Chemistry and Microphysics of the Troposphere: Chemical and Microphysical Aerosol Model

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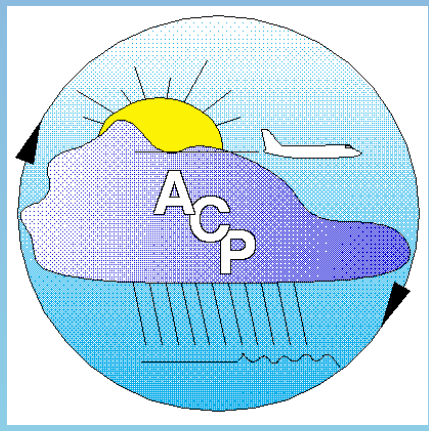


Background

★ **HYPOTHESIS**

#Anthropogenic emissions of SO_2 influence cloud and clear-sky albedo and can thus influence climate.

★ **MECHANISMS**

#Direct: enhancement of clear-sky albedo.

#Indirect: enhancement of average cloud albedo .

★ **PROPERTIES**

#Both forcing and results are highly variable in time and space.

★ **EXISTING CONDITIONS**

#Few observational data are available.

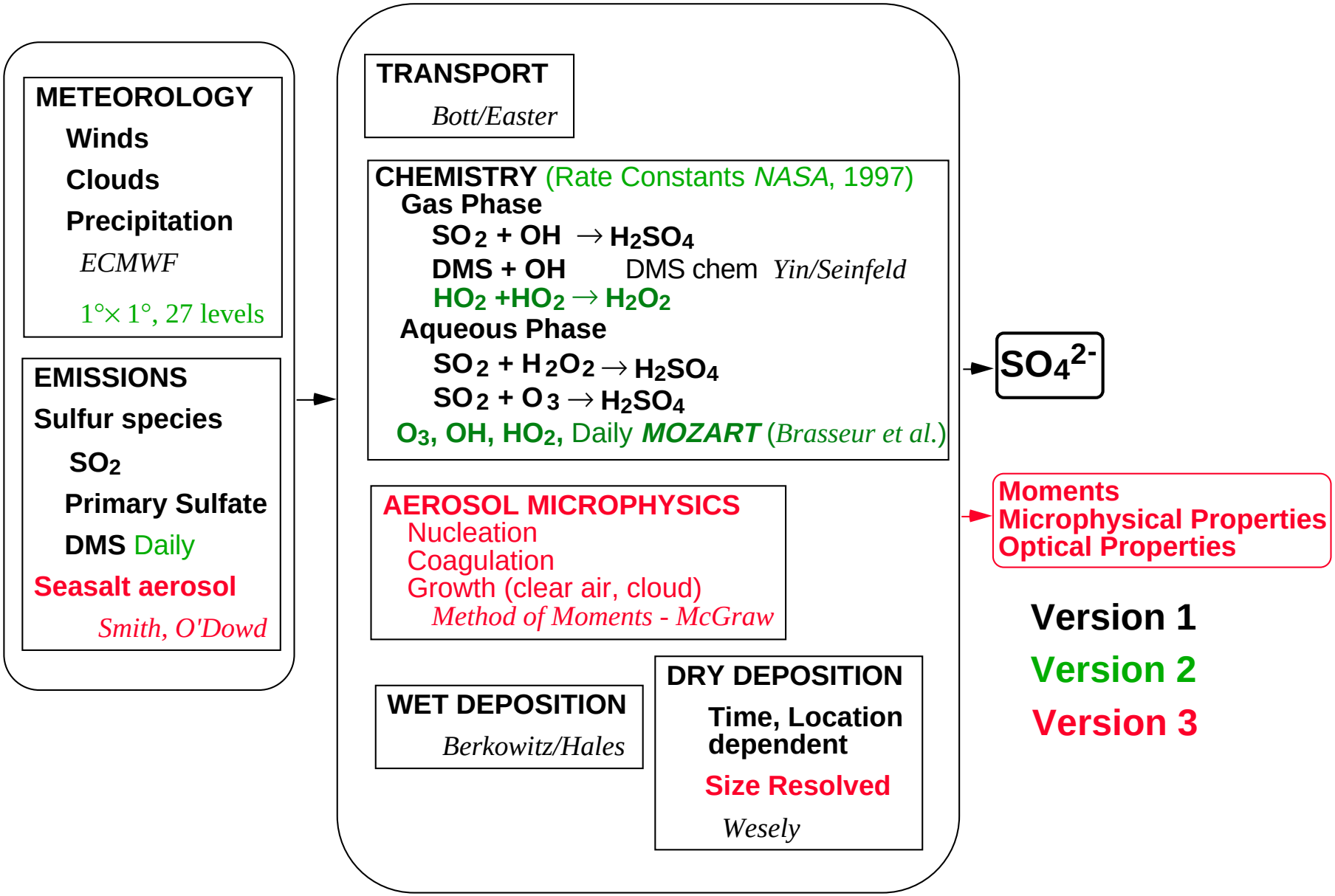
#Consistent estimates need modeling studies.

★ **METHOD OF STUDY**

#Model sulfate concentrations explicitly with high space and time resolution.

Aerosol Chemical Transport Model GChM-O

Global Chemistry Model Driven by Observation-Derived Meteorological Data



GChM-O Version 1

Specifications

★ **Meteorology**

#ECMWF unitialized analyses.

#1.125º x 1.125º horizontal resolution.

#15 vertical levels.

★ **Transport**

#Bott advection scheme as modified by Easter.

★ **Dry deposition**

#Time and location dependent using the methodology of Wesely and the ECMWF meterological data.

★ **Wet deposition:**

Berkowitz/Hales.

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Specifications

★ **Chemistry:**

- # Gas-phase
  - ! SO_2 + OH → Sulfate
  - ! DMS + OH → Sulfate, MSA (Yin/Seinfeld mechanism)
- # Aqueous-phase (precipitating clouds only)
  - ! $SO_2 + H_2O_2$ → Sulfate
  - ! $SO_2 + O_3$ → Sulfate
  - ! Cloud water pH = 4.5
- # Oxidants
  - ! [OH] from Spivakovsky et al.
  - ! Seasonal [H_2O_2] and [O_3]
- # Rate constants from NASA (1991).

Species Represented by the Model

Source ↓	**SO_2**		**Sulfate**			**DMS**	**MSA**
Process →	Primary	Secondary Gas Phase	Primary	Secondary Gas Phase	Secondary Aqueous Phase	Primary	Secondary Gas Phase
Anthropogenic 140°W to 30°W	X		X	X	X		
Anthropogenic 30°W to 60°E	X		X	X	X		
Biogenic		X		X	X	X	X
External		X		X	X		

Primary = directly emitted

Secondary = formed by atmospheric chemistry

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Simulations

★ **Temporal domain**

- #June 28 to July 31, 1986.
- #October 14 to November 15, 1986.
- #January 28 to February 28, 1987.
- #March 28 to April 30, 1987.

★ **Geographic domain**

- #140ºW to 68ºE.
- #12ºN to 81ºN.

★ **Emissions**

- #Anthropogenic SO_2 and primary sulfate
  - ! NAPAP for North America
  - ! EMEP for Europe
  - ! Dignon (1989) for all other areas
- #Biogenic DMS from Bates et al. (1992); ocean DMS distributed using Costal Zone Color Scanner (CZCS).

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Analysis of Model Input and Results

★ **Meterology**

#Wind vector plots.

#Synoptic charts.

★ **Comparison with observations**

#Time series.

#Histograms.

★ **Graphics**

#Maps of sulfate column burdens.

#Time series of relative contribution of sources.

★ **Autocorrelation studies**

#1/e decay of temporal and spatial autocorrelation.

