AEROSOLS, CLOUDS, AND CLIMATE CHANGE

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Nucleation & Atmospheric Aerosols
Kyoto, Japan on July 26-30, 2004

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

OUTLINE

The global energy balance: Radiative forcing of climate change.

The Twomey effect: Physical basis and sensitivity

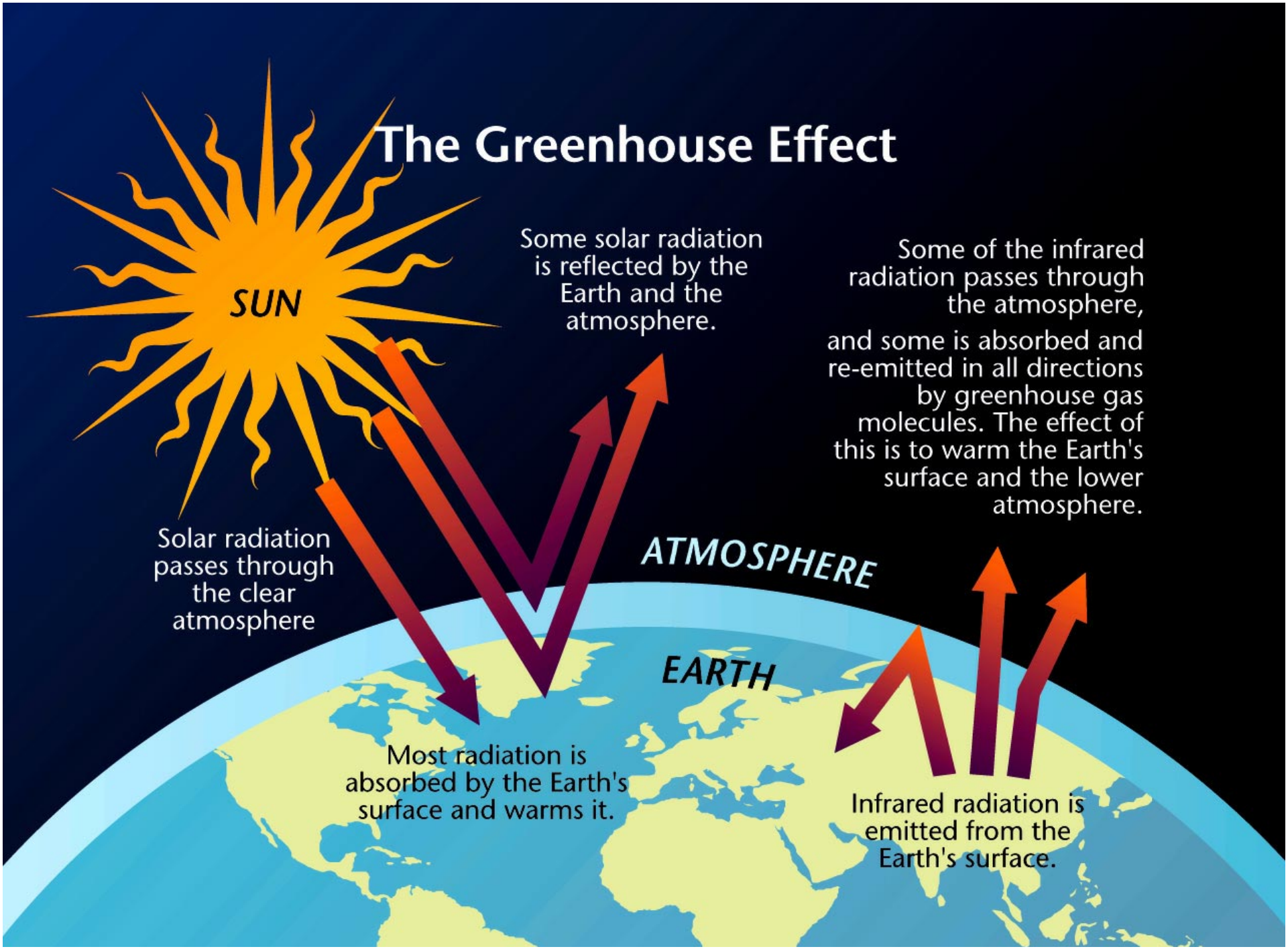
Relation between aerosol concentrations and cloud drop concentrations

Search for indirect effect in interhemispheric comparisons

Using results from a chemical transport model to identify situations of high aerosol loading to pinpoint aerosol indirect effect

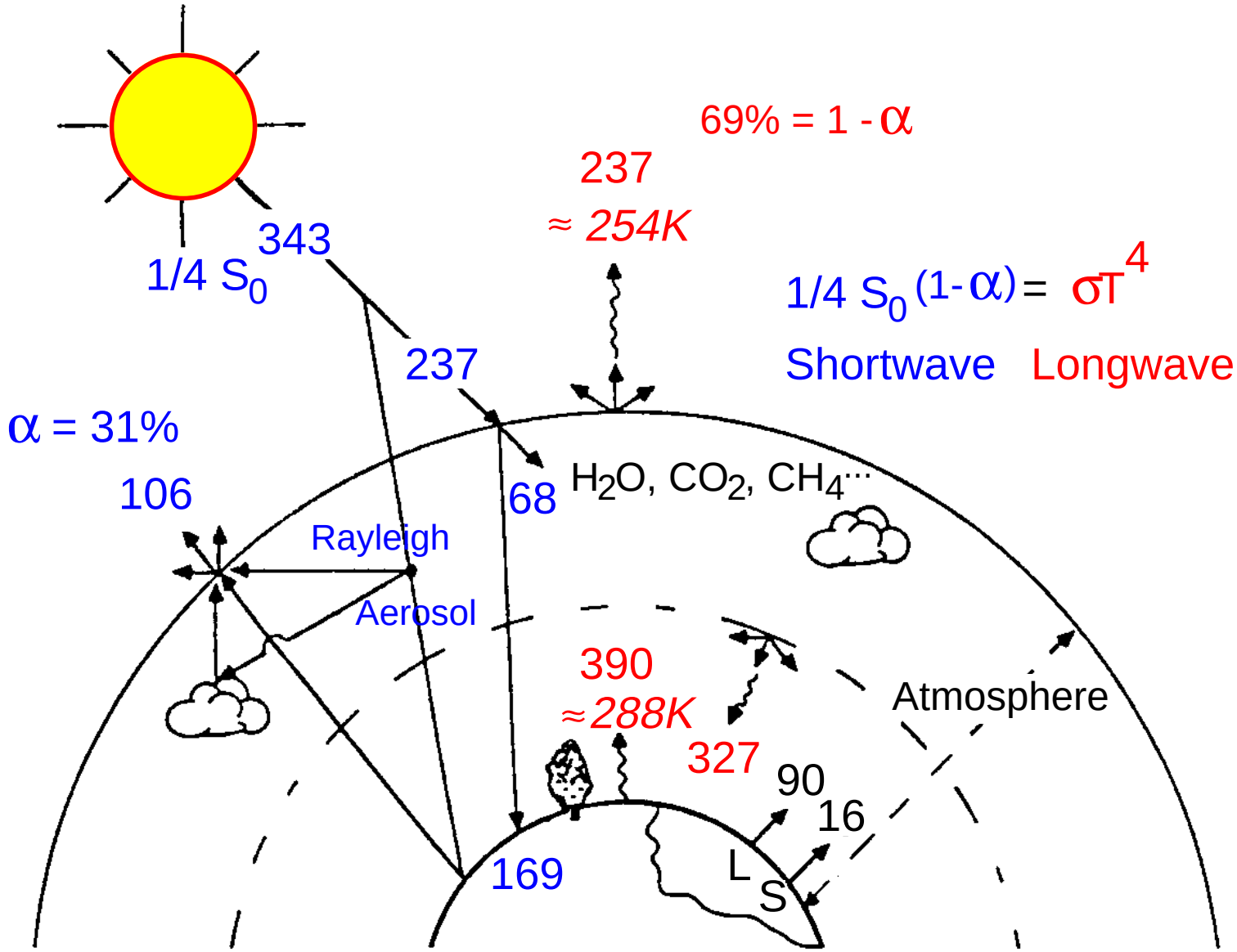
Quantification of aerosol indirect forcing in ground-based remote sensing

Concluding remarks


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

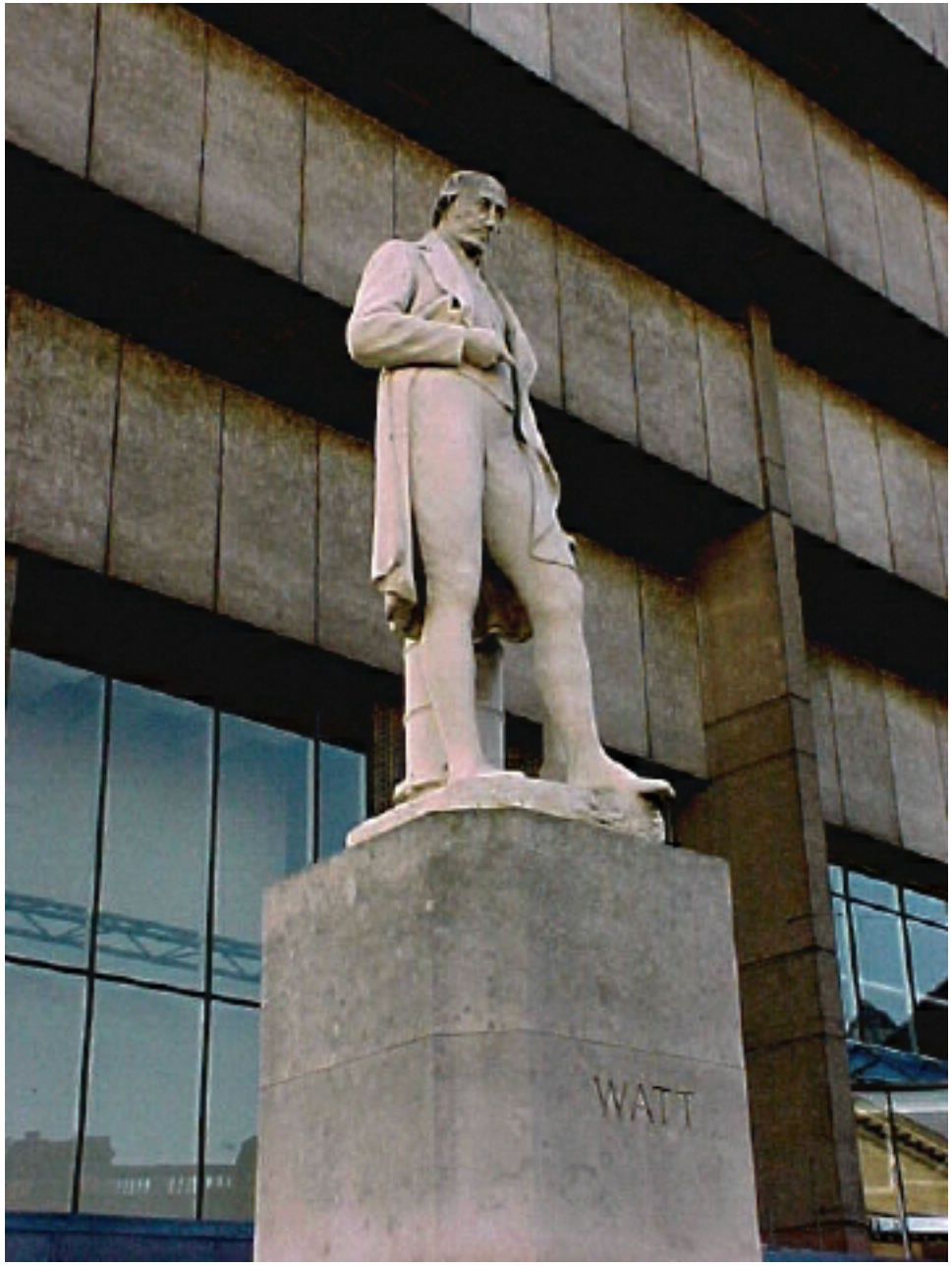
Energy per area per time

Power per area

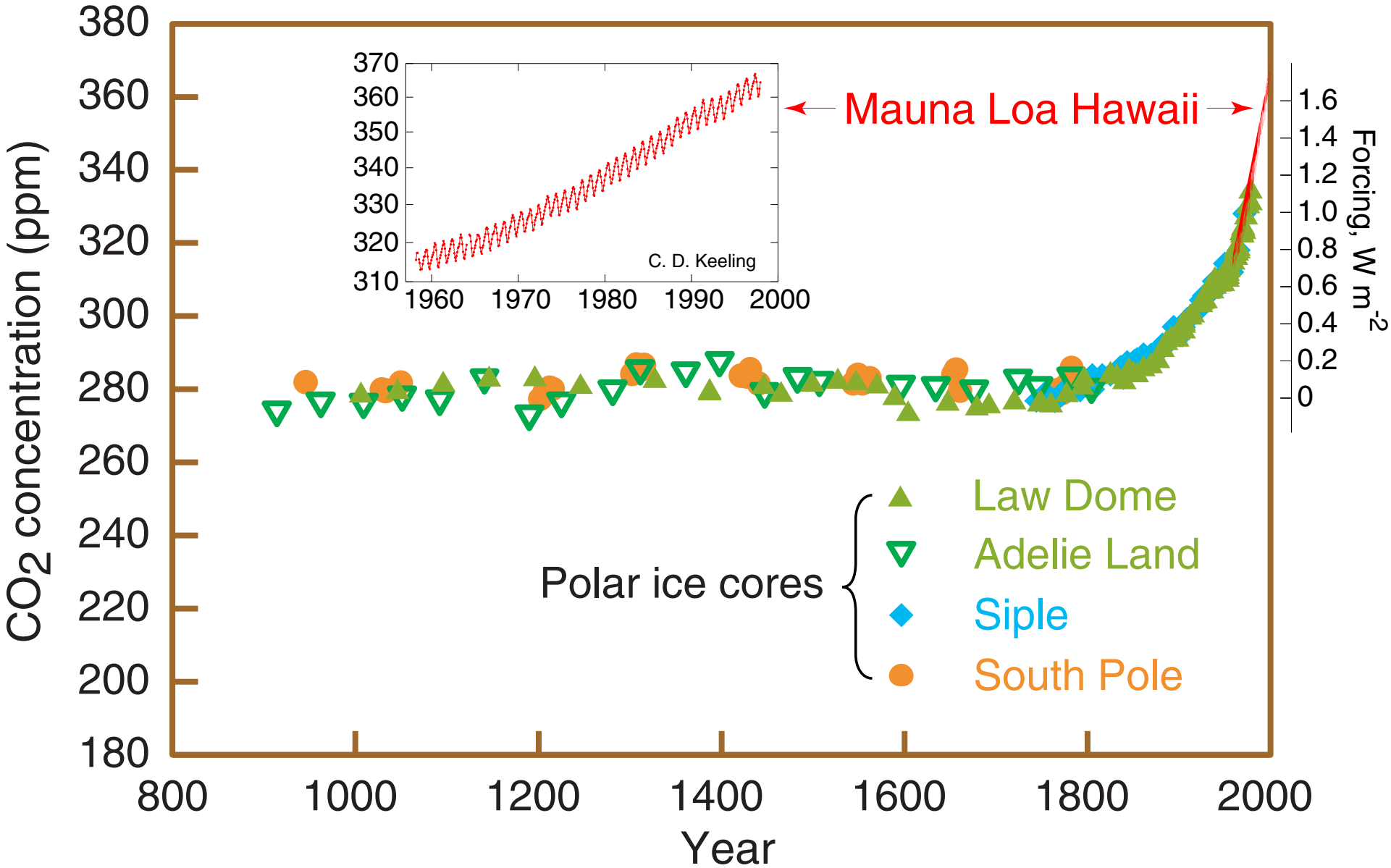
Unit:

Watt per square meter

W m^{-2}



ATMOSPHERIC CARBON DIOXIDE IS INCREASING



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

RADIATIVE FORCING OF CLIMATE CHANGE

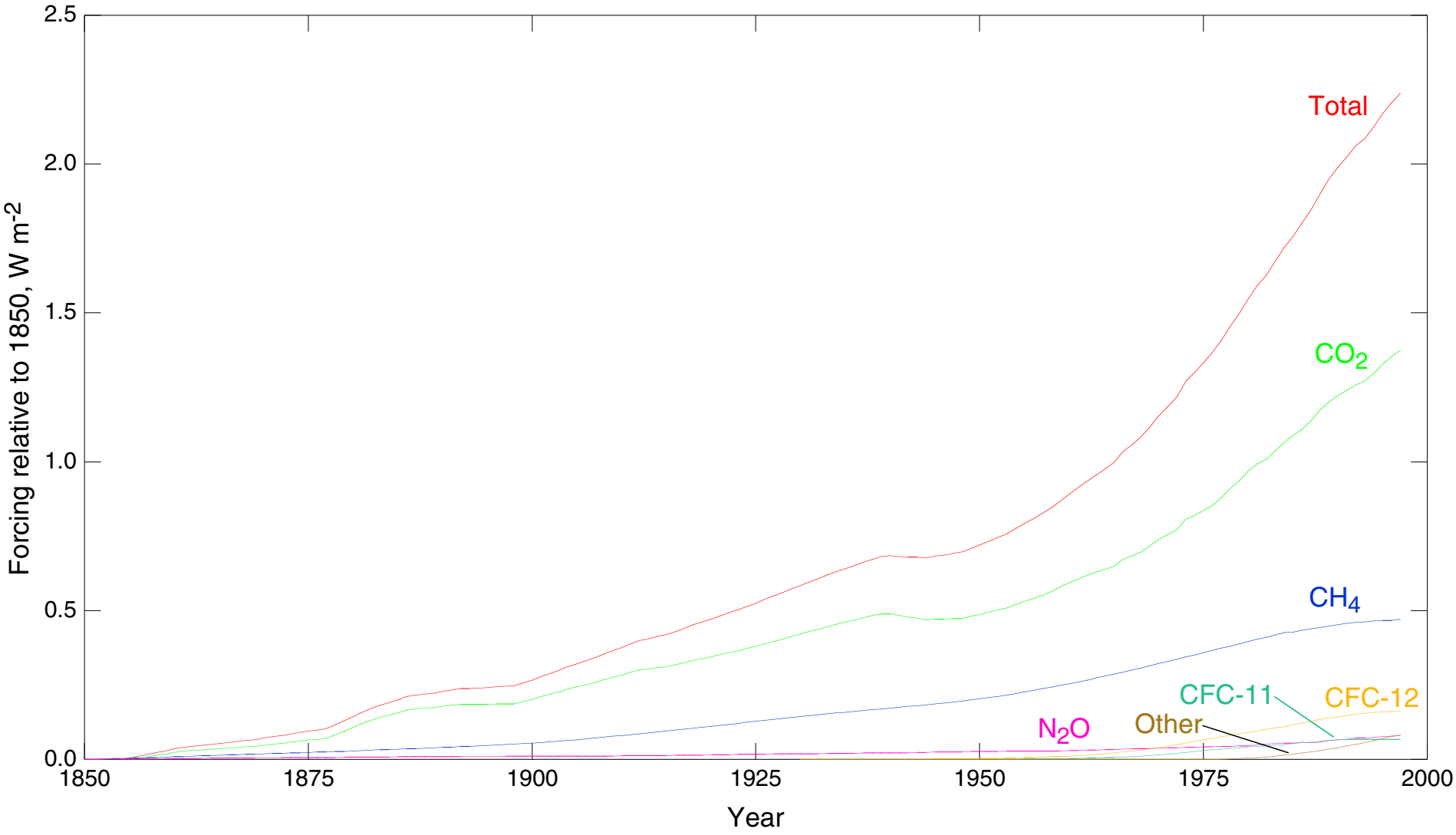
A ***change*** in a radiative flux term in Earth's radiation budget, F, W m^{-2}.

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.

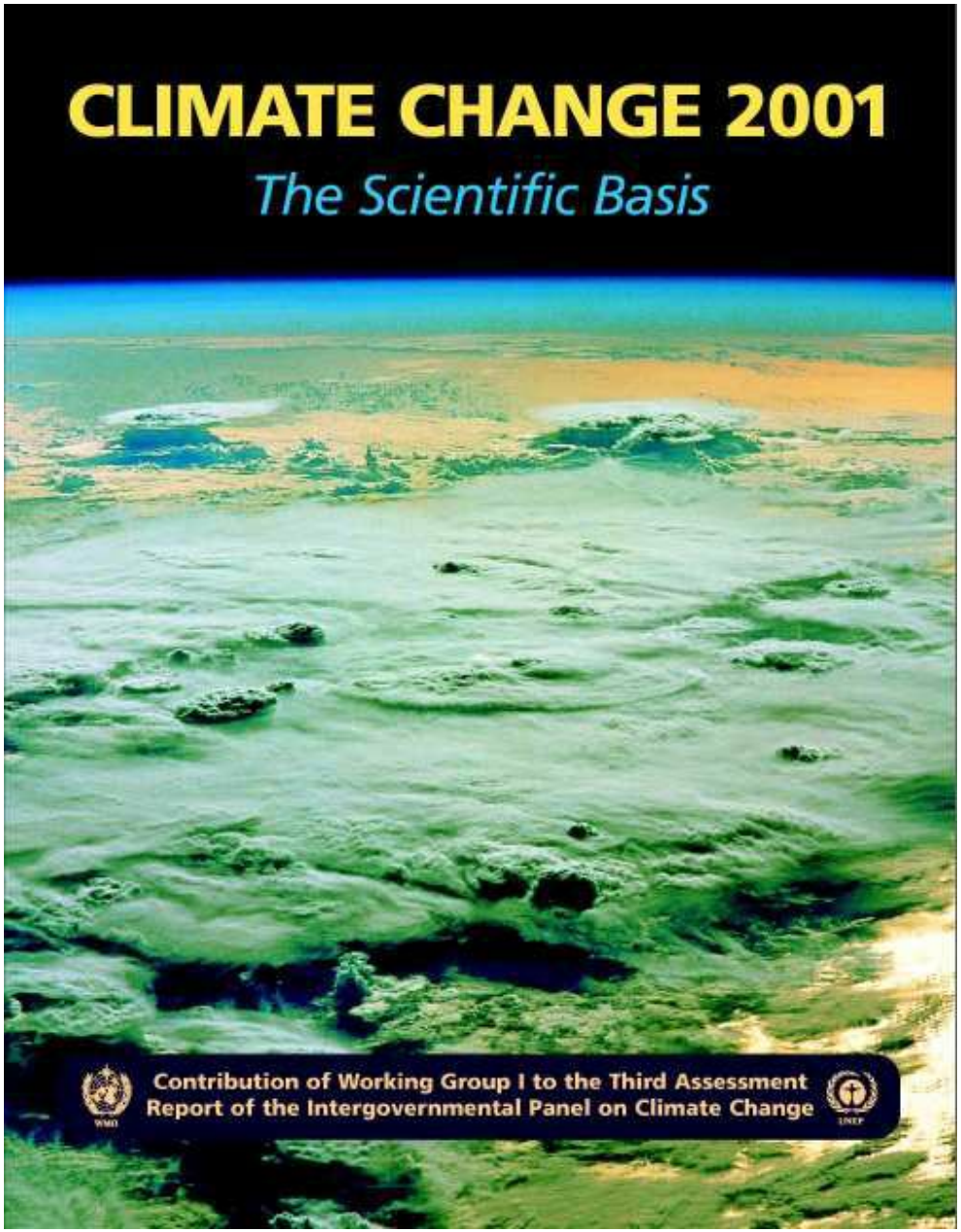
GREENHOUSE GAS FORCINGS OVER THE INDUSTRIAL PERIOD



Data: GISS

THE “BIBLE” OF CLIMATE CHANGE RESEARCH

WMO

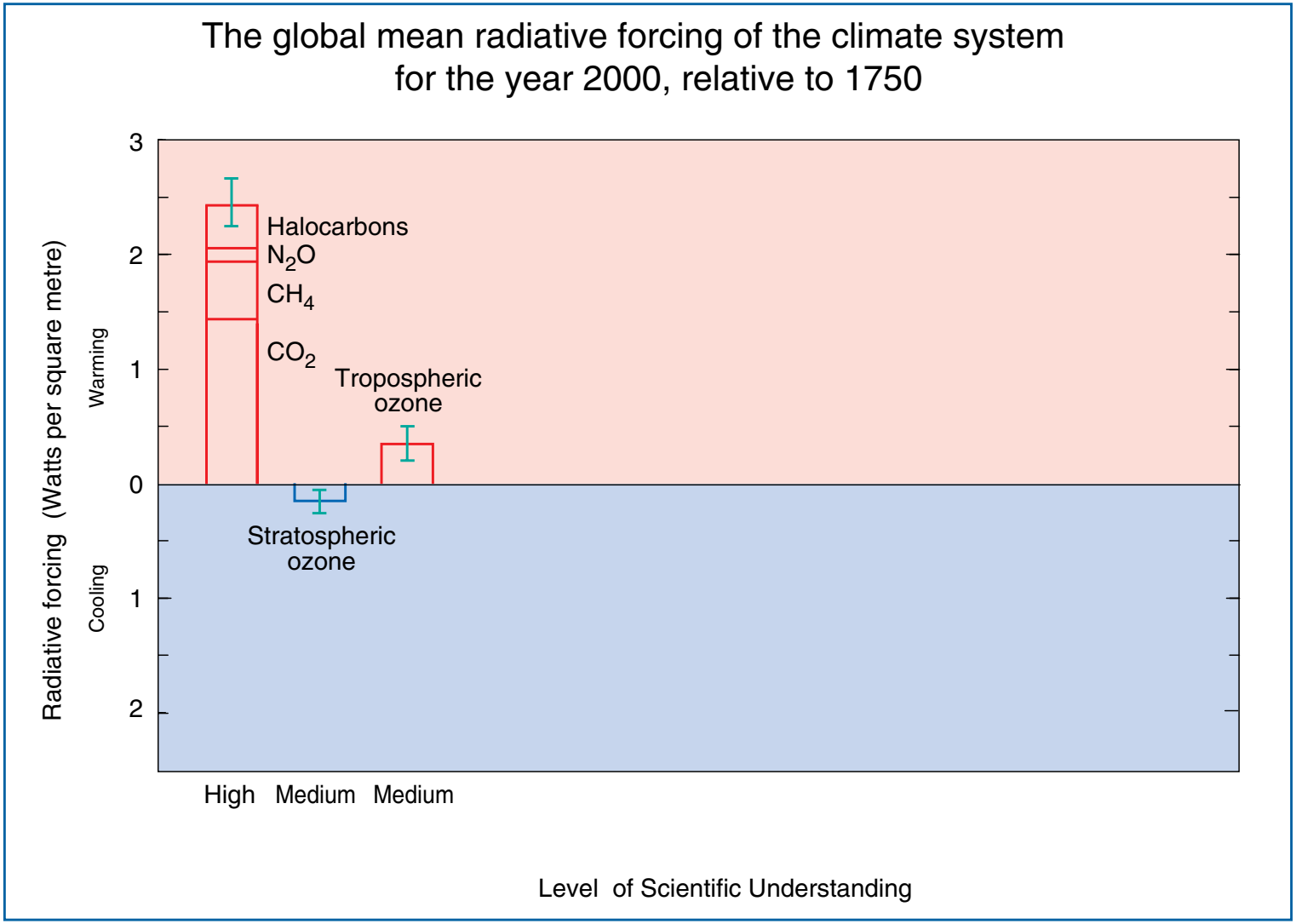


UNEP

Cambridge University Press, 2001

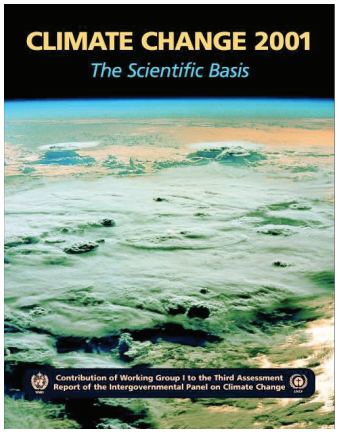
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)

Greenhouse gases only



Summary for Policymakers

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



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)

