VARIATION OF AEROSOL OPTICAL PROPERTIES AND RADIATIVE IMPLICATIONS

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Upton, Long Island, NY

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Aerosol Metrology for Climate Workshop
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www.ecd.bnl.gov/steve

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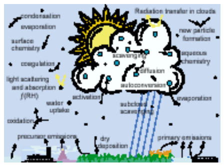
OVERVIEW



Aerosol processes that must be understood

CLASSES OF AEROSOL PROPERTIES AND PROCESSES

It is essential to distinguish . . .

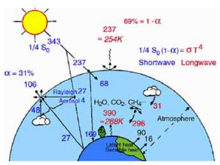
Aerosol chemical and microphysical properties.

Aerosol optical properties.

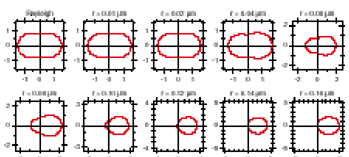
Aerosol radiative influences.

Aerosol radiative forcing.

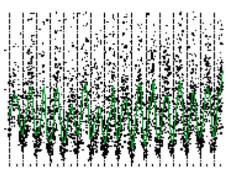
Classes of aerosol properties and processes



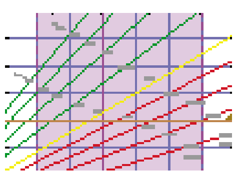
Relation to climate change



Aerosol optical properties



Aerosol radiative influences



Some implications for climate change

DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE AEROSOL

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

$\overline{\Delta F_R}$ 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

$\theta_{SO_4^{2-}}$ 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

AEROSOL DIRECT SHORTWAVE FORCING

Global Average for ***Nonabsorbing*** Aerosol

$$\Delta F = -\frac{1}{2} F_0 T^2 (1 - A_c)(1 - R)^2 \bar{\beta} \tau \qquad \tau = \int \alpha C dz = \int \sigma_{sp} dz$$

Light Scattering Coefficient

Mass Concentration

Mass Scattering Efficiency

Aerosol Optical Depth

Mean Upscatter Fraction

Surface Reflectance

Cloud Fraction

Atmospheric Transmittance

Solar Constant

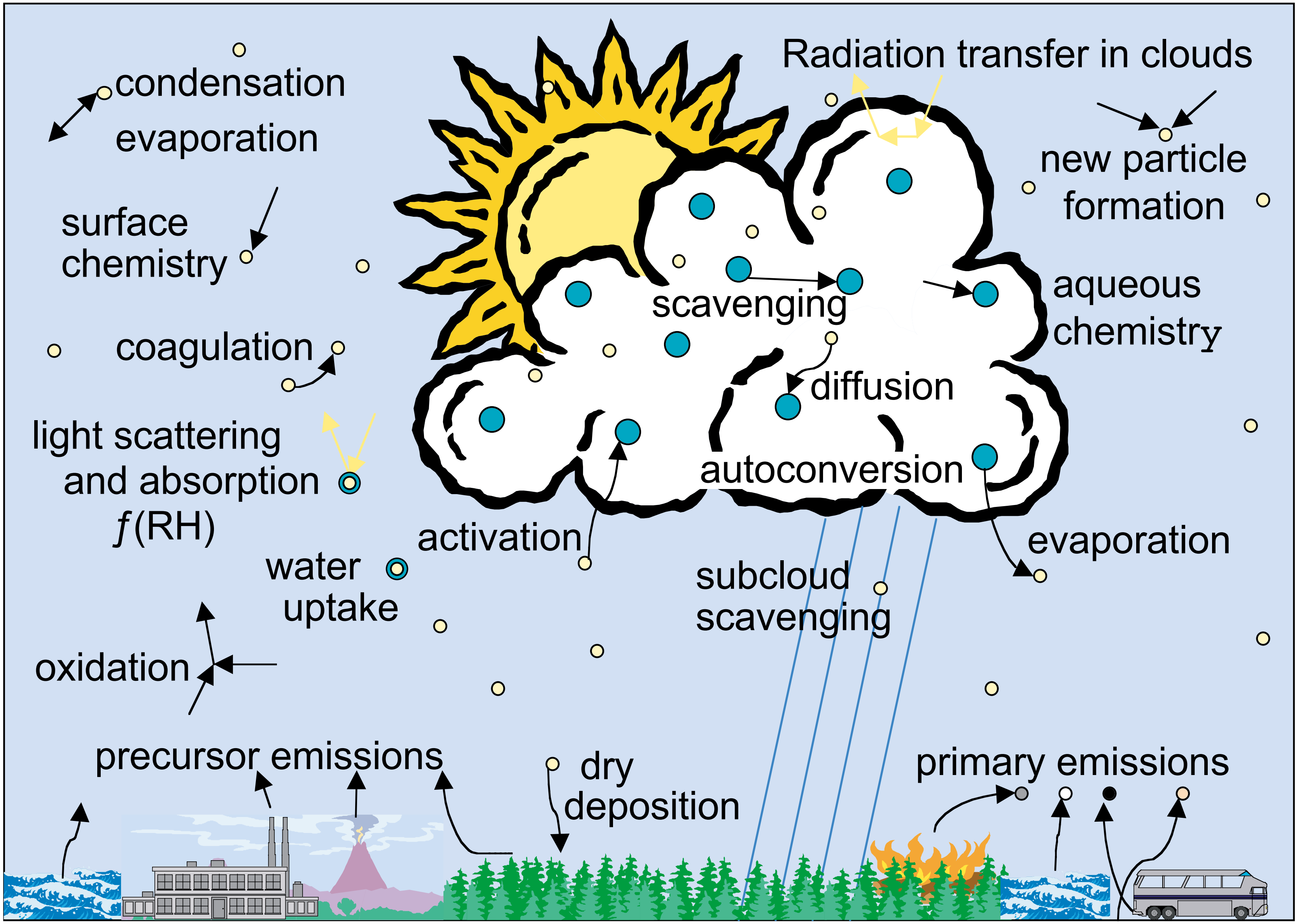
Change in Net TOA Flux

Global Average for ***Absorbing*** Aerosol

$$\Delta F = -\frac{1}{2} F_0 T^2 (1 - A_c)(1 - R)^2 \bar{\beta} \tau \omega \left\{ 1 - \frac{2R}{(1 - R)^2} \frac{(1 - \omega)}{\bar{\beta} \omega} \right\}$$

Single Scattering Albedo

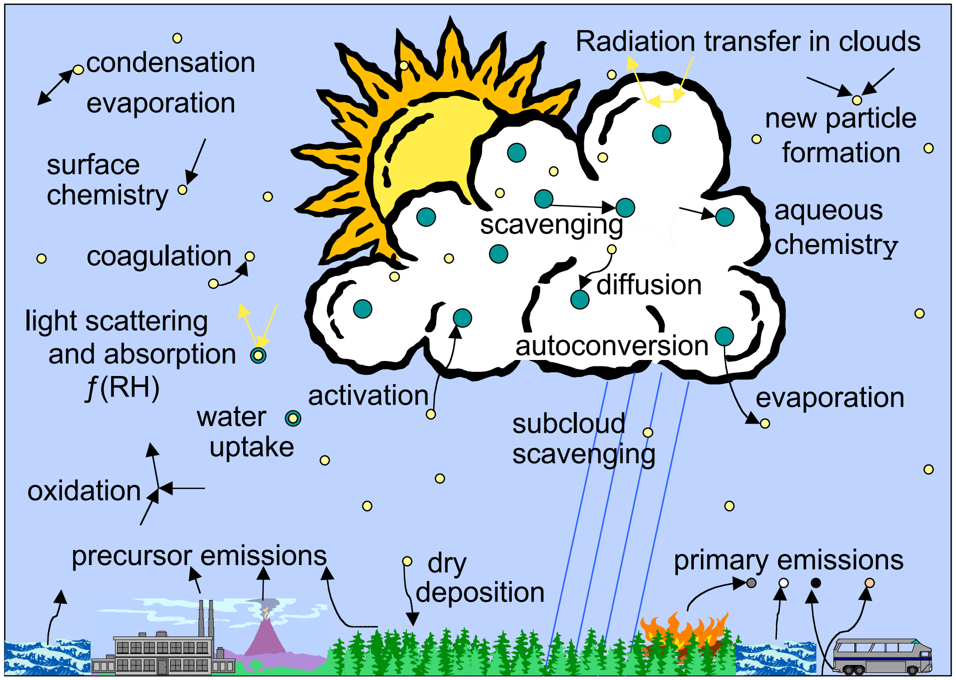
AEROSOL PROCESSES THAT MUST BE UNDERSTOOD AND REPRESENTED IN MODELS



Modified from Ghan and Schwartz, Bull. Amer. Meterol. Soc., 2007

APPROACH TO DETERMINE AEROSOL FORCING

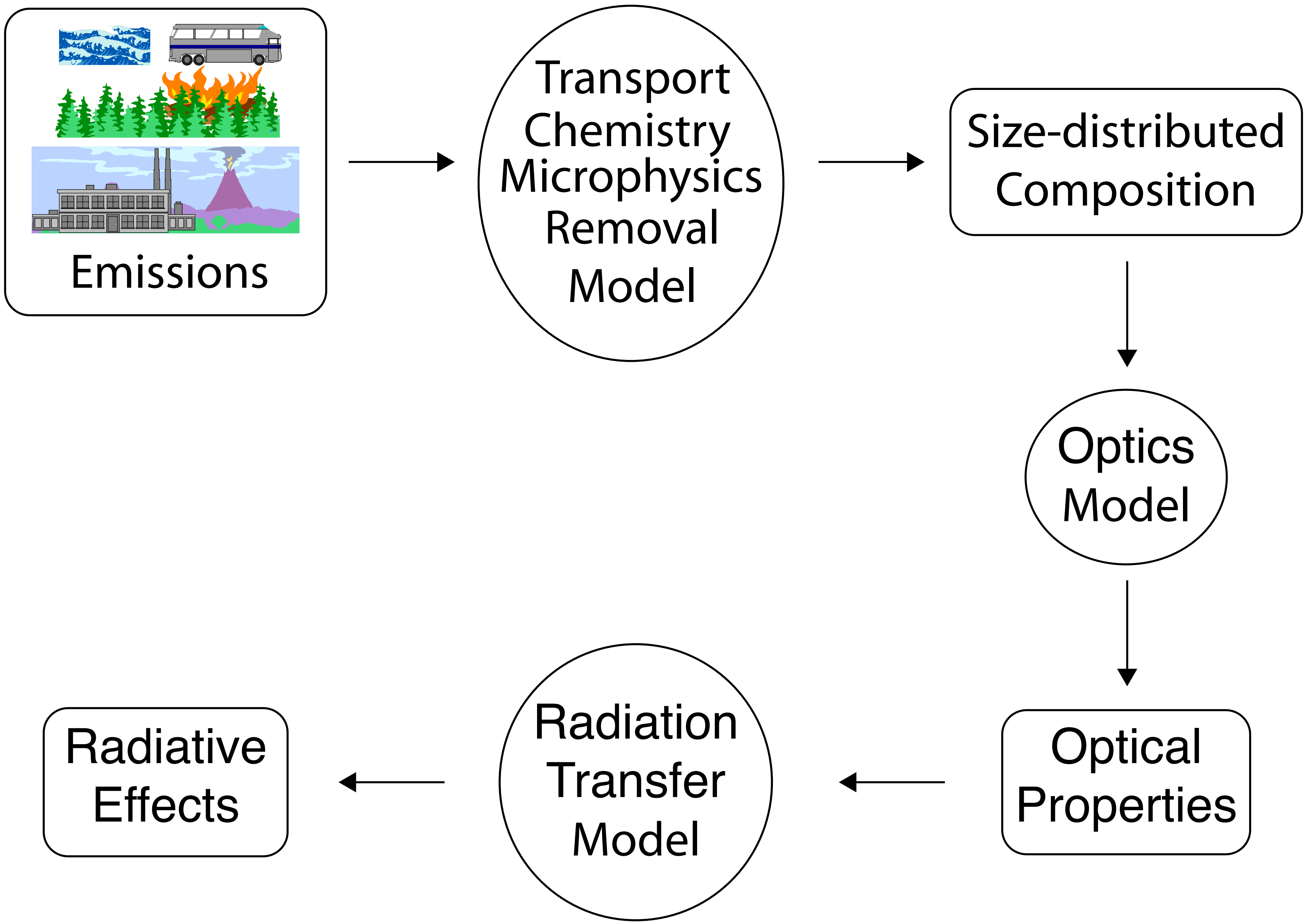
Numerical simulation of physical processes



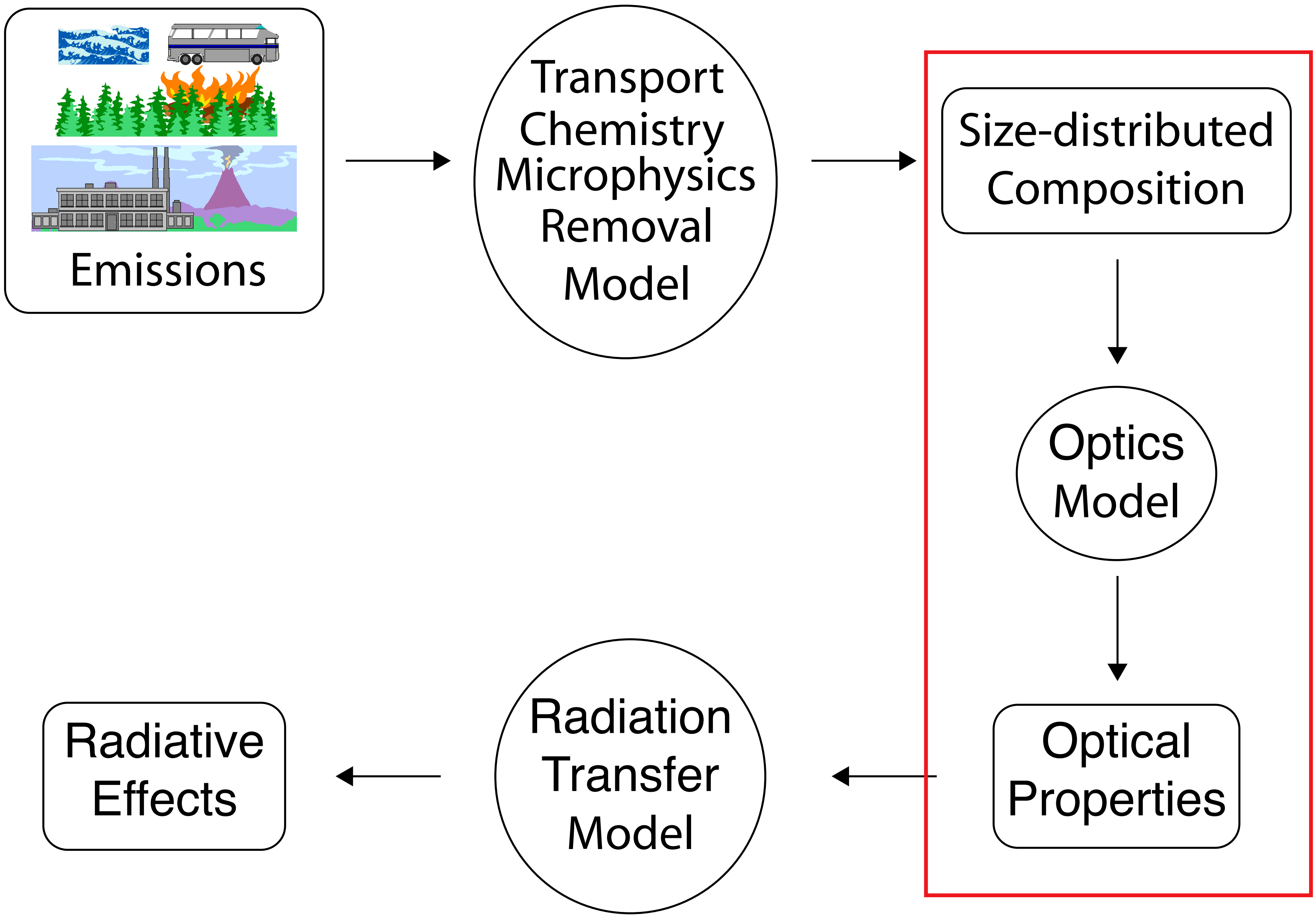
Isomorphism of processes to computer code

Modeling aerosol processes requires understanding these processes, developing and testing their numerical representations, and incorporating these representations in global scale models.

APPROACH TO MODELING AEROSOL RADIATIVE EFFECTS



APPROACH TO MODELING AEROSOL RADIATIVE EFFECTS



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AEROSOL CHEMICAL AND MICROPHYSICAL PROPERTIES

Size distributed chemical composition: dry, or at reference ***relative humidity*** (RH).

Dependence on RH: hygroscopic growth

Liquid *vs* solid; homogeneous spheres? crystalline *vs* amorphous . . .

Internal *vs* external mixture:

- Fully internally mixed: All particles have same composition.
- Size-dependent internal mixture: Particles of a given size have same composition.
- External mixture: Particles of same size may exhibit different composition.

SINGLE-PARTICLE OPTICAL PROPERTIES

Size

Real and imaginary components of index of refraction

Homogeneous, *vs* spherically symmetric, *vs* inhomogeneous

Mie scattering theory allows calculation of angular dependent scattering for homogeneous or for radially dependent, spherically symmetric refractive index (*e.g*. spherical shells).

Nonspherical particles: Approximate methods for spheroids, etc. Numerical methods.

Reference: Mishchenko, Hovenier, Travis, *Light Scattering by Nonspherical Particles*, Academic Press, 2000.

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AEROSOL OPTICAL PROPERTIES

Necessary aerosol optical properties for calculating radiative influences are:

{ Scattering coeff, Absorption coeff } <=> { Extinction coeff, Single scat albedo }

Asymmetry parameter: Average of cosine of scattering angle.

Dependence on ***wavelength***.

Dependence on ***relative humidity***.

Also relevant:

Phase function – Intensity of scattering as function of scattering angle.

Polarization – Spherical vs crystal, pertinent to source.

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RADIATIVE INFLUENCES

A change in radiative flux due to of the presence of aerosol.

Requires specification of the radiative situation: solar zenith angle, surface albedo, height of aerosol in atmosphere, cloud-free vs all-sky, . . .

Examples:

Aerosol optical depth: Δ optical depth due to aerosol.

Δ Direct normal solar irradiance.

Δ Diffuse downwelling shortwave irradiance at surface.

Δ ***Absorbed*** downwelling shortwave irradiance at surface.

Δ Upwelling shortwave irradiance at top of atmosphere (TOA).

Might be instantaneous and local; might be temporal and/or spatial average.

RADIATIVE *FORCING*

A specific ***difference***; a change in radiative flux between two specified conditions.

Examples:

Forcing by the ***total*** aerosol: Flux with aerosol minus flux without aerosol.

