DO AEROSOLS CHANGE CLOUD COVER AND AFFECT CLIMATE?

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DO AEROSOLS CHANGE CLOUD COVER AND AFFECT CLIMATE?

Yes

IN WHAT WAYS?

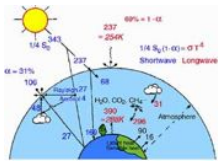
Direct (scattering, absorption)
Indirect (albedo, lifetime)
Semi-direct, ...
Autoconversion, latent heat, ...

HOW MUCH?

???????

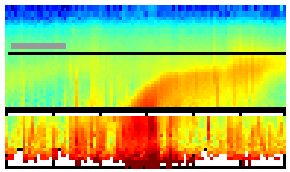
OVERVIEW

Aerosol influences on climate and climate change

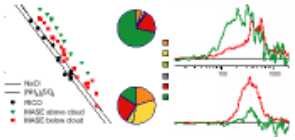


Relation to climate change over the industrial era

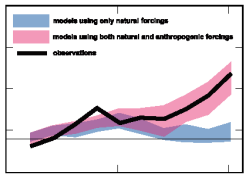
Aerosol properties and processes pertinent to climate influences



Recent studies examining aerosol processes



Recent studies examining aerosol indirect effects



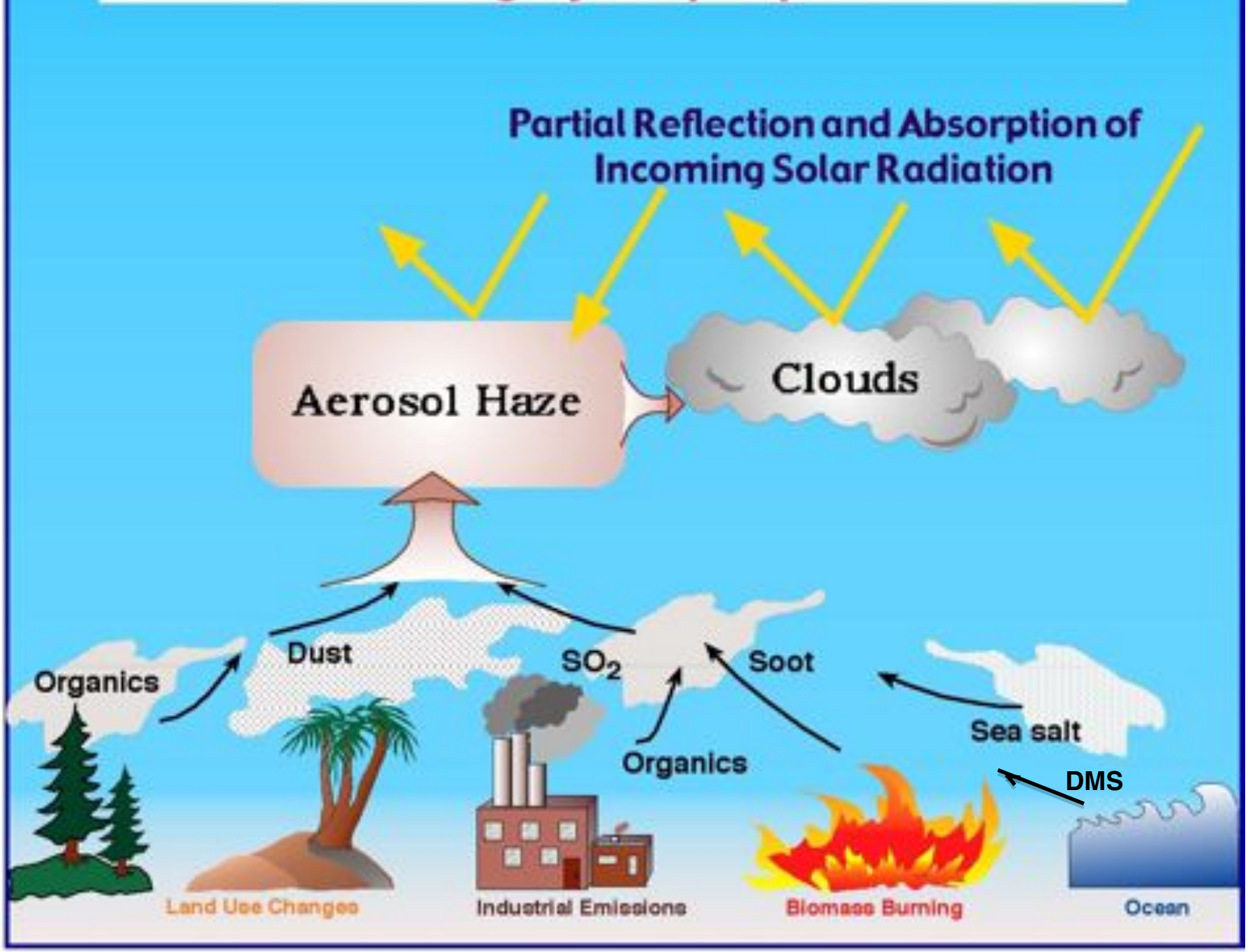
Implications of aerosol forcing on interpretation of climate change

The path forward

Concluding remarks

AEROSOL INFLUENCES ON CLIMATE AND CLIMATE CHANGE

Radiative Forcing by Tropospheric Aerosol



AEROSOL IN MEXICO CITY BASIN

AEROSOL IN MEXICO CITY BASIN

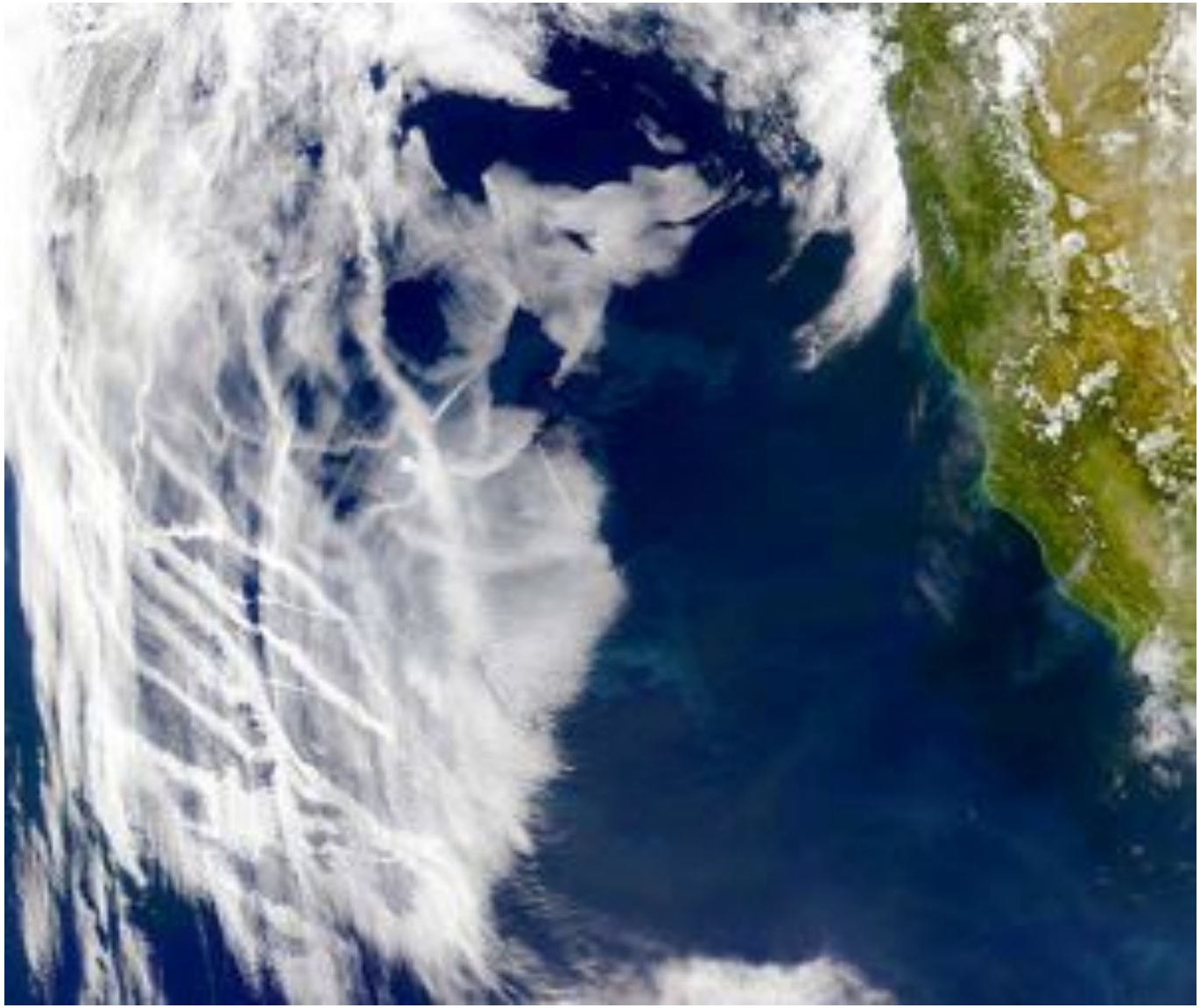
Light scattering by aerosols decreases absorption of solar radiation.

AEROSOLS AS SEEN FROM SPACE

Fire plumes from southern Mexico transported north into Gulf of Mexico.

CLOUD BRIGHTENING BY SHIP TRACKS

Satellite photo off California coast

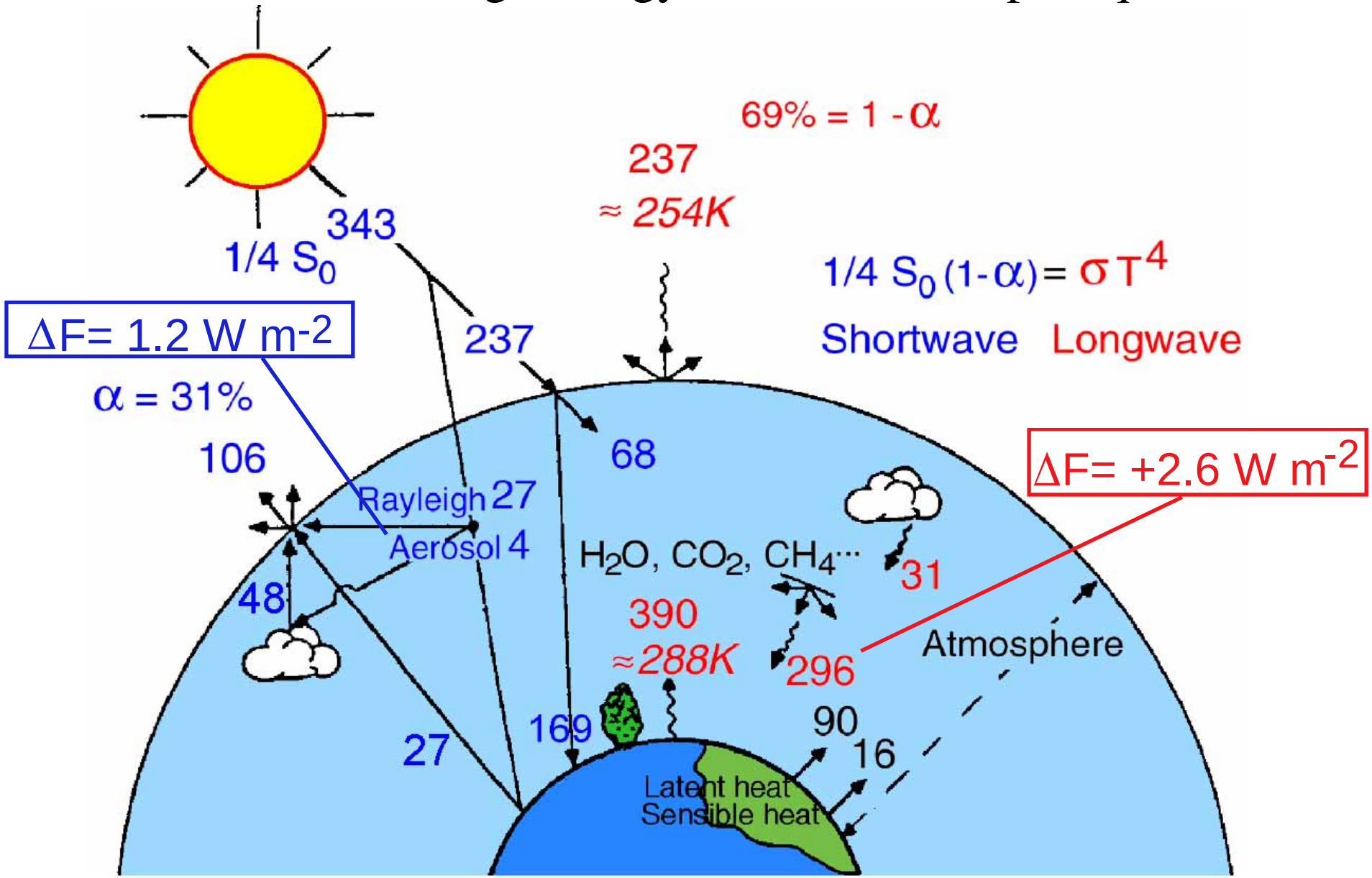


Aerosols from ship emissions enhance reflectivity of marine stratus.

RELATION TO CLIMATE CHANGE OVER THE INDUSTRIAL ERA

GLOBAL ENERGY BALANCE

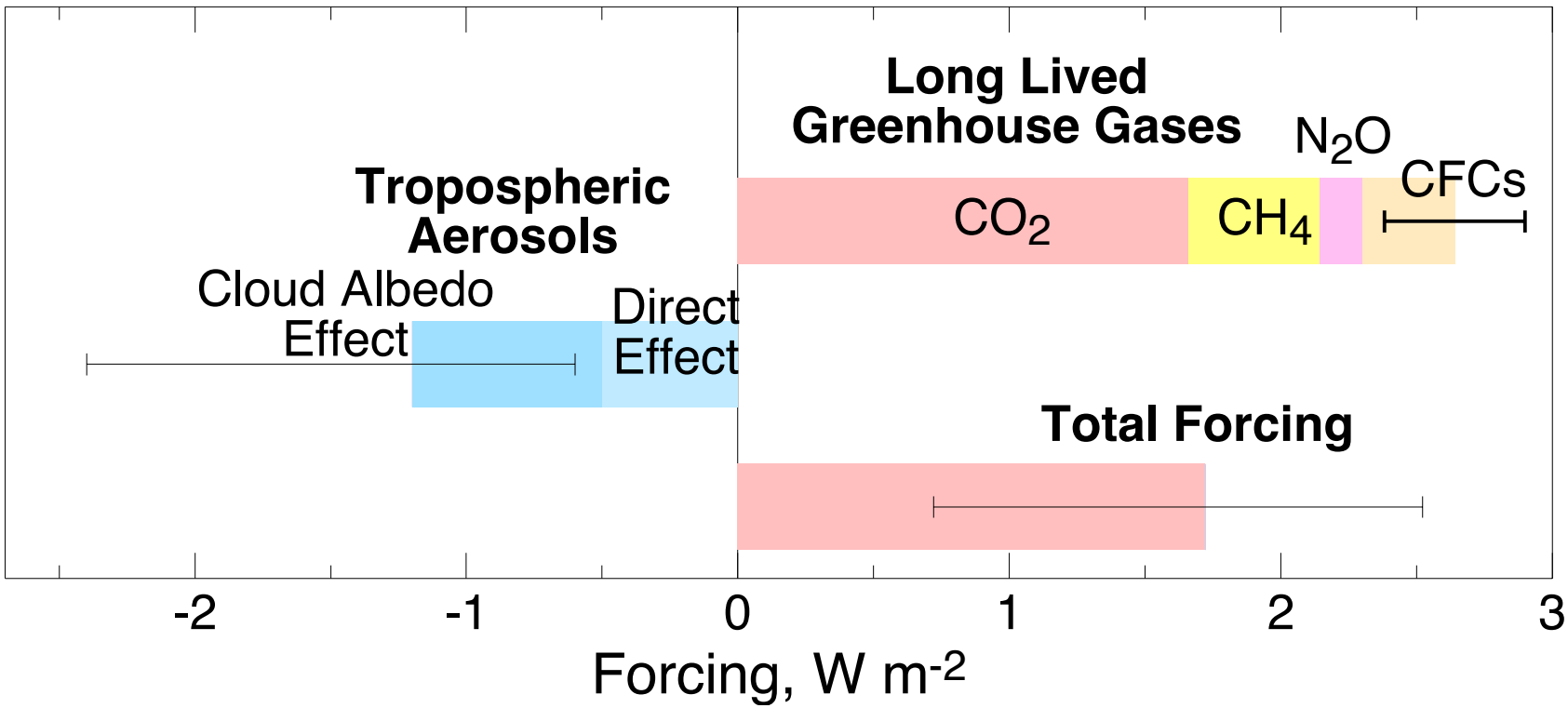
Global and annual average energy fluxes in watts per square meter



