AEROSOLS AND ARM

Stephen E. Schwartz

On behalf of the ARM Aerosol Community
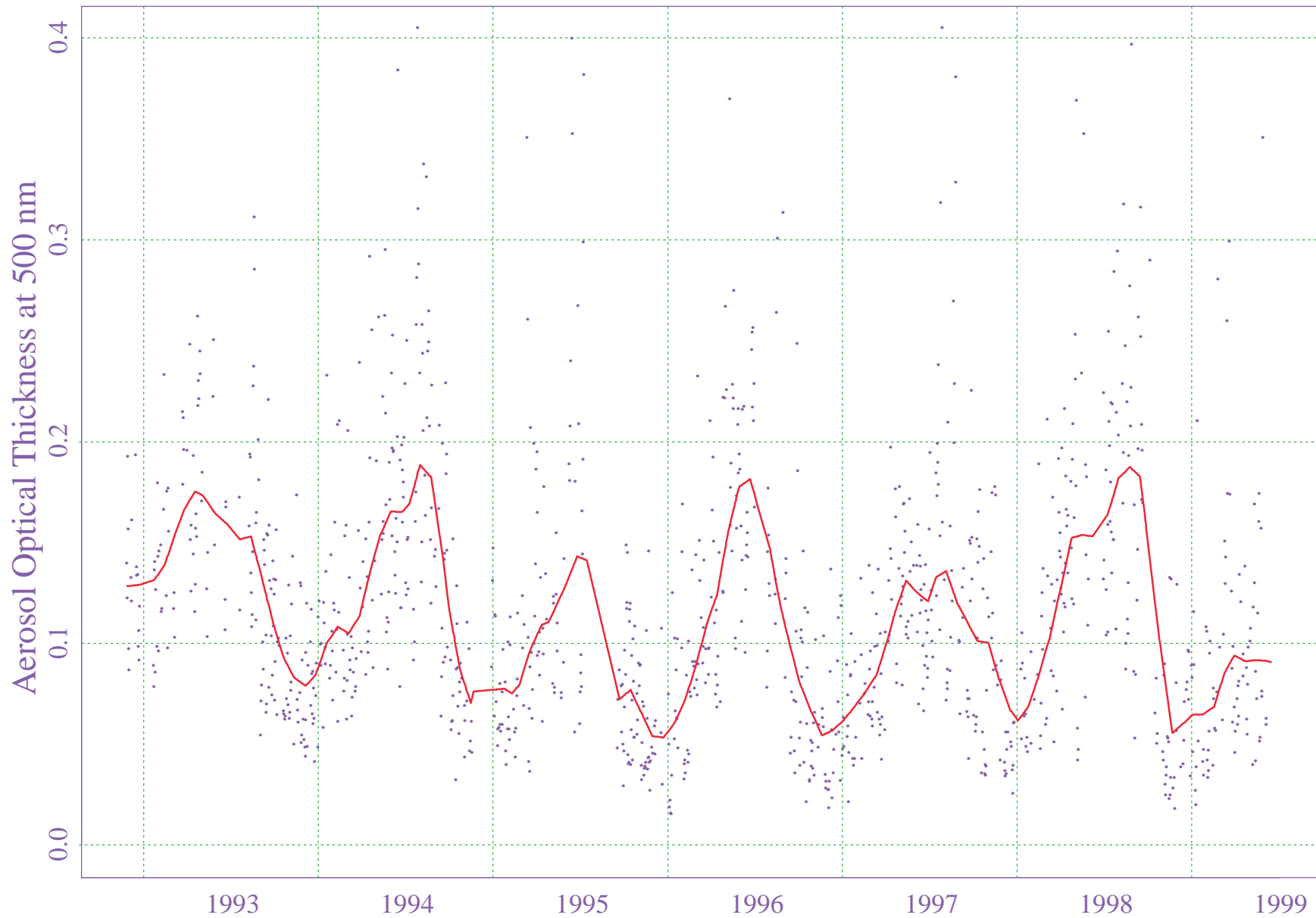
Importance

What have we learned (especially from IOPs)?

What should we be doing in the future?

"The Revenge of the Dirt Boys"

Aerosol Optical Thickness, North Central Oklahoma, 1993-1999
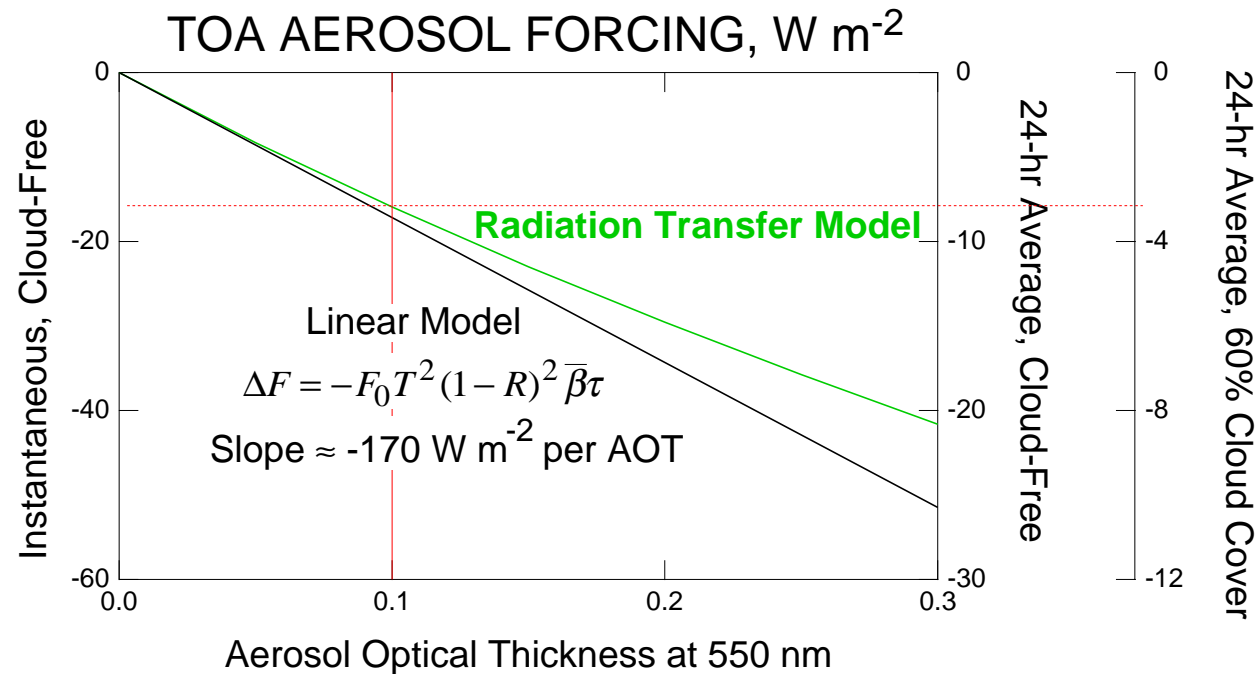Daytime average, DOE ARM Southern Great Plains site

J. Michalsky et al., JGR, to be submitted, 2000

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 ω_0 = 1.

TOA AEROSOL FORCING, W m^{-2}

Instantaneous, Cloud-Free

24-hr Average, Cloud-Free

24-hr Average, 60% Cloud Cover

Radiation Transfer Model

Linear Model

$$\Delta F = -F_0 T^2 (1-R)^2 \, \overline{\beta} \tau$$

Slope $\approx$ -170 W m^{-2} per AOT

Aerosol Optical Thickness at 550 nm

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

AEROSOL IOPs AT SGP

Date	In Conjunction with
April 1997	Cloud Radar IOP
September 15 - October 5, 1997	Integrated Fall IOP
August 3 - 28, 1998	Shortwave IOP

Vertical aircraft profiles, typically to 5-6 km.

Instrument package typically consists of:

Nephelometer (aerosol light scattering coefficient, 3 wavelengths, backscatter)

Particle absorption photometer

Optical particle counter (nominally number *vs*. size, 0.2 - 2 µm diameter)

Forward scattering probe (number *vs*. size, 2 - 20 µm diameter)

Meteorological state parameters (temperature, humidity, ...)

KEY STUDIES

Investigators	Topic
Harrison *et al.*	Spectral Measurement Diffuse-Direct Ratio
Mlawer, Clough *et al.*	Spectral Model Diffuse-Direct Ratio
Daum, Liu	Size distribution - Nephelometer Closure
Schwartz, Bergin *et al.*	Aerosol Optical Thickness Closure
Kato *et al.*	Aerosol Optical Thickness Closure
Schwartz, Halthore	Aerosol Surface Forcing
Daum, Liu	Dispersion & Effective Radius of Cloud Drops

SPECTRAL MEASUREMENTS OF DIFFUSE/DIRECT RATIOS BY ROTATING SHADOWBAND RADIOMETER

Lee Harrison, Mark Beauharnois, Jerry Berndt, Peter Kiedron, Joseph Michalsky, and Quilong Min, SUNY Albany

Questions-

How accurately do atmospheric radiation transfer models represent the spectral diffuse irradiance in cloud-free sky?

Do spectral diffuse irradiance measurements identify the wavelength region of the "clear-sky diffuse anomaly"?

Approach-

Examination of Direct/Diffuse Ratio removes dependence on absolute calibration and absolute solar spectrum.

Model depends on assumed or measured aerosol properties.