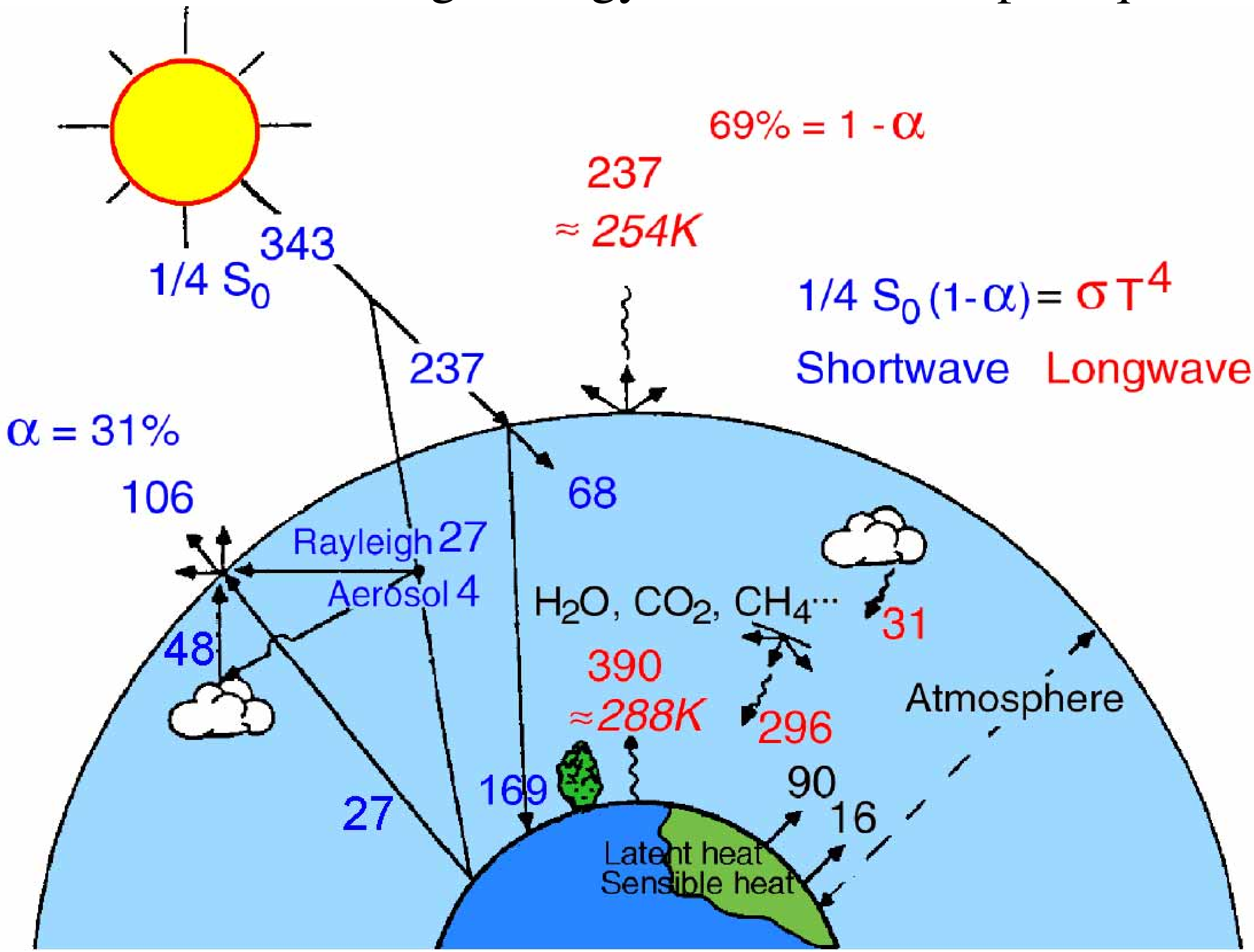
Anthropogenic aerosol forcing: Flux with anthropogenic aerosol minus flux without.

Might be instantaneous, local; might be temporal and/or spatial
Might be instantaneous, local; might be temporal and/or spatial average; cloud-free or all-sky; might be surface or TOA.

RELATION TO CLIMATE CHANGE OVER THE INDUSTRIAL ERA

GLOBAL ENERGY BALANCE

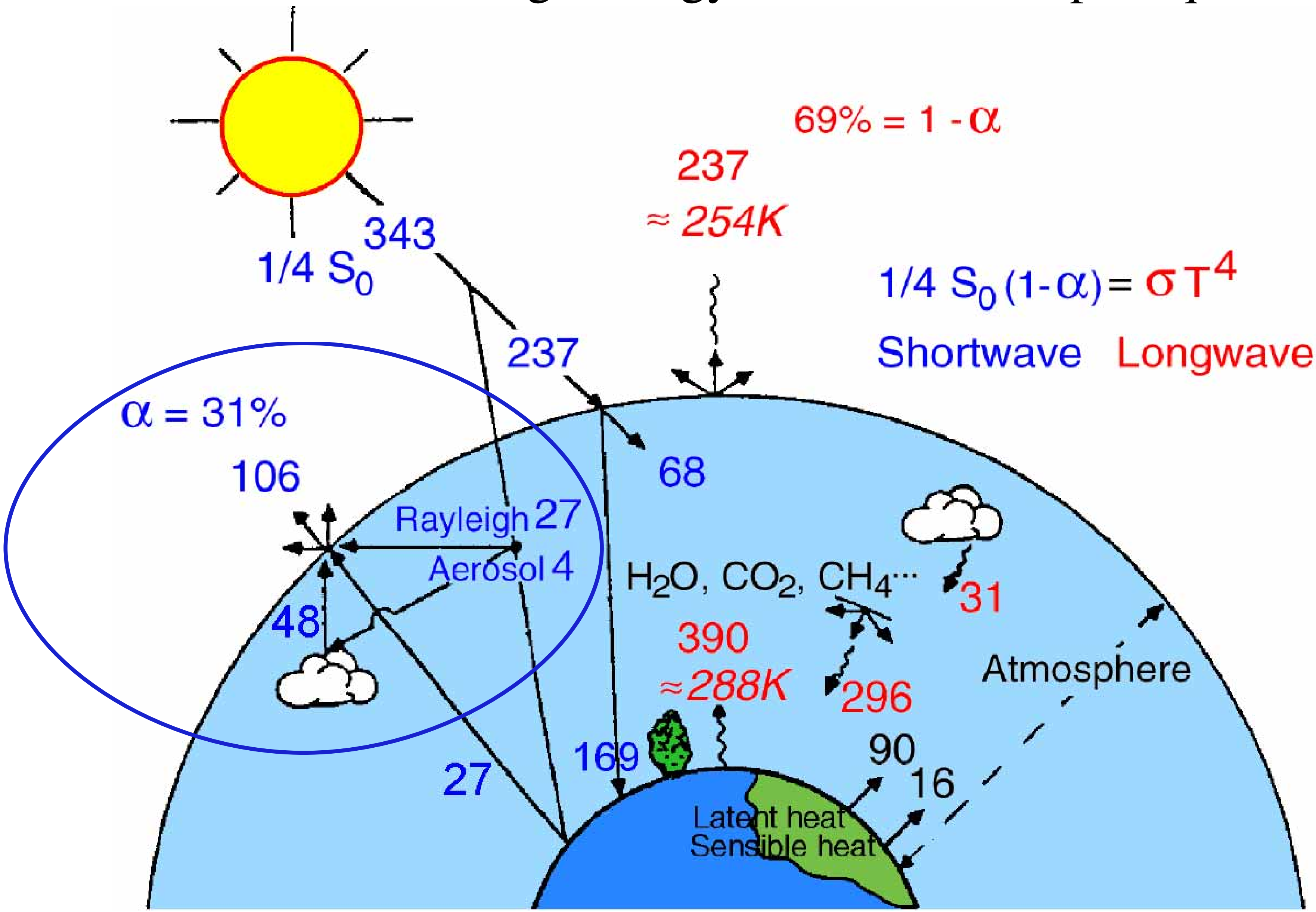
Global and annual average energy fluxes in watts per square meter



Schwartz, 1996, modified from Ramanathan, 1987

GLOBAL ENERGY BALANCE

Global and annual average energy fluxes in watts per square meter



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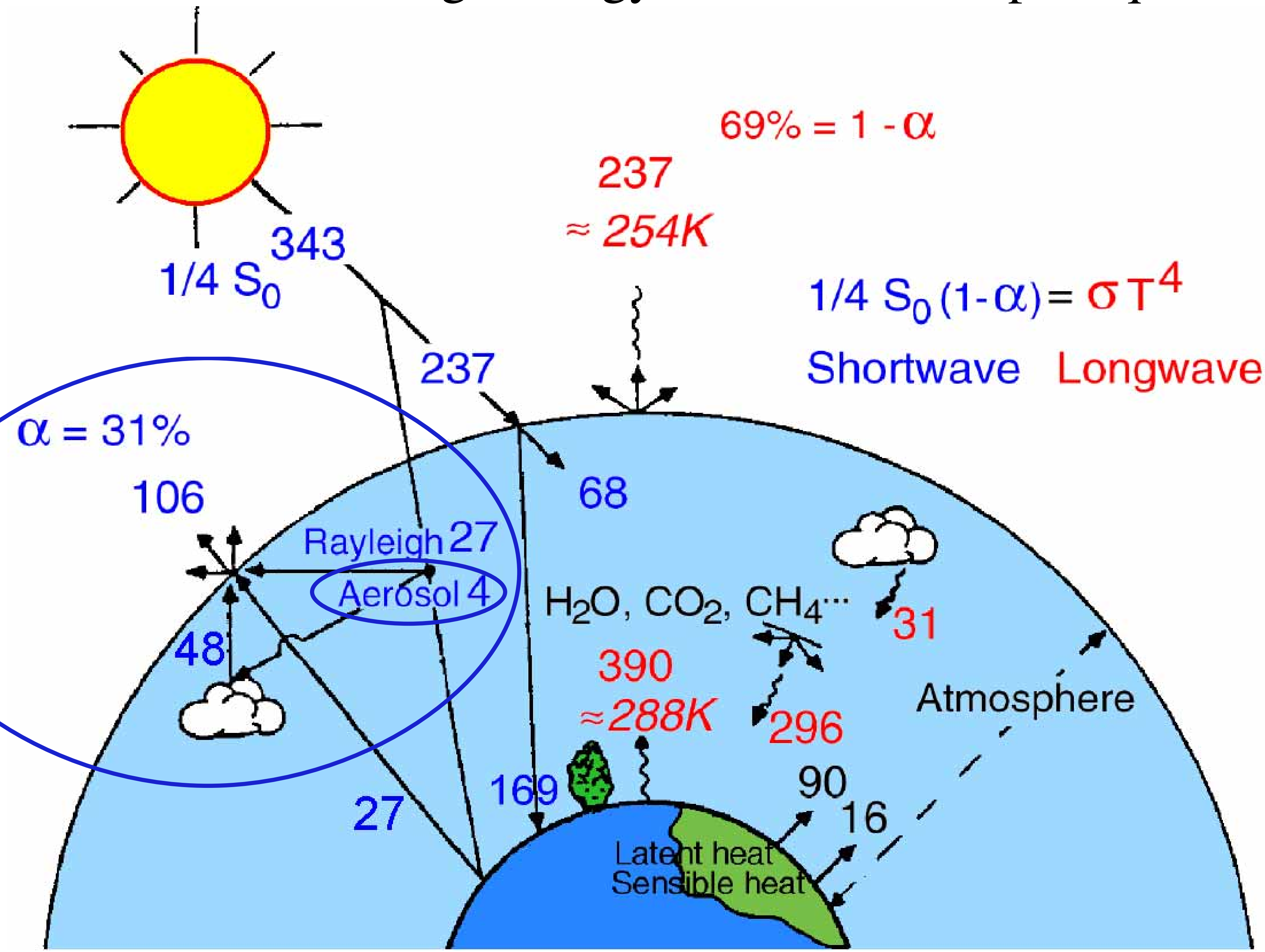

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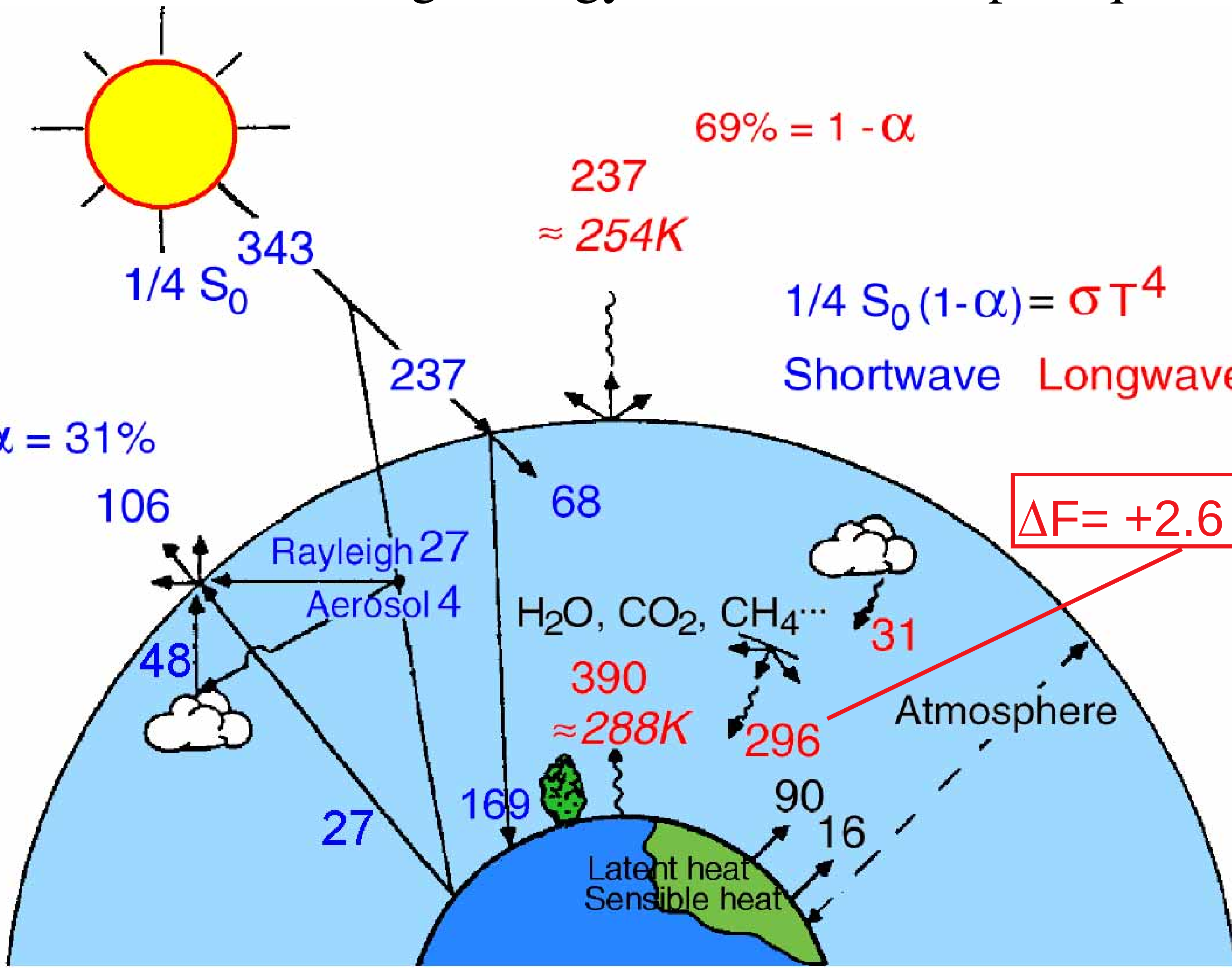

