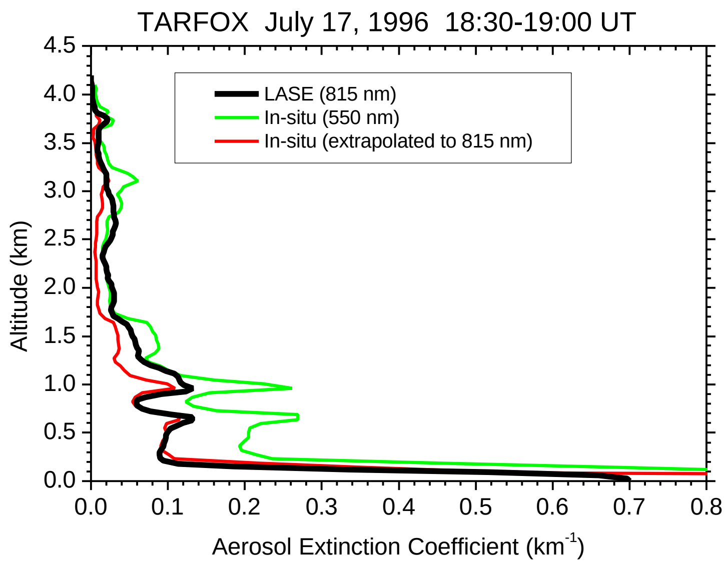
General Approach:

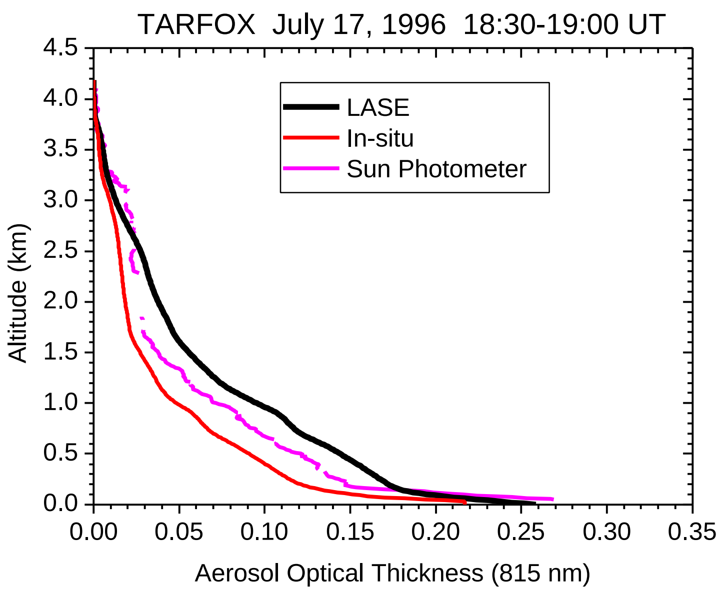
1. Use airborne Sun photometer to measure aerosol extinction, optical thickness profiles
2. Use additional airborne in situ measurements of scattering, absorption, extinction
3. Acquire airborne measurements of aerosol humidification factors

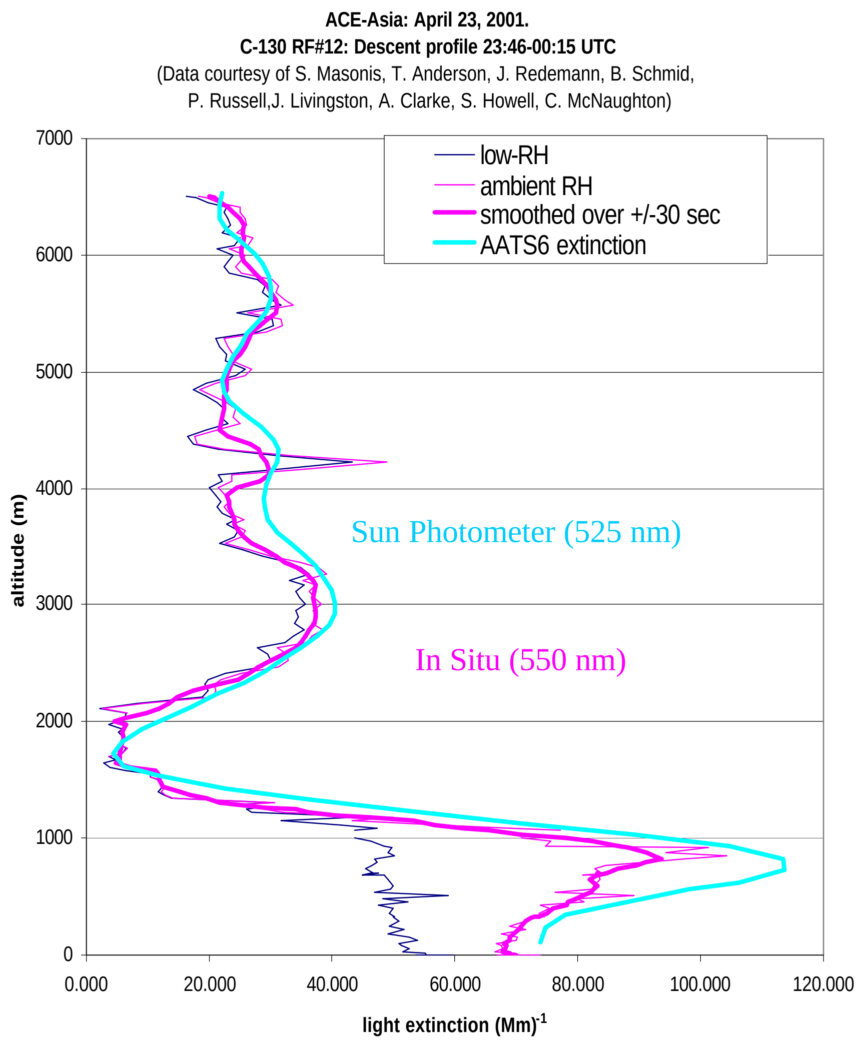
Goal:

Accurately characterize ability of routine ARM SGP measurements to measure profiles of aerosol optical thickness, extinction. Assess ability of models to recreate aerosol optical profiles.

Examples of Aerosol Extinction Closure Experiments







Aerosol IOP: Diffuse Flux Closure

Hypothesis:

Can closure between measurements and models of diffuse radiation be achieved under low AOT conditions with accurate measurements of the aerosol single scattering albedo?

Background:

Considerable uncertainty in the values of aerosol absorption and single scattering albedo ω_o that have been derived (measured vs. model) from various methods

General Approach:

1. Use photoacoustic method, which measures the sound pressure produced in an acoustic resonator caused by light absorption, to "calibrate" the surface PSAP measurements as well as a PSAP on an aircraft.
2. Measurements of diffuse radiation (under cloud-free conditions) would be acquired on days with simultaneous measurements of ω_o acquired by both (i.e. PSAP filter and photoacoustic) types of surface measurements discussed above as well as airborne profiles of aerosol absorption, scattering, and extinction.
3. Acquire measurements of aerosol absorption at several wavelengths to determine the validity of the common assumption that aerosol absorption is constant in the visible part of spectrum.
4. Acquire airborne (low-altitude) measurements of surface albedo.

Goal:

Accurately constrain the lower limit on ω_o throughout the atmospheric profile during periods of low AOT and then compare the measured and modeled diffuse radiation.

Aerosol IOP AOT and Diffuse Flux “Closure” Experiments

- Aerosol Extinction (Surface)
    - AOS (Scattering+Absorption)*Humidification/Extinction
- Aerosol Absorption (Surface)
    - AOS PSAP/Photoacoustic cell
- Humidification Factor (Profile)
    - AOS Surface + IAP (single elevated RH)/Aircraft Humidigraph
- Aerosol Extinction Profiles Derived from SGP Facility Instruments
    - Raman, MPL Lidars/Airborne Sun Photometer
    - Raman, MPL Lidars/Aircraft [In Situ (Scattering+Absorption), Extinction]
    - IAP/Airborne Sun Photometer
    - IAP/Aircraft [In Situ (Scattering+Absorption)*Humidification, Extinction]
- Aerosol Absorption Profile
    - IAP/Aircraft [In situ (Calibrated Absorption)]
- Aerosol Scattering/Absorption/Extinction Profiles
    - Lidar/Sun Photometer, Aircraft In Situ/Aircraft In Situ Model (Aerosol Size + Composition)
- Diffuse Downwelling (Broadband)
    - Measured (Shaded Pyranometer)/Model (with aerosol/gas inputs)
- Diffuse Downwelling (Spectral)
    - Measured (RSS,SSFR)/Model (with aerosol/gas inputs)
- Diffuse/Direct Ratio (Spectral)
    - Measured/Model (with aerosol/gas inputs)

CCN and Clouds

(Coordination with Steve Ghan and others from Cloud Parameterization and Modeling Group)

Issues:

- influence of aerosol on droplet number concentration
  - –CCN measurements at cloud base
  - –updraft model or measurements
  - –droplet measurements
- influence of aerosol on cloud liquid water content
  - –above, plus liquid water measurements
- influence of aerosol on cloud optical depth/albedo
  - –above, plus
    - cloud thickness measurements
    - cloud optical depth/radiative forcing measurements

Lack of Measurements:

- *In situ* CCN measurements available only one ARM IOP
- During that IOP, single-layer warm clouds (required for remote sensing retrieval) only formed on one day.
- On that day clouds dissipated before aircraft measurements of droplet number were obtained.