Schwartz, 1996, modified from Ramanathan, 1987

CLIMATE FORCINGS OVER THE INDUSTRIAL PERIOD

Extracted from IPCC AR4 (2007)

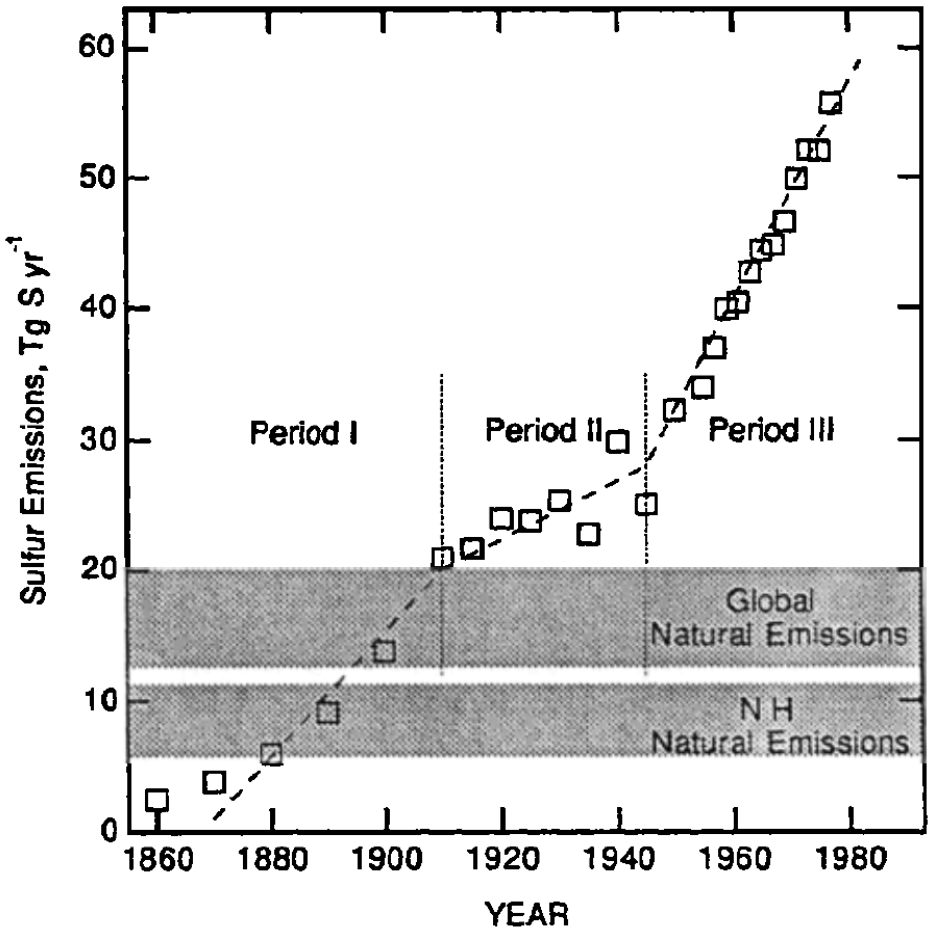


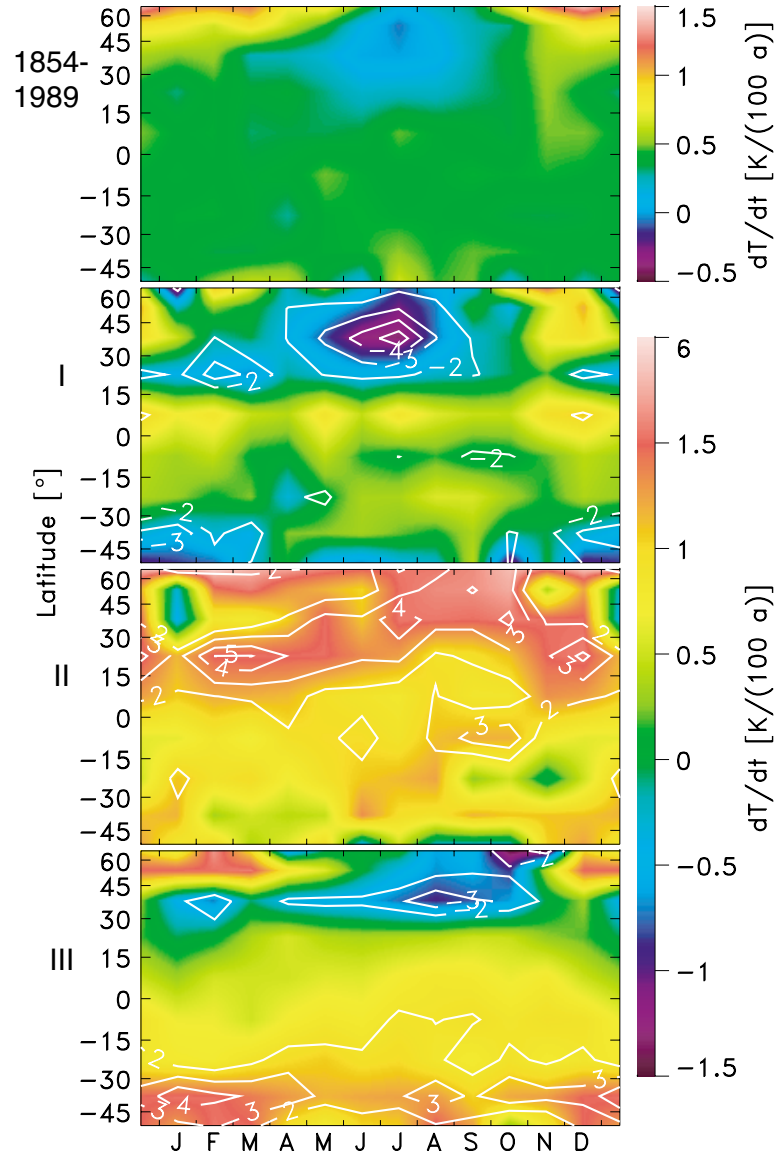
Total forcing includes other anthropogenic and natural (solar) forcings.

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

Uncertainty in aerosol forcing dominates uncertainty in total forcing.

LATITUDINAL, ANNUAL DEPENDENCE OF TEMPERATURE CHANGE





Hunter, Schwartz, Wagener & Benkovitz, GRL 1993

Greatest cooling is midlatitude NH summer during rapid increase of sulfur emission.

REQUIREMENTS TO QUANTIFY AEROSOL FORCING

Quantify change in Earth radiation budget due to aerosols as f(time).

- ***Direct*** radiative influences; ***Indirect*** influences, on clouds and precip.
- Input to climate models.
- Required accuracy in forcing *ca*. 0.3 W m^{-2}.

Relate quantitatively to emissions of primary aerosols and precursor gases.

- Necessary for past, present & projected future forcing as $f(x, y, z, t)$.

Represent processes governing aerosol radiative influences in models.

- Chemical transport models & climate models.
- Required ***optical*** properties: Extinction, single scattering albedo, phase function as $f(\lambda; x, y, z, t)$.
- Required ***cloud-nucleating*** properties: CCN, IFN conc as f(supersat'n)

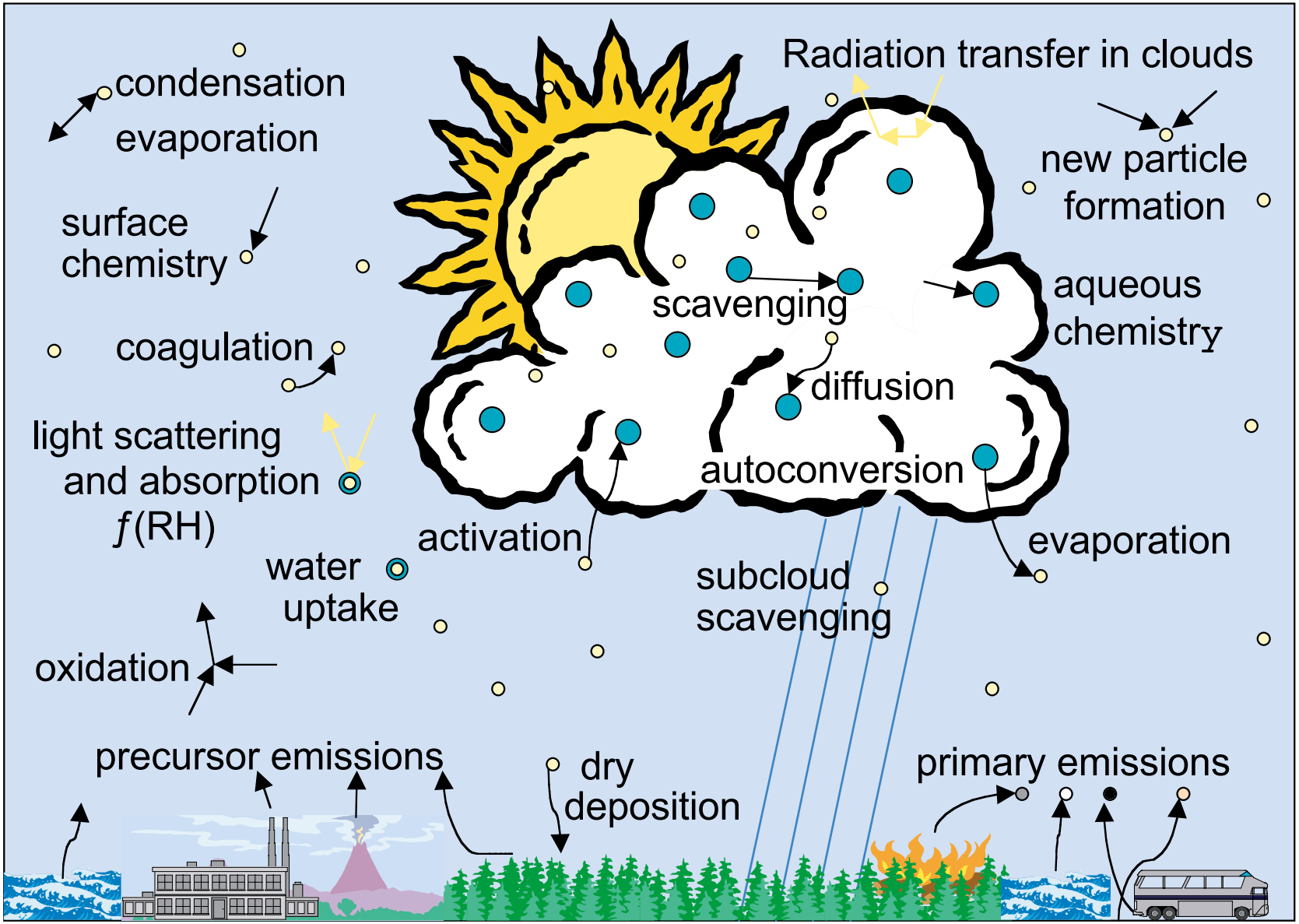
Understand ***processes*** governing aerosol amount and chem & microphys properties as $f(x, y, z, t)$.

Calculate aerosol ***optical*** and ***cloud nucleating*** properties from size-distrib composition.

Evaluate ***accuracy*** of models by observation.

AEROSOL PROPERTIES AND PROCESSES

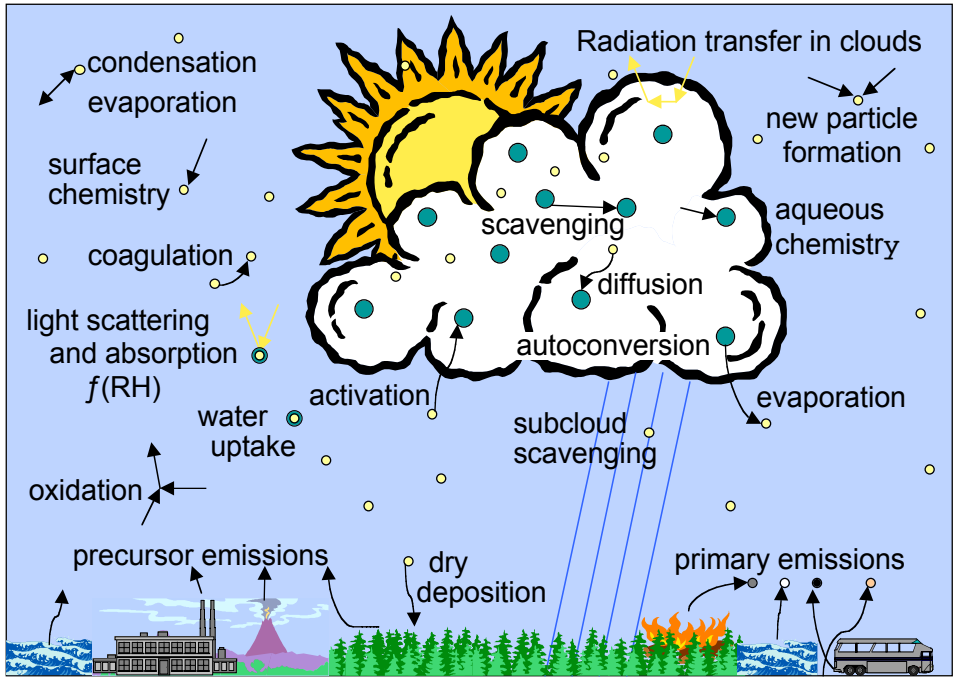
AEROSOL PROCESSES THAT MUST BE UNDERSTOOD AND REPRESENTED IN MODELS

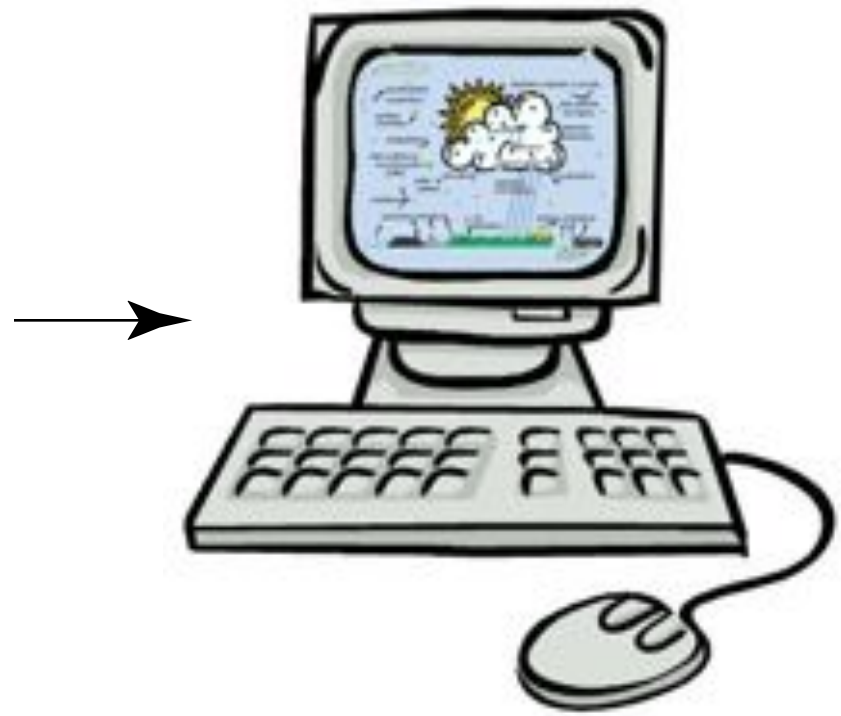


Modified from Ghan and Schwartz, Bull. Amer. Meterol. Soc., 2007

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.

ARE WE THERE YET?

Much research: ***hundreds of papers per year***.

- Process research - field studies, lab studies, theory, modeling.
- Several major field campaigns per year.
- Observations - surface, satellite, *in-situ*.

Poor understanding of primary aerosol emissions: anthro & natural.

Pretty good model-observation agreement in some observables, *e.g.*, optical depth; compensation in models.

Poor agreement in attribution to chemical substances; anthro vs. natural.

Forcing is still quite uncertain.

- IPCC AR4 (2007): direct, ±0.4 W m^{-2}; first indirect, +0.4, -0.8 W m^{-2}.
- Other indirect effects even more poorly understood and quantified.

