ATMOSPHERIC SCIENCE

Atmospheric Sciences Division

MEETS NANOSCIENCE

Center for Functional Nanomaterials

THE ROLE OF AEROSOLS IN CLIMATE CHANGE

Stephen E. Schwartz
Atmospheric Sciences Division, BNL
http://www.ecd.bnl.gov/steve

BNL Center for Functional Nanomaterials Department Retreat
Oldfield NY, July 9, 2008

OVERVIEW

What is an aerosol?

Earth's climate and climate change

Importance of aerosols to climate and climate change

Key processes of atmospheric aerosols

Some recent research findings

Important directions for laboratory studies

Concluding remarks

ATMOSPHERIC AEROSOLS

Aerosol: Colloidal suspension of solid or liquid particles in air

Characterized by size, composition, morphology, sources . . .

- Radius 1 nm to 10 μm
- Twelve orders of magnitude in volume

Sources:

- Natural: Dust, sea salt, haze (Great Smokies, Blue Ridge Mtns.)
- Anthropogenic: Smoke, smog

Importance

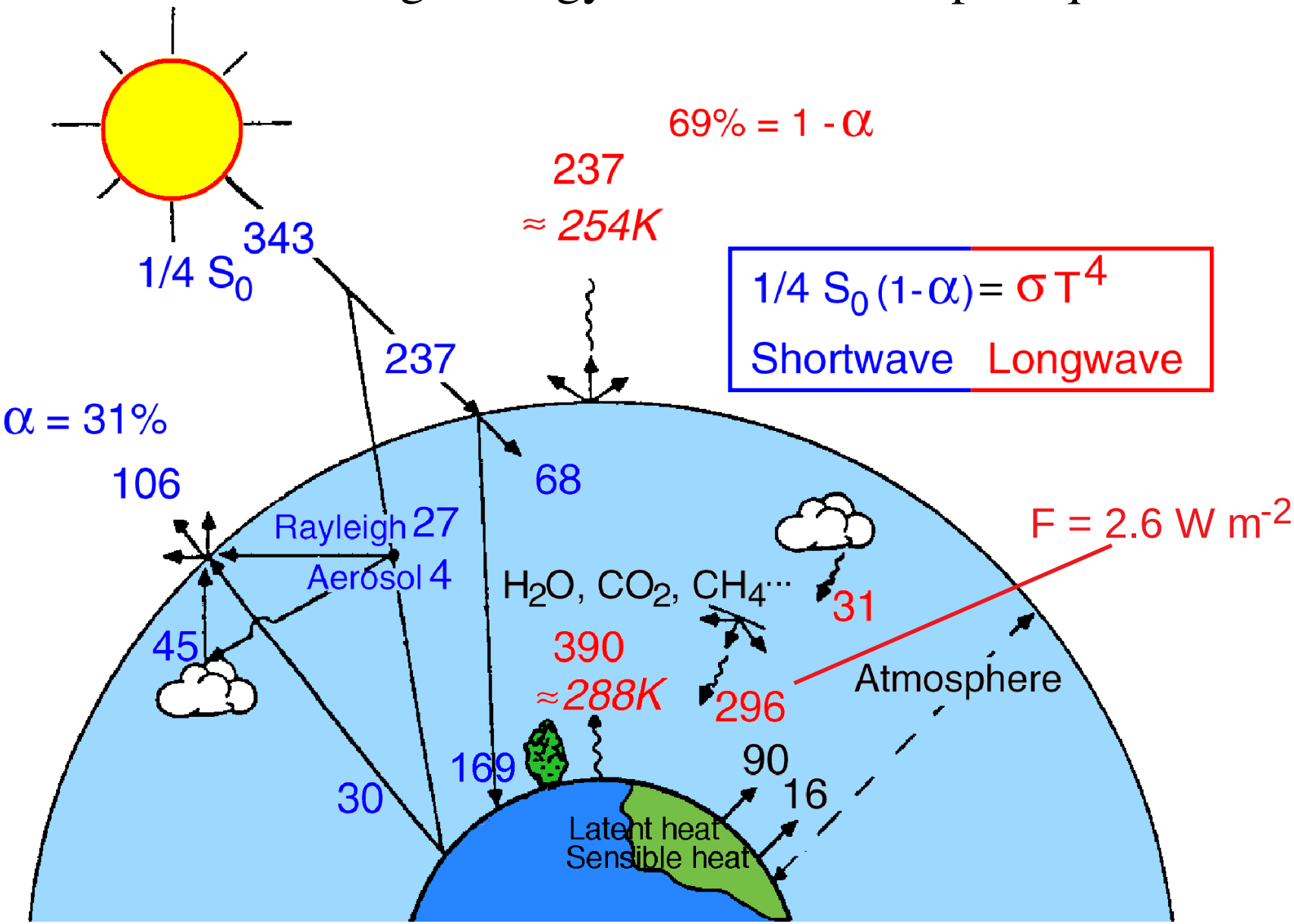
- Clouds – Aerosol particles as cloud condensation nuclei, ice nuclei
- Human health: inhalation
- Acid deposition
- Visibility impairment: light scattering
- Influence on climate and climate change

AEROSOLS – A SIGN OF PROSPERITY

The Lackawanna Valley, George Inness, ca. 1850

GLOBAL ENERGY BALANCE

Global and annual average energy fluxes in watts per square meter



Schwartz, 1996, modified from Ramanathan, 1987

RADIATIVE FORCING

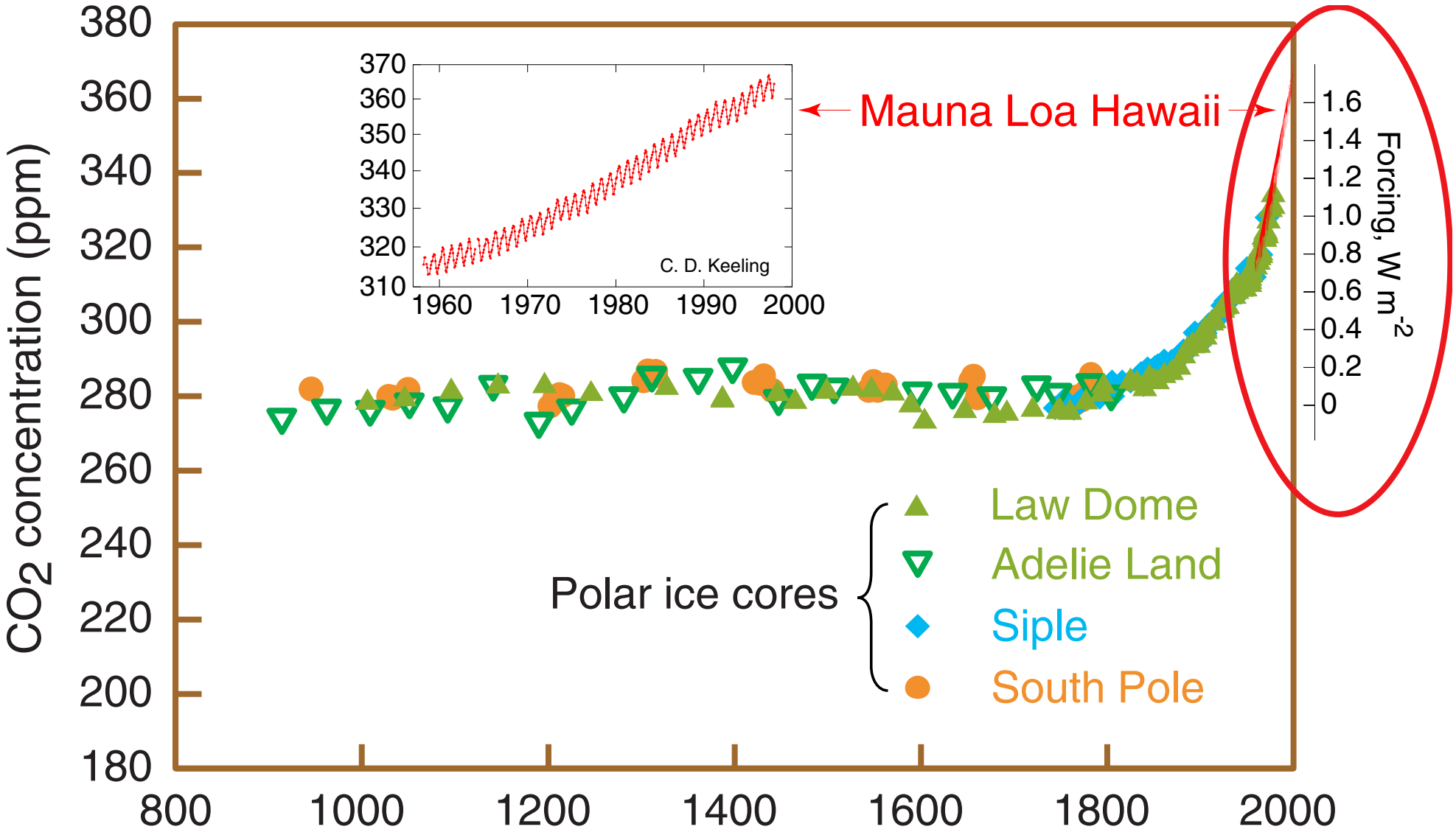
A ***change*** in a radiative flux term in Earth's radiation budget, ΔF, W m^{-2}.

Working hypothesis:

On a global basis radiative forcings are additive and fungible.

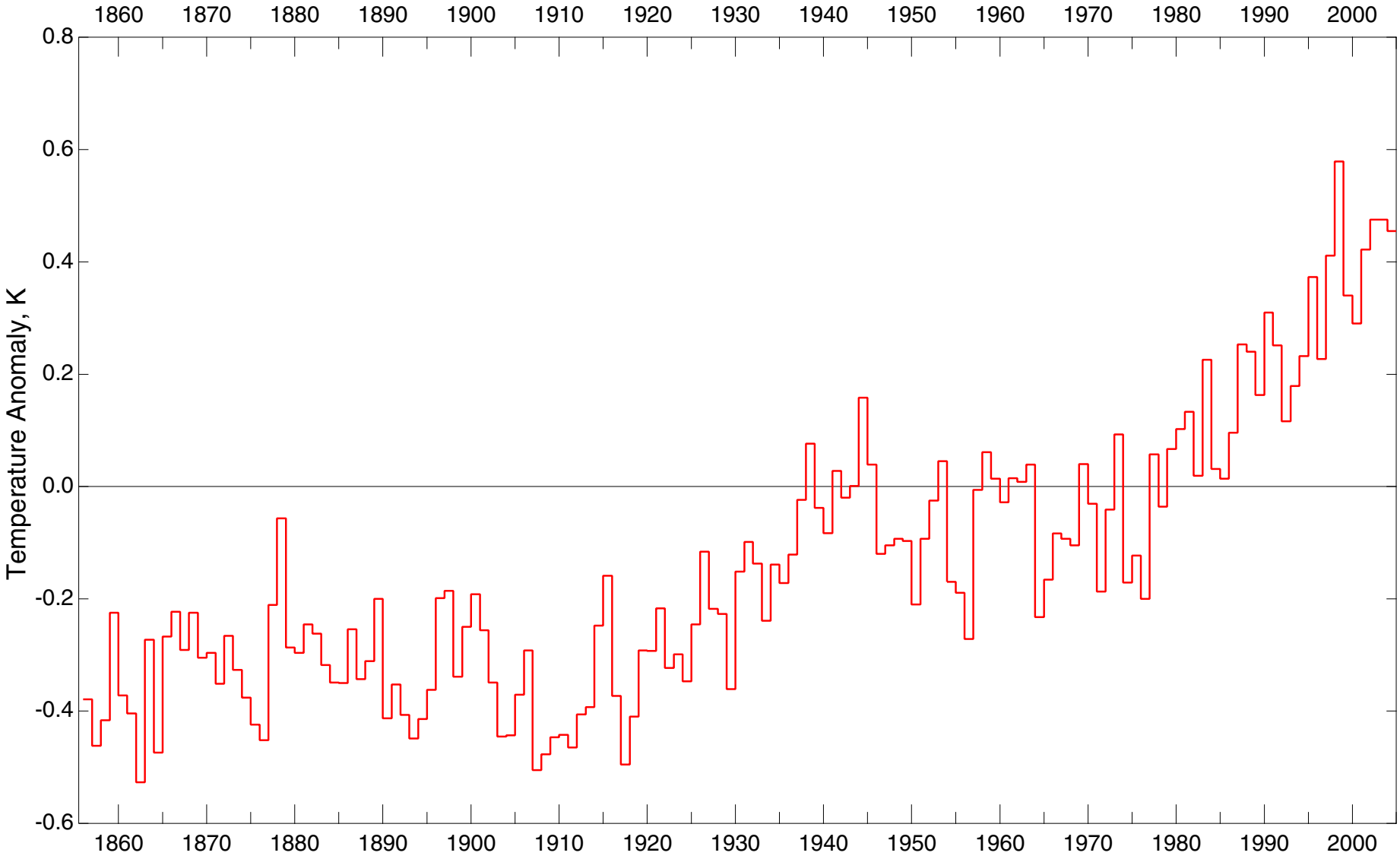
- This hypothesis is fundamental to the radiative forcing concept.
- This hypothesis underlies much of the assessment of climate change over the industrial period.

