AEROSOLS AND CLIMATE CHANGE
CERTAINTIES AND UNCERTAINTIES

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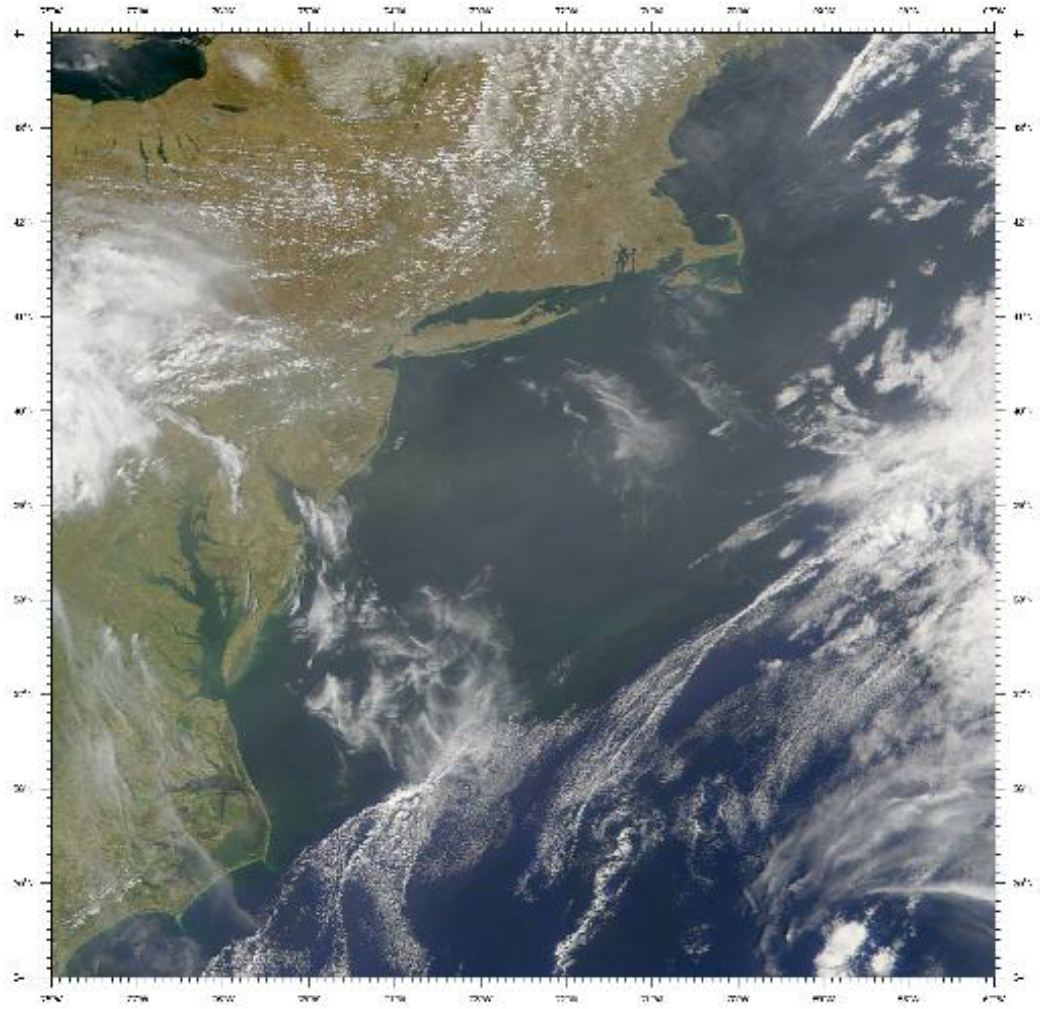
September 4, 2003

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

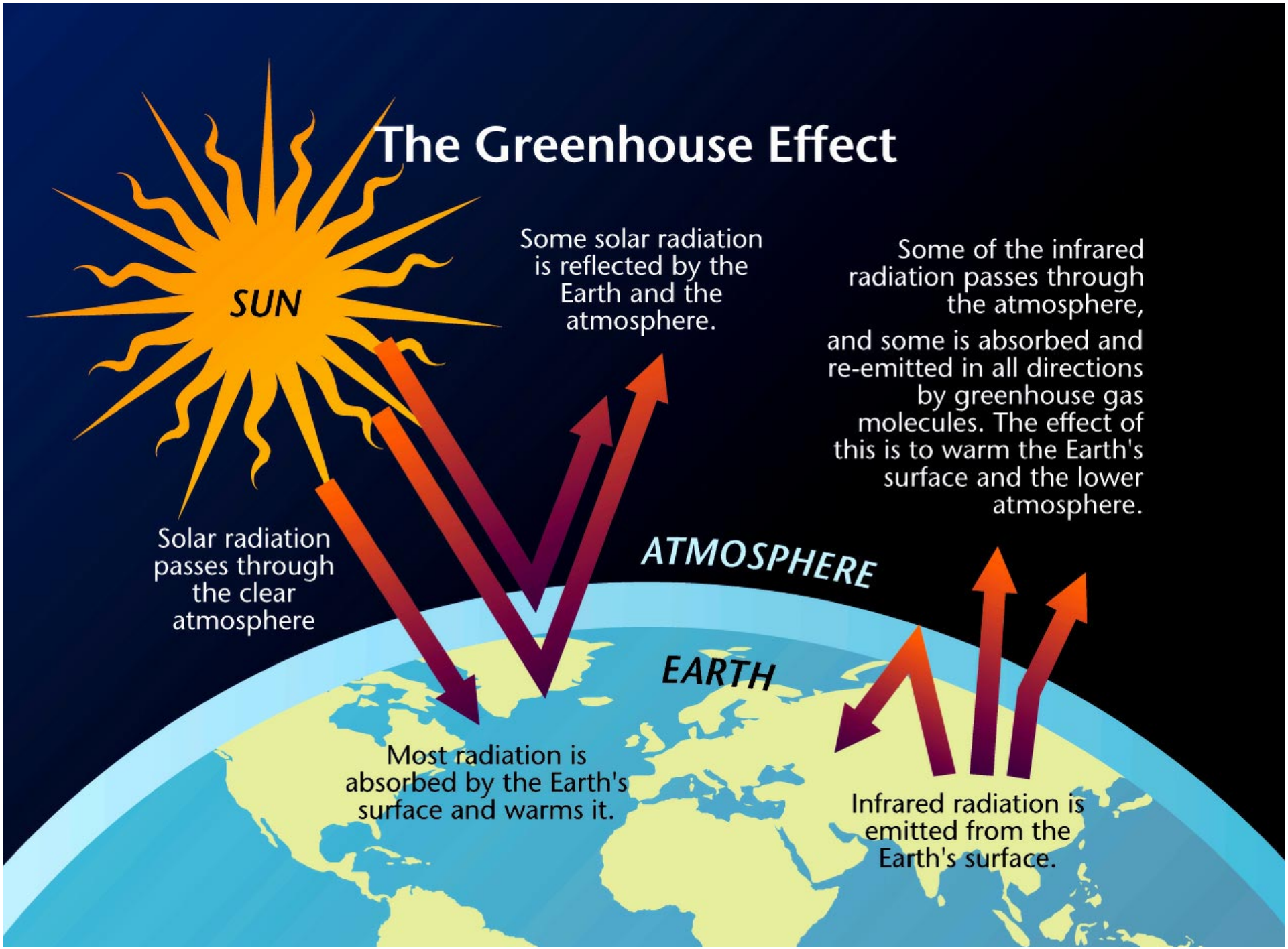
AEROSOL: A suspension of particles in air



2001-04-22-17:28

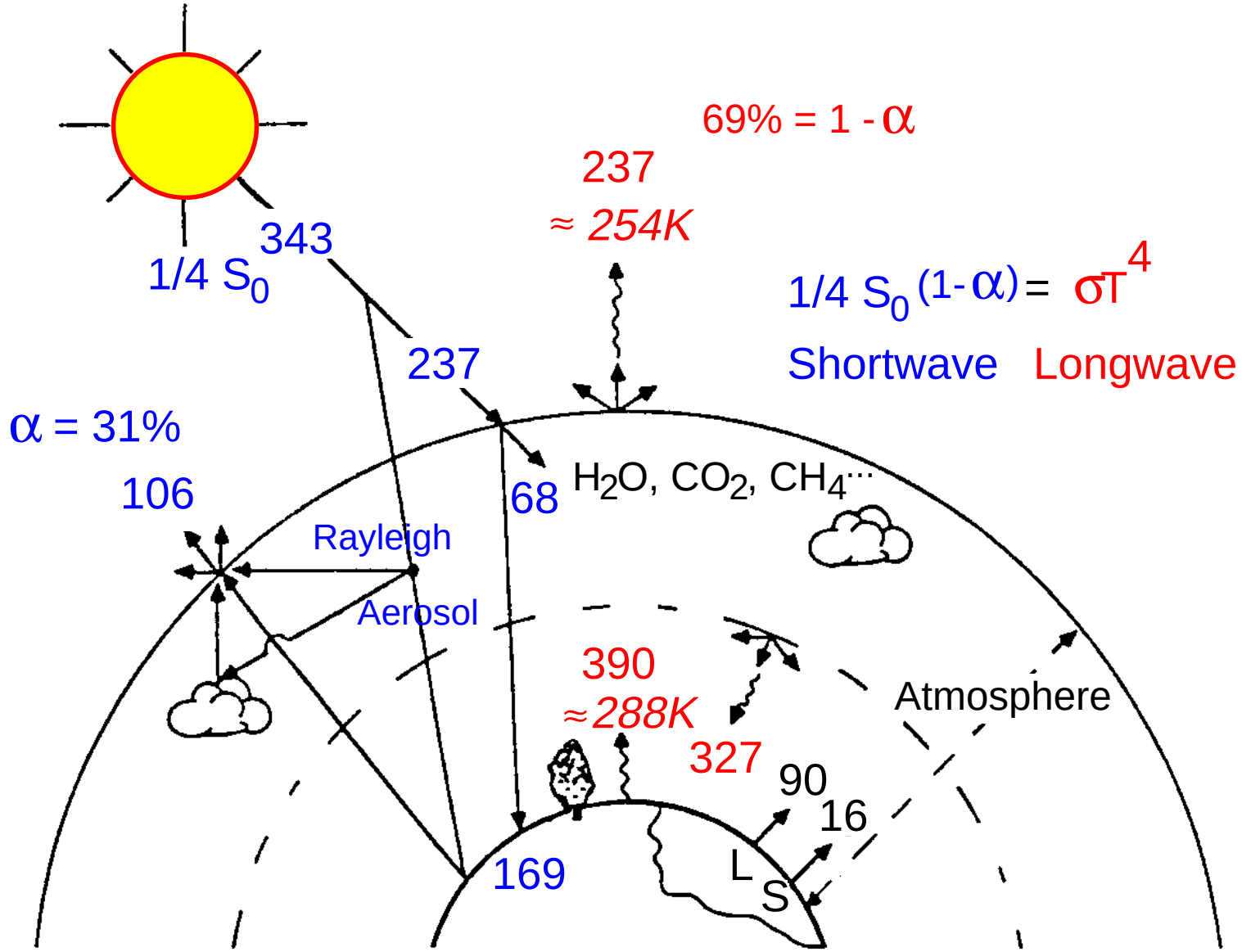
SeaWiFS Project, NASA/Goddard Space Flight Center, and ORBIMAGE

Atmospheric aerosols may result from primary emissions (dust, smoke) or from gas to particle conversion in the atmosphere (haze, smog).


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 RADIATION

Energy per area per time

Power per time

Unit:

Watt per square meter

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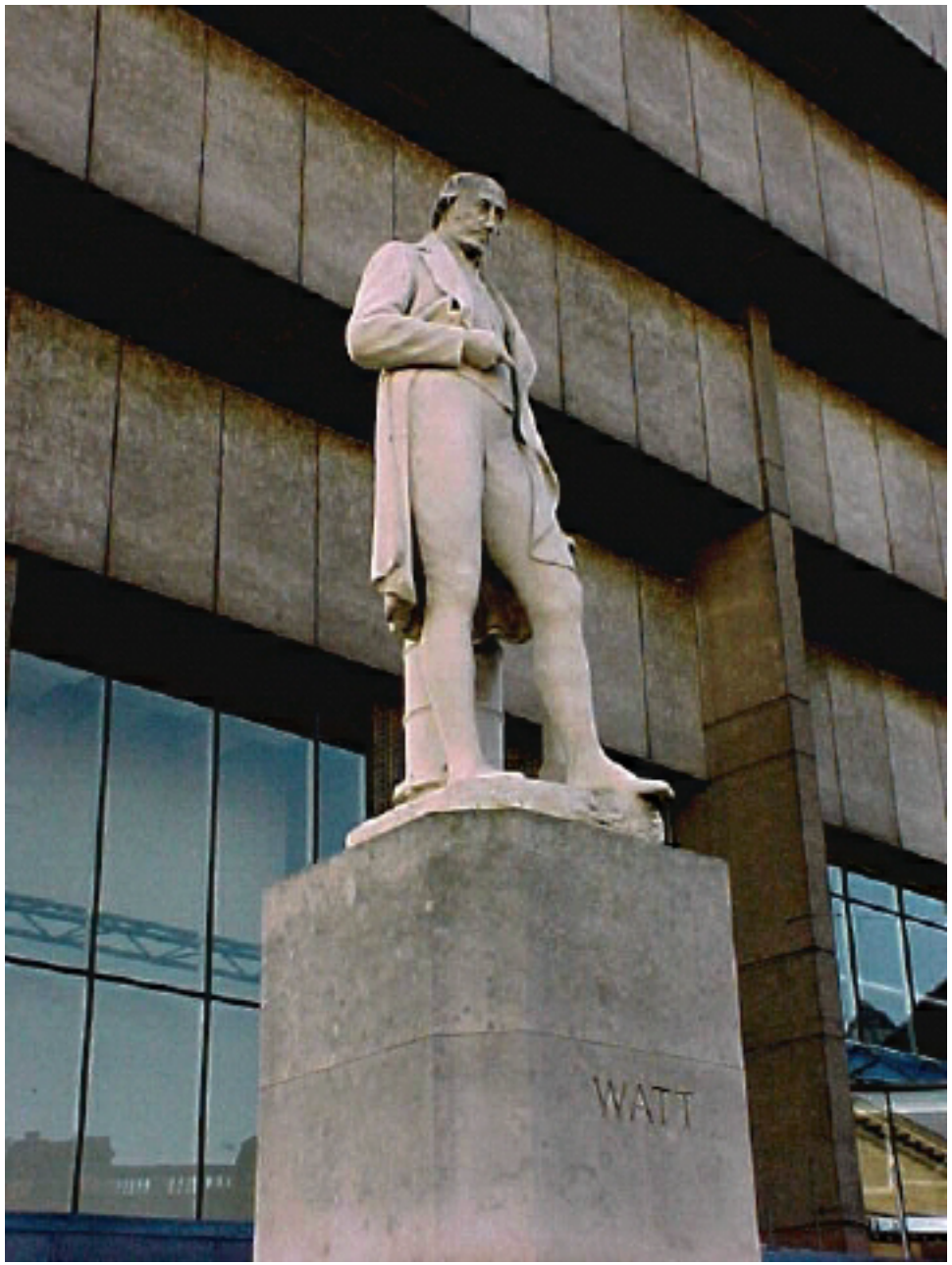


RADIATIVE FORCING

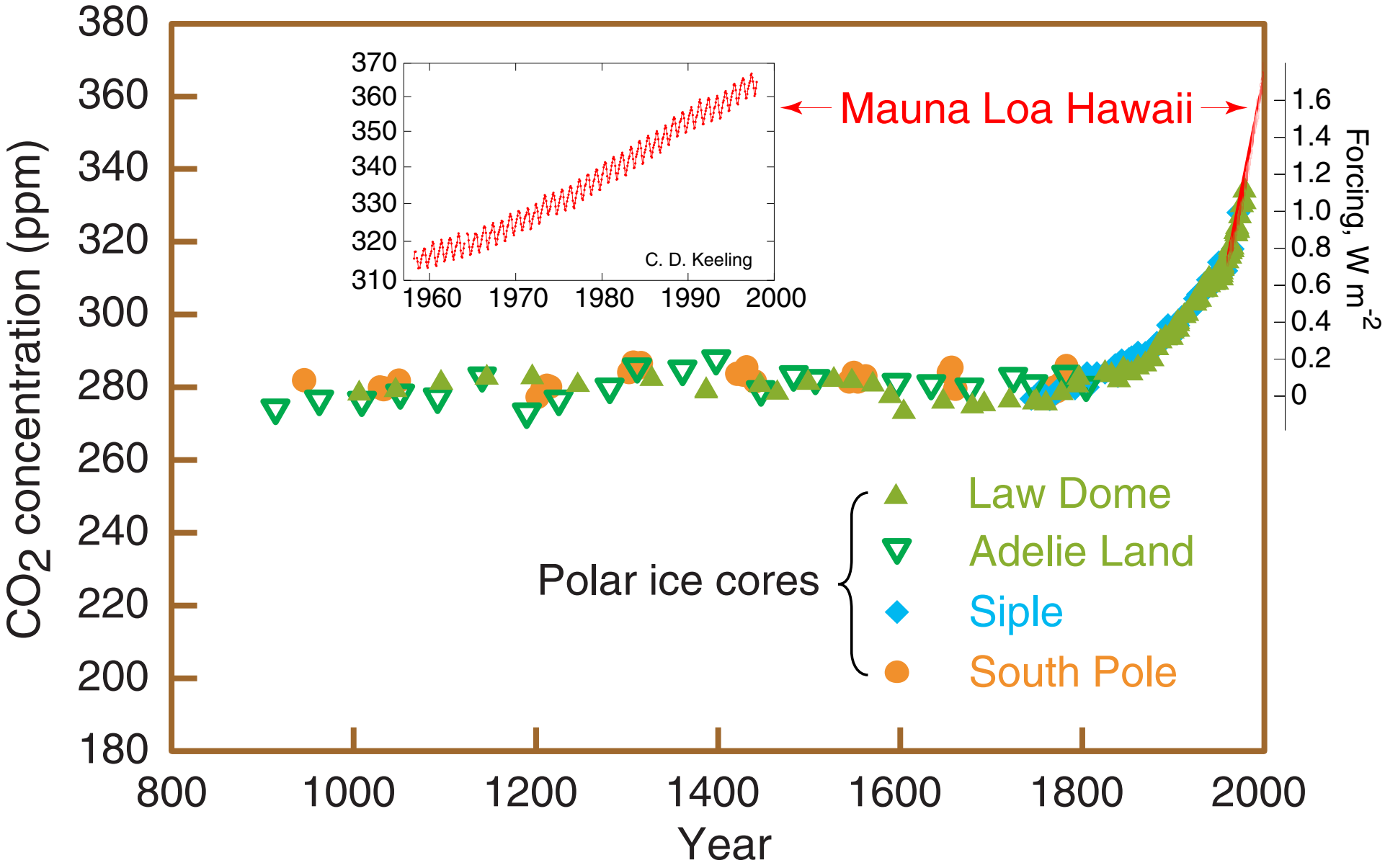
A ***change*** in a radiative flux term in the Earth's radiation budget, F, W m^{-2}.

Working hypothesis:

On a global basis radiative forcings are additive and fungible.

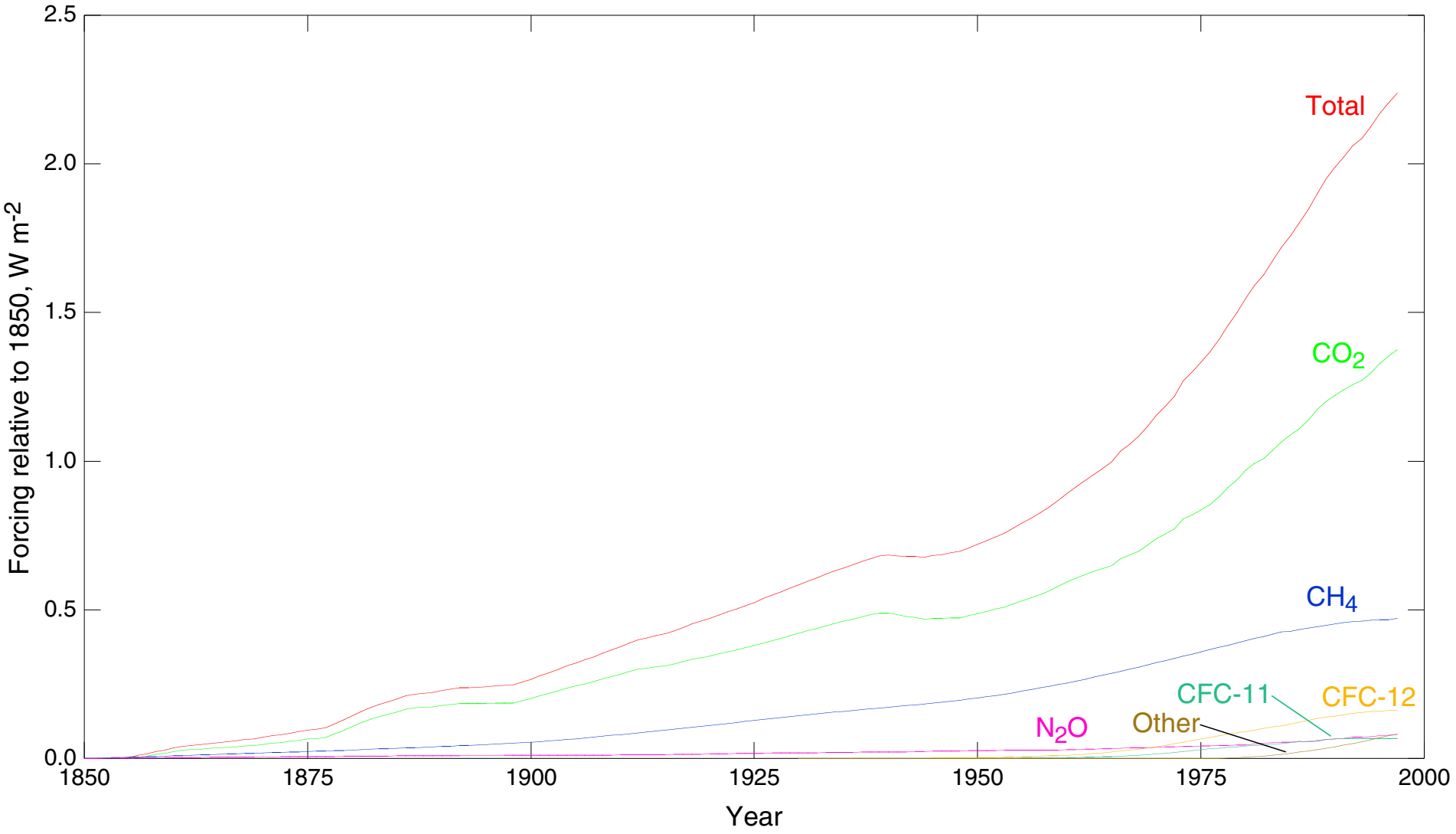
- This hypothesis is fundamental to the radiative forcing concept.
- This hypothesis underlies much of the assessment of climate change over the industrial period.

