HEMISPHERIC-SCALE CHEMICAL AND MICROPHYSICAL AEROSOL MODEL

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OUTLINE

Recap on GChM-O (Global Chemical Model driven by Observation-derived meteorological data)

Application of GChM sulfur chemistry in CCM3 (Collaboration with NCAR)

Disproportionate influence of volcanic sulfur on sulfate burden

Recent model advances

Recap on using the Method of Moments to represent aerosol microphysics

Obtaining aerosol integral properties from moments

Application of Method of Moments to hemispheric scale calculations in GChM-O

Representing cloud microphysics by the Method of Moments

Theory: Examination of ternary NH_3 - H_2SO_4 - H_2O nucleation

Ongoing and future work and Collaborations

CHEMICAL AND MICROPHYSICAL AEROSOL MODEL

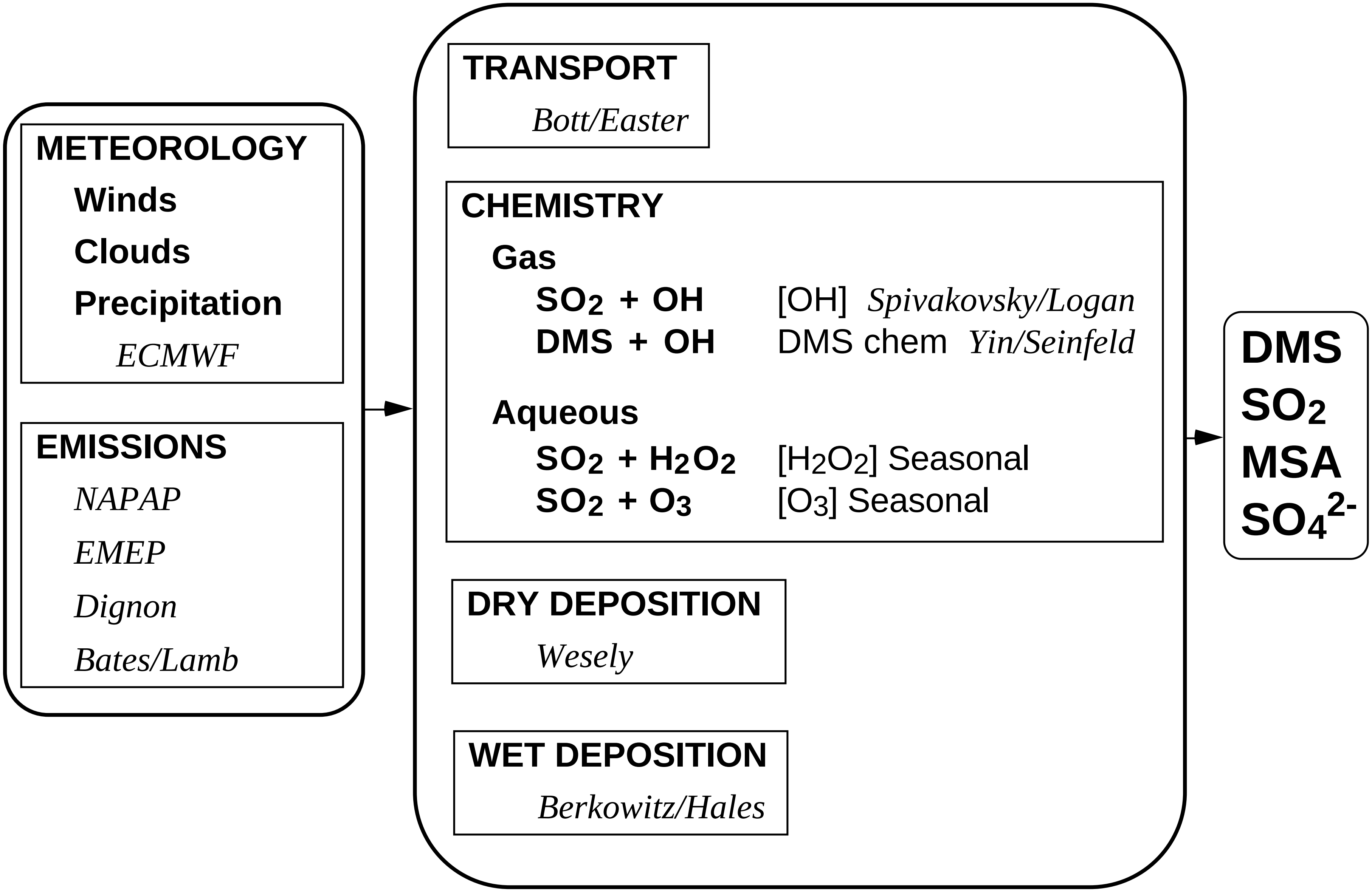
DRIVEN BY

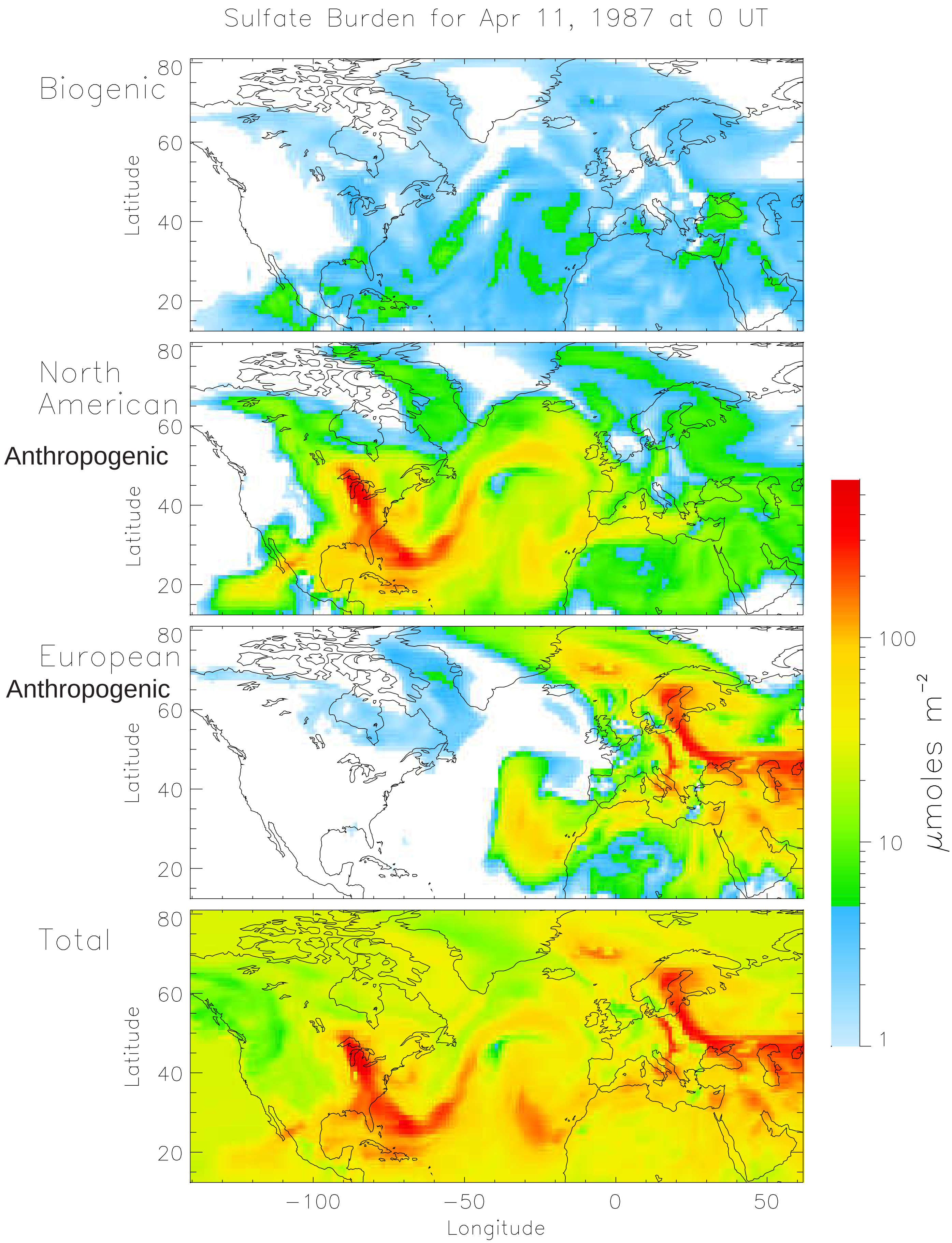
OBSERVATION-DERIVED METEOROLOGICAL DATA

OBJECTIVES

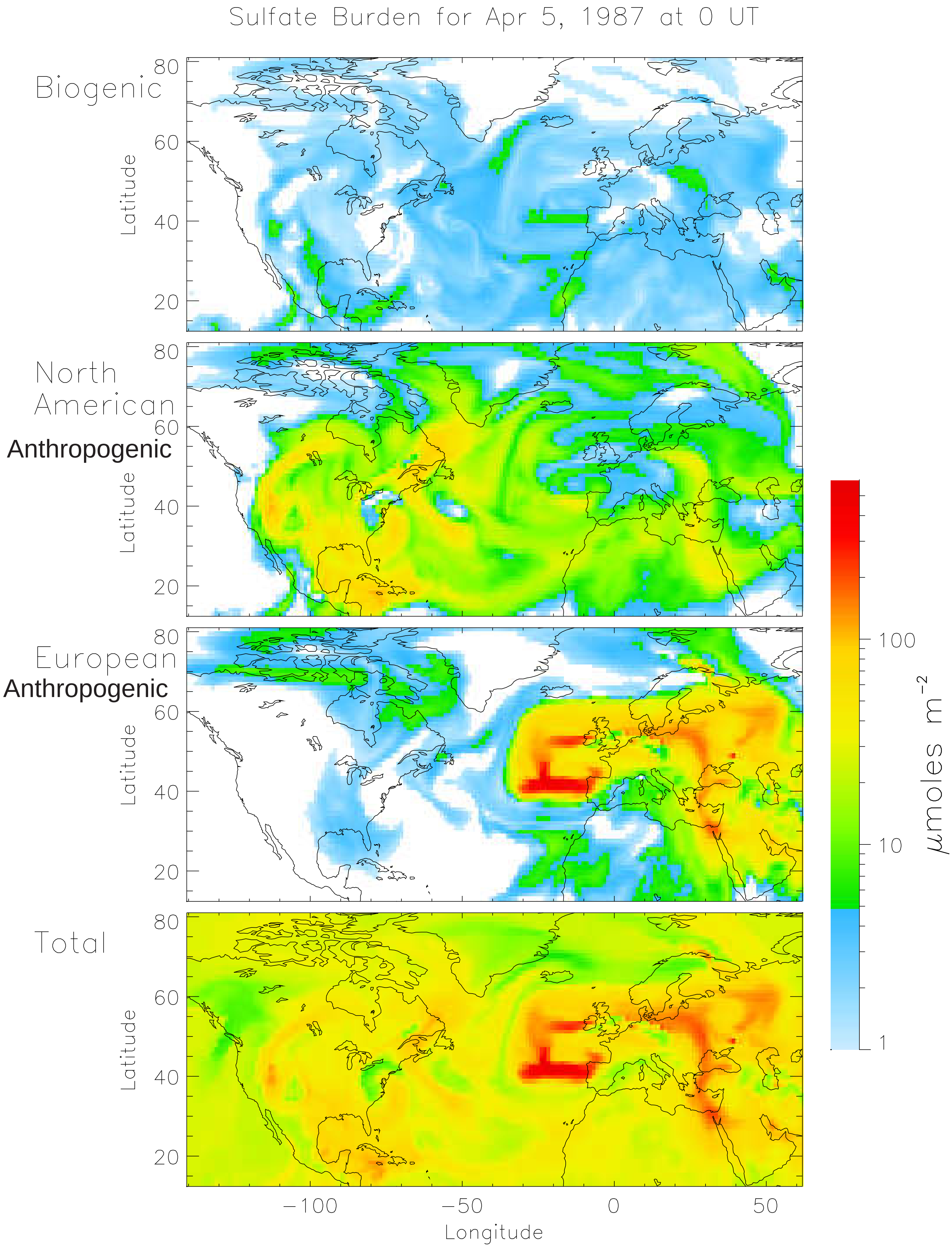
***Develop**, **evaluate**, and **apply** models describing the chemical and microphysical properties of atmospheric aerosols resulting from energy related and other activities on a hemispheric geographical scale.*

