DESCRIPTION OF ~~ATMOSPHERIC~~ AEROSOL DYNAMICS BY THE ***QUADRATURE*** METHOD OF MOMENTS

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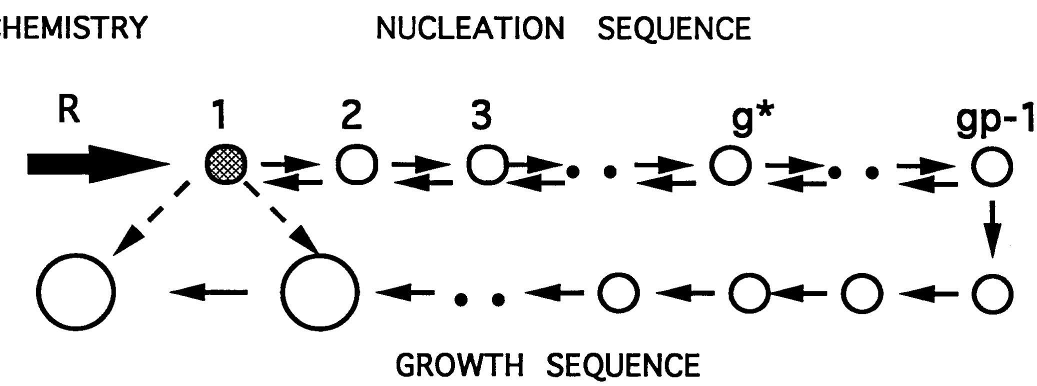
OUTLINE

1. Introduction.
   - Kinetics of coupled nucleation and growth.
   - Advantages and limitations of the conventional method of moments (*MOM*).
   - Aerosol optical properties directly from the moments (with Peter Huang).

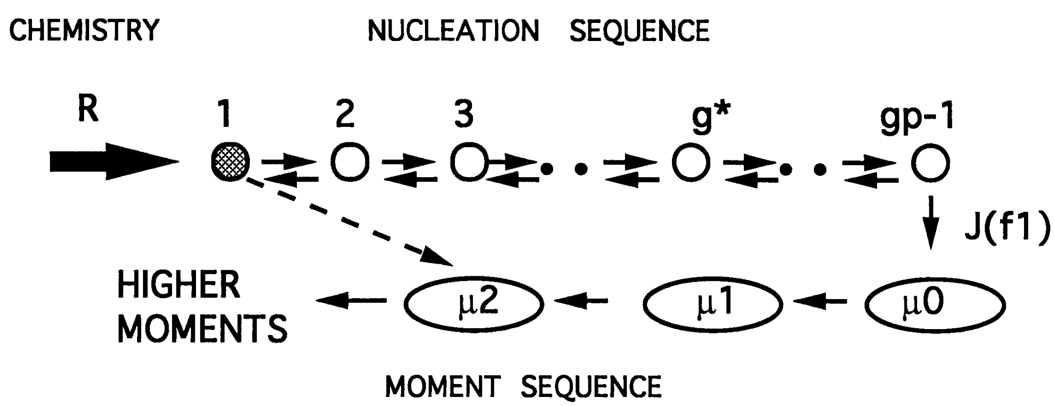
2. Quadrature method of moments (*QMOM*).
   - Illustrative calculations for diffusion controlled growth.

3. Properties of aerosol size distributions having identical radial moments (with Seth Nemesure).

KINETICS OF COUPLED NUCLEATION AND GROWTH



Method of moments (MOM): Replace the infinite growth sequence with the lower-order moment sequence:



<u>MOMENTS</u> $$\mu_k = \int_0^\infty r^k f(r)dr$$

where *f(r)* is the (generally unknown) particle size distribution.

