REPRESENTING AEROSOL DYNAMICS AND PROPERTIES IN ATMOSPHERIC CHEMICAL TRANSPORT MODELS BY THE METHOD OF MOMENTS

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Upton, New York, U.S.A.

Symposium on Environmental Chemistry in Multiphasic Systems in honor of

Michael R. Hoffmann

ACS Awardee for Creative Advances in Environmental Science and Technology

American Chemical Society 221st National Meeting

Division of Environmental Chemistry

San Diego CA April 1-5, 2001

OUTLINE

- Introduction: Importance of atmospheric aerosols, key properties
- Moments of the aerosol particle size distribution
- Obtaining aerosol properties from the moments
- The Method of Moments (MOM) and the ***Quadrature*** Method of Moments (QMOM) for calculating aerosol evolution
- Evaluations of QMOM
- Application of QMOM in a large scale chemical transport model
- Summary

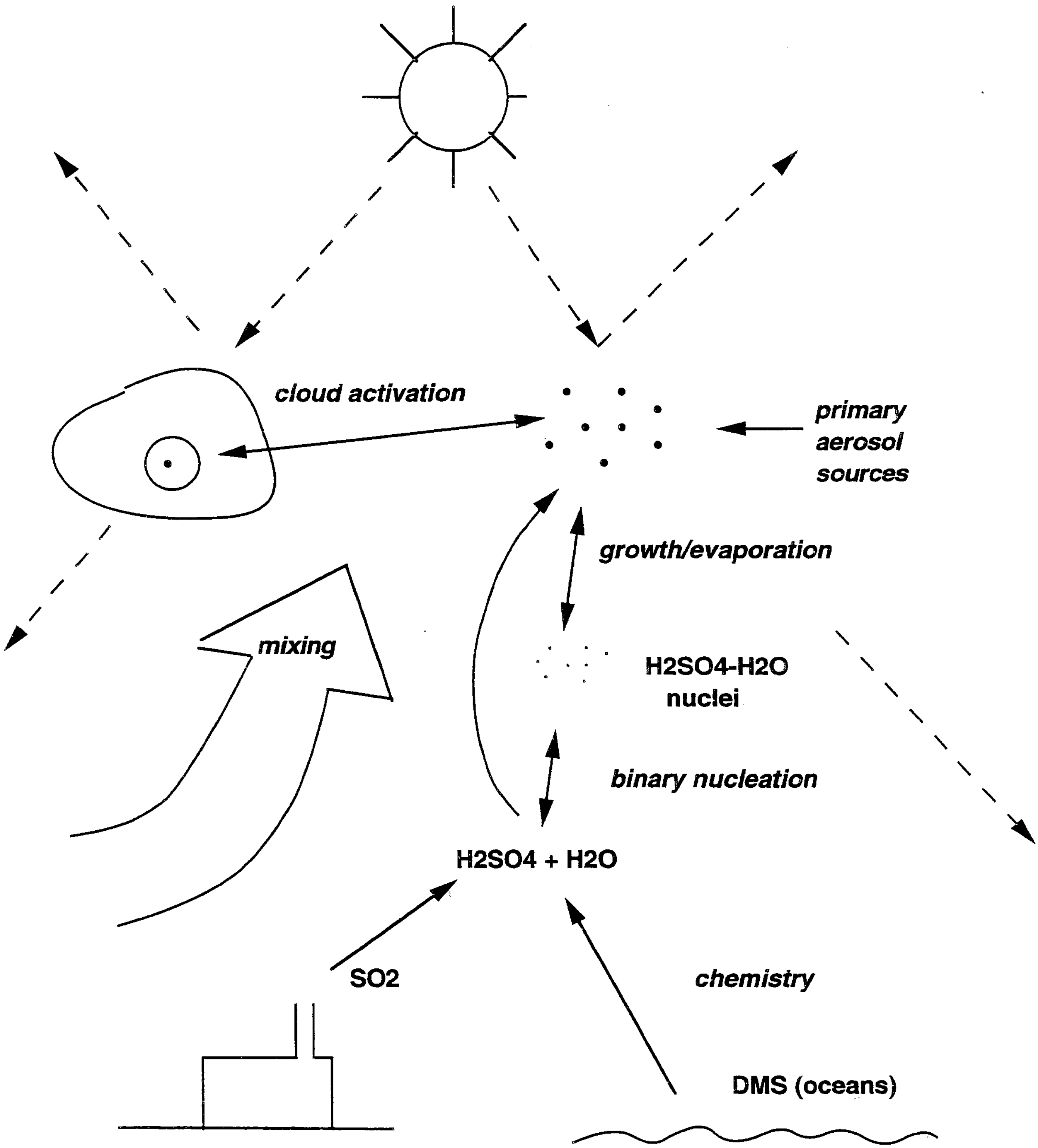
IMPORTANCE OF ATMOSPHERIC AEROSOLS

- Human health - impairment via inhalation
- Light scattering and absorption - visibility, climate change
- Heterogeneous reactions - stratosphere, troposphere
- Modification of cloud physical properties - hydrology and climate
- Modification of fog, cloud, and precipitation composition

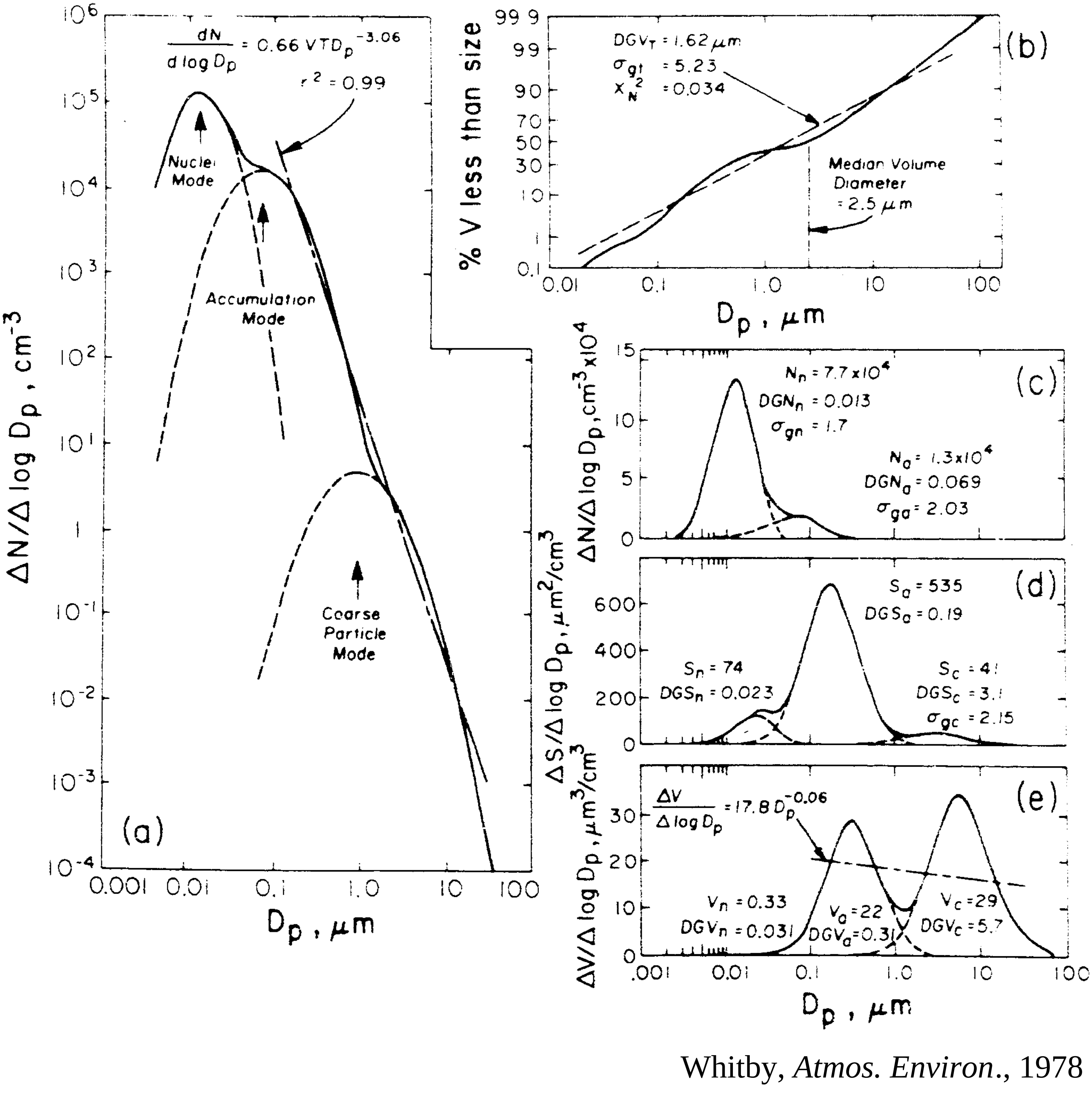
Understanding and describing the role of aerosols in these phenomena require ***size- and composition-dependent treatment*** of chemical and physical processes involving aerosols.

This descriptive capability must be represented in atmospheric transport and transformation models on a variety of scales from regional to global.