COMPONENTS OF THE TRANSPORT - TRANSFORMATION - REMOVAL MODEL

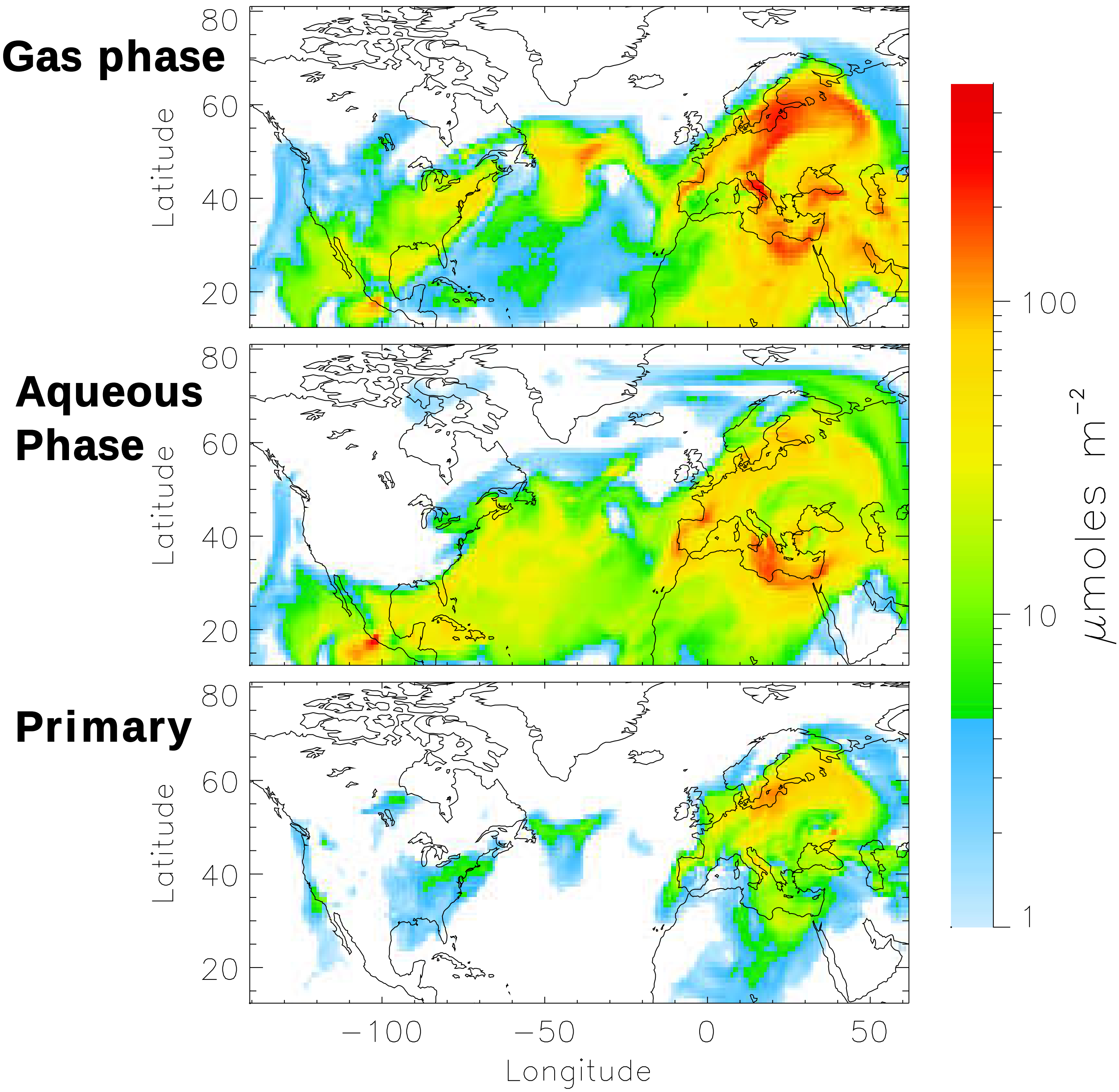




Sulfate Burden for Apr 11, 1987 at 0 UT
Biogenic
North American Anthropogenic
European Anthropogenic
Total
Latitude
80
60
40
20
−100
−50
0
50
Longitude
μmoles m⁻²
100
10
1


Sulfate Burden for Apr 5, 1987 at 0 UT
Biogenic
North American Anthropogenic
European Anthropogenic
Total
Latitude
80
60
40
20
−100
−50
0
50
Longitude
1
10
100
μmoles m⁻²

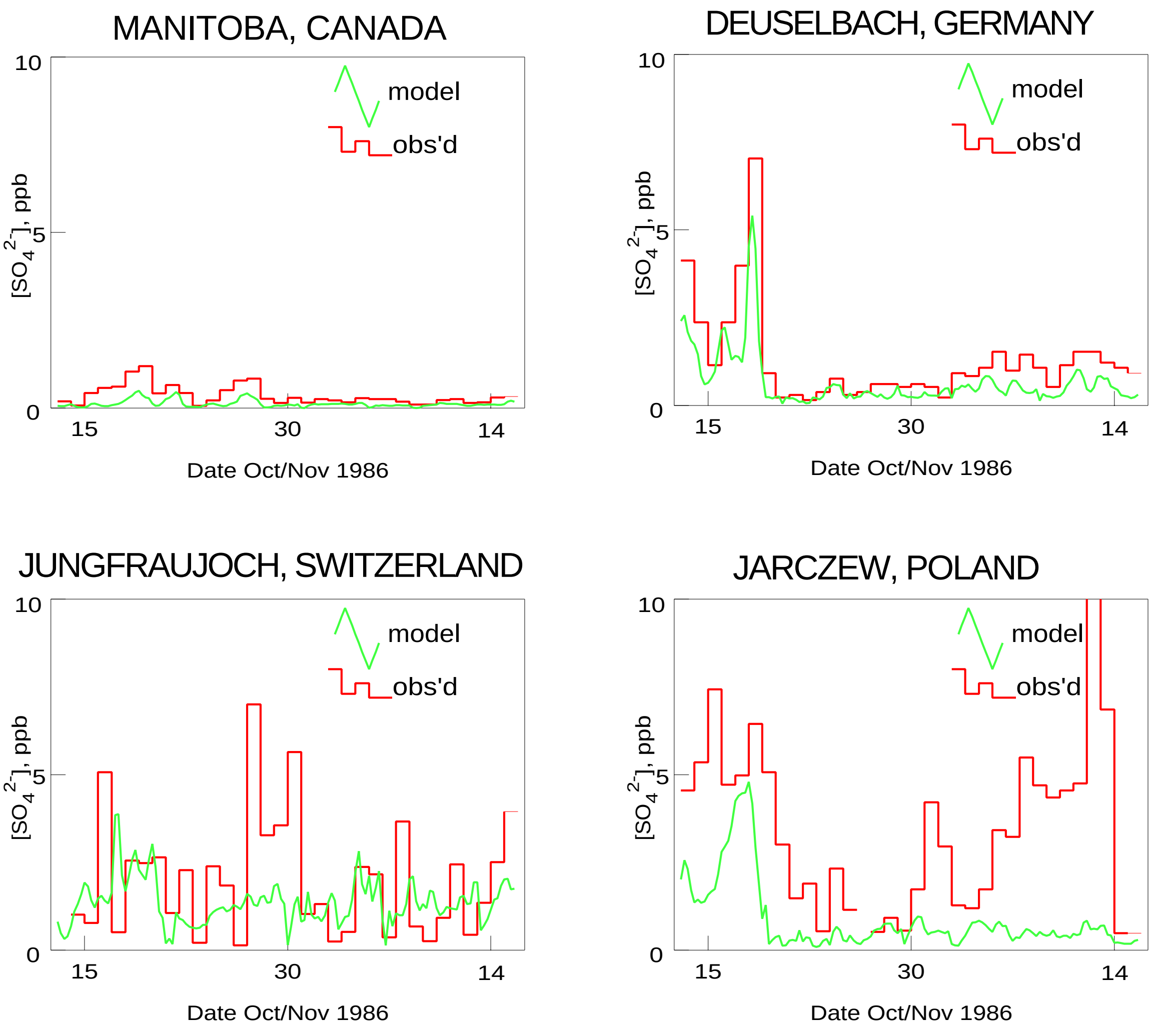
Sulfate Column Burden by Production Mechanism



