Retrieval of Black Carbon Concentration from the Aerosol Robotics Network (AERONET)

Greg Schuster, *NASA LaRC*
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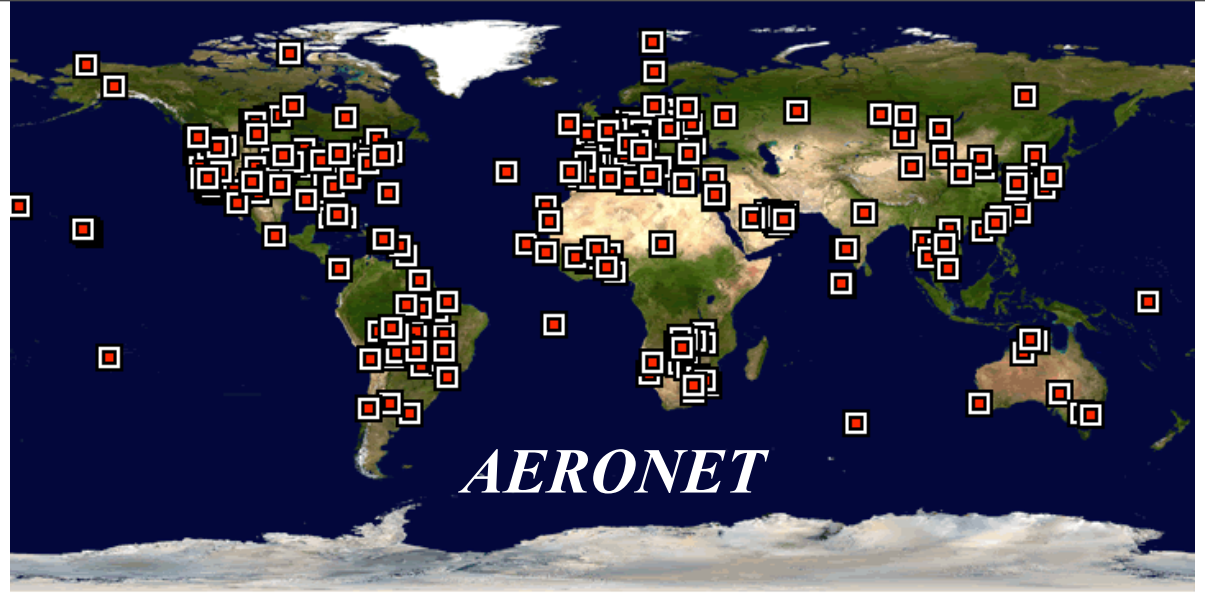
Brent Holben, *NASA GSFC*

Worldwide black carbon concentration measurements are needed to assess the efficacy of the carbon emissions inventory and transport model output. This requires long-term measurements in many regions, as model success in one region or season does not apply to all regions and seasons. AERONET is an automated network of more than 180 surface radiometers located throughout the world. The sky radiance measurements obtained by AERONET are inverted to provide column-averaged aerosol refractive indices and size distributions for the AERONET database, which we use to derive column-averaged black carbon concentrations and specific absorptions that are constrained by the measured radiation field. This provides a link between AERONET sky radiance measurements and the elemental carbon concentration of transport models without the need for an optics module in the transport model. Knowledge of both the black carbon concentration and aerosol absorption optical depth (i.e., input and output of the optics module) will enable improvements to the transport model optics module.



Black Carbon Concentration from Worldwide Aerosol Robotic Network (AERONET) Measurements

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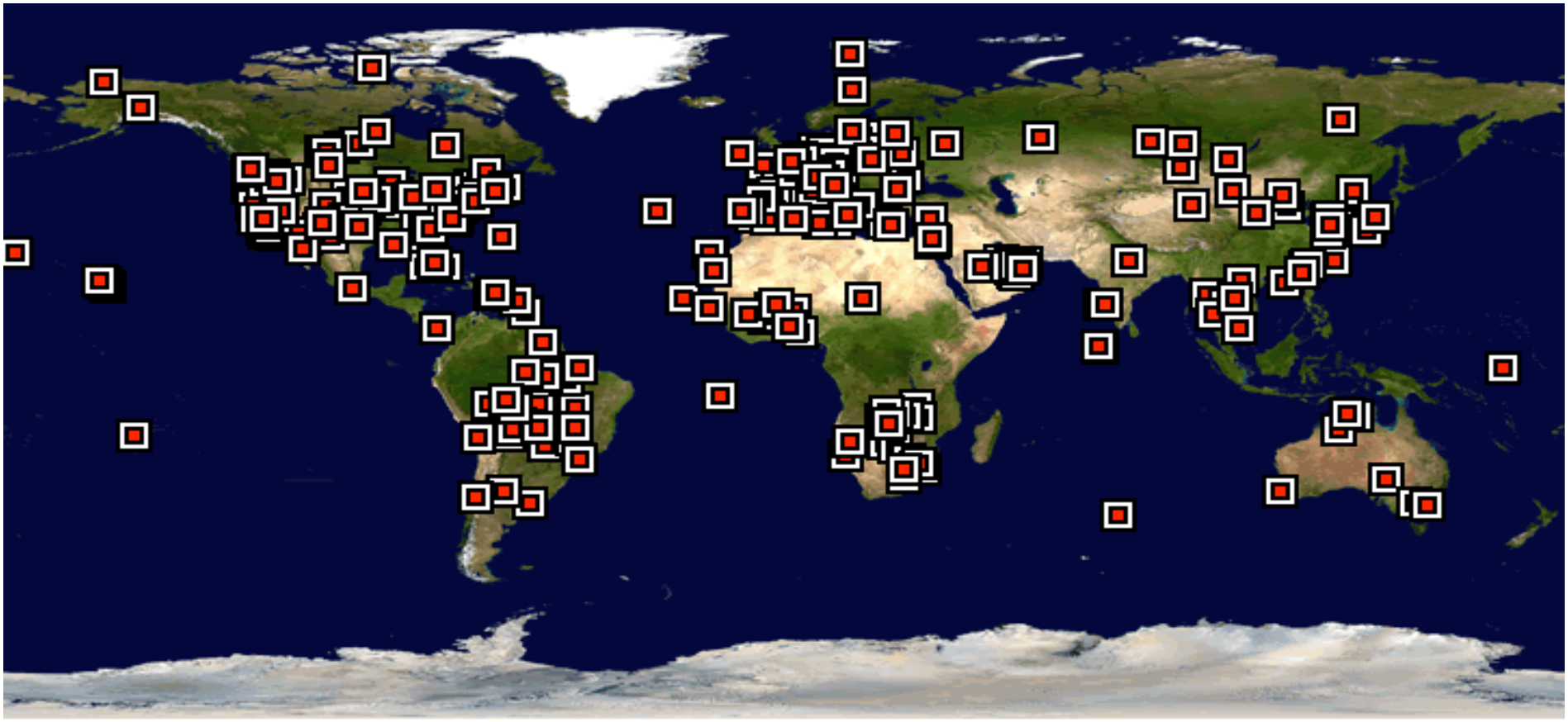
Brent Holben, *NASA GSFC*

Eugene Clothiaux, *Penn State*

- Details at Schuster, et al., *J. Geophys. Res., 110, 2005*.

Outline

- Motivation
- AERONET product
- Maxwell Garnett effective medium approximation
- Description of black carbon retrieval and results
- Validation and sensitivity study



Modeling Global Black Carbon Absorption

Carbon Emissions Inventory

Uncertainty factor > 2

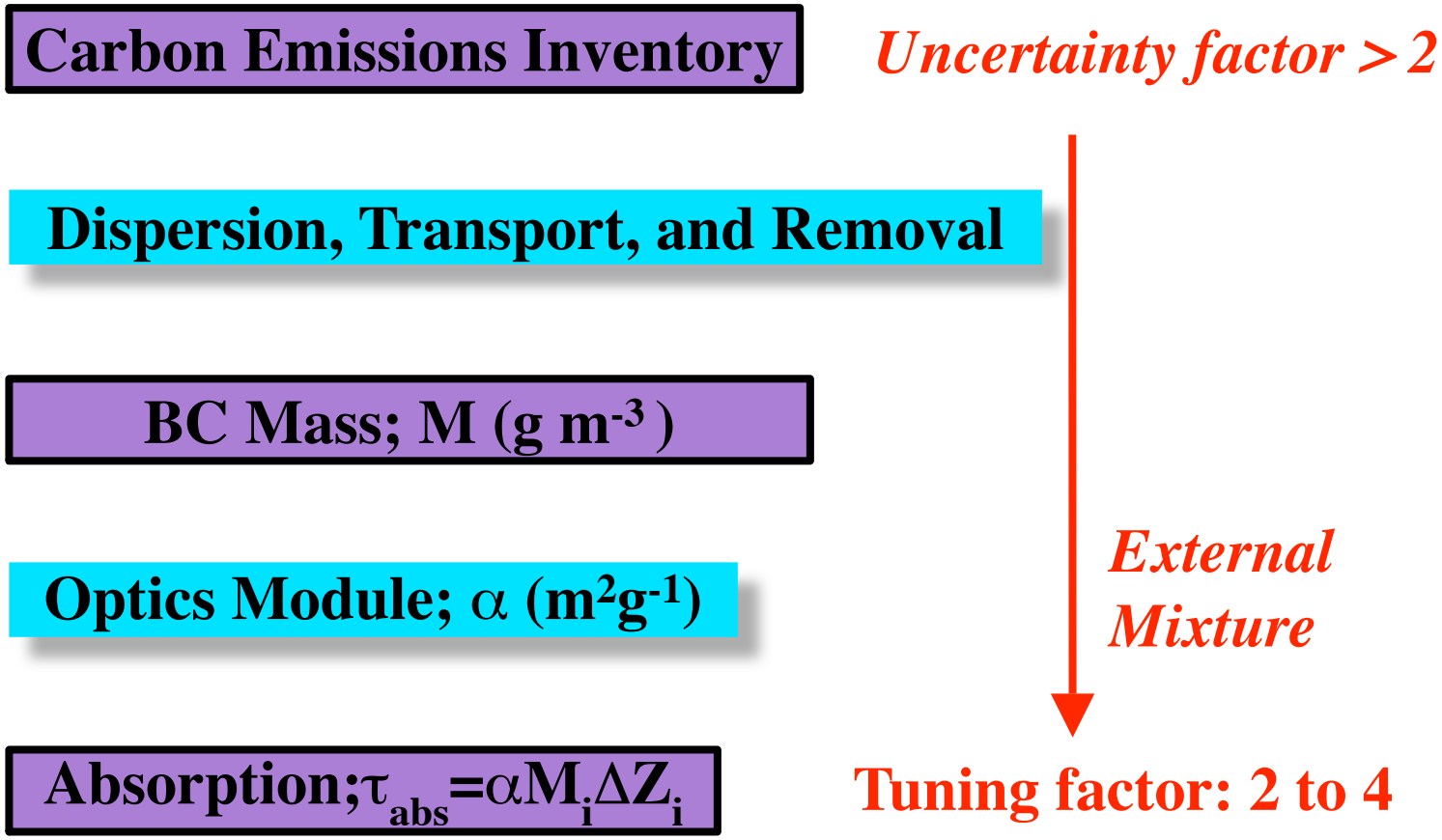
Dispersion, Transport, and Removal

BC Mass; M (g m^{-3})

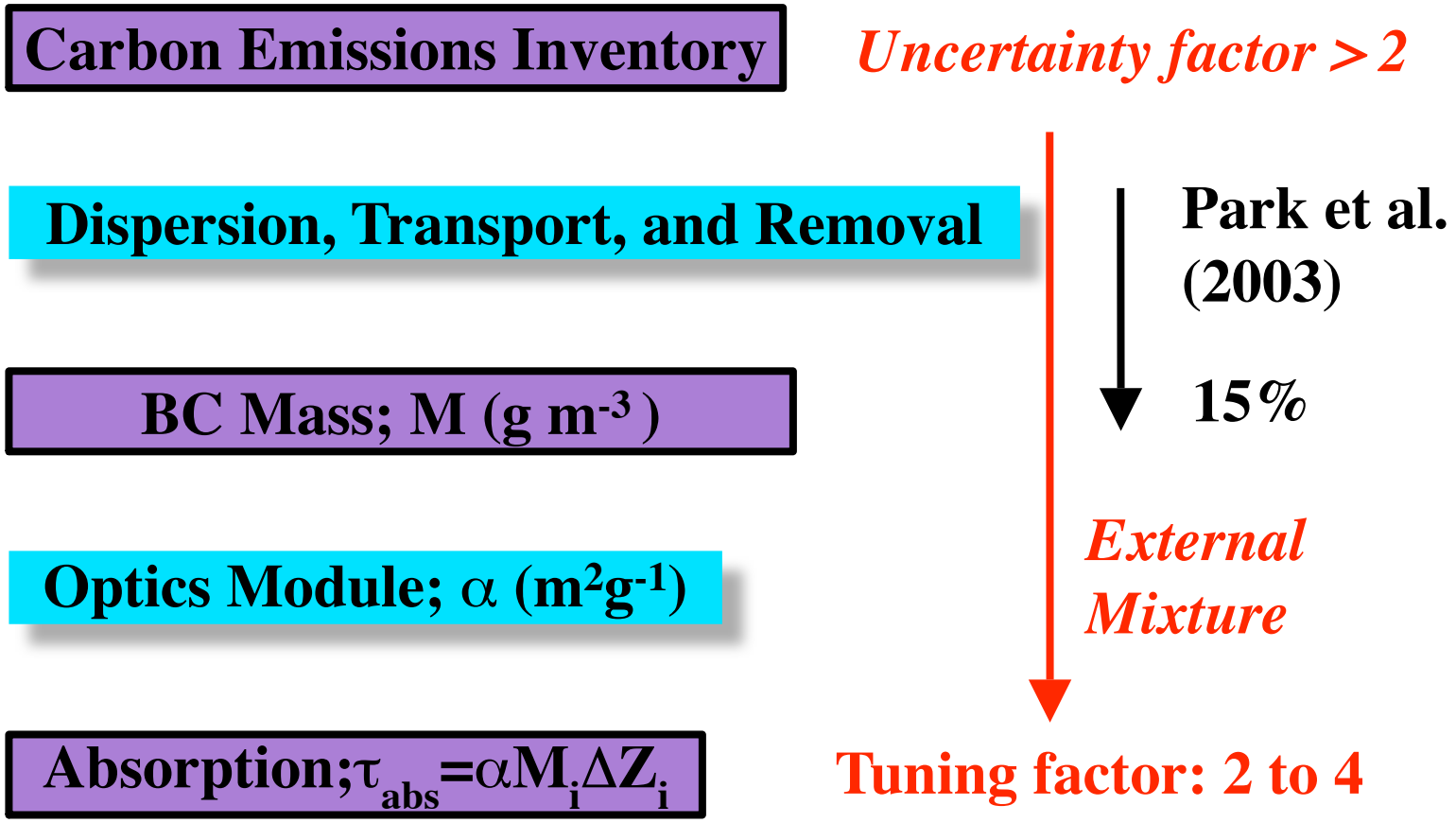
Optics Module; α ($m^2 g^{-1}$)

Absorption; $\tau_{abs} = \alpha M_i \Delta Z_i$

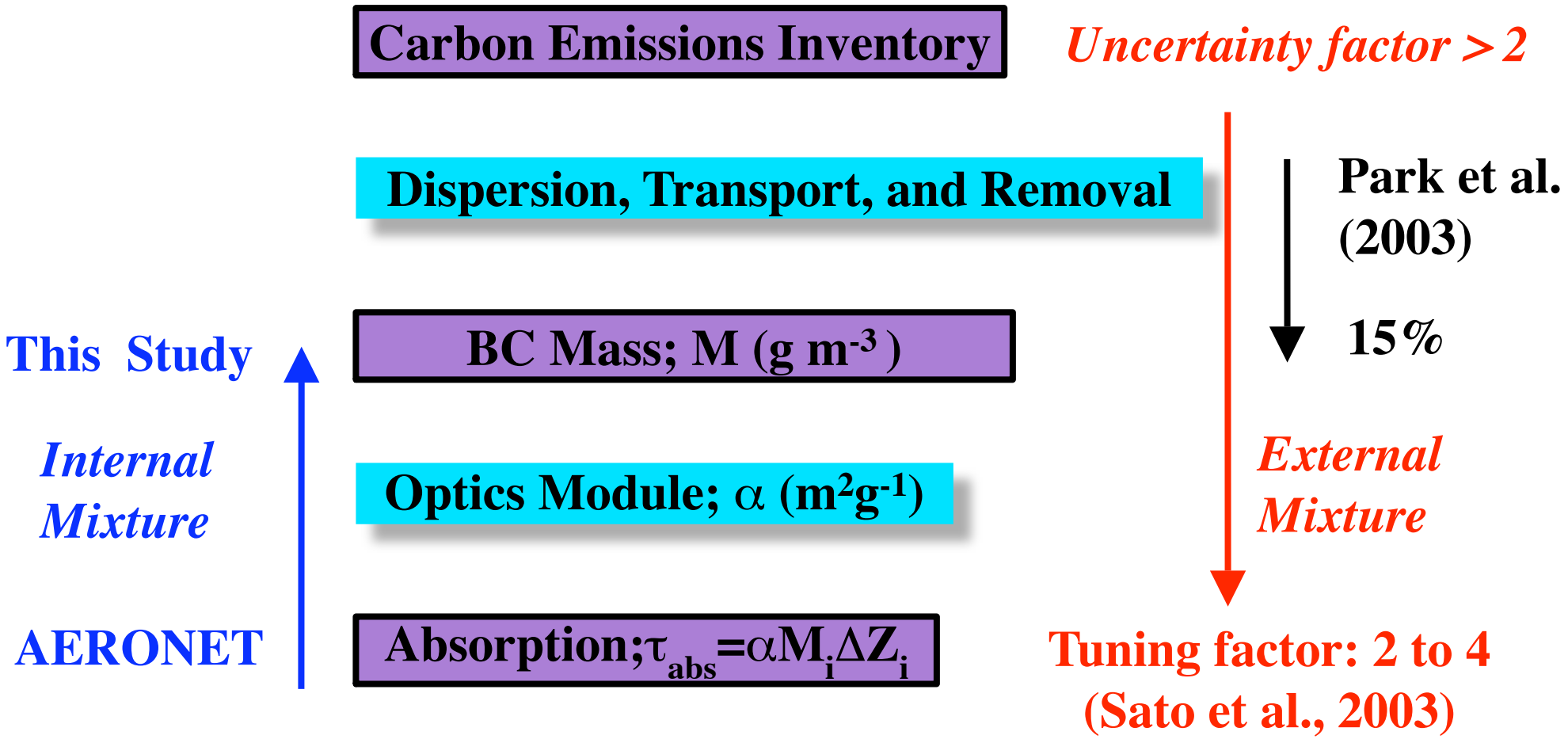
Modeling Global Black Carbon Absorption



Modeling Global Black Carbon Absorption



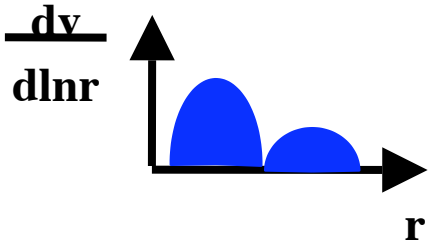
Modeling Global Black Carbon Absorption



AERONET Aerosol Retrieval

Based upon almucantar sky radiance scan
Avg residual radiance errors < 5%, 21 angles