To date modeling has used "standard" aerosol types rather than aerosol properties measured coincidentally with the photometry.
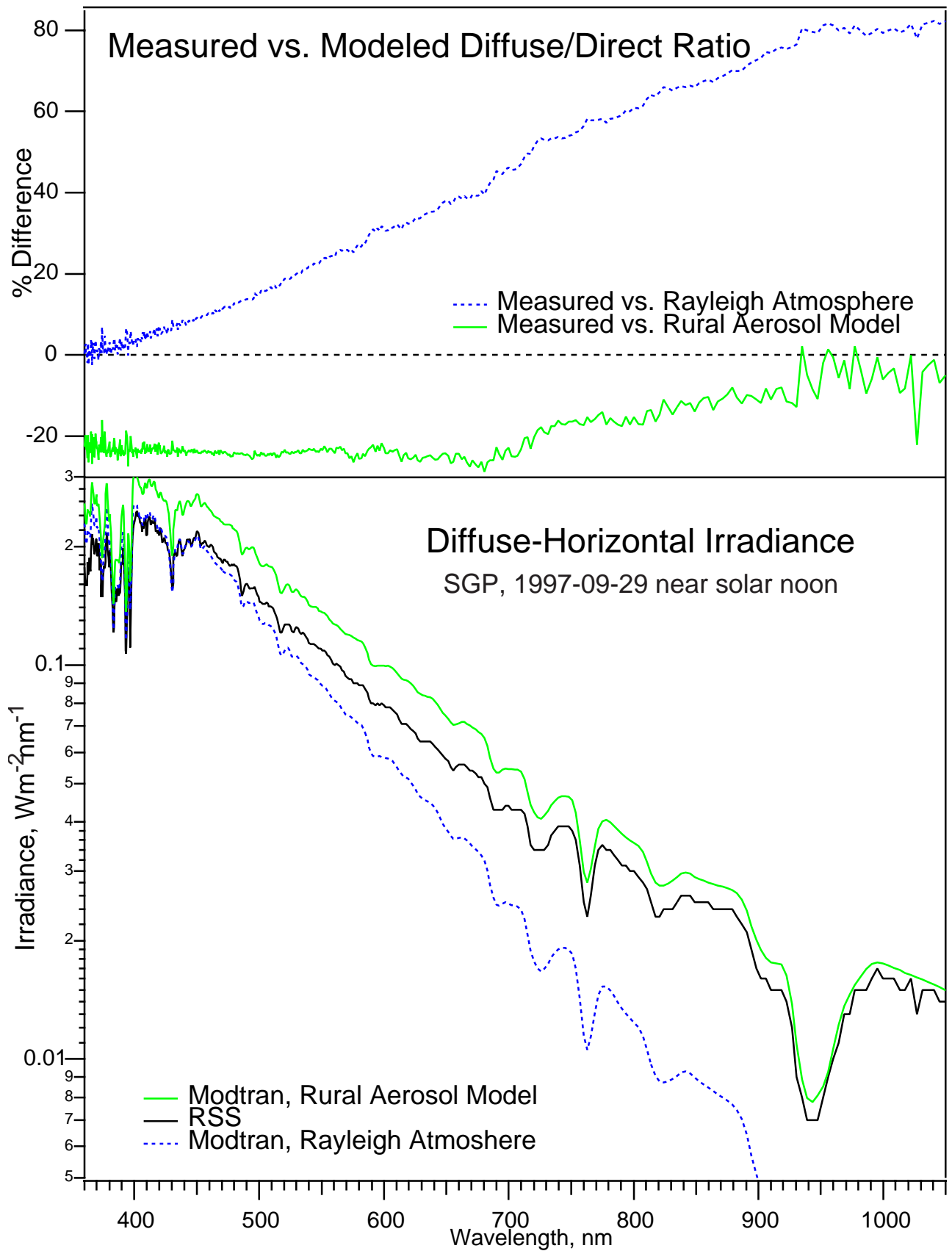
Measurements-

Morning Langley Optical Depth Spectrum, SGP, Nov. 2 1999
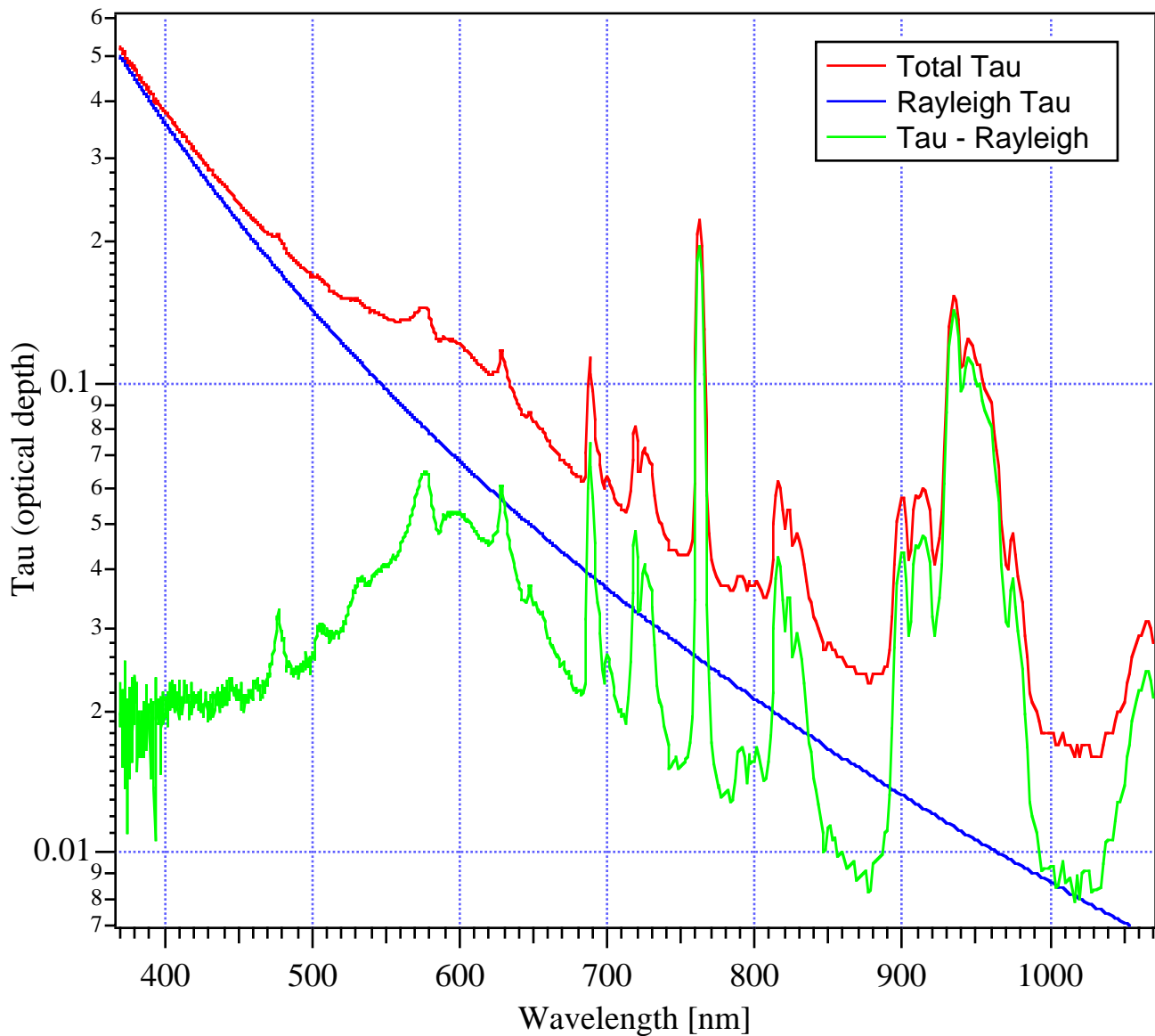
- Extremely clear and cloud-free day (confirmed by all-sky camera).

- Chappuis O_3 is not removed from the green line.

- Aerosol optical depth at 500 nm is ~ 0.015.

Measured vs. Modeled Diffuse/Direct Ratio

- Measured vs. Rayleigh Atmosphere
- Measured vs. Rural Aerosol Model

Diffuse-Horizontal Irradiance

SGP, 1997-09-29 near solar noon

- Modtran, Rural Aerosol Model
- RSS
- Modtran, Rayleigh Atmoshere

Conclusions-

The departure of measurement and model looks like a modest "clear sky anomaly" as suggested by Arking, Kato *et al.*, and Halthore *et al.*

Integration over the spectrum yields a deficit of ~12 W m^{-2} in the diffuse irradiance from the model calculation using the rural aerosol parameterization.

The spectral signature of the "diffuse anomaly" is bland and increases to shorter wavelengths. This is strongly suggestive of an aerosol, not a gas as hypothesized by Halthore *et al.*

Measurements of aerosol optical properties aloft would go a long way to settle this issue, but are hindered by the rarity of suitable skies, and the difficulty of measuring the aerosol single-scattering albedo.

L. Harrison, M. Beauharnois, J. Berndt, P. Kiedron,
J.J. Michalsky, and Q. Min, GRL, 1999

COMPARISON OF SPECTRAL DIRECT AND DIFFUSE SOLAR IRRADIANCE MEASUREMENTS AND CALCULATIONS FOR CLOUD-FREE CONDITIONS

Eli Mlawer, Patrick Brown, Shepard Clough, AER

Lee Harrison, Joseph Michalsky, and Piotr Kiedron, SUNY Albany

Tim Shippert, PNNL

Questions-

- How accurately does a line-by-line radiation transfer model represent the spectral direct and diffuse irradiance in cloud-free sky?

- Do comparisons of measured and modeled direct and diffuse irradiance shed light on the nature and origin of unmodeled atmospheric absorption?

Approach-

- Compare model with absolutely calibrated spectral radiometric measurements.

- Model assumes a particular solar spectrum.

- Model depends on assumed or measured aerosol properties.