October 15, 1986 at 6 UT

COMPARISON OF MODEL AND OBSERVATIONS

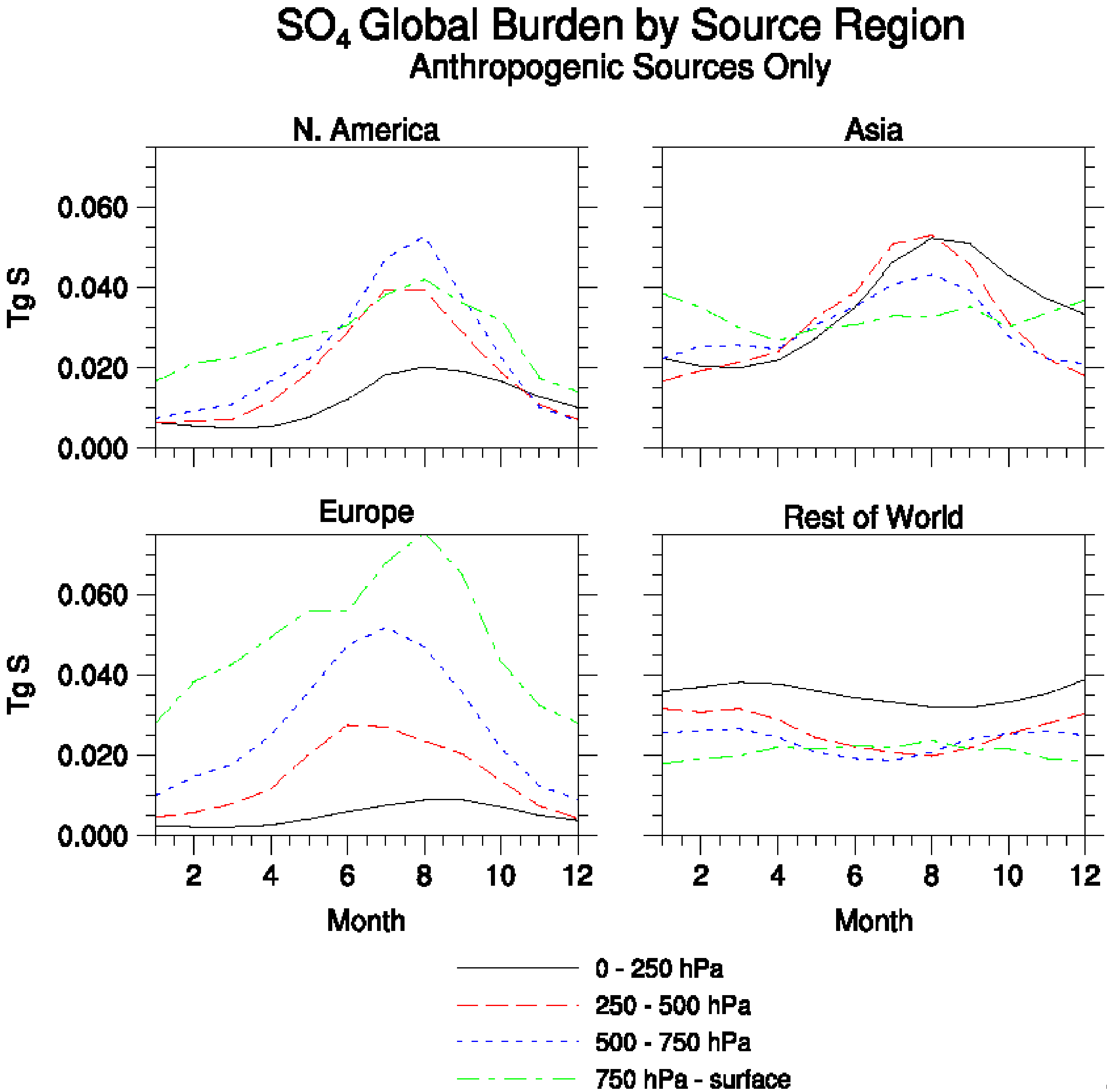
Typical comparisons for 24-hr Sulfate mixing ratio at surface



Statistics of Comparisons

	N	Median Spread
Obs-Obs	503	1.5
Model-Obs Same locations	503	1.9
Model-Obs All locations	7907	2.3