CCN and Clouds

(Coordination with Steve Ghan and others from Cloud Parameterization and Modeling Group)

Hypotheses:

- What is the relationship between CCN number concentration (at several supersaturations in the range ~0.1 - 1%) and aerosol size distribution, at the surface and at cloud base?
- How well can the cloud nucleating properties of particles just below cloud base be represented using surface measurements of cloud nucleating properties of particles along with profiles of relative humidity and aerosol extinction?
- What is the relationship between the cloud base CCN number concentrations and size distributions, cloud base turbulence, and cloud droplet number concentrations and size distributions?

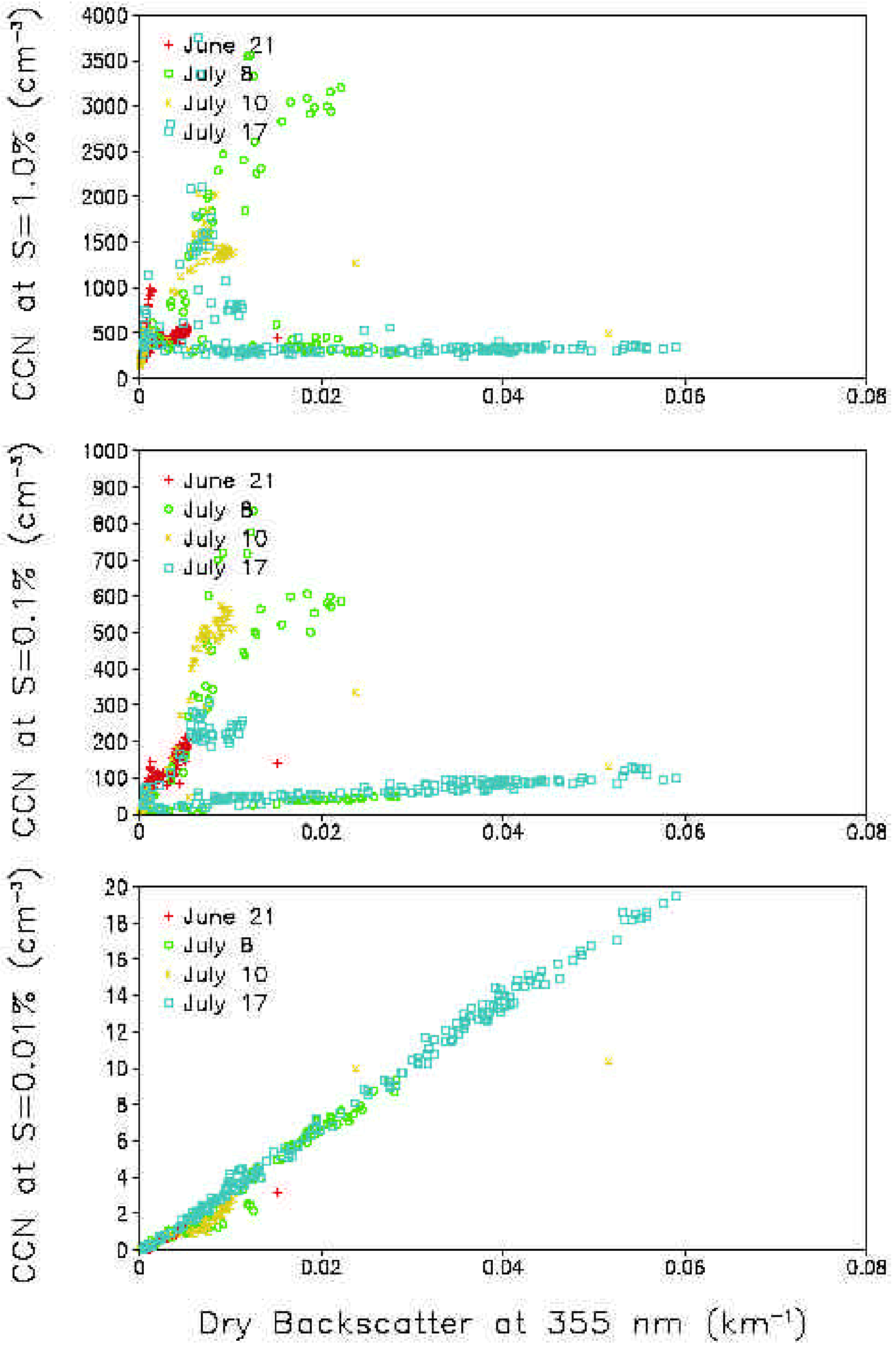
Approach:

1. Acquire both airborne and surface CCN measurements.
2. Acquire vertical profiles of humidification factor, cloud drop number, size distributions
3. Combine surface *in situ* measurements of CCN and f(RH) with lidar profiles of RH and extinction or backscatter to estimate CCN profiles
4. Use cloud radar/MWR/ceilometer to estimate cloud updraft velocity, droplet number concentration, cloud thickness, and liquid water content
5. Sample conditions with similar updrafts but very different CCN concentrations at cloud base

Goal:

Test ability to determine CCN at cloud base with surface CCN measurements. Determine ability to produce a profile from Raman lidar extinction and humidification factor.

CCN retrieval tests



Tests using ACE-2 aerosol data

• The retrieval of CCN at low supersaturations is insensitive to variations in the size distribution, but the retrieval of CCN at high supersaturations is sensitive.

• Retrieval of CCN at low supersaturations from surface measurements appears to be achievable under a wide variety of conditions, but retrieval at high supersaturations is only possible when the size distribution varies little with altitude.

CCN and Cloud “Closure” Experiments

- CCN (Surface)
  - CCN (spectrometer, diffusion chamber)/Aerosol size distribution/composition

- CCN (Cloud base)
  - CCN (spectrometer, diffusion chamber)/Aerosol size distribution/composition

- CCN Profile
  - CCN Surface+Lidar extinction profiles+Humidification+RH/CCN aircraft

- Cloud transmittance
  - Surface measurements of optical depth/Model+LWP+drop concentration

- Cloud drop concentration
  - Model from radar/Aircraft In situ

- Cloud liquid water path
  - microwave radiometer+LWC (radar+model)/aircraft (vertical integrals of drop concentration and LWC)

Additional Airborne Aerosol Measurements during Aerosol IOP

CIRPAS Twin Otter

- Aerosol size distribution (PCASP, FSSP, CAPS)
- Total aerosol number distribution (CNC)
- Aerosol scattering (TSI Nephelometer, 3 wavelengths) (Univ. Washington)
- Aerosol absorption (PSAP, photoacoustic) (Univ. Wash., DRI)
- Aerosol humidification factor (humidigraph) (Univ. Wash)
- Aerosol extinction (Cavity ringdown) (NASA Ames)
- Aerosol optical thickness, extinction (Sun photometer) (NASA/Ames)
- Aerosol size distribution (10 nm – 1 μm at 2 RH) (TDMA)
- Up and Down solar and IR broadband irradiance (CM-22, CG-4) (NRL, Sandia)
- Up and Down solar spectral irradiance (SSFR) (NASA Ames)
- CCN (Cal Tech), (possibly additional measurements on another small aircraft (DRI))
- Cloud liquid water (Johnson probe on CAPS, Gerber probe)
- Meteorological state variables

Additional Surface Aerosol Measurements during Aerosol IOP

- Aerosol absorption (photoacoustic, aethalometer) (DRI, NOAA/CMDL)
- Aerosol extinction (cavity ringdown)
- Sky radiance, polarization, aerosol optical thickness (Sun-sky-surface sensor) (NASA/GSFC)
- broadband irradiance (broadband cavity radiometer) (PNNL)
- UV diffuse/direct irradiance (UVRSS) (SUNY Albany)
- Column ozone (UVRSS) (SUNY Albany)
- CCN (DRI)
- Aerosol size distribution (20-500 nm) (SMPS) (DRI)
- Polarization?

Coordination with DOE Atmospheric Chemistry and Tropospheric Aerosol Programs

- Aerosol composition
- Aerosol size

Summary

- Aerosol absorption and extinction characterization experiment
    - June 2002 at DRI (Lab)
    - Characterize uncertainties associated with existing ARM SGP surface and airborne measurements of aerosol absorption and extinction
    - Develop methodology to employ new instruments to more accurately measure profiles of aerosol absorption, scattering, and extinction under low AOT conditions during Aerosol IOP
- Aerosol IOP
    - May 2003 at SGP
    - Closure experiments to address
        - AOT and aerosol humidification
        - Aerosol absorption and diffuse irradiance closure
        - CCN
    - Additional surface measurements
    - Aircraft measurements

- (See poster by Schwartz et al. for more details and discussion)