NUCLEATION AND GROWTH PROCESSES OF ATMOSPHERIC AEROSOLS AND CLOUDS



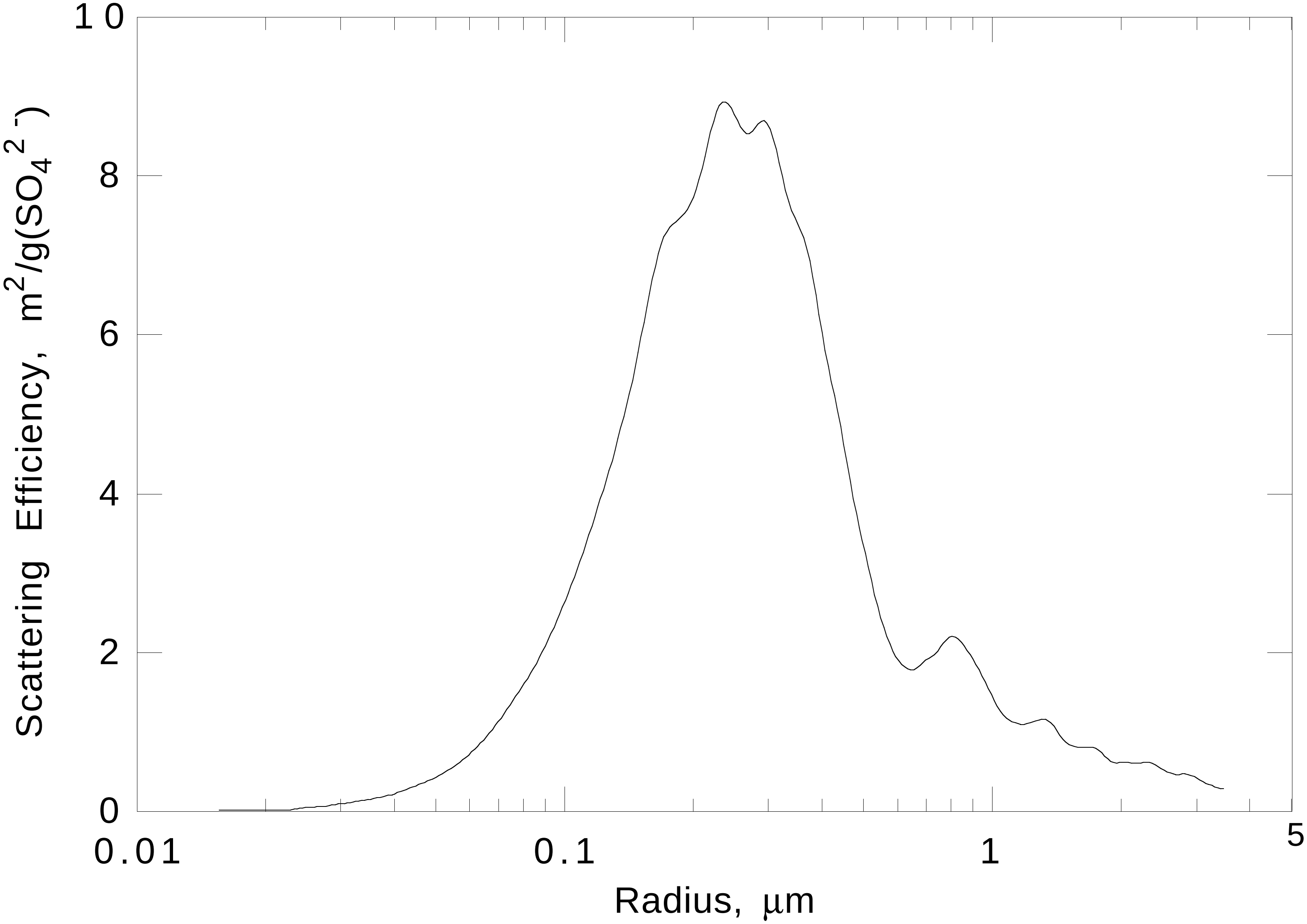
REPRESENTATIONS OF AEROSOL SIZE DISTRIBUTION



Whitby, *Atmos. Environ.*, 1978

SIZE MATTERS

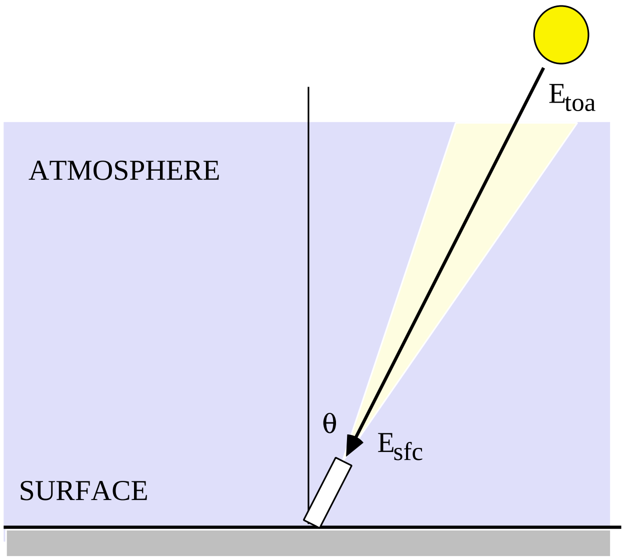
Light scattering efficiency of ammonium sulfate vs. radius



Data of Ouimette and Flagan, *Atmos. Environ.*, 1982

KEY AEROSOL OPTICAL PROPERTIES

Aerosol optical thickness, τ_{aerosol}



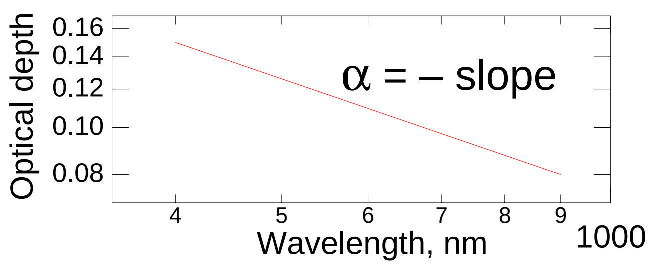
$$\frac{E_{\mathrm{sfc}}}{E_{\mathrm{toa}}} = \exp(-\tau_{\mathrm{meas}} / \cos\theta)$$

$$\tau_{\mathrm{aerosol}} = \tau_{\mathrm{meas}} - (\tau_{\mathrm{Rayleigh}} + \tau_{\mathrm{gas\ abs}})$$

τ_{aerosol} depends on aerosol loading and properties.

Ångström exponent, α

$$\alpha = -\frac{d \log \tau_{\mathrm{aerosol}}}{d \log wavelength}$$



α is largest for smallest particle size.

Aerosol radiative forcing

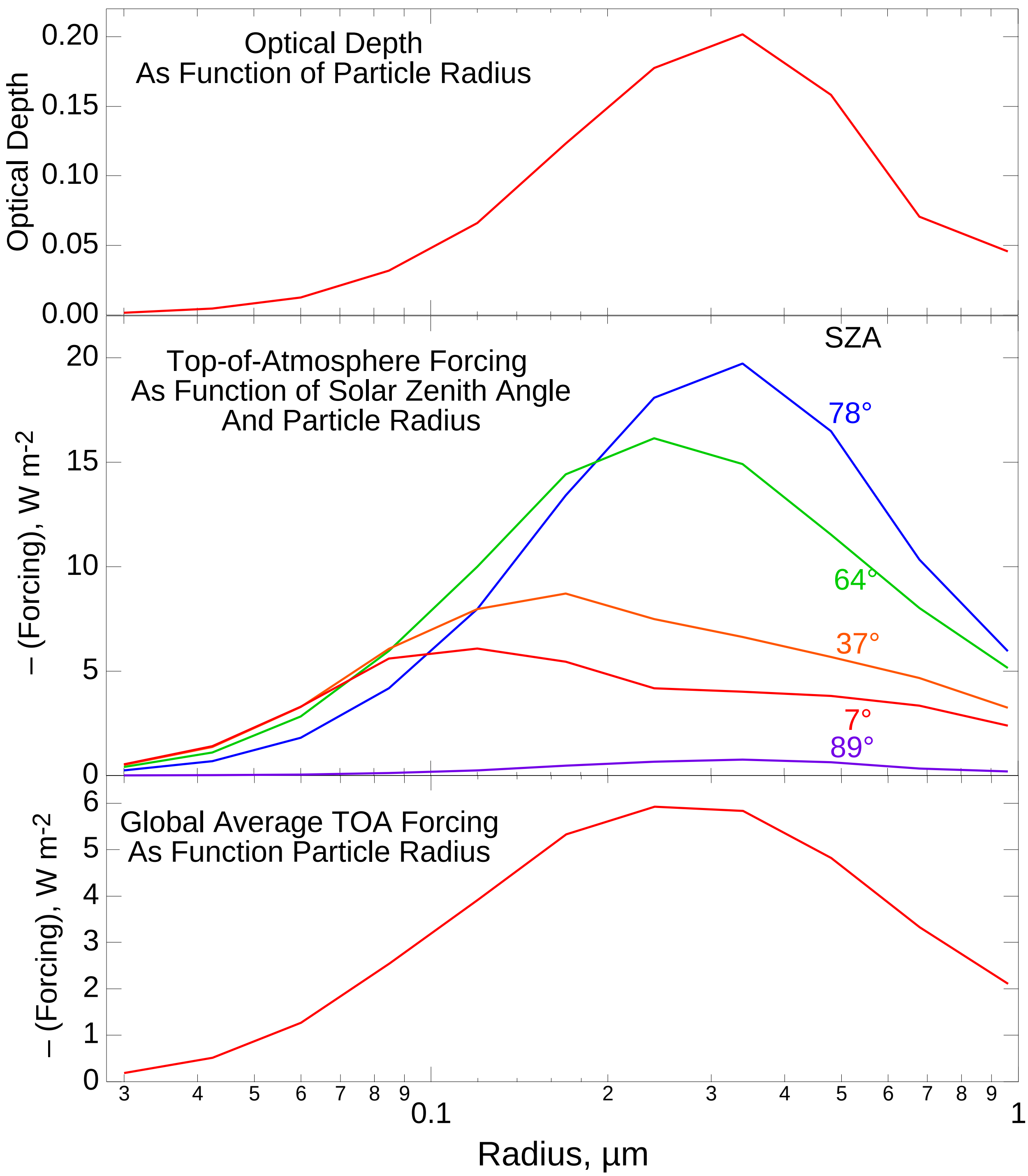
Forcing = Flux(aerosol) - Flux(no aerosol)

Flux may be at top of atmosphere or surface.

