# AEROSOLS AND CLIMATE CHANGE

## A TUTORIAL

Stephen E. Schwartz

BROOKHAVEN NATIONAL LABORATORY

Atlanta, Georgia AAAR 04

October 4, 2004

*(Viewgraphs revised 2004-10-06)*

http://www.ecd.bnl.gov/steve/schwartz.html

# AEROSOLS AND CLIMATE CHANGE

*A Tutorial*

## OUTLINE

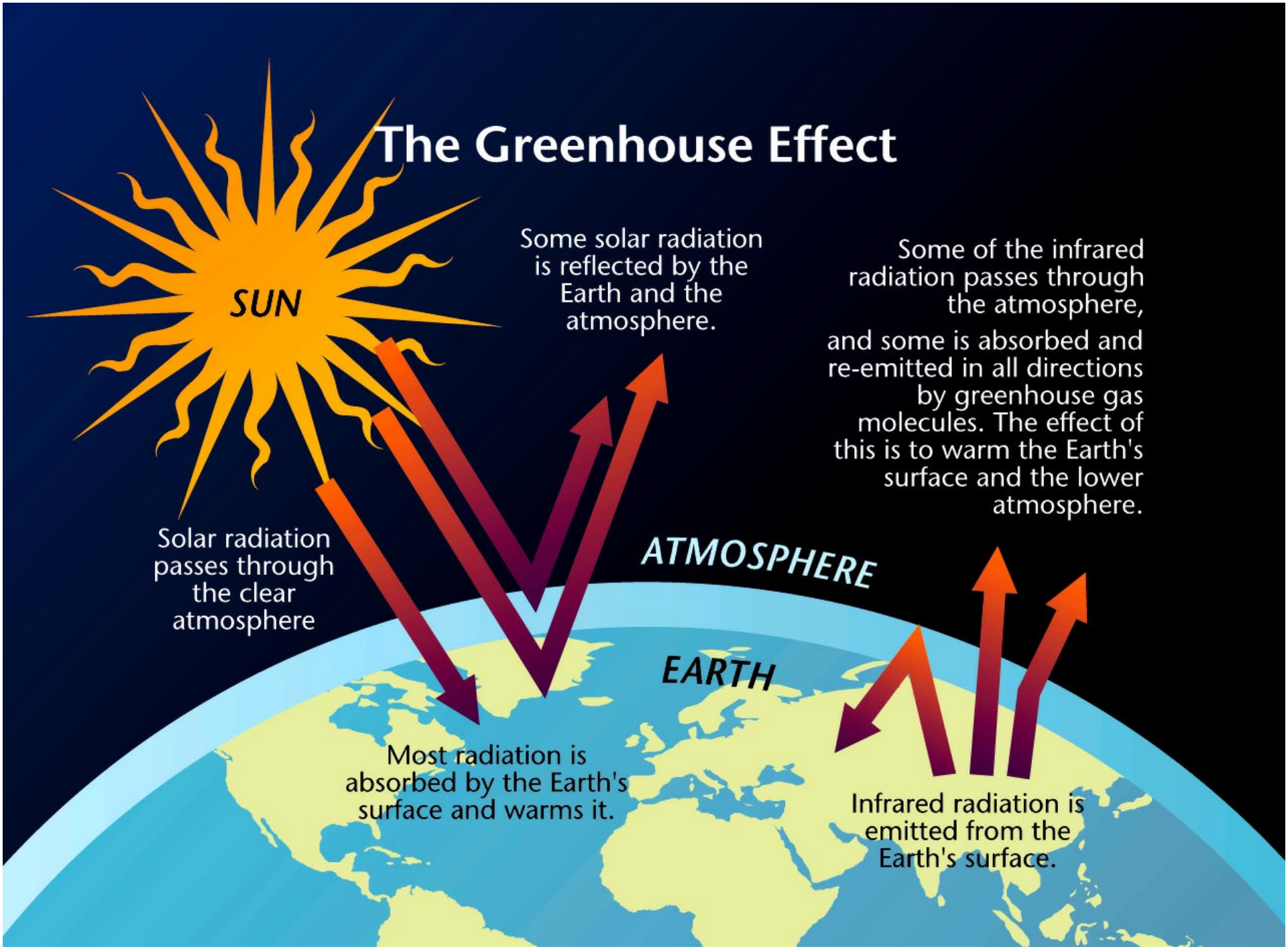
Climate and ***climate change***

***Forcing*** of climate change and climate ***sensitivity***

Aerosol influences on climate change: ***mechanisms and magnitudes***

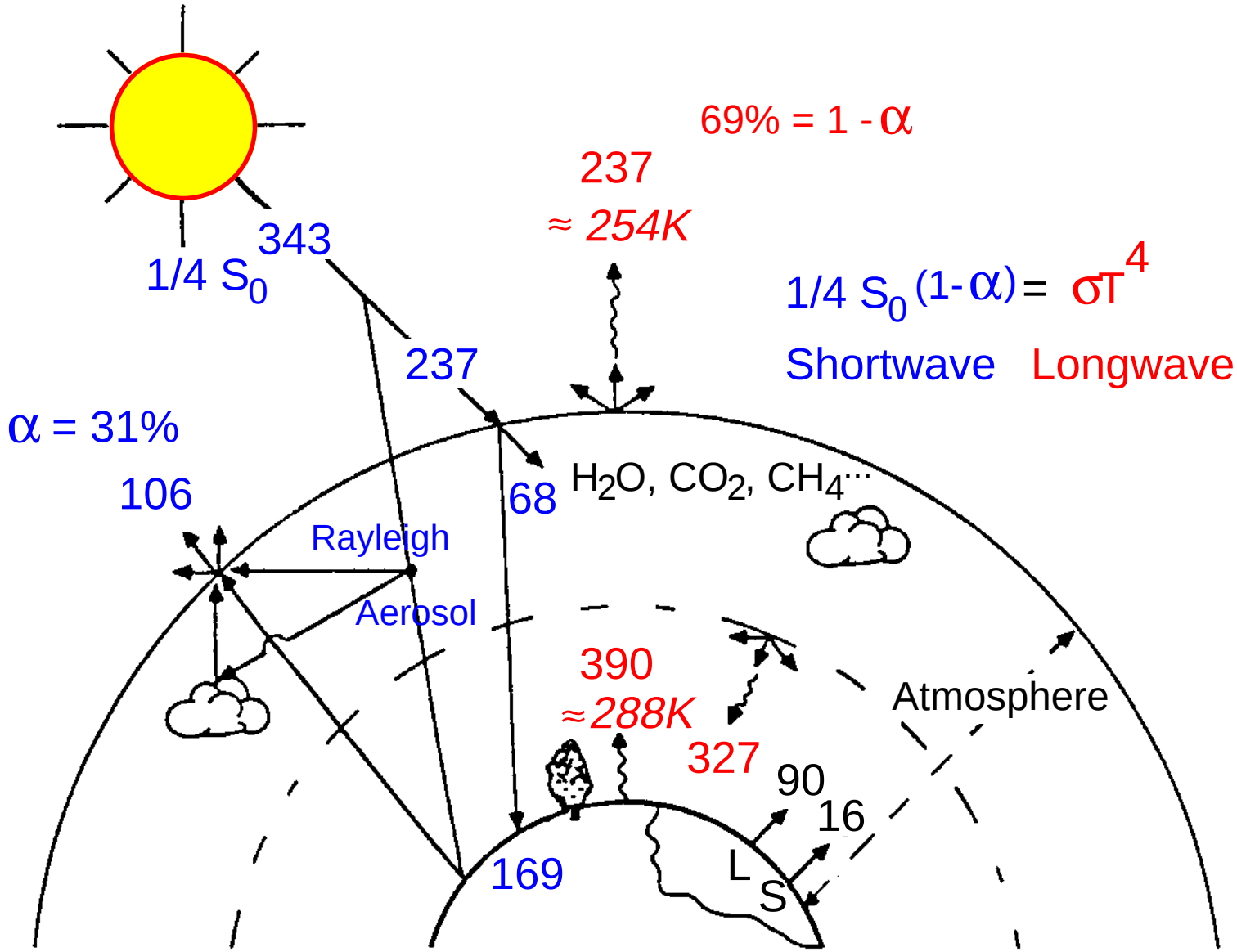
- Direct effects
- Indirect effects

***Implications of uncertainties***


The Greenhouse Effect
SUN
Some solar radiation is reflected by the Earth and the atmosphere.
Some of the infrared radiation passes through the atmosphere,
and some is absorbed and re-emitted in all directions by greenhouse gas molecules. The effect of this is to warm the Earth's surface and the lower atmosphere.
Solar radiation passes through the clear atmosphere
ATMOSPHERE
EARTH
Most radiation is absorbed by the Earth's surface and warms it.
Infrared radiation is emitted from the Earth's surface.

# GLOBAL ENERGY BALANCE

Global and annual average energy fluxes in watts per square meter




*Schwartz, 1996, modified from Ramanathan, 1987*

# *ATMOSPHERIC RADIATION*

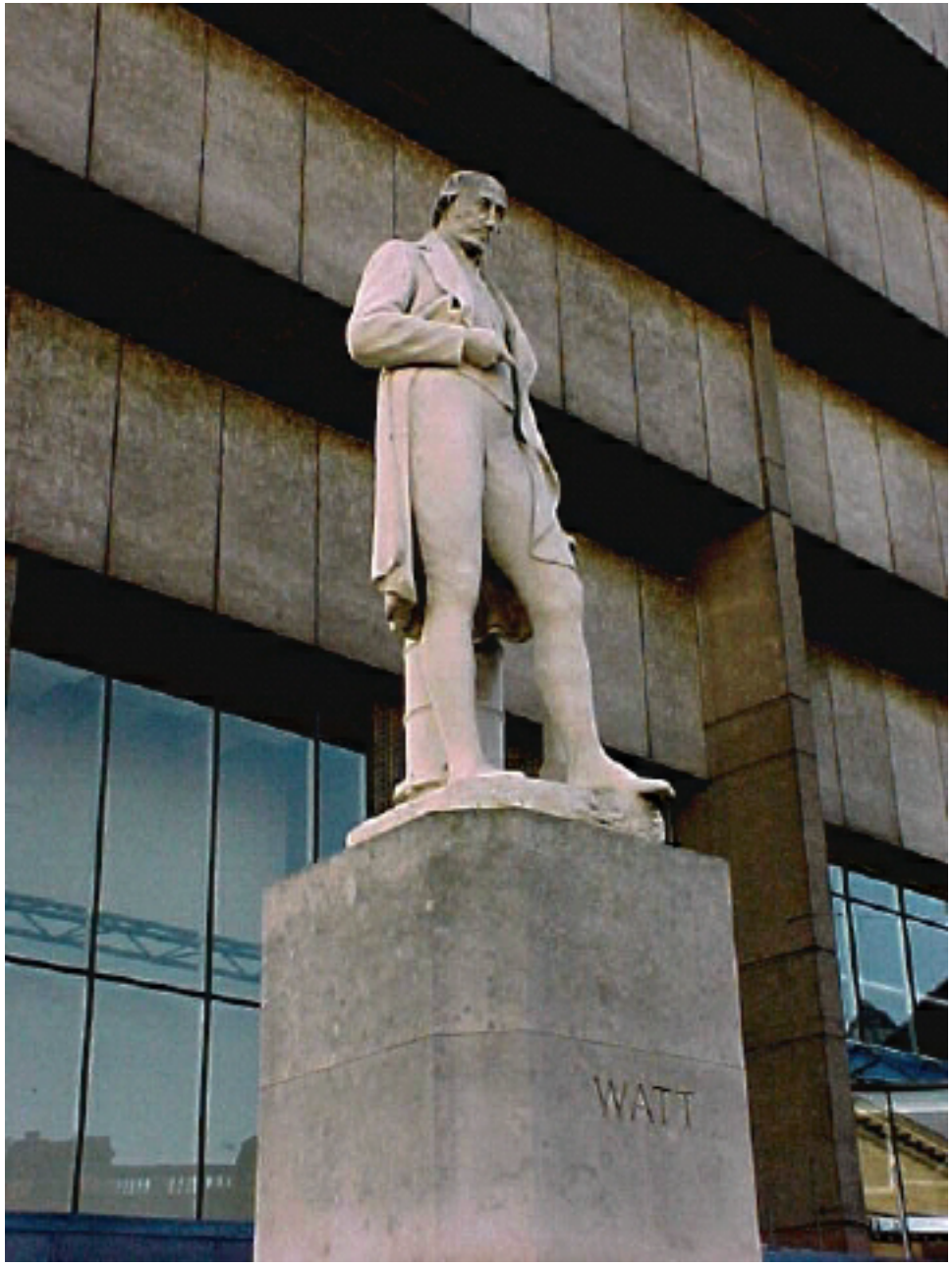
***Energy per area per time***

***Power per time***

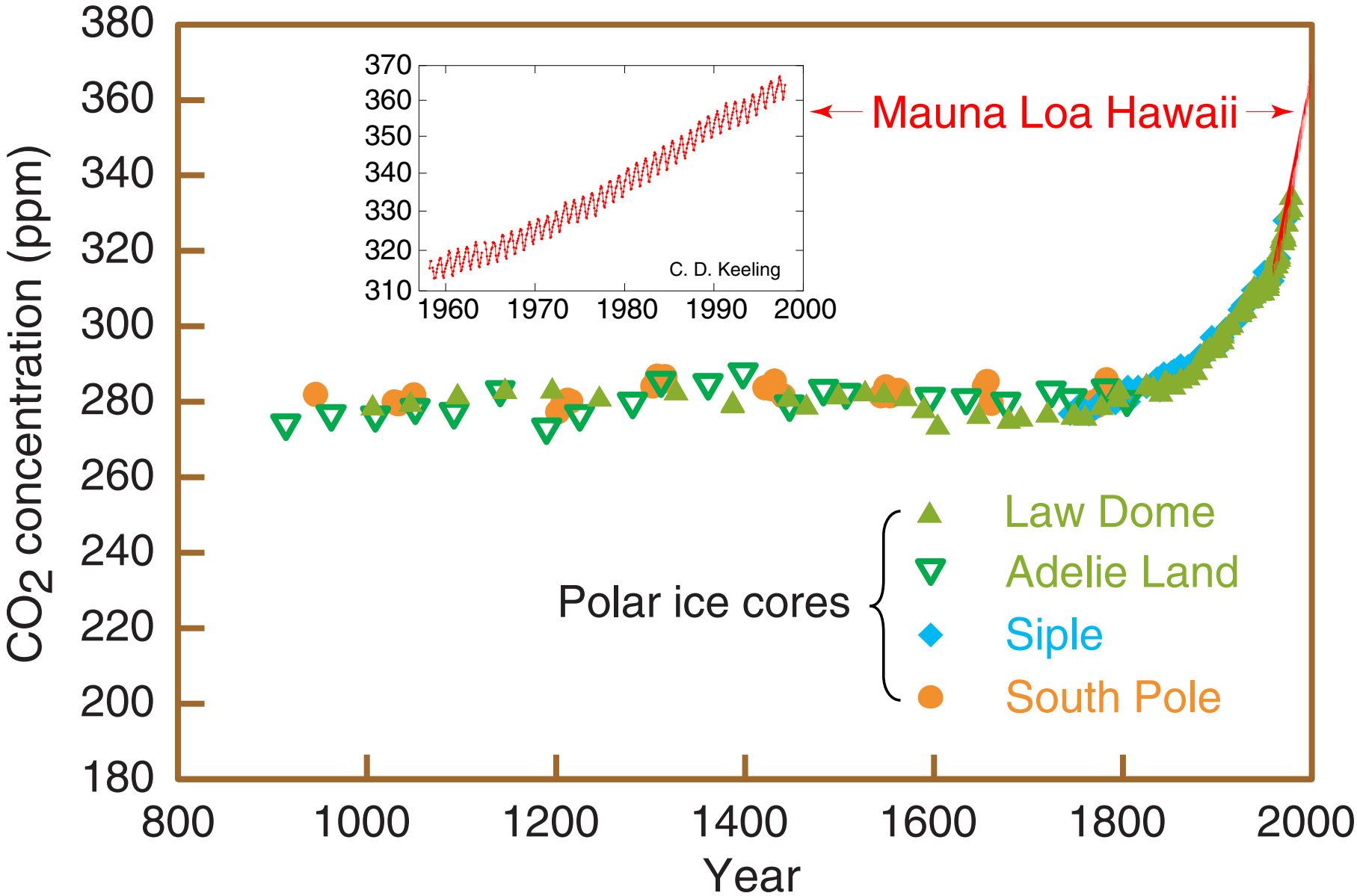
***Unit:***

***Watt per square meter***

***W $m^{-2}$***



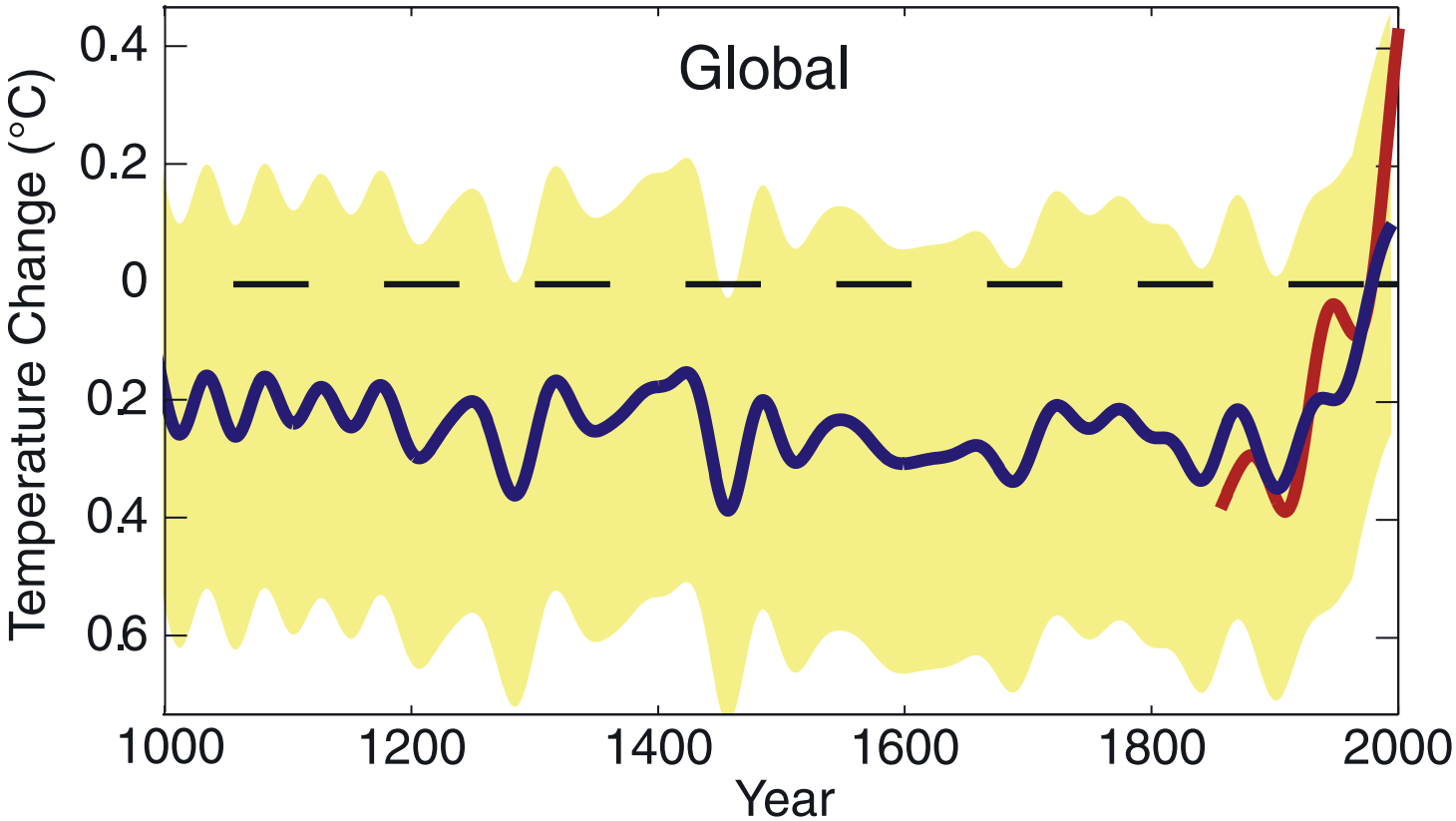
# ATMOSPHERIC CARBON DIOXIDE IS INCREASING




Global carbon dioxide concentration over the last 1000 years

# GLOBAL TEMPERATURE TREND (1000-2000)

From tree-ring, coral, and ice-core proxy records
As calibrated by instrumental measurements




Instrumental record

Proxy reconstruction

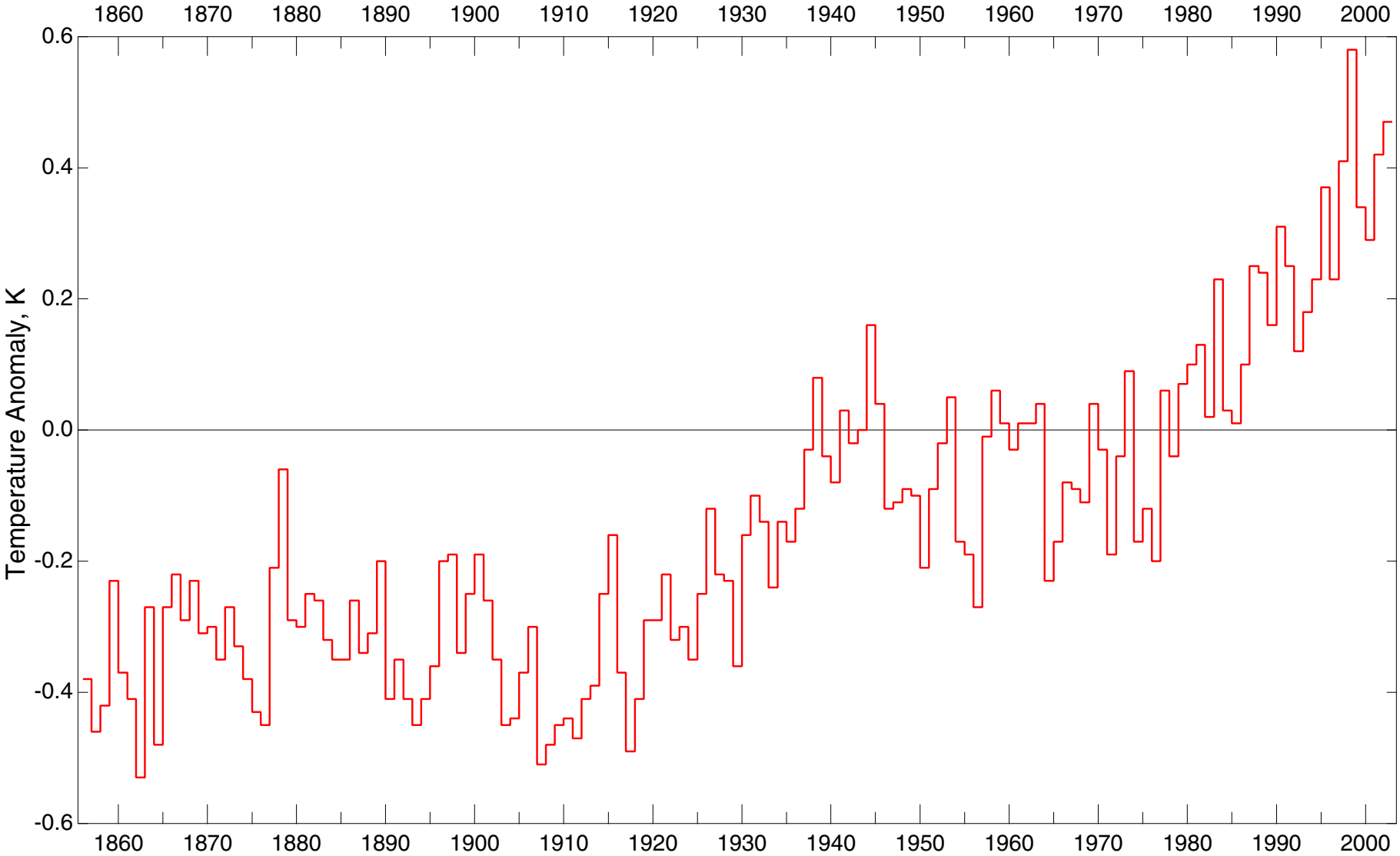
Uncertainty in proxy reconstruction

*Jones & Mann, Revs. Geophys., 2004*

# GLOBAL AVERAGE TEMPERATURE TREND

## 1856-2002

### Temperature Anomaly Relative to Base Period 1961-1990




*Climate Research Unit, East Anglia UK*

# *RADIATIVE FORCING OF CLIMATE CHANGE*

-- An ***externally imposed perturbation*** in the radiative energy budget of Earth's climate system.

Pertinent to climate change over the industrial period is ***radiative forcing of the surface-troposphere system.***

EXAMPLES:

Shortwave forcing due to change in ***solar constant***

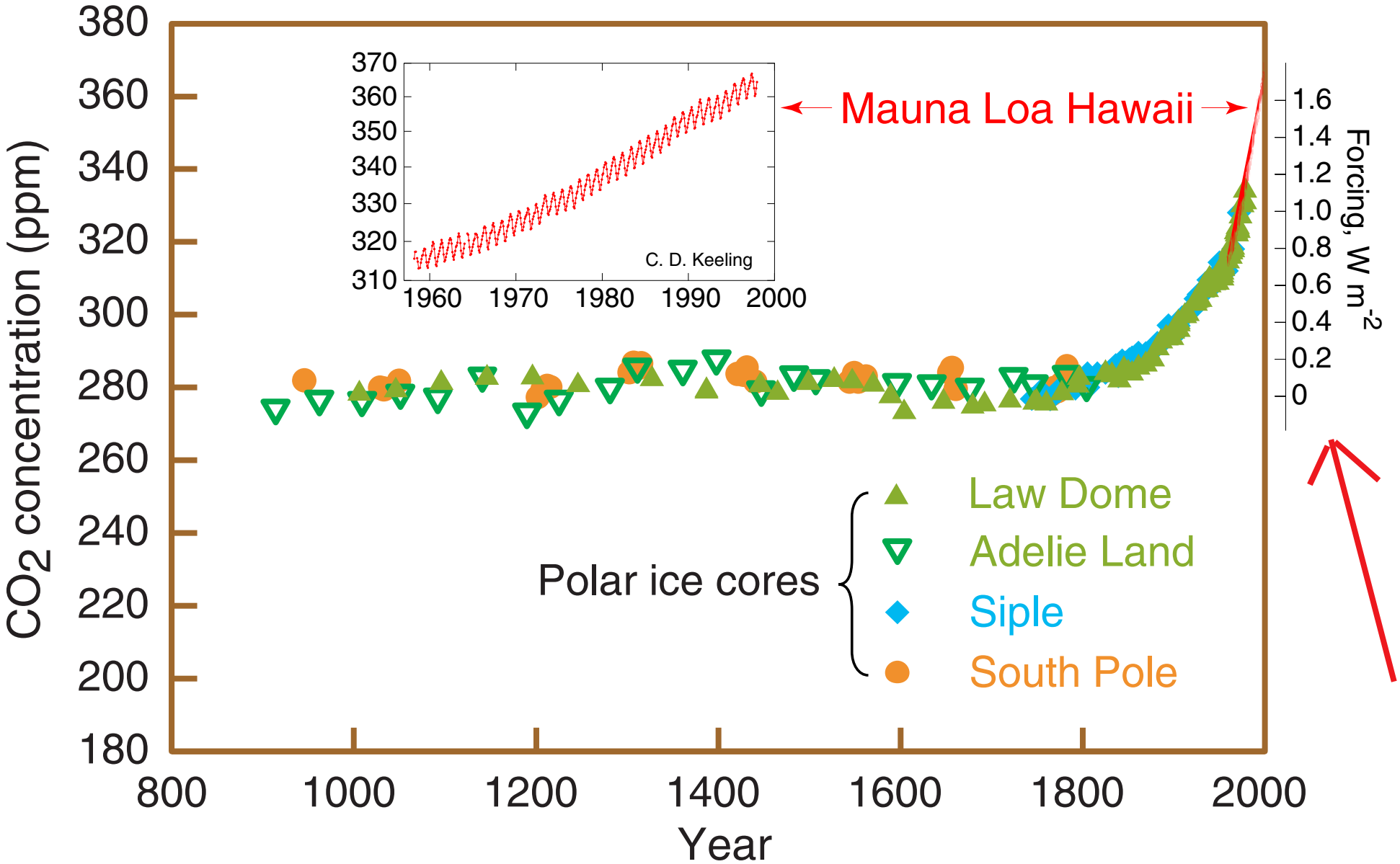
Longwave (thermal infrared) forcing due to increased ***greenhouse gas concentrations***

Shortwave forcing by changes in ***surface albedo***

Shortwave forcing due to enhanced light scattering by ***aerosols***

UNITS: Watts per square meter, W $m^{-2}$

# ATMOSPHERIC CARBON DIOXIDE IS INCREASING




Global carbon dioxide concentration and infrared radiative forcing over the last thousand years

# *CLIMATE RESPONSE*

The ***change*** in global and annual mean temperature, $\Delta T$, K, resulting from a given radiative forcing.