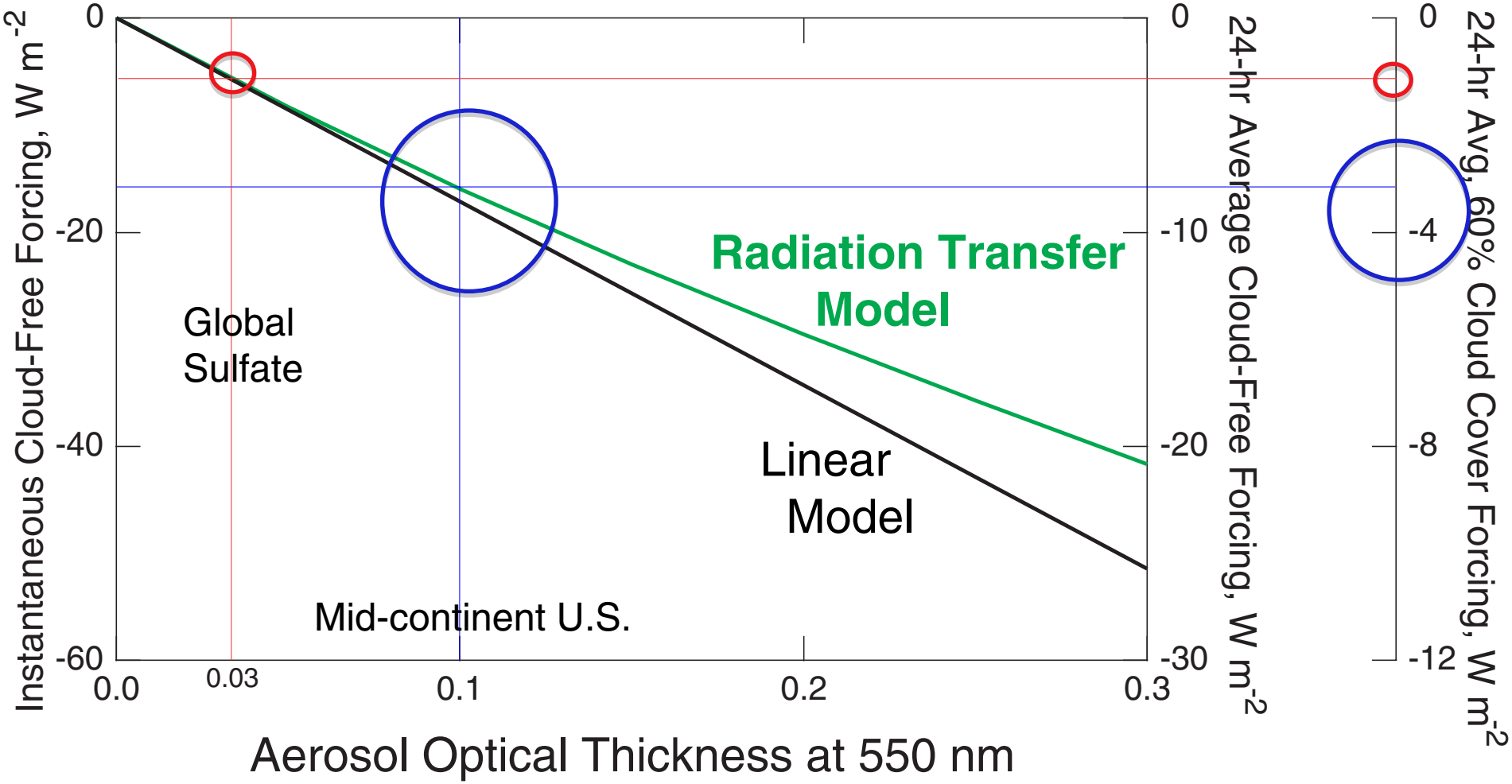
Still in discovery stage: chemistry, microphysics.

- New particle formation, involving not the usual suspects, affecting aerosol dynamics and number concentration.
- Large contribution of secondary organics.
- New effects, especially involving clouds, autoconversion.

Models are lagging the research.

ESTIMATES OF AEROSOL DIRECT FORCING

By linear model and by radiation transfer modeling

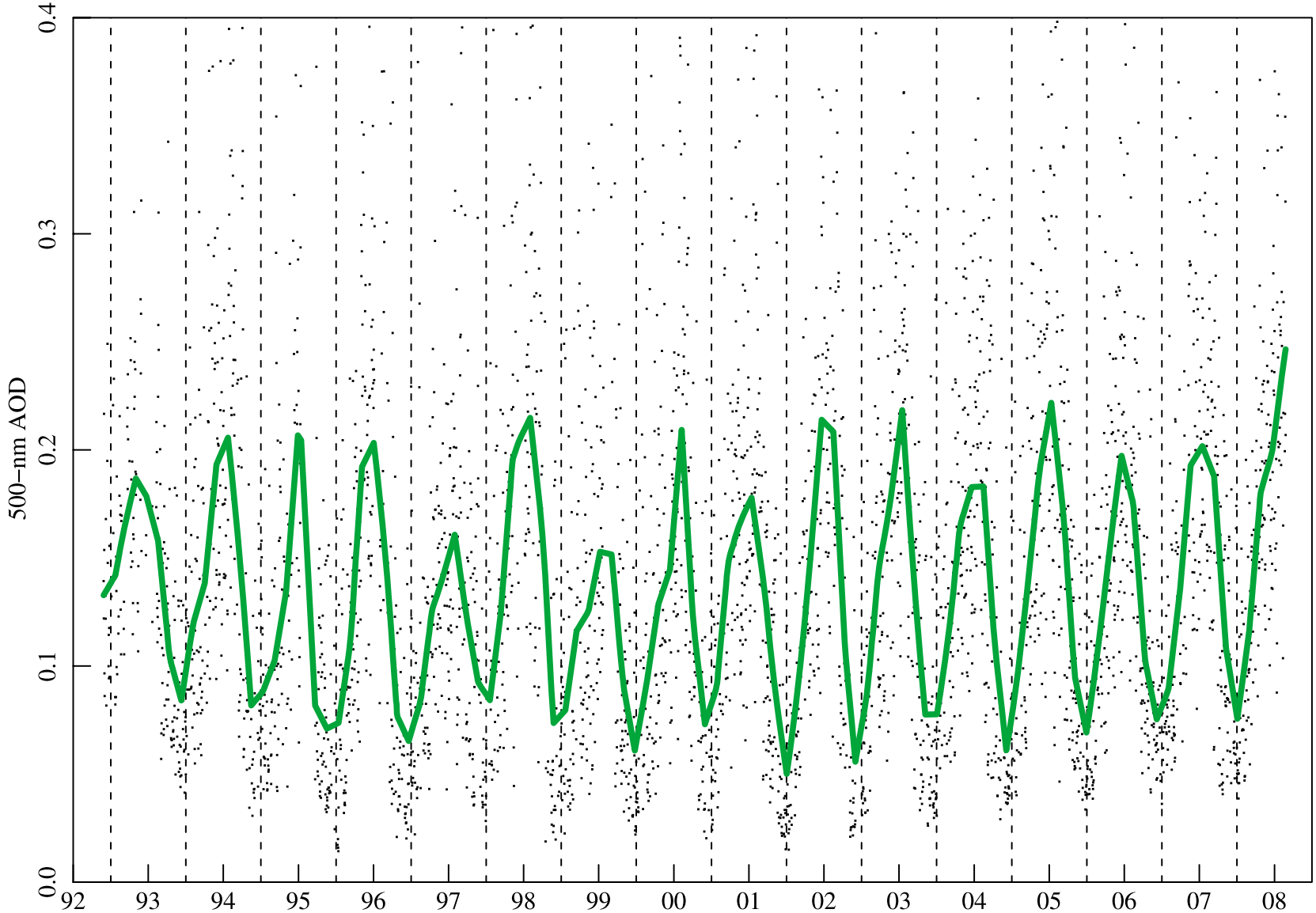


Global average sulfate optical thickness is 0.03: ***1 W m^{-2} cooling***.

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

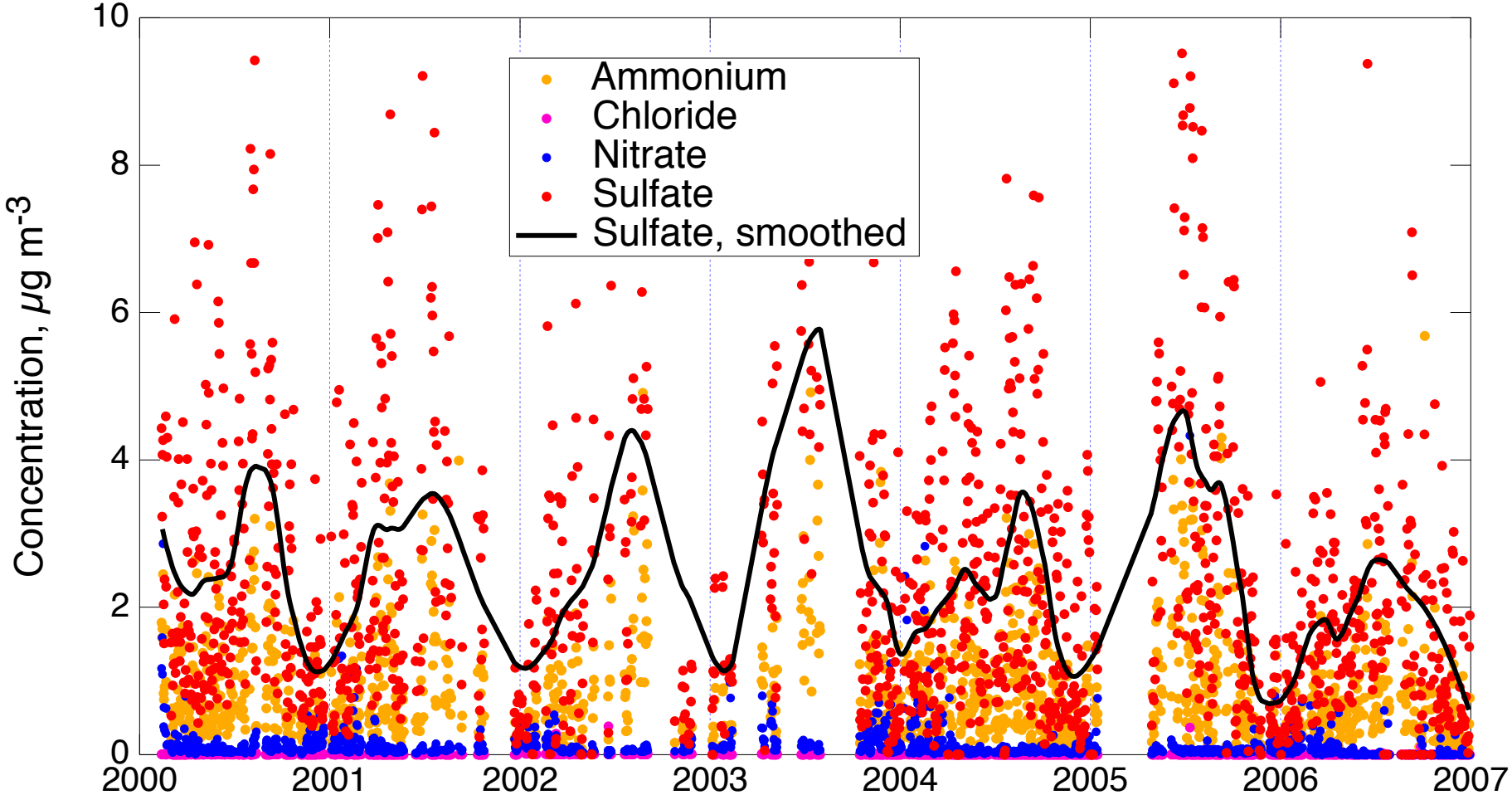


Michalsky, Denn, Flynn, Hodges, Kiedron, Koontz, Schlemmer, Schwartz, JGR, 2010

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

AEROSOL COMPOSITION AT ARM SGP

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

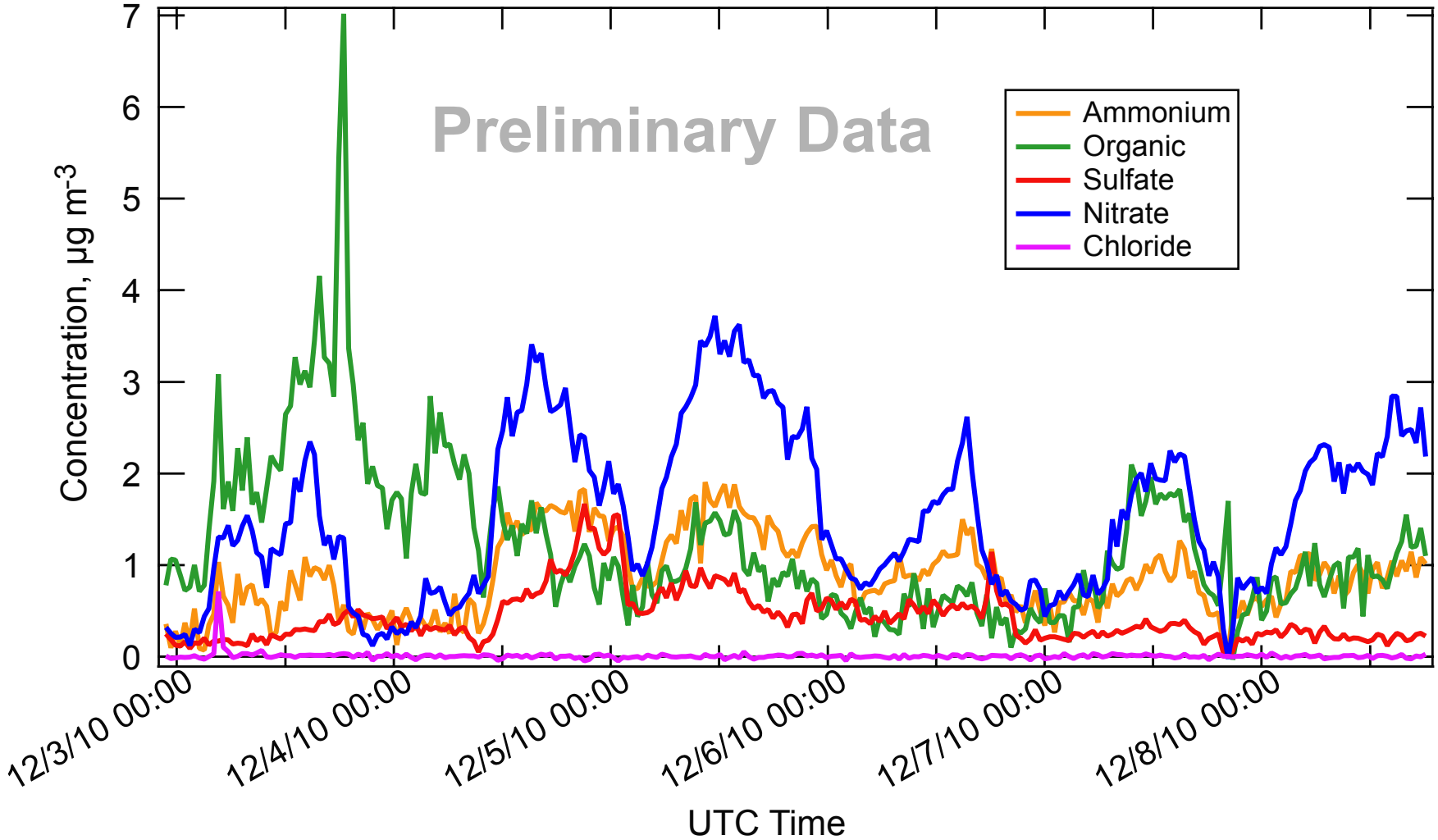


Measurements of P. Quinn, NOAA, PMEL

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

AEROSOL COMPOSITION AT ARM SGP

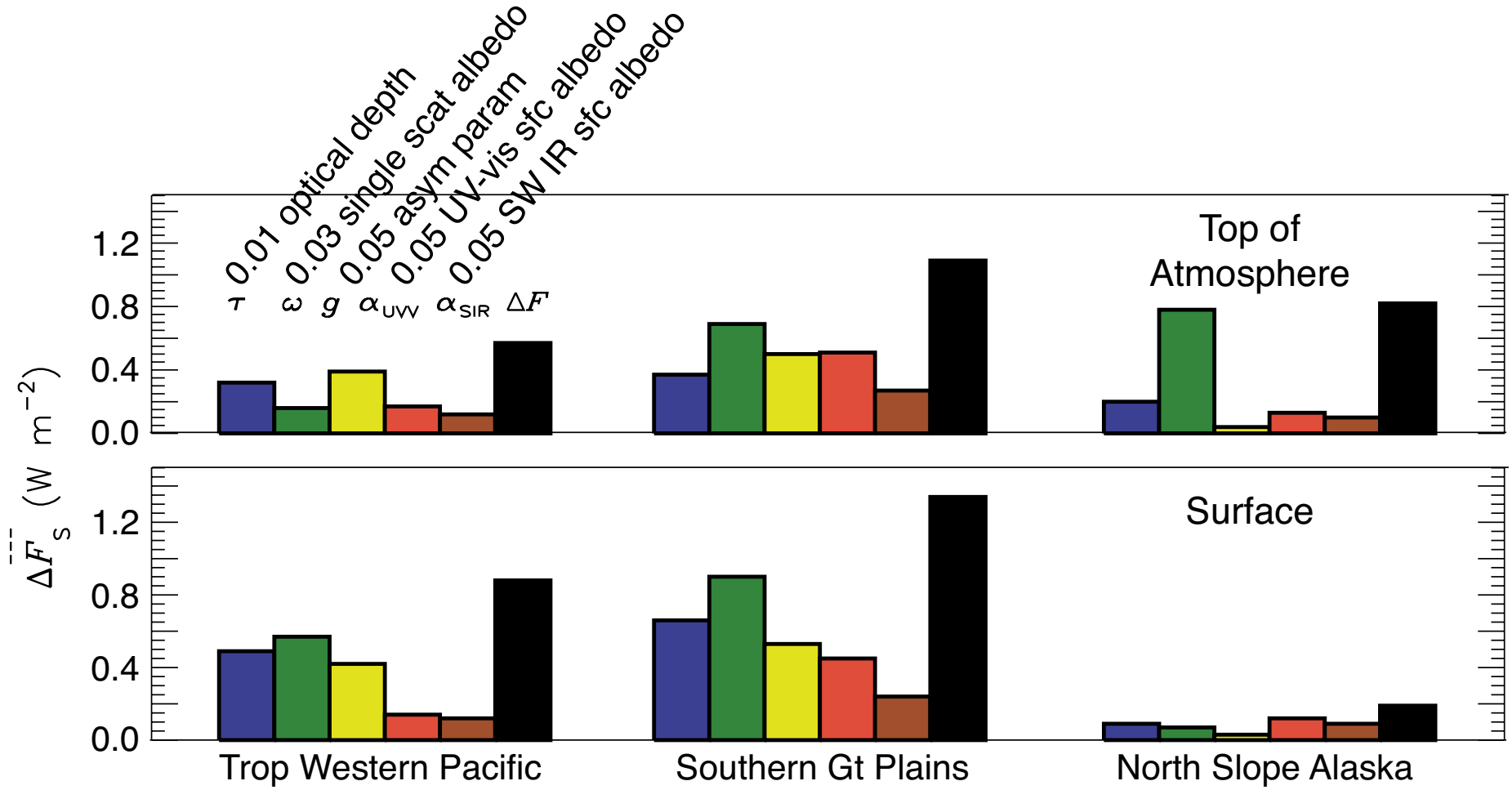
Six days of ***30-minute average composition*** in North Central Oklahoma



- 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.