NUCLEATION AND GROWTH IN COMPLEX FLOWS

MOMENT EVOLUTION EQUATIONS

Coupled set of general dynamic equations (GDE's) suitable for describing aerosol formation in complex flowfields:

$$\frac{\partial}{\partial t} f_1 = R - S - \nabla \cdot D \nabla f_1 - \nabla \cdot (f_1 \mathbf{v}) + \left(\frac{\partial}{\partial t} f_1 \right)_{nucl} - \underbrace{\frac{4\pi}{v_1} \int r^2 \phi(r) f(r) dr}_{\text{loss of monomer}}$$

$$\frac{\partial}{\partial t} \mu_0 = -\nabla \cdot D \nabla \mu_0 - \nabla \cdot (\mu_0 \mathbf{v}) + \int J(r) dr$$

$$\frac{\partial}{\partial t} \mu_k = -\nabla \cdot D \nabla \mu_k - \nabla \cdot (\mu_k \mathbf{v}) + \underbrace{\int r^k J(r) dr}_{\text{nucleation}} + \underbrace{k \int r^{k-1} \phi(r) f(r) dr}_{\text{growth from monomer addition}}, \; k \geq 1$$

where:

f_1 is the monomer concentration,
R and *S* are source and sink rates, respectively, for monomer,
D is the eddy diffusion constant for turbulent mixing,
v is the local flow velocity,
v_1 is the volume per monomer,
J(r) is the nucleation rate, and
$\phi(r)$ is the particle growth law, $\phi(r) \equiv dr / dt$.

- *How to evaluate integrals over the unknown distribution function???*

LIMITATION OF THE CONVENTIONAL METHOD OF MOMENTS

GROWTH LAW RESTRICTION

Necessary and sufficient condition for exact closure of the moment evolution equations is a growth law of the form:

$$\phi(r) \equiv \frac{dr}{dt} = a + br$$

where *a* and *b* are independent of *r*. Then integral evaluation proceeds as follows:

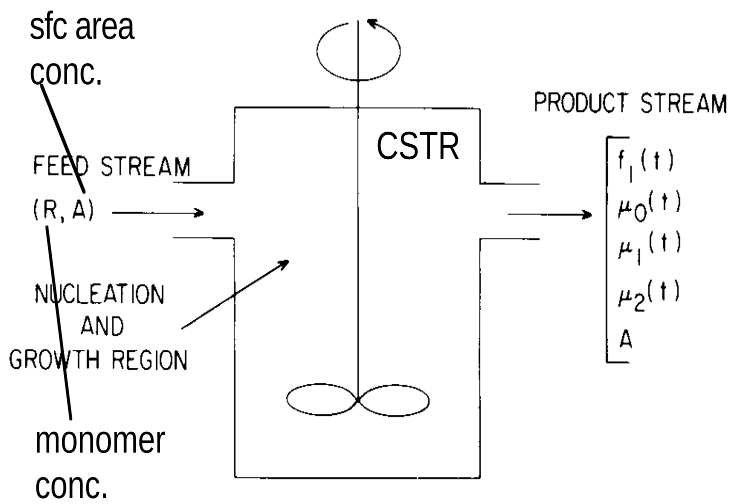
$$k\int r^{k-1}\phi(r)\,f(r)dr = ak\int r^{k-1} f(r)dr + bk\int r^{k} f(r)dr = ak\mu_{k-1} + bk\mu_{k}.$$

- *The important case b = 0 represents free-molecular growth.*

- *The QMOM replaces exact closure with an* ***approximate but much less restrictive closure condition.***

AN APPLICATION OF THE MOMENT METHOD

(R. McGraw and J. H. Saunders, *Aerosol Sci. and Tech.* **3**, 367, 1984)



$$\frac{df_1}{dt} = k_D R - k_D f_1 - \beta(1) f_1 \mu_2,$$

collision loss of monomer

$$\frac{d\mu_0}{dt} = J(f_1) - k_D \mu_0,$$

$$\frac{d\mu_1}{dt} = r_{g_p} J(f_1) + v_1 \beta(1) f_1 \mu_0 - k_D \mu_1,$$

nucleation growth by add'n of monomer

$$\frac{d\mu_2}{dt} = s_{g_p} J(f_1) + 8\pi v_1 f_1 \beta(1) \mu_1 - k_D \mu_2.$$

