ARM - ACP AEROSOL IOP

Chemical - Microphysical - Optical - Radiative Closure Experiments

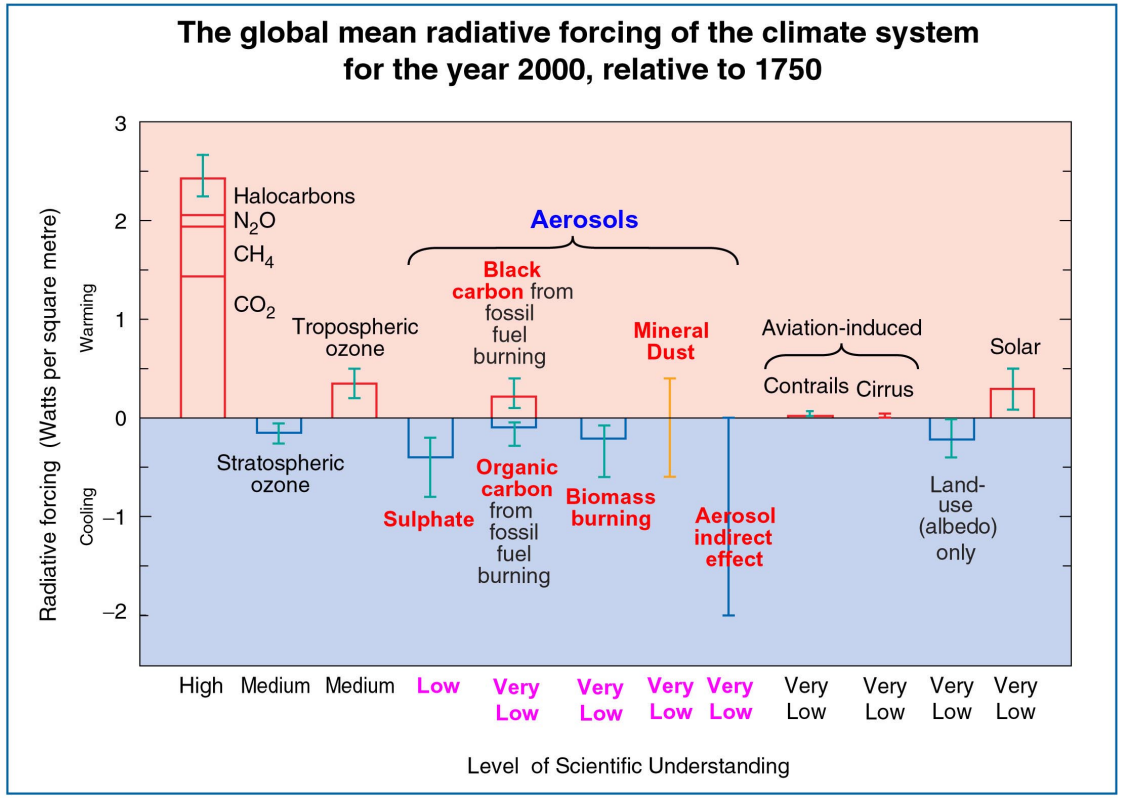
Steve Schwartz and Dan Imre

ACP Annual Meeting
Orlando FL March 4-5, 2003

www.tap.bnl.gov/arm_acp_aerosol_iop

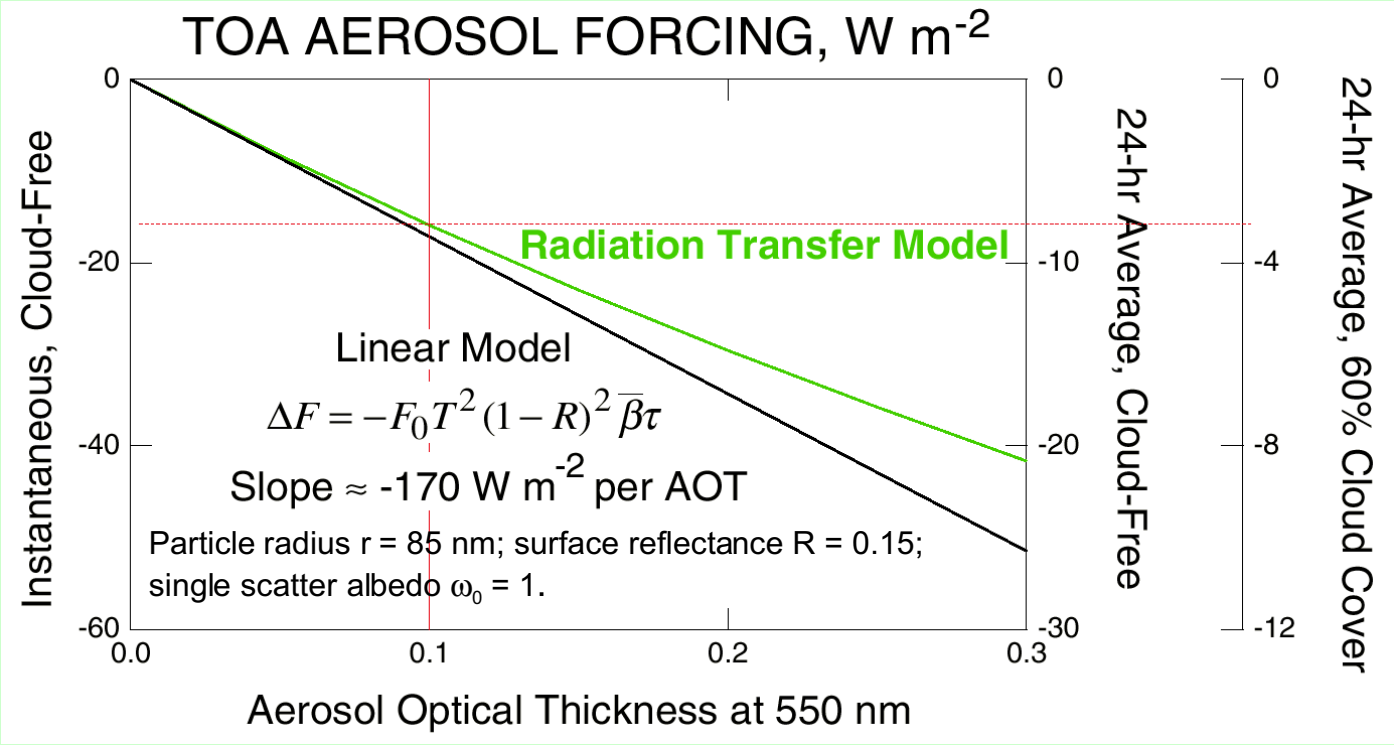
ARM ACP AEROSOL IOP

Motivation: Aerosols are the greatest source of present uncertainty in radiative forcing of climate change.