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Analysis of Model Input and Results

★ **Animations**

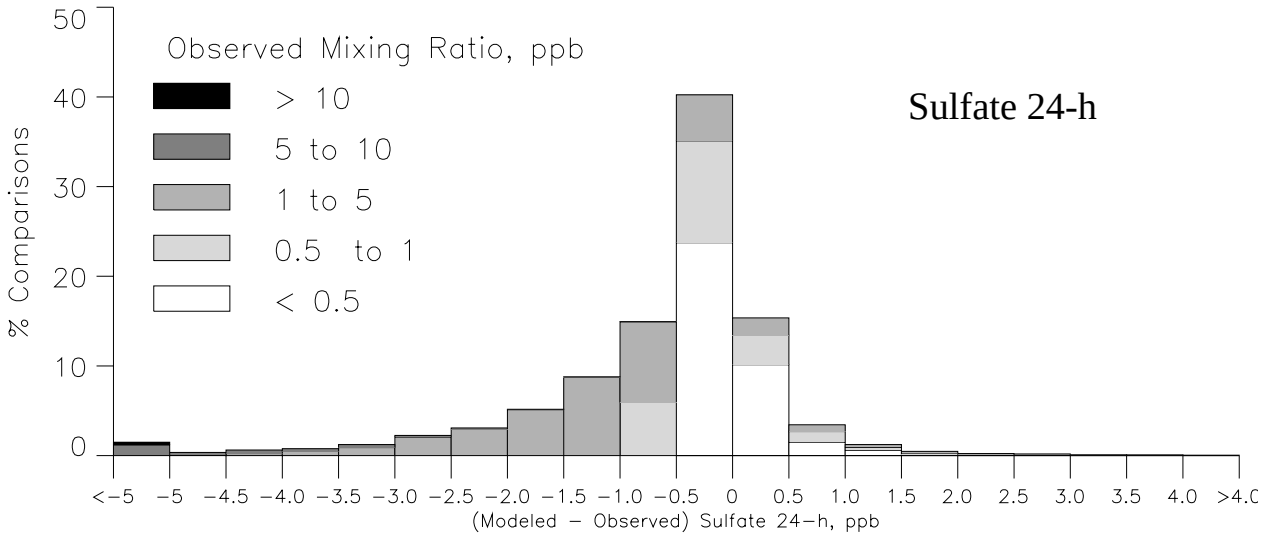
- #Animations of the development of the sulfate column burden.
- #Web site: *http://www.ecd.bnl.gov/sulfate_model.html*

★ **Analysis of meteorological influences on sulfate column burdens.**

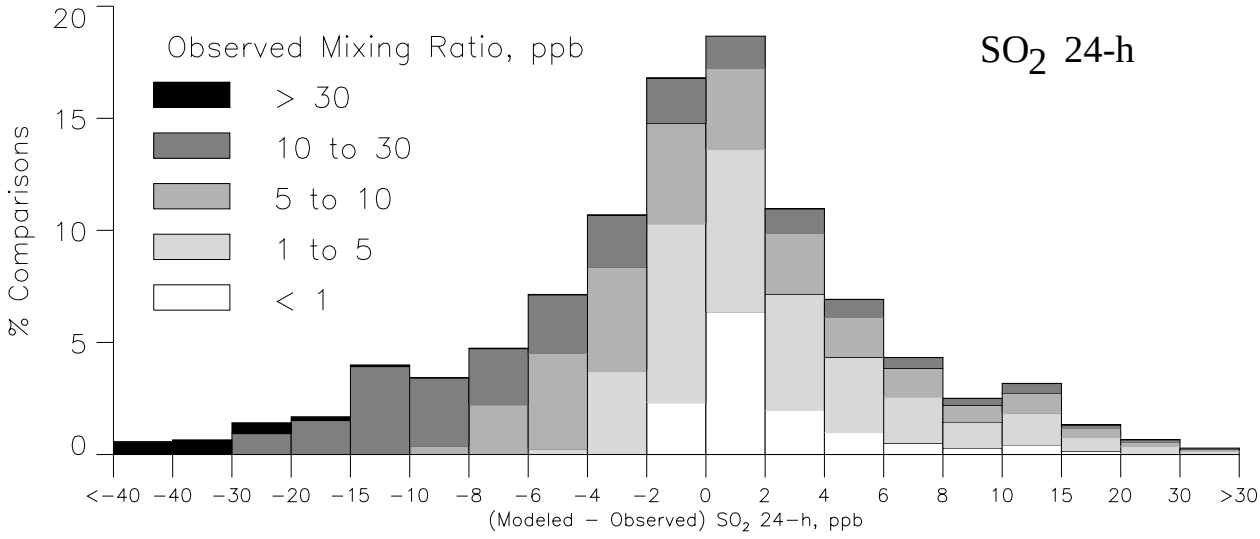
★ **References**

- #Benkovitz et al., JGR 1994.
- #Benkovitz & Schwartz, JGR 1997.
- #Barth et al., JGR (submitted).
- #Rasch et al., JGR (submitted).
- #Benkovitz et al., (in preparation).

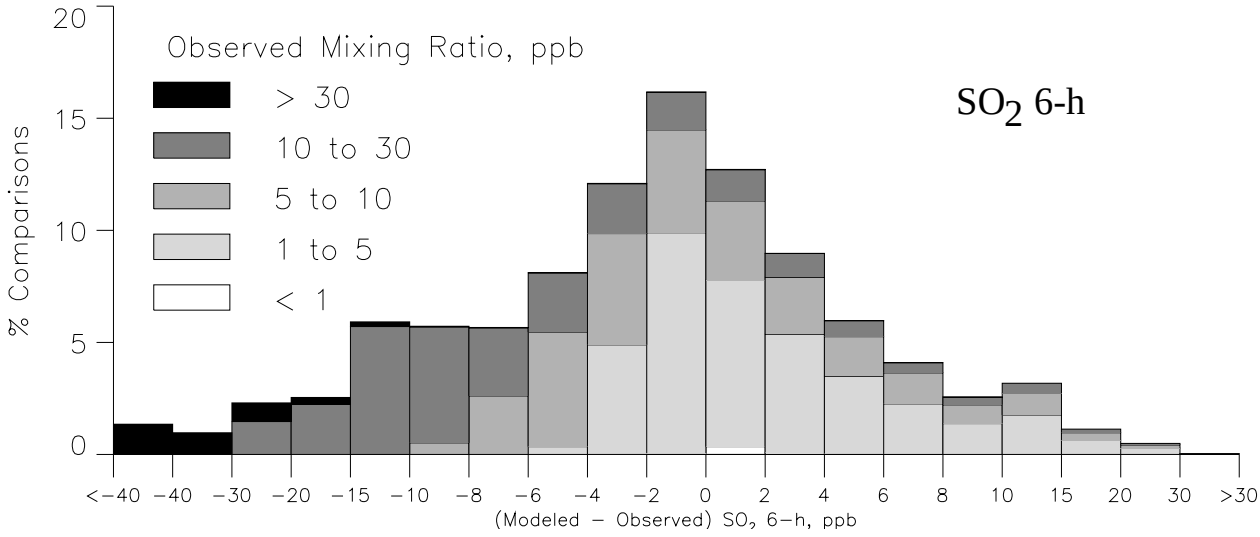
Comparison of Modeled and Observed Surface Mixing Ratios