ATMOSPHERIC CARBON DIOXIDE IS INCREASING



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

CHANGE IN GLOBAL MEAN SURFACE TEMPERATURE 1855-2004



Climate Research Unit, University of East Anglia, UK

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 is proportional to the forcing, but independent of its nature and spatial distribution.

$$\Delta T = \lambda \, \Delta F$$

CLIMATE SENSITIVITY

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

$$\lambda = \Delta T / \Delta F.$$

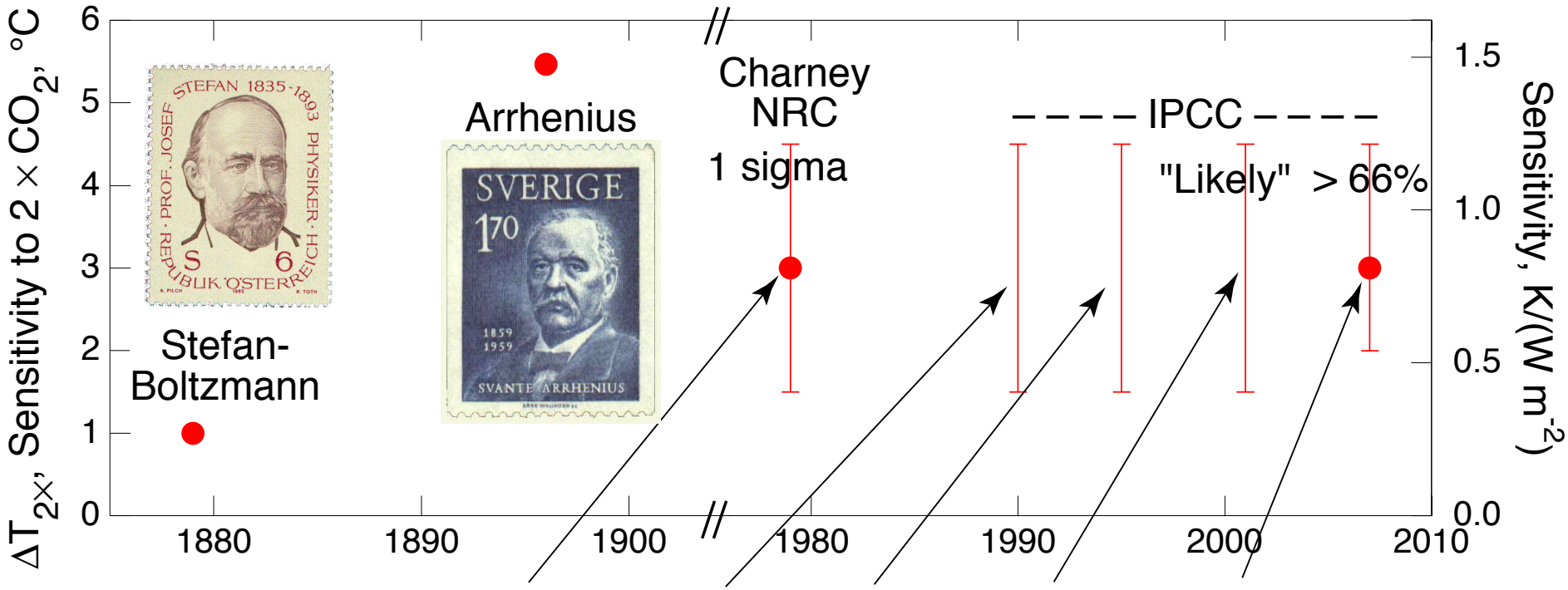
Climate sensitivity is not known and is the objective of much current research on climate change.

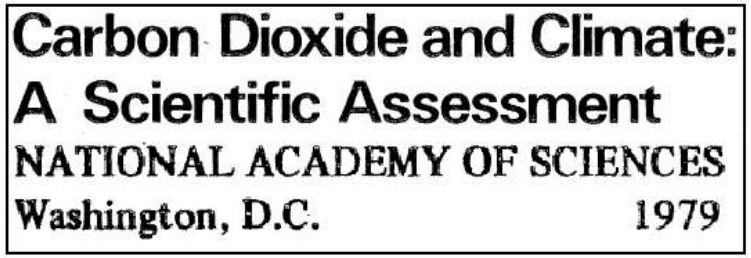
Climate sensitivity is often expressed as the temperature for doubled CO_2 concentration $\Delta T_{2\times}$.

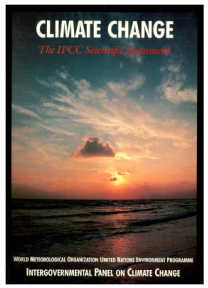
$$\Delta T_{2\times} = \lambda \, \Delta F_{2\times}$$

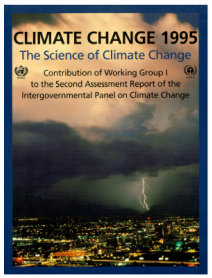
CLIMATE SENSITIVITY ESTIMATES THROUGH THE AGES

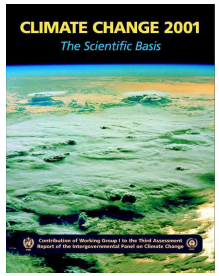
Estimates of central value and uncertainty range from major national and international assessments

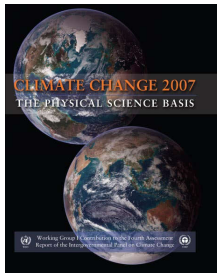








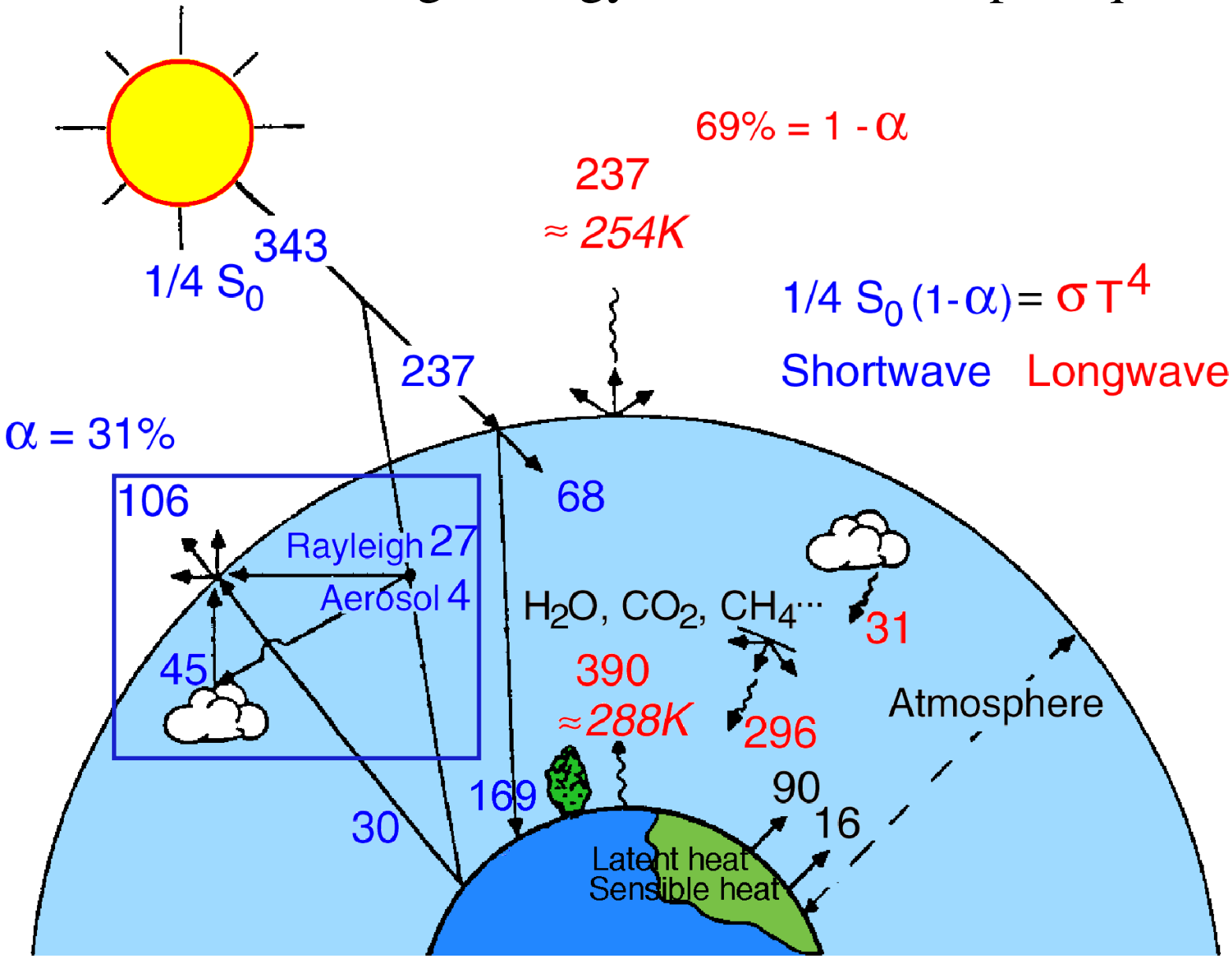




Despite extensive research, climate sensitivity remains ***highly uncertain***.

