REPRESENTING AEROSOL MICROPHYSICS IN HEMISPHERIC-SCALE CHEMICAL TRANSPORT AND TRANSFORMATION MODELS

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ABSTRACT

Direct light scattering of solar radiation and enhanced cloud albedo due to anthropogenic sulfate aerosol are hypothesized to lead to a shortwave radiative forcing of climate by about -1 W m-2 global average (that is, exerting a cooling influence), comparable but opposite to longwave forcing by anthropogenic greenhouse gases. Inclusion of this forcing in climate models requires improved estimates of the geographical distribution of this forcing, and in turn of sulfate concentrations. This can be achieved only with models. Here we report results of applying our subhemispheric scale transport and transformation model for aerosol sulfate and comparison with observations, and outline the approach to incorporating aerosol microphysical properties in the model.

The model has been described previously (Benkovitz et al., 1994). The distinguishing feature of the model is that it is driven by observationally derived meteorological data, thereby allowing comparisons with observations not just at specific locations, as is customary, but also at specific times. These comparisons indicate that a substantial fraction of the temporal variance at any given location is accounted for by the temporal variability of the model, all of which is due to temporal variability in the meteorological field driving the model. Examples of comparisons of SO2 and sulfate mixing ratios are presented illustrating the temporal variability and the ability of the model to capture this variability. The comparisons include observations from multiple observing stations within a single model grid cell (1.125˚ × 1.125 ˚) illustrating substantial subgrid spatial variability. The model permits attribution of sulfate to source regions and to chemical production mechanism.

It is intended to represent aerosol microphysical properties in the model in terms of the moments of the radial size distribution. The moments accurately represent observable physical properties of the aerosol (McGraw et al., 1995). Under limited (but important) conditions the moments evolve according to a closed set of differential equations and may thus be treated as chemical species (McGraw and Saunders, 1984), but in general this set of equations is not closed. Recently we have developed an extension of this approach that permits the set of equations to be closed by an approximation involving gaussian quadratures (McGraw, 1996). Initial tests indicate that this approximation is highly accurate. This quadrature method of moments should thus be of great utility in representing aerosol dynamics in regional to hemispheric or global scale models.

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OVERVIEW

- The hemispheric scale transport and transformation model

- Representing aerosol nucleation and growth processes

OBJECTIVES

Develop, **evaluate**, and **apply** models describing the chemical and microphysical properties of atmospheric aerosols resulting from energy related and other activities on a hemispheric geographical scale.

MOTIVATION

- Direct light scattering of solar radiation and enhanced cloud albedo due to anthropogenic sulfur aerosol are hypothesized to lead to a shortwave radiative forcing of climate by about -1 W m^{-2} global average (that is, exerting a cooling influence), comparable but opposite to longwave forcing by anthropogenic greenhouse gases.
- Inclusion of this forcing in climate models requires improved estimates of the geographical distribution of this forcing, and in turn of sulfate concentrations.
- This can be achieved only with models.

APPROACH

- Eulerian transport and transformation model for atmospheric sulfate and its precursors, **driven by operational meteorological data** (Benkovitz et al., JGR, 1994).

- Present model represents sulfate molar mixing ratio with no explicit treatment of aerosol microphysics.

- We are developing approaches to representing aerosol microphysics via the moments of the radial size distribution.

MODEL ATTRIBUTES

- Horizontal grid cell is 1.125°.
- Terrain following eta (η) coordinates in the vertical; 15 levels between surface and 100 mbar.
- Time resolution is six hours.
- Advection per Bott (Mon. Wea. Rev., 1989); subgrid vertical convective transport scheme.

INPUTS TO THE MODEL

- Winds and precipitation from the 6-hour forecast model of the European Center for Medium-Range Weather Forecast (ECMWF).

- Anthropogenic emissions from the NAPAP and EMEP 1985 inventories.

- Biogenic emissions based on Bates et al. (J. Atmos. Chem., 1992); ocean emissions of DMS scaled to Coastal Zone Color Scanner chlorophyll.

CHEMICAL REACTIONS

- Gas-phase oxidation of SO_2 and DMS by OH.
- Climatological concentration field for OH.

