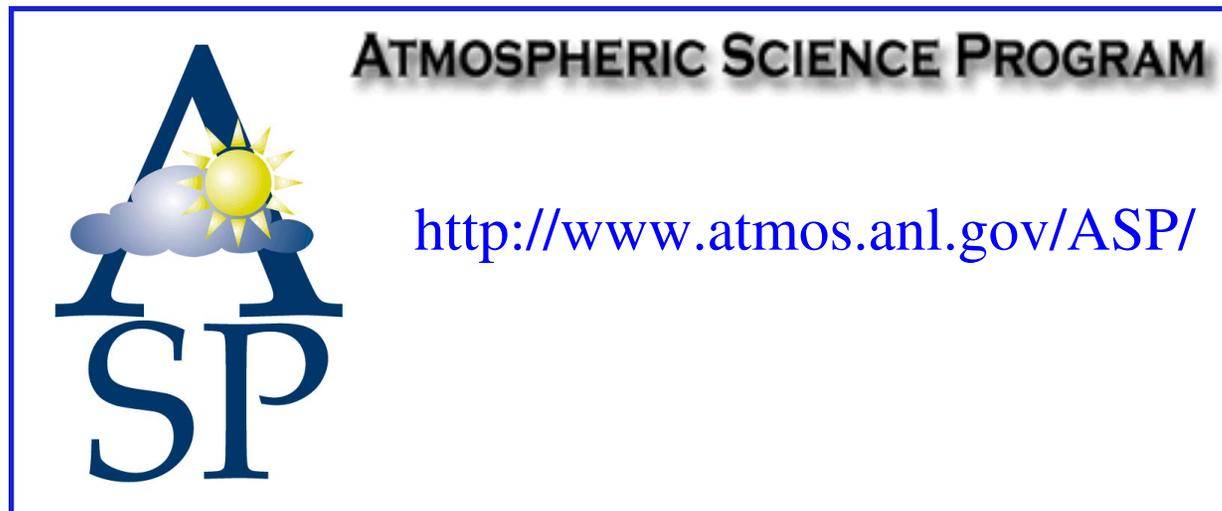




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Stephen E. Schwartz

ARM Aerosol Working Group

Albuquerque NM

March 22, 2004

NEW ASP SCIENCE FOCUS

Aerosol radiative forcing of climate

Enhance the scientific knowledge needed to simulate and predict radiative forcing and other climatic effects of aerosols



AEROSOL FORCING OF CLIMATE AND CLIMATE CHANGE

Aerosol forcing of climate is difference between radiative flux with aerosol minus radiative flux without aerosol.

Aerosol forcing of climate change is difference between radiative flux with present aerosol minus radiative flux with preindustrial aerosol.

Determination of aerosol forcing of climate change requires:

- *Attribution of aerosol to anthropogenic vs. natural, and*
- *Determination of aerosol forcing for natural and total aerosol.*

This inherently involves atmospheric chemistry [and aerosol microphysics and optical properties and cloud microphysics] as well as atmospheric radiation.

ARM *Atmospheric Radiation Measurement Program*

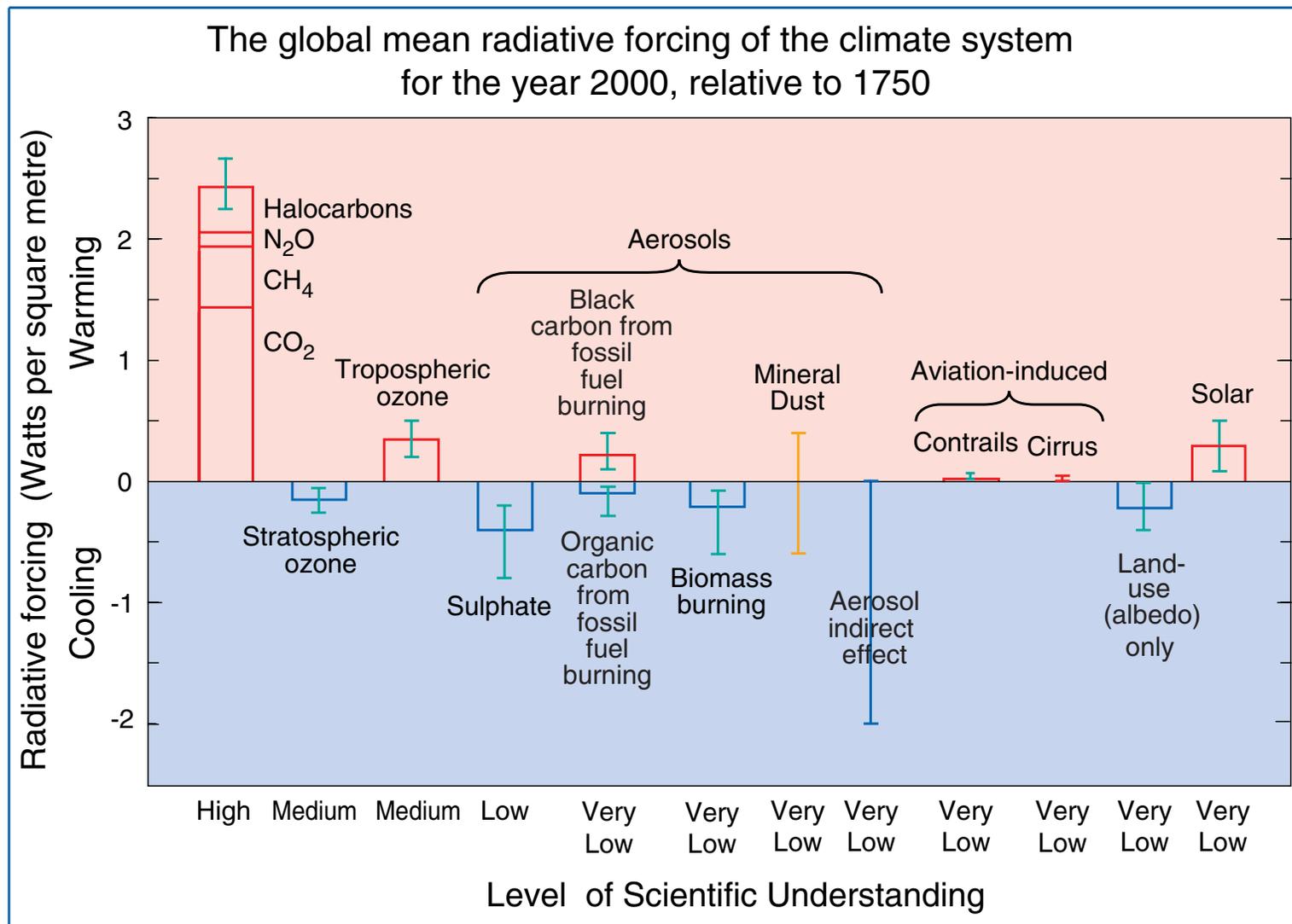


ATMOSPHERIC SCIENCE PROGRAM



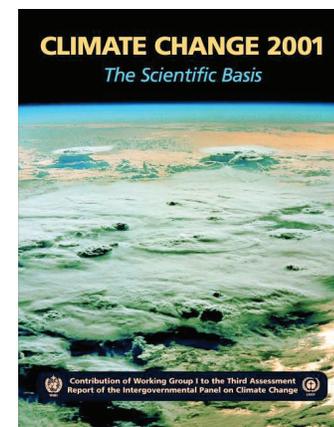
<http://www.atmos.anl.gov/ASP/>

RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD IPCC (2001)



Summary for Policymakers

A Report of Working Group I of the Intergovernmental Panel on Climate Change



DIRECT EFFECT

DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE AEROSOL

$$\overline{\Delta F_R} = -\frac{1}{2} F_T T^2 (1 - A_c)(1 - R_s)^2 \cdot \overline{\beta} \alpha_{\text{SO}_4^{2-}} f(\text{RH}) \cdot Q_{\text{SO}_2} Y_{\text{SO}_4^{2-}} \left(\frac{\text{MW}_{\text{SO}_4^{2-}}}{\text{MW}_S} \right) \tau_{\text{SO}_4^{2-}} / A$$

Geophysics Aerosol Optical Depth
Aerosol Microphysics Column Burden Atmospheric Chemistry

$\overline{\Delta F_R}$ is the area-average shortwave radiative forcing due to the aerosol, W m^{-2}

F_T is the solar constant, W m^{-2}

A_c is the fractional cloud cover

T is the fraction of incident light transmitted by the atmosphere above the aerosol

R_s is the albedo of the underlying surface

$\overline{\beta}$ is upward fraction of the radiation scattered by the aerosol,

$\alpha_{\text{SO}_4^{2-}}$ is the scattering efficiency of **sulfate and associated cations** at a reference low relative humidity, $\text{m}^2 (\text{g SO}_4^{2-})^{-1}$

$f(\text{RH})$ accounts for the relative increase in scattering due to relative humidity

Q_{SO_2} is the source strength of anthropogenic SO_2 g S yr^{-1}

$Y_{\text{SO}_4^{2-}}$ is the fractional yield of emitted SO_2 that reacts to produce sulfate aerosol

MW is the molecular weight

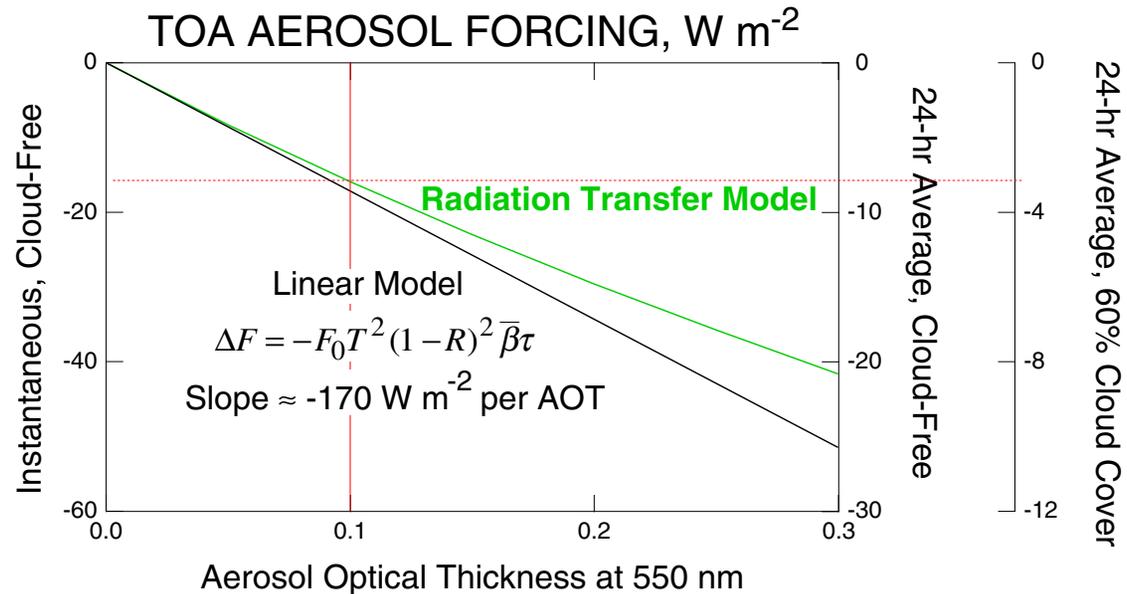
$\tau_{\text{SO}_4^{2-}}$ is the sulfate lifetime in the atmosphere, yr

A is the area of the geographical region under consideration, m^2

DIRECT AEROSOL FORCING

Comparison of linear formula and radiation transfer model

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



Forcing is highly sensitive to modest aerosol loadings.