Provides columnar size distribution
at 22 radii from 0.05 to 15 μm



Complex refractive index at 4 wavelengths $n(\lambda), k(\lambda)$

Internal mixture

Cloud-screening: temporal, spatial, and symmetry constraints

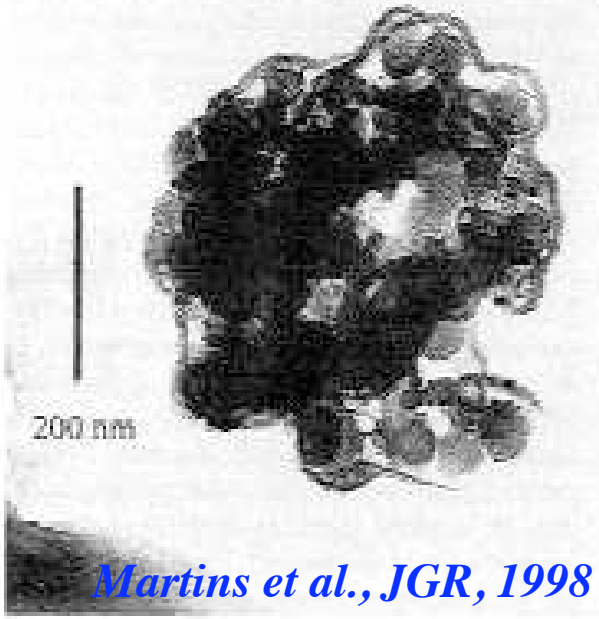
Maxwell Garnett effective medium approximation

Maxwell Garnett refractive index:

$$m_{MG}\left(m_{host}, m_j, f_j\right)$$

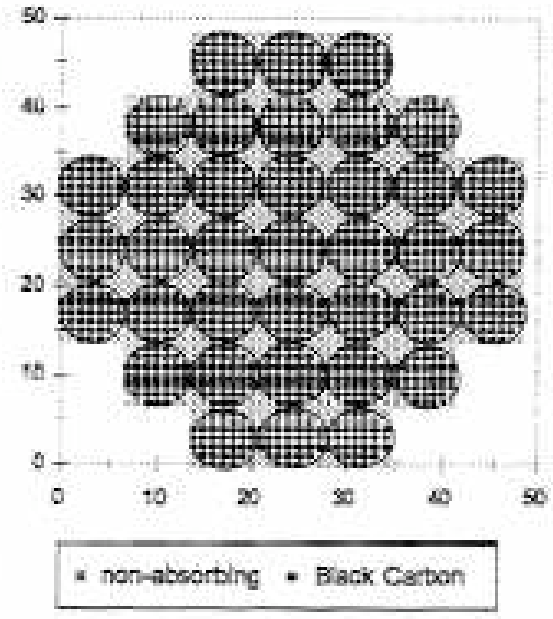
f_j = **inclusion volume fraction**

Assumptions:
(small, spherical inclusions)



Martins et al., JGR, 1998

Electron microscope photograph



Discrete dipole model

Maxwell Garnett effective medium approximation

Maxwell Garnett refractive index:

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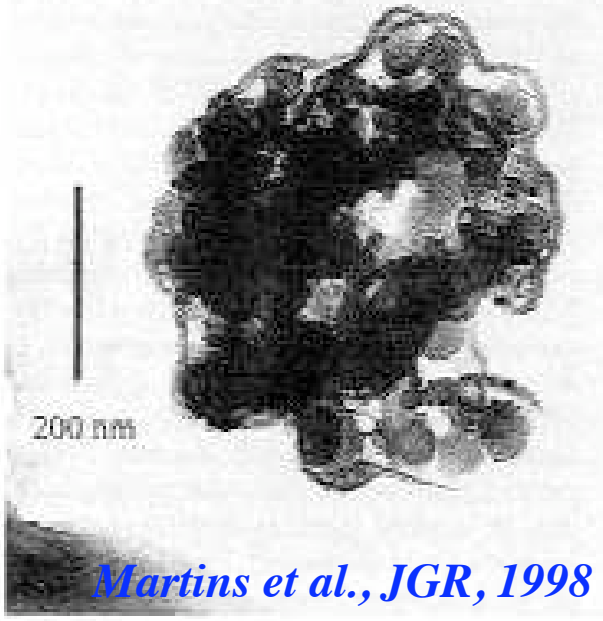
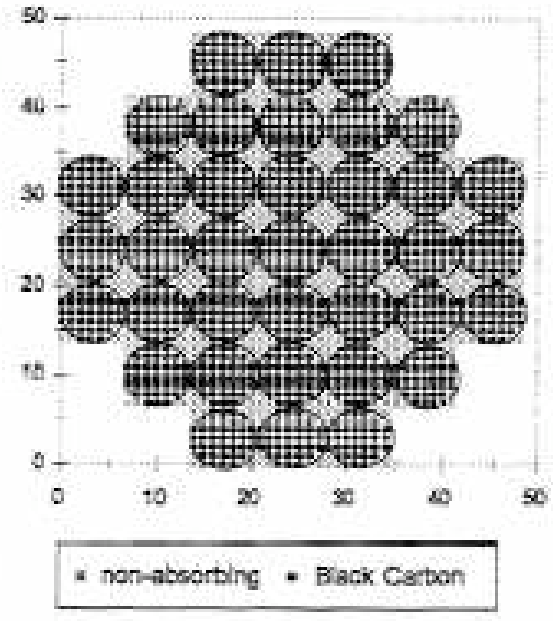
f_j = **inclusion volume fraction**

Assumptions:
(small, spherical inclusions)



Martins et al., JGR, 1998

Electron microscope photograph



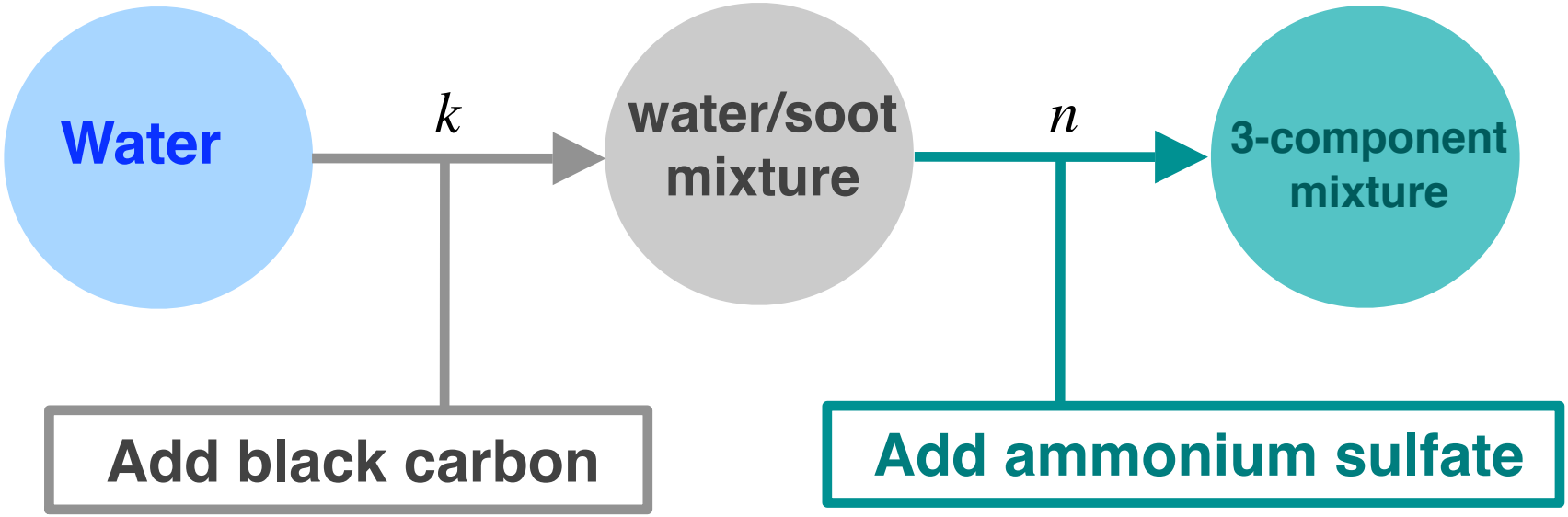
Discrete dipole model

52% Black carbon, monomer radii up to 57 nm, particle radii 0.05-0.4 μm

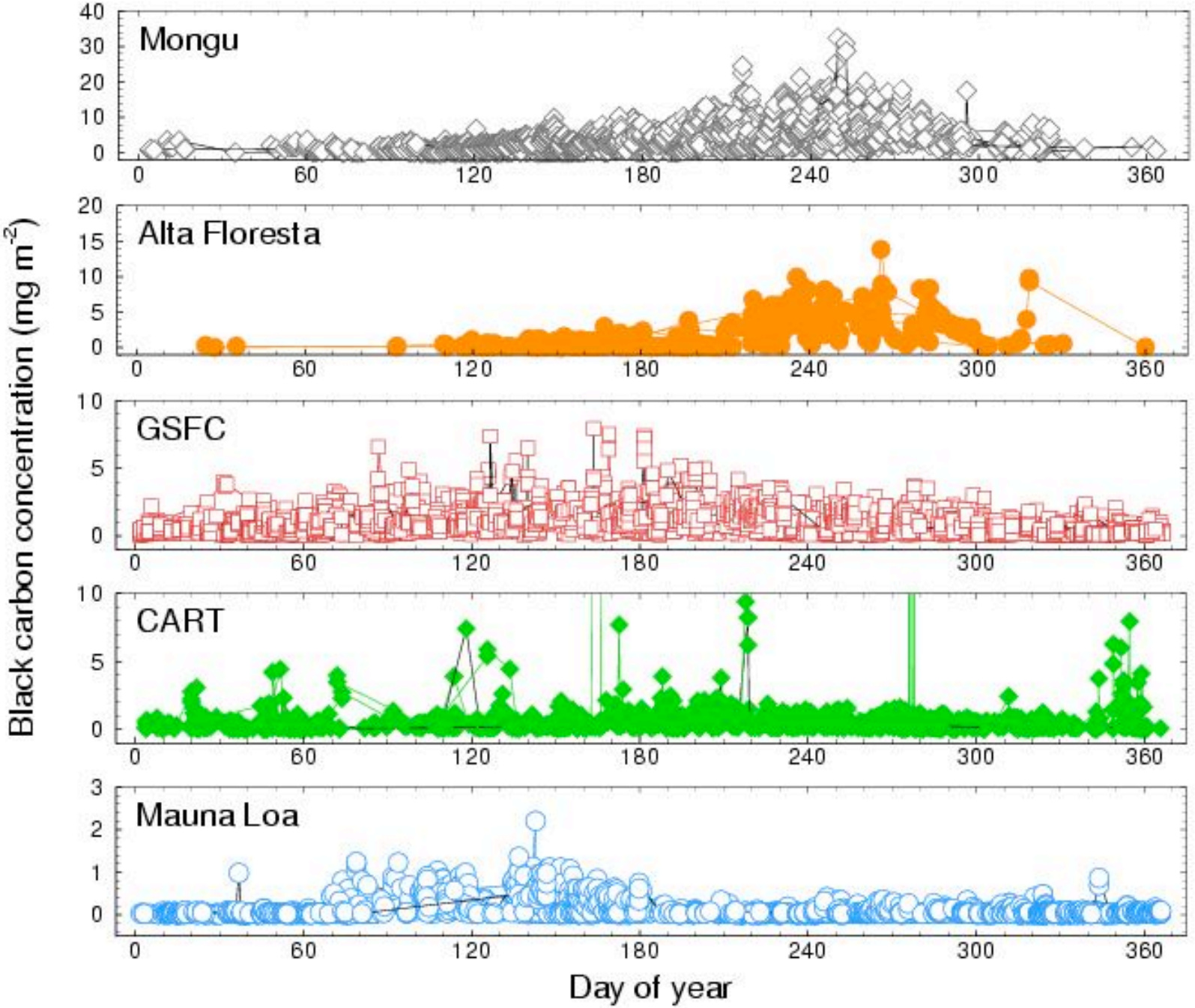
Maxwell Garnett specific absorption consistent with discrete dipole model to within ~10%.

Black carbon content from AERONET retrievals

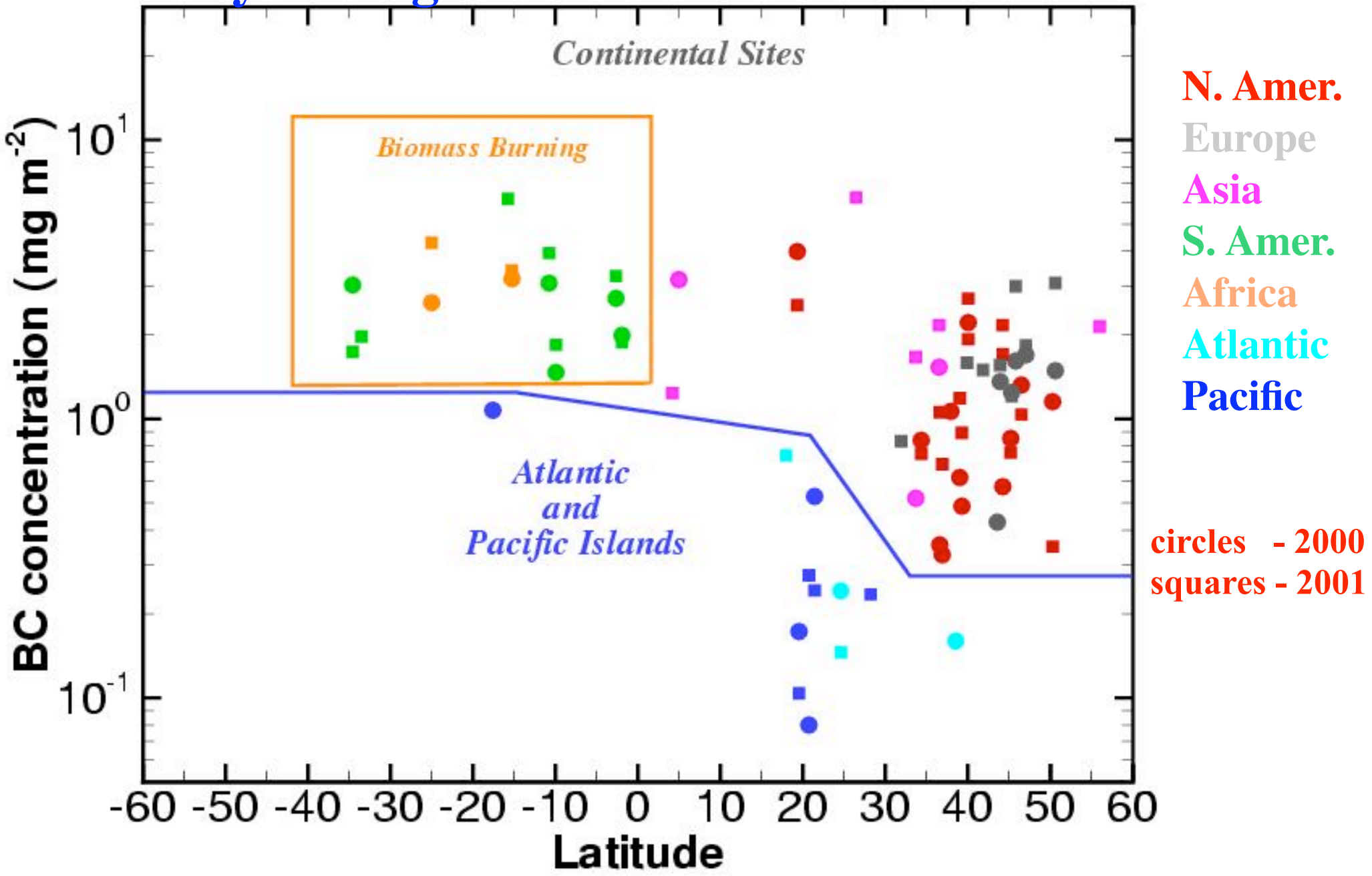
$$\chi^2 = \sum_{l=1}^{4\ \text{wvlns}} \frac{\left(m_l^{rtrv} - m_l^{mix}\right)^2}{m_l^{rtrv}} \Rightarrow 0$$