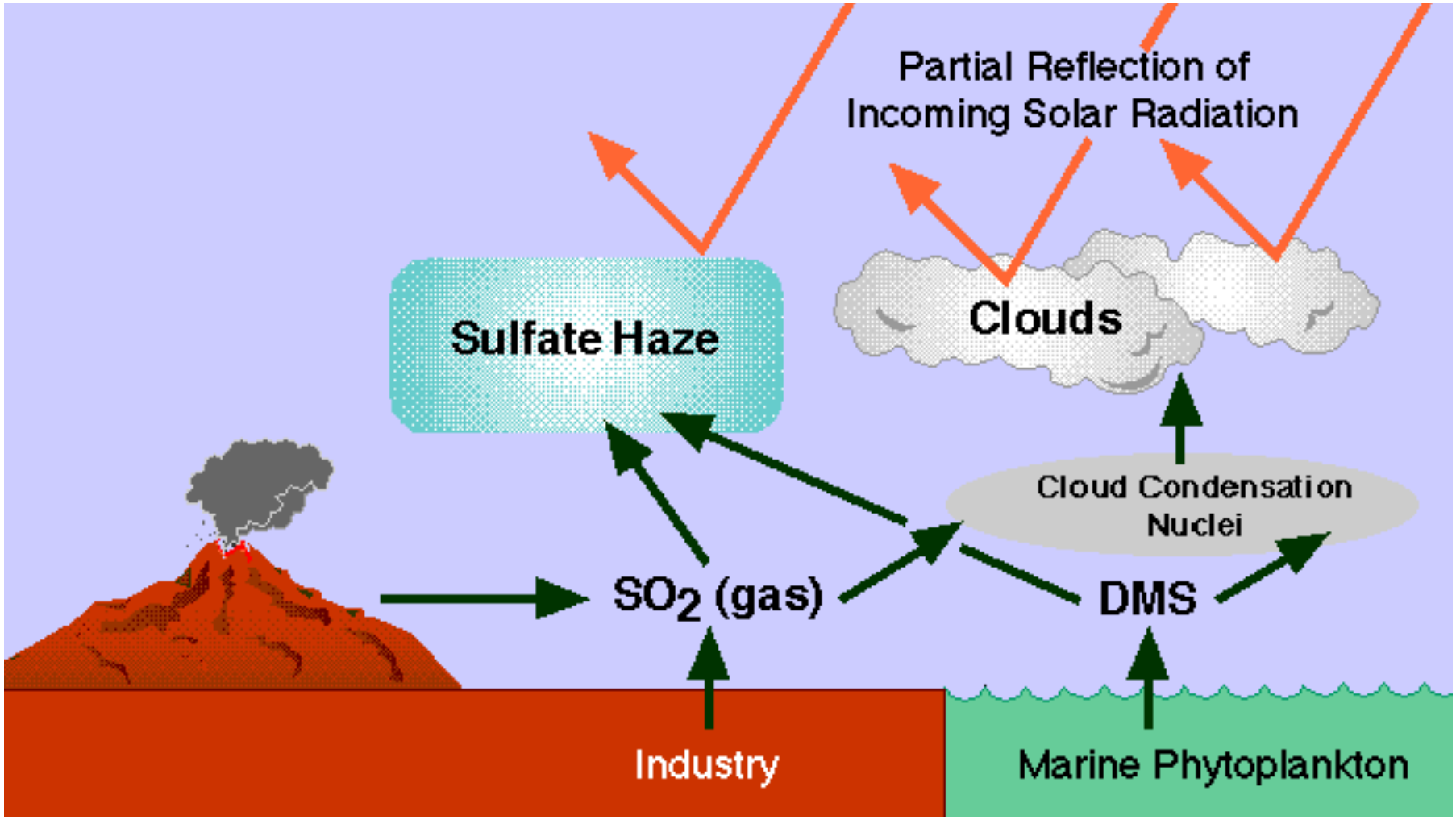
More droplets -- Enhanced cloud lifetime (Albrecht)

Semi-Direct Effect

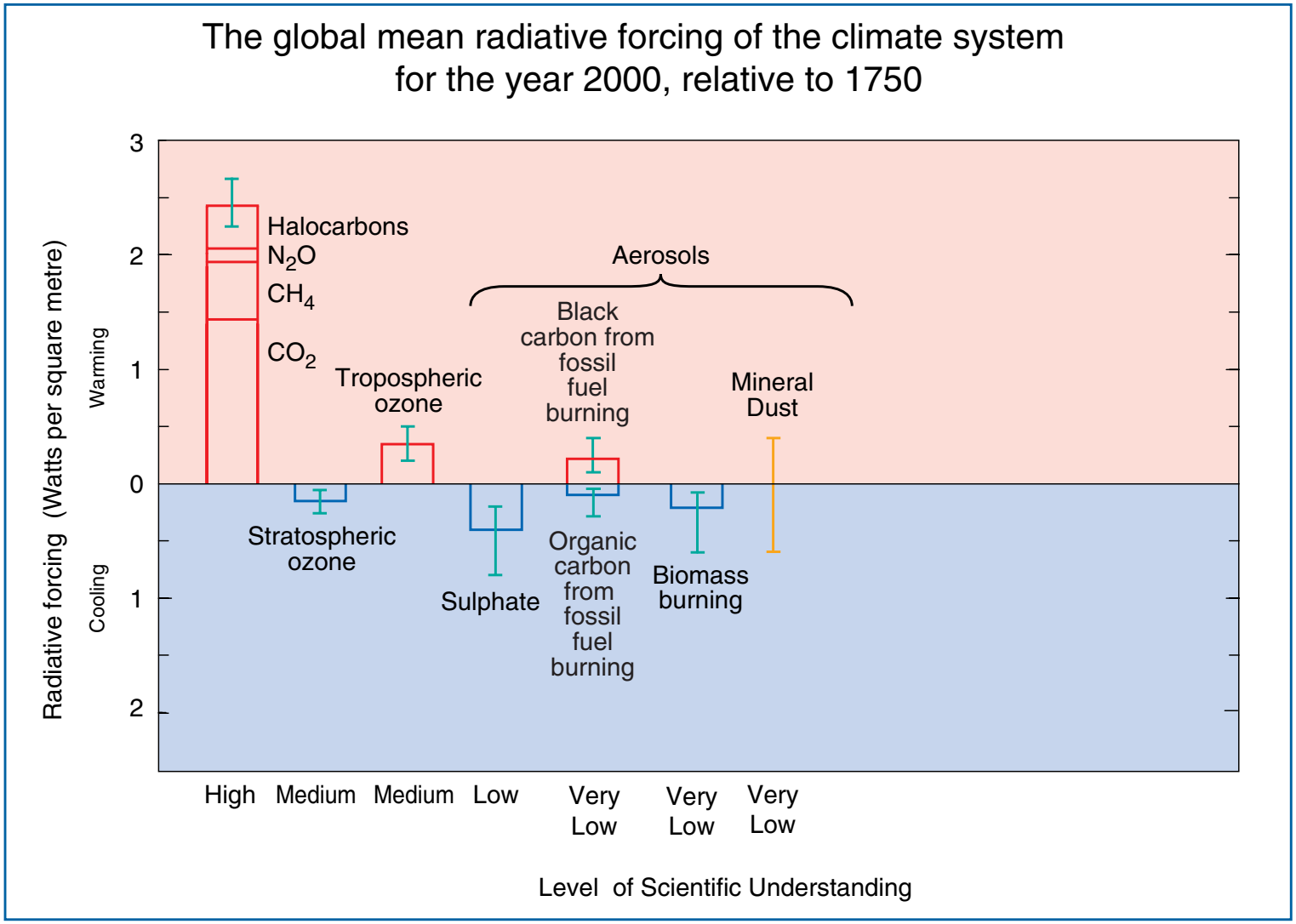
Absorbing aerosol heats air and evaporates clouds

THE “WHITEHOUSE EFFECT”

RADIATIVE FORCING OF CLIMATE CHANGE BY AEROSOLS



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



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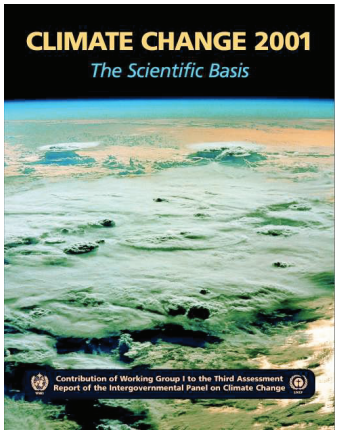
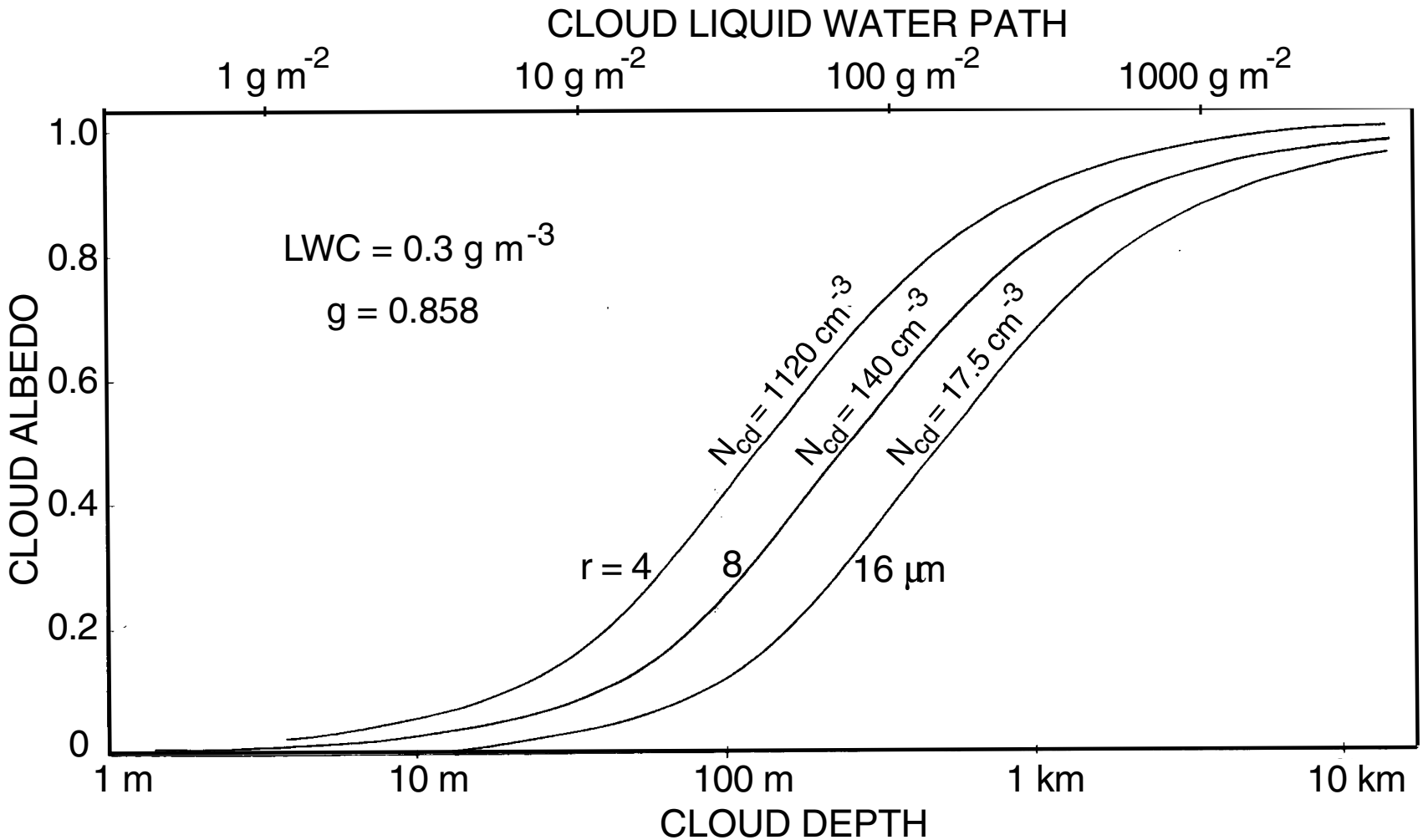


THE TWOMEY EFFECT

PHYSICAL BASIS AND SENSITIVITY

DEPENDENCE OF CLOUD ALBEDO ON CLOUD DEPTH

Influence of Cloud Drop Radius and Concentration

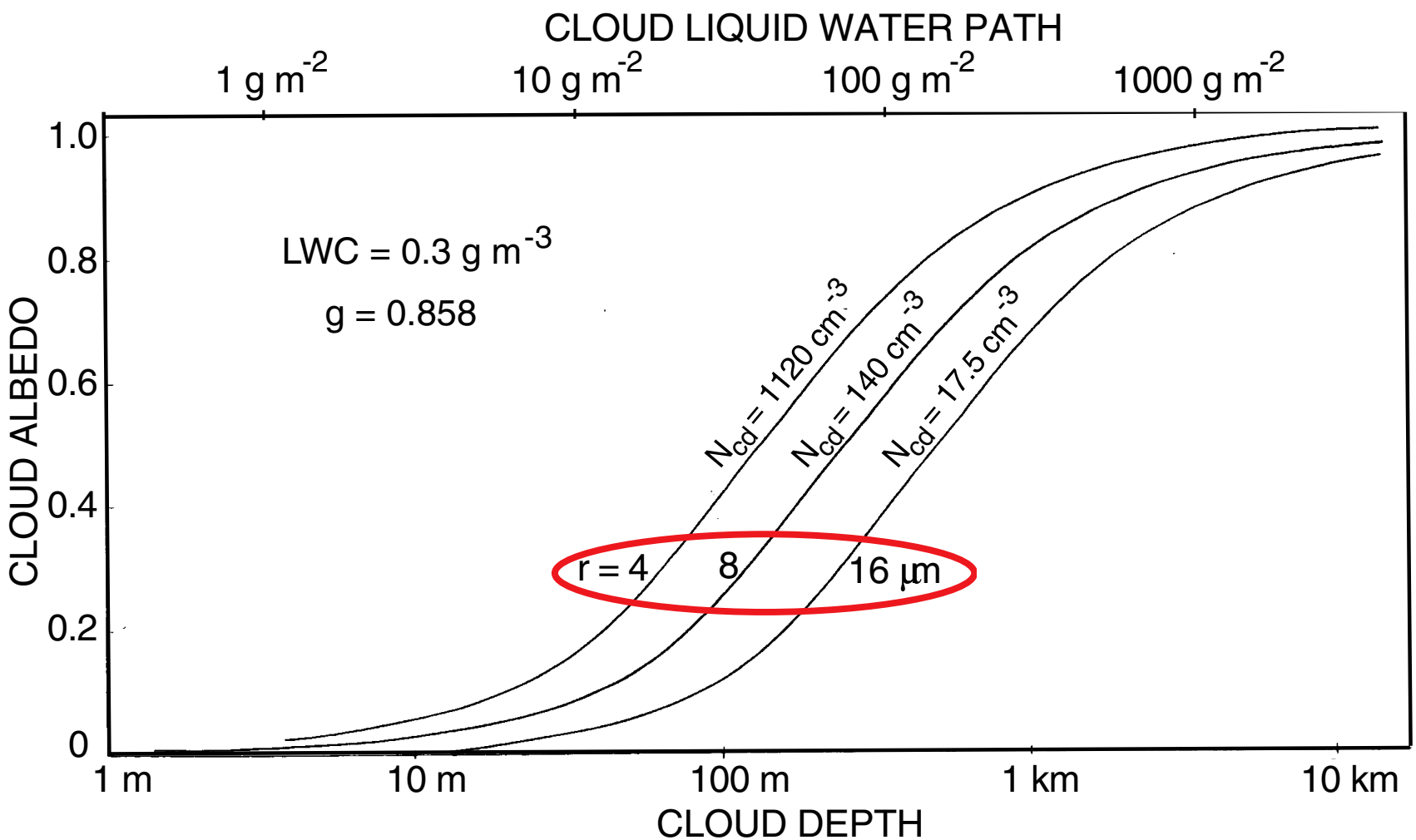


Twomey, *Atmospheric Aerosols*, 1977

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

DEPENDENCE OF CLOUD ALBEDO ON CLOUD DEPTH

Influence of Cloud Drop Radius and Concentration



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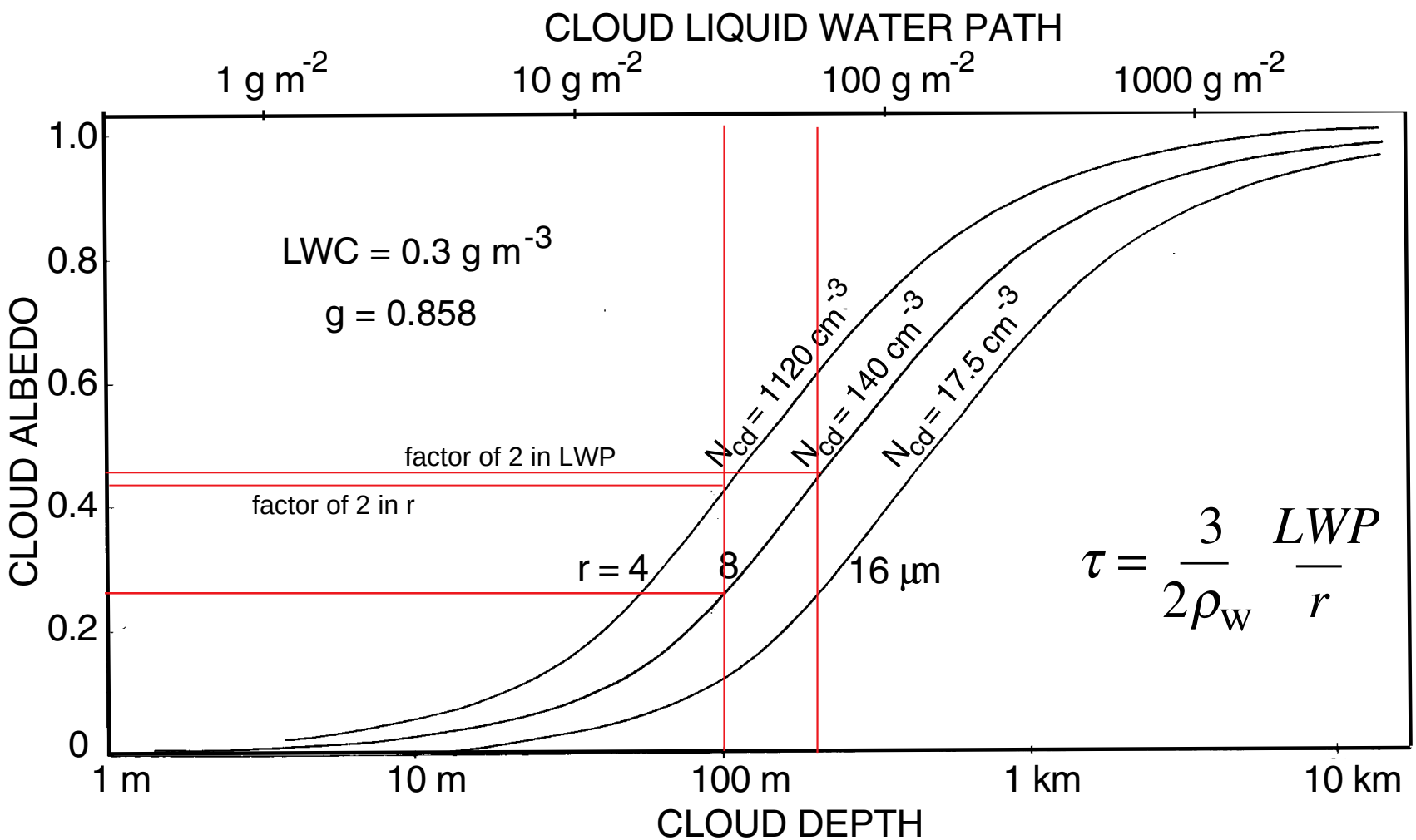
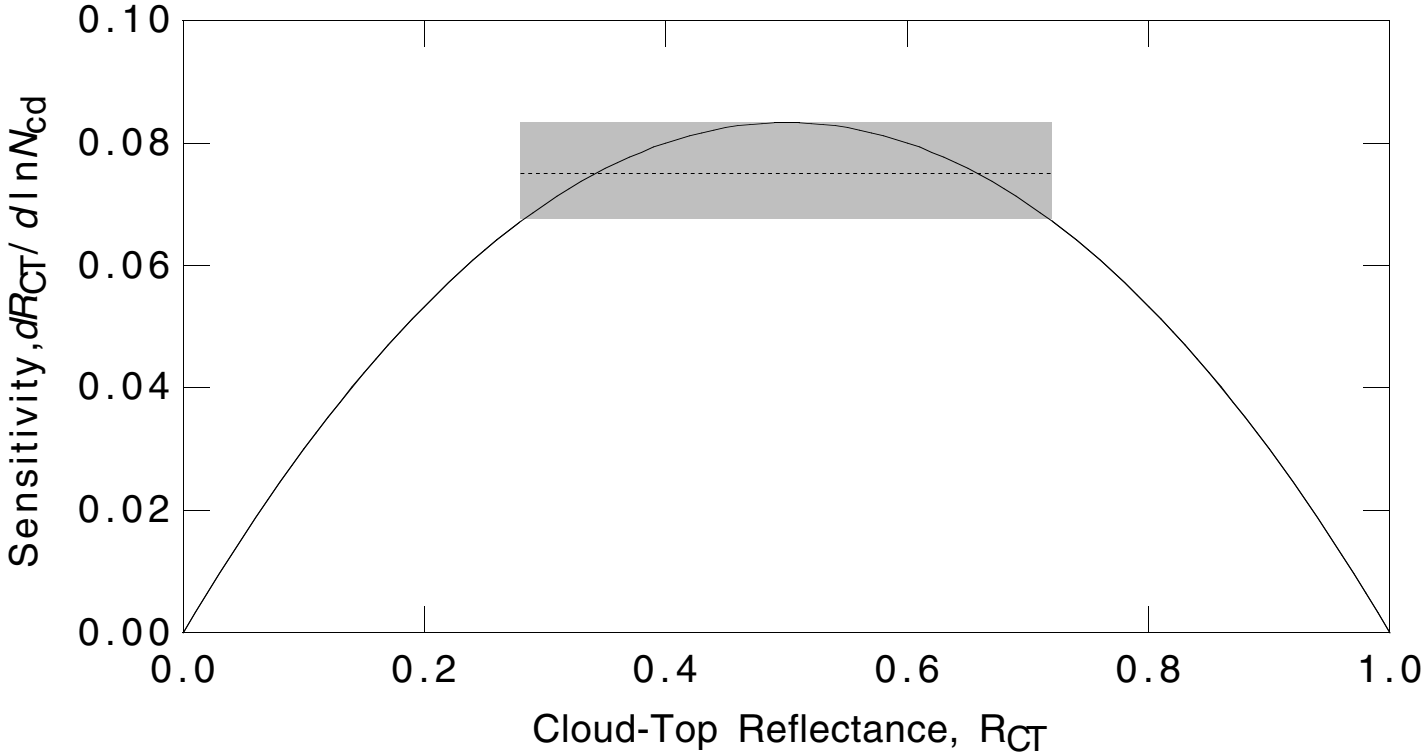


Twomey, *Atmospheric Aerosols*, 1977

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

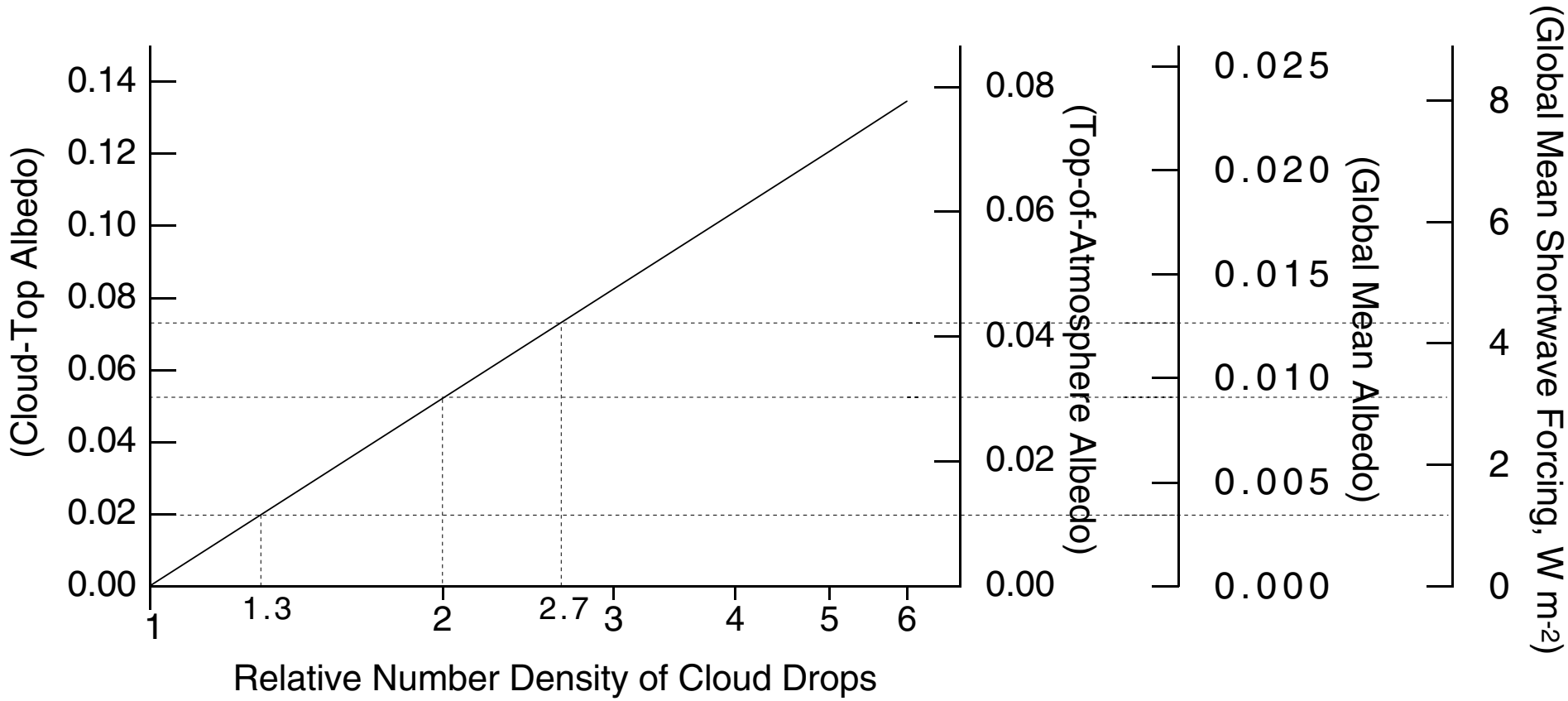


Modified from Charlson et al., 1992

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_{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.