ARM ACP AEROSOL IOP

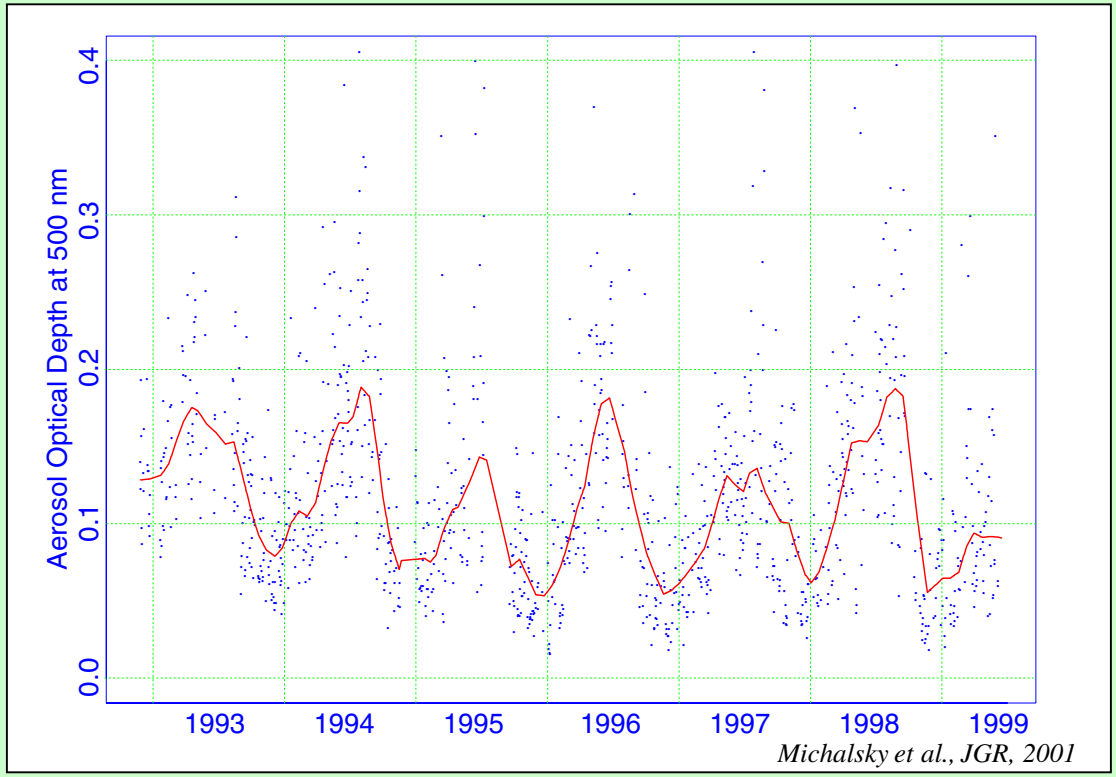
Motivation: Radiative flux components in cloud-free air are very sensitive to aerosol loading and properties.



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

ARM ACP AEROSOL IOP

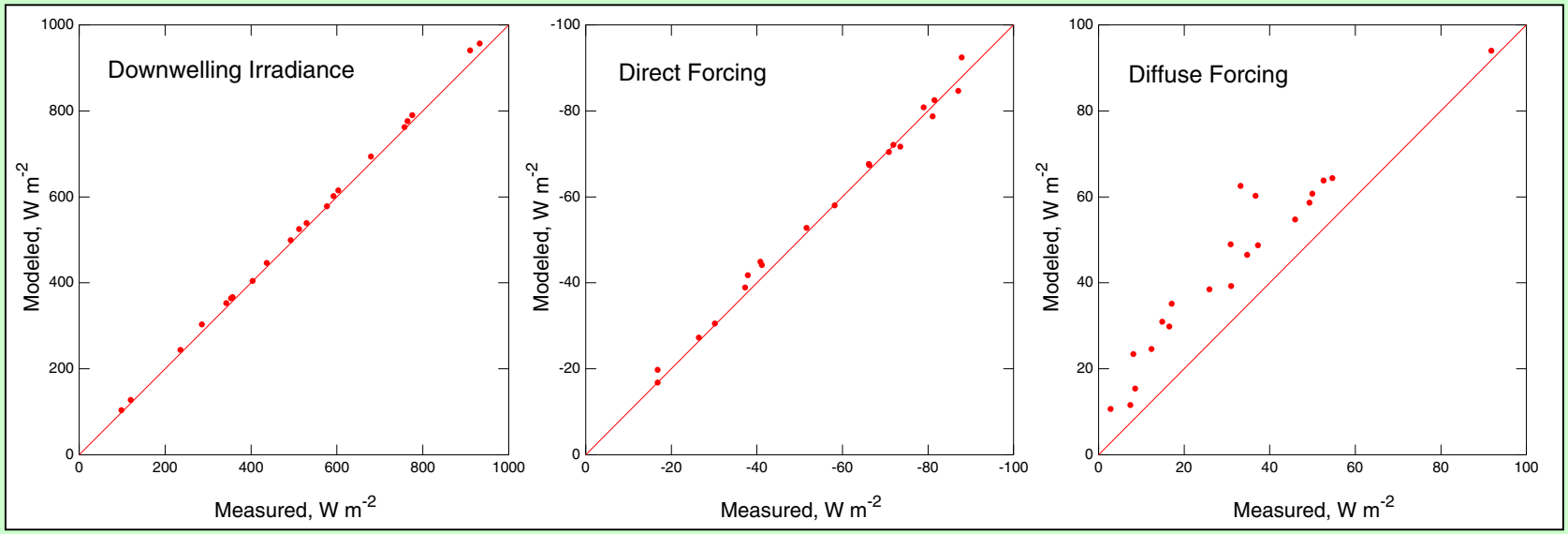
Motivation: Aerosol optical depths leading to substantial direct forcing are not uncommon.



Multiple-year record of aerosol optical depth at ARM SGP site in north-central Oklahoma.