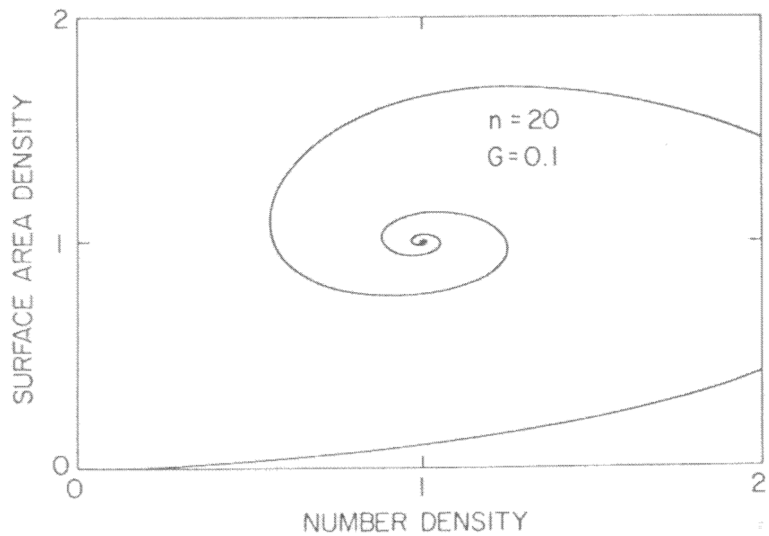
nucleation growth by add'n of monomer

The moments evolve according to a ***closed set of differential equations*** *having the same structure as rate equations governing the evolution of reacting chemical species.*

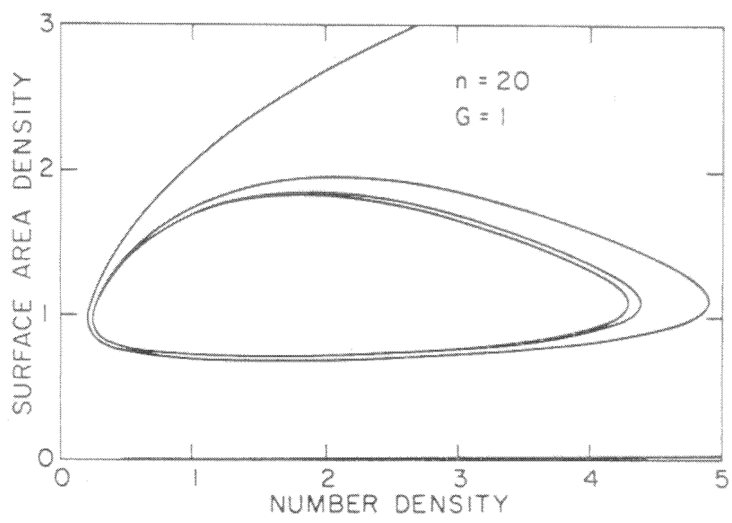
Representation of nucleation and growth processes in models is thus reduced mathematically to the ***simulation of a few coupled reacting chemical species*** *in the same flow.*

NONLINEAR EFFECTS FROM CONDENSATION FEEDBACK

(R. McGraw and J. H. Saunders, *Aerosol Sci. and Tech.* **3**, 367, 1984)