***Working hypothesis:***

*The change in global mean temperature depends on the magnitude of the forcing, not its nature or its spatial distribution.*

$$\Delta T = \lambda F$$

# *CLIMATE SENSITIVITY*

The ***change*** in global and annual mean temperature per unit forcing, $\lambda$, K/(W m$^{-2}$).

# TOP-LEVEL QUESTION IN CLIMATE CHANGE SCIENCE

- ***How much will the global mean temperature change?***

$$\Delta T = \lambda F$$

where $F$ is the ***forcing*** and $\lambda$ is the ***climate sensitivity***.

- A ***forcing*** is a change in a radiative flux component, W $m^{-2}$.
- Forcings are thought to be ***additive*** and ***fungible***.

- ***What is Earth's climate sensitivity?***

***- National Academy Report (Charney, 1979):*** $F$ = 4 W $m^{-2}$

"We estimate the most probable global warming for a doubling of $CO_2$ to be ***near 3 degrees C***, with a probable error of ***plus or minus 1.5 degrees***.

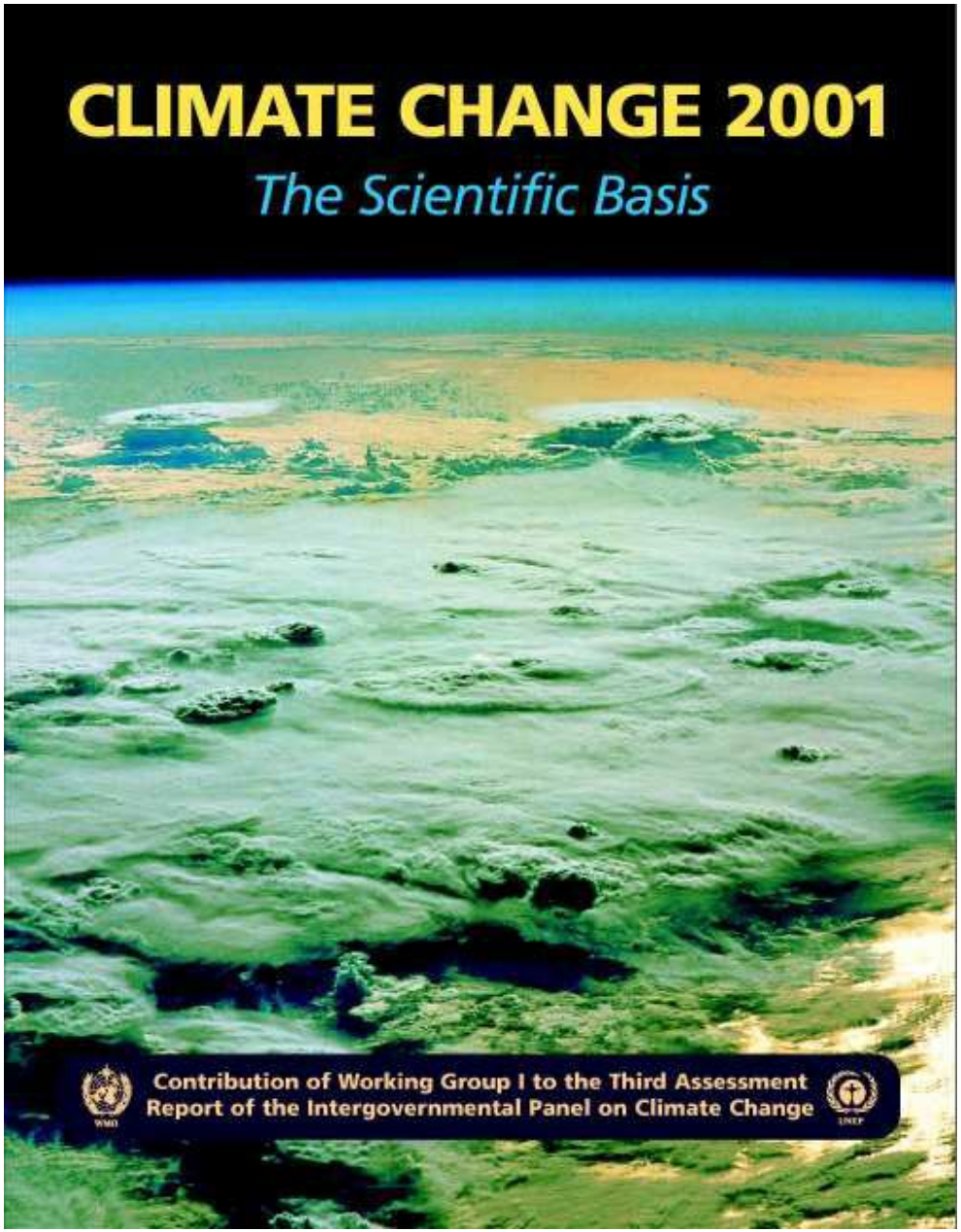
***- Intergovernmental Panel on Climate Change (IPCC, 2001):***

"Climate sensitivity [to $CO_2$ doubling] is likely to be in the range ***1.5 to 4.5˚C***.

***This level of uncertainty is not very useful for policy planning.***

# THE “BIBLE” OF CLIMATE CHANGE RESEARCH

WMO

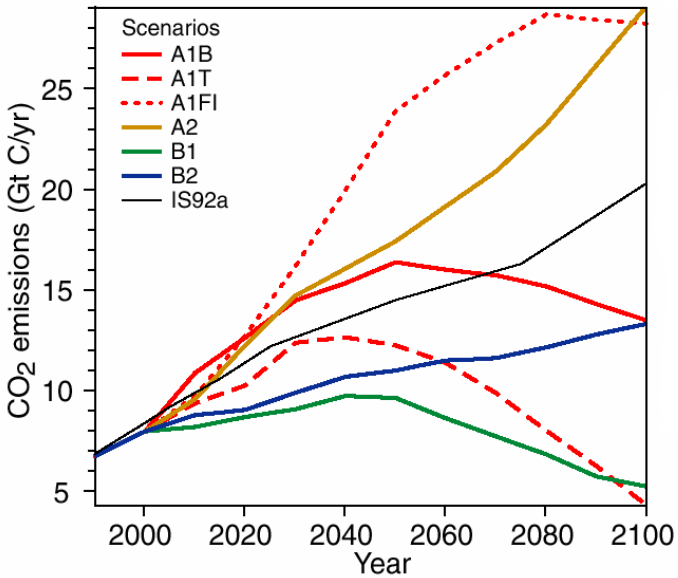


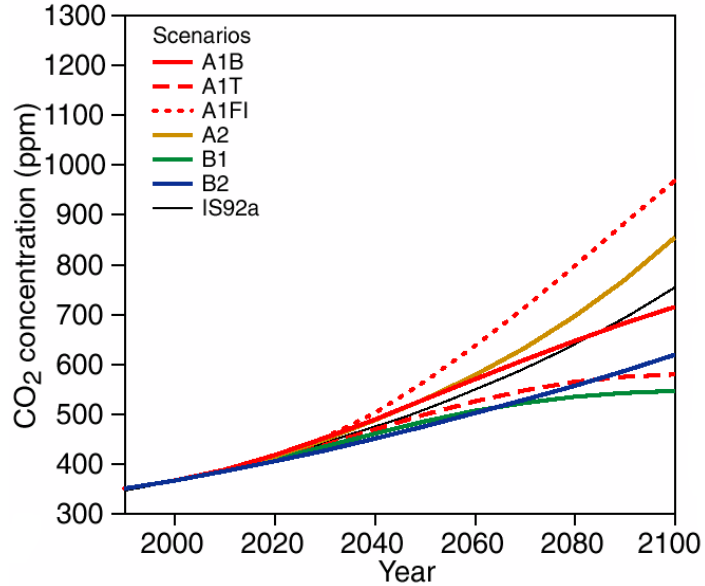

UNEP

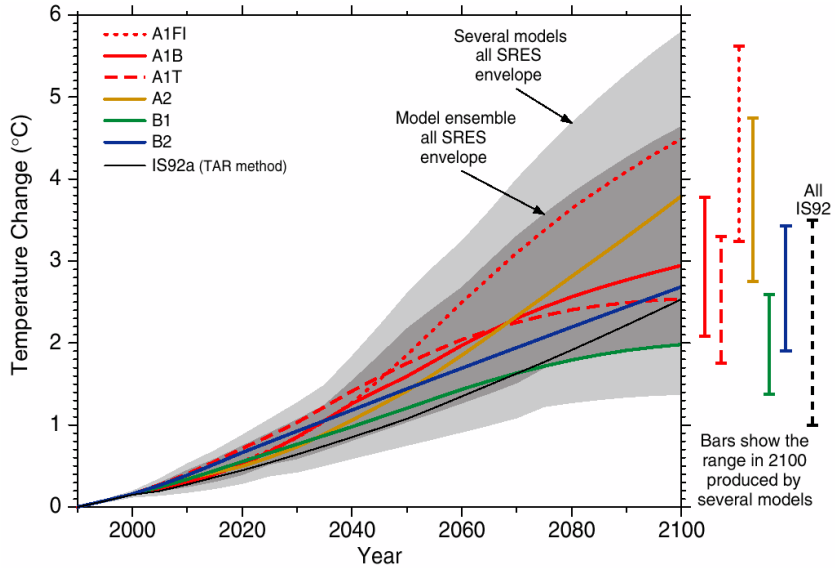
Cambridge University Press, 2001

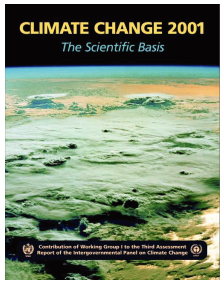
http://www.grida.no/climate/ipcc_tar/wg1/

# FUTURE CLIMATE IS HIGHLY UNCERTAIN



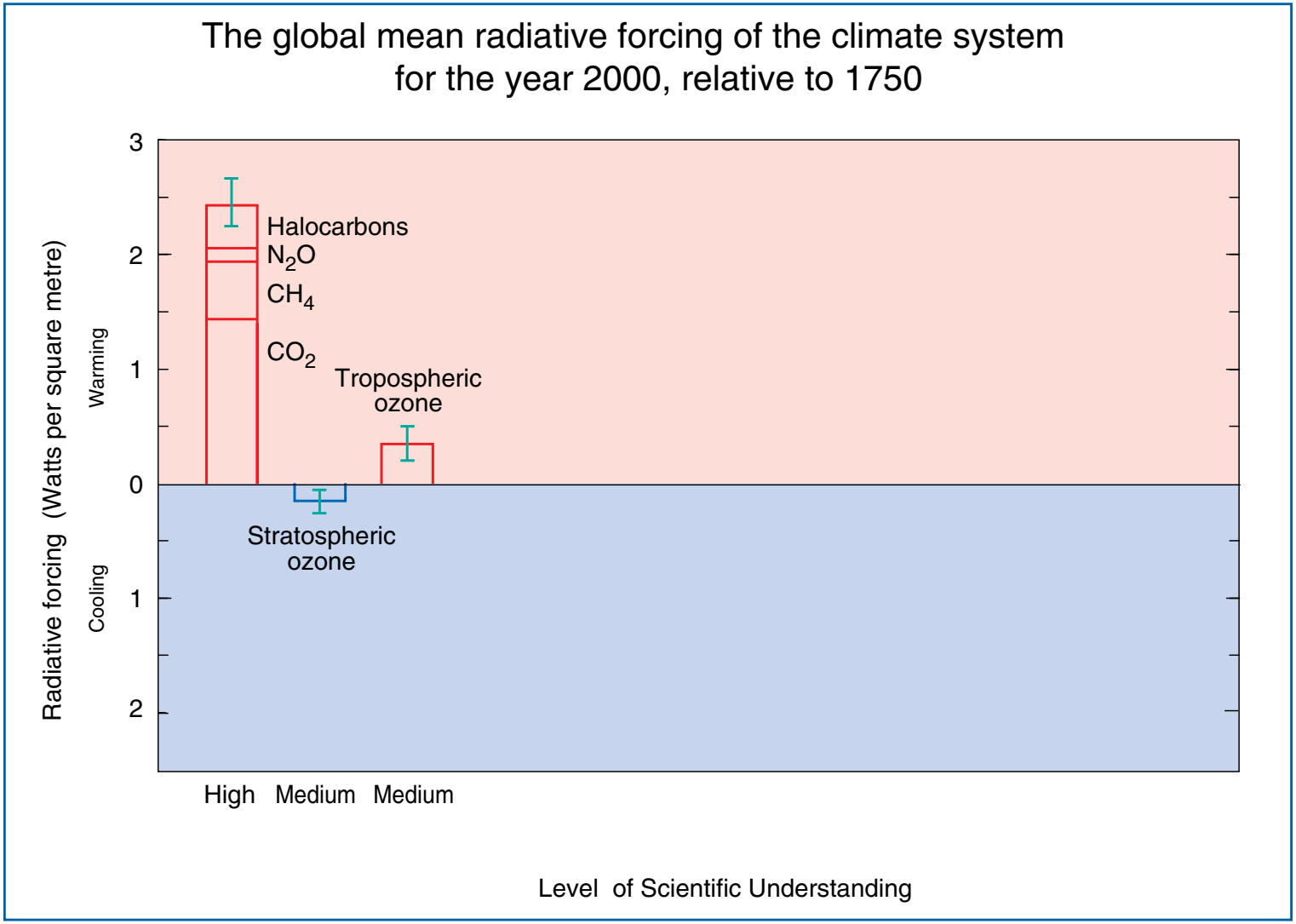




(*IPCC, 2001*)

Contributors to uncertainty in future temperature include ***emissions***, ***concentrations***, and Earth's ***climate sensitivity***. ***And other forcings.***

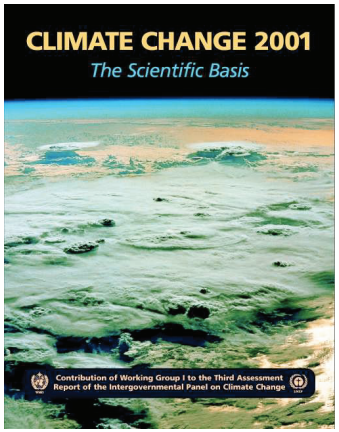
# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
# IPCC (2001)

## Greenhouse gases only




Summary for Policymakers

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



# AEROSOL INFLUENCES ON RADIATION AND CLIMATE

***Direct Shortwave Radiative Effects (Clear sky)***

Light scattering → Cooling influence

Light absorption → Warming influence, depending on surface

***Indirect Shortwave Radiative Effects–Aerosols influence cloud properties***

More droplets → Brighter clouds (Twomey)

More droplets → Enhanced cloud lifetime (Albrecht)

More droplets → Narrowing of drop distribution -- warming (Liu)

***Semi-Direct Shortwave Radiative Effect***

Absorbing aerosol heats air and evaporates clouds (Hansen)

***Longwave Radiative Effect (Clear sky)***

Greenhouse effect of aerosol particles (Vogelmann)

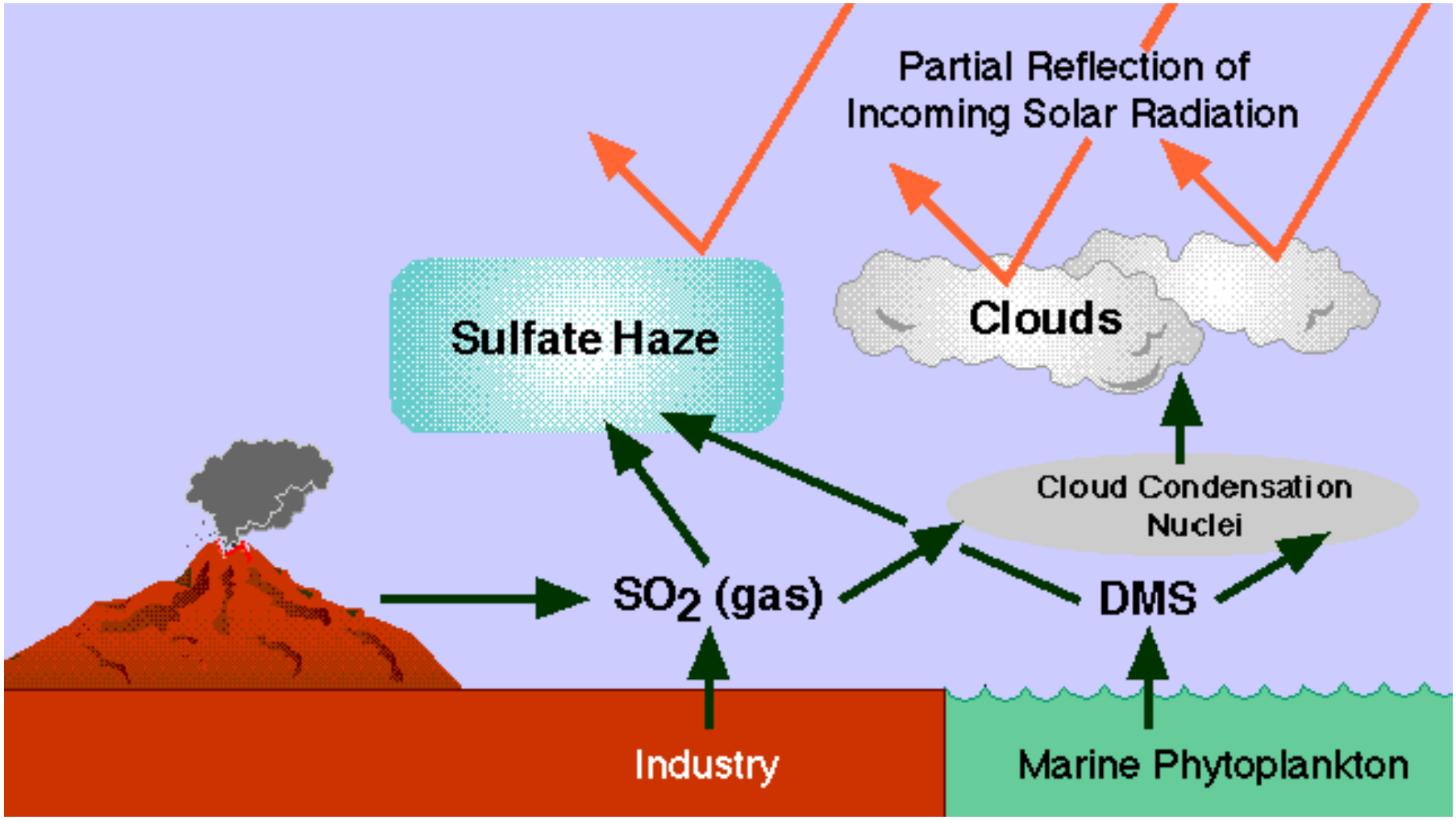
***Hydrological Effects***

Suppressed surface evaporation -- Spinning down the water cycle

Displaced precipitation -- Clouds last longer or evaporate (Rosenfeld)

# THE “WHITEHOUSE EFFECT”

## RADIATIVE FORCING OF CLIMATE CHANGE BY AEROSOLS



# DIRECT EFFECT

# AEROSOL DIRECT SHORTWAVE FORCING

Local, instantaneous, cloud-free for absorbing aerosol

Single-scattering limit (optically thin aerosol)

$$\boxed{\Delta F = -F_0 T(\theta_0) T'(1-R)^2 \beta(\theta_0) \tau \Omega}$$

***Master Equation***

- $\Omega$ — Absorption Factor
- $\tau$ — Aerosol Optical Depth
- $\beta$ — Upscatter Fraction
- $R$ — Surface Reflectance
- $T'$ — Transmittance (upward)
- $\theta_0$ — Solar Zenith Angle
- $T$ — Atmospheric Transmittance (downward)
- $F_0$ — Solar Constant
- $\Delta F$ — Change in Net TOA Flux

Aerosol Optical Depth

$$\tau = \int \sigma_{ep} dz = \int \alpha C dz$$

- $C$ — Mass Concentration
- $\alpha$ — Mass Extinction Efficiency
- $\sigma_{ep}$ — Light Extinction Coefficient

Absorption Factor

$$\Omega = \omega \left\{ 1 - \frac{2R}{(1-R)^2} \frac{(1-\omega)}{\beta\omega} \right\}$$

For $\omega = 1,\ \Omega = 1$

# AEROSOL DIRECT SHORTWAVE FORCING

Local, instantaneous, cloud-free for absorbing aerosol

Single-scattering limit (optically thin aerosol)

$$\boxed{\Delta F = -F_0 T(\theta_0) T'(1-R)^2 \beta(\theta_0) \tau \Omega}$$

***Master equation***

- $\Omega$ — Absorption Factor
- $\tau$ — Aerosol Optical Depth
- $\beta$ — Upscatter Fraction
- $R$ — Surface Reflectance
- $T'$ — Transmittance (upward)
- $\theta_0$ — Solar Zenith Angle
- $T$ — Atmospheric Transmittance (downward)
- $F_0$ — Solar Constant
- $\Delta F$ — Change in Net TOA Flux

Aerosol Optical Depth

$$\tau = \int \sigma_{ep} dz = \int \alpha C dz$$

- $C$ — Mass Concentration
- $\alpha$ — Mass Extinction Efficiency
- $\sigma_{ep}$ — Light Extinction Coefficient

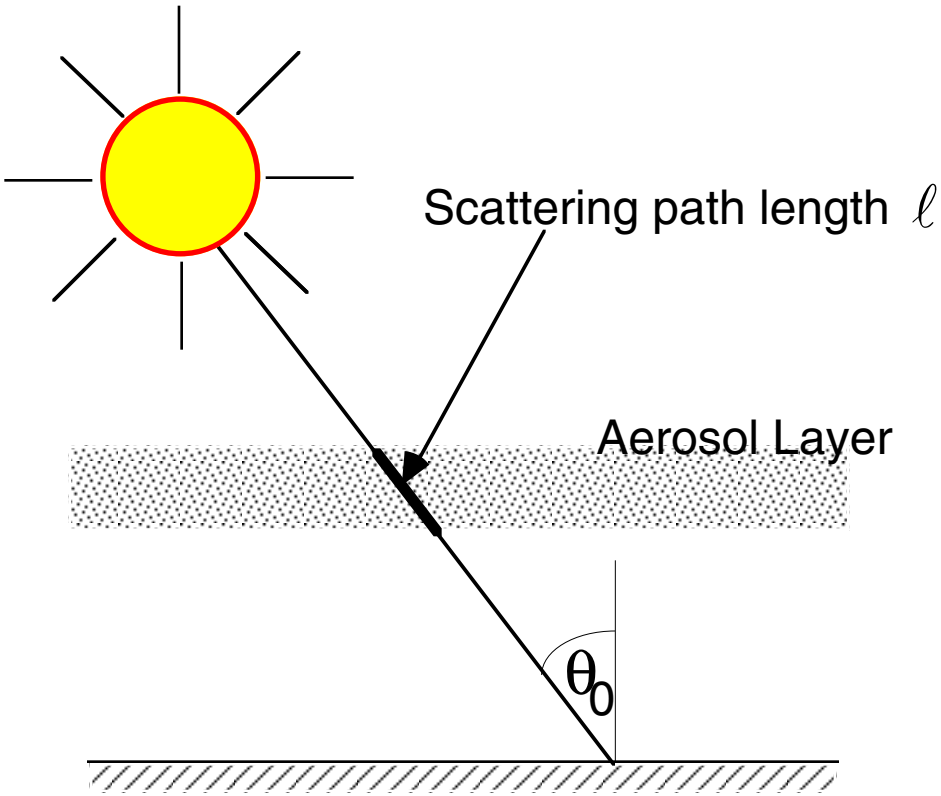
Absorption Factor

$$\Omega = \left\{ 1 - \frac{2R}{(1-R)^2} \frac{(1-\omega)}{\beta\omega} \right\}$$

For $\omega = 1,\ \Omega = 1$

# DIRECT AEROSOL FORCING

Dependence on solar zenith angle




The scattering path length $\ell$ varies as sec $\theta_0$, ***canceling the* cos $\theta_0$ *dependence*** of direct beam irradiance, so . . .

To first approximation, the direct aerosol forcing is ***independent of solar zenith angle* $\theta_0$**.

# DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE AEROSOL

$$\overline{\Delta F_R} = \overbrace{-\frac{1}{2} F_T T^2 (1 - A_c)(1 - R_s)^2}^{\text{Geophysics}} \cdot \overbrace{\underbrace{\bar{\beta}\alpha_{SO_4^{2-}} f(RH)}_{\text{Aerosol Microphysics}} \cdot \underbrace{Q_{SO_2} Y_{SO_4^{2-}} \left(\frac{MW_{SO_4^{2-}}}{MW_S}\right) \tau_{SO_4^{2-}} \Big/ A}_{\text{Column Burden Atmospheric Chemistry}}}^{\text{Aerosol Optical Depth}}$$

$\overline{\Delta F_R}$ 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

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

$\alpha_{SO_4^{2-}}$ is the scattering efficiency of **sulfate and associated cations** at a reference low relative humidity, $m^2$ (g $SO_4^{2-}$)$^{-1}$

$f$(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 $yr^{-1}$

$Y_{SO_4^{2-}}$ is the fractional yield of emitted $SO_2$ that reacts to produce sulfate aerosol

MW is the molecular weight

$\tau_{SO_4^{2-}}$ is the sulfate lifetime in the atmosphere, yr

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

*Charlson, Schwartz, Hales, Cess, Coakley, Hansen & Hofmann, Science, 1992*

# EVALUATION OF GLOBAL MEAN DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE

| Quantity | Central Value | Units | Uncertainty Factor |
|---|---|---|---|
| $F_T$ | 1370 | W m$^{-2}$ | 1.004 |
| $1\text{-}A_c$ | 0.4 | —— | 1.1 |
| $T$ | 0.76 | —— | 1.15 |
| $1\text{-}R_s$ | 0.85 | —— | 1.1 |
| $\bar{\beta}$ | 0.29 | —— | 1.3 |
| $\alpha_{SO_4^{2-}}$ | 5 | m$^2$ (g $SO_4^{2-}$)$^{-1}$ | **1.5** |
| $f$(RH) | 1.7 | —— | 1.2 |
| $Q_{SO_2}$ | 80 | Tg S yr$^{-1}$ | 1.15 |
| $Y_{SO_4^{2-}}$ | 0.4 | —— | **1.5** |
| $\tau_{SO_4^{2-}}$ | 0.02 | yr | **1.5** |
| $A$ | $5 \times 10^{14}$ | m$^2$ | — |
| $\overline{\Delta F_R}$ | -1.1 | W m$^{-2}$ | 2.4 |

$\alpha^* = 8.5$ m$^2$ (g $SO_4^{2-}$)$^{-1}$

Column Burden 4 mg $SO_4^{2-}$ m$^{-2}$

Optical Depth = 0.03

Total uncertainty factor evaluated as $f_t = \exp\left[\sum (\log f_i)^2\right]^{1/2}$

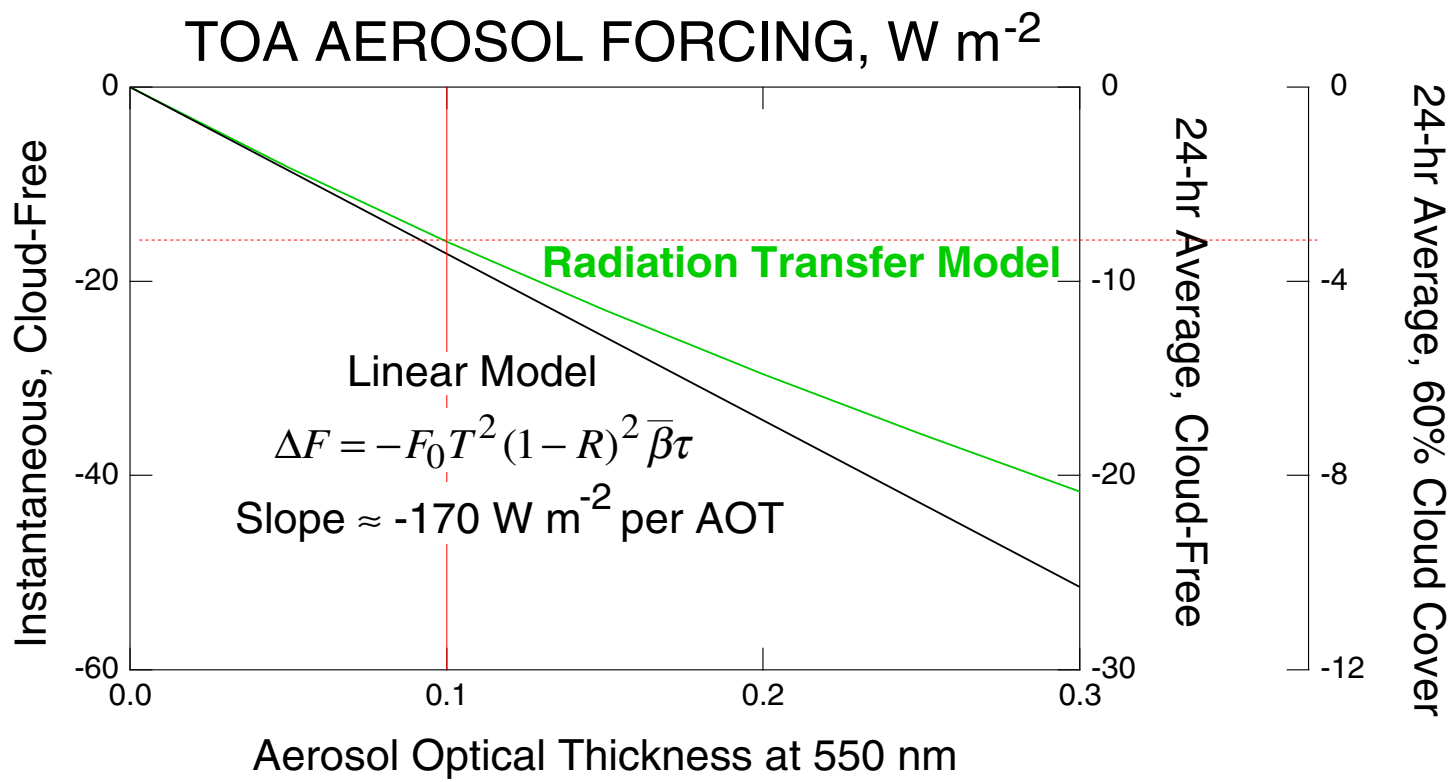
*Penner, Charlson, Hales, Laulainen, Leifer, Novakov, Ogren, Radke, Schwartz & Travis, BAMS, 1994*

# 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.




*Forcing is highly sensitive to modest aerosol loadings.*

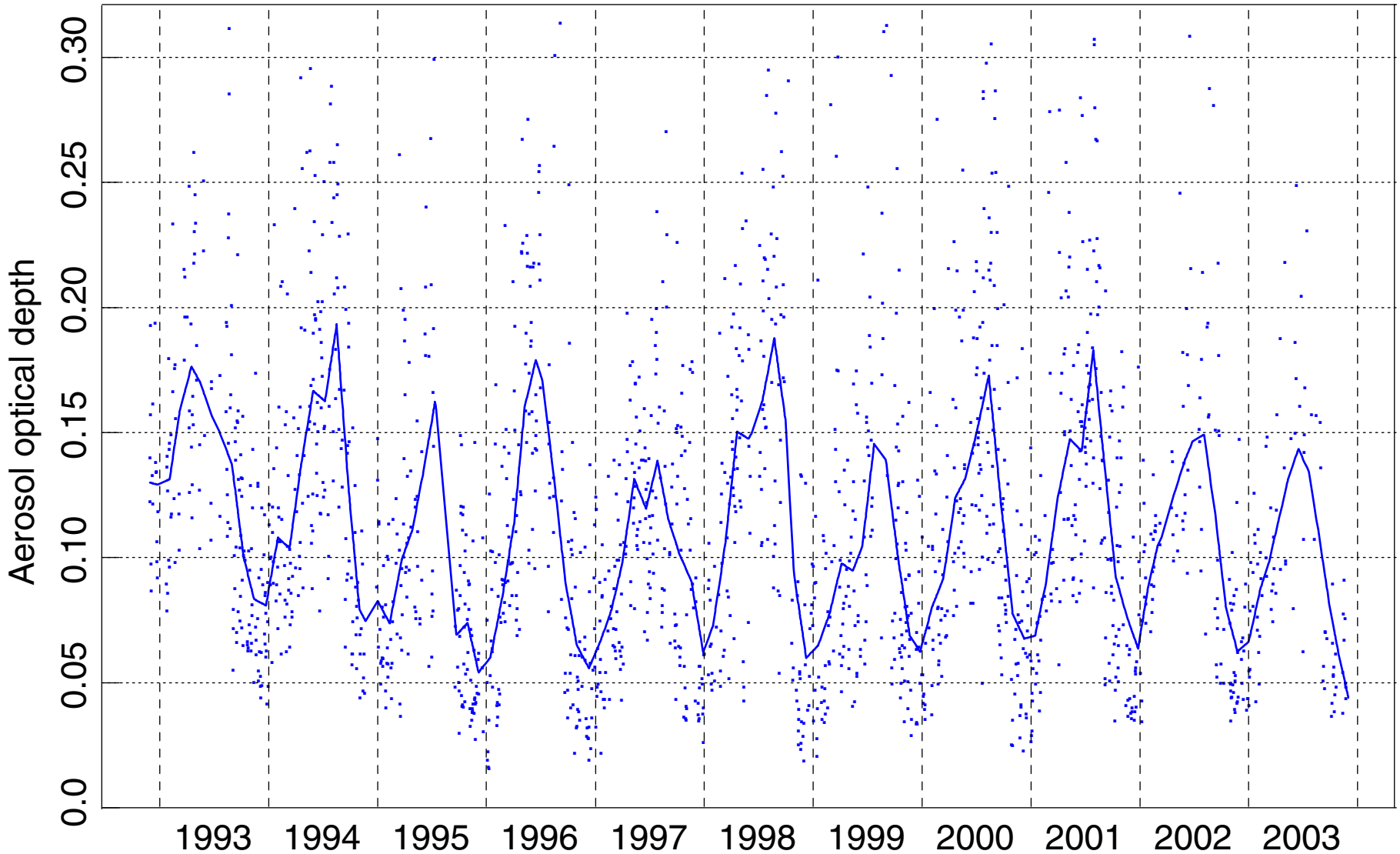
*Global-average AOT 0.1 corresponds to global-average forcing -3.2 W m$^{-2}$.*

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

# AEROSOL OPTICAL DEPTH

## Determined by sunphotometry

## North central Oklahoma - Daily average at 500 nm




*J. Michalsky et al., JGR, 2001*

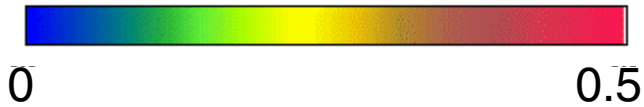
*Variability is due to variability in tropospheric aerosols.*
*Optical depth variability of 0.1 is common even at a rural mid-continental site.*

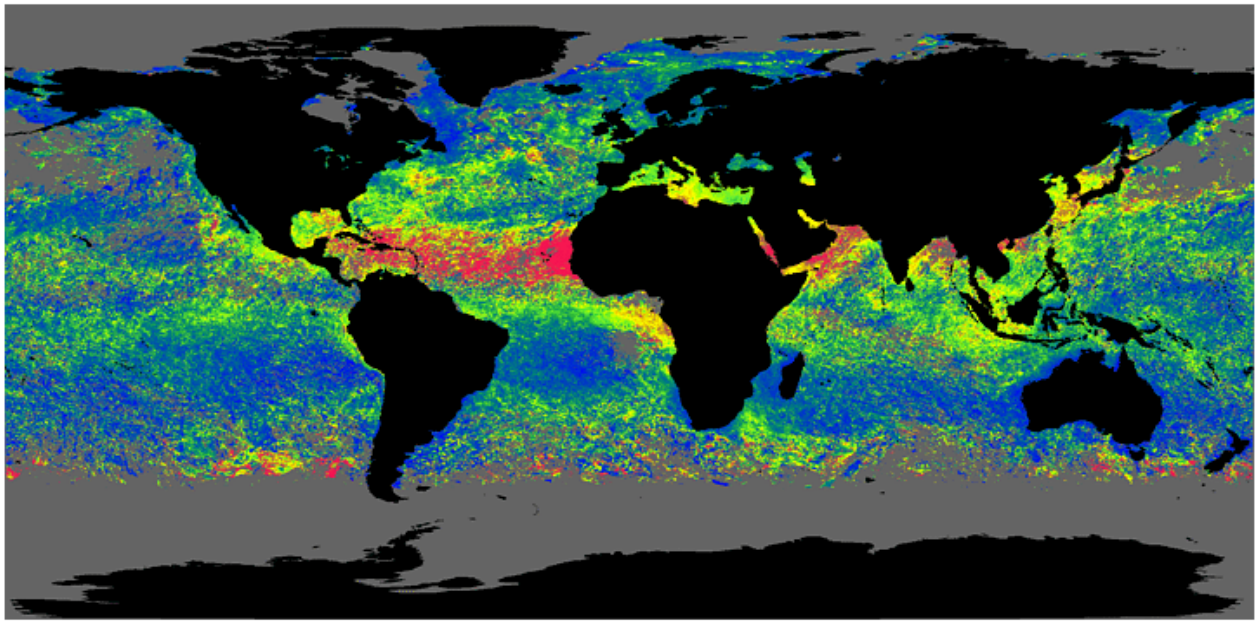
# MONTHLY AVERAGE AEROSOL JUNE 1997

## Polder radiometer on Adeos satellite

Optical Thickness $\tau$

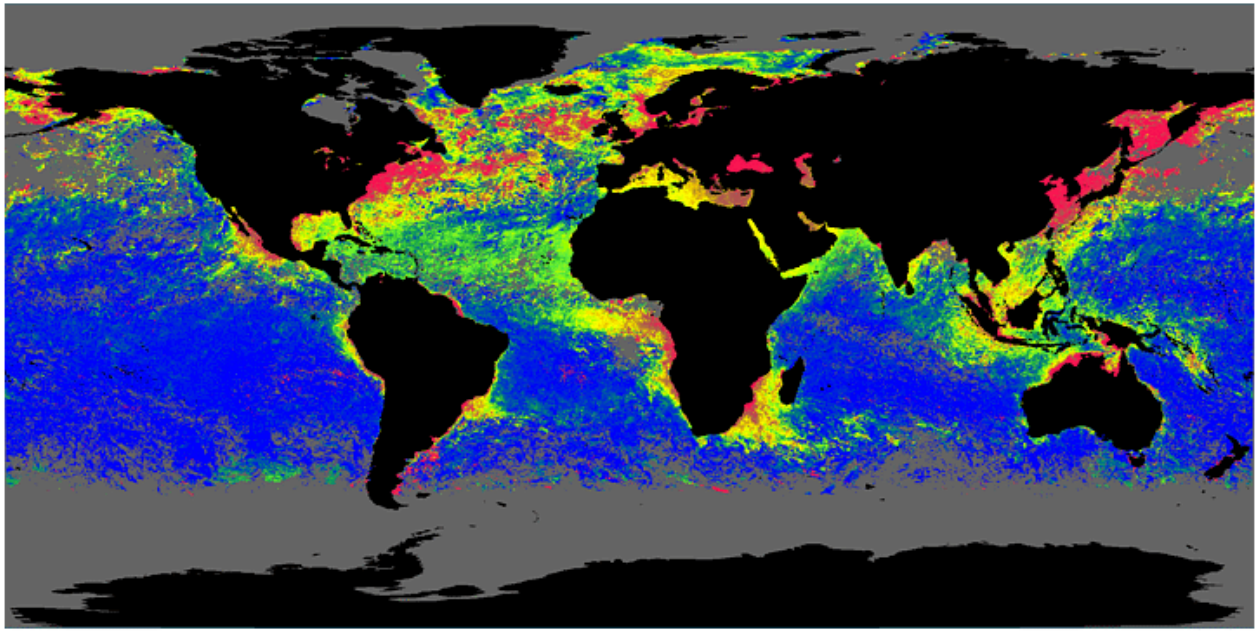
$\lambda = 865$ nm



Ångström Exponent $\alpha$

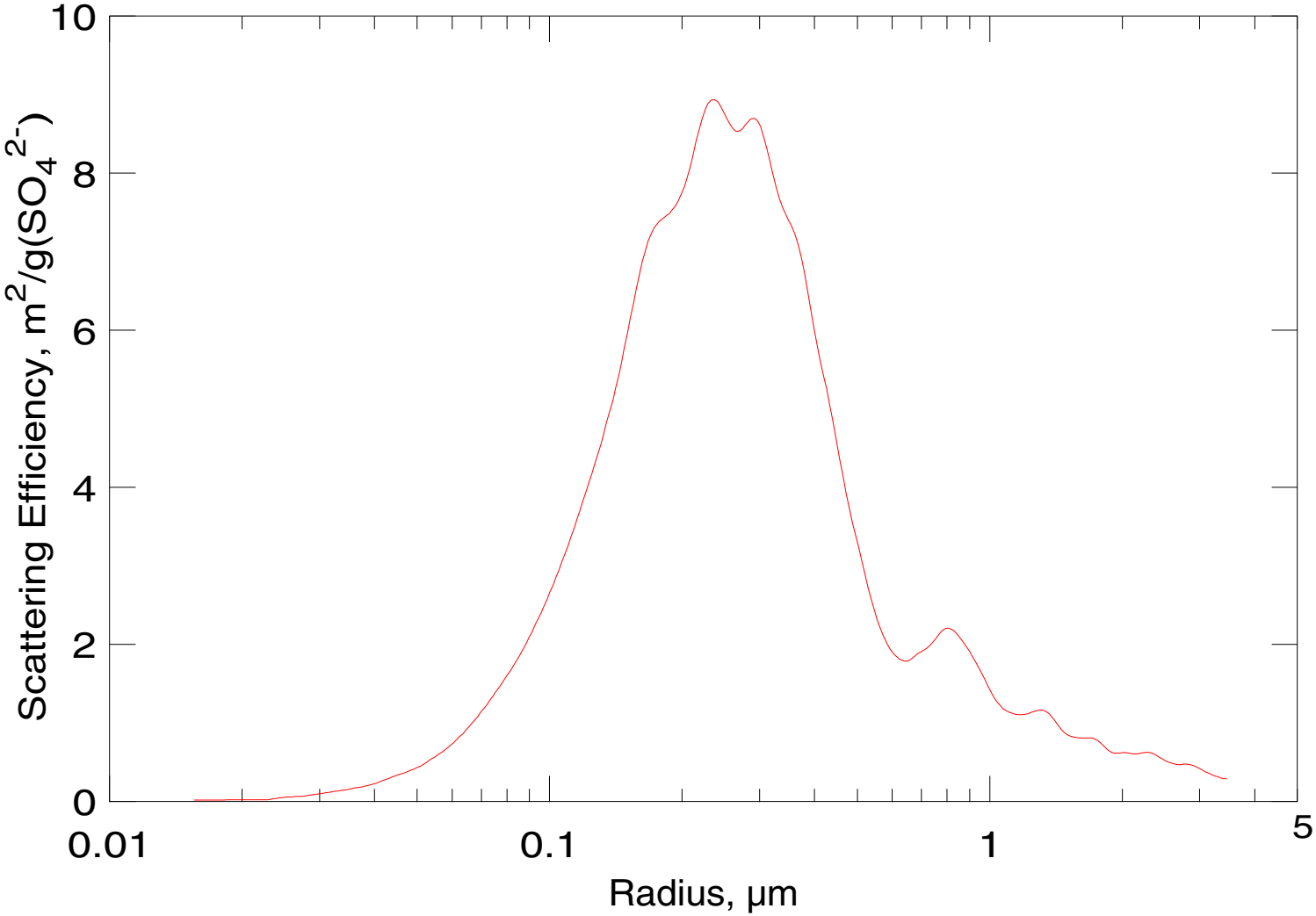
$$\alpha = -d \ln \tau / d \ln \lambda$$



# LIGHT SCATTERING EFFICIENCY

Dependence on particle radius -- Size matters!

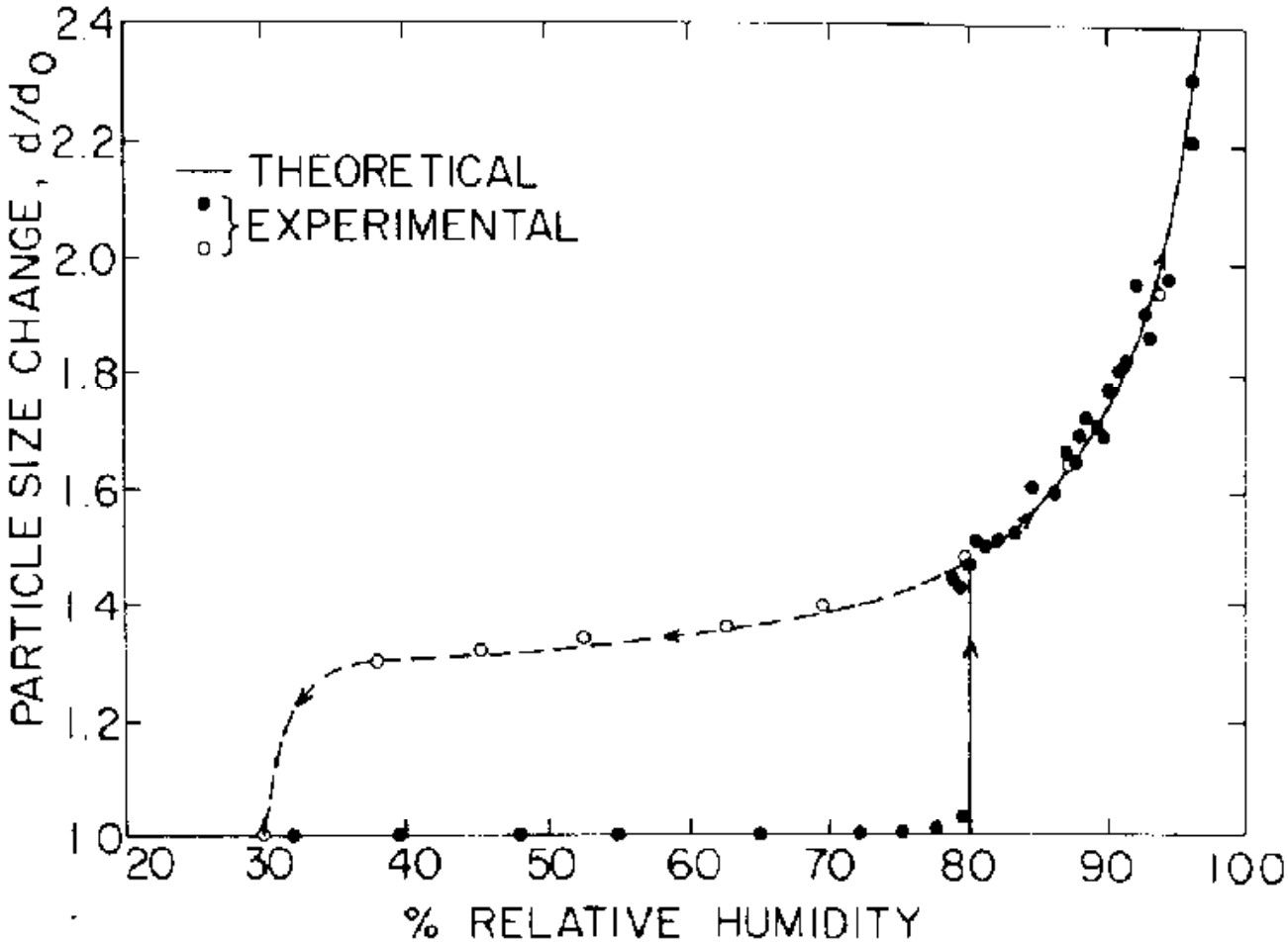
Ammonium Sulfate, 530 nm




Ouimette and Flagan, 1982

# DELIQUESCENCE, SIZE CHANGE, AND EFFLORESCENCE OF HYGROSCOPIC PARTICLES

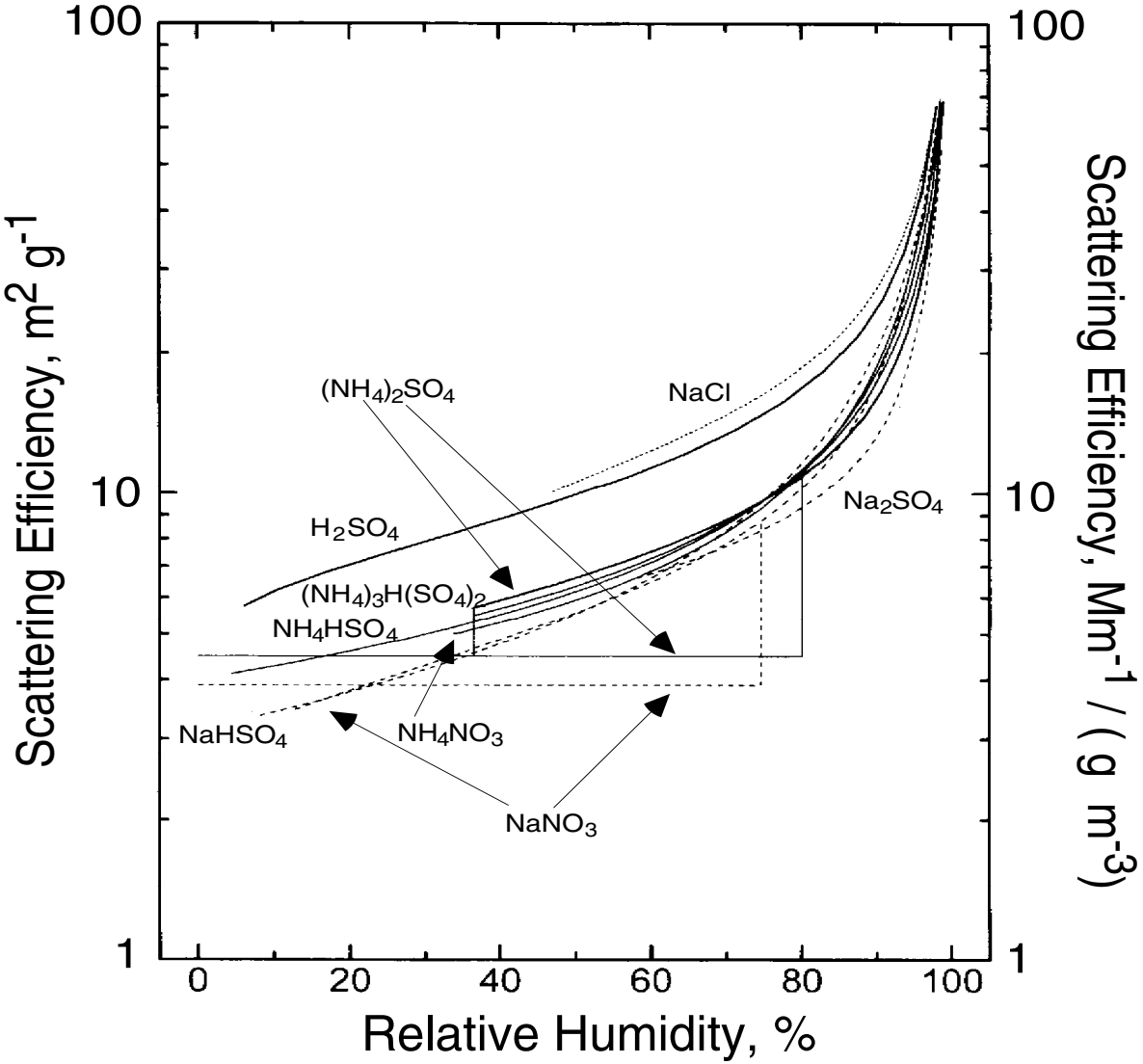
## Ammonium Sulfate at 25°C




*Tang and Munkelwitz, 1984*

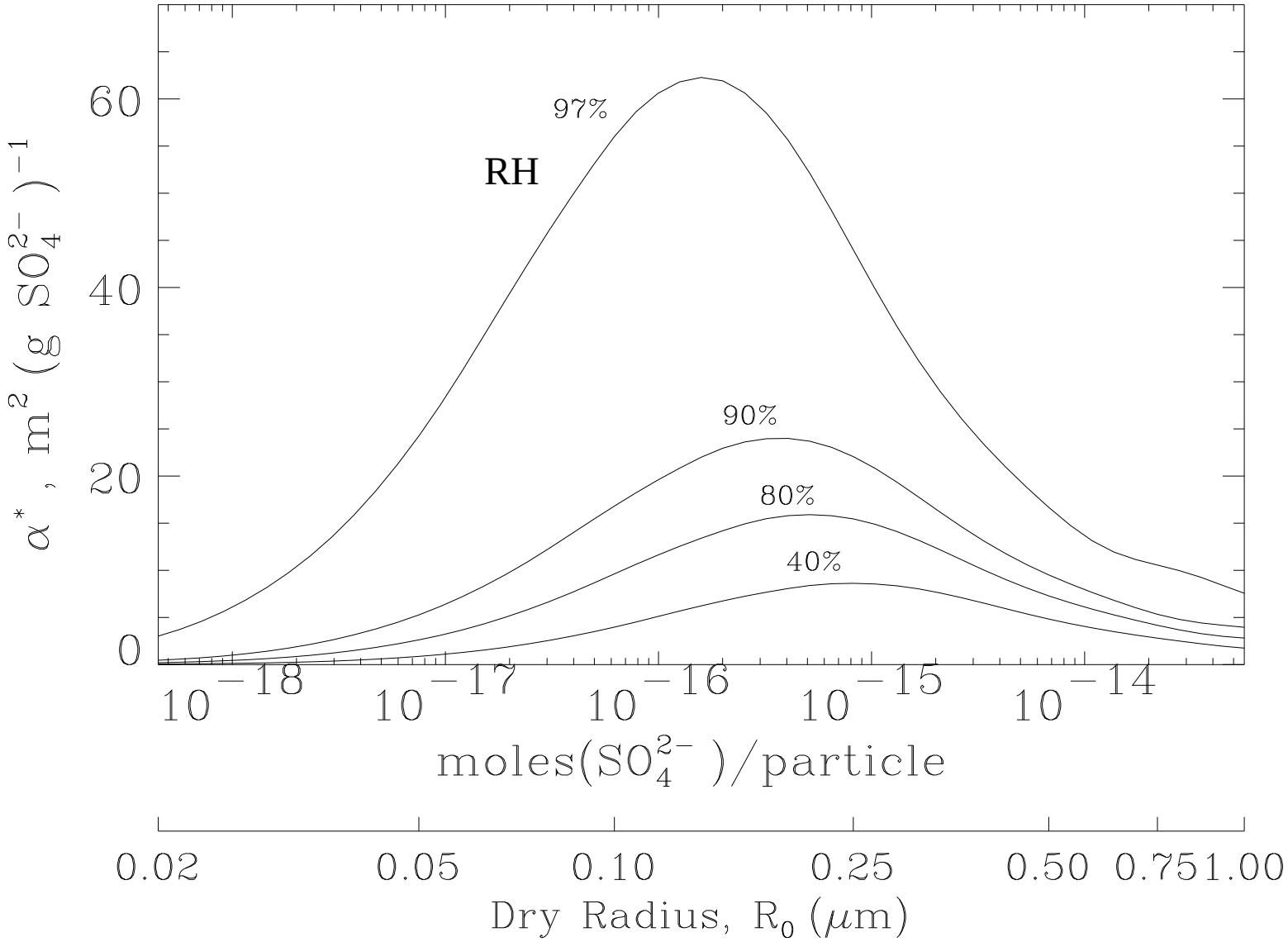
# WATER UPTAKE AND LIGHT SCATTERING COEFFICIENT

Dependence on relative humidity for various substances




*Lognormal, $D_g$ = 0.3 μm; $\sigma_g$ = 1.5; I. Tang, JGR, 1996*

# LIGHT SCATTERING EFFICIENCY OF $(NH_4)_2SO_4$ DEPENDENCE ON PARTICLE SIZE AND RH

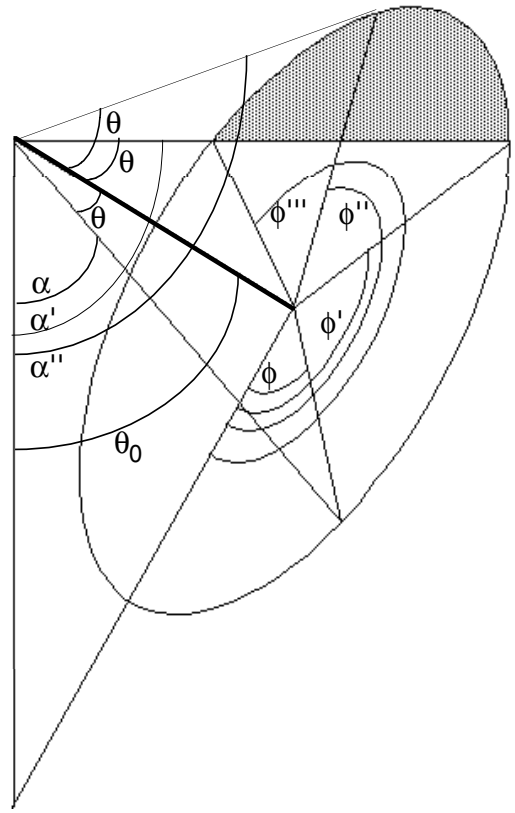



*Nemesure, Wagener & Schwartz, JGR, 1995*

*Scattering, optical depth, and forcing are highly sensitive to particle size and to hygroscopic growth, which depends on composition.*