Sulfate 24-h

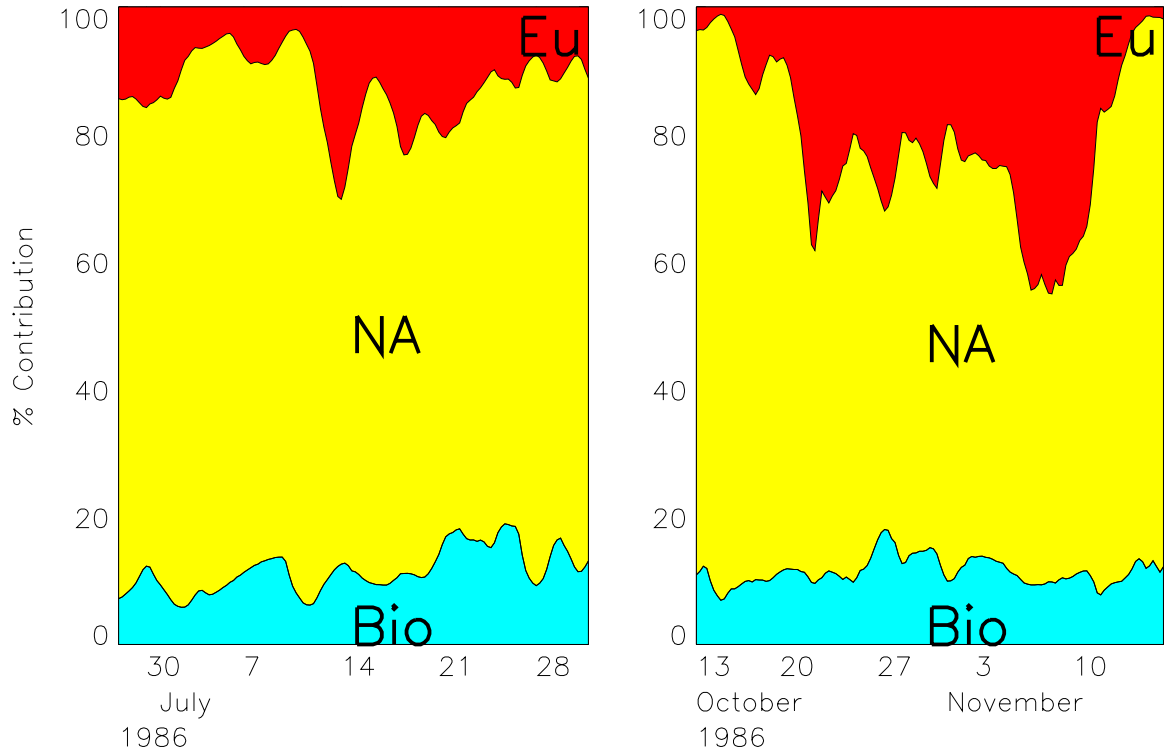


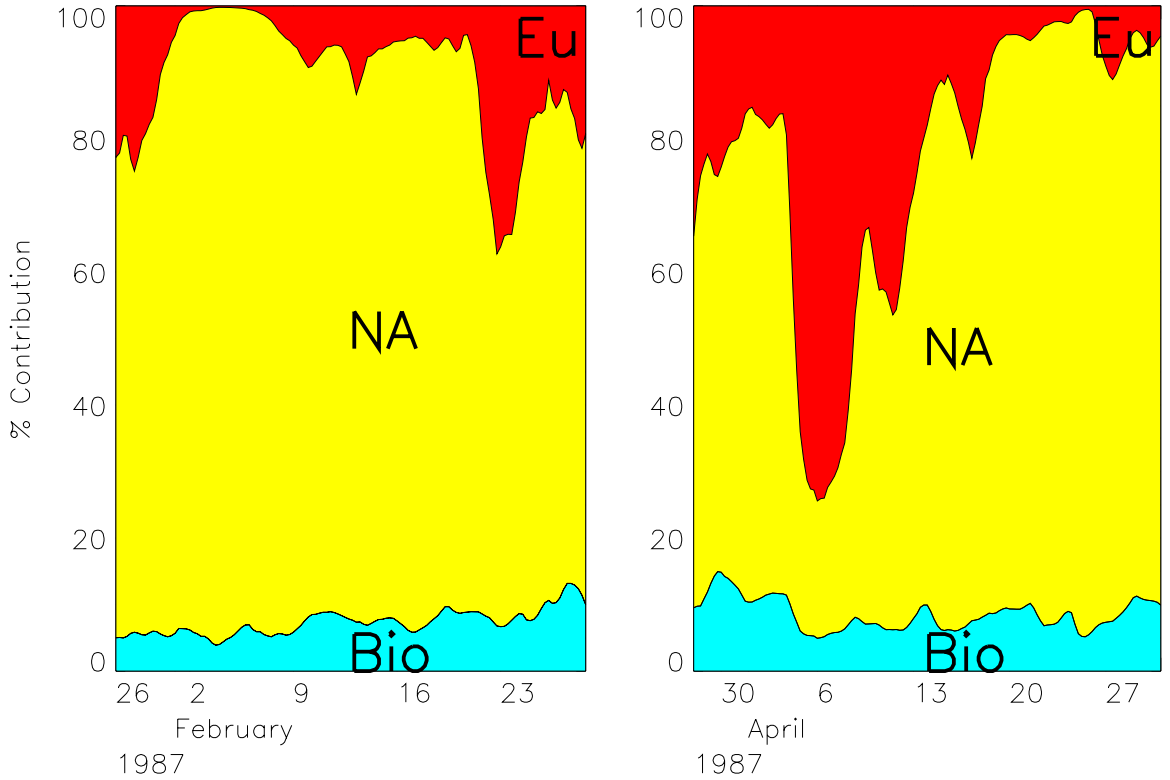
SO_2 24-h

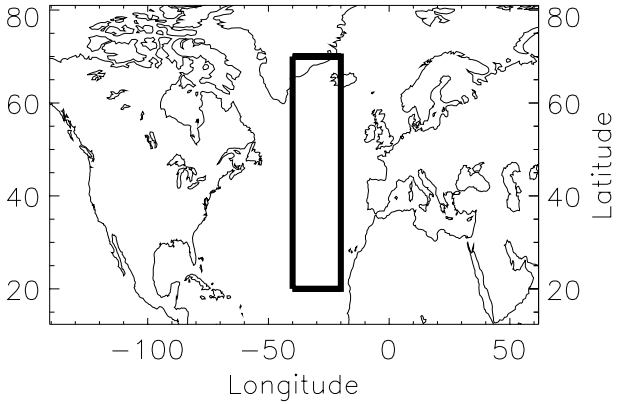


SO_2 6-h

Contribution of Sources to Sulfate Burden

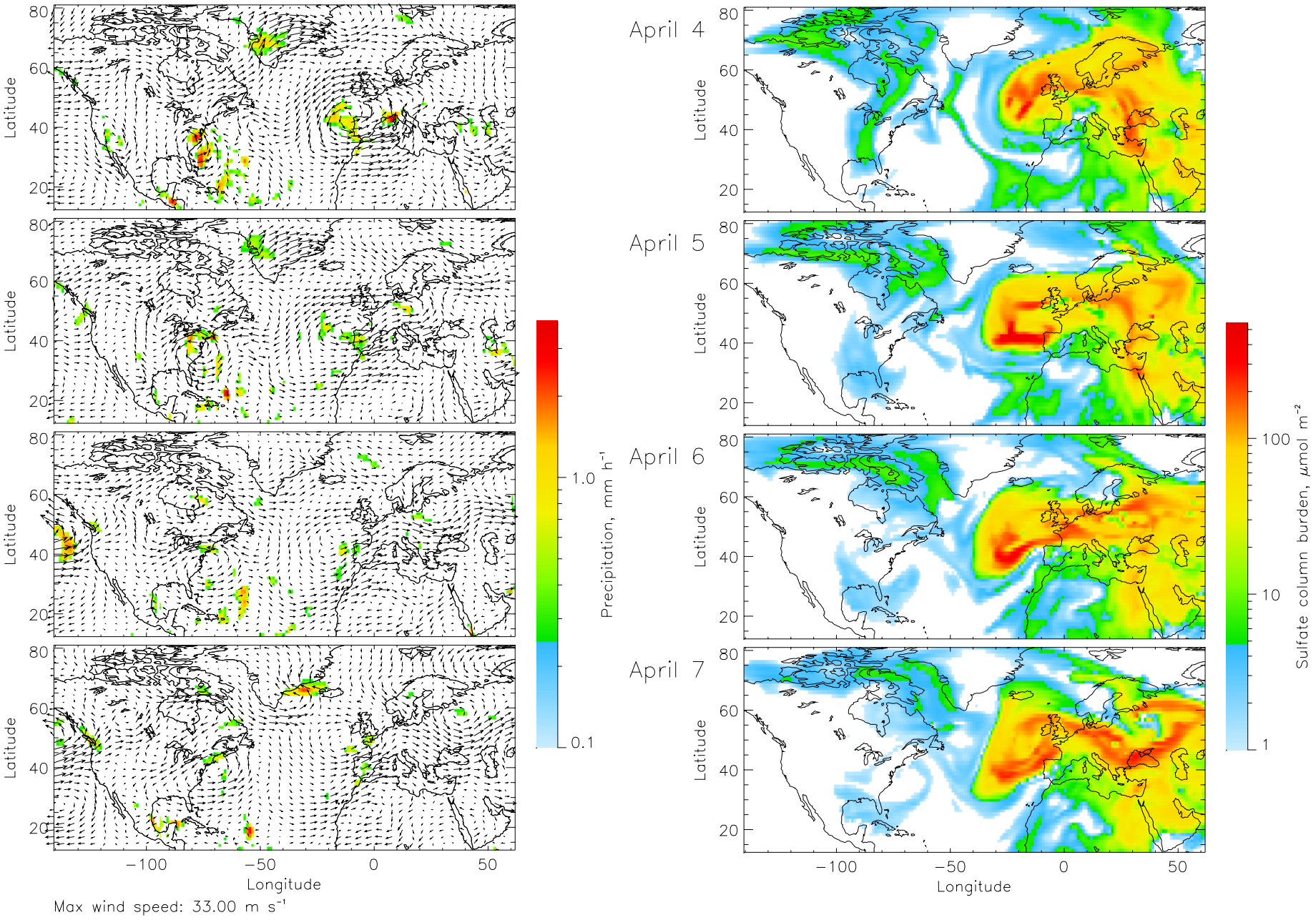






Bio=biogenic sources
NA=anthropogenic sources
west of 30W
Eu=anthropogenic sources
east of 30W

Evolution of the European Contribution to Sulfate Column Burden
April 4 to 7, 1987



Max wind speed: 33.00 m s^{-1}

Winds at ~ 850 hPa.

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★ **Meteorology**

#ECMWF

! Initialized analysis surface and upper air data.

! Surface forecast data (precipitation and radiation).

#1° x 1° horizontal resolution.

#27 vertical levels.

★ **Transport**

#Bott advection scheme as modified by Easter.

★ **Dry deposition**

#Time and location dependent using the methodology of Wesely with the ECMWF meterological data.

★ **Wet deposition**

#Berkowitz/Hale.

GChM-O Version 2

★ Chemistry

#Gas phase

- ! SO_2 + OH → Sulfate
- ! DMS + OH → Sulfate, MSA (Yin/Seinfeld mechanism)
- ! HO_2 + HO_2 → H_2O_2 (limited by MOZART mixing ratios)

#Aqueous phase (all clouds)

- ! SO_2 + H_2O_2 → Sulfate
- ! SO_2 + O_3 → Sulfate