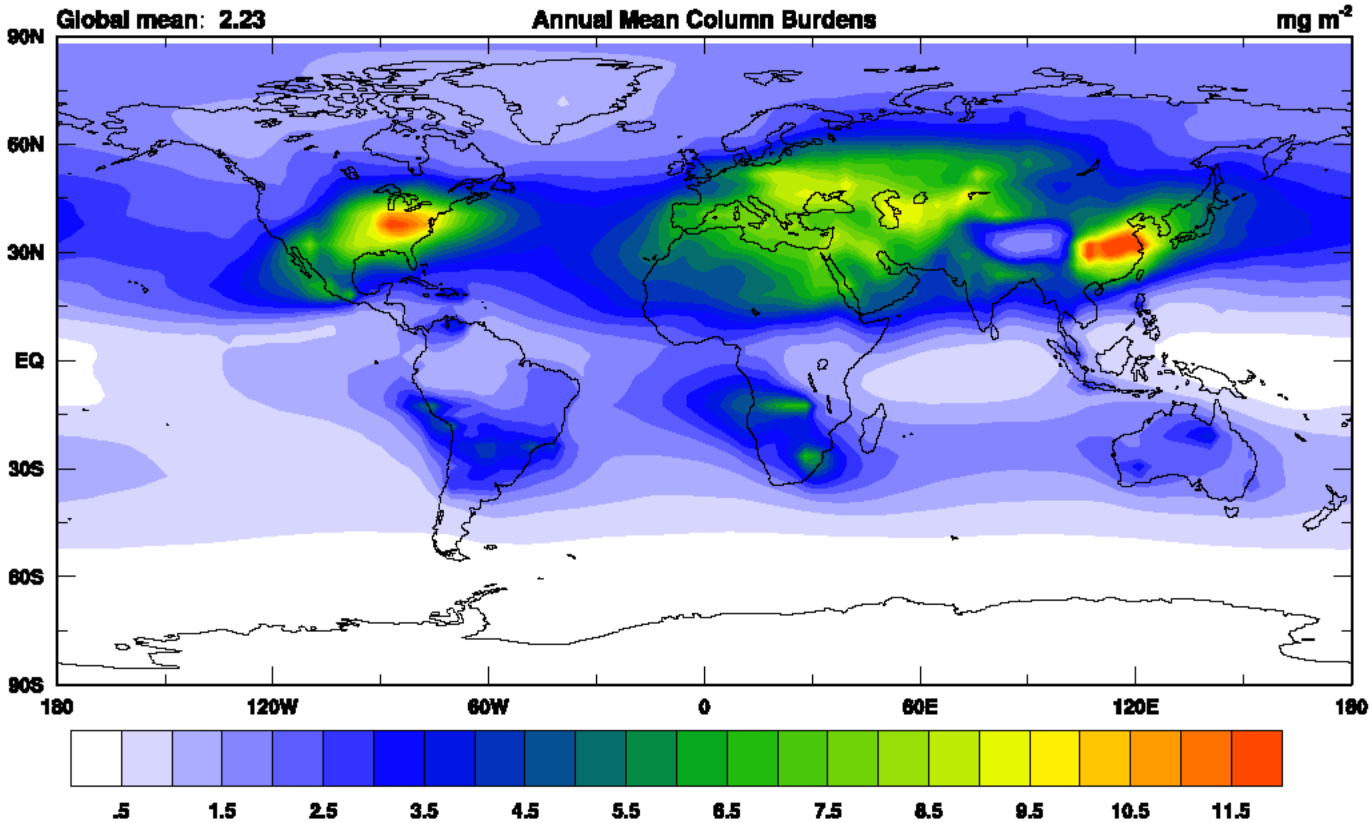
Benkovitz and Schwartz, *JGR*, 1997

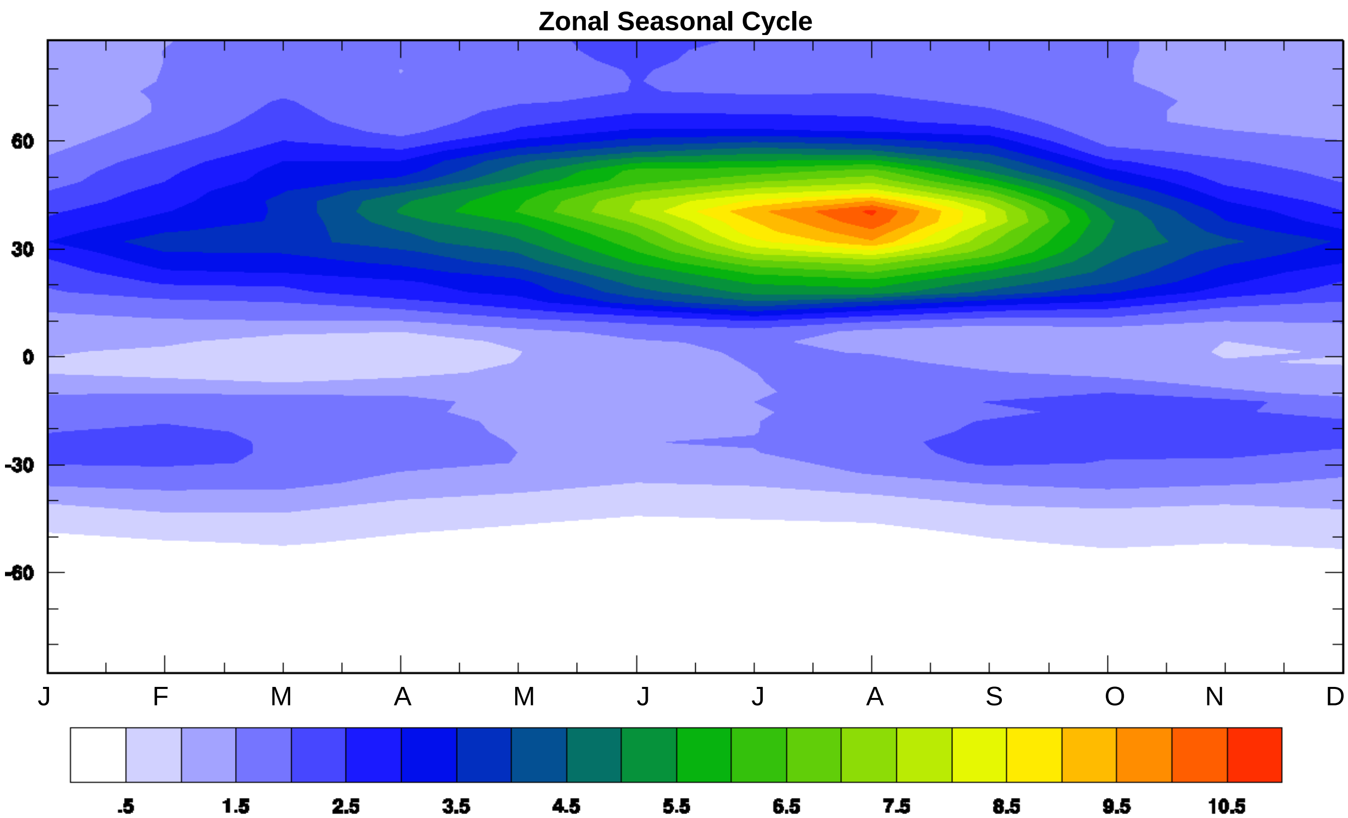


Rasch, Barth, Kiehl, Schwartz, Benkovitz, *JGR*, in press, 1999

ANTHROPOGENIC SULFATE COLUMN BURDEN

BNL Sulfate Model in NCAR CCM3



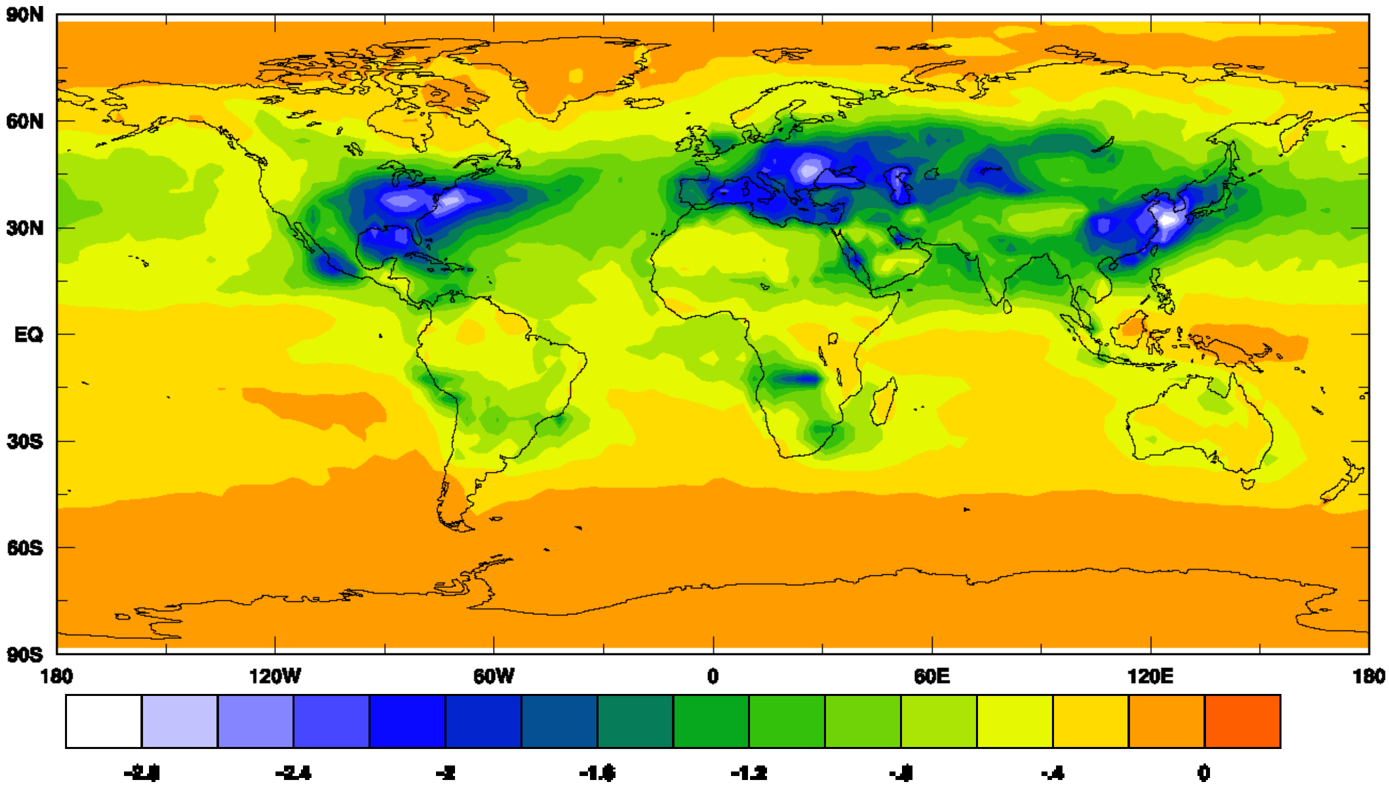


Kiehl et al., JGR, in press

ANTHROPOGENIC SULFATE DIRECT SHORTWAVE FORCING, ANNUAL AVERAGE

BNL Sulfate Model in NCAR CCM3

Global Mean, -0.56 W m^{-2}



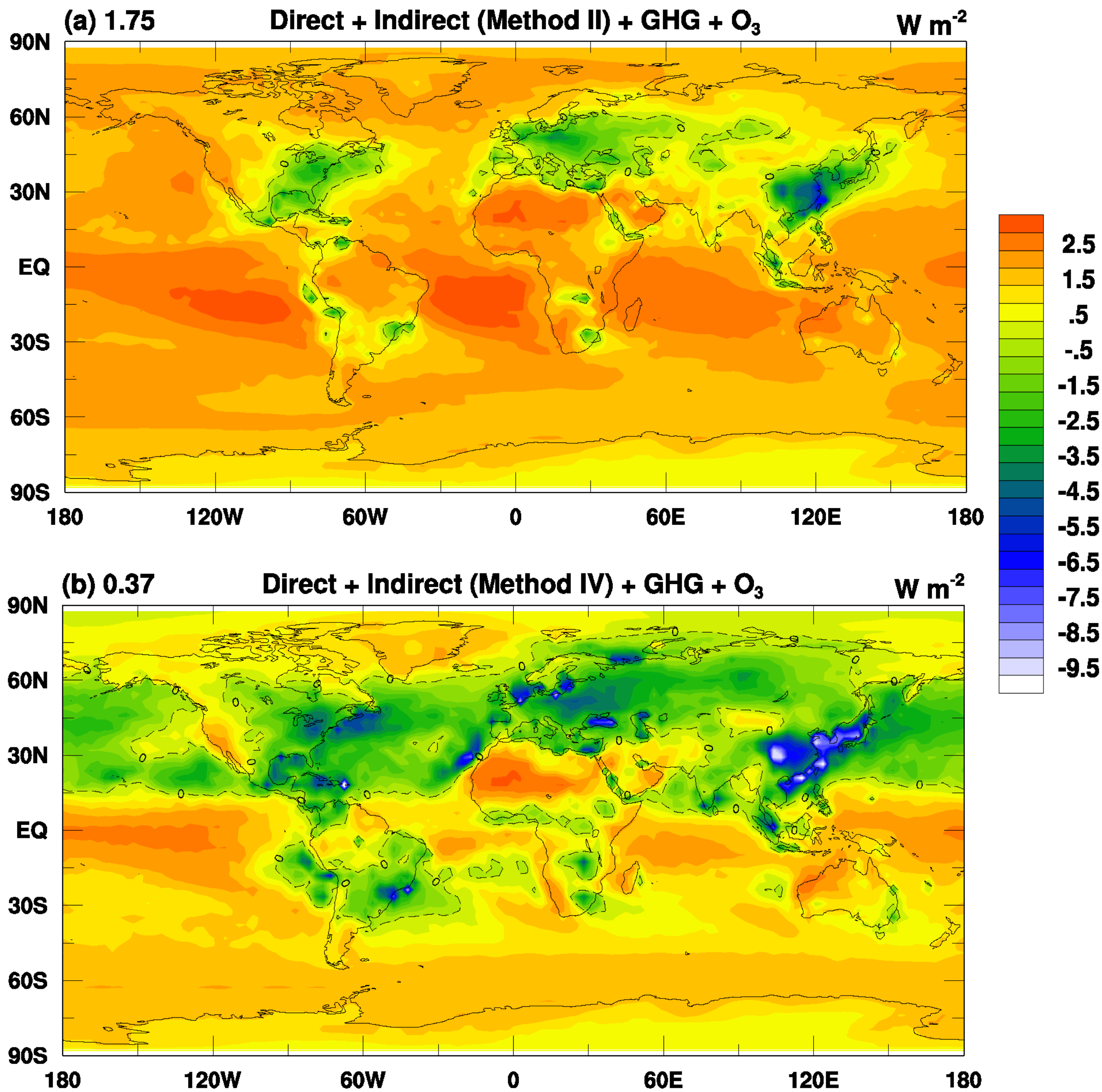
Kiehl et al., JGR, in press

SHORTWAVE FORCING, ANNUAL AVERAGE

BNL Sulfate Model in NCAR CCM3

GHG's + O_3 + Sulfate (Direct and Indirect)

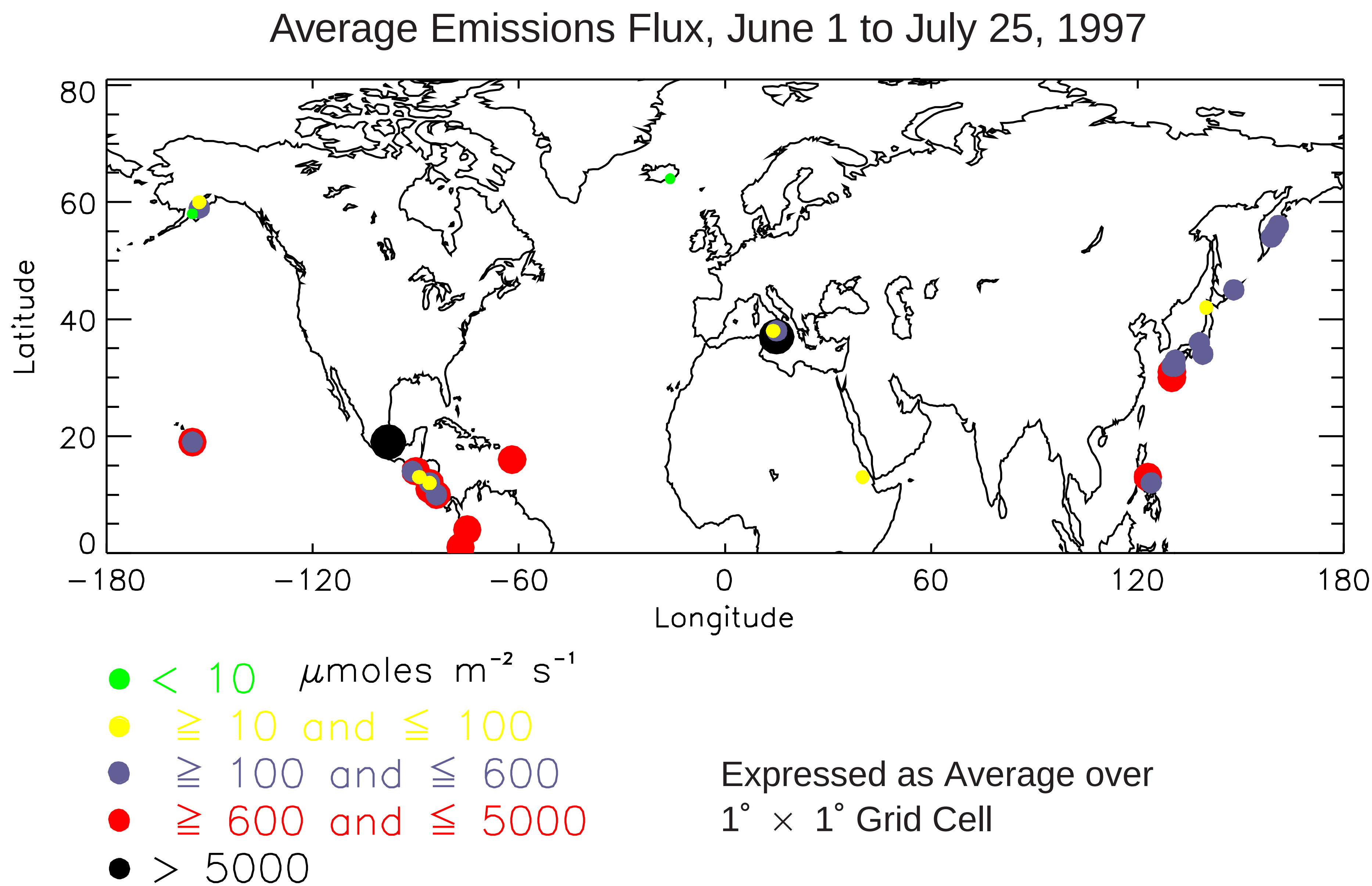
Two Formulations of Cloud Droplet Concentration