Global-average AOT 0.1 corresponds to global-average forcing $\sim -3 \text{ W m}^{-2}$.

Linear model is accurate and convenient, especially for error budgets.

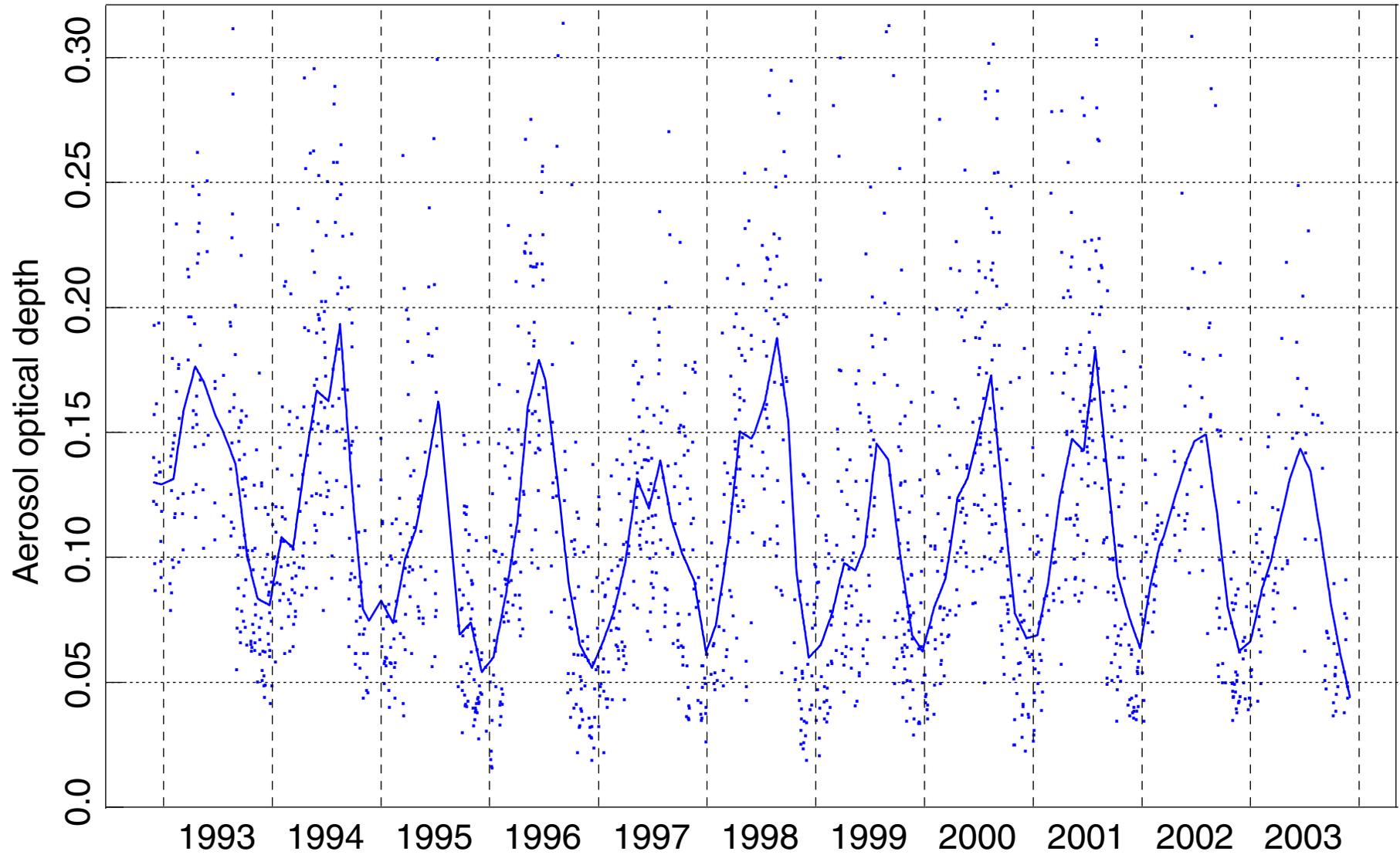
Forcing per optical depth depends on particle size.

Top-of-atmosphere forcing depends on single scattering albedo and surface reflectance.

AEROSOL OPTICAL DEPTH

Determined by sunphotometry

North central Oklahoma - Daily average at 500 nm

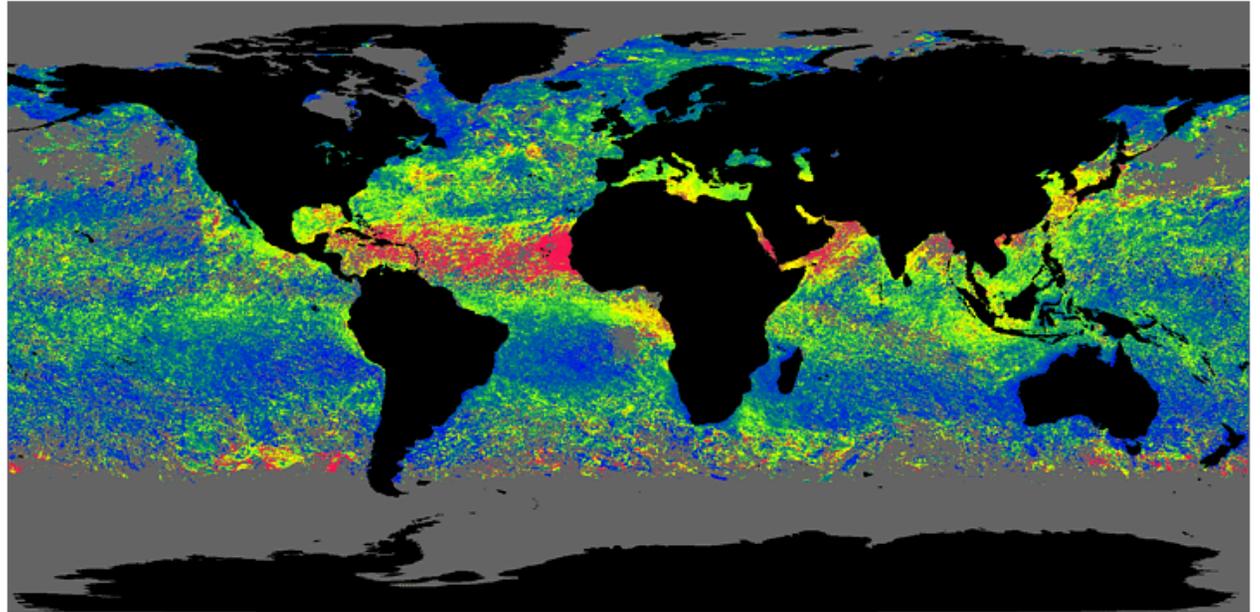
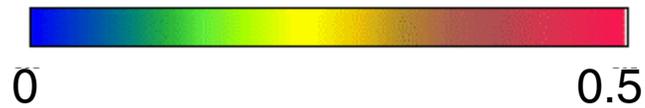


MONTHLY AVERAGE AEROSOL JUNE 1997

Polder radiometer on Adeos satellite

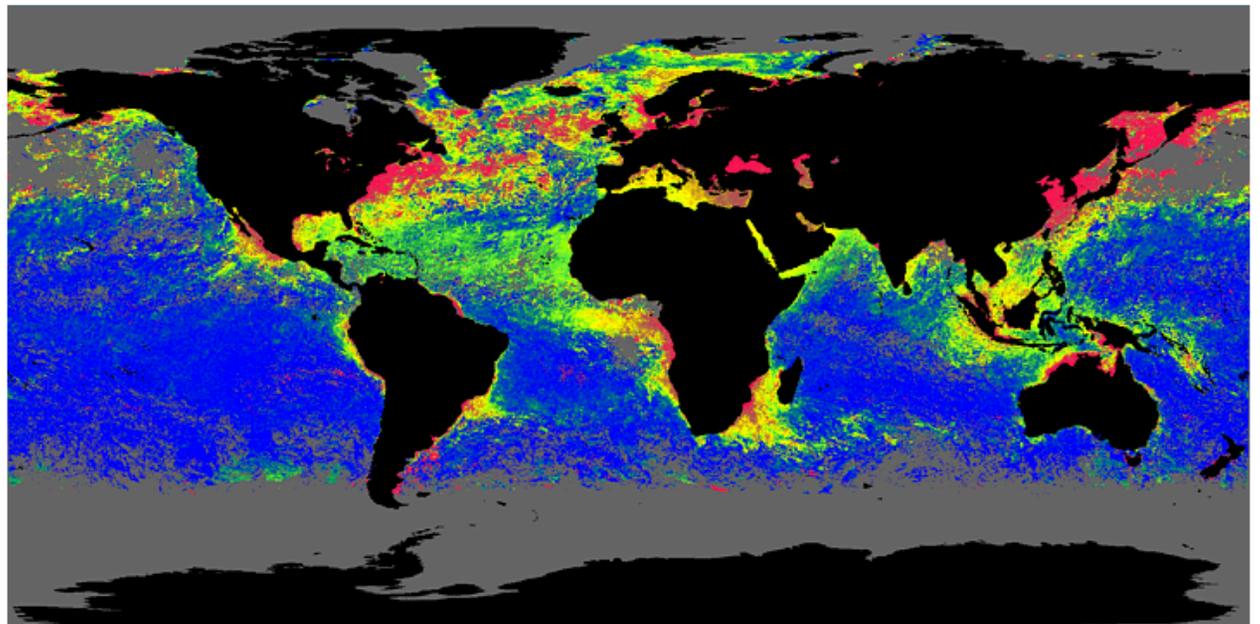
Optical Thickness τ

$\lambda = 865 \text{ nm}$



Ångström Exponent α

$\alpha = -d \ln \tau / d \ln \lambda$



UNCERTAINTY BUDGET FOR *DIRECT* FORCING BY INDUSTRIAL AEROSOLS

Quantity	Central Value	2/3 Uncertainty Range	Uncertainty Factor
Total emission of anthropogenic OC from fossil fuel burning (Tg/yr)	20	10 to 30	3.0
Atmospheric burden of OC from fossil fuels (Tg)	0.48	0.33 to 0.70	2.1
Total emission of anthropogenic BC from fossil fuel burning (Tg/yr)	7	4.67 to 10.5	2.2
Atmospheric burden of BC from fossil fuel burning (Tg)	0.133	0.11 to 0.16	1.5
Total emission of anthropogenic sulfate from fossil fuel burning (Tg/yr)	69	57.5 to 82.8	1.4
Atmospheric burden of sulfate from fossil fuel burning (Tg S)	0.525	0.35 to 0.79	2.3
Fraction of light scattered into upward hemisphere, $\bar{\beta}$	0.23	0.17 to 0.29	1.7
Aerosol mass scattering efficiency (m^2g^{-1}), α_s	3.5	2.3 to 4.7	2.0
Aerosol single scattering albedo, co-albedo (dry), ω_0 , $1 - \omega_0$	0.92	0.85 to 0.97	1.1, 5
T_a , atmospheric transmittance above aerosol layer	0.87	0.72 to 1.00	1.4
Fractional increase in aerosol scattering efficiency due to hygroscopic growth at RH=80%	2.0	1.7 to 2.3	1.4
Fraction of Earth not covered by cloud	0.39	0.35 to 0.43	1.2
Mean surface albedo, co-albedo	0.15	0.08 to 0.22	2.8, 1.2
Result: If central value is -0.6 Wm^{-2} the 2/3 uncertainty range is from 0.1 to 1.0 Wm^{-2} .			10.0