# UPSCATTER FRACTION

## SCATTERING OF SOLAR RADIATION BY AEROSOL PARTICLE

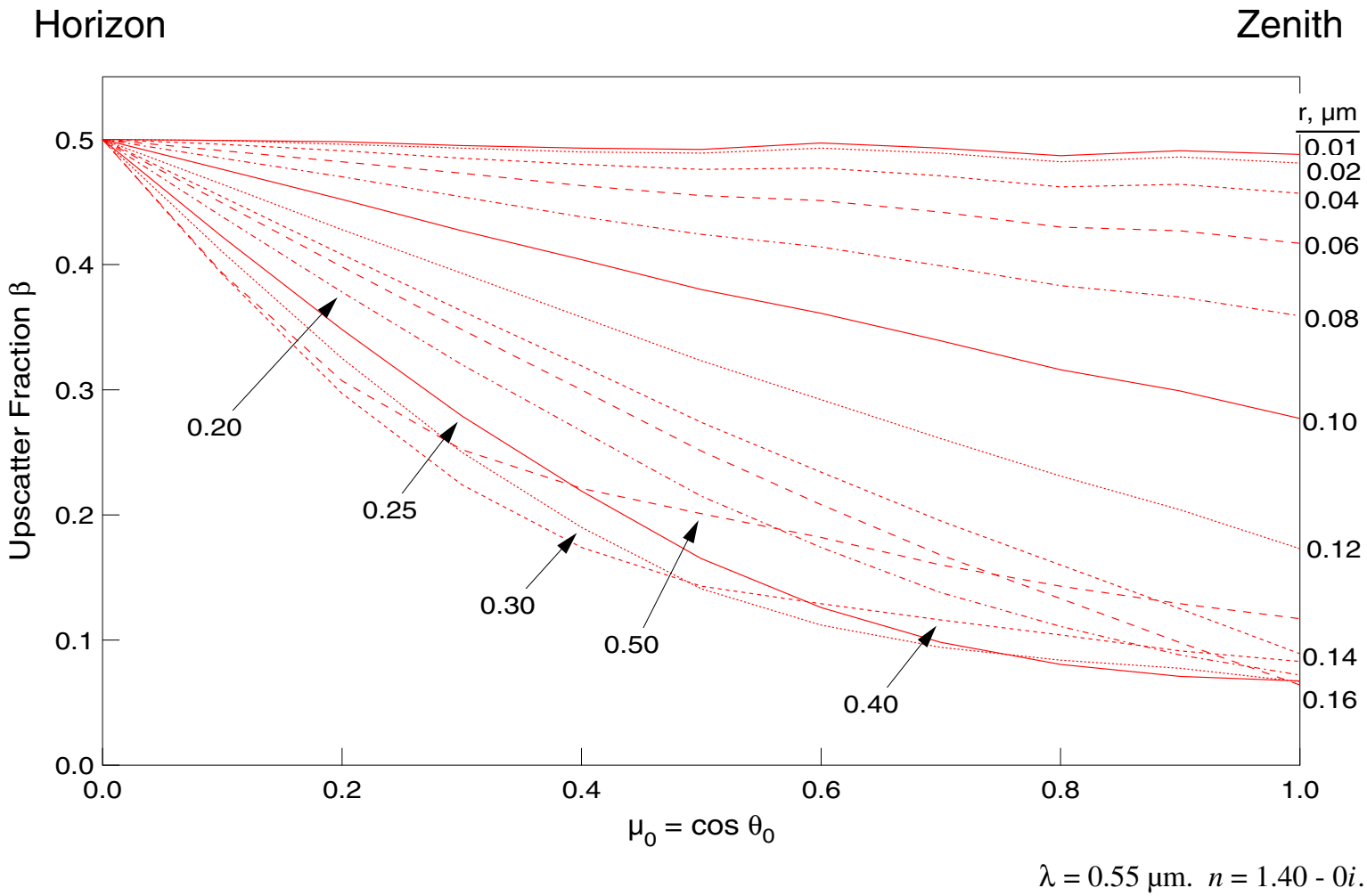



*Upscatter fraction* β is the fraction of radiation scattered into the upward hemisphere.

$$\beta = \int_{\substack{\text{upward}\\ \text{hemisphere}}} P(\theta,\phi)\,d\Omega \Big/ \int_{4\pi} P(\theta,\phi)\,d\Omega = \int_{\cos\alpha<0} P(\theta,\phi)\,d\Omega \Big/ 4\pi$$

# UPSCATTER FRACTION

## Dependence on solar zenith angle and particle radius



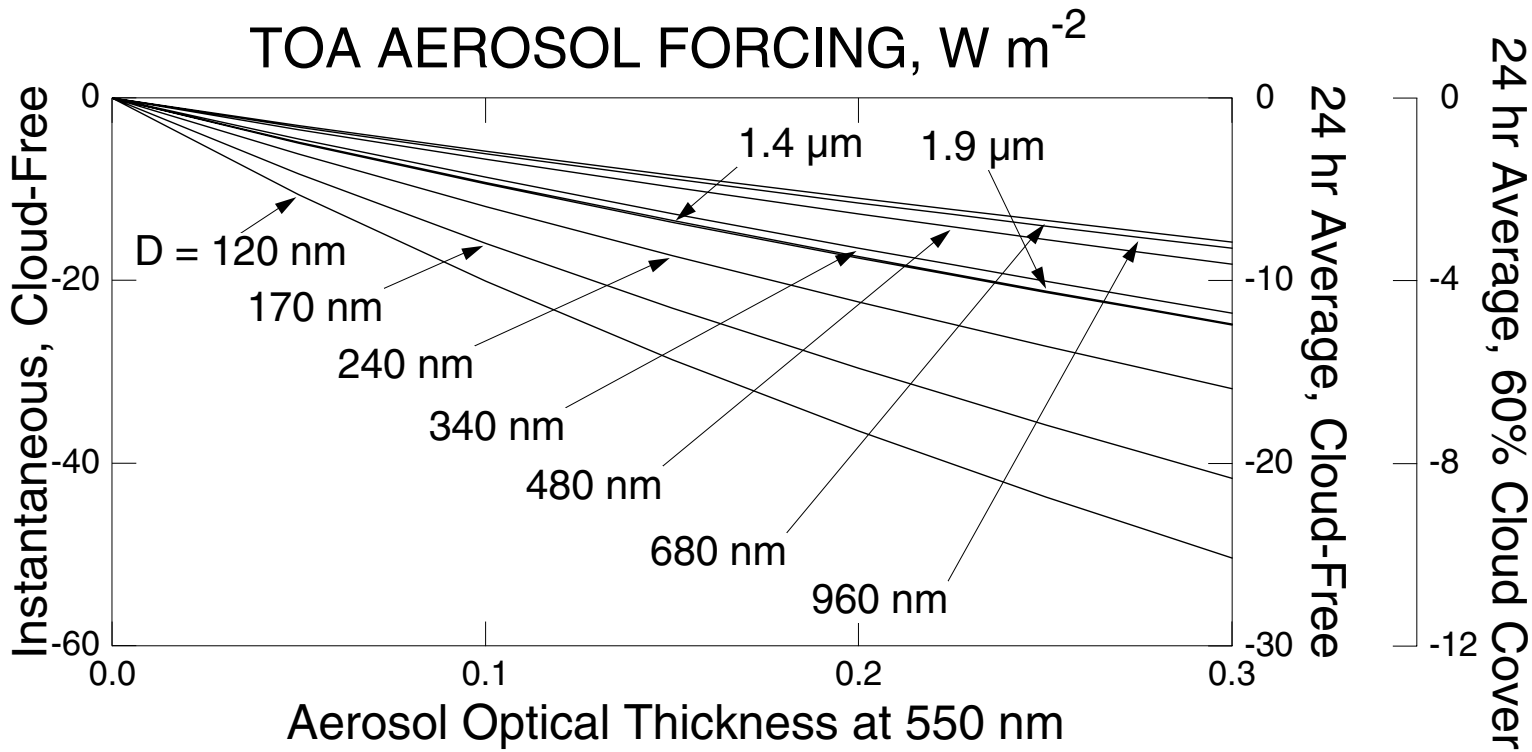

For sun at horizon $\beta = 0.5$ (by symmetry).

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

# DIRECT AEROSOL TOA FORCING

## Dependence on particle diameter

Surface reflectance $R = 0.15$; single scatter albedo $\omega_0 = 1$.

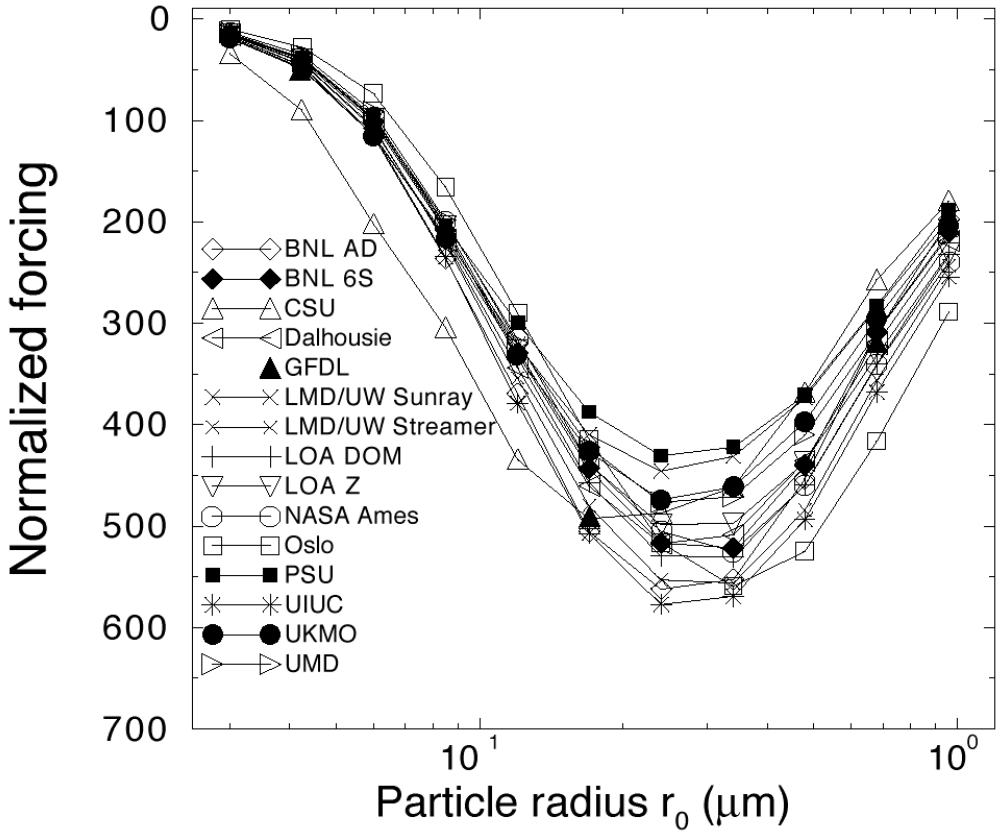



*Forcing per optical depth depends on particle size.*

# INTERCOMPARISON OF BROADBAND SHORTWAVE FORCING BY AMMONIUM SULFATE AEROSOL

Normalized global-average forcing: W $m^{-2}$ / g($SO_4^{2-}$) $m^{-2}$ or W /g($SO_4^{2-}$)

Aerosol optical depth 0.2; surface albedo 0.15



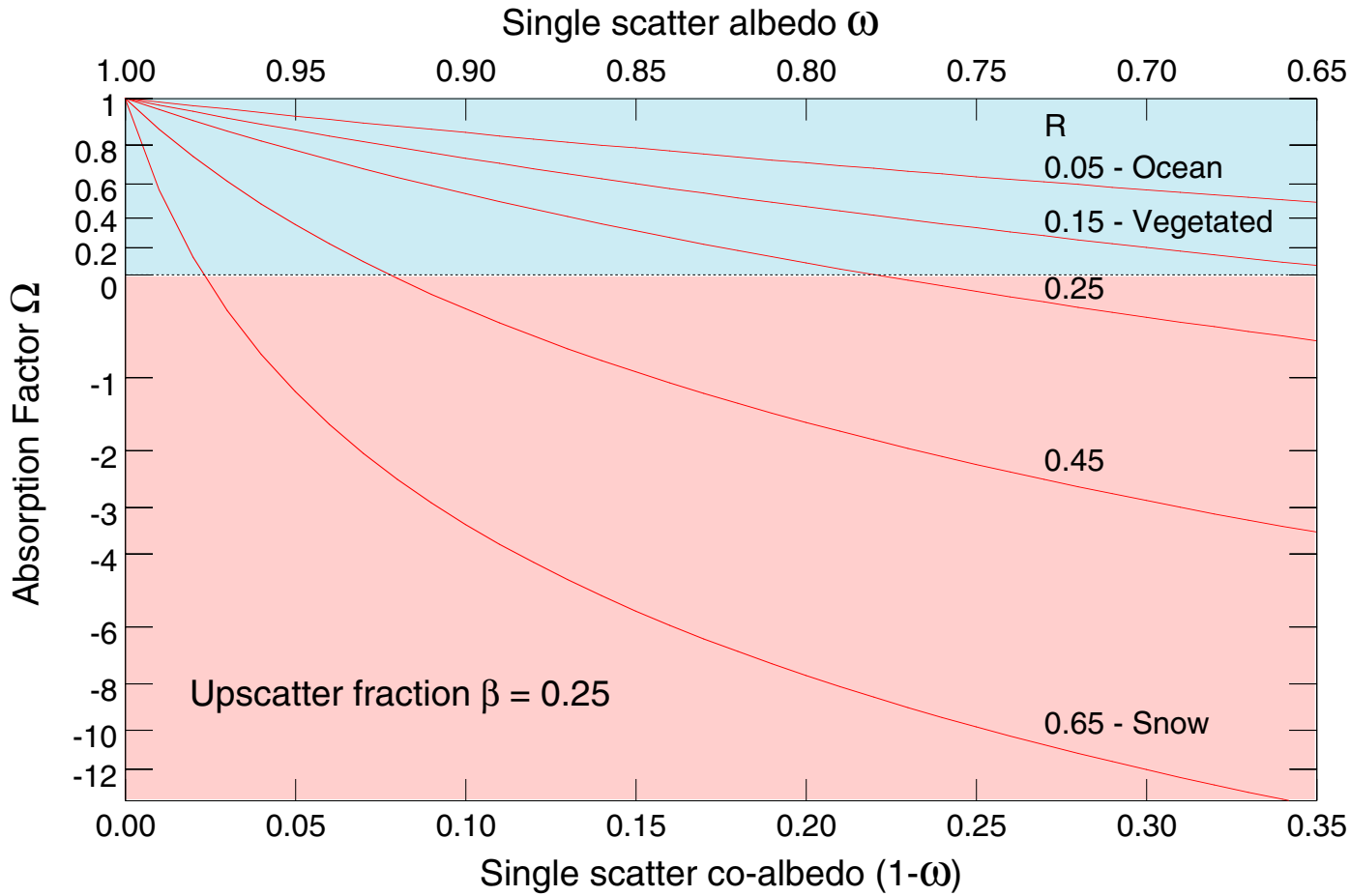

Standard deviation ~8% for 15 models at radius ~ 200 nm.

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

*Radiation transfer models agree closely for well specified aerosol.*

# DECREASE IN FORCING DUE TO ABSORPTION

Dependence on single scattering albedo ω and surface reflectance *R*



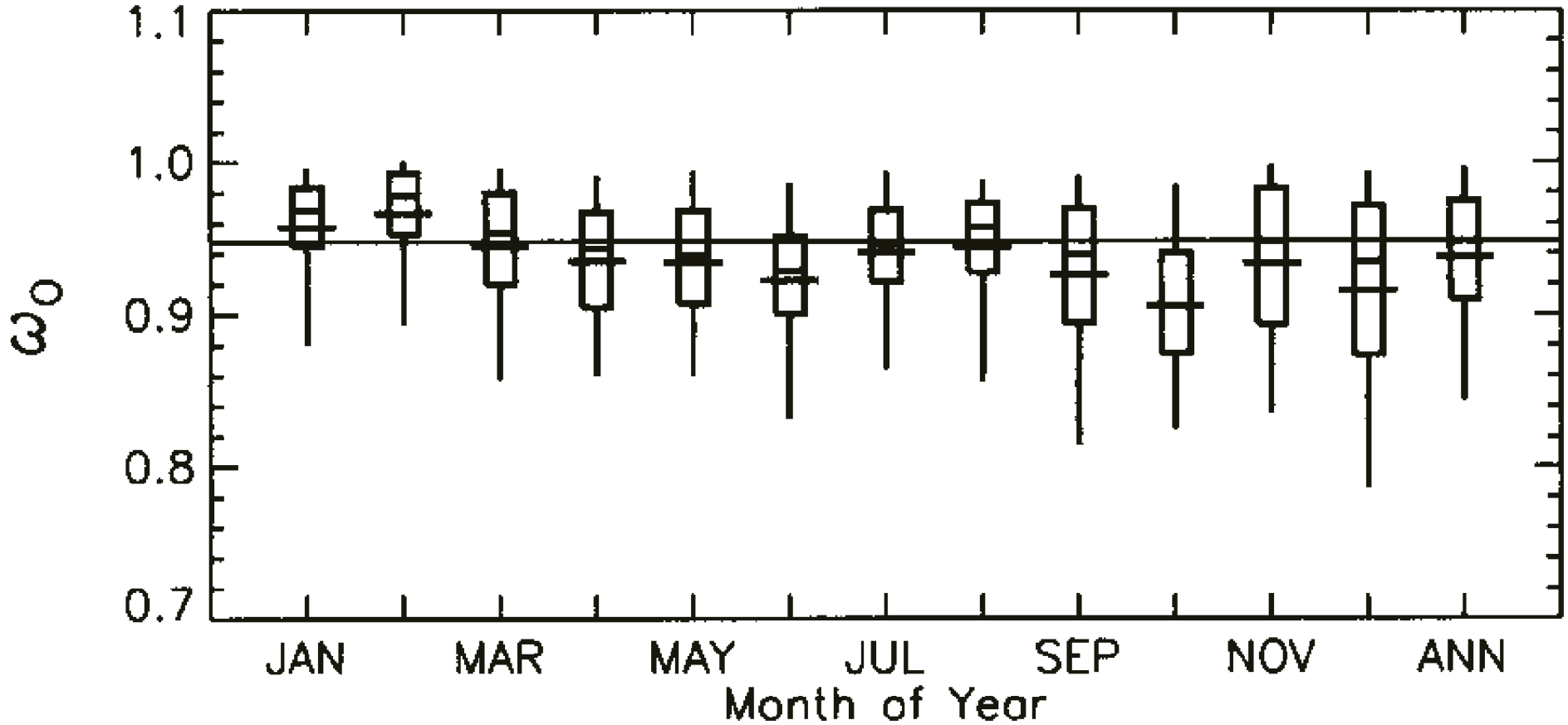

Note nonlinear scale.

Negative absorption factor results in positive (warming) forcing.

Forcing can be strongly warming over snow or clouds.

# AEROSOL SINGLE SCATTERING ALBEDO

North central Oklahoma, 4 years of hourly data, sub 10 μm diameter




*Sheridan, Delene & Ogren, JGR, 2001*

Median $\omega = 0.95$

For great majority of data $\omega > 0.9$

# AEROSOL INFORMATION REQUIRED TO DETERMINE DIRECT FORCING

***Time-dependent 3-D map*** of size-dependent particle concentration, composition, and morphology.

***Needed*** for computation of optical properties, cloud-nucleating properties, and radiative and hydrological influences.

***This can be obtained only by chemical transport modeling of aerosols . . .***

Based on ***understanding*** of the controlling processes.

***Evaluated*** by comparison with observations.

# REPRESENTING AEROSOL PROCESSES IN CTMs

***Sources*** of primary particles and precursor gases

***3-D transport*** as controlled by governing meteorology

***Gas to particle conversion*** (clear air and cloud)
New particle formation
Accretion onto existing particles

***Particle size evolution*** (clear air and cloud)

Particle ***morphology*** and intra-particle structure

***Removal*** of particles and precursor gases, controlled by meteorology

Aerosol properties as ***bivariate functions of particle size and composition***

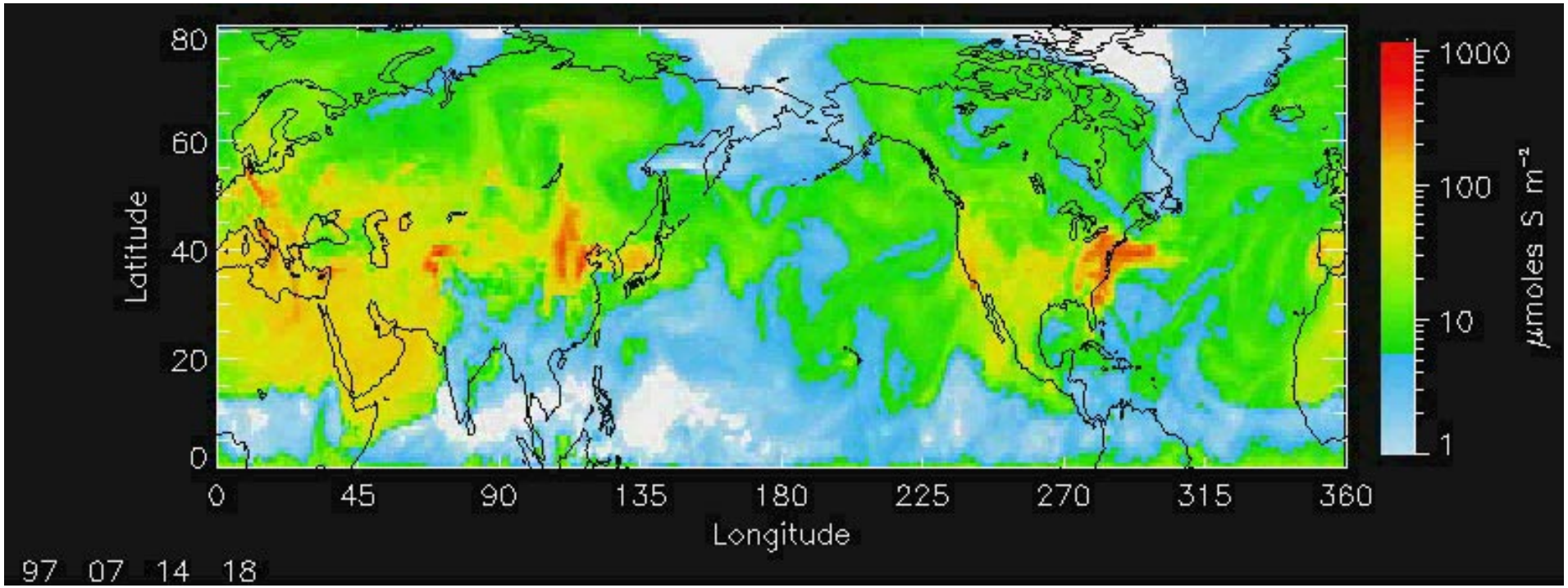
Aerosol optical properties (extinction coefficient, single scatter albedo, asymmetry parameter) as integrals over single particle properties (Mie)