ARM ACP AEROSOL IOP

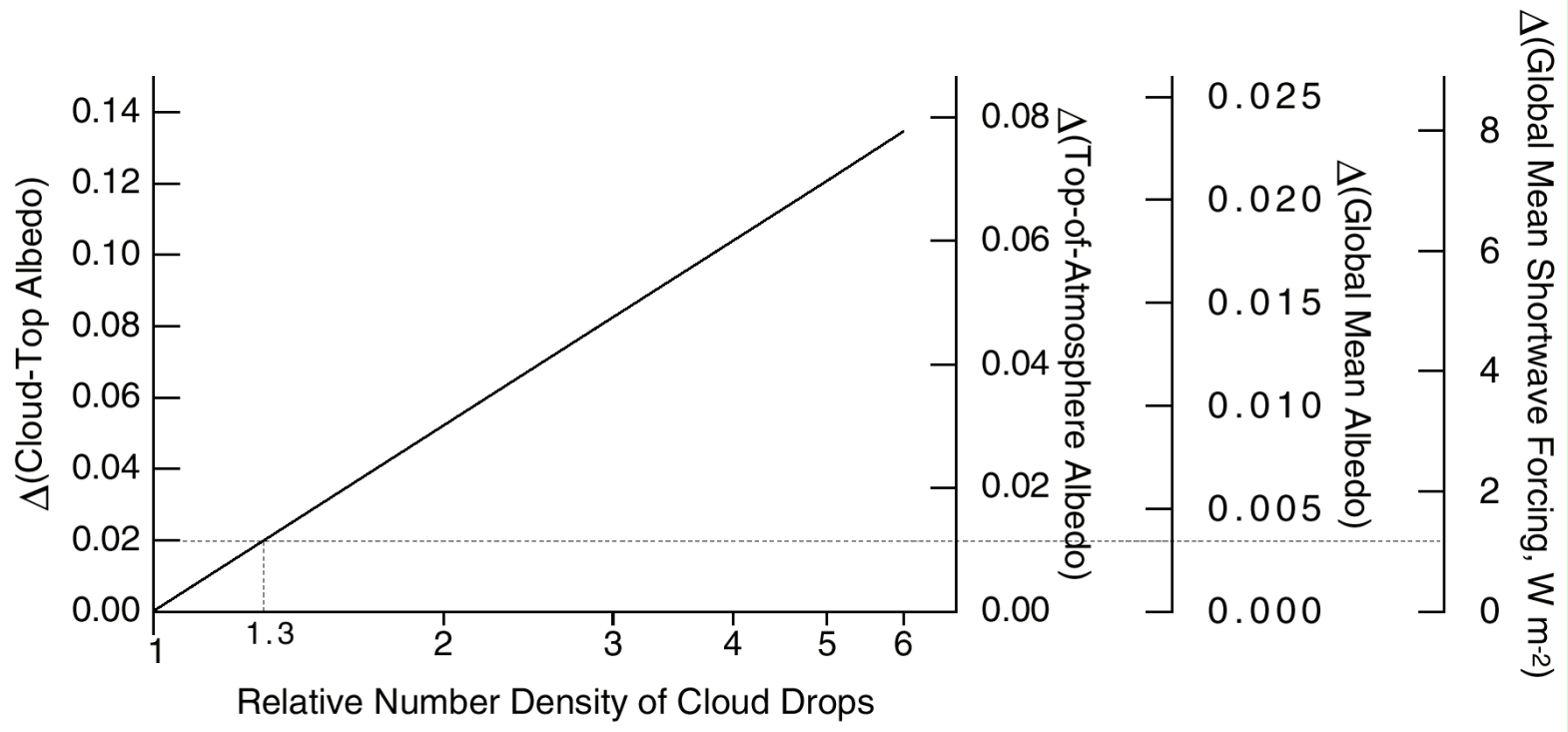
Motivation: There are substantial discrepancies between measured and modeled radiation components.



Measurements during cloud-free conditions at ARM SGP site in north-central Oklahoma.

ARM ACP AEROSOL IOP

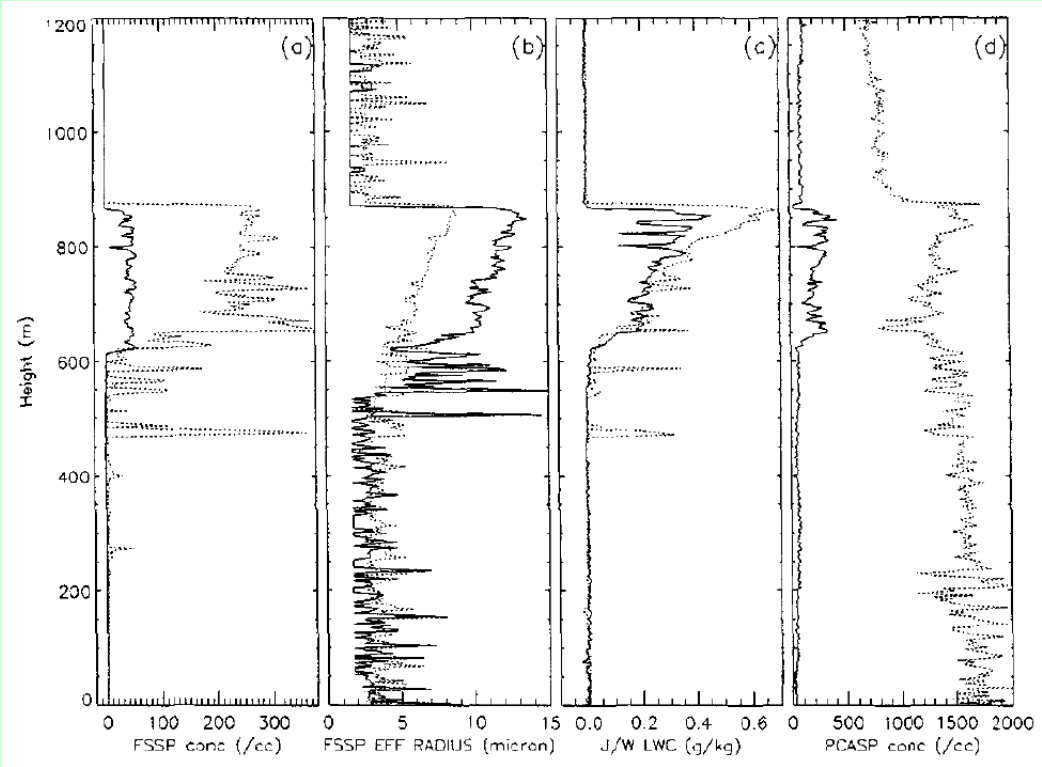
Motivation: Aerosol indirect forcing is highly sensitive to number concentration of cloud drops.



A 30% enhancement in cloud drop concentration in marine stratus clouds results in a global-mean forcing of -1 W m^{-2}.

ARM ACP AEROSOL IOP

Motivation: Anthropogenic enhancement of CCN and cloud-drop concentrations by several fold are common.



Measurements of aerosol and cloud drop concentrations in North Atlantic (ASTEX, 1992)

AEROSOLS & CLIMATE: THE HOLY GRAIL

Sources of Aerosols & Precursors

↓

Aerosol Chemical & Optical Properties

↓

Chemical Transport Models

↓

Detailed Aerosol Loading ($f(t, X, Y, Z)$)

↓

Radiation Transfer Models

↓

Global and Regional Climate Models

ARM ACP AEROSOL IOP

An ARM *Intensive Operational Period* to be conducted at the Southern Great Plains site in north central Oklahoma in May 2003.

IOP Objectives: Evaluate present understanding of aerosol influences on atmospheric radiation and clouds.

IOP Requirements: Detailed characterization of aerosol optical properties and influences on radiation and clouds, at the surface and in the vertical by aircraft and remote sensing.

ARM Objectives

- "Closure experiments" will compare measurement of a quantity with modeled quantity based on measured input variables.
- Careful measurement of aerosol optical properties will remove "wiggle room" in modeling aerosol radiative forcing: extinction coefficient, single scattering albedo and asymmetry parameter.
- Measure vertical distributions of aerosol optical properties and radiative flux components. Compare aerosol extinction with $d\tau/dz$.
- Measurement of CCN spectra and relation to size distribution, composition, and hygroscopic growth will allow development and testing of parametrizations for cloud drop concentration.
- Multiple measurements will constrain uncertainties and identify weaknesses in measurement.