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.

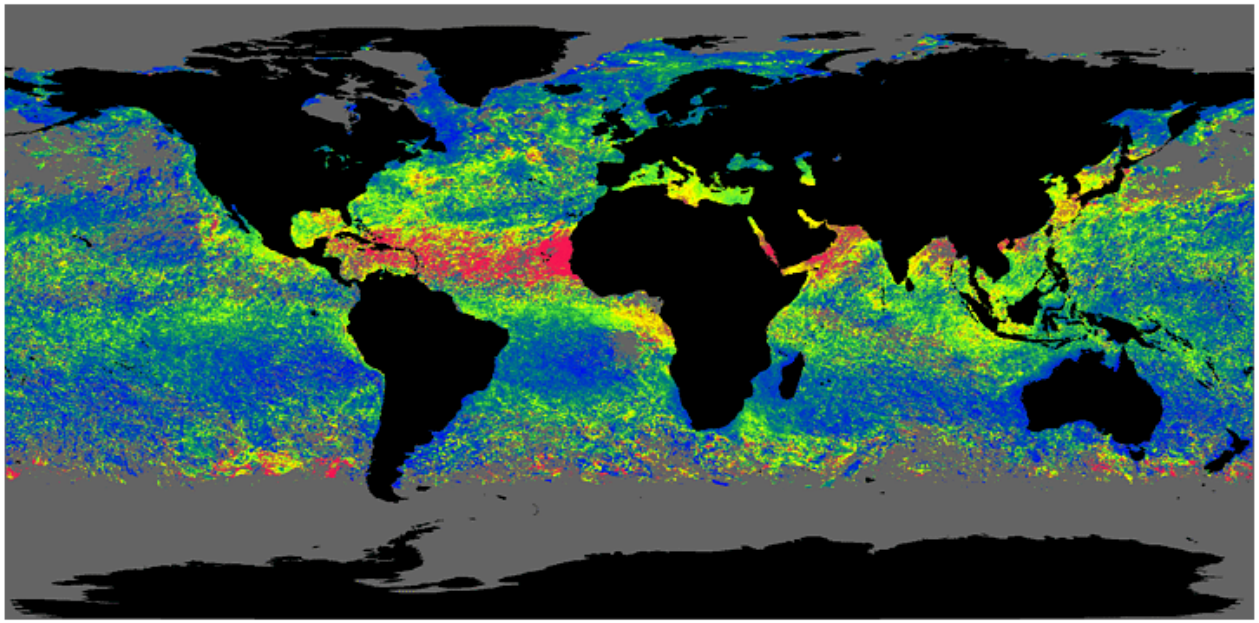
Uncertainties are substantial in context of forcings over industrial period.

MONTHLY AVERAGE AEROSOL JUNE 1997

Polder radiometer on Adeos satellite

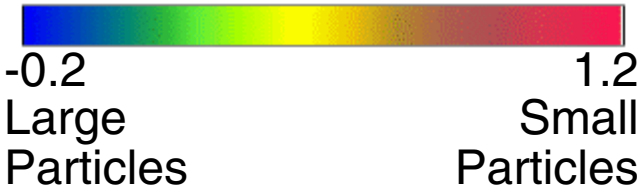
Optical Thickness τ

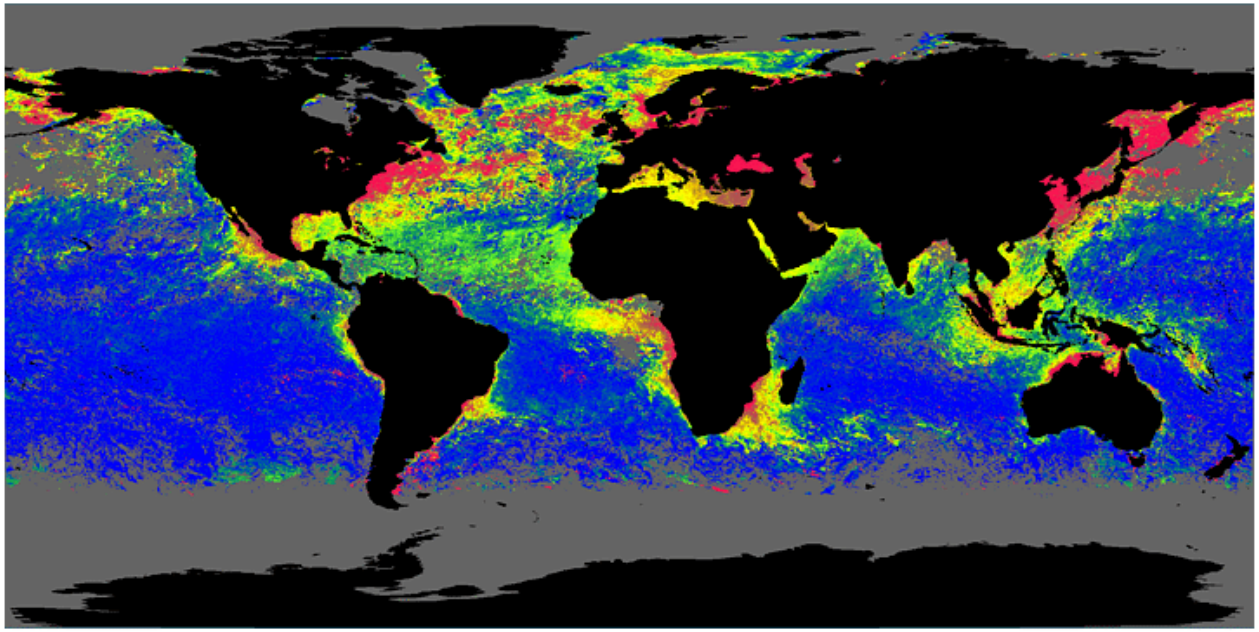
$\lambda = 865$ nm



Ångström Exponent α

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

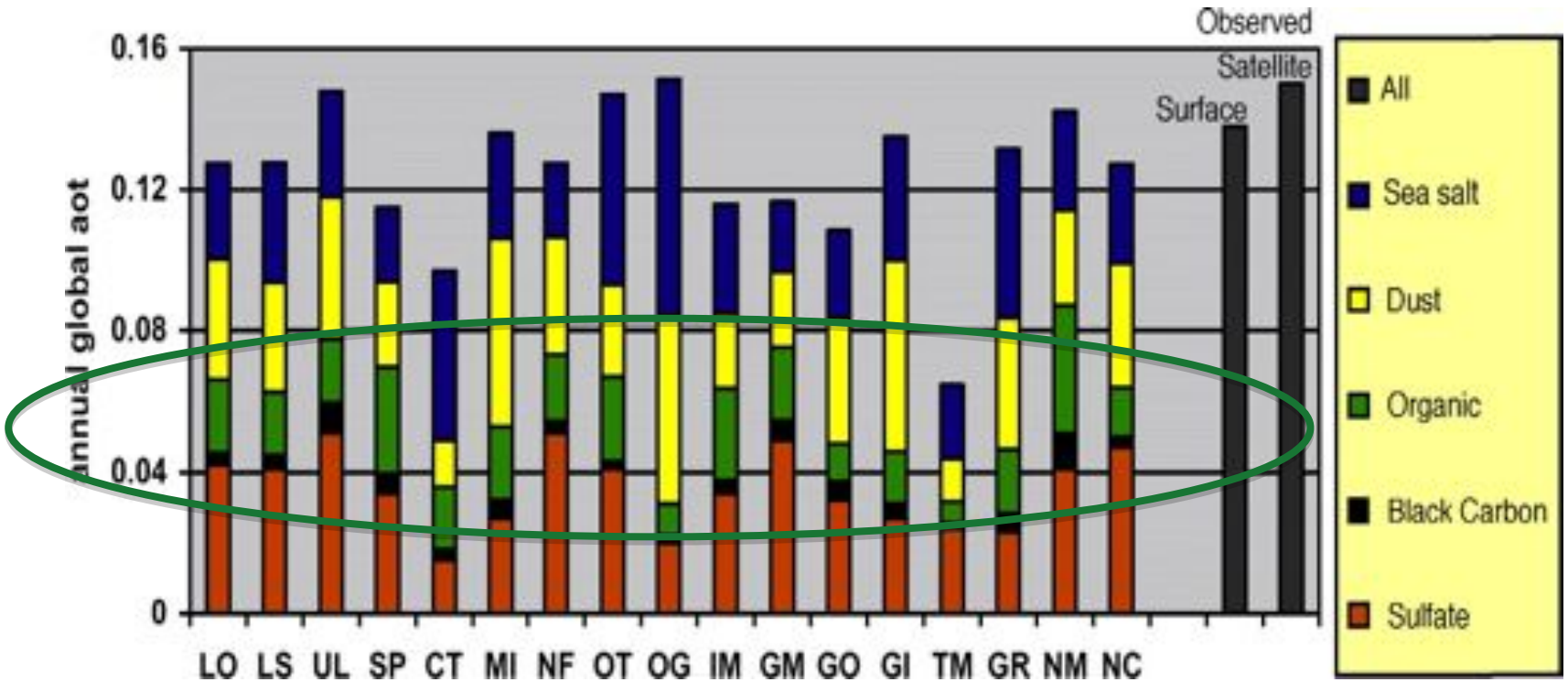




Small particles are from gas-to-particle conversion.

AEROSOL OPTICAL DEPTH IN 17 MODELS (AEROCOM)

Comparison also with surface and satellite observations



Kinne et al., ACP, 2006

Surface measurements: AERONET network.

Satellite measurements: composite from multiple instruments/platforms.

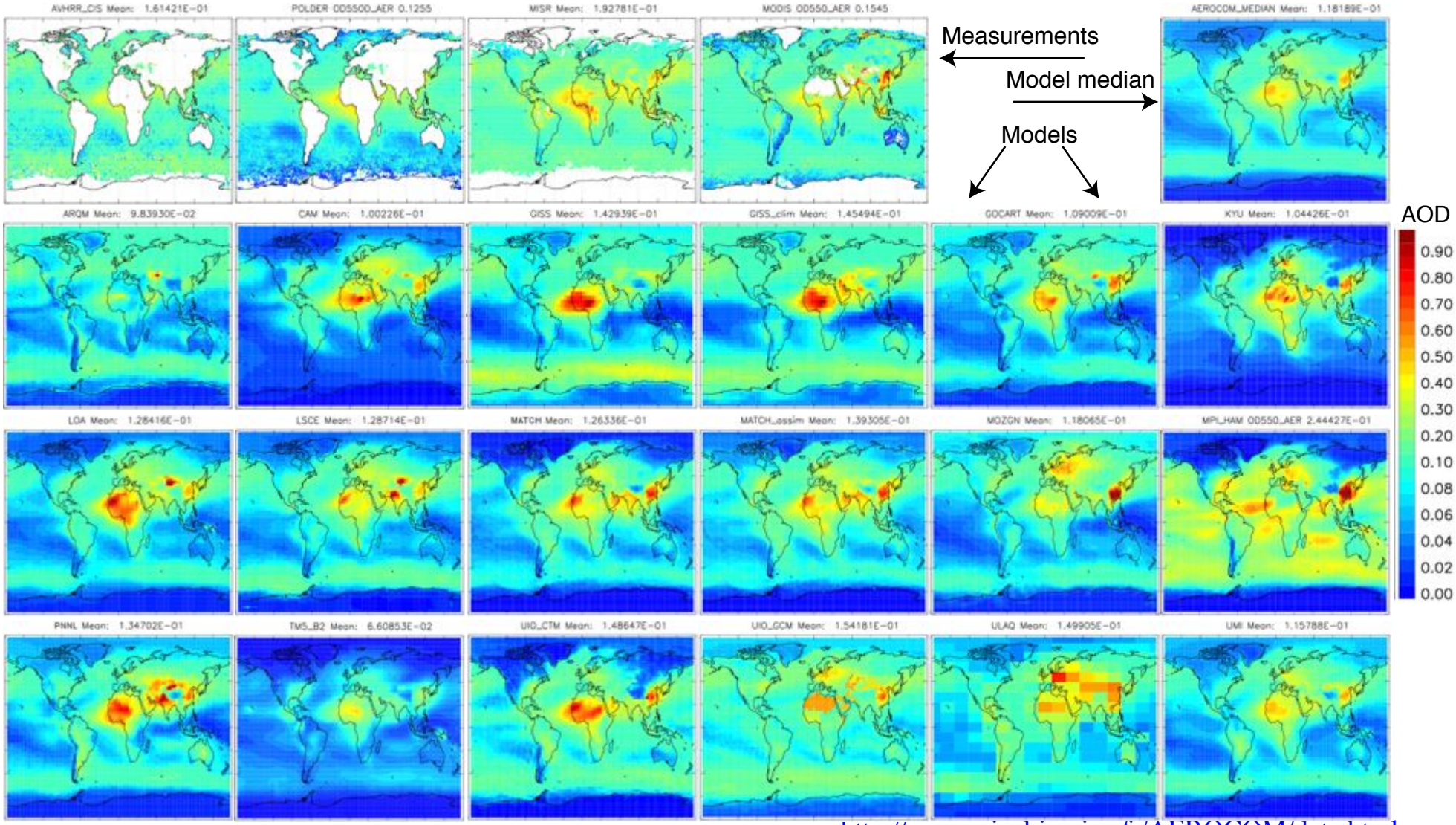
Are the models getting the "right" answer for the wrong reason?

Are the models getting the "right" answer because the answer is known?

Are the satellites getting the "right" answer because the answer is known?

