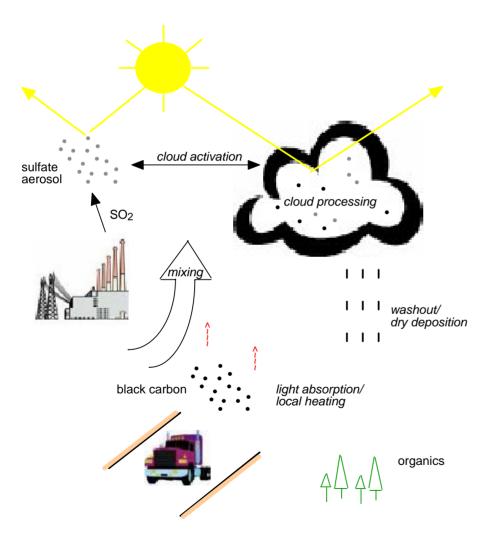
Robert McGraw and James Wegrzyn Brookhaven National Laboratory Upton, NY 11973

**Overview:** 

- Survey of emissions and BC aerosol processes
- New developments in modeling (aerosols, clouds, and climate)
- Future studies

# AEROSOL PROCESSES AND THEIR IMPACT ON CLIMATE

Understanding and modeling the importance of diesel emissions reduction on climate



### SOME KEY QUESTIONS:

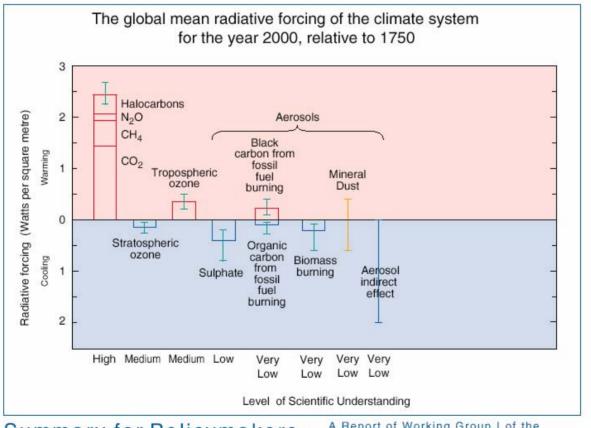
Emissions: What is the contribution of diesel as a primary BC aerosol source? (including impact of clean diesel technology and lower emissions of CO<sub>2</sub> on climate)

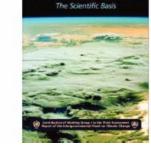
What is the subsequent mixing state of the BC aerosol? (e.g. does BC become coated with sulfate?)

Is mixing state important to the assessment of BC aerosol optical properties, cloud properties, and climate effects?

# RADIATIVE FORCING OVER THE INDUSTRIAL PERIOD IPCC (2001)

GHG's and aerosol direct and indirect effects





CLIMATE CHANGE 2001

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

## **EMISSIONS**

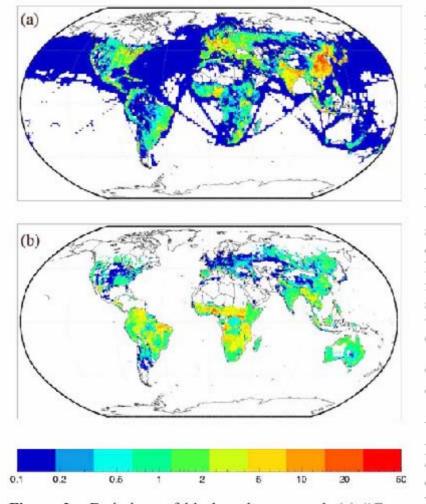
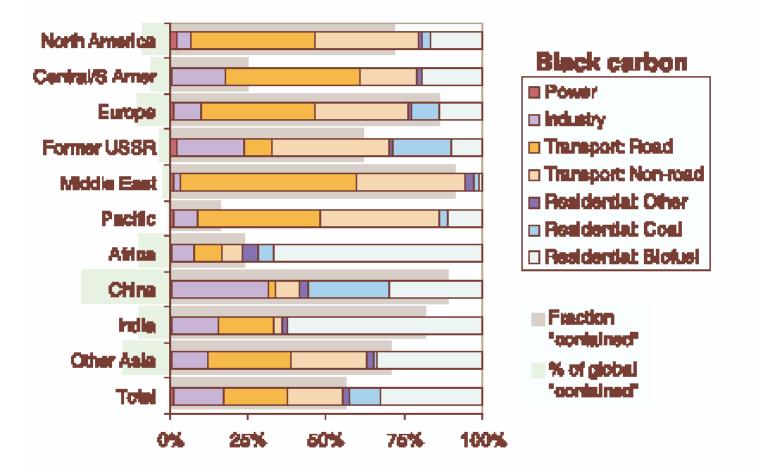