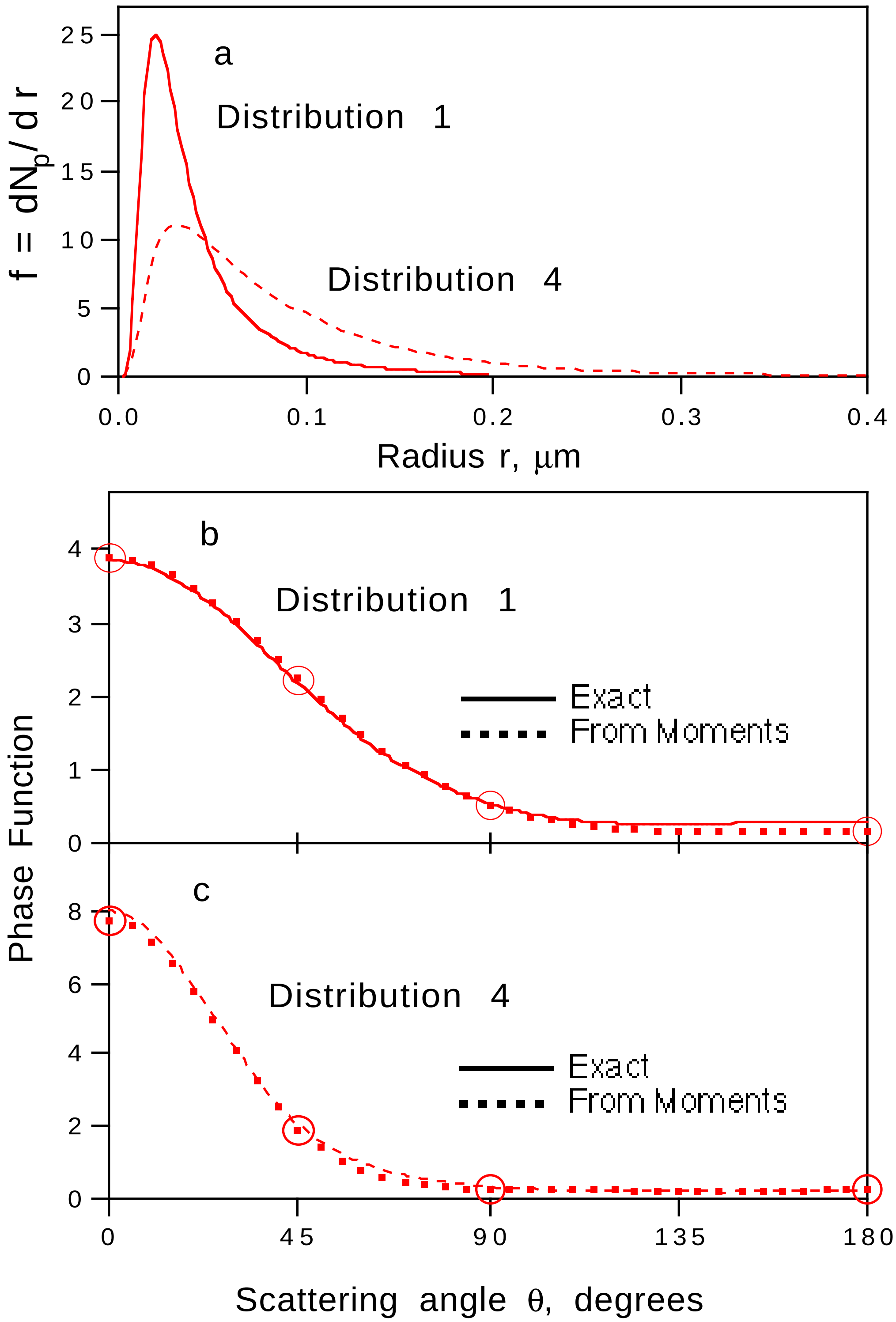
Aerosol surface area density vs. number density in the stable regime (low rate of monomer generation).



Similar plot in the unstable regime (high rate of monomer generation leads to oscillatory behavior).

CALCULATION OF OPTICAL PROPERTIES DIRECTLY FROM MOMENTS

(McGraw R., Huang P. I., and Schwartz S. E. *Geophys. Res. Lett*. **22**, 2929, 1995)



QUADRATURE METHOD OF MOMENTS

INTEGRAL APPROXIMATION VIA n-POINT GAUSSIAN QUADRATURE:

$$k\int r^{k-1}\phi(r)f(r)dr \cong k\sum_{i=1}^{n} r_i^{k-1}\phi(r_i)w_i, \quad k \geq 1.$$

- *Essence of quadrature-based closure lies in the fact that the abcissas (r_i) and weights (w_i) are completely specified in terms of the lower-order moments of f(r).*
- *The moments themselves may be written in this form:*

$$\mu_k = \int r^k f(r)dr = \sum_{i=1}^{n} r_i^k w_i$$

MOMENT INVERSION

An efficient algorithm has been developed (McGraw, *Aerosol Sci. and Tech.*, submitted, 1996) for rapid conversion of the lower-order moment sequence to quadrature abcissas and weights. For 3-point quadrature (requiring 6 moments):

$$\{\mu_0,\mu_1,\mu_2,\mu_3,\mu_4,\mu_5\} \Leftrightarrow \{r_1,w_1;r_2,w_2;r_3,w_3\}$$