ARM ACP AEROSOL IOP

Some Closure Experiments During ARM Aerosol IOP

Optical properties $f(z)$	$\longrightarrow$	Radiative flux components
Optical properties $f(z)$	$\longrightarrow$	dFlux/dz
CCN f(supersaturation)	$\longrightarrow$	Cloud drop concentration
Composition & Size Distribution	$\longrightarrow$	Optical properties f(RH)
Composition & Size Distribution	$\longrightarrow$	CCN f(supersaturation)

ARM ACP AEROSOL IOP

What is the role of ACP In this IOP?

ARM ALONE

Sources of Aerosols & Precursors

Aerosol Chemical & Optical Properties

Chemical Transport Models

Detailed Aerosol Loading

↓

Radiation Transfer Models

↓

Global and Regional Climate Models

ACP + ARM

Sources of Aerosols & Precursors

↓

Aerosol Chemical & Optical Properties

↓

Chemical Transport Models

↓

Detailed Aerosol Loading

↓

Radiation Transfer Models

↓

Global and Regional Climate Models

ARM ACP AEROSOL IOP

It is impossible to generalize what will be learned at the ARM site during IOP without the combined knowledge of aerosol chemical, optical, and radiative properties.

ACP Objectives

1. Quantify the relationships between aerosol composition and microphysics and aerosol optical properties on the 1 m^3 scale.

2. Quantify the relationships between aerosol composition and microphysics and atmospheric radiation on the 1 to 2 km column scale.

Aerosol Characterization by ACP

- TDMA system + OPC
  - Rapid size distribution - (120 nm - 3 µm) PCASP and (3 nm - 1 µm) DMA*
  - Particle refractive index - OPC+DMA*
  - Particle hygroscopicity - Humidified Tandem DMA*
- Single particle size, composition - SPLAT*
- Single particle size, composition, density, and hygroscopicity - SPLAT-MS/HTDMA*
- Aerosol size resolved composition - AMS
- Aerosol total soluble organic carbon - PILS-TOC*
- Aerosol ion content - PILS-IC*
- Elemental and Organic Carbon - Light spectrometer and thermal analysis*
- Soot photometer - SP2 by Droplet Measurement Technology
- Total aerosol mass - TEOM

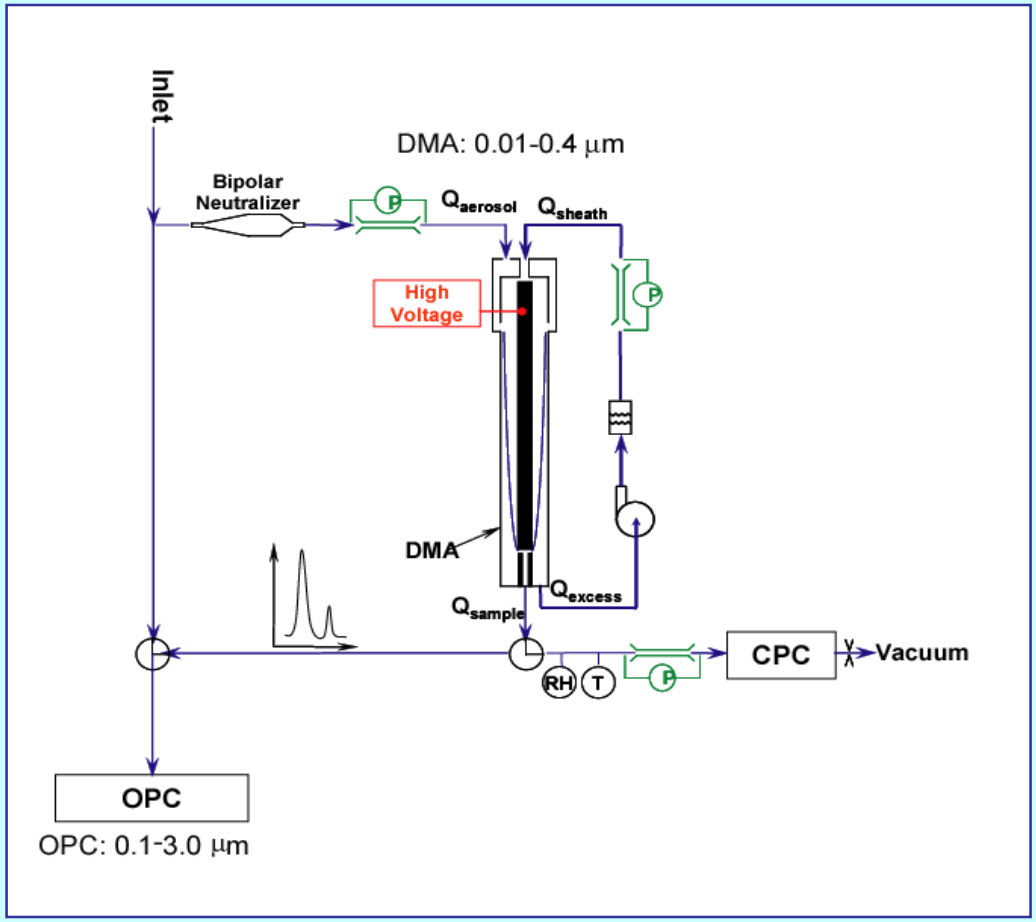
***One-of-a-kind system designed and constructed by ACP scientists**

Scanning Mobility Particle Sizer And Index of Refraction

(Jian Wang)

(1) Measure particle size distribution with high temporal resolution (60 s), using DMA to cover the 10 nm to 400 nm, and