Figure 2. Emissions of black carbon aerosol. (a) "Contained" combustion, based on 1996 activity data. (b) Open burning, annual average. The color coding is an approximately logarithmic scale. Units are ng/m<sup>2</sup>/s (1 ng/m<sup>2</sup>/s  $\sim$ 32 kg/km<sup>2</sup>/yr).

T. Bond et al., "A technologybased inventory of black carbon and organic carbon emissions from combustion. J. Geophys. Res. 109, D14203 (2004)

# **EMISSIONS (CONTINUED)**



From Bond et al. J. Geophys. Res. <u>109</u>, D14203 (2004)



Figure from Andreae M. O. "The dark side of aerosols", *Nature* <u>409</u>, 671-672 (2001).

Importance of mixing state: Sulfate coated-BC can have double the contribution to global warming relative to the uncoated form (Jacobson, 2002) Chater contribution to warming than previously thought chater beneficial impact from BC reduction than previously thought.

Optical absorption properties: these were based on calculations for a spherical BC core/sulfate mantle particle. Observed particles have more complicated shapes.

# Indirect effects: Aerosols Clouds Climate

• Aerosols, by providing more seed particles for cloud condensation, partition cloud water into more droplets of smaller size, resulting in optically thicker and more reflecting clouds (Twomey effect or first indirect effect).

• This same partitioning into smaller drops leads to a decrease in precipitation resulting in longer cloud lifetimes and greater cloud cover (second indirect effect). *Clouds tend to persist longer under polluted conditions than in the marine environment.* 

• However, the issue is further complicated with BC due to absorption: By warming cloudy air, BC can result in enhanced evaporation resulting in dissipation or partial dissipation of clouds (semi-direct effect).

## ABSORBING PHENOMENA



Aircraft photo taken during INDOEX 1999 over the tropical Indian Ocean shows small cumulus clouds embedded in an absorbing aerosol layer. Absorption of solar radiation by the aerosol heats the air surrounding the clouds, causing them to evaporate. (From Schwartz and Buseck, Science 288, 989 (2000)).

## IMPACT ASSESSMENT CONSIDERATIONS



10-nm soot carbon spherules can be found embedded within much larger sulfate particles. *Figure from Buseck and Posfai (1999).* 

The inhomogeneity in properties and geographical distributions of BC aerosols make it difficult to characterize their influences on climate and to represent these influences in models.

Our approach: Use a statistically-based representation of generallymixed aerosols, and their physical and optical properties, suitable for use in regional to hemispheric scale atmospheric models.

## NEW DEVELOPMENTS IN THE MODELING OF GENERALLY MIXED PARTICLE POPULATIONS USING THE METHOD OF MOMENTS

# MOMENTS OF THE PARTICLE SIZE DISTRIBUTION

$$\mu_k \equiv \int_0^\infty r^k (\frac{dN}{dr}) dr \quad (\text{not normalized})$$

Moment	Physical Interpretation	Unit
$\mu_0$	Particle number concentration	cm <sup>-3</sup>
$\mu_1$	Total radius per unit volume	cm cm <sup>-3</sup>
$\mu_2$	$(4\pi)^{-1}$ × Area per unit volume	$\rm cm^2 cm^{-3}$
$\mu_3$	$(\frac{4\pi}{3})^{-1}$ × Volume per unit volume	cm <sup>3</sup> cm <sup>-3</sup>

The *quadrature* method of moments evaluates the moment evolution equation

$$\frac{d}{dt}\mu_k = k \int_0^\infty r^{k-1} \phi(r) f(r) dr$$

by Gaussian quadratures:

$$\frac{d}{dt}\mu_k \cong k \sum_{i=1}^3 r_i^{k-1} \phi(r_i) w_i$$



This approach is *completely general and highly accurate*.

R. McGraw, Aerosol Sci. Technol. (1997)

*Host model*: MAQSIP - Multiscale Air Quality Simulation Platform prototype of US EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System

*Model domain*: 72 by 74 square, 36-km grid cells, 22 layers, sigma coordinate, from the surface to ~160 hPa.