- Aqueous-phase oxidation of SO_2 by H_2O_2 and O_3.
- Climatological formation rate of H_2O_2; depletion by in-cloud reaction.
- O_3 25 - 50 ppb (seasonal); cloudwater pH 4.5.

REMOVAL PROCESSES

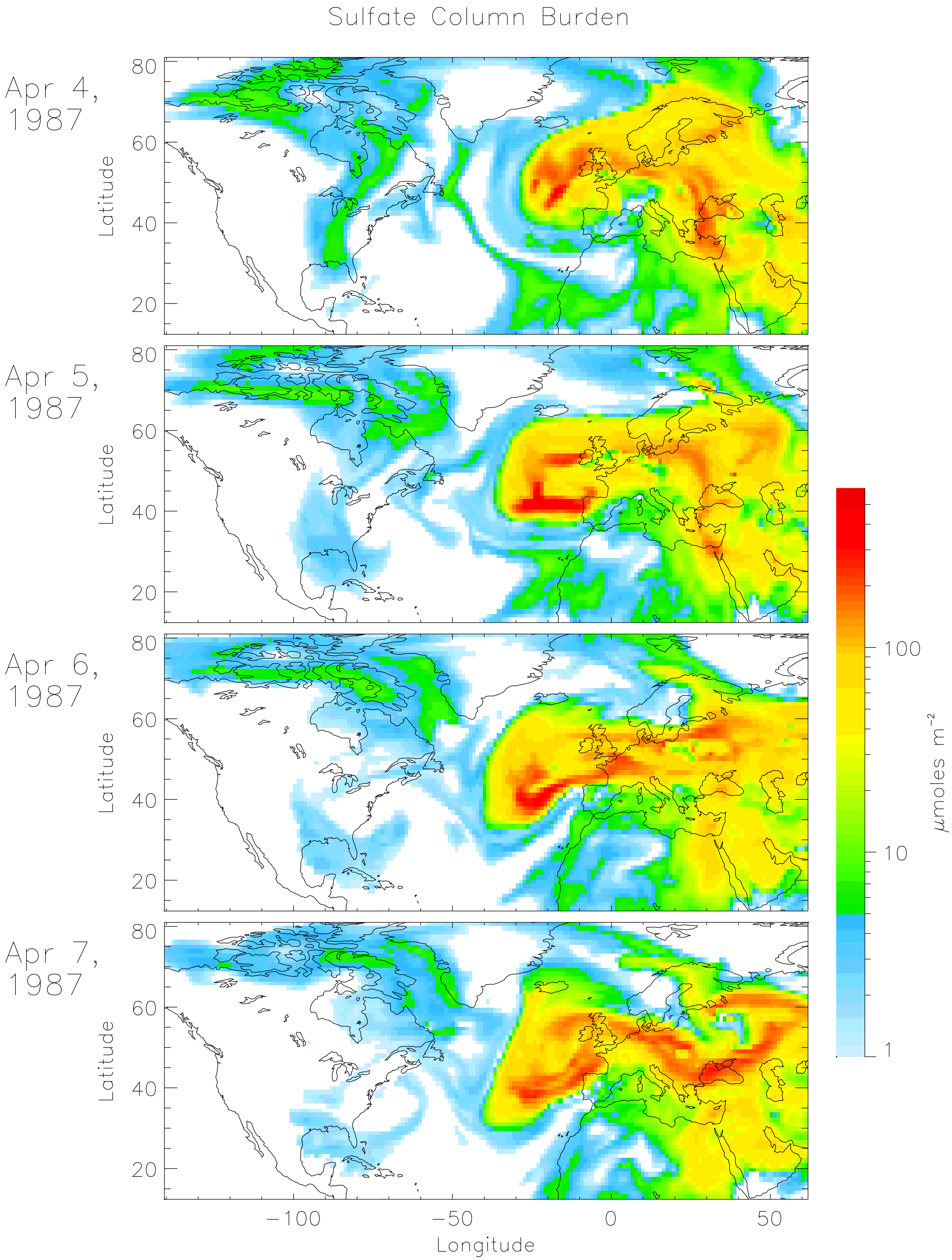
- Dry deposition velocities of SO_2 and sulfate calculated per Wesely and colleagues (M. Wesely, Atmos. Environ. 1989).