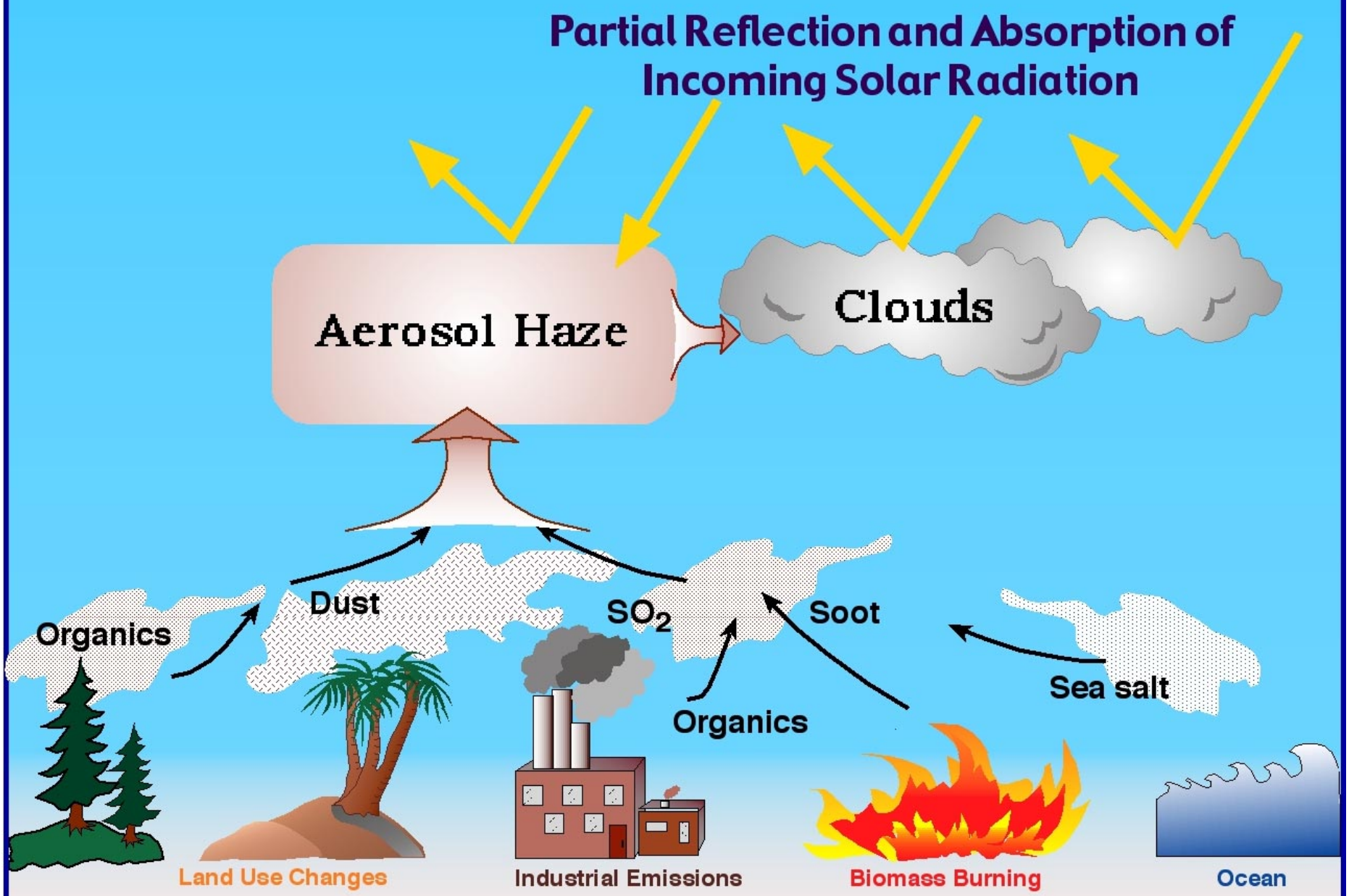
GLOBAL ENERGY BALANCE

Global and annual average energy fluxes in watts per square meter



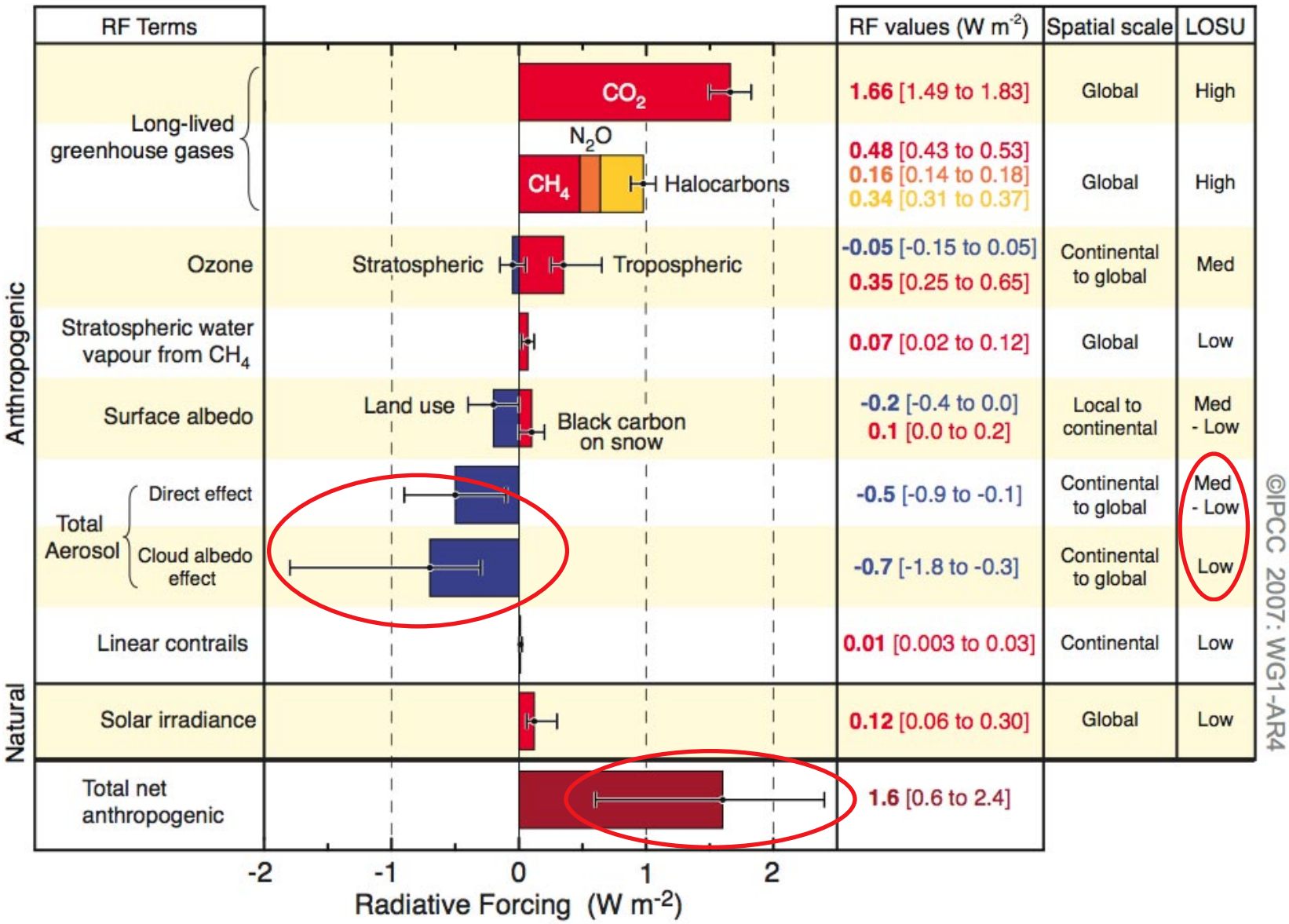
Schwartz, 1996, modified from Ramanathan, 1987

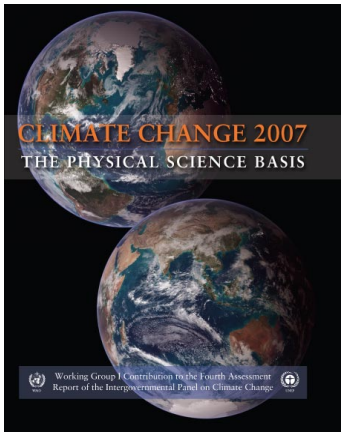
Radiative Forcing by Tropospheric Aerosol



GLOBAL-MEAN RADIATIVE FORCINGS (RF)

Pre-industrial to present (Intergovernmental Panel on Climate Change, 2007)





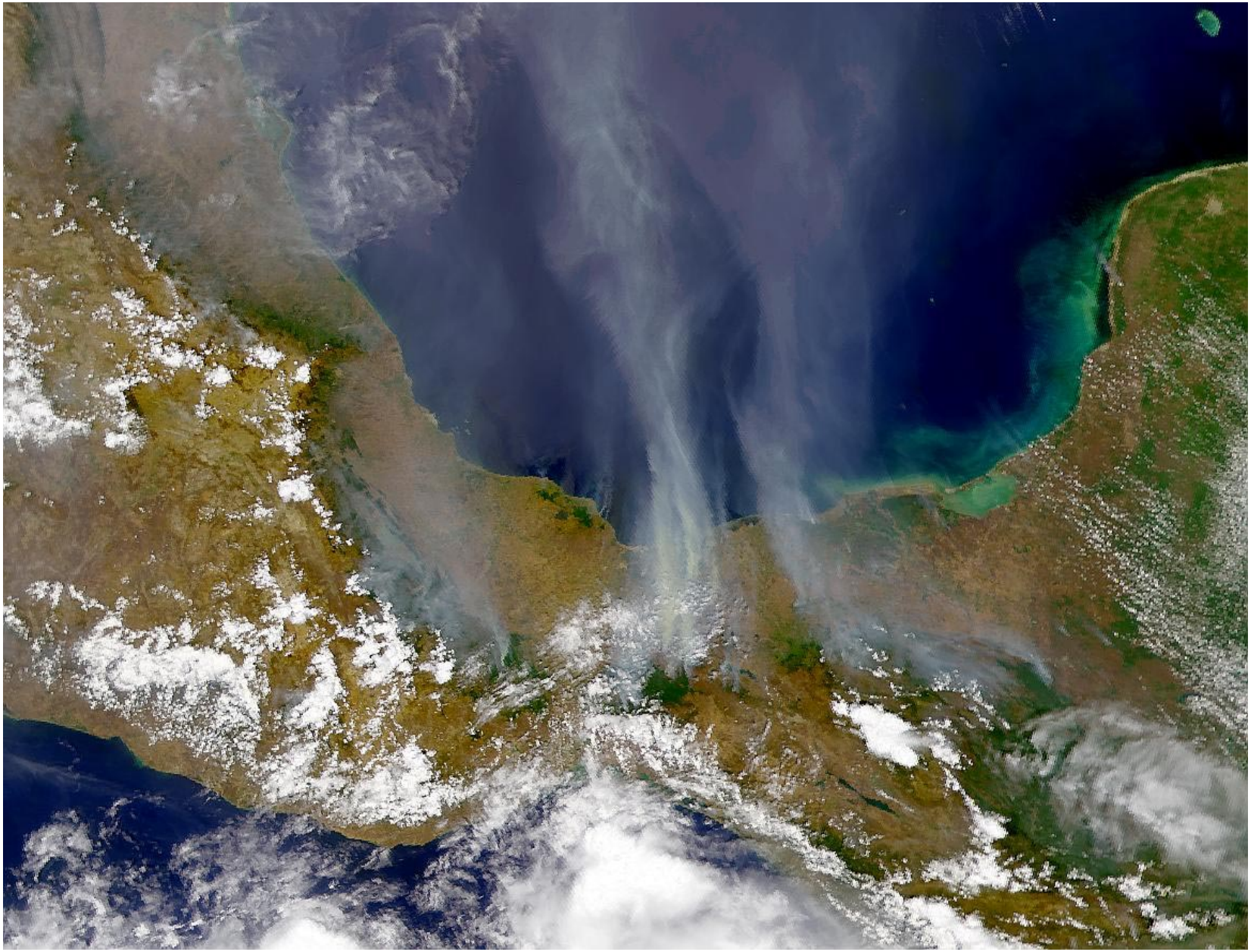
LOSU denotes level of scientific understanding.

AEROSOL IN MEXICO CITY BASIN

AEROSOL IN MEXICO CITY BASIN

Mexico City is a wonderful place to study aerosol properties and evolution.