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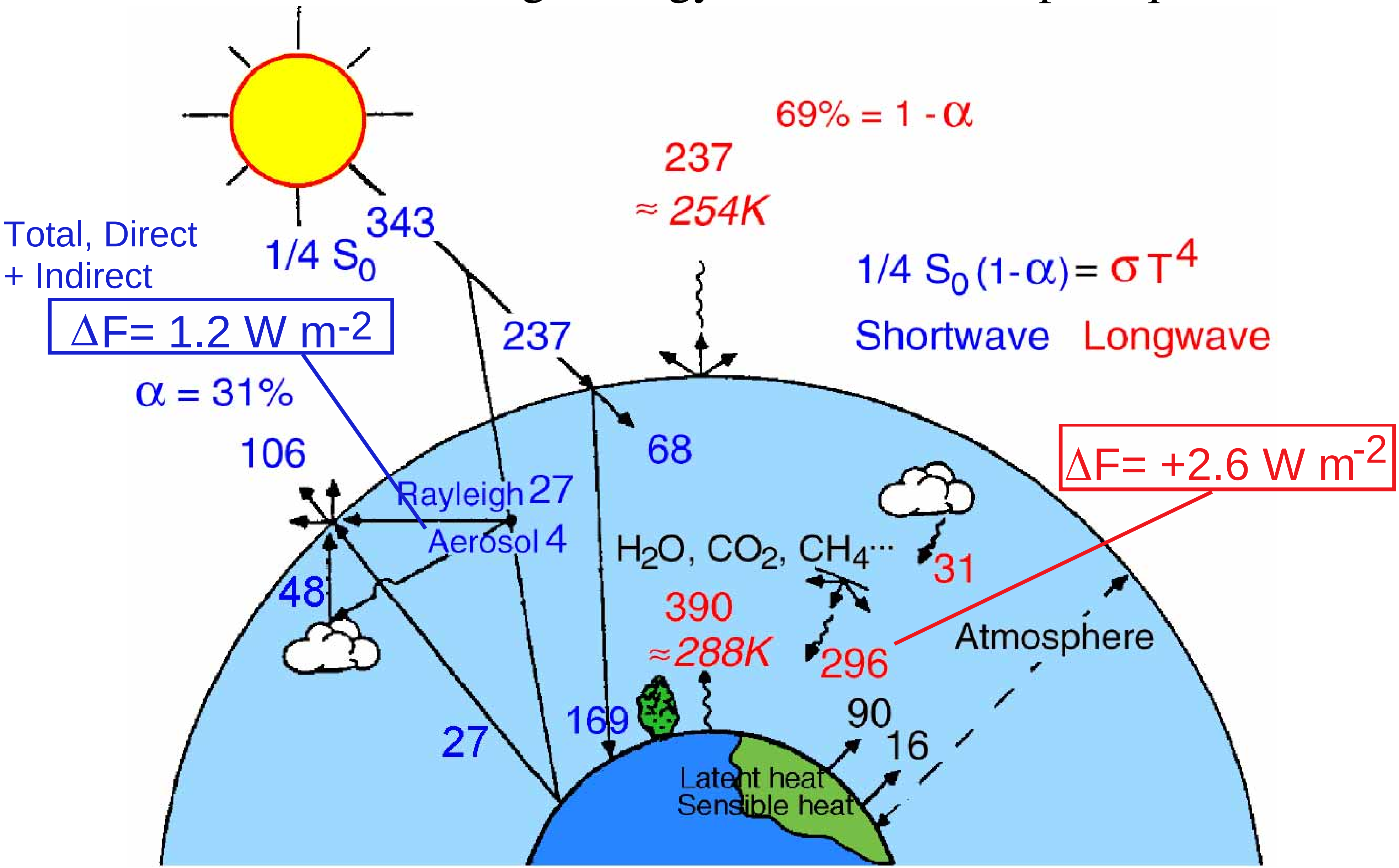
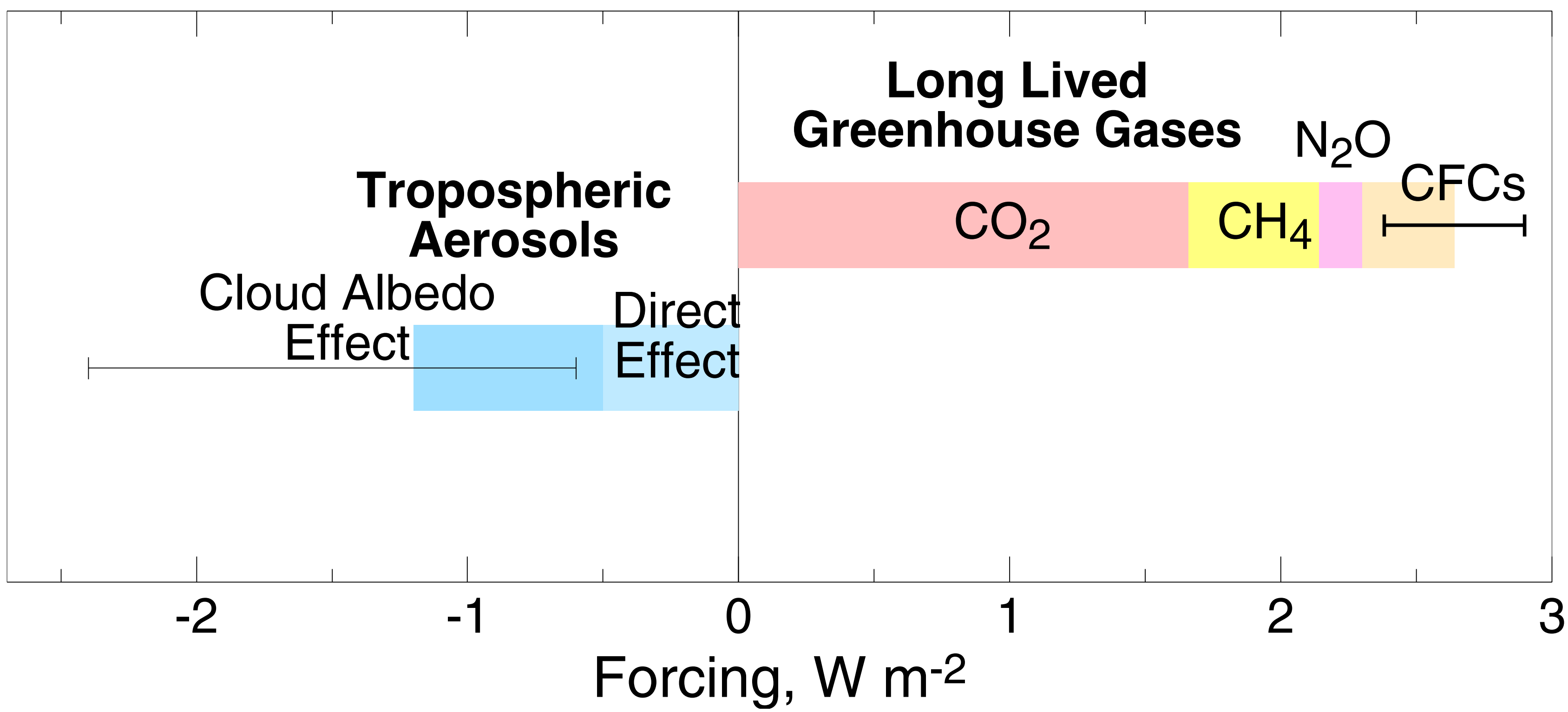


Schwartz, 1996, modified from Ramanathan, 1987

CLIMATE FORCINGS OVER THE INDUSTRIAL PERIOD

Extracted from IPCC AR4 (2007)

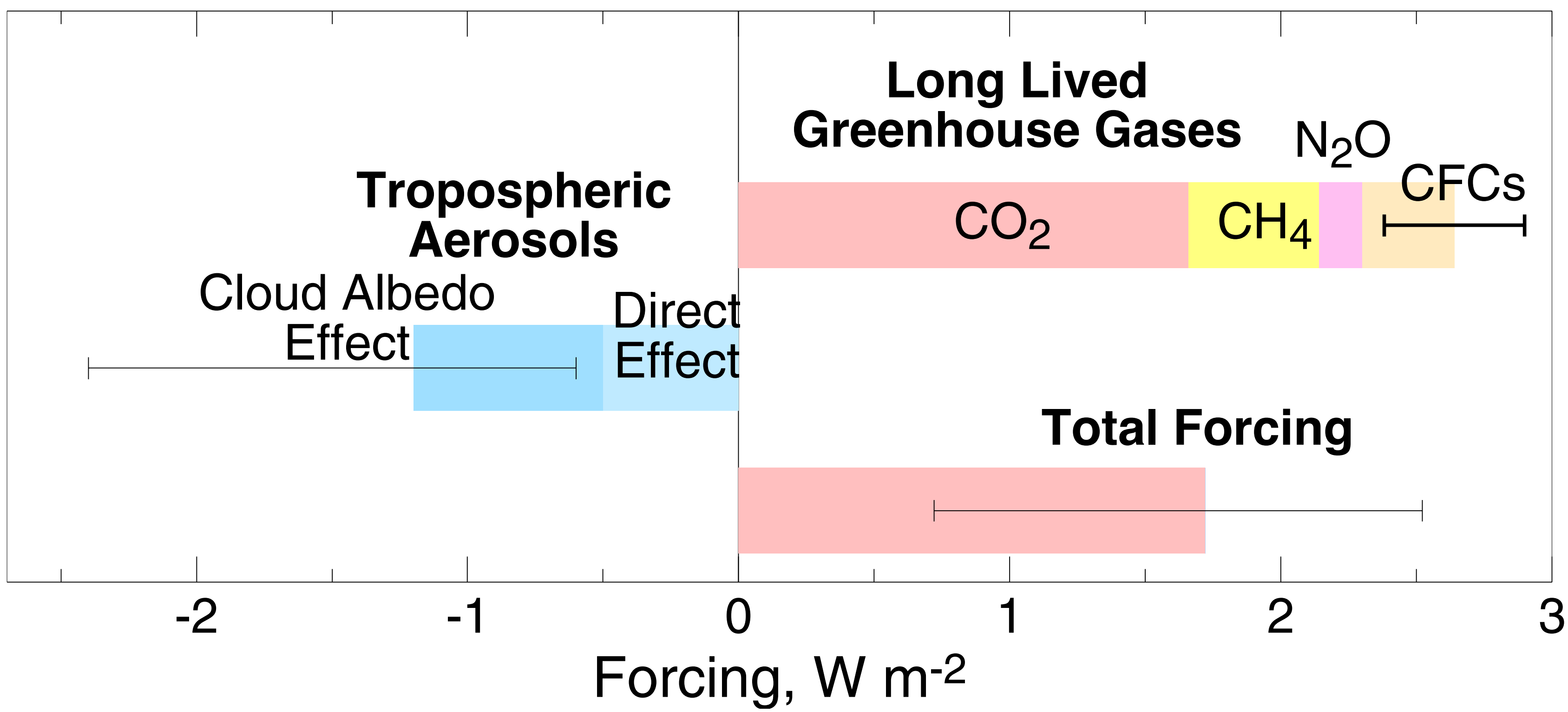


Negative aerosol forcing substantially offsets GHG forcing.

Aerosol forcing is highly uncertain.

CLIMATE FORCINGS OVER THE INDUSTRIAL PERIOD

Extracted from IPCC AR4 (2007)



Total forcing includes other anthropogenic and natural (solar) forcings.

Forcing by tropospheric ozone, ~0.35 W m^{-2}, is the greatest of these.

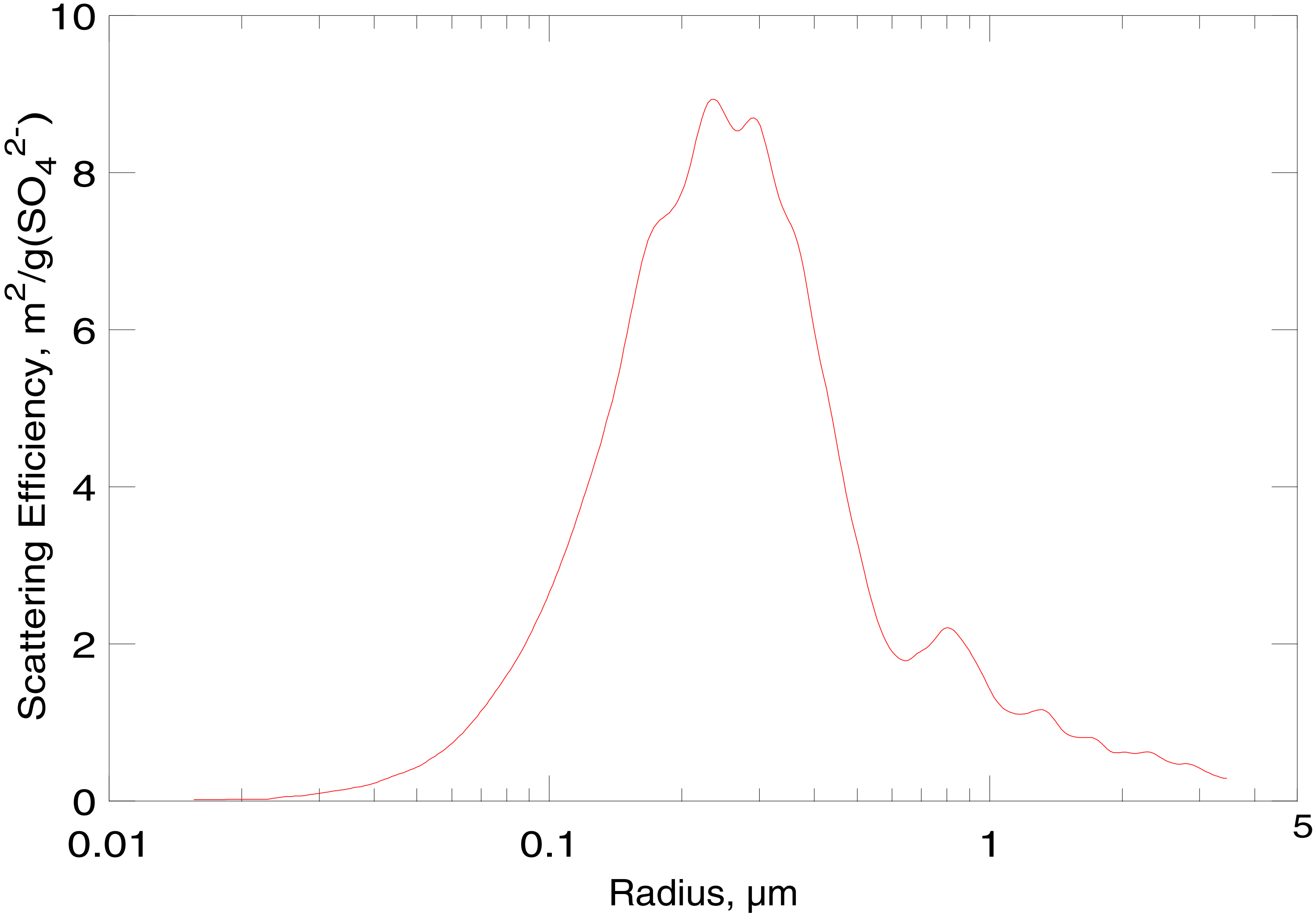
Uncertainty in aerosol forcing dominates uncertainty in total forcing.

AEROSOL OPTICAL PROPERTIES

LIGHT SCATTERING EFFICIENCY

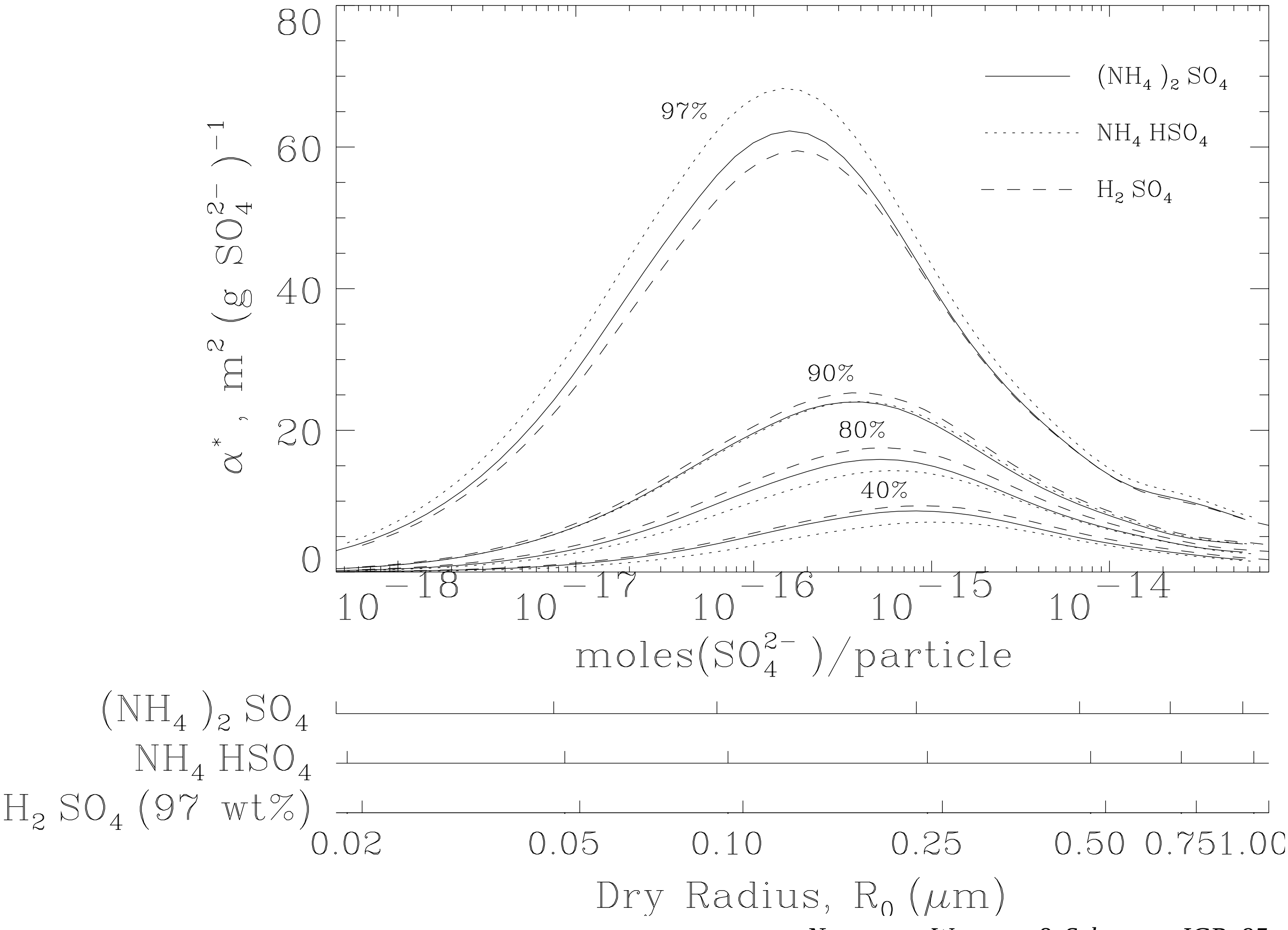
Dependence on particle radius

Ammonium Sulfate, 550 nm



Data of Ouimette and Flagan, 1982

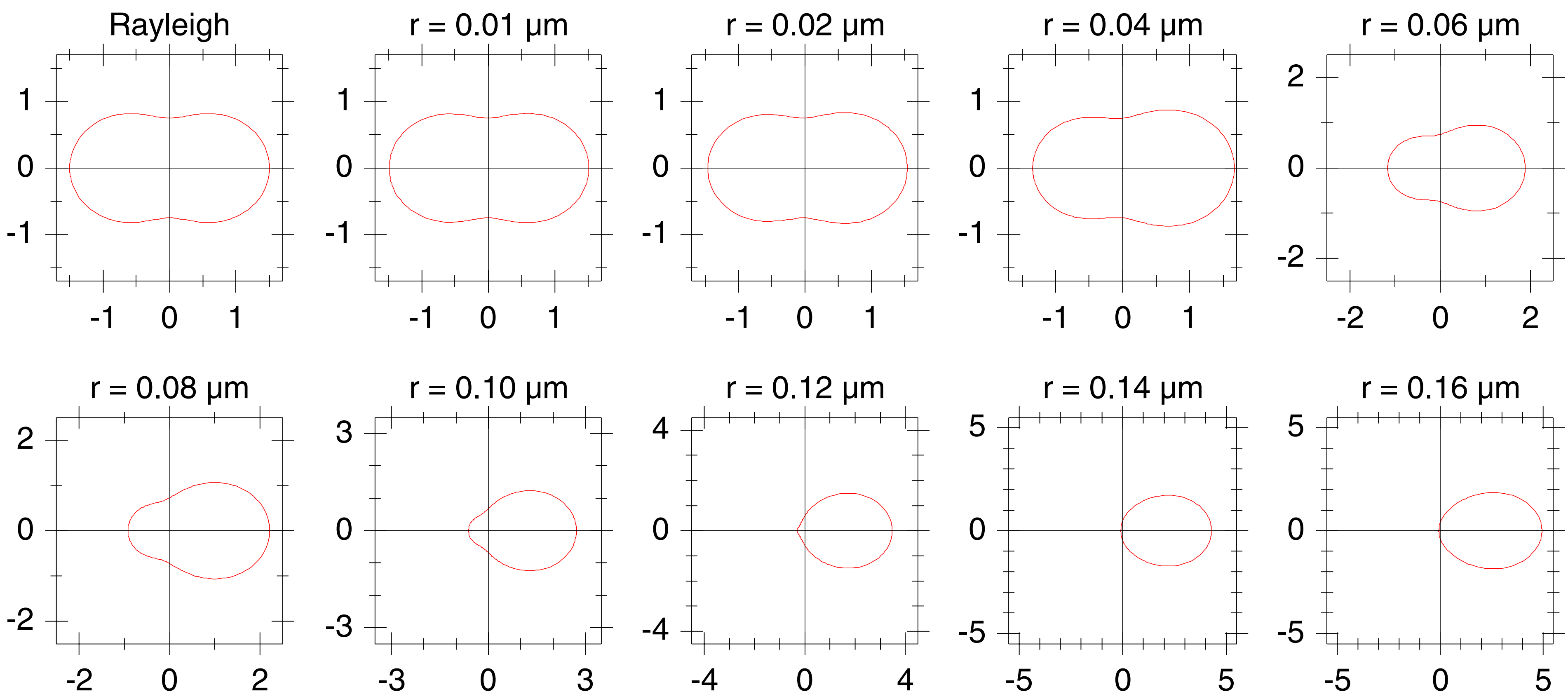
LIGHT SCATTERING CROSS SECTION: DEPENDENCE ON PARTICLE SIZE, COMPOSITION AND RH



Nemesure, Wagener & Schwartz, JGR, 95

LIGHT SCATTERING BY AEROSOL PARTICLES

Dependence of angular distribution of scattering (phase function) on particle size