ATMOSPHERIC CARBON DIOXIDE IS INCREASING



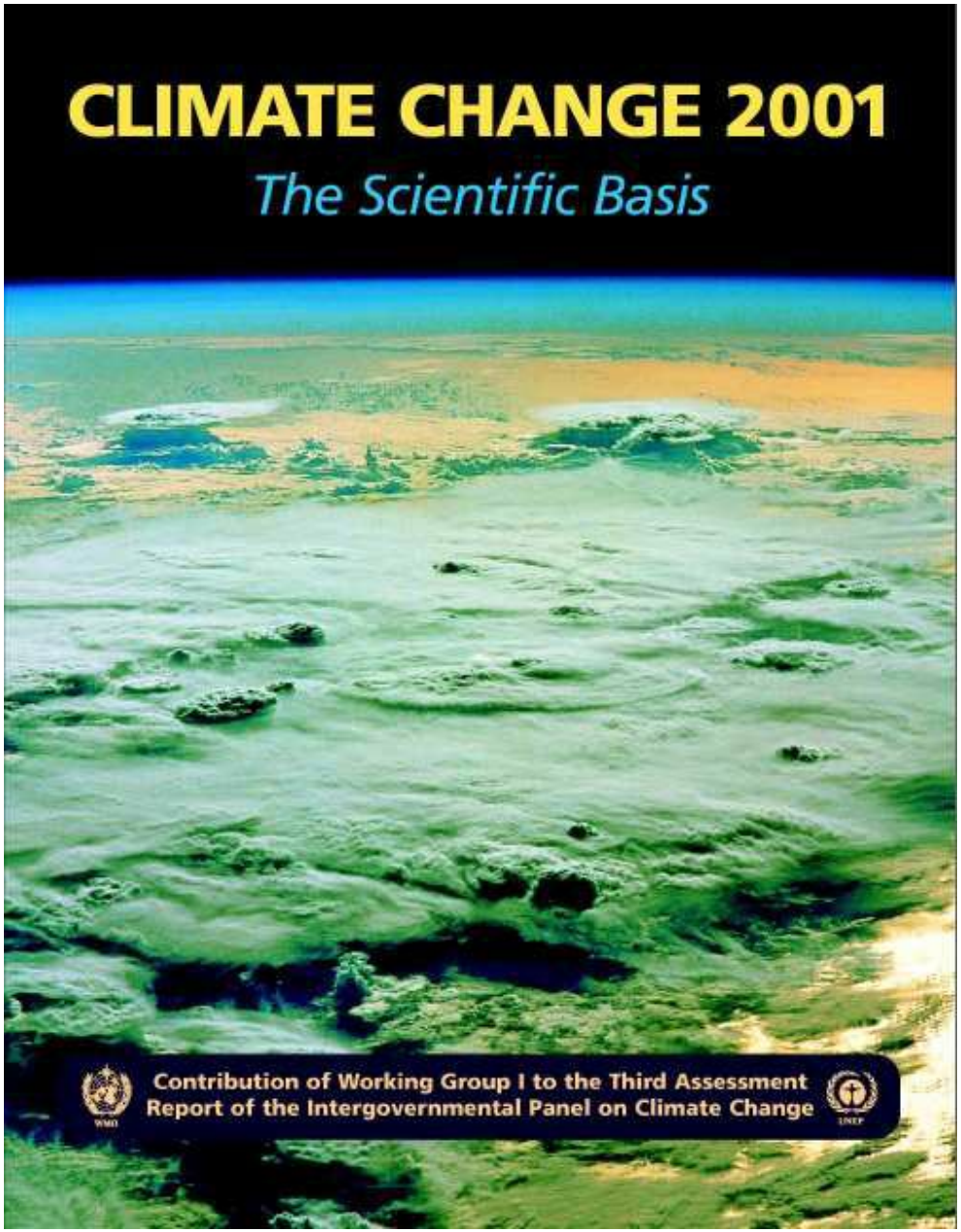
Global carbon dioxide concentration and infrared radiative forcing over the last thousand years

GREENHOUSE GAS FORCINGS OVER THE INDUSTRIAL PERIOD



Data: GISS

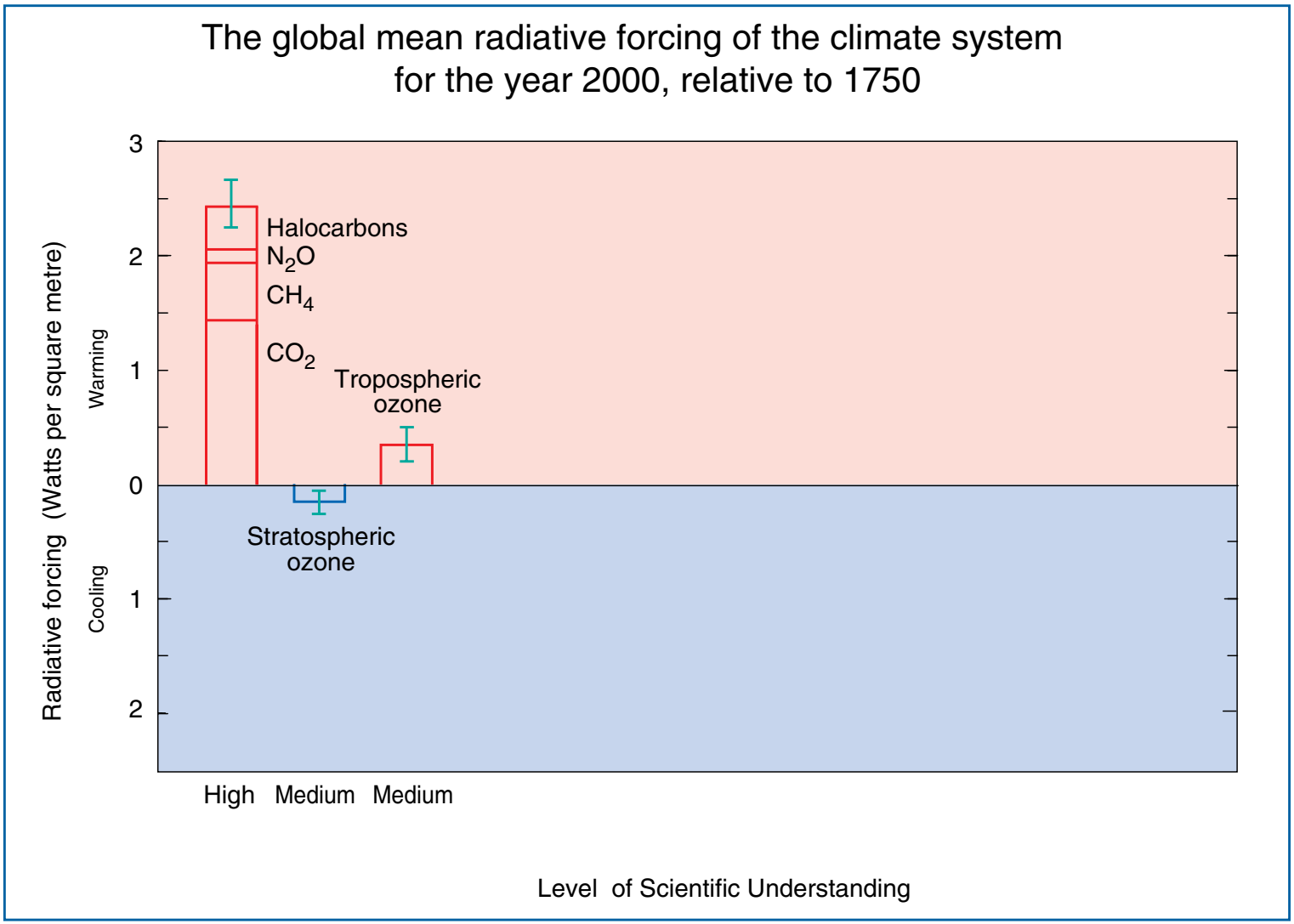
THE "BIBLE" OF CLIMATE CHANGE RESEARCH



Cambridge University Press, 2001

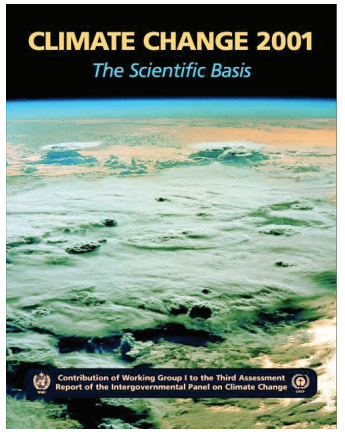
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)

Greenhouse gases only



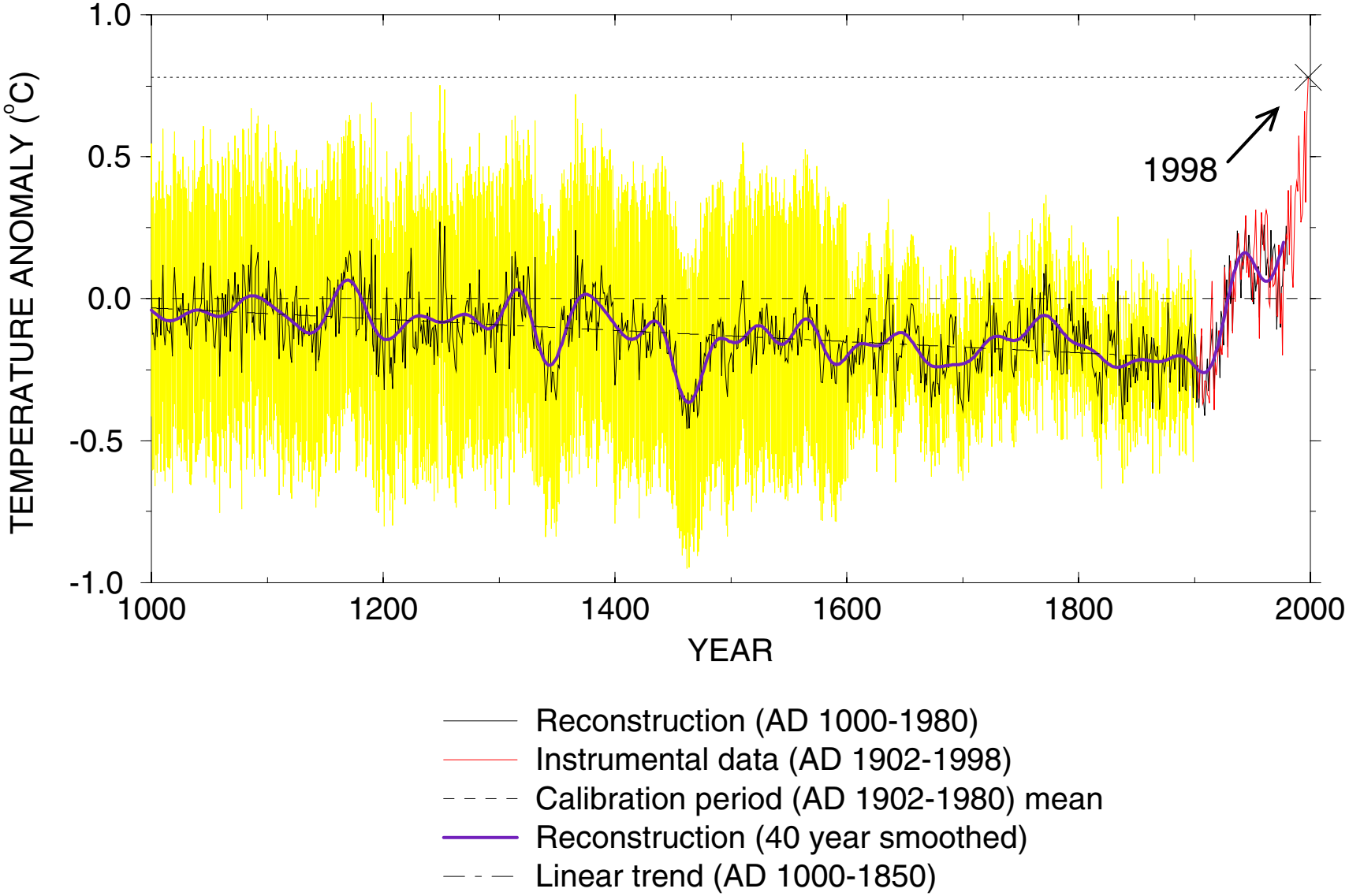
Summary for Policymakers

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



NORTHERN HEMISPHERE TEMPERATURE TREND (1000-1998)

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



Mann et al., GRL, 1999

CLIMATE RESPONSE

The ***change*** in global and annual mean temperature, ΔT, 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, λ, K/(W m^{-2}).

EMPIRICAL CLIMATE SENSITIVITY

Greenhouse forcing over the industrial period is 2.5 W m^{-2}

Temperature increase over the industrial period is 0.6 K.

Empirical Sensitivity: $\lambda = \frac{dT}{dF} = \frac{0.6 \text{ K}}{2.5 \text{ W m}^{-2}} = 0.24 \text{ K / (W m}^{-2})$

CLIMATE CHANGE SENSITIVITY

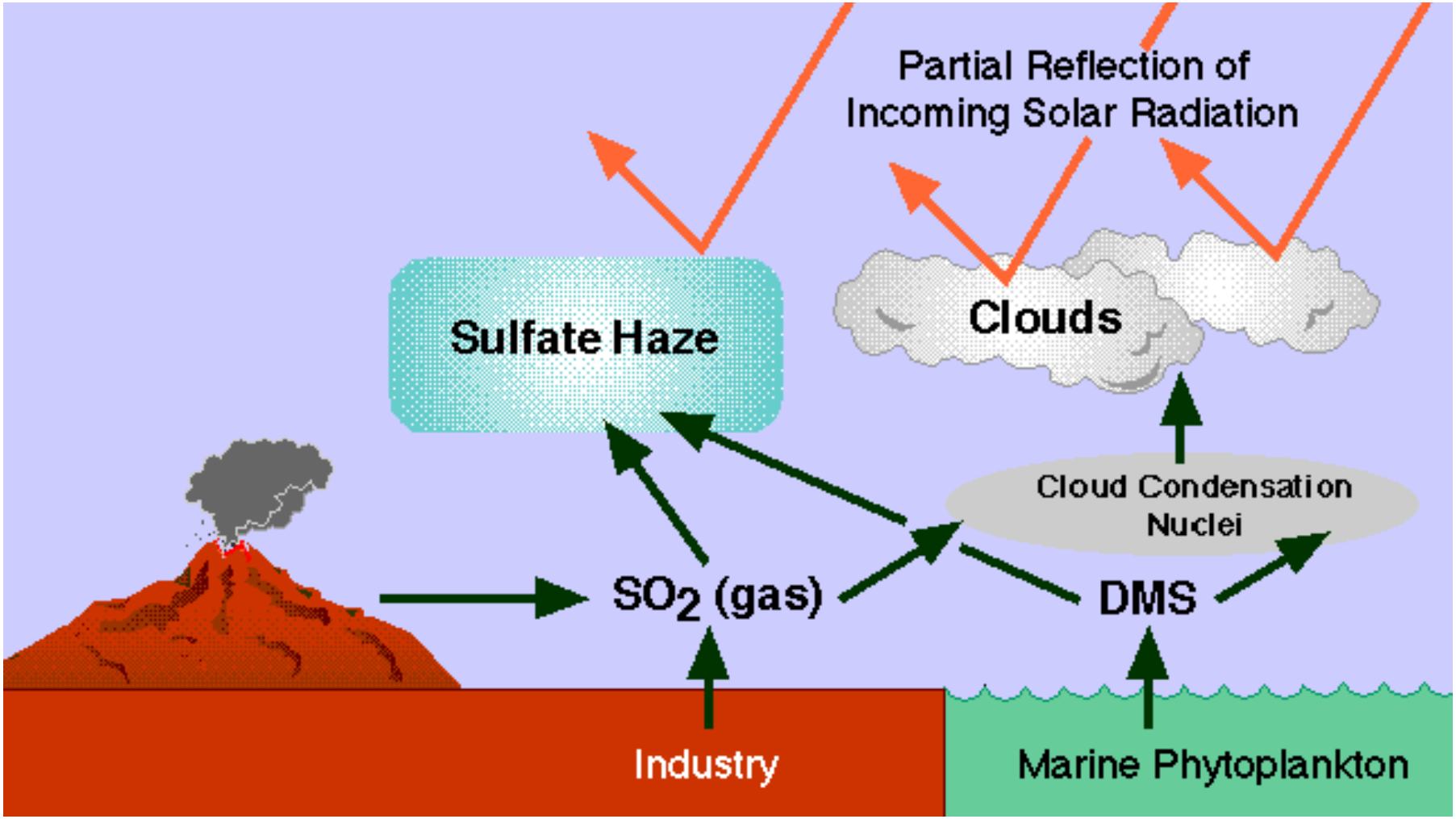
Summary of Current Models

Units: K / (W m^{-2})

Number of Models	Mean	Standard Deviation	Range
15	0.87	0.23	0.5 - 1.25

IPCC, Climate Change 2001, Cambridge University Press, 2001

RADIATIVE FORCING OF CLIMATE CHANGE BY AEROSOLS



AEROSOL INFLUENCES ON RADIATION BUDGET AND CLIMATE

Direct Effect (Cloud-free sky)

Light scattering -- Cooling influence

Light absorption -- Warming influence, depending on surface

Indirect Effects (Aerosols influence cloud properties)

More droplets -- Brighter clouds (Twomey)

More droplets -- Enhanced cloud lifetime (Albrecht)

Semi-Direct Effect

Absorbing aerosol heats air and evaporates clouds

DIRECT EFFECT

BIOMASS BURNING AND WIDESPREAD AEROSOL
Northeastern Oklahoma, 2000-12-01

DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE AEROSOL

Geophysics

Aerosol Optical Depth

$$\overline{\Delta F_R} = -\frac{1}{2} F_T T^2 (1 - A_c)(1 - R_s)^2 \cdot \overline{\beta} \alpha_{SO_4^{2-}} f(RH) \cdot Q_{SO_2} Y_{SO_4^{2-}} \left(\frac{MW_{SO_4^{2-}}}{MW_S} \right) \tau_{SO_4^{2-}} \Big/ A$$

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,

$\alpha_{SO_4^{2-}}$ 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}	—
$1-A_c$	0.4	——	1.1
T	0.76	——	1.15
$1-R_s$	0.85	——	1.1
$\overline{\beta}$	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	5×10^{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 $f_t = \exp\left[\sum (\log f_i)^2\right]^{1/2}$

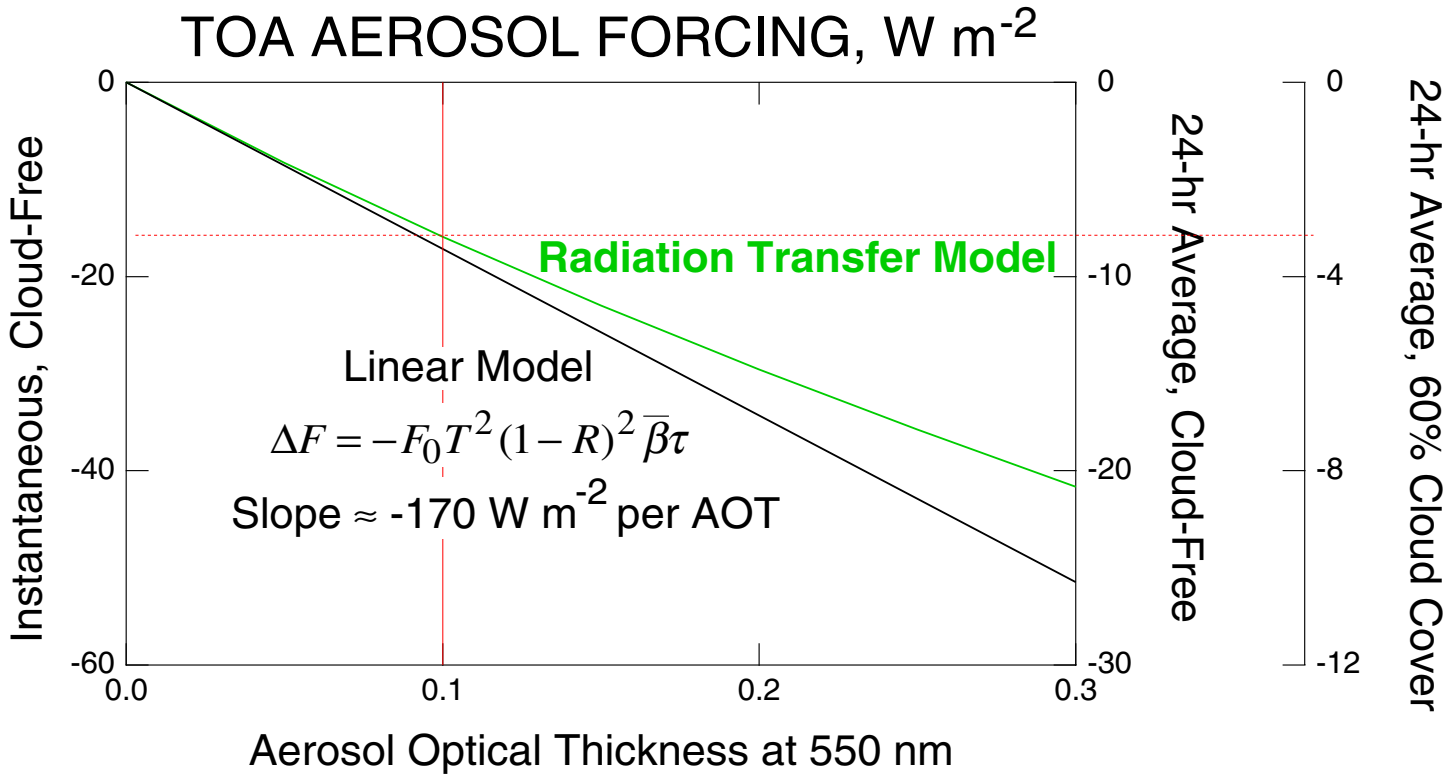
Penner, Charlson, Hales, Laulainen, Leifer, Novakov, Ogren, Radke, Schwartz & Travis, BAMS, 1994

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 ω_0 = 1.

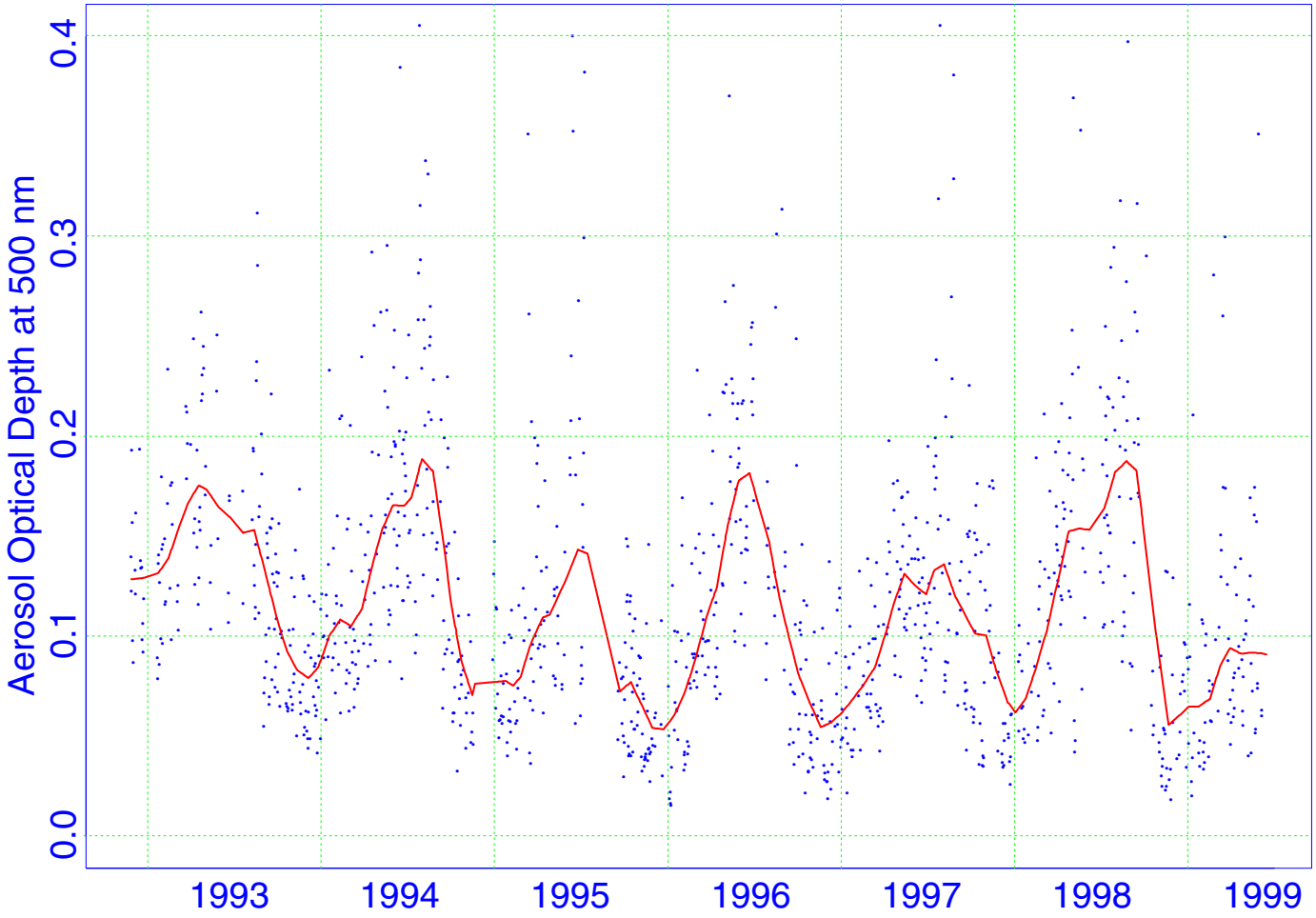


Global-average AOT 0.1 corresponds to global-average forcing -3.2 W m^{-2}.