1993-2002

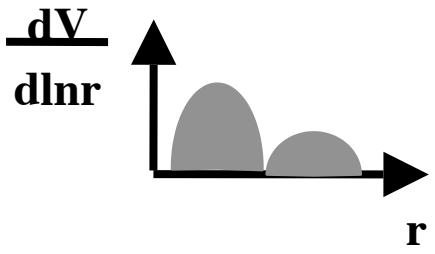

Mongu
Alta Floresta
GSFC
CART
Mauna Loa
Black carbon concentration (mg m-2)
Day of year
0
60
120
180
240
300
360

Yearly averaged black carbon concentration



Black Carbon Specific Absorption

$$\alpha = \frac{\tau_a}{[BC]} = \frac{\tau_a\left(m_{mix}\left(f_{BC}\right)\right)}{f_{BC}\rho_{BC}\int \frac{dV}{d\ln r} d\ln r}$$



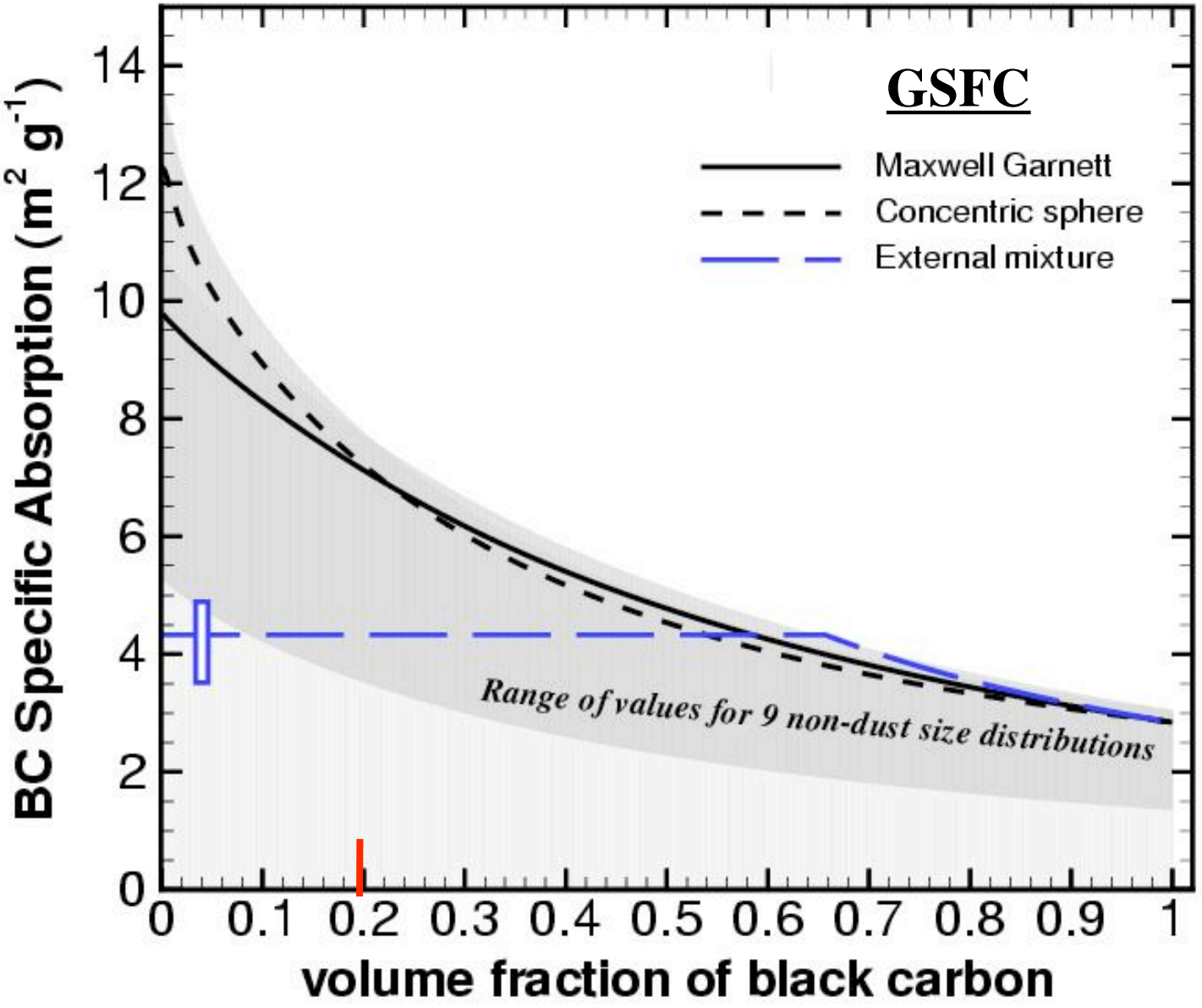
τ_a **absorption AOT**

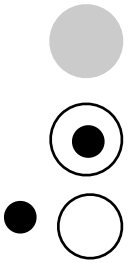
f **component fraction**

m **refractive index**

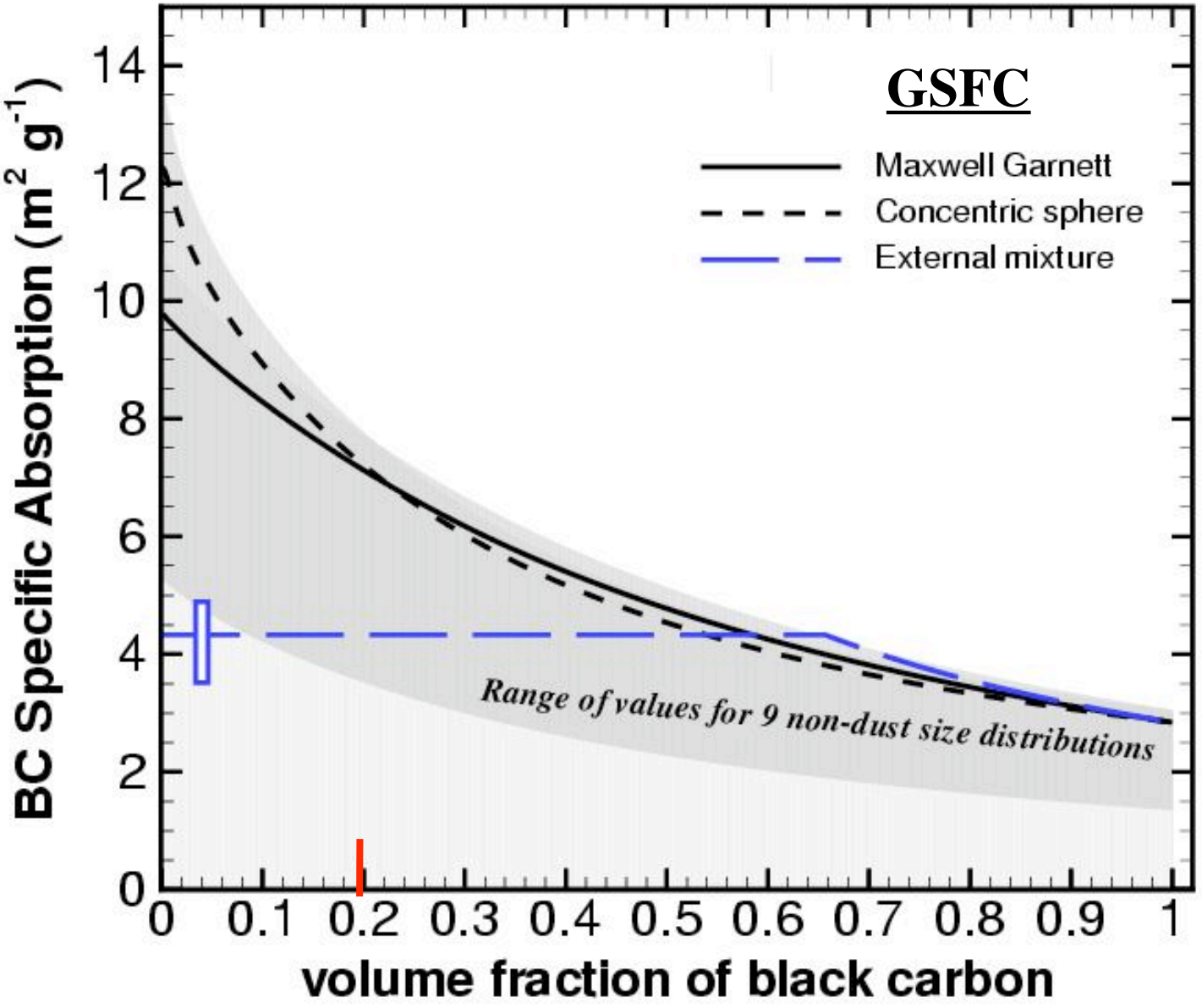
ρ_{BC} **density of black carbon**

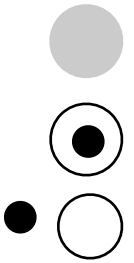
BC Specific Absorption for Nine Nondust Climatologies



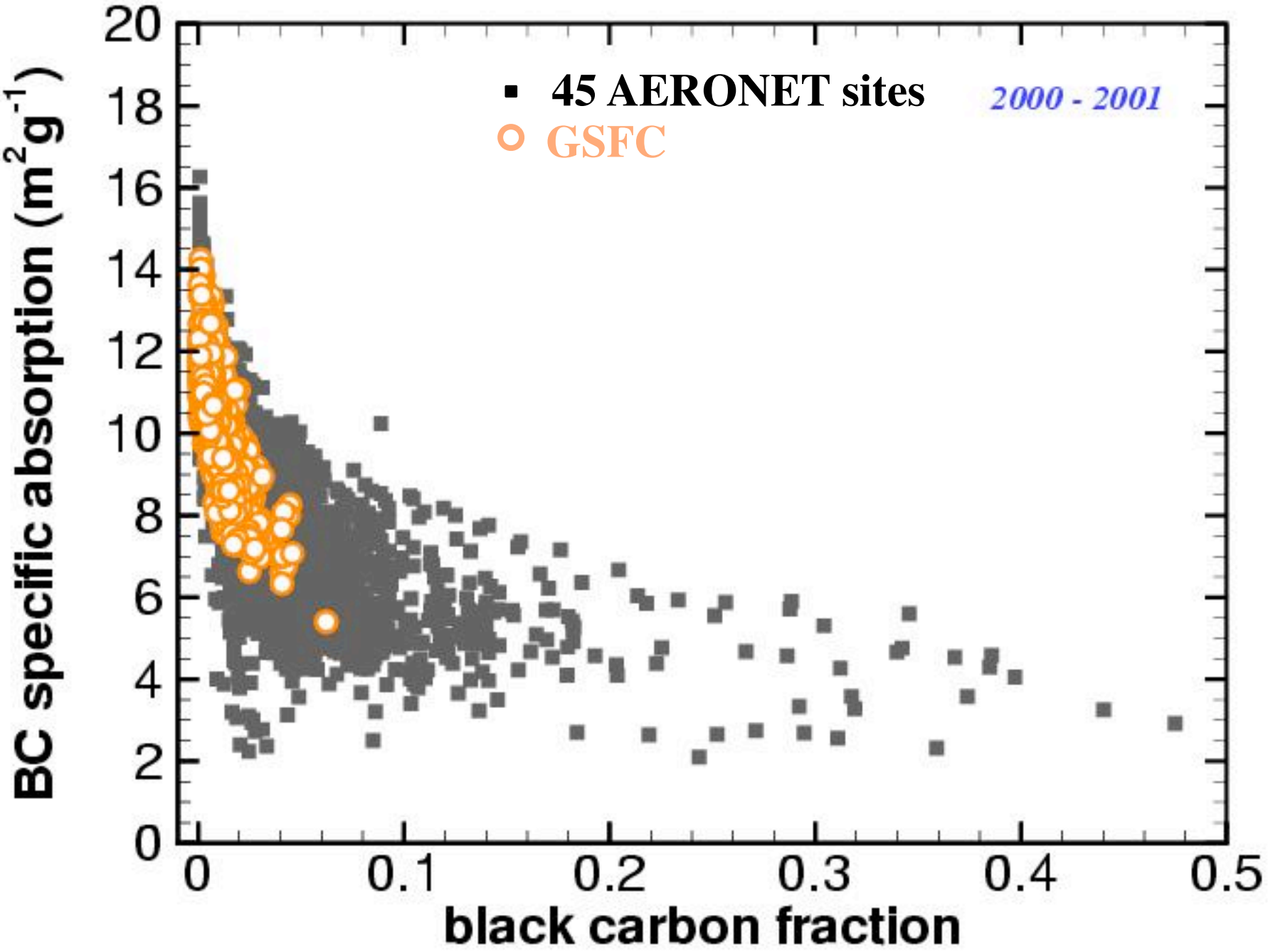


BC Specific Absorption for Nine Nondust Climatologies

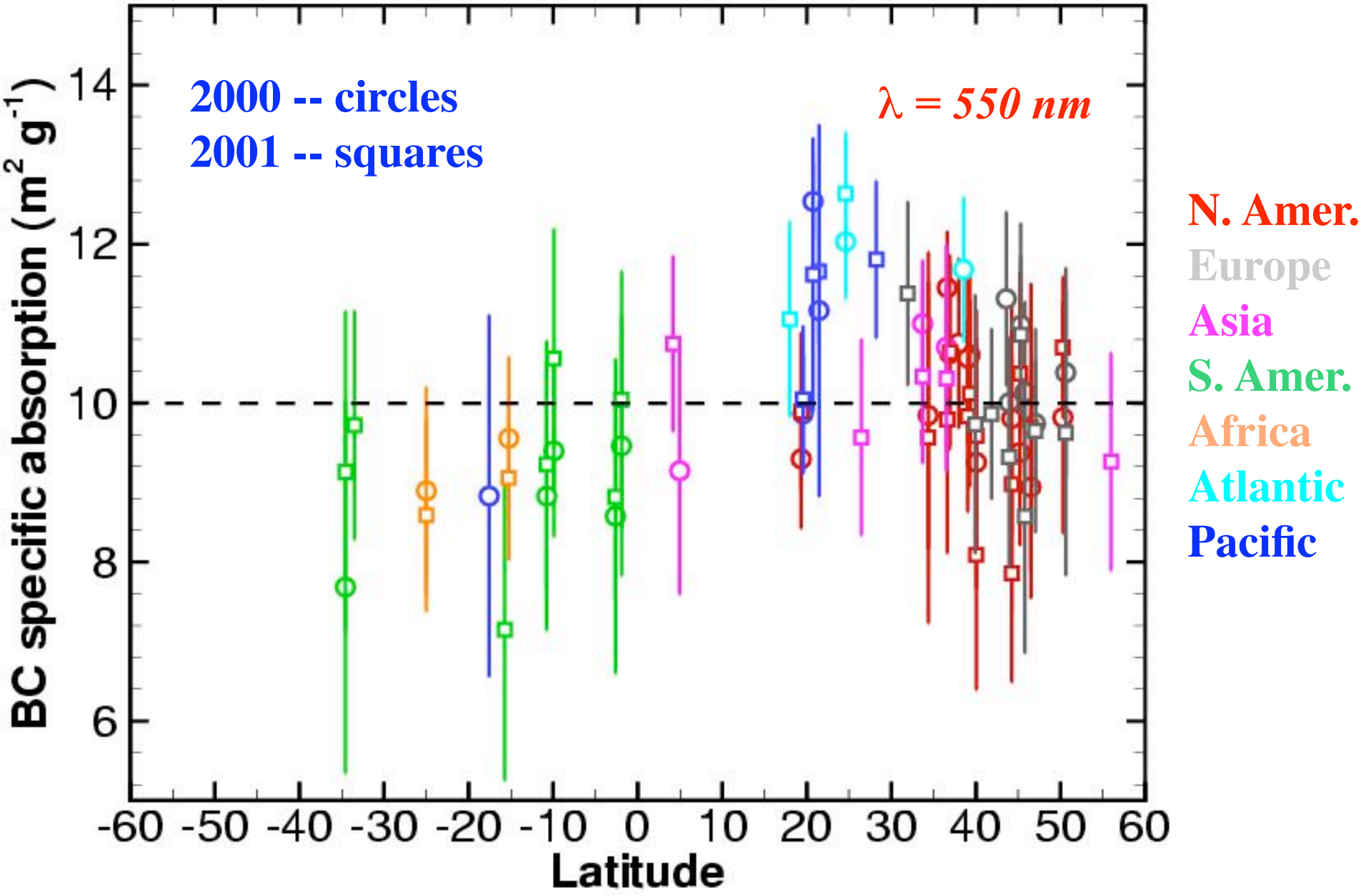




Also reported by Petzold et al., 1997; Neusuß et al., 2002


λ = 550 nm
45 AERONET sites
GSFC
2000 - 2001
BC specific absorption (m²g⁻¹)
black carbon fraction
0
2
4
6
8
10
12
14
16
18
20
0
0.1
0.2
0.3
0.4
0.5