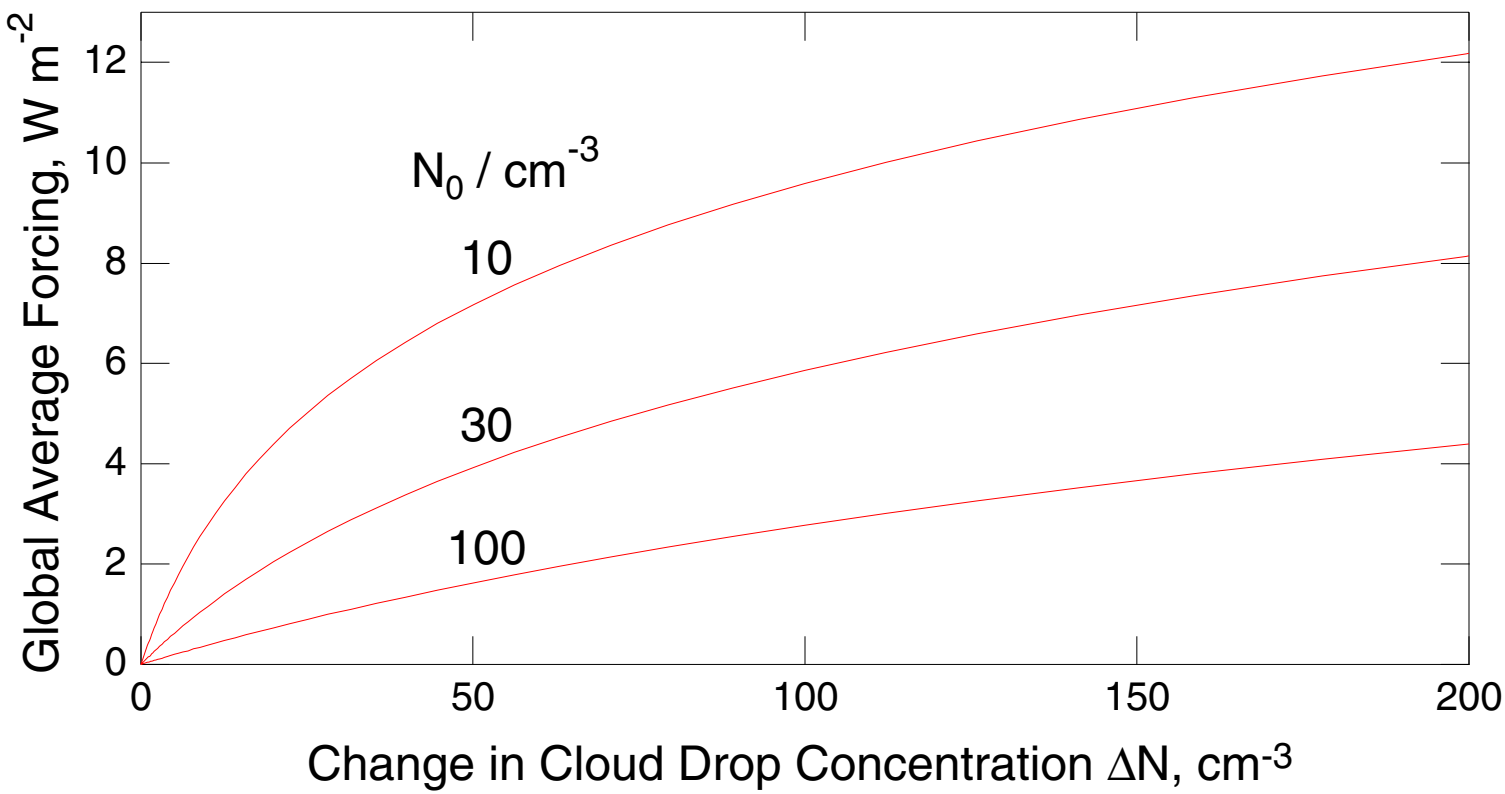
A 30% increase in cloud drop concentration results in a forcing of ~1 W m^{-2}.

INDIRECT (TWOMEY) FORCING

Dependence on incremental cloud drop concentration ΔN and

Sensitivity to initial cloud drop concentration N_0

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

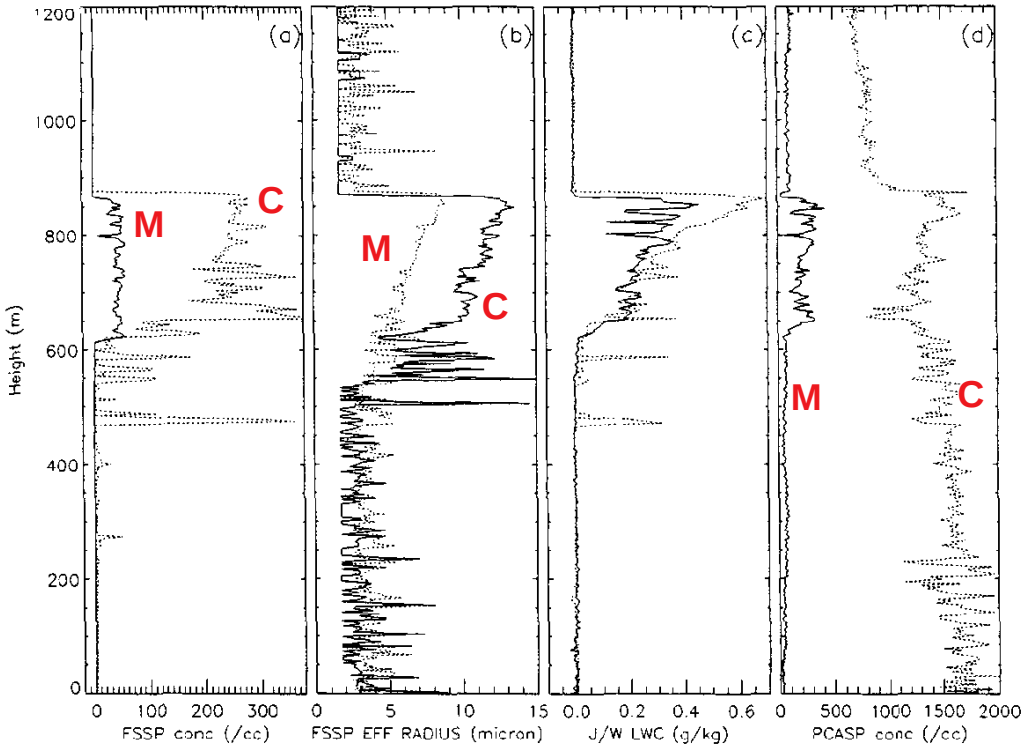


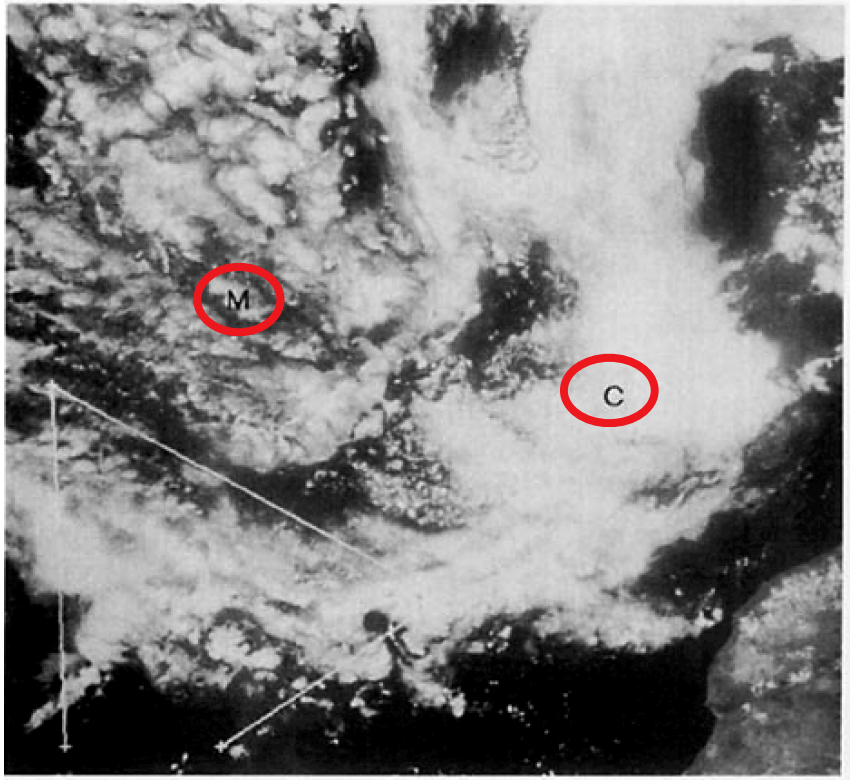
Aerosol indirect forcing is highly sensitive to background CCN concentration.

RELATION BETWEEN AEROSOL CONCENTRATIONS AND CLOUD DROP CONCENTRATIONS

CLOUD MICROPHYSICAL PROPERTIES AND SATELLITE VISIBLE RADIANCE

ASTEX, Northeast Atlantic, June, 1992

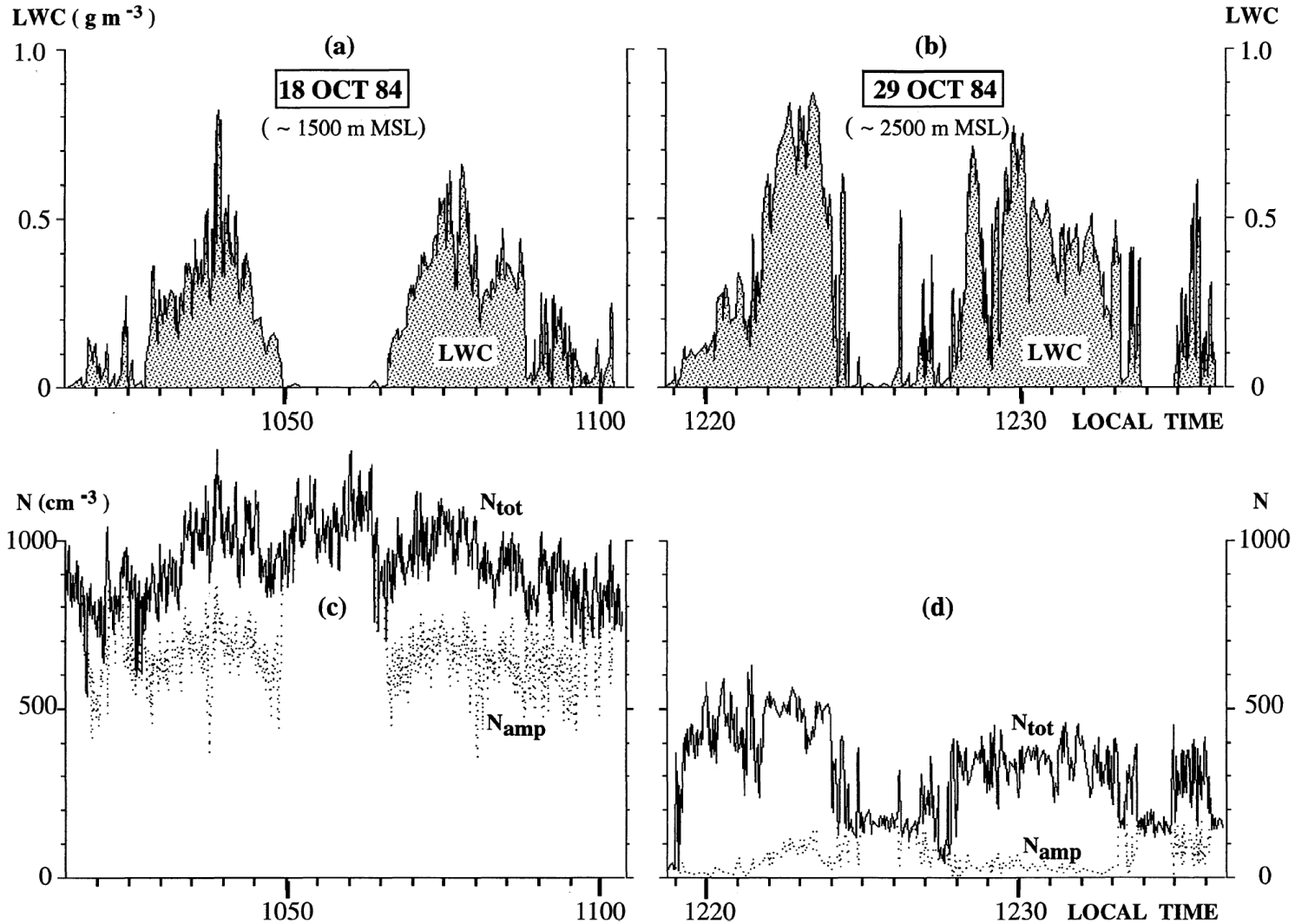




Albrecht et al., BAMS, 1995

FRACTIONAL UPTAKE OF ACCUMULATION MODE PARTICLES INTO CLOUDWATER

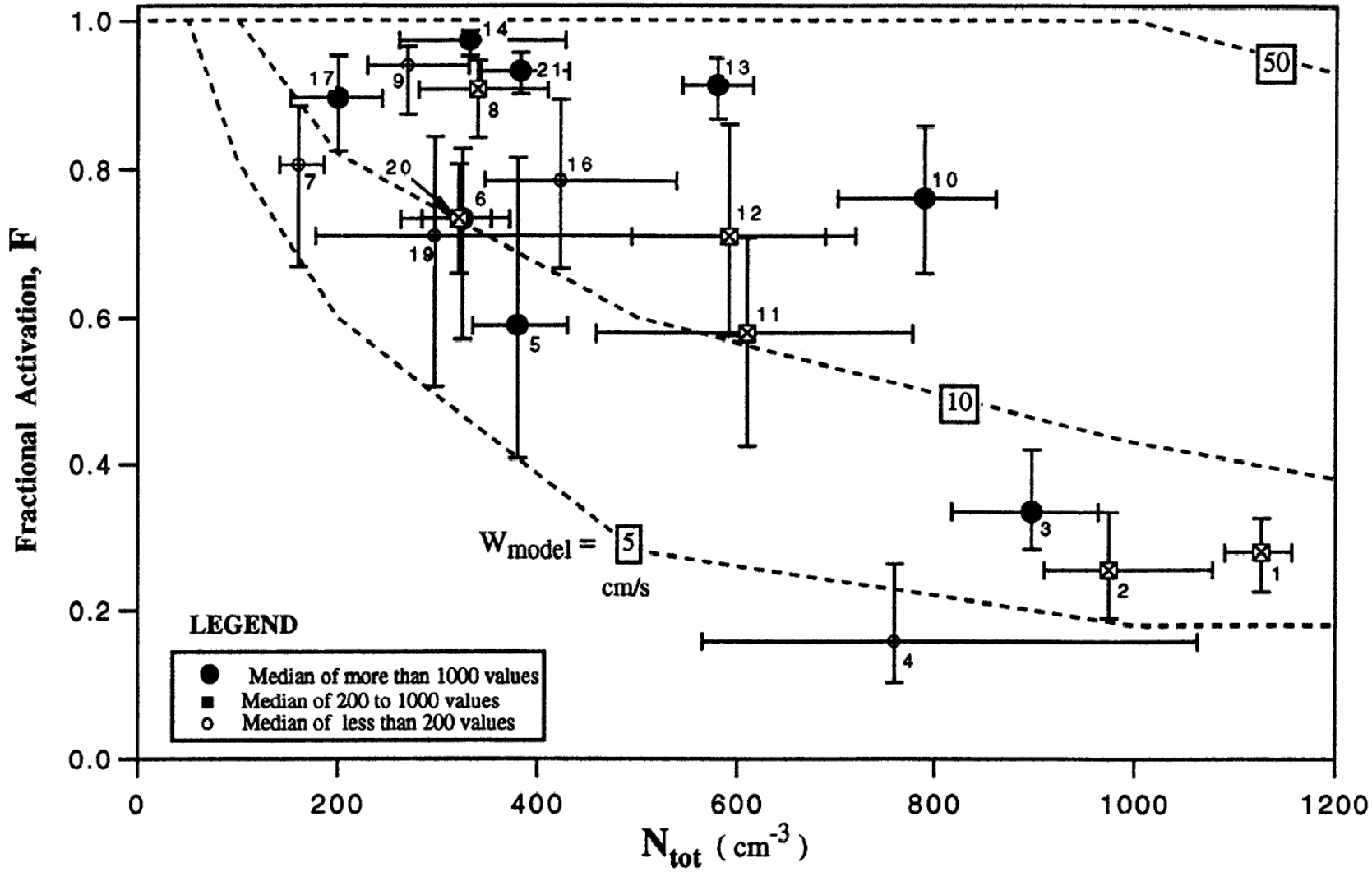
Aircraft measurements of ASASP (0.17 - 2 μm diameter) and FSSP (2 - 35 μm diameter) particles



Gillani et al., JGR, 1995

FRACTION OF ACTIVATED AEROSOL PARTICLES

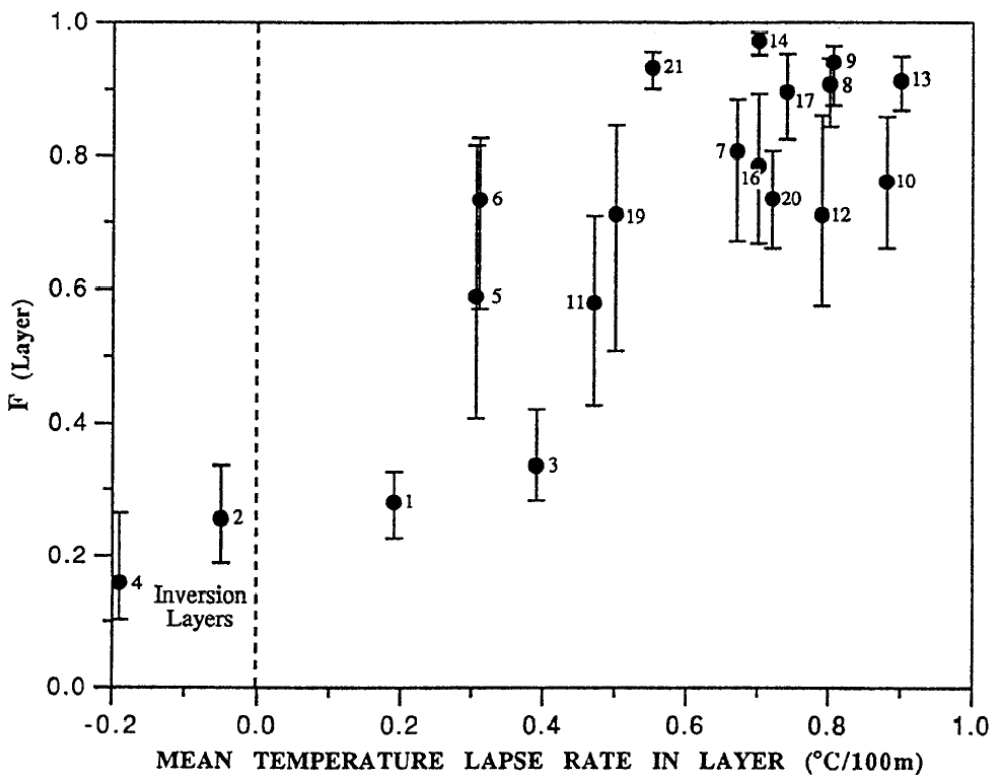
Dependence on particle number concentration

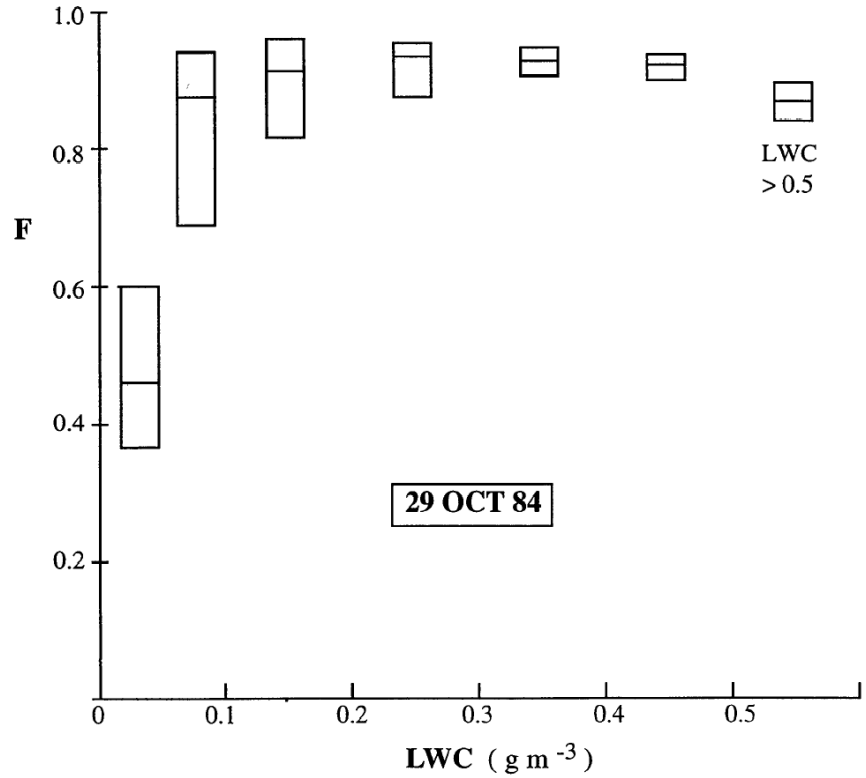


Gillani et al., JGR, 1995

FRACTION OF ACTIVATED AEROSOL PARTICLES

Dependence on Lapse Rate and Liquid Water Content



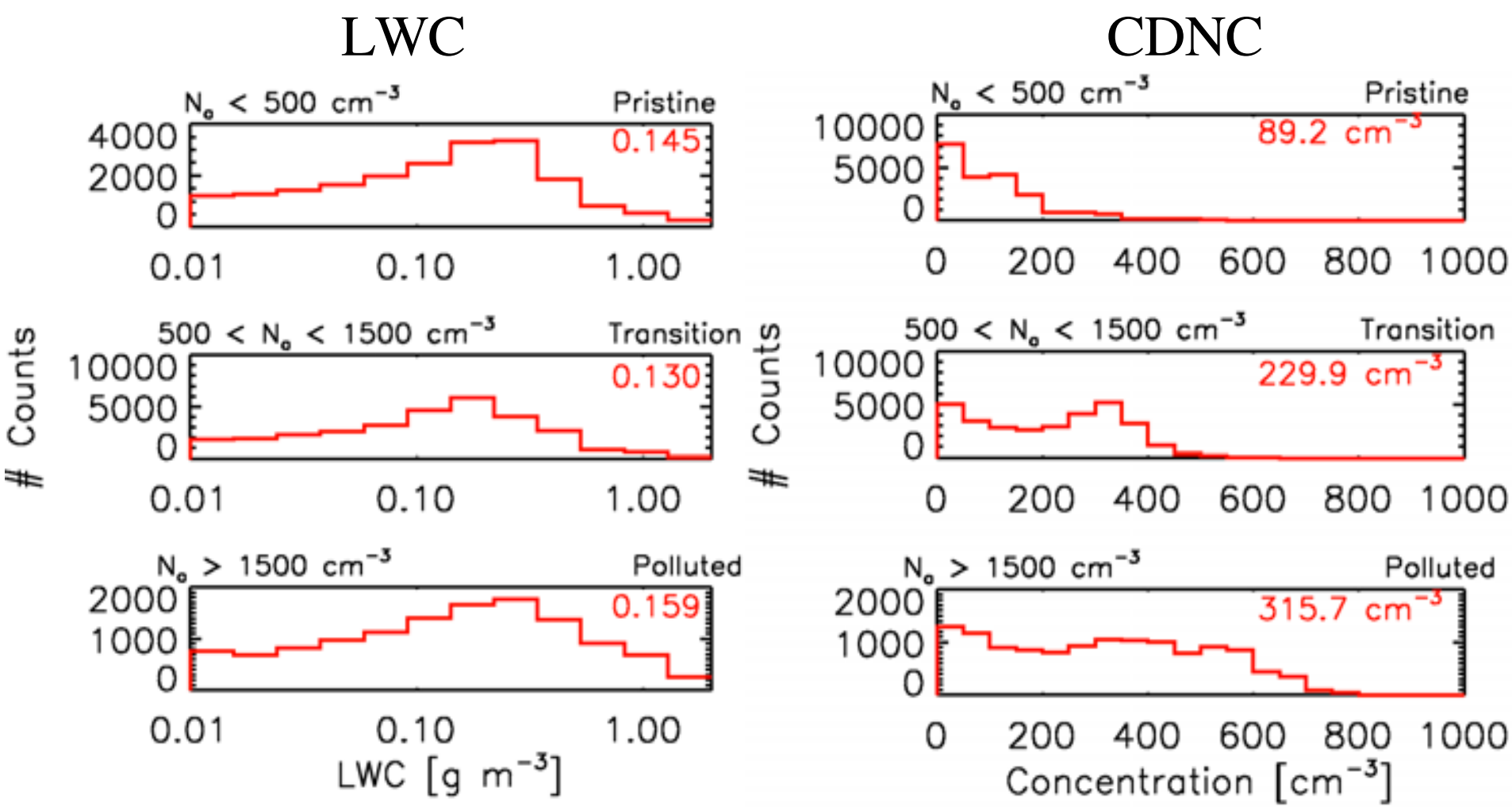


Gillani et al., JGR, 1995

PRISTINE *vs*. POLLUTED CLOUDS IN INDOEX

In-situ aircraft measurements during cross-Equatorial transects

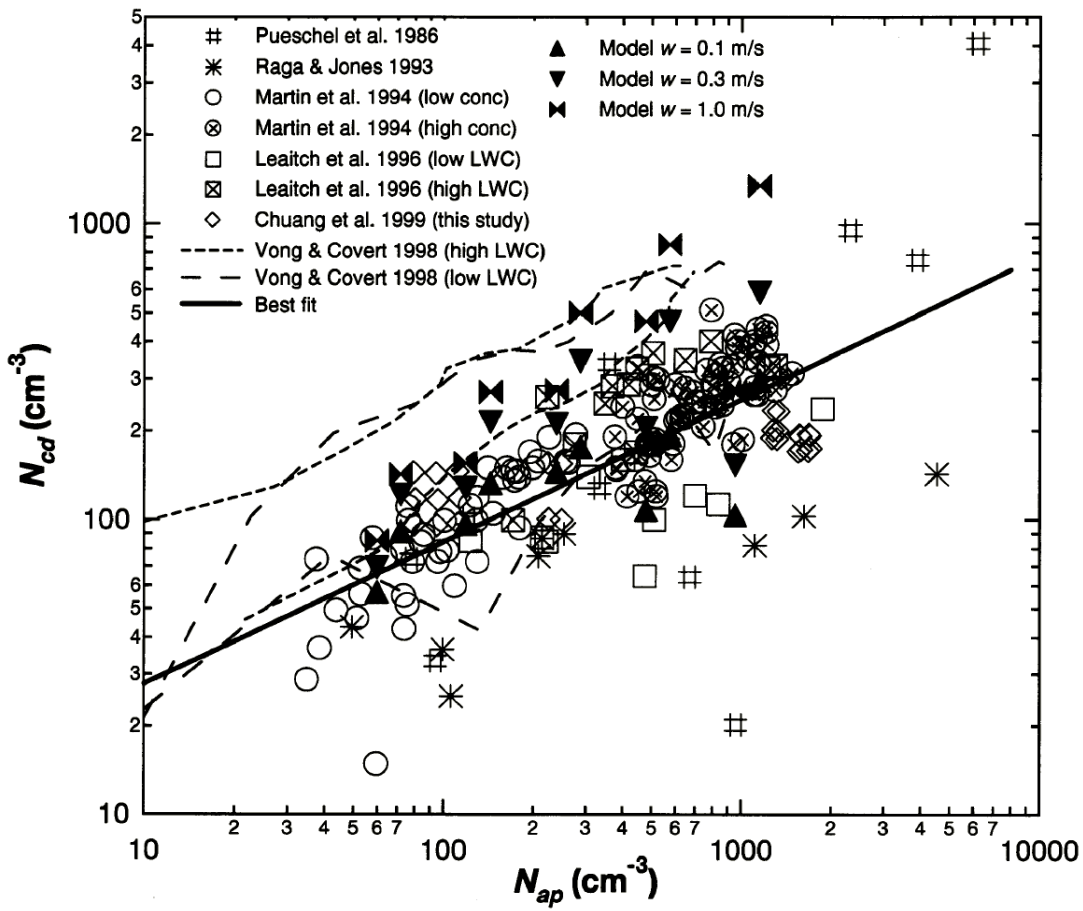
Classified by concentration of aerosol particles