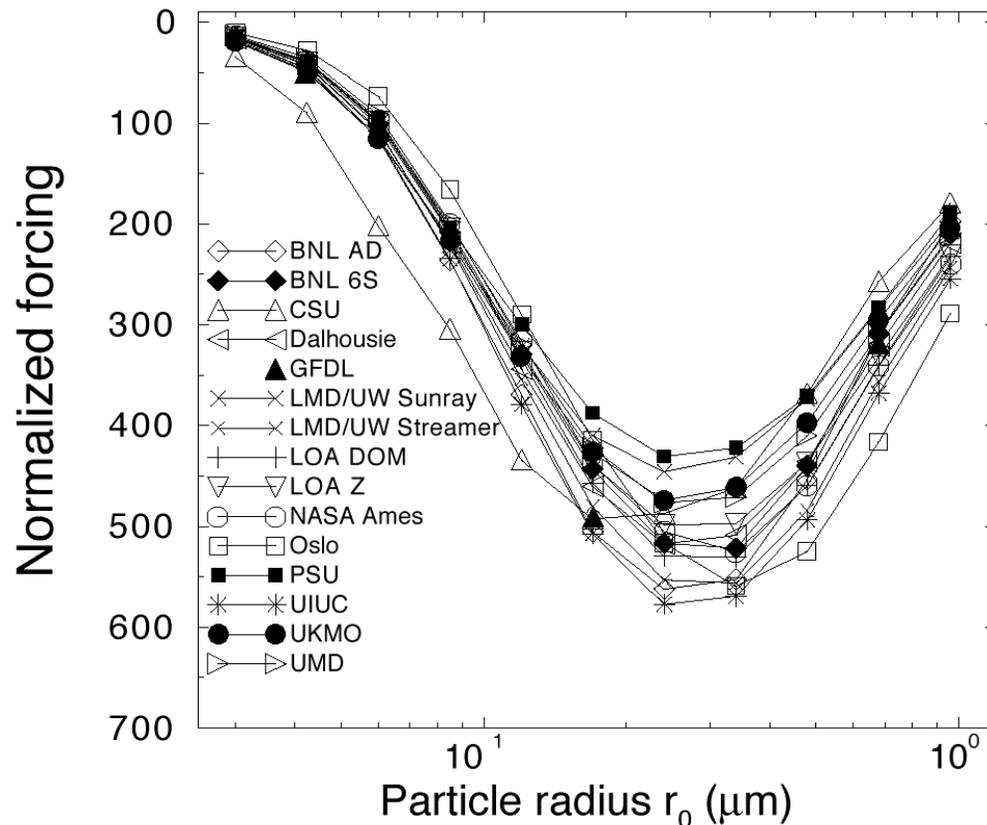
Modified from Penner et al., IPCC, 2001

- *Greatest uncertainties are in chemical, microphysical, and optical properties.*

INTERCOMPARISON OF BROADBAND SHORTWAVE FORCING BY AMMONIUM SULFATE AEROSOL

Normalized global-average forcing: $W \text{ m}^{-2} / \text{g}(\text{SO}_4^{2-}) \text{ m}^{-2}$ or $W / \text{g}(\text{SO}_4^{2-})$

Aerosol optical depth 0.2; surface albedo 0.15



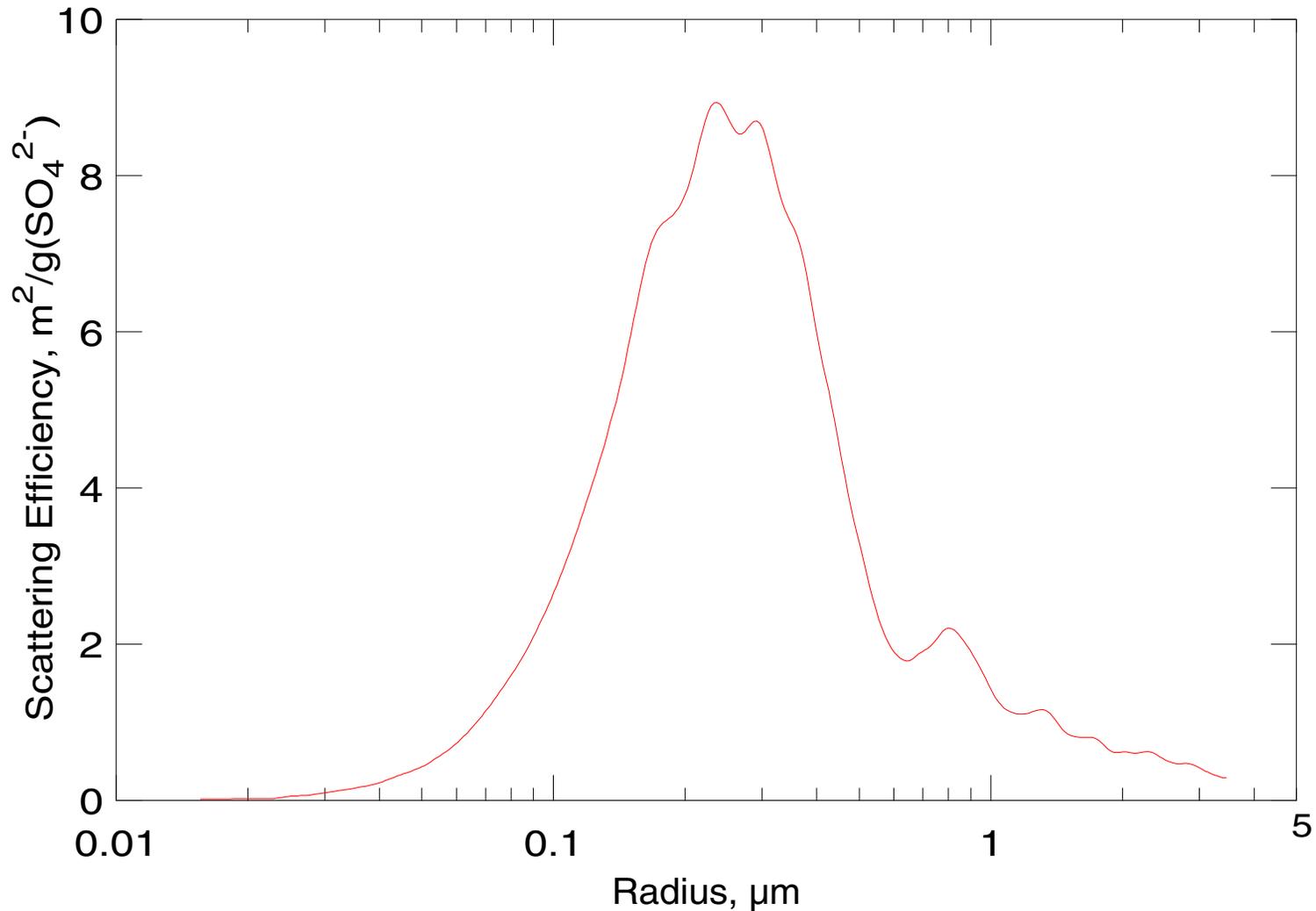
Standard deviation $\sim 8\%$ for 15 models at radius $\sim 200 \text{ nm}$.

Boucher, Schwartz and 28 co-authors, JGR, 1998

LIGHT SCATTERING EFFICIENCY

Dependence on particle radius -- Size matters!