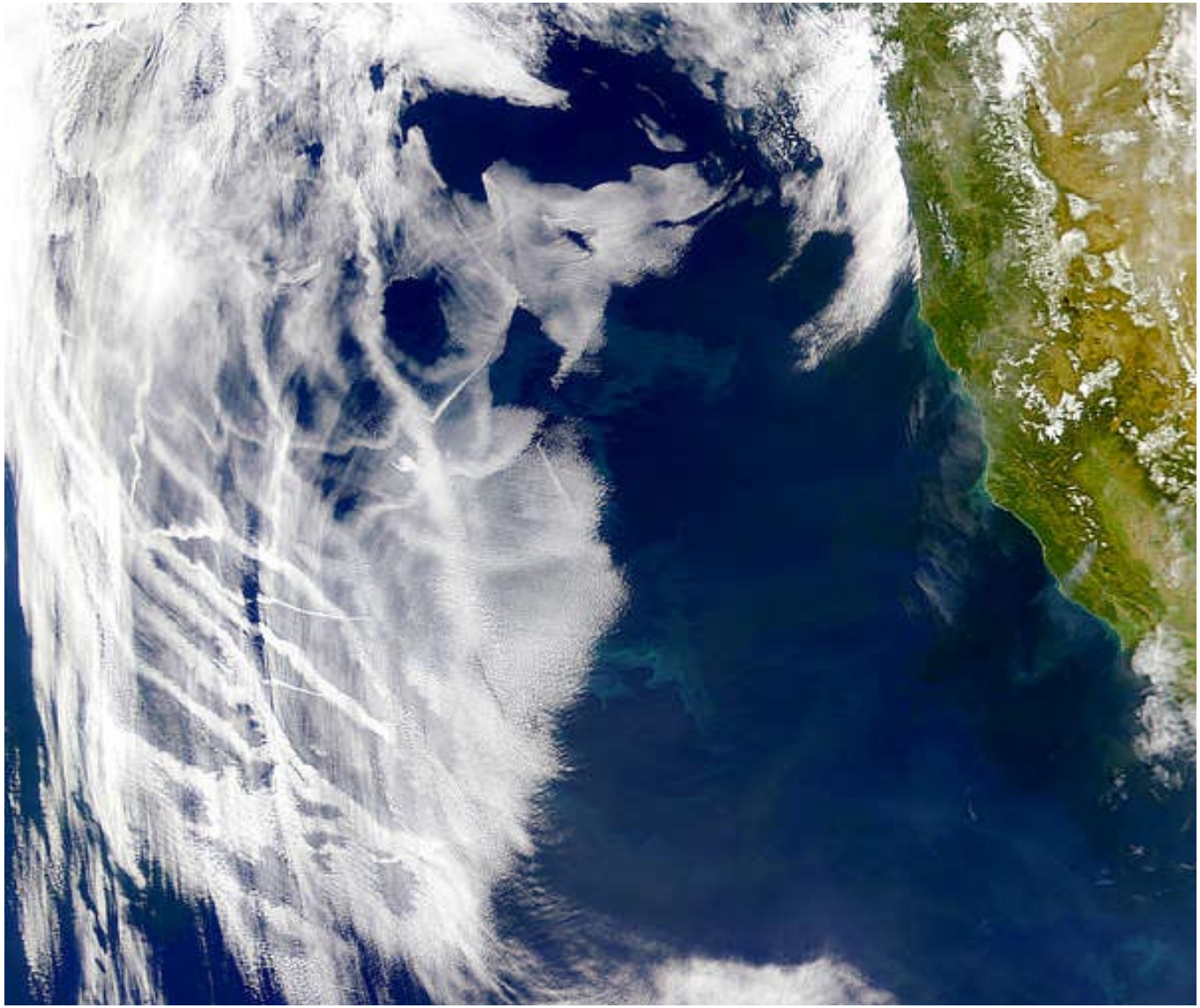
AEROSOLS AS SEEN FROM SPACE



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

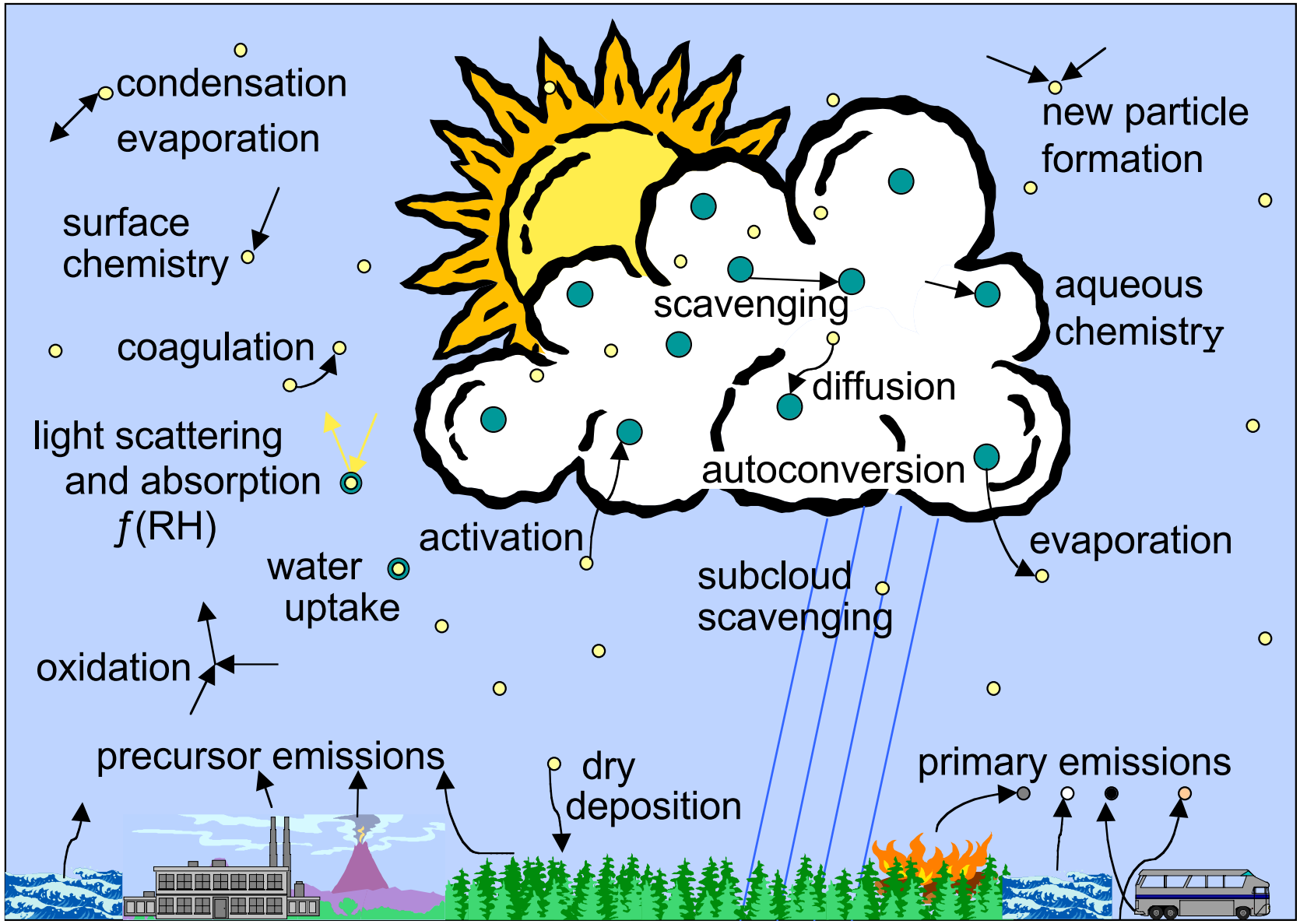
CLOUD BRIGHTENING BY SHIP TRACKS

Satellite photo off California coast



Aerosols from ship emissions enhance reflectivity of marine stratus.

AEROSOL PROCESSES THAT MUST BE UNDERSTOOD AND REPRESENTED IN MODELS



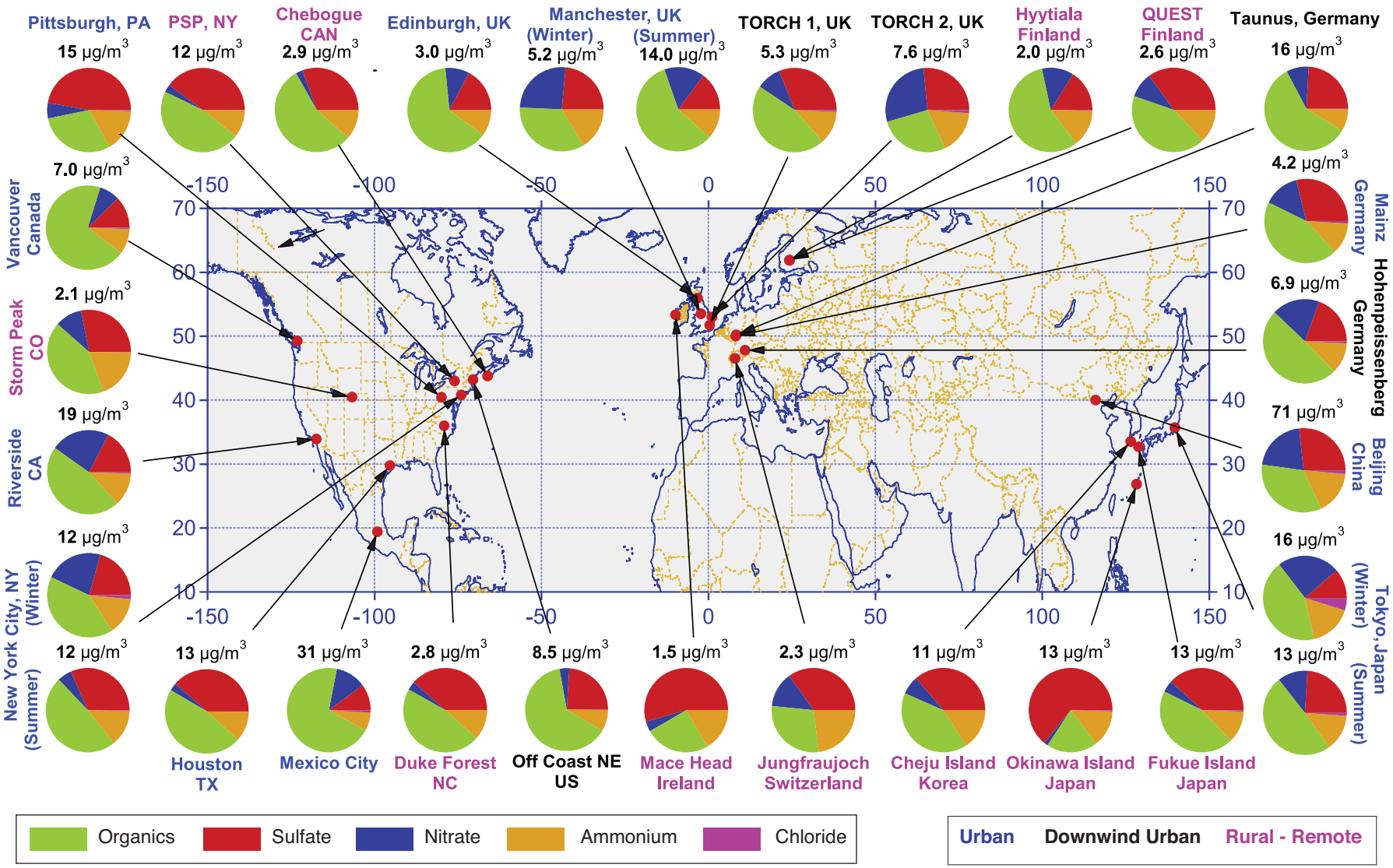
Ghan and Schwartz, Bull. Amer. Meterol. Soc., 2007

RECENT FINDINGS FROM THE DEPARTMENT OF ENERGY'S ATMOSPHERIC SCIENCE PROGRAM

For more information please visit
www.asp.bnl.gov

DOMINANCE OF ORGANIC AEROSOL

Measurements by aerosol mass spectrometer

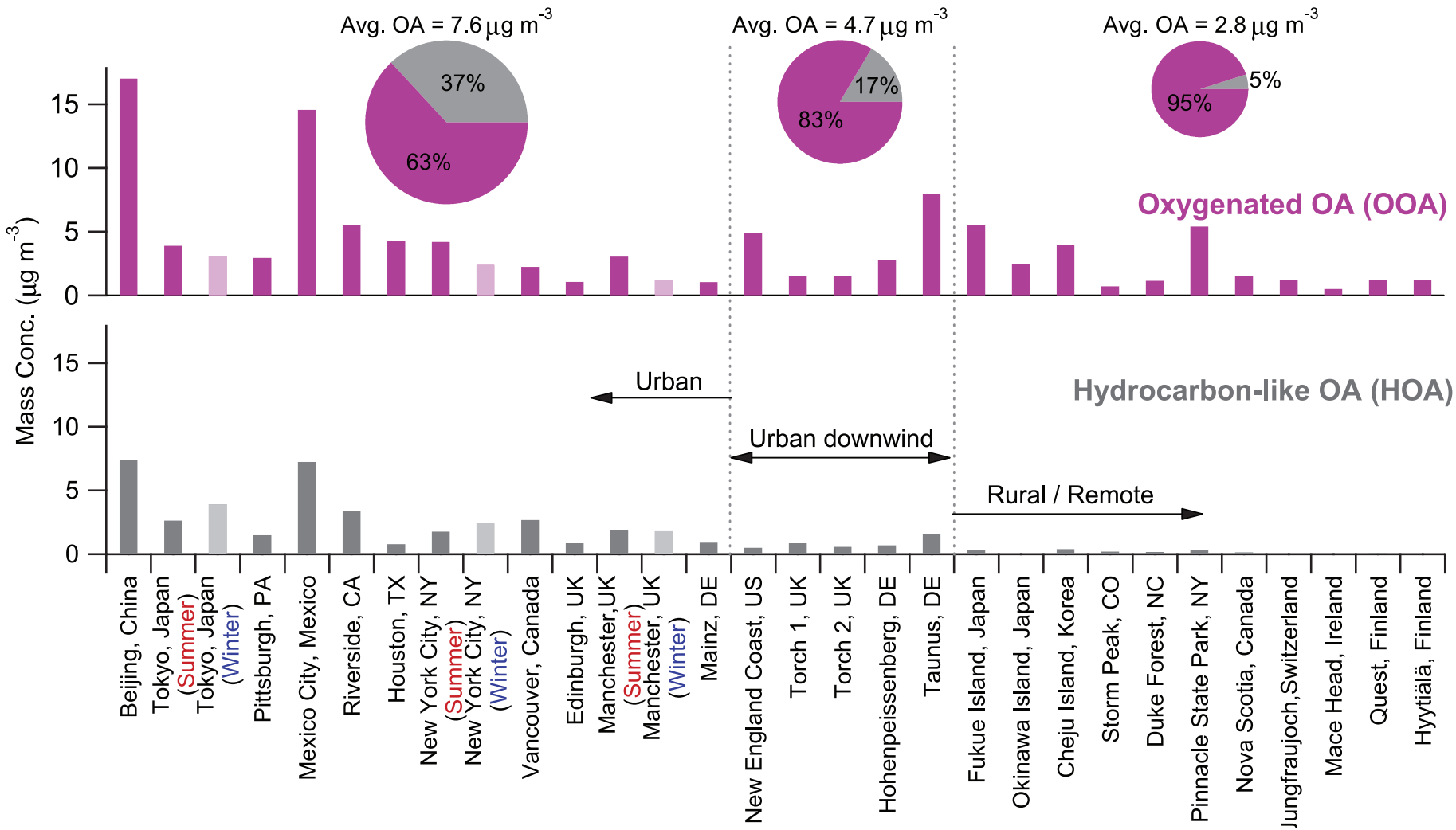


Zhang et al., GRL, 2007

Organic aerosol is major or dominant species throughout the anthropogenically influenced Northern Hemisphere.

PRIMARY AND SECONDARY ORGANIC AEROSOL BY LOCATION TYPE

Area of pie scaled to organic aerosol concentration

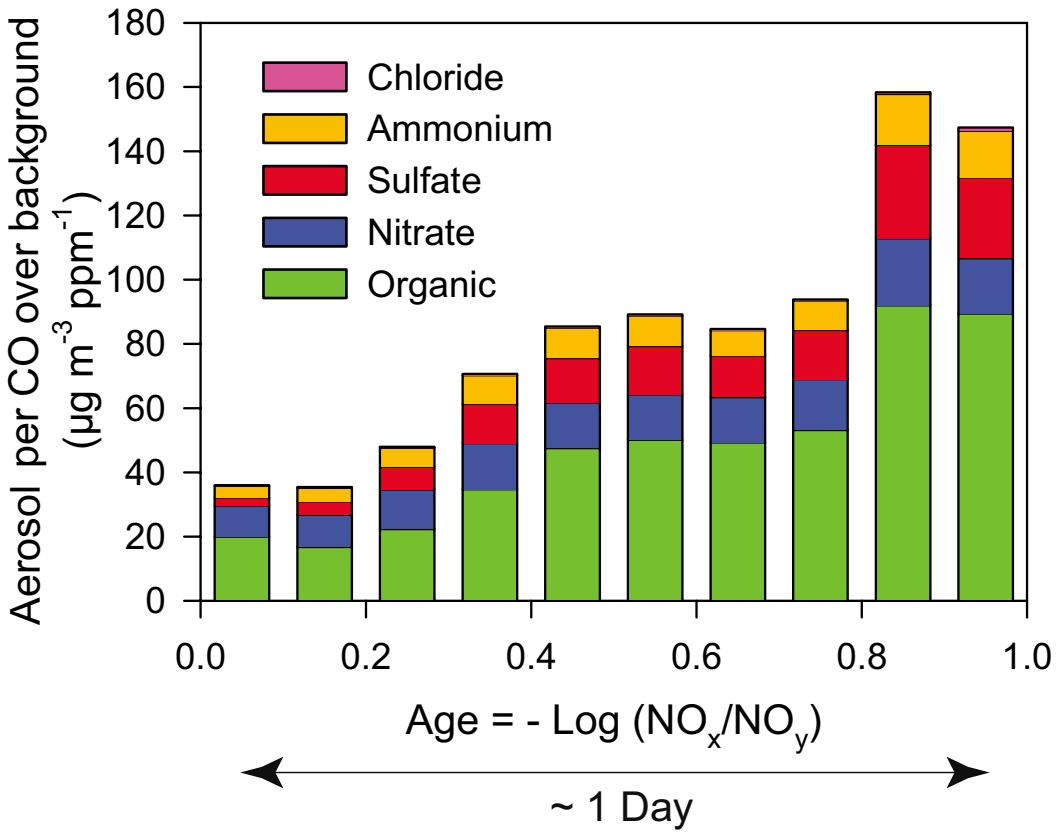


Zhang et al., GRL, 2007

Secondary fraction increases with increasing distance from urban sources.