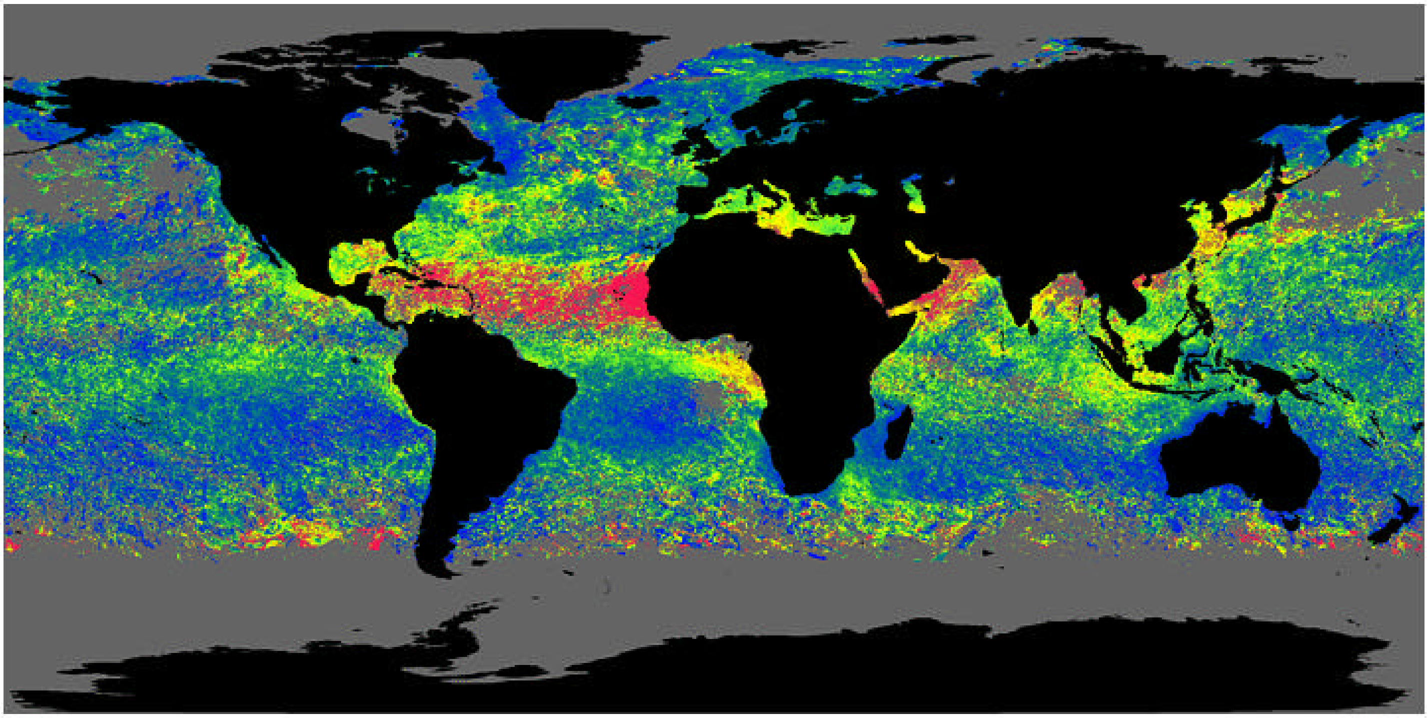
SIZE DEPENDENCE OF AEROSOL OPTICAL DEPTH AND TOA RADIATIVE FORCING

Ammonium Sulfate, 10 mg (sulfate) m^{-2} at 80% RH

Cloud-free Sky, Surface Reflectance 0.15

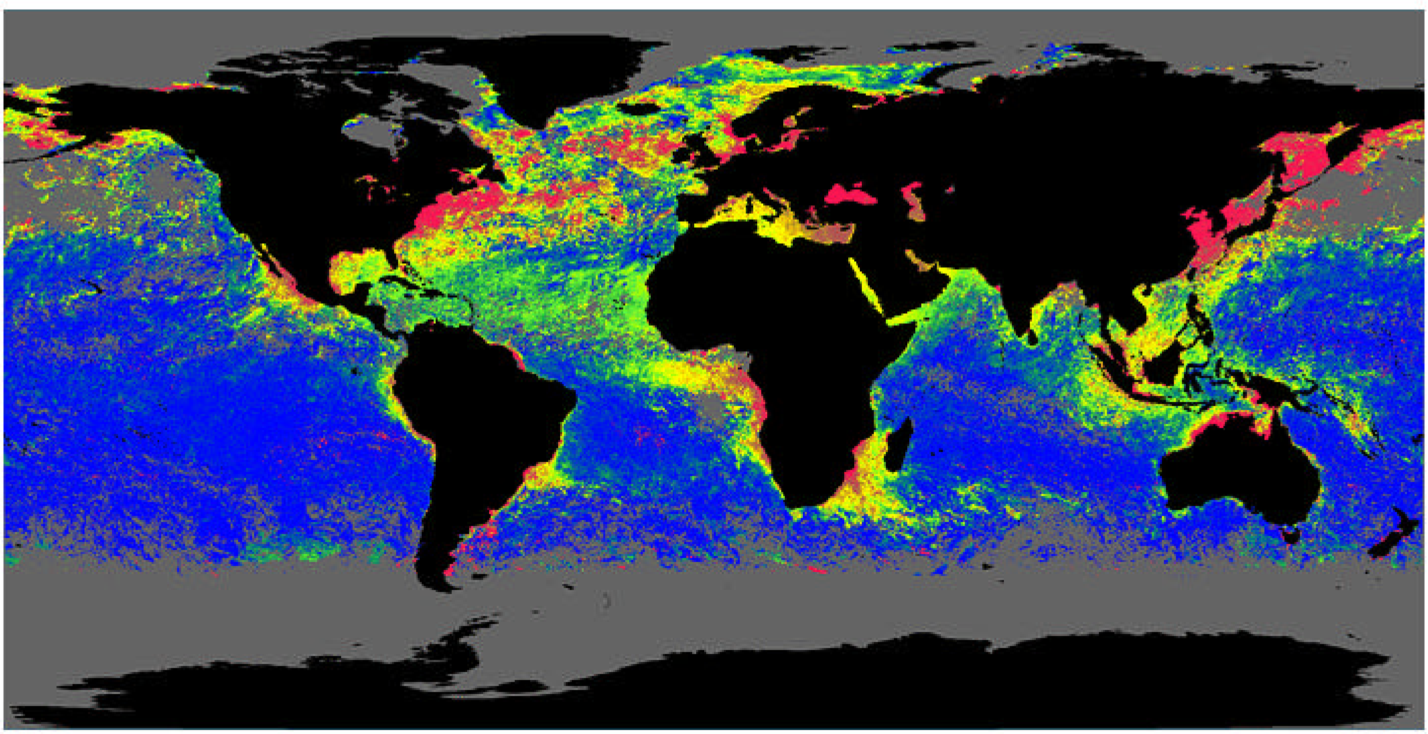


MONTHLY AVERAGE AEROSOL JUNE 1997



Optical Thickness at 865 nm

0.0 0.5



Ångström Exponent

−0.2 1.2

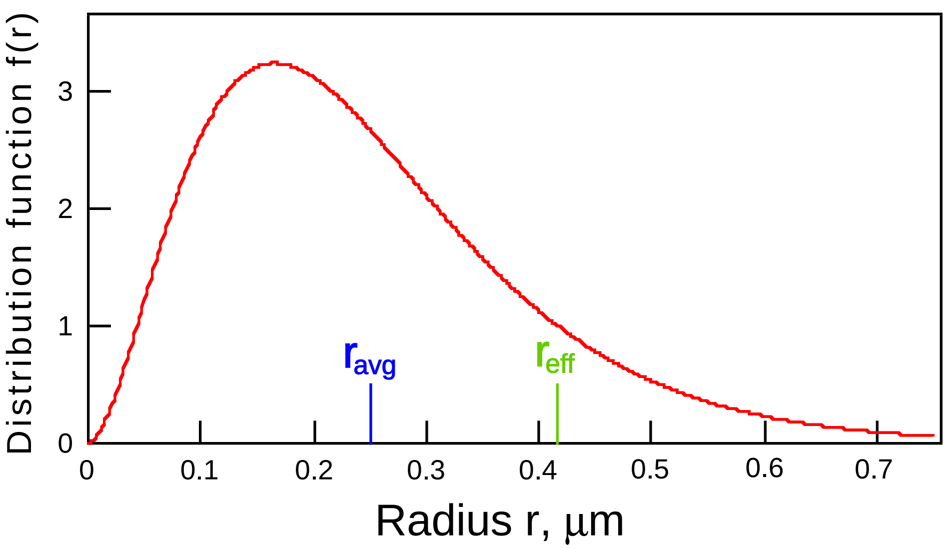
Polder/Adeos CNES/NASDA LOA/LSCE

MOMENTS OF THE PARTICLE SIZE DISTRIBUTION

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

Moment	Physical Interpretation	Unit
μ_0	Particle number concentration	cm^{-3}
μ_1	Total radius per unit volume	$cm\ cm^{-3}$
μ_2	$(4\pi)^{-1} \times$ Area per unit volume	$cm^2\ cm^{-3}$
μ_3	$(\frac{4\pi}{3})^{-1} \times$ Volume per unit volume	$cm^3\ cm^{-3}$

Particle Size Distribution



Average radius: $r_{avg} = \mu_1/\mu_0$.

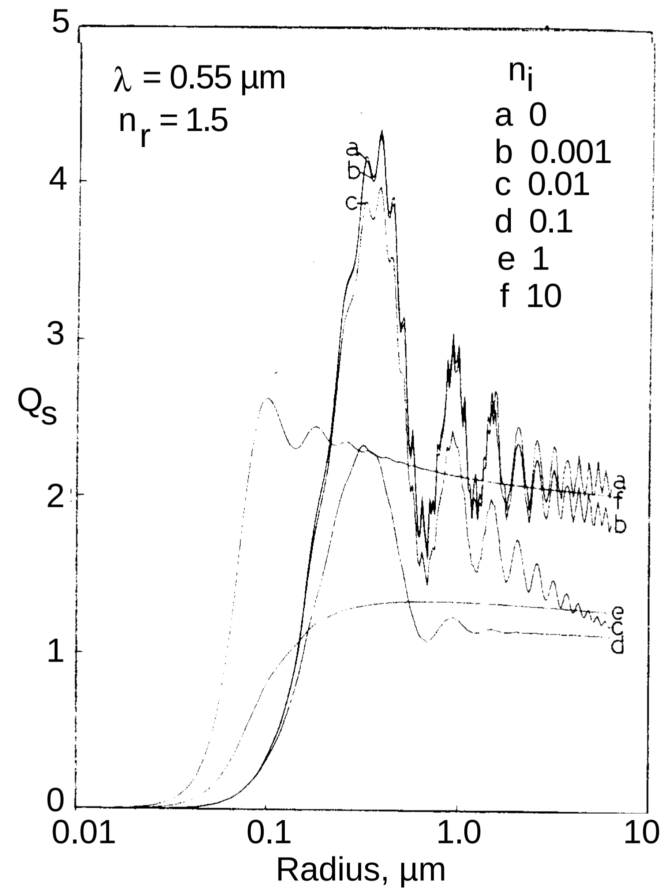
Effective radius: $r_{eff} = \mu_3/\mu_2$.

EVALUATION OF AEROSOL PROPERTIES

Aerosol properties are integrals of kernel function over size distribution

$$P = \int_0^\infty \sigma(r) f(r) dr$$

Property (P)	Kernel (σ)
Particle number density	*1*
Surface area density	r^2
Volume concentration	r^3
Optical properties	*Optical kernels*
scattering	$r^2 \times Q_s$
forcing	
Ångström exponent	
effective radius	μ_3/μ_2
Cloud activation	*Step function*
PM 2.5	$H(r-r_0)$
Concentration at specific radius r_0	*Delta function* $\delta(r-r_0)$



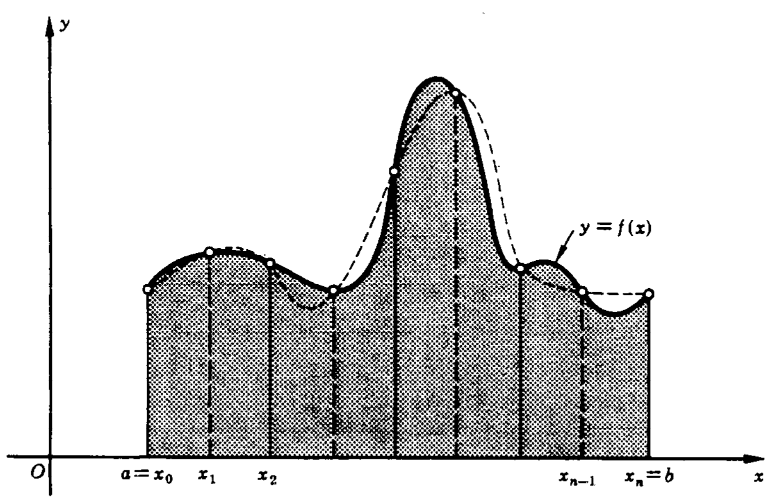
AEROSOL PHYSICAL PROPERTIES
HOW TO OBTAIN FROM MOMENTS?

Aerosol physical or optical properties are an integral over the size distribution, requiring integrals like

$$P = \int_0^\infty \sigma(r) f(r) dr$$

where the kernel function $\sigma(r)$ describes the property of interest.