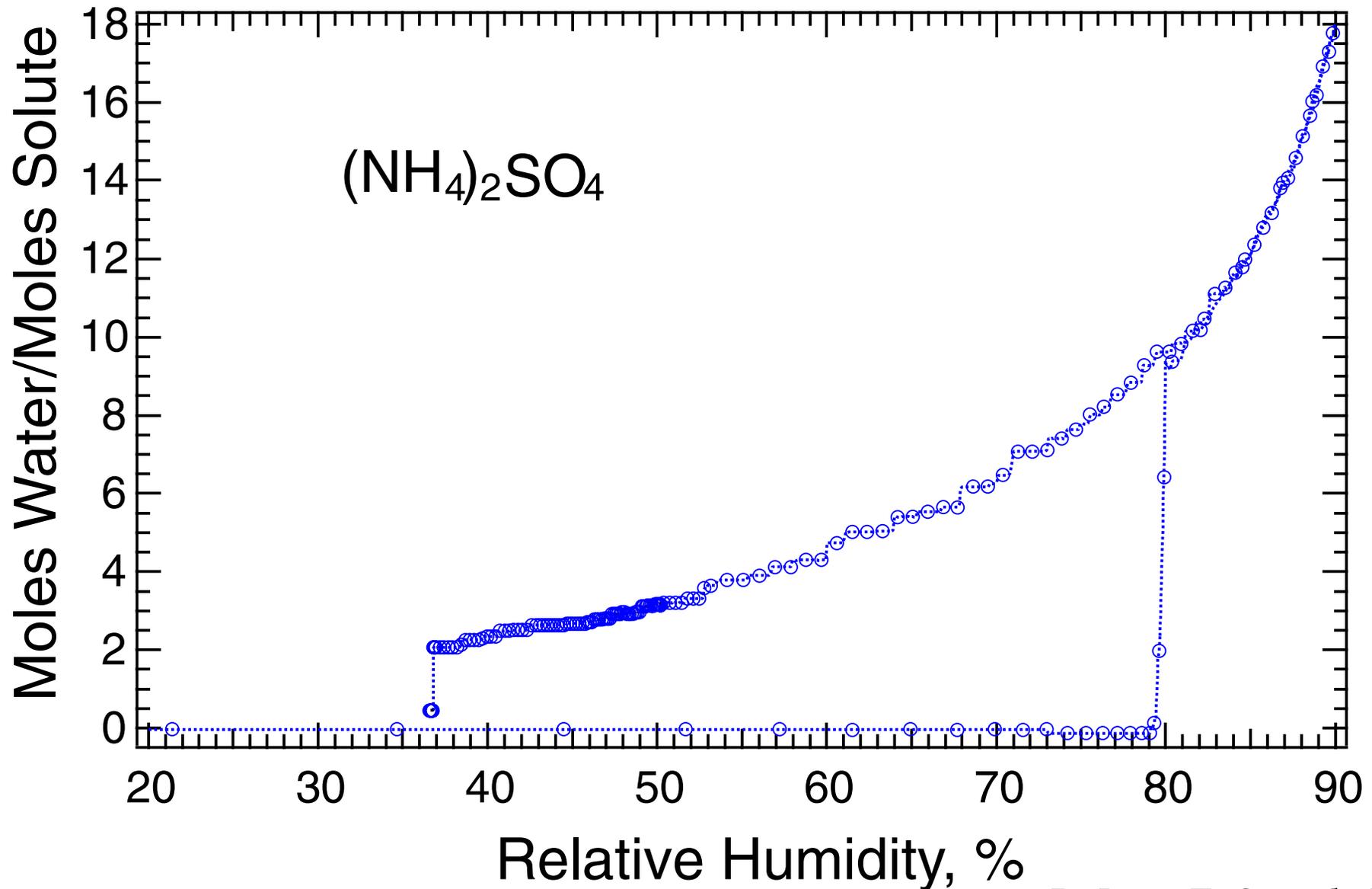
Ammonium Sulfate, 530 nm



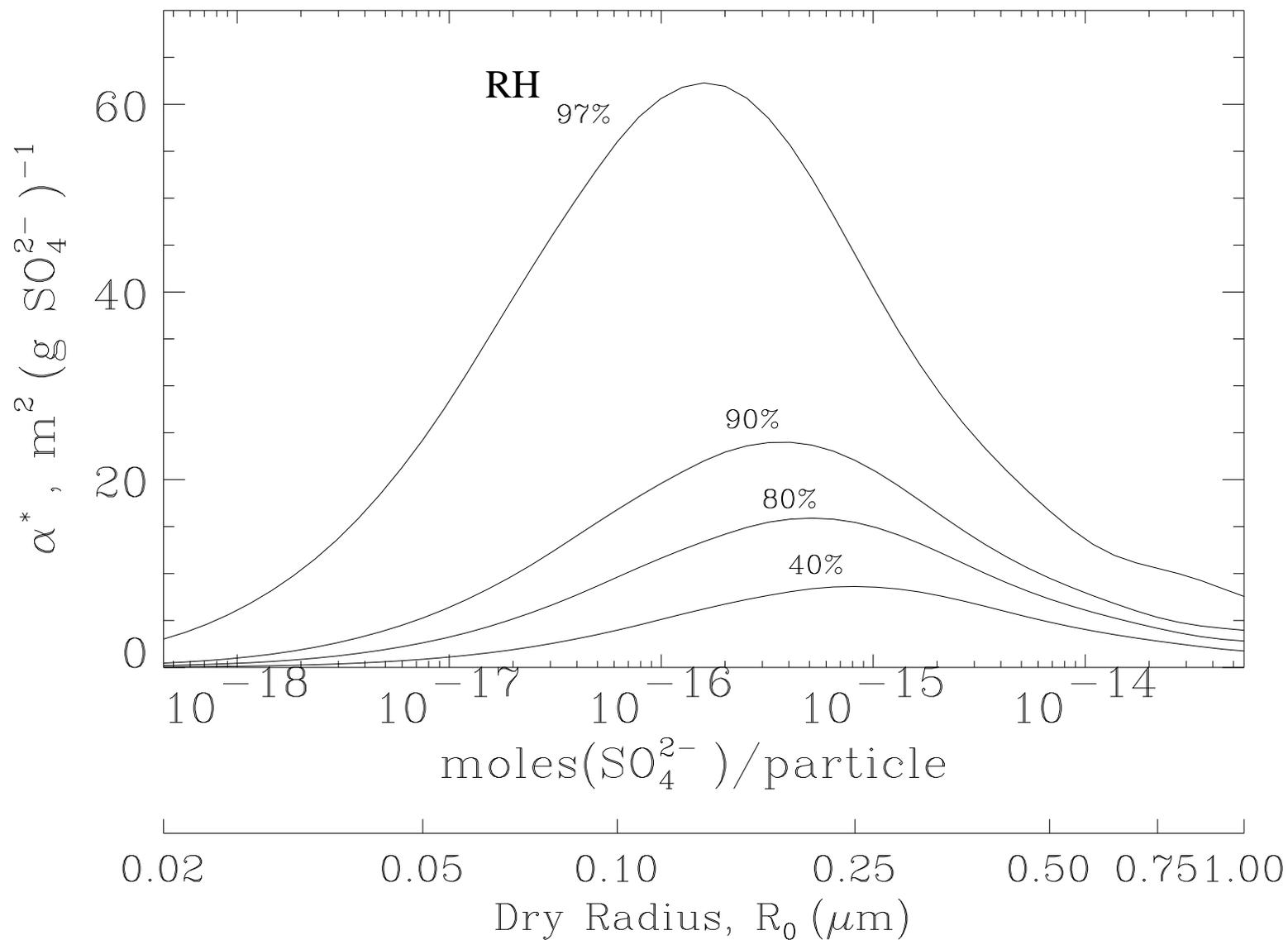
Data of Ouimette and Flagan, 1982

WATER UPTAKE BY HYGROSCOPIC PARTICLE

Dependence on relative humidity



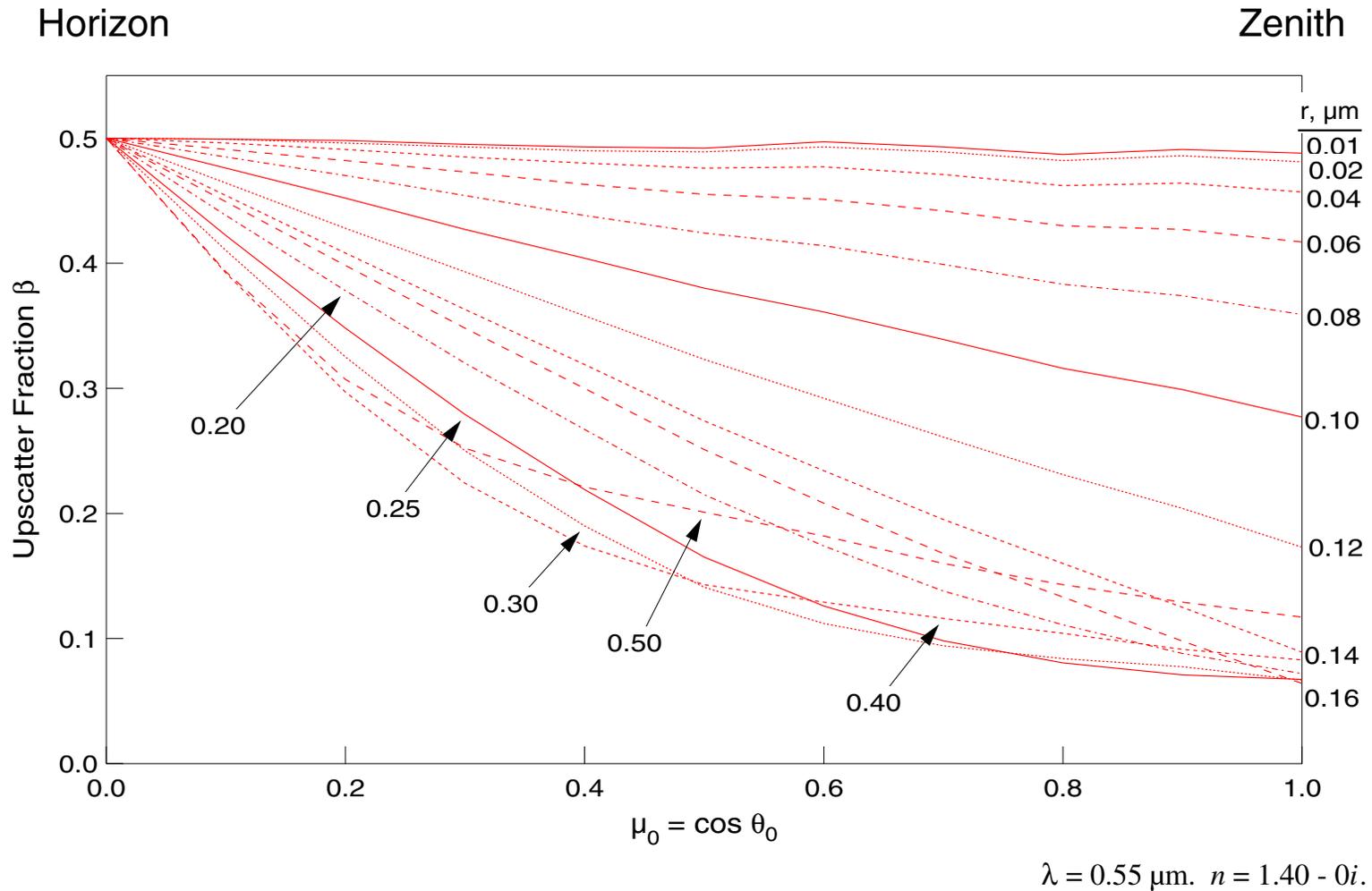
LIGHT SCATTERING EFFICIENCY OF $(\text{NH}_4)_2\text{SO}_4$ DEPENDENCE ON PARTICLE SIZE AND RH