SECONDARY AEROSOL PRODUCTION

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



Kleinman et al, ACP, 2008

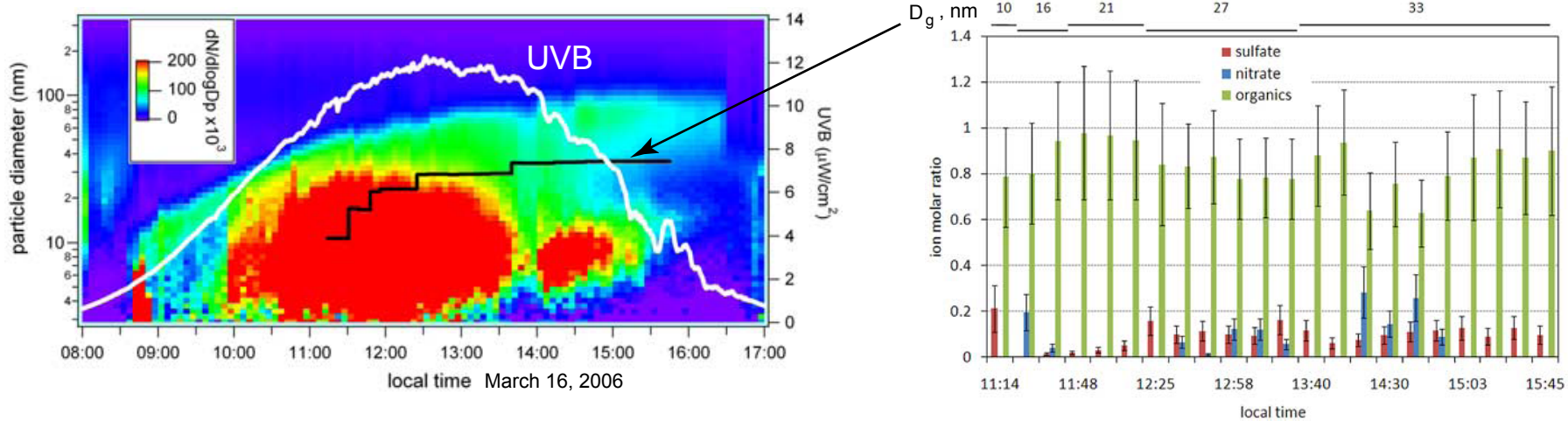
Parcel photochemical age measured using $-\mathrm{Log}(NO_x/NO_y)$ as clock.

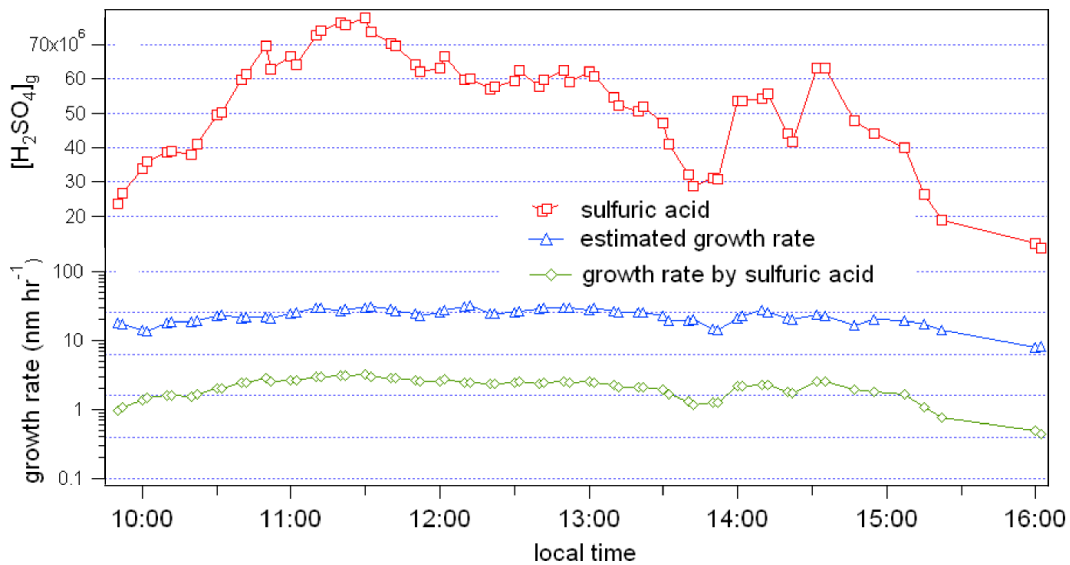
Aerosol normalized to CO above background to account for dilution.

Fivefold increase in organic aerosol.

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

Following the growth of particles formed from nucleation at Tecamac, Mexico





Sulfate accounts for only ~10% of particulate mass.

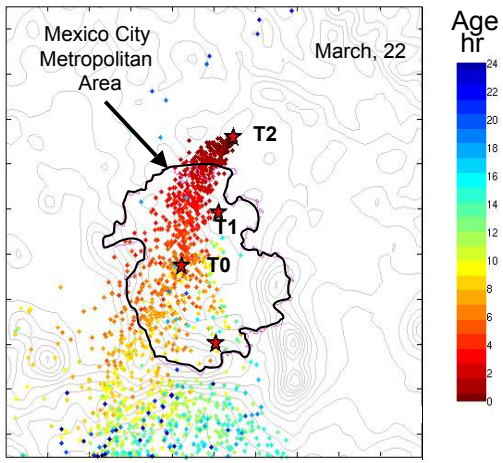
Growth rate exceeds that from sulfuric acid by order of magnitude.

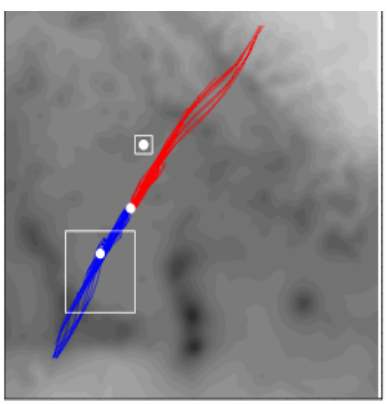
Smith, McMurry et al., GRL, 2008

AEROSOL TRANSPORT AND EVOLUTION

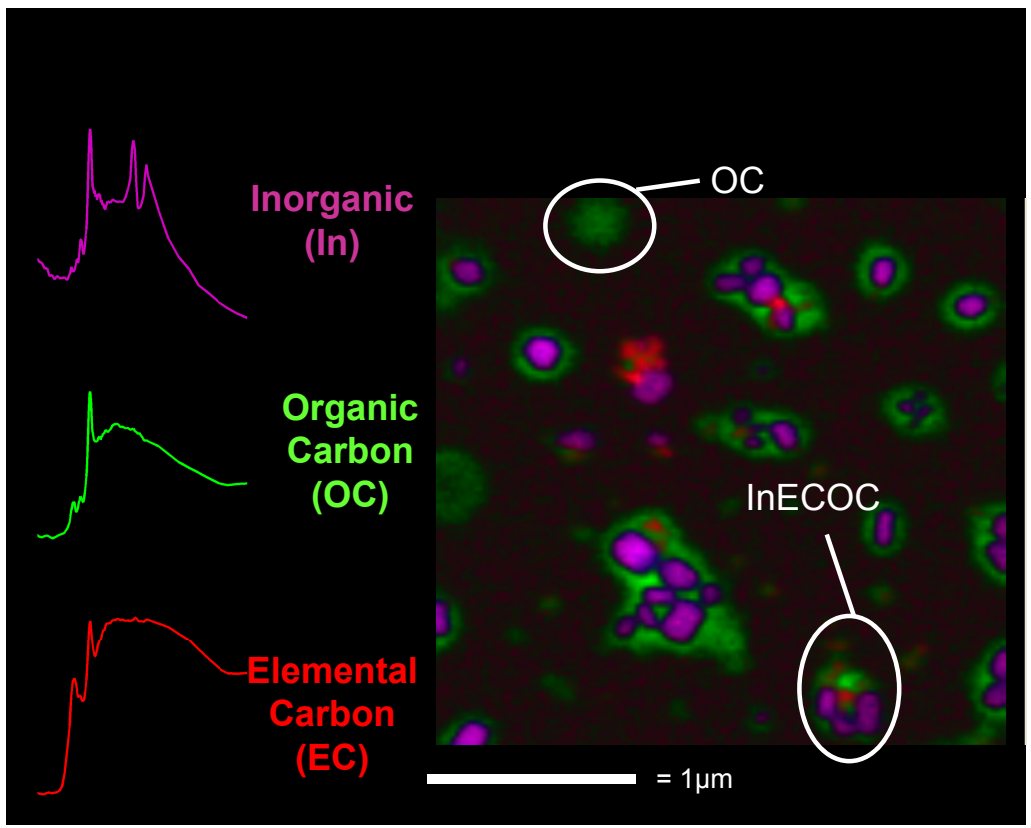
Mexico City, March 22, 2006

FLEXPART age and back trajectories

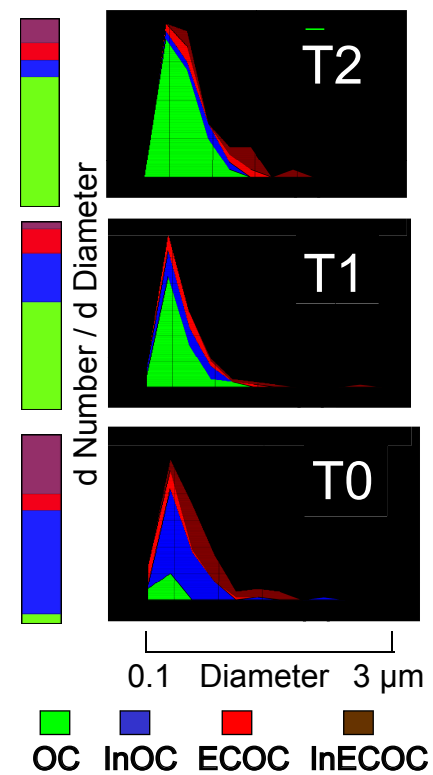




Component ID by scanning transmission electron microscopy



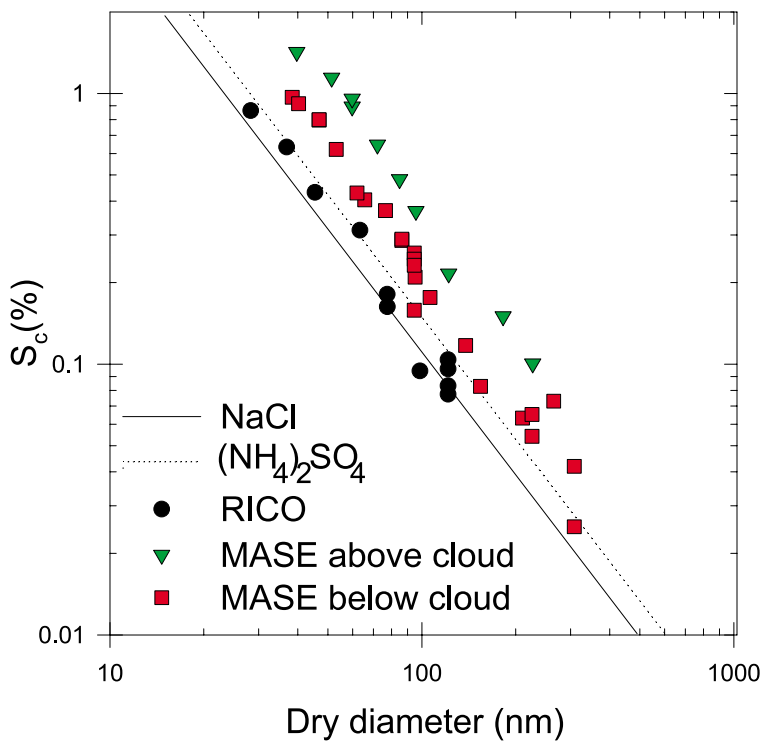
Number fraction and number size distribution