Results-

MEASURED AND MODELED DIRECT AND DIFFUSE SURFACE IRRADIANCE

SGP 1997-09-18

4.6 Airmasses, PW = 4.2 cm, AOT(700 nm) = 0.375, ω_0 = 0.85
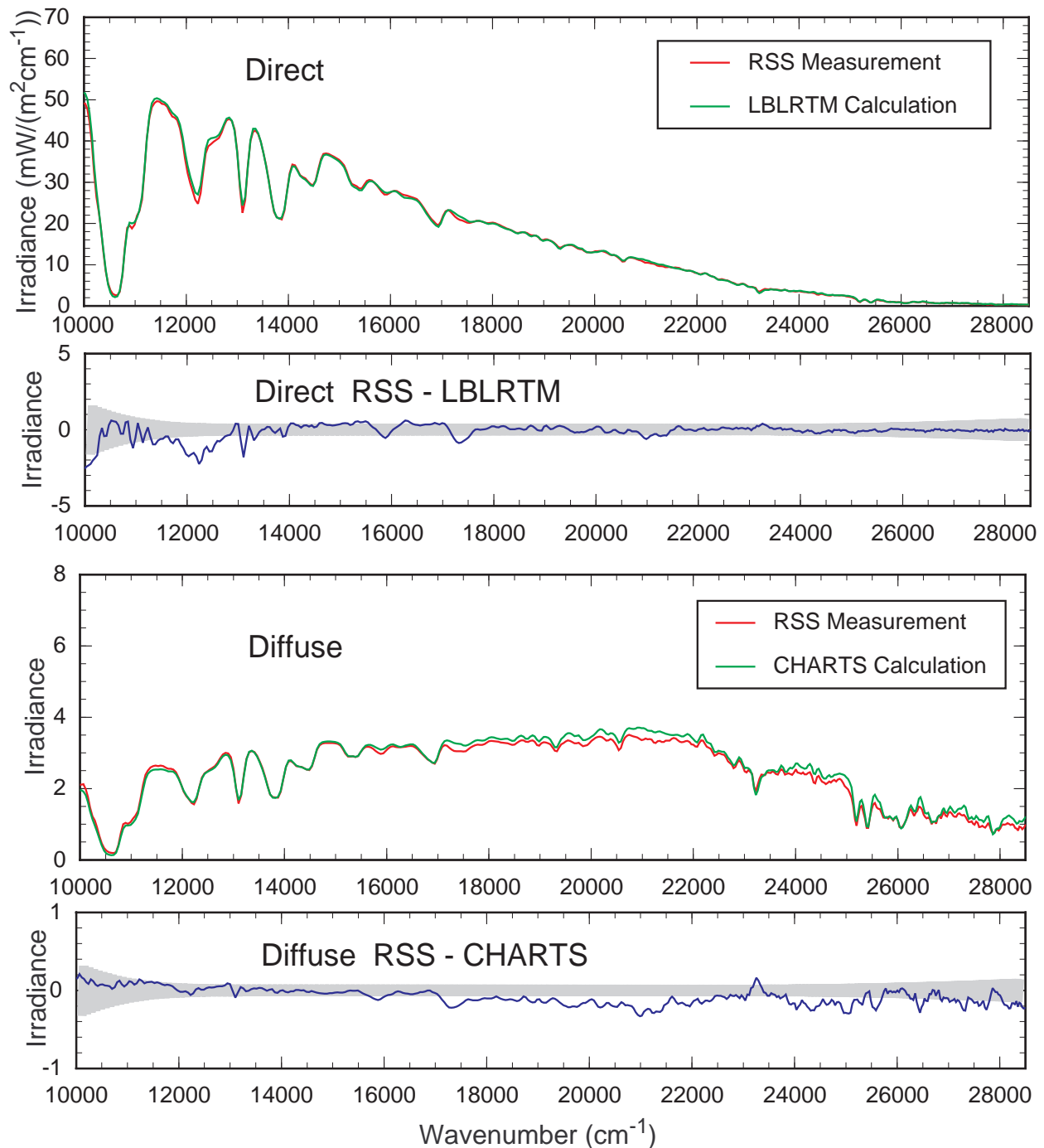
Conclusions-

- Comparisons between direct and diffuse RSS measurements and calculations provide persuasive evidence against unknown molecular absorption of significance in the spectral range of the RSS.

- State-of-the-art radiative transfer models accurately account for atmospheric absorption between 550-28500 cm^{-1} (18.2-0.35 µm).

- The most likely cause of the unexplained discrepancies between measurements and calculations reported previously [Kato *et al.*, Pilewskie *et al.*, Halthore and Schwartz] is the use of aerosol single-scattering albedos that are too large

- The values of single-scattering albedo used in this work (0.60 - 0.85) are lower than usually assumed, and, if generally valid would represent a substantial source of unmodeled atmospheric absorption.

E. J. Mlawer, P. D. Brown, S. A. Clough, L. C. Harrison, J. J. Michalsky, P. W. Kiedron, T. Shippert, GRL, submitted (2000)

SIZE DISTRIBUTIONS AND LIGHT SCATTERING COEFFICIENTS DERIVED FROM OPTICAL PARTICLE COUNTERS

Peter Daum and Yangang Liu, Brookhaven

A closure experiment-

Does the light measured aerosol scattering coefficient equal that calculated by integration over the size distribution?

$$\sigma_{\text{sp}} \overset{?}{=} \int \pi r^2 Q_{\text{S}}(r,m) \frac{dn}{dr} dr$$

Issue-

- Light scattering coefficients measured directly using an integrating nephelometer during the Fall and Spring 1997 Aerosol IOPs exceed by a factor of two those calculated from measurements of number concentrations and size distributions.
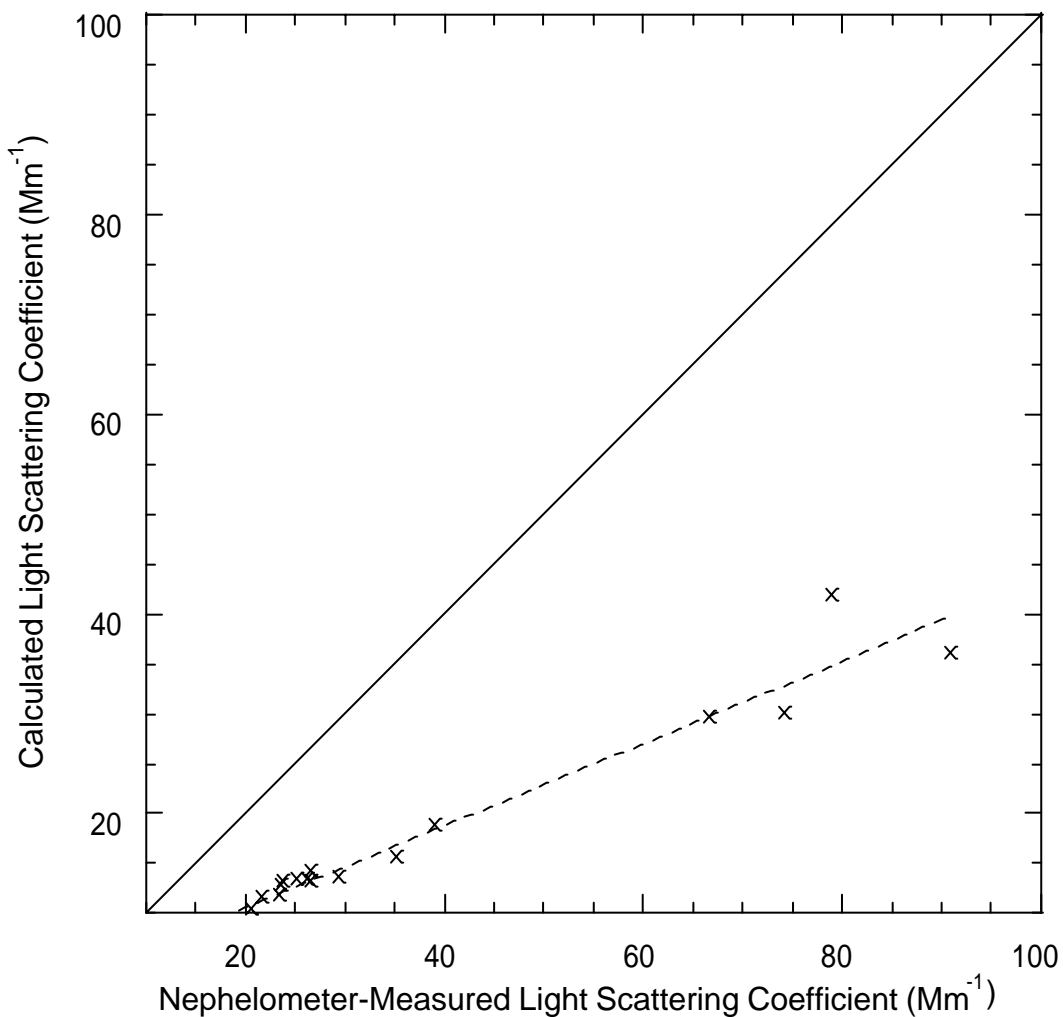
- This discrepancy is found by other investigators using optical particle counters, but not with non-optical particle sizing instruments.

Possible explanation-

- Optical particle counters typically undersize ambient particles because they are calibrated with particles having a refractive index much larger than ambient particles.

- Calculated light scattering coefficient is sensitive to refractive index.
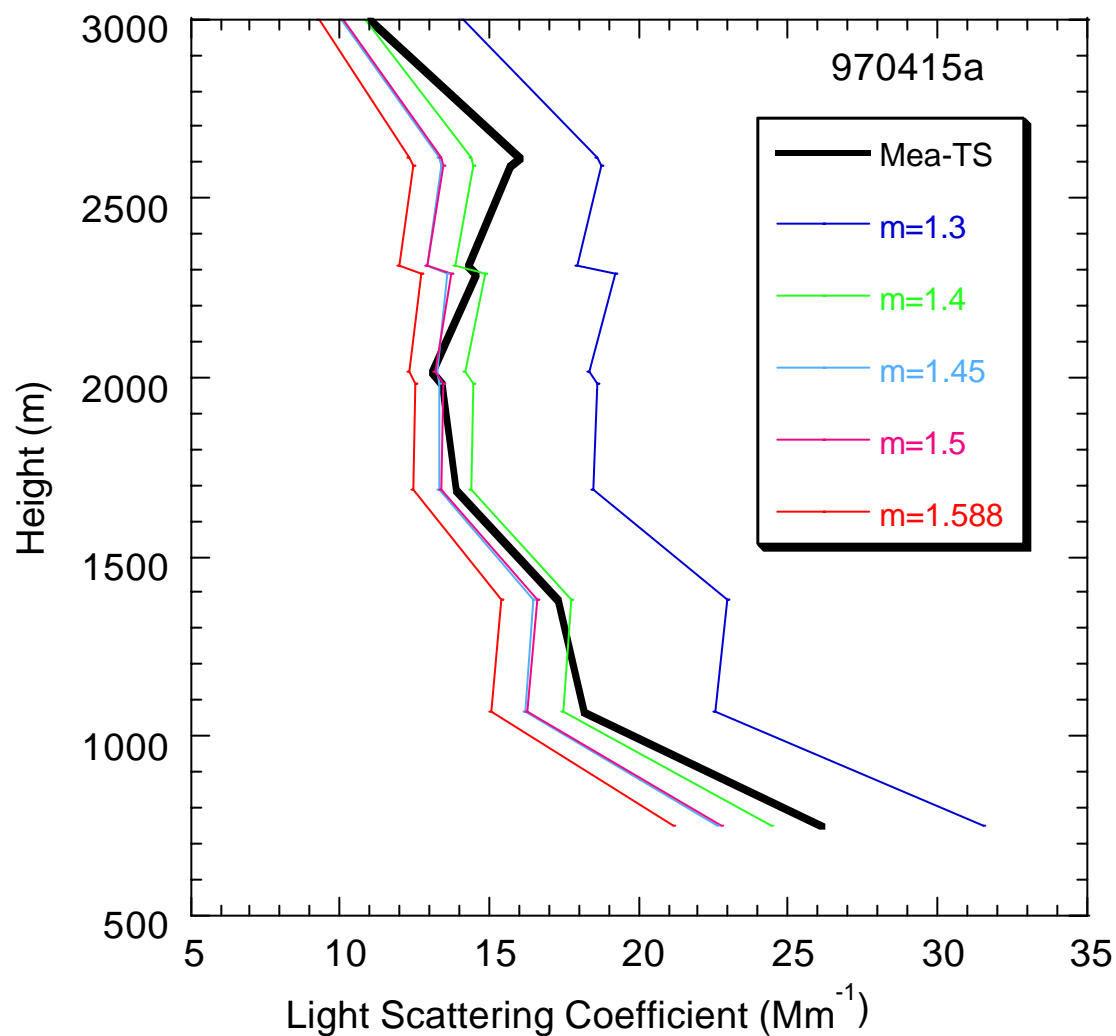
Approach-

- Derive a formulation for the effect of refractive index on the response of optical particle counters.

- Use the formulation to derive an algorithm for correcting size distributions for the difference between the refractive index of calibration and measured particles.