Nemesure et al., JGR, 1995

UPSCATTER FRACTION

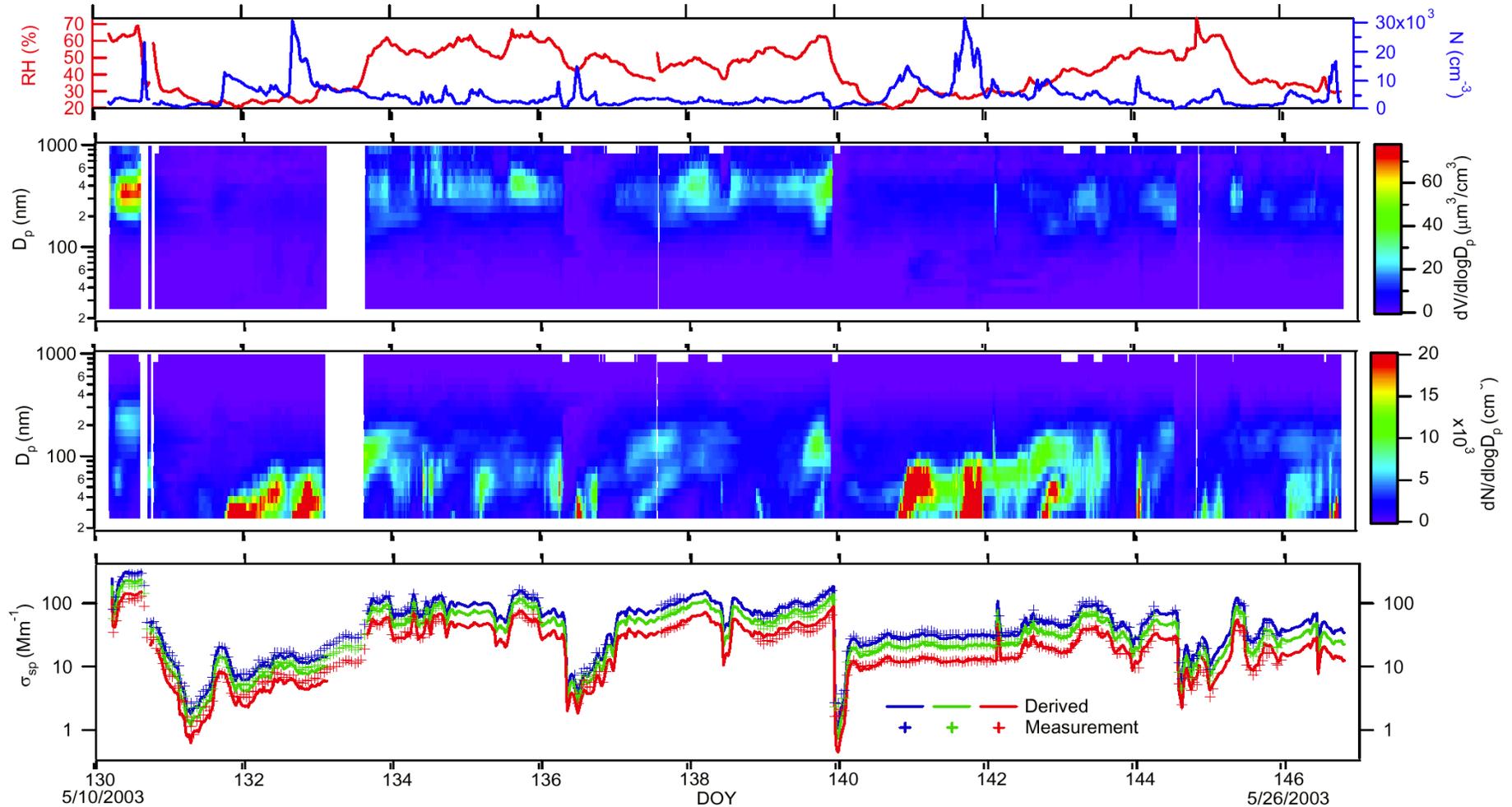
Dependence on solar zenith angle and particle radius



For sun at horizon $\beta = 0.5$ (by symmetry).

For small particles, $r \ll \lambda$, upscatter fraction approaches that for Rayleigh scattering (0.5).

AEROSOL OPTICAL AND MICROPHYSICAL PROPERTIES DURING ARM ACP AEROSOL IOP MAY 2003



N from condensation nucleus counter.

RH as measured in nephelometer.

Index of refraction from optical particle counter on particles selected by DMA; RH dependence from TDMA.

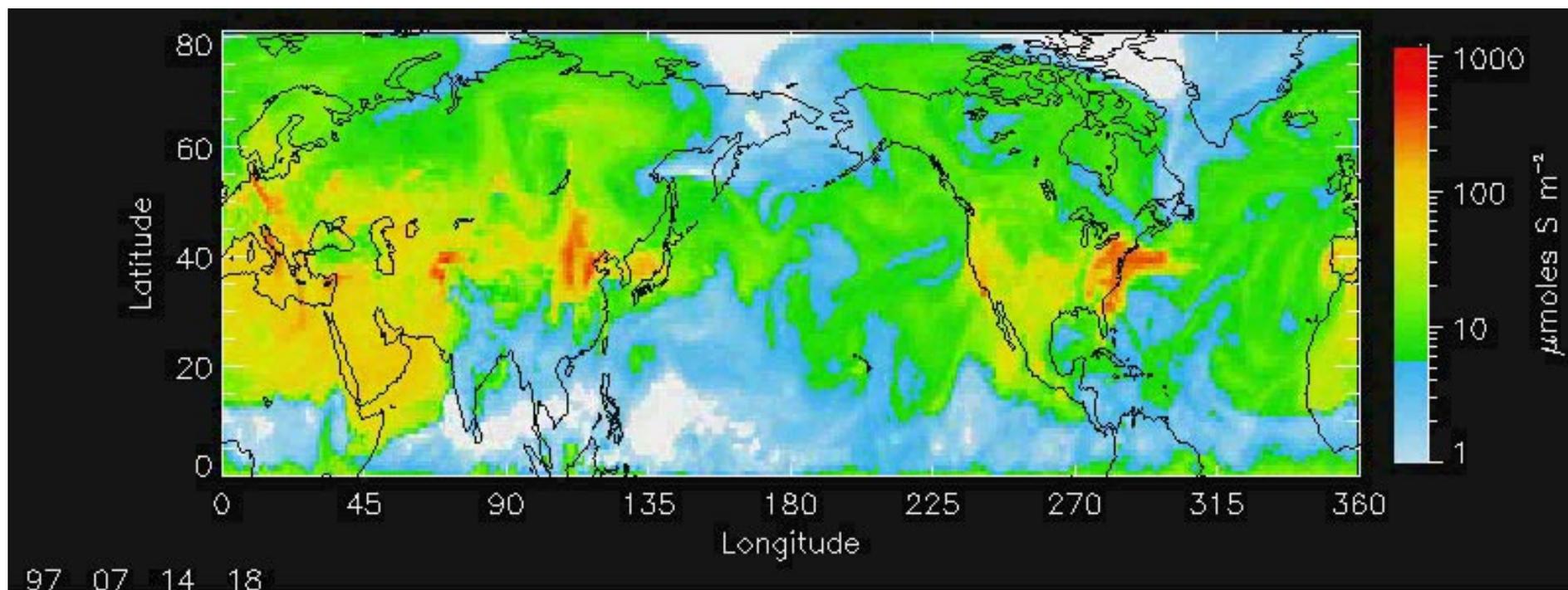
Wang (BNL); Collins, Gasparini (TAMU); Ogren, Sheridan (NOAA)

Aerosol loading, and microphysical and optical properties exhibit a rich variability, which must be understood and represented in models.