- ! Cloud water pH estimated assuming sulfate is present as NH_4HSO_4 + the contribution of MSA.

#Mixing ratios of HO_2, OH, H_2O_2, and O_3 from MOZART model (Brasseur, JGR 103, 1998)

#Rate constants from NASA (1997)

Species Represented by the Model

Source ↓	**SO_2**		**Sulfate**			**DMS**	**MSA**
Process →	Primary	Secondary Gas Phase	Primary	Secondary Gas Phase	Secondary Aqueous Phase	Primary	Secondary Gas Phase
Anthropogenic 140°W to 30°W	X		X	X	X		
Anthropogenic 30°W to 60°E	X		X	X	X		
Anthropogenic 60°E to 140°W	X		X	X	X		
Volcanic	X			X	X		
Biogenic		X		X	X	X	X
External		X		X	X		

Primary = directly emitted

Secondary = formed by atmospheric chemistry

GChM-O Version 2

Simulations

★ **Temporal Domain**

- November 1 to December 15, 1995 (ACE-1).
- June 1 to July 25, 1997 (ACE-2).

★ **Geographic domain**

- Southern Hemisphere to 81ºS (ACE-1).
- Northern Hemisphere to 81ºN (ACE-2).

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Emissions

★ **Anthropogenic SO_2**

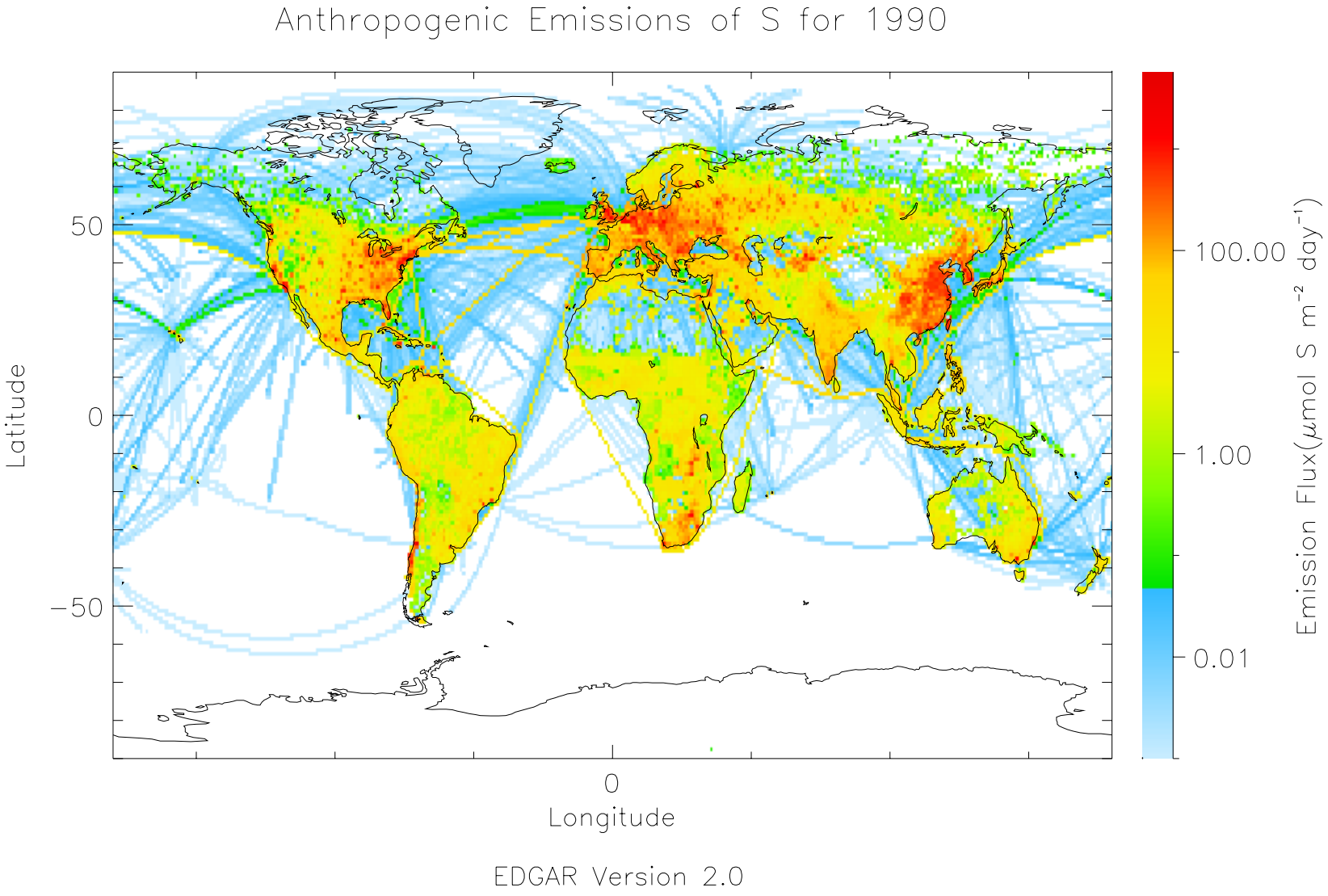
- ! EDGAR 1990 inventory, Olivier et al., 1995.
- ! Regional emissions for Australia (F. Carnovale) and New Zealand (H. Plume, personal communication).