- Calculated scattering coefficients using corrected size distributions.

- Compared measured and calculated scattering coefficients.

Results-
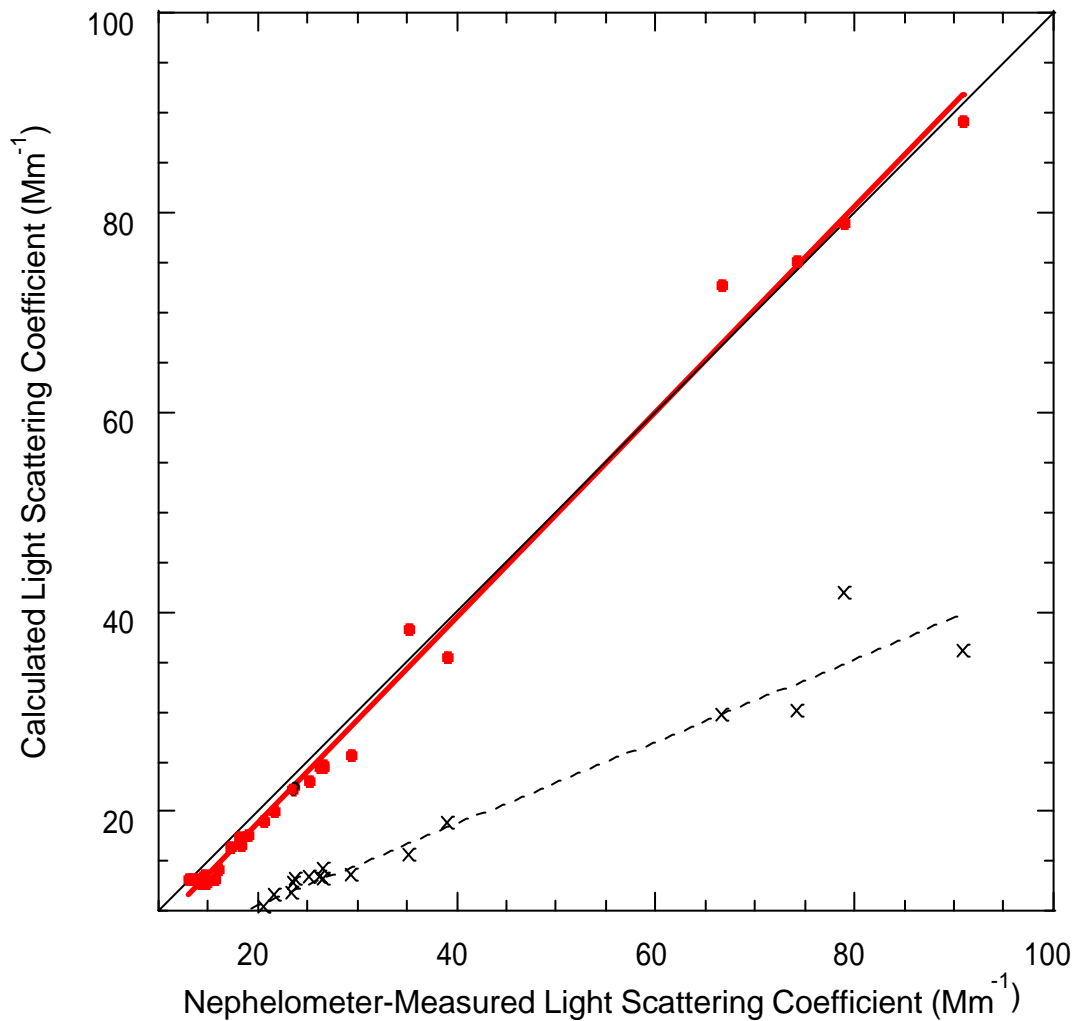
This approach permits confident determination of size distribution and refractive index from optical particle counter data.

Y. Liu and P. H. Daum, J. Aerosol Sci., in press, (2000).

COMPARISON OF AEROSOL OPTICAL DEPTH INFERRED FROM SURFACE MEASUREMENTS AND SUN PHOTOMETRY

Stephen Schwartz, Rangasayi Halthore, Brookhaven
Michael Bergin, NOAA CMDL, Brookhaven
John Ogren, NOAA CMDL
Dennis Hlavka, NASA/GSFC, Science Systems & Applications, Inc.

A closure experiment-

- Does the aerosol optical depth determined from surface aerosol properties and aerosol vertical profile equal that measured by sun photometry?

$$\tau \stackrel{?}{=} \int \sigma_{\text{ext}} dz$$

- Is the mixed layer height a good surrogate for the vertical distribution?

$$\tau \stackrel{?}{=} \sigma_{\text{ext}}(\text{sfc}) H_{\text{ML}}$$

Approach-

- Determine aerosol optical depth by sunphotometry.

- Determine aerosol vertical profile by micropulse Lidar backscatter.

- Measure aerosol scattering and absorption coefficients at surface.

- Calculate aerosol optical depth by integral over vertical profile.

Issues-

- Vertical distribution of aerosol.
- Representativeness of surface aerosol properties of aerosol in vertical column.
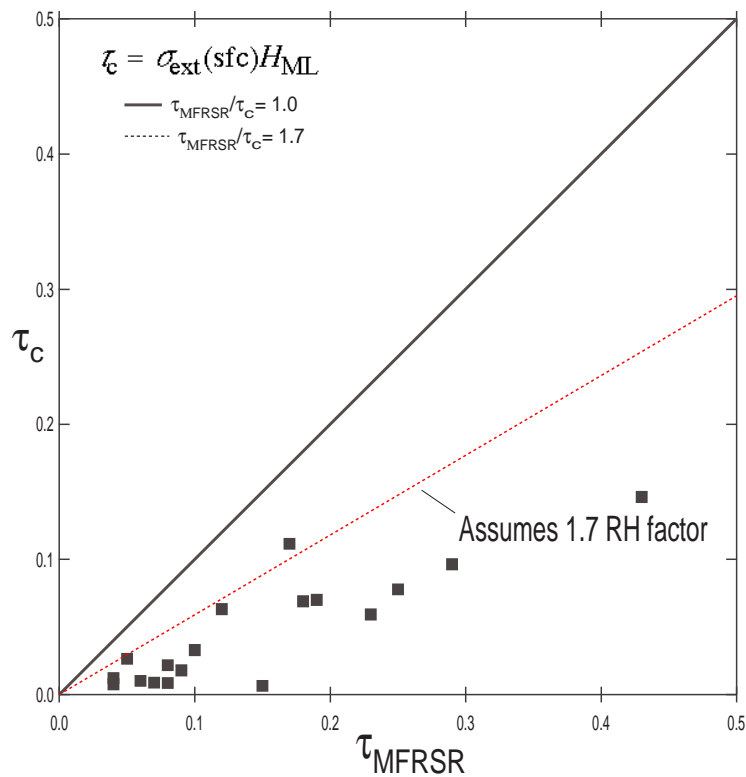- Relative humidity profile and growth of aerosol as function of height.

Observations-

Aerosol Lidar Backscatter on Cloud-Free Days at SGP
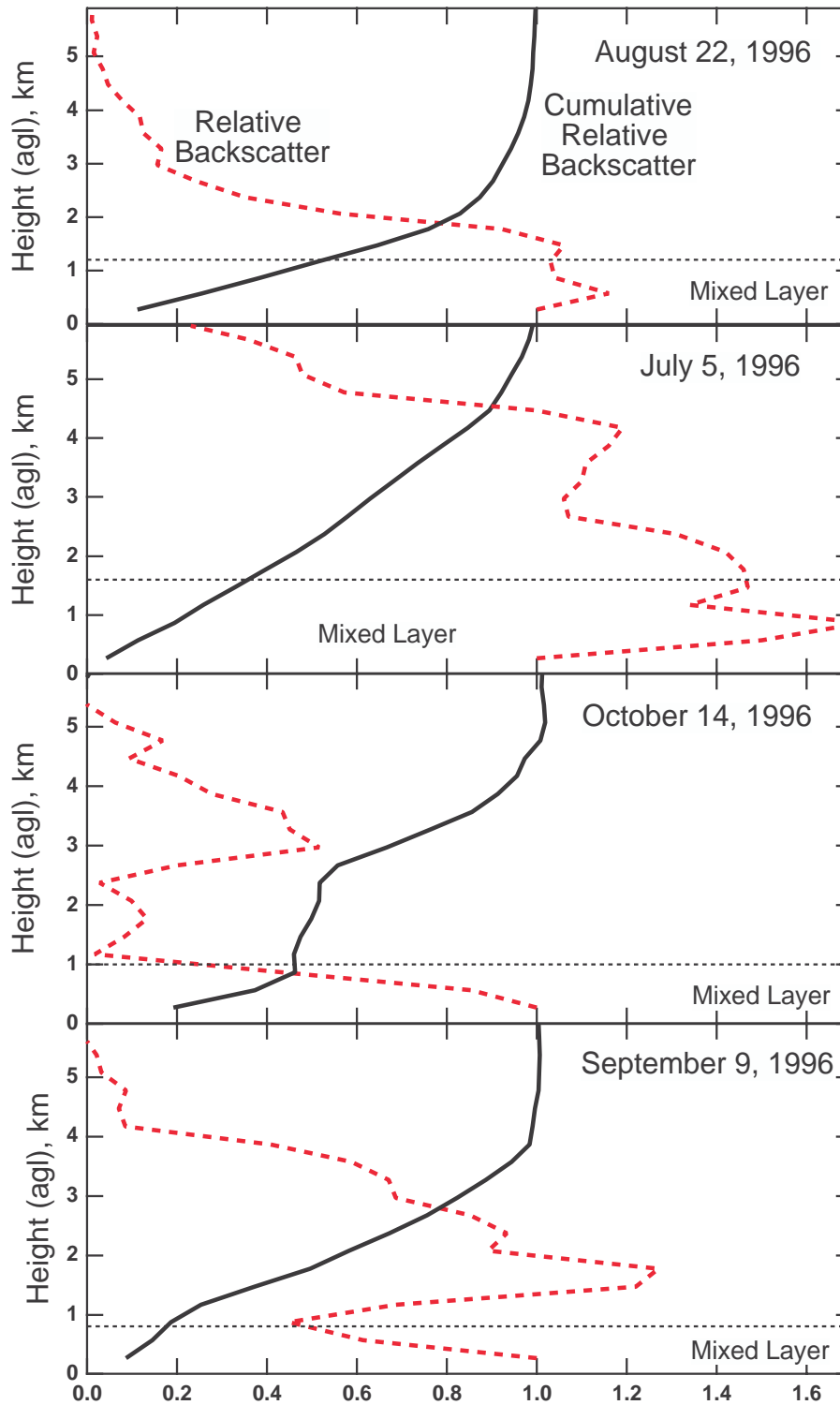
Correlation of Aerosol Optical Thickness and Surface Extinction Coefficient