Kiehl et al., JGR, in press

VOLCANIC SULFUR EMISSIONS

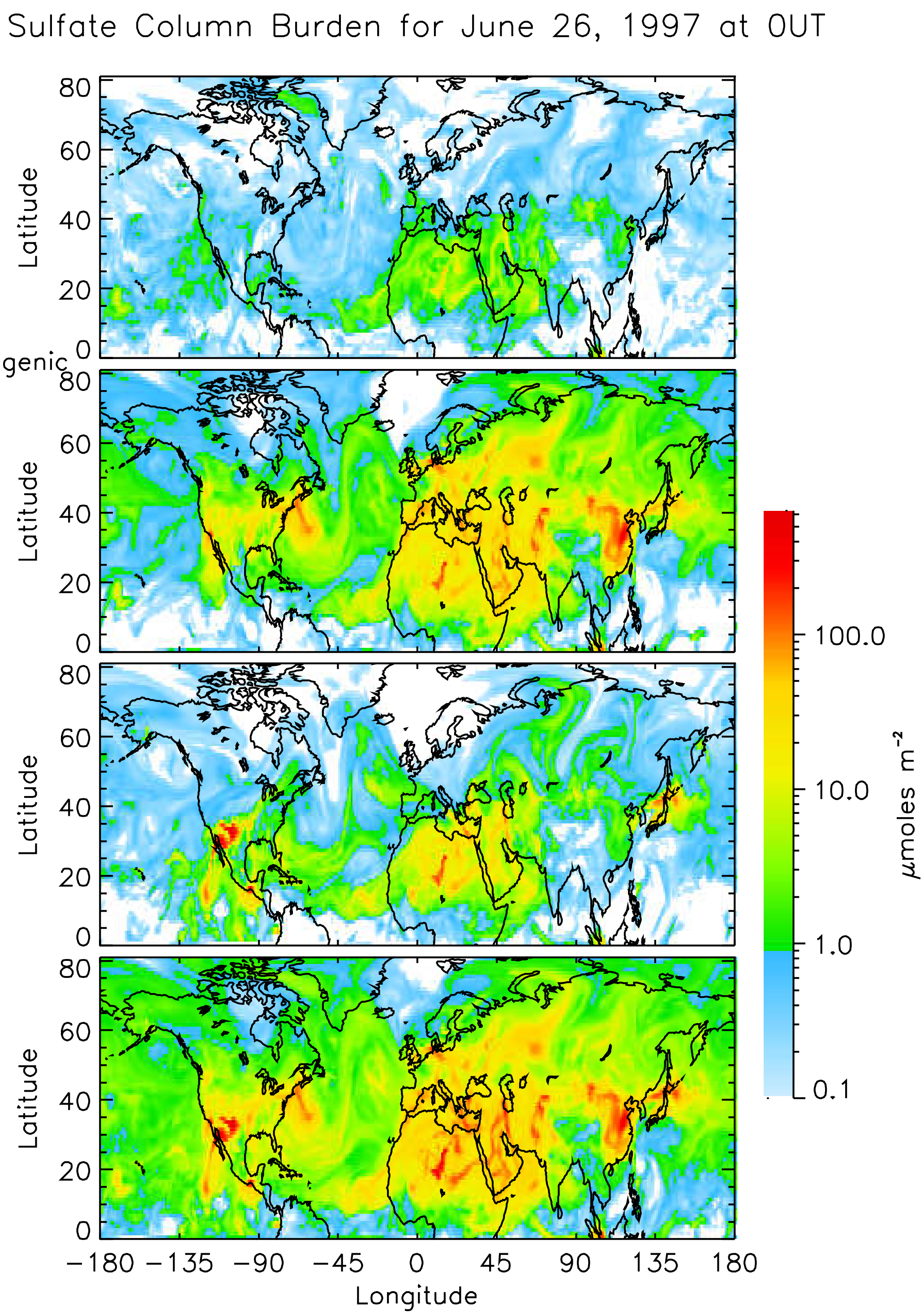
Location and Magnitude during ACE-2 Time Frame



Expressed as Average over
1° × 1° Grid Cell

Volcanos Contribute Disproportionately to Sulfate Burden

Contribution to Emissions	Contribution to Sulfate Burden	
12%	6%	Biogenic
84%	66%	Anthropogenic
4%	28%	Volcanos
		Total



Aerosol Chemical Transport Model GChM-O

Global Chemistry Model Driven by Observation-Derived Meteorological Data

METEOROLOGY
Winds
Clouds
Precipitation
ECMWF
1°× 1°, 27 levels

EMISSIONS
Sulfur species
SO_2
Primary Sulfate
DMS Daily
Seasalt aerosol
Smith, O'Dowd

TRANSPORT
Bott/Easter

CHEMISTRY (Rate Constants *NASA*, 1997)
Gas Phase
$SO_2 + OH \rightarrow H_2SO_4$
DMS + OH DMS chem *Yin/Seinfeld*
$HO_2 + HO_2 \rightarrow H_2O_2$
Aqueous Phase
$SO_2 + H_2O_2 \rightarrow H_2SO_4$
$SO_2 + O_3 \rightarrow H_2SO_4$
O_3, OH, HO_2, Daily ***MOZART*** (*Brasseur et al.*)

AEROSOL MICROPHYSICS
Nucleation
Coagulation
Growth (clear air, cloud)
Method of Moments - McGraw

WET DEPOSITION
Berkowitz/Hales

DRY DEPOSITION
Time, Location dependent
Size Resolved
Wesely

DMS
SO_2
MSA
SO_4^{2-}

Moments
Microphysical Properties
Optical Properties

Version 1
Version 2
Version 3

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.

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)

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

$$\text{Rate} = kmA$$

Aerosol surface area density is

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

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

LIMITATION TO THE METHOD OF MOMENTS

GROWTH LAW RESTRICTION

For exact closure of the moment evolution equations the growth law must be of the form:

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

where *a* and *b* are independent of *r*. Then integral is evaluated as:

$$\int r^{k-1}\phi(r) f(r)dr = a\int r^{k-1} f(r)dr + b\int r^k f(r)dr$$

$$= a\mu_{k-1} + b\mu_k$$

- *This includes the important case of free-molecular growth, for which b = 0.*

- *For other growth laws the* ***Quadrature Method of Moments*** *replaces exact closure with an approximate but much less restrictive closure condition.*

QUADRATURE METHOD OF MOMENTS

INTEGRAL APPROXIMATION VIA n-POINT GAUSSIAN QUADRATURE

- *The moments are evaluated as*

$$\mu_k \equiv \int_0^\infty r^k f(r)dr \cong \sum_{i=1}^{n} r_i^k w_i$$

The abcissas (r_i) and weights (w_i) are specified in terms of the low-order moments of f(r).

- *Quadrature-based closure is obtained by the approximation*

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

MOMENT INVERSION

Low order moments μ_k can be efficiently converted to quadrature abcissas r_i and weights w_i. For 3-point quadrature, 6 moments are required.

- *Once the abcissas and weights have been determined, the unknown distribution function integrals are obtained by the summation.*

McGraw, *Aerosol Sci. Technol.* **27**, 255 (1997)

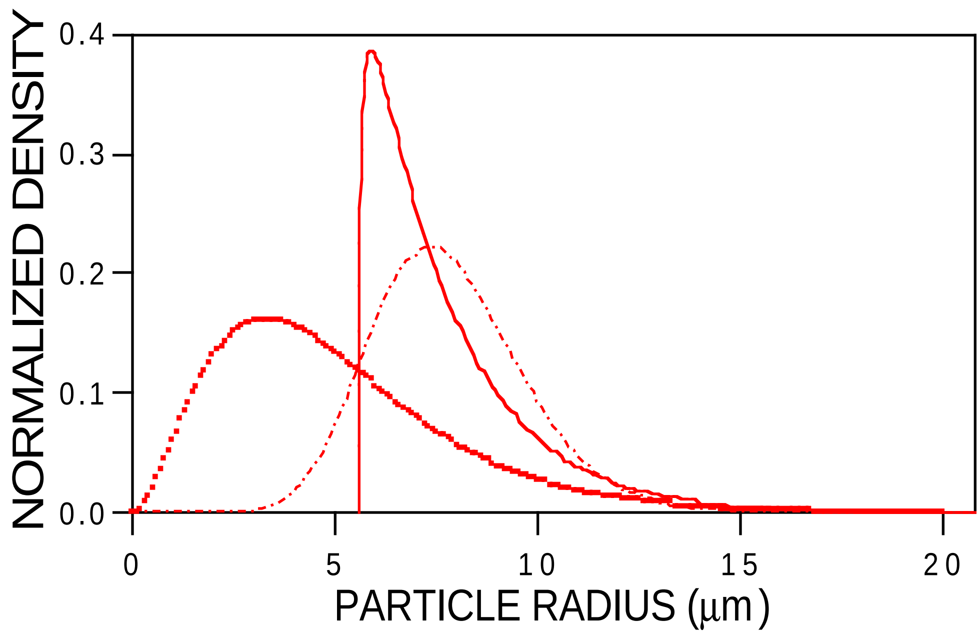
CALCULATIONS FOR DIFFUSION CONTROLLED GROWTH

• *For diffusional growth*

$$dr/dt = k/r$$

the moment evolution equations are not in closed form. One approach has been to use assumed distributions parameterized in terms of moments (e.g. Laguerre).

Diffusion controlled growth of water drops for 20 s at T = 278 K and fixed saturation of 101%.



Dotted curve: Initial normalized Khrgian-Mazin distribution with mean particle radius of 5 µm.