HEMISPHERIC DISTRIBUTION OF SULFATE COLUMN BURDEN

Vertical integral of concentration

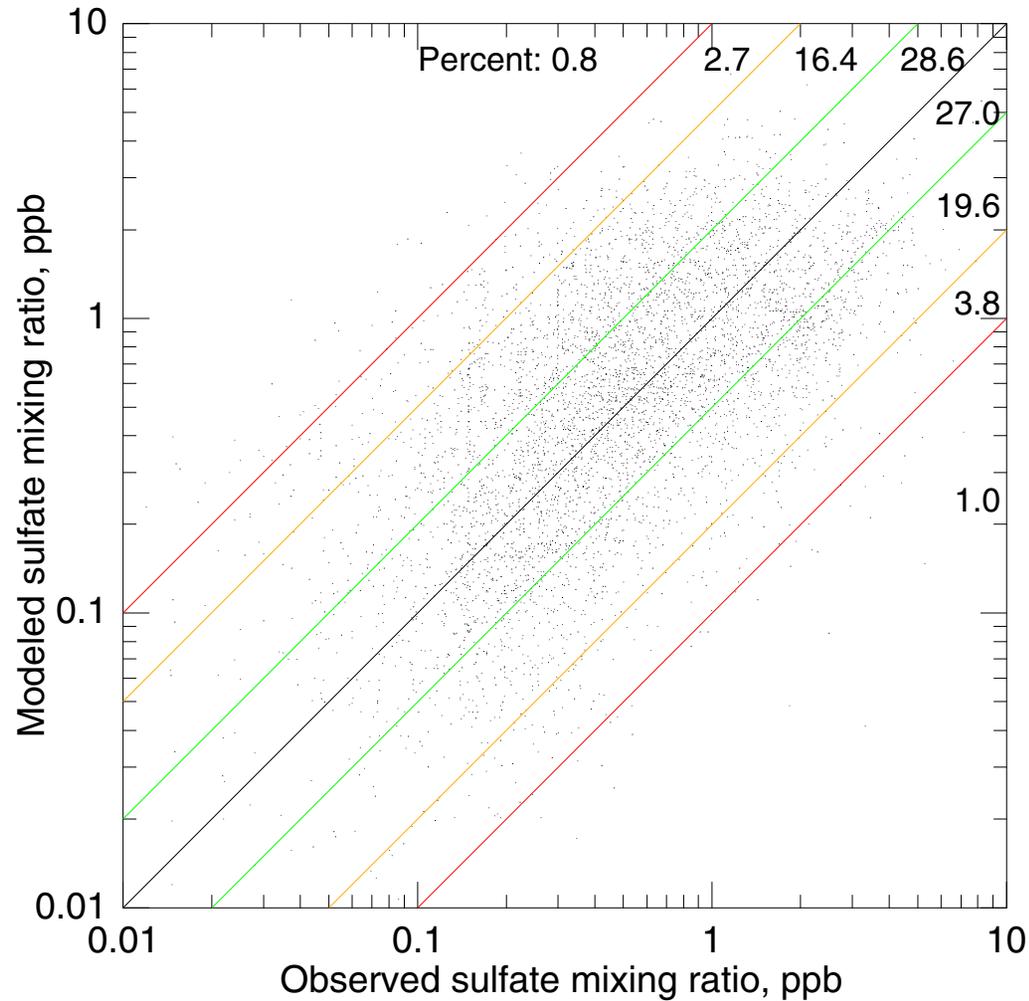
July 14, 1997, 1800 UTC



Brookhaven National Laboratory Chemical Transport Model

MODEL-OBSERVATION COMPARISONS

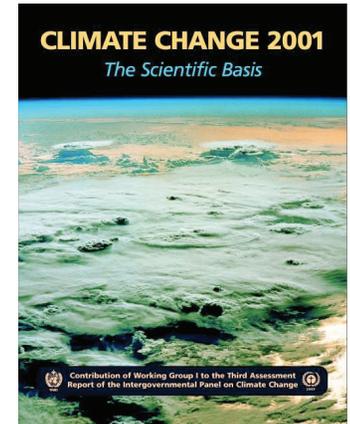
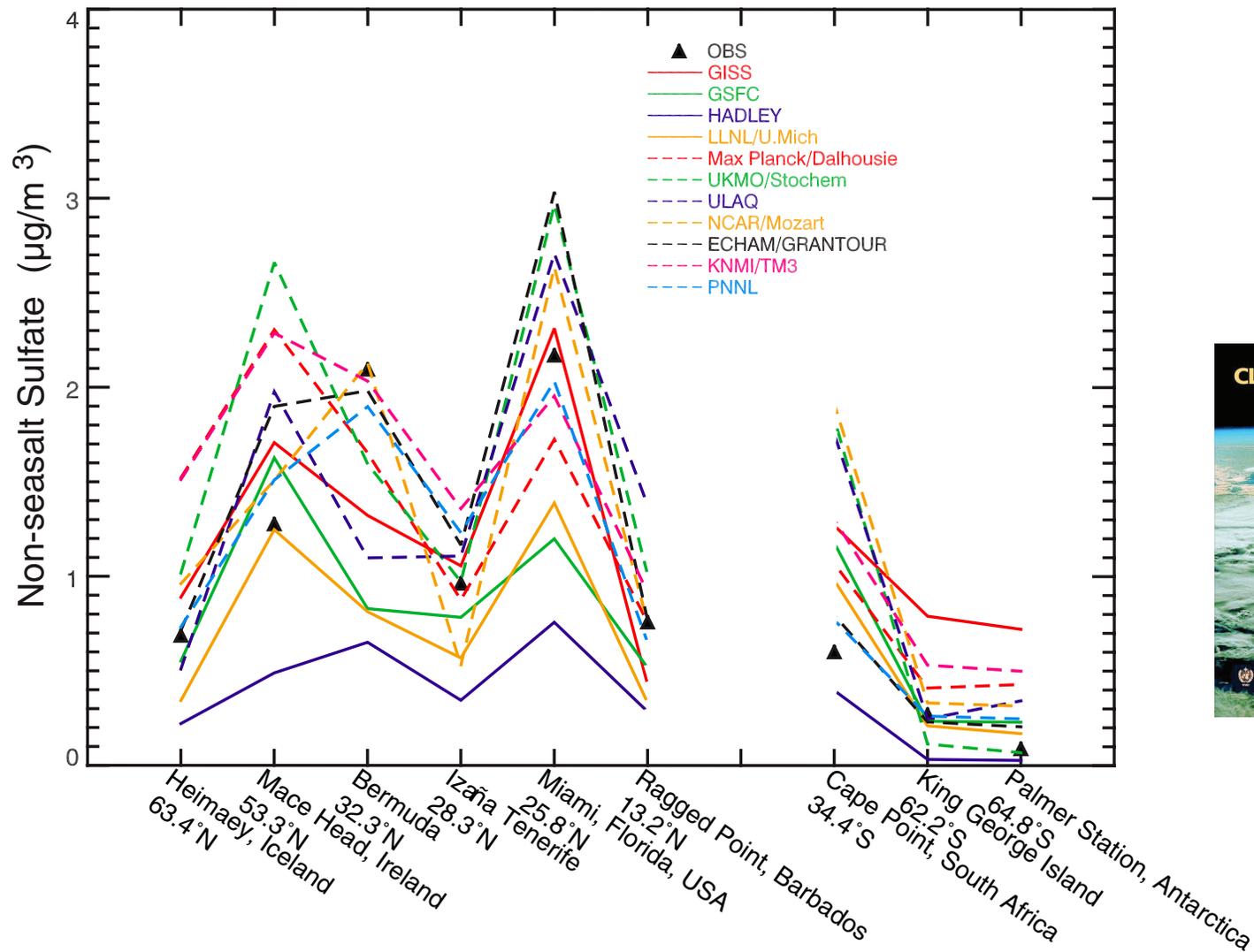
5083 24-Hour sulfate mixing ratio in BNL CTM driven by assimilated meteorological data - June-July 1997



56% of comparisons within factor of 2. 92% within factor of 5.

SULFATE MODEL INTERCOMPARISON

Annual average non-seasalt sulfate in 11 chemical transport models and comparison with observations at nine stations



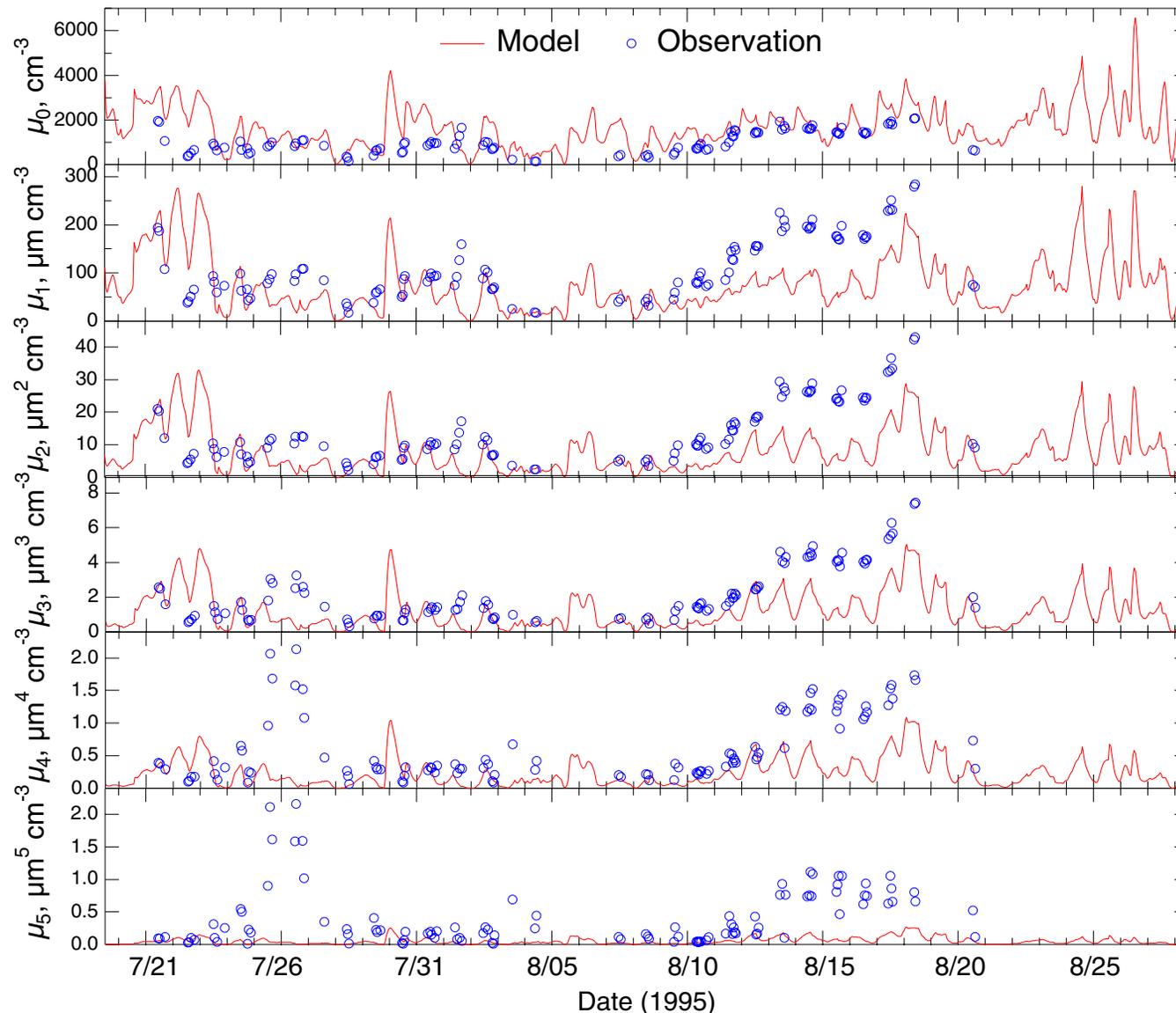
Penner et al., IPCC, 2001

“Most models predict surface-level seasonal mean sulphate aerosol mixing ratios to within 20%.”

“We cannot be sure that these models achieve reasonable success for the right reasons.”

TIME SERIES COMPARISON FOR AEROSOL MOMENTS

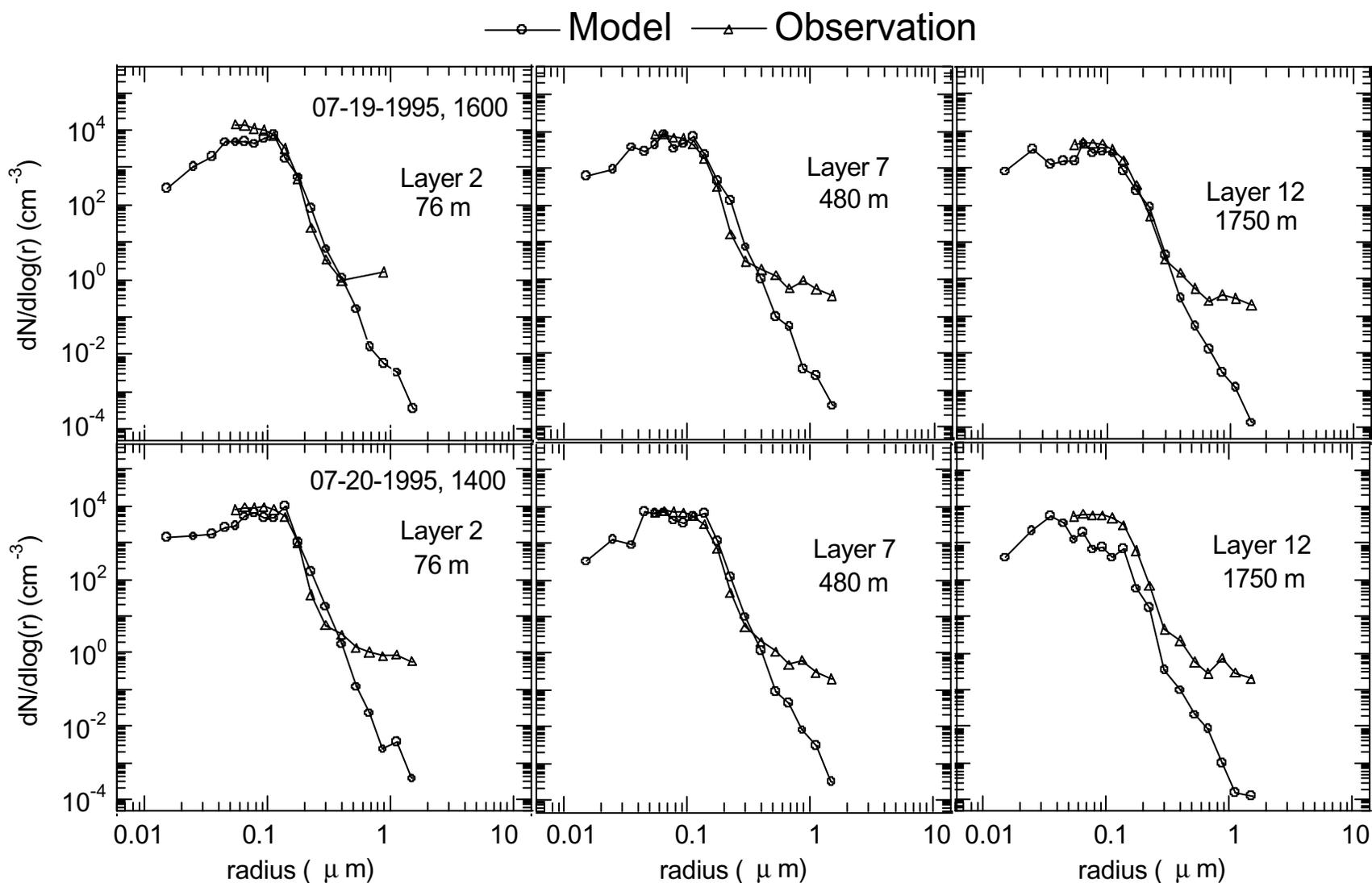
Look Ridge, Great Smoky Mountains TN (84° W, 36° N; 900 m) during SEAVS