Heymsfield and McFarquhar, JGR, 2001

CLOUD DROP NUMBER CONCENTRATION

Dependence on accumulation-mode aerosol particle concentration

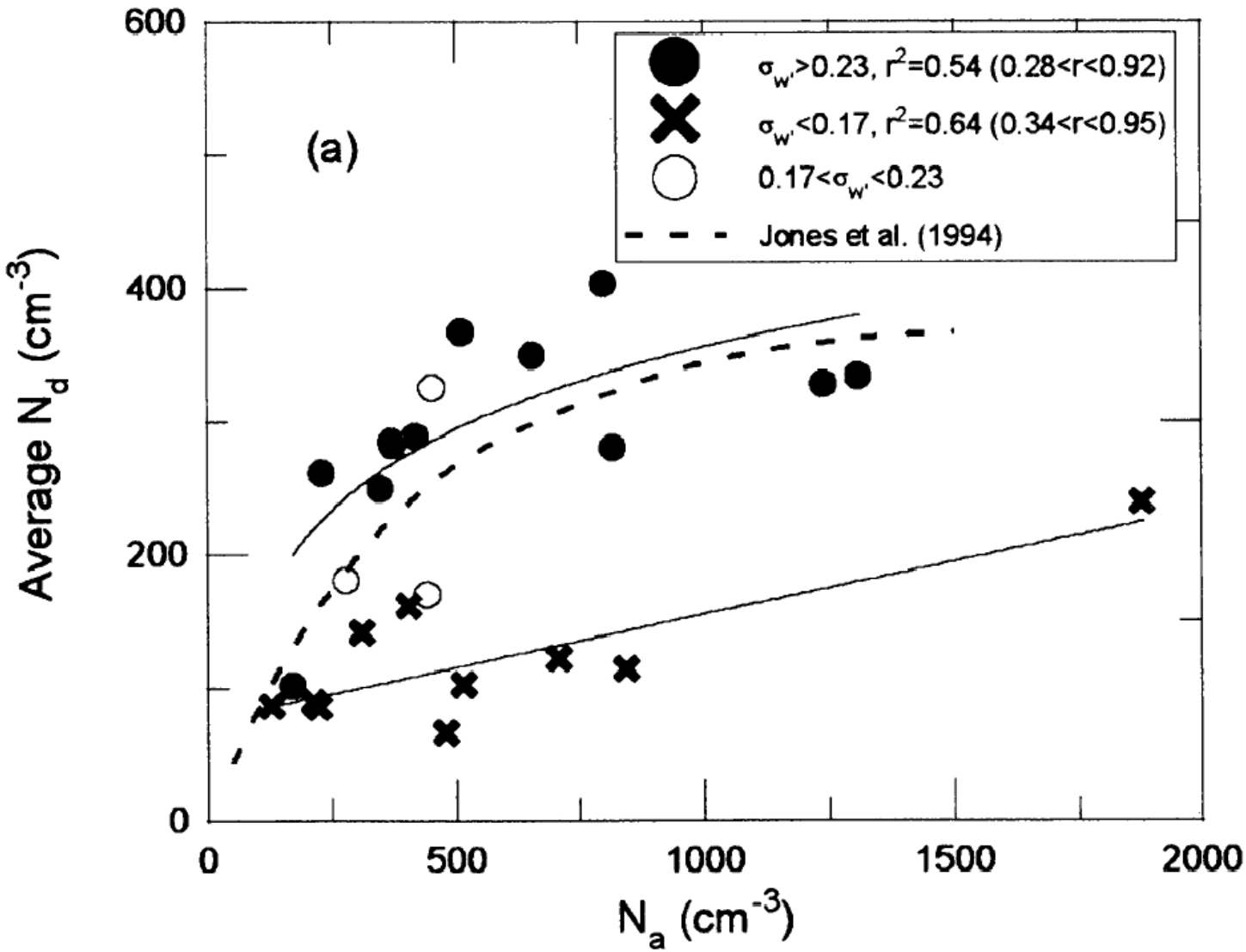


Chuang et al., 2000

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

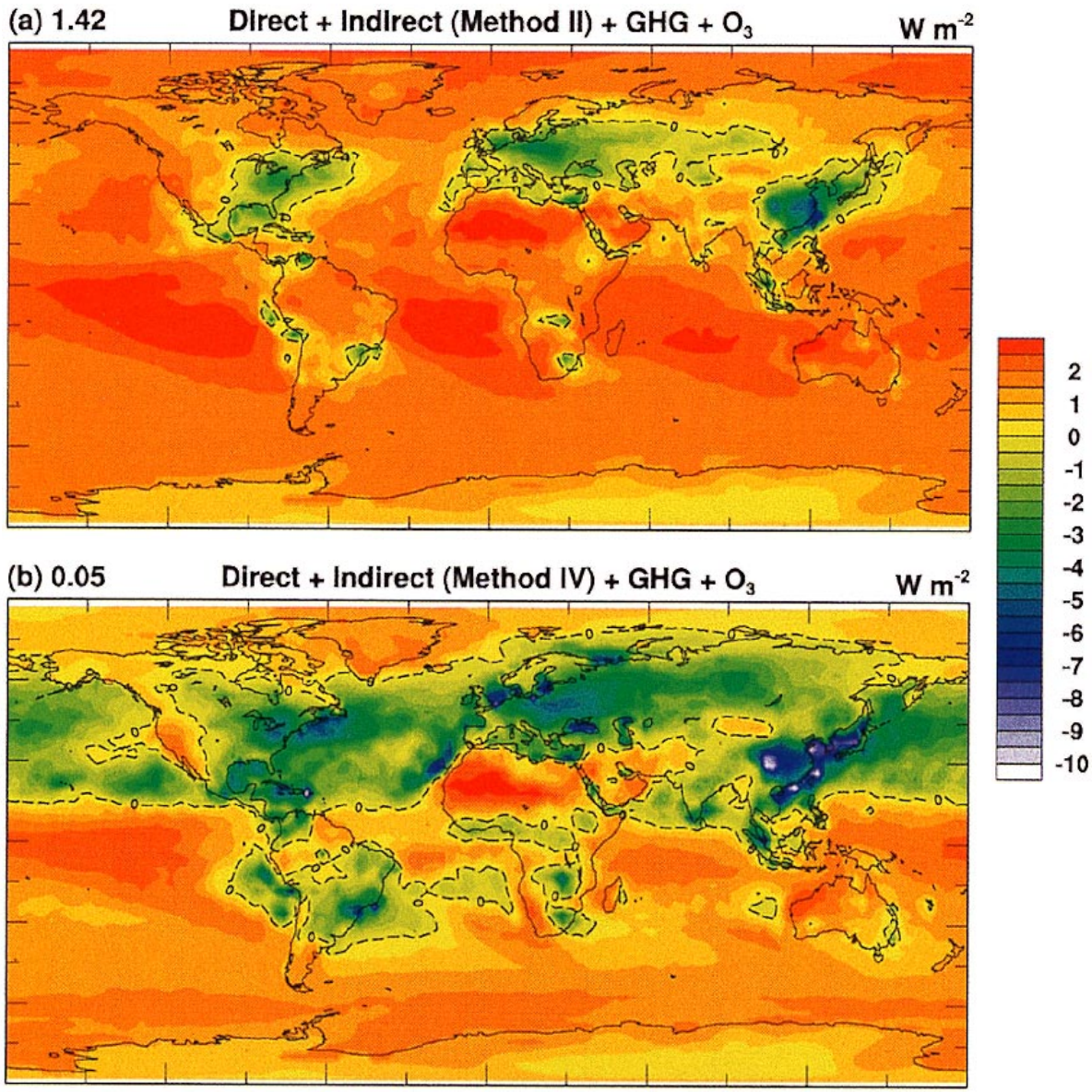


Leaitch et al., JGR, 1996

SHORTWAVE FORCING, ANNUAL AVERAGE

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

Two Formulations of Cloud Droplet Concentration



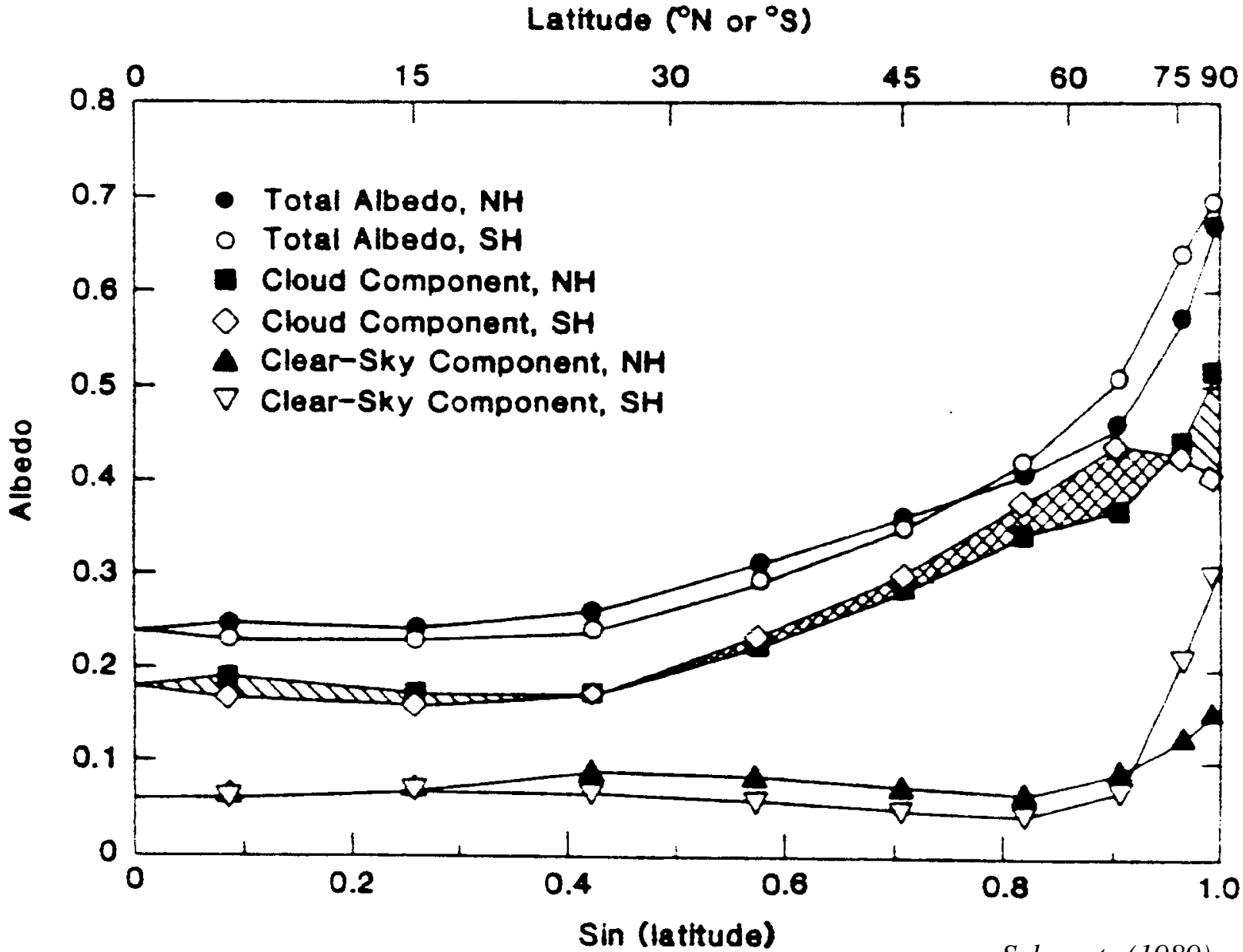
Kiehl et al., JGR, 2000

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

SEARCH FOR INDIRECT EFFECT IN INTERHEMISPHERIC COMPARISONS

INTERHEMISPHERIC COMPARISON OF ALBEDO COMPONENTS

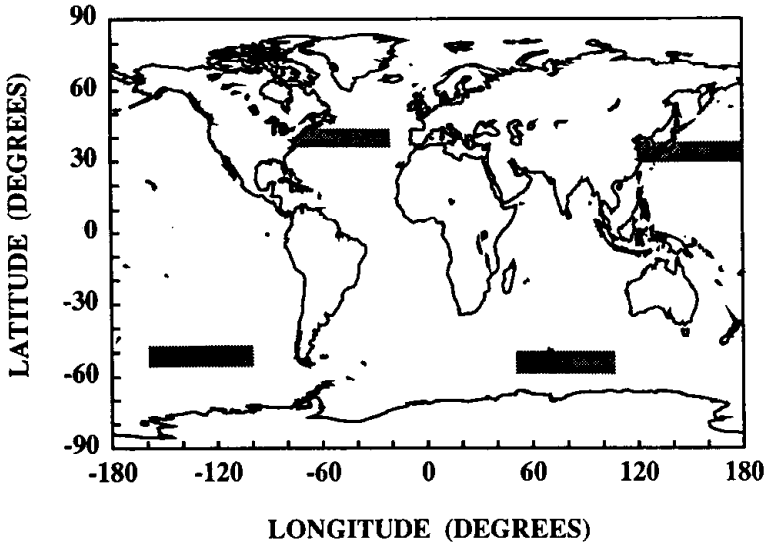
Data from Nimbus 4

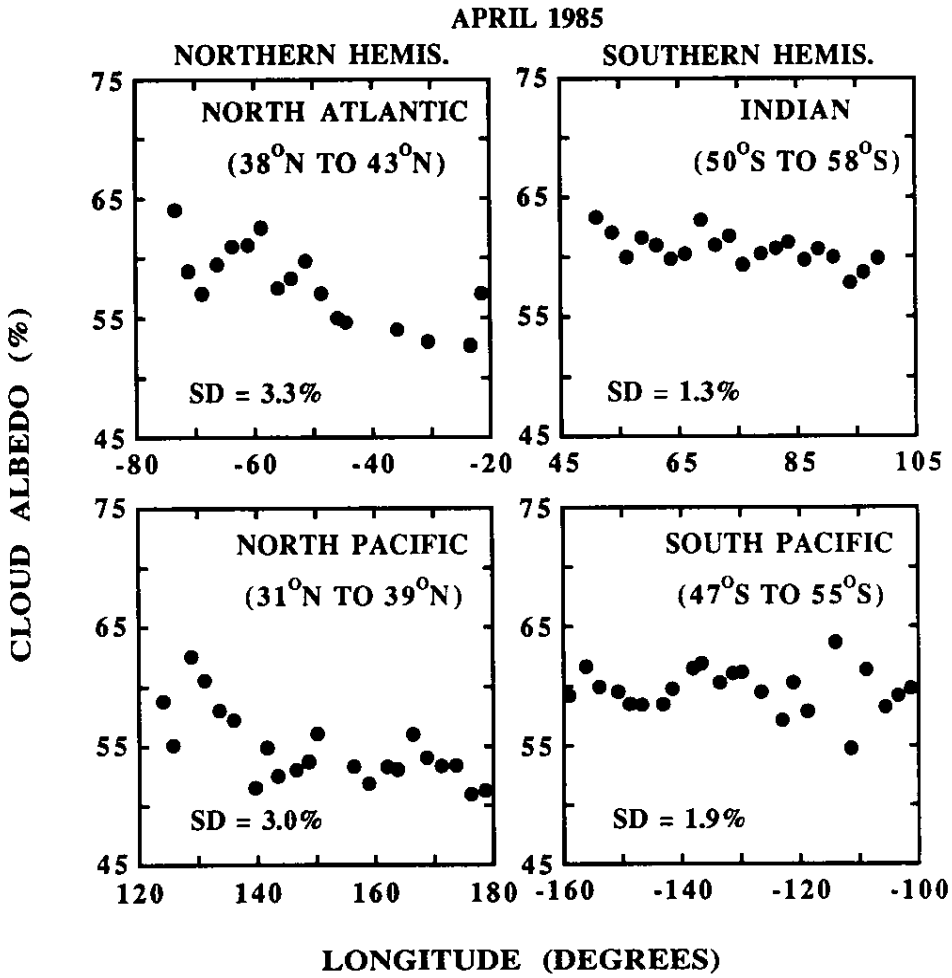


Schwartz (1989)

LONGITUDE DEPENDENCE OF CLOUD ALBEDO

Test for Anthropogenic Influence in Northern Hemisphere vs. Southern Hemisphere as Control

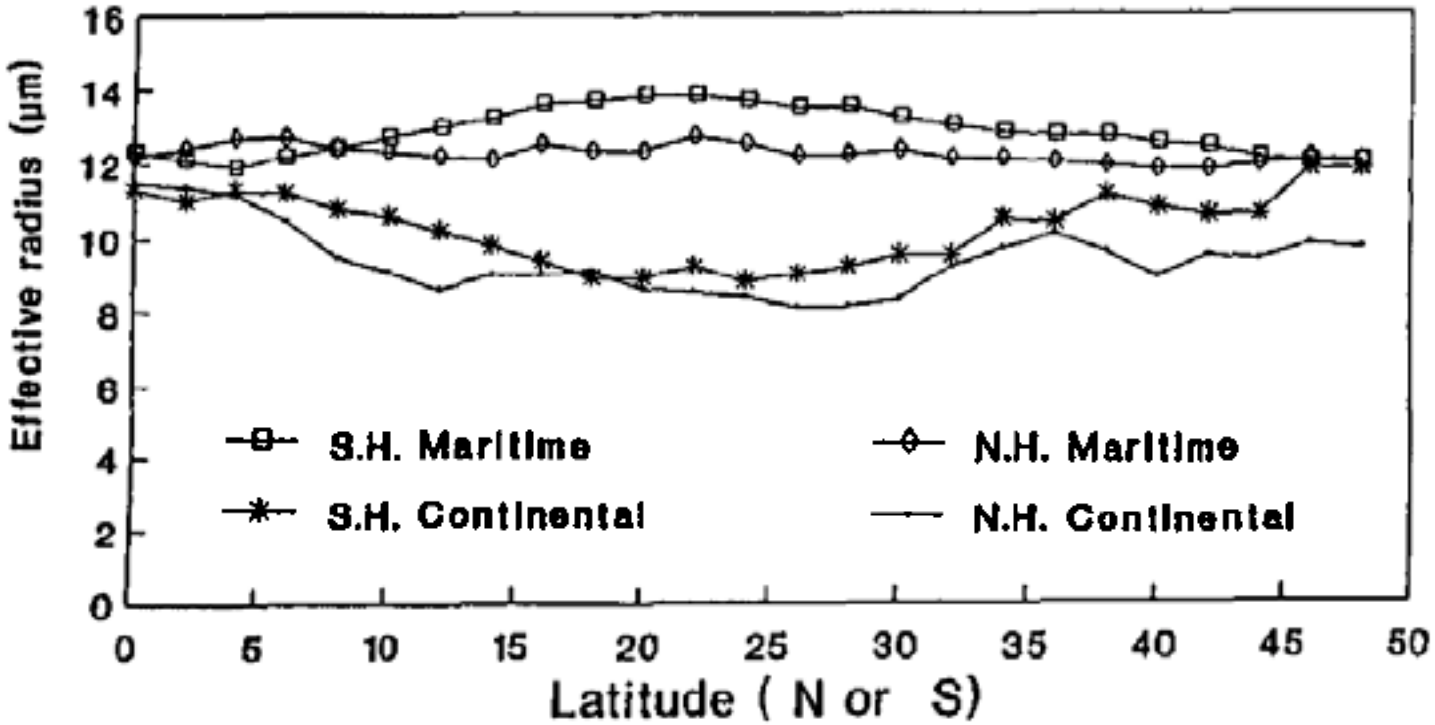




Kim and Cess, JGR, 1994

EXAMINATION FOR INDIRECT EFFECT IN INTERHEMISPHERIC COMPARISONS

Zonal-mean cloud drop effective radius

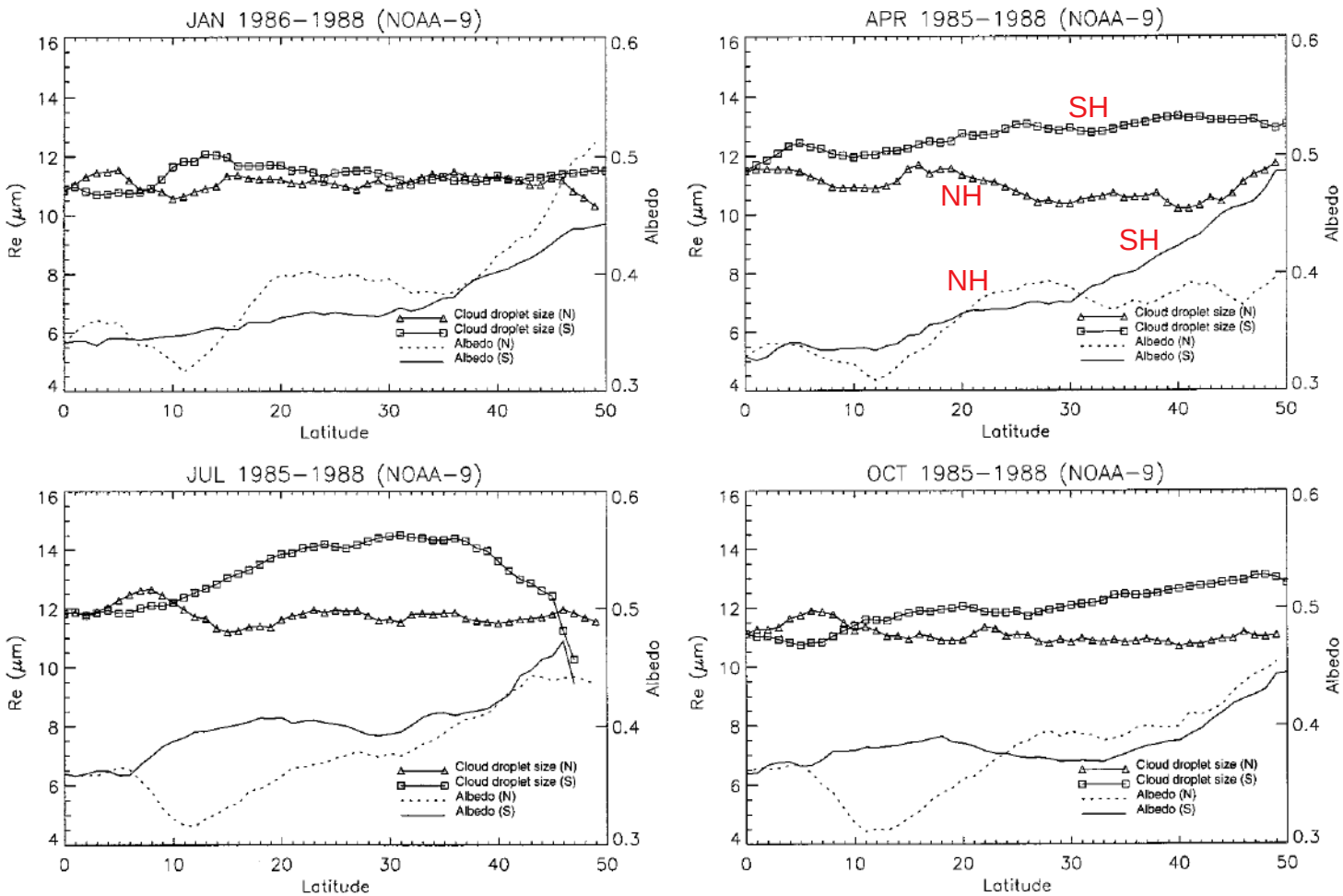


Han, Rossow, and Lacis, 1994

Smaller effective radius in NH would be indicative of greater cloud drop concentration due to industrial aerosol.

EXAMINATION FOR INDIRECT EFFECT IN INTERHEMISPHERIC COMPARISONS

Zonal-mean cloud drop effective radius and cloud albedo

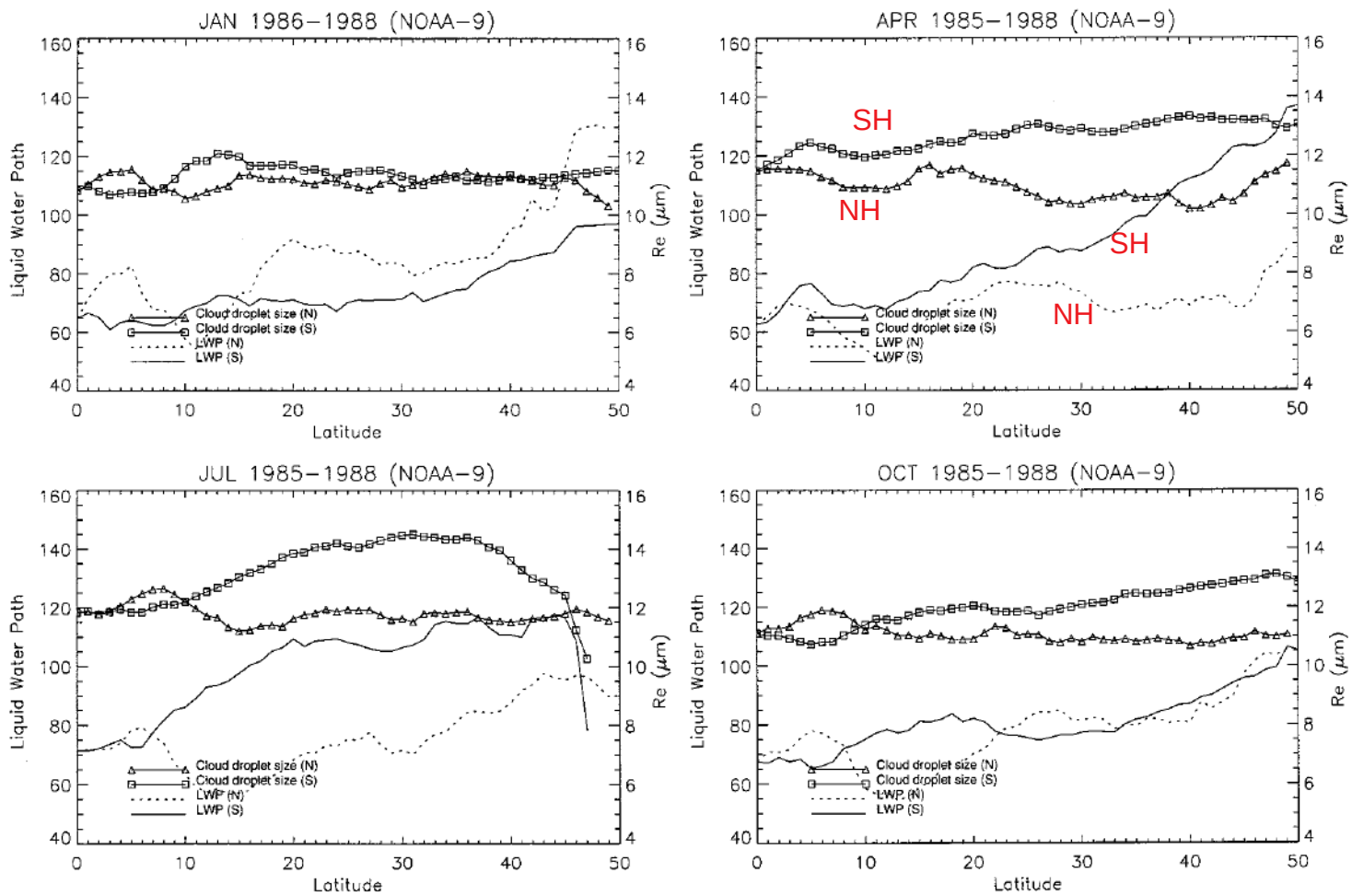


Han et al., 1998

Interhemispheric difference in effective radius is not exhibited in cloud albedo.