23 VIEWS OF EARTH'S AEROSOL OPTICAL DEPTH

Annual mean in 4 Satellites, 18 Models and Model Median

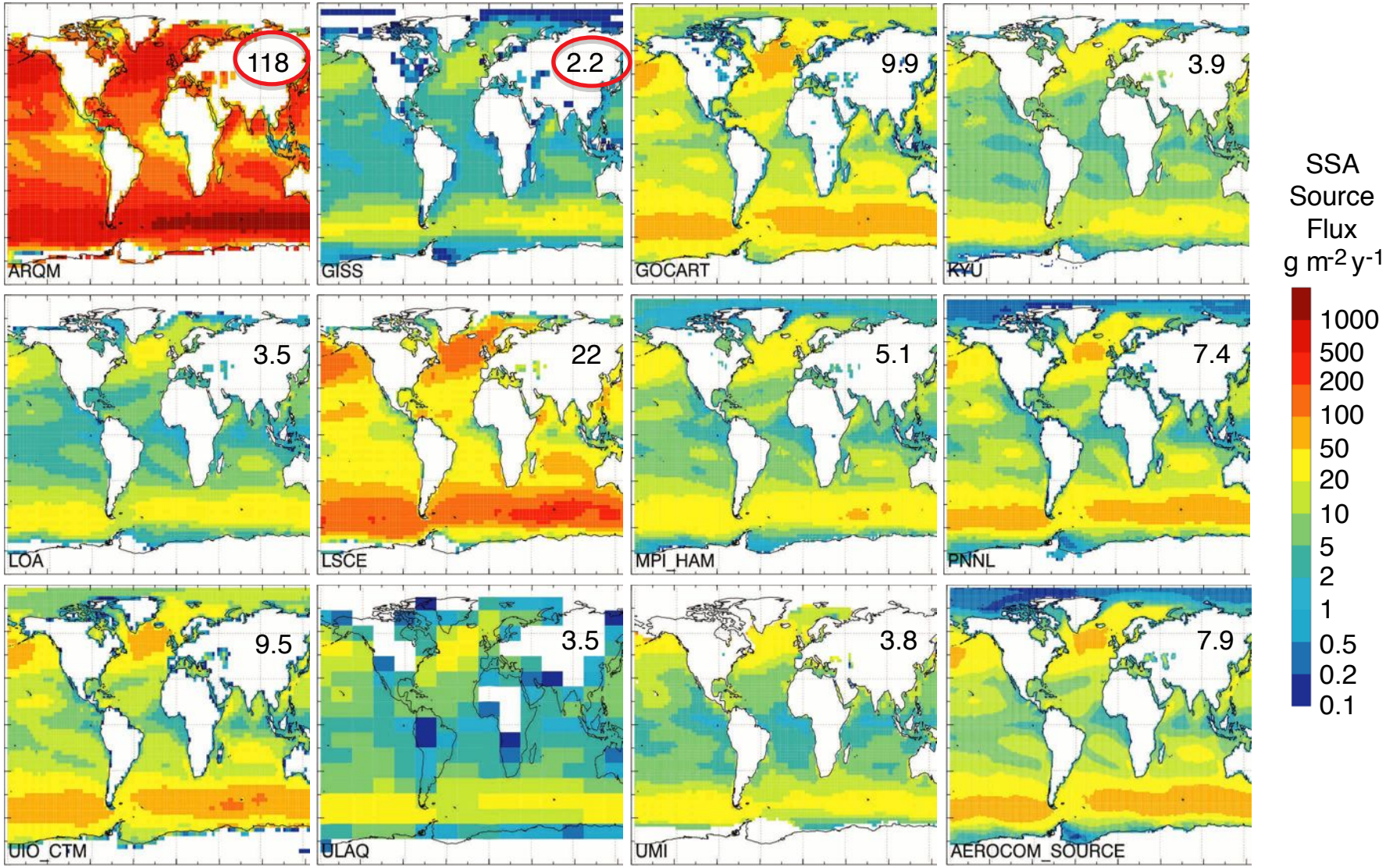


http://nansen.ipsl.jussieu.fr/AEROCOM/data.html

- Measurements: 0.12 – 0.19.
- Models: Median 0.12; range 0.07 – 0.24.

SEA SALT AEROSOL MASS EMISSIONS

Annual average in AEROCOM models; 10^{12} kg yr^{-1}



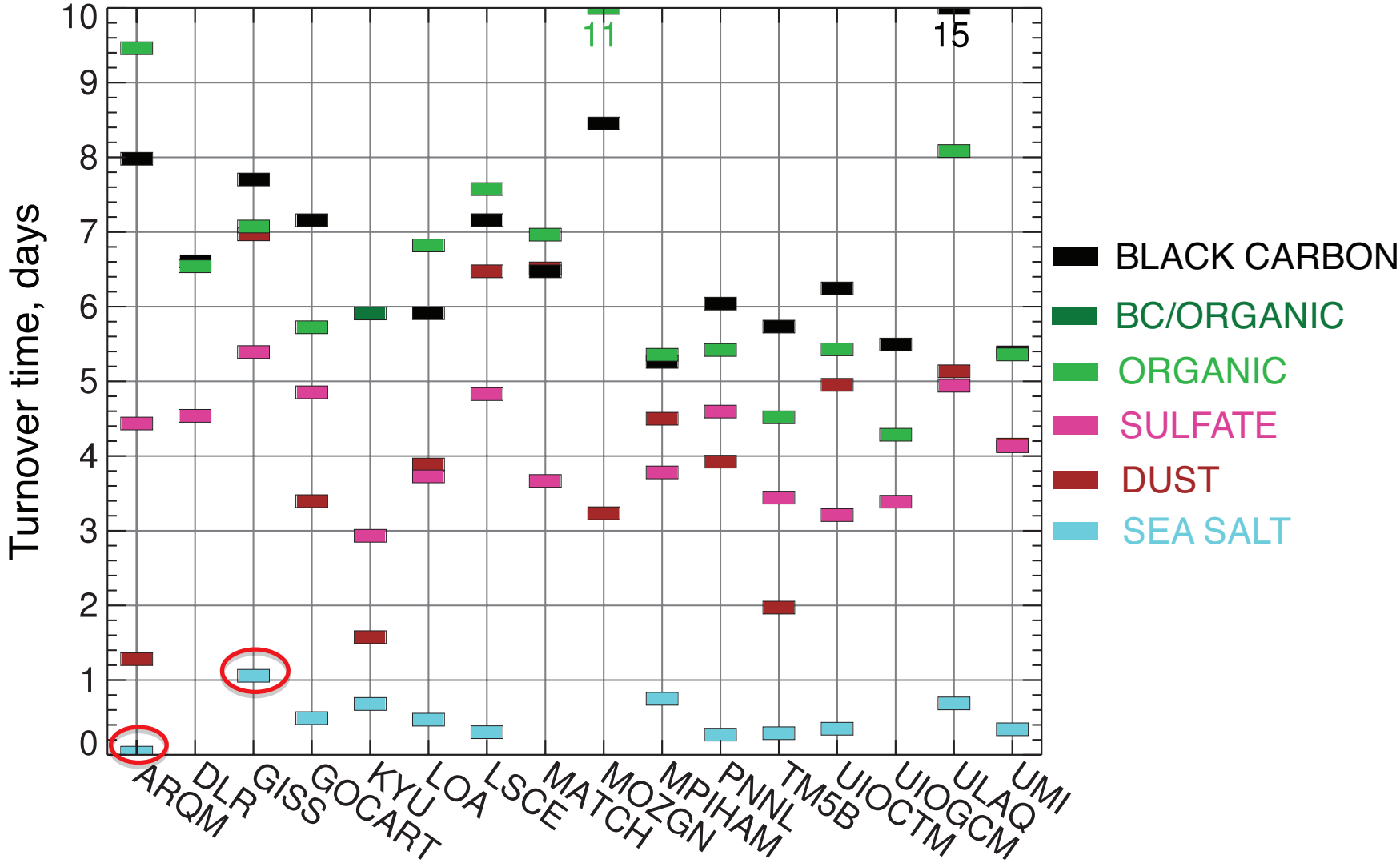
Textor et al., ACP 2006; courtesy Michael Schulz

http://dataipsl.ipsl.jussieu.fr/cgi-bin/AEROCOM/aerocom/aerocom_work_annualrs.pl

Range of global annual mean is a factor of 50.

TURNOVER TIME AS INTENSIVE VARIABLE

Five aerosol species in 16 global chemical transport models

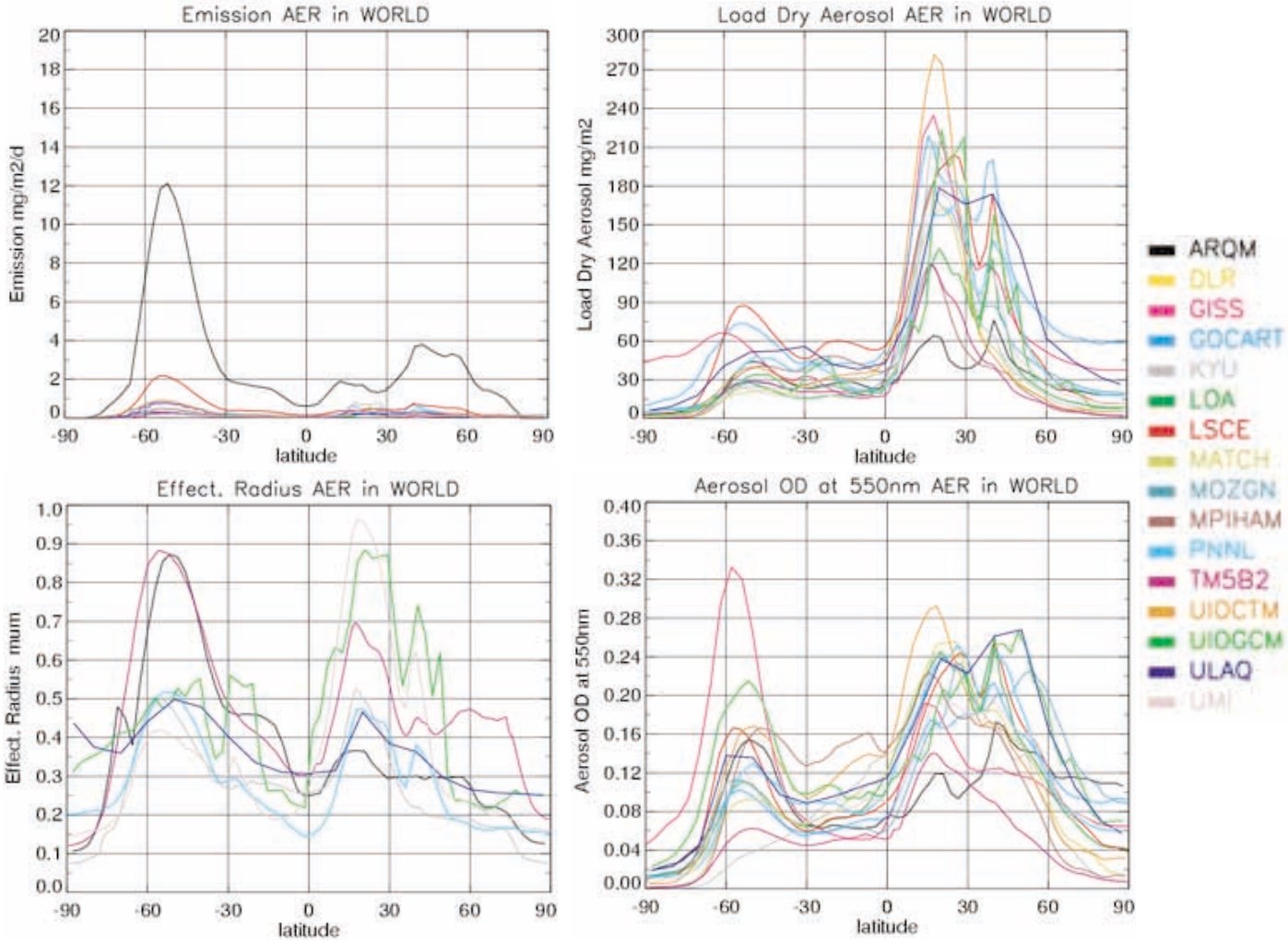


Textor et al., ACP, 2006

Characterize and compare processes in chemical transport models.
Turnover time displays ***wide model-to-model variance***.

AEROSOL PROPERTIES IN 16 MODELS

Zonal mean emissions, loading, effective radius, optical depth



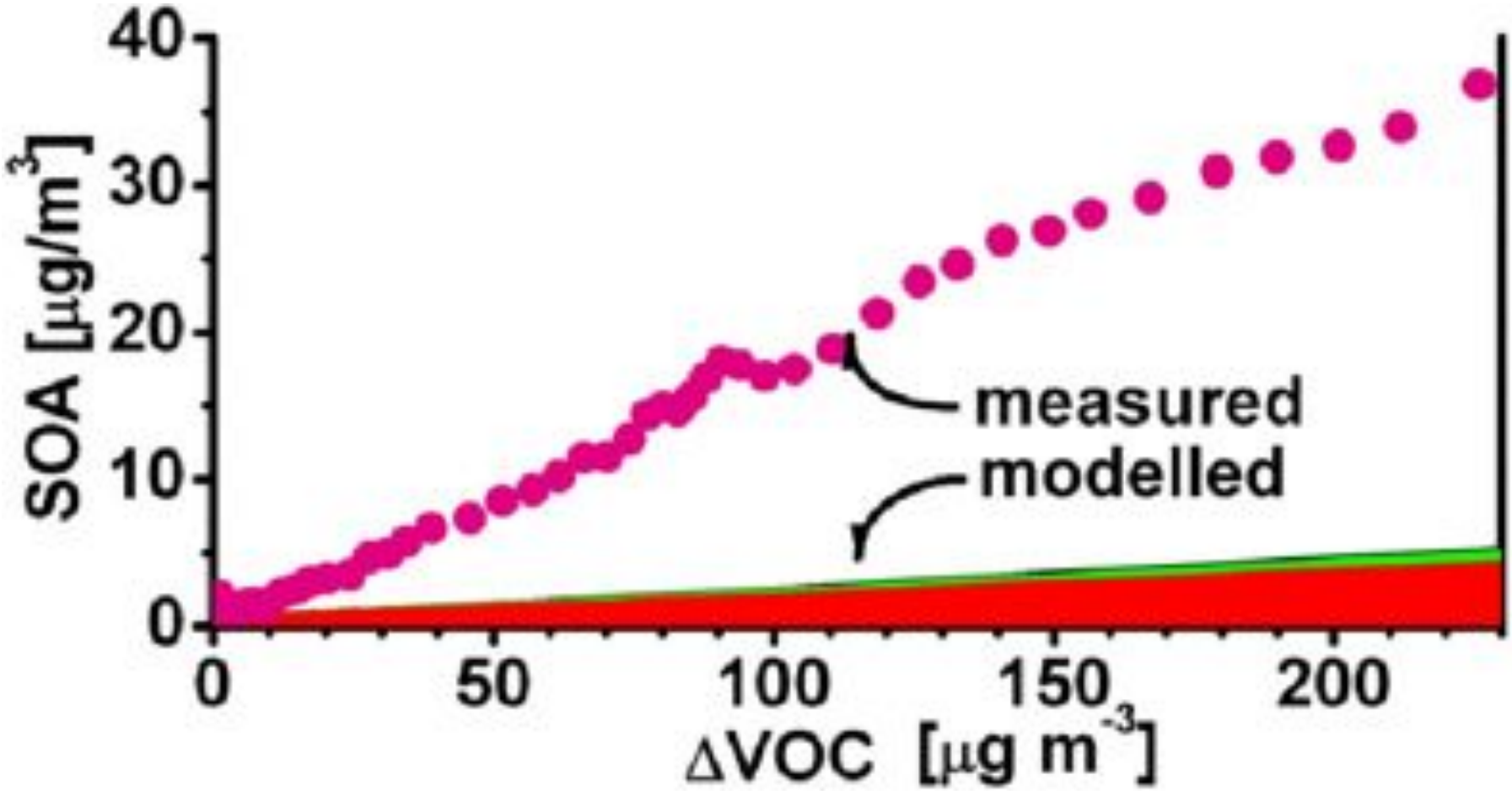
http://nansen.ipsl.jussieu.fr/AEROCOM/data.html

- Emissions and optical properties differ much more than optical depth because of compensating effects of different variables.

RECENT STUDIES EXAMINING AEROSOL PROCESSES

MEASURED ORGANIC AEROSOL GREATLY EXCEEDS MODELED

Mexico City, April 9, 2003, prior to 2 p.m.

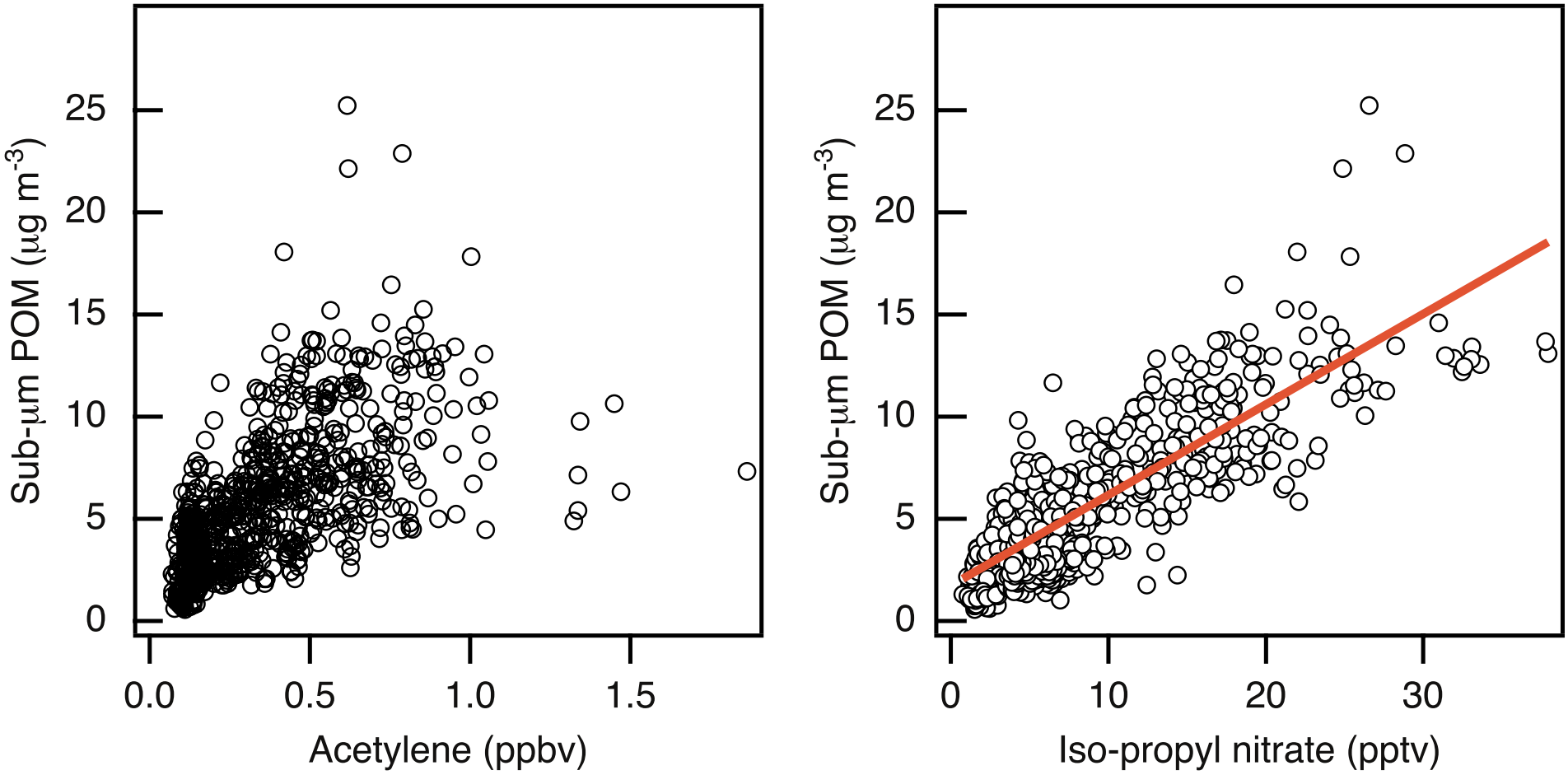


Modified from Volkamer et al., GRL, 2006

Comparison of measured oxygenated organic aerosol (OOA) and modeled secondary organic aerosol vs. decrease in volatile organic carbon.