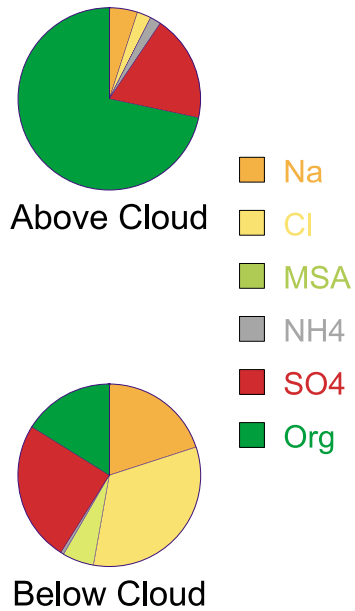


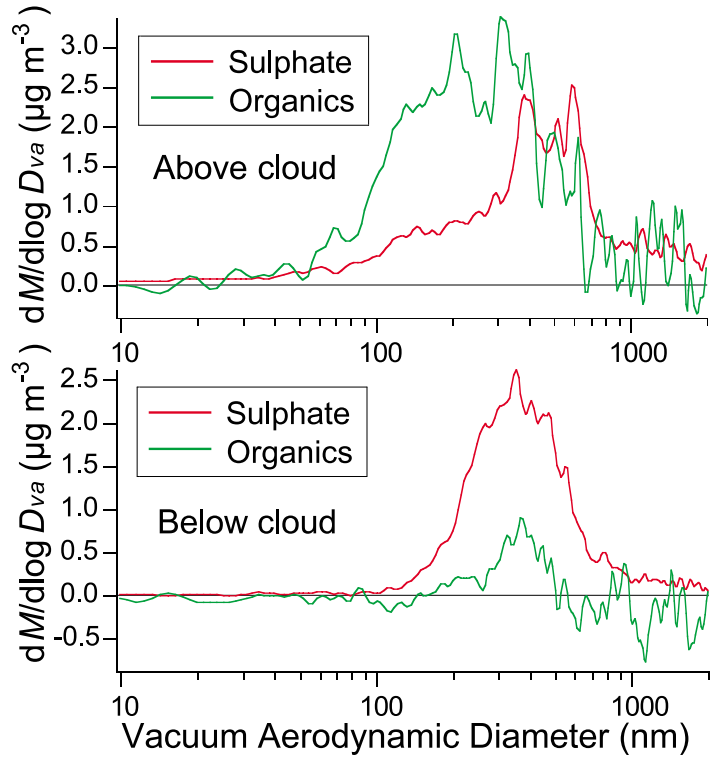
Moffet, Tivanski, Hopkins, Desyaterik, Fast, Barnard, Laskin, Gilles; PNNL, LBL

COMPOSITION MATTERS

Size dependent critical supersaturation of aerosol particles







J. Hudson, Y.-N. Lee, M. Alexander

Measurements below (110-170 m) and above (400-470 m) clouds off the coast of California, north of San Francisco, on July 25, 2005.

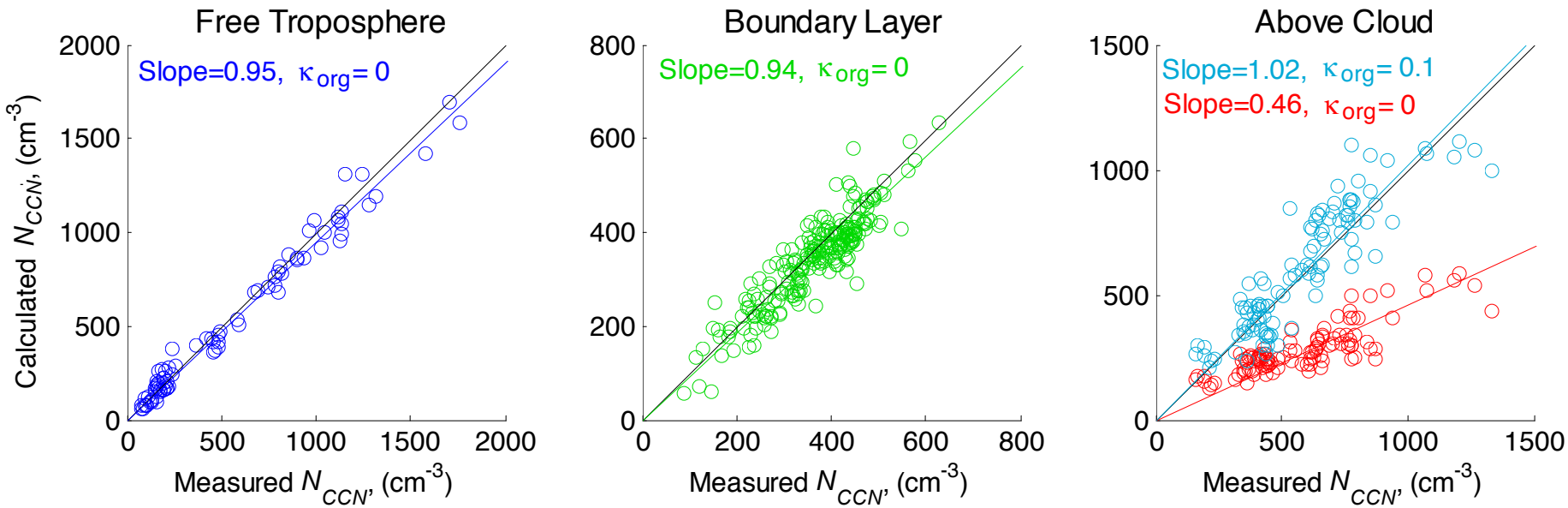
Higher supersaturation is required to activate particles with greater organic fraction.

Bulk composition determined by PILS (particle into liquid sampler).

Size-dependent composition determined by aerosol mass spectrometer.

CLOSURE STUDY ON CCN CONCENTRATION

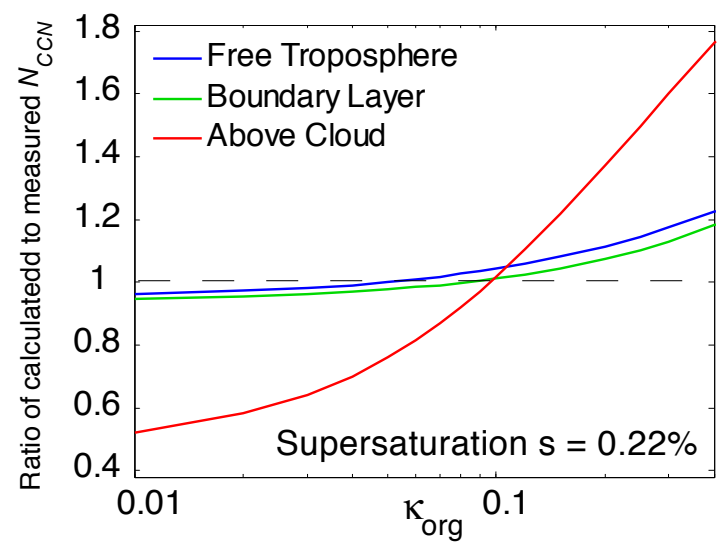
CCN concentration at 0.22% supersaturation for 9 flights during MASE, July 2005



$$S = \frac{D^3 - D_p^3}{D^3 - D_p^3\left(1-\kappa\right)} \exp\left(\frac{4\sigma_w M_w}{RT\rho_w D}\right) \qquad \kappa = \sum_i x_i \kappa_i$$

Strong dependence for above cloud aerosol because of high organic fraction, ~ 80%.

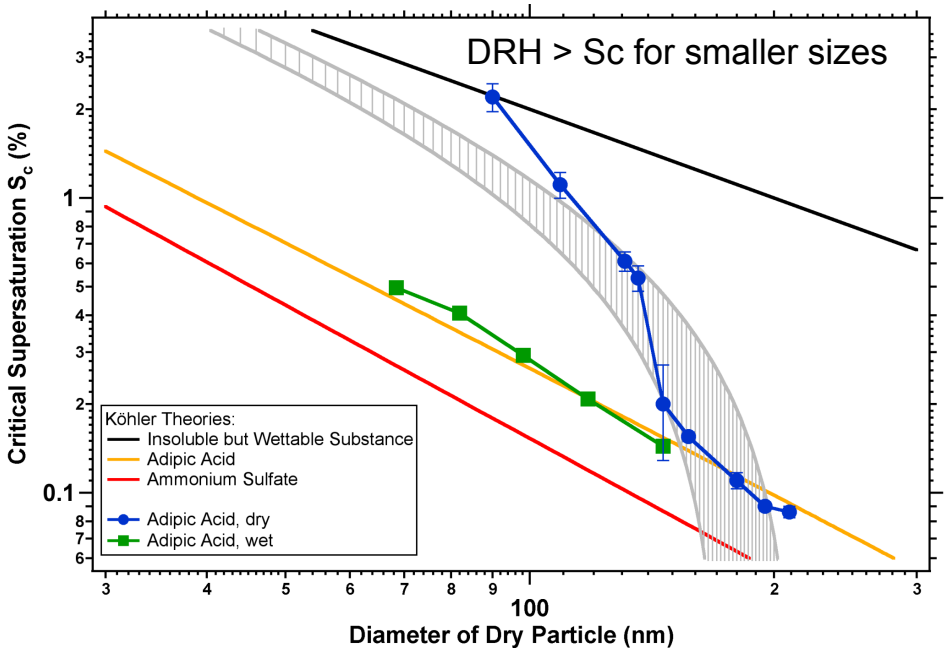
Weak dependence on κ_{org} for free troposphere and boundary layer aerosol because of lower organic fraction, ~ 50%.

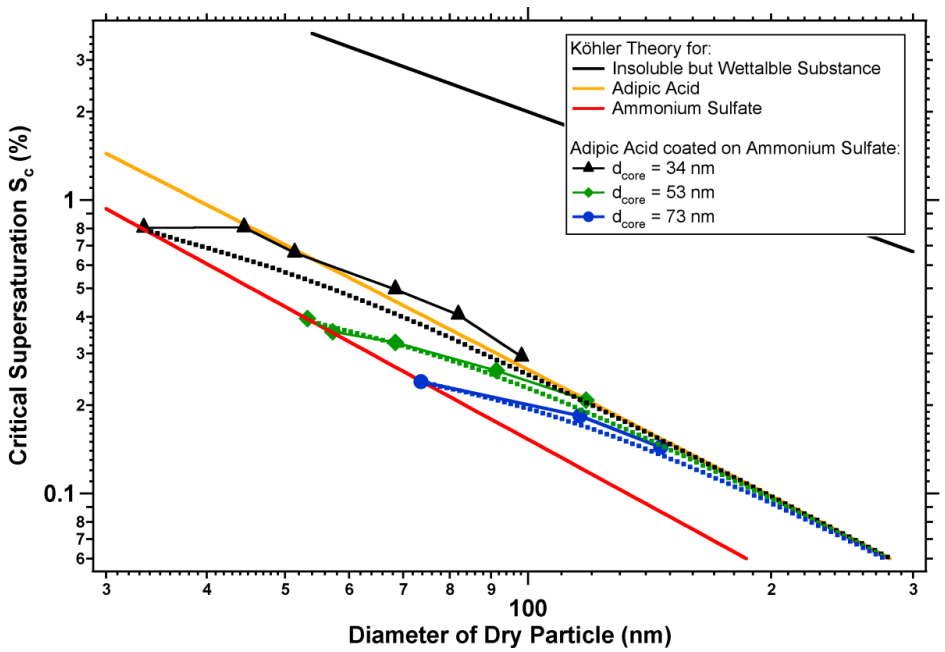


Wang, Lee, Daum, BNL; Alexander, PNNL; Jayne, Aerodyne

LABORATORY STUDIES OF ACTIVATION OF MIXED ADIPIC ACID – AMMONIUM SULFATE PARTICLES

Sparingly soluble organic acid – soluble inorganic salt





Koehler theory works well mixed adipic acid and sulfate particles.

For slightly soluble compounds initial particle phase is very important.

P. Davidovits, Boston Coll.; T. Onasch, Aerodyne, Inc., et al.

WATER WATER EVERYWHERE

Water is taken up by hygroscopic aerosol particles increasing their light scattering.