EXAMINATION FOR INDIRECT EFFECT IN INTERHEMISPHERIC COMPARISONS

Zonal-mean cloud drop effective radius and liquid water path

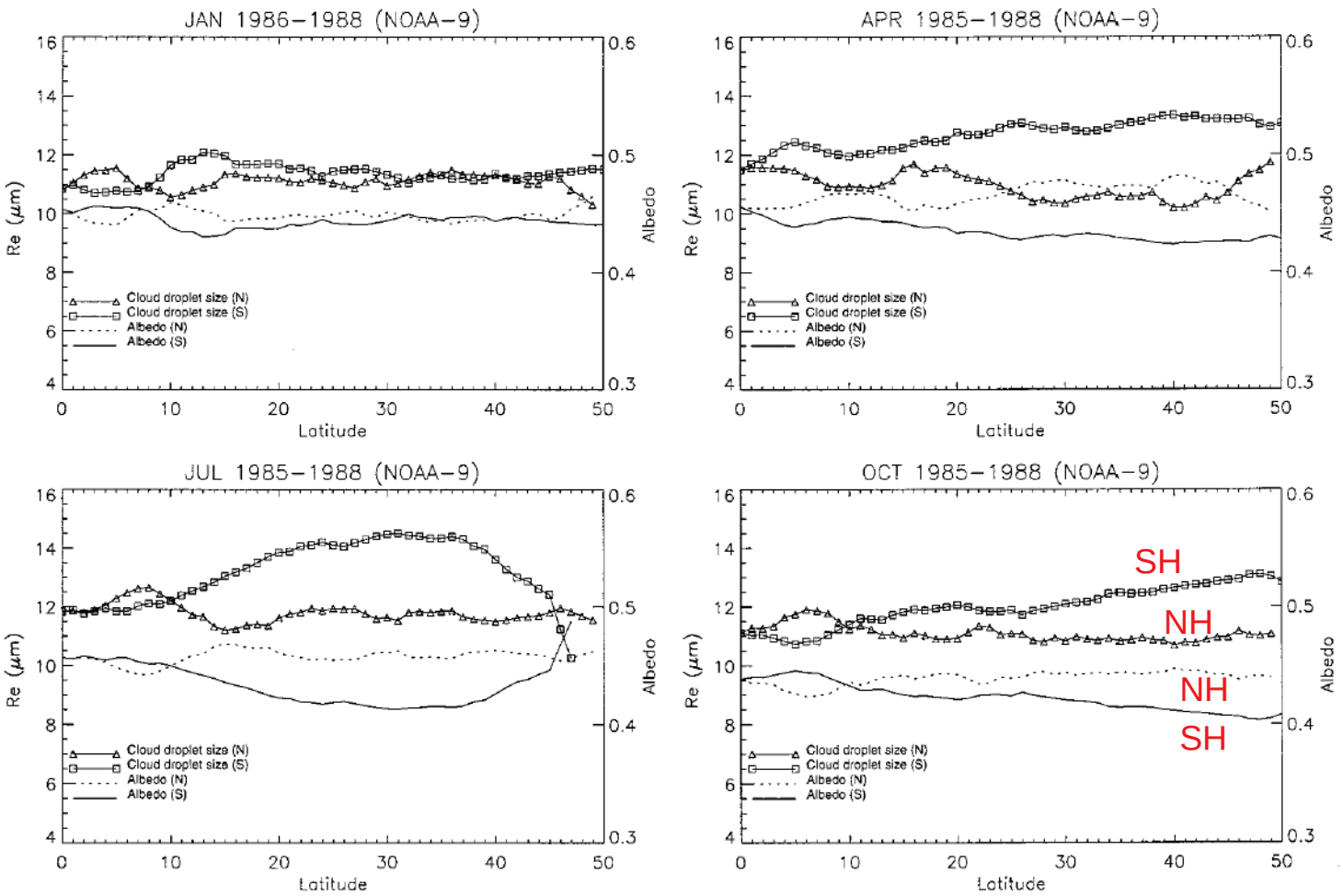


Han et al., 1998

Smaller effective radius in NH is negated by lower LWP.

EXAMINATION FOR INDIRECT EFFECT IN INTERHEMISPHERIC COMPARISONS

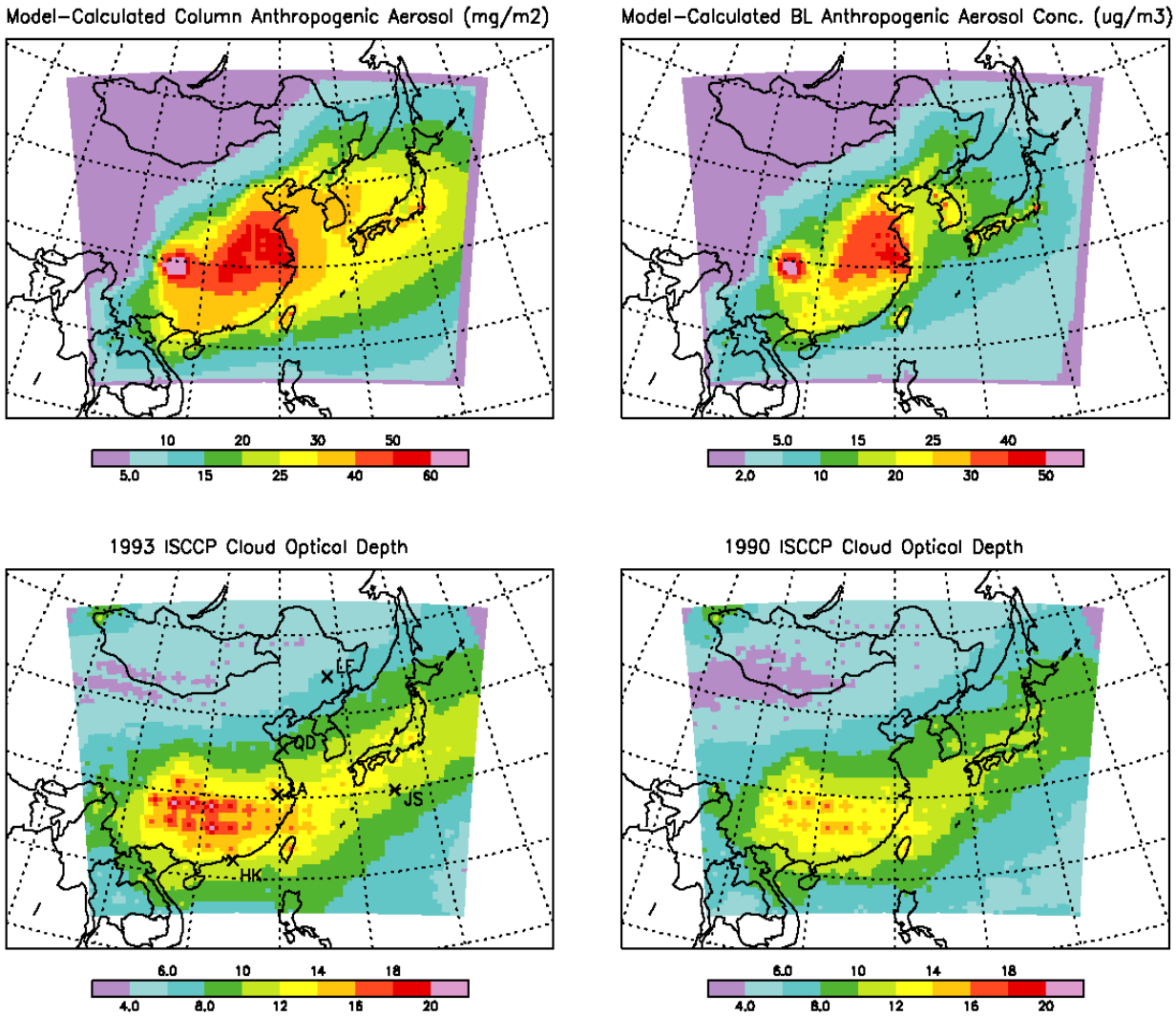
Zonal-mean cloud drop effective radius and albedo *for assumed constant LWP*



Han et al., 1998

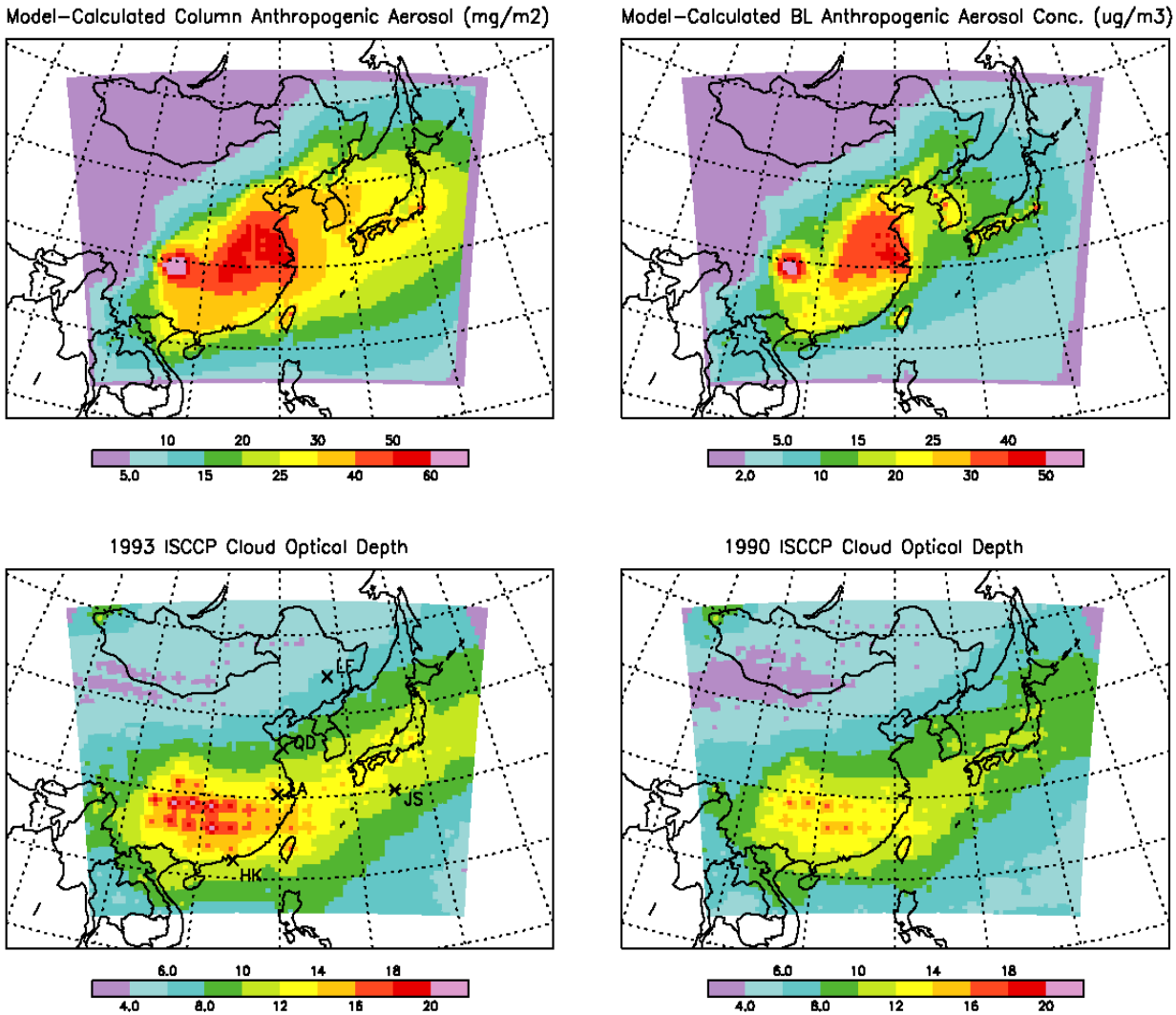
USING RESULTS FROM A CHEMICAL TRANSPORT MODEL TO IDENTIFY SITUATIONS OF HIGH AEROSOL LOADING TO PINPOINT AEROSOL INDIRECT EFFECT

ASSOCIATION DOES NOT NECESSARILY EQUAL CAUSALITY



Chameides et al., 2002

ASSOCIATION DOES NOT NECESSARILY EQUAL CAUSALITY



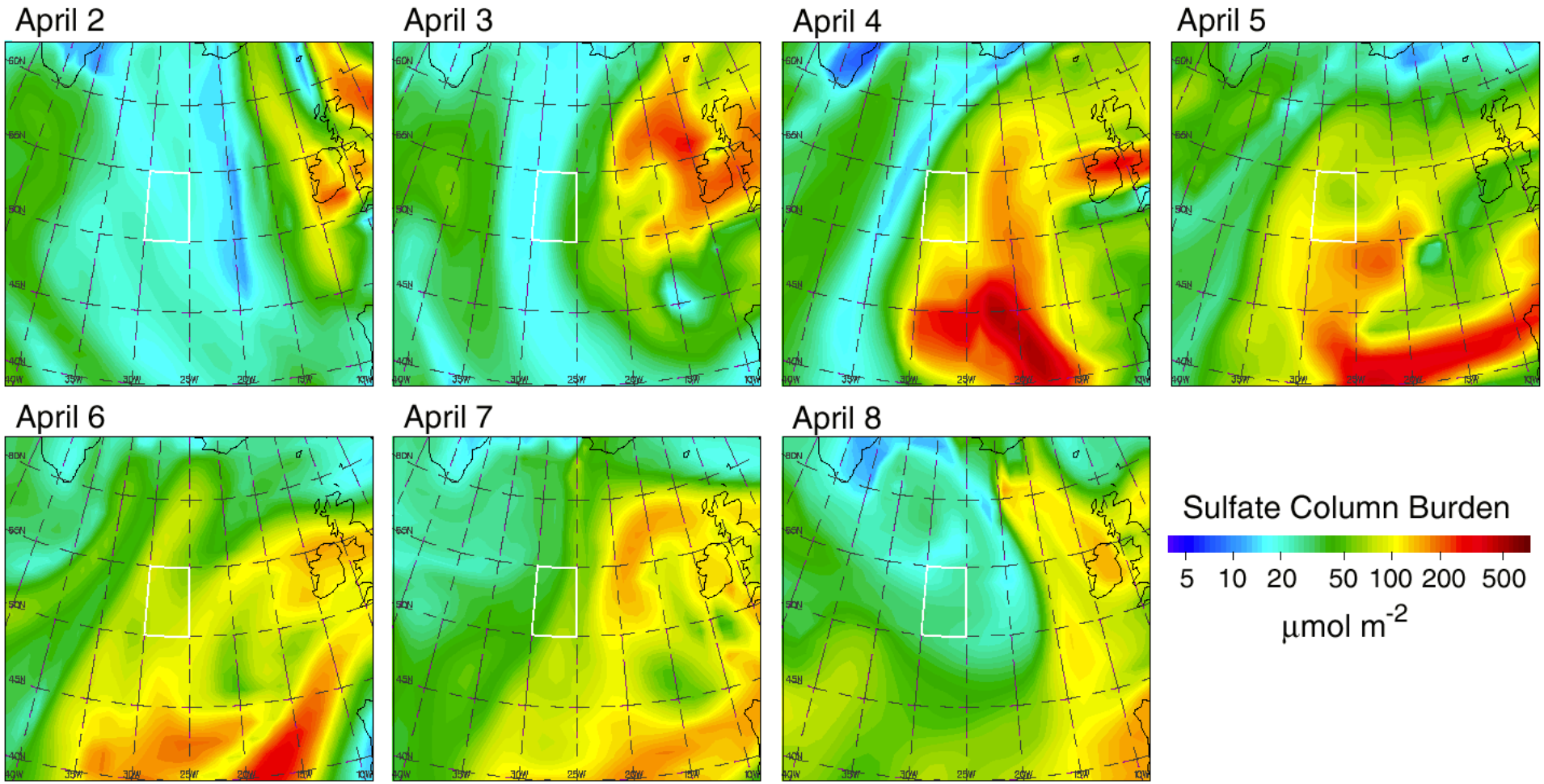
Chameides et al., 2002

"The dog in the sky eats the sun."

MODELED SULFATE COLUMN BURDEN

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

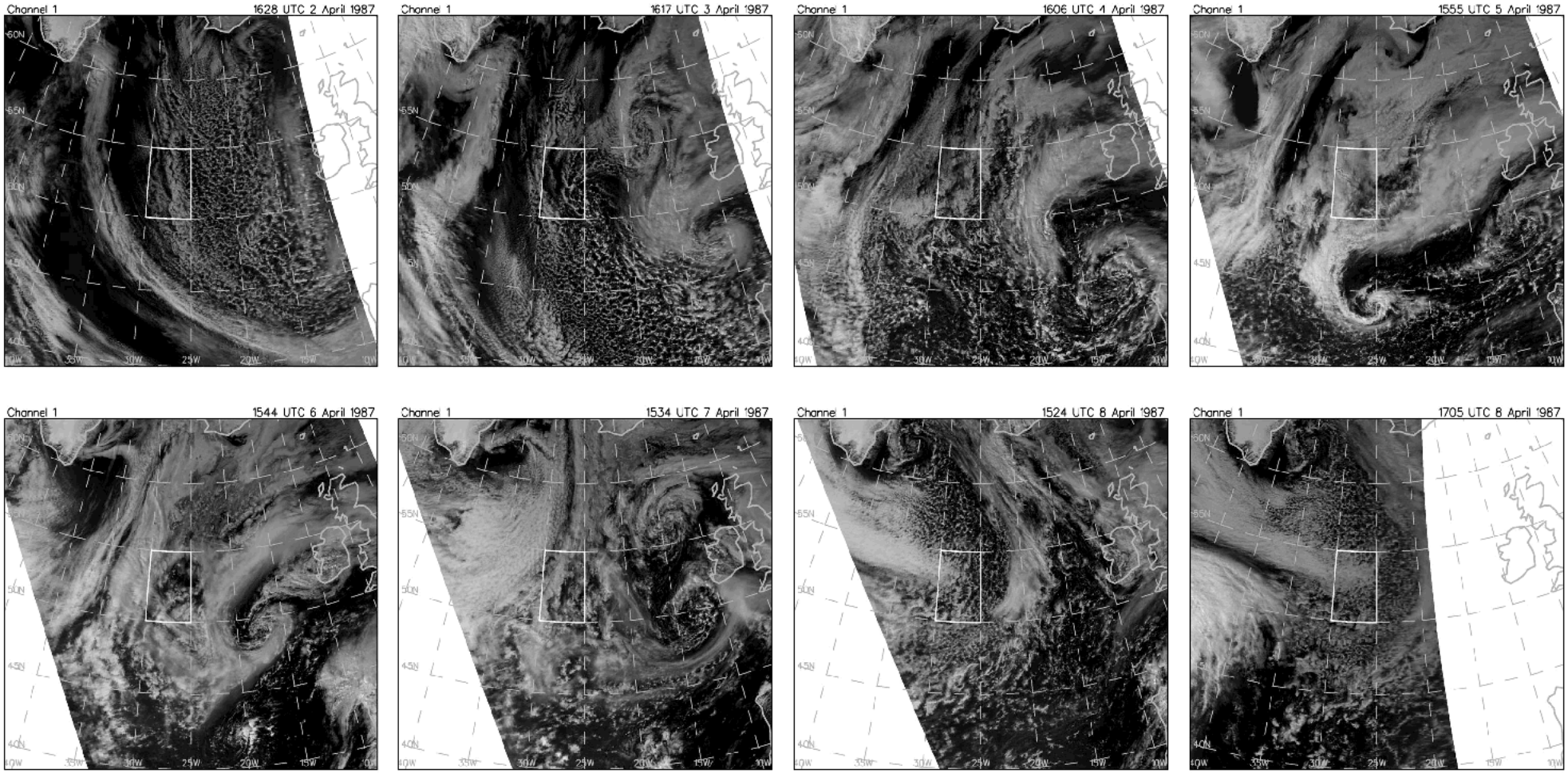
April 2-8, 1987



Schwartz, Harshvardhan & Benkovitz, PNAS, 2002

AVHRR IMAGES APRIL 2-8, 1987

Channel 1, Visible, 0.58-0.68 μm

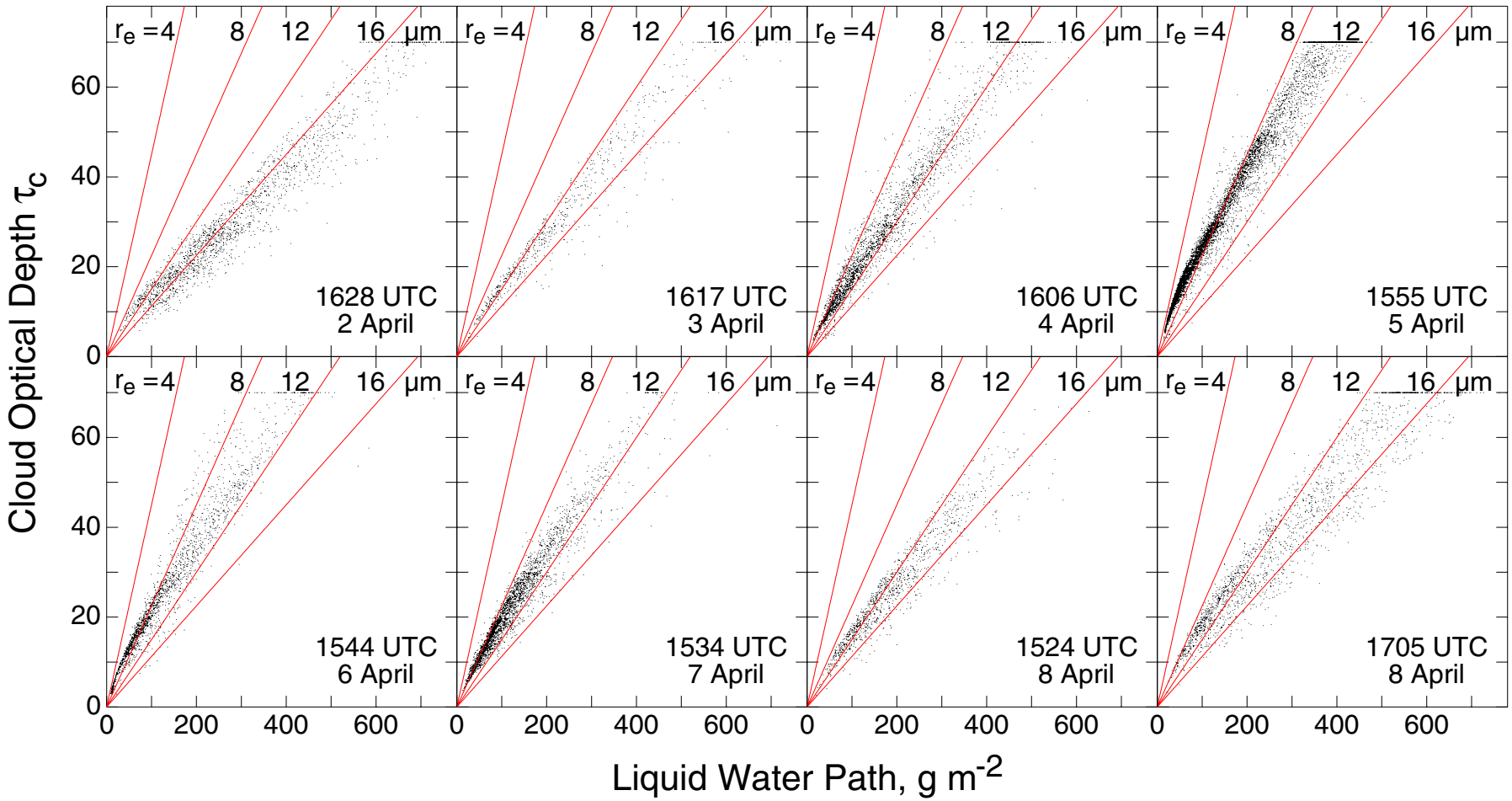


Harshvardhan, Schwartz, Benkovitz and Guo, J Atmos Sci, 2002

CLOUD OPTICAL DEPTH

Dependence on Liquid Water Path

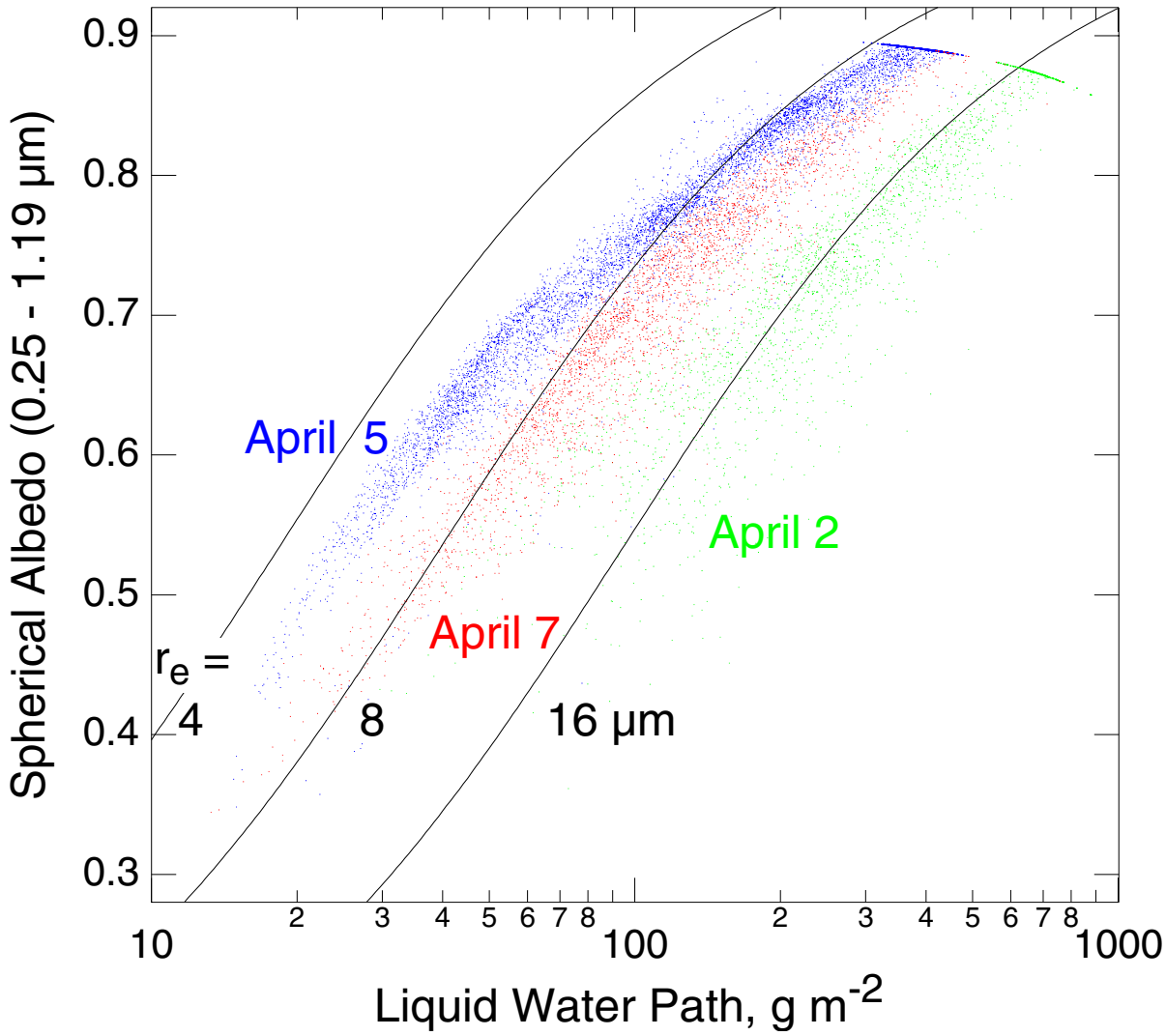
25°-30°W, 50°-55°N April 2-8, 1987



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

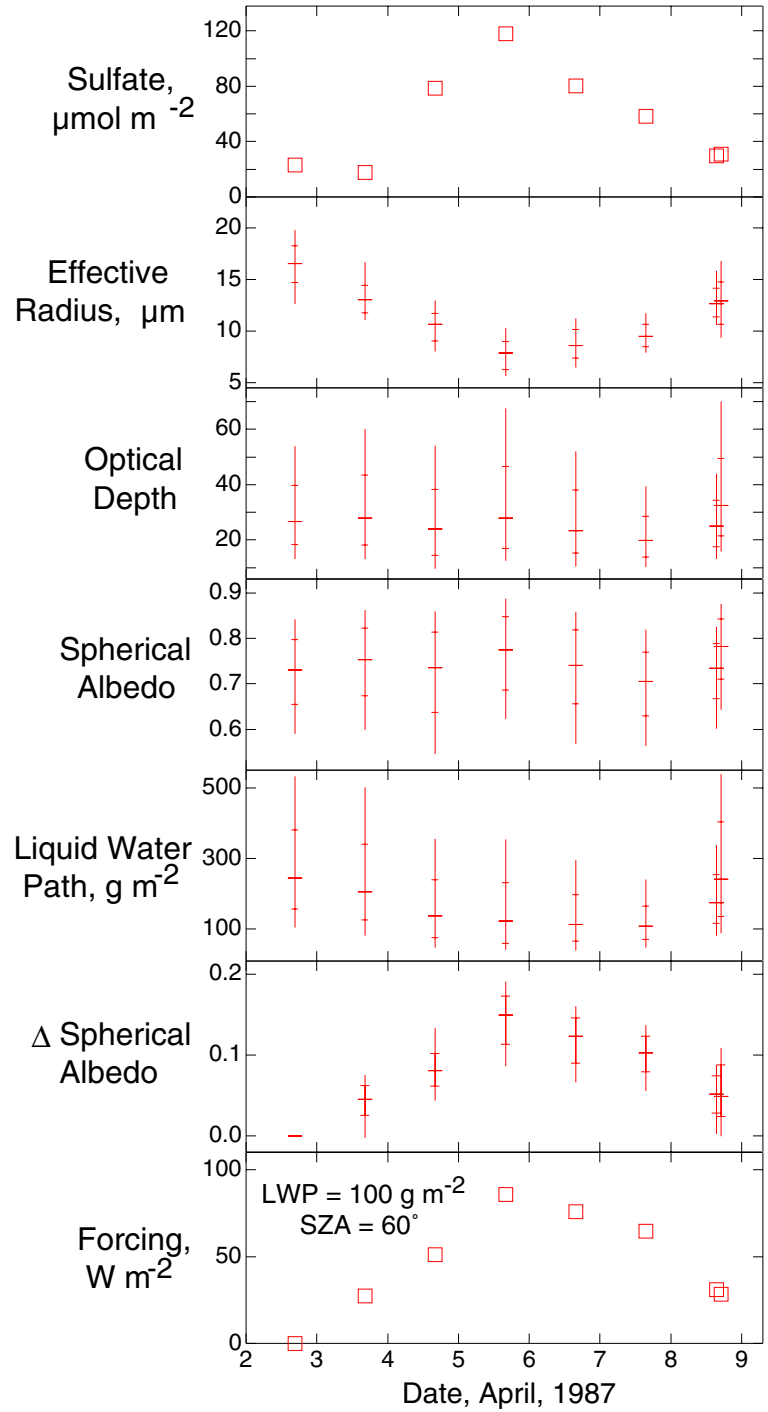
Mid North Atlantic (25-30°W, 50-55°N), April 2-8, 1987