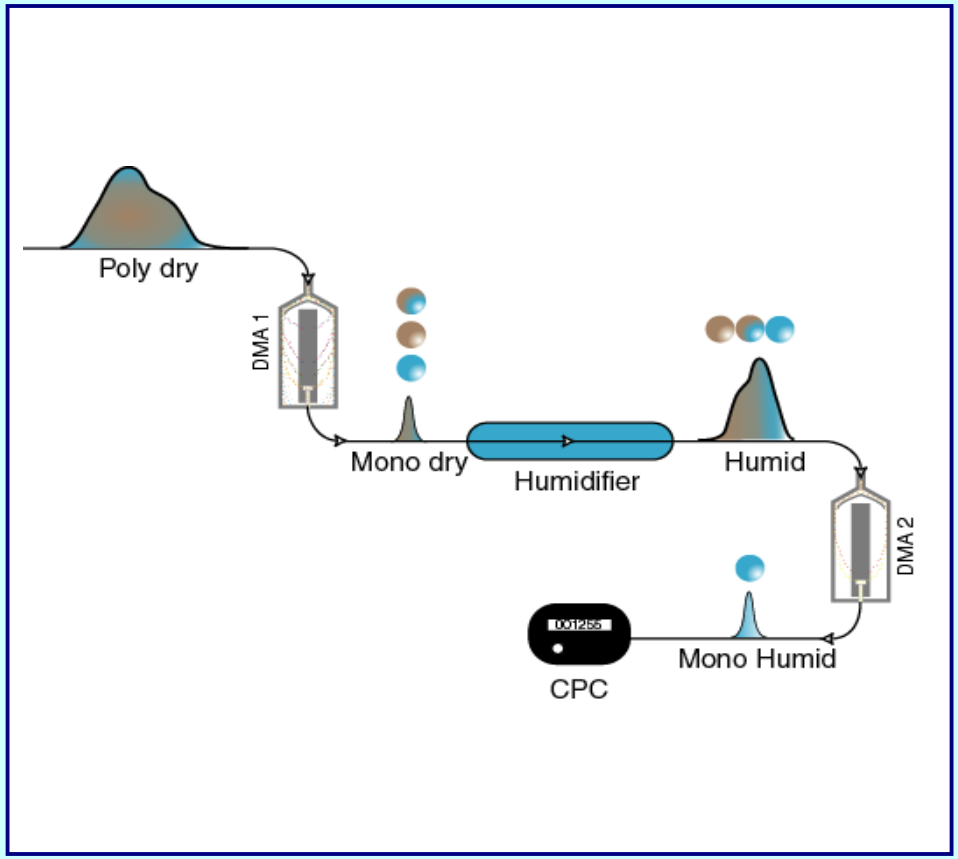
(2) In-situ calibration of OPC and refractive index of particle in the range of $0.1 < D_p < 0.4$ µm



HTDMA – Particle Hygroscopicity

(Jian Wang)

Measure particle water uptake in real-time. The Hydrated Tandem DMA system uses two identical DMAs and a humidity conditioner in-between.



PILS-IC:
Aerosol PM2.5 Ionic Composition
(Yin Nan Lee)

PILS-IC: (Particle Into Liquid Sampler - Ion Composition)

Rapid in-situ determination of aerosol chemical composition

Ionic Species:

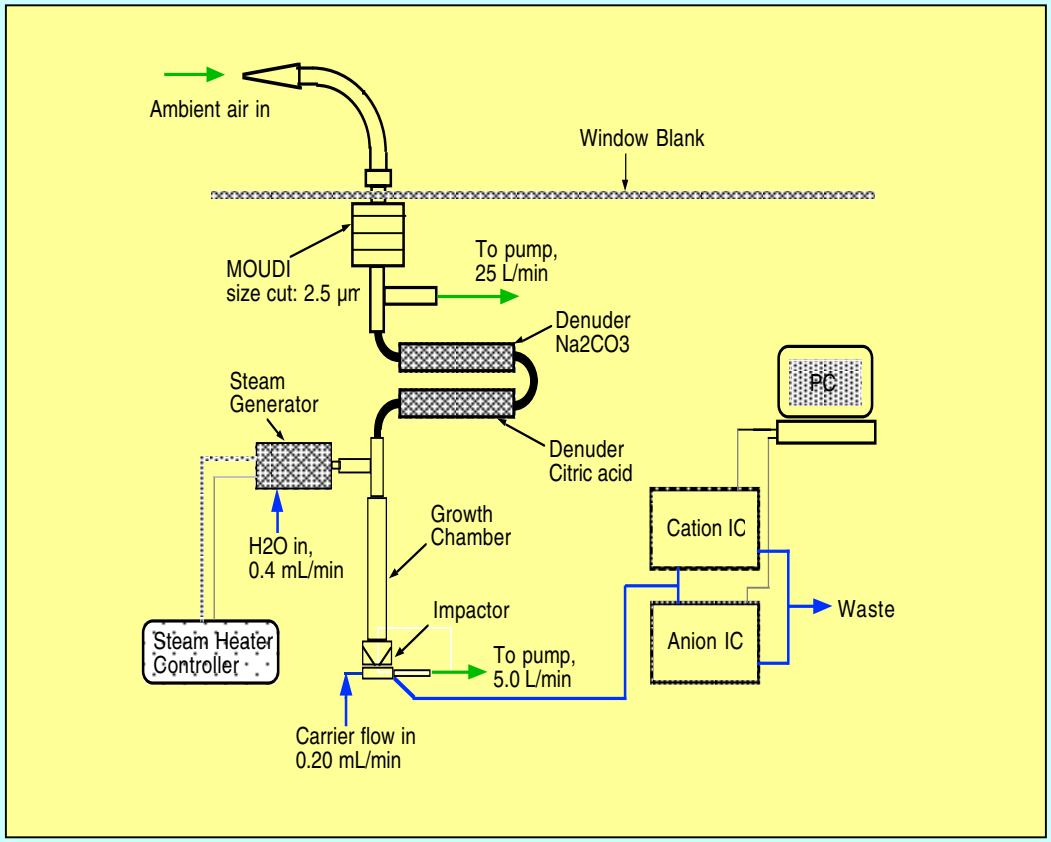
NH_4^+, NO_3^-, SO_4^{2-}, K^+, Ca^{2+}, Na^+, Mg^{2+}, Cl^-

Time response:

3 minutes

Sensitivity:

0.1 μg m^{-3}



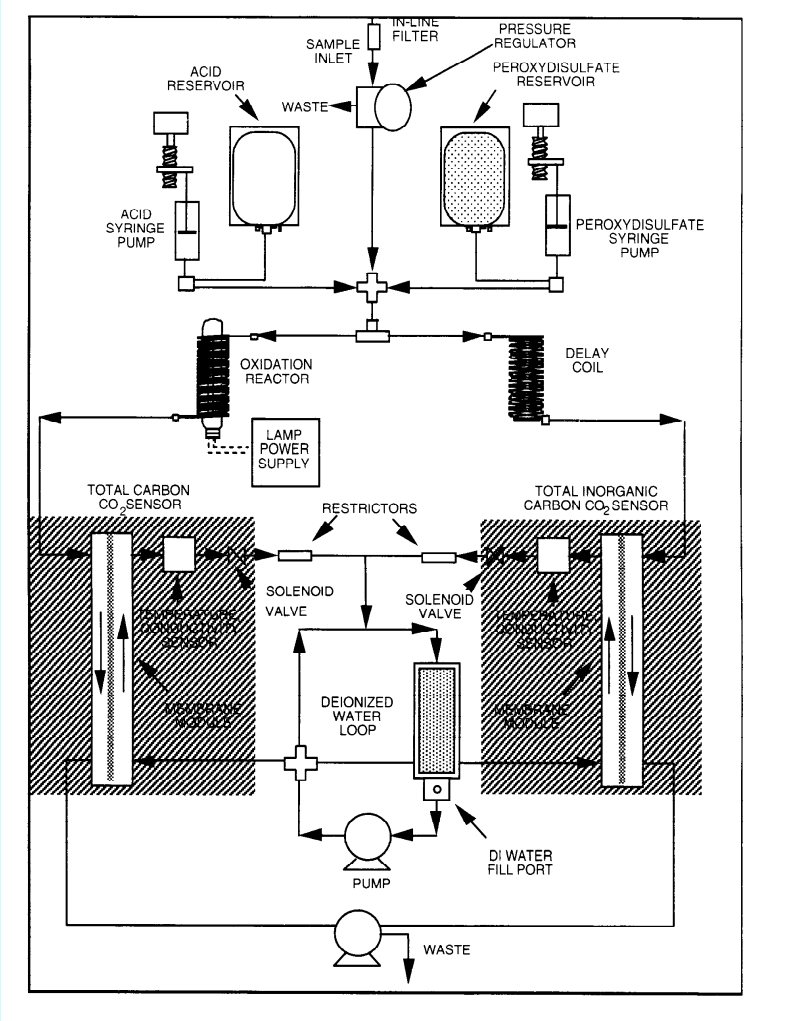
PILS-TOC:
Aerosol Total Organic Carbon Composition
(Yin Nan Lee)