- Wet deposition of dissolved species including aerosol according to removal of liquid water in rain.

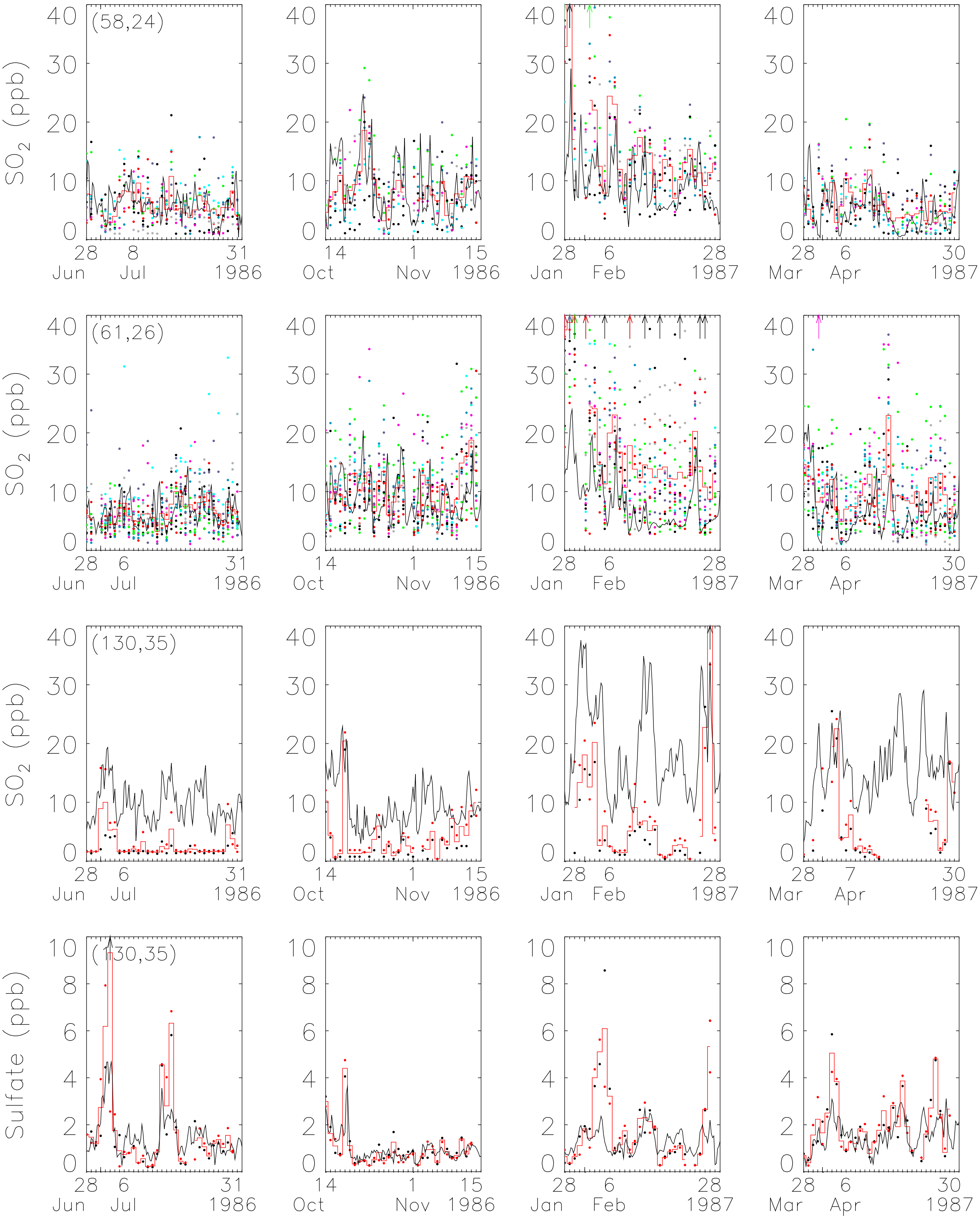
MODEL OUTPUT

- Concentrations of sulfate aerosol (n mol per mol of air, ppb) as function of location (*x*, *y*, *z*) and time.
- Column burdens of sulfate aerosol (mol m^{-2}) as function of location (*x*, *y*) and time.