AEROSOL OPTICAL DEPTH

Determined by Sunphotometry

North Central Oklahoma



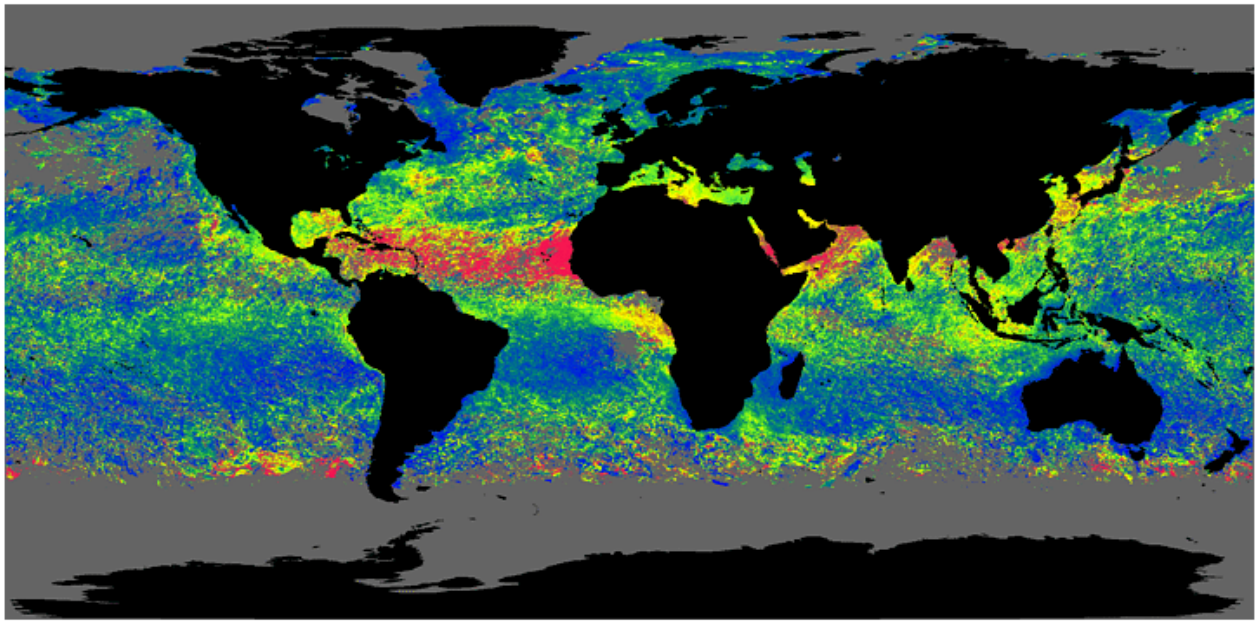
J. Michalsky et al., JGR, 2001

MONTHLY AVERAGE AEROSOL JUNE 1997

Polder radiometer on Adeos satellite

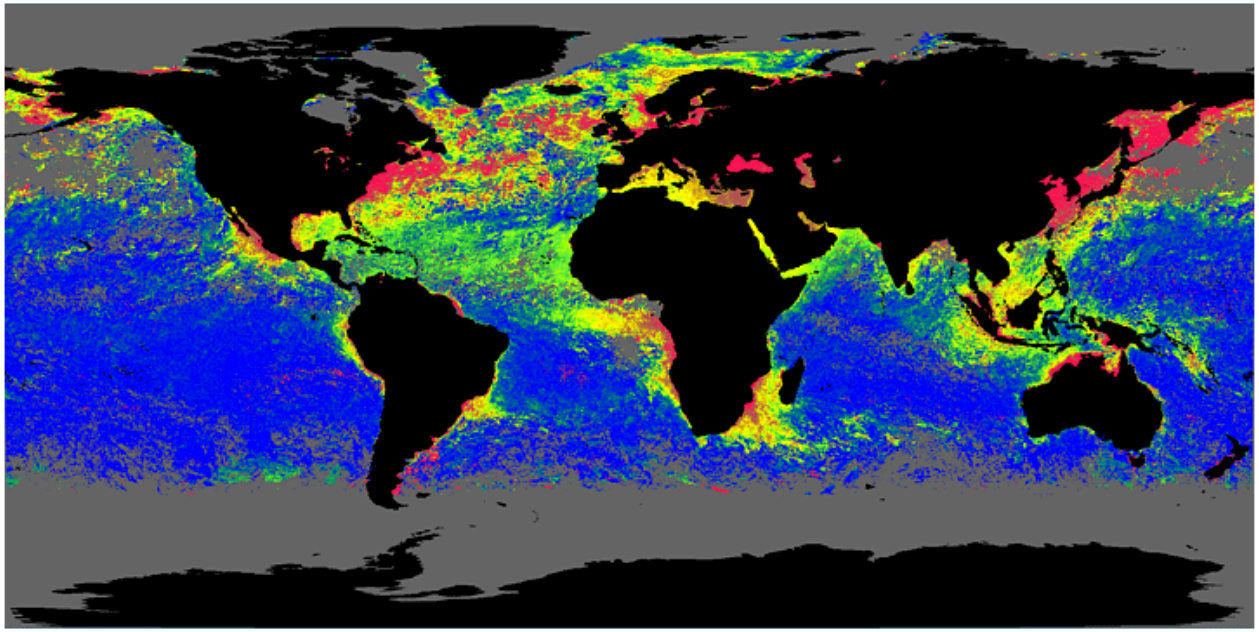
Optical Thickness τ

$\lambda = 865$ nm

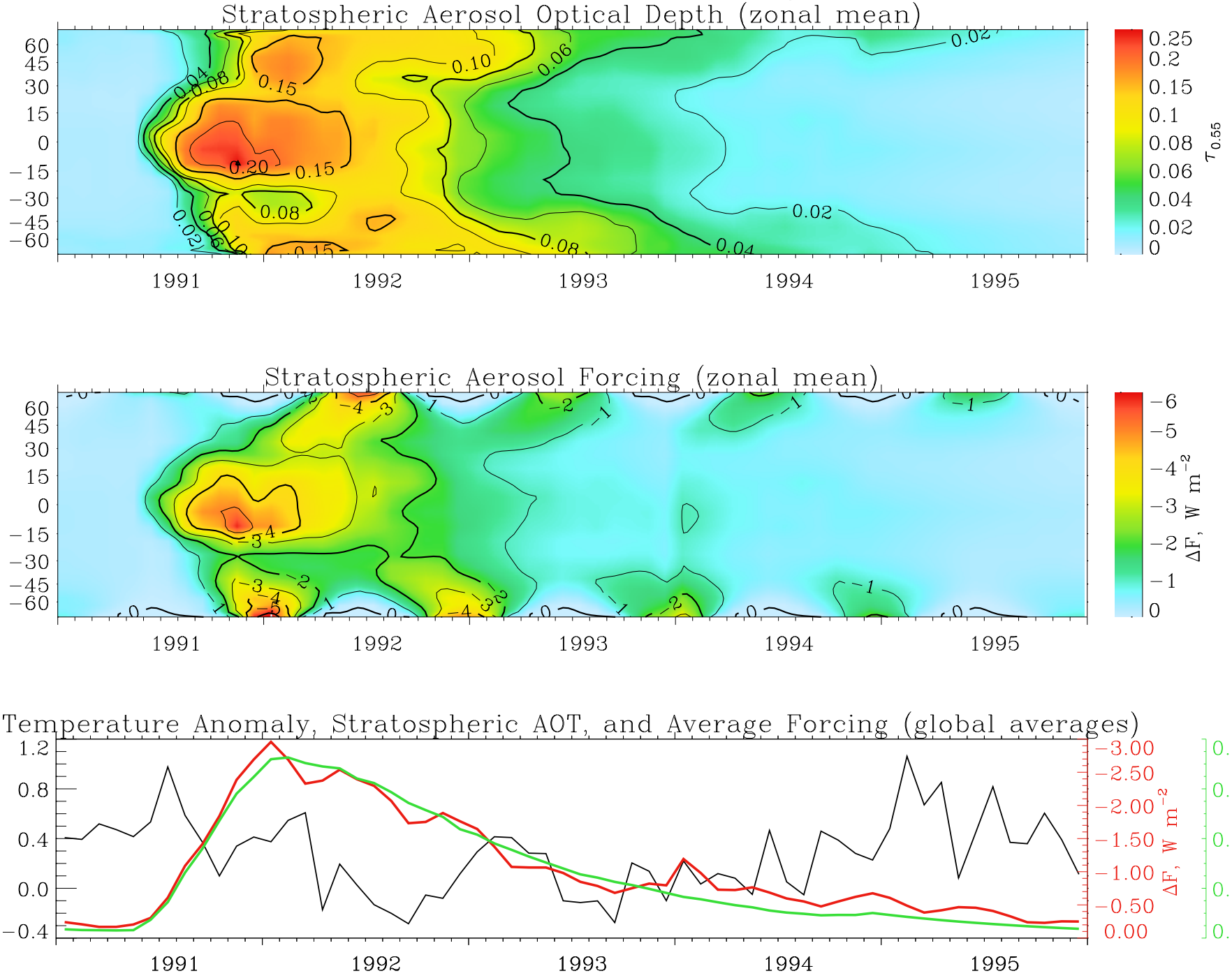


Ångström Exponent α

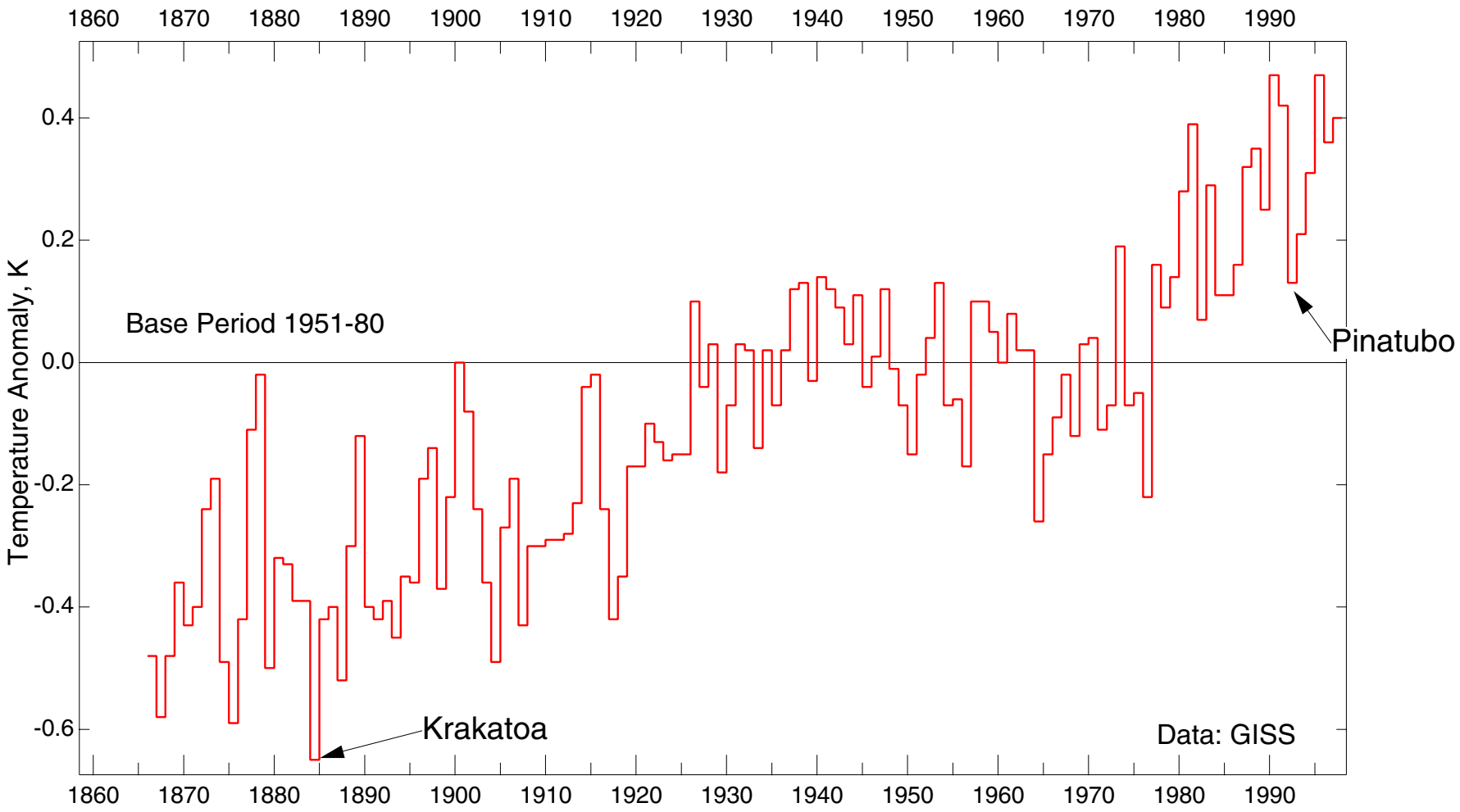
$\alpha = -d \ln \tau / d \ln \lambda$



Influence of Pinatubo Eruption on Aerosol Forcing and Global Temperature



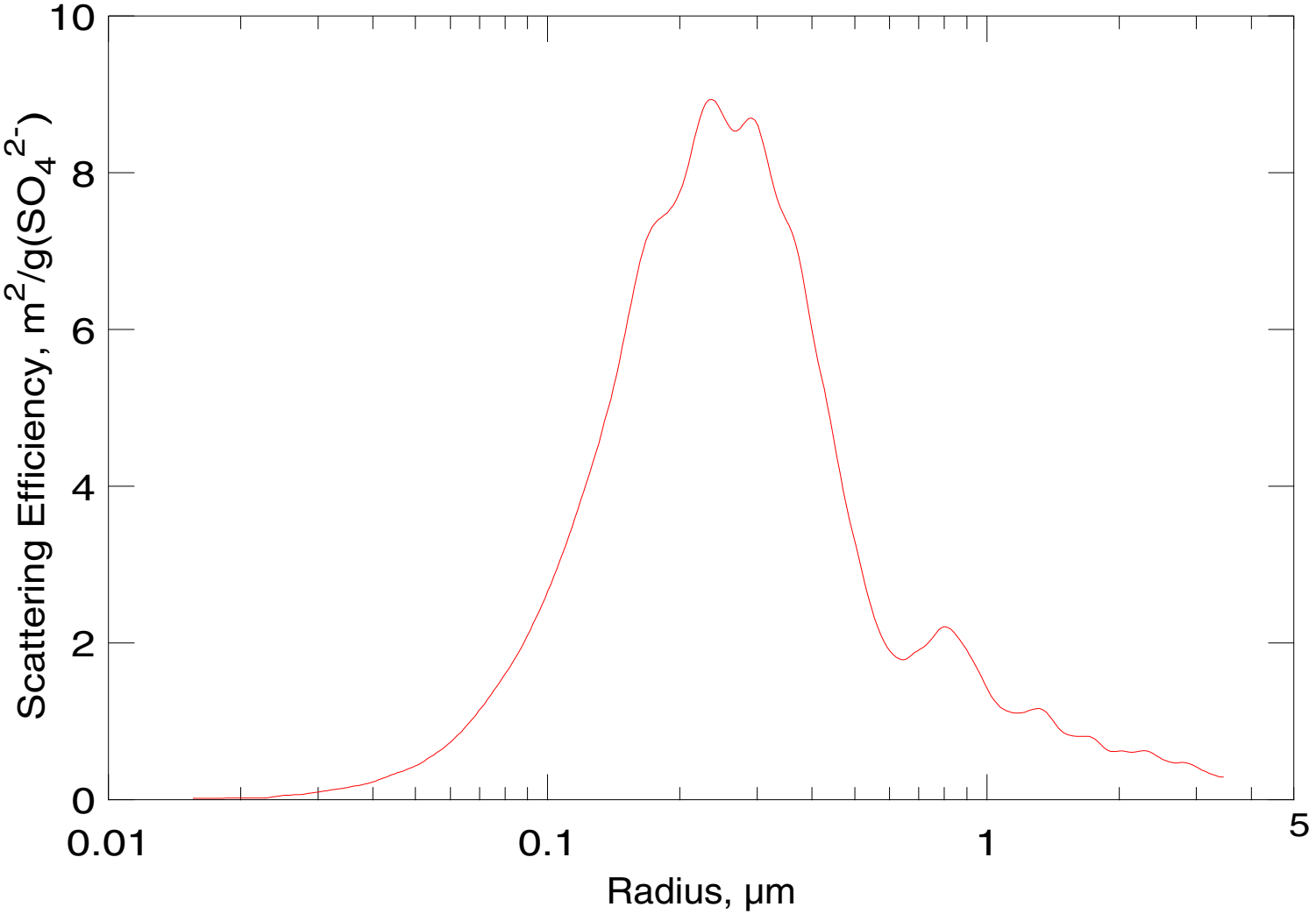
GLOBAL TEMPERATURE TREND OVER THE INDUSTRIAL PERIOD



LIGHT SCATTERING EFFICIENCY

Dependence on particle radius

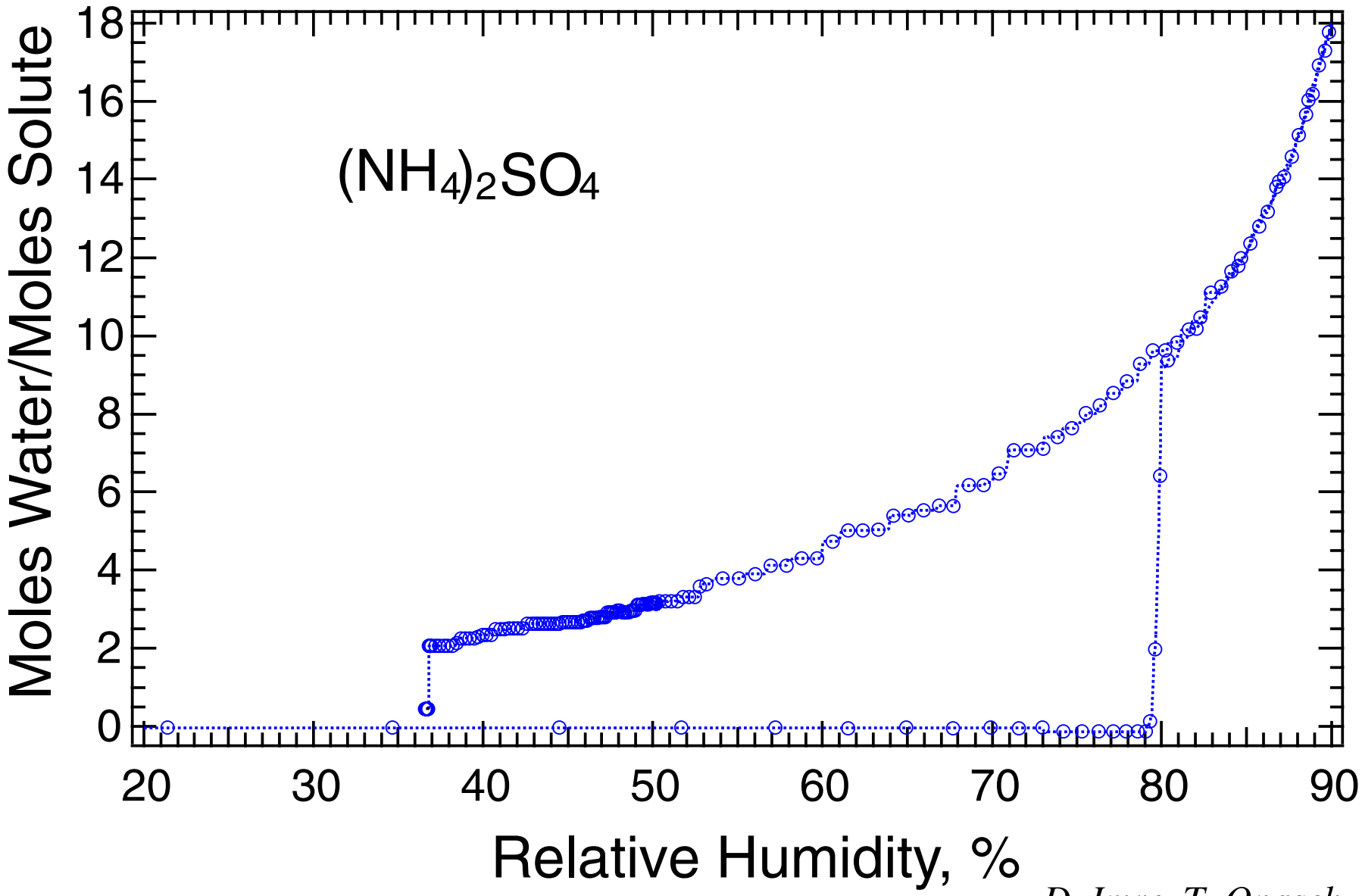
Ammonium Sulfate, 530 nm



Data of Ouimette and Flagan, 1982

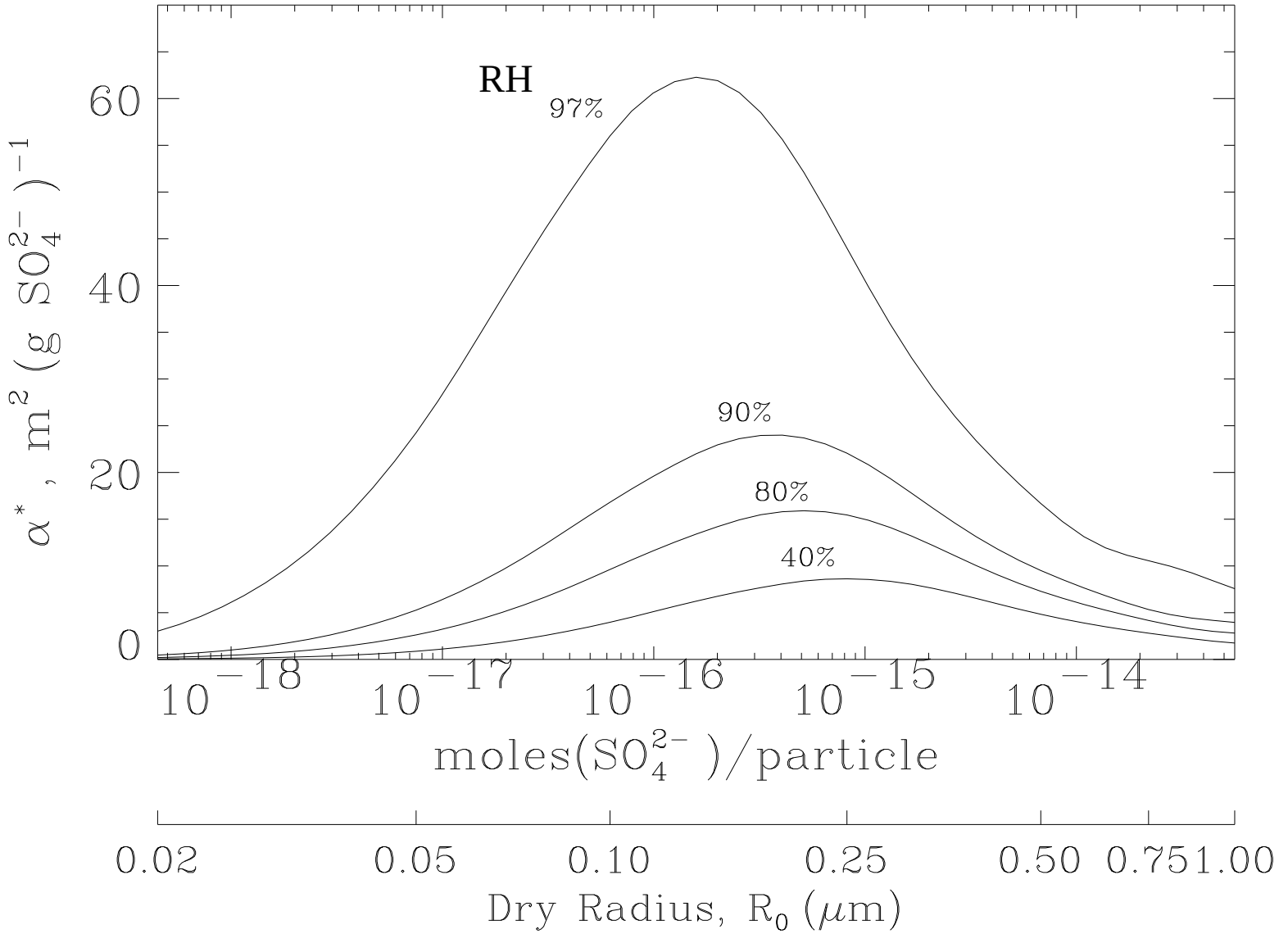
WATER UPTAKE BY HYGROSCOPIC PARTICLE

Dependence on relative humidity



D. Imre, T. Onasch, BNL

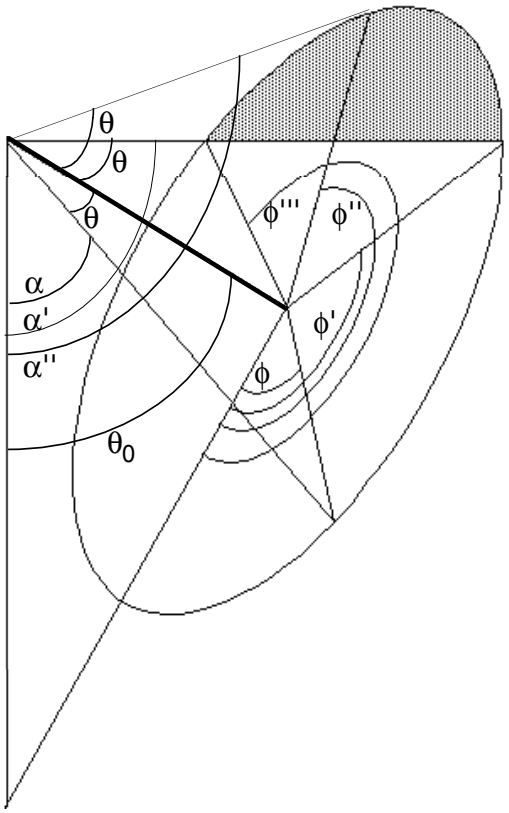
LIGHT SCATTERING EFFICIENCY OF $(NH_4)_2SO_4$ DEPENDENCE ON PARTICLE SIZE AND RH



Nemesure et al., JGR, 1995

UPSCATTER FRACTION

SCATTERING OF SOLAR RADIATION BY AEROSOL PARTICLE

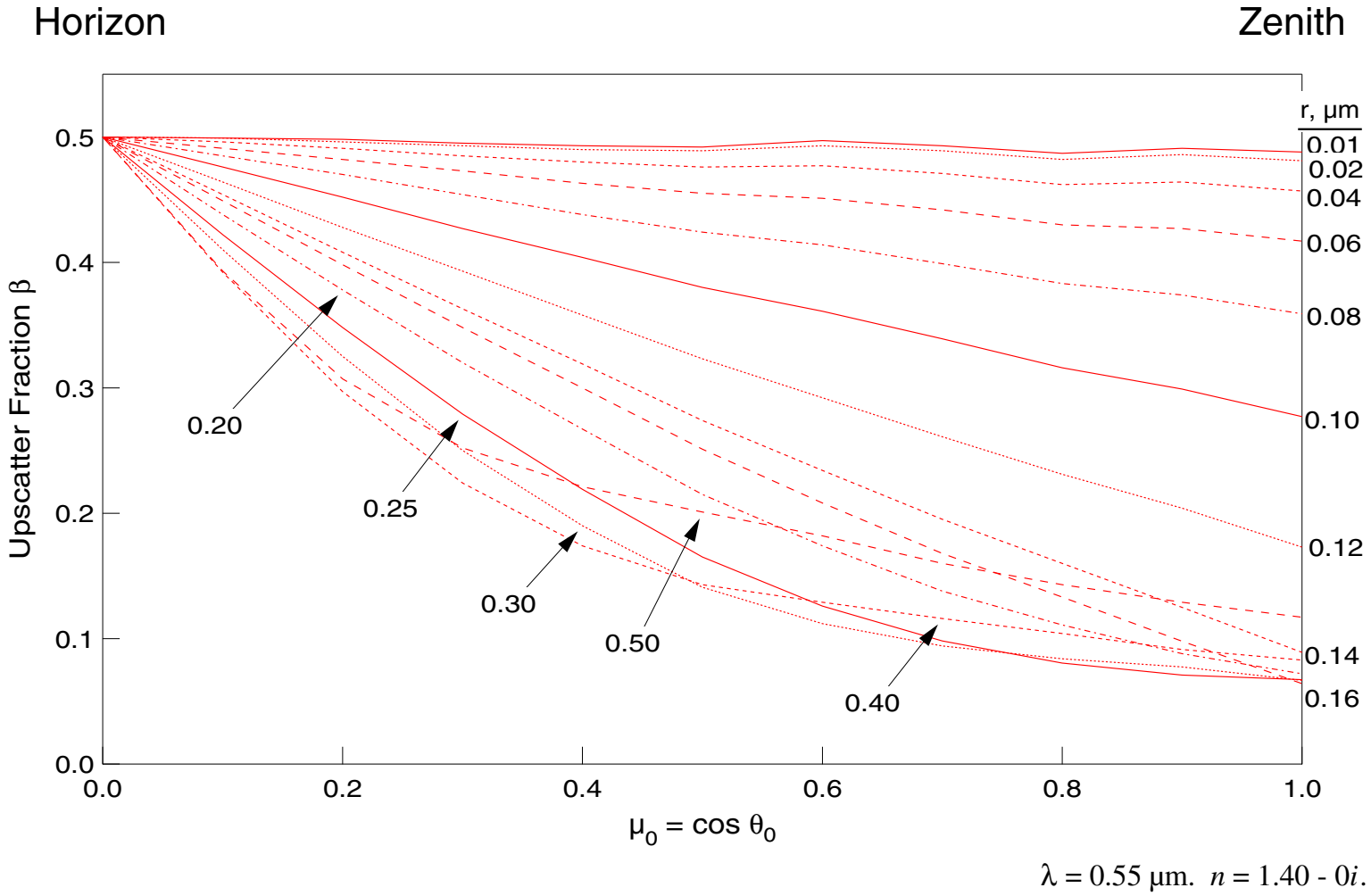


Upscatter fraction β is the fraction of radiation scattered into the upward hemisphere.

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

UPSCATTER FRACTION

Dependence on solar zenith angle and particle radius



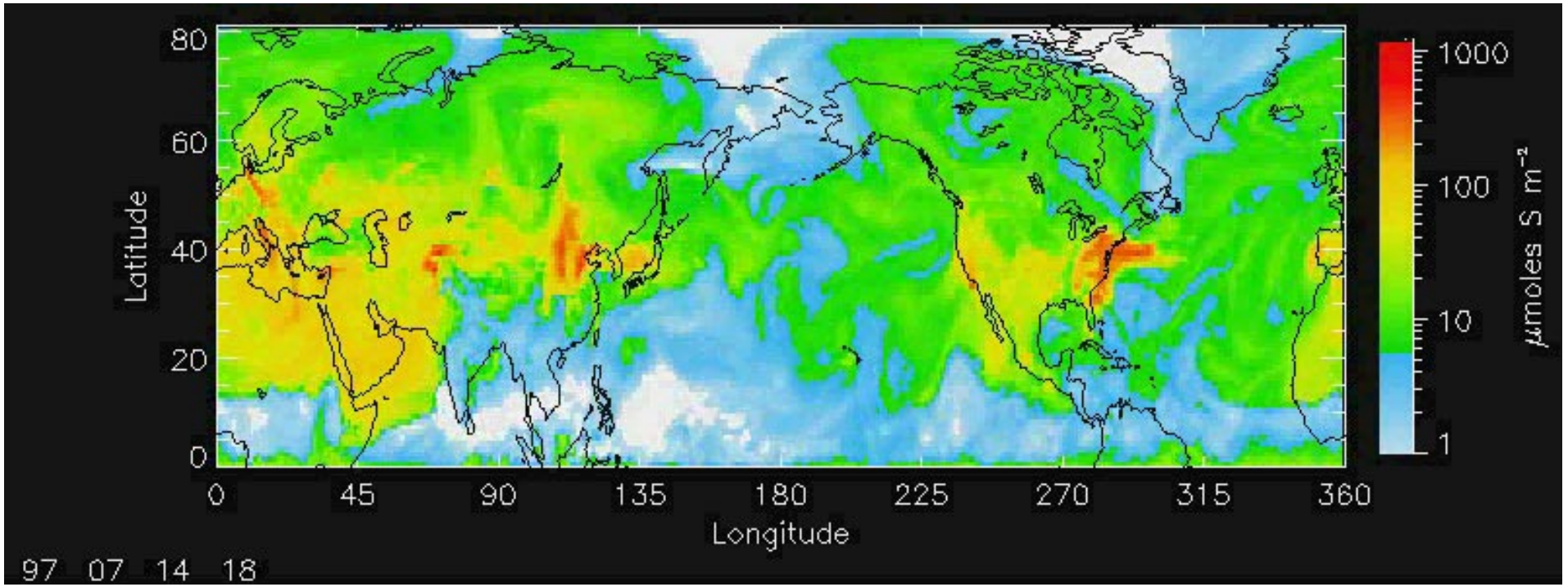