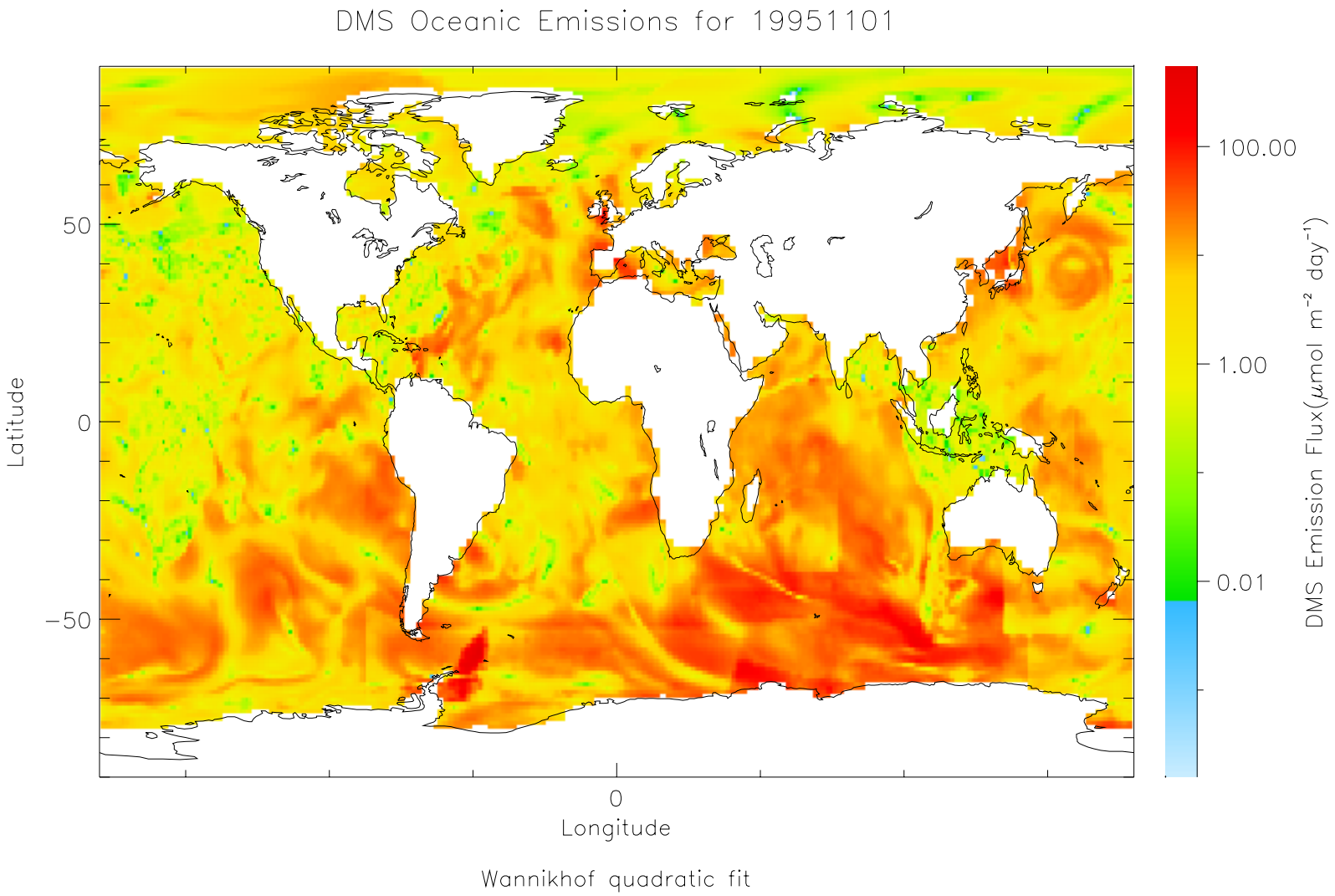
★ **Biogenic DMS**

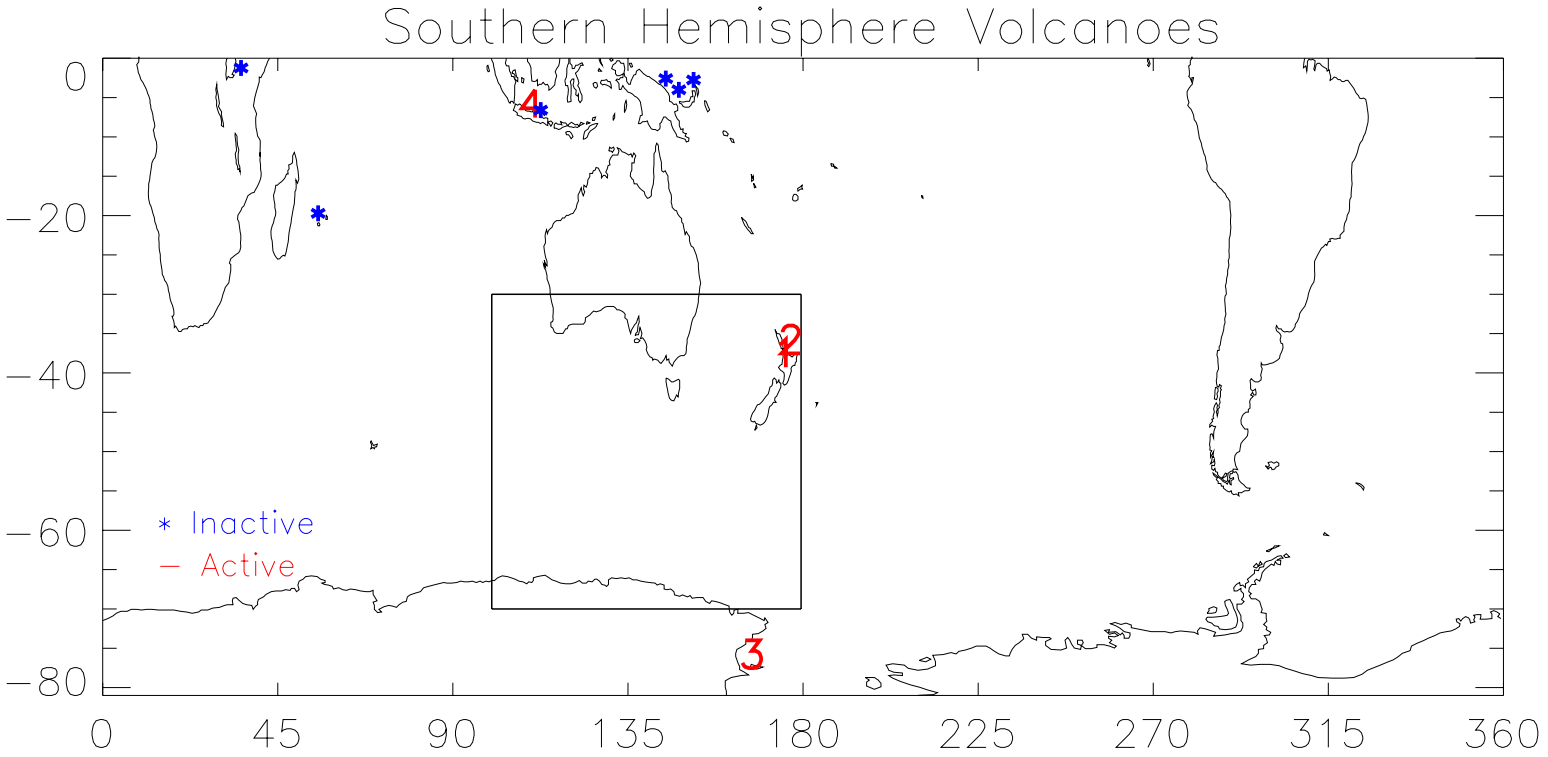
- ! Oceanic: Kriging ocean measurements from Kettle et al., 1996. Kriging ACE-1 measurements to improve values in the ACE-1 experimental region. Kriging ACE-2 measurements to improve values in the ACE-2 experimental region.
- ! Land: B. Lamb (personal communication).