$r^2 = 0.55$

slope = 2982 m $\pm$ 593 m

τ_{MFRSR}

$\sigma_{ep,dry}(\text{Mm}^{-1})$
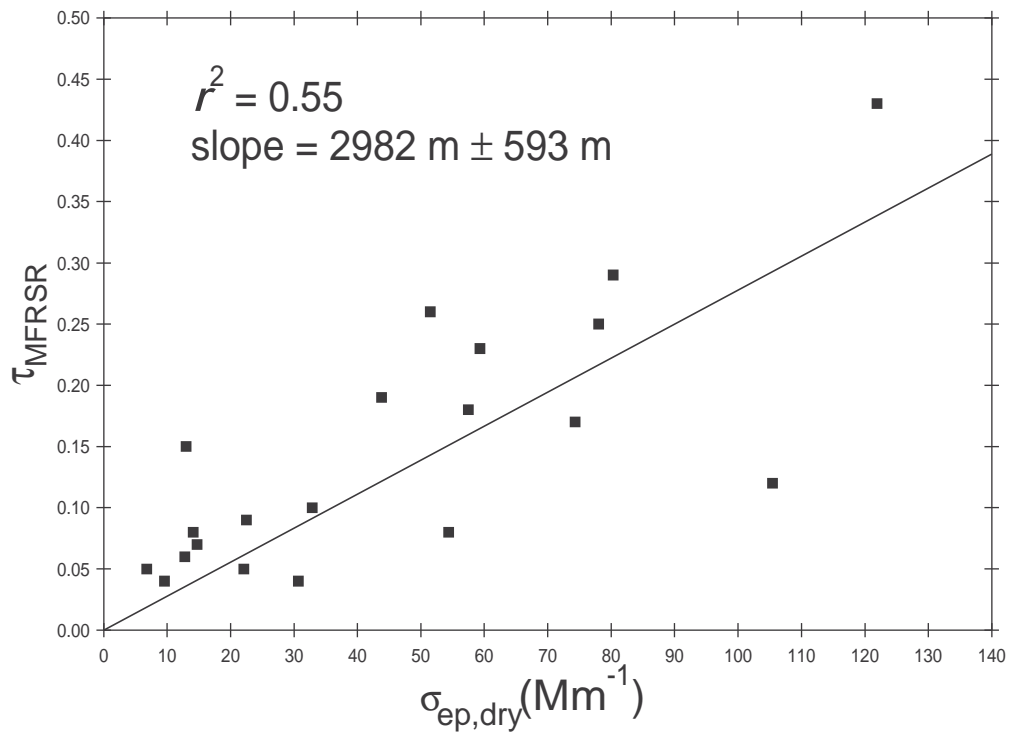
Aerosol Optical Thickness based on Surface Extinction and Mixed Layer Height

$\tau_c = \sigma_{ext}(\text{sfc})H_{ML}$

—— $\tau_{MFRSR}/\tau_c = 1.0$

········ $\tau_{MFRSR}/\tau_c = 1.7$

Assumes 1.7 RH factor

τ_c

τ_{MFRSR}

Aerosol Optical Thickness based on Surface Extinction and Aerosol Profiles
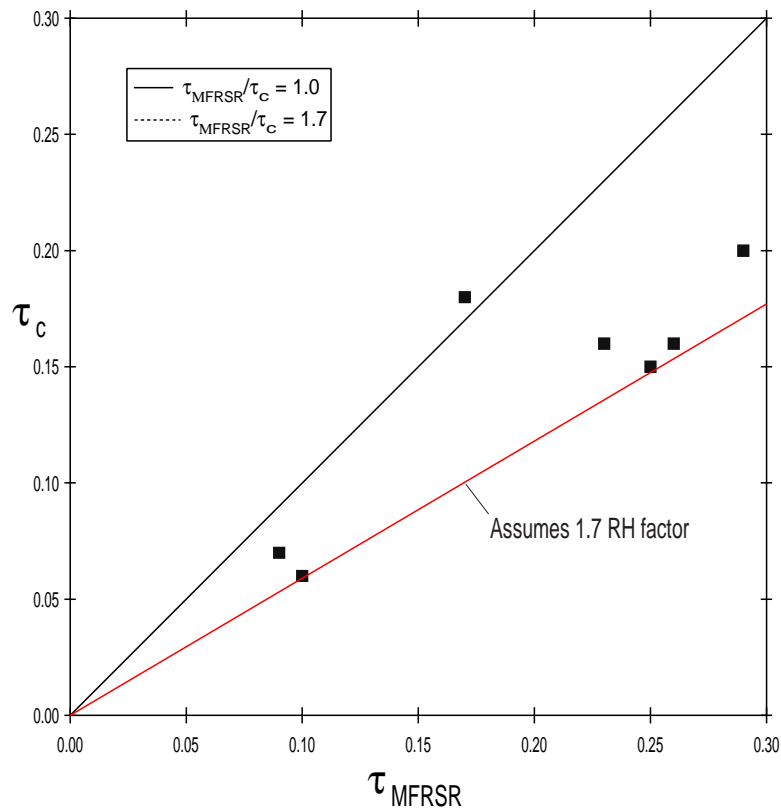
Conclusions-

- Much of the aerosol extinction is above the mixed layer.

- Properties of the vertically distributed aerosol are requires to accurately obtain closure in aerosol optical depth.

M. H. Bergin, S. E. Schwartz, R. N Halthore, J. A. Ogren, and D. L. Hlavka, JGR , in press, (2000).

COMPARISON OF THE AEROSOL THICKNESS DERIVED FROM GROUND-BASED AND AIRBORNE MEASUREMENTS

Seiji Kato, Hampton U. and NASA Langley

Michael Bergin, Georgia Tech

Thomas Ackerman, Nels Laulainen, David Turner, PNNL

Thomas Charlock, Richard Ferare, NASA Langley

Eugene Clothiaux, Penn State

Rangasayi Halthore, Brookhaven

Gerald Mace, Univ. Utah

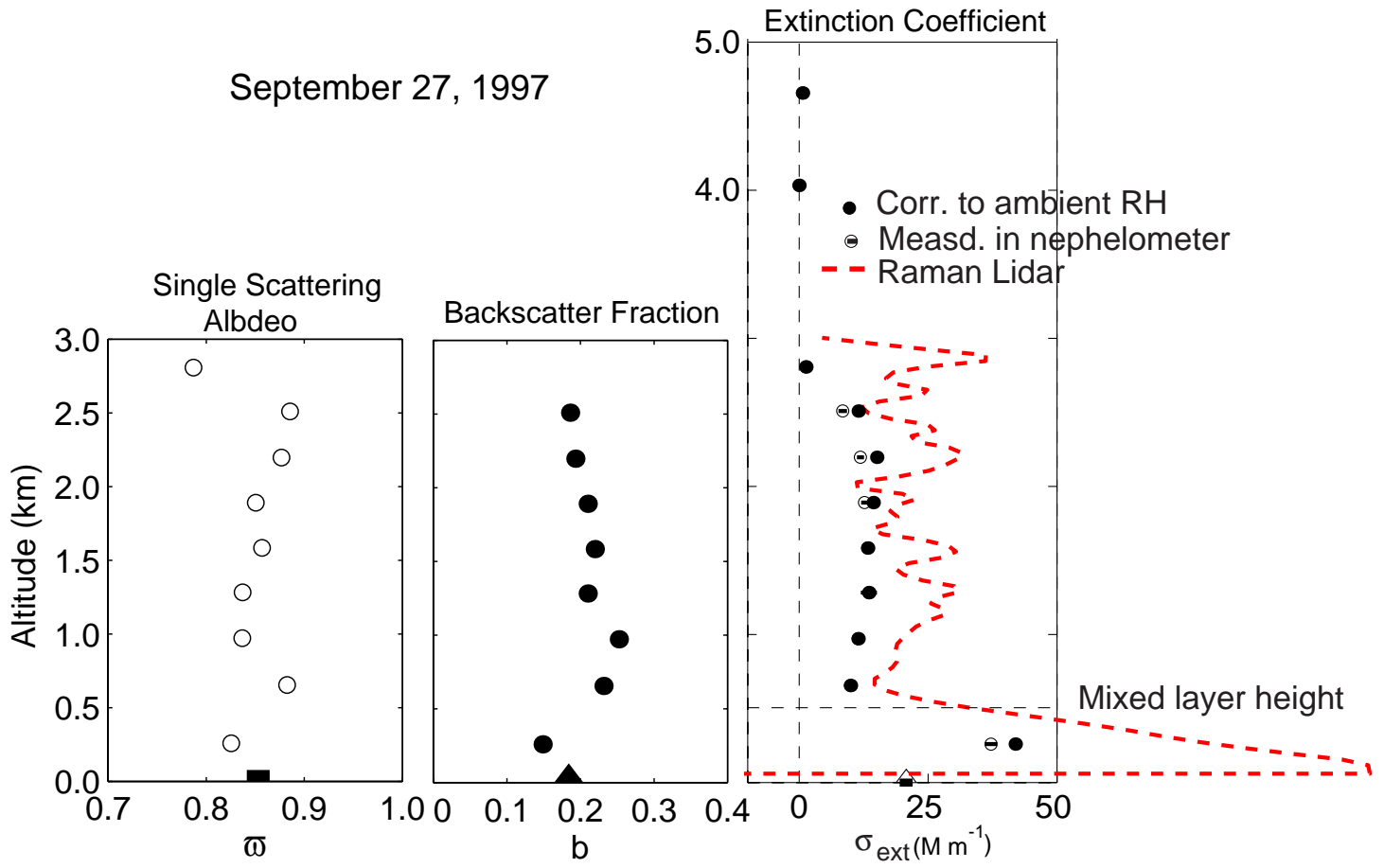Joseph Michalsky, SUNY Albany

A closure experiment-

- Does aerosol optical depth determined as the vertical integral of aerosol scattering and extinction equal that measured by sun photometry?

$$\tau_{\mathrm{p}}(\lambda, \mathrm{RH}) \overset{?}{=}$$

$$\int_{\mathrm{sfc}}^{5\mathrm{km}} [\sigma_{\mathrm{sp}}(\lambda, z) f_{\mathrm{s}}(\lambda, \mathrm{RH}) + \sigma_{\mathrm{ap}}(\lambda, z) f_{\mathrm{a}}(\lambda, \mathrm{RH})] dz$$