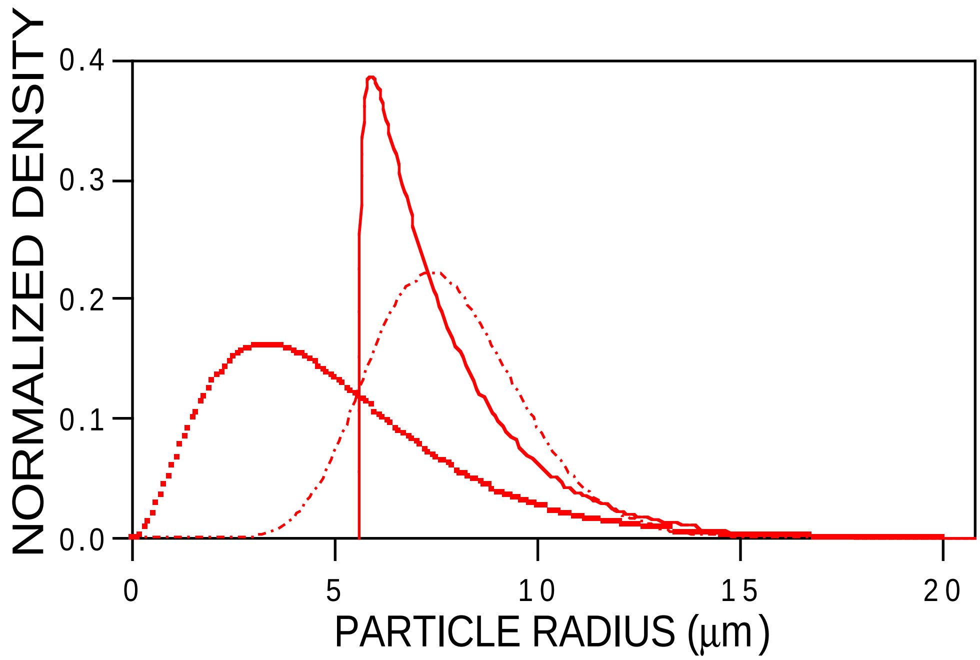
- *Once the abcissas (r_i) and weights (w_i) have been determined (from the moments), the unknown distribution function integrals are obtained by the summation indicated on the right hand side of the first equation above.*

CALCULATIONS FOR DIFFUSION CONTROLLED GROWTH

• *The diffusional growth law:*

$$dr/dt = k/r$$

results in moment evolution equations that are not in closed form. Only approach until now has been to use assumed distributions parameterized in terms of moments (e.g. Laguerre).