★ **Volcanic SO_2 compiled at BNL.**

- ! Daily for November 1 to December 15, 1995.
- ! Daily for June 1 to July 25, 1997.


Anthropogenic Emissions of S for 1990
50
0
−50
Latitude
0
Longitude
100.00
1.00
0.01
Emission Flux(μmol S m⁻² day⁻¹)
EDGAR Version 2.0

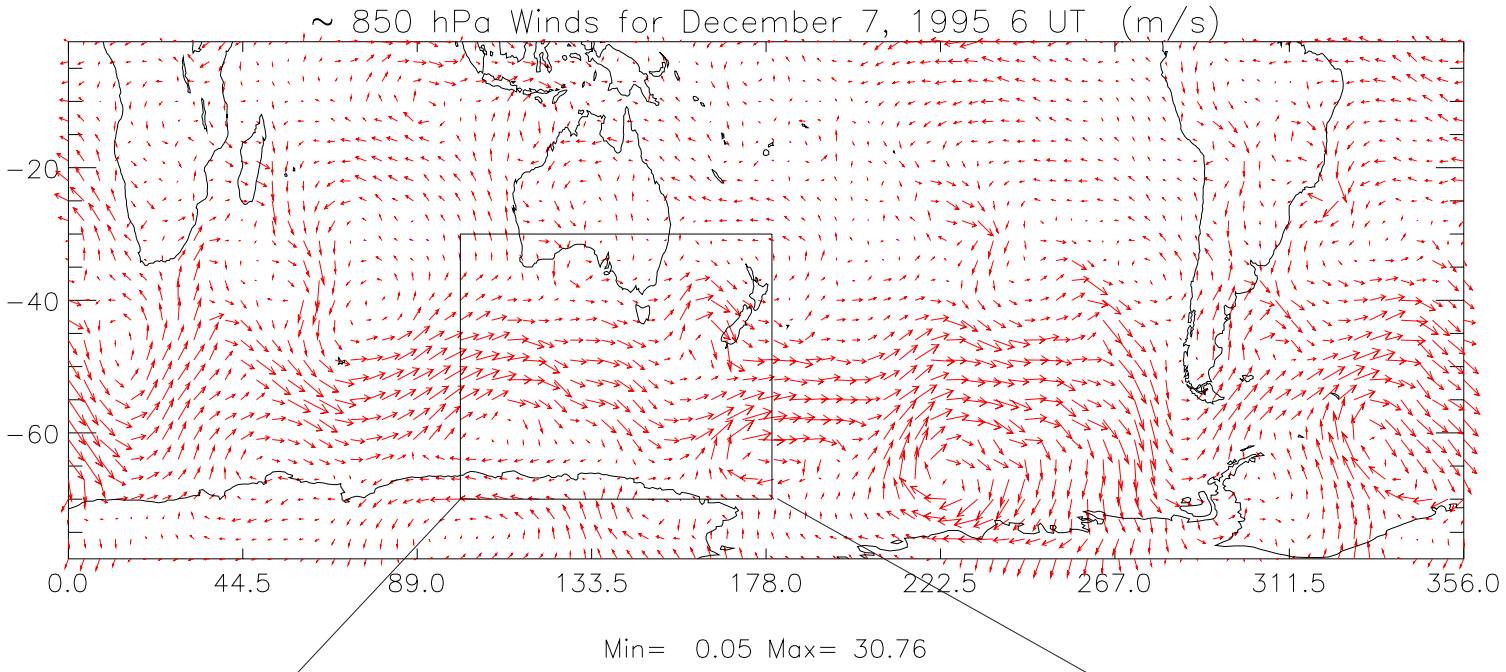

DMS Oceanic Emissions for 19951101
Latitude
50
0
−50
0
Longitude
100.00
1.00
0.01
DMS Emission Flux(μmol m⁻² day⁻¹)
Wannikhof quadratic fit

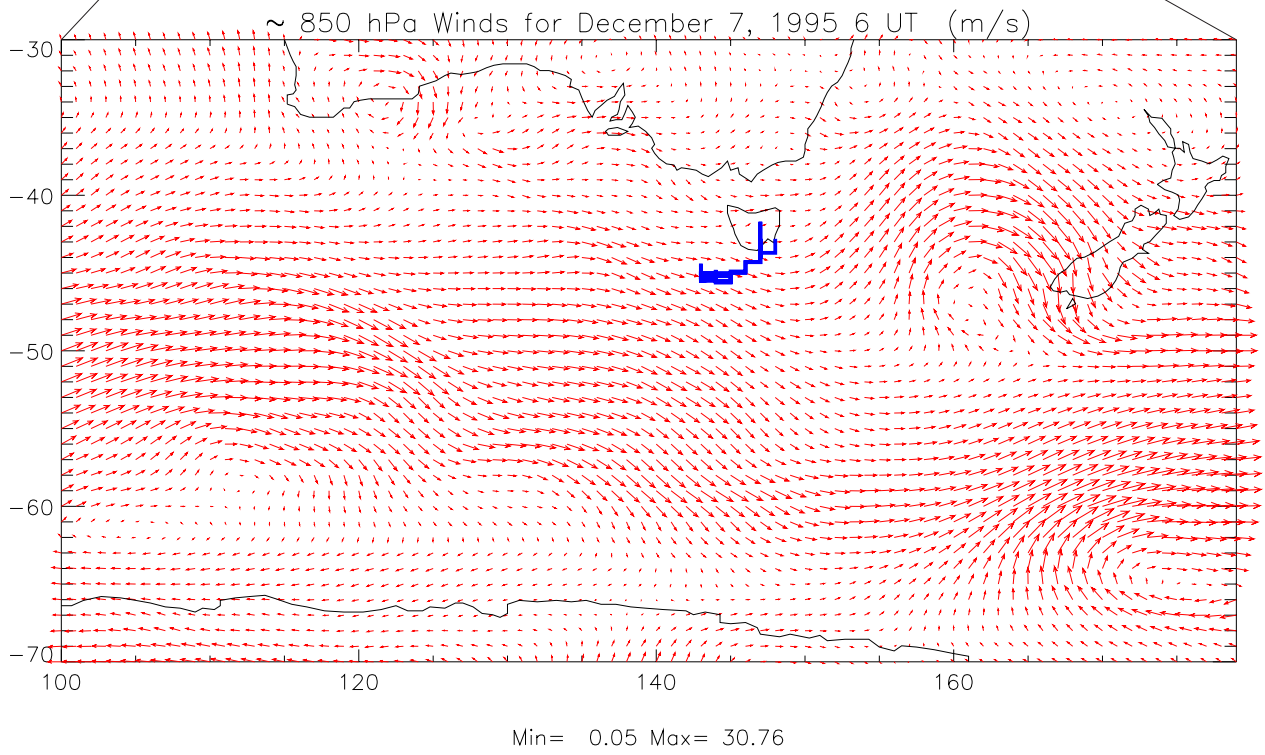


Emissions for Nov 1 to Dec 31, 1995

	Name	10^6 g SO_2
1	Ruapehu	111010
2	White Island	19093
3	Erebus	3355
4	Merapi	5020