Example model output showing contributions to sulfate column burden (vertical integration of the concentration) for April 6, 1987. (Top) Burden resulting from biogenic emissions. (Second) Burden resulting from anthropogenic emissions west of 30^{o}W. (Third) Burden resulting from anthropogenic emissions east of 30^{o}W. (Bottom) Total burden (resulting from emissions above, plus material transported from outside the model domain). Note the recirculating band of high burdens from European emissions over the northeast Atlantic ocean. An occluded cyclone is centered to the southeast of the British Isles; an extensive band of easterly winds associated with the warm conveyor belt is causing this recirculating band.


Sulfate Column Burden
Apr 4, 1987
Apr 5, 1987
Apr 6, 1987
Apr 7, 1987
Latitude
80
60
40
20
−100
−50
0
50
Longitude
100
10
1
μmoles m^{-2}

Example comparisons of observed and modeled mixing ratios. Panels show time series of the SO_2 24-h average mixing ratio (first 3 rows) and of the observed sulfate 24-h mixing ratio (last row) at grid cells ($1.125^{o} \times 1.125^{o}$) for which observations are available from multiple stations for four simulations (June 28 to July 31, 1986; October 14 to November 14, 1986; January 28 to February 28, 1987 and March 28 to April 30, 1987). Points of a given color represent observations at a single station. The spread of points of the same color represents the time variability at a station; the vertical spread of different color points on a given date represents the subgrid spatial variability within the grid cell. The arrows indicate one or more observations outside the limits of the plot. The grid-cell average (red line) observed mixing ratio and the 6-h modeled mixing ratio (black line) are also shown.


(58,24)
(61,26)
(130,35)
(130,35)
SO_2 (ppb)
SO_2 (ppb)
SO_2 (ppb)
Sulfate (ppb)
0
10
20
30
40
0
2
4
6
8
10
28 Jun
8 Jul
6 Jul
31 1986
14 Oct
1 Nov
15 1986
28 Jan
6 Feb
28 1987
28 Mar
6 Apr
7 Apr
30 1987

DESCRIPTION OF AEROSOL PROPERTIES AND 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 size distribution.

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

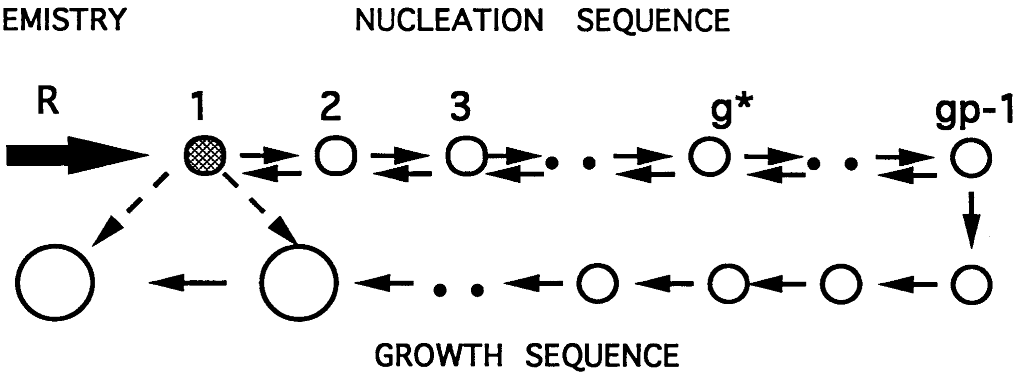
Aerosol ***properties*** (e.g., light scattering coefficient) can be accurately represented as simple functions of low order moments.