Measurements-

Aerosol Properties

September 27, 1997

Vertical Profile of RH

September 27, 1997

Legend:
- Sounding
- Raman Lidar
- Ambient (in-situ)
- In Nephelometer

Altitude (km) vs. Relative Humidity (%)
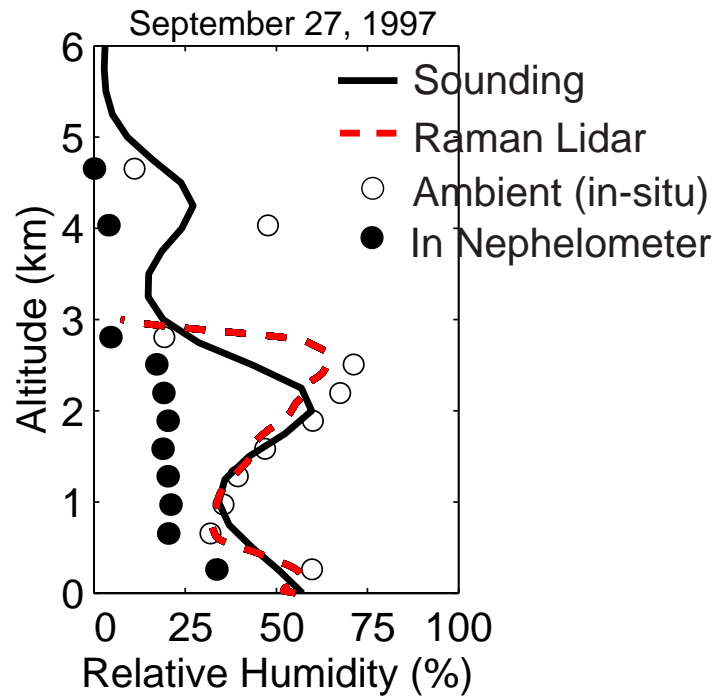
RH Dependence of Scattering coefficient (Surface measurements at SGP)

450 nm

$\sigma_{sp} / \sigma_{sp,ref}$ vs. Relative Humidity (%)
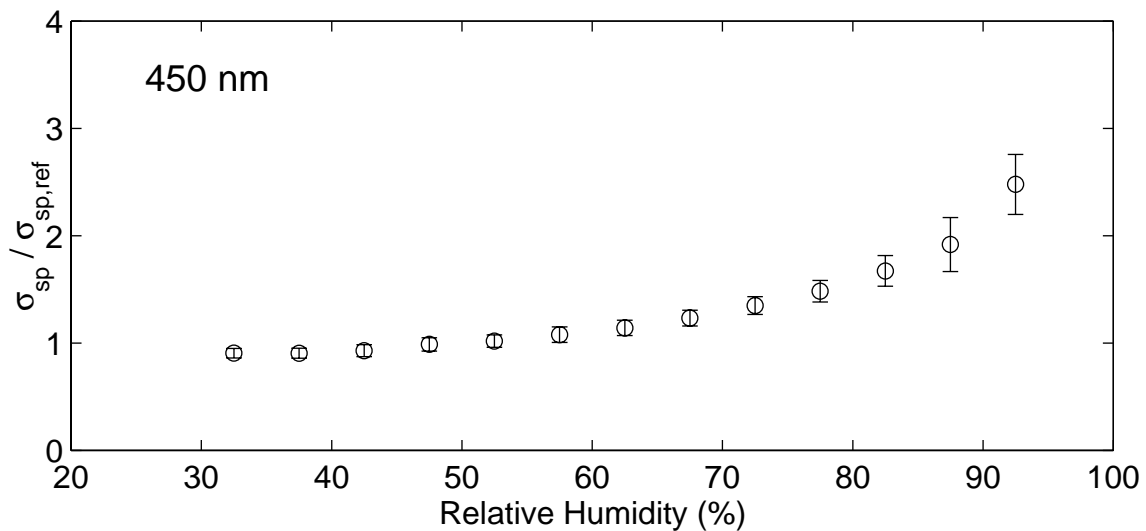
Comparisons-
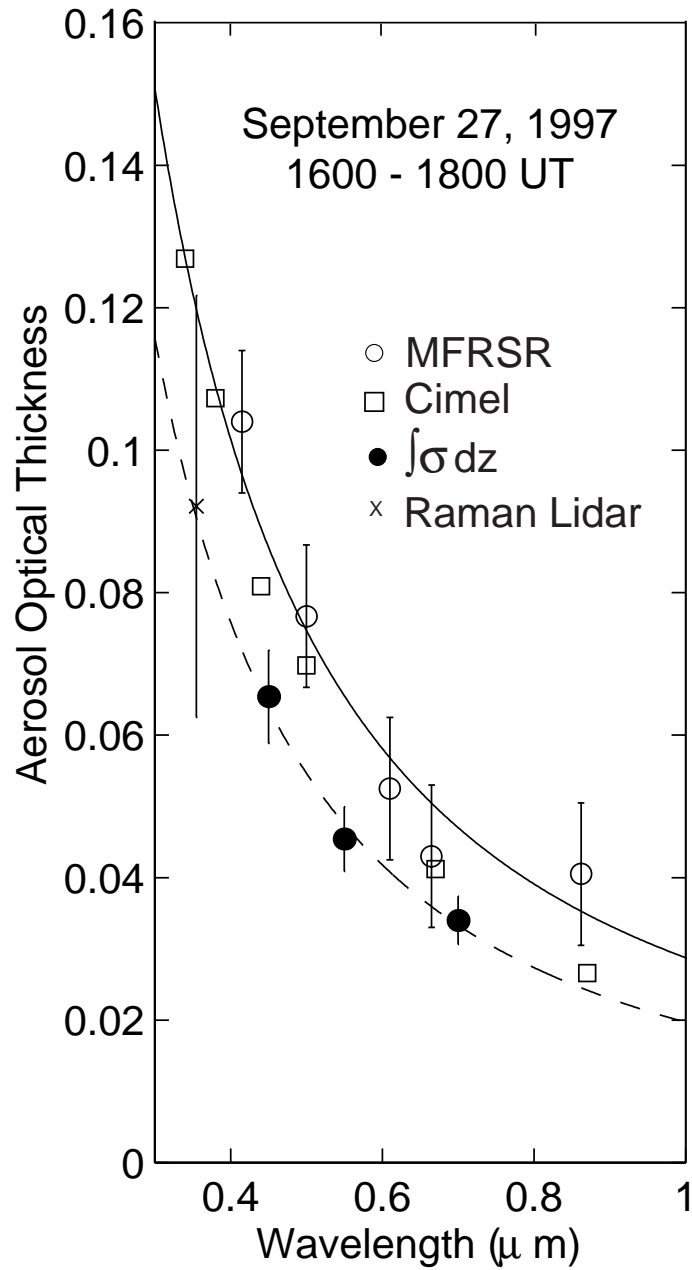
Conclusions-

- For April 1997 and September 1997 the difference between the optical thickness from sun photometry and vertical profiles is not significant.

- For August 1998 cases (high boundary layer RH) the optical thickness from sun photometry exceeds that from vertical profiles by 0.03 to 0.07 (25% to 31%).

- Based on these comparisons, the single-scattering albedo of particles in the lower troposphere is between 0.84 and 0.97.

S. Kato, M. H. Bergin, T. P. Ackerman, T. P. Charlock, E. E. Clothiaux, R. A. Ferrare, R. N. Halthore, N. Laulainen, G. G. Mace, J. Michalsky, D. D. Turner, JGR, in press (2000)

MEASUREMENT OF AEROSOL DIRECT FORCING OF SURFACE IRRADIANCE

Stephen Schwartz and Rangasayi Halthore, Brookhaven

Question-

What is the magnitude of direct radiative forcing of surface irradiance by aerosols?

Approach-

- Aerosol **direct** forcing is difference between surface irradiance with and without aerosol.

- For cloud-free, aerosol-free (Rayleigh) atmosphere, surface irradiance is *calculated* (**direct** and diffuse components) for specified illumination geometry, surface reflectance.

- Surface irradiance is *measured* (**direct** and diffuse components) in the presence of aerosol of measured optical thickness (sun photometry), for cloud-free sky.

- **Direct** Aerosol Forcing is *measured* (**direct** and diffuse components) as function of aerosol optical thickness as the difference between measurement and Rayleigh calculation.