SECONDARY AEROSOL FORMATION

Correlation of organic aerosol with acetylene (primary) and isopropyl nitrate (secondary) during New England Air Quality Study

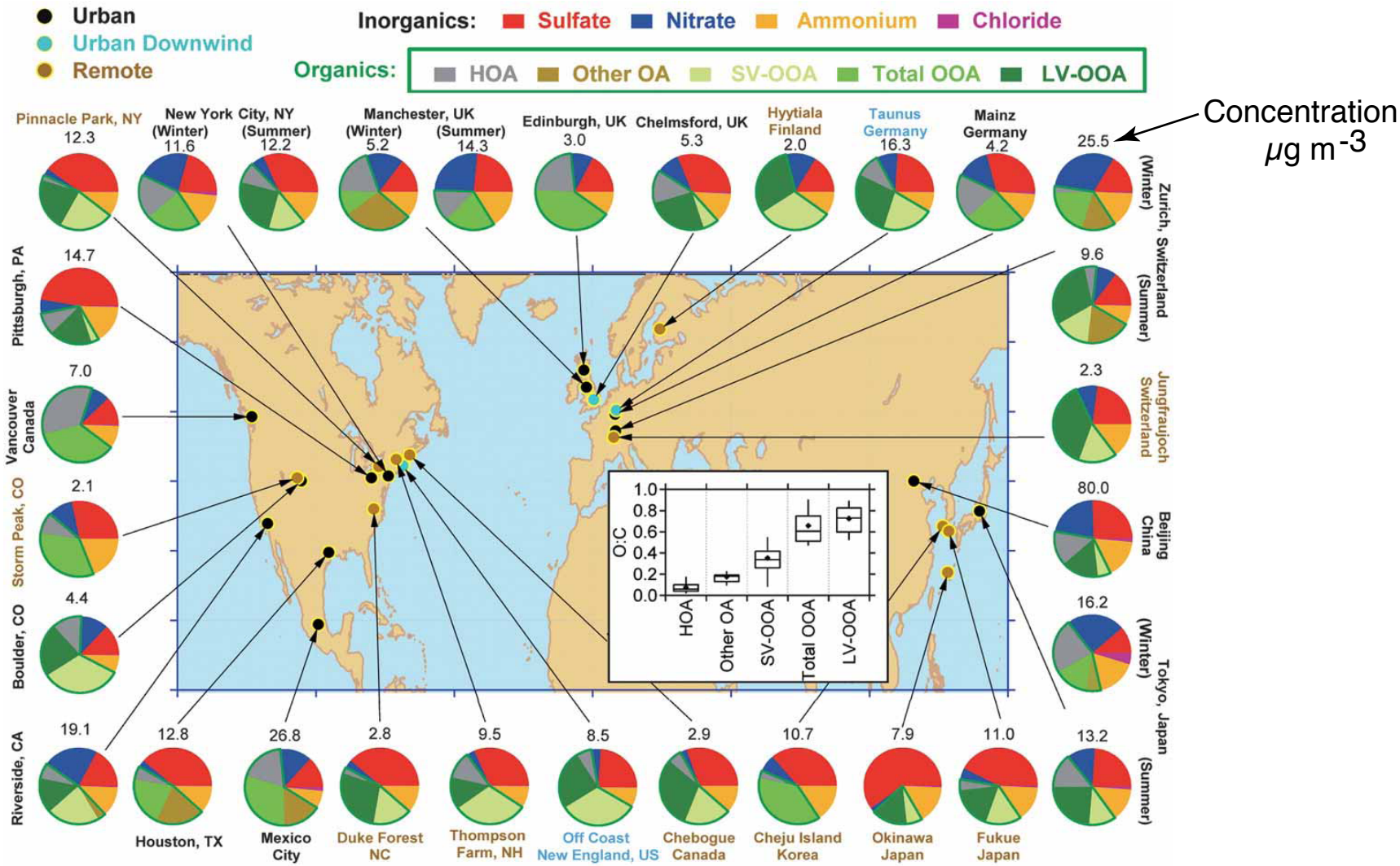


Modified from De Gouw et al., JGR, 2005

Tight correlation with isopropyl nitrate shows organic aerosol is largely secondary.

ORGANIC CONTRIBUTIONS TO TROPOSPHERIC AEROSOL

Mass-spec determination of primary vs secondary organics

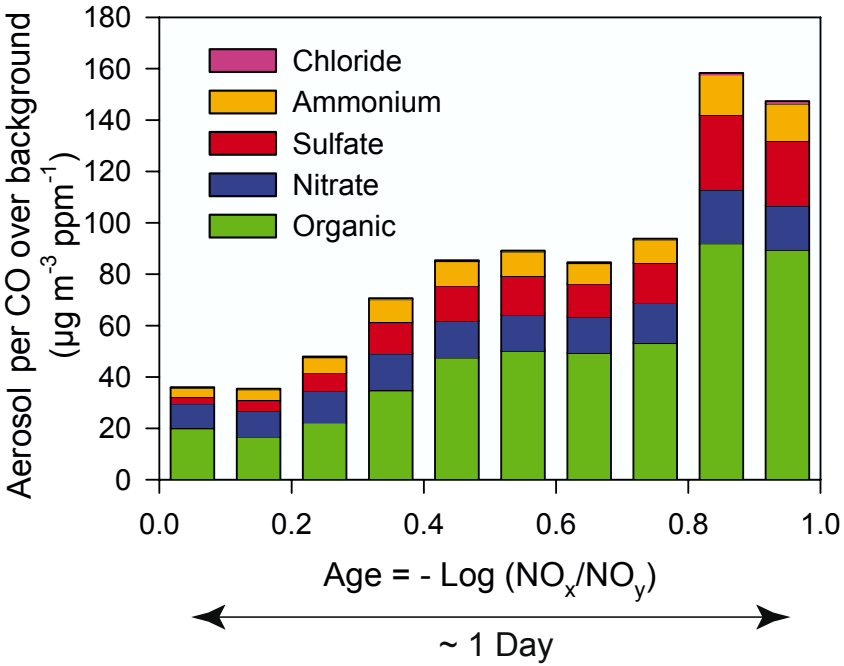


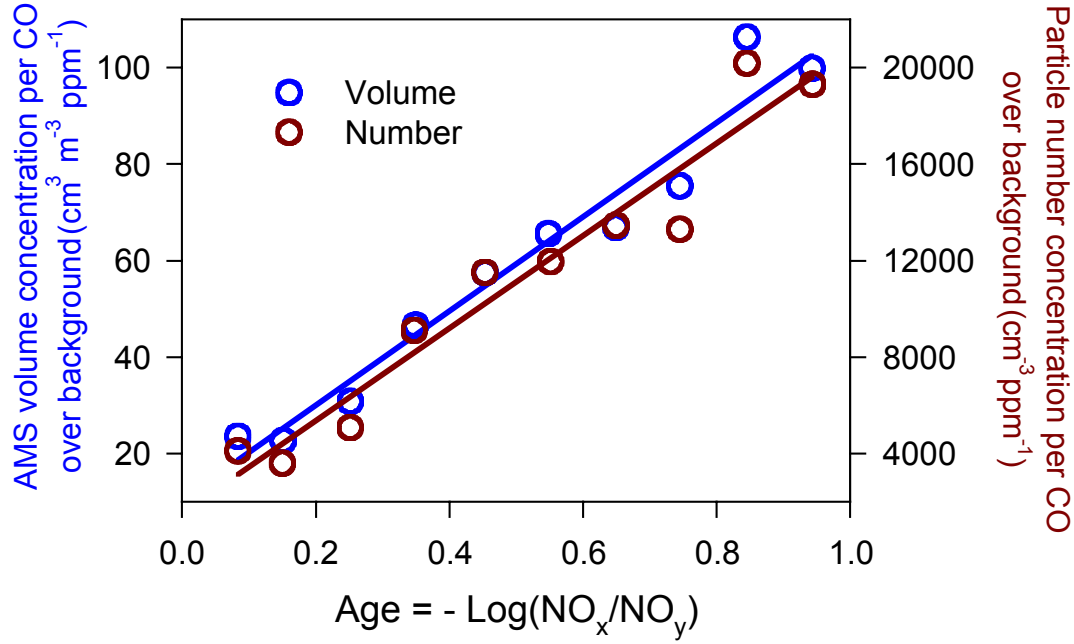
Evolution of Organic Aerosols in the Atmosphere
J. L. Jimenez, *et al. Science* **326**, 1525 (2009)

New analytical techniques permit identification of formation mechanisms.

SECONDARY AEROSOL PRODUCTION

Eight aircraft flights above and downwind of Mexico City, March 2006





Kleinman et al, ACP, 2008, 2009

Parcel photochemical age measured using $-\mathrm{Log}(NO_x/NO_y)$ as clock.

Aerosol normalized to CO above background to account for dilution.

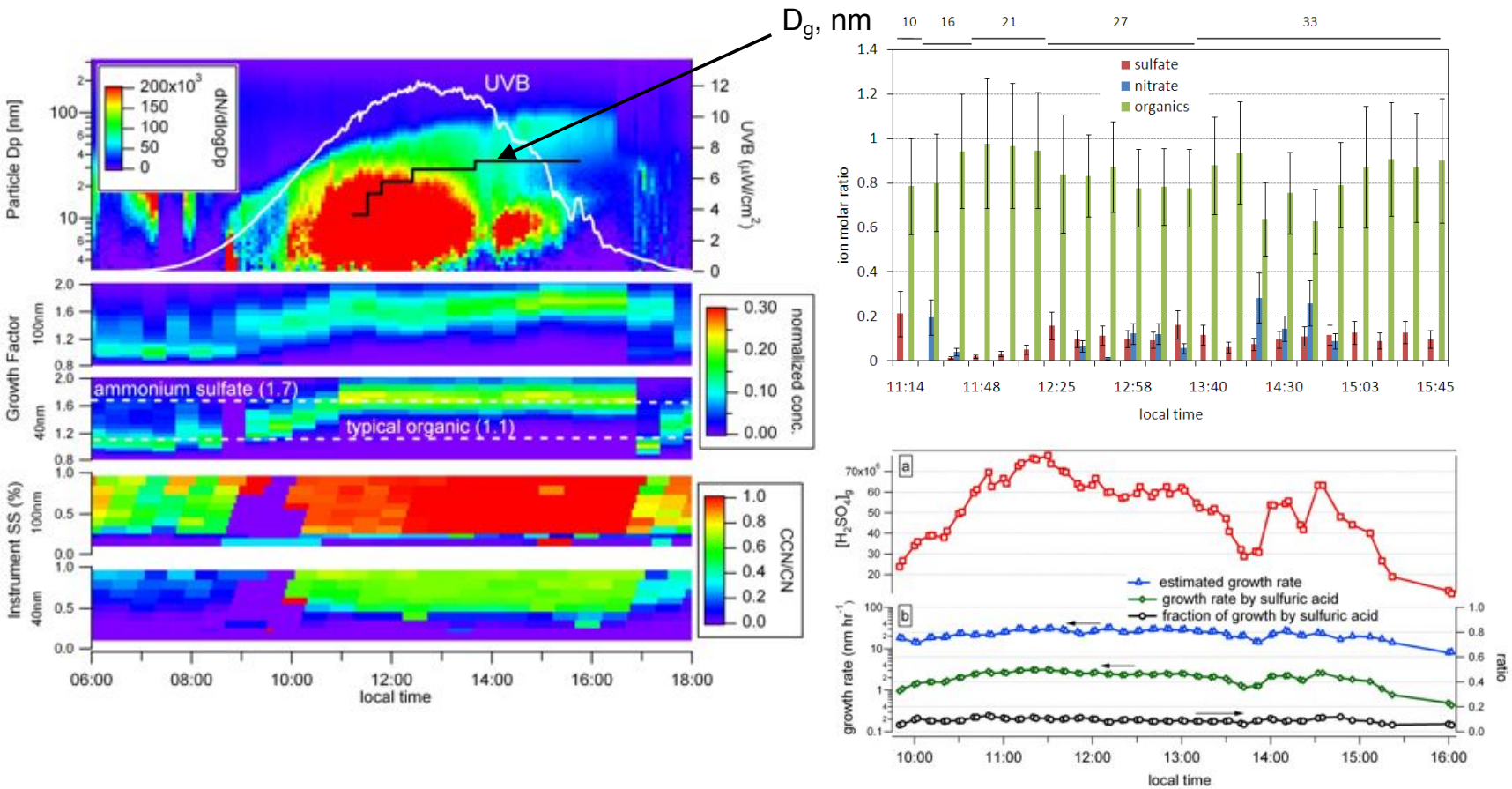
Fivefold increase in organic aerosol.

Volume and number scales are proportional, both indicating ~ 5-fold increase with age over period corresponding to ~ 1 day.

Measured increase in organic aerosol exceeds modeled based on laboratory experiments and measured volatile organic carbon ***tenfold***.

NEW PARTICLE FORMATION EVENT

Mexico City, March 16, 2006



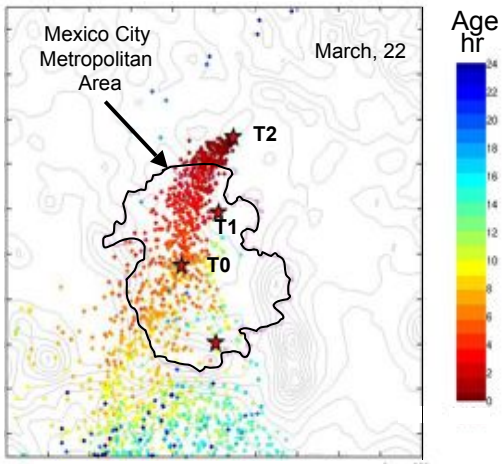
Smith, Dunn, VanReken, Iida, Stolzenburg, McMurry, & Huey, GRL, 2008

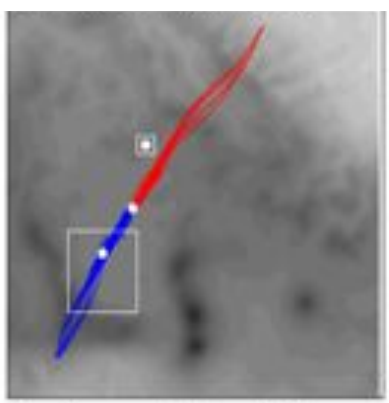
Mass spec shows composition of new particles is ***dominated by organics***.
Particle growth rate ***exceeds that by sulfuric acid*** by order of magnitude.
New particles show ***hygroscopic growth characteristic of soluble material***.
Particles grow to ***CCN active range*** (100 nm diameter) in hours.
Large fraction of 100 nm particles are CCN active at 0.5% supersaturation.