Most integration (quadrature) methods require that the integrand be known as some set of points, for example Simpson's rule:



In our case $\sigma(r)$ is known, but the distribution function $f(r)$ is not known, only a few of its moments.

CALCULATING AEROSOL PROPERTIES BY GAUSSIAN QUADRATURES

Problem: *How to evaluate integrals over the aerosol size distribution when only the lower-order moments of the distribution are known???*

Solution: *Gaussian quadrature*

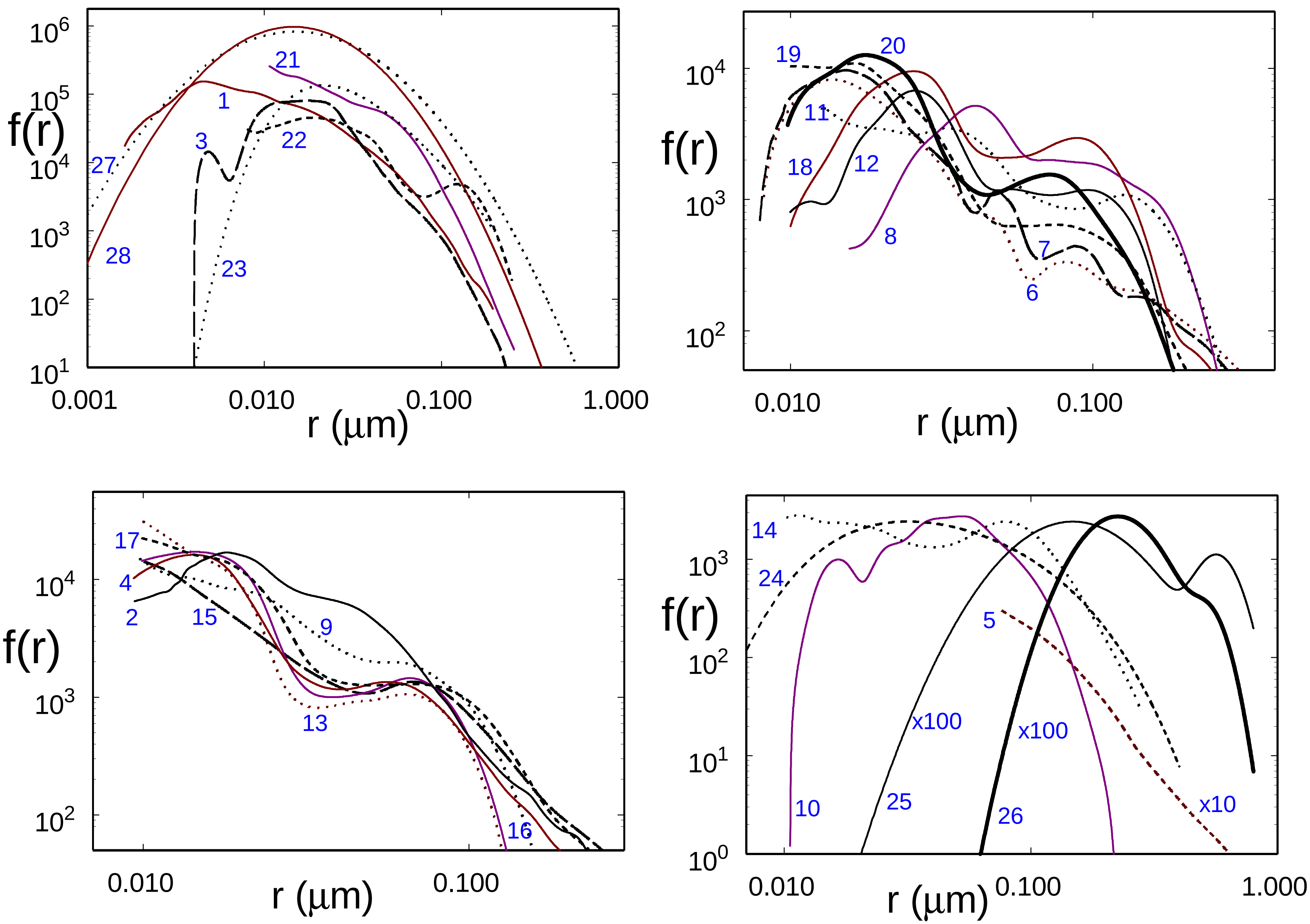
$$\int_0^\infty \sigma(r) f(r) dr \approx \sum_{i=1}^{N} \sigma(r_i) w_i$$

Here $\sigma(r)$ *is the known kernel function and* $f(r)$ *is the unknown size distribution.*

The N abcissas $\{r_i\}$ and N weights $\{w_i\}$ are determined from $2N$ moments of $f(r)$ by inversion of

$$\mu_k \equiv \int_0^\infty r^k f(r) dr = \sum_{i=1}^{N} r_i^k w_i \quad k = 0, 1, \ldots, 2N\text{-}1.$$