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

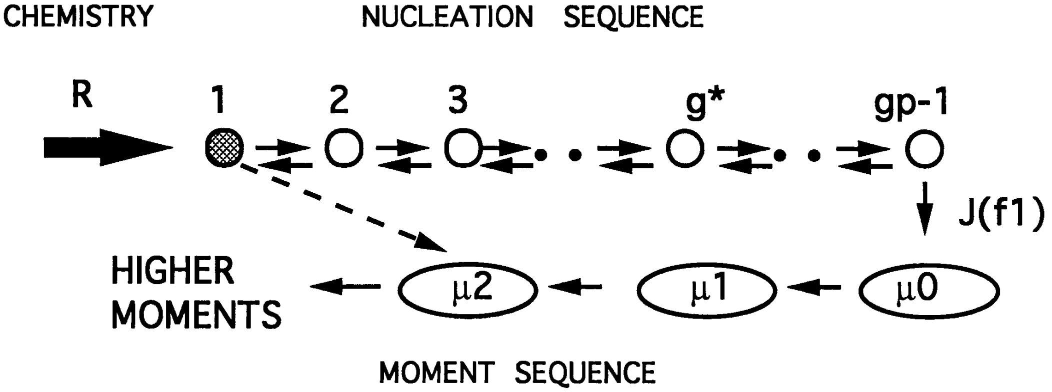
The moments advect and mix just like chemical species--they are conserved and additive.

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

KINETICS OF COUPLED NUCLEATION AND GROWTH



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



MOMENTS

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

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:

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

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

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

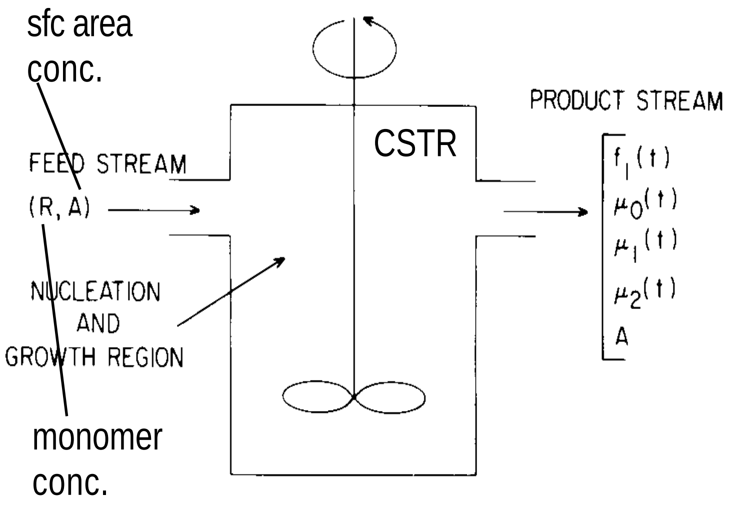
where:

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,
$\mathbf{v}$ 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???*