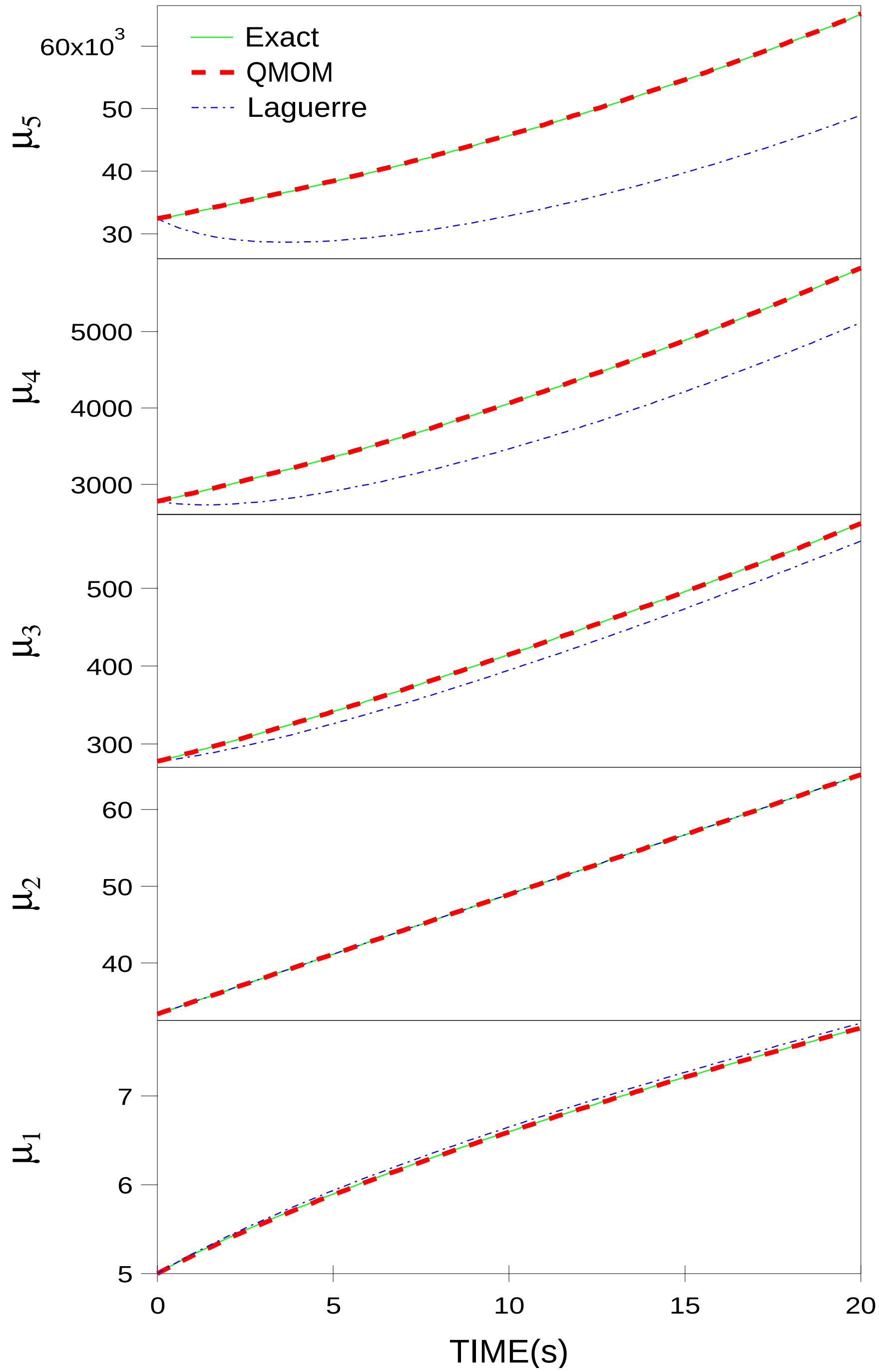
Solid curve: Exact evolved distribution obtained by numerical calculation.

Dashed-dotted curve: Laguerre distribution parameterized by the moments 0 through 2 obtained by the Laguerre closure method.

McGraw, *Aerosol Sci. Technol.* **27**, 255 (1997)

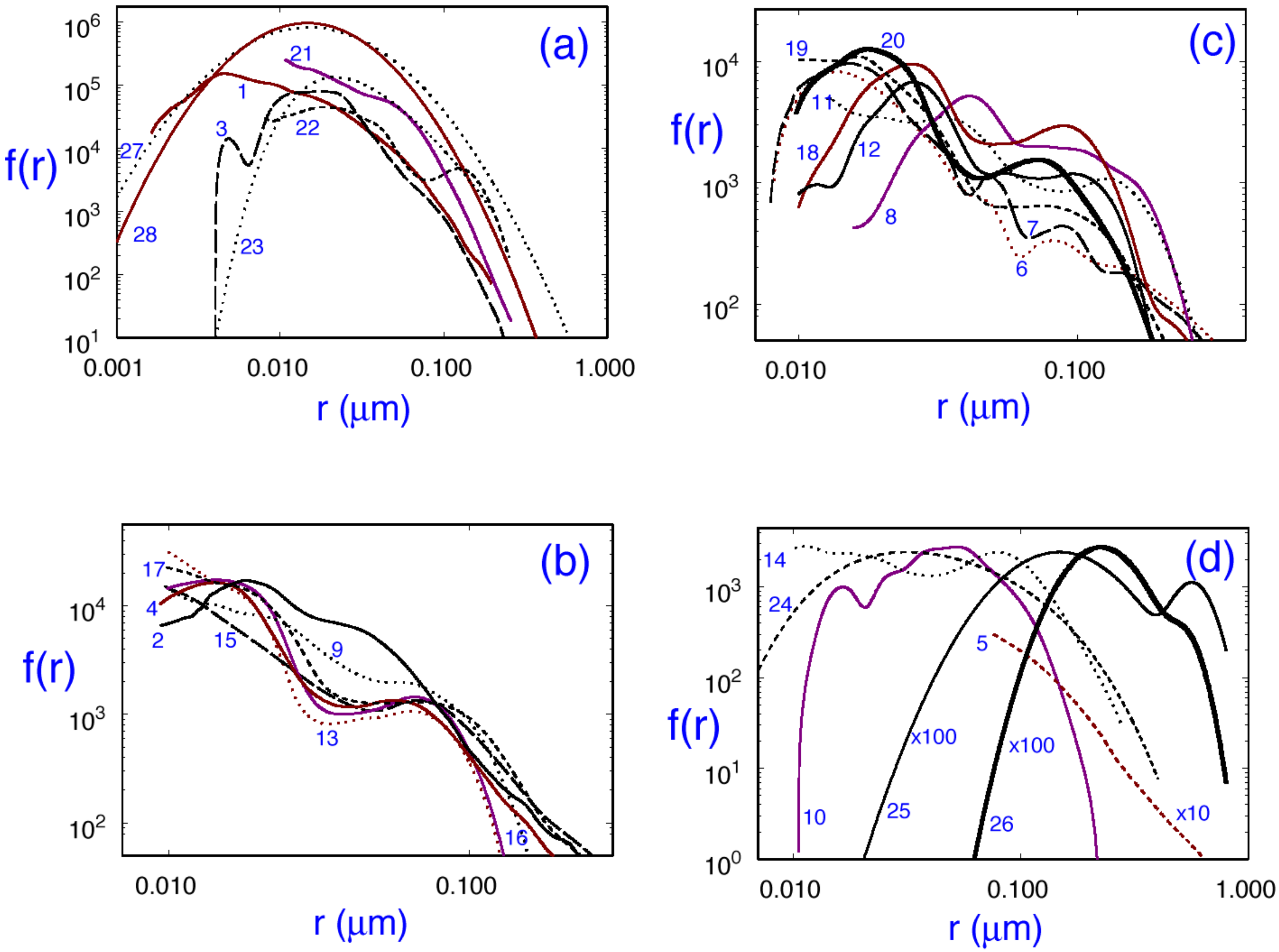
QMOM CALCULATIONS FOR DIFFUSION CONTROLLED GROWTH

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



McGraw, *Aerosol Sci. Technol.* **27**, 255 (1997)

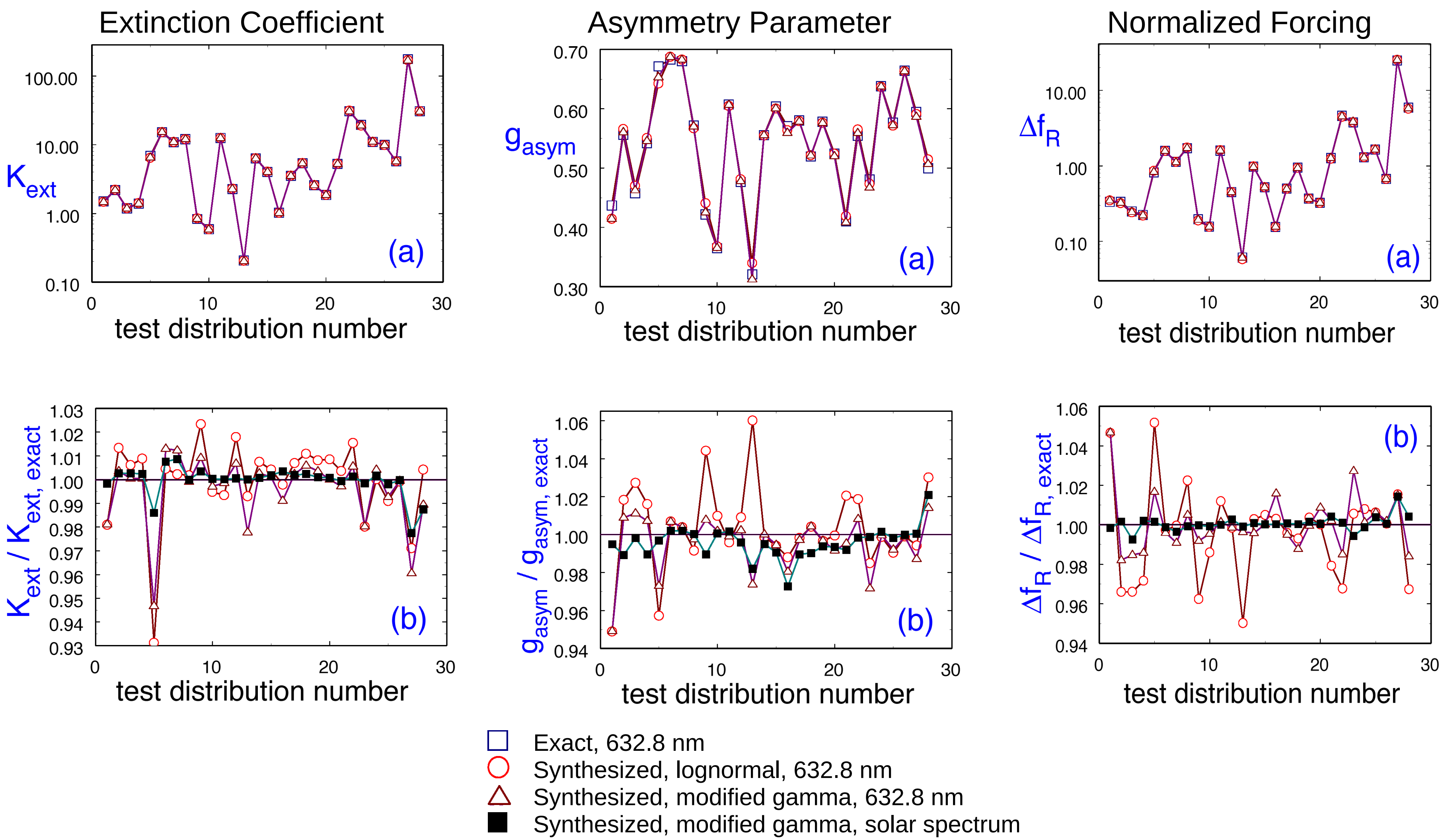
DISTRIBUTIONS USED IN TEST OF RETRIEVAL OF OPTICAL PROPERTIES FROM MOMENTS



D. L. Wright, Jr., *J. Aerosol Sci.*, in press, 1999

SYNTHESIS OF OPTICAL PROPERTIES FROM MOMENTS OF SIZE DISTRIBUTION

Multiple Isomomental Distribution Aerosol Synthesis (MIDAS) Method
(Six moments, μ_0 - μ_5)