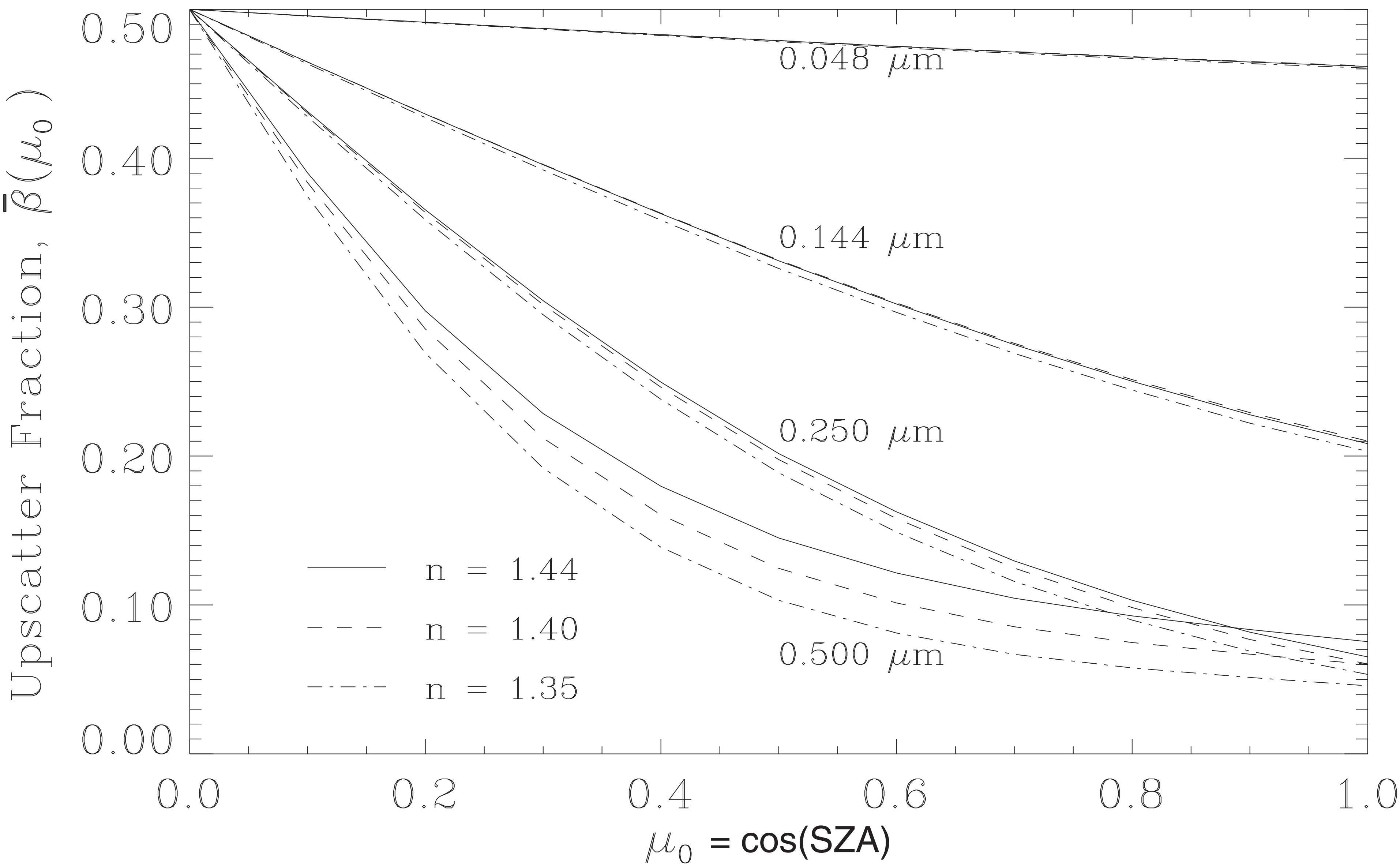
Calculations by R. Wagener, BNL

Larger particles scatter increasingly in the forward direction.

AEROSOL RADIATIVE INFLUENCES

UPSCATTER FRACTION

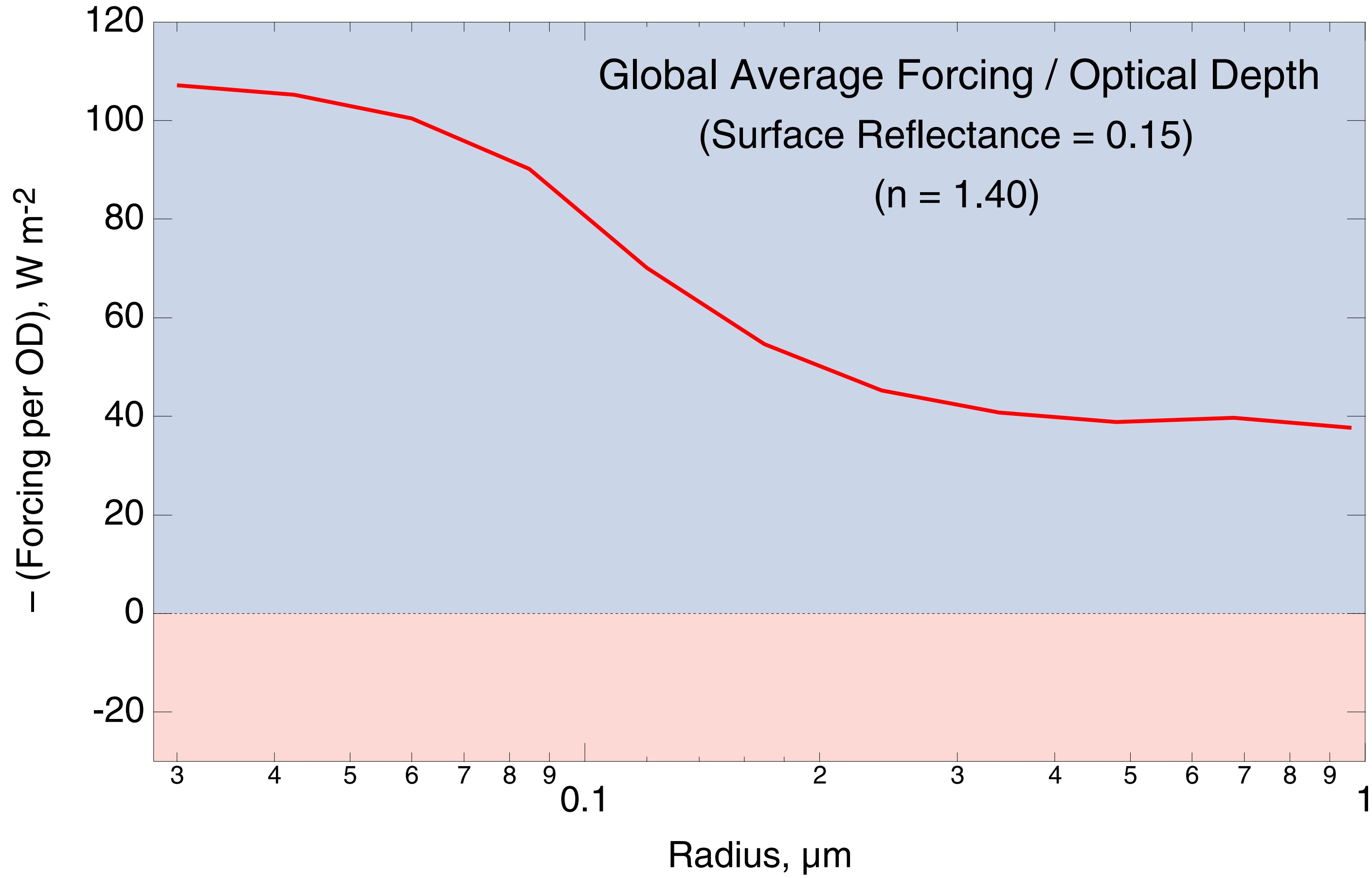
Dependence on solar zenith angle, particle radius, and refractive index



Nemesure, Wagener & Schwartz, JGR, 95

FORCING PER OPTICAL DEPTH

Global average, cloud-free sky - Scattering aerosol

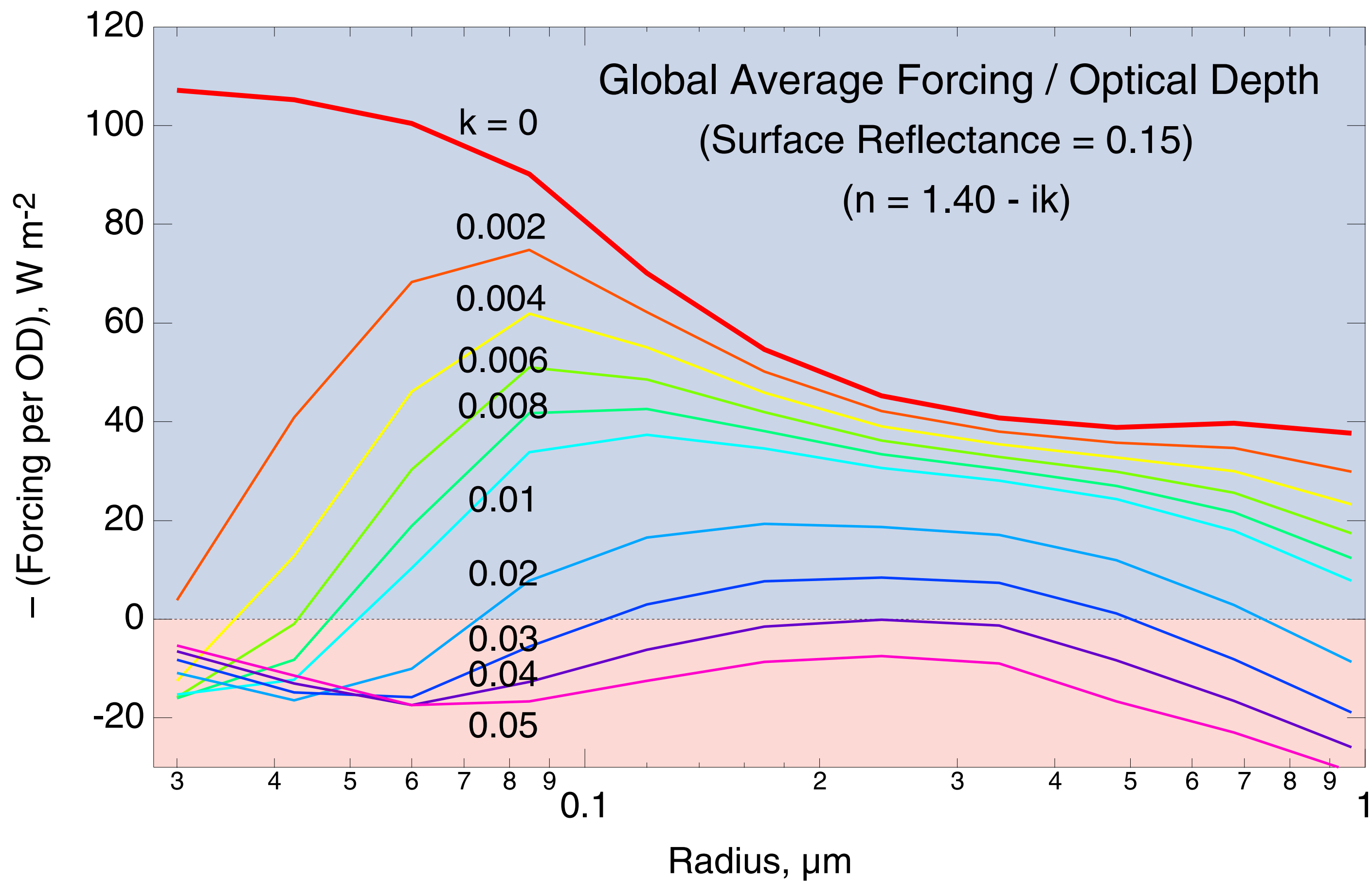


Forcing per optical depth depends rather strongly on particle size.

Forcing accuracy 0.5 W m^{-2} requires optical depth accuracy 0.005 - 0.01 (0.01 - 0.02 for 60% cloud cover).

FORCING PER OPTICAL DEPTH

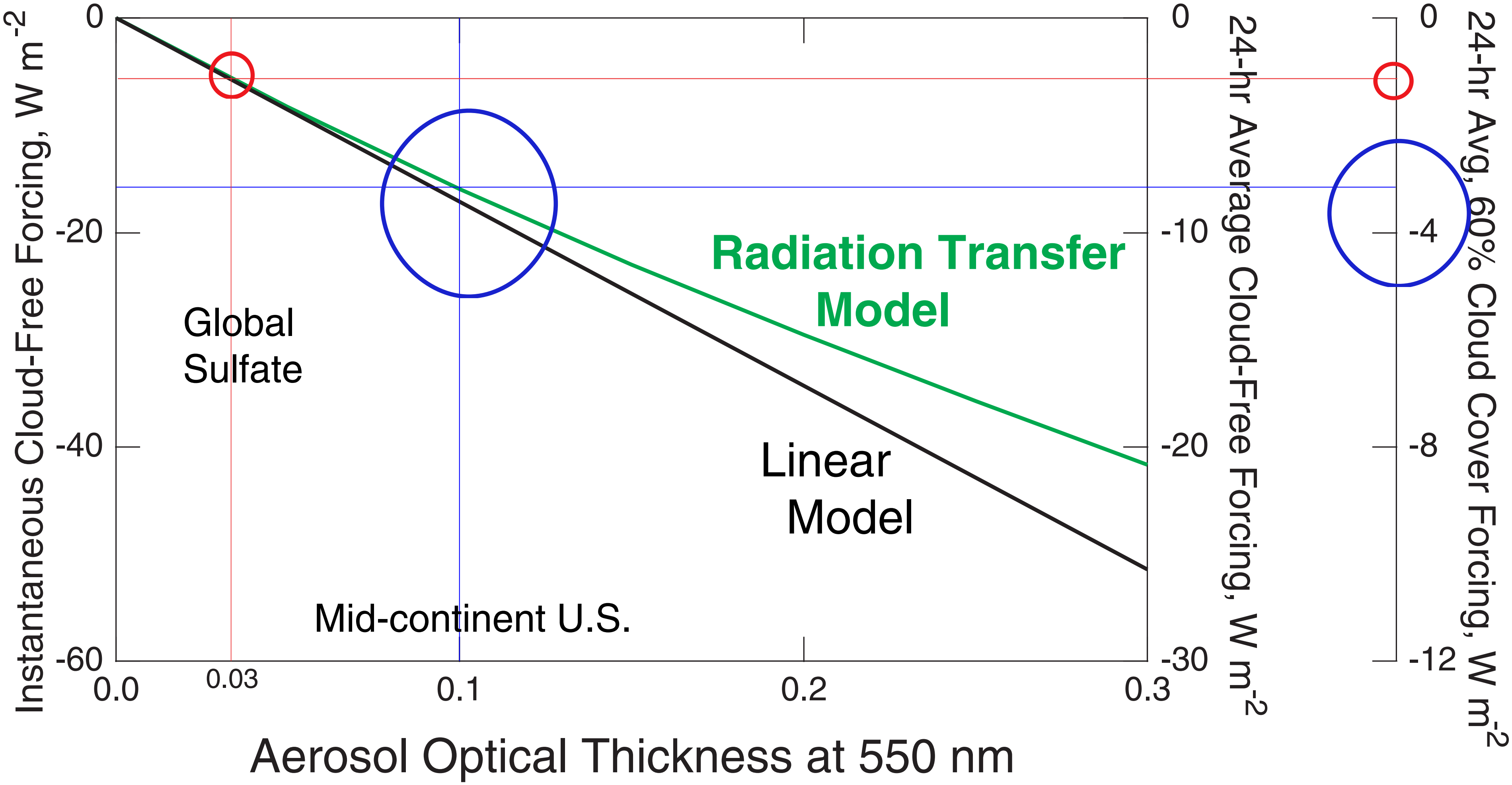
Global average, cloud-free sky - Absorbing aerosol



Forcing per optical depth depends rather strongly on particle size – and also rather strongly on aerosol absorption.