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)

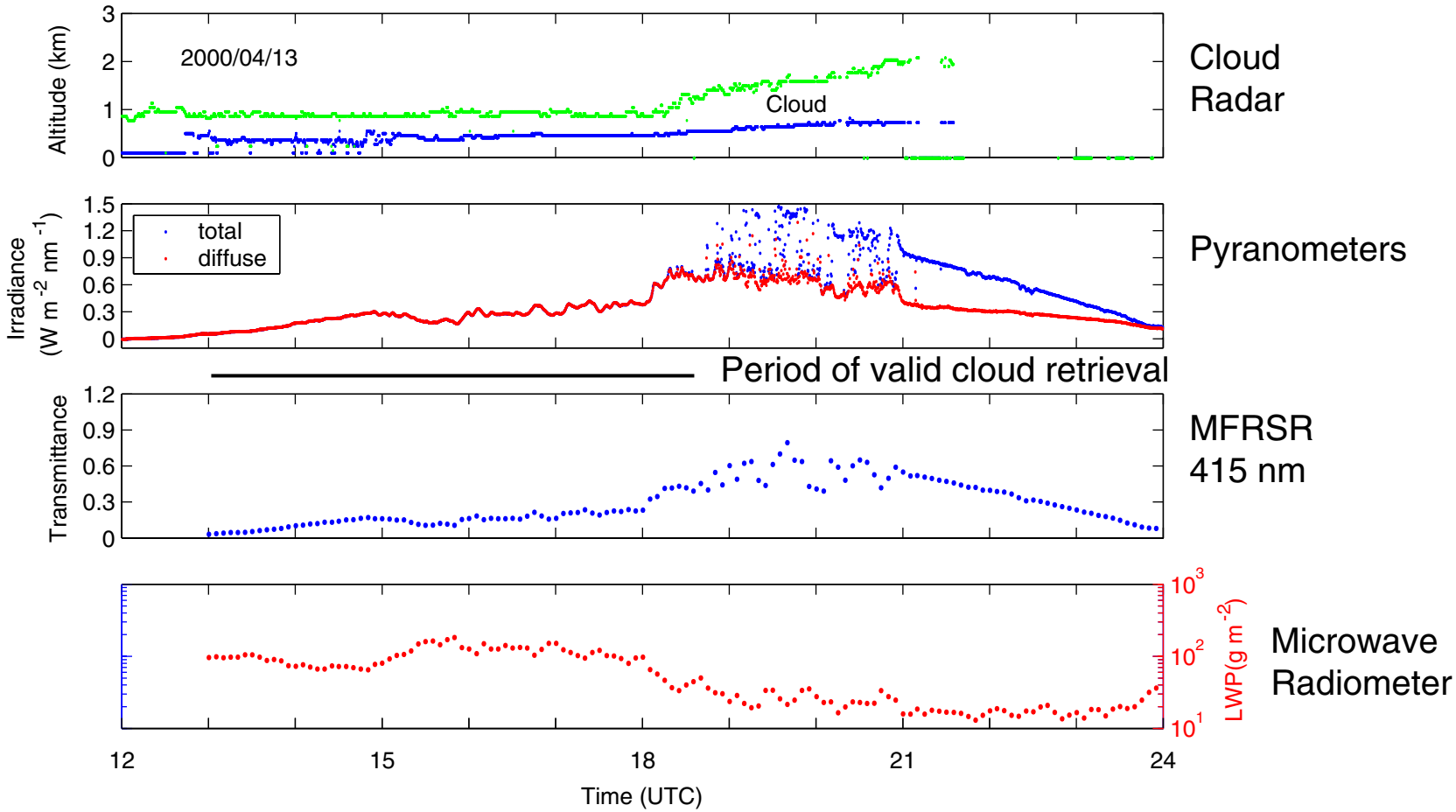
Forcing is calculated for median effective radius relative to April 2; solar zenith angle 60°; LWP 100 g m^{-2}



QUANTIFICATION OF AEROSOL INDIRECT FORCING IN GROUND-BASED REMOTE SENSING

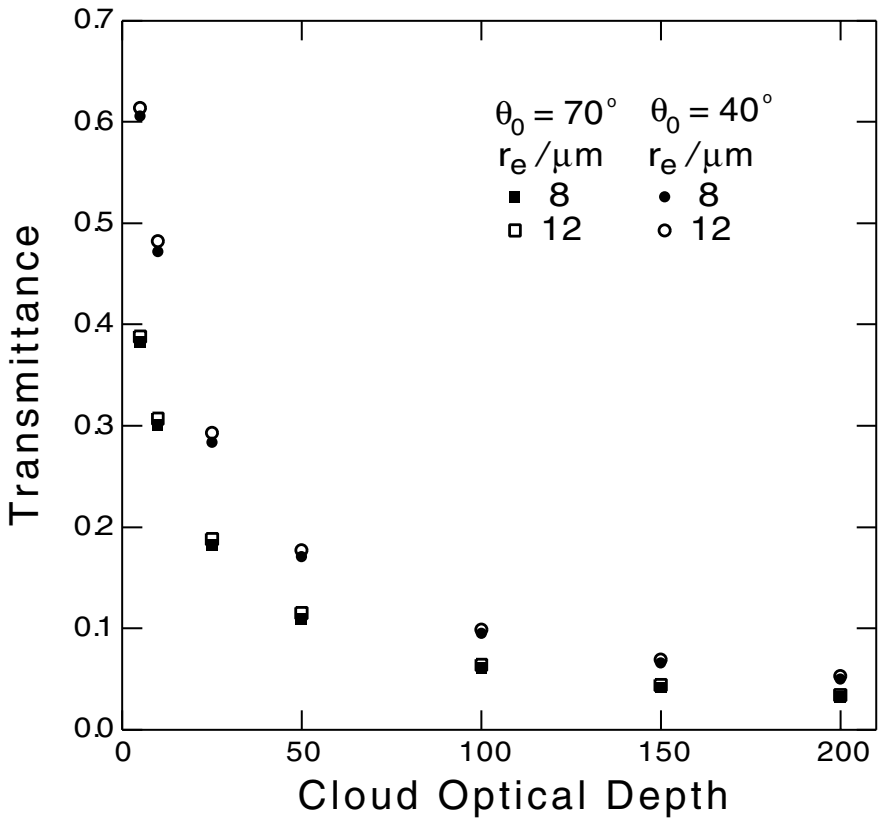
GROUND BASED REMOTE SENSING OF CLOUD PROPERTIES

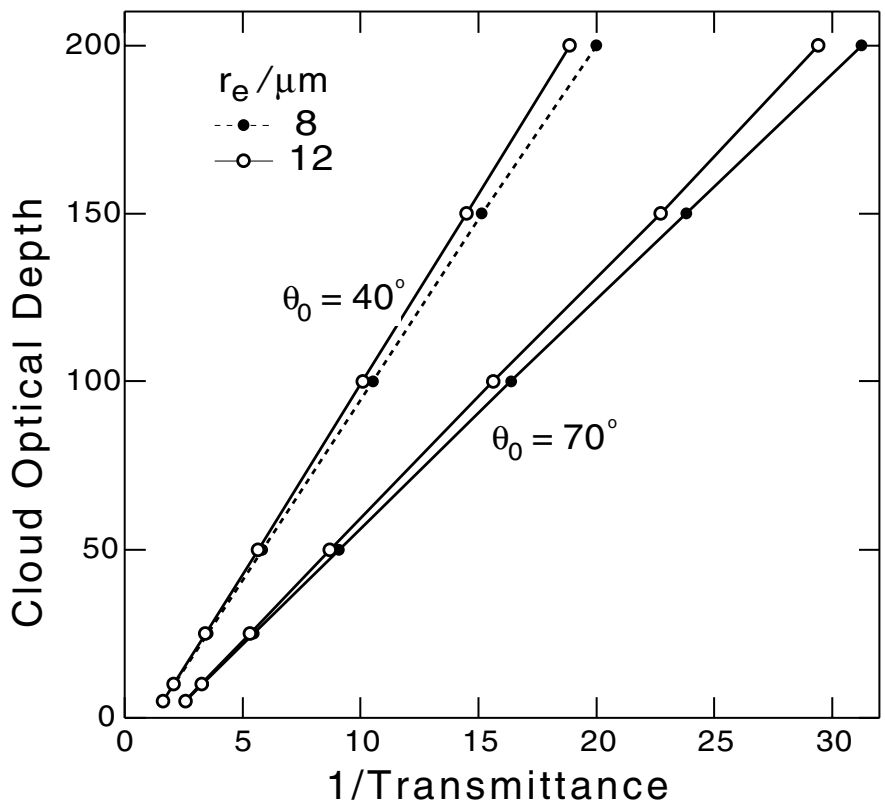
North Central Oklahoma, April 13, 2000 Local time = UTC - 6h



Kim, Schwartz, Miller, and Min, JGR, 2003

RELATION BETWEEN ATMOSPHERIC TRANSMITTANCE AND CLOUD OPTICAL DEPTH



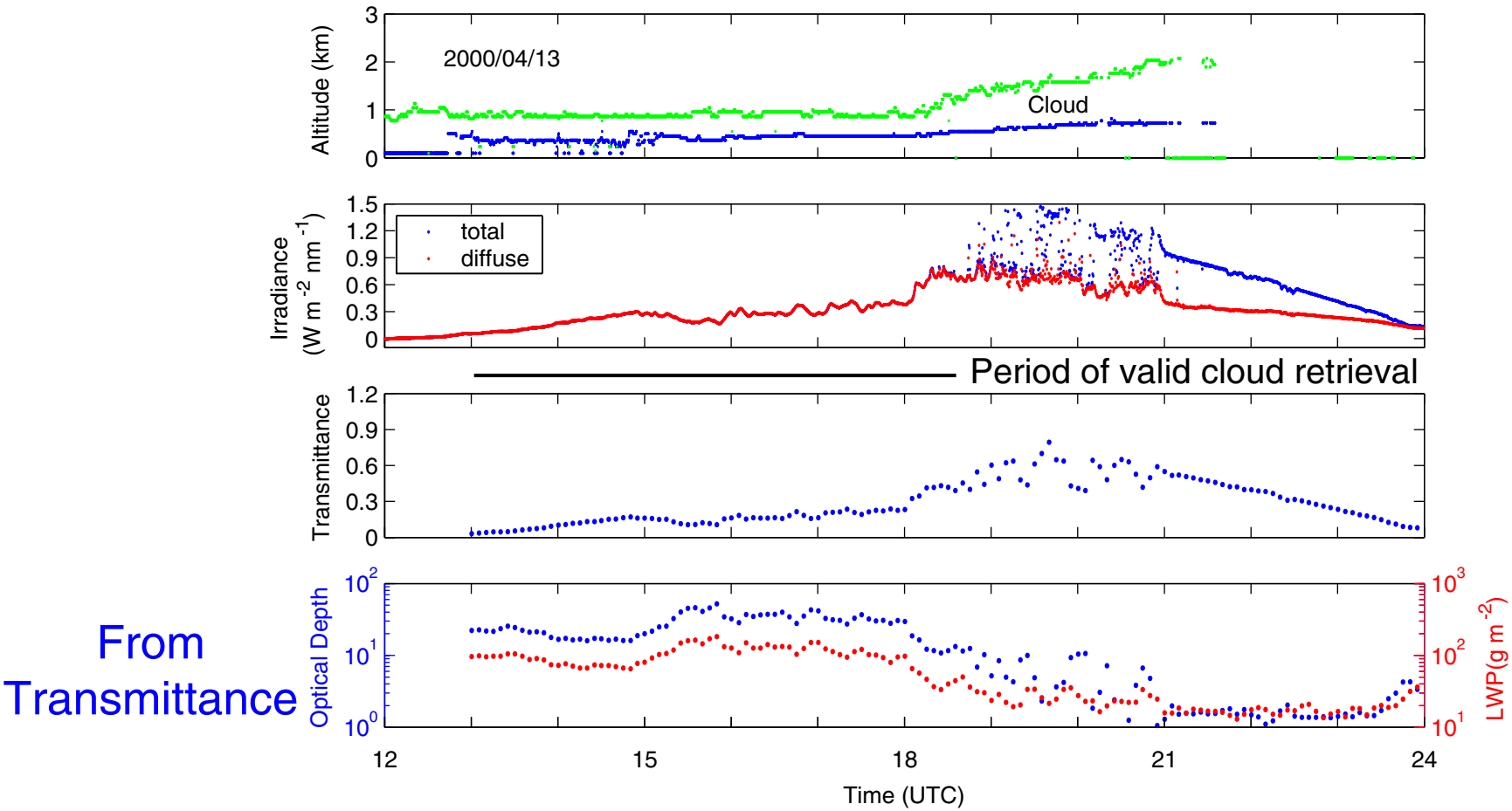


Kim, Schwartz, Miller, and Min, JGR, 2003

SBDART Radiation transfer model

GROUND BASED REMOTE SENSING OF CLOUD PROPERTIES

North Central Oklahoma, April 13, 2000 – Local time = UTC - 6



Kim, Schwartz, Miller, and Min, JGR, 2003

MEASURING CLOUD DROP EFFECTIVE RADIUS BY GROUND BASED REMOTE SENSING

Effective radius: Cloud or aerosol property important for radiative transfer

For a homogeneous volume

$$r_e \equiv \frac{\mu_3}{\mu_2} \equiv \frac{\int N(r) r^3 dr}{\int N(r) r^2 dr}$$

For a cloud

$$r_e = \frac{\iint N(r,z) r^3 dr dz}{\iint N(r,z) r^2 dr dz}$$

Cloud liquid water path (LWP) (microwave radiometer)

$$L = \frac{4\pi}{3} \rho_w \int\int r^3 N(r,z) dr dz$$

Cloud optical depth (MFRSR)

$$\tau_c = \iint \pi r^2 Q_e(r) N(r,z) dr dz$$

Mie scattering efficiency

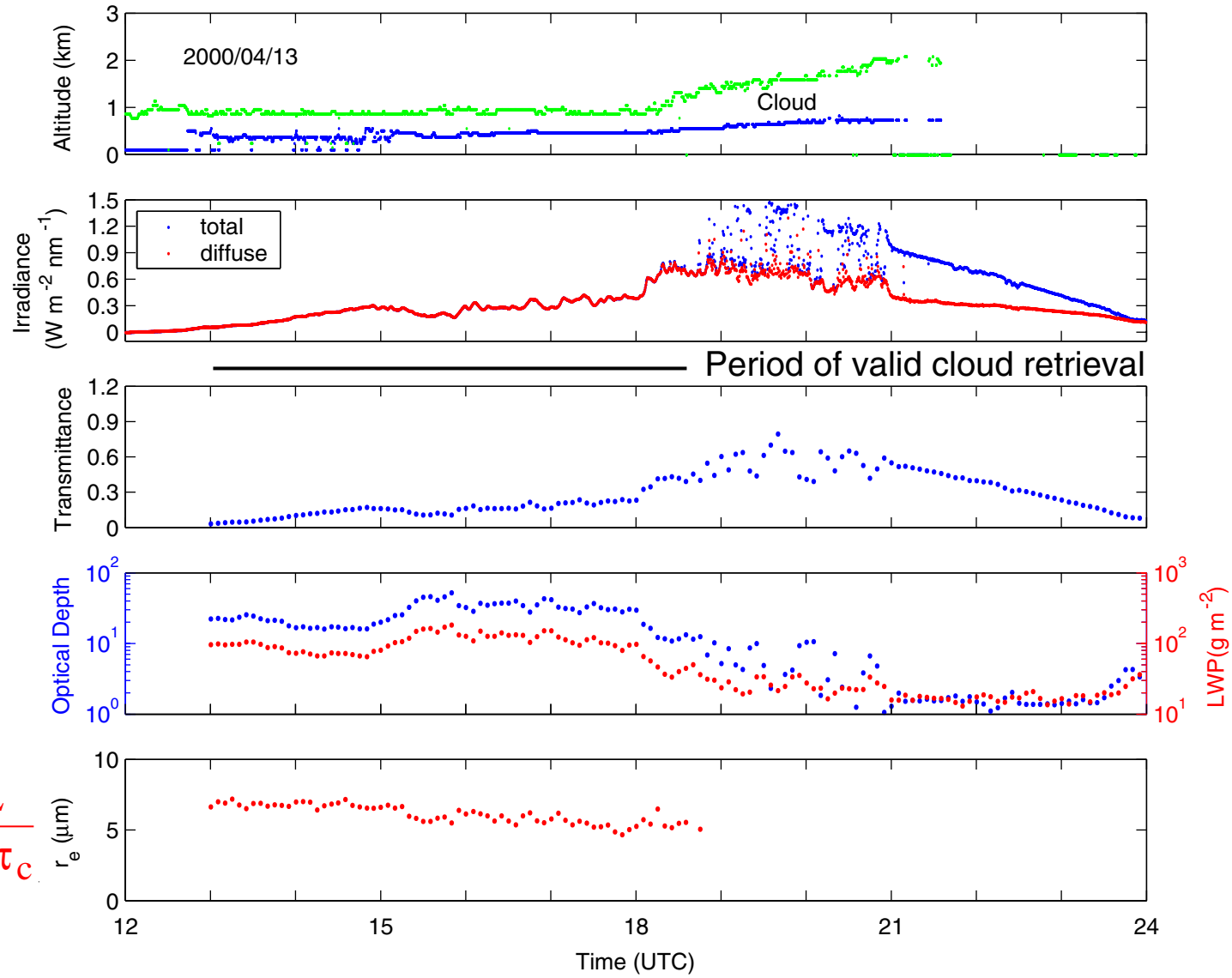
$$Q_e \approx 2 \text{ for } r >> \lambda$$

Whence

$$r_e \approx \frac{3}{2} \frac{L}{\rho_w \tau_c}$$

GROUND BASED REMOTE SENSING OF CLOUD PROPERTIES

North Central Oklahoma, April 13, 2000 – Local time = UTC - 6



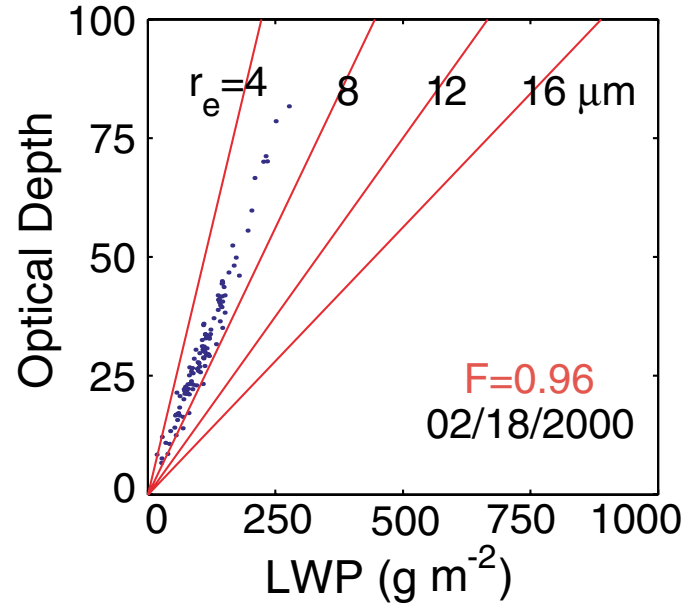
$$r_e \approx \frac{3}{2}\frac{L}{\rho_w \tau_c}$$

Kim, Schwartz, Miller, and Min, JGR, 2003

CLOUD OPTICAL DEPTH VS. LIQUID WATER PATH

North Central Oklahoma, 2000

$$\tau_c \approx \frac{3}{2}\frac{L}{\rho_w r_e}$$



Kim, Schwartz, Miller, and Min, JGR, 2003

Optical depth is highly correlated with and strongly dependent on liquid water path.

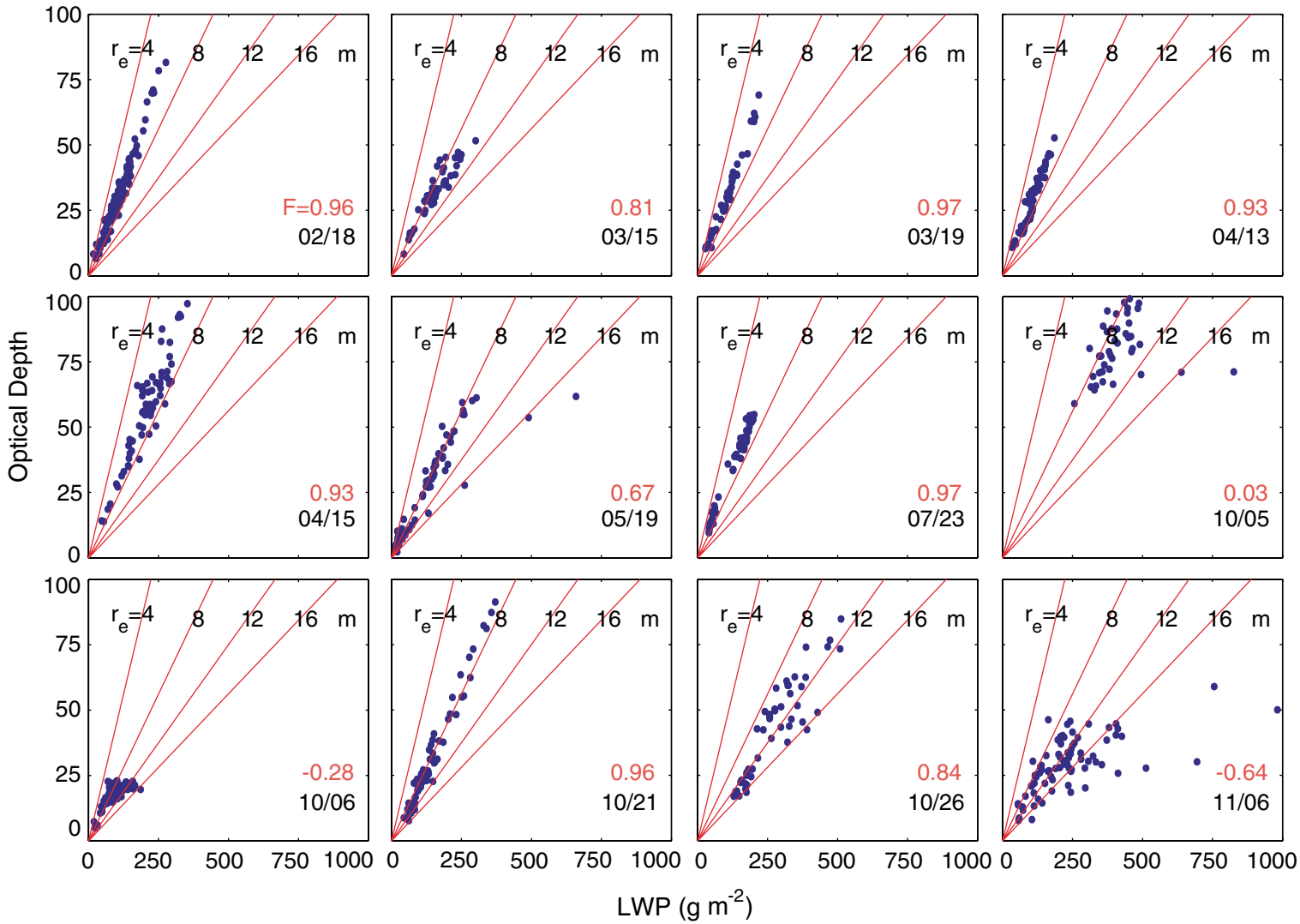
Tight cluster of points about a diagonal line through the origin is indicative of constant effective radius over the day.

Slope is inversely proportional to effective radius.

F, fraction of variance accounted for by regression = 96%.

CLOUD OPTICAL DEPTH VS. LIQUID WATER PATH

North Central Oklahoma, 2000



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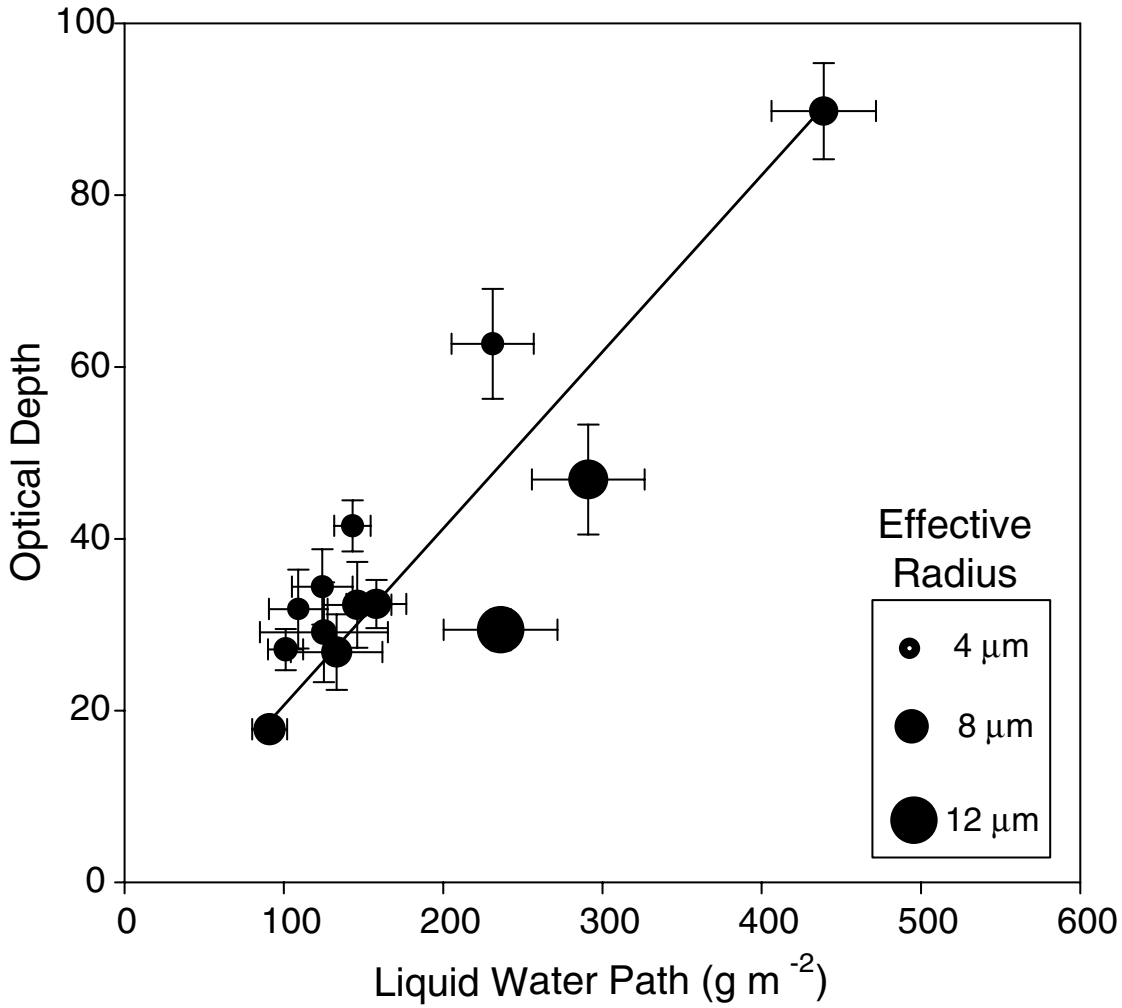
F, fraction of variance accounted for by regression, mainly > 80%.