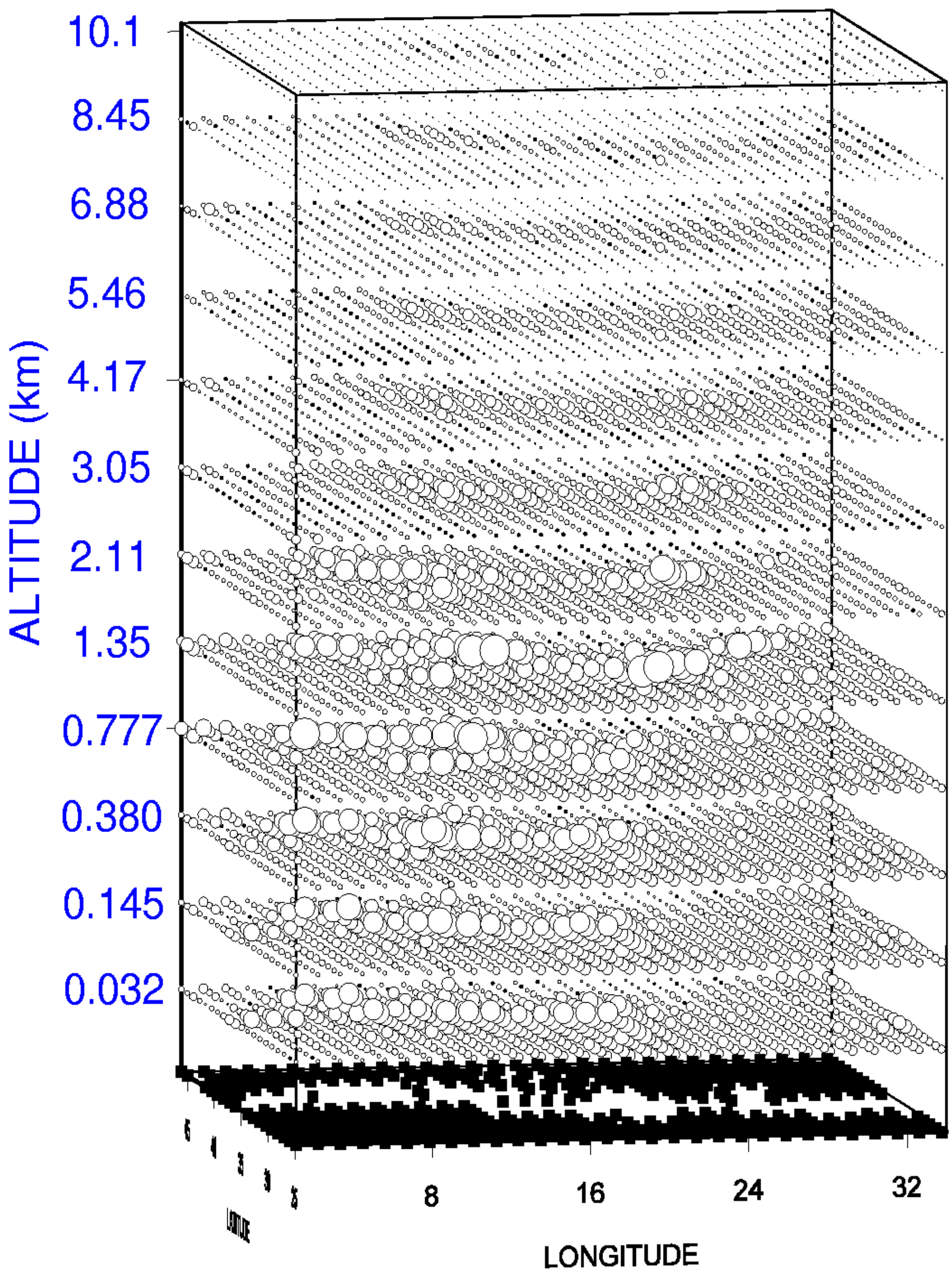


D. L. Wright, Jr., *J. Aerosol Sci.*, in press, 1999

SIZE OF SULFATE PARTICLES

Dependence on Latitude, Longitude, Altitude

Mediterranean Sea, October 15, 1986, 1200 UTC

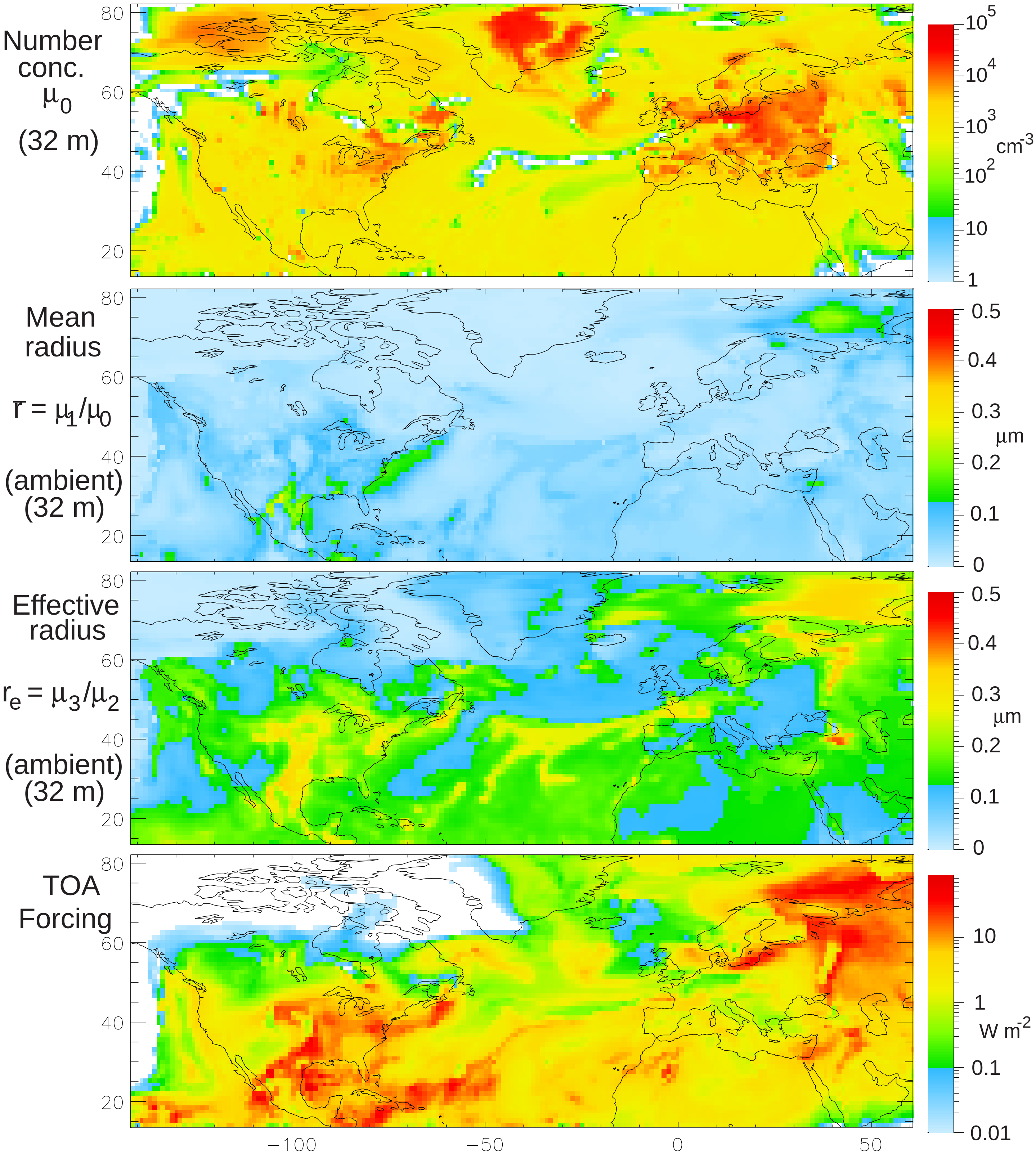


Particle mean radius is proportional to radius of disk, 10-250 nm.

D. Wright, unpublished

AEROSOL SULFATE, October 22, 1986

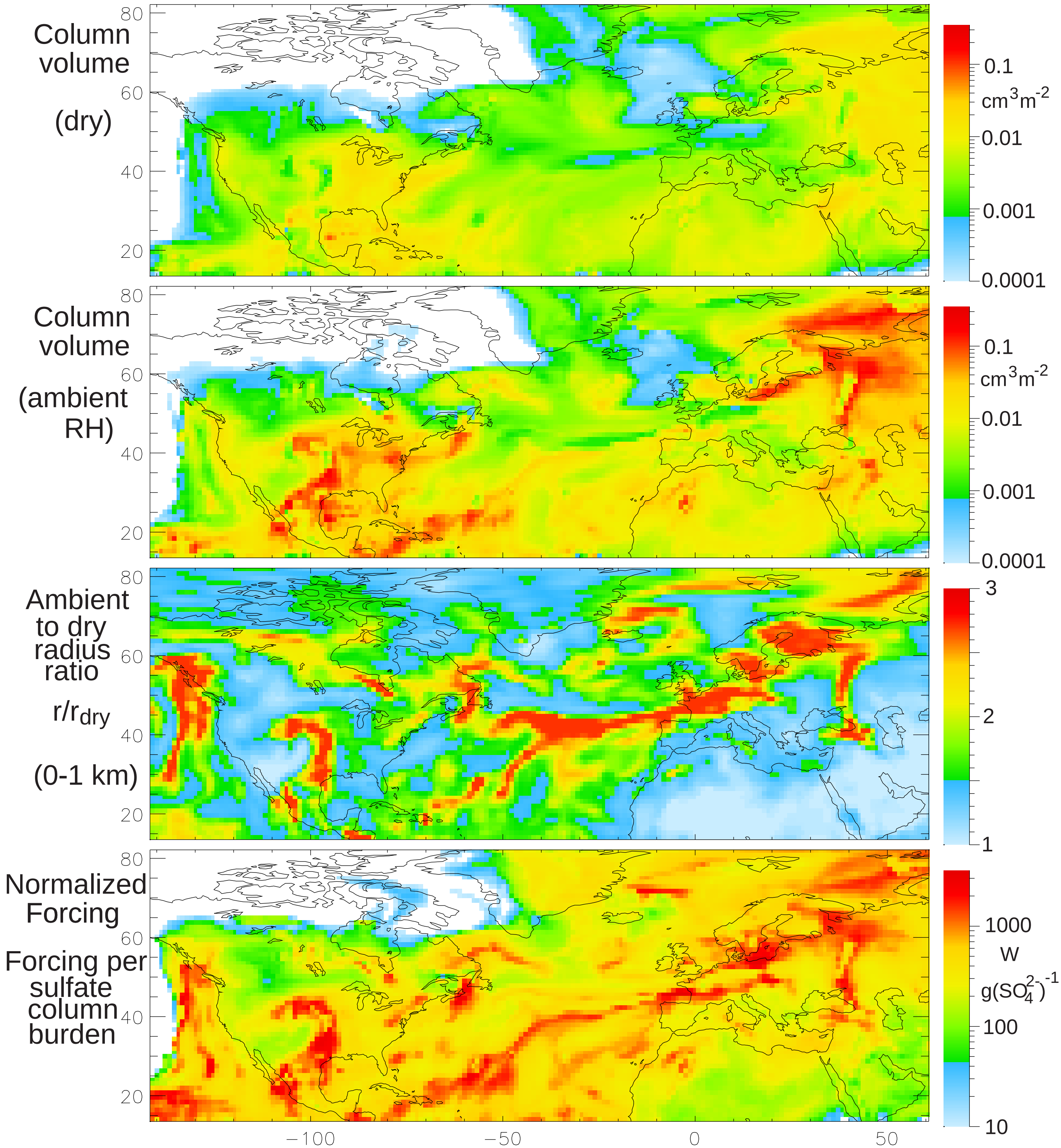
Method of Moments in Subhemispheric Model