PILS-TOC: (Particle Into Liquid Sampler Total Organic Carbon)

Rapid in-situ determination of aerosol total carbon composition

Time response:
1 minute

Sensitivity:
0.2 μg m^{-3}



SPLAT-MS:
Single Particle Size and Composition
(Dan Imre and Alla Imre)

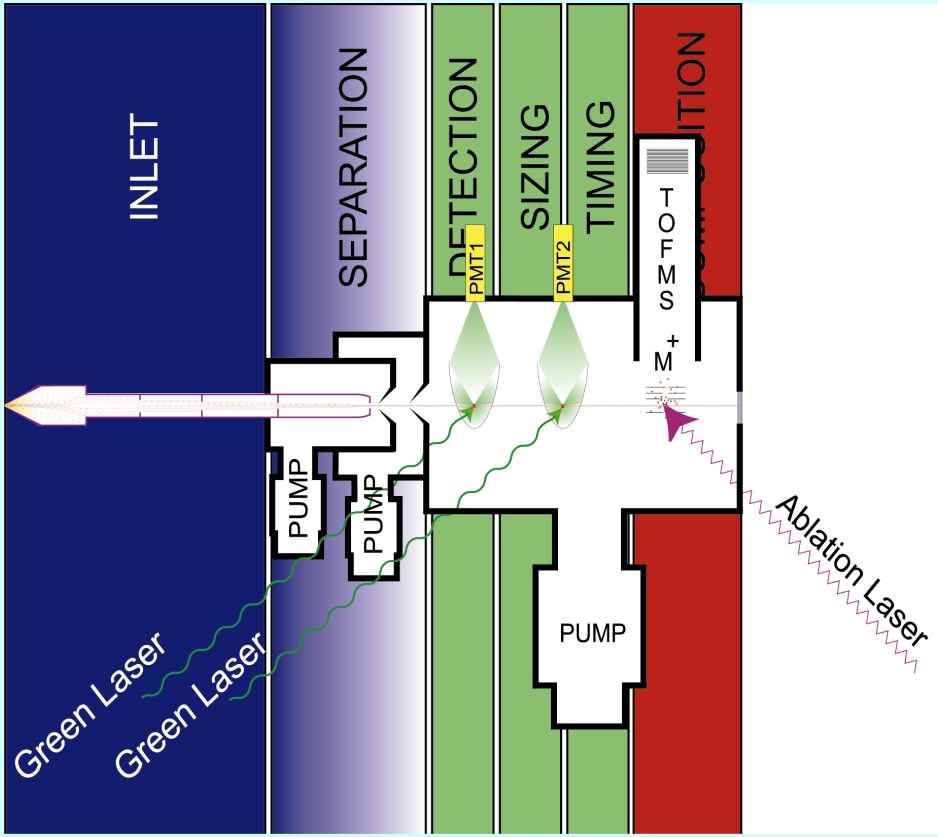
SPLAT-MS: Single Particle Laser Ablation Time of Flight Mass Spectrometer

Real-time determination of individual particle size and composition

Time resolution:
~20 particles s^{-1}

Size range: 50 nm to 3.5 μm

Size resolution: 3 nm



Single Particle Size, Composition, Density, and Hygroscopicity

(Dan Imre, Alla Imre, Jian Wang)

SPLAT-MS/HTDMA:

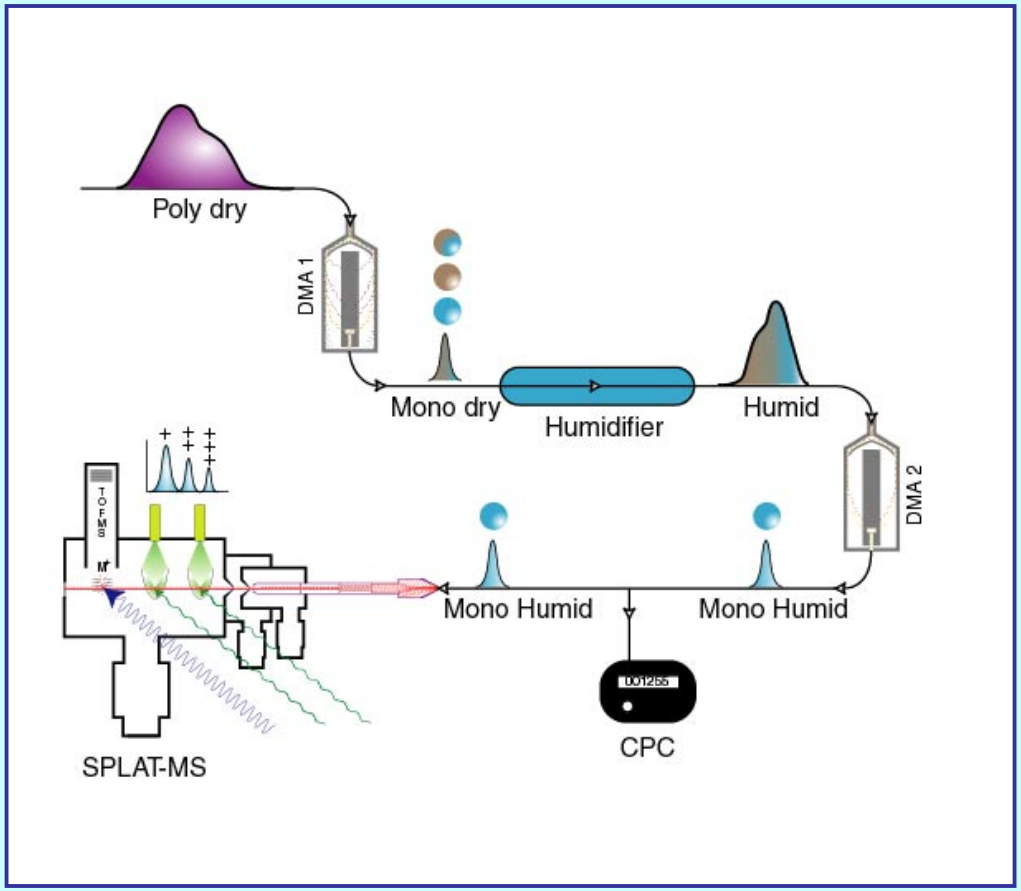
Real-time determination of individual particle size, composition, hygroscopicity, and density