ACE-1
Location of Flight 24
December 7, 1995





Flight 24

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Incorporation of Aerosol Microphysical Processes

★ **Quadrature Method of Moments (QMOM)**

- #Full Shugard- Heist-Reiss (SHR) model for binary nucleation of H_2SO_4-water aerosol.
- #Aerosol growth laws of arbitrary form.
- #Closure of aerosol moment evolution equation via QMOM.

★ **Aerosol dynamics**

- #Supplementation of QMOM with Lagrangian interpolation method.
- #Multicomponent nucleation of NH_3-H_2SO_4-water.
- #Coagulation dynamics.

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Incorporation of Aerosol Microphysical Processes

★ **Coupled aerosol and cloud processes**

Explicit treatment of the size of cloud droplets pertinent to indirect forcing.

Model will now include two coupled sets of six moments each: one set for aerosol and one set for the cloud distribution.

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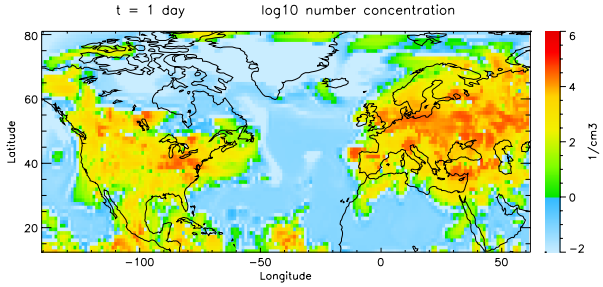
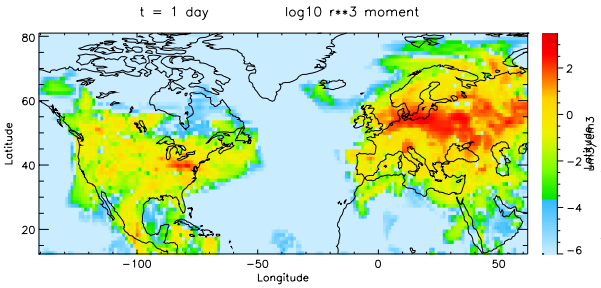
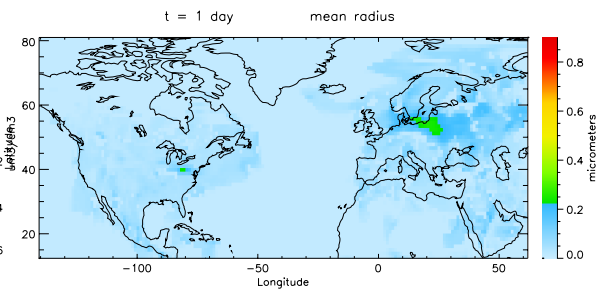
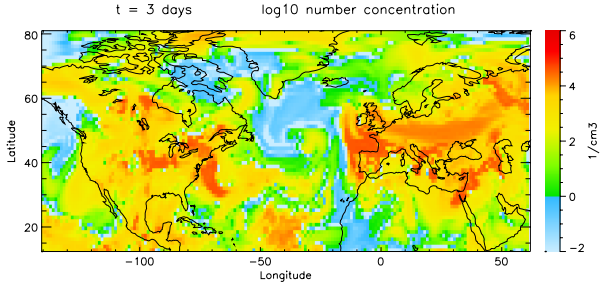
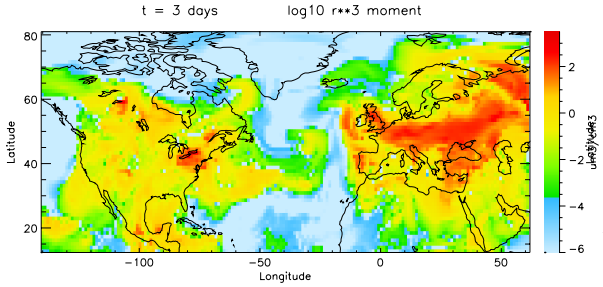
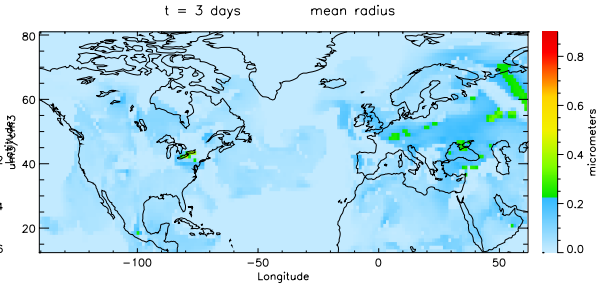
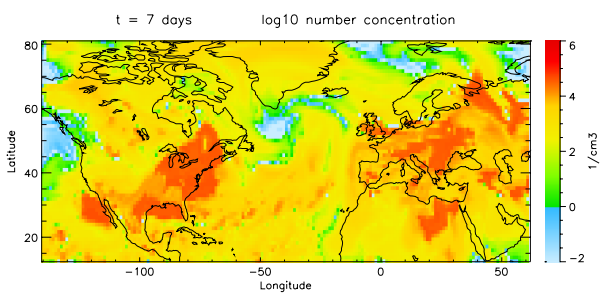
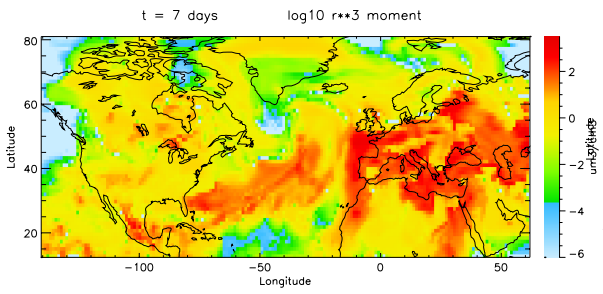
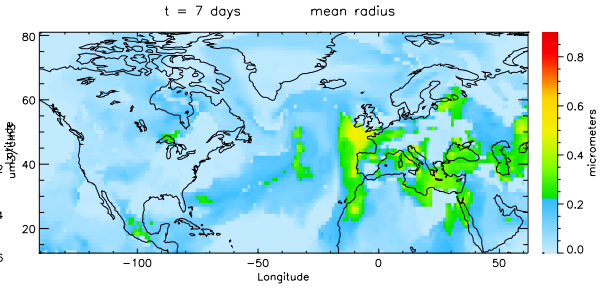
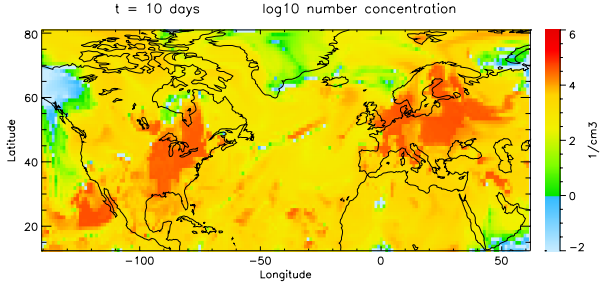
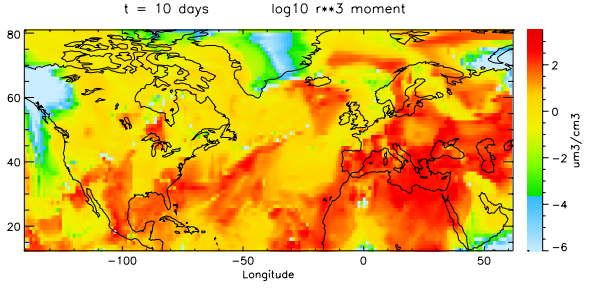
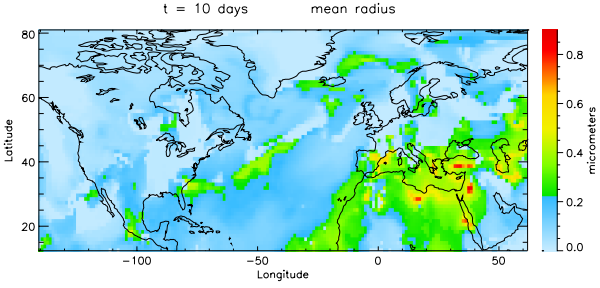
Ten-Day Simulation Includes