ESTIMATES OF AEROSOL DIRECT FORCING

By linear model and by radiation transfer modeling

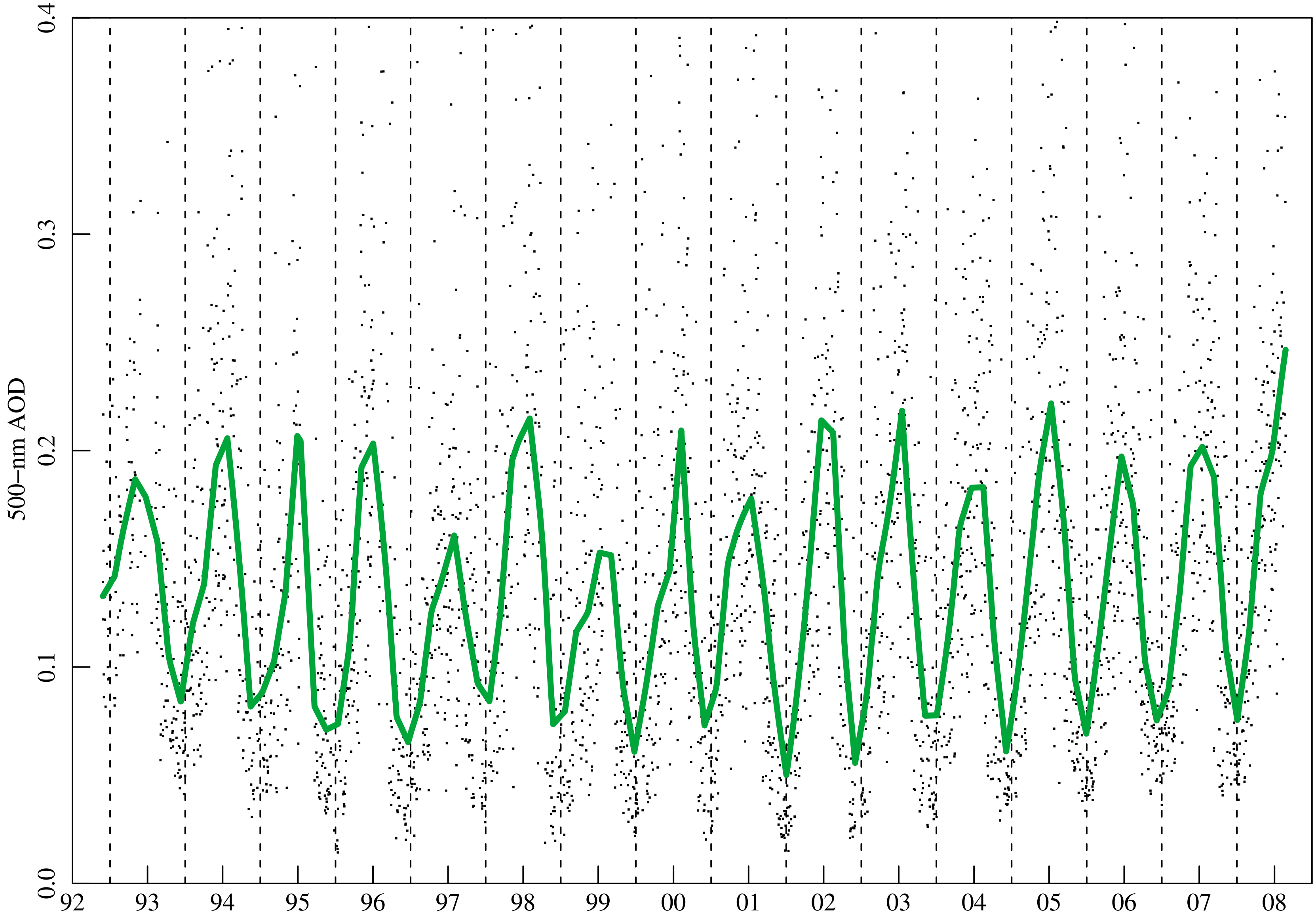


Global average sulfate optical thickness is 0.03: ***1 W m^{-2} cooling***.

In ***continental U. S.*** typical aerosol optical thickness is 0.1: ***3 W m^{-2} cooling***.

AEROSOL OPTICAL DEPTH AT ARM SGP

Fifteen years of daily average 500 nm AOD in North Central Oklahoma

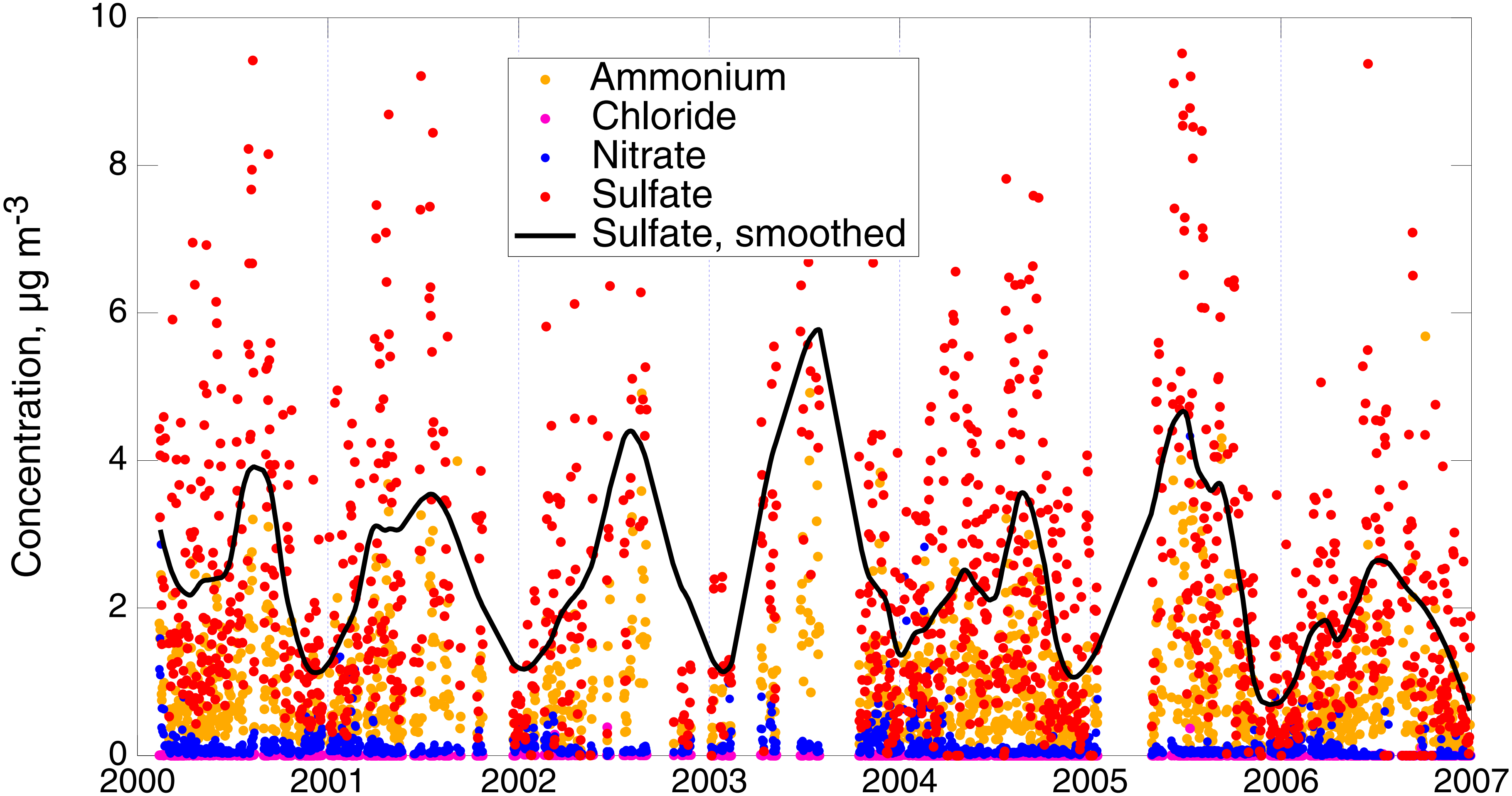


Michalsky, Denn, Flynn, Hodges, Kiedron, Koontz, Schlemmer, Schwartz, JGR, 2010

Green curve is LOWESS (locally weighted scatterplot smoothing) fit.

AEROSOL COMPOSITION AT ARM SGP

Seven years of ***daily average composition*** in North Central Oklahoma

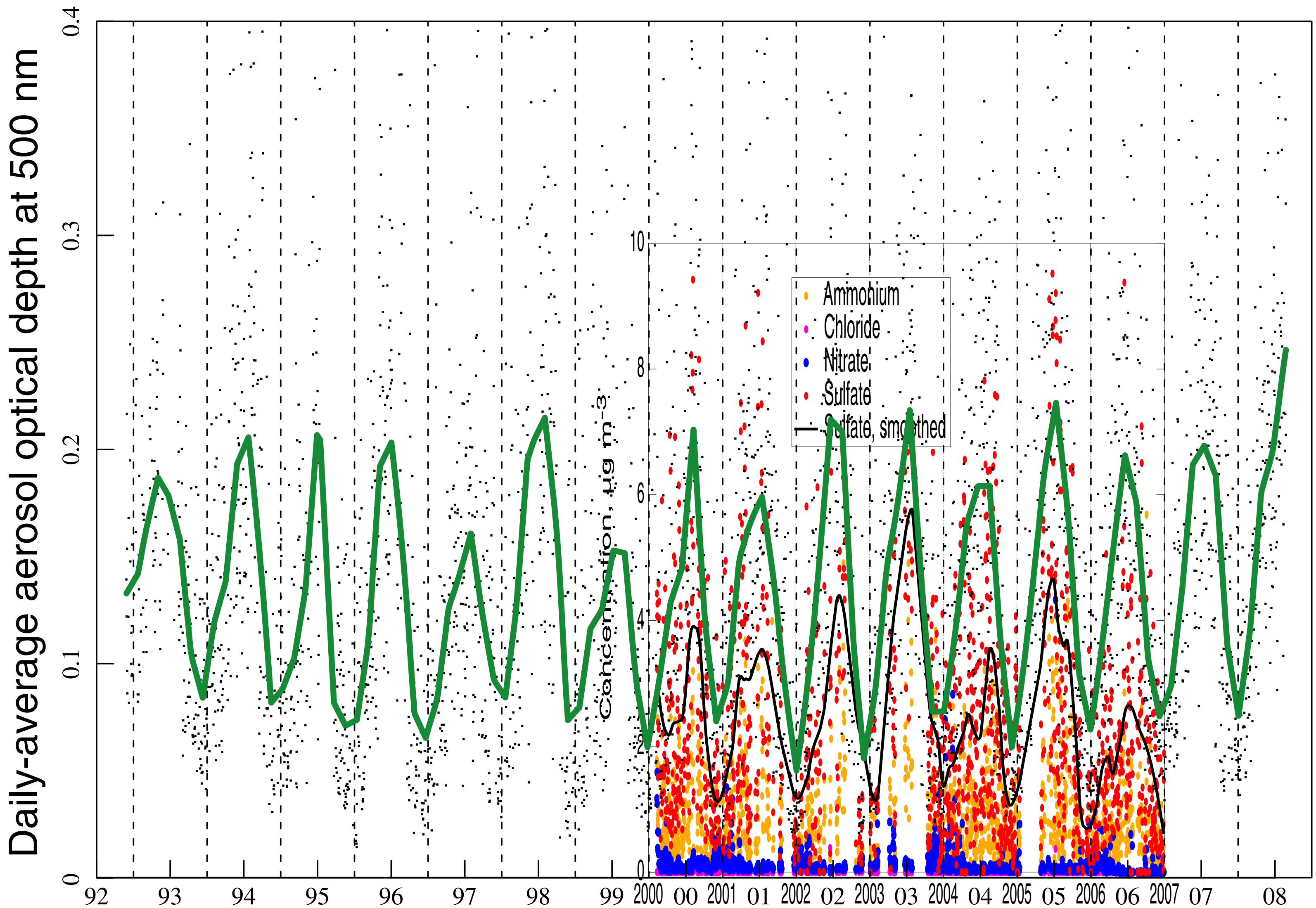


Measurements of P. Quinn, NOAA, PMEL

Black curve is LOWESS (locally weighted scatterplot smoothing) fit. Note summertime peak of sulfate.

AEROSOL OPTICAL DEPTH AND COMPOSITION AT ARM SGP

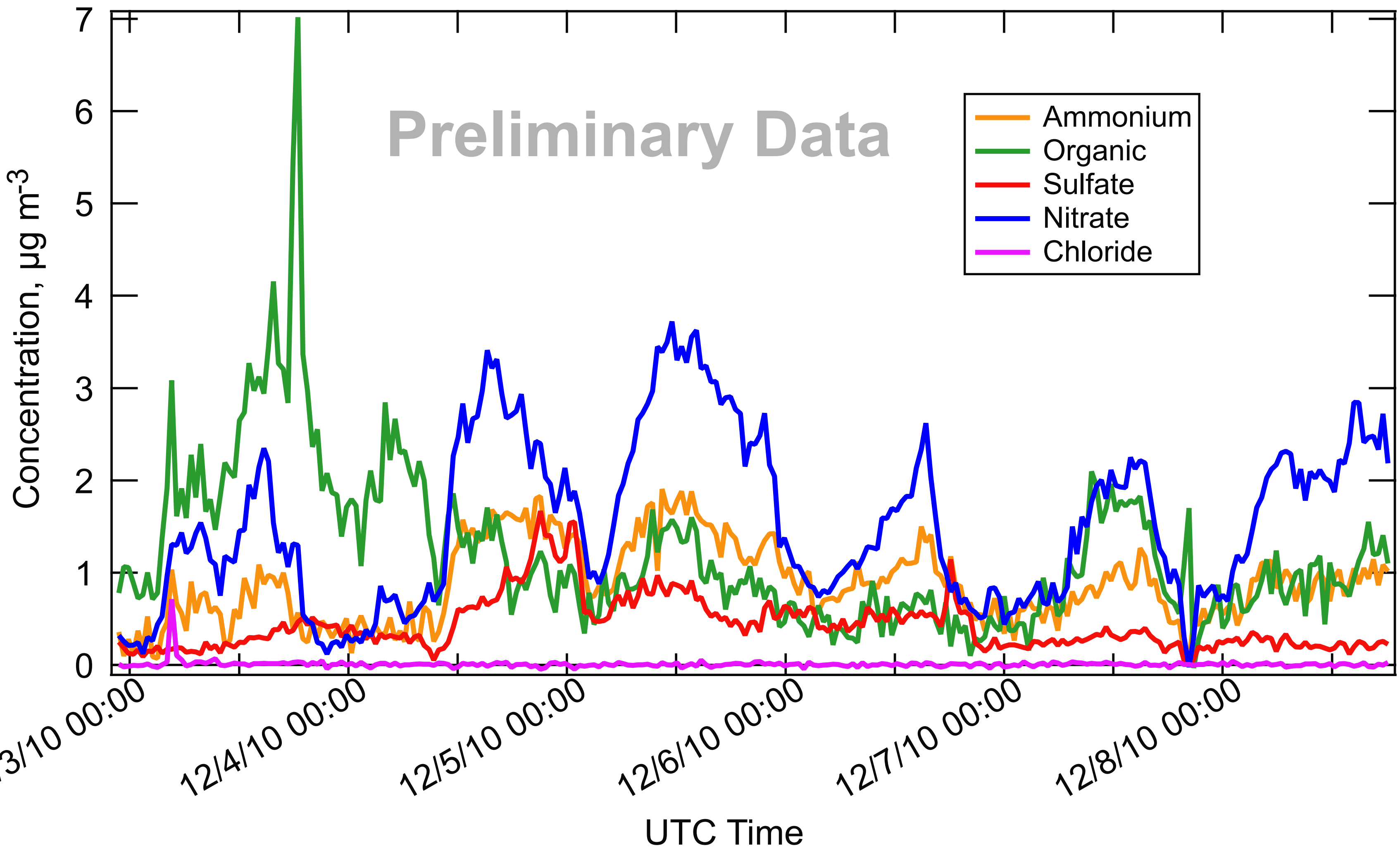
Daily average optical depth and composition in North Central Oklahoma



Summertime sulfate peak coincides with maximum optical depth.

AEROSOL COMPOSITION AT ARM SGP

Six days of ***30-minute average composition*** in North Central Oklahoma

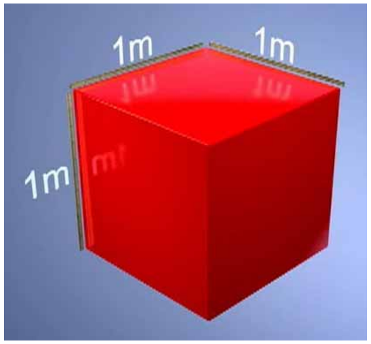


- First data from new aerosol mass spectrometer installed at SGP.
- Continuous data; for particles with diameter ≤ 0.5 μm.
- Note high nitrate compared to sulfate; substantial organic component.

CLOSURE EXPERIMENTS

"Cubic meter experiments"

Compare measured optical properties with those modeled based on measured aerosol composition and size distribution.



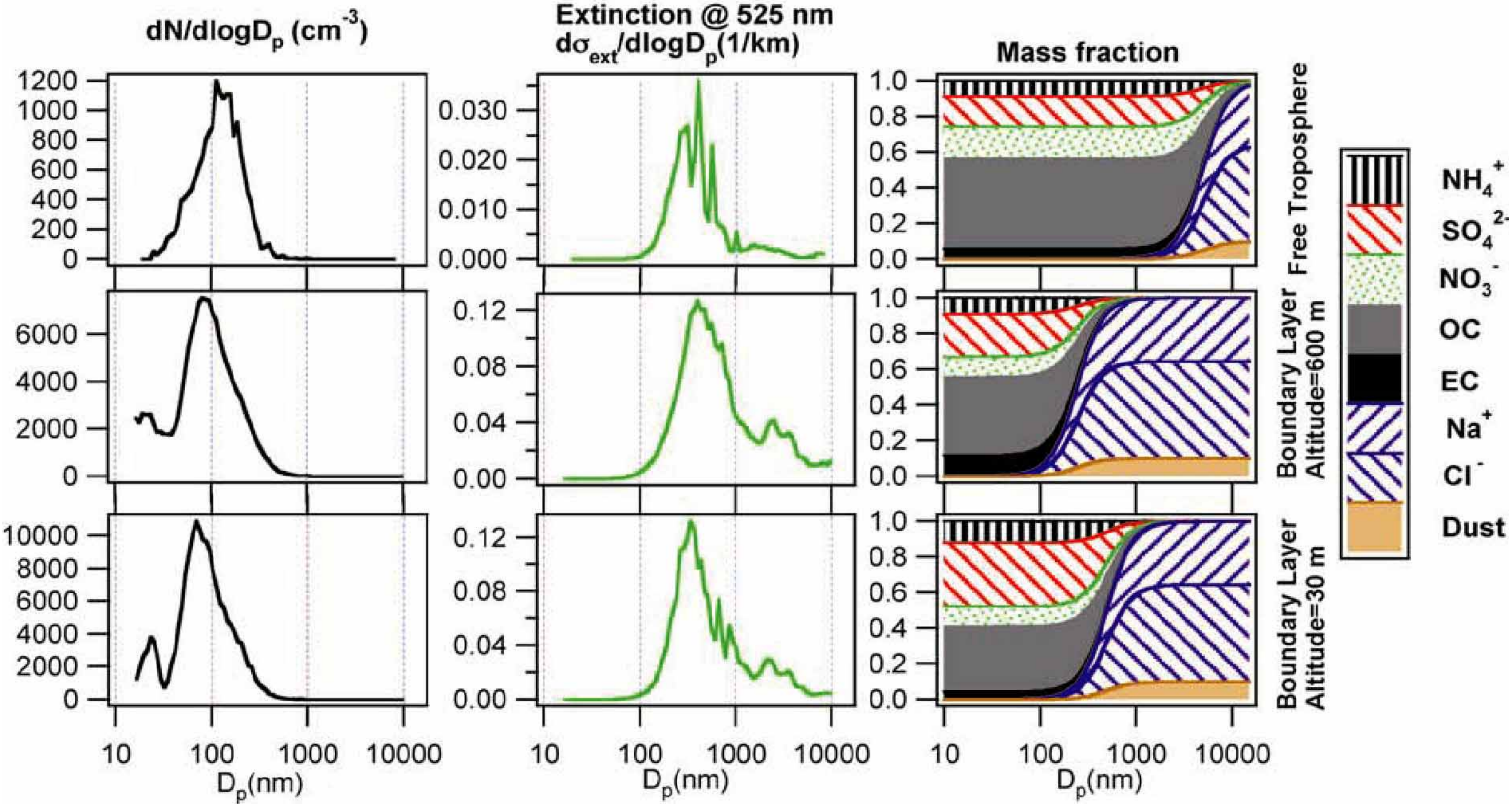
Can be done locally. Require time series of composition, size distribution, and optical properties.