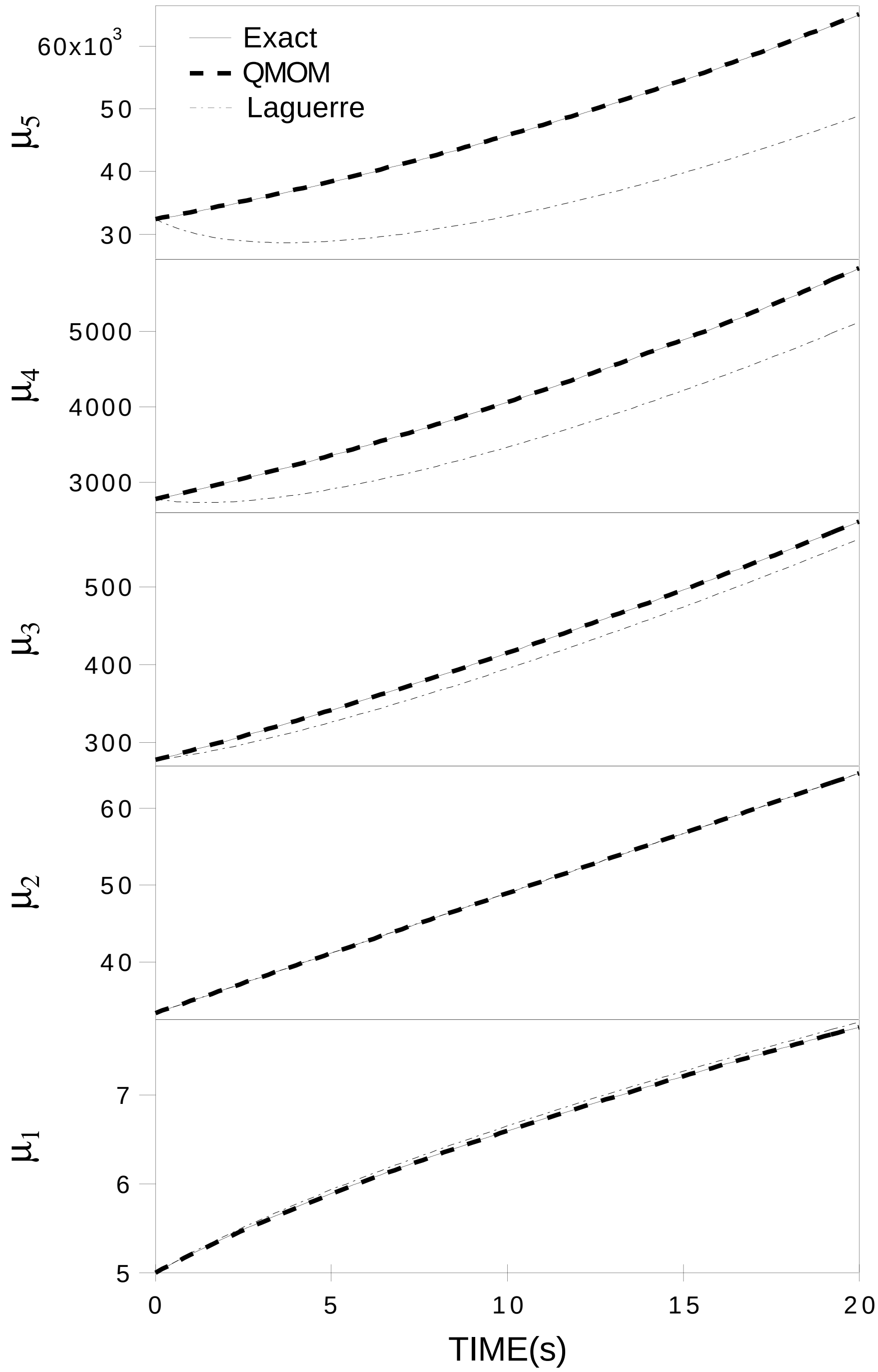


Particle size distributions. Diffusion controlled growth of water drops at T = 278 K and fixed supersaturation of 101% $(S = 1.01)$. *Dotted curve, initial normalized K-M distribution with mean particle radius of 5 mm. Solid curve, exact evolved distribution after 20s. Dashed-dotted curve, Laguerre distribution parameterized by the moments 0 through 2 after propagation to t=20s using the Laguerre closure method.*

QMOM CALCULATIONS FOR DIFFUSION CONTROLLED GROWTH

(McGraw R., *Aerosol Sci. and Tech.*, submitted, 1996)