For sun at horizon $\beta = 0.5$ (by symmetry).

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

HEMISPHERIC DISTRIBUTION OF SULFATE COLUMN BURDEN

Vertical integral of concentration

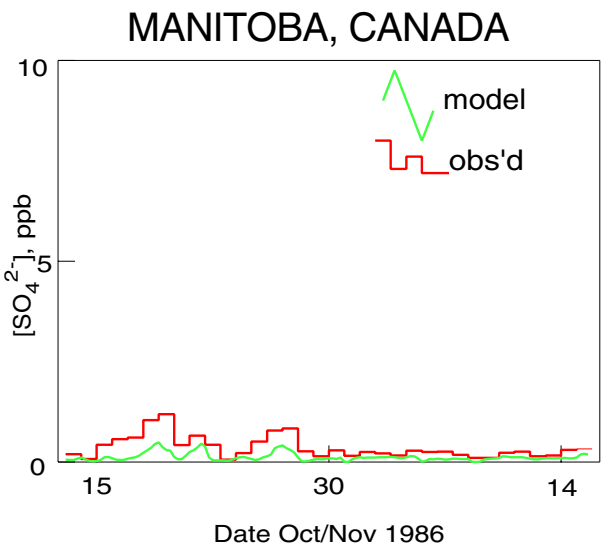
July 14, 1997, 1800 UTC

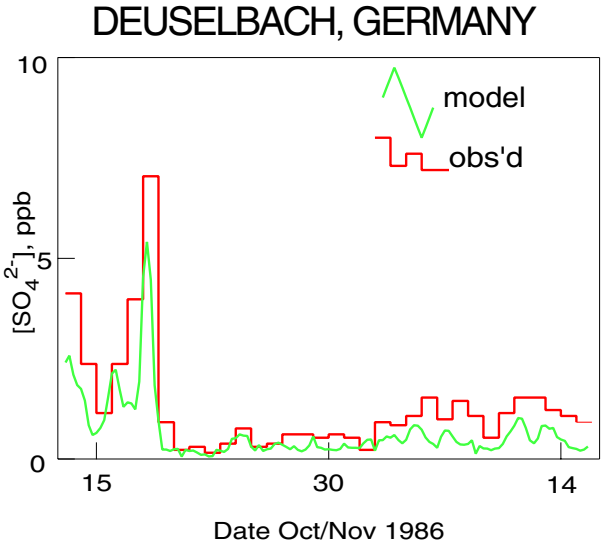


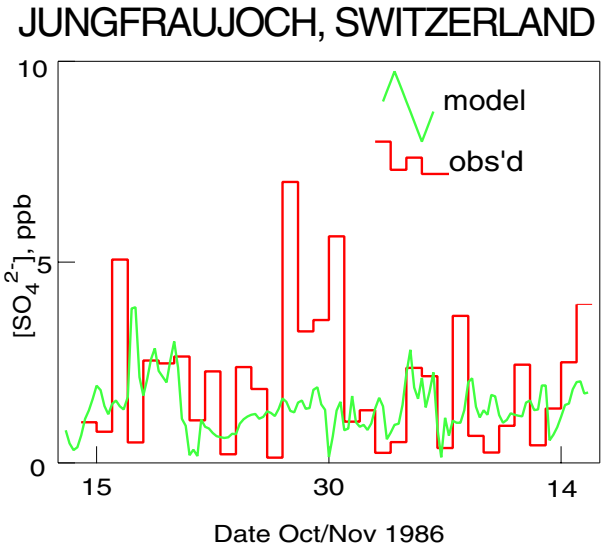
Brookhaven National Laboratory Chemical Transport Model

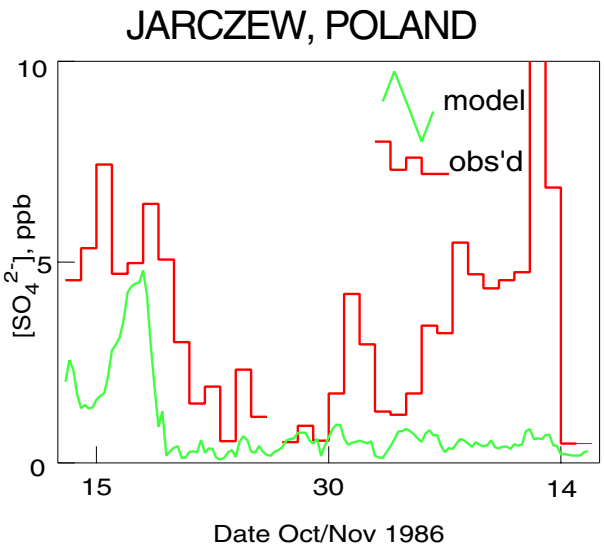
COMPARISON OF MODEL AND OBSERVATIONS

Comparisons for 24-hr sulfate mixing ratio at surface









COMPARISON OF MODEL AND OBSERVATIONS

Statistics of Comparisons

	N	Median Spread
Obs-Obs	503	1.5
Model-Obs Same locations	503	1.9
Model-Obs All locations	7907	2.3

Benkovitz and Schwartz, *JGR*, 1997

DO YOU HAVE A FEW MOMENTS?

The Problem

How to represent the size-distribution of atmospheric aerosols and its evolution in chemical transport models

The Solution

Represent the size distribution in terms of its low-order moments

$$\mu_k \equiv \int_0^\infty r^k \left(\frac{dN}{dr}\right) dr$$

APPLICATION TO SULFATE IN EASTERN NORTH AMERICA

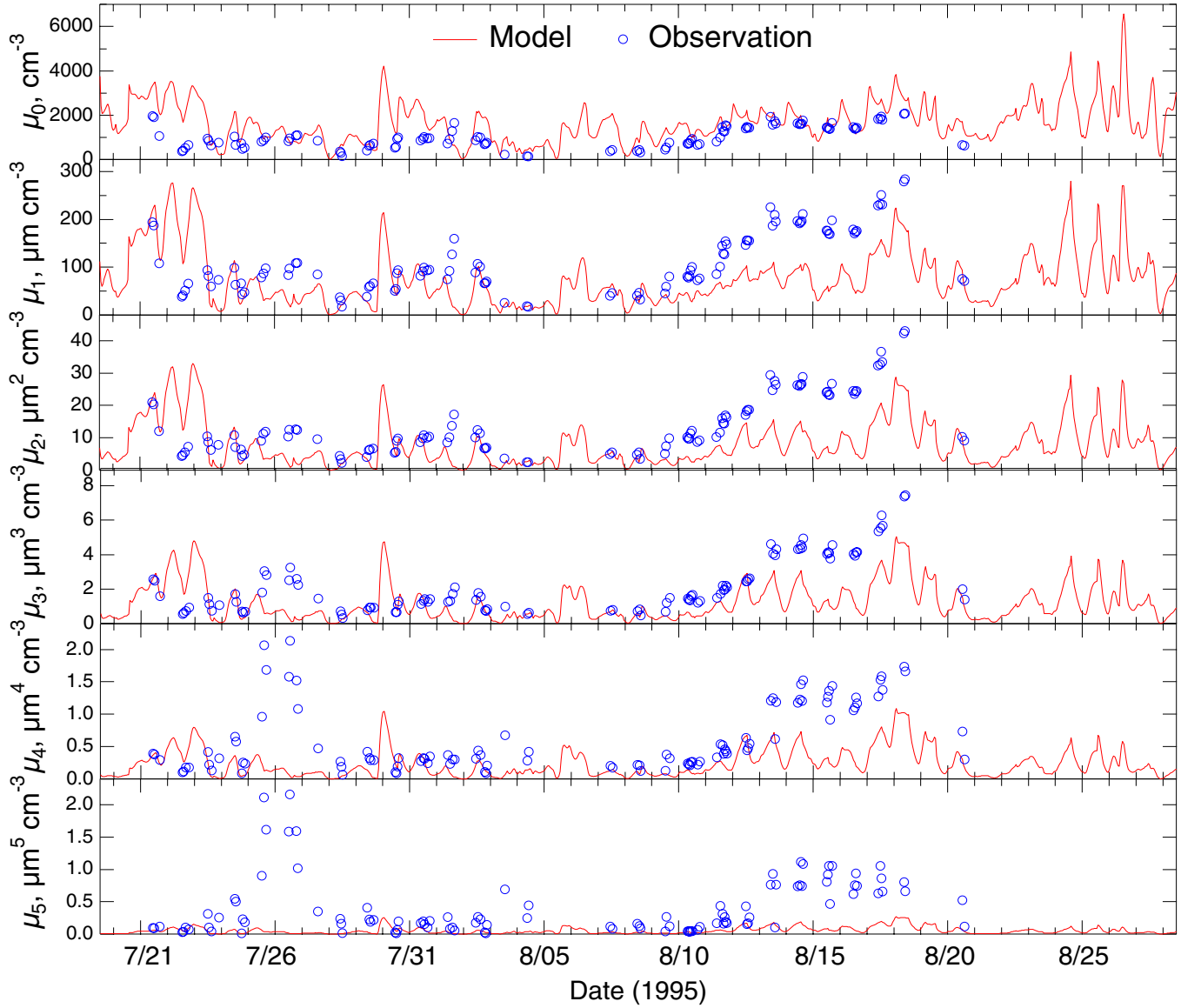
Simulations: 40 days, 19 July to 28 August 1995.

Comparison with observations: Sulfate mass concentration, aerosol number concentration and size distributions at the Great Smoky Mountains National Park during Southeastern Aerosol and Visibility Study.

Limitation: Model is for sulfate only; size measurements are for entire aerosol, not just sulfate.