All as a function of ***3-D location and secular time***

# HEMISPHERIC DISTRIBUTION OF SULFATE COLUMN BURDEN

## Vertical integral of concentration

### July 14, 1997, 1800 UTC

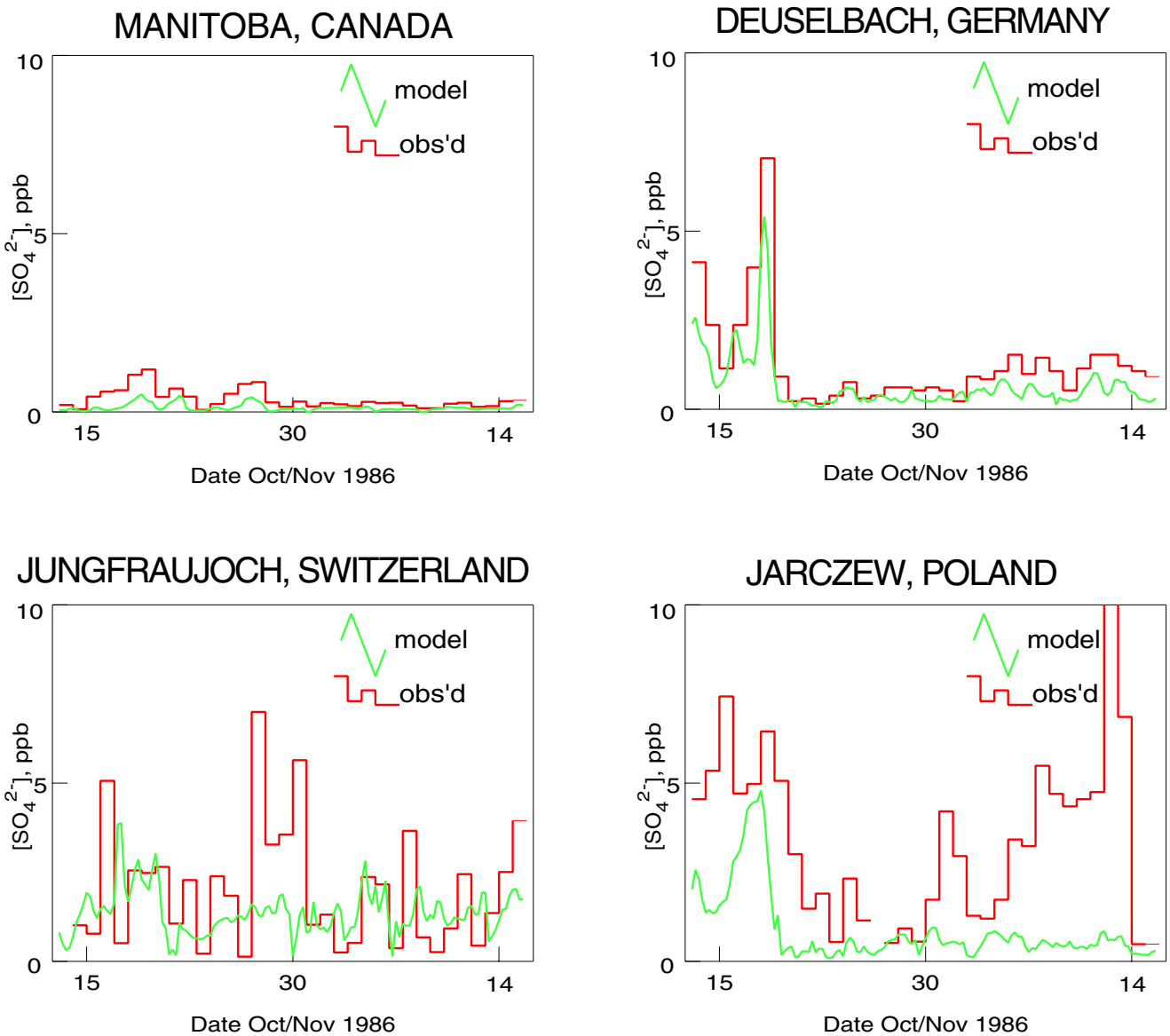



Brookhaven National Laboratory Chemical Transport Model

*Benkovitz & Schwartz, JGR, In press 2004*

# COMPARISON OF MODEL AND OBSERVATIONS

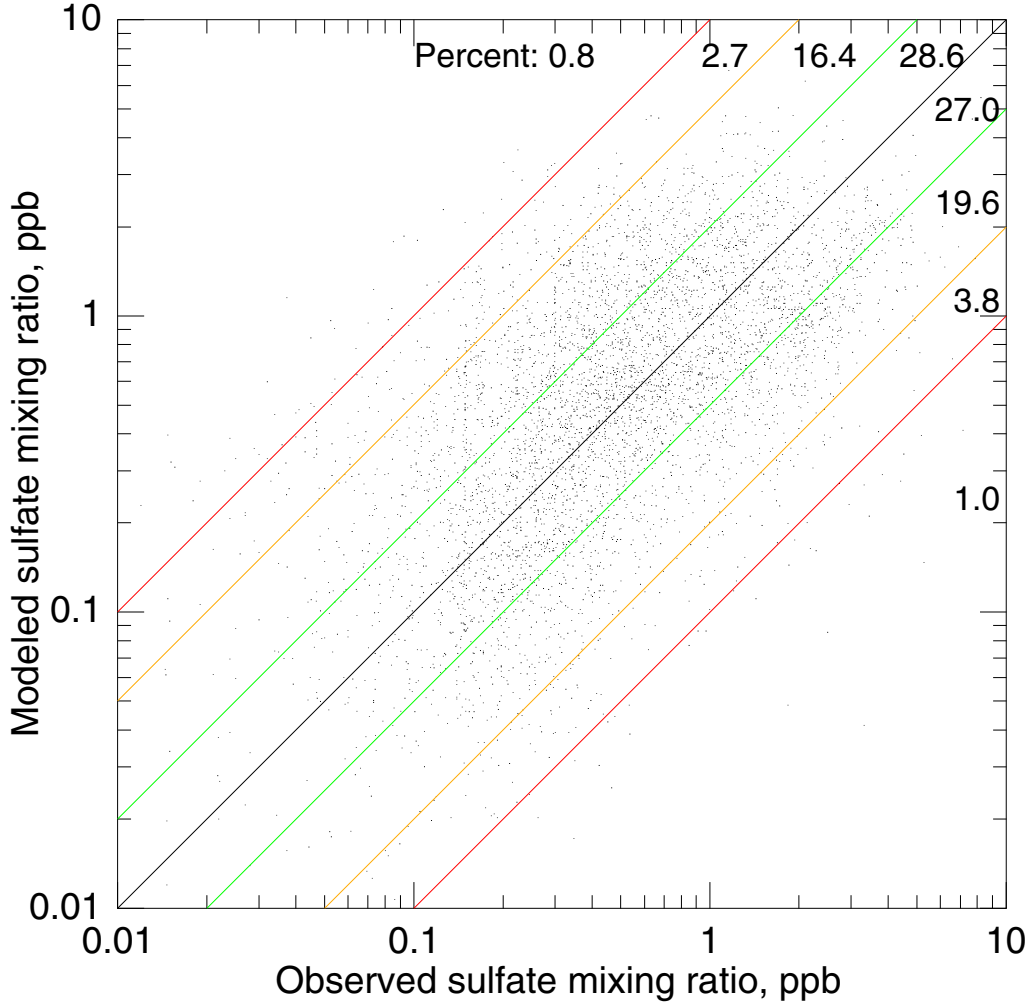
## Comparisons for 24-hr sulfate mixing ratio at surface




*Benkovitz et al., JGR, 1994*

# 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.

*Benkovitz & Schwartz, JGR, In press 2004*

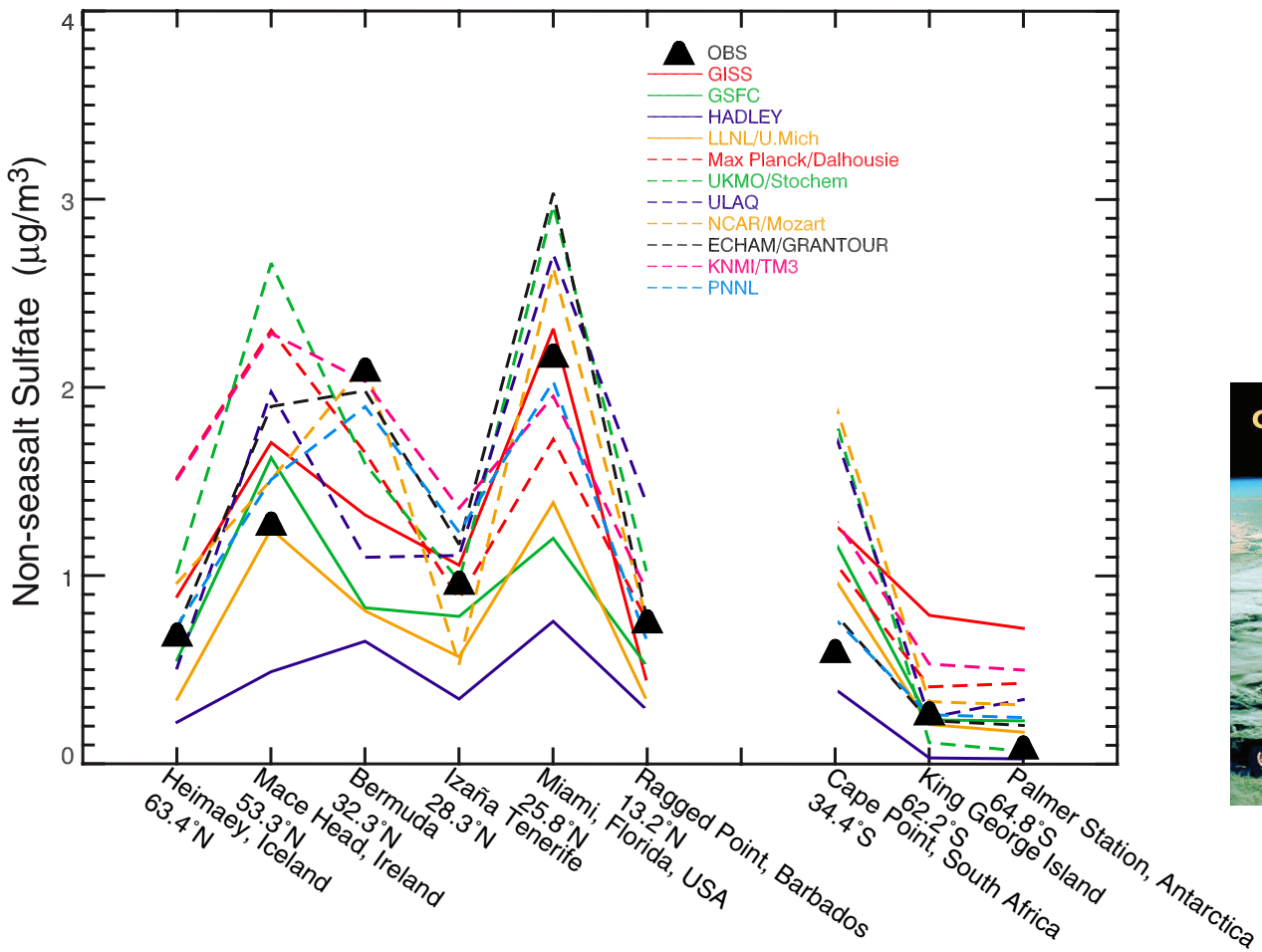
# STATISTICS OF SEVERAL SULFATE CTMs

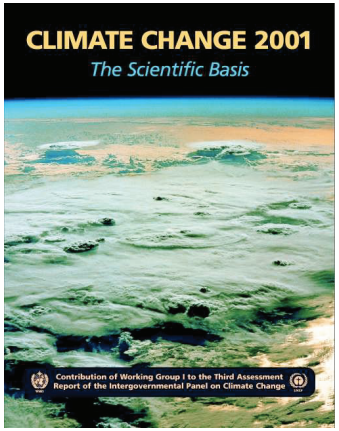
| Model | F96 | L97 | R98 | K99 | R00 | C00 | B97 | B04 |
|---|---|---|---|---|---|---|---|---|
| **$SO_2$ Sink Rate (%/day)** | | | | | | | | |
| Dry Deposition | 26 | 10 | 8 | 17 | 16 | 26 | 12 | 24 |
| Wet Deposition | 5 | 0 | 0 | 0 | 1 | 7 | 0 | $4 \times 10^{-4}$ |
| Gas Conversion | 11 | 7 | 8 | 6 | 6 | 9 | 8 | 14 |
| Aqueous Conversion to sulfate | 22 | 26 | 27 | 15 | 29 | 15 | 16 | 30 |
| Oxidation & immediate wet dep | | | | | | | | 21 |
| **Sulfate Sink Rate (%/day)** | | | | | | | | |
| Dry Deposition | 3 | 5 | 5 | 4 | 2 | 2 | 3 | 2 |
| Wet Deposition | 20 | 14 | 17 | 14 | 23 | 15 | 18 | 13 |
| **Inverse Lifetime (%/day)** | | | | | | | | |
| $SO_2$ | 63 | 43 | 42 | 38 | 53 | 56 | 36 | 90 |
| Sulfate | 23 | 19 | 21 | 18 | 25 | 17 | 21 | 15 |
| **Sulfate yield, %** | | | | | | | | |
| | 51 | 76 | 82 | 55 | 68 | 43 | 66 | 50 |
| **Burden (Tg S)** | | | | | | | | |
| $SO_2$ | 0.33 | 0.56 | 0.61 | 0.56 | 0.4 | 0.43 | | 0.20 |
| Sulfate | 0.43 | 1.05 | 0.96 | 0.73 | 0.60 | 0.63 | | 0.60 |
| **Sulfate Potential (days)** | | | | | | | | |
| | 2.1 | 4.4 | 3.7 | 3.4 | 2.5 | 2.7 | | 3.3 |

*Benkovitz & Schwartz, 2004*

# SULFATE MODEL INTERCOMPARISON

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



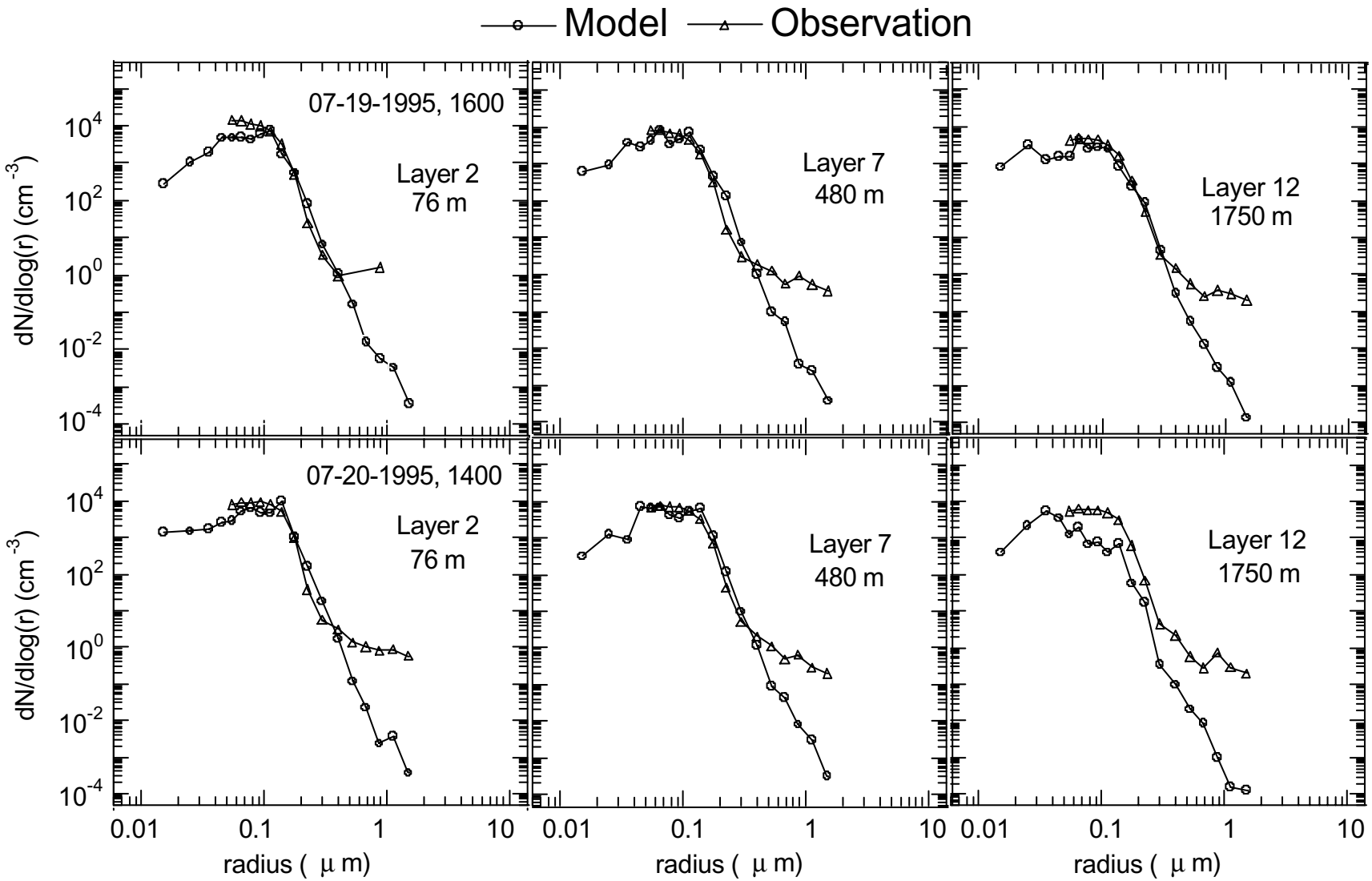

*Penner et al., IPCC, 2001*

*"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."*

# SIZE DISTRIBUTIONS

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

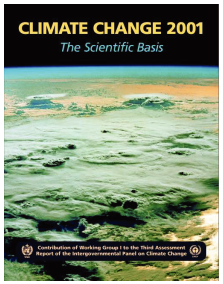



Yu, Kasibhatla, Wright, Schwartz, McGraw & Deng, *JGR*, 2003

# UNCERTAINTY BUDGET FOR *DIRECT* FORCING BY INDUSTRIAL AEROSOLS

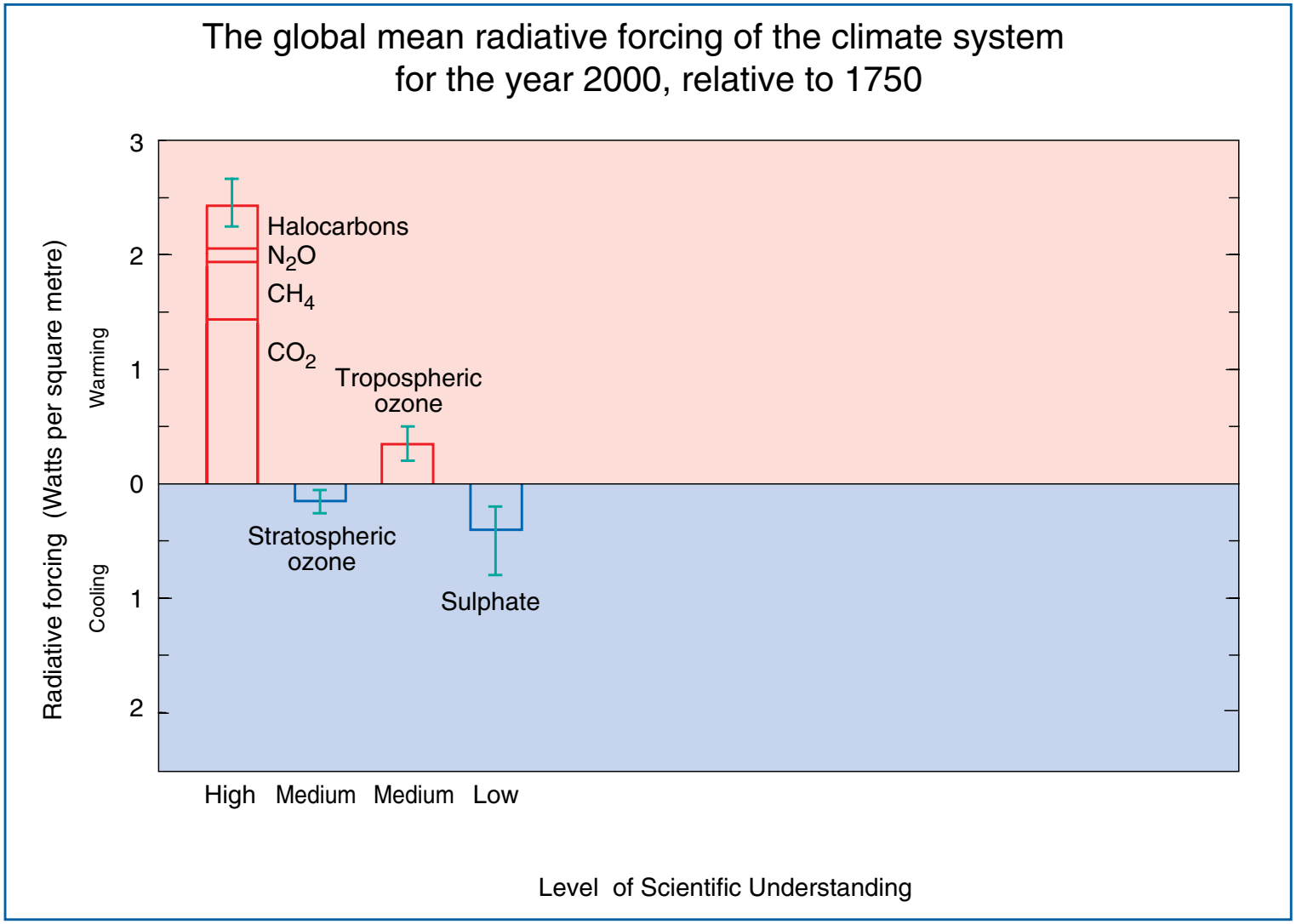
| Quantity | Central Value | 2/3 Uncertainty Range | Uncertainty Factor |
|---|---|---|---|
| | | | |
| | | | |
| | | | |
| | | | |
| Total emission of anthropogenic sulfate from fossil fuel burning (Tg/yr) | 69 | 57.5 to 82.8 | 1.4 |
| Atmospheric burden of sulfate from fossil fuel burning (Tg S) | 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 ($m^2 g^{-1}$), $\alpha_s$ | 3.5 | 2.3 to 4.7 | **2.0** |
| Aerosol single scattering albedo, co-albedo (dry), $\omega_0$, $1-\omega_0$ | 1 | | |
| $T_a$, 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: Central value of forcing is $-0.5\ Wm^{-2}$ ; the uncertainty range is from -0.25 to -1 $Wm^{-2}$. | | | **4** |

*Modified from Penner et al., IPCC, 2001*



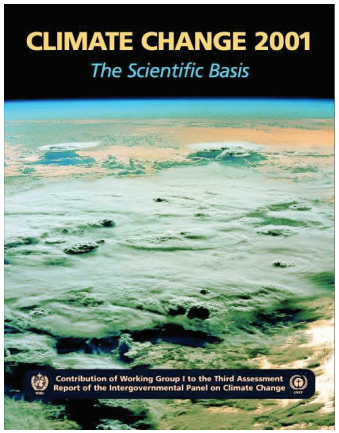

Greatest uncertainties are in burden and optical properties.

# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
# IPCC (2001)
## GHG's and sulfate aerosol direct effects




Summary for Policymakers

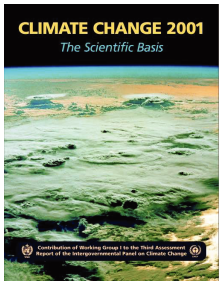
A Report of Working Group I of the Intergovernmental Panel on Climate Change



# UNCERTAINTY BUDGET FOR *DIRECT* FORCING BY INDUSTRIAL AEROSOLS

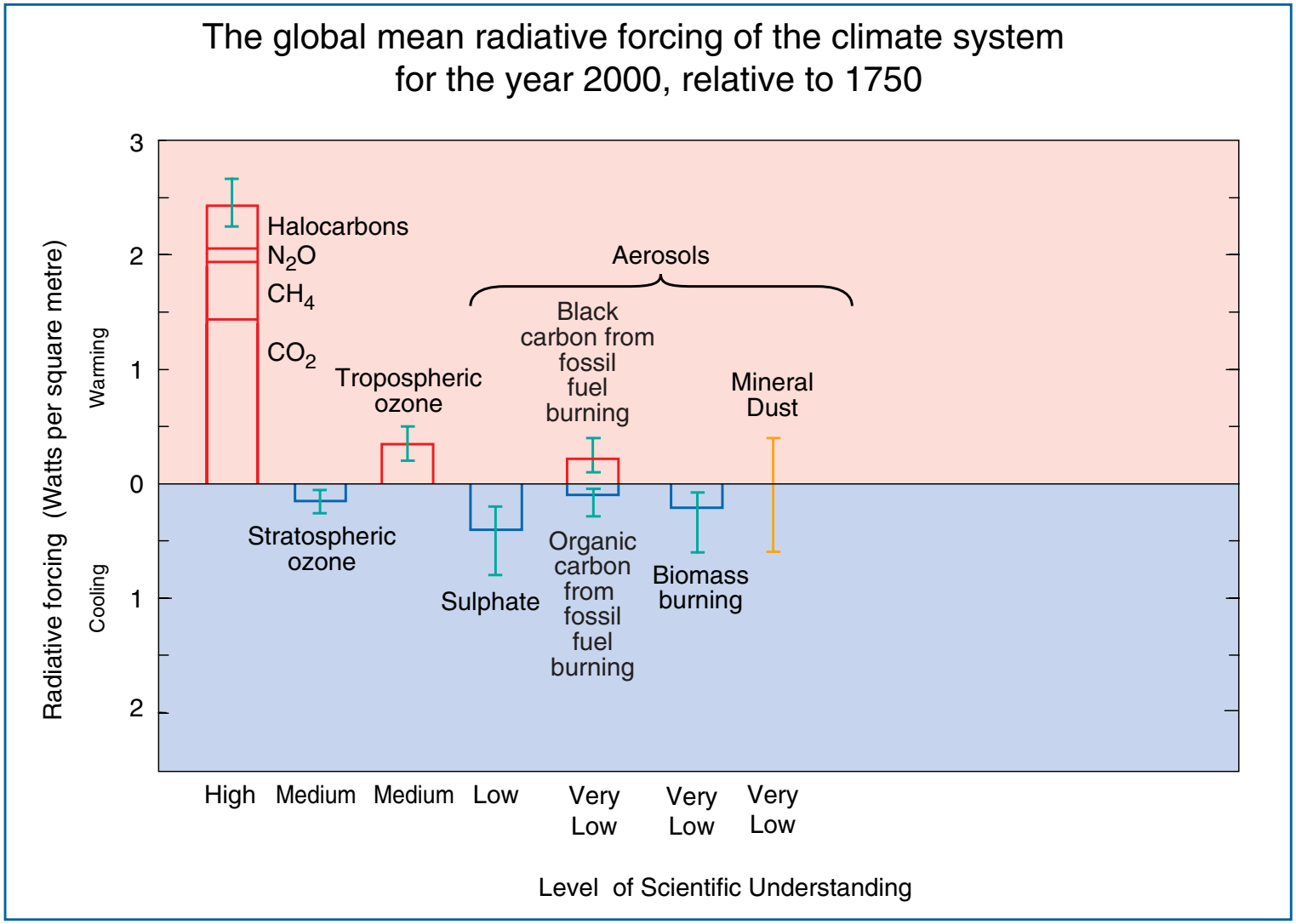
| Quantity | Central Value | 2/3 Uncertainty Range | Uncertainty Factor |
|---|---|---|---|
| Total emission of anthropogenic OC from fossil fuel burning (Tg/yr) | 20 | 10 to 30 | **3.0** |
| Atmospheric burden of OC from fossil fuels (Tg) | 0.48 | 0.33 to 0.70 | **2.1** |
| Total emission of anthropogenic 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 ($m^2 g^{-1}$), $\alpha_s$ | 3.5 | 2.3 to 4.7 | **2.0** |
| Aerosol single scattering albedo, co-albedo (dry), $\omega_0$, $1-\omega_0$ | 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*



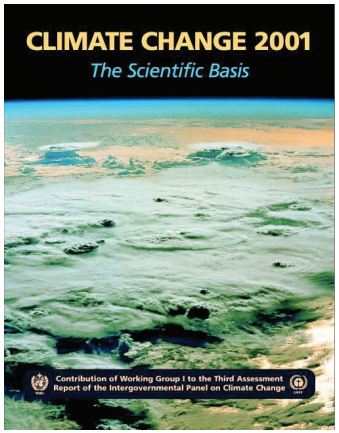

Greatest uncertainties are in chemical, microphysical, and optical properties.

# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
# IPCC (2001)
## GHG's and aerosol direct effects




Summary for Policymakers

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

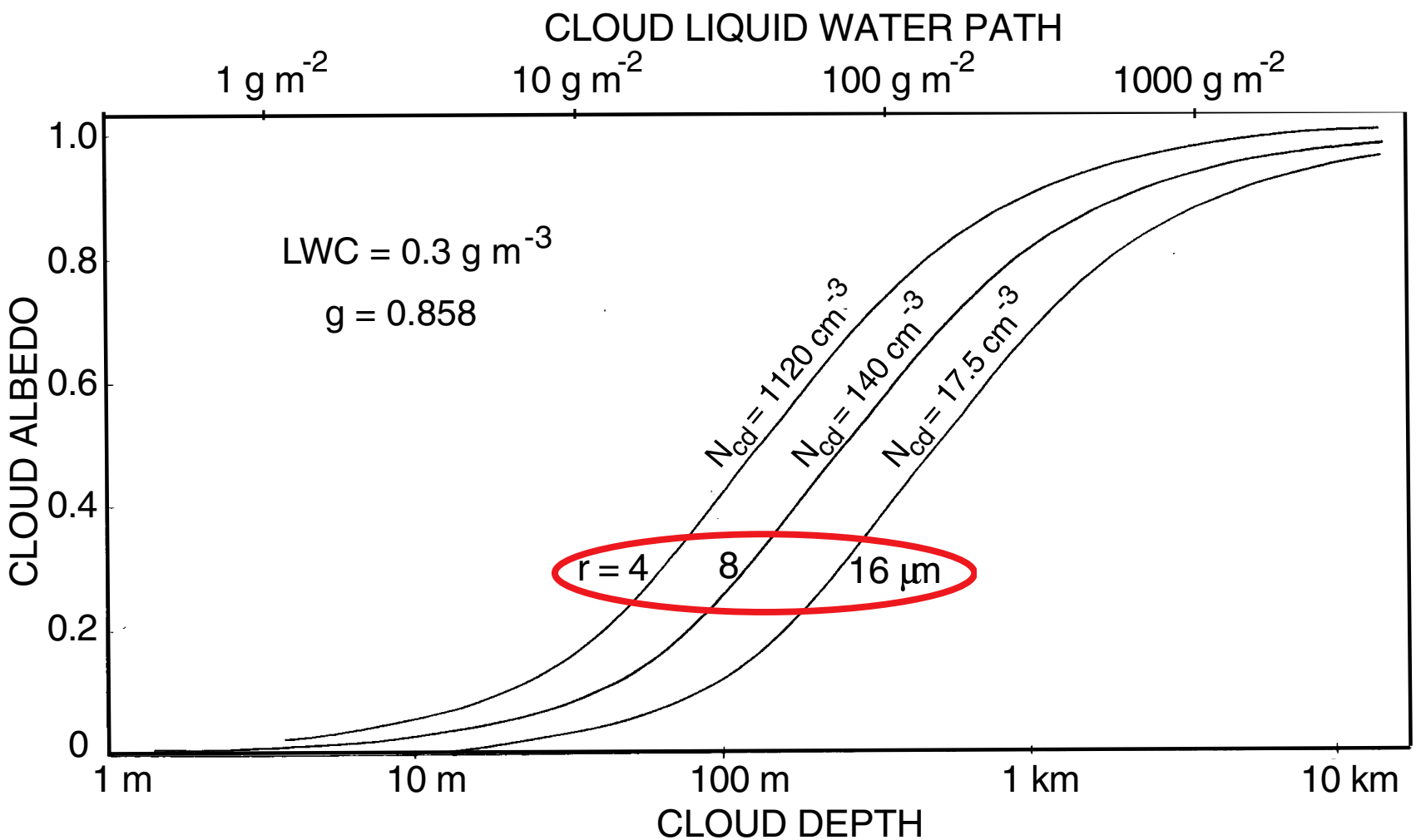


# THE TWOMEY EFFECT

# PHYSICAL BASIS AND SENSITIVITY

# DEPENDENCE OF CLOUD ALBEDO ON CLOUD DEPTH

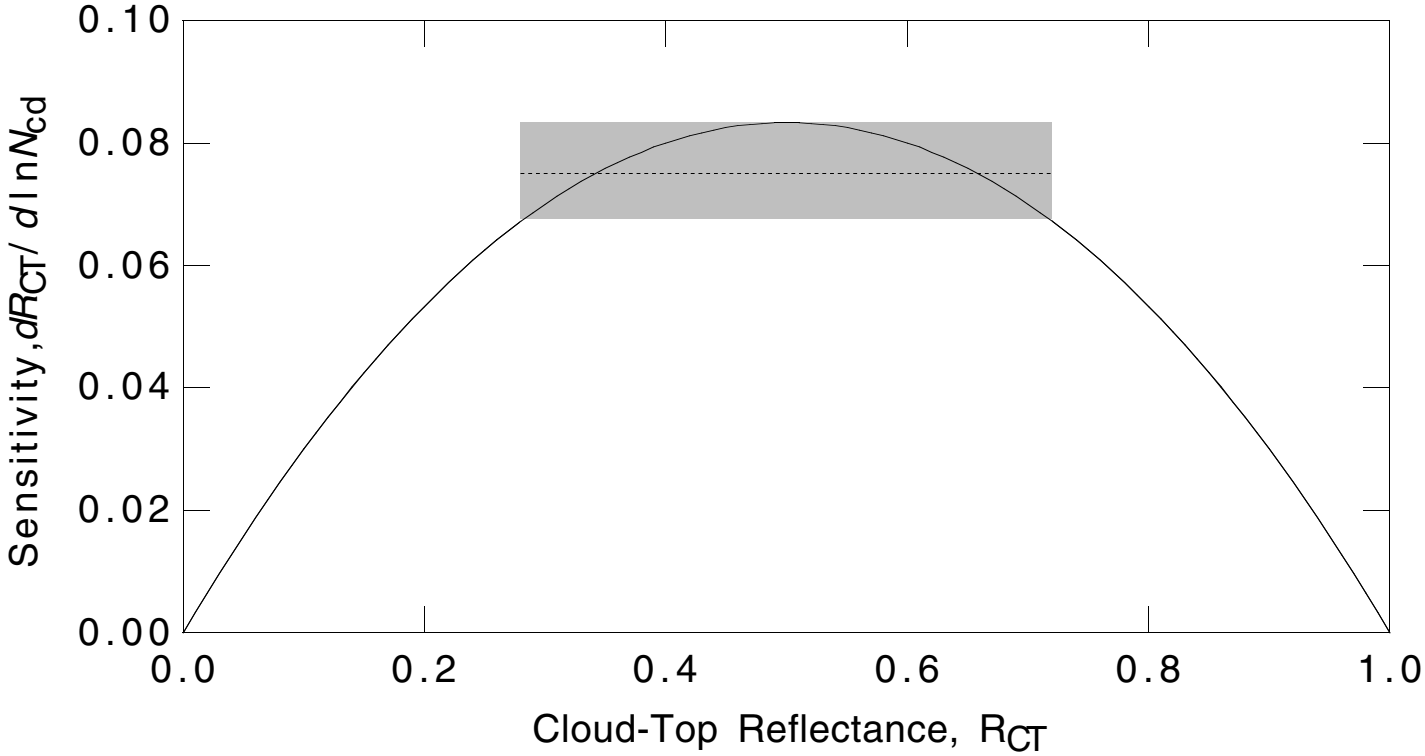
## Influence of Cloud Drop Radius and Concentration




Twomey, *Atmospheric Aerosols*, 1977

*For a given liquid water path, cloud albedo is highly sensitive to cloud drop number concentration or radius.*

# TWOMEY SENITIVITY
## Dependence on cloud-top reflectance

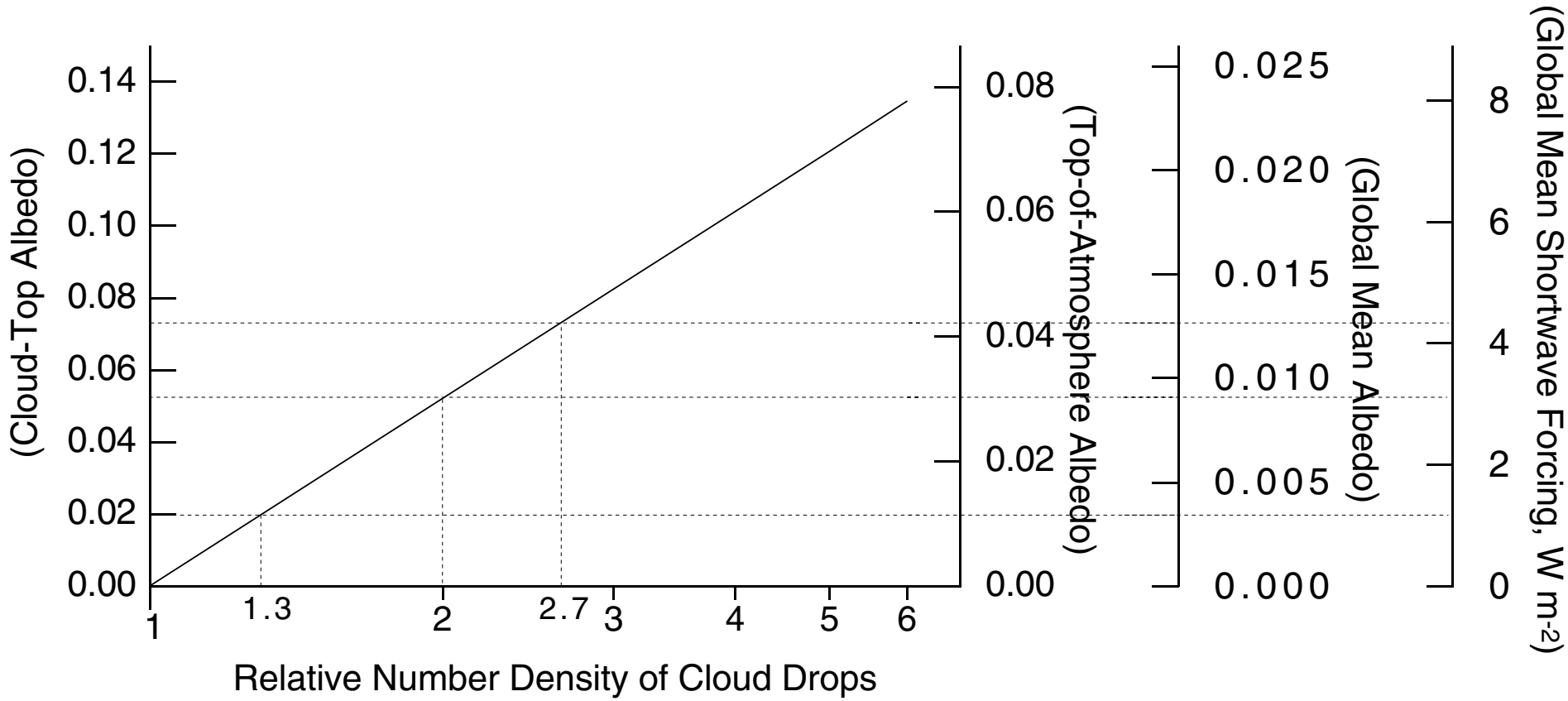



*Modified from Charlson et al., 1992*

Sensitivity is greatest for clouds of intermediate optical depth.

For $dR_{\mathrm{CT}} / d\ln N_{\mathrm{cd}} = 0.08 = 0.08$, a 10% increase in $N_{\mathrm{cd}}$ increases cloud-top reflectivity by 0.008.

# SENSITIVITY OF ALBEDO AND FORCING TO CLOUD DROP CONCENTRATION




*Schwartz and Slingo (1996)*

*Indirect forcing is highly sensitive to small perturbations in cloud drop concentration.*

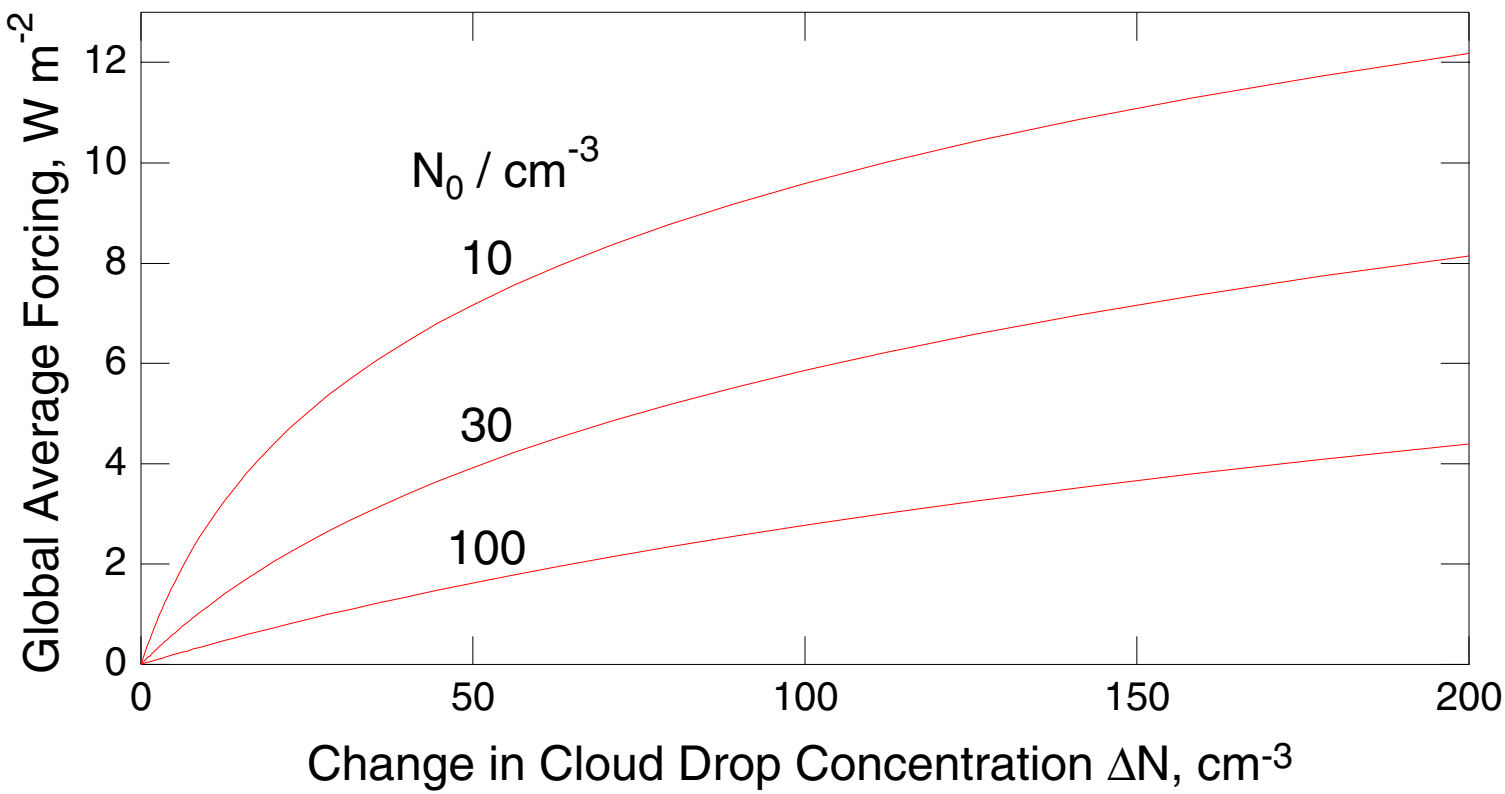
*A 30% increase in cloud drop concentration results in a forcing of ~1 W m$^{-2}$.*

# INDIRECT (TWOMEY) FORCING

Dependence on incremental cloud drop concentration $\Delta N$ and

Sensitivity to initial cloud drop concentration $N_0$

$$F\,/\,(\mathrm{W\ m^{-2}}) \approx 4\ln\left(\frac{N_0+\Delta N}{N_0}\right) = 4\ln\left(1+\frac{\Delta N}{N_0}\right)$$

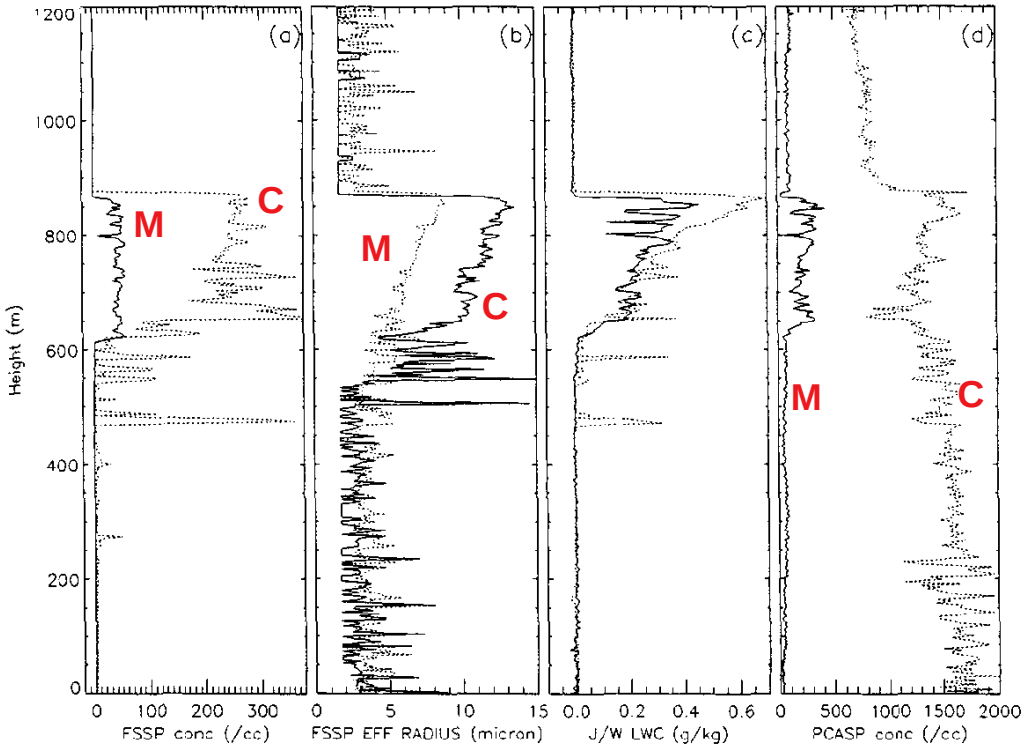


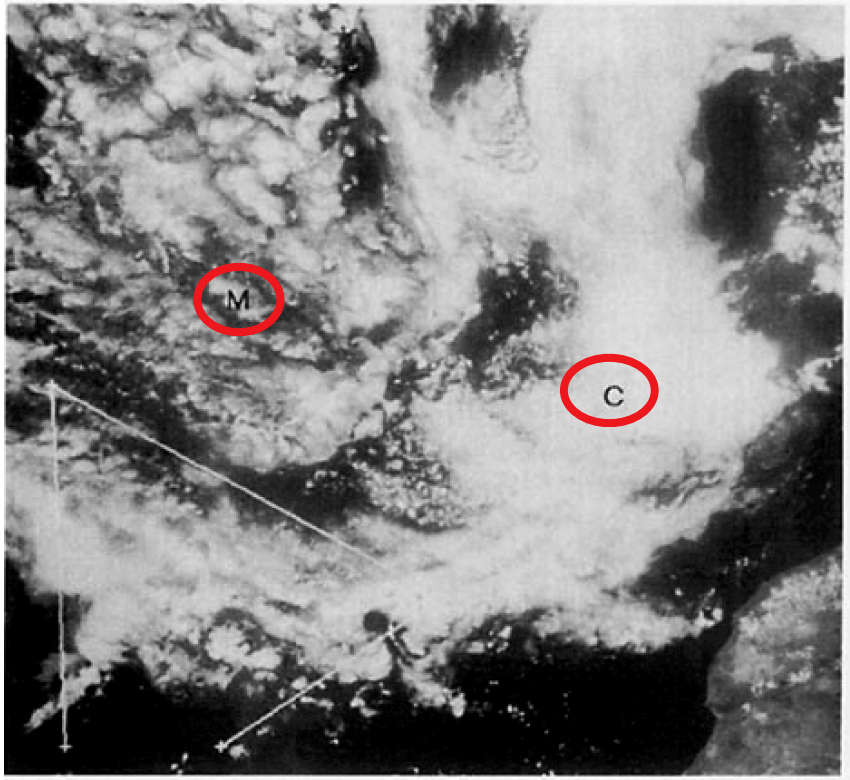

*Aerosol indirect forcing is highly sensitive to background CCN concentration.*

# RELATION BETWEEN AEROSOL CONCENTRATIONS AND CLOUD DROP CONCENTRATIONS

# CLOUD MICROPHYSICAL PROPERTIES AND SATELLITE VISIBLE RADIANCE

## ASTEX, Northeast Atlantic, June, 1992

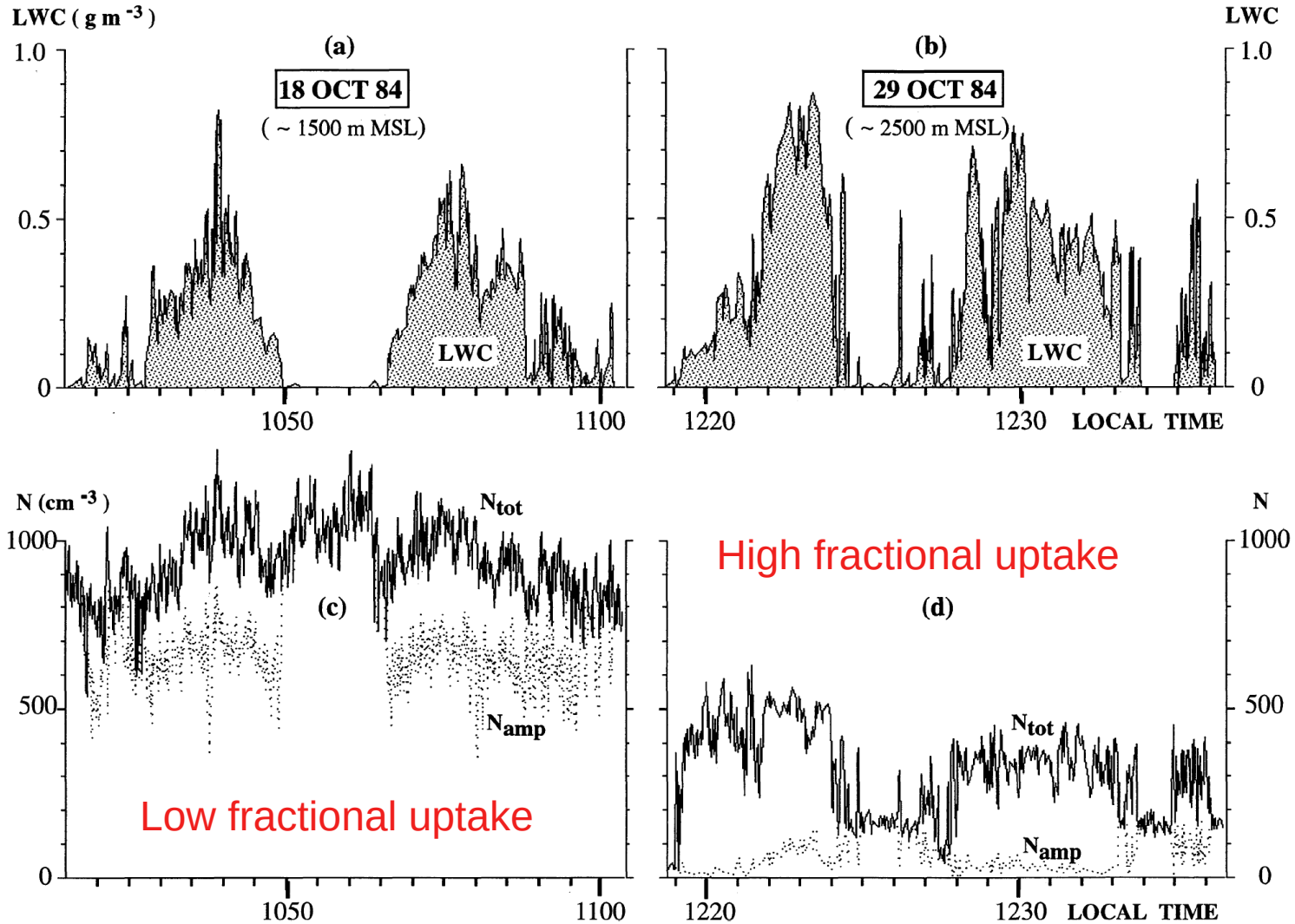



*Albrecht et al., BAMS, 1995*

# FRACTIONAL UPTAKE OF ACCUMULATION MODE PARTICLES INTO CLOUDWATER

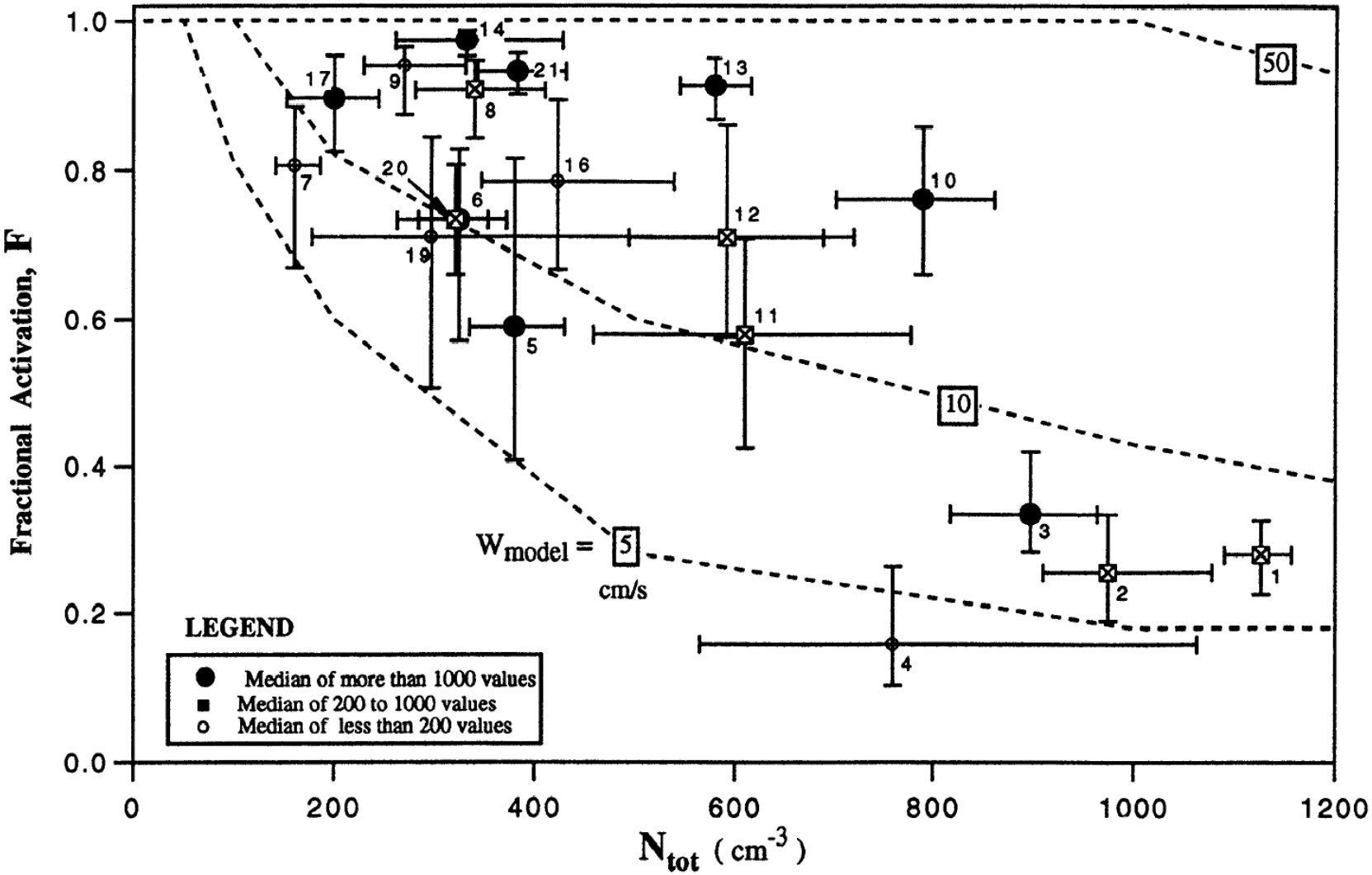
## Aircraft measurements of ASASP (0.17 - 2 μm diameter) and FSSP (2 - 35 μm diameter) particles