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

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

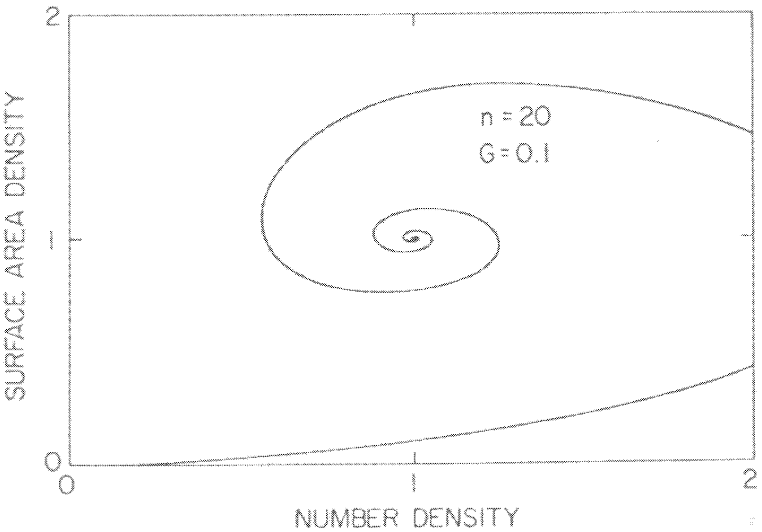
nucleation growth by add'n of monomer

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.*

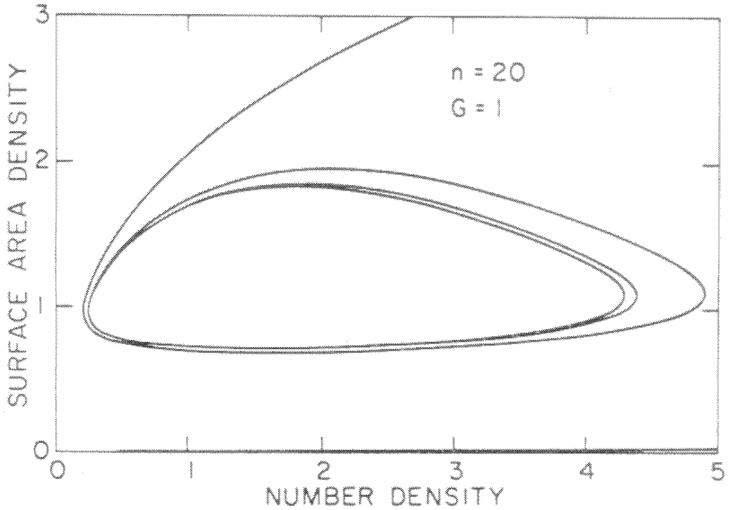
Representation of nucleation and growth processes in models is thus reduced mathematically to the ***simulation of a few coupled reacting chemical species*** *in the same flow.*

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).

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.***

QUADRATURE METHOD OF MOMENTS

INTEGRAL APPROXIMATION VIA n-POINT GAUSSIAN QUADRATURE:

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

- *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):

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

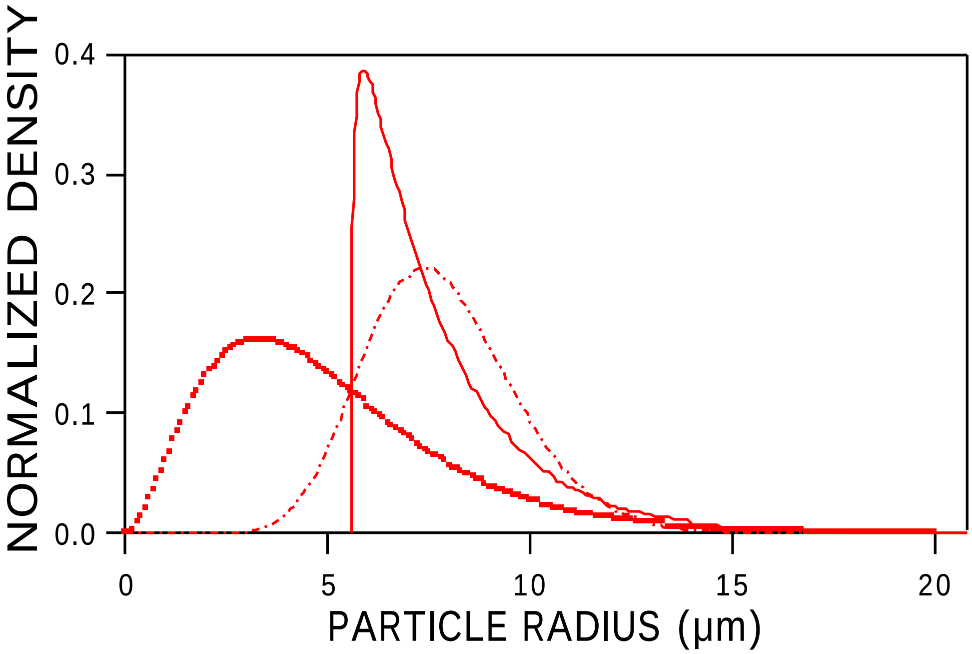
- *Once the abcissas (r_i) and weights (w_i) have been determined (from the moments), the unknown distribution function integrals are obtained by the summation indicated on the right hand side of the first equation above.*

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).