TIME SERIES COMPARISON FOR AEROSOL MOMENTS

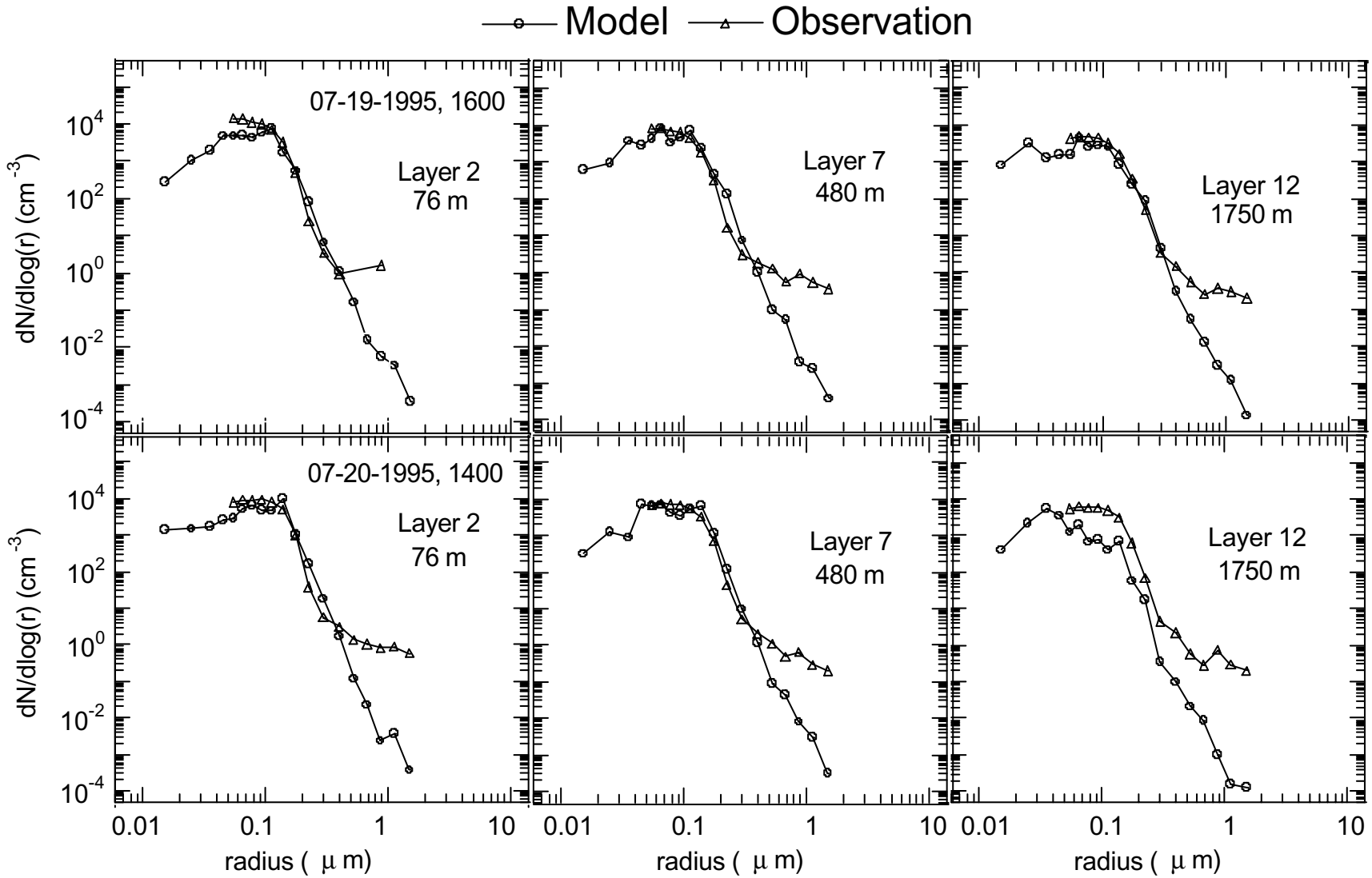
Look Ridge, Great Smoky Mountains TN (84° W, 36° N; 900 m) during SEAVS



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

SIZE DISTRIBUTIONS

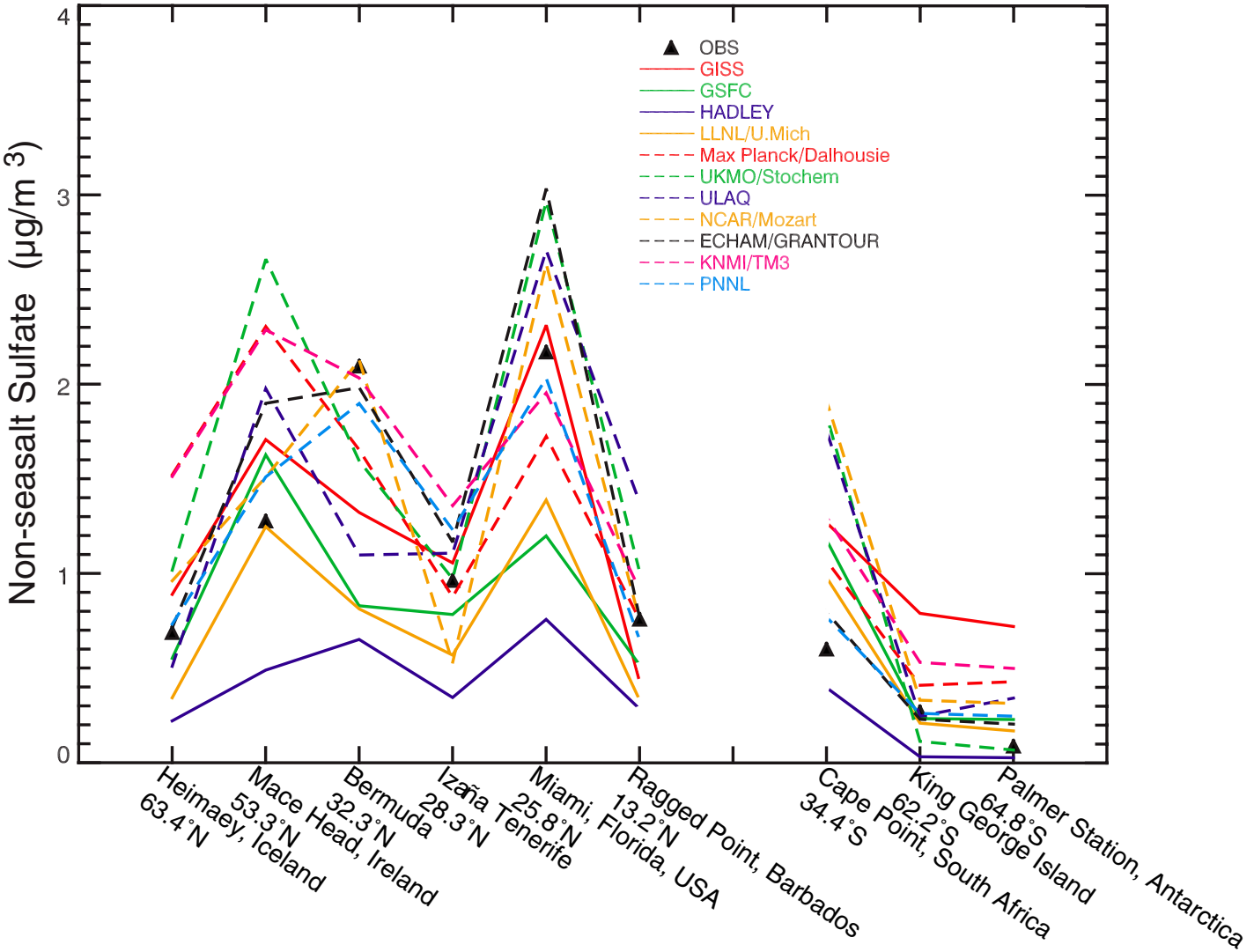
Comparison of Measurement and Retrieval from Model At 3 Altitudes near Nashville TN



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

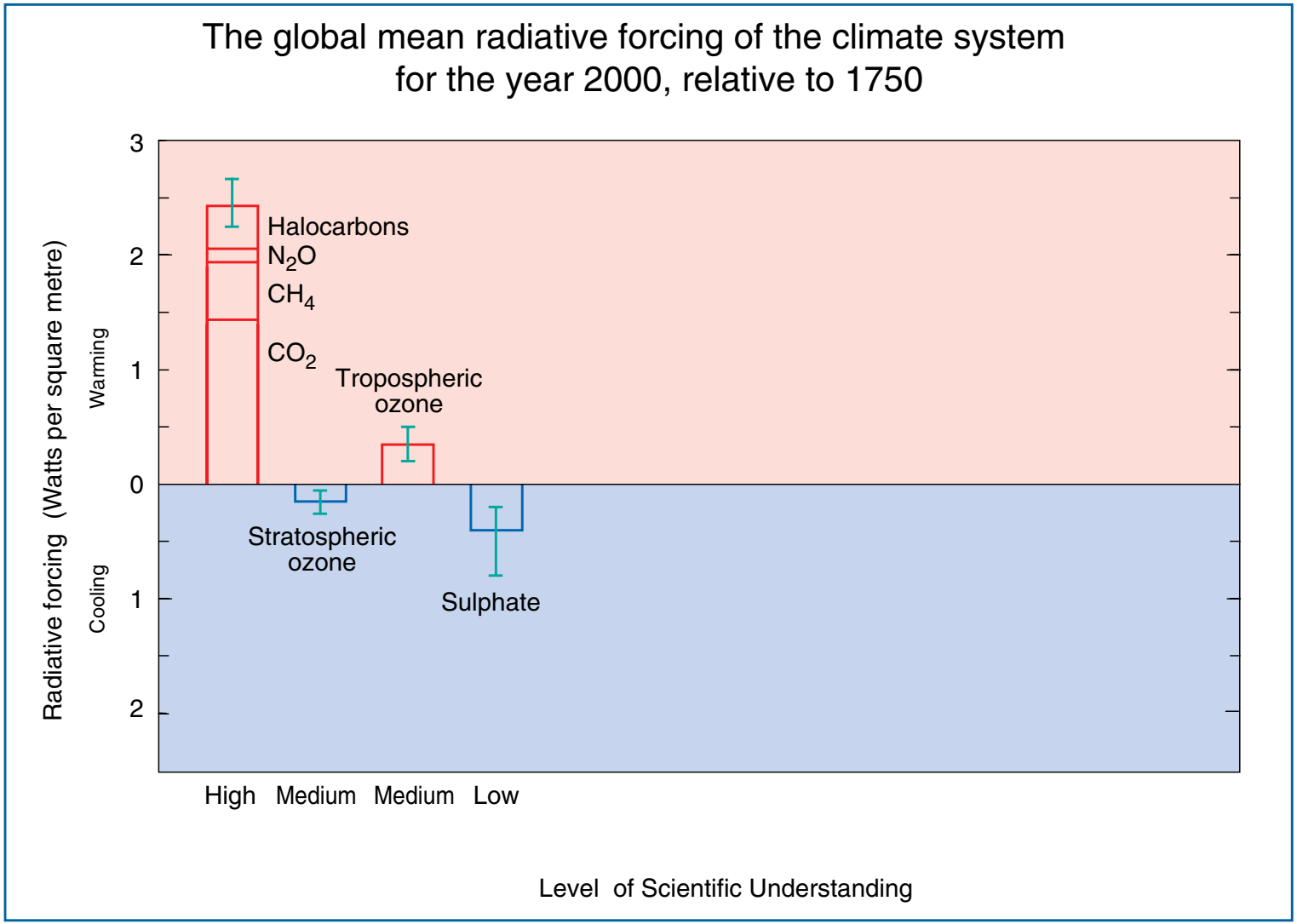
SULFATE MODEL INTERCOMPARISON

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



Penner et al., IPCC, 2001

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



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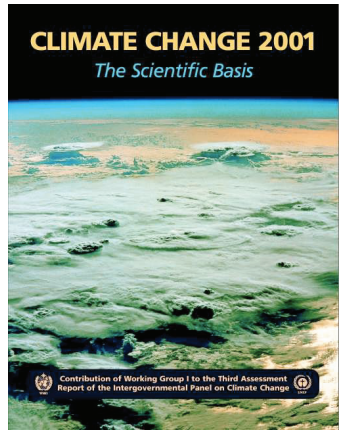
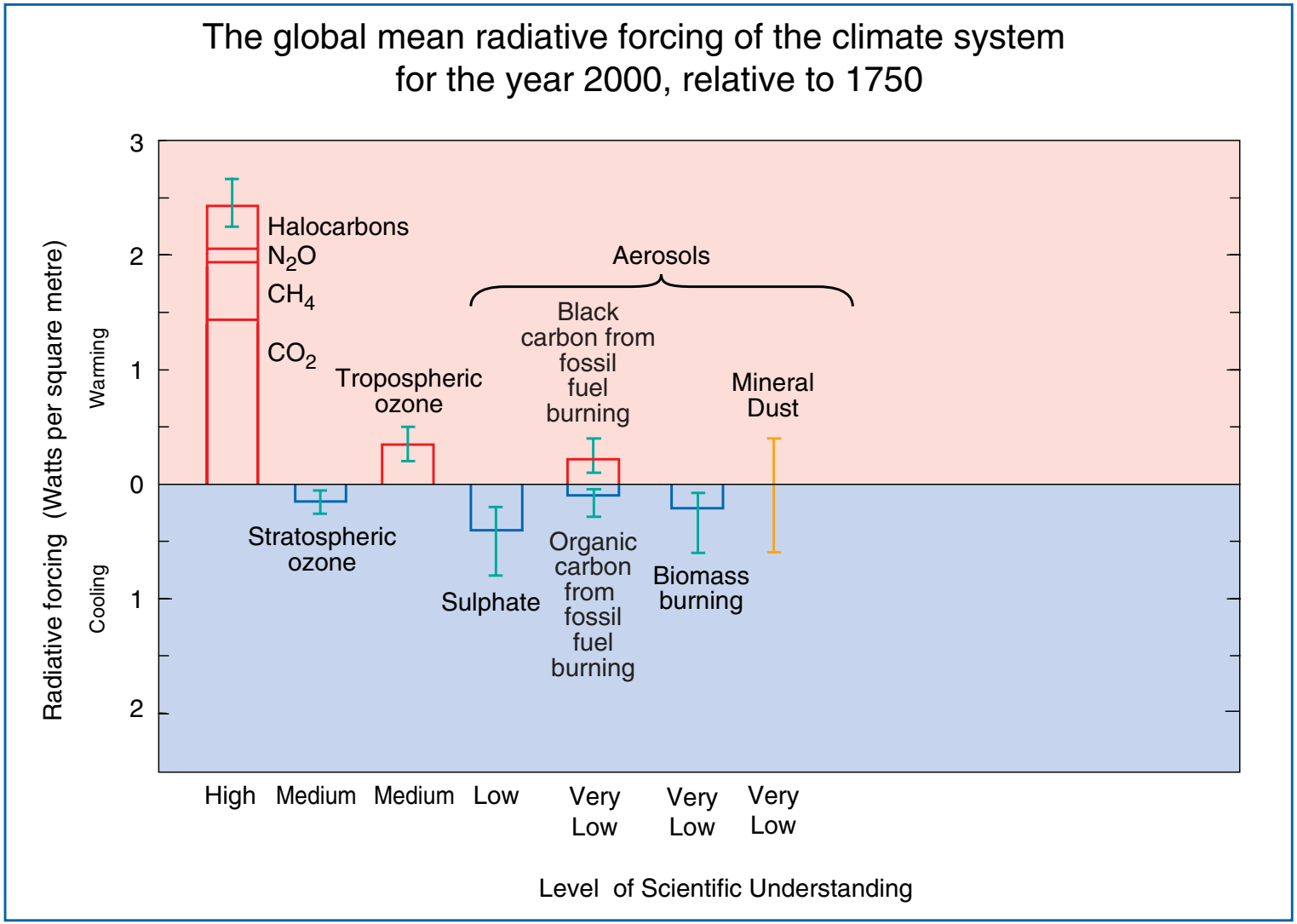


RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)
GHG's and aerosol direct effects



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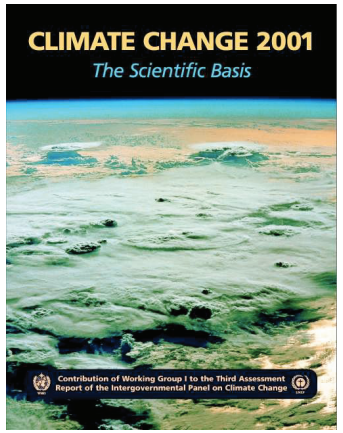
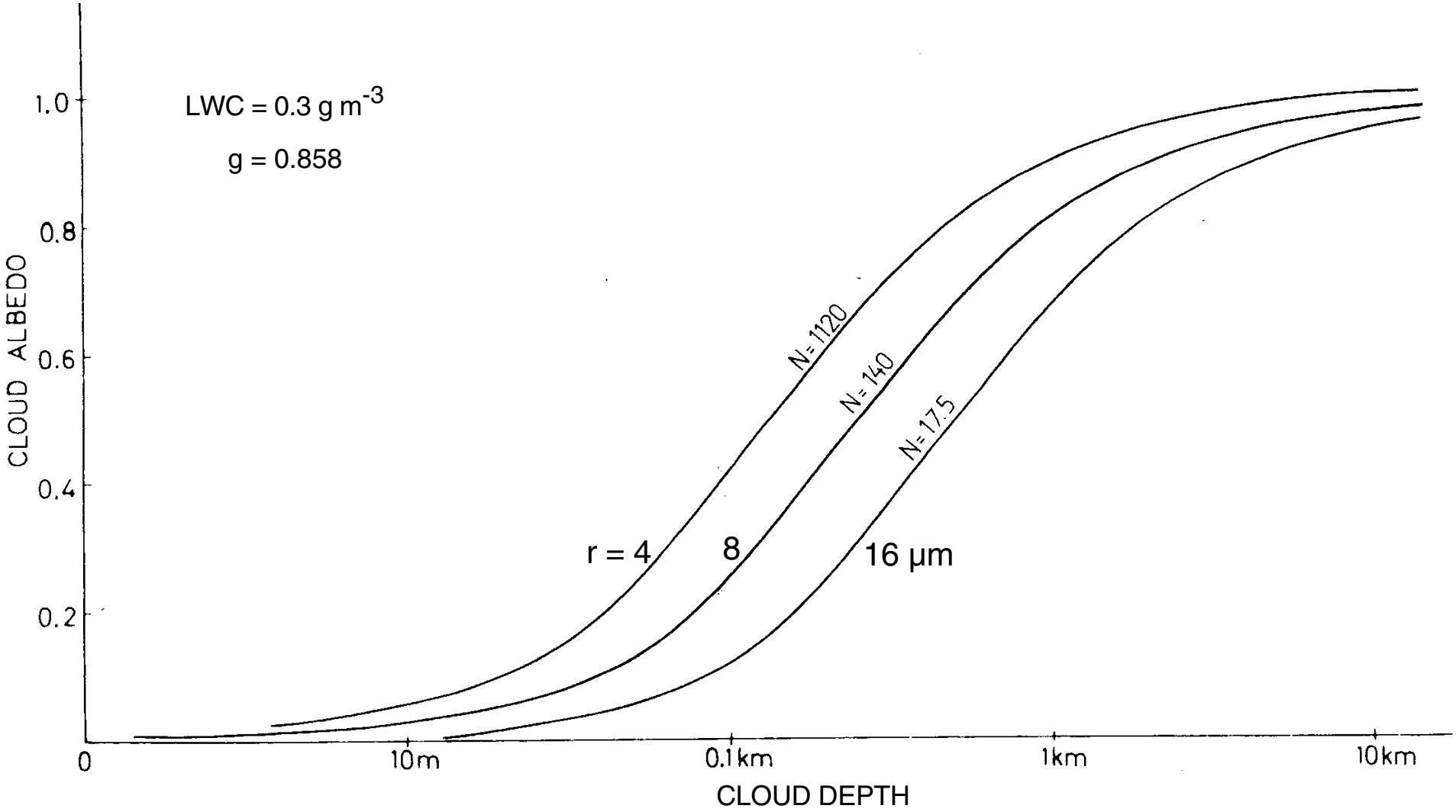


INDIRECT EFFECT

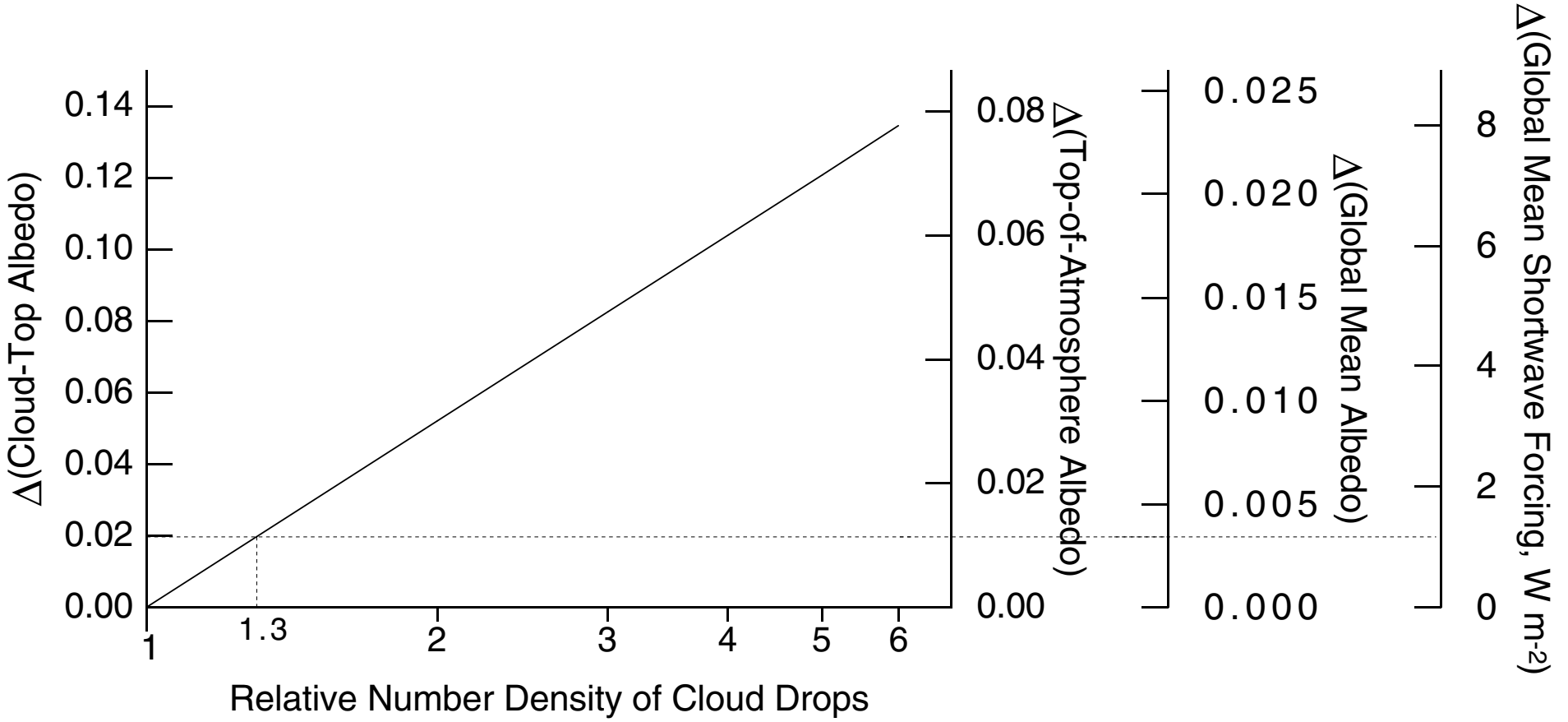
DEPENDENCE OF CLOUD ALBEDO ON CLOUD DEPTH

Influence of Cloud Drop Radius and Concentration



Twomey, *Atmospheric Aerosols*, 1977

SENSITIVITY OF ALBEDO AND FORCING TO CLOUD DROP CONCENTRATION

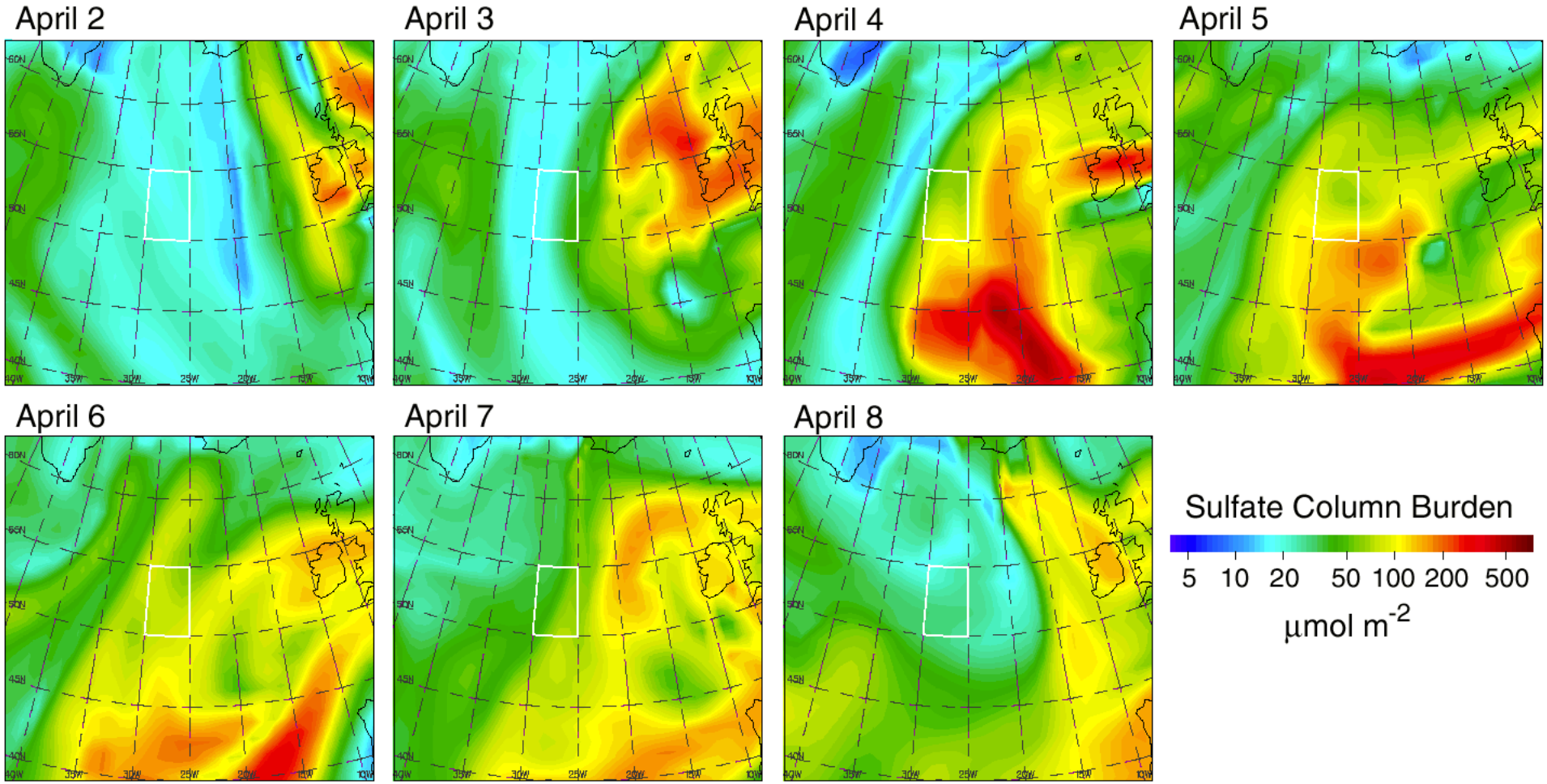


Schwartz and Slingo (1996)