*Meteorological driver*: MM5 meteorological model.

Emissions: 1990 EPA National Emissions Trends (NET90) Inventory.

*Chemistry*: SO<sub>2</sub> oxidation by OH in air and oxidation by H<sub>2</sub>O<sub>2</sub> in cloud droplets.

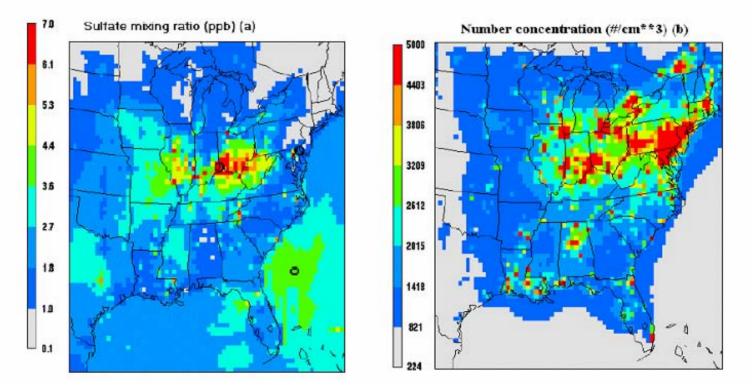
*Clouds*: Sub-grid convective (precipitating and non-precipitating) and grid-scale resolved.

Particle activation, wet and dry deposition: Dependent on particle size.

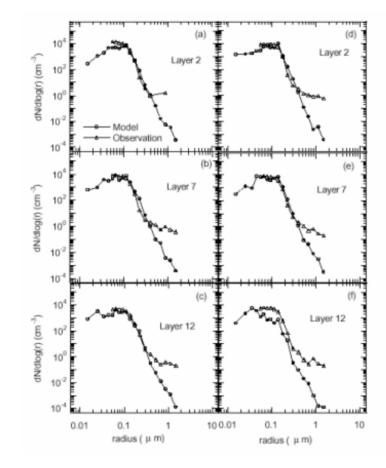
#### MOMENT-BASED SIMULATION OF MICROPHYSICAL PROPERTIES OF SULFATE AEROSOLS IN THE EASTERN UNITED STATES

Shaocai Yu, Prasad S. Kasibhatla, Douglas L. Wright, Stephen E. Schwartz, Robert McGraw, Aijun Deng, J. Geophysical Res. <u>108</u>, D12, 4353 (2003).

Distributions of mean sulfate mixing ratios and number concentrations.