Yu, Kasibhatla, Wright, Schwartz, McGraw & Deng, *JGR*, 2003

SIZE DISTRIBUTIONS

Comparison of Measurement and Retrieval from Model
At 3 Altitudes near Nashville TN

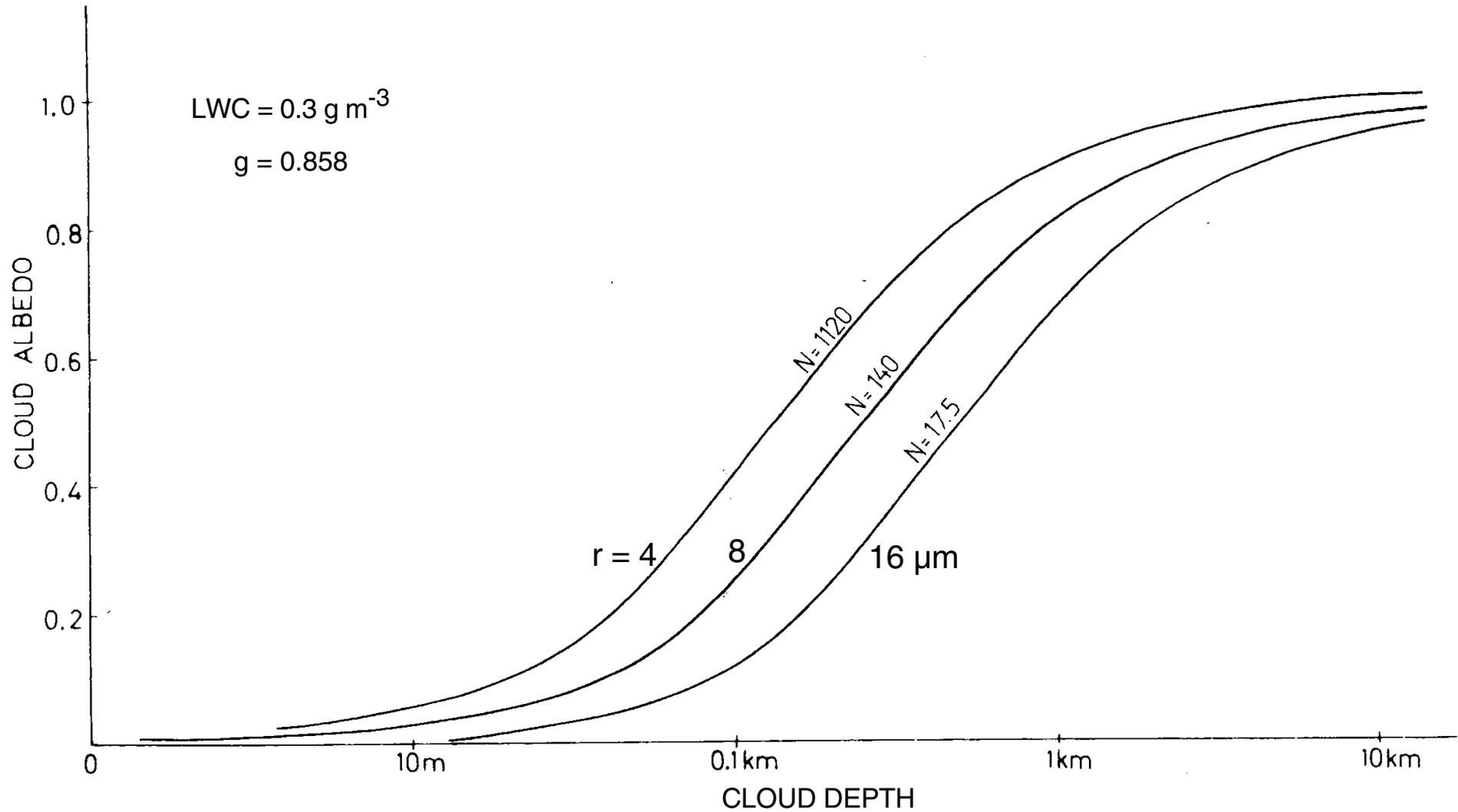


Yu, Kasibhatla, Wright, Schwartz, McGraw & Deng, *JGR*, 2003

INDIRECT EFFECT

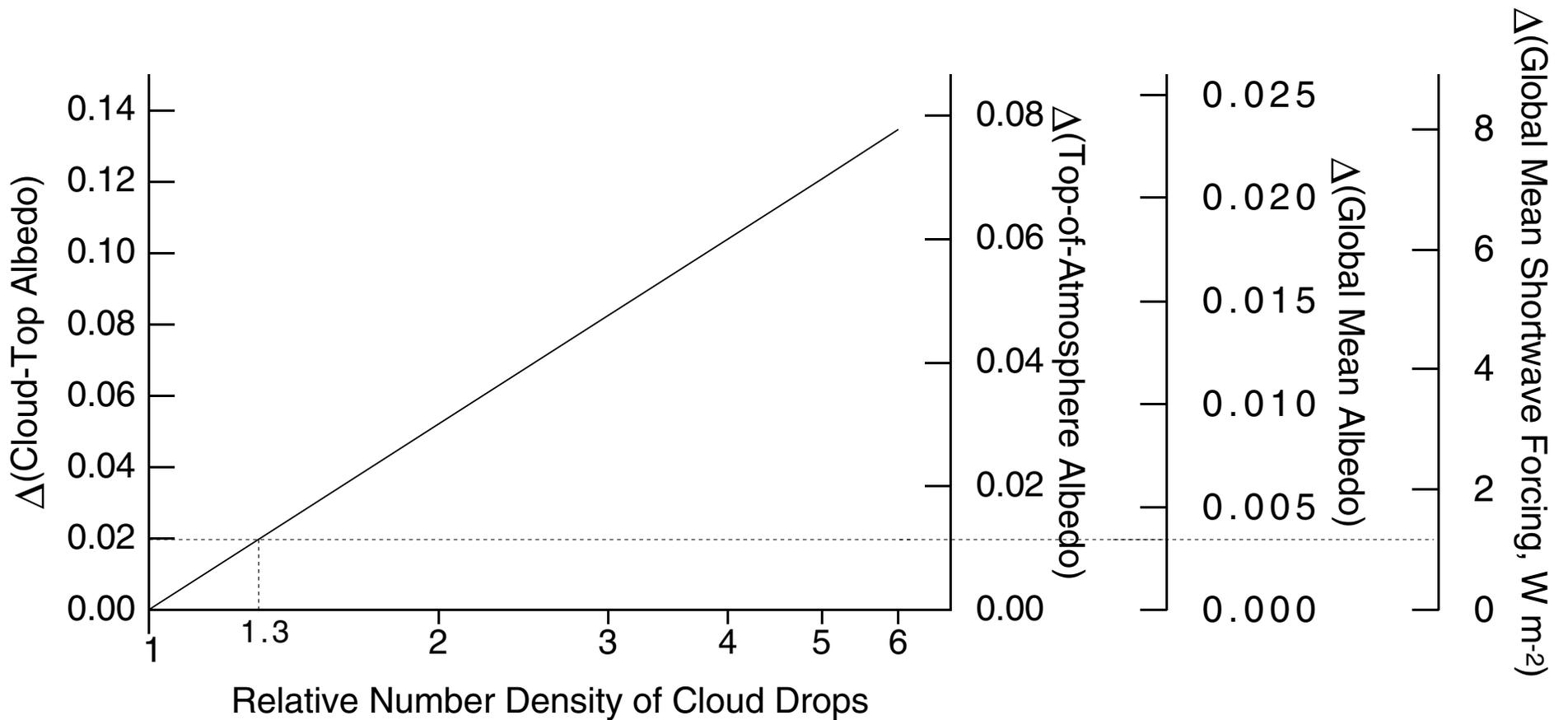
DEPENDENCE OF CLOUD ALBEDO ON CLOUD DEPTH

Influence of Cloud Drop Radius and Concentration



Twomey, *Atmospheric Aerosols*, 1977

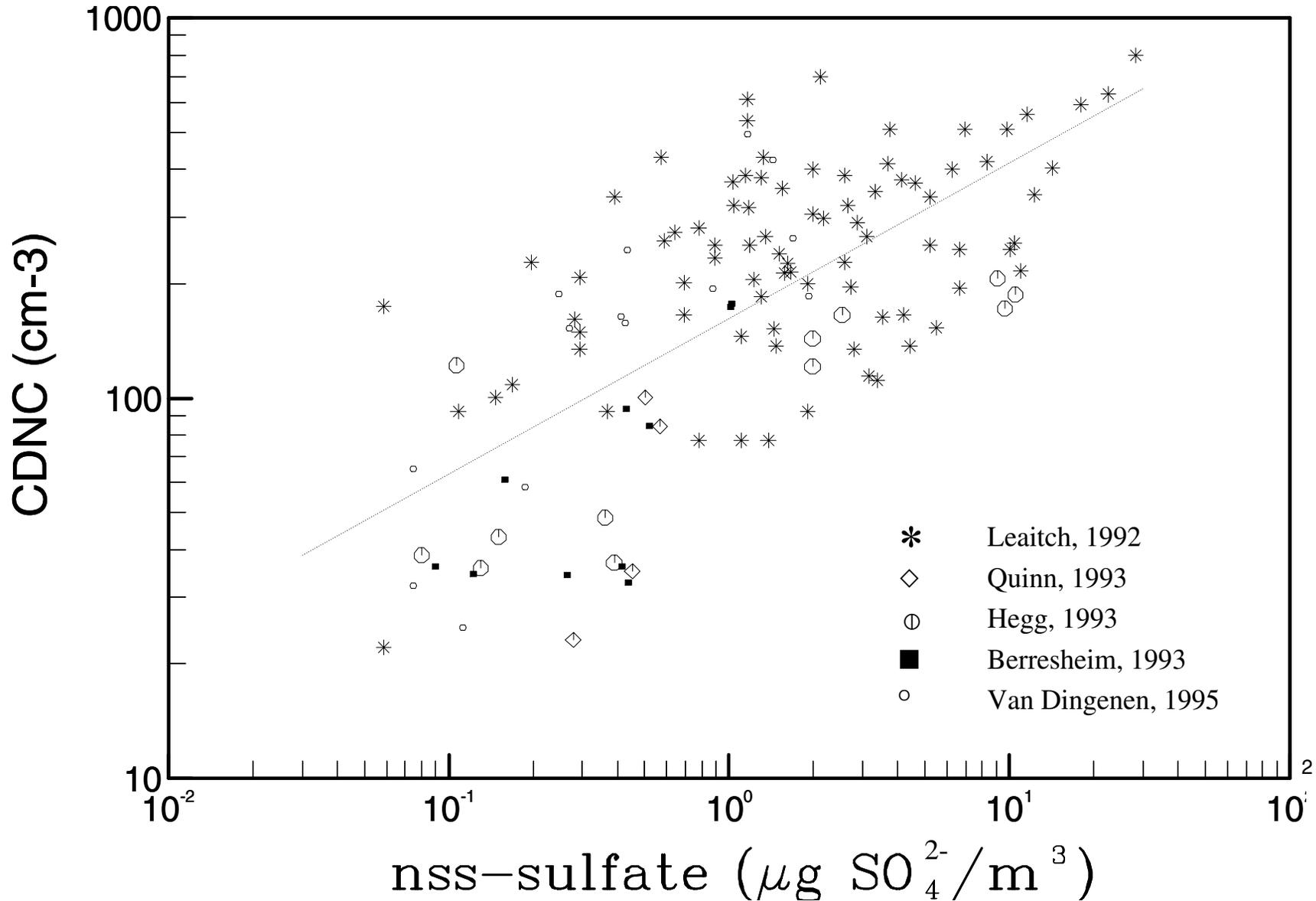
SENSITIVITY OF ALBEDO AND FORCING TO CLOUD DROP CONCENTRATION



Schwartz and Slingo (1996)