*Gillani et al., JGR, 1995*

# FRACTION OF ACTIVATED AEROSOL PARTICLES

## Dependence on particle number concentration

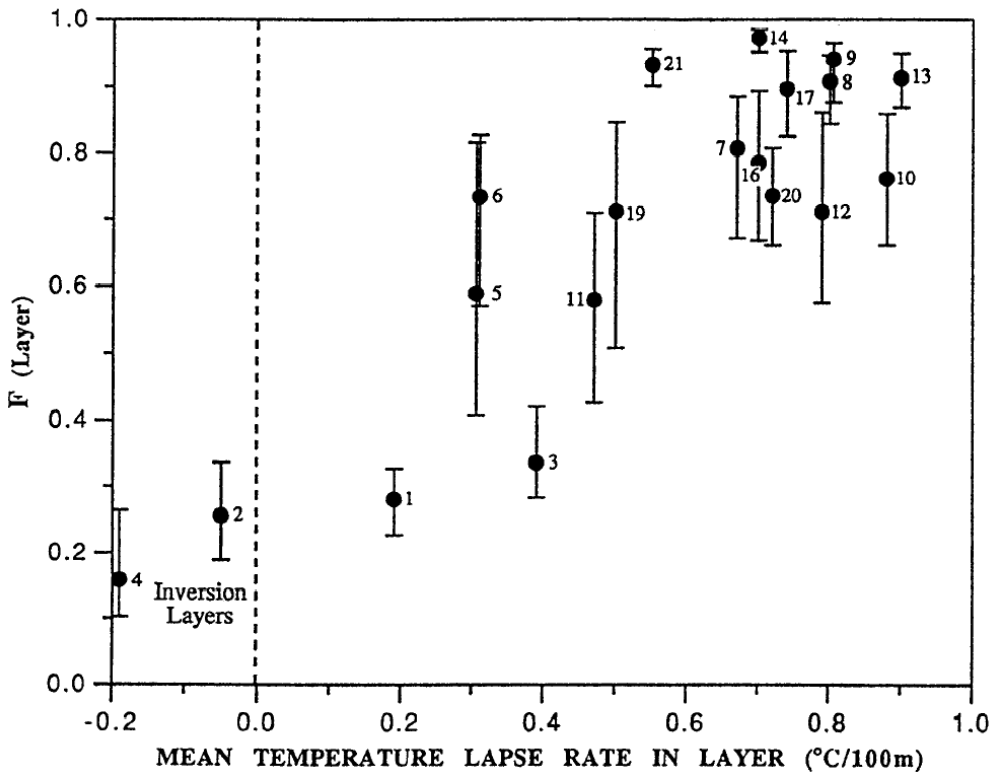


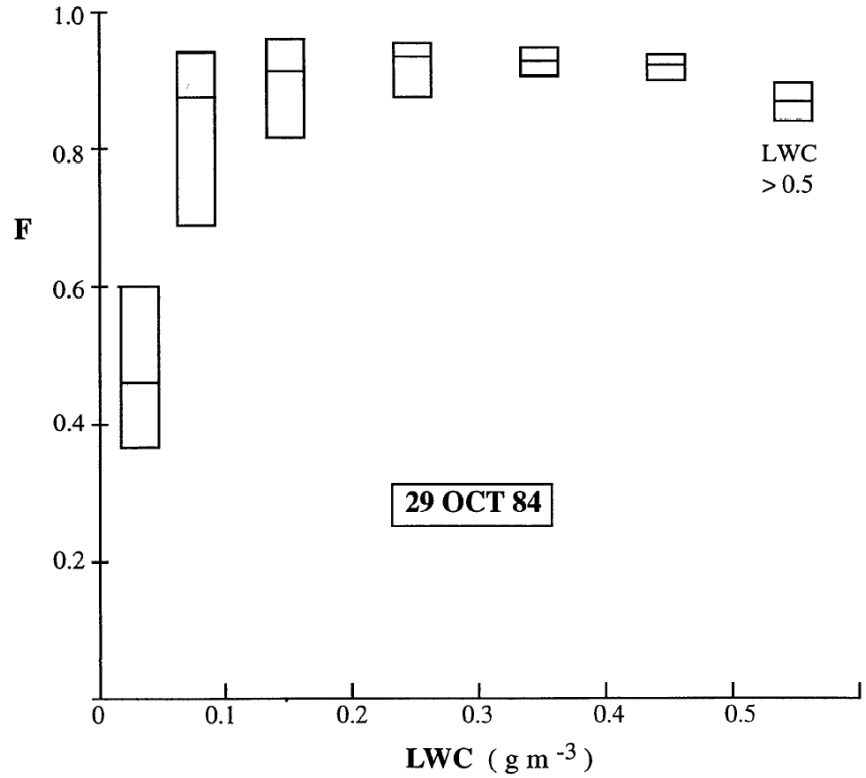

*Gillani et al., JGR, 1995*

Uptake decreases with increasing number concentration of aerosol particles

# FRACTION OF ACTIVATED AEROSOL PARTICLES

## Dependence on Lapse Rate and Liquid Water Content




*Gillani et al., JGR, 1995*

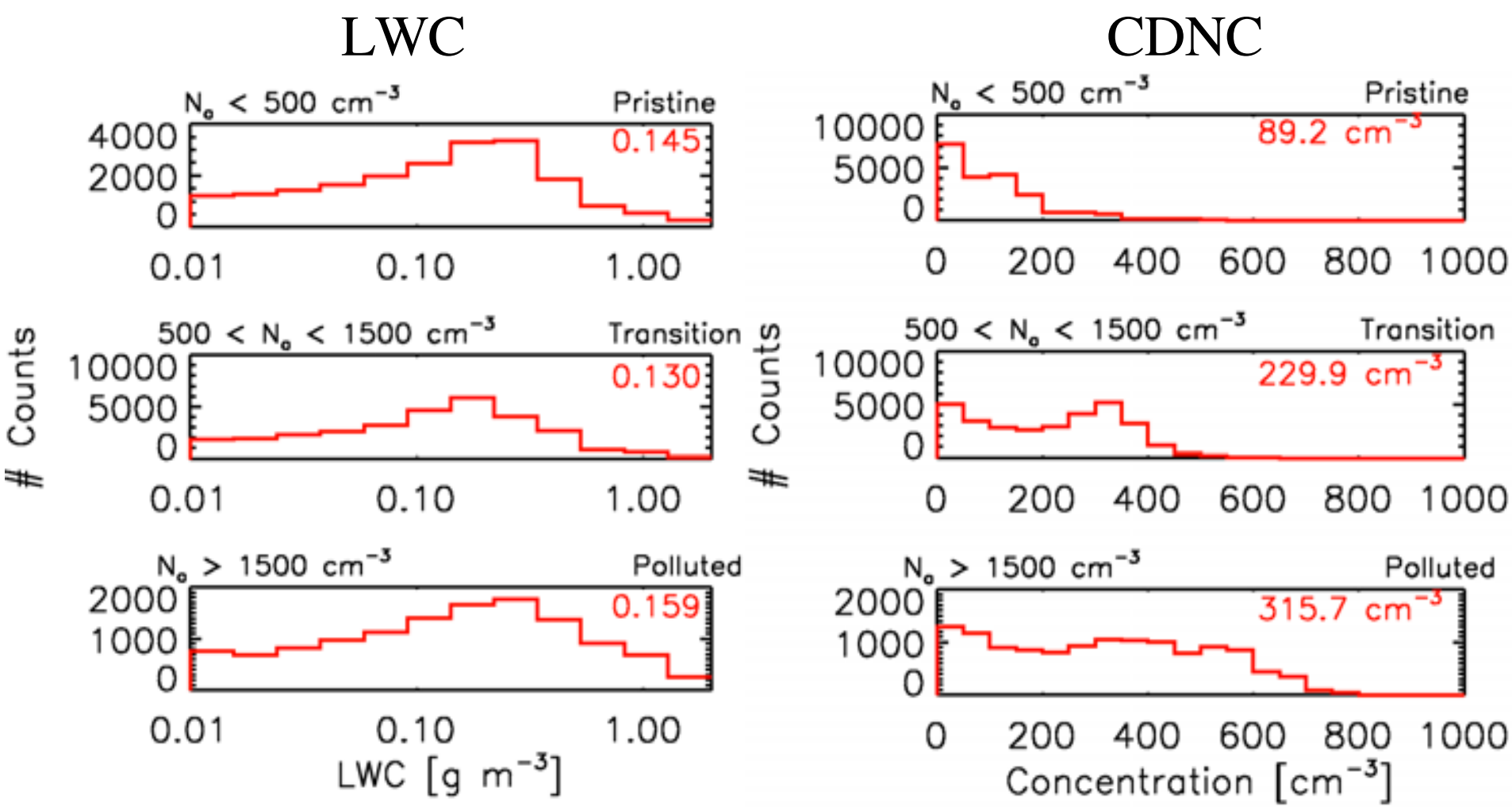
Uptake increases with increasing instability

Uptake increases with increasing liquid water content

# PRISTINE *vs*. POLLUTED CLOUDS IN INDOEX

In-situ aircraft measurements during cross-Equatorial transects

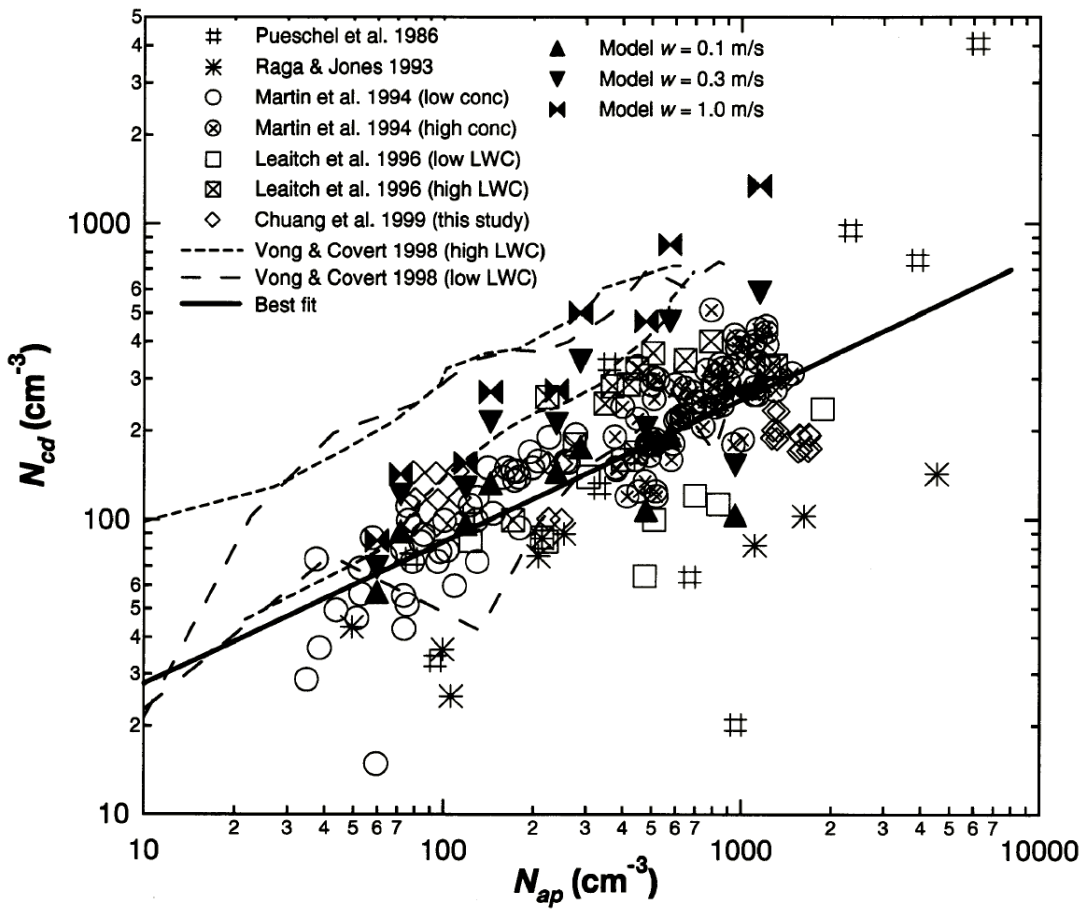
Classified by concentration of aerosol particles




*Heymsfield and McFarquhar, JGR, 2001*

# CLOUD DROP NUMBER CONCENTRATION

## Dependence on accumulation-mode aerosol particle concentration

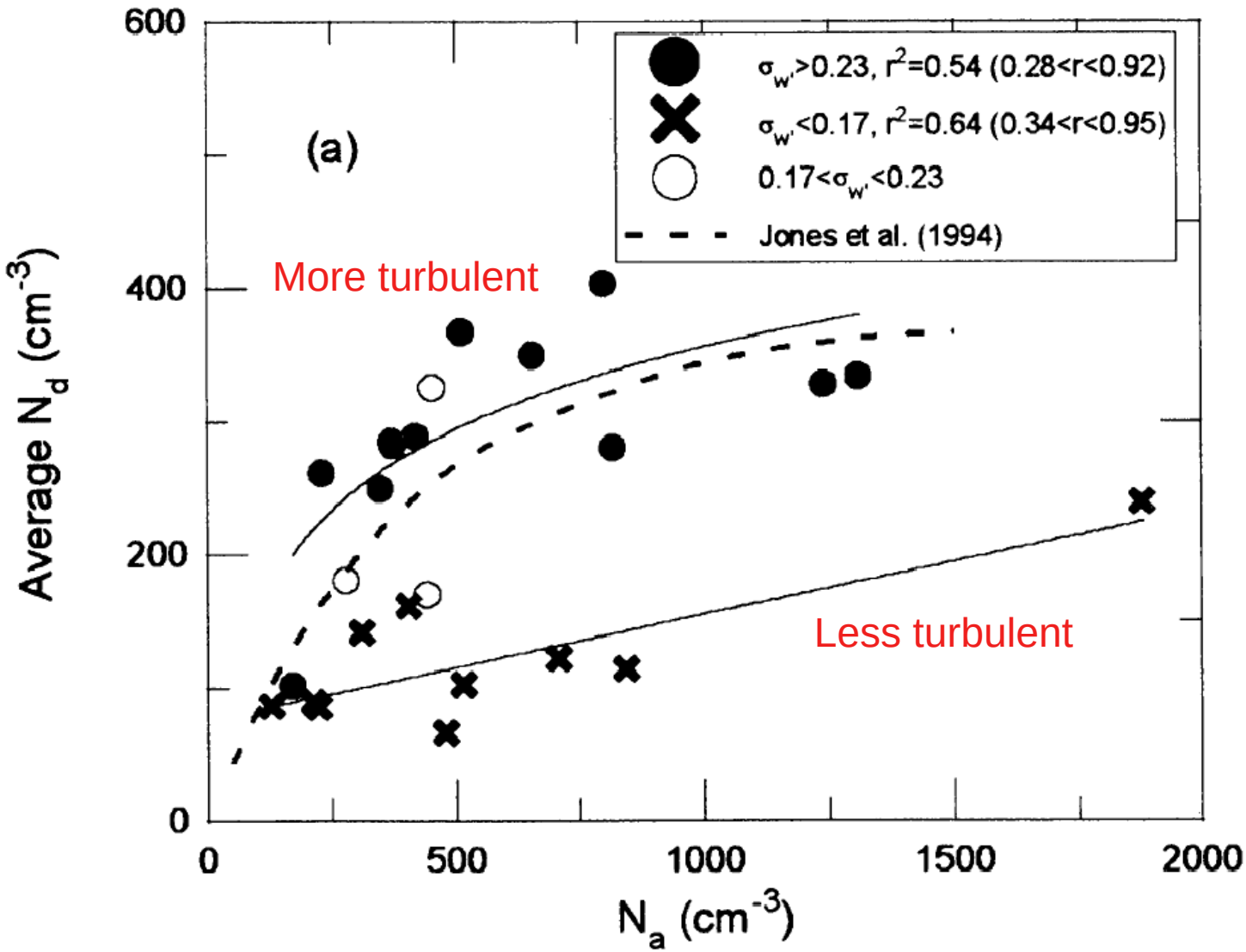



*Chuang et al., 2000*

$N_{cd}$ increases with increasing $N_{ap}$, but scatter at any $N_{ap}$ is comparable to increase in $N_{cd}$ over range of $N_{ap}$.

# CLOUD DROP NUMBER CONCENTRATION

Dependence on below-cloud aerosol particle concentration
Stratified by turbulent intensity

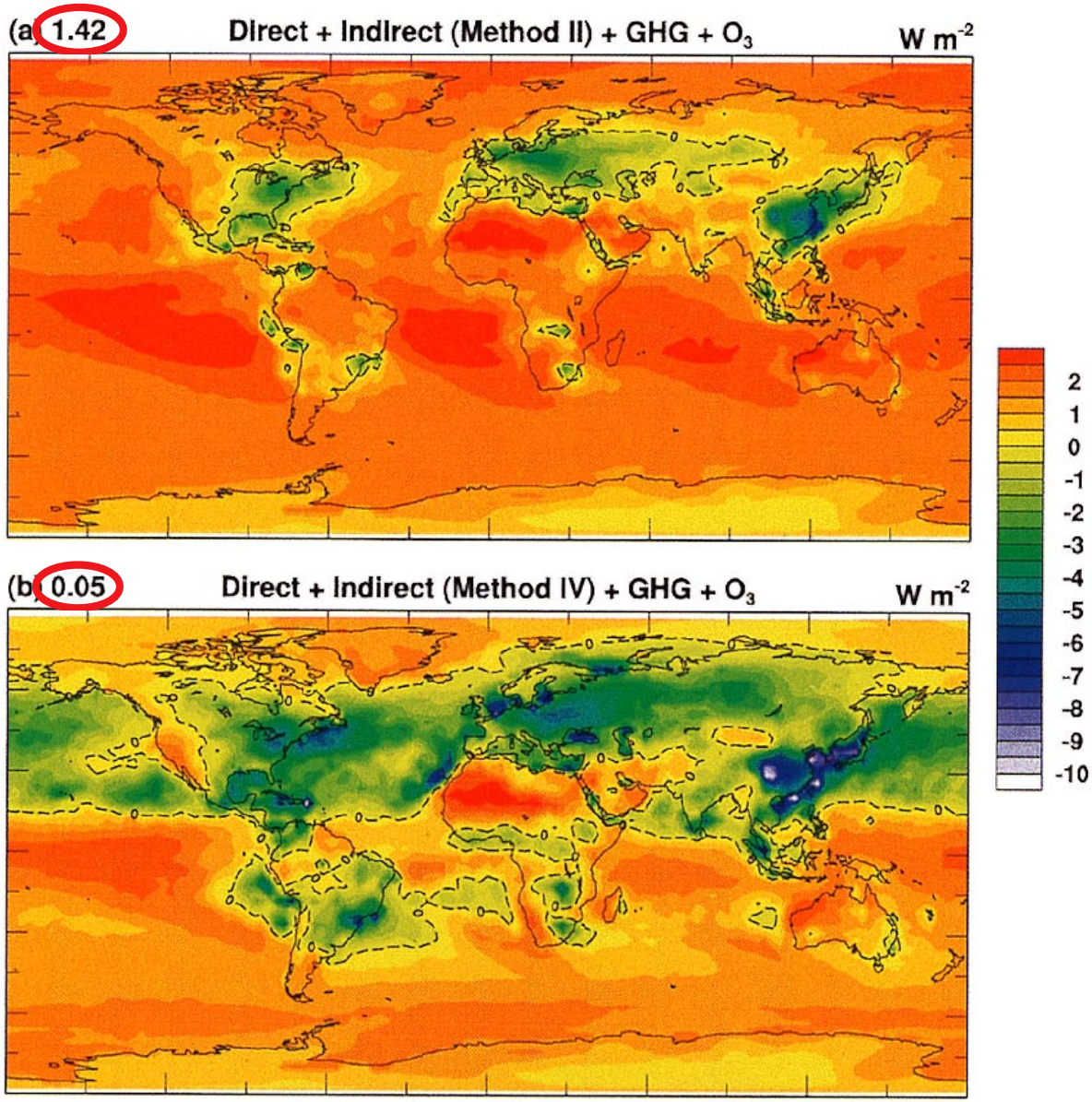



*Leaitch et al., JGR, 1996*

# SHORTWAVE FORCING, ANNUAL AVERAGE

GHG's + $O_3$ + 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.*

# ESTIMATES OF AEROSOL INDIRECT FORCING

## Global-annual average; from IPCC (2001)

***Table 6.6:*** *The global mean annual average aerosol indirect radiative forcing from different global studies. Letters P (prescribed) and C (computed) refer to off-line and on-line sulphate aerosol calculations, respectively. CCN and CDN stand for cloud condensation nuclei and cloud droplet number, respectively. In studies indicated by an asterisk, the estimate in flux change due to the indirect effect of aerosols was computed as the difference in top of atmosphere fluxes between two distinct simulations and therefore does not represent a forcing in the strict sense (see text). When several simulations are performed in the same study, base indicates the baseline calculation, while the range of estimates is given in parenthesis.*

| Reference | Aerosol Type | Forcing estimate ($Wm^{-2}$) | | | | Remarks |
|---|---|---|---|---|---|---|
| | | First effect | Second effect | Both effects | | |
| Boucher and Rodhe (1994)* | Sulphate | | | -0.65 to -1.35 | P | Uses 3 relationships between sulphate mass and CCN/CDN concentrations. |
| Chuang *et al.* (1994) | Sulphate | -0.47 | | | C | Includes a parametrization of cloud nucleation processes. |
| Jones *et al.* (1994) | Sulphate | -1.3 | | | P | Uses a relationship between aerosol and droplet number concentrations. |
| Boucher and Lohmann (1995) | Sulphate | -0.5 to -1.4 | | | P | LMD GCM — Uses 4 different relationships between sulphate mass and CCN/CDN concentrations (A, B, C, and D). |
| Boucher and Lohmann (1995) | Sulphate | -0.45 to -1.5 | | | P | ECHAM |
| Jones and Slingo (1996) | Sulphate | -0.3 to -1.5 | | | P | Uses 2 different sulphate distributions. Follows Jones *et al.* (1994), Hegg (1994), Boucher and Lohmann (1995) D . |
| Kogan *et al.* (1996) Kogan *et al.* (1997) | Sulphate | -1.1 | | | | Uses a cloud climatology rather than GCM-simulated clouds. |
| Chuang *et al.* (1997) | Sulphate | -0.4 to -1.6 | | | C | Includes a parametrization of cloud nucleation processes. Uses a mixture of pre-existing aerosols. |
| Feichter *et al.* (1997) | Sulphate | -0.76 | | | C | Uses Boucher and Lohmann (1995) A relationship. |

*cont'd*

# ESTIMATES OF AEROSOL INDIRECT FORCING

## Global-annual average; from IPCC (2001), *cont'd*

| Reference | Aerosol Type | Forcing estimate ($Wm^{-2}$) First effect | Second effect | Both effects | | Remarks |
|---|---|---|---|---|---|---|
| Jones and Slingo (1997) | Sulphate | -0.55 to -1.50 | | | P | Uses 2 different versions of the Hadley Centre model. |
| Lohmann and Feichter (1997)* | Sulphate | -1 | | -1.4 to -4.8 | C | Uses Boucher and Lohmann (1995) A relationship. |
| Rotstayn (1999)* | Sulphate | base -1.2 (-1.1 to -1.7) | base -1.0 (-0.4 to -1.0) | base -2.1 (-1.6 to -3.2) | P | Includes a (small) long-wave radiative forcing. |
| Jones *et al.* (1999)* [a] | Sulphate | -0.91 | base -0.66 | 1.18 | C | Includes a (small) long-wave radiative forcing. The two effects add non-linearly. |
| Kiehl *et al.* (2000) | Sulphate | -0.40 to -1.78 | | | C | |
| Ghan *et al.* (2001a)* | Sulphate | ~50% for base | ~50% for base | base -1.7 (-1.6 to -3.2) | C | Includes a parametrization of cloud nucleation. Predicted aerosol size distribution. |
| Lohmann *et al.* (2000)* | Sulphate | | | base -0.4 (0 to -0.4) | C | Includes a parametrization of cloud nucleation processes. |
| | Carb. | | | base -0.9 (-0.9 to -1.3) | C | |
| | Sulphate and Carb. | 40% for base | 60% for base | base -1.1 (-1.1 to -1.9) | C | |
| Chuang *et al.* (2000b) | Sulphate | -0.30 | | | C | Includes a parametrization of cloud nucleation processes. Includes the effect of BC absorption in clouds. |
| | Carb. | base -1.51 (-1.27 to -1.67) | | | C | |
| | Sulphate and Carb. | -1.85 | | | C | |

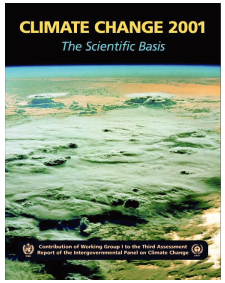
Estimates vary enormously within a given study (sensitivity to parameters) and between studies.

# 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 ($\mu g\ m^{-3}$) | 1.5 | 0.85 to 2.15 | **2.5** |
| Cloud layer thickness (m) | 200 | 100 to 300 | 3.0 |
| Perturbed sulfate concentration ($\mu g\ m^{-3}$) | 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*

The greatest uncertainties are in aerosol and cloud microphysics properties, such as dependence of cloud drop concentration on ***aerosol composition, loading, and microphysical properties.***




***These uncertainties are not well quantified.***

# ISSUES IN DETERMINING AEROSOL INDIRECT FORCING

1. Enhancement in ***aerosol particle concentration*** (and size, composition, etc.) between preindustrial and present, as function of location.
2. Relation between aerosol particle concentration (and size, composition, etc.) and ***cloud droplet concentration***.
3. Relation between cloud drop concentration and ***cloud reflectance***.
4. Aerosol influences on ***LWP, cloud lifetime***, etc., in addition to reflectance.