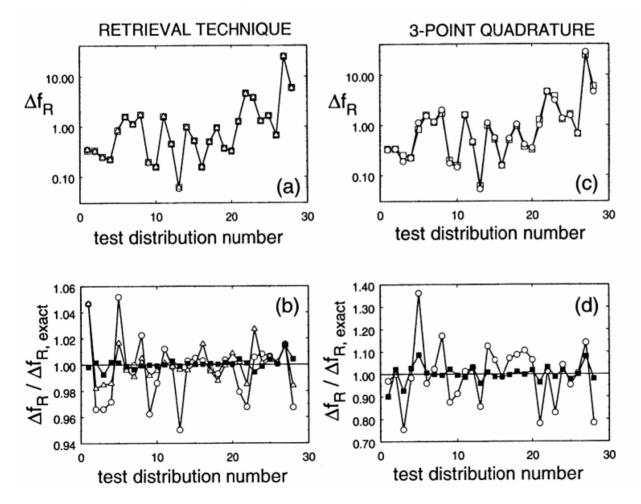


## PREDICTED AEROSOL SIZE DISTRIBUTIONS FROM MOMENTS AND COMPARISON WITH OBSERVATIONS



## **OPTICAL PROPERTIES**

# Results, illustrated here for forcing and 28 test distribution, obtainable directly from six moments

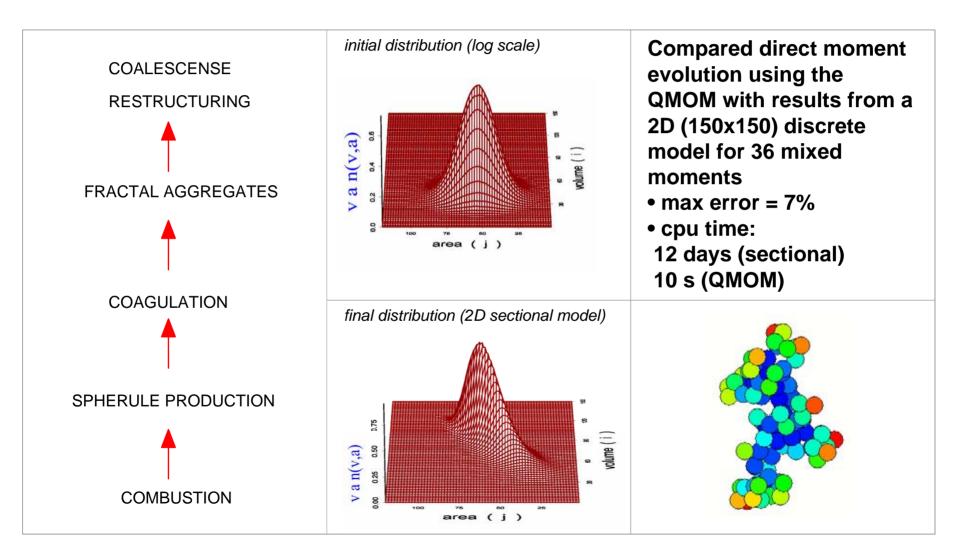


Squares: exact results calculated from size distribution

Circles: Results obtained from six radial moments

#### BIVARIATE EXTENSION OF THE QMOM FOR MODELING SIMULTANEOUS COAGULATION AND SINTERING OF PARTICLE POPULATIONS

#### Wright, McGraw, and Rosner, J. Coll. Interface Sci. 236, 242 (2001)



## NEW DEVELOPMENTS IN THE MODELING OF GENERALLY MIXED AEROSOLS



Aerosol Science 35 (2004) 561-576

Journal of Aerosol Science

www.elsevier.com/locate/jaerosci

# Representation of generally mixed multivariate aerosols by the quadrature method of moments: I. Statistical foundation

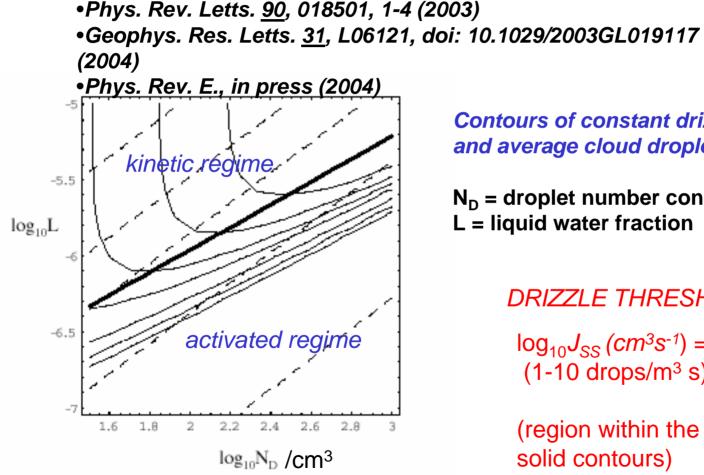
Choongseok Yoon<sup>a,b,1</sup>, Robert McGraw<sup>a,\*</sup>

<sup>a</sup>Atmospheric Sciences Division, Brookhaven National Laboratory, Upton, NY 11973, USA <sup>b</sup>Department of Applied Mathematics and Statistics, State University of New York, Stony Brook, NY 11794-3600, USA

Aerosol Science 35 (2004) 577-598

Representation of generally mixed multivariate aerosols by the quadrature method of moments: II. Aerosol dynamics

### New model for drizzle formation, R. McGraw and Y. Liu



Contours of constant drizzle rate (solid) and average cloud droplet radius (dashed)

 $N_{\rm D}$  = droplet number concentration L = liquid water fraction

## DRIZZLE THRESHOLD

 $\log_{10} J_{SS} (cm^3 s^{-1}) = -6 \text{ to } -5$  $(1-10 \text{ drops/m}^3 \text{ s})$ 

(region within the two lowest solid contours)

• Simulation of BC aerosol and cloud processes using a multi-scale (regional to hemispheric scale) chemical transport model.

• These models will include the latest inventories for diesel BC emissions and projected emissions reduction achievable through clean diesel technology. The objective will be to quantify the favorable climate impact now achievable through BC particulate emissions reduction