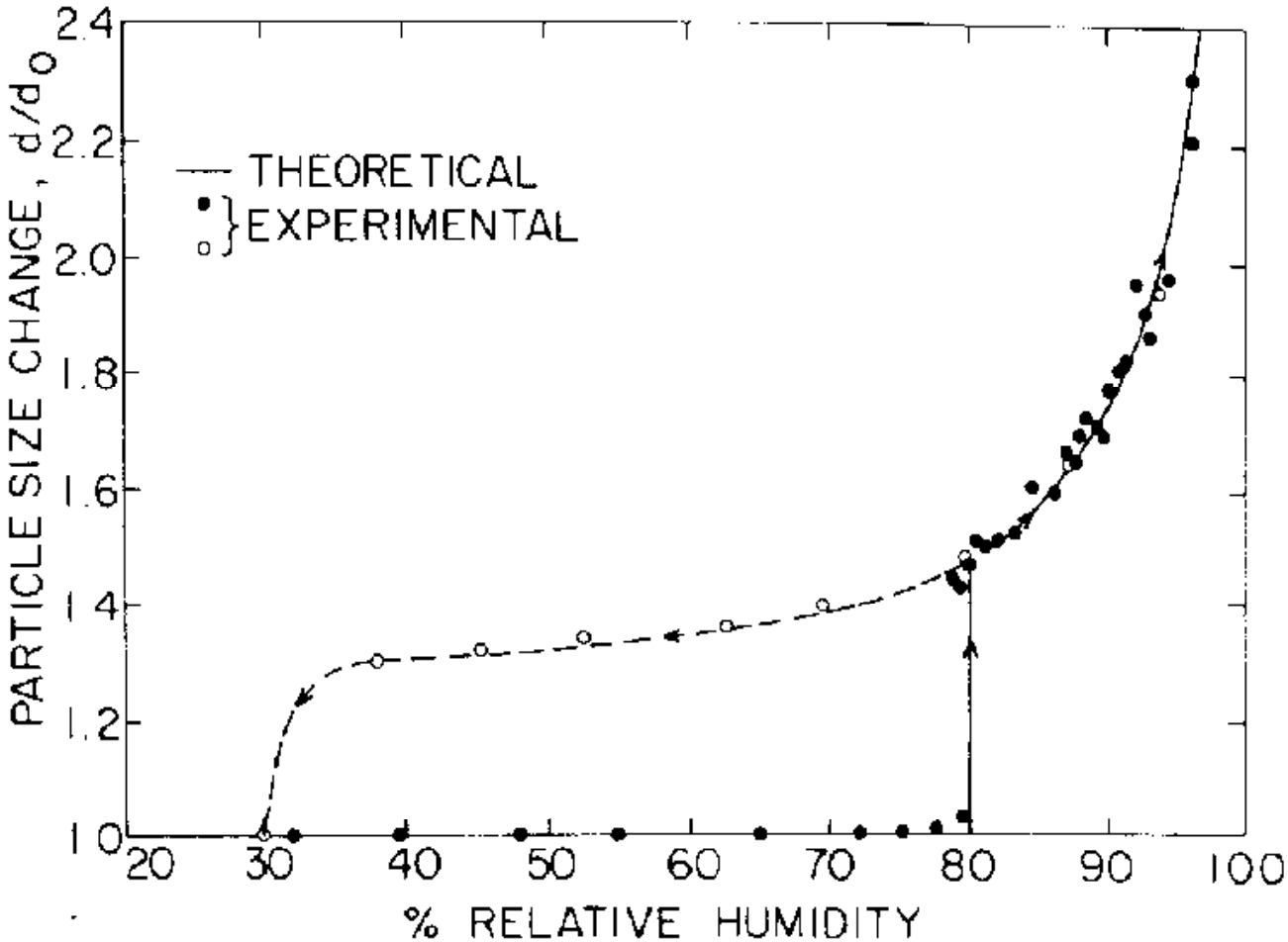
Water content depends on composition, relative humidity, and history.

CCN activity depends on composition and surface properties.

Understanding interactions with water is central to describing aerosol optical and cloud nucleating properties.

DELIQUESCENCE, SIZE CHANGE, AND EFFLORESCENCE OF HYGROSCOPIC PARTICLES

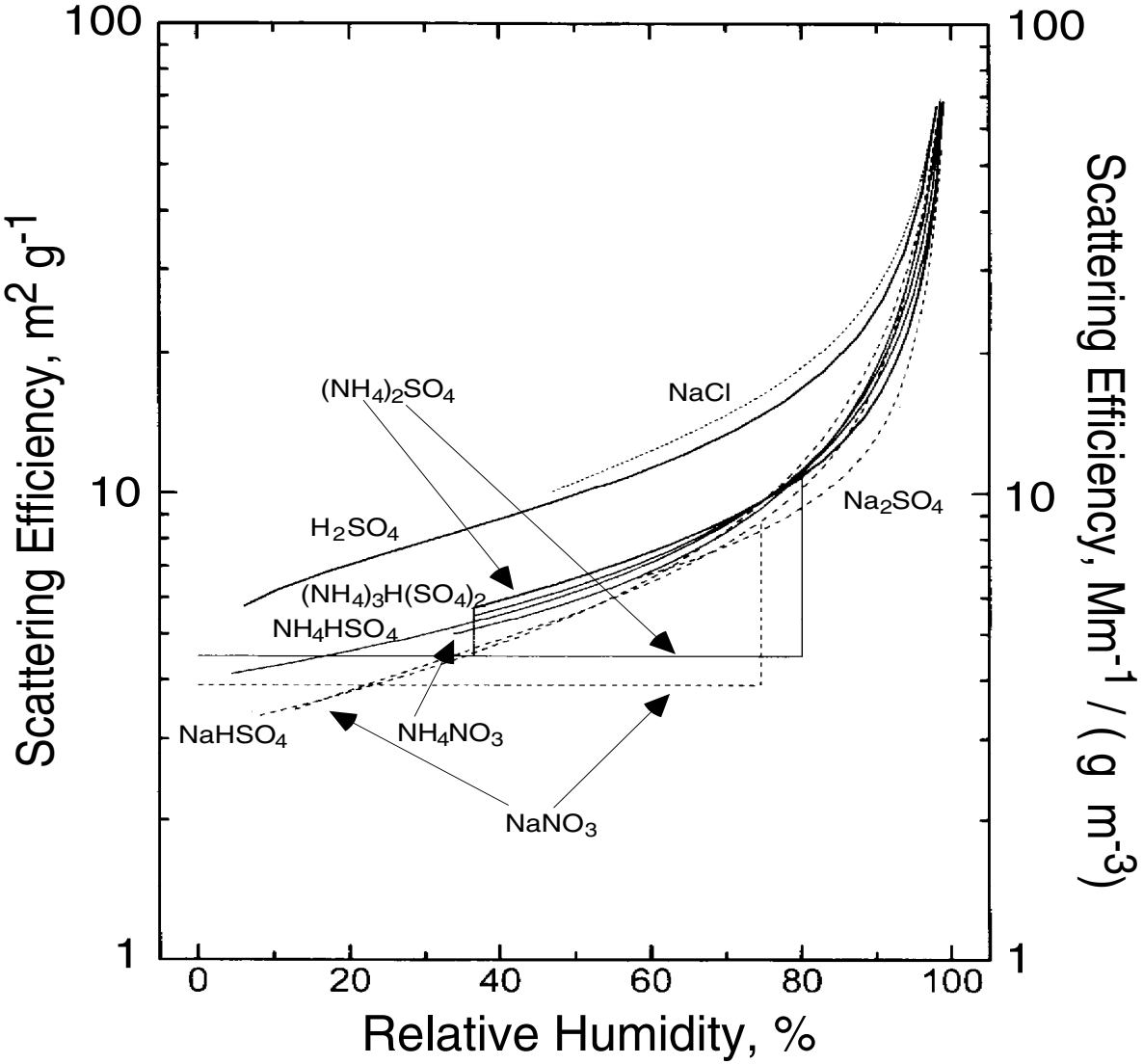
Ammonium Sulfate at 25°C



Tang and Munkelwitz, 1984

WATER UPTAKE AND LIGHT SCATTERING COEFFICIENT

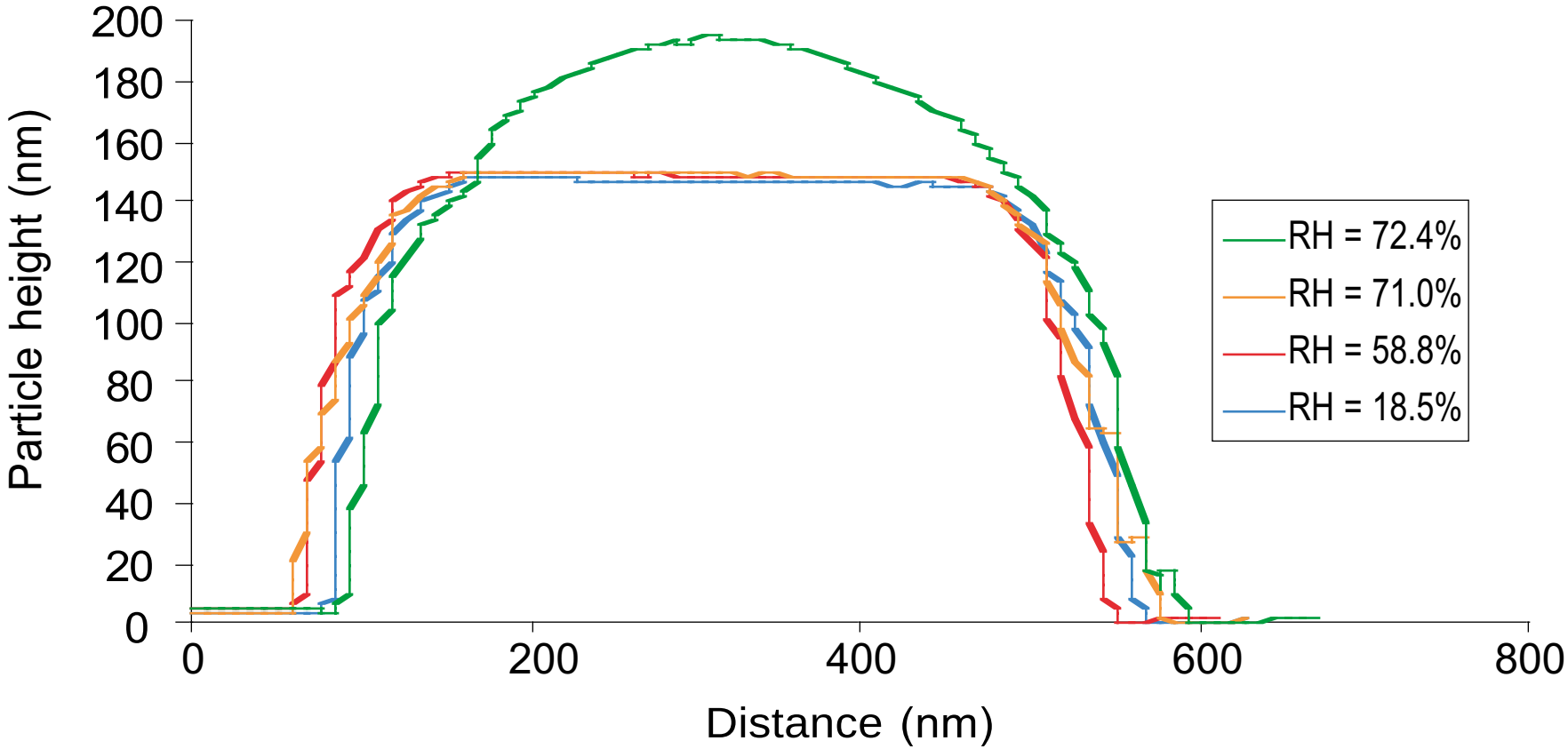
Dependence on relative humidity for various substances



Lognormal, D_g = 0.3 μm; σ_g = 1.5; I. Tang, JGR, 1996

WETTING OF SODIUM CHLORIDE

Change of particle shape studied by Atomic Force Microscopy



Oatis, Strasberg, Checco, Ocko; BNL Material Sci. Cond. Matt. Phys. Dept.

Representative AFM line profiles of a single NaCl salt nanoparticle on flat silicon oxide terminated surface.

No apparent uptake of water below ~ 71% RH.

Significant change in morphology observed near deliquescence RH 75%.

SOOT HAPPENS

Elemental carbon is a ubiquitous component of atmospheric aerosols.

Significant absorber of atmospheric radiation.

Often emitted as small chain aggregates that are modified by uptake of semivolatiles changing morphology and optical properties.

Characterization of soot is a challenge in field measurements.