Time resolution: ~20 particles s^{-1}

Size range: 100 nm to 1 µm

Size resolution: 3 nm

Density resolution: 0.05 g cm^{-3}



Aerosol Mass Spectrometer
(Aerodyne)

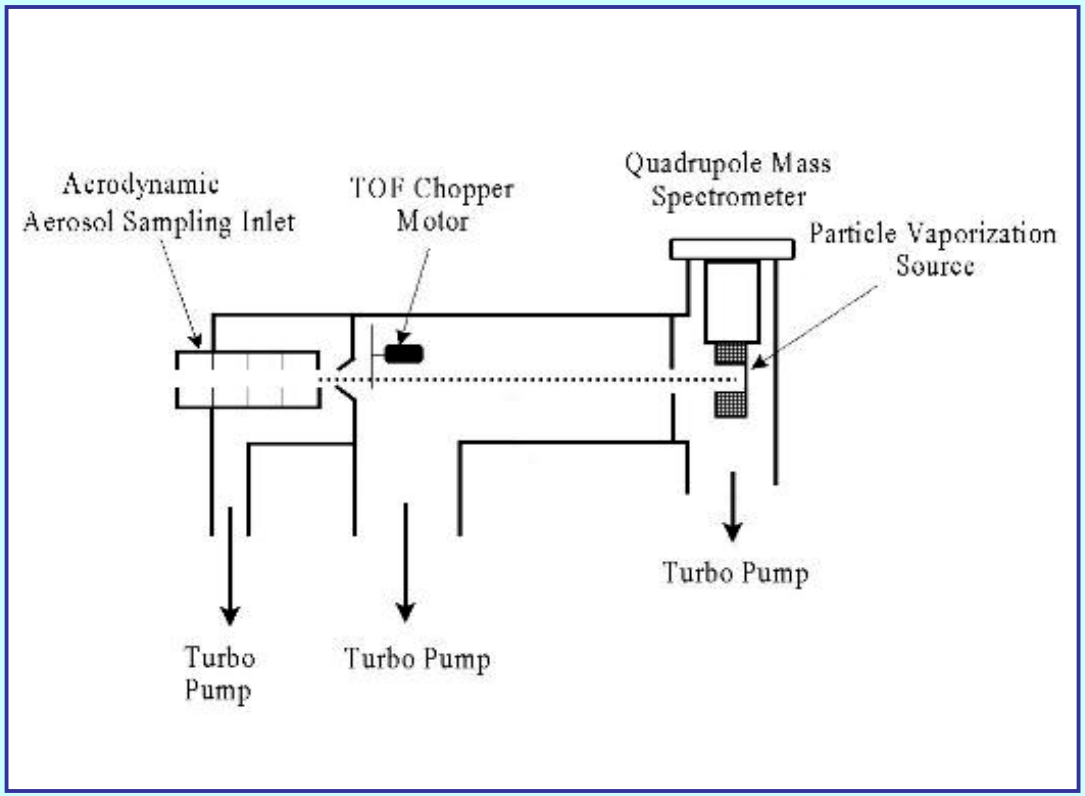
AMS

Real-time determination of aerosol size and composition

Time resolution: ~20 particles s^{-1}

Size range: 30 nm to 3 µm

Size resolution: 10 nm

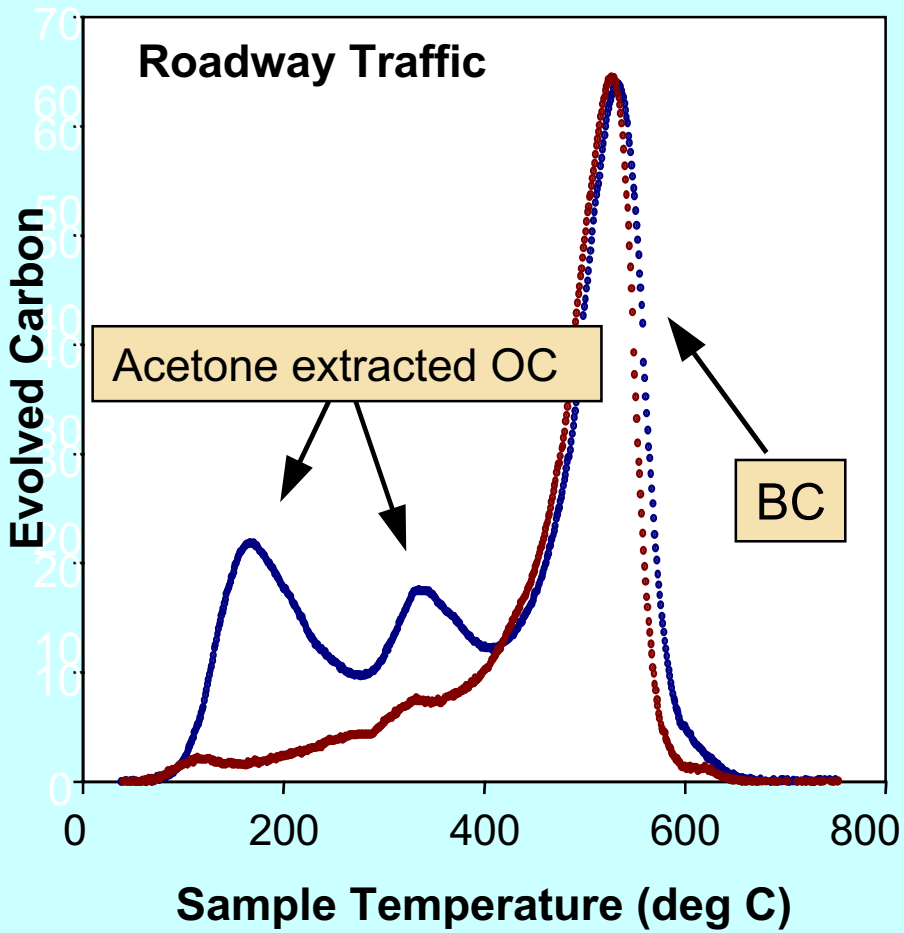


Single Particle Soot Photometer
(Droplet Measurement Tech., Inc.)