CLOUD OPTICAL DEPTH VS. LIQUID WATER PATH

North Central Oklahoma, 2000, aggregated by days



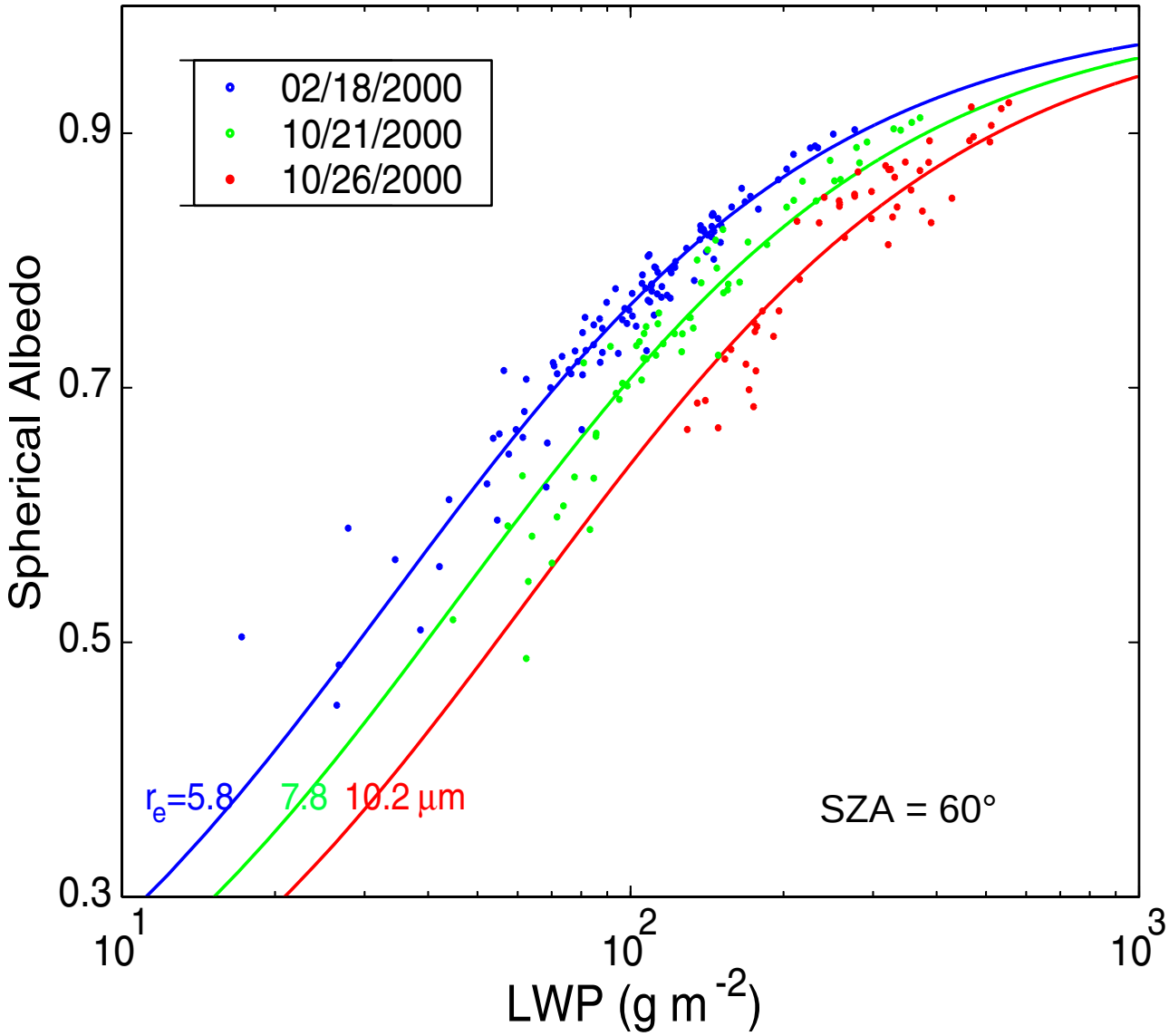
Kim, Schwartz, Miller, and Min, JGR, 2003

Fraction of variance accounted for by regression, 74%.

Days with smaller radii have a greater optical depth for a given LWP.

CLOUD ALBEDO CALCULATED FROM MEASURED EFFECTIVE RADIUS AND LIQUID WATER PATH

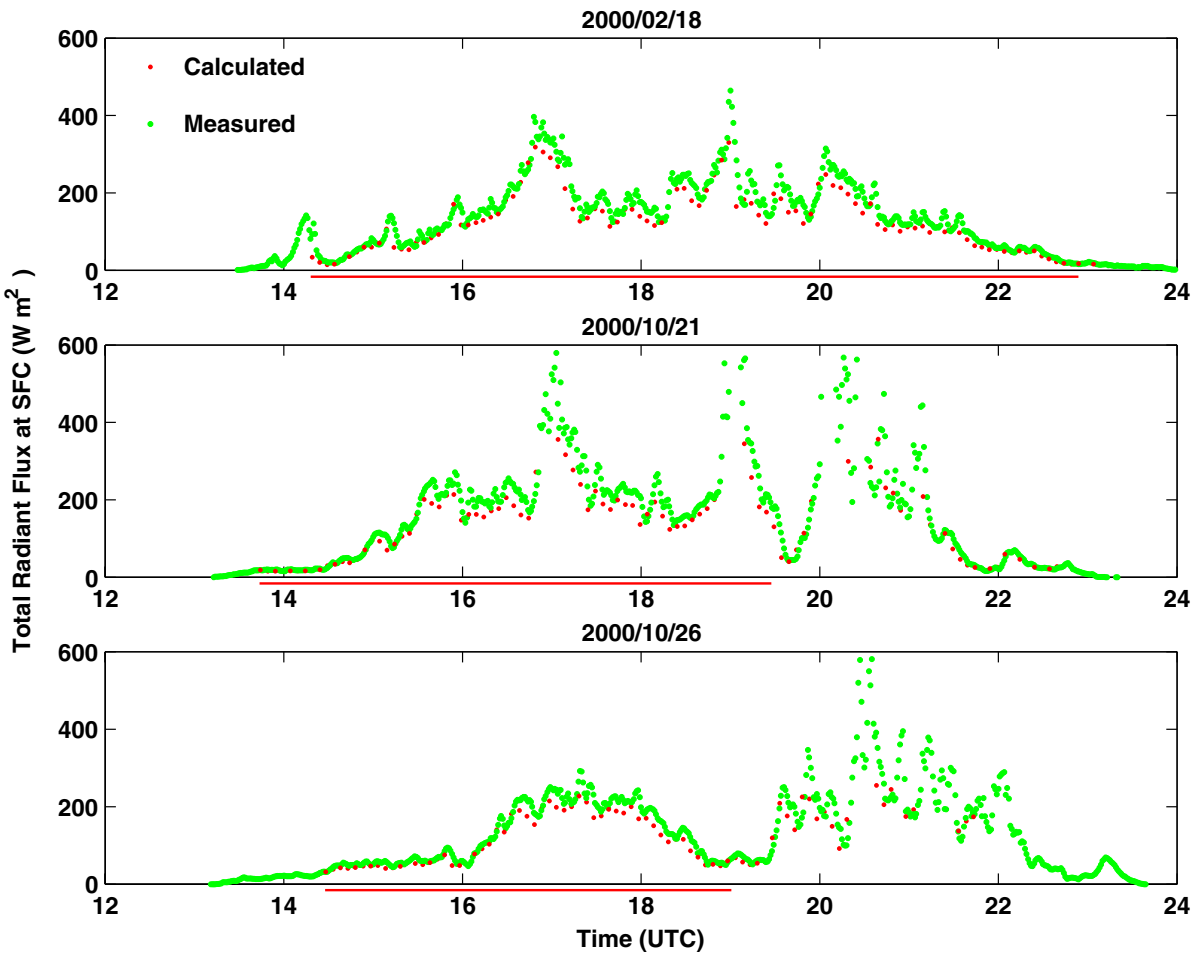
North Central Oklahoma



Kim, Schwartz, Miller, and Min, JGR, 2003

COMPARISON OF MEASURED AND MODELED DOWNWELLING SURFACE IRRADIANCE

North Central Oklahoma, uniform overcast sky

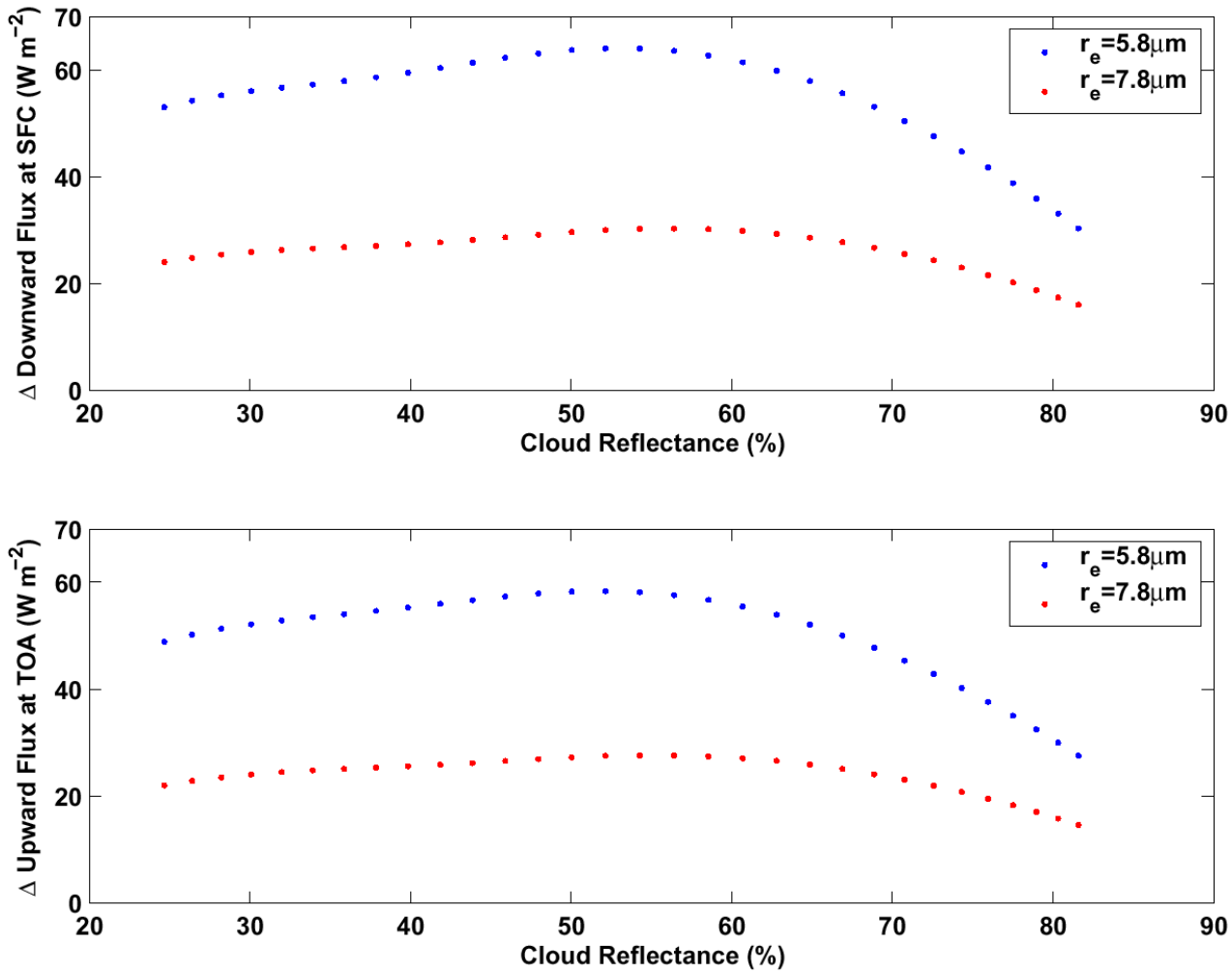


Kim and Schwartz, in preparation, 2004

SBDART Radiation transfer model.
LWP from microwave radiometer; optical depth from narrow band radiometry at 415 nm.

CALCULATED SURFACE AND TOP-OF-ATMOSPHERE FORCING RELATIVE TO REFERENCE EFFECTIVE RADIUS (r_e = 10.2 μm)

Dependence on Cloud-top Reflectance

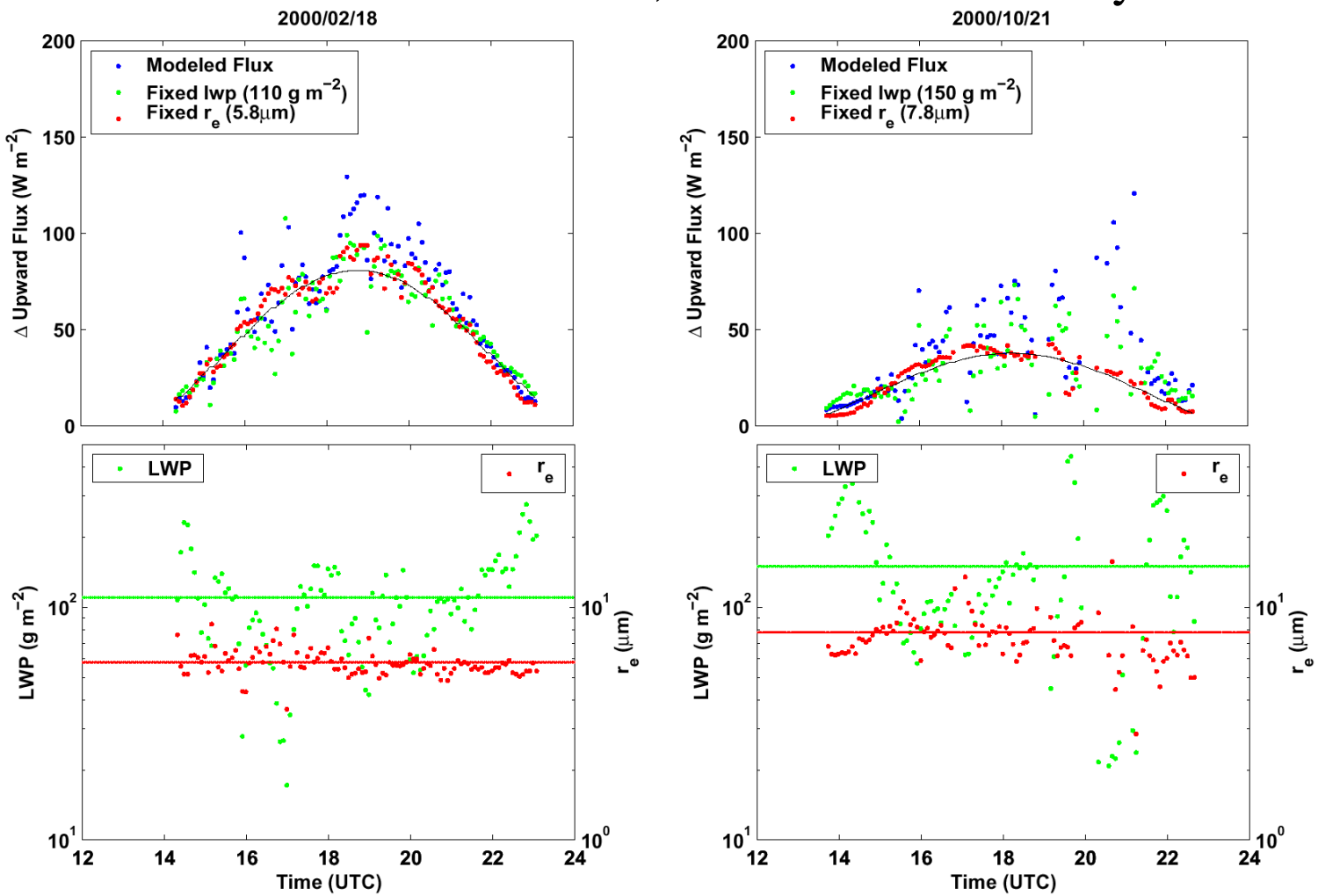


Kim and Schwartz, in preparation, 2004

SBDART Radiation transfer model; SZA = 60˚.

CALCULATED TOP-OF-ATMOSPHERE FORCING RELATIVE TO REFERENCE EFFECTIVE RADIUS (r_e = 10.2 μm)

North Central Oklahoma, uniform overcast sky



Kim and Schwartz, in preparation, 2004

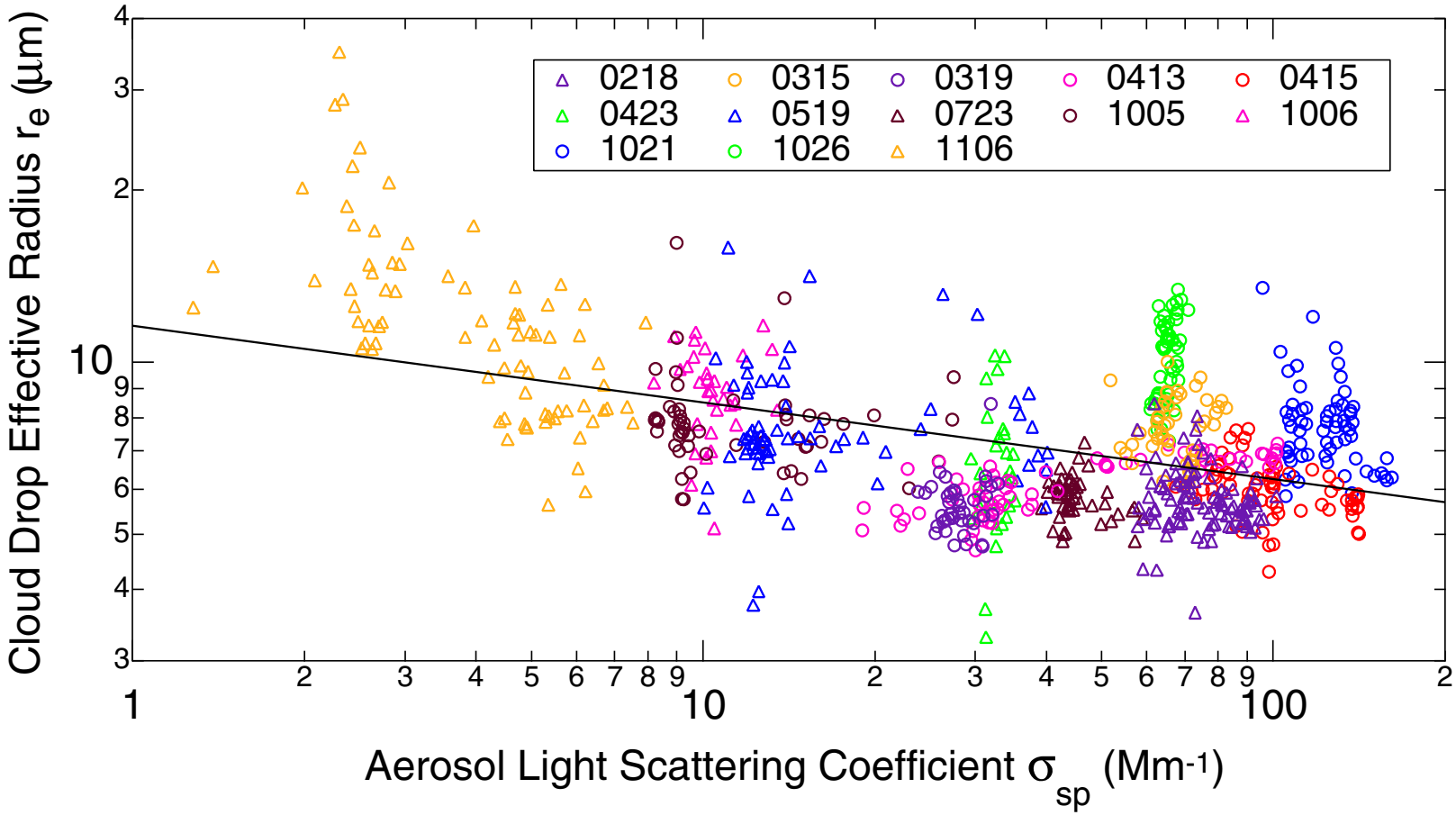
SBDART Radiation transfer model.
LWP from microwave radiometer; optical depth from narrow band radiometry at 415 nm.

CORRELATION OF CLOUD DROP EFFECTIVE RADIUS AND AEROSOL LIGHT SCATTERING COEFFICIENT

North Central Oklahoma

All days in 2000 meeting complete overcast criterion

$R^2 = 0.24$



Kim, Schwartz, Miller, and Min, JGR, 2003

CONCLUDING REMARKS

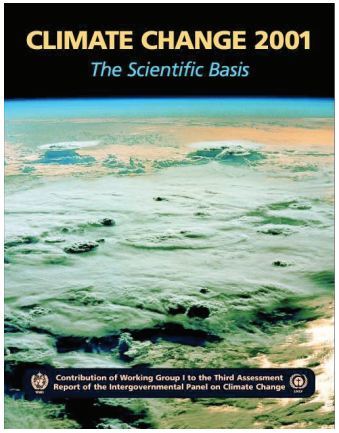
UNCERTAINTY BUDGET FOR *INDIRECT* FORCING BY INDUSTRIAL AEROSOLS

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}.			**∞**

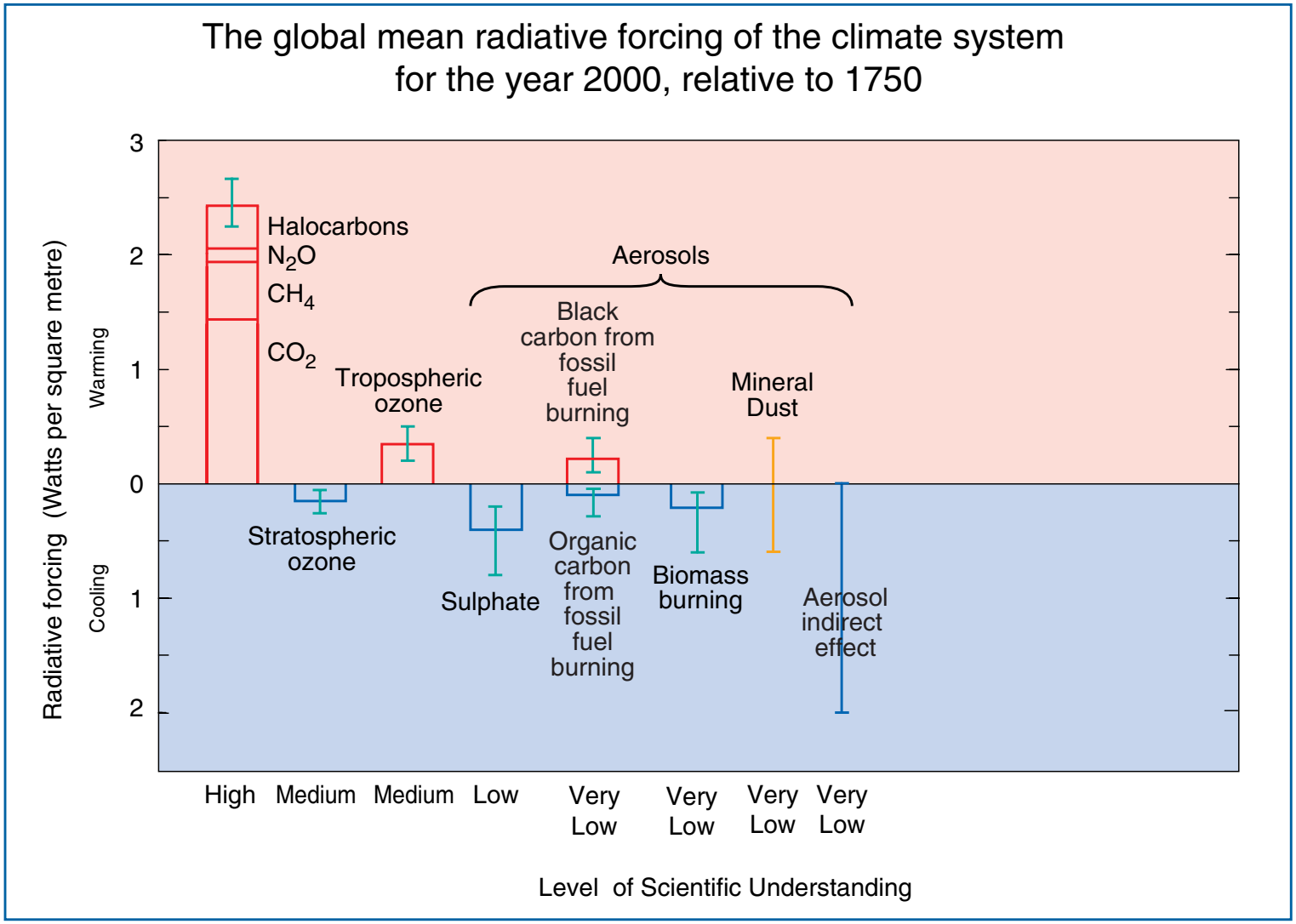
Modified from Penner et al., IPCC, 2001

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.