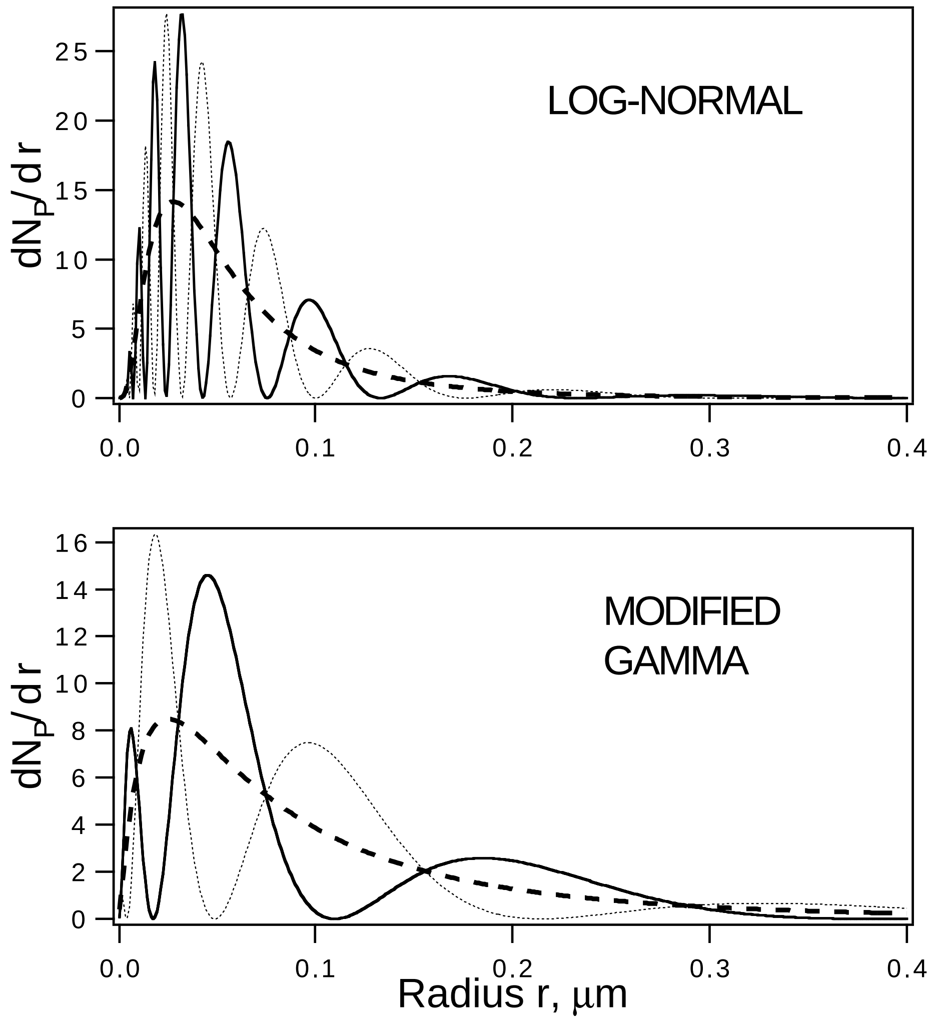
• *Quadrature MOM permits calculation of the evolution of the moments directly, without a priori assumptions about the form of the evolving distribution.*