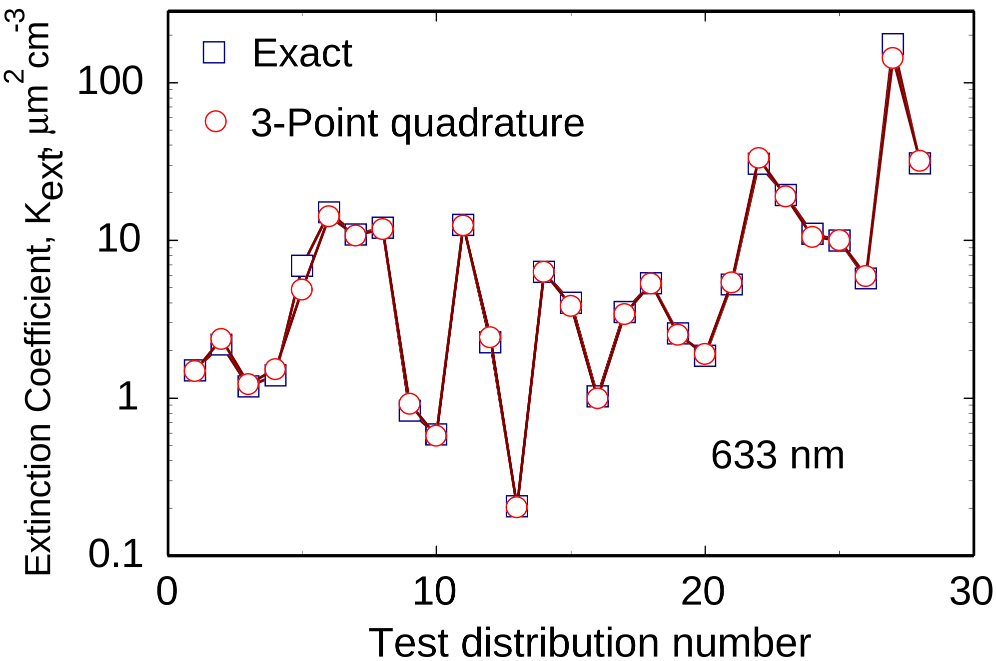
DISTRIBUTIONS USED IN TEST OF RETRIEVAL OF OPTICAL PROPERTIES FROM MOMENTS

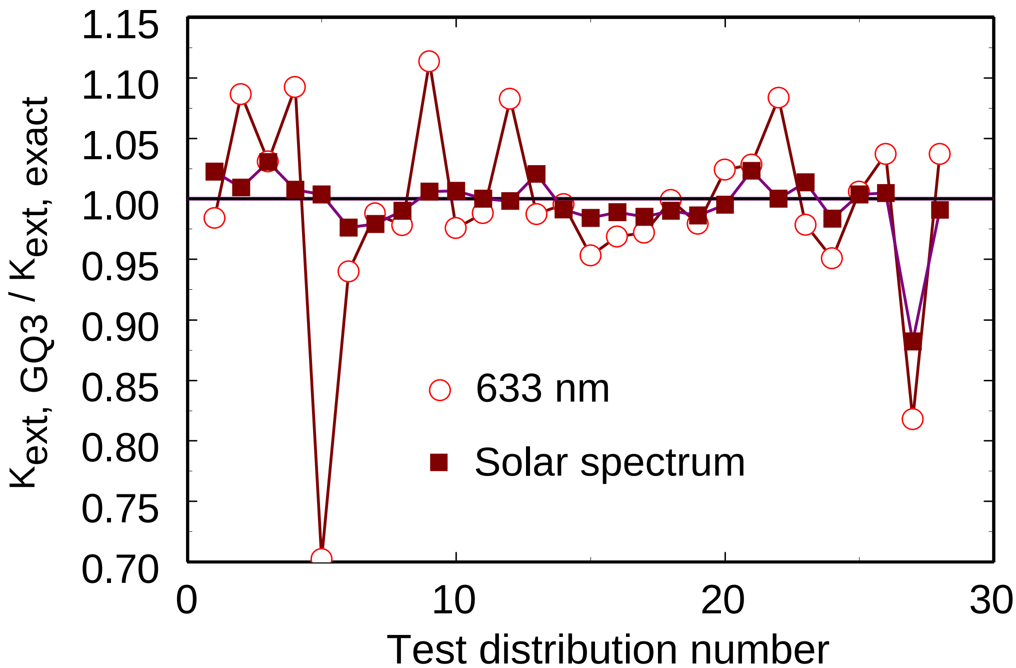


D. L. Wright, Jr., *J. Aerosol Sci.*, 2000

SKILL OF GAUSSIAN QUADRATURES
TO OBTAIN AEROSOL OPTICAL PROPERTIES
FOR 28 TEST DISTRIBUTIONS

Index of refraction *n* = 1.55 - 0*i*

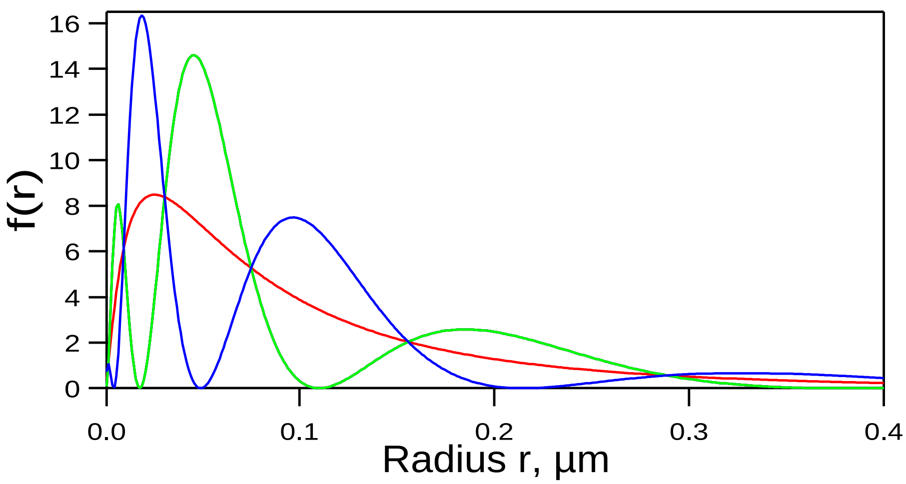




D. L. Wright, Jr., *J. Aerosol Sci.*, 2000

THE MOMENTS DO NOT UNIQUELY DEFINE THE DISTRIBUTION

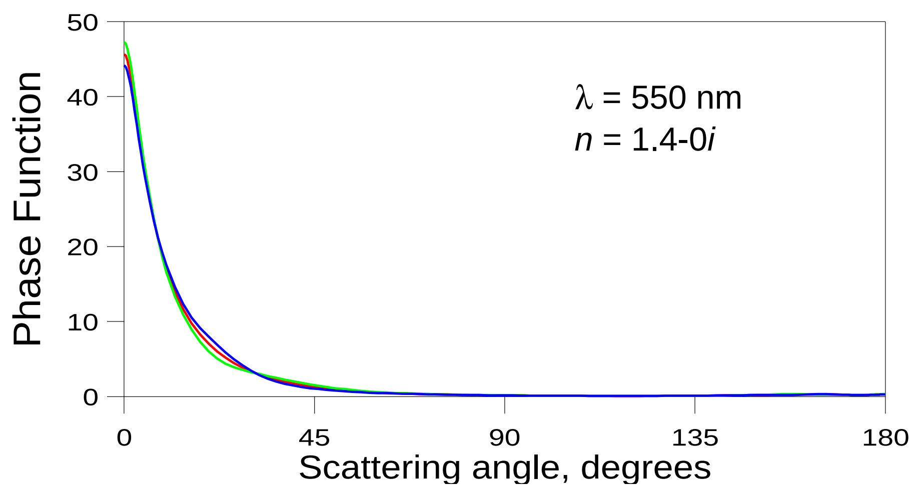
Question: How many identical moments do the following distributions have in common?



Answer: Infinitely many! These distributions have identical moments for all non-negative integer values of *k*.

$$\mu_k \equiv \int_0^\infty r^k f(r) dr$$

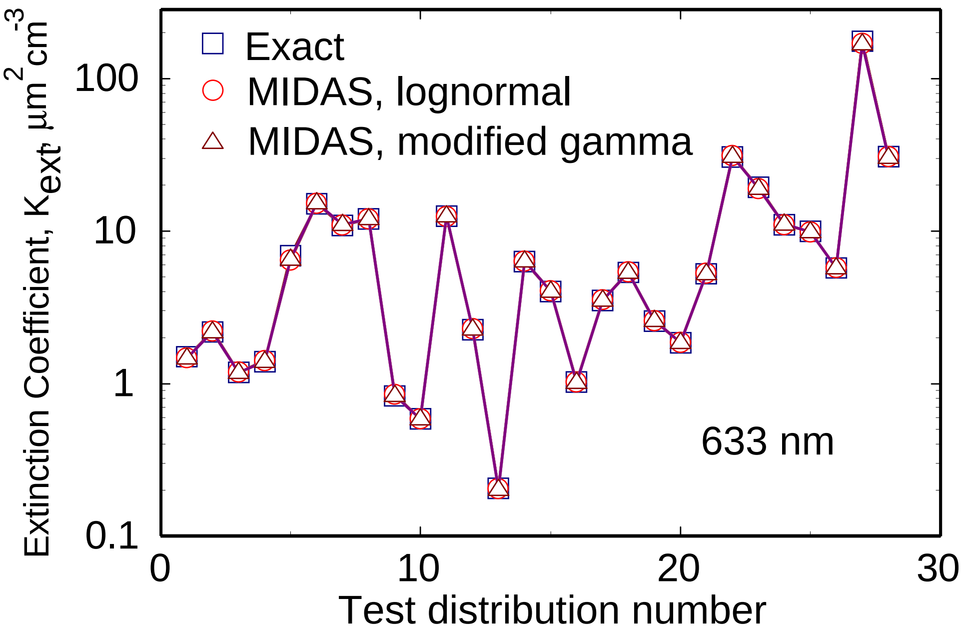
Despite the differences in the distributions, the optical properties are virtually identical!

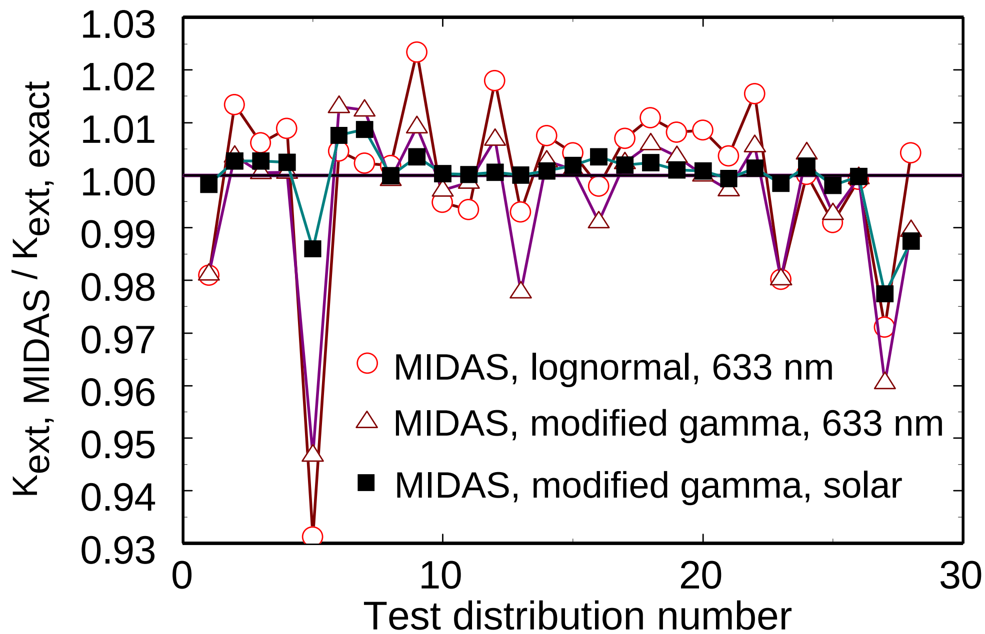


McGraw et al., *J. Aerosol Sci.* (1998)

SKILL OF MIDAS METHOD
(Multi Isomomental Distribution Aerosol Simulator)
TO OBTAIN AEROSOL OPTICAL PROPERTIES
FOR 28 TEST DISTRIBUTIONS

Index of refraction n = 1.55 - 0i





D. L. Wright, Jr., *J. Aerosol Sci.*, 2000

AEROSOL DYNAMICS BY THE METHOD OF MOMENTS

The ***method of moments*** is an approach to describing aerosol properties and dynamics in terms of the moments μ_k of the radial number size distribution $f(r)$.

$$\mu_k = \int_0^\infty r^k f(r) dr$$

Aerosol ***properties*** (e.g., light scattering coefficient) can be accurately represented as simple functions of low order moments.

Aerosol ***dynamics*** can be represented by growth laws (differential equations) in the moments.

The moments advect and mix just like chemical species--they are conserved and additive.

Hence representing aerosol properties and dynamics in 3-D transport models is equivalent to representing a small number of additional chemical species -- the low order moments.

METHOD OF MOMENTS
Heuristic Description

Consider accretion of monomer by existing aerosol.

This can be considered a ***reaction*** between monomer (m) and aerosol surface area (A)

$$m + A \rightarrow \text{slightly larger distribution}$$

$$\text{Rate} = kmA$$

Aerosol surface area density is

$$A = \int_0^\infty 4\pi r^2 f(r)dr = 4\pi\mu_2$$

So accretion of monomer by existing aerosol is a ***reaction*** between monomer and second moment.

METHOD OF MOMENTS
General Description

Consider the growth law for particles of radius *r*:

$$\frac{dr}{dt}\left(r \right) = \phi(r)$$

The corresponding ***moment evolution equation*** is:

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

Closure of the moment evolution equations requires the growth law to be of the form:

$$\phi(r) = a + br$$

where *a* and *b* are independent of *r*. In this case moment evolution is evaluated as:

$$\begin{aligned}\frac{1}{k}\frac{d}{dt}\mu_k &= \int r^{k-1}\phi(r)f(r)dr \\ &= a\int r^{k-1}f(r)dr + b\int r^k f(r)dr \\ &= a\mu_{k-1} + b\mu_k\end{aligned}$$

This case includes free-molecular growth, $\phi(r) = a$.

QUADRATURE METHOD OF MOMENTS

The requirement that the growth law be of the form

$$\phi(r) = a + br$$

is not generally satisfied.

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

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

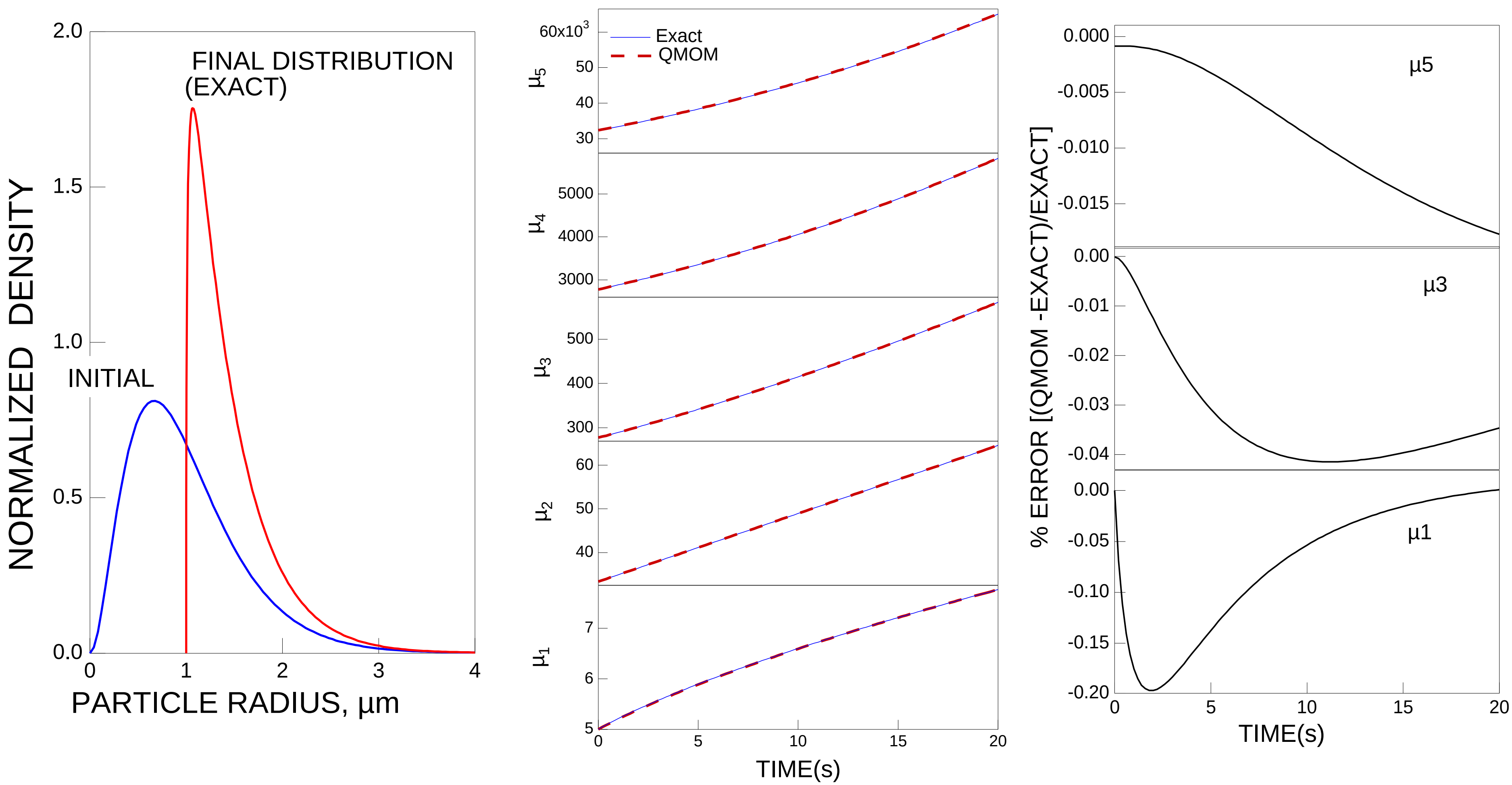
by Gaussian quadratures:

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

This approach is completely general and highly accurate.