Yearly-averaged black carbon specific absorption



Pseudo-validation

Model Comparison to RSS irradiance at ARM SGP site

Description of RSS (Rotating Shadowband Spectroradiometer)

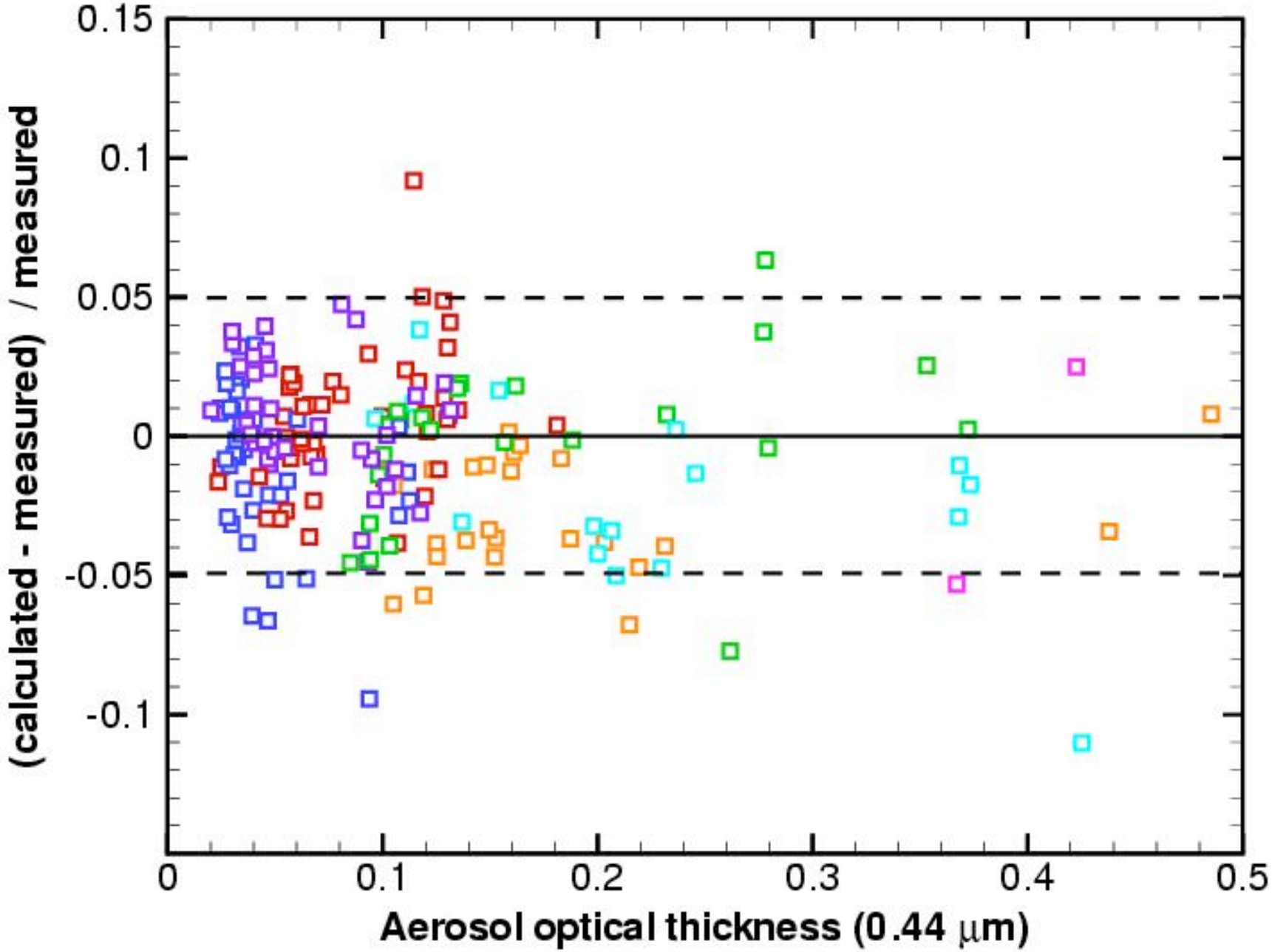
- **Spectral irradiance, 0.36-1.1 μm**
- **193 measurements coincident w/ AERONET retrievals in 2000-2002**

Model Atmosphere

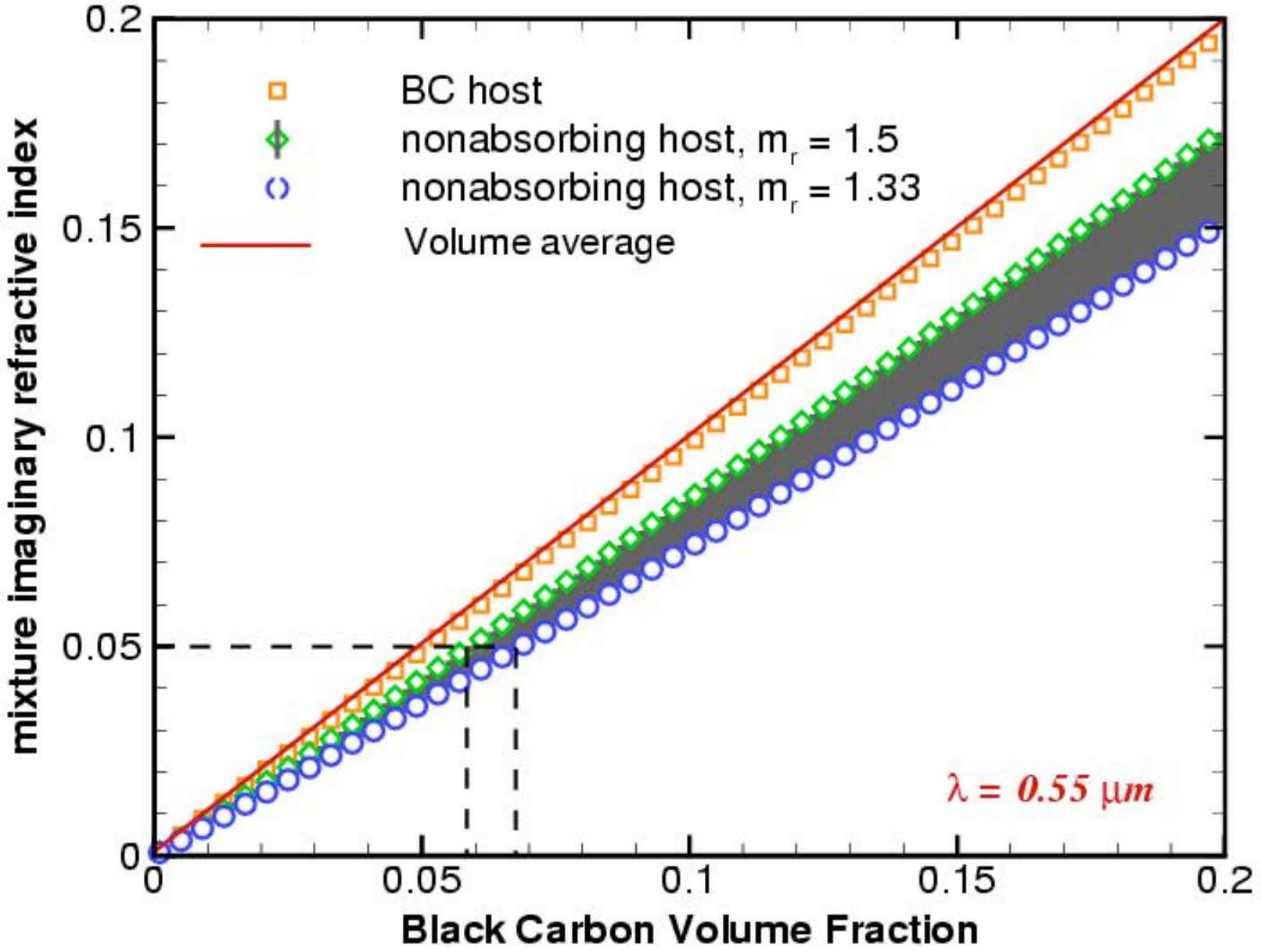
- **Correlated-k distribution for gas absorption (Kato et al., 1999)**
- **Microwave radiometer and other instruments: P(Z), T(Z), H_2O_v(Z)**
- **Molecular extinction and O_2 absorption scaled to P(Z)**
- **AERONET size distribution, constrained to 2-km boundary layer**
- **Minimum χ^2 fit for AERONET aerosol refractive index**

Model comparison with RSS irradiance

Kato et al., 1999 CKD; 0.35-1.05 μm,



Sensitivity of retrieved black carbon to choice of host aerosol



III. Sensitivity to Assumptions

	[BC] Error (%)	Specific Absorption Error (%)
Nonwater host	+15	-15
Coarse mode	??	??
OC	+10	-10
Maxwell Garnett	+/- 10	-/+ 10
BC density	+/- 5	-/+ 5
	-15 to + 40	-40 to +15

- Improvement over factor of 2+ uncertainty in BC emissions inventories
- Some cancellation of errors is likely

  Avg specific absorption for 19,591 retrievals is 9.9 m^2g^{-1}

  Accurately calculates surface radiation

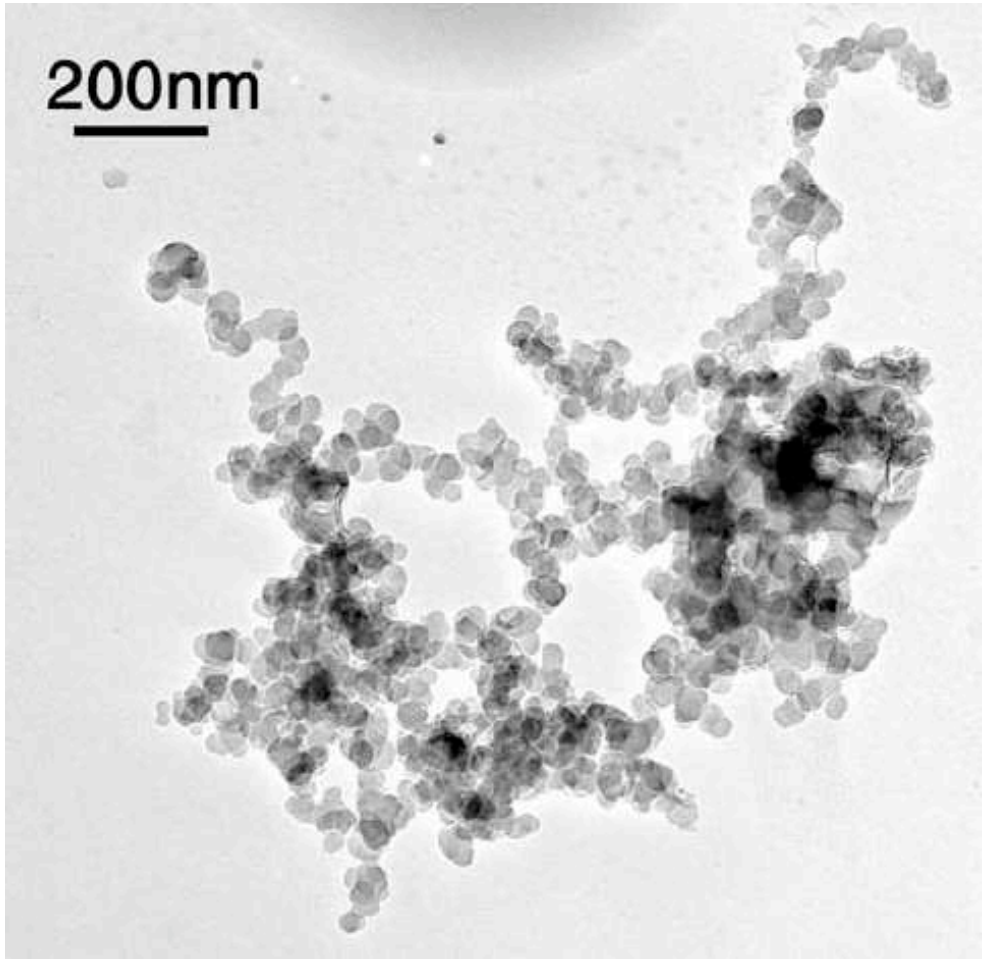
CONCLUSIONS

- **BC concentrations and specific absorptions at 46 AERONET sites**
- **Results look reasonable**
- **"Pseudo-validated" with independent radiation measurements**
- **Internally-mixed BC absorption is sensitive to the details of the size distribution and the fraction of black carbon in the mixture**
- **Volume averaging for internal mixtures produces refractive indices that are too high; Maxwell Garnett equations are easily parameterized.**
- gregory.l.schuster@nasa.gov
- Details at Schuster, et al., *J. Geophys. Res., 110, 2005*.

Acknowledgements:
This work was supported by NASA ESE and the CERES project. We appreciate the efforts of the entire AERONET team, and the Baltimore Supersite data provided by P. Hopke and M. Adam.

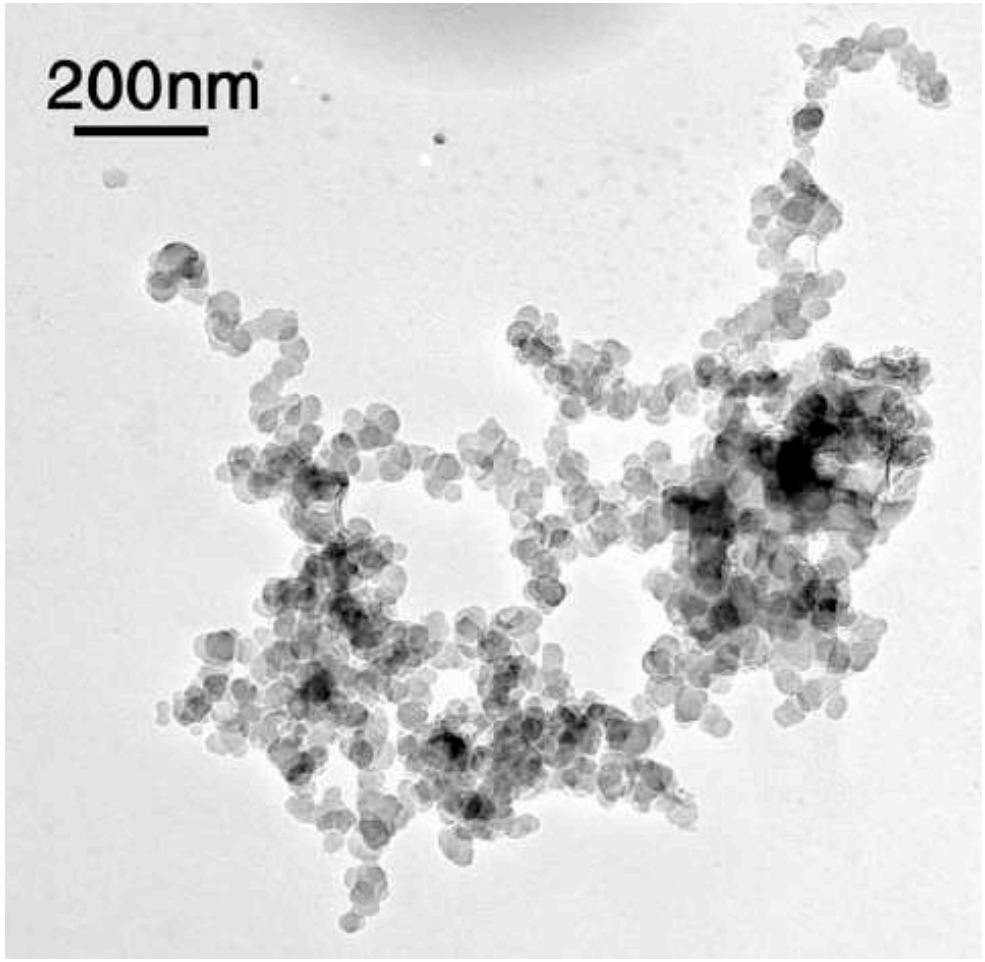
Appendix

Black carbon is not a sphere



Aggregate soot at Sagres, Portugal;
Li et al., JGR (2003)