AEROSOL SIZE DISTRIBUTIONS HAVING IDENTICAL RADIAL MOMENTS

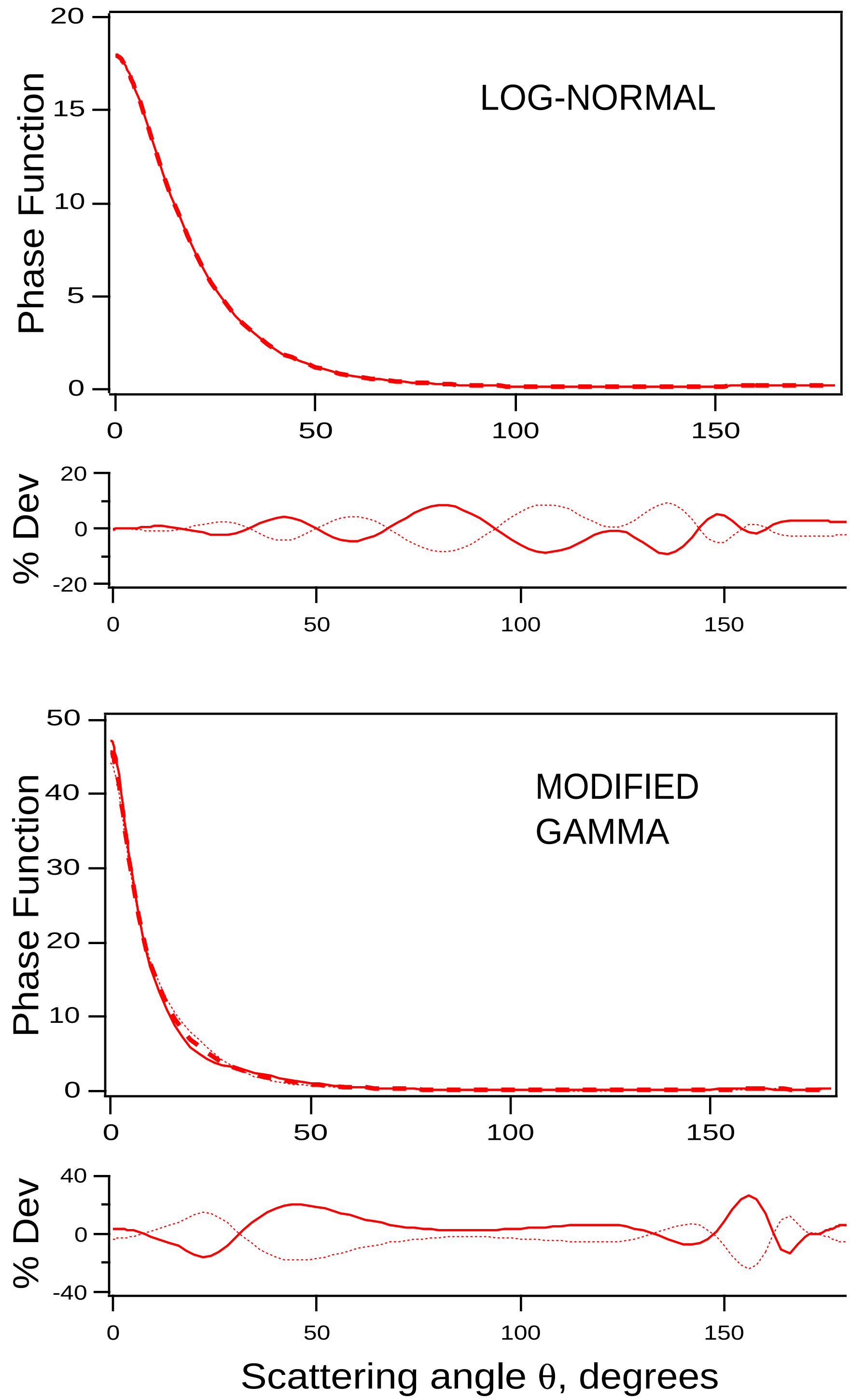
(McGraw R., Nemesure S., and Schwartz S. E., *Aerosol Sci. and Tech.*, to be submitted, 1996)

Two classes of size distributions having identical moments:



Class distribution form is $g(r) = f(r)[1 + wF(r)]$ where $f(r)$ is the parent distribution (e.g. lognormal or modified gamma) and $F(r)$ is periodic with properties such that the moments of the product $f(r)F(r)$ vanish. The parameter w ($|w| \le 1$ such that $g(r)$ is nonnegative) is here set to 0 and ± 1.

PHASE FUNCTIONS OF DIFFERENT AEROSOL SIZE DISTRIBUTIONS HAVING IDENTICAL RADIAL MOMENTS



Phase functions for the distributions $w = 0$ and $w = \pm 1$. Percent deviations from the parent distributions ($w = 0$) are also shown.

CONCLUSIONS

- Aerosol optical properties are well represented by the moments of the radial size distribution or by functions of the moments, *e.g.*, parametrization of radiation transfer in terms of the "effective radius" $r_e = \mu_3/\mu_2$, and the phase function as shown here.

- The ***Conventional*** Method of Moments is well suited for describing the evolution of the moments of an aerosol size distribution, but only for highly restrictive growth laws.

- The ***Quadrature*** Method of Moments extends the method of moments to arbitrary growth laws and should thus greatly extend the applicability of the method, for example application in atmospheric transport models.

- Different distributions having the identical sets of moments seems to be of little practical concern.