Results-

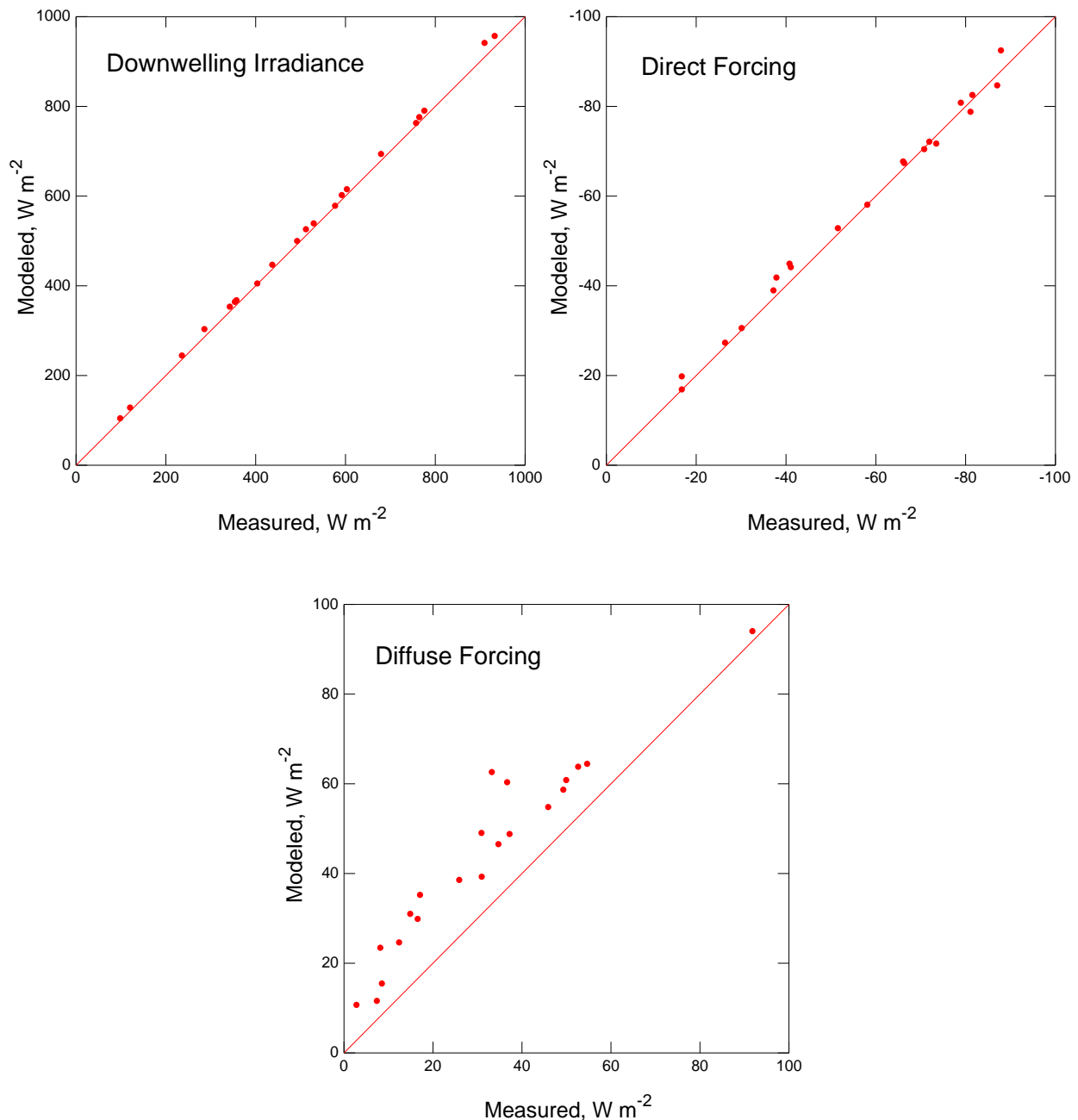## AEROSOL FORCING OF SURFACE IRRADIANCE
Cloud-free sky, SGP
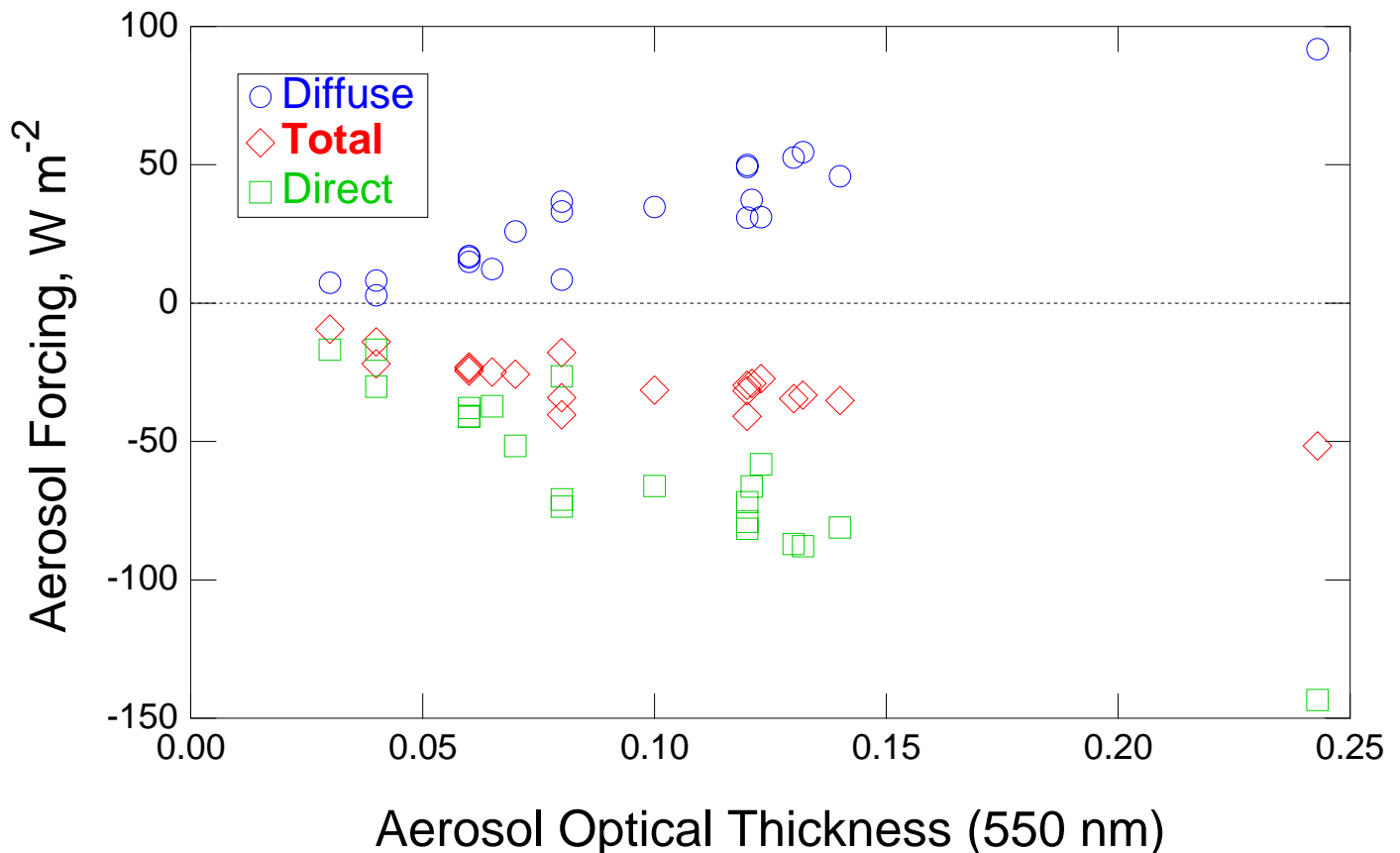
- Aerosol scattering **decreases direct** irradiance, **increases diffuse** irradiance.

- Aerosols **decrease total surface irradiance** (**direct** + **diffuse**).

AEROSOL FORCING OF SURFACE IRRADIANCE
Cloud-free sky, SGP
Comparison with Radiation Transfer Model

Model systematically overestimates diffuse forcing, as revealed also in Halthore *et al*. (*GRL*, 1998) and Halthore and Schwartz (*JGR*, submitted, 2000).

Conclusions-

- Surface forcing can be readily measured. The only "assumption" is the ability to calculate direct and diffuse irradiance for Rayleigh atmosphere.

- Aerosols exert substantial instantaneous surface forcing under cloud-free sky at SGP (-30 W m^{-2} for AOT $= 0.10$).

- Present models (MODTRAN-3.5, DISORT) accurately estimate direct beam forcing but overestimate the magnitude of diffuse forcing for AOT inferred from sunphotometry and reasonable aerosol properties.

R. N. Halthore and S. E. Schwartz, JGR, submitted (2000)

S. Schwartz and R. Halthore, Poster, ARM Science Team Meeting, San Antonio, 2000

SPECTRAL DISPERSION OF CLOUD DROPLET SIZE DISTRIBUTIONS AND PARAMETERIZATION OF CLOUD DROPLET EFFECTIVE RADIUS

Peter Daum and Yangang Liu, Brookhaven

Cloud Liquid Water Content: $L = \dfrac{4\pi}{3}\int r^3 \dfrac{dN}{dr}dr$

If monodisperse: $L = \dfrac{4\pi}{3}r^3$

Effective Radius: $r_e \equiv \mu_3 / \mu_2 = \int r^3 \dfrac{dN}{dr}dr \Big/ \int r^2 \dfrac{dN}{dr}dr$

If monodisperse:

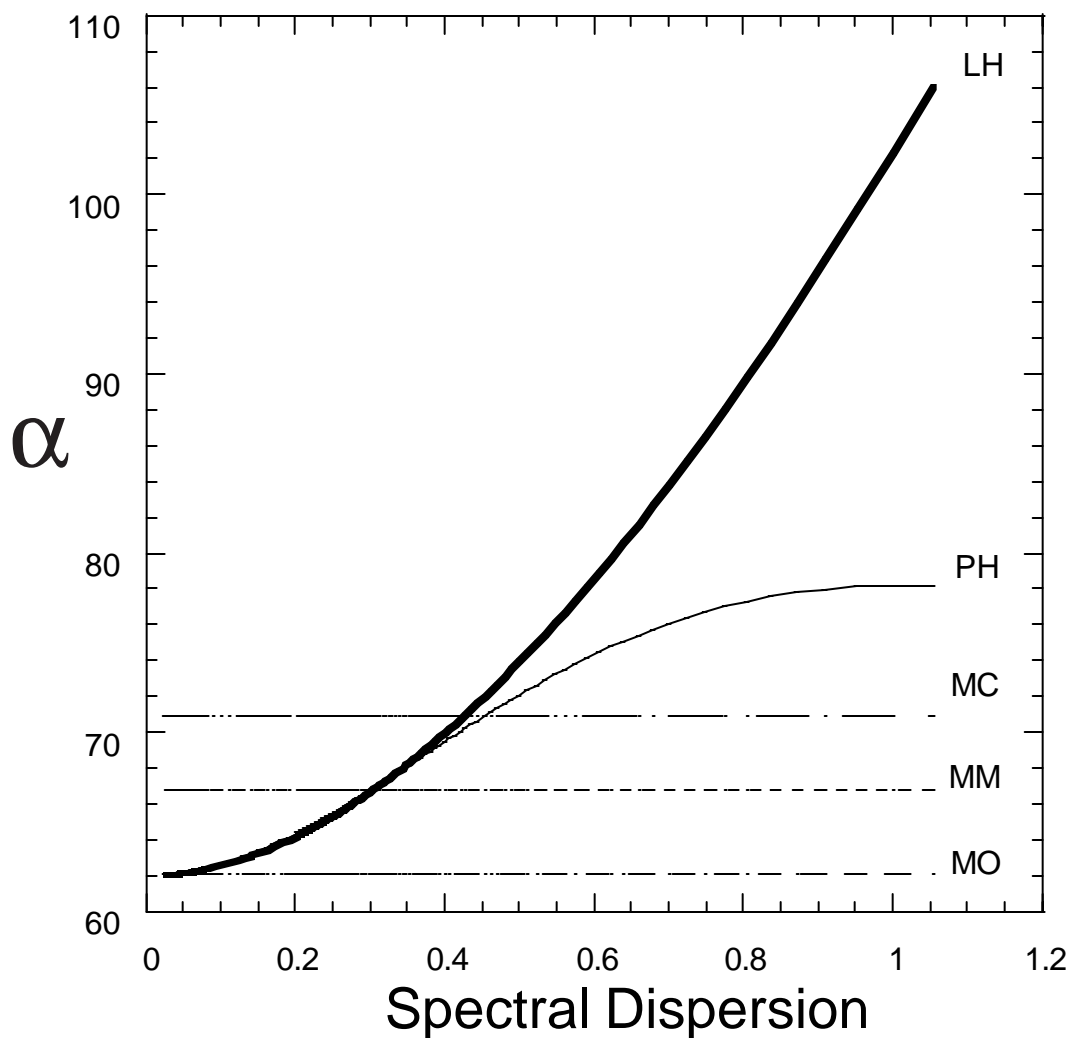$$r_e = 100\left(\frac{4\pi}{3}\right)^{1/3}\left(\frac{L}{N}\right)^{1/3} \approx 62.035\left(\frac{L}{N}\right)^{1/3}$$