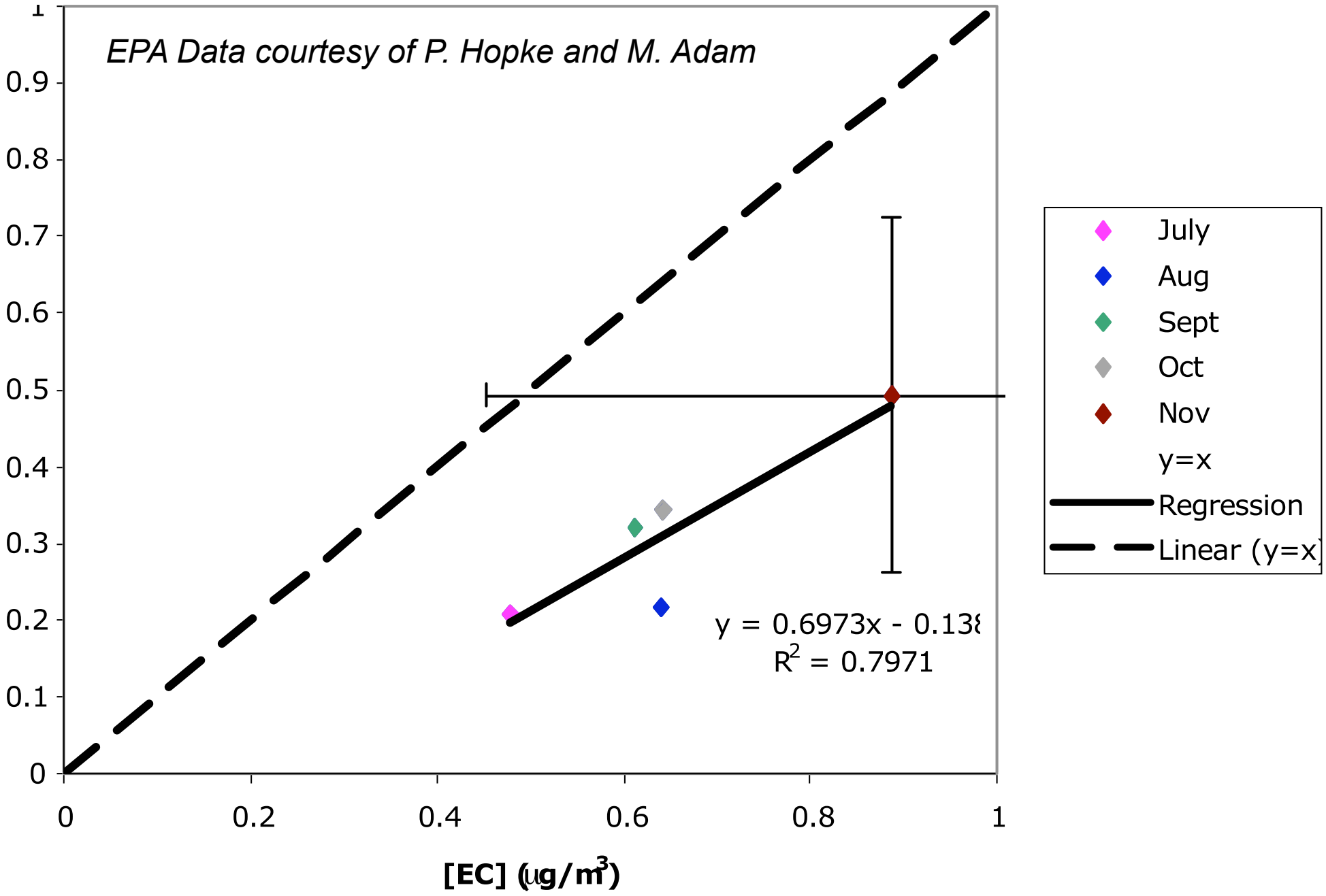
Black carbon is not a sphere



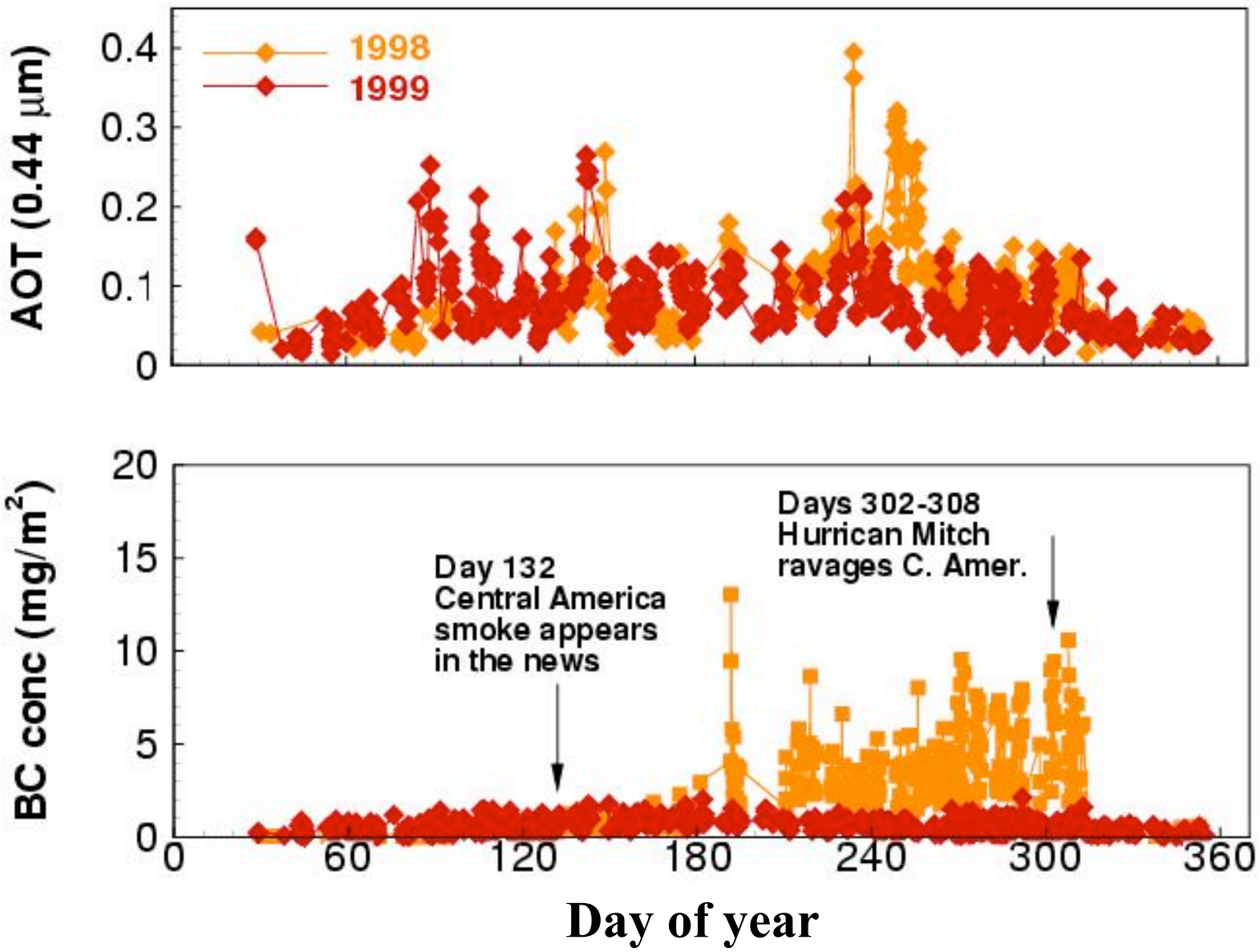
Aggregate soot at Sagres, Portugal;
Li et al., JGR (2003)

Aggregate absorption can be modeled as a loose collection of spheres to within 10-14%
(Mulholland et al,1994; Fuller,1995)

Comparison with Sunset Labs EC/OC Analyzer at Baltimore EPA Supersite

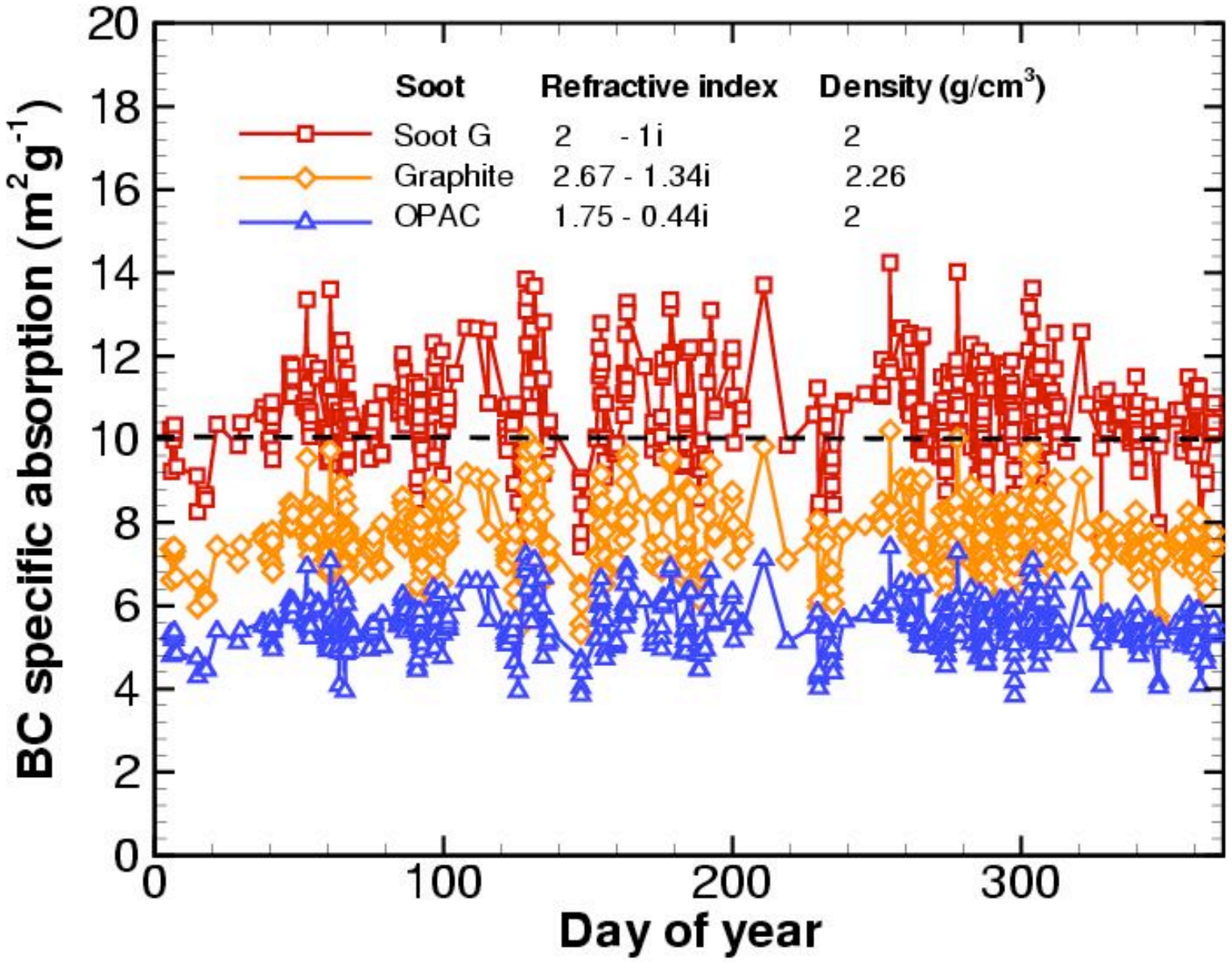


Sevilleta, New Mexico



Sensitivity of specific absorption (550 nm)to BC refractive index

GSFC, 2000



Sensitivity of specific absorption (550 nm) to BC refractive index

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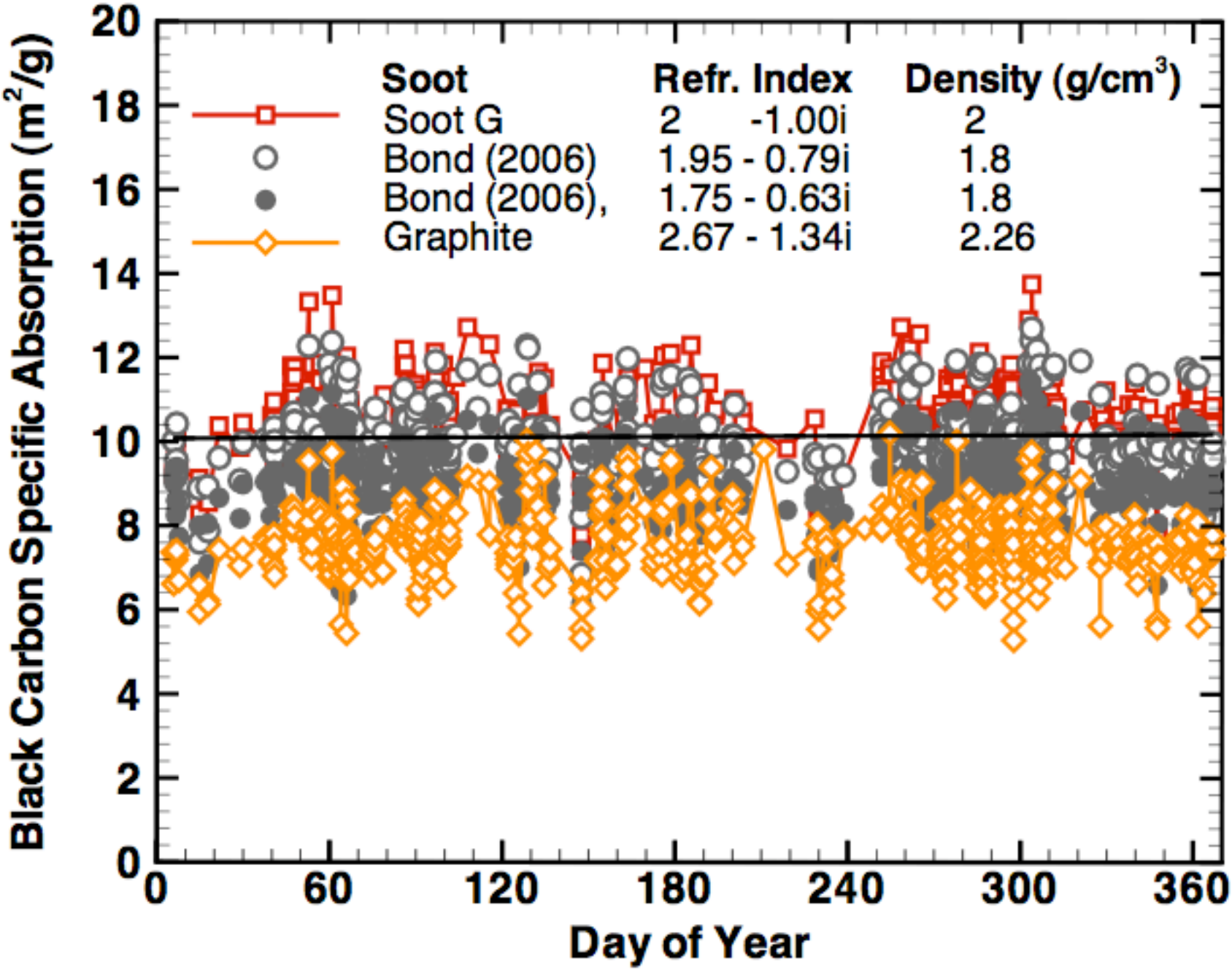
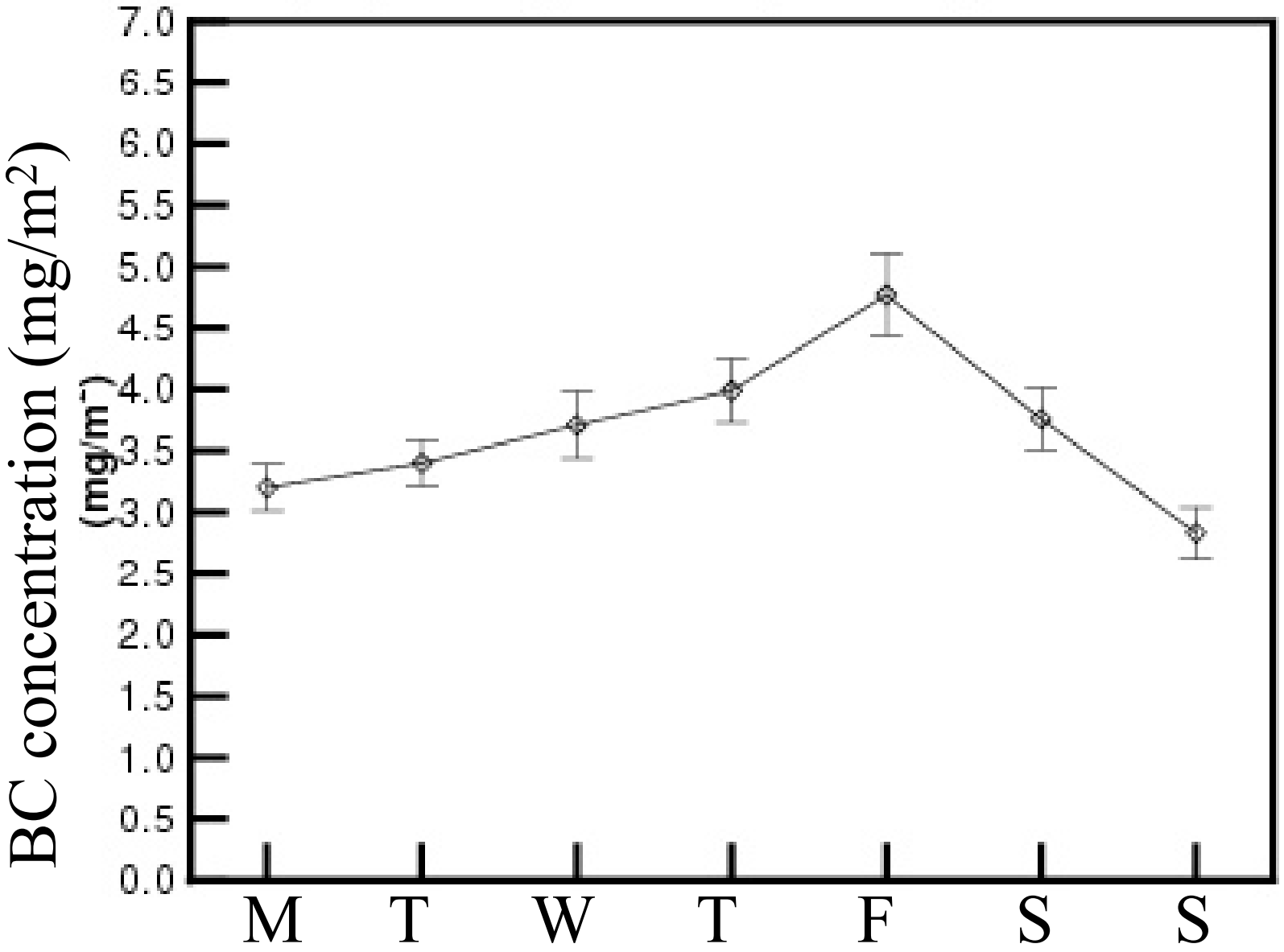
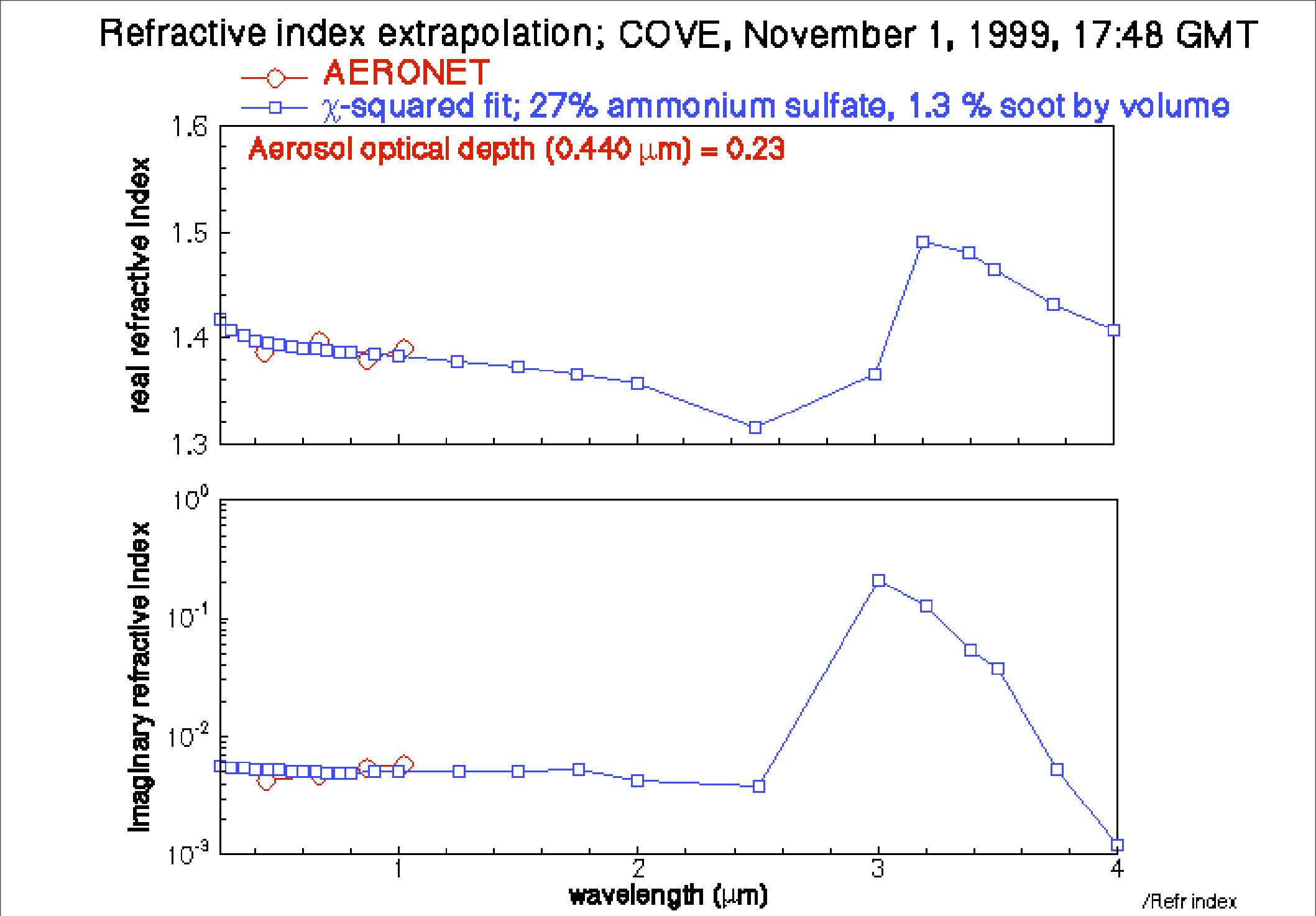