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

COMPARISON OF EXACT AND QUADRATURE MOMENT EVOLUTION



Initial size distribution: Normalized Khrgian-Mazin distribution with mean particle radius of 5 µm.

Diffusion controlled growth by accretion of water vapor for 20 s at 278 K and fixed saturation of 101%.

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

CONDITIONS OF TESTS FOR SKILL OF QUADRATURE METHOD OF MOMENTS

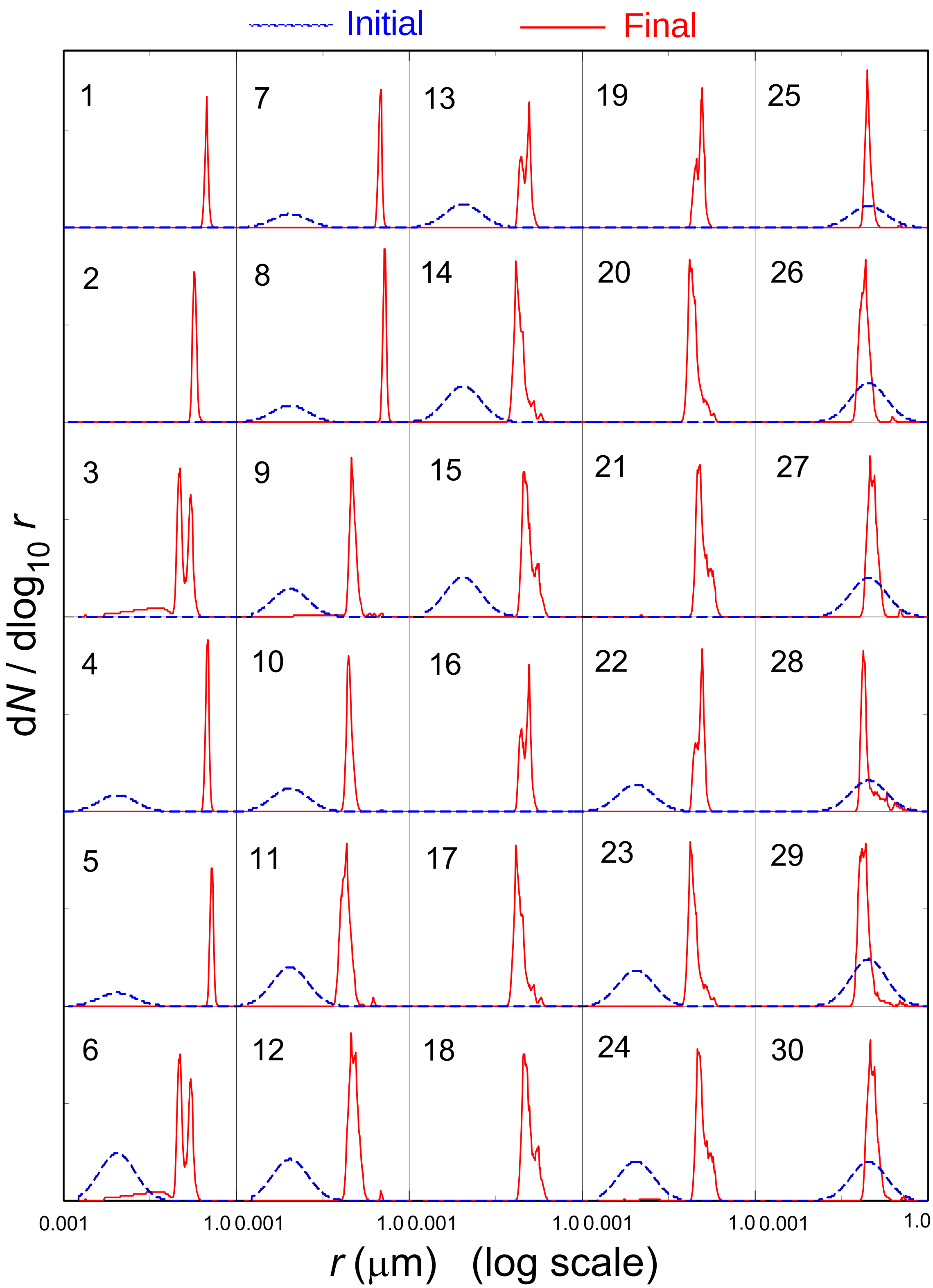
QMOM was extensively compared in box-model calculations to results obtained by discrete integration of the PSD.

Test case number	Dry deposition	Cloud type	Initial aerosol			Initial concentrations	
			N_0 cm^{-3}	r_g µm	σ_g	$[H_2SO_4]_0$ mol cm^{-3}	$[SO_2]_0$ mol cm^{-3}
1-3	none	Cumulus	0	0	0	0	0
4-6	none	Cumulus	100	0.01	2.0	0	0
7-9	none	Stratiform	100	0.01	2.0	0	0
10-12	none	Stratiform	100	0.01	2.0	5.0×10^{15}	1.0×10^{12}
13-15	none	Cumulus	100	0.01	2.0	5.0×10^{15}	1.0×10^{12}
16-18	none	Cumulus	0	0	0	5.0×10^{15}	1.0×10^{12}
19-21	to land (W = 5.0 m/s)	Cumulus	0	0	0	5.0×10^{15}	1.0×10^{12}
22-24	to ocean (W = 10 m/s)	Stratiform	100	0.01	2.0	5.0×10^{15}	1.0×10^{12}
25-27	none	Stratiform	100	0.10	2.0	5.0×10^{15}	1.0×10^{12}
28-30	none	Stratiform	1000	0.10	2.0	5.0×10^{15}	1.0×10^{12}

Each aerosol test case was run for three sets of meteorological conditions.