MODELED SULFATE COLUMN BURDEN

$$\int [SO_4^{2-}] dz$$

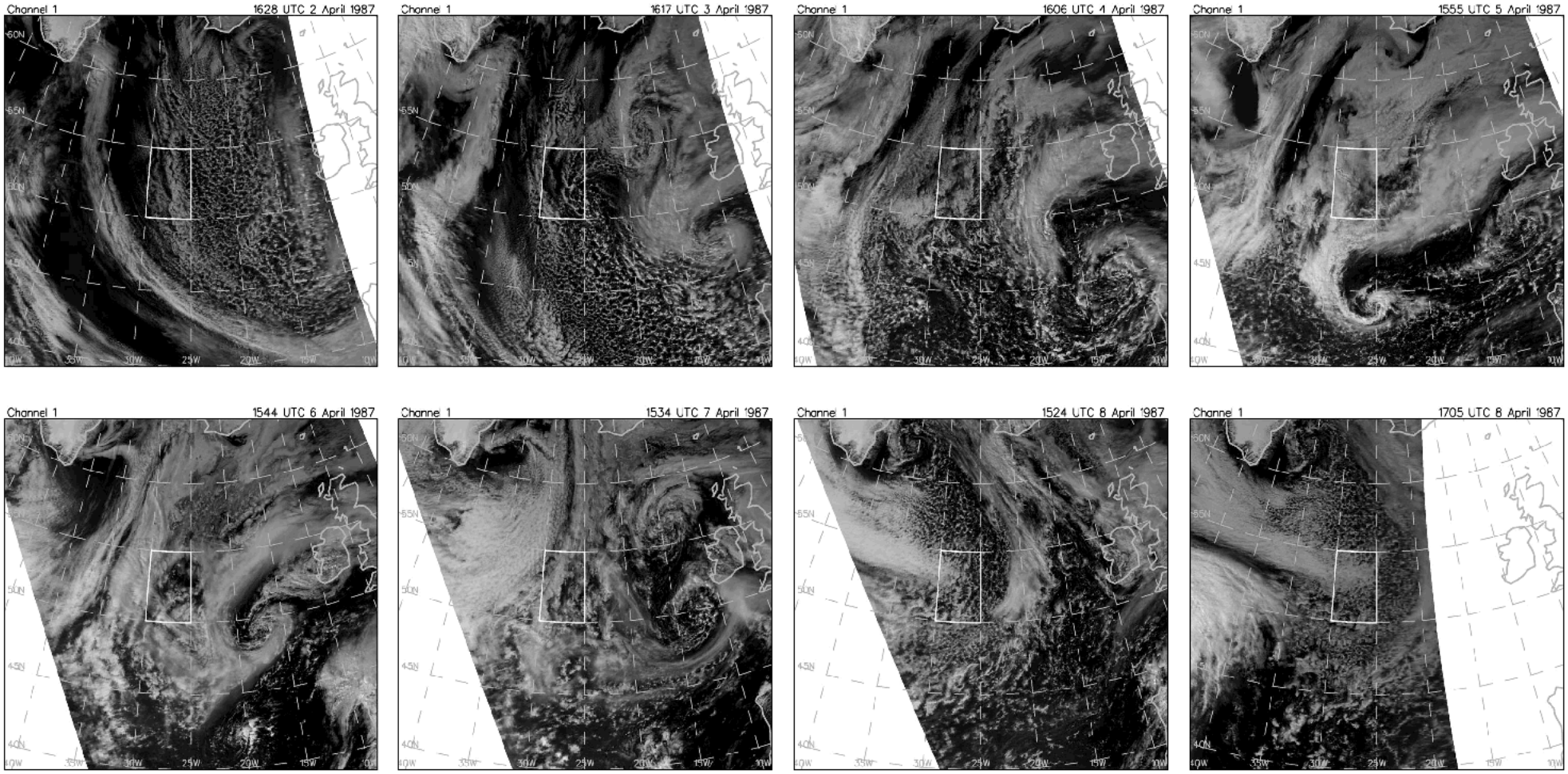
April 2-8, 1987



Schwartz, Harshvardhan &Benkovitz, PNAS, 2002

AVHRR IMAGES APRIL 2-8, 1987

Channel 1, Visible, 0.58-0.68 μm

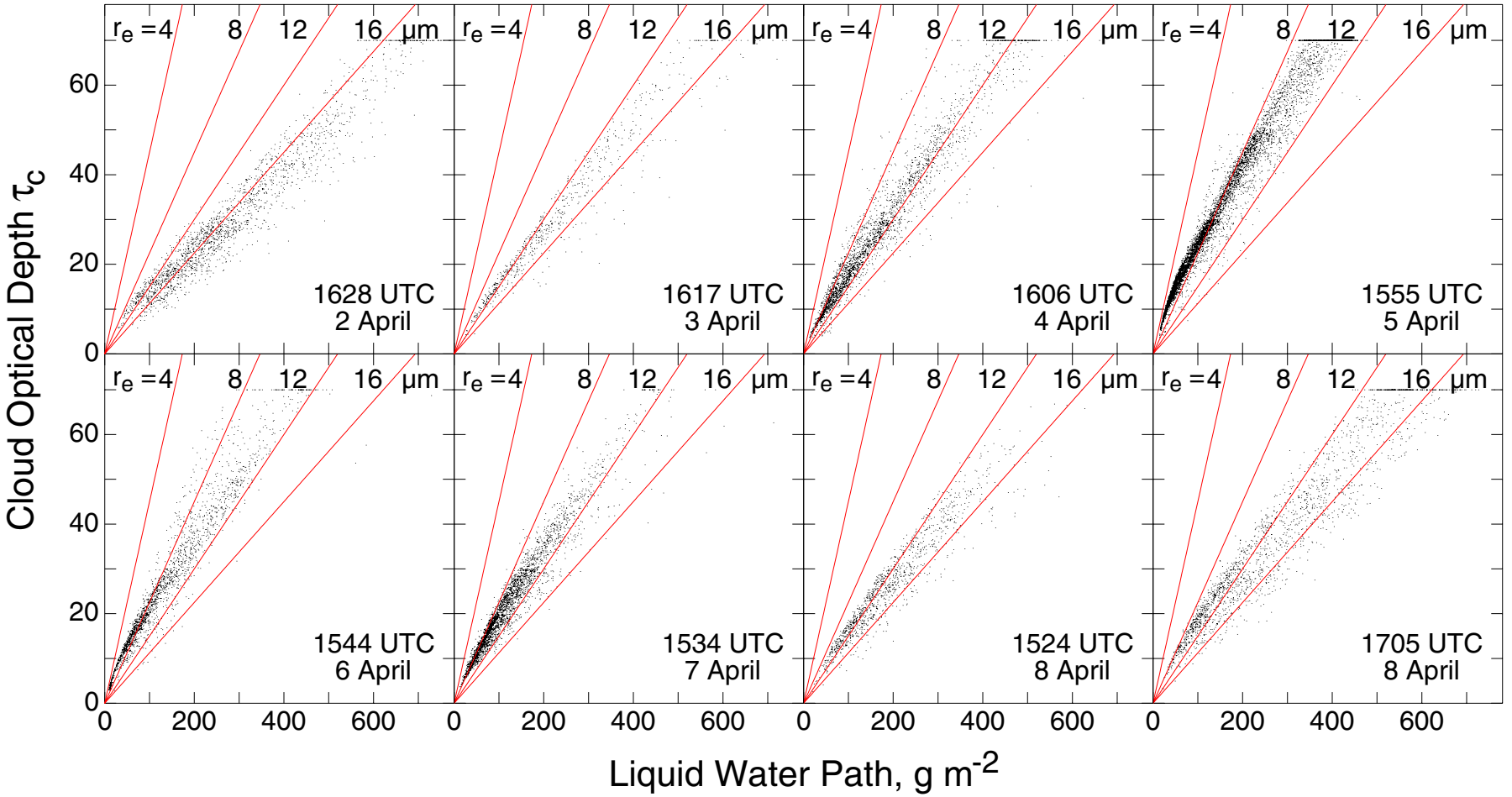


Harshvardhan, Schwartz, Benkovitz and Guo, J Atmos Sci, 2002

CLOUD OPTICAL DEPTH

Dependence on Liquid Water Path

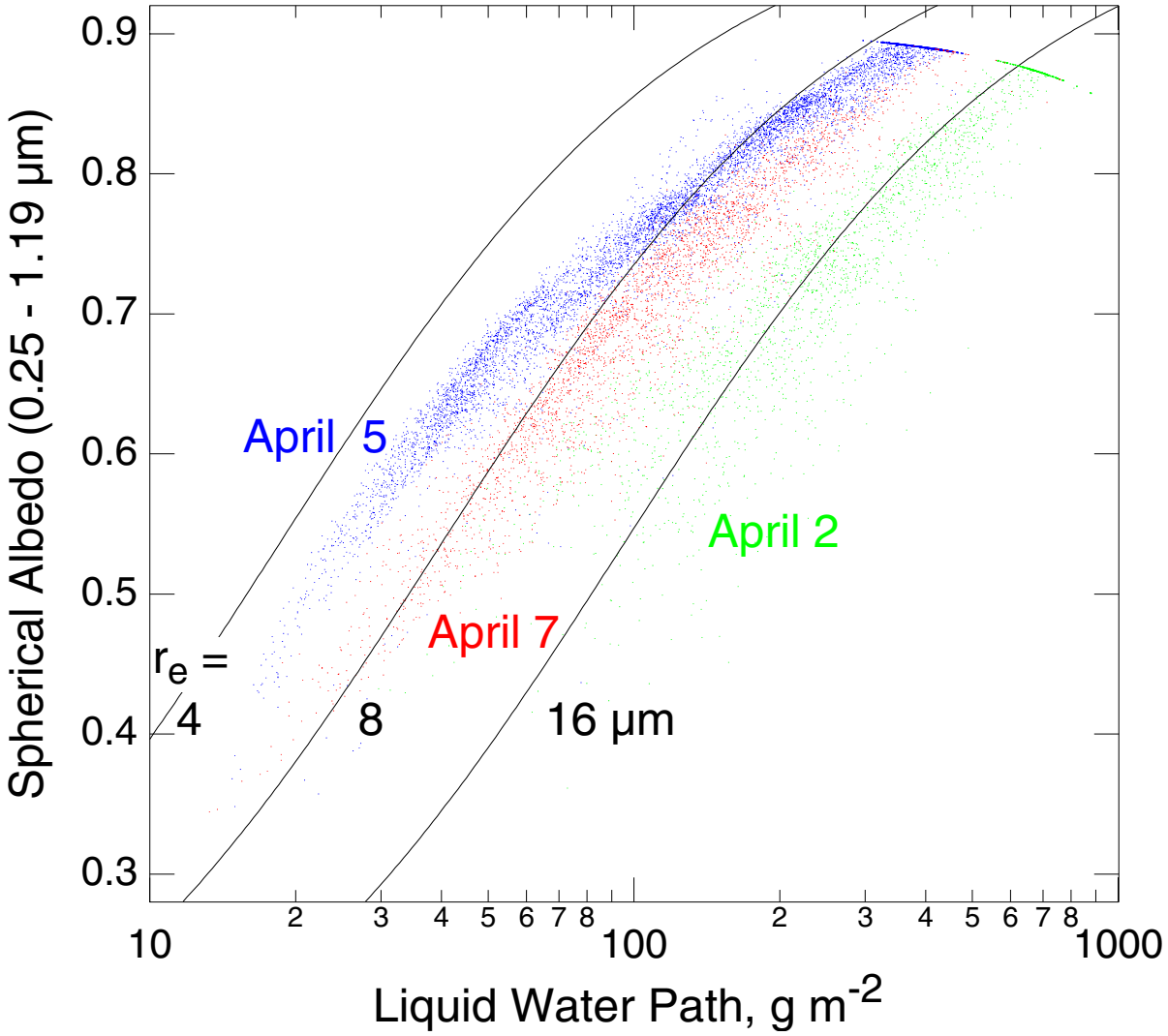
25°-30°W, 50°-55°N April 2-8, 1987



CLOUD-TOP ALBEDO

Dependence on Liquid Water Path

25°-30°W, 50°-55°N April 2, 5 and 7,1987



Schwartz, Harshvardhan & Benkovitz, PNAS, 2002

SULFATE COLUMN BURDEN, CLOUD PROPERTIES AND INDIRECT FORCING

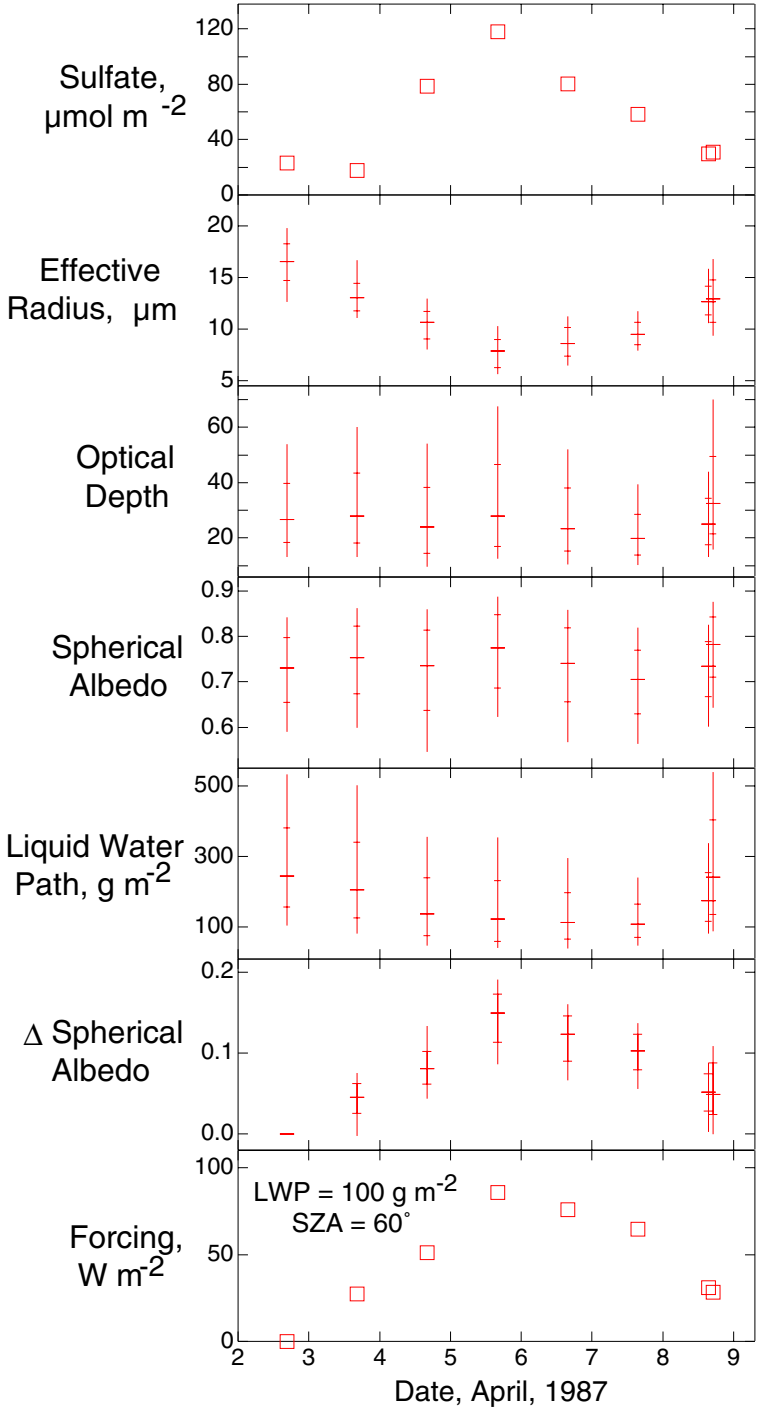
Mid North Atlantic (25-30°W, 50-55°N), April 2-8, 1987

Sulfate from chemical transport model
(Benkovitz et al., *JGR*, 1997)

Cloud drop effective radius and cloud optical depth from satellite retrievals
(Harshvardhan et al., *JAS*, 2002)

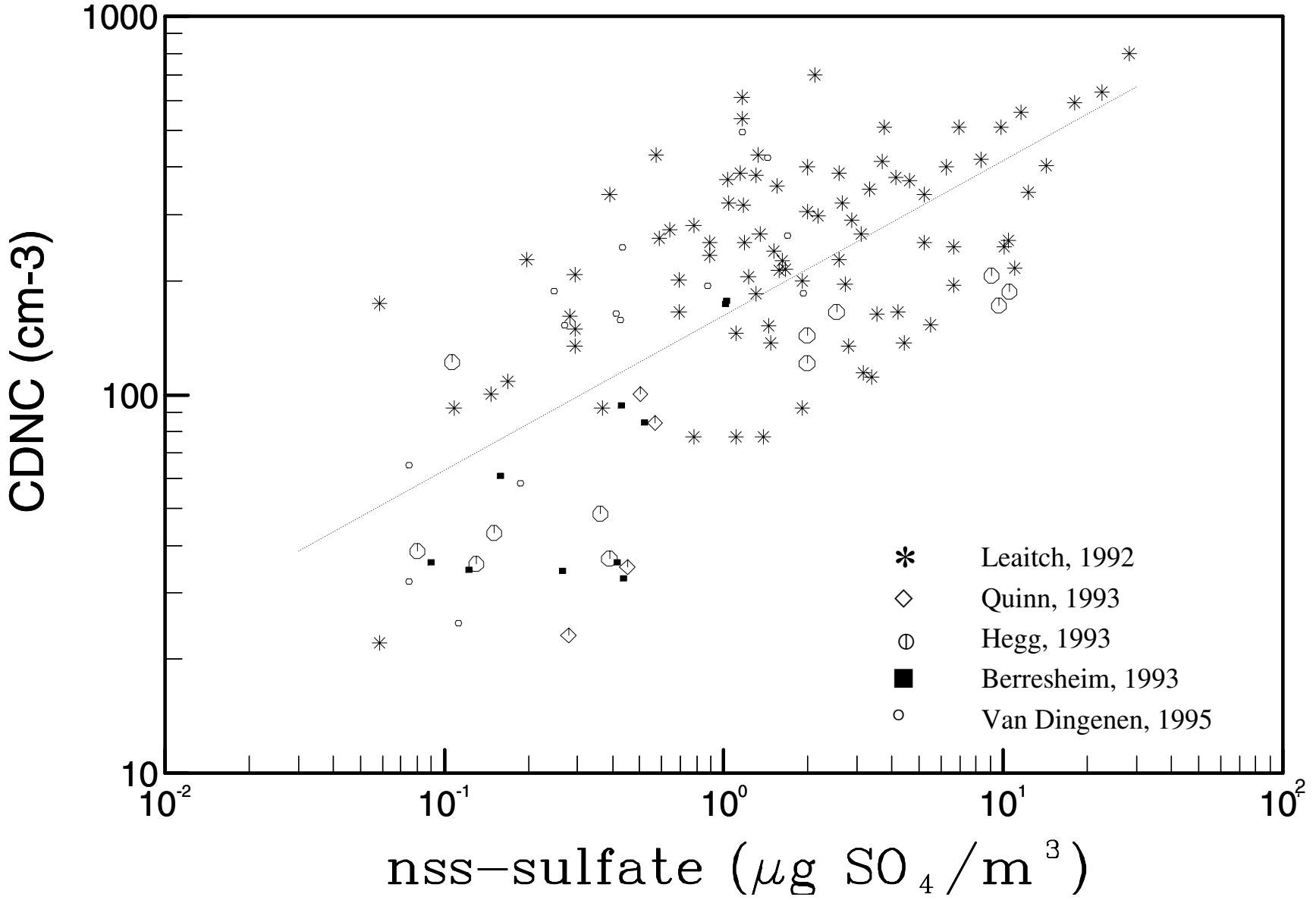
Δ spherical albedo is calculated relative to median effective radius on April 2 (16.5 μm) for retrieved LWP
(Schwartz et al., *PNAS*, 2002)

Forcing is calculated for median effective radius relative to April 2; solar zenith angle 60°; LWP 100 g m^{-2}