- Nd:YAG Intracavity laser induced particle incandescence
- Size determination by light scattering (soot and non soot particles)
- Highly specific for soot
- Single particle sensitivity down to 80 nm equivalent diameter
- Capable of analyzing 5000 particles cm^{-3}

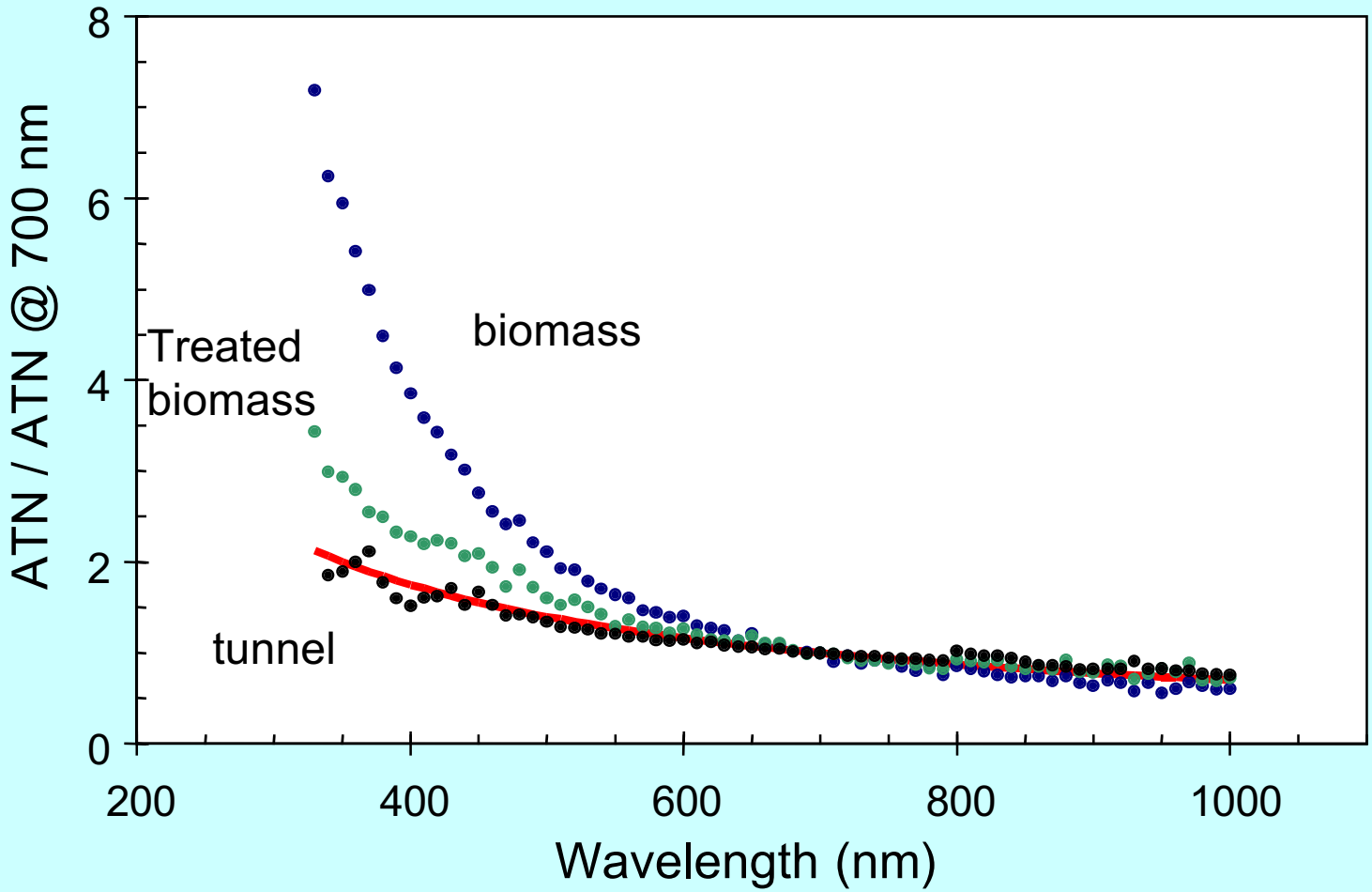
Thermal Analysis TC/OC/BC

(Tom Kirchstetter and Tica Novakov)



Thermal Analysis TC/OC/BC

(Tom Kirchstetter and Tica Novakov)



Closure of Aerosol Optical And Microphysical Properties (Bob McGraw)

APPROACH:

- *Calculate effective refractive index of each particle from its measured composition using mixing rules.*

  The difference in optical properties between a homogeneous vs. a core/shell particle is largest for mixtures involving black carbon, but even here differences are typically less than 5% (Chylek, 2002).

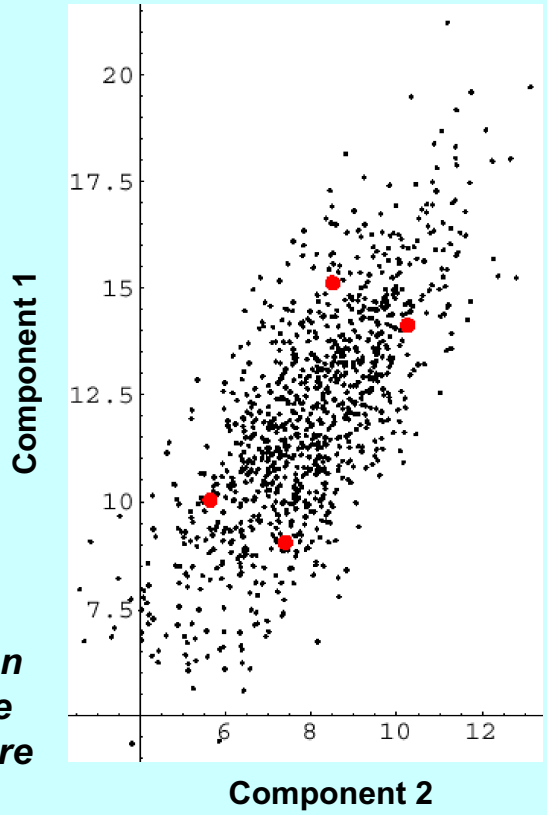
- *Compute aerosol optical properties through statistical analysis of ground-based single-particle composition and size measurements.*

- *Compare results with ground-based and air-to-ground optical measurements under well-mixed boundary layer conditions for closure (taking into account water uptake as a function of RH).*

Statistical Methods for Closure of Optical Properties from Measured Composition and Size Distribution (Bob McGraw and Yangang Liu)

Aerosol optical properties will be calculated from statistical properties of the measured particle size/composition distribution, including its ***multivariate mixed moments***, using advanced statistical methods developed in collaboration with the USB/AMS Dept.).

Optical properties of ***non-spherical particles*** will be computed at optimal surrogate compositions using *T*-matrix methods.



Example: Multivariate classification of a particle distribution using Principal Components Analysis (PCA). Red points are optimal surrogate compositions obtained by PCA-quadrature methods.

CONCLUSION

- The ARM-ACP IOP will provide the most complete aerosol chemical, microphysical, optical, and radiative characterization ever attempted.

- Only through the measurements conducted by ACP scientists will it be possible to understand and generalize the observed optical and radiative aerosol properties.

- It will establish a much needed relationship between two arms of OBER in which the sum of the two is much greater than the individual parts alone.