Wright, Kasibhatla, McGraw and Schwartz, *JGR*, in press, 2001

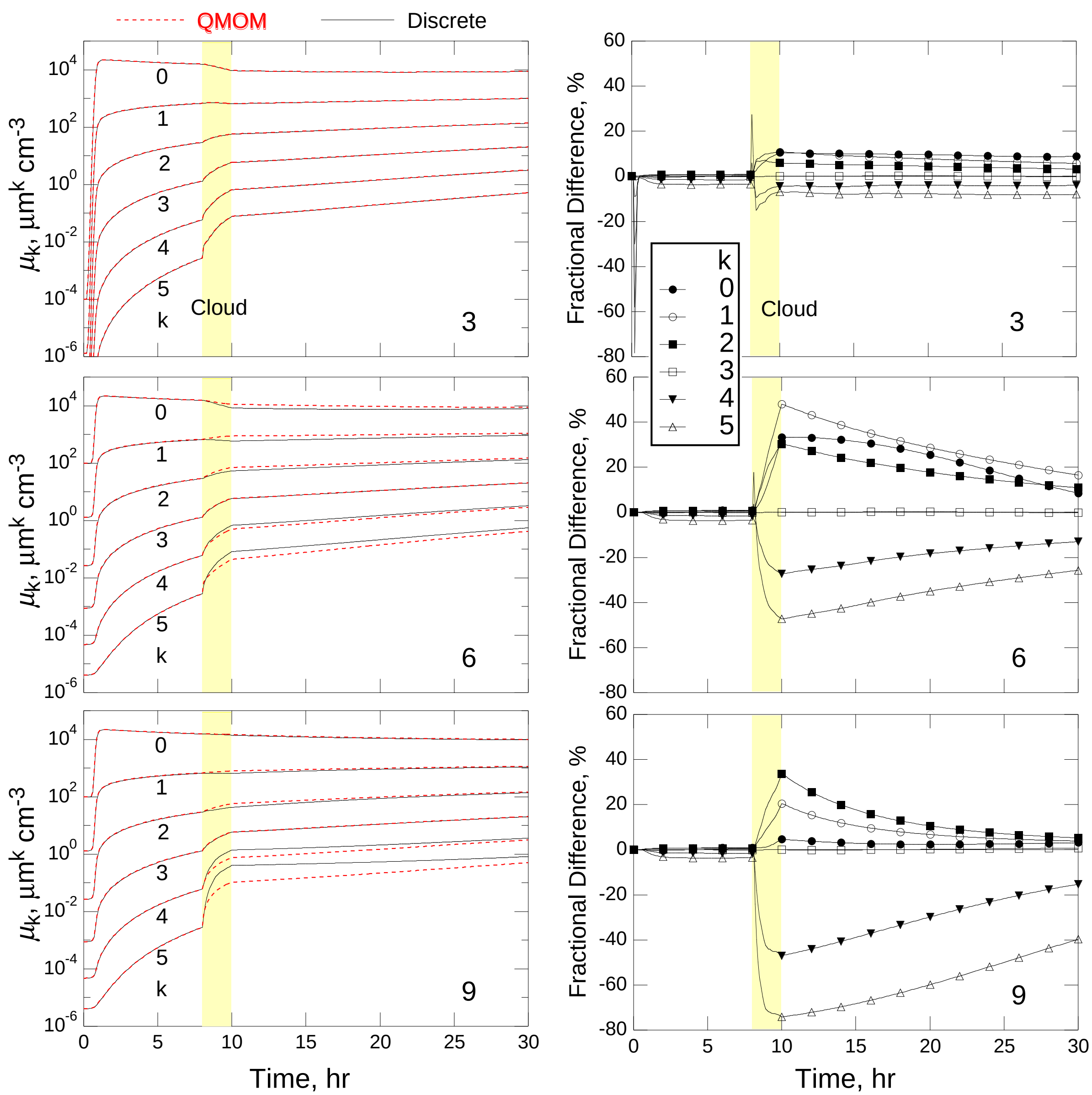
DISTRIBUTIONS IN TESTS OF SKILL OF QUADRATURE METHOD OF MOMENTS



Wright, Kasibhatla, McGraw and Schwartz, *JGR*, in press, 2001

MOMENT EVOLUTION AND ERRORS IN BOX MODEL TESTS OF SKILL OF QUADRATURE METHOD OF MOMENTS

Comparison of QMOM *vs*. discrete integration of PSD

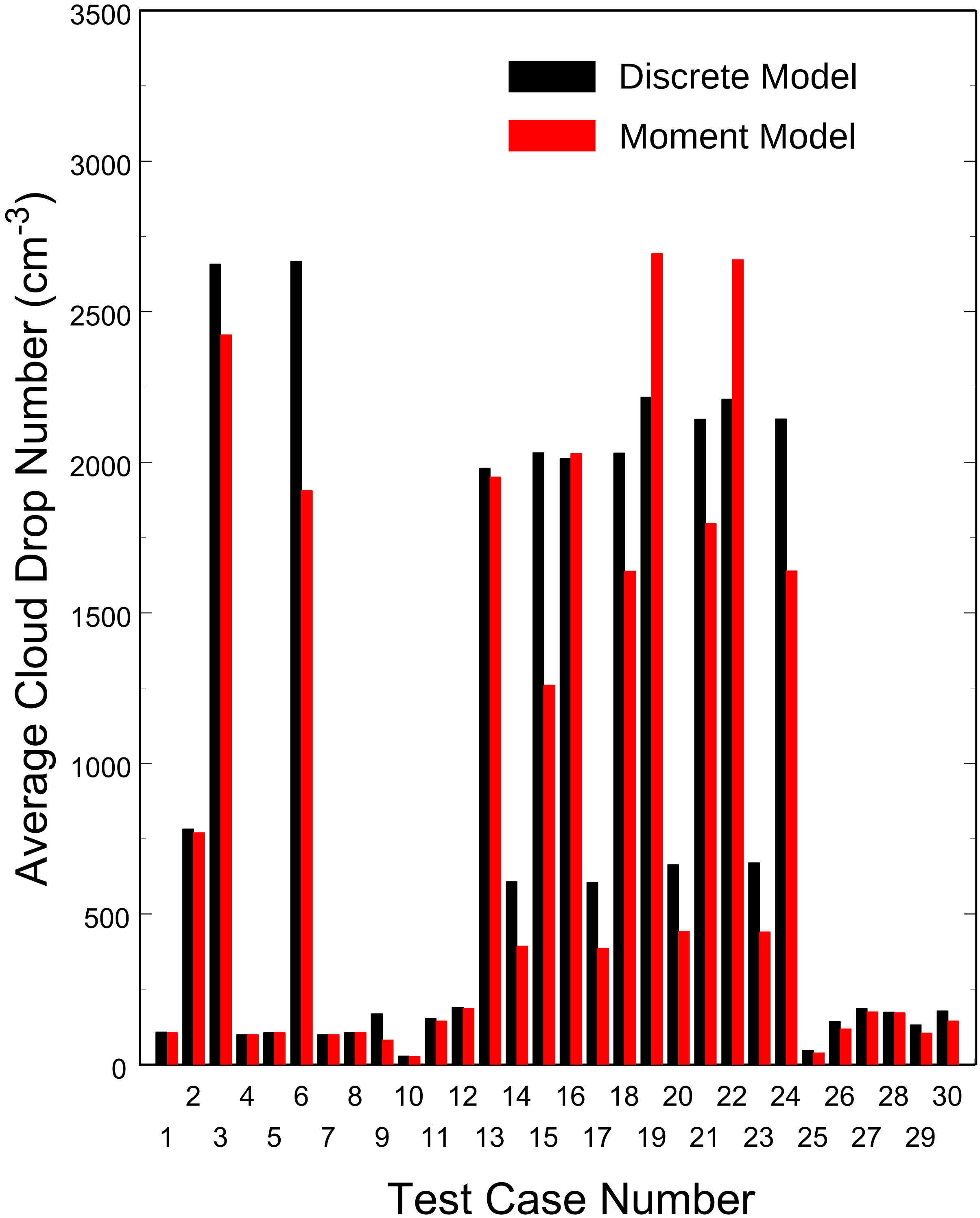


Departures are associated mainly with cloud events.

Some of the departures are attributable to errors in discrete integration.

Wright, Kasibhatla, McGraw and Schwartz, *JGR*, in press, 2001

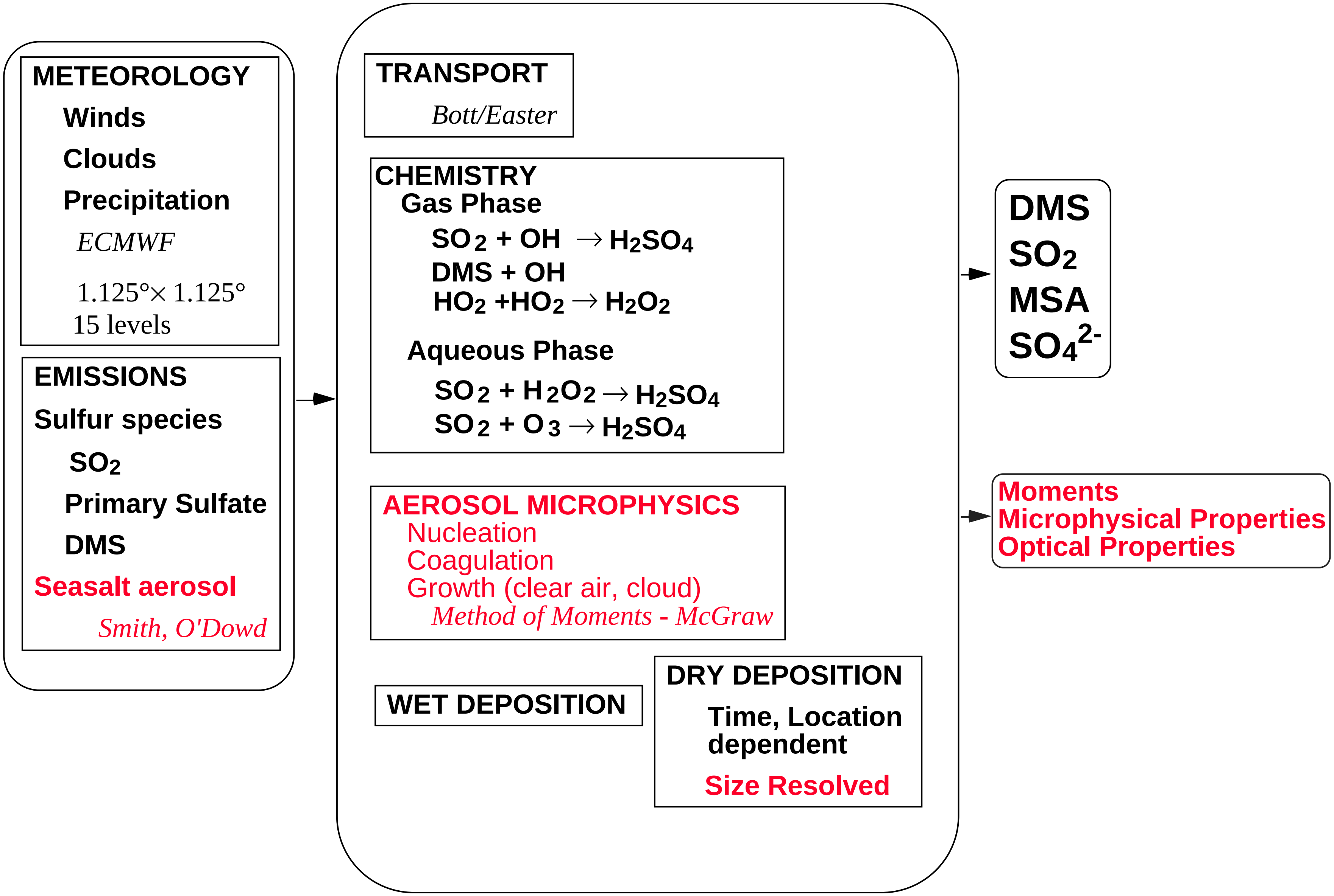
COMPARISON OF CLOUD DROPLET NUMBER CONCENTRATION BETWEEN MOMENT AND DISCRETE MODELS



Wright, Kasibhatla, McGraw and Schwartz, *JGR*, in press, 2001

Aerosol Chemical Transport Model GChM-O

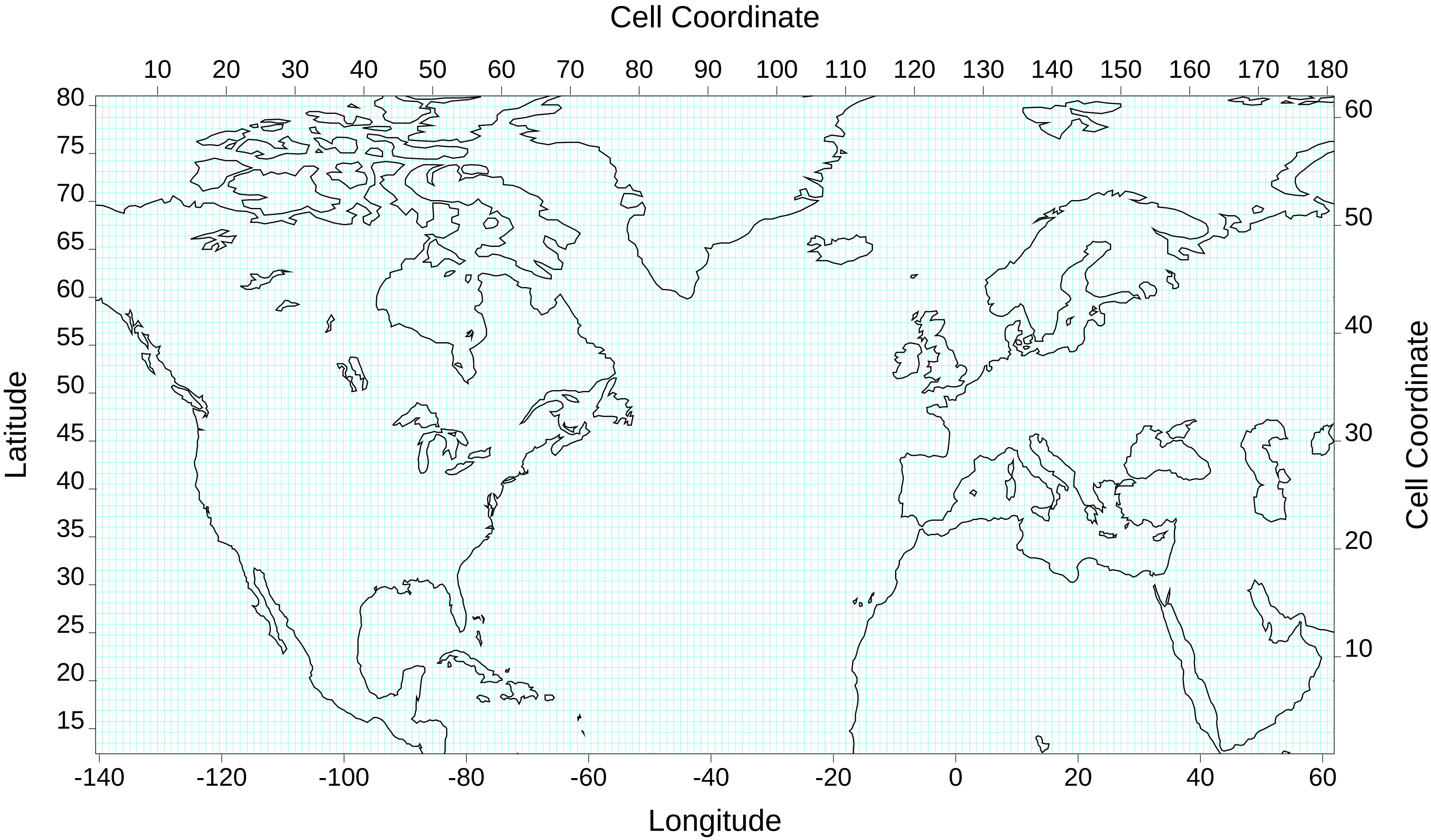
Global Chemistry Model Driven by Observation-Derived Meteorological Data