"Column closure experiments"

Compare optical depth, direct beam irradiance, diffuse irradiance; total and/or spectral, at surface or $f(z)$. Require characterization of aerosol optical properties vertically.

LOCAL OPTICAL CLOSURE EXPERIMENT

Aircraft measurements, south of Japan, ACE-Asia, 2001

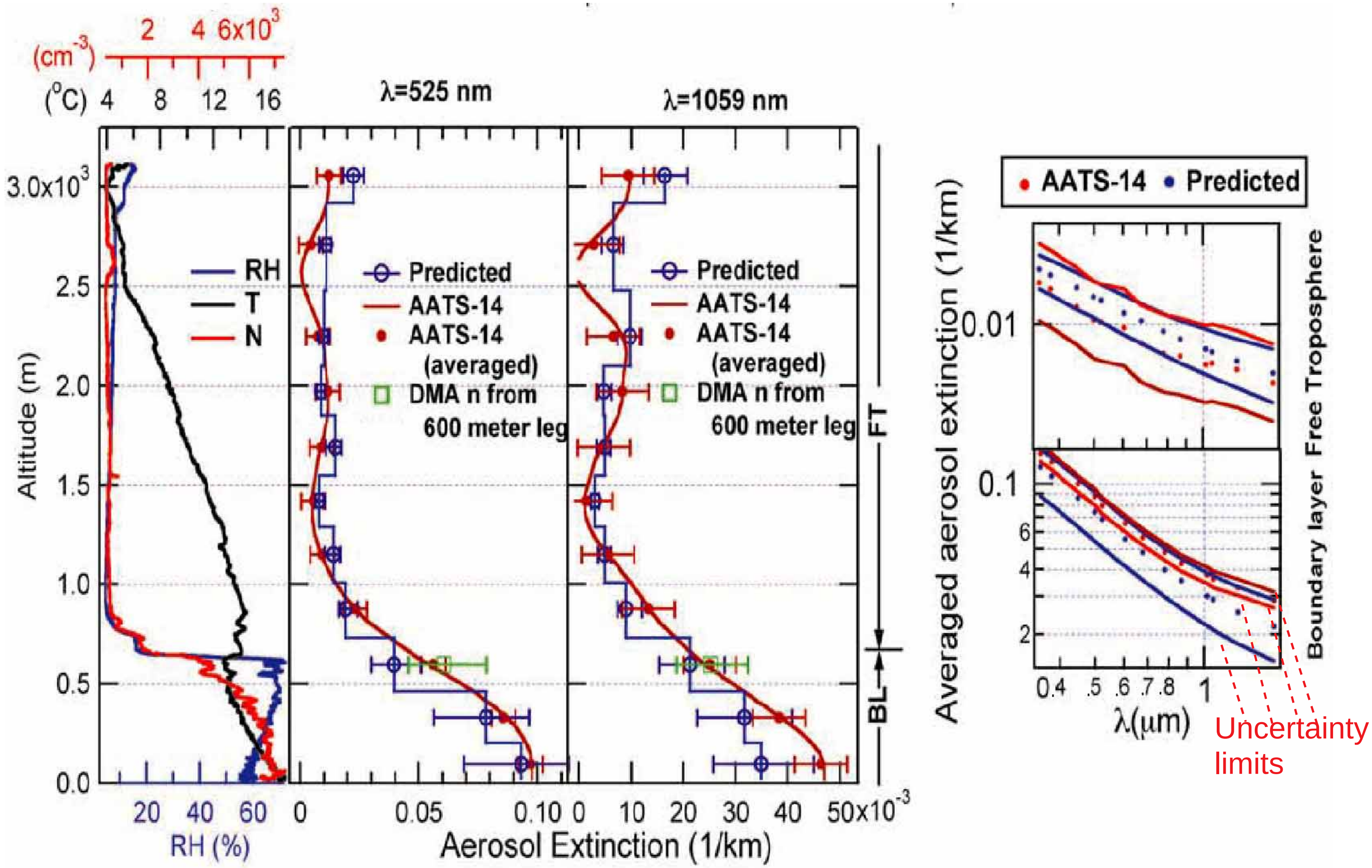


Wang et al., JGR 02

$d\sigma_{ext}/d\log D_p$ calculated by Mie theory for measured composition; index of refraction from Bruggeman mixing rule.

LOCAL OPTICAL CLOSURE EXPERIMENT

Aircraft measurements, south of Japan, ACE-Asia, 2001



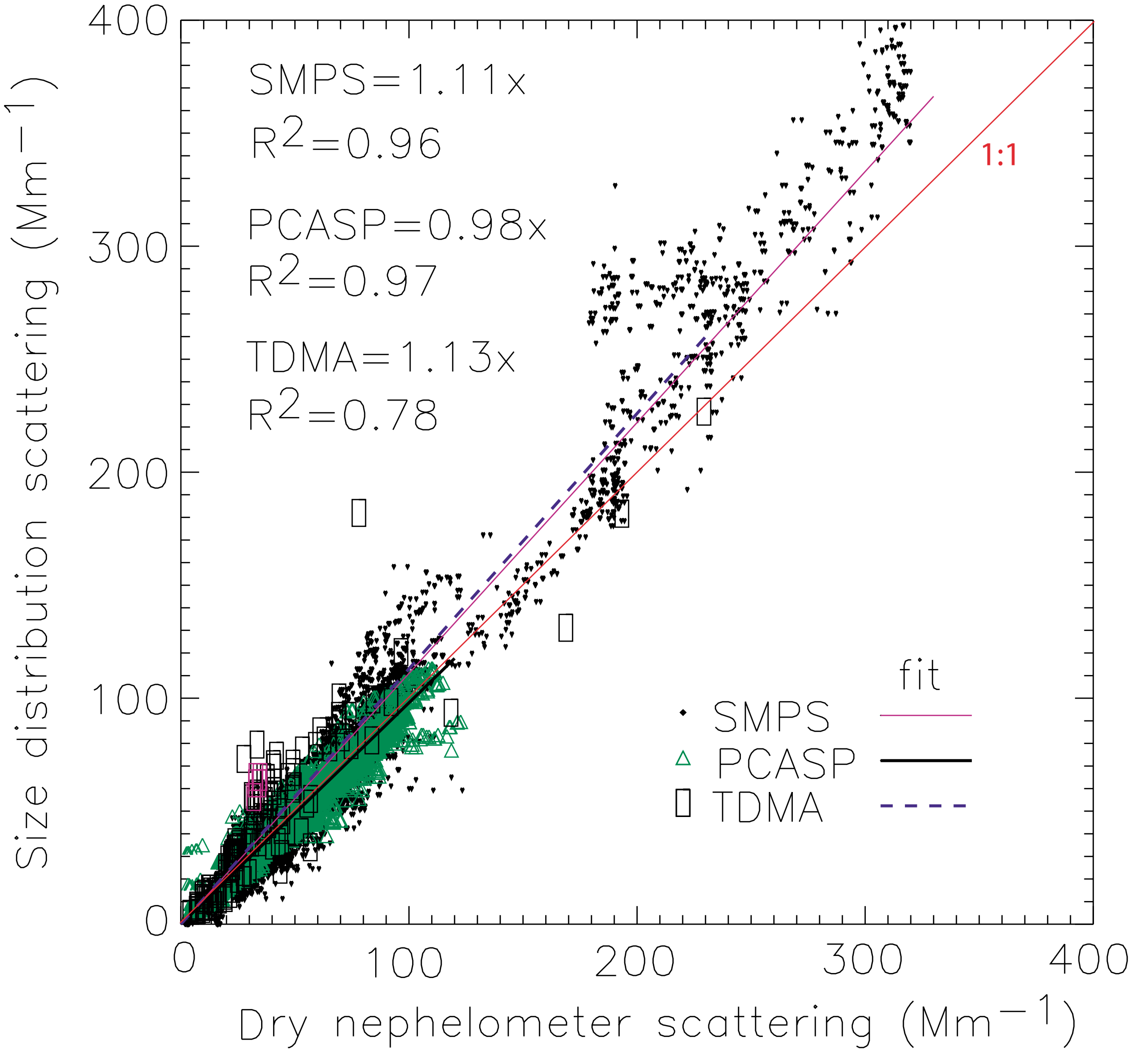
Wang et al., JGR 02

Calculated extinction coefficient and wavelength dependence agreed with observations within ***fairly broad uncertainty limits***.

MEASURED AND MODELED SCATTERING COEFFICIENT

North Central Oklahoma, May 2003

Submicrometer aerosol, low RH



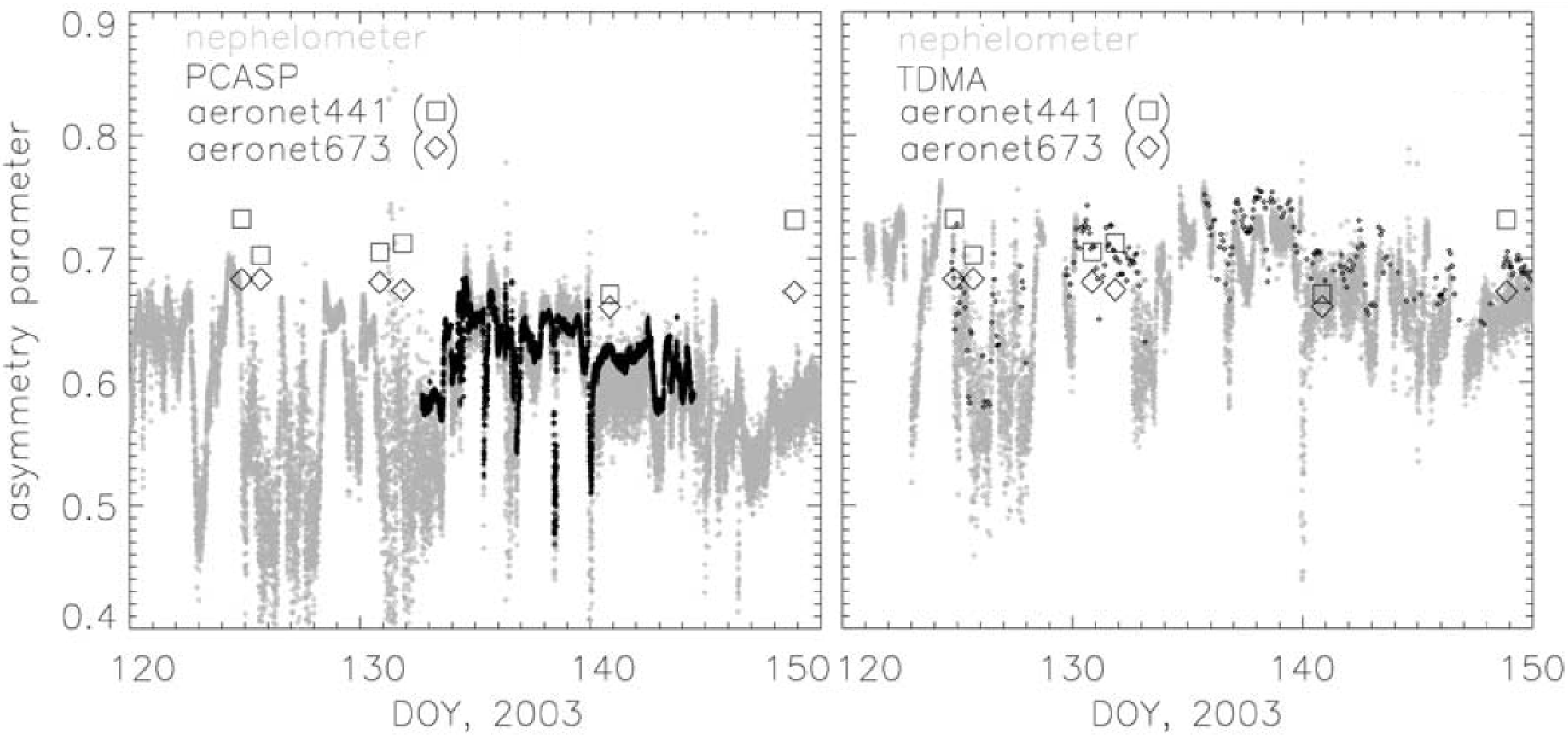
Modified from Andrews et al. JGR, 2006

MEASURED AND MODELED ASYMMETRY PARAMETER

North Central Oklahoma, May 2003

PCASP, Neph dry TDMA, 85% RH; Neph, Ambient

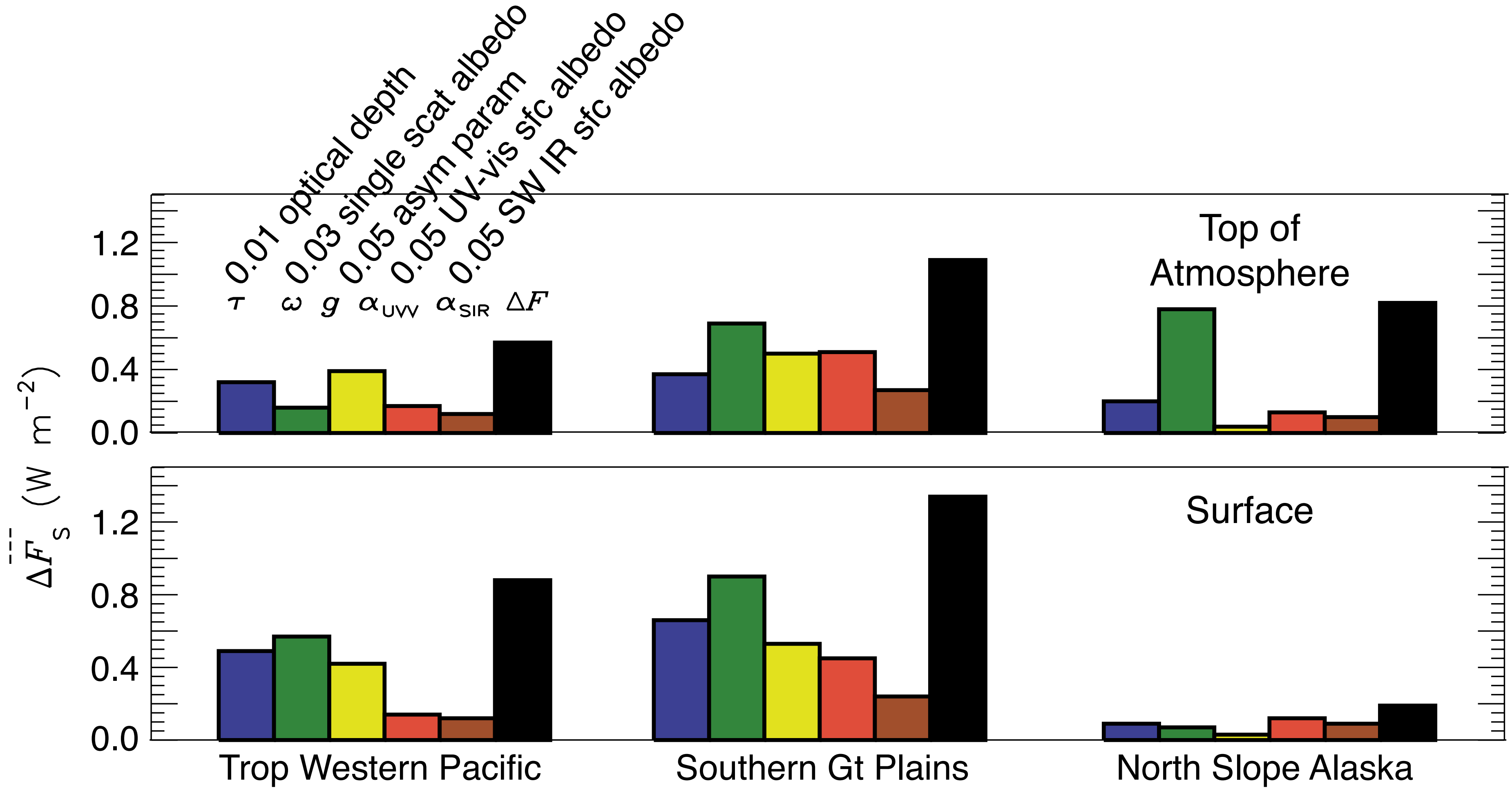
AERONET, ambient column



Andrews et al. JGR, 2006

UNCERTAINTY IN AEROSOL DIRECT FORCING

Resulting from typical uncertainty in measurements of input variables



McComiskey, Schwartz, Schmid, Guan, Lewis, Ricchiazzi, & Ogren, JGR, 2008

Colored bars denote uncertainties in 24-hr average forcing at equinox resulting from uncertainties in the individual parameters.

Black bar denotes resultant uncertainty in forcing.

Uncertainties are substantial in context of forcings over industrial period.

SOME IMPLICATIONS FOR CLIMATE CHANGE

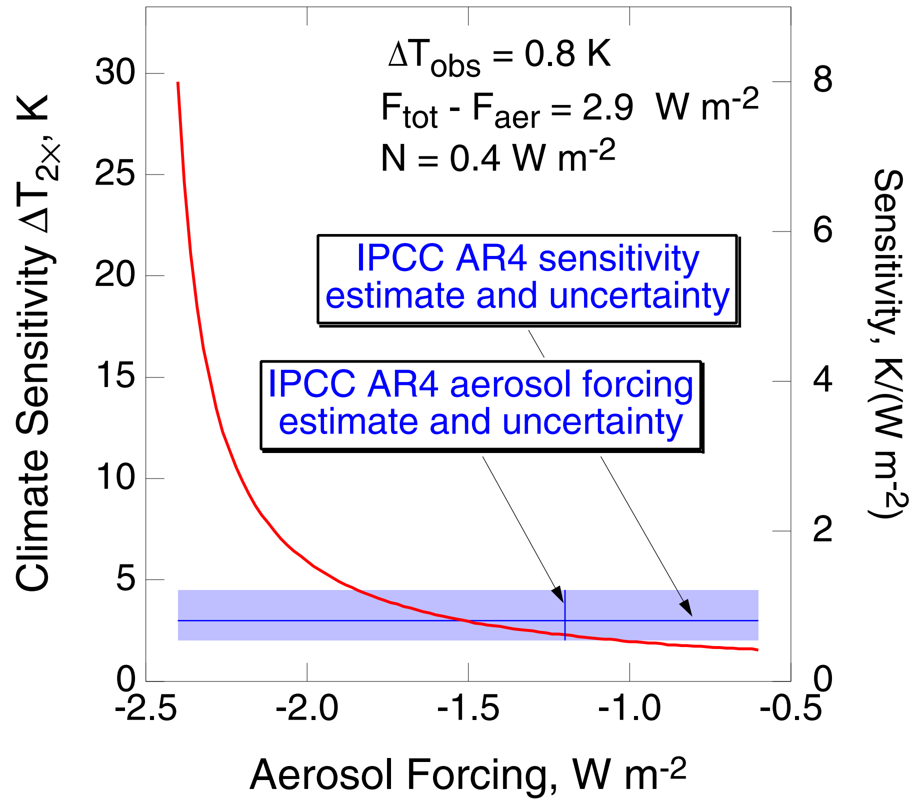
EMPIRICAL DETERMINATION OF EARTH'S CLIMATE SENSITIVITY

Dependence on aerosol forcing

$$S = \frac{\Delta T_{\mathrm{obs}}}{F_{\mathrm{eff}}}$$

$$S = \frac{\Delta T_{\mathrm{obs}}}{F_{\mathrm{tot}} - N}$$

$$S = \frac{\Delta T_{\mathrm{obs}}}{F_{\mathrm{aer}} + (F_{\mathrm{tot}} - F_{\mathrm{aer}}) - N}$$



F_{tot} - F_{aer} is due mainly to LLGHGs.