AEROSOL TRANSPORT AND EVOLUTION

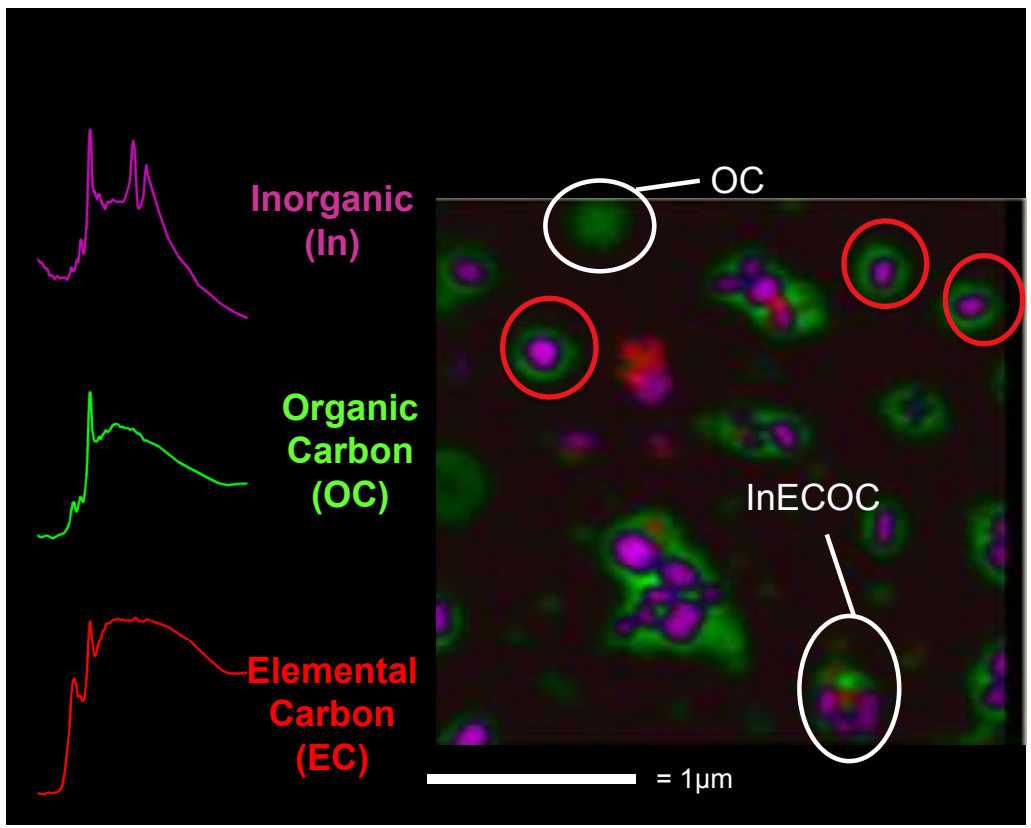
Mexico City, March 22, 2006

FLEXPART age and back trajectories

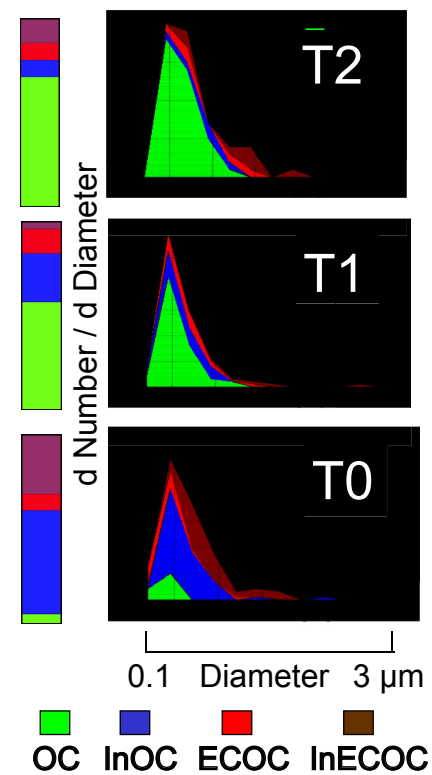




Component ID by scanning transmission electron microscopy

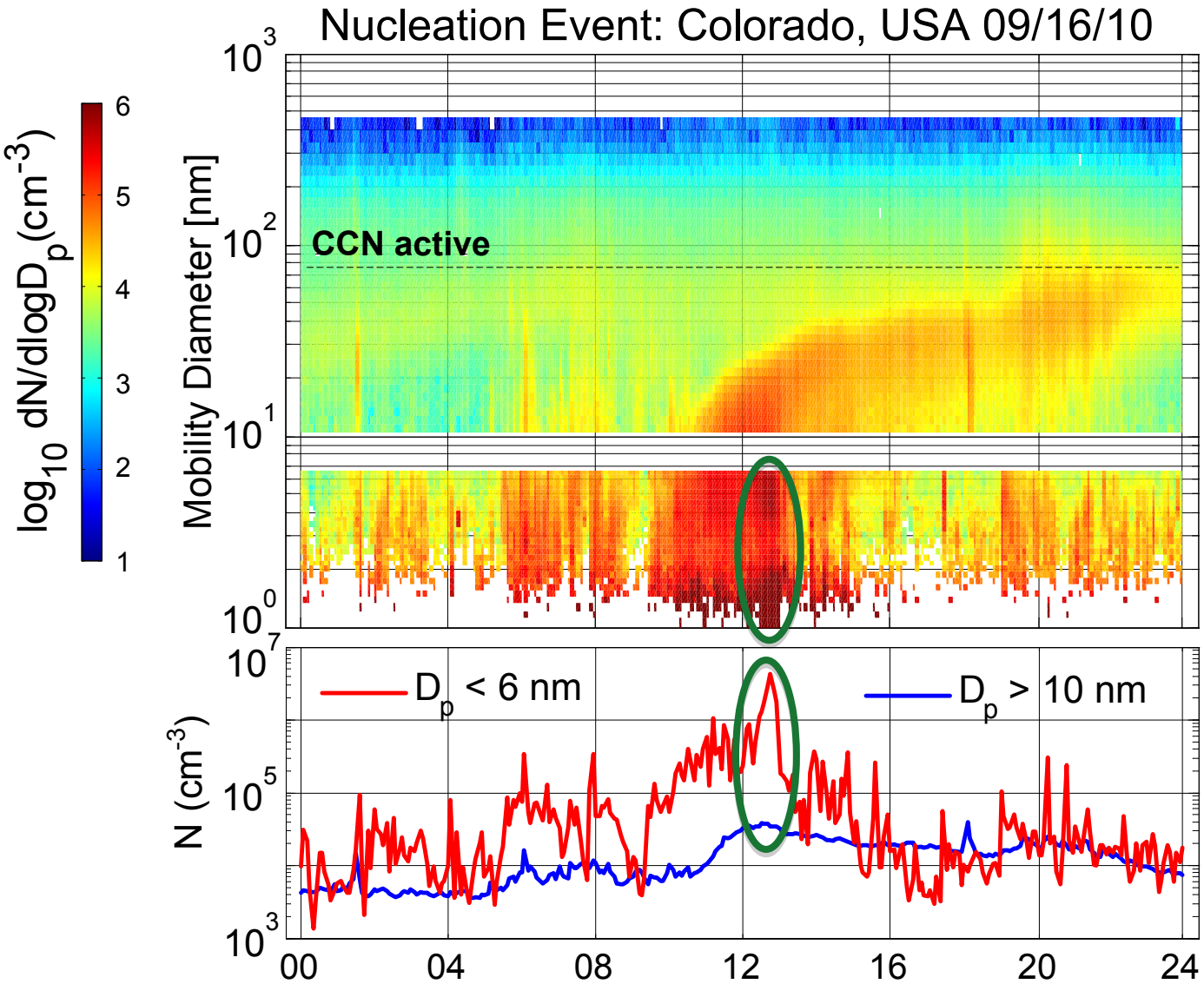


Number fraction and number size distribution



Moffet, Tivanski, Hopkins, Desyaterik, Fast, Barnard, Laskin, Gilles; PNNL, LBL

NEW MEASUREMENTS YIELD NEW INSIGHTS



conventional measurements **[GROWTH]**

new sub 6 nm measurements **[NUCLEATION]**

- **$N_{1\text{-}6\ nm}$ ~ 100X $N_{10\text{-}500\ nm}$**
- Highly dynamic ➔ need size/time dep. meas.

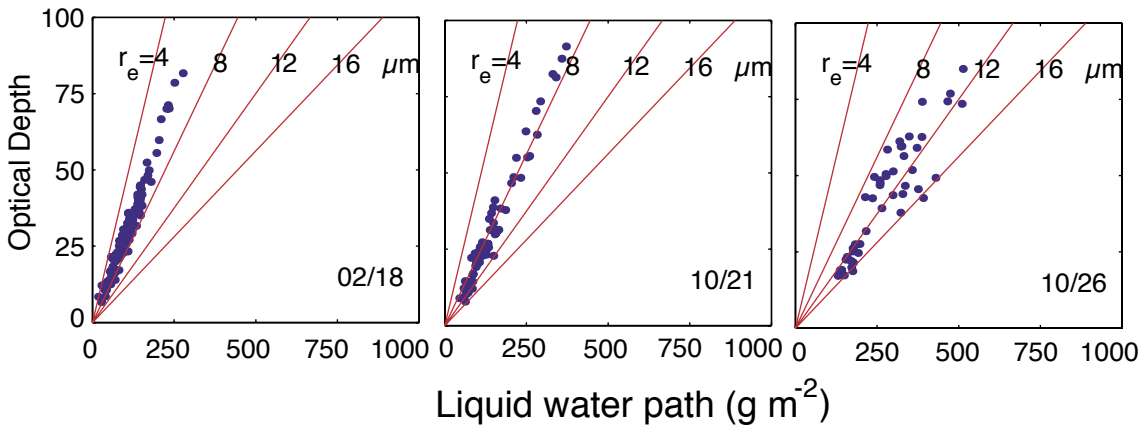
Chongai Kuang, BNL

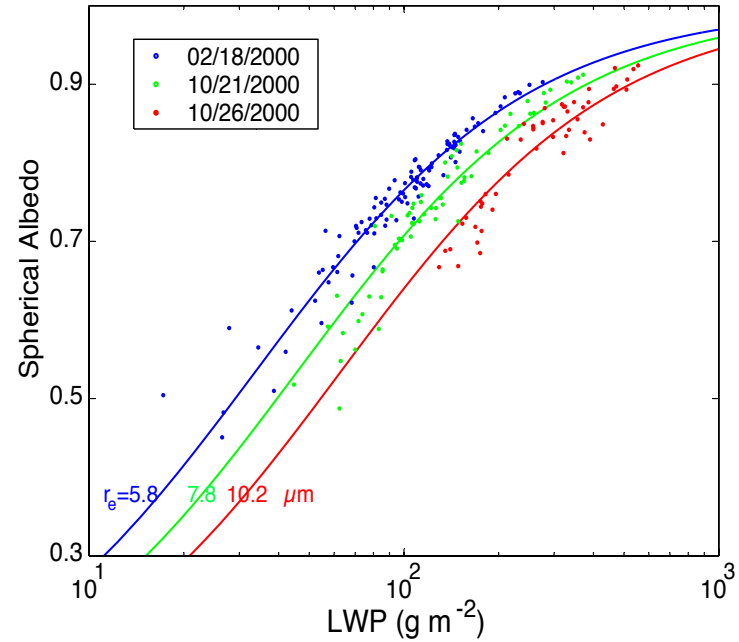
RECENT STUDIES EXAMINING AEROSOL INDIRECT EFFECTS

CLOUD ALBEDO AND FORCING CALCULATED FROM MEASURED EFFECTIVE RADIUS AND LIQUID WATER PATH

North Central Oklahoma

Effective radius determined from slope of Optical depth vs. Liquid water path





Cloud albedo is calculated for observed data and for average effective radius for each day. Forcing is calculated for indicated conditions relative to October 26.

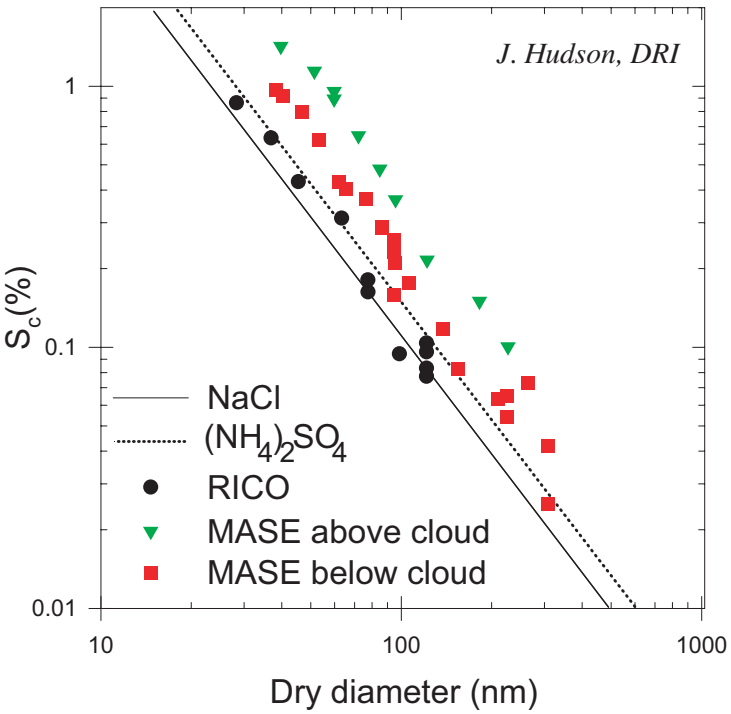
Radiative forcing for solar zenith angle 60° and liquid water path 100 g m^{-2}

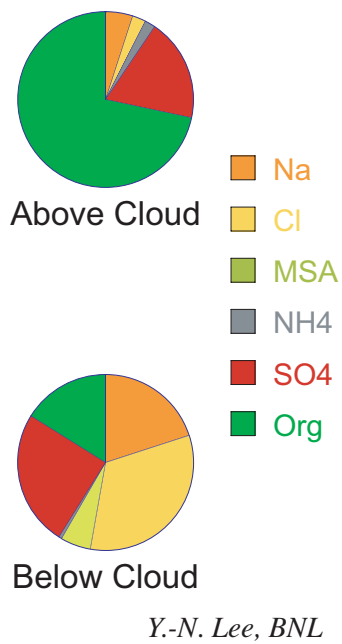
Date, 2000	Effective radius r_e, μm	Optical Depth	Net flux at TOA W m^{-2}	Forcing relative to 10/26, W m^{-2}
10/26	10.2	15.1	293	—
10/21	7.8	20.8	266	27
02/18	5.8	28.3	240	53

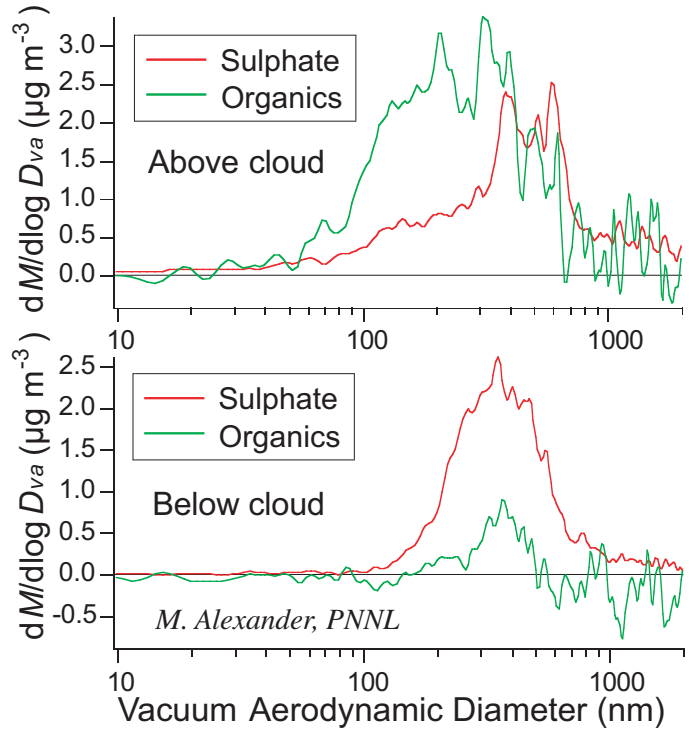
Kim, Schwartz, Miller, and Min, JGR, 2003

CRITICAL SUPERSATURATION

Dependence on particle size and composition







Particles above cloud layer showed greater increase in supersaturation than particles below cloud.

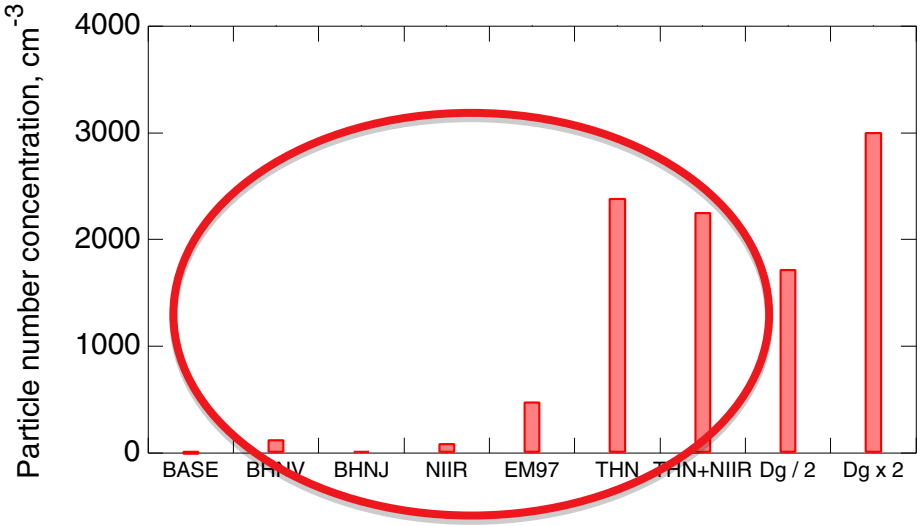
Composition measured with PILS (particle into liquid sampler) showed ***high organic fraction*** in above cloud aerosol.