CLOUD DROPLET NUMBER CONCENTRATION

Dependence on Non-Seasalt Sulfate



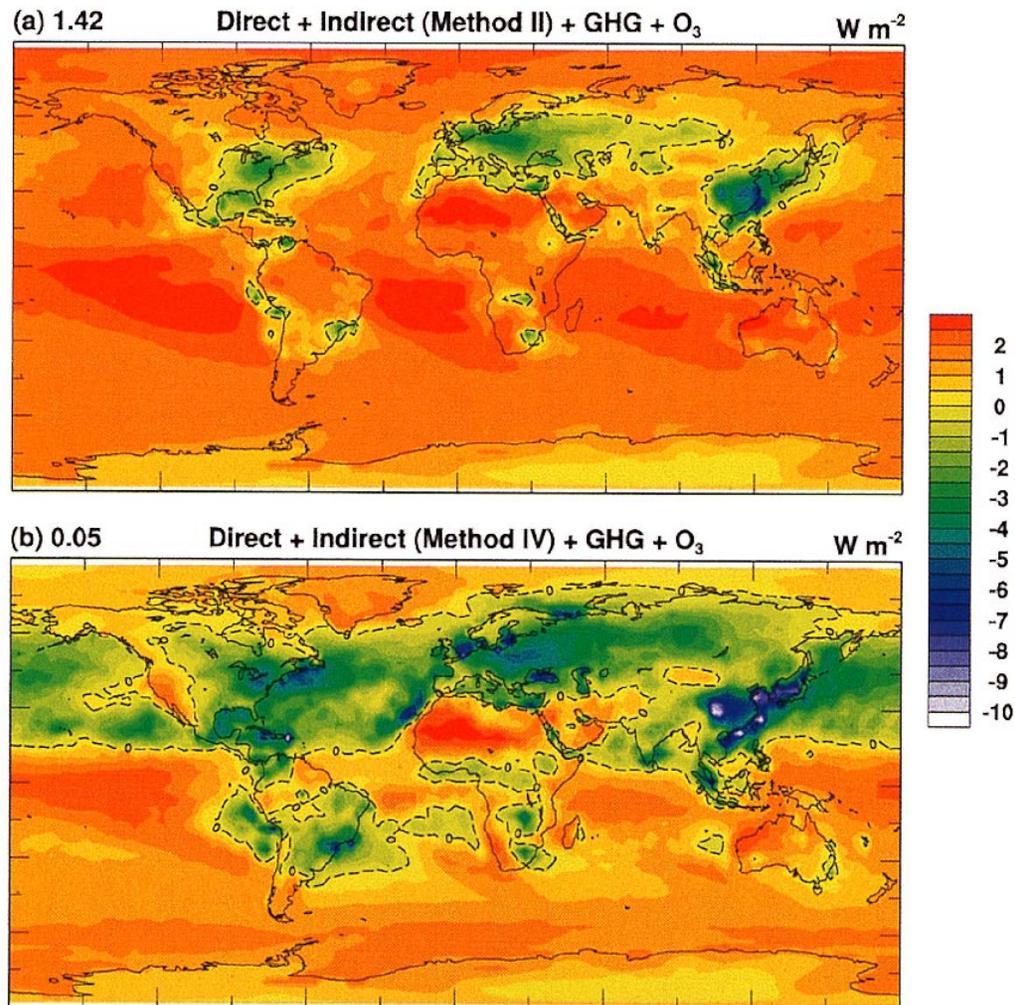
Boucher and Lohmann, 1995

CDNC depends on sulfate concentration but much other variance is not accounted for.

SHORTWAVE FORCING, ANNUAL AVERAGE

GHG's + O₃ + Sulfate (Direct and Indirect)

Two Formulations of Cloud Droplet Concentration

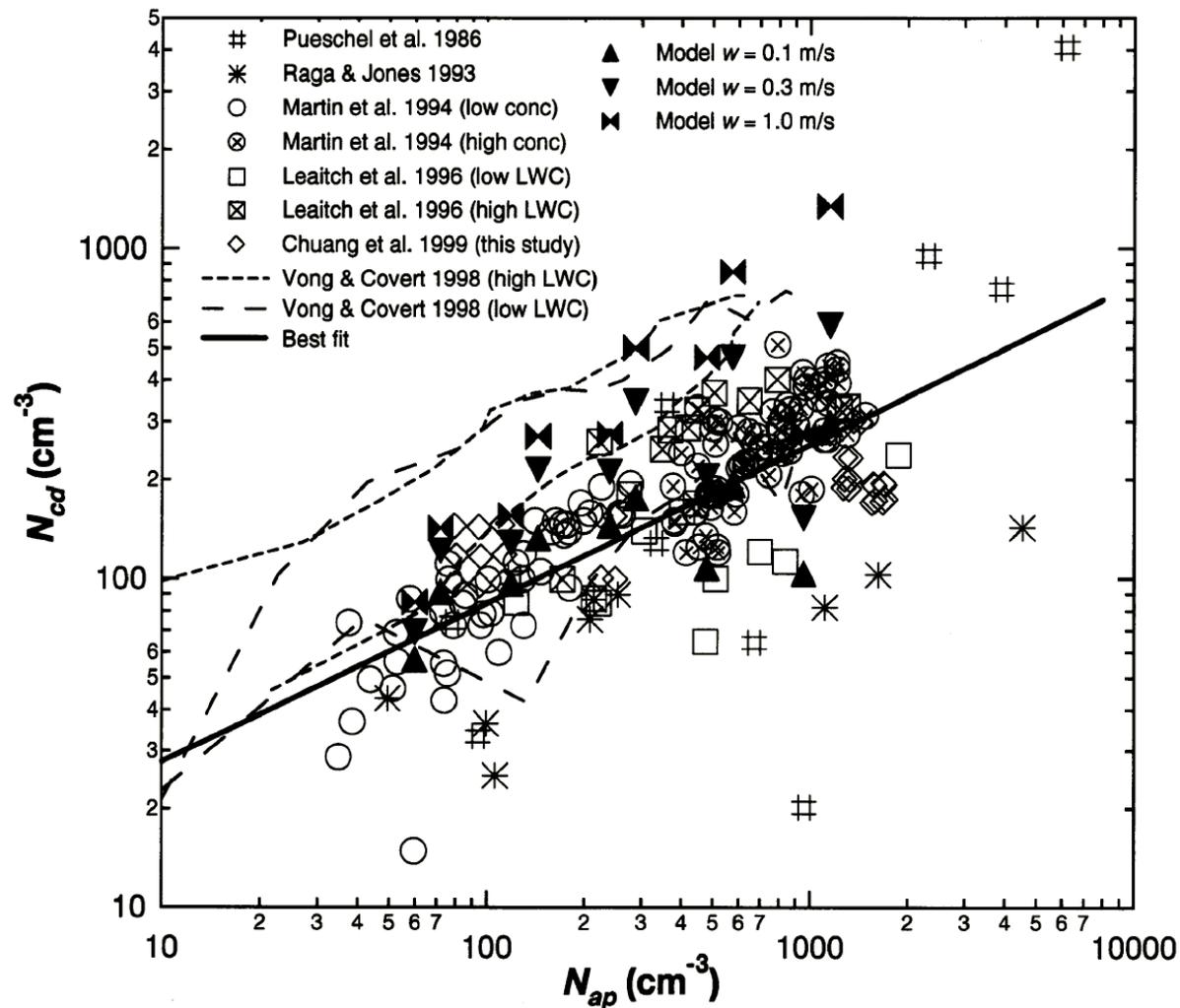


Kiehl et al., JGR, 2000

Indirect forcing is highly sensitive to the assumed relation between sulfate concentration and cloud droplet number concentration.

CLOUD DROP NUMBER CONCENTRATION

Dependence on aerosol particle concentration



Chuang et al., *Tellus*, 2000

The variables controlling CDNC must be understood and represented in models much better than at present.

UNCERTAINTY BUDGET FOR *INDIRECT* FORCING BY INDUSTRIAL AEROSOLS

Quantity	Central Value	2/3 Uncertainty Range	Uncertainty Factor
Background N_d for Northern Hemisphere marine (cm^{-3})	140	66 to 214	3.2
Perturbed N_d for Northern Hemisphere marine (cm^{-3})	217	124 to 310	2.5
Cloud mean liquid water content (LWC) (g m^{-3})	0.225	0.125 to 0.325	2.6
Background sulfate concentration ($\mu\text{g m}^{-3}$)	1.5	0.85 to 2.15	2.5
Cloud layer thickness (m)	200	100 to 300	3.0
Perturbed sulfate concentration ($\mu\text{g m}^{-3}$)	3.6	2.4 to 4.8	2.0
Susceptible cloud fraction, f_c	0.24	0.19 to 0.29	1.5
Atmospheric transmission above cloud layer, T_a	0.92	0.78 to 1.00	1.3
Mean surface albedo	0.06	0.03 to 0.09	3.0, 1.1
Result: If central value is -1.4 Wm^{-2} the 2/3 uncertainty range is from 0 to -2.8 Wm^{-2} .			∞

Modified from Penner et al., IPCC, 2001

- *Many of the greatest quantified uncertainties are in chemical properties.*
- *Some key uncertainties are at the interface of aerosols and clouds, such as relation between cloud drop concentration and aerosol loading, microphysical properties, and composition. **These uncertainties are not quantified.***

AEROSOL RESEARCH REQUIREMENTS BY CCRD COMPONENT

Atmospheric Science Program

Abundance, composition, mixing state and optical and cloud-nucleating properties of atmospheric aerosols

Observe - Model - Compare (local closure experiments)

Sources of aerosols and aerosol precursors (mass rates and size-dependent composition and mixing state)

Measure - Understand - Quantify

Atmospheric chemical and microphysical transformation processes and three dimensional mixing and transport processes

Experiment - Understand - Model - Compare (model evaluation)

Aerosol direct and indirect radiative forcing

Model - Estimate

Atmospheric Radiation Measurement Program (ARM)

Aerosol-radiation interactions

Aerosol-cloud-radiation interactions

Climate Change Prediction Program (CCPP)

Represent aerosol influences in climate models

Determine climate sensitivity in models and compare with observations