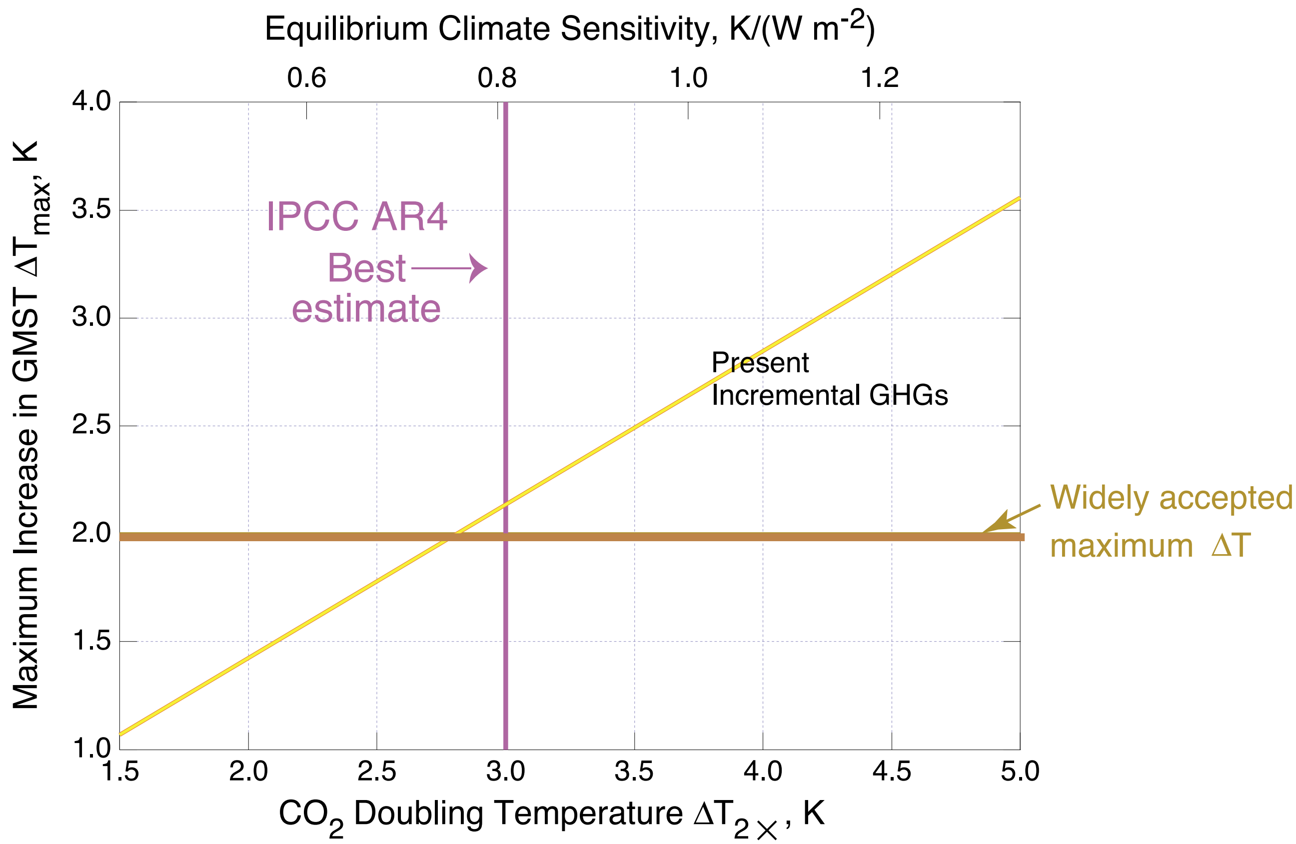
Heating rate N determined from rate of increase of ocean heat content.

Graph is plotted for uncertainty range of aerosol forcing from IPCC AR4.

Sensitivity increases strongly for aerosol forcing > 2 W m^{-2}.

ALLOWABLE FUTURE CO_2 EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

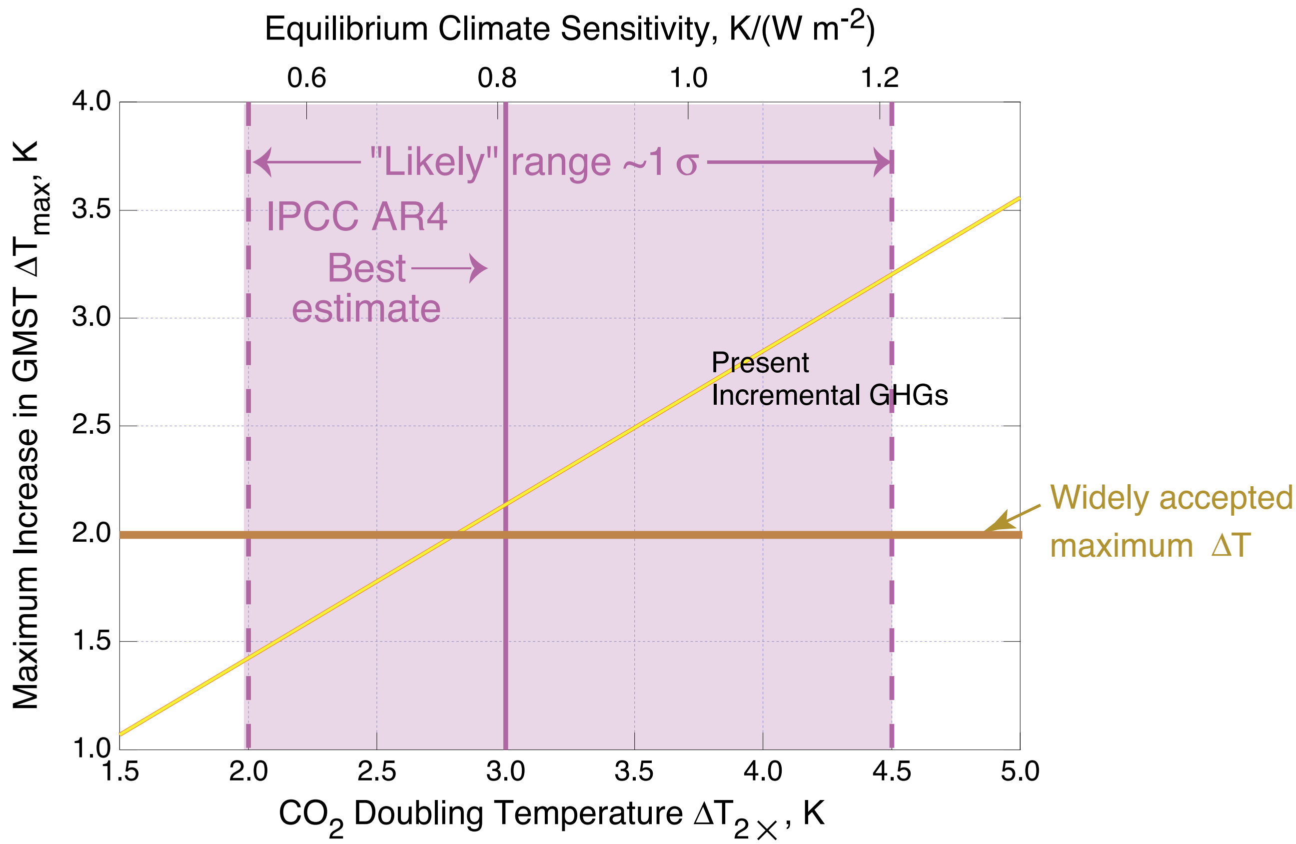


For $\Delta T_{max} = 2$ K,

If sensitivity $\Delta T_{2\times}$ is 3 K, ***no further emissions!***

ALLOWABLE FUTURE CO_2 EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

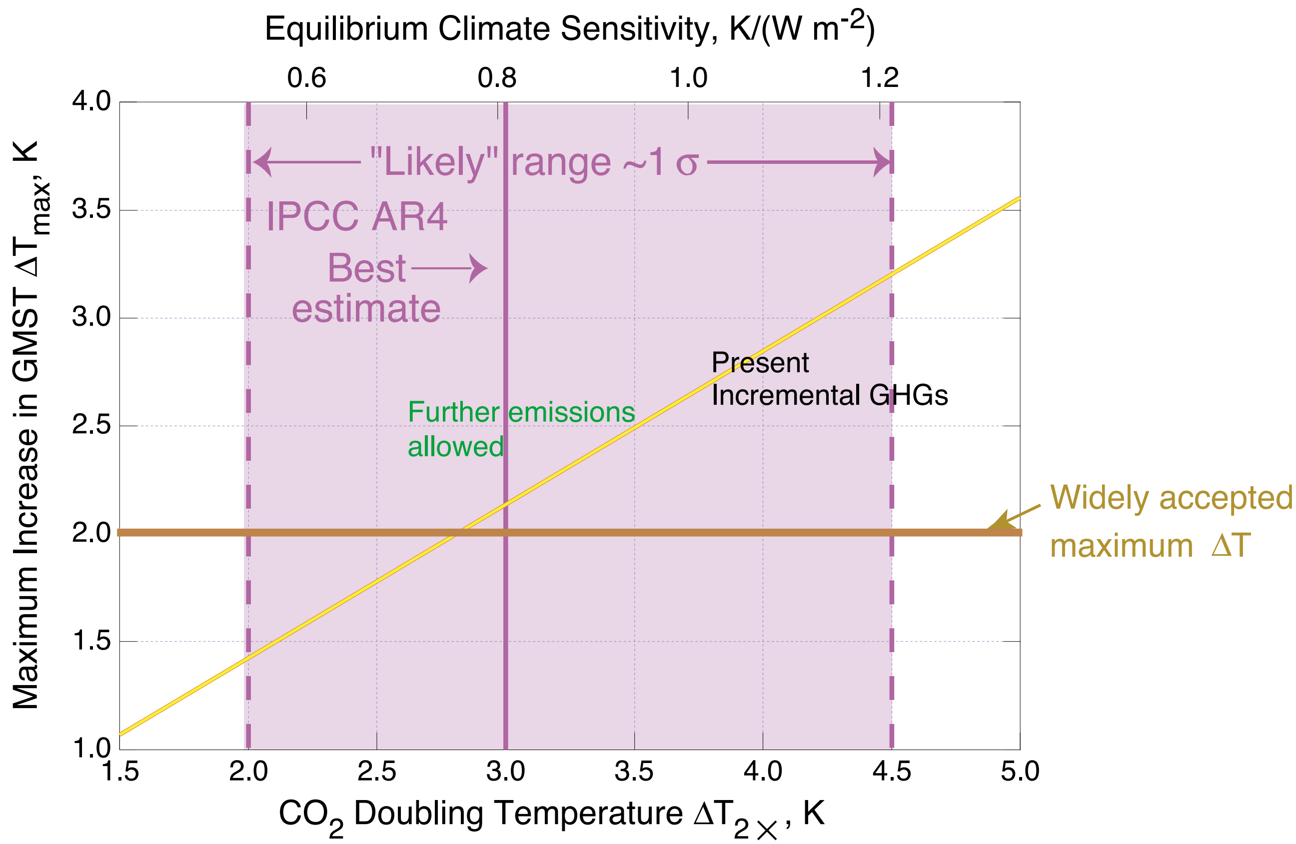


For $\Delta T_{max} = 2$ K,

Allowability of future emissions depends on climate sensitivity.

ALLOWABLE FUTURE CO_2 EMISSIONS

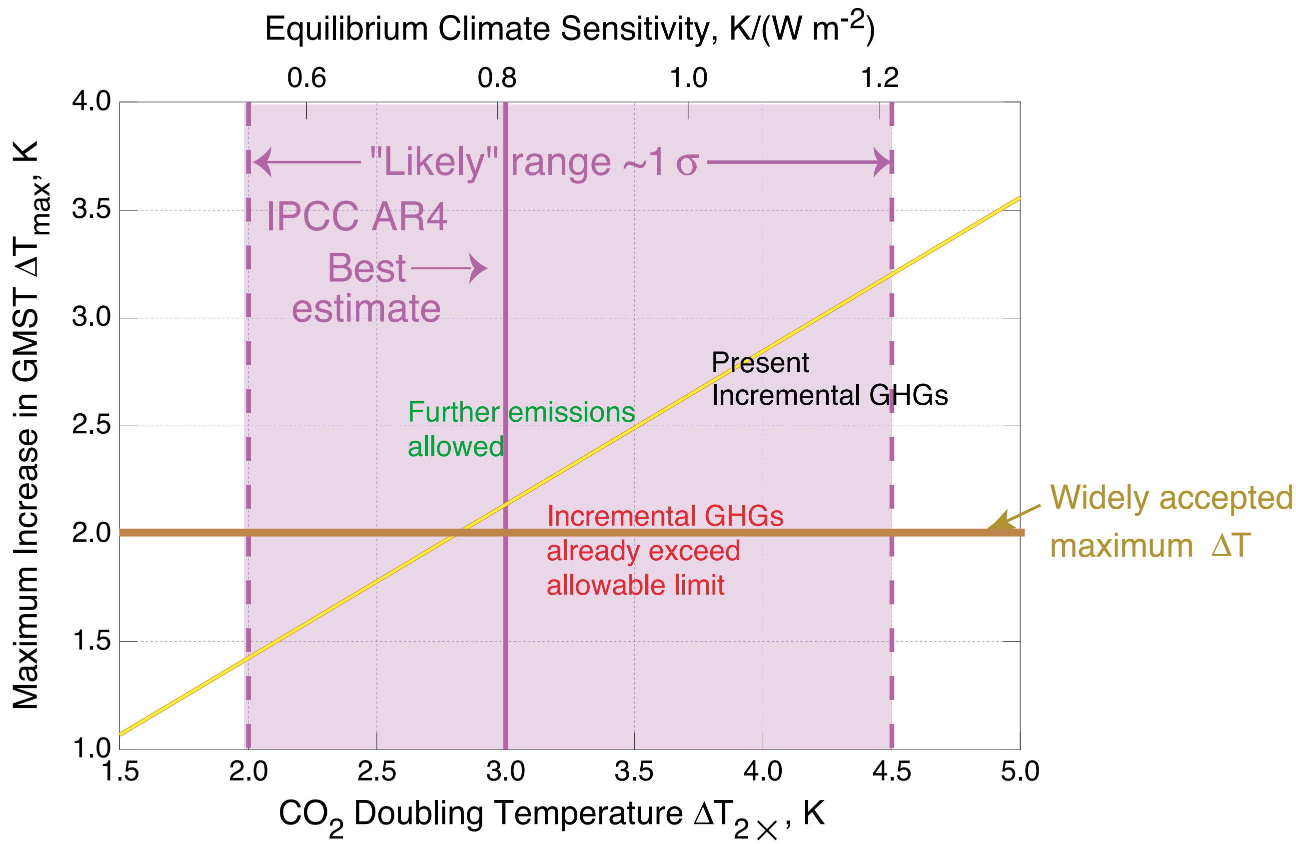
Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial



If $\Delta T_{max} > 2.1$ K and/or sensitivity $\Delta T_{2\times} < 3$ K, further emissions are allowed without exceeding ΔT_{max}.

ALLOWABLE FUTURE CO_2 EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial



If $\Delta T_{max} > 2.1$ K and/or sensitivity $\Delta T_{2\times} < 3$ K, further emissions are allowed without exceeding ΔT_{max}.

If $\Delta T_{max} < 2.1$ K and/or sensitivity $\Delta T_{2\times} > 3$ K, committed temperature increase already exceeds ΔT_{max}.