More generally: $r_e = \alpha\left(\dfrac{L}{N}\right)^{1/3}$

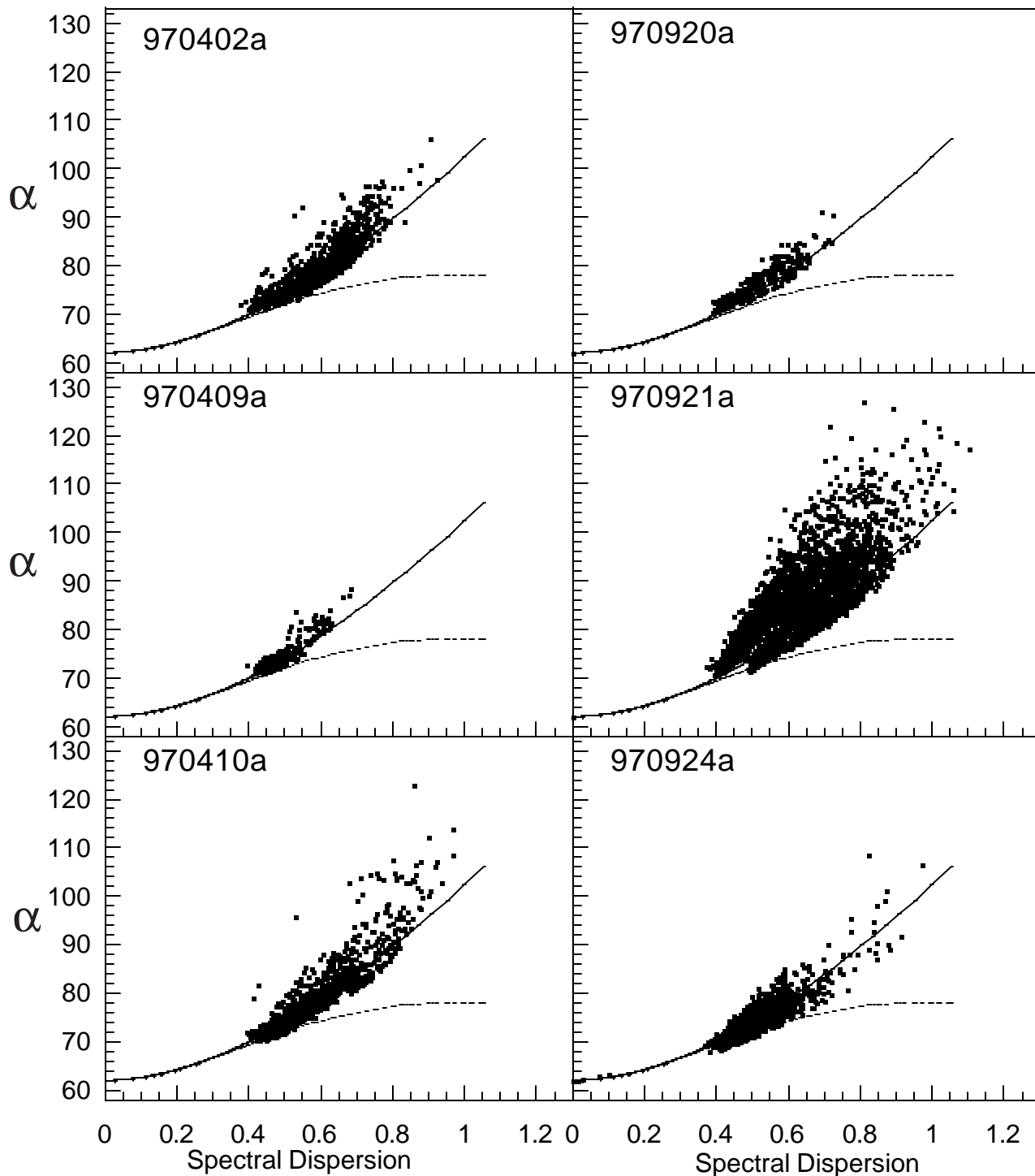For radiation transfer modeling it is necessary to know α.

Theory and previous observations-

There are two competing theories for dependence of α on dispersion of size distribution and limited previous observations of α and several previous field measurements.
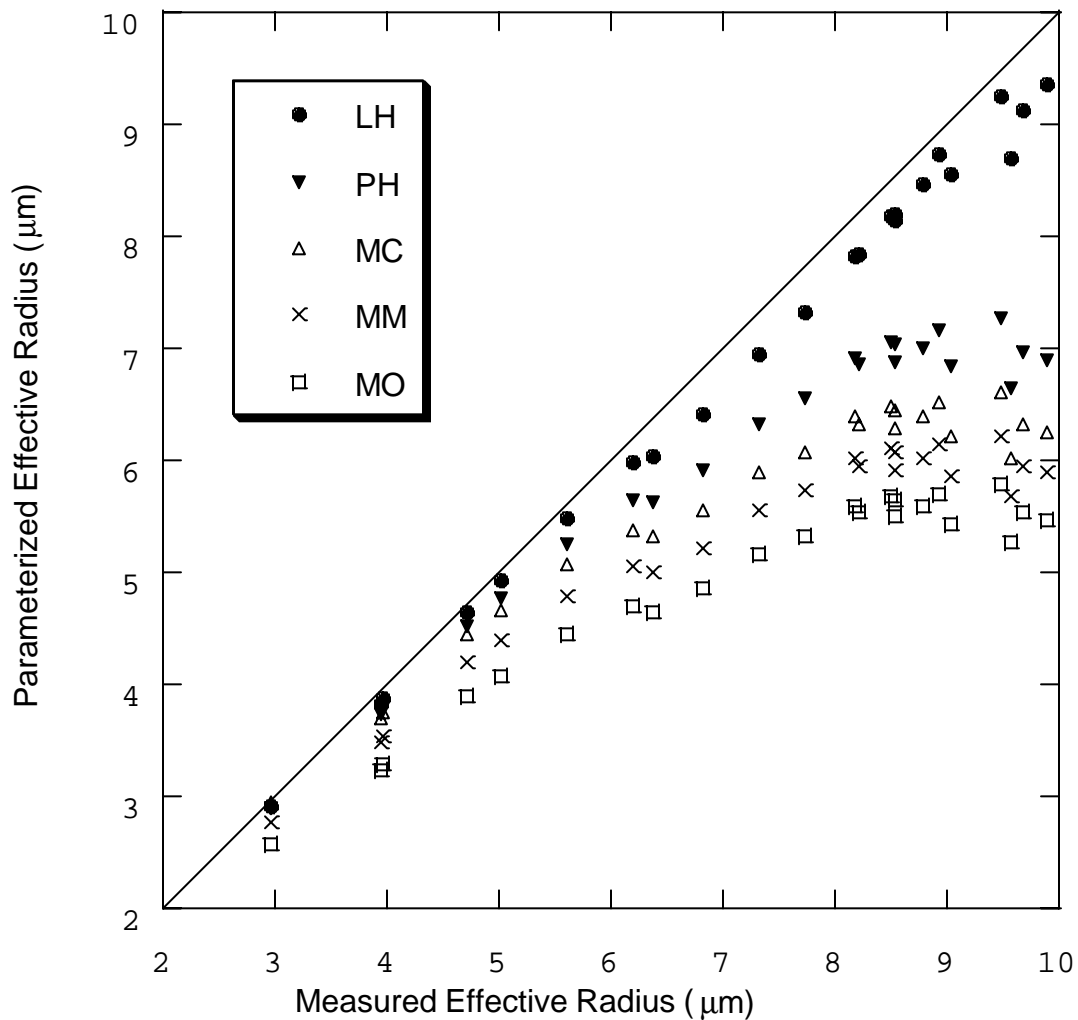
Observations-

From measurements of cloud droplet size distributions at SGP spring and fall 1997.

Parametrization of effective radius-

This study leads to enhanced confidence in parametrization of effective radius of cloud droplets.

Y. Liu and P. H. Daum, GRL, submitted, 2000

THE FUTURE

Regularly conducted vertical profiling at SGP Central Facility of:

Aerosol scattering coefficient σ_{sp} (3-wavelength with backscatter shutter).

Aerosol absorption coefficient σ_{ap}.

    Extinction coefficient ($\sigma_{ep} = \sigma_{sp} + \sigma_{ap}$) can be compared to Raman Lidar and Sun photometry.

    Single scattering albedo $\omega_0 = \sigma_{sp} / (\sigma_{sp} + \sigma_{ap})$ and backscatter fraction are required for radiative transfer calculations.

Altitude range 0.15 - 3.3 km. 2 - 3 flights per week, weather permitting.

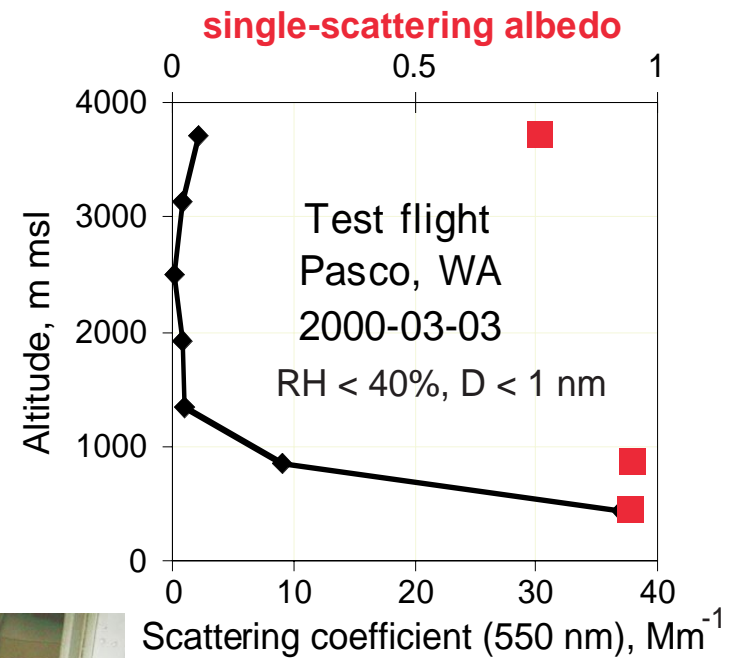Methodology will be the same as at SGP surface site:

    Sub-1 μm; RH $< 40\%$.

In-Situ Aerosol Profiling at SGP

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

Measurements: σ_{sp} (3λ), σ_{ap} (1λ), identical instruments and sample conditioning as at surface site

2-3 profiles/week for 1 year

single-scattering albedo

Test flight
Pasco, WA
2000-03-03
RH < 40%, D < 1 nm

Altitude, m msl

Scattering coefficient (550 nm), Mm^{-1}

Sample Inlet

Port view of rack

J. Ogren

CONCLUSIONS

- Characterization of aerosol is essential to modeling shortwave radiation at SGP.

- Aerosol research is alive and well at SGP.

- The shortwave and aerosol research groups will be able to make good use of forthcoming vertical profile data, relate to Raman Lidar profiles, to surface aerosol measurements.

- A remaining issue is RH dependence of aerosol aloft and/or f(RH) measurement.

- Aerosol-cloud microphysics coupling is subcritical at SGP.

- Aerosol microphysics is subcritical at NSA and TWP.