***These requirements can be met only by models . . .***

***Based on improved understanding and evaluated by comparison with observation:***

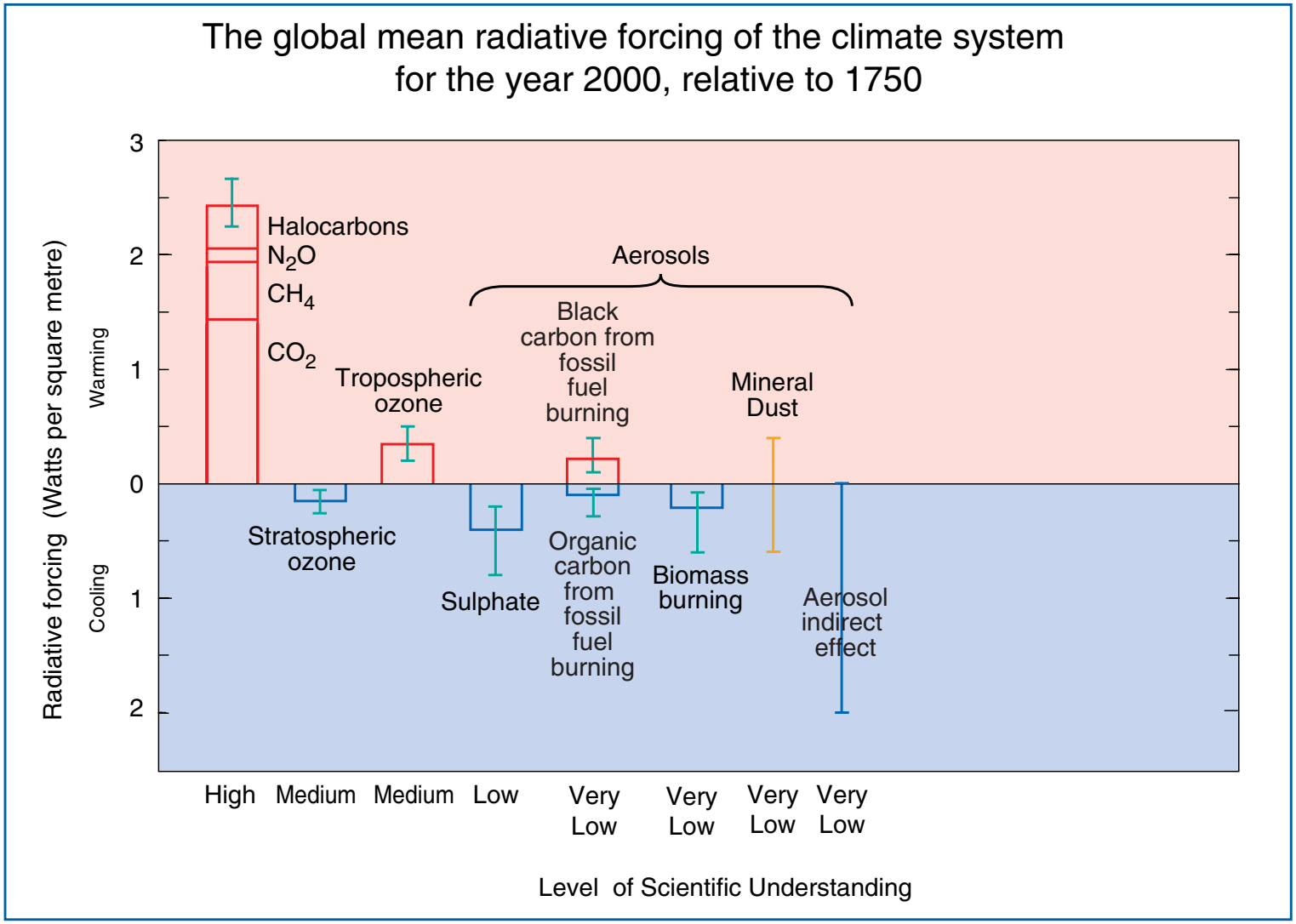
***Emissions***

***Chemical transport and transformation***

***Cloud drop activation and microphysics***

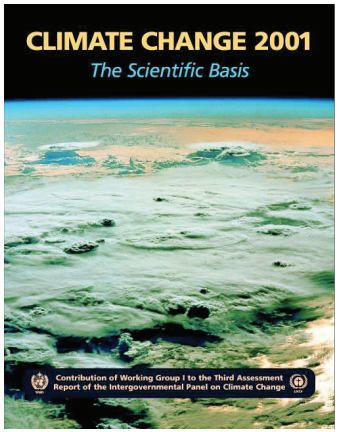
***Radiation transfer***

# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
# IPCC (2001)
## GHG's and aerosol direct and indirect effects

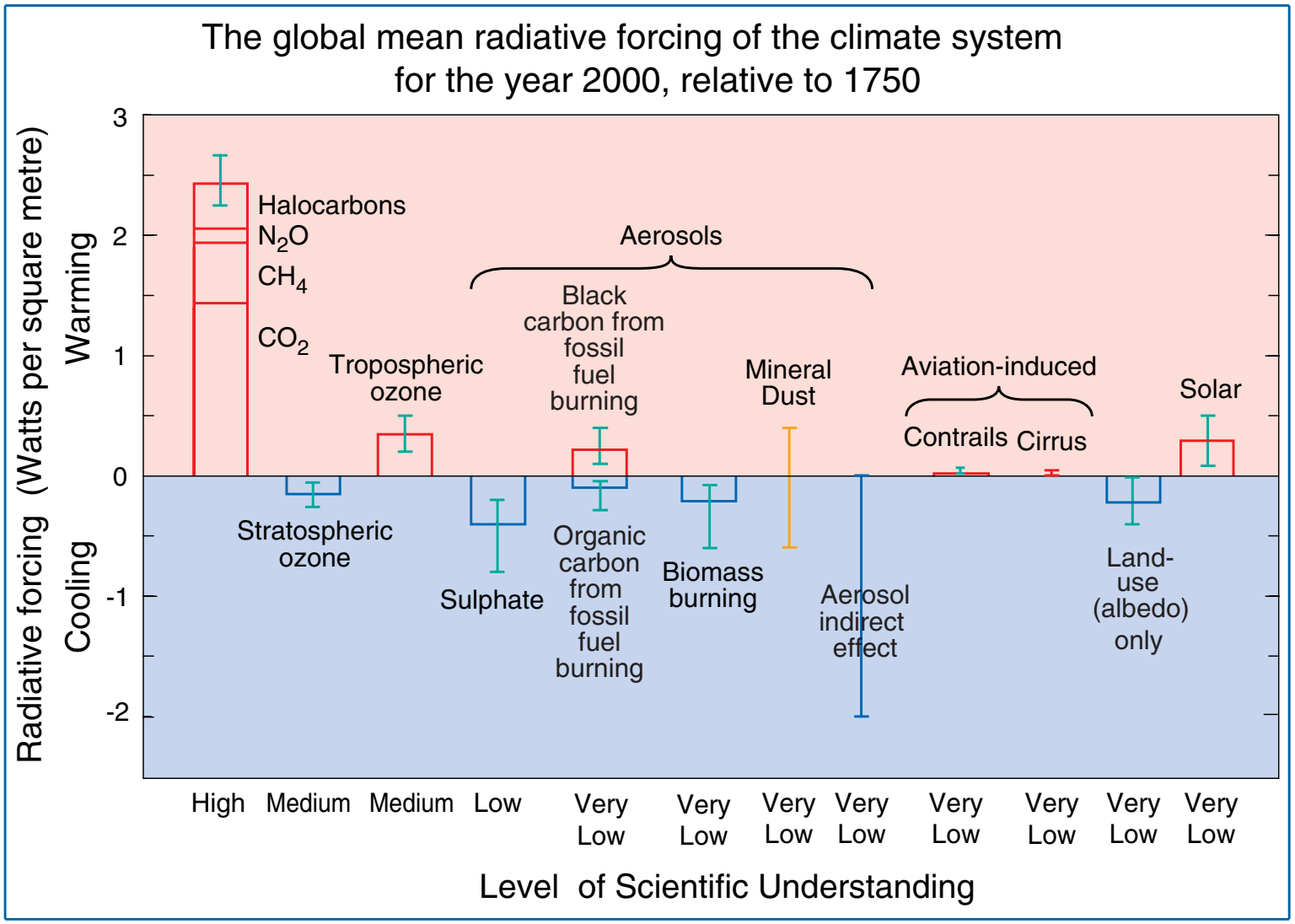



Summary for Policymakers

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

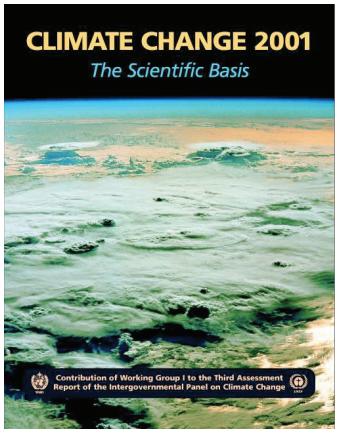


# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
# IPCC (2001)




Summary for Policymakers

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



# WHY SO LARGE UNCERTAINTY IN AEROSOL FORCING?

- ***Uncertainties in knowledge of atmospheric composition***

  ***Mass loading and chemical and microphysical properties and cloud nucleating properties of anthropogenic aerosols, and geographical distribution.***

  ***At present and as a function of secular time.***

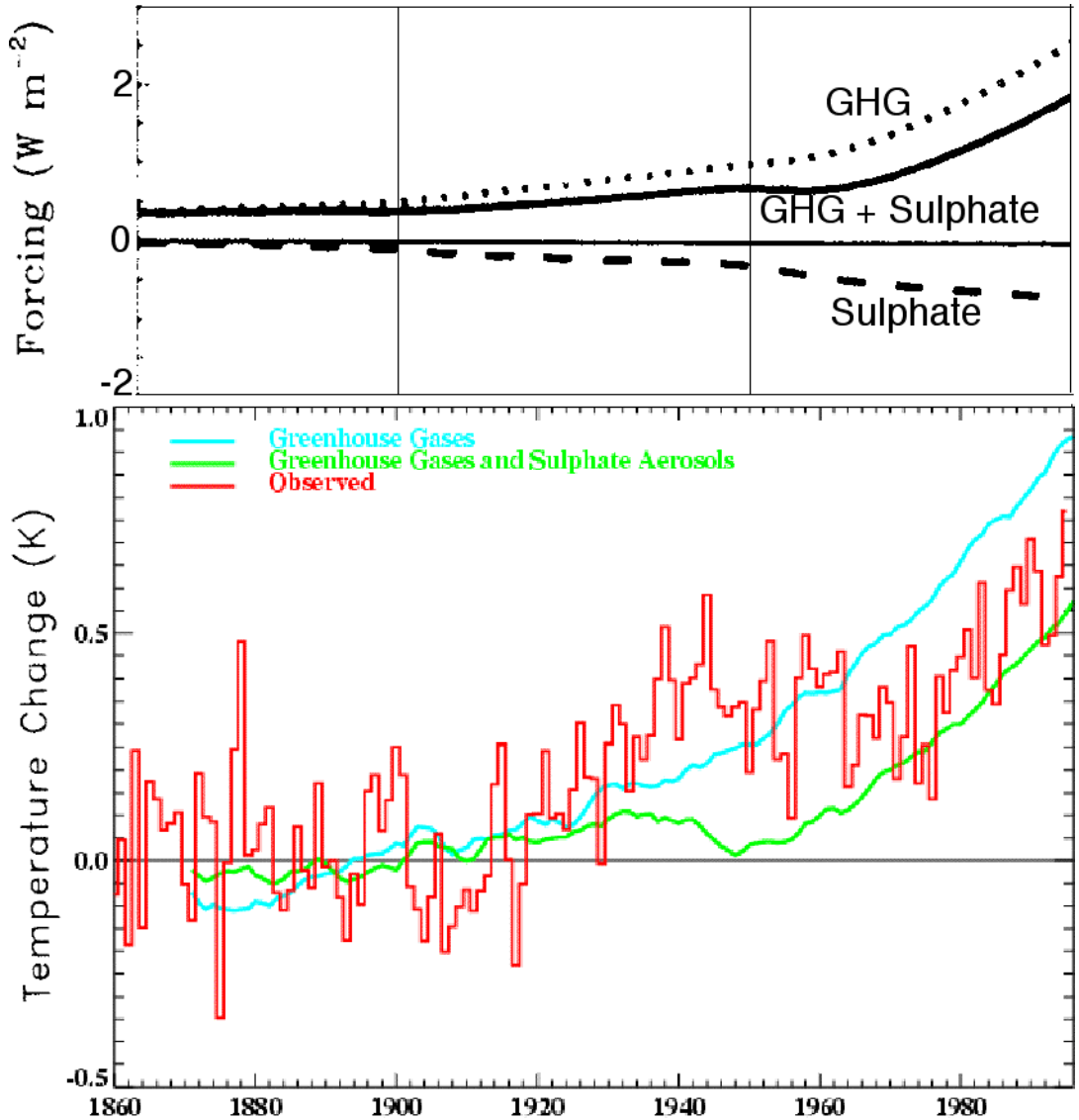
- ***Uncertainties in knowledge of atmospheric physics of aerosols***

  ***Relating direct radiative forcing and cloud modification by aerosols to their loading and their chemical and microphysical properties.***

# REPRESENTING AEROSOL FORCING IN CLIMATE MODELS

# FORCING AND RESPONSE IN THE UK MET OFFICE MODEL (1995)

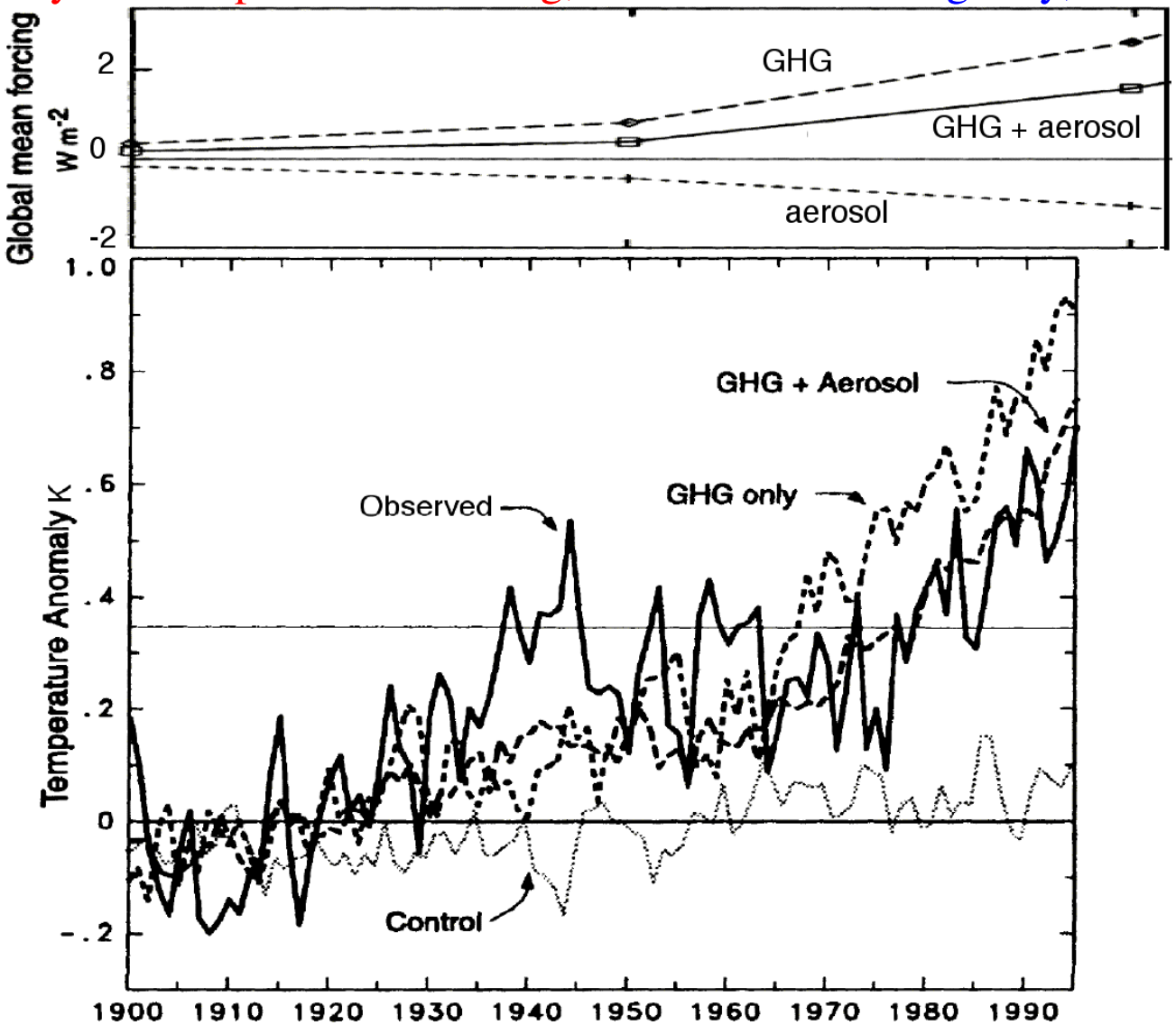
Model sensitivity = 2.5 K per $CO_2$ doubling; sulfate direct forcing only, -0.6 W $m^{-2}$ (1990)




"Inclusion of sulphate aerosol forcing ***improves the simulation*** of global mean temperature over the last few decades." -- *Mitchell, Tett, et al., Nature, 1995*

# FORCING AND RESPONSE IN THE CANADIAN CLIMATE MODEL (2000)

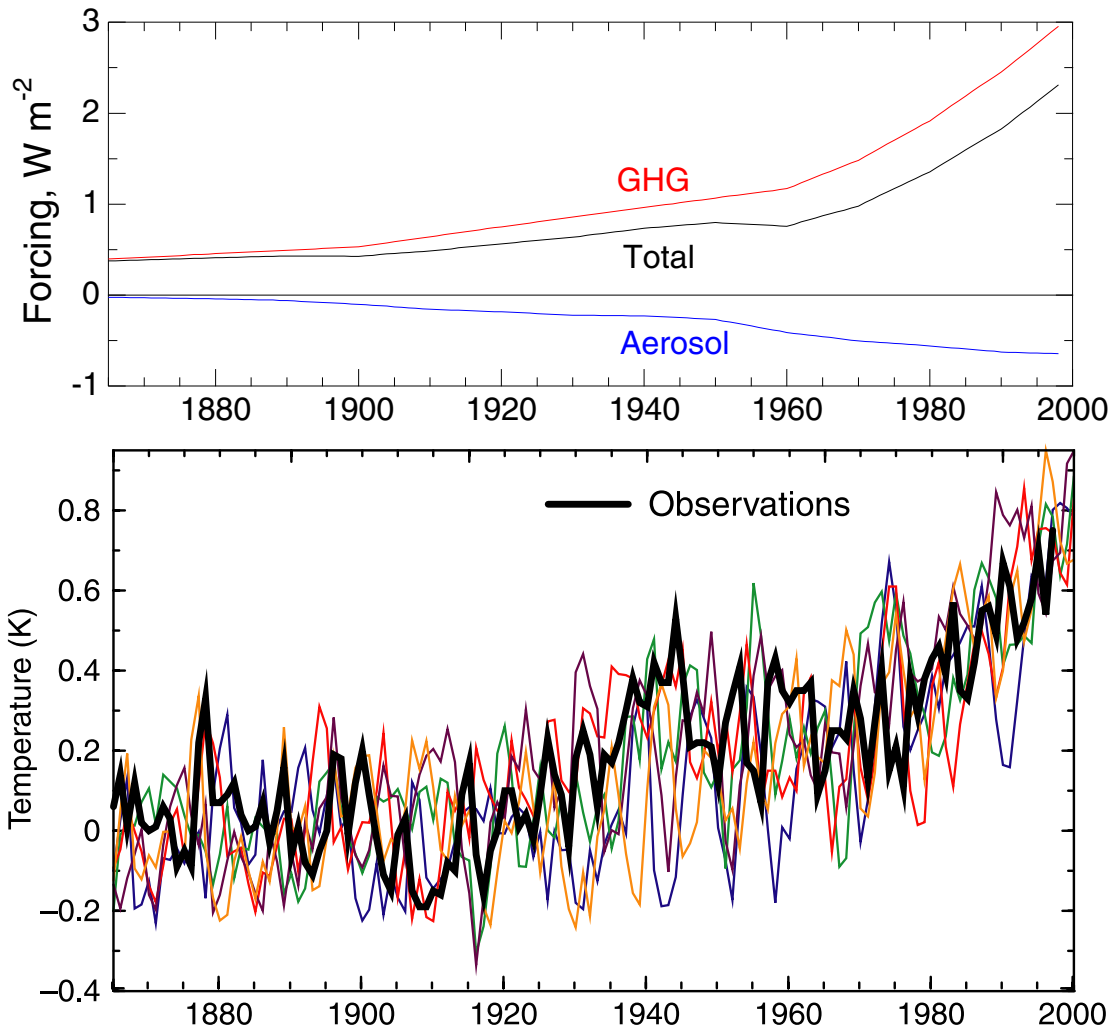
Model sensitivity = 3.5 K per $CO_2$ doubling; sulfate direct forcing only, -1.0 W $m^{-2}$ (1990)




"Observed global mean temperature changes and those simulated for GHG + aerosol forcing show ***reasonable agreement***." -- *Boer, et al., Climate Dynamics, 2000*

# FORCING AND RESPONSE IN THE GFDL MODEL (2000)

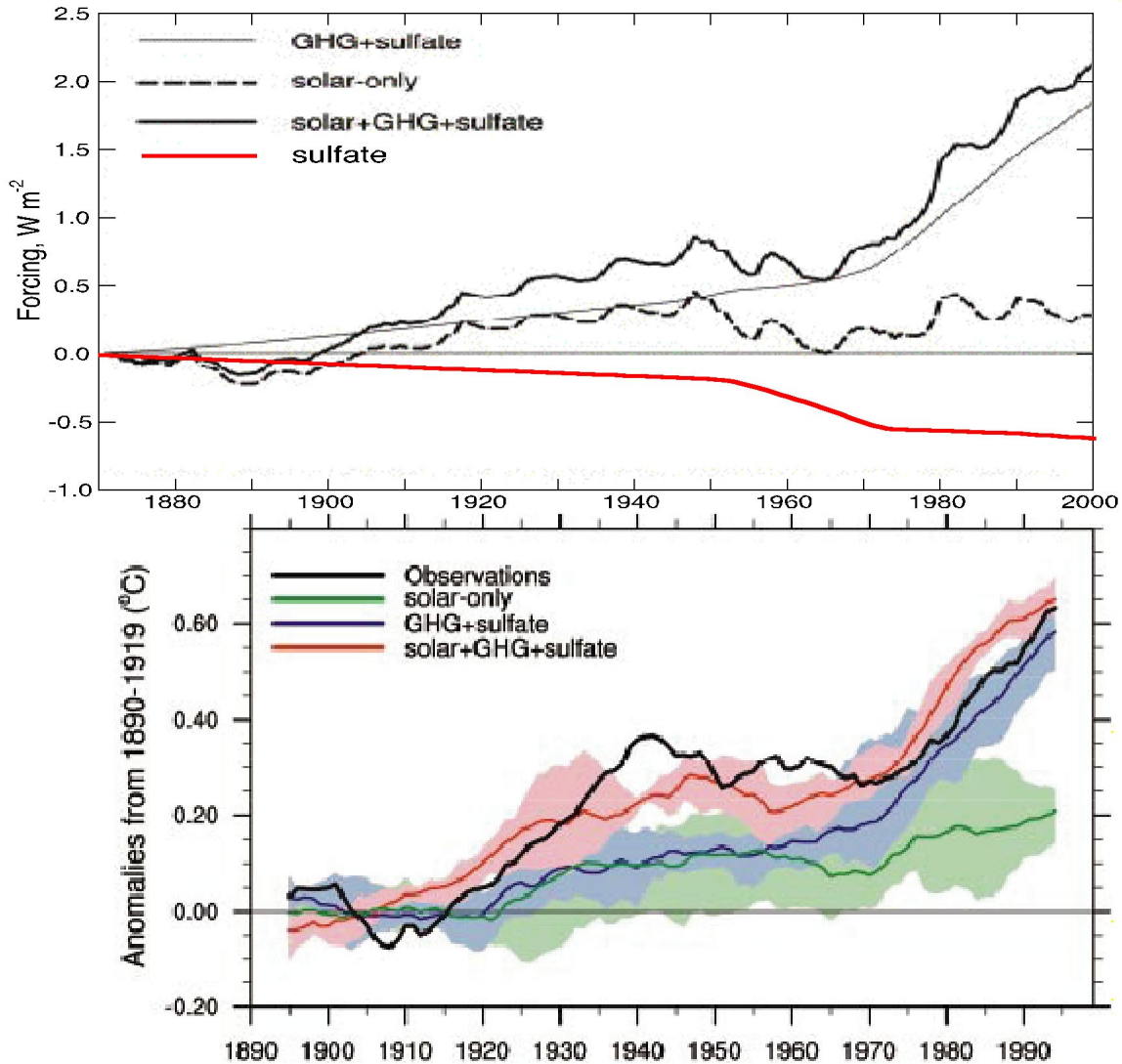
Model sensitivity = 3.4 K per $CO_2$ doubling; sulfate forcing, -0.62 W $m^{-2}$ (1990)




"The surface temperature time series from the five GHG-plus-sulfate integrations show an increase over the last century, which is ***broadly consistent*** with the observations." -- *Delworth & Knutson, Science, 2000*

# FORCING AND RESPONSE IN THE NCAR MODEL (2003)

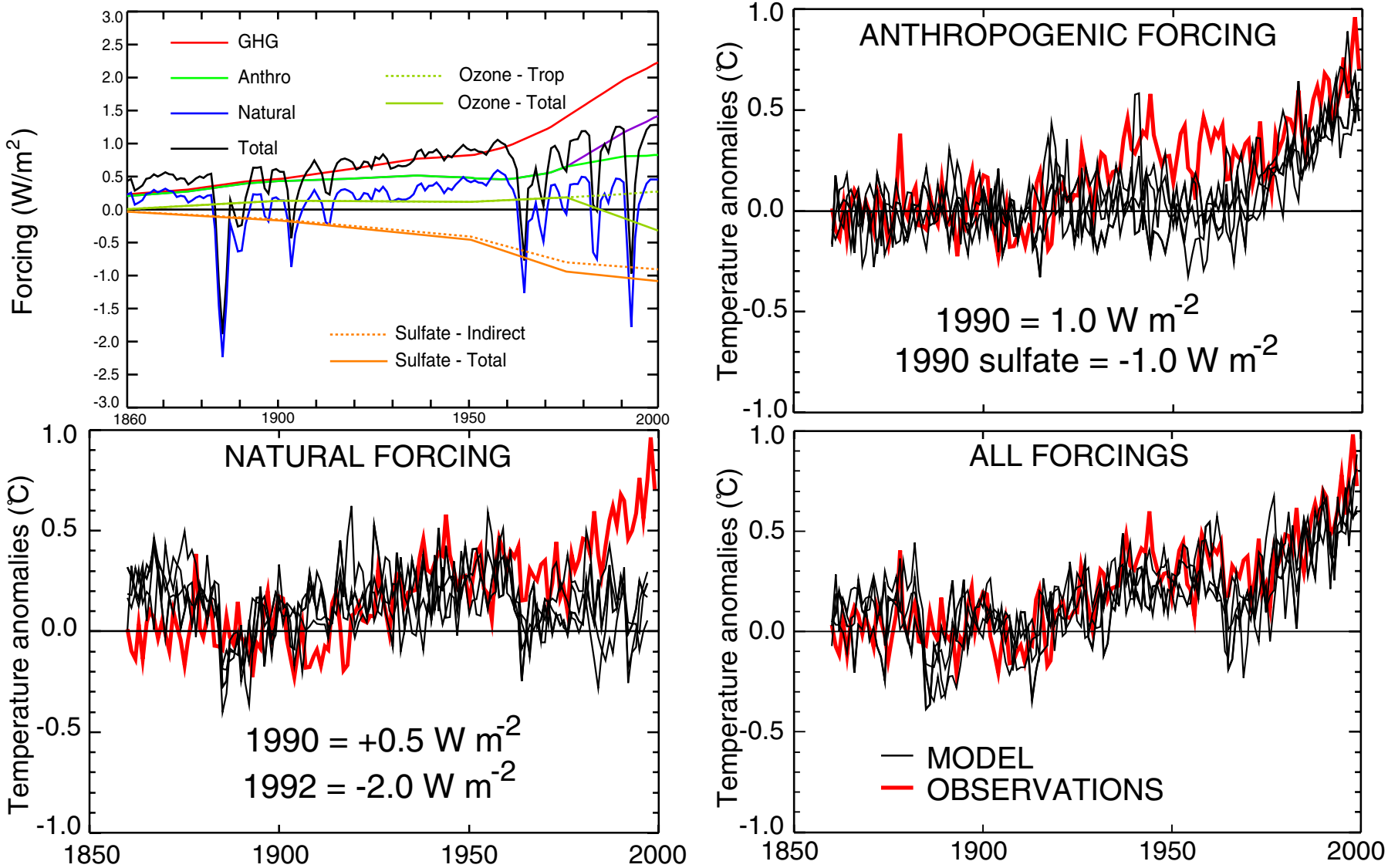
Model sensitivity = 2.18 K per $CO_2$ doubling; sulfate direct forcing only, -0.6 W $m^{-2}$ (1990)




"The time series from GHG + sulfates + solar shows ***reasonable agreement*** with the observations." -- *Meehl, Washington, Wigley et al., J. Climate, 2003.*

# FORCING AND RESPONSE IN THE UK MET OFFICE MODEL (2000)

Model sensitivity = 3.45 K per $CO_2$ doubling; sulfate + indirect forcing, -1.1 W $m^{-2}$ (1990)



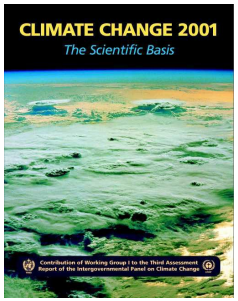

"The ALL ensemble ***captures the main features*** of global mean temperature changes observed since 1860." -- *Stott, Tett, Mitchell, et al., Science, 2000*

# IPCC-2001 STATEMENTS ON DETECTION AND ATTRIBUTION OF CLIMATE CHANGE

“ ***Simulations that include estimates of natural and anthropogenic forcing reproduce the observed large-scale changes in surface temperature over the 20th century.***

“ ***Most model estimates that take into account both greenhouse gases and sulphate aerosols are consistent with observations over this period.***

UNEP




WMO

# IMPLICATIONS OF UNCERTAINTY IN AEROSOL FORCING

Present uncertainty in forcing of climate change, caused largely by uncertainty in forcing by aerosols, ***precludes evaluation of climate models and empirical determination of climate sensitivity***.

Uncertainty in aerosol forcing must be reduced ***at least three-fold*** (~0.5 W $m^{-2}$) for uncertainty in climate sensitivity to be meaningfully reduced and bounded.

Such a reduction in uncertainty in aerosol forcing presents an ***enormous challenge*** to the aerosol research community.

Atlanta, Georgia
AAAR 04

# *Thank You*

Stephen E. Schwartz

BROOKHAVEN
NATIONAL LABORATORY

http://www.ecd.bnl.gov/steve/schwartz.html