Measurements with aerosol mass spectrometer showed organic material in CCN size range.

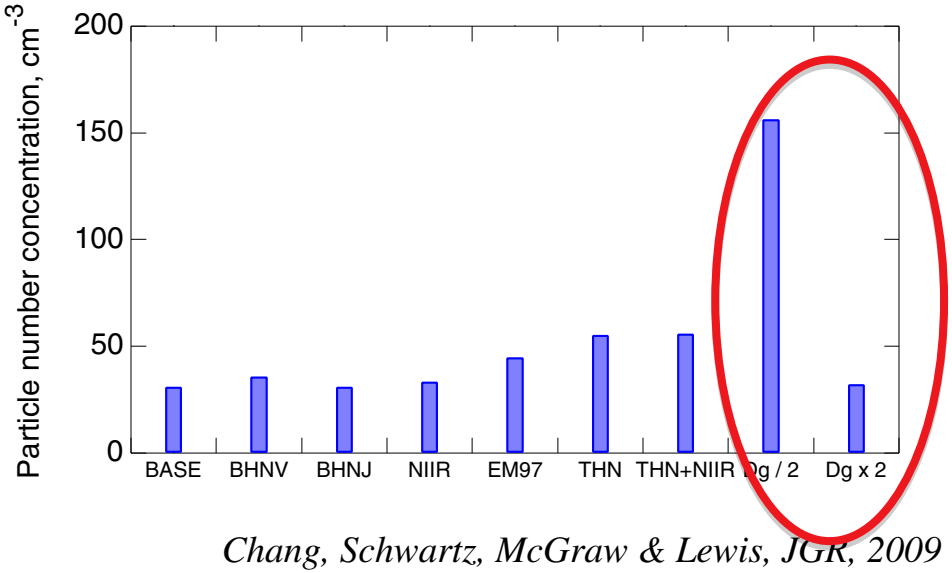
AEROSOL PARTICLE NUMBER CONCENTRATION

Average particle number concentrations North America, July 2004

Aitken mode particles ($D \leq 100$ nm)



Accumulation mode particles ($D \geq 100$ nm)



Chang, Schwartz, McGraw & Lewis, JGR, 2009

Strong dependence on new particle formation mechanism

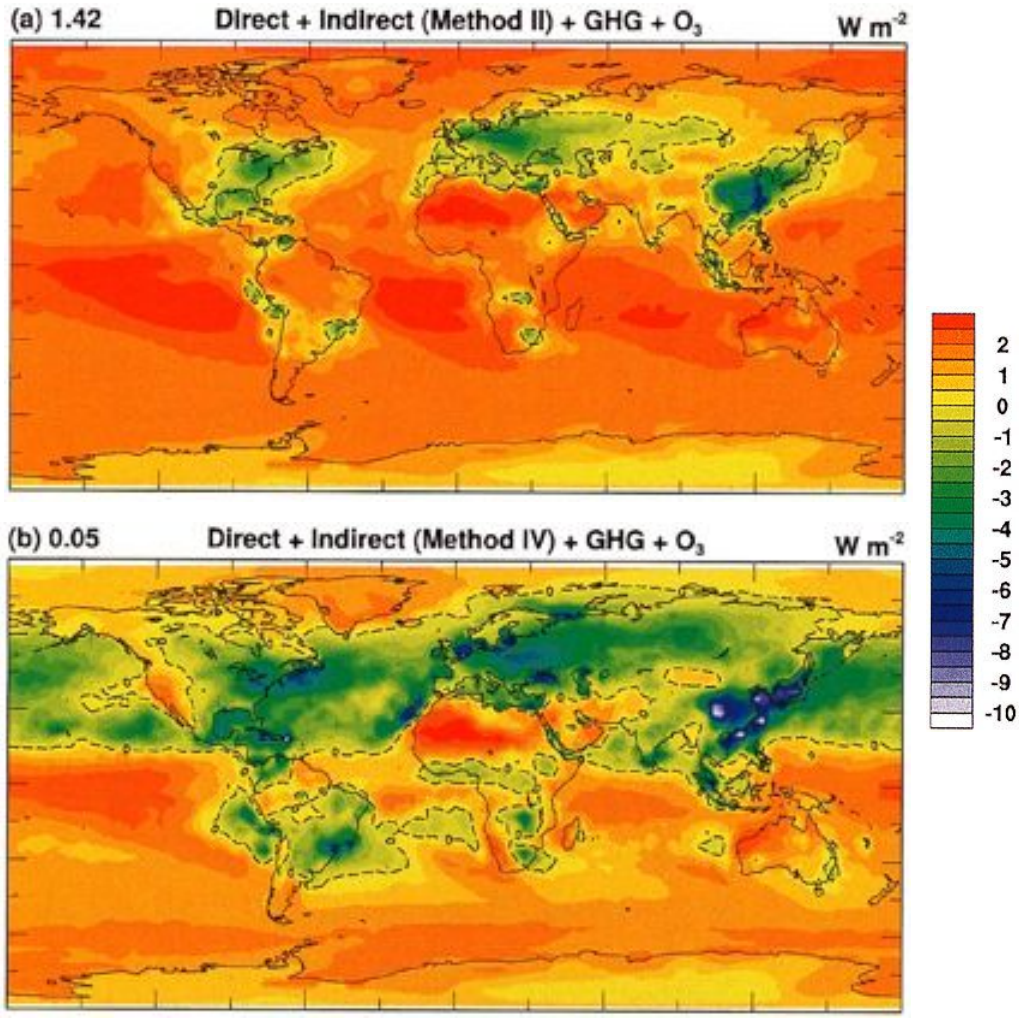
Strong dependence on size of primary emissions

Accurate representation of number concentrations and aerosol indirect effects requires improved knowledge of ***new particle formation rate*** and ***size distributed emissions***.

TOTAL FORCING, ANNUAL AVERAGE

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

Two Formulations of Cloud Droplet Concentration



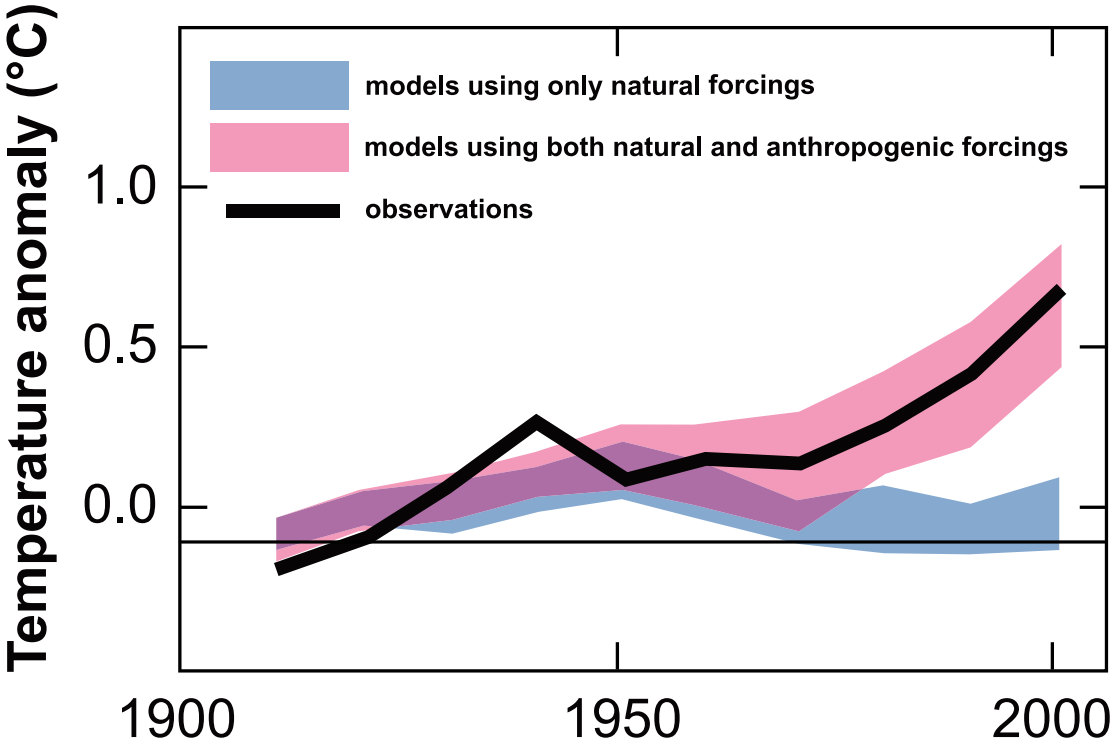
Kiehl et al., JGR, 2000

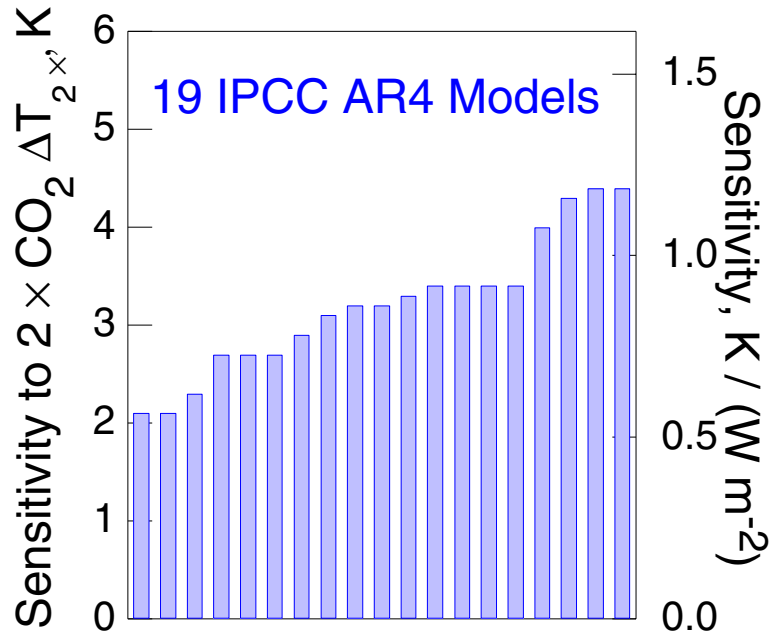
Indirect forcing is highly sensitive to the assumed relation between sulfate concentration and cloud droplet number concentration.

IMPLICATIONS

TOO ROSY A PICTURE?

Ensemble of 58 model runs with 14 global climate models





- Simulations that incorporate anthropogenic forcings, including increasing greenhouse gas concentrations and the effects of aerosols, and that also incorporate natural external forcings provide a ***consistent explanation of the observed temperature record***.
- These simulations used models with ***different climate sensitivities, rates of ocean heat uptake and magnitudes and types of forcings***.

IPCC AR4, 2007

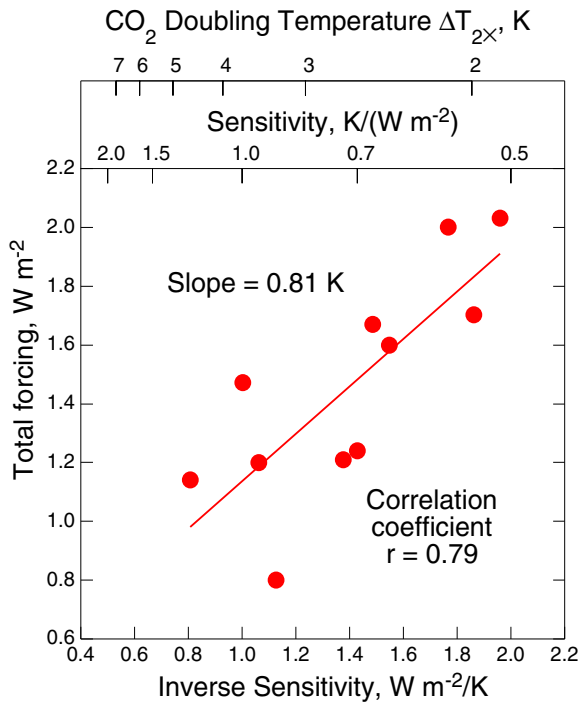
How can this be?

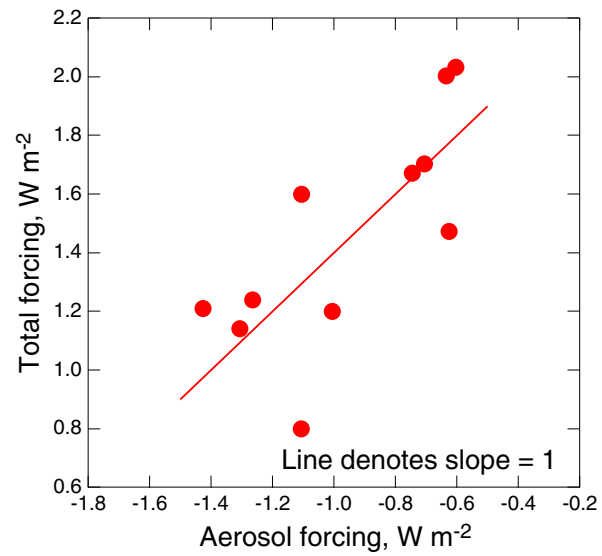
CORRELATION OF AEROSOL FORCING, TOTAL FORCING, AND SENSITIVITY IN CLIMATE MODELS

Nine coupled ocean-atmosphere models; two energy balance models

$$S = \Delta T / F$$

$$F = \Delta T \, S^{-1}$$





Modified from Kiehl, GRL, 2007

Total forcing is linearly correlated with inverse sensitivities of the models.

Climate models with lower sensitivity (higher inverse sensitivity) employed a greater total forcing.

Slope (0.8 K) is approximately equal to observed temperature change. Models accurately reproduce known temperature change.

Greater total forcing is due to smaller (less negative) aerosol forcing.

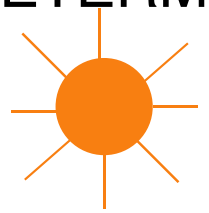
THE PATH FORWARD

Determine aerosol forcing with high accuracy.

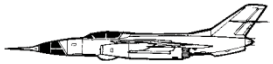
Multiple approaches are required:

- ***Laboratory studies*** of aerosol processes.
- ***Field measurements*** of aerosol processes and properties: emissions, new particle formation, evolution, size distributed composition, optical properties, CCN properties, removal processes . . .
- Represent aerosol processes in ***chemical transport models***.
- Evaluate models by ***comparison with observations***.
- ***Satellite measurements*** for spatial coverage.
- Calculate forcings in ***chemical transport models and GCMs***.
- ***Measurement based determination of aerosol forcings***.

DIRECT DETERMINATION OF AEROSOL FORCINGS AT ARM SITES



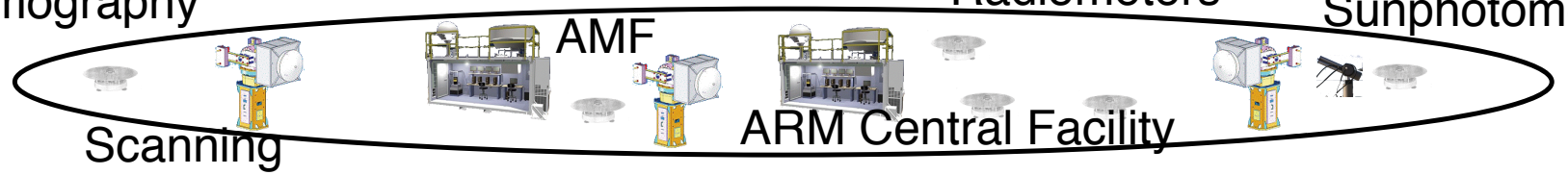
Measurements 24-7-365



Drone
Net SW and LW at TOA

3-D Characterization of Aerosol and Cloud Properties

Characterization of 3-D Cloud Properties by Radars, Tomography



~50 km

TAKE HOME MESSAGES

Aerosol forcing is substantial in the context of forcing over the industrial period.

This forcing is quite uncertain in that context.

This uncertainty has major implications on the interpretation of climate change over the industrial period and projected future climate change.

Many aerosol processes are not well understood. We are still in discovery stage.

The modeling is way ahead of the understanding.

Measurement techniques have greatly improved in the past few years, leading to important insights and capabilities.

This situation calls for greatly enhanced effort in quantifying aerosol influences on radiation and cloud & precipitation processes.