MODEL DOMAIN AND GRID CELL STRUCTURE

1.125° latitude and longitude; 15 terrain-following vertical levels

$61 \times 180 \times 15 = 164{,}700$ grid cells



MODELING EVOLUTION OF AEROSOL LOADING AND PROPERTIES

Moment-based representation of aerosol microphysics

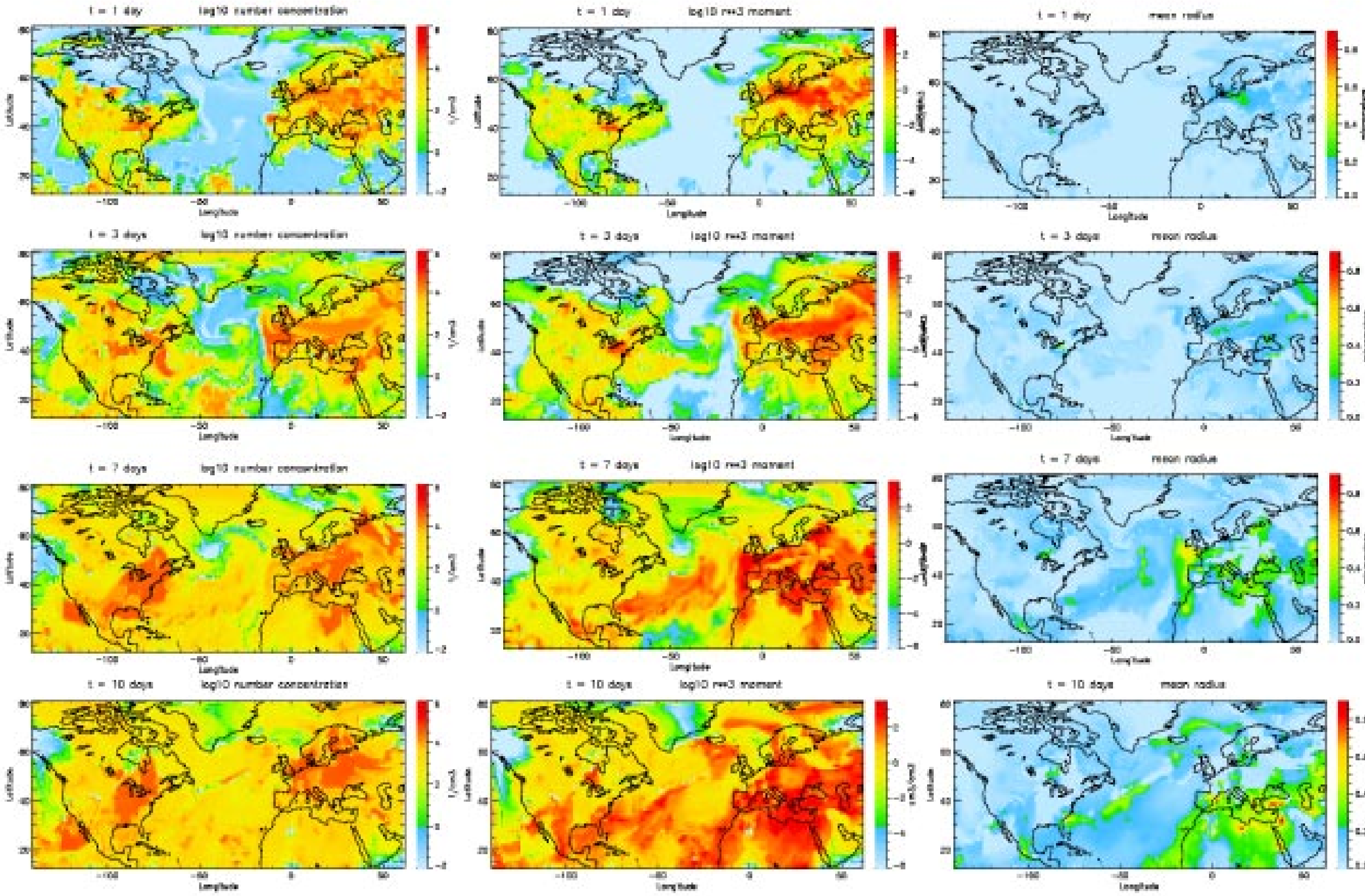
Day $\quad$ $\log_{10} N$ $\quad$ $\log_{10} \mu_3$ $\quad$ Mean Radius

1

3

7

10



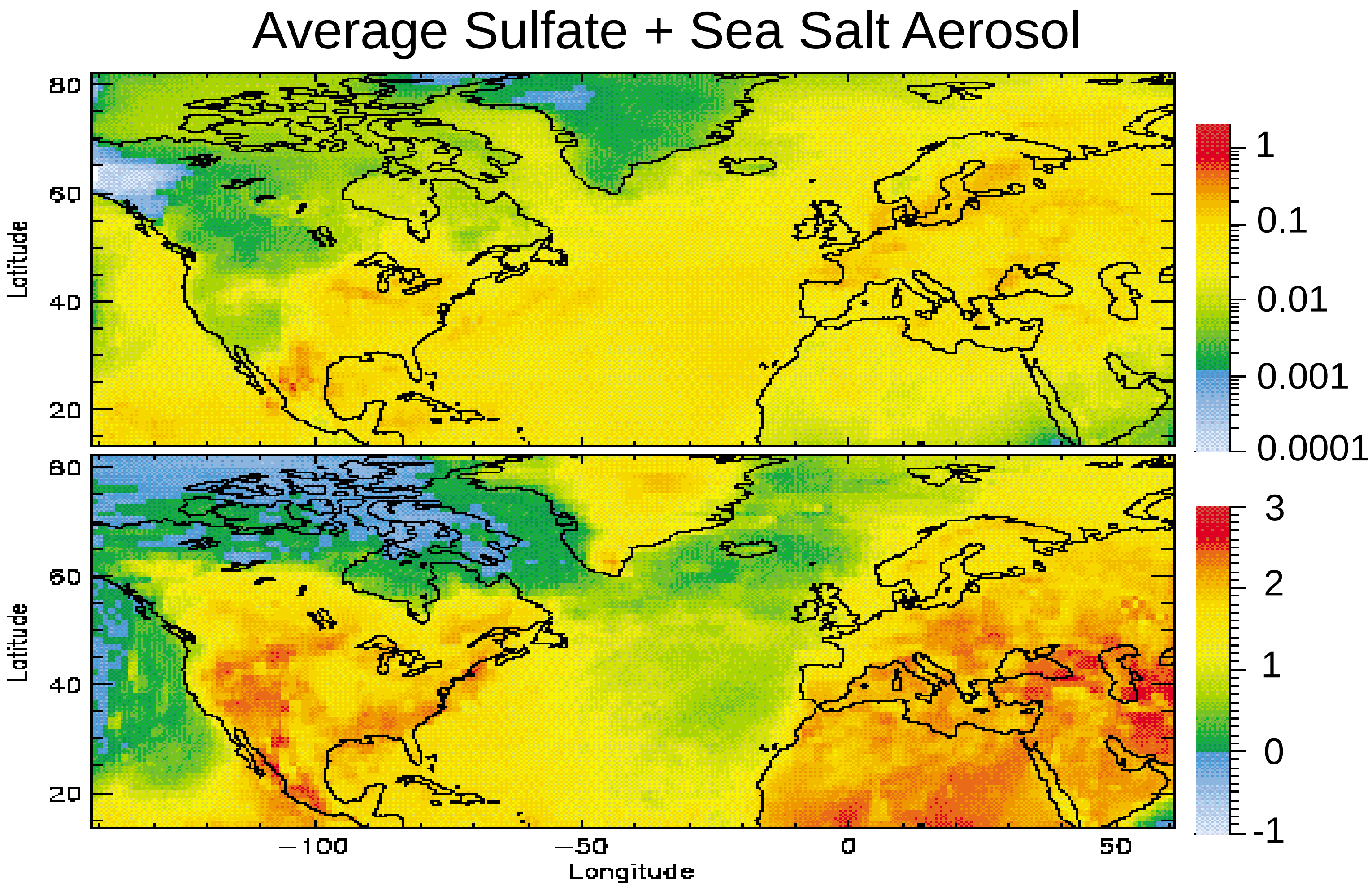
Based on Wright, McGraw, Benkovitz, and Schwartz, *GRL*, 2000

AEROSOL PROPERTIES FROM SUBHEMISPHERIC SULFATE TRANSPORT AND TRANSFORMATION MODEL

835 nm   October 14-31, 1986

Aerosol optical depth

Ångström exponent



APPROACHES TO REPRESENTING AEROSOL SIZE DISTRIBUTION AND ITS EVOLUTION IN MODELS

	Conventional	Moment Methods
Size distribution represented by...	Number of particles in bins as function of radius $N_i(r_i)$	Moments, $\mu_k = \int r^k N(r) dr$
Aerosol evolution represented by...	Differential equations in $N_i(r_i)$	Differential equations in low-order moments, $k = 0 \ldots 5$
Aerosol properties represented by...	Direct integration over kernel function, $\sigma = \int \sigma(r) N(r) dr$	Gaussian quadrature, $\sigma = \sum w_i \sigma(r_i)$; radii r_i and weights w_i determined from moments
Advantages	Explicit knowledge of size distribution	Compact representation
Issues	Numerical diffusion between bins; many variables $N_i(r_i)$ required	Closure of the set of moment equations; non-uniqueness of distributions

SUMMARY

There is a need for accurate and efficient representation of aerosol microphysical properties in chemical transport models.

The Method of Moments (MOM) meets this need. It is orders of magnitude more efficient than discrete aerosol models and highly accurate.

MOM does ***not*** provide the particle size distribution, but it tells you (almost) everything else you want to know about the aerosol.

MOM is beginning to see application in atmospheric models by several groups.

MOM is also being applied to combustion aerosols. It has many potential applications.

For further information see **www.ecd.bnl.gov/steve/schwartz.html**