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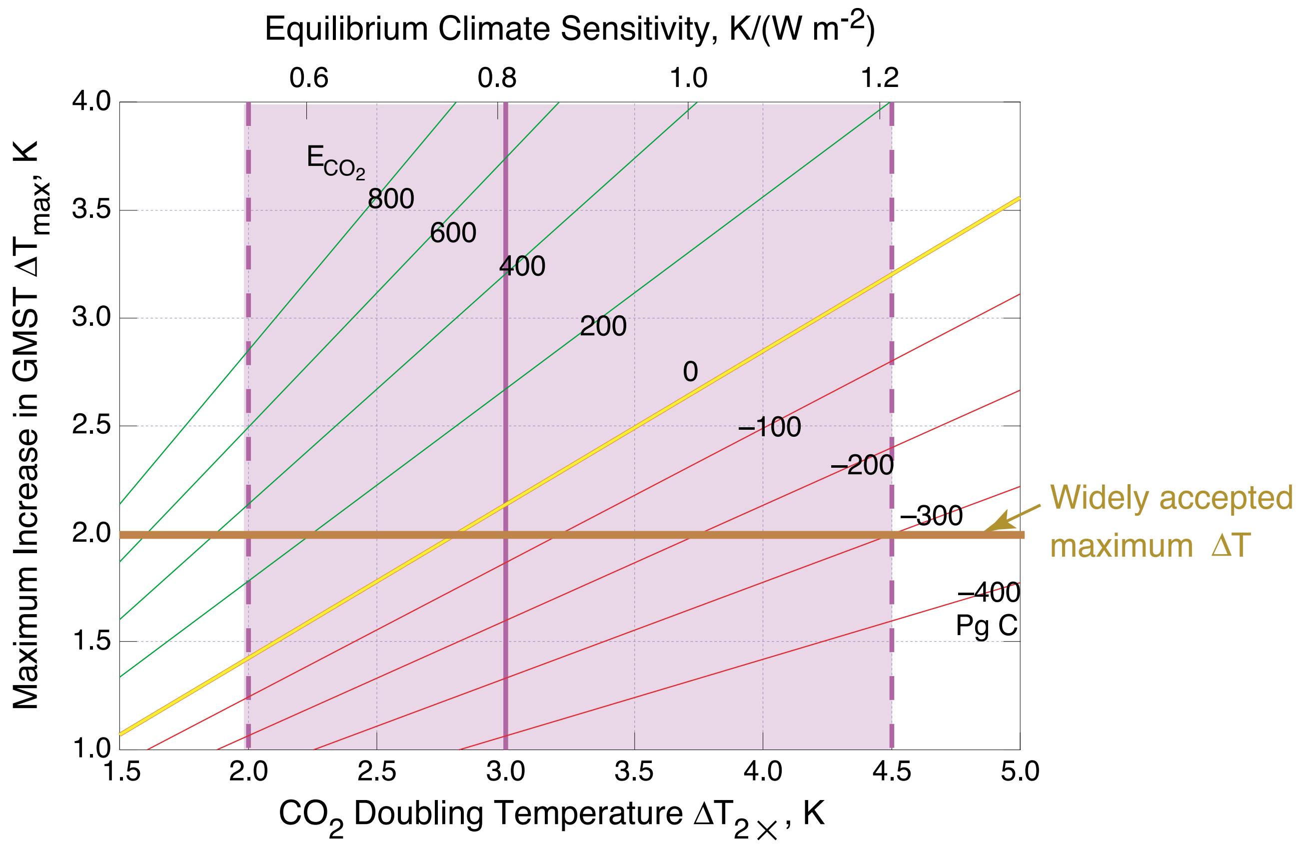
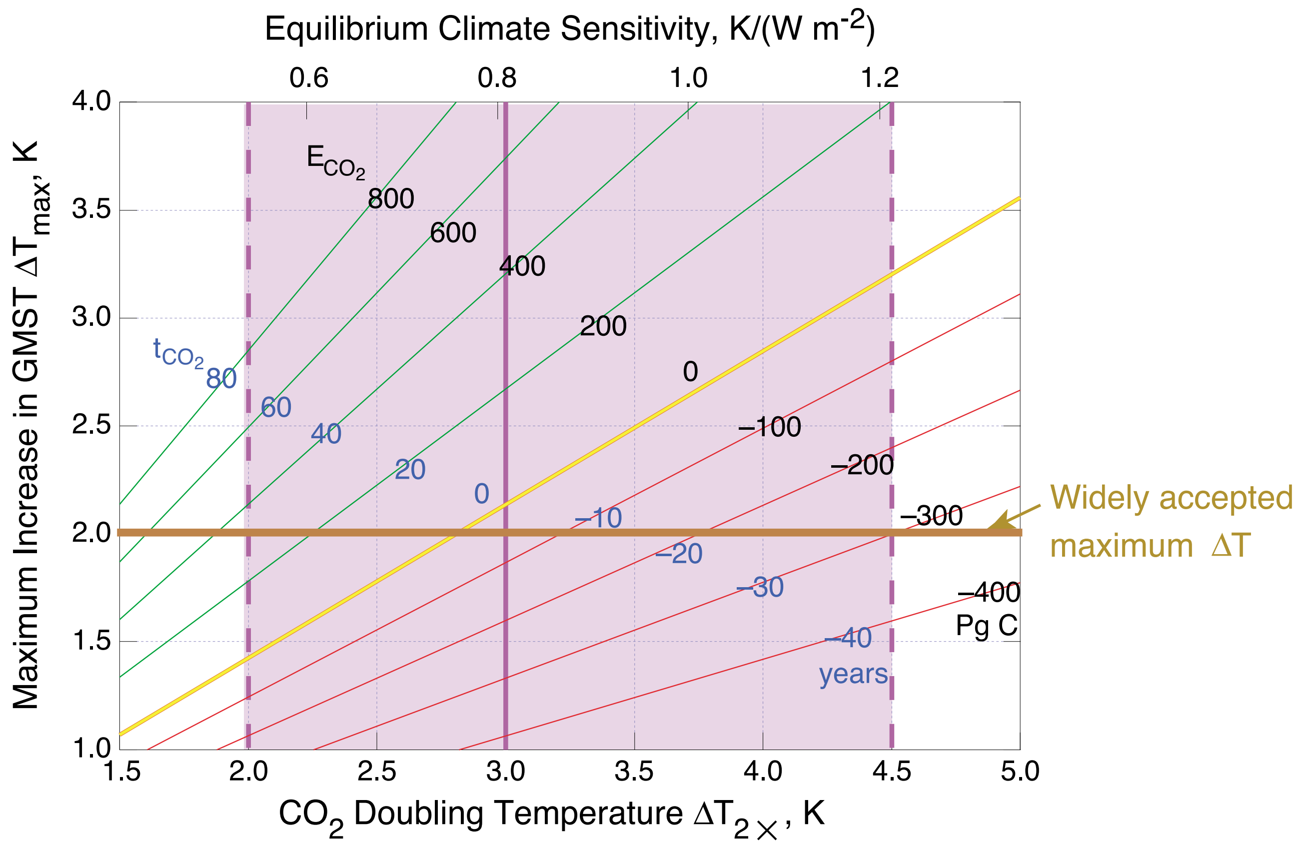


Allowable future emissions ***or amount by which present GHGs exceed the allowable threshold*** depend on climate sensitivity and ΔT_{max}.

ALLOWABLE FUTURE CO_2 EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial



Schwartz, Charlson, Kahn, Ogren & Rodhe, J. Clim. 2010

For $\Delta T_{max} = 2$ K . . .

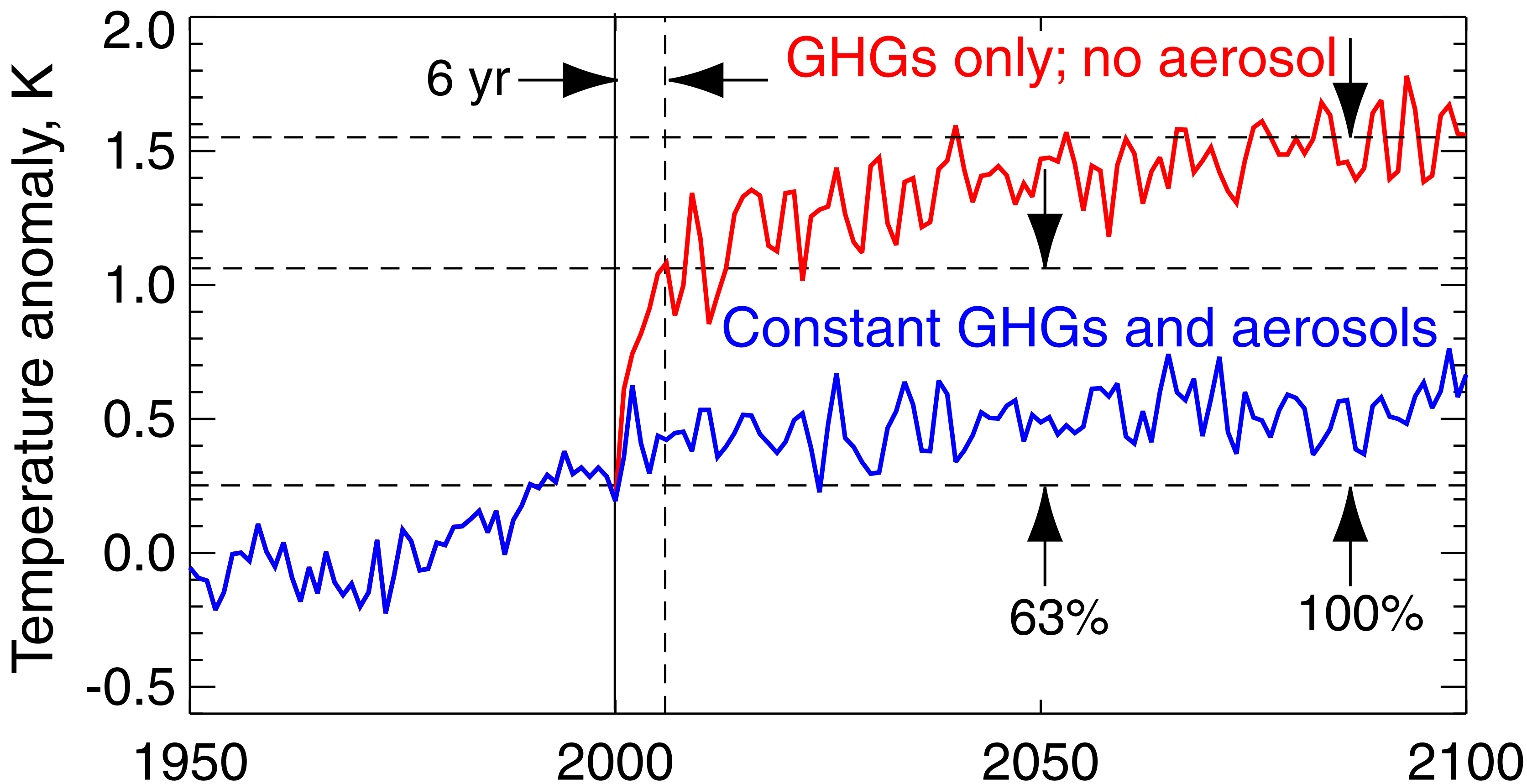
If sensitivity $\Delta T_{2\times}$ is 3 K, ***no more emissions***.

If sensitivity $\Delta T_{2\times}$ is 2 K, ~ ***30 more years of emissions at present rate***.

If sensitivity $\Delta T_{2\times}$ is 4.5 K, ***threshold is exceeded by ~30 years***.

GCM TEMPERATURE RESPONSE TO REMOVAL OF AEROSOL SOURCES

Experiment with ECHAM-5 GCM



Modified from Brasseur and Roeckner, GRL, 2005

Aerosols are removed from atmosphere in days to weeks.

Global temperature rapidly increases following removal of aerosol forcing.

Time constant for climate response to step-change in forcing is about 6 years.

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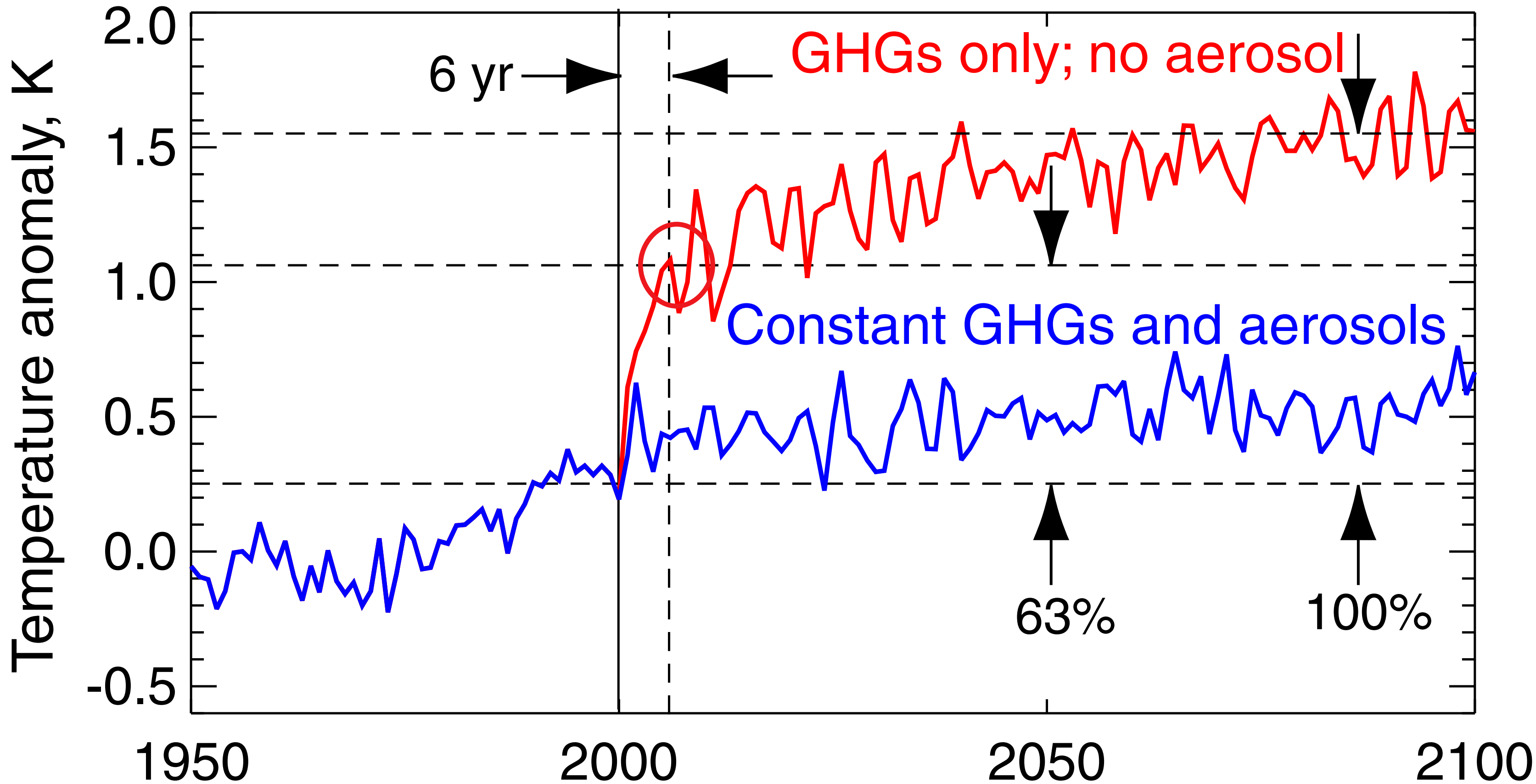

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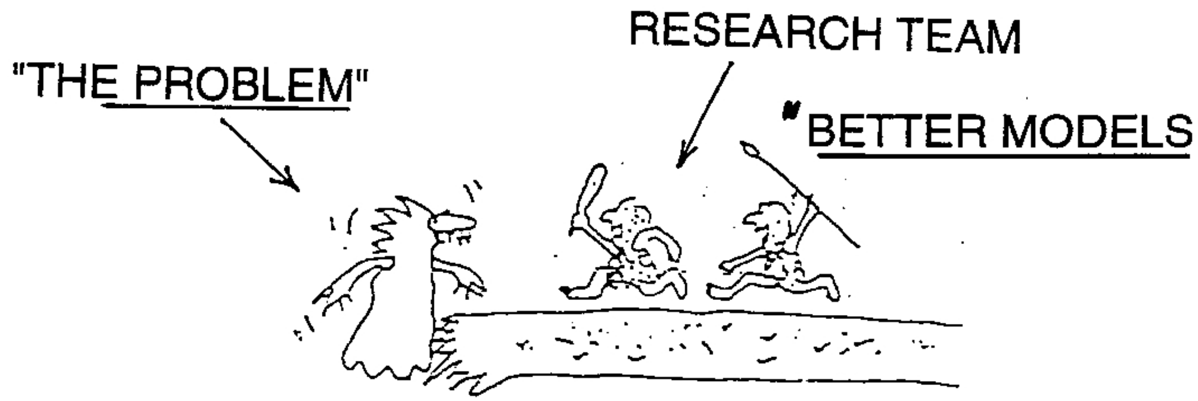
"THE CLIMATE PREDICTION PROBLEM"



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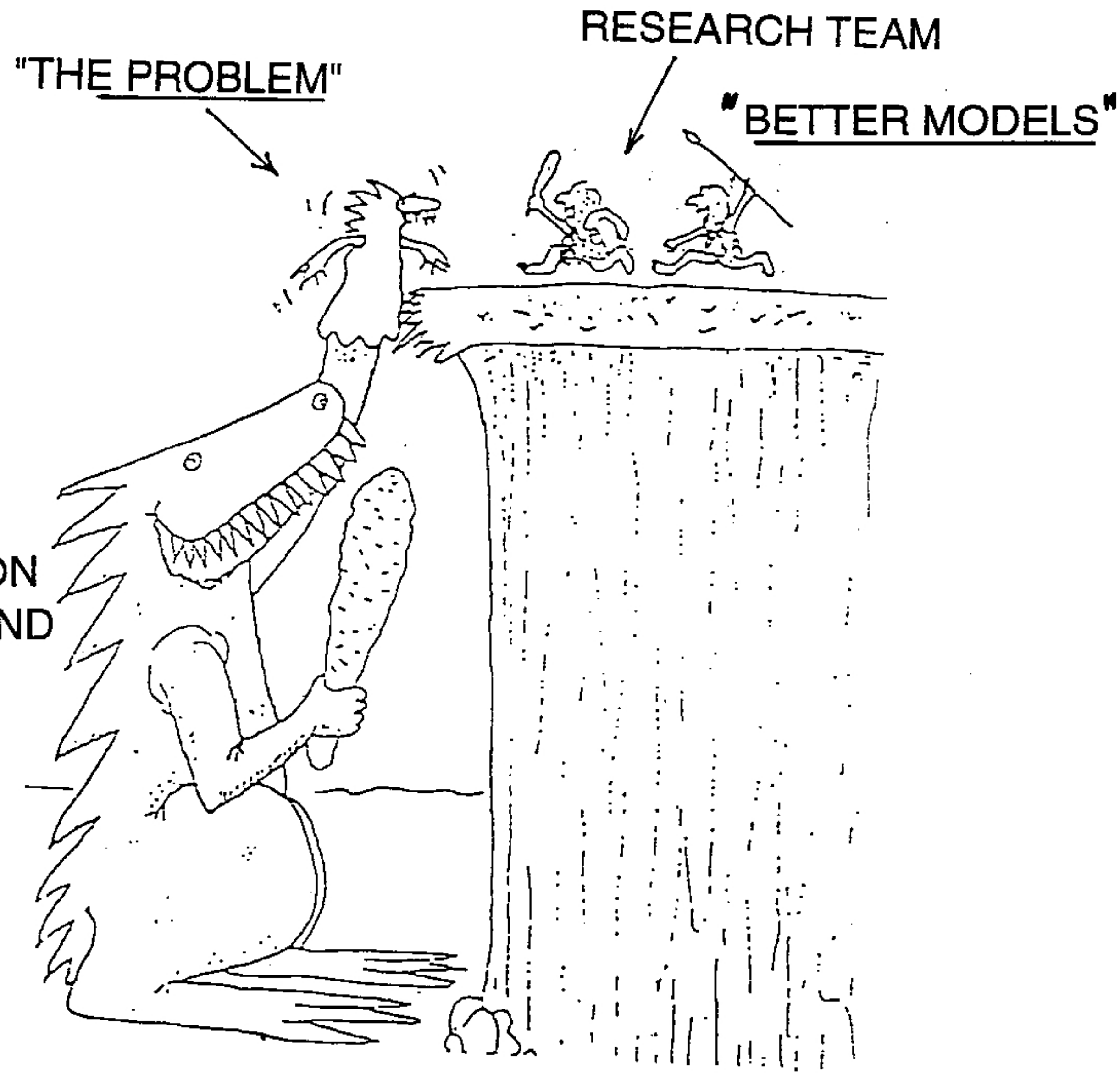

THE REAL PROBLEM

INADEQUATE KNOWLEDGE
OF THE GLOBAL DISTRIBUTION
OF *AEROSOL* PROPERTIES AND
PROCESSES!