Wright, McGraw, Benkovitz & Schwartz, GRL, submitted, 1999

AEROSOL SULFATE, October 22, 1986

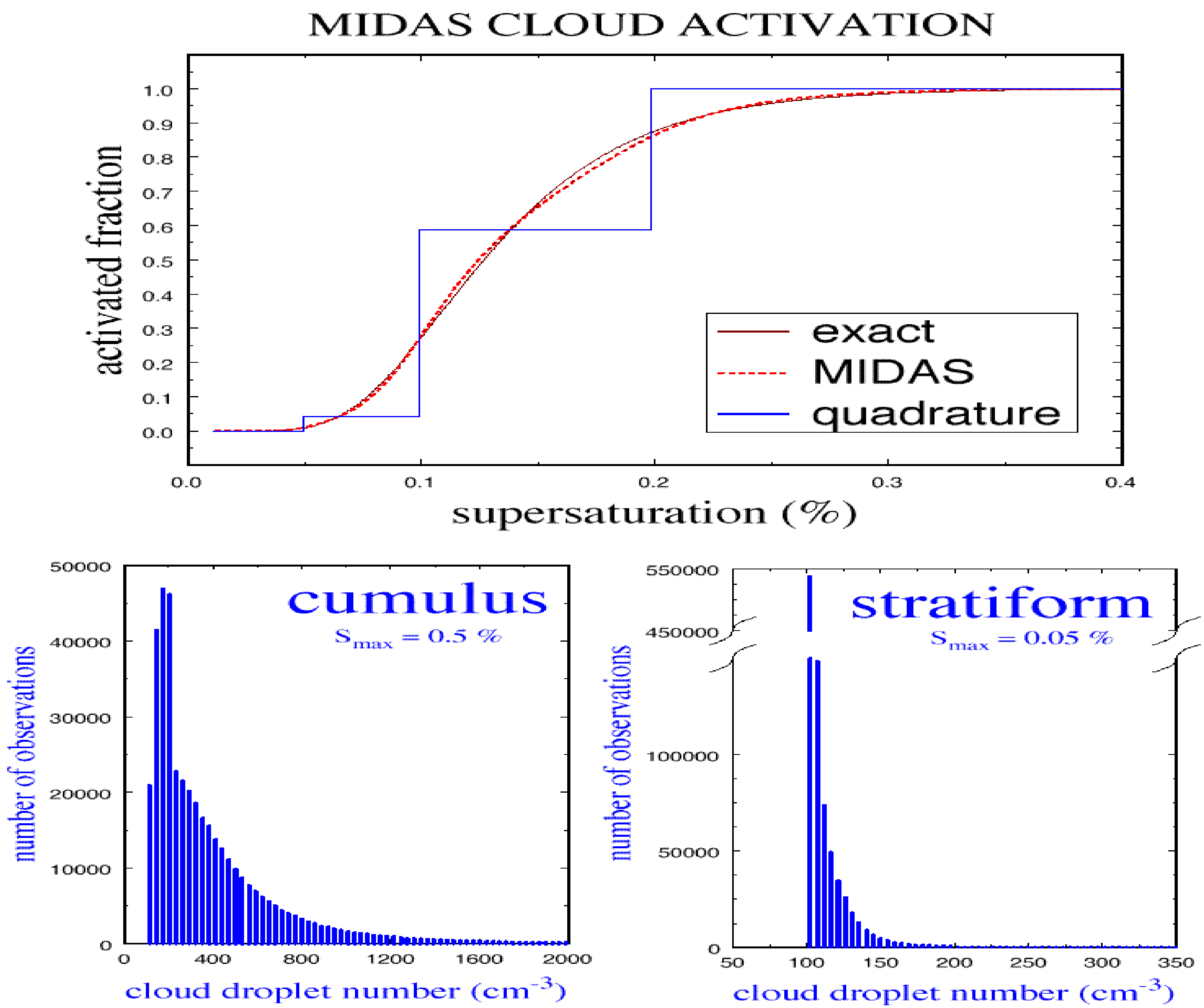
Method of Moments in Subhemispheric Model



Wright, McGraw, Benkovitz & Schwartz, GRL, submitted, 1999

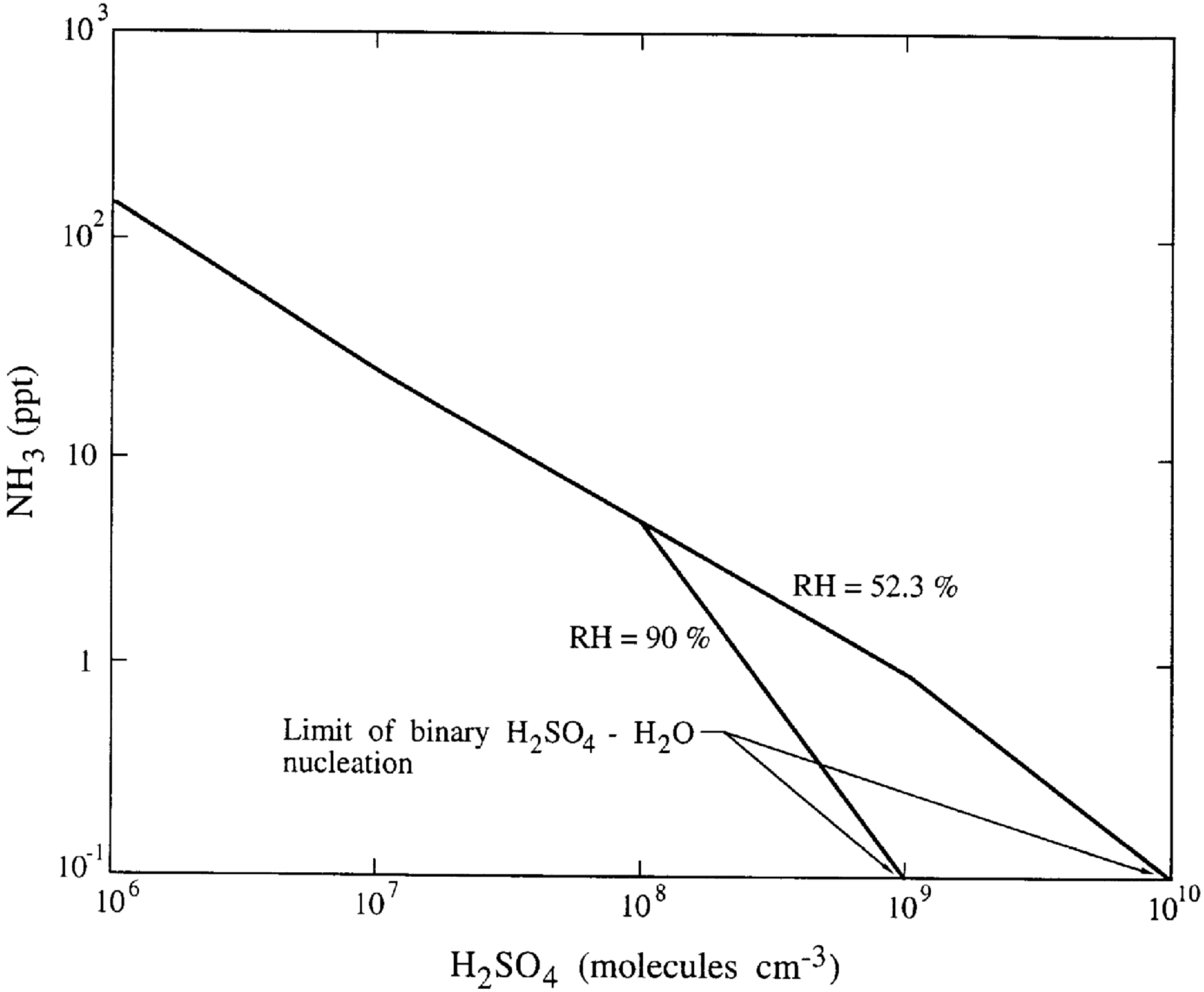
CLOUD DROPLET ACTIVATION FROM MOMENTS OF AEROSOL SIZE DISTRIBUTION

Multiple Isomomental Distribution Aerosol Synthesis (MIDAS) Method



D. L. Wright and R. L. McGraw, to be submitted

AMMONIA INFLUENCE ON SULFURIC ACID NUCLEATION THRESHOLD



Korhonen P., M. Kulmala, A. Laaksonen, Y. Viisanen, R. McGraw, and J.H. Seinfeld, Ternary nucleation of H_2SO_4, NH_3, and H_2O in the atmosphere, *JGR*, accepted, 1999.

COLLABORATIONS

Helsinki, Kuopio, London, Caltech	Examine nucleation mechanisms and rates
NCAR	Incorporate BNL chemistry model in CCM3 to examine direct and indirect forcing
Purdue	Evaluate model by comparison of model output and satellite data
NOAA PMEL	Interpret modeling results during ACE-2 campaign
Caltech, Irvine	Incorporate moment methods into regional aerosol models and compare with alternative approaches
Duke	Incorporate moment method into GFDL GCM to examine aerosol forcing for various scenarios
	Compare moment and bin methods in low-dimensional calculations
	Apply moment methods to evaluation of regional scale PM 2.5
LLNL	Implement aerosol microphysics in LLNL chemical transport model

CURRENT ACTIVITIES/FUTURE PLANS

- GChM-O Version 2 (Sulfate Mass, Enhanced Chem, 360° Long, 0-81° Lat)

  ACE-2 (June 1 - July 25, 1997, Northern Hemisphere). Model runs underway.

  ACE-1 (Nov 1 - December 15, 1995, Southern Hemisphere). Shortly.
- Develop enhancements to representation of aerosol microphysical properties.
- Incorporate additional species: Seasalt, Dust, Carbonaceous
- GChM-O Version 3 (QMOM for Aerosol Properties)

  ACE-2 Sulfate, Seasalt, Dust

  ACE-1 Sulfate and Seasalt
- Collaborate with others in applying these methods.

COMPARISON WITH OBSERVATIONS!