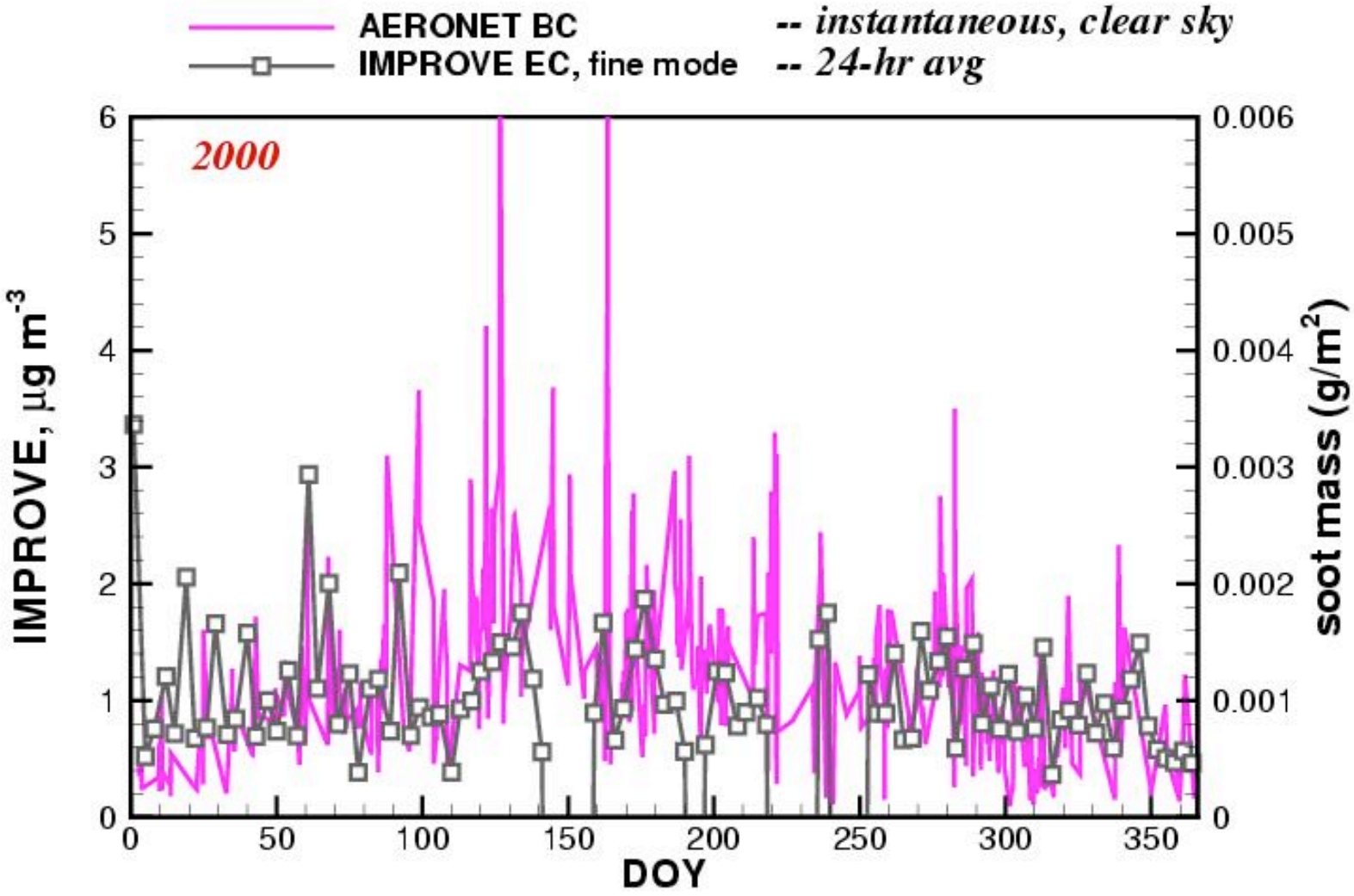
Day of Week in Mexico City

1999-2002

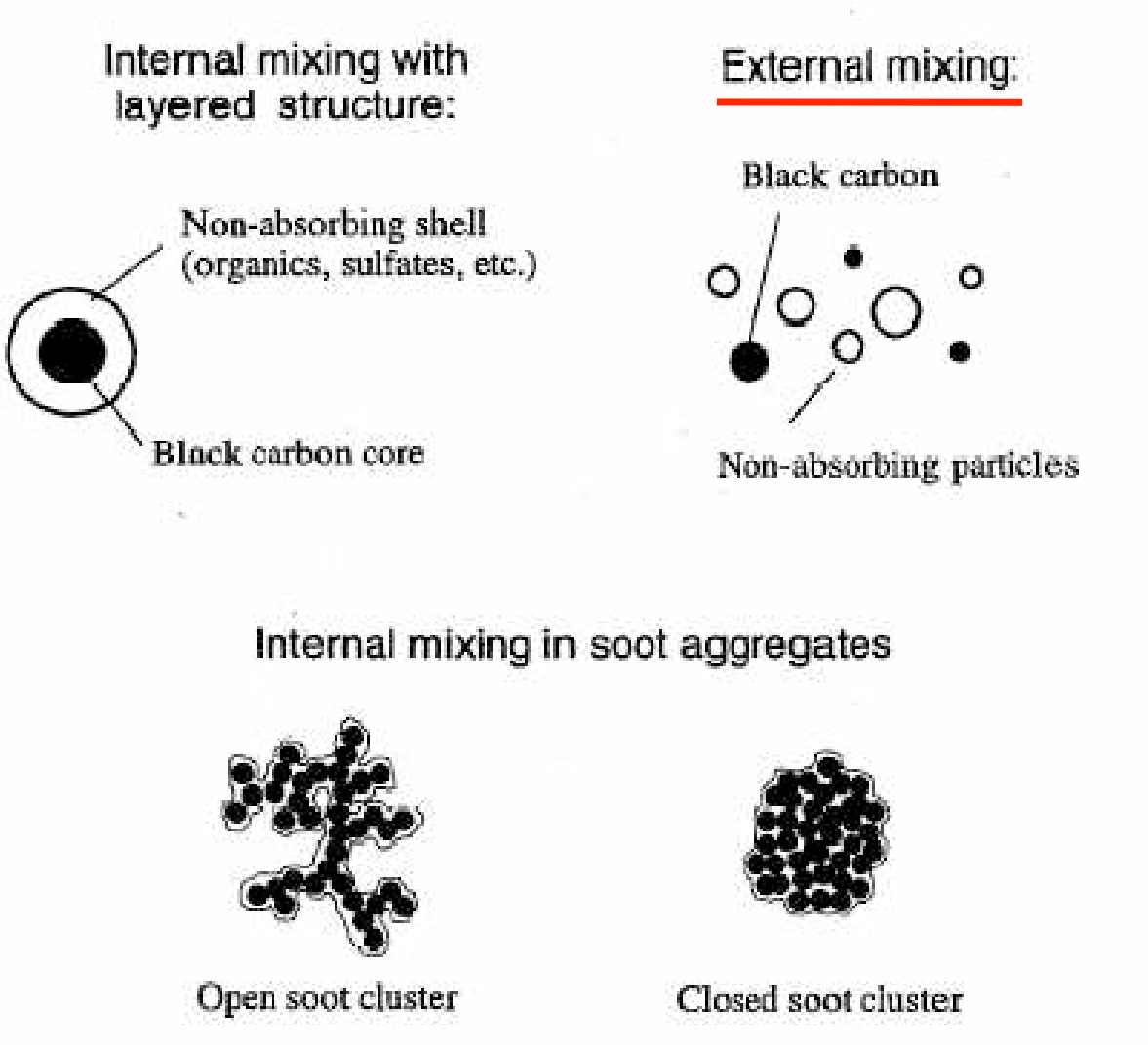


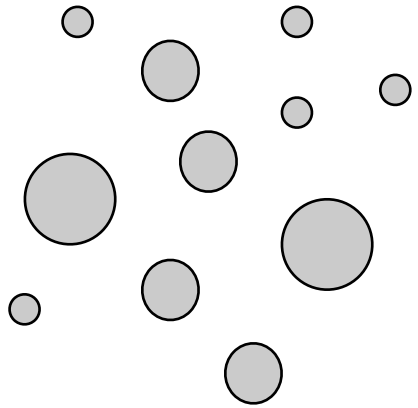

Refractive index extrapolation; COVE, November 1, 1999, 17:48 GMT
AERONET
χ-squared fit; 27% ammonium sulfate, 1.3 % soot by volume
Aerosol optical depth (0.440 μm) = 0.23
real refractive index
1.6
1.5
1.4
1.3
imaginary refractive index
10^0
10^-1
10^-2
10^-3
1
2
3
4
wavelength (μm)
/Refr index