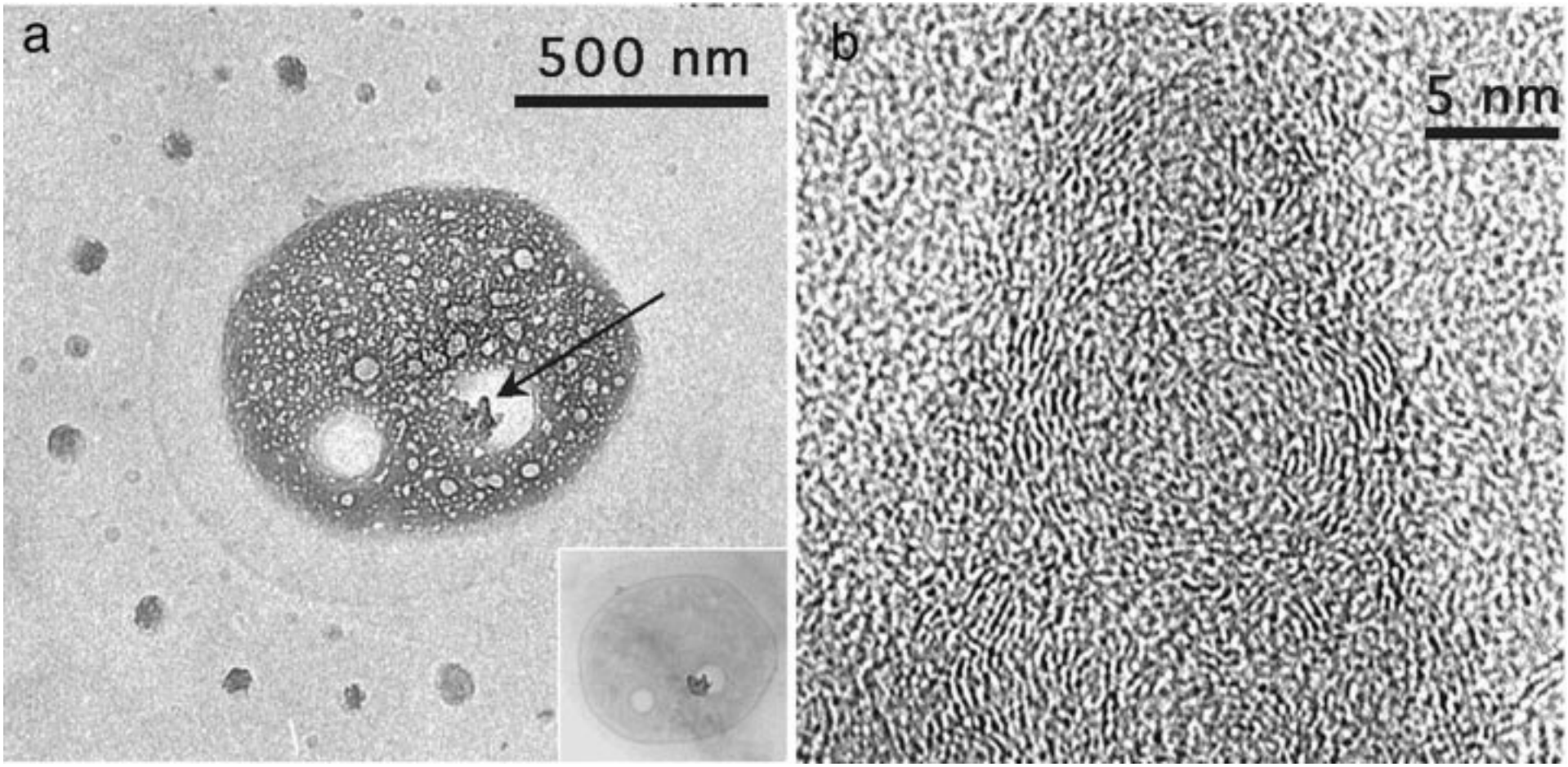
Understanding soot morphology and its changes is central to describing its optical properties and fate in atmosphere.

HOW BLACK IS BLACK CARBON?

TRANSMISSION ELECTRON MICROGRAPH OF INDIVIDUAL PARTICLE

Soot inclusion in single particle consisting mainly of $(NH_4)_2SO_4$

Unpolluted air near Tasmania.



Buseck and Pósfai, PNAS, 1999

ELECTRON TOMOGRAM OF SINGLE SOOT PARTICLE

Clicking on the image should download and show a movie showing rotation of the image. Alternatively enter the link below the picture (which is to Peter Buseck's web page server) into a browser.

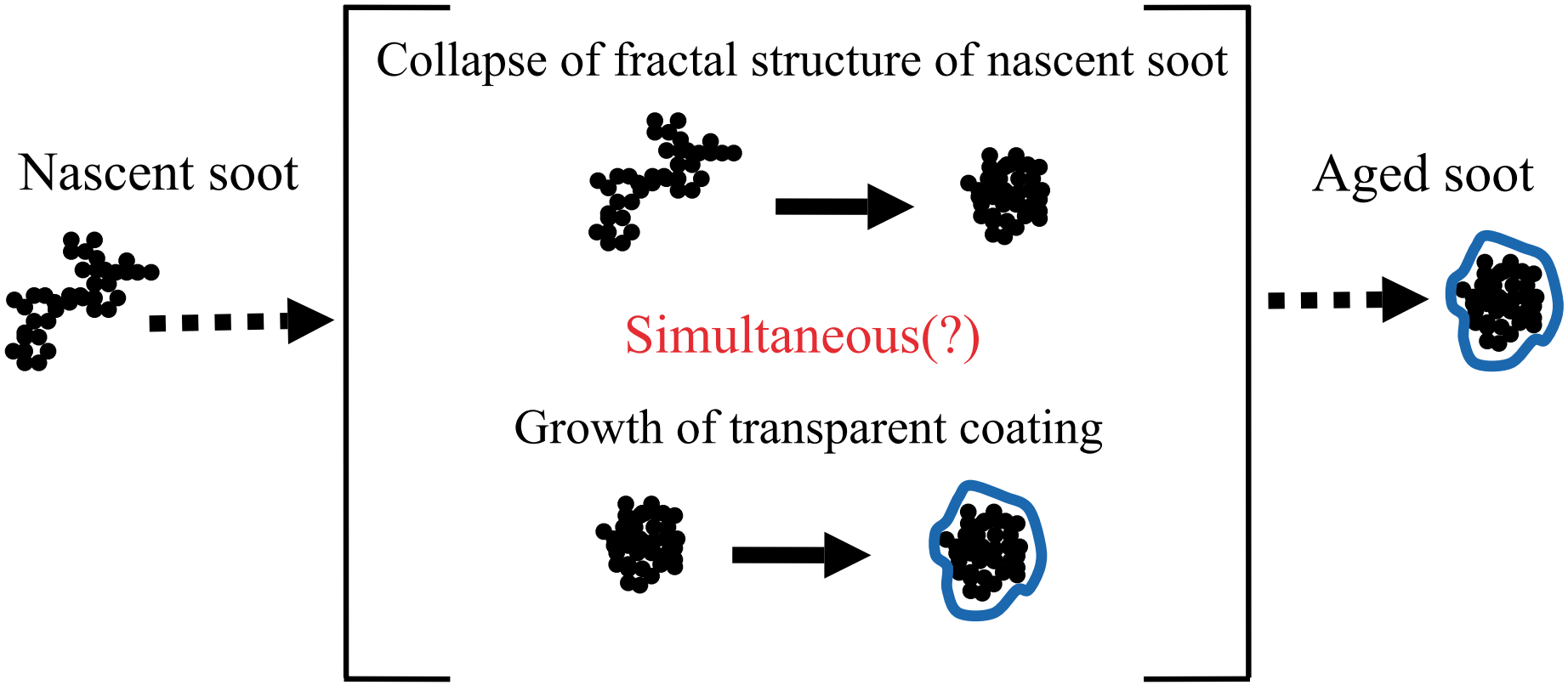
200 nm

Van Poppel, Friedrich, Spinsby, Chung, Seinfeld, and Buseck, Geophys. Res. Lett., 2005

http://7starm.asu.edu/movies/DieselSoot_lvp184.mpg

ATMOSPHERIC AGING OF SOOT

Condensation of semi-volatile secondary particulate matter and collapse of fractal structure under influence of surface tension



Collapse decreases absorption efficiency because of screening.

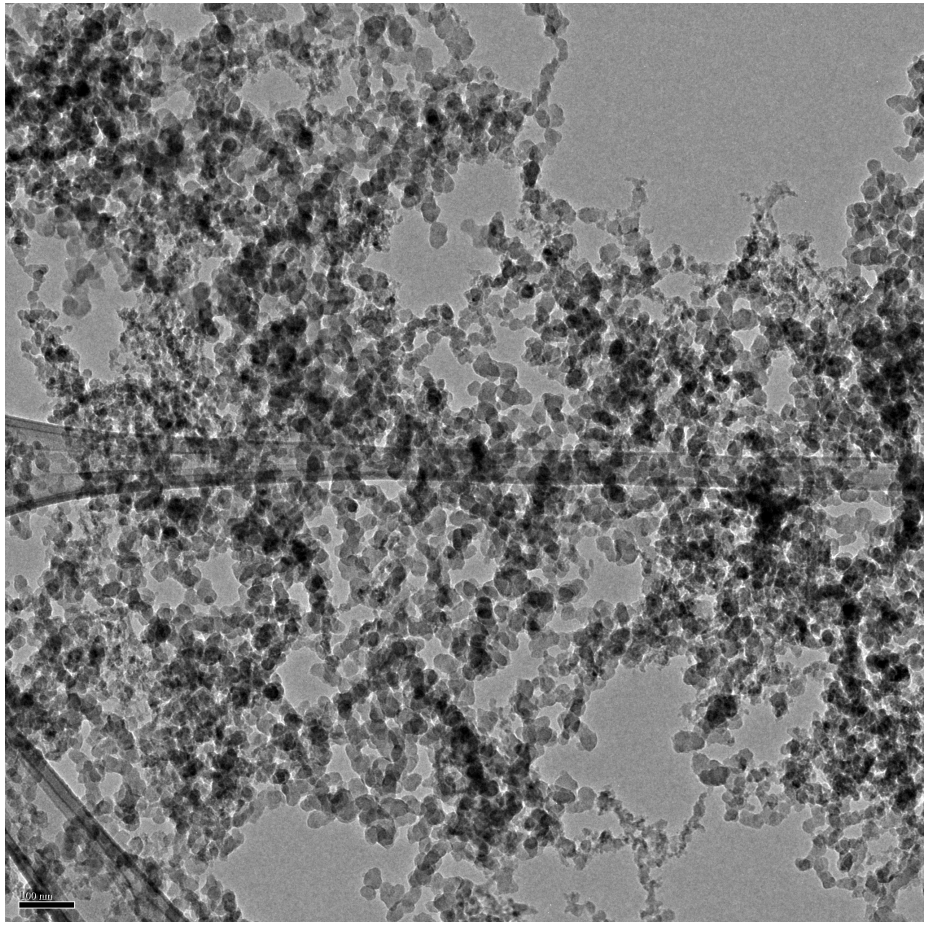
Transparent coating increases absorption efficiency because of lensing.

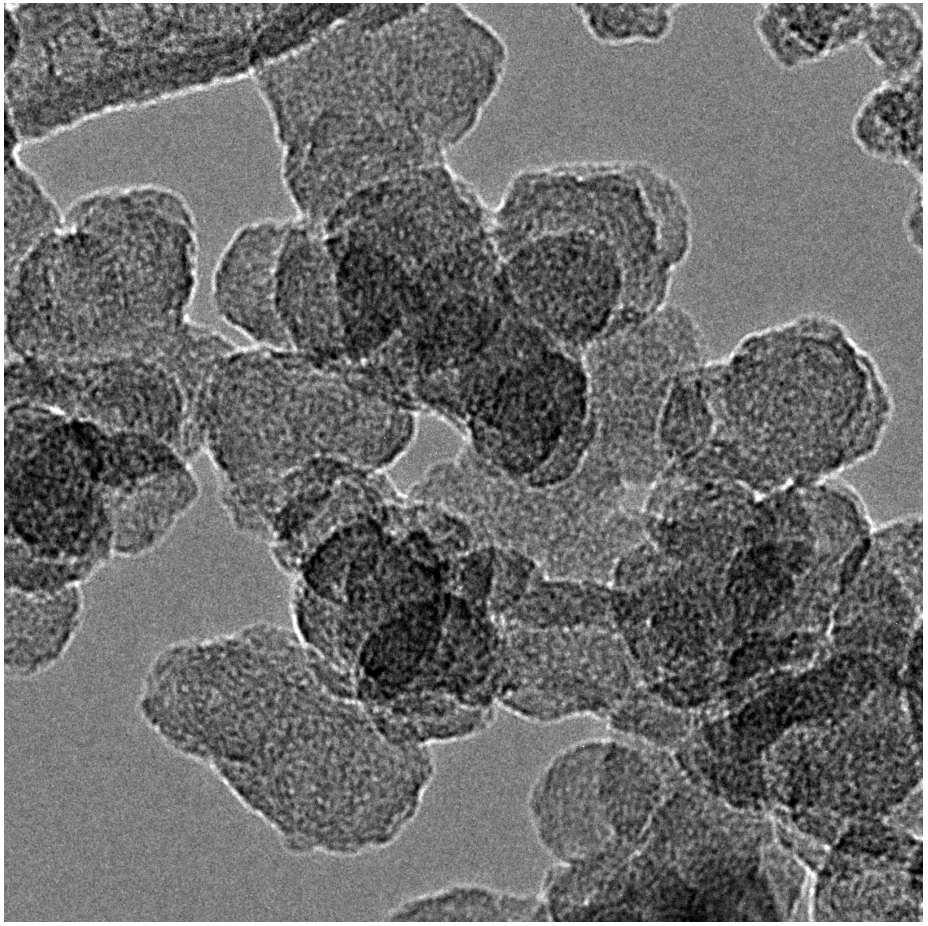
Hygroscopic coating would make particle more active cloud condensation nucleus.

Art Sedlacek, ASD, BNL

FRESHLY GENERATED FRACTAL SOOT

TEM images of acetylene-air soot





100 nm

Lihua Zhang, CFN; Art Sedlacek, Jason Olfert, ASD

CONCLUDING REMARKS

Atmospheric aerosols are dysfunctional nanomaterials.

Heterogeneous: space, time, sources, composition, morphology.

Important to climate and climate change.

Necessary to develop methods to characterize properties and evolution, and to represent these in climate models.

Understanding aerosol properties and evolution would benefit greatly from advanced experimental methods.