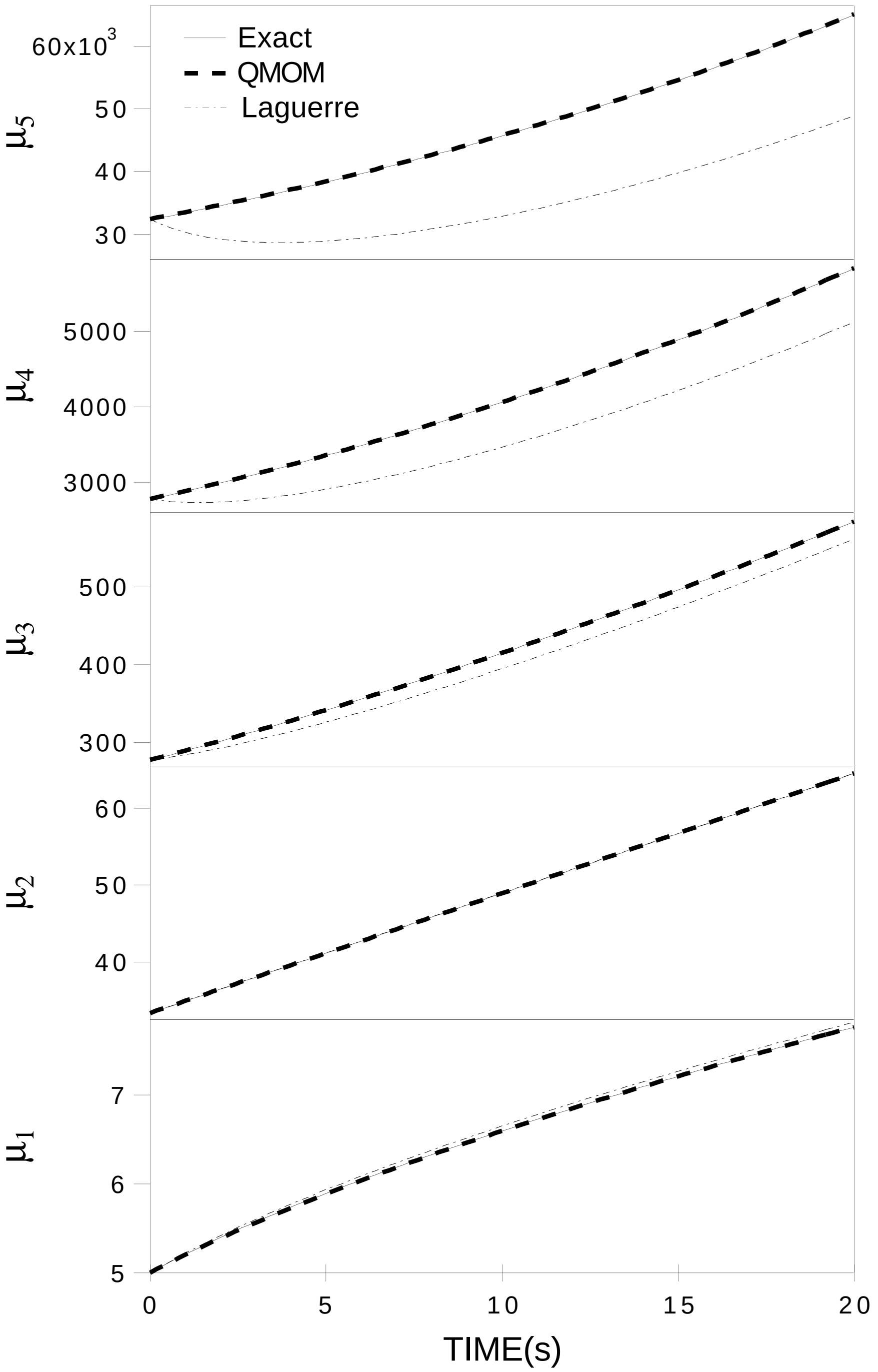


Particle size distributions. Diffusion controlled growth of water drops at T = 278 K and fixed supersaturation of 101% ($S = 1.01$). *Dotted curve, initial normalized K-M distribution with mean particle radius of 5 mm. Solid curve, exact evolved distribution after 20s. Dashed-dotted curve, Laguerre distribution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



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