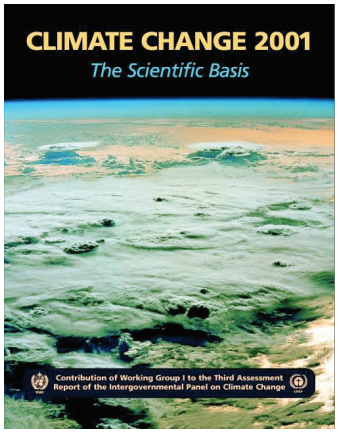


RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)
GHG's and aerosol direct and indirect effects



Summary for Policymakers

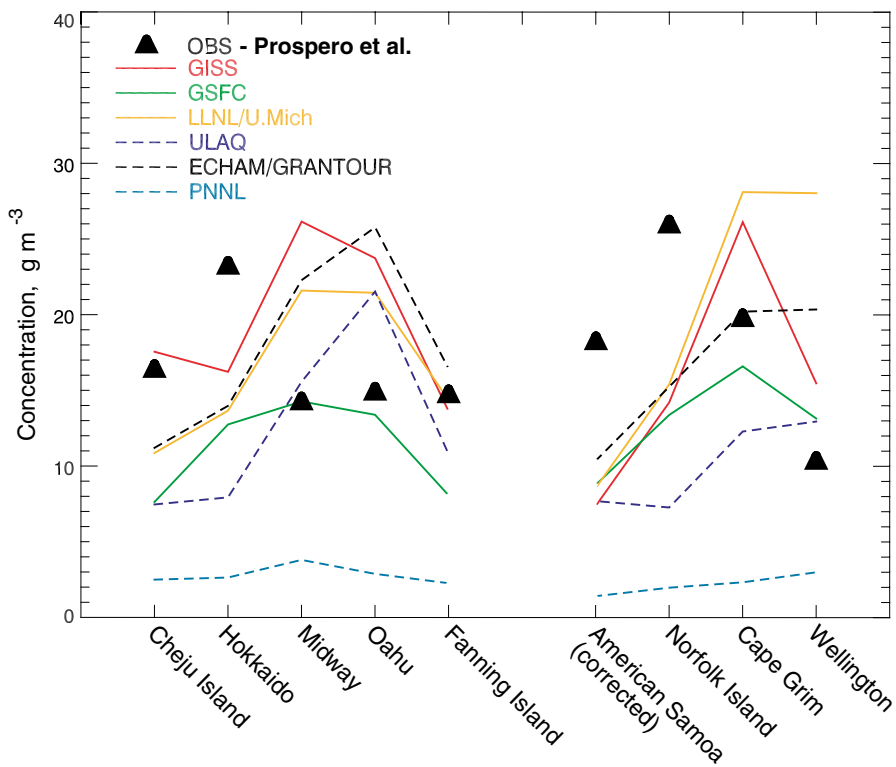
A Report of Working Group I of the Intergovernmental Panel on Climate Change

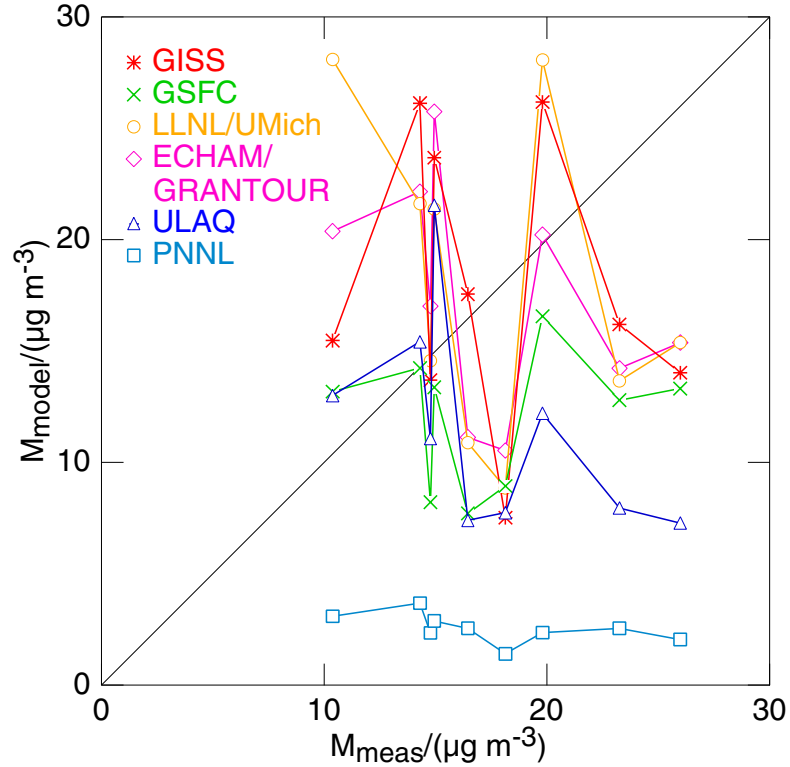


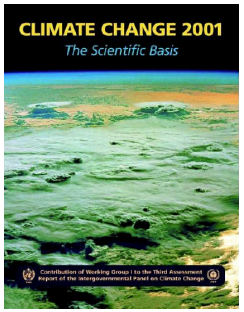
SEASALT AEROSOL MASS CONCENTRATION

Modeled and observed annual concentrations

From IPCC (2001) intercomparison







"After throwing out the burdens from models that were outliers in terms of their comparison with observations, the model results for sea salt still differed by a factor of 4.9 and 5.3, for diameter less than and greater than 2 µm, respectively. . . . In the upper troposphere . . . the range increased to as much as a factor of 20 or more."

WHY SO LARGE UNCERTAINTY IN AEROSOL FORCING?

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

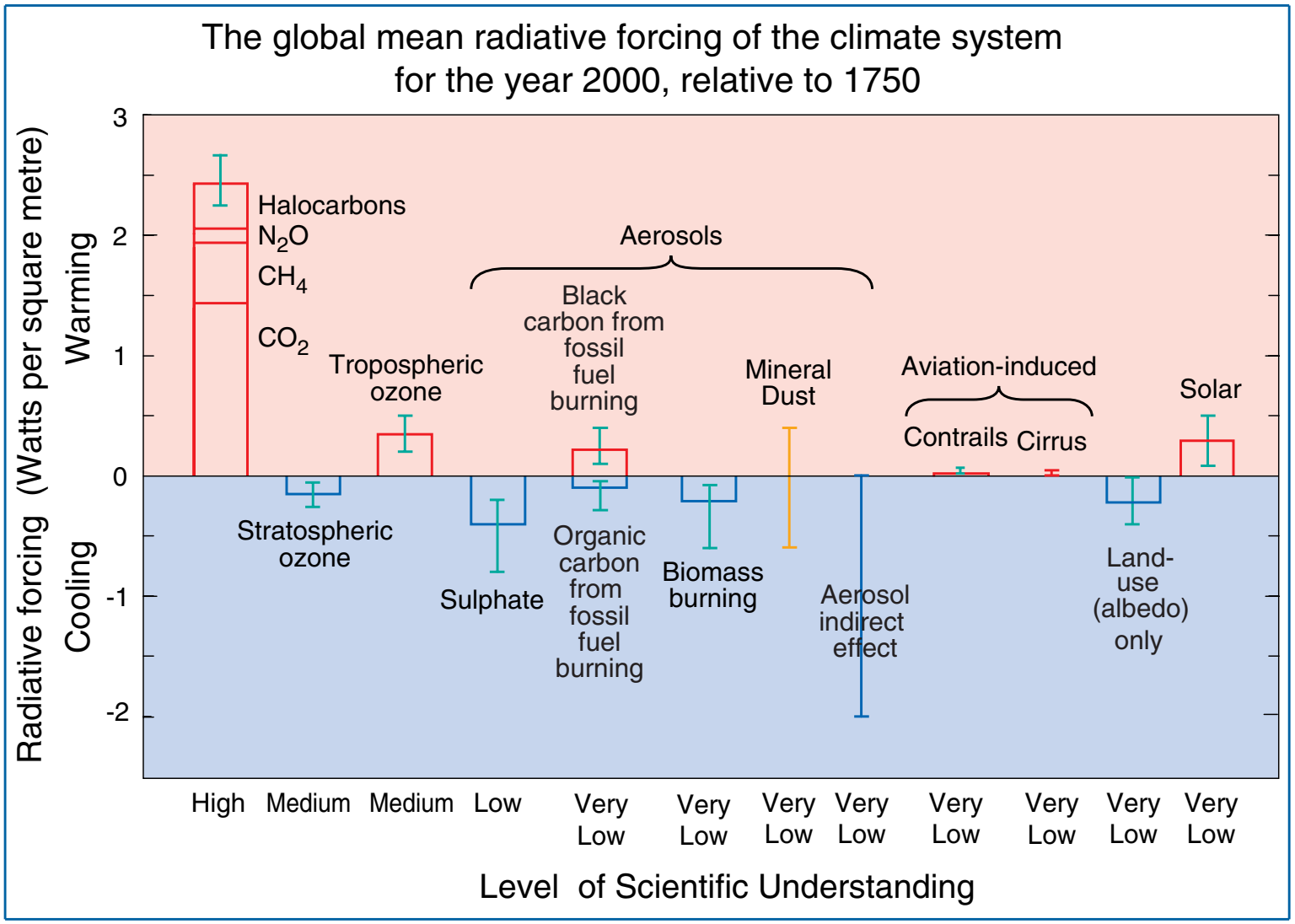
  Mass loading and chemical and microphysical properties and cloud nucleating properties of anthropogenic aerosols, and geographical distribution.

  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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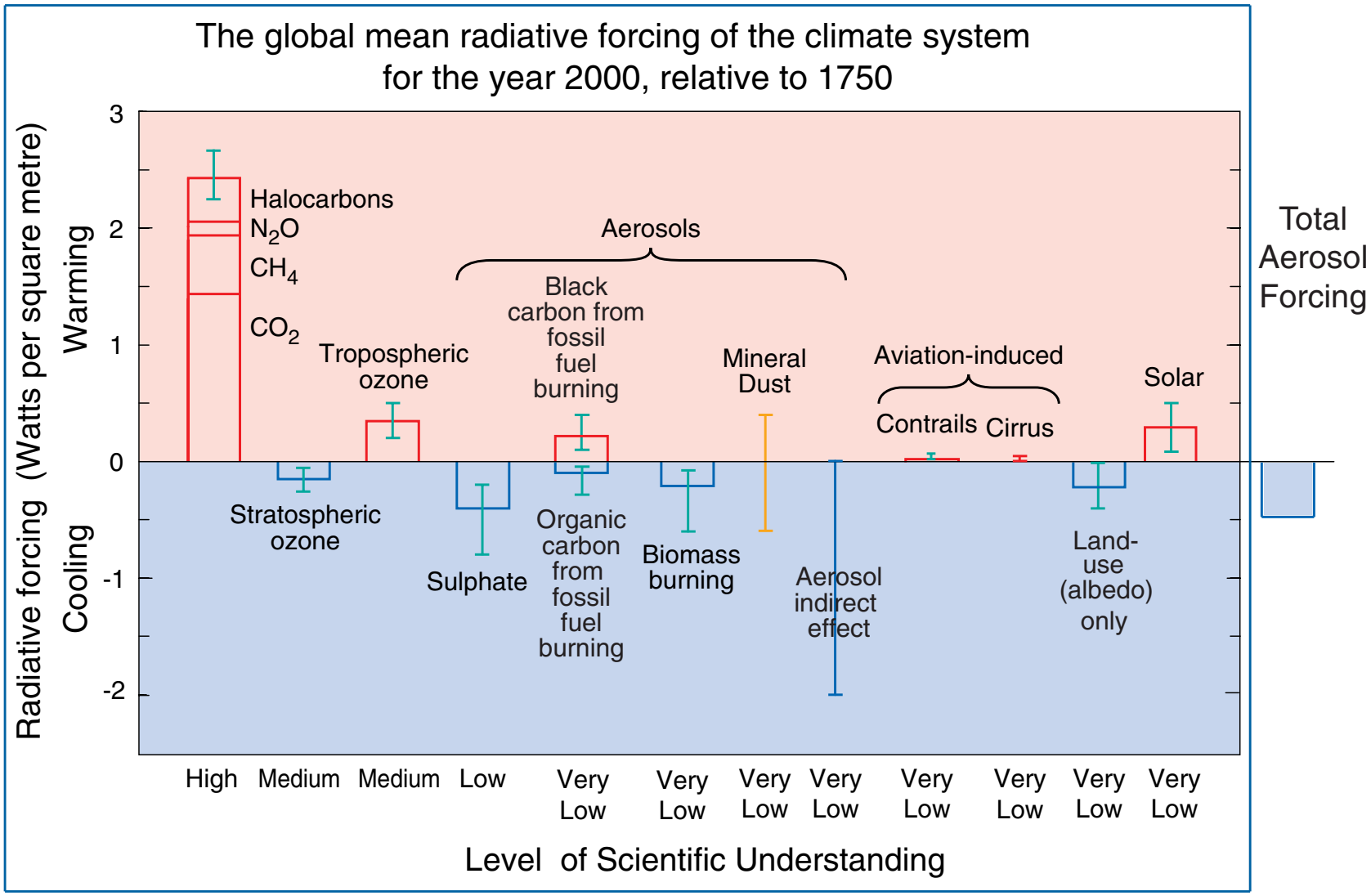
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)

With total aerosol forcing



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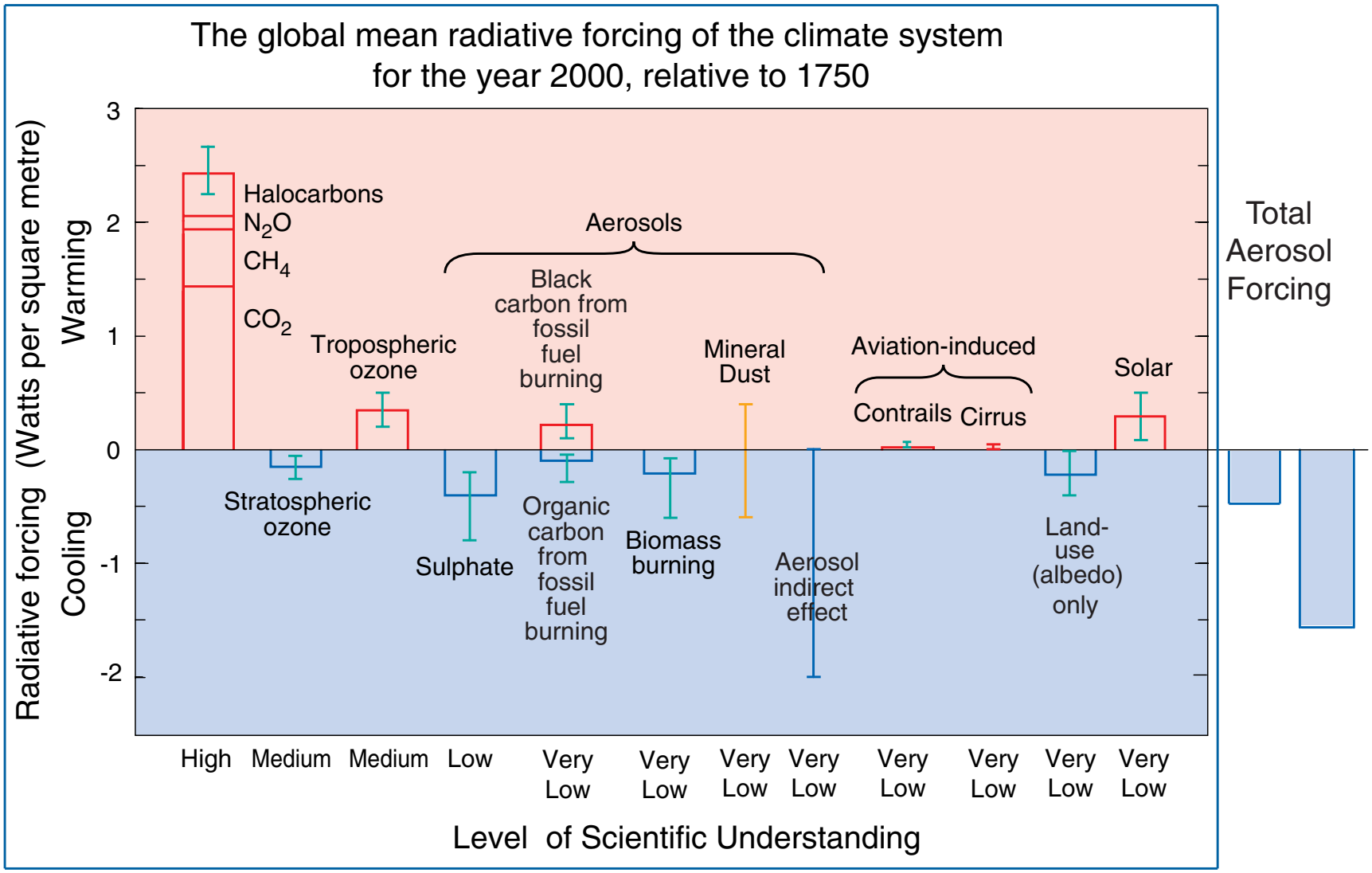
IPCC (2001)

With total aerosol forcing



Summary for Policymakers

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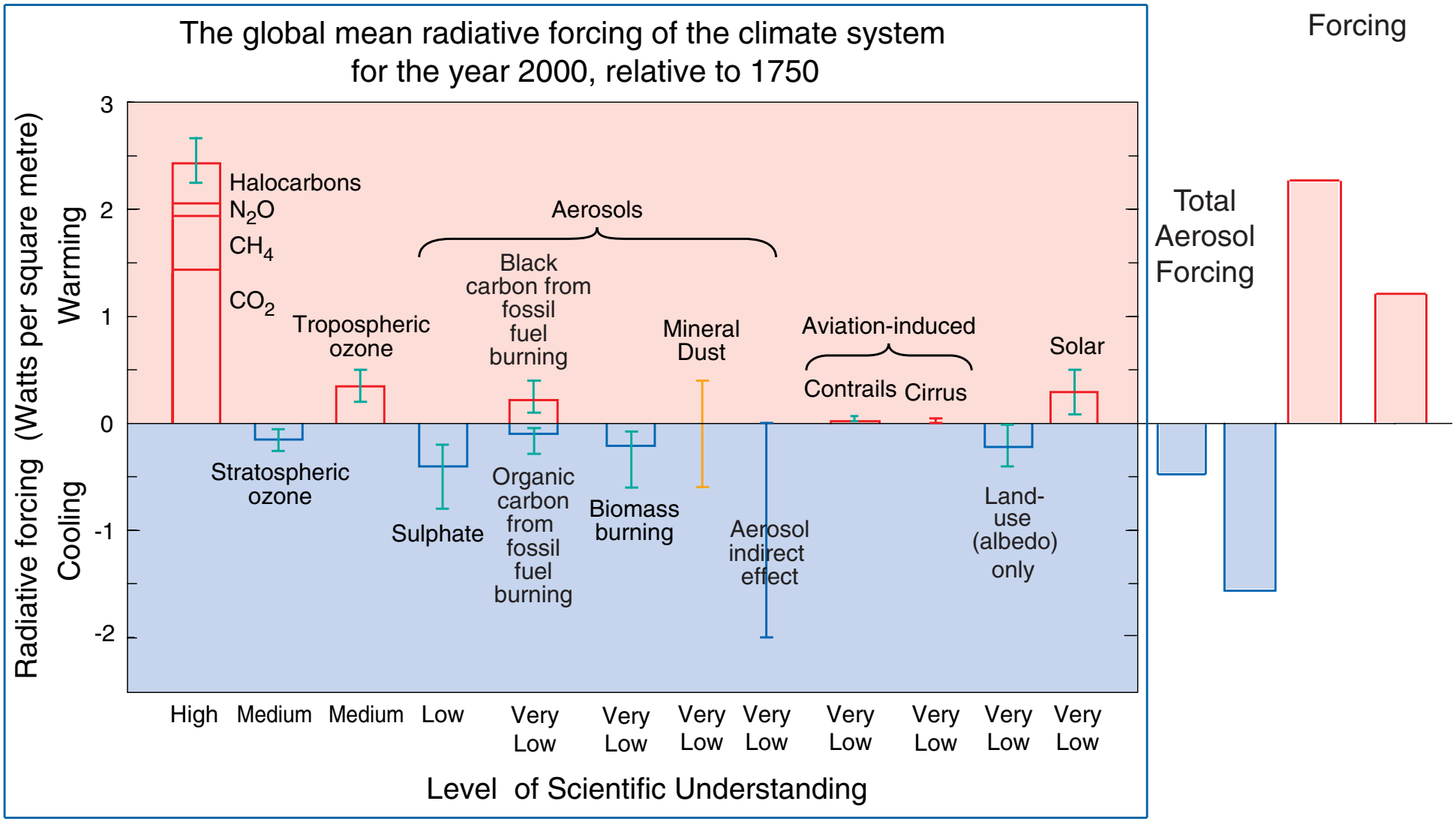
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)

With total aerosol forcing and total forcing



Summary for Policymakers

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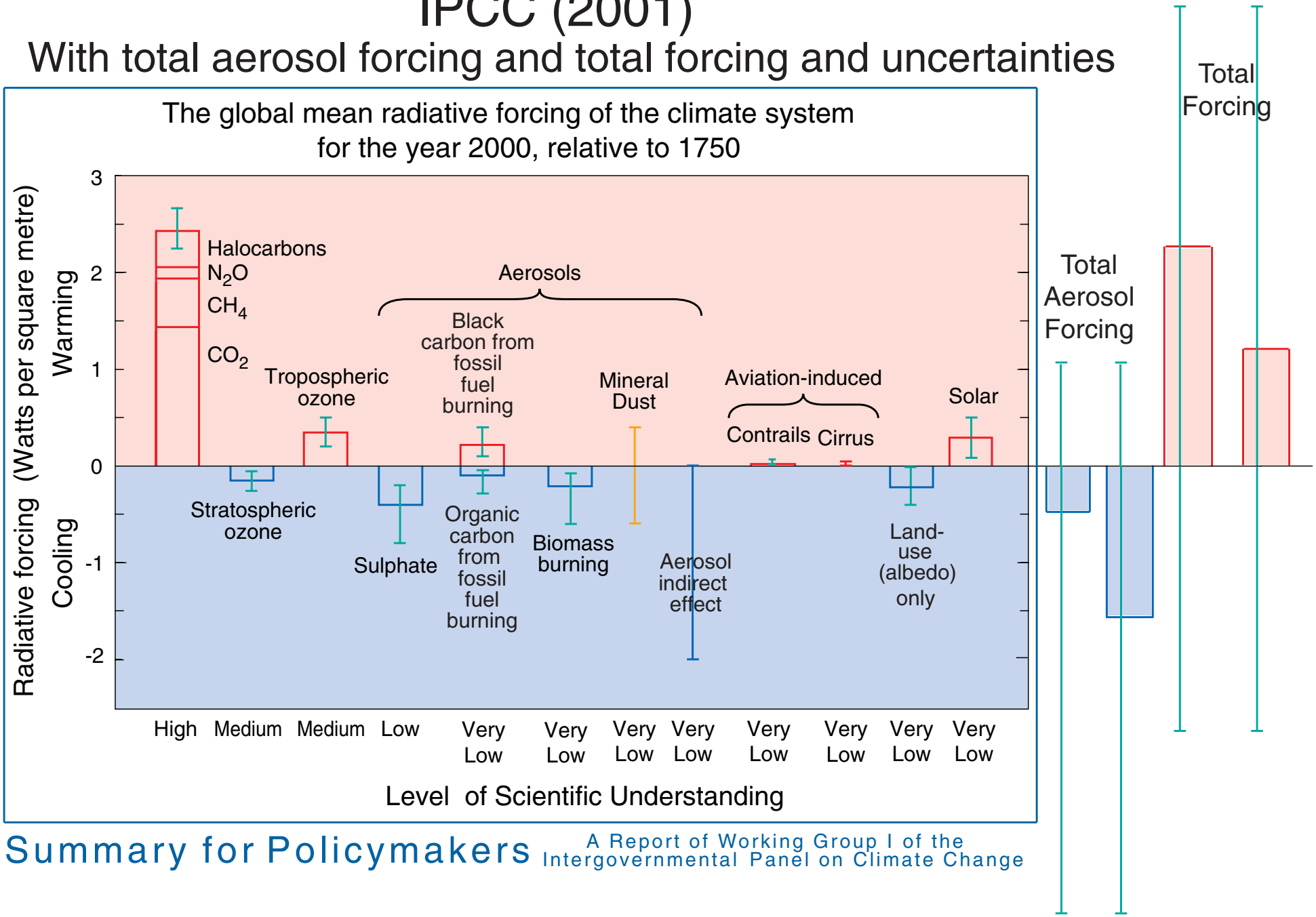
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)
With total aerosol forcing and total forcing and uncertainties



Summary for Policymakers

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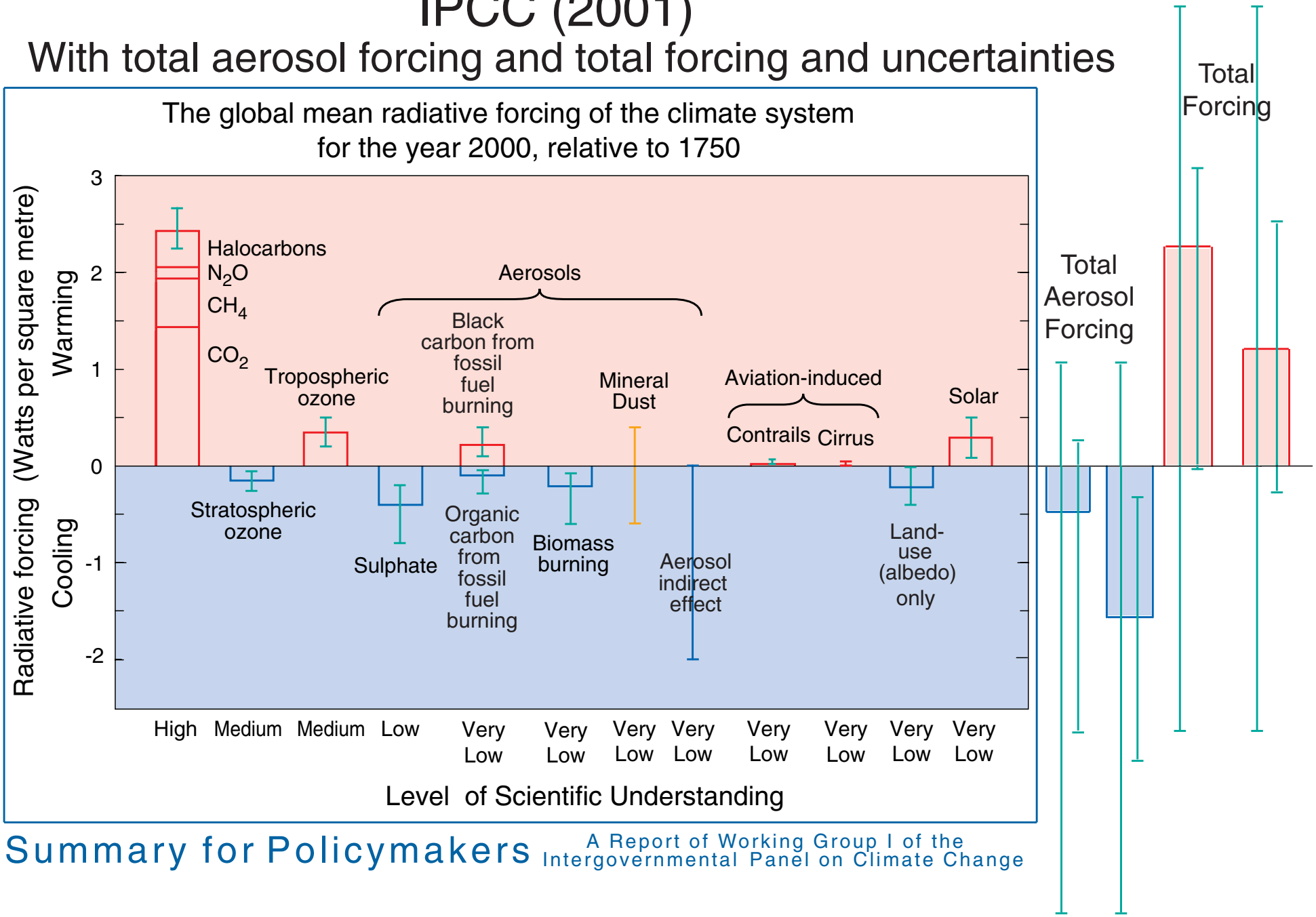
RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD
IPCC (2001)
With total aerosol forcing and total forcing and uncertainties



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Quantities and processes governing aerosol indirect forcing of climate change and the quantities on which their dependence must be known

Quantity/Process	Symbol	Dependence on
Particle and gaseous precursor emissions	$e(\boldsymbol{x}, t, r_{ap}, \chi)$	Radius r_{ap}, composition χ
Transport, chemical reaction, microphysical evolution	$\downarrow$	Concentrations of precursors and other reagents, solar intensity; size dependent concentrations of other aerosol species; 3-D winds, clouds . . .
Aerosol particle number concentration	$n_{ap}(\boldsymbol{x}, t, r_{ap}, \chi)$	Radius, composition
Supersaturation spectrum	$n_{ccn}(s)$	Radius, composition, supersaturation s
Cloud formation and dissipation	$\downarrow$	$n_{ccn}(s)$, updraft velocity, turbulent intensity, precipitation development, heating rate, entrainment . . .
Cloud drop number concentration and properties	$n_{cd}(\boldsymbol{x}, t, r_{cd}, \omega(\lambda))$	Radius, single scatter albedo ω, wavelength λ
Cloud optics	$\downarrow$	Cloud drop size distribution, Mie scattering
Cloud drop scattering and absorption coefficients	$\{\sigma_{sc}, \sigma_{ac}\}(\boldsymbol{x}, t, \lambda)$	Absorption by dissolved and suspended materials
Vertical integral	$\downarrow$	Updrafts, entrainment
Cloud scattering and absorption optical depth	$\{\tau_{sc}, \tau_{ac}\}(\boldsymbol{x}, t, \lambda)$	Cloud physical depth, liquid water path
Radiation transfer (3D)	$\downarrow$	Cloud geometry, surface reflectance
Net spectral flux at top of atmosphere	$F_{toa}(\boldsymbol{x}, t, \lambda)$	

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 – models that are evaluated by comparison with observation

Emissions models

Chemical transport and transformation models

Cloud drop activation and microphysics models

Radiation transfer models

Thank you!

Stephen E. Schwartz

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