GSFC and WASH1 comparison
21 km between sites



Internal and External Aerosol Mixtures

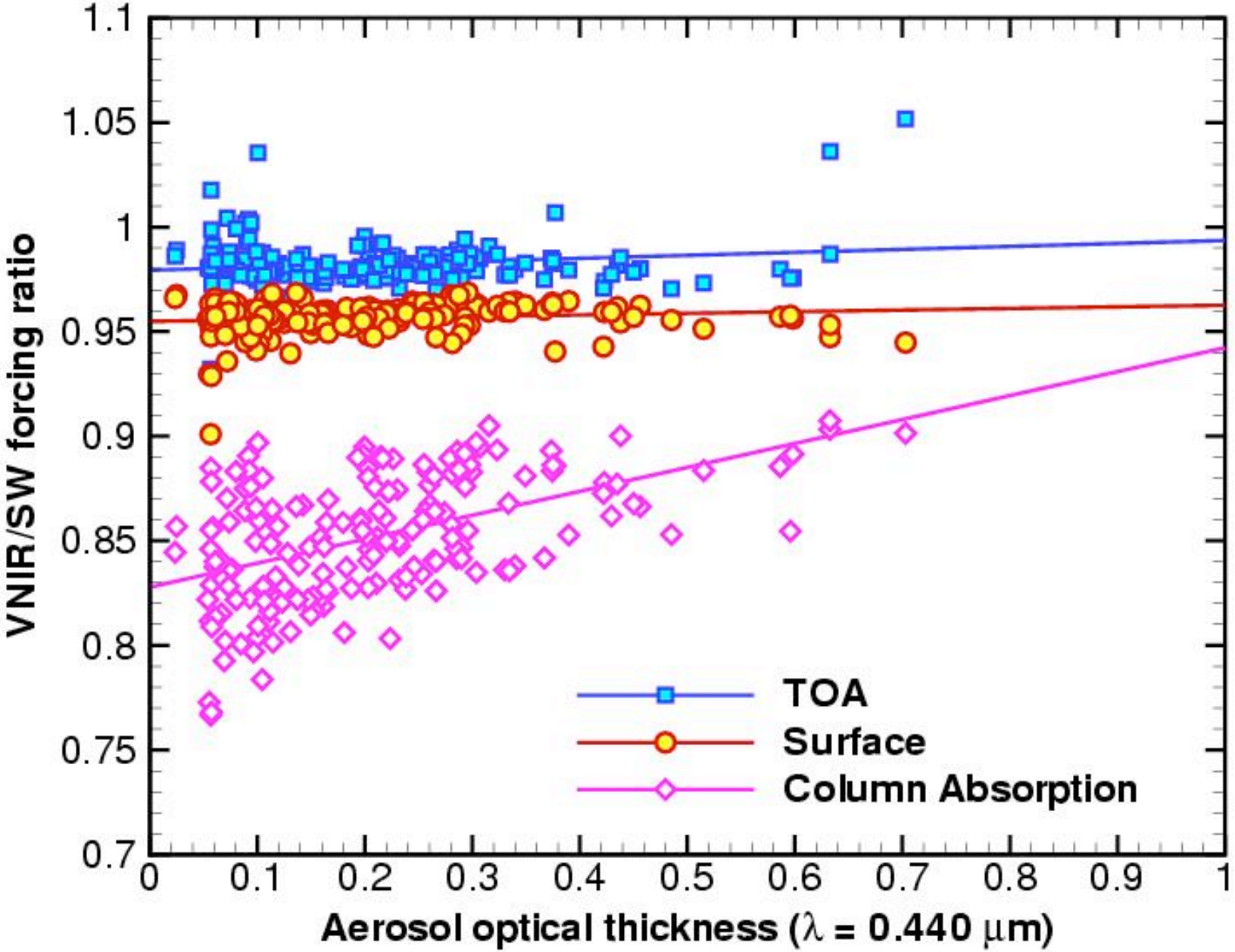




Internal mixture

Martins et al., JGR, 1998

Fraction of aerosol forcing at RSS wavelengths

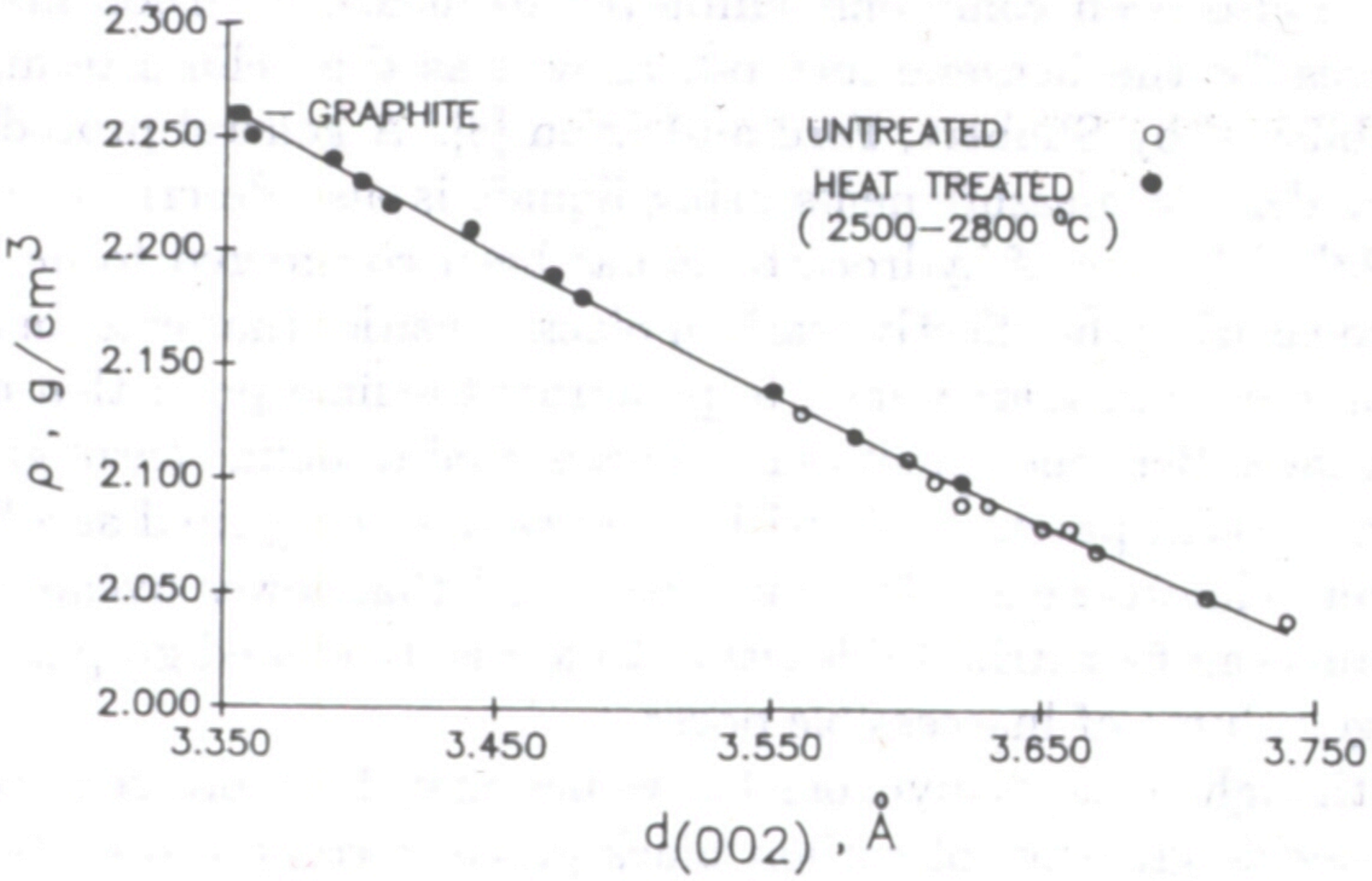


Model comparison to principle plane radiances

- **Principle plane represents an independent measurement**
- **Homogenous troposphere of aerosols and molecules**
- **Ozone absorption in stratosphere using TOMS dataset**

Compare average calculated radiance to measurements at four scanning wavelengths (0.44, 0.67, 0.87, 1.02 μm)

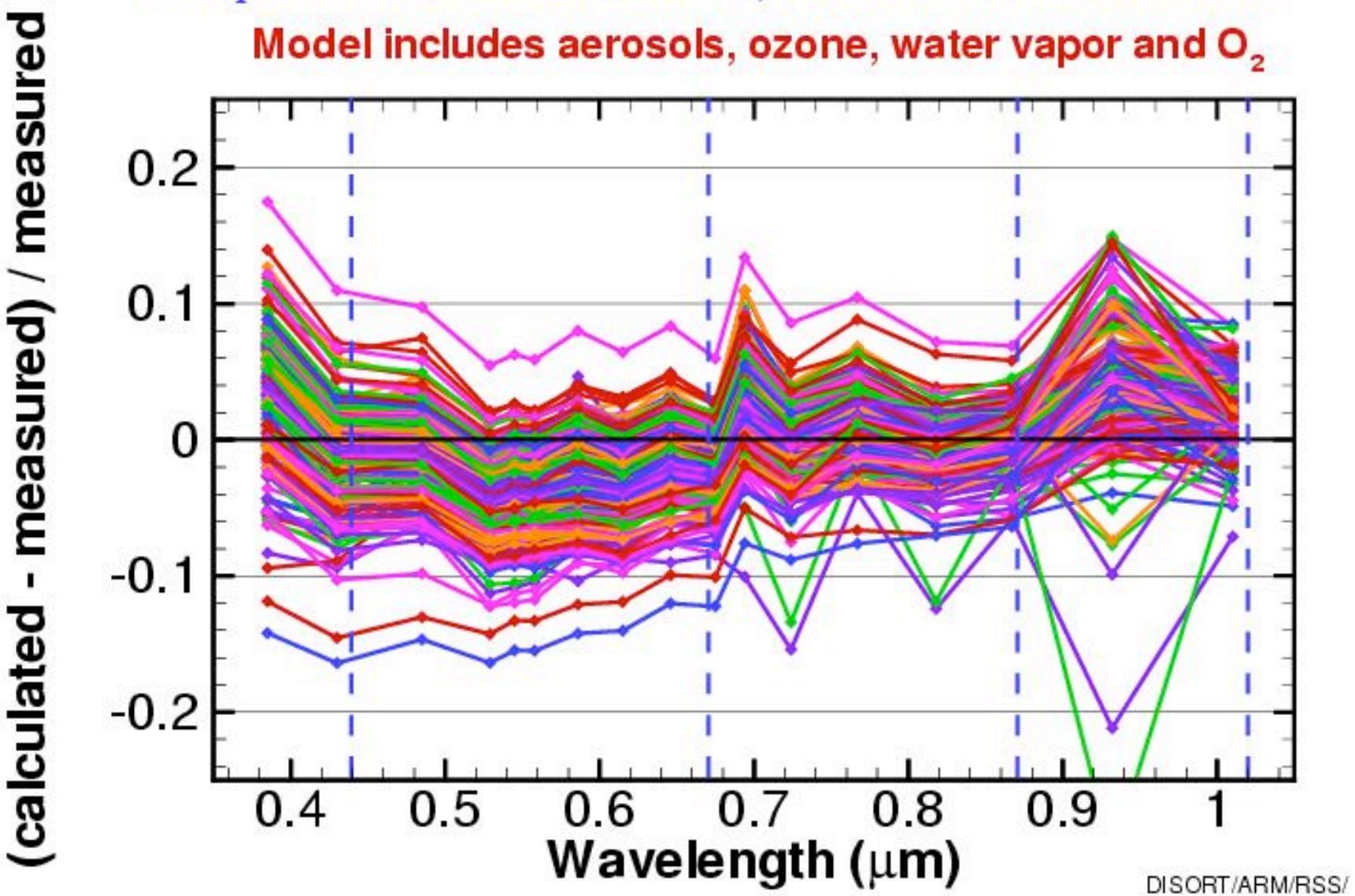
$$\overline{Error} = \frac{1}{4}\sum_{j=1}^{4}\frac{I_j^{calc} - I_j^{meas}}{I_j^{meas}}$$


2.300
2.250
2.200
2.150
2.100
2.050
2.000
ρ , g/cm3
— GRAPHITE
UNTREATED
HEAT TREATED
(2500–2800 °C)
3.350
3.450
3.550
3.650
3.750
d(002) , Å

Irradiance errors in the k-distribution bands

Comparison with RSS at ARM CF, 193 retrievals in 2000-2002.

Model includes aerosols, ozone, water vapor and O_2



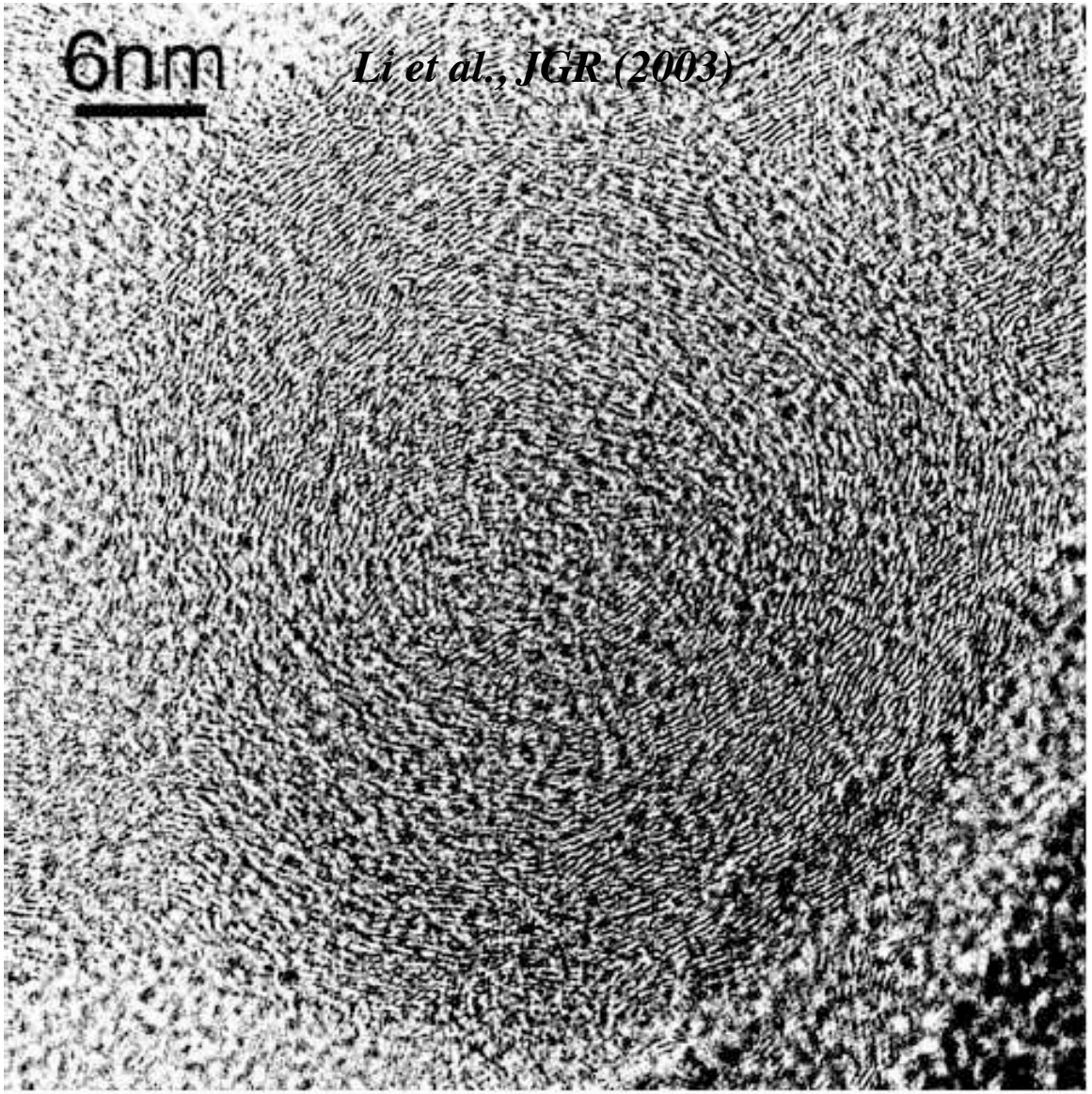
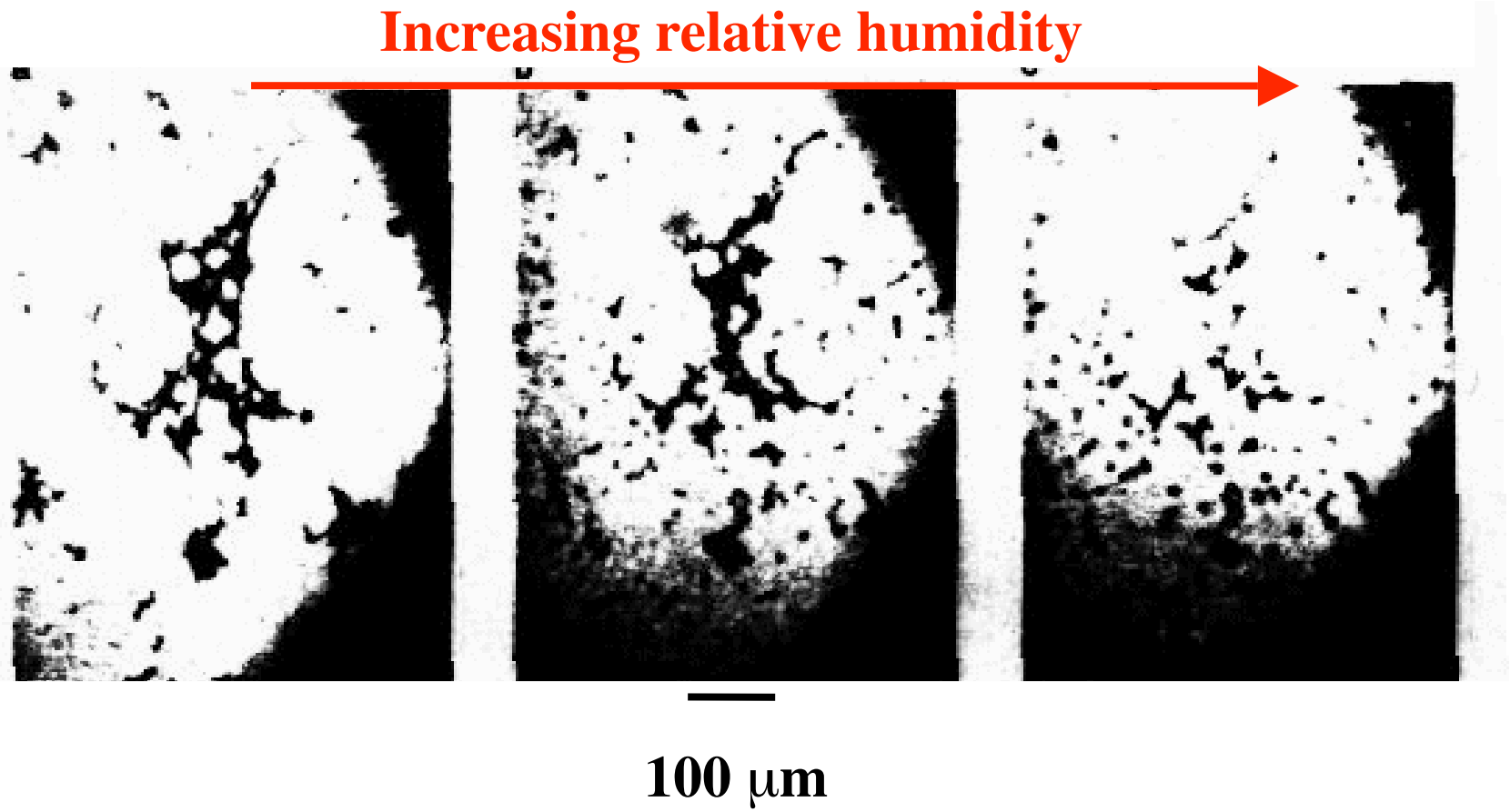


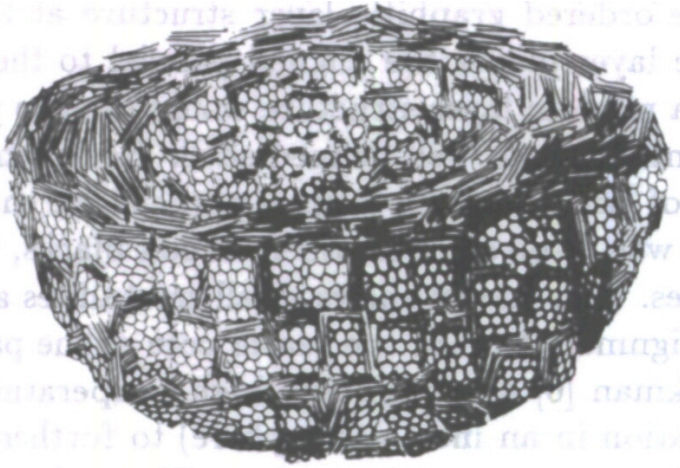
Figure 8. High-resolution TEM image of soot from Sagres showing the discontinuous onion-like structure of graphitic layers.

Relative Humidity Effects on Soot Aggregates



Hallet et al., Aerosol Sci. Tech., 1989

What is Black Carbon?