CLOUD DROPLET NUMBER CONCENTRATION

Dependence on Non-Seasalt Sulfate

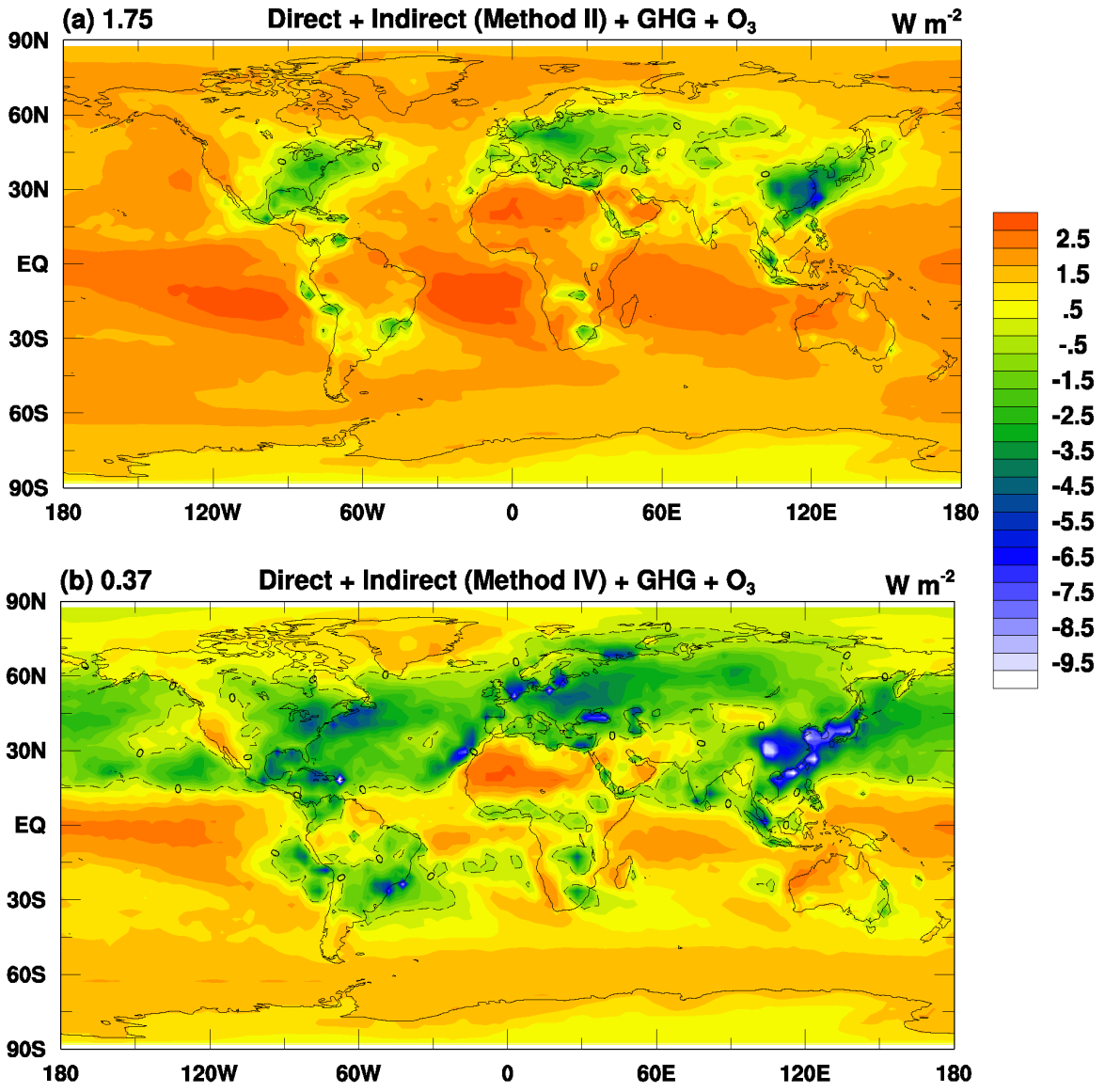


Boucher and Lohmann, 1995

SHORTWAVE FORCING, ANNUAL AVERAGE

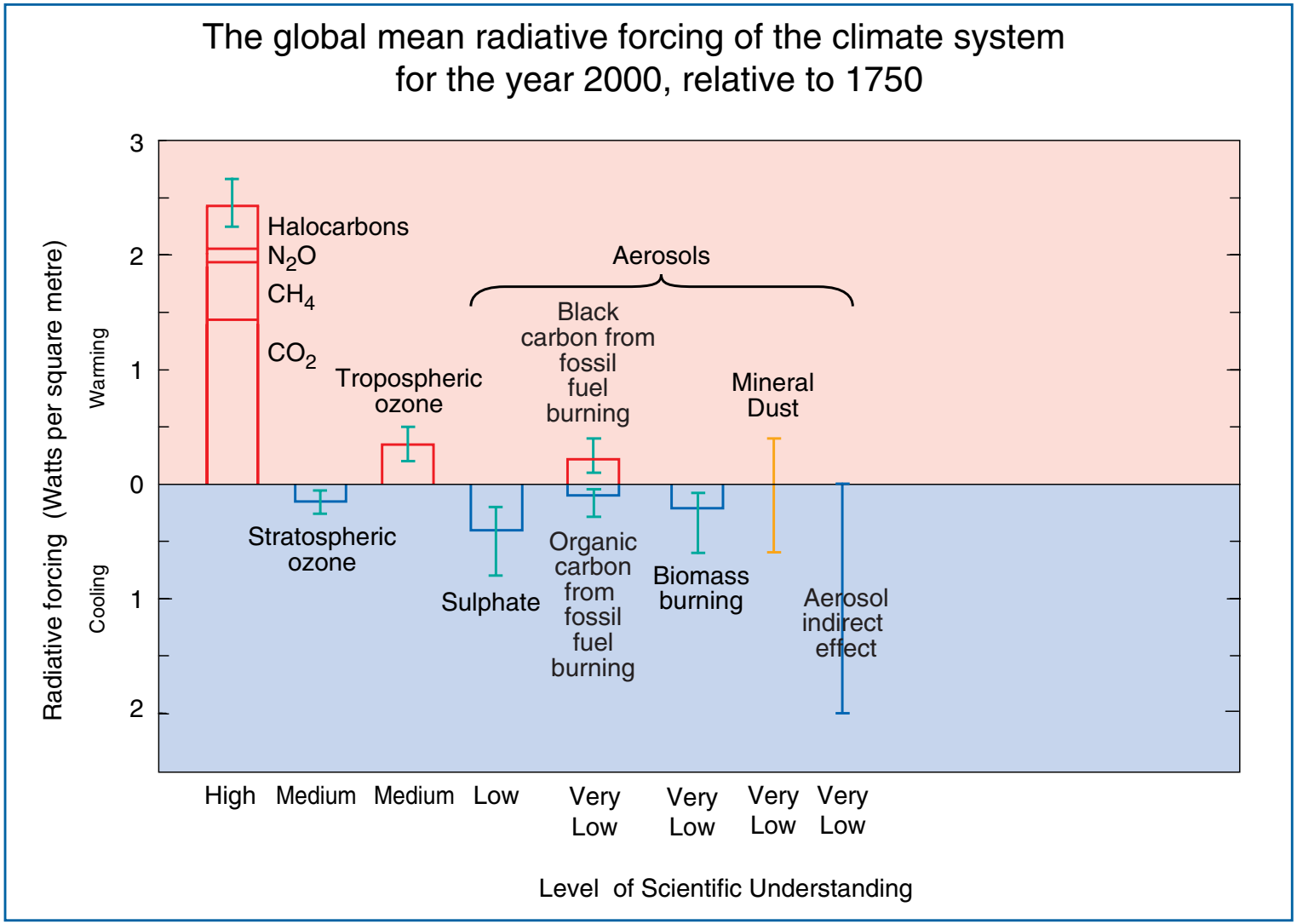
GHG's + O_3 + Sulfate (Direct and Indirect)

Two Formulations of Cloud Droplet Concentration



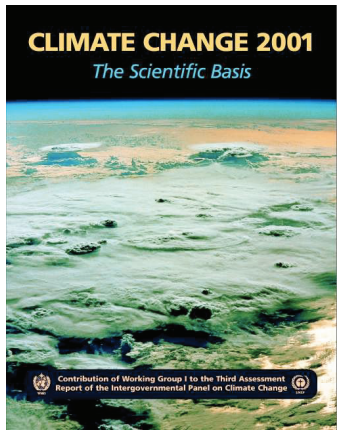
Kiehl et al., JGR, 2000

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



WHY SO LARGE UNCERTAINTY IN AEROSOL FORCING?

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

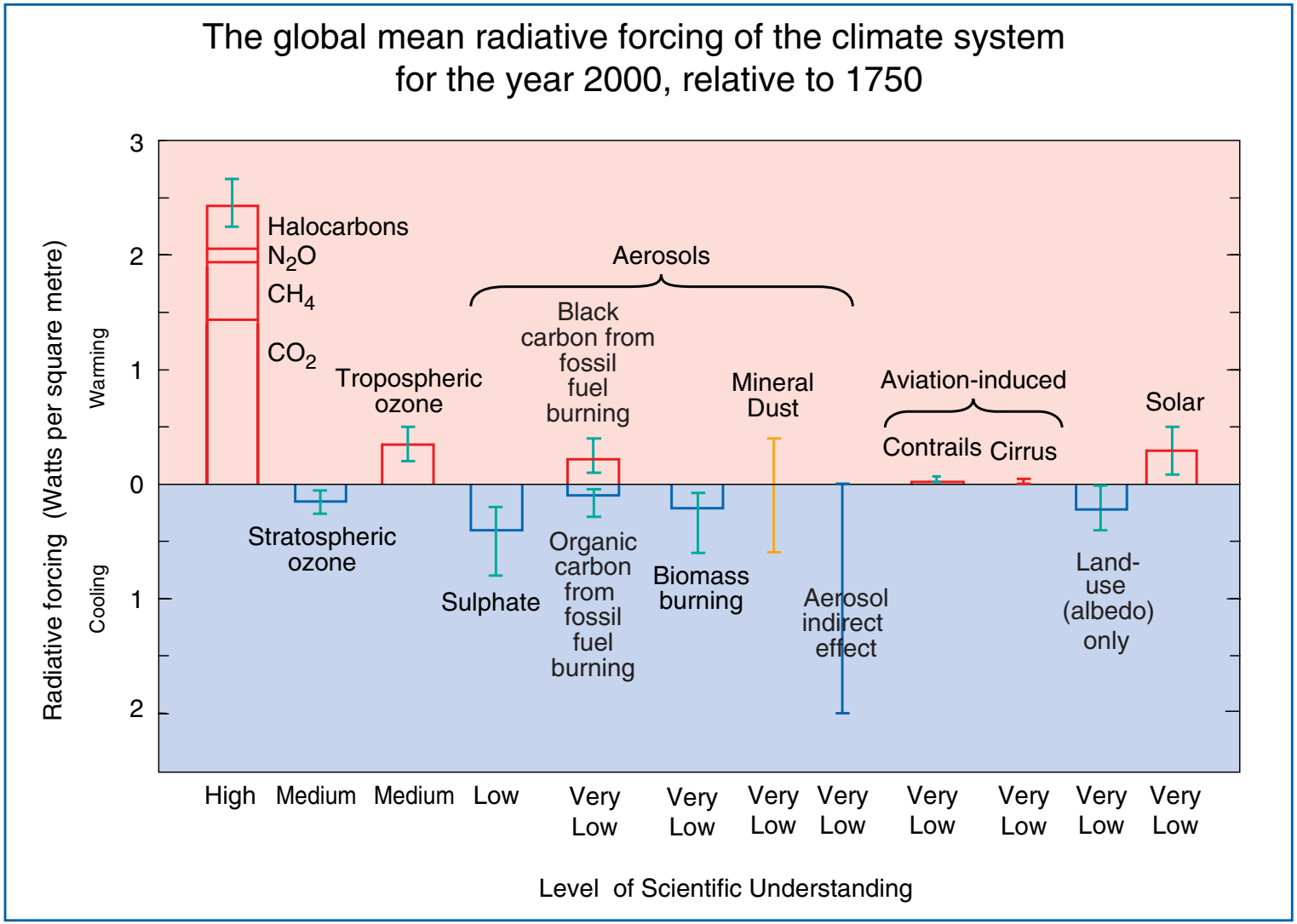
  Mass loading and chemical and microphysical properties and cloud nucleating properties of anthropogenic aerosols, and geographical distribution.

  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.

RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)



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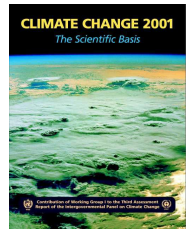
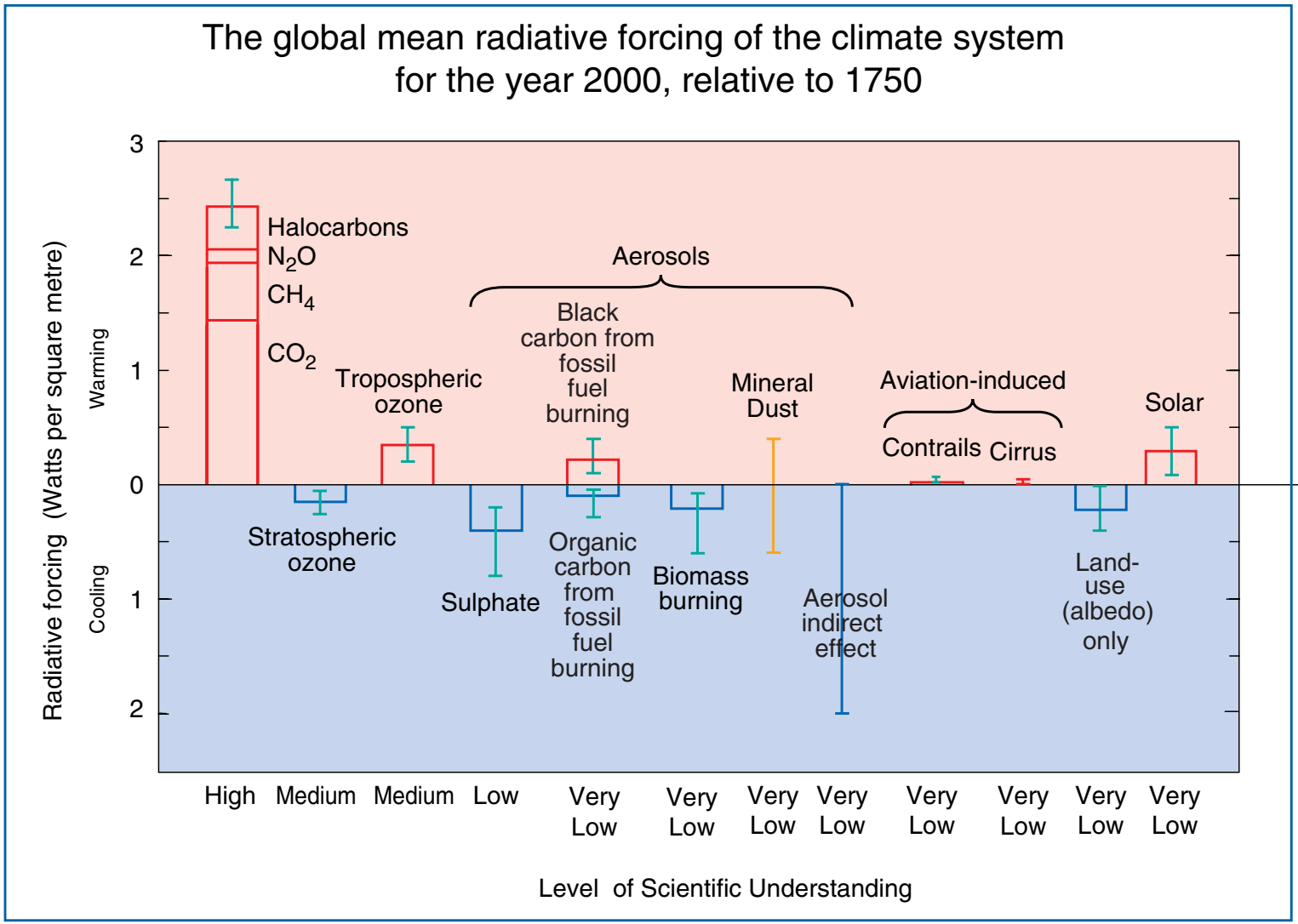
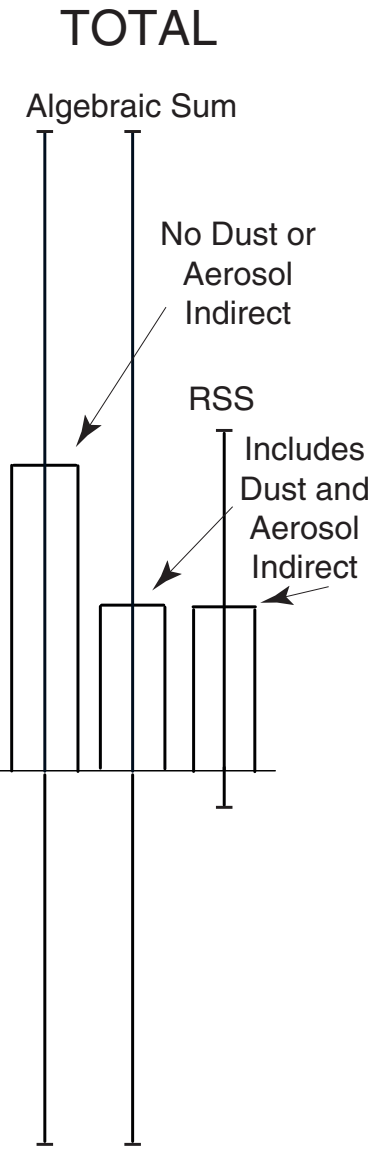


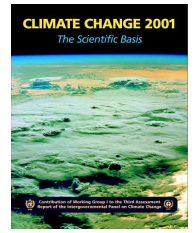
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)
With totals and overall uncertainties by 3 approaches





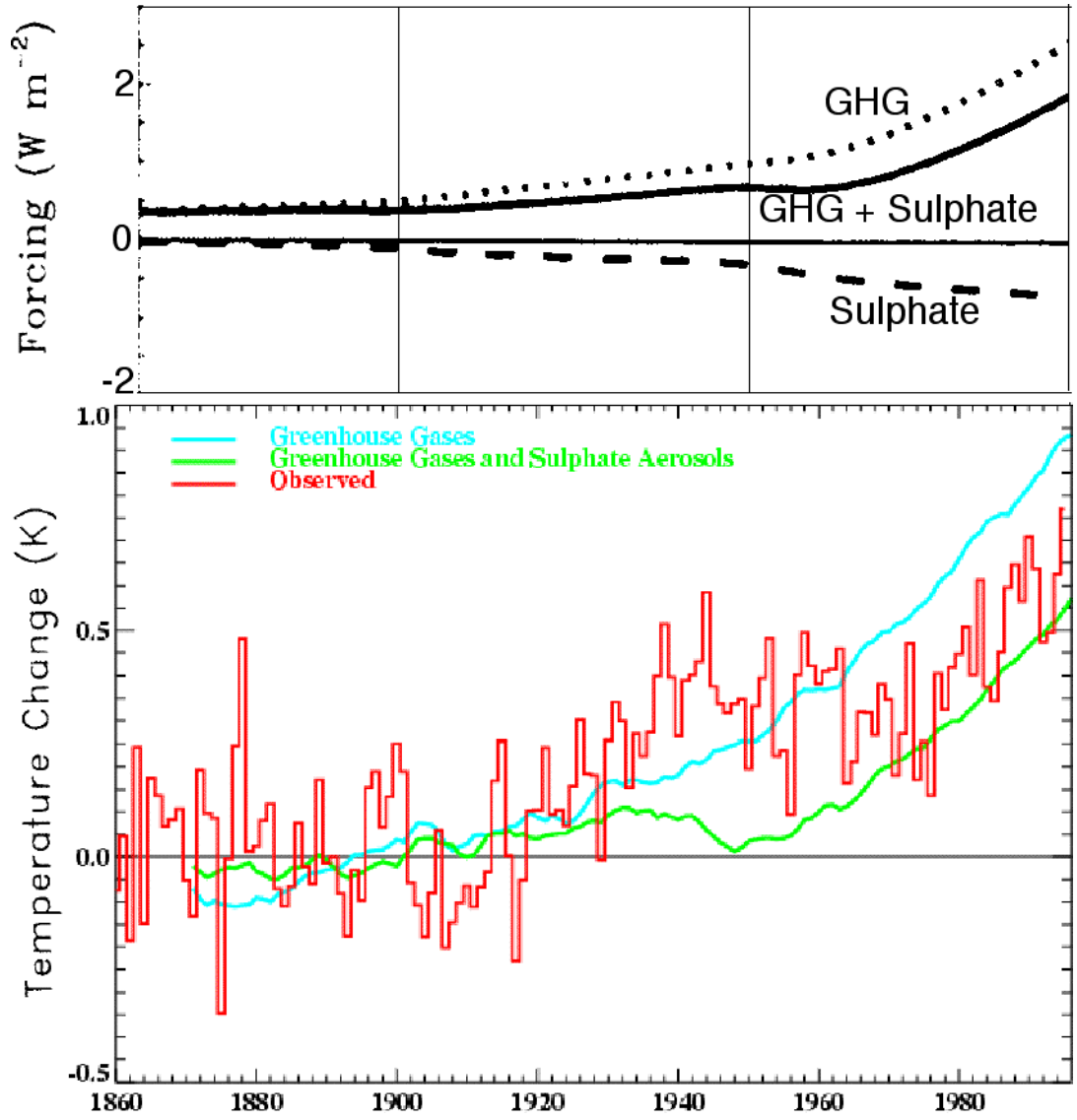
Summary for Policymakers

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



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

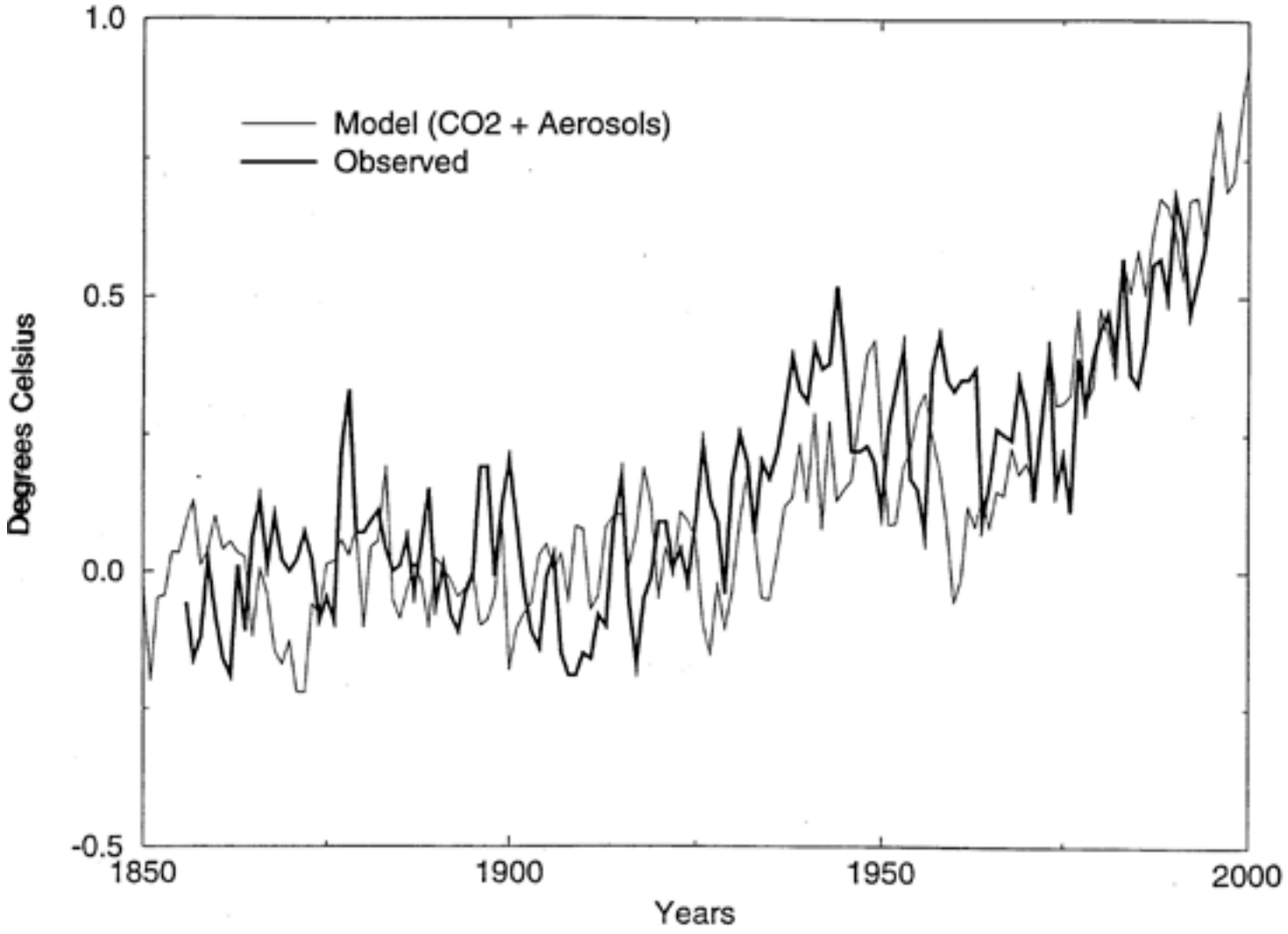
Model sensitivity = 2.5 K per CO_2 doubling; sulfate direct forcing only, -0.6 W m^{-2} (1990)



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

CLIMATE RESPONSE IN THE GFDL MODEL (1997)

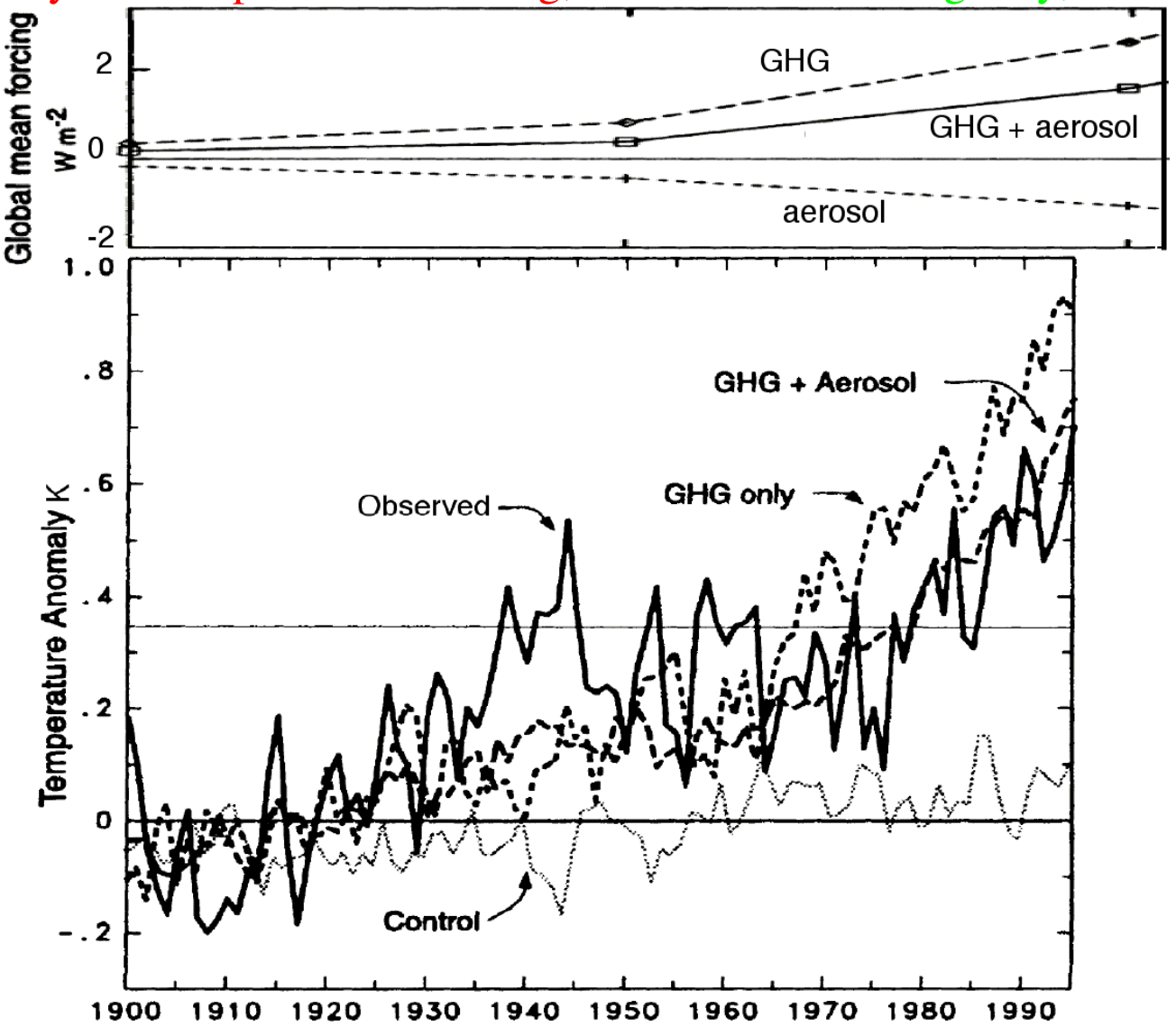
Model sensitivity = 3.7 K per CO_2 doubling; sulfate direct forcing only, -0.6 W m^{-2} (1990)



"The global average SAT trend from the model [is] in ***reasonable agreement*** with the observations." -- *Haywood, Ramaswamy et al., Geophys. Res. Lett, 1997*

FORCING AND RESPONSE IN THE CANADIAN CLIMATE MODEL (2000)

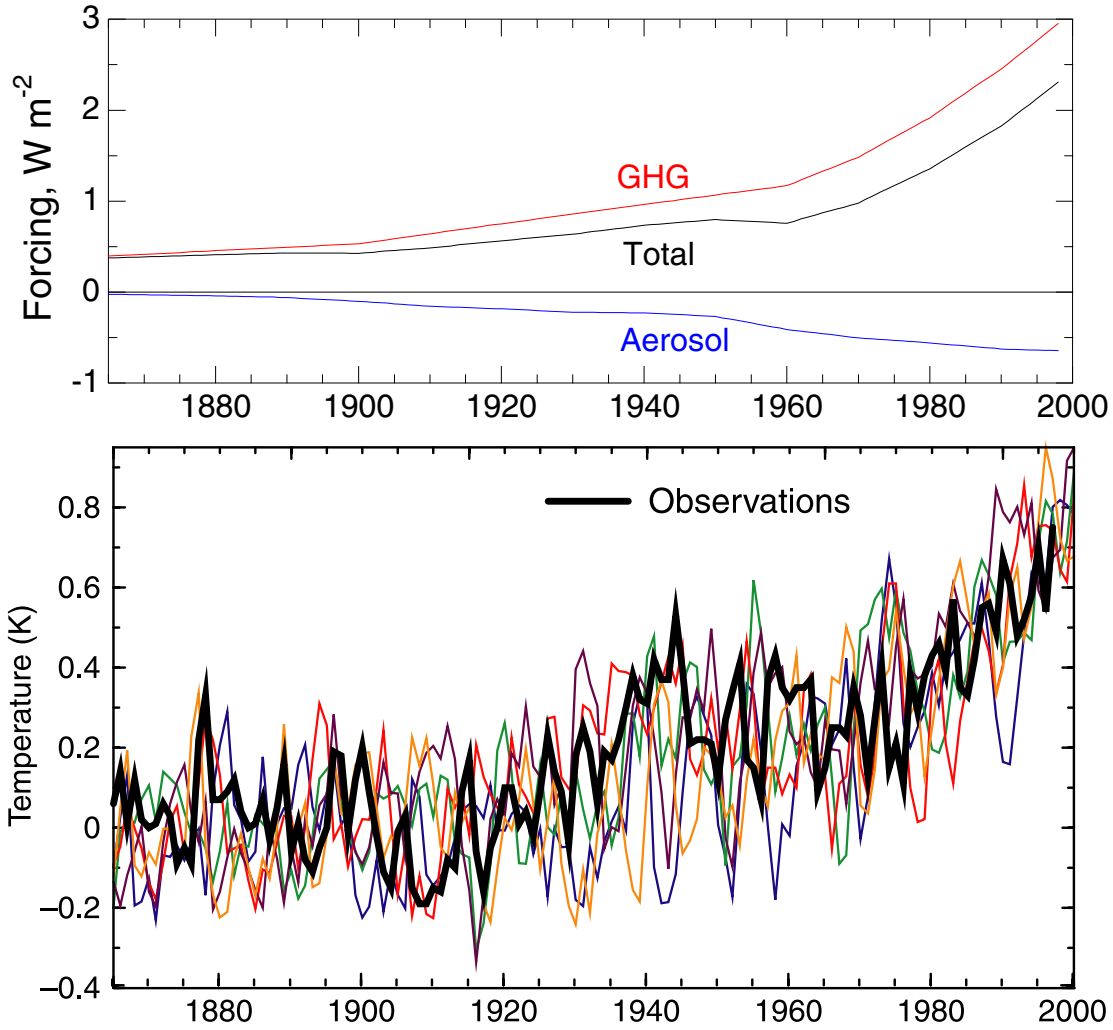
Model sensitivity = 3.5 K per CO_2 doubling; sulfate direct forcing only, -1.0 W m^{-2} (1990)



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

CLIMATE RESPONSE IN THE GFDL MODEL (2000)

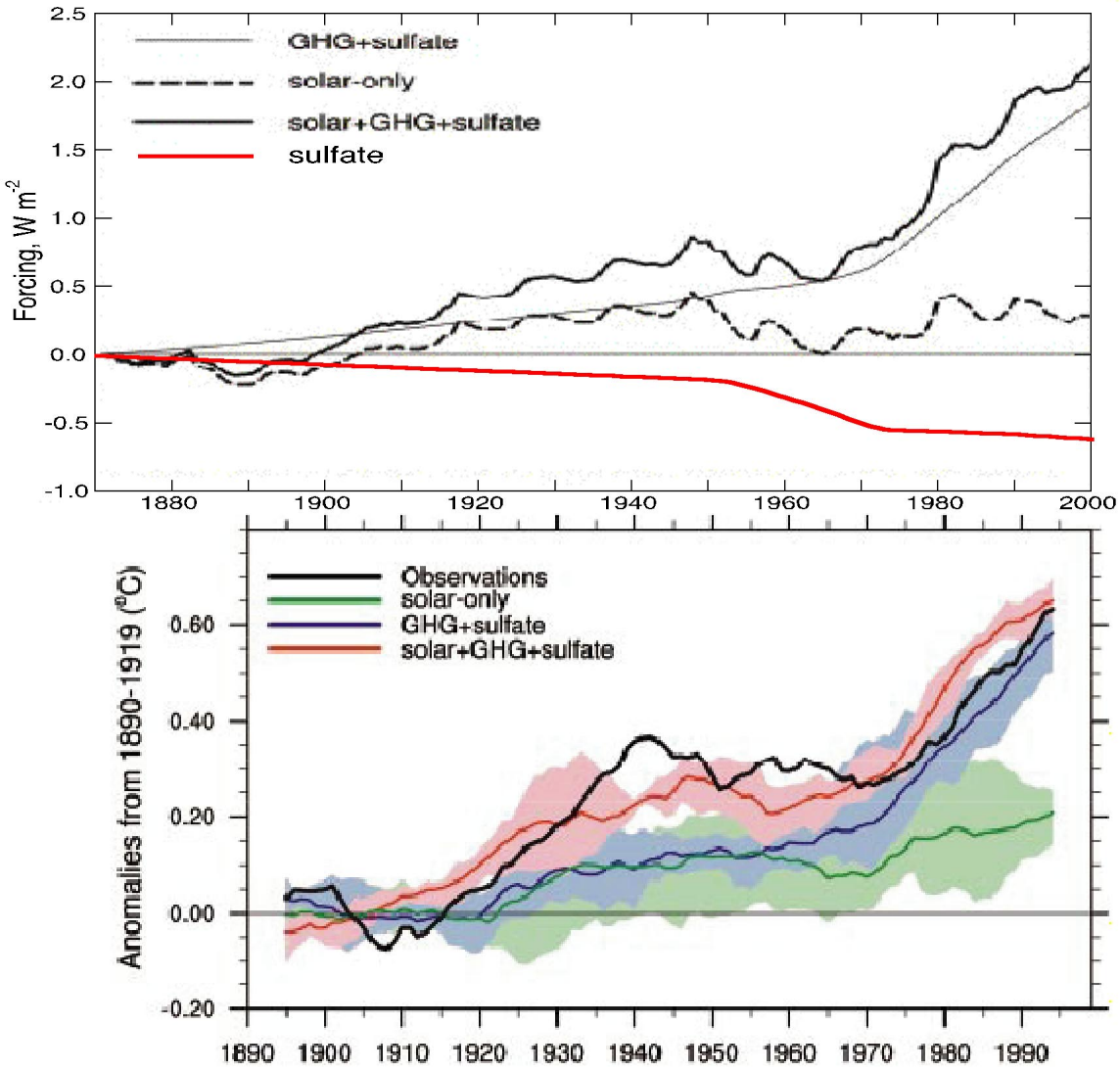
Model sensitivity = 3.4 K per CO_2 doubling; sulfate forcing, -0.62 W m^{-2} (1990)



"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)

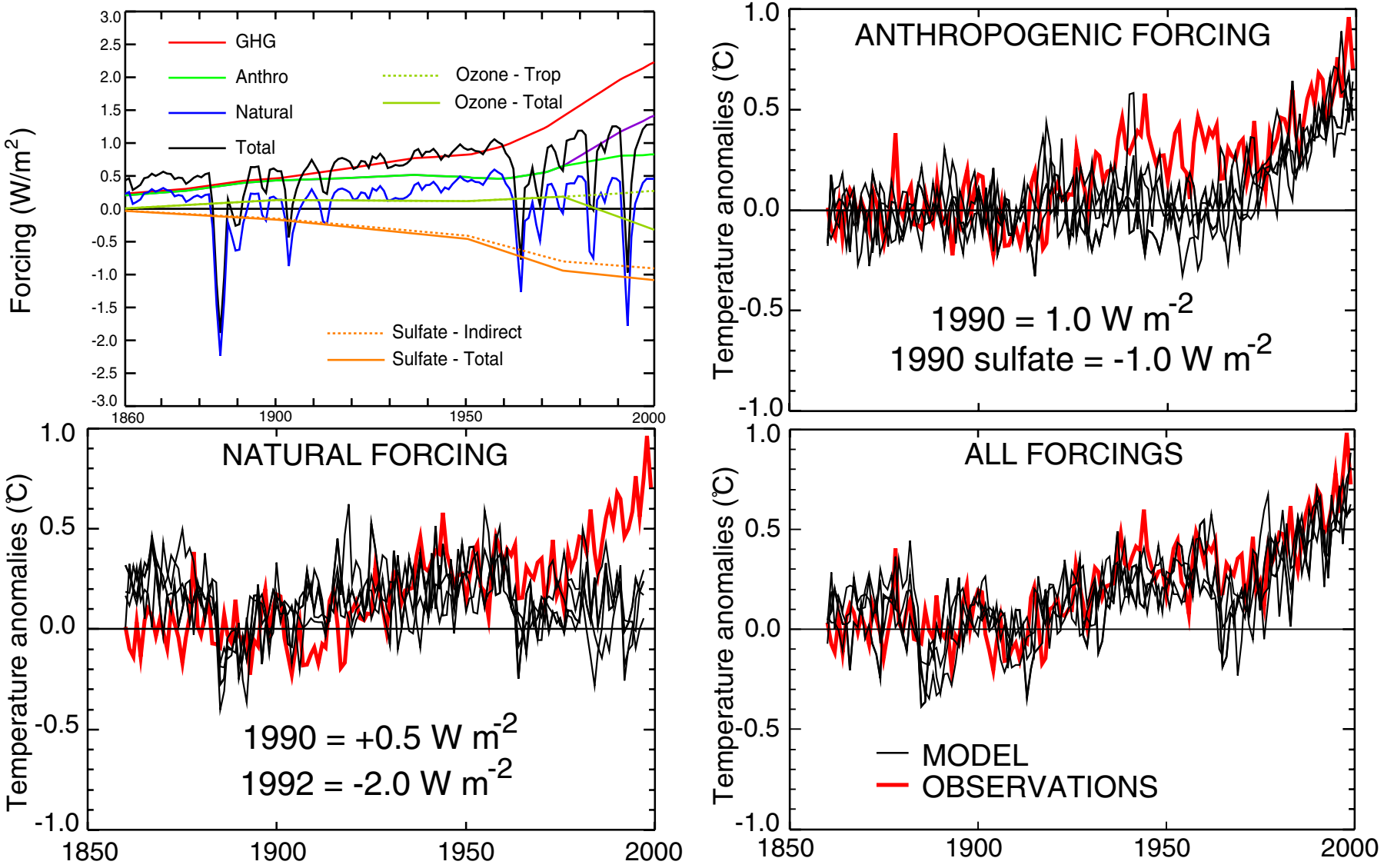
Model sensitivity = 2.18 K per CO_2 doubling; sulfate direct forcing only, -0.6 W m^{-2} (1990)



"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)

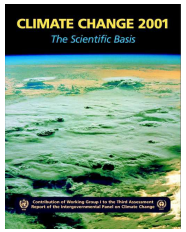
Model sensitivity = 3.45 K per CO_2 doubling; sulfate + indirect forcing, -1.1 W m^{-2} (1990)



"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 of these studies find that, over the last 50 years, the estimated rate and magnitude of warming due to increasing concentrations of greenhouse gases alone are comparable with, or larger than, the observed warming. ***Furthermore, most model estimates that take into account both greenhouse gases and sulphate aerosols are consistent with observations over this period.***



OUR SIMULATIONS THAT INCLUDE ESTIMATES OF NATURAL AND ANTHROPOGENIC FORCING REPRODUCE THE OBSERVED LARGE-SCALE CHANGES IN SURFACE TEMPERATURE OVER THE 20TH CENTURY.
BUT MOM, DON'T THE GCM CALCULATIONS REQUIRE ACCURATE ESTIMATES OF FORCING?
SHHHH!! THE EMPEROR MIGHT HEAR YOU.
IPCC GCM's
WMO
UNEP
The Emperor's New Clothes

UNCERTAINTY PRINCIPLES

Climate sensitivity $\lambda = \Delta T / F$

The fractional uncertainty in climate sensitivity λ is evaluated from fractional uncertainties in temperature change ΔT and forcing F as:

$$\frac{\delta\lambda}{\lambda} = \sqrt{\left(\frac{\delta\Delta T}{\Delta T}\right)^2 + \left(\frac{\delta F}{F}\right)^2}$$

A reasonable target uncertainty might be:

$$\frac{\delta\lambda}{\lambda} = 30\%, \textit{e.g.,}\ \Delta T_{2\times CO_2} = (3 \pm 1)\ \mathrm{K}$$

This would require uncertainties in temperature anomaly and forcing:

$$\frac{\delta\Delta T}{\Delta T} \approx \frac{\delta F}{F} \approx 20\%.$$

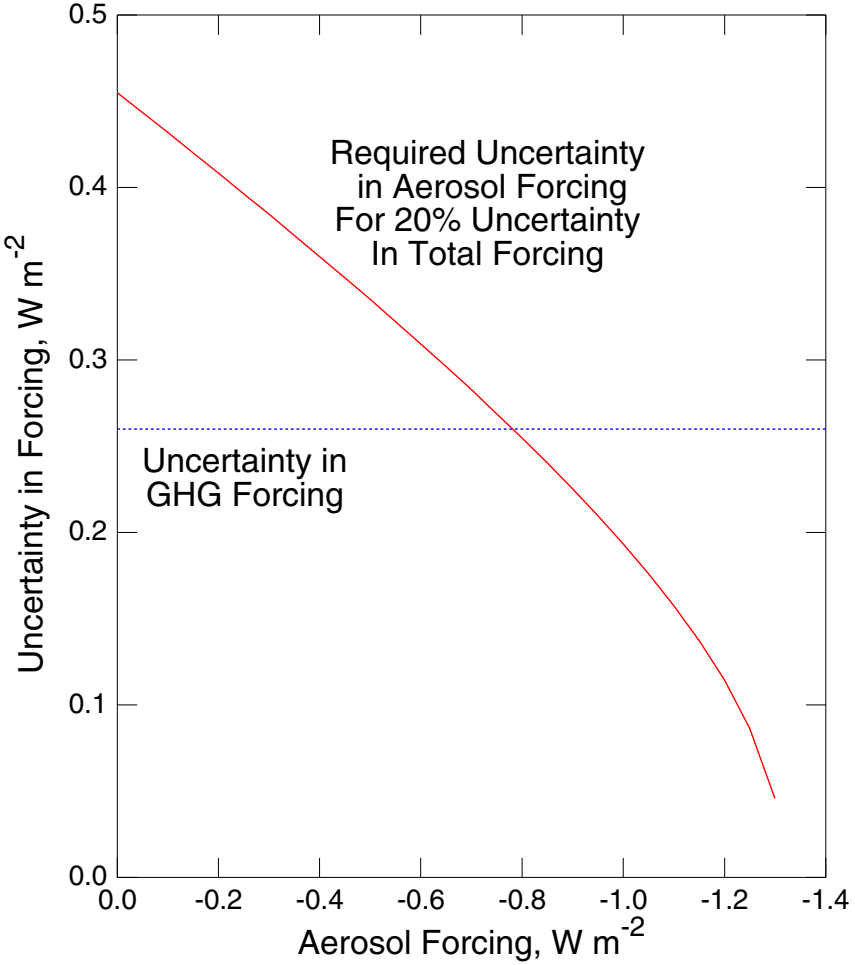
This imposes ***stringent requirements on uncertainty in aerosol forcing!***

REQUIRED UNCERTAINTY IN AEROSOL FORCING

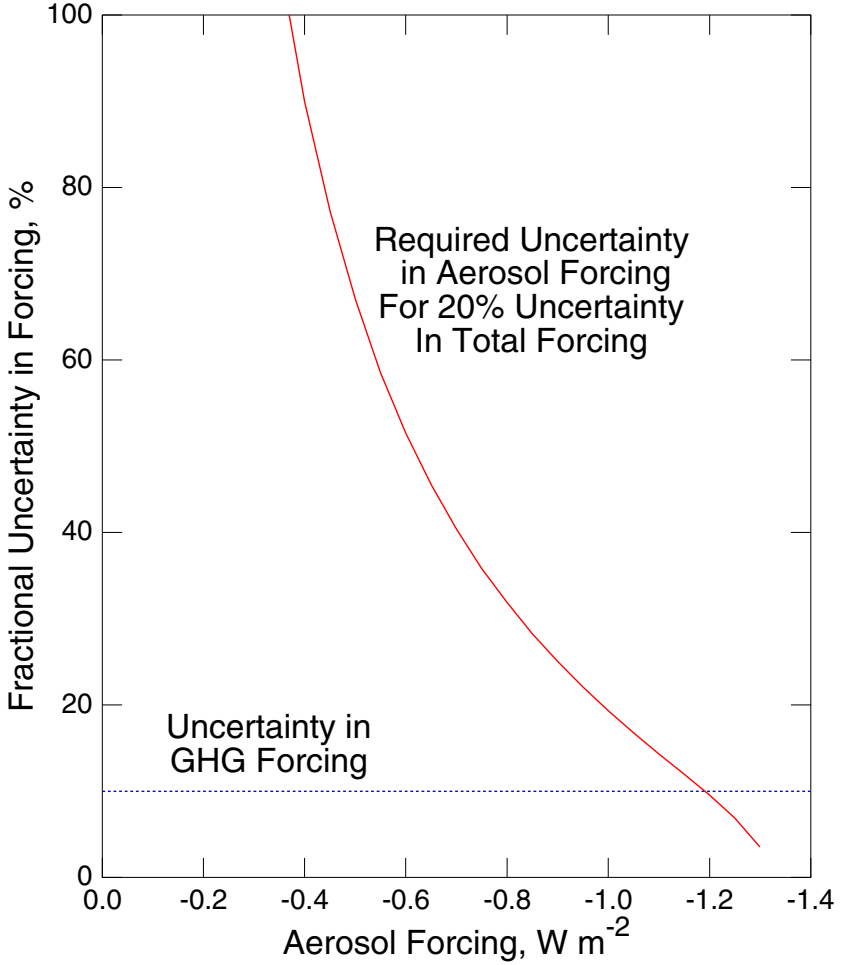
Uncertainty in total forcing not to exceed 20%

GHG Forcing (well mixed gases + strat and trop O_3) = 2.6 W m^{-2} ± 10%

Absolute, W m^{-2}



Percent



KEY REQUIREMENTS FOR FUTURE RESEARCH

- ***Abundance, composition, and mixing state and optical and cloud-nucleating properties of atmospheric aerosols as a function of location and time***

  Observation

  - *In-situ* measurements.
  - Ground-based and satellite-based remote sensing.

  Chemical transport modeling

  - Evaluate by comparison with observation.

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

  Measurement

  - As a function of location and controlling variables.
  - For anthropogenic *and* natural aerosols.

  Develop emission factors and emission inventories

- ***Atmospheric chemical and microphysical transformation processes***

  Laboratory, theory, field measurements and modeling

cont'd . . .

KEY REQUIREMENTS FOR FUTURE RESEARCH (*cont'd*)

- ***Wet and dry removal processes***
  Size and composition dependence.
- ***Representation of aerosols in chemical transport models***
  Mass loading as a function of location and secular time.
  Size-dependent composition and mixing state.
  - Optical properties
  - Hygroscopic properties
  - Cloud nucleating properties
- ***Aerosol-radiation interactions***
  Quantify aerosol influences on short- and longwave radiation in cloud-free skies.
- ***Aerosol - cloud interactions***
  Quantify the effects of changes in aerosol abundance and composition on cloud formation, persistence, and amount, on precipitation amounts, and on cloud radiative properties.
- ***Uncertainties in all the above***

SOME CONCLUDING OBSERVATIONS

- GHG concentrations and forcing are increasing. GHGs persist in the atmosphere for decades to centuries.
- Aerosol forcing is comparable to greenhouse gas forcing but much more uncertain.
- Hence total forcing over the industrial period is highly uncertain.
- Hence the sensitivity of the climate system remains highly uncertain.
- Climate sensitivity will remain uncertain unless and until aerosol uncertainty is substantially decreased.
- Decisions must be made in an uncertain world. (Lack of controls on GHG emissions is also a decision).