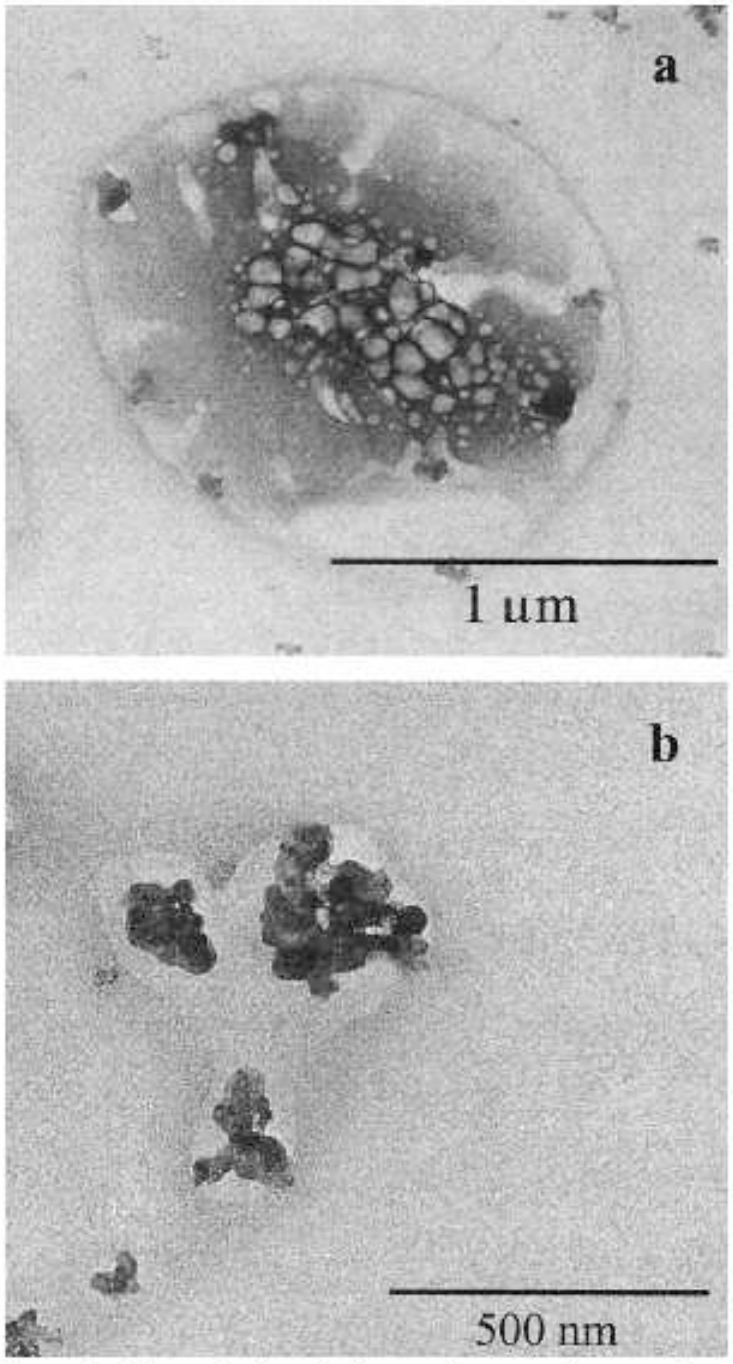


Hess and Herd, Carbon Black (1993)

- **Byproduct of incomplete combustion**
  - **fossil fuel burning**
  - **biomass burning**
- **Graphitized**
- **Other names:**

carbon blacks	**produced in controlled conditions**
soot	**atmospheric; contains impurities**
elemental carbon	**measured by thermal analysis**
black carbon	**measured by optical absorption**

Internal mixtures at an urban location

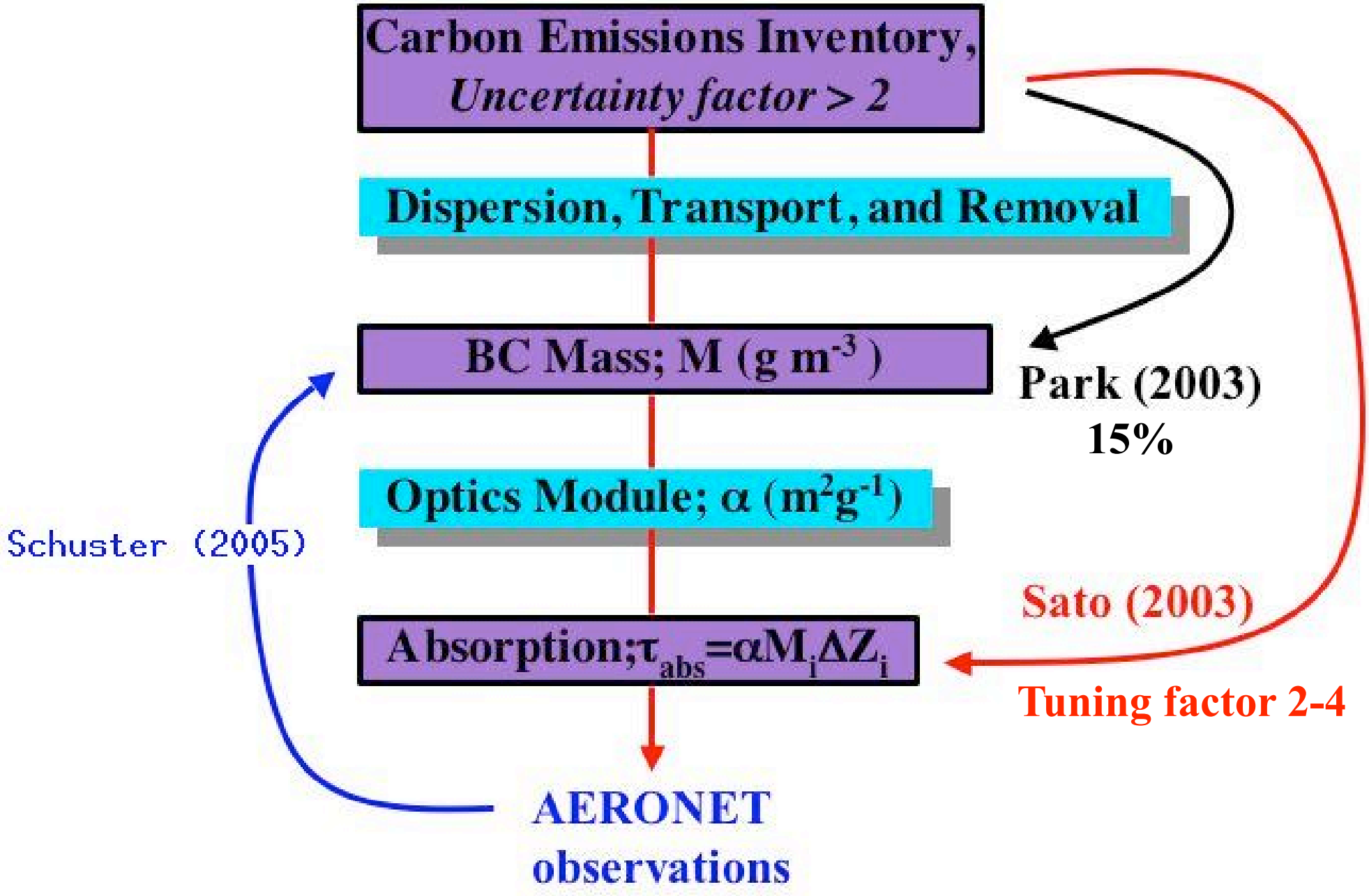


Ammonium sulfate particles with soot inclusions

- **Lindenberg Aerosol Characterization Experiment (LACE 98)**
- **70 km southeast of Berlin**
- **carbon/sulfate mixtures found in 4-49 volume percent of all particles**
- **typical soot fraction is 5-10 % by volume; values up to 50% observed**

Ebert et al., JGR, 2002

Modeling Global Black Carbon Absorption



What do we know about global black carbon concentrations and absorption?

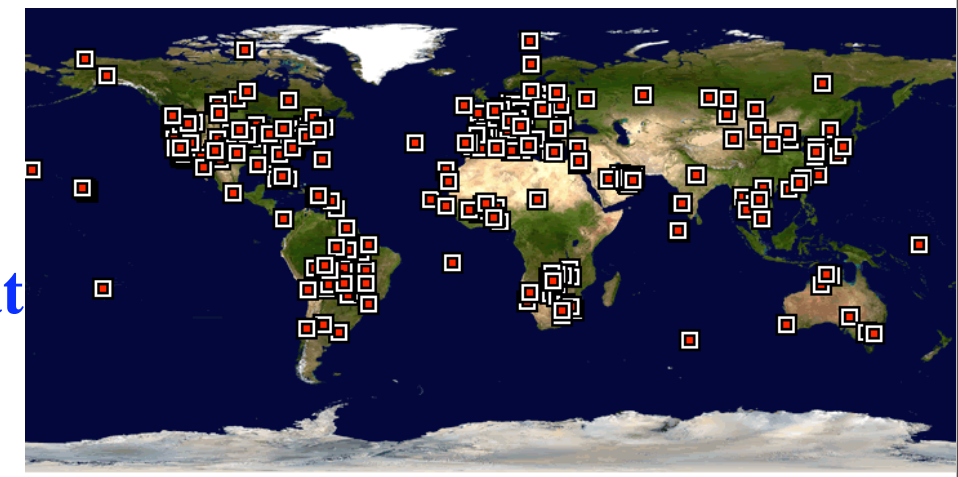
- **Satellite measurements unavailable, so we rely on models**
- **Modeled emission inventories are uncertain by *at least* a factor of 2**
- **Measurements for testing models are inadequate**

  Not enough surface measurements

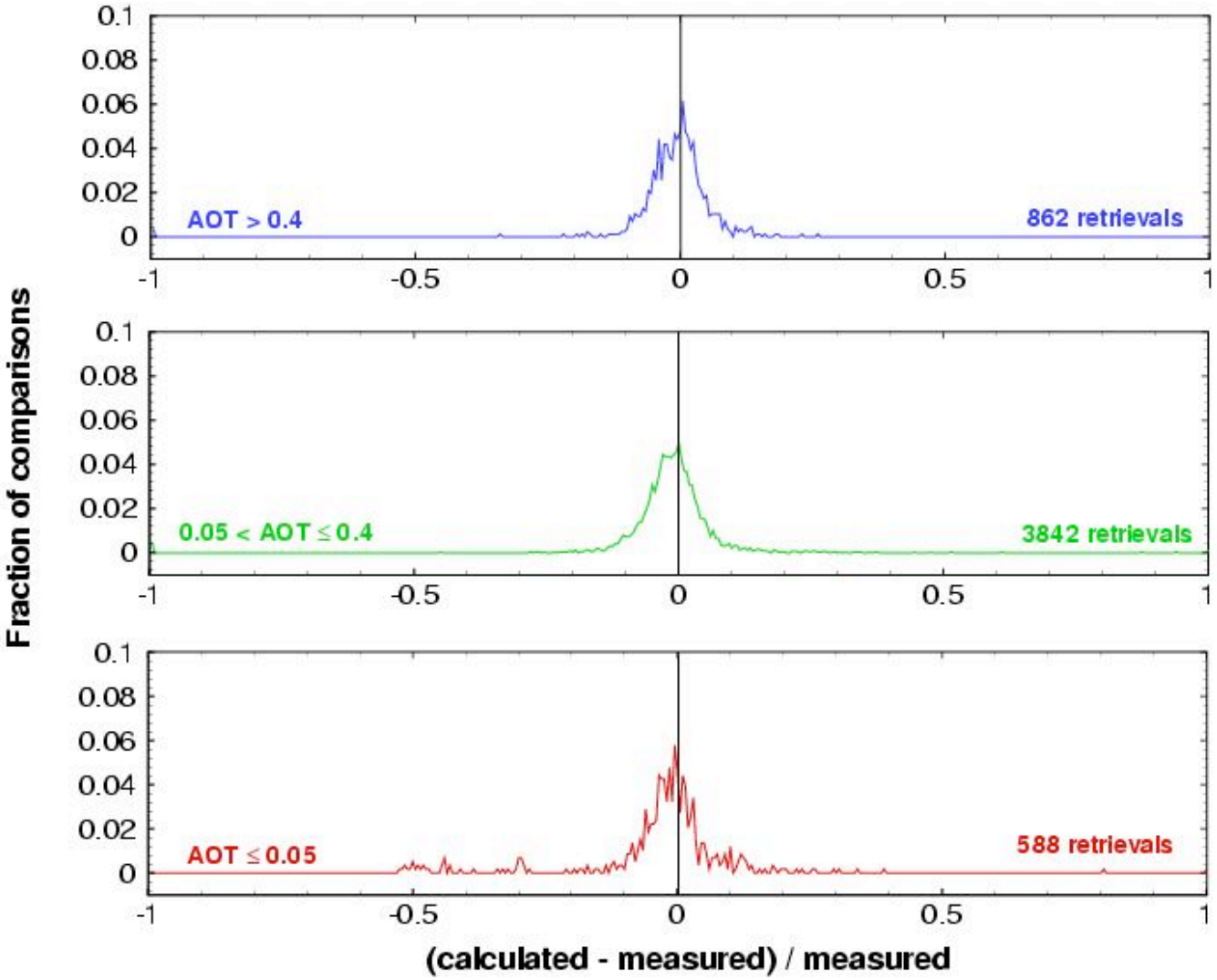
  Vertical distribution available only during field missions

  More measurements are needed

- **AERONET provides radiometric retrievals of column aerosol size distributions and refractive index at 180+ locations**

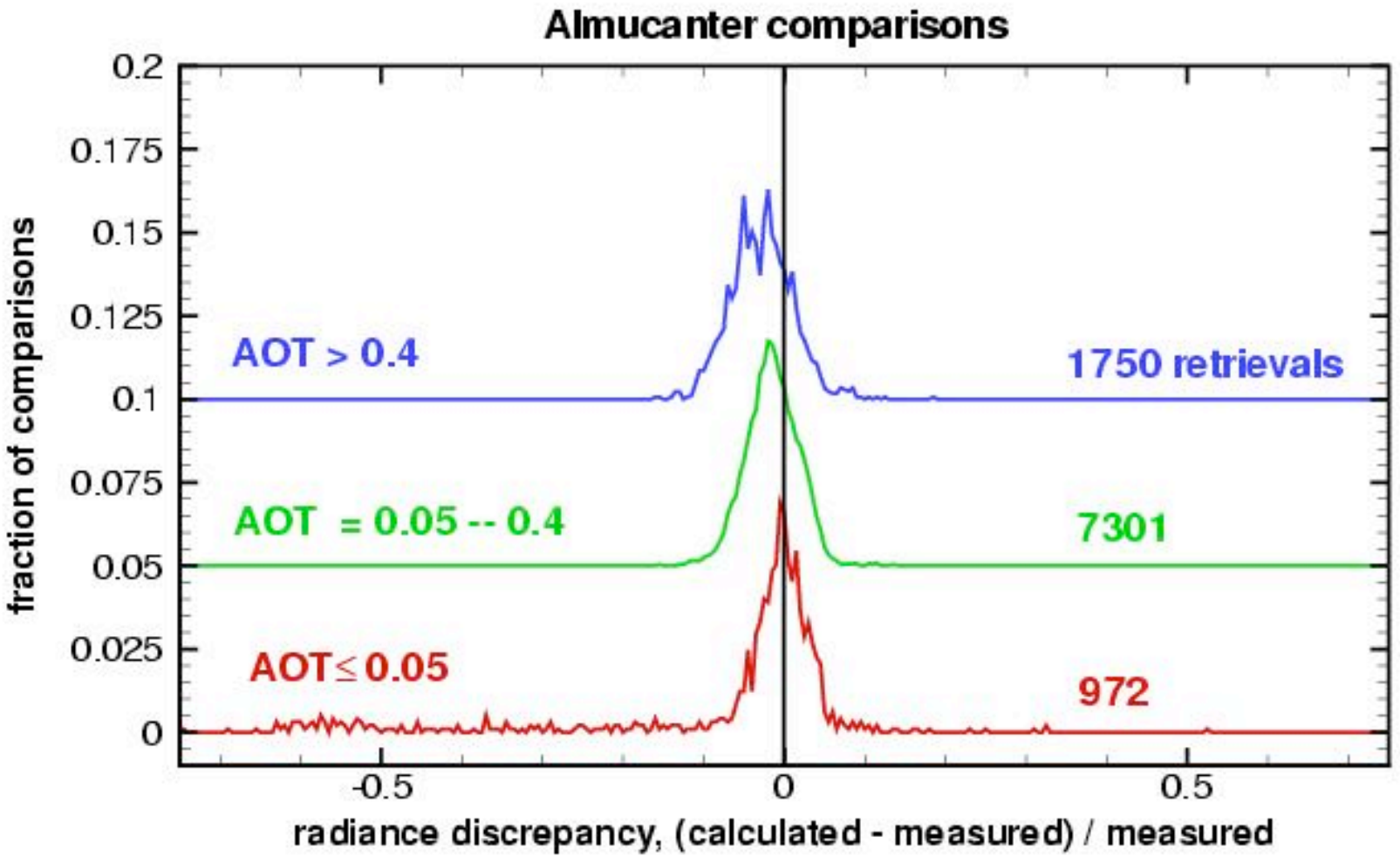


Principle plane comparisons



AOT	Fraction of errors <5%	<10%	< 20%
> 0.4	0.73	0.94	0.99
0.05-0.4	0.68	0.90	0.98
< 0.05	0.63	0.83	0.91

Averaged over all angles and all wavelengths



Comparison of calculated and measured almucantar radiances for three ranges of aerosol optical thickness. Fraction of errors less than 5, 10, and 20 percent shown below.

AOT	5%	10%	20%
> 0.4	0.66	0.96	0.99
0.05-0.4	0.83	0.98	0.99
< 0.05	0.74	0.82	0.86