- ★ Sulfate and sea salt aerosols.
- ★ Water uptake and release with changing RH.
- ★ Nucleation of H_2SO_4/water aerosol via Jaecker-Voirol &Mirabel (1989).
- ★ Sea salt source via Smith et al. (1993).
- ★ Condensational growth.
- ★ Coagulation.
- ★ Size-resolved dry deposition.
- ★ Wet removal in precipitating clouds.

Results of a Ten Day Simulation

	Log_{10} Number Concentration	Log_{10} Third Moment	Mean Radius
Day 1			
Day 3			
Day 7			
Day 10			

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Retrieval of Aerosol Optical Properties From Moments of The Particle Size Distribution

★ **The Multiple Isomomental Distribution Aerosol Surrogate (MIDAS) Technique**

- **Retrieves families of isomomental surrogate distributions for the true size distribution from its lower moments.**
- **Each distribution is a multimodal lognormal or modified gamma distribution.**
- **For retrieved lognormals, the MIDAS technique 'broadens' the abscissas obtained from Gaussian quadrature into 'modes' of finite width.**
- **Optical properties are computed from an average of the retrieved distributions.**

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Reduced forcing per unit volume ($\mu m^2\ cm^{-3}$) is defined as:

$$\Delta f_r = p \int r^2 Q_{sca}(r) b_U(r, q_0) f(r) dr$$

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Figure Caption.

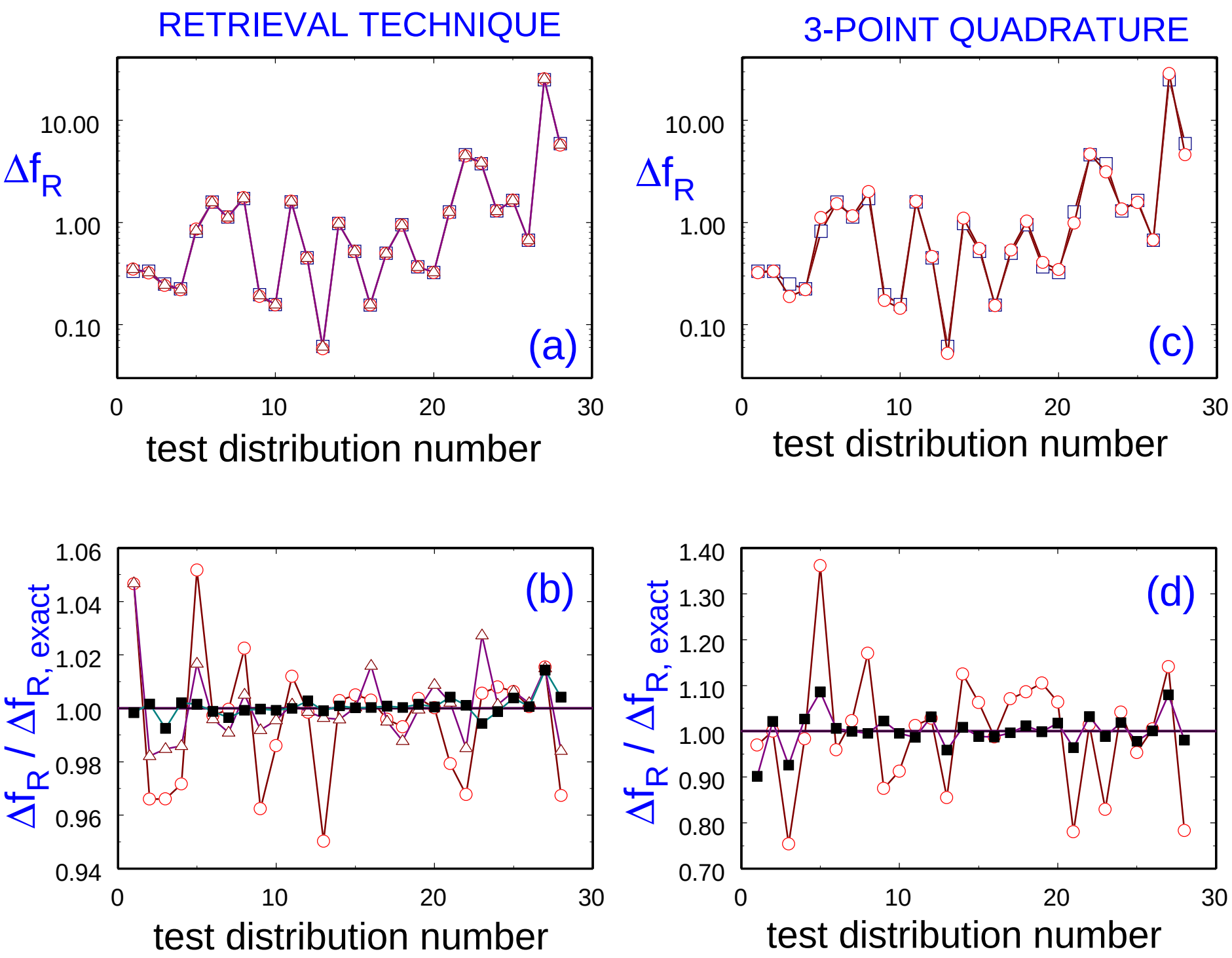
Reduced forcing per unit volume ($\mu m^2\ cm^{-3}$) for each of the test distributions.

(a) **Q = exact results, " = retrieved lognormal results, Δ = retrieved modified gamma results, at λ = 632.8 nm and *n* =1.55-0 *i*.**

(b) **Ratio of retrieved result to exact result. " = lognormals, Δ = modified gammas, # = modified gammas averaged over the solar spectrum of *Coakley et al.* (1983).**

(c) **Q = exact results, " = 3-point quadrature.**

(d) **Ratio of the quadrature result to the exact result. " = the single wavelength results, # = the results averaged over the solar spectrum.**


RETRIEVAL TECHNIQUE
3-POINT QUADRATURE
Δf_R
10.00
1.00
0.10
(a)
(c)
0
10
20
30
test distribution number
Δf_R / $\Delta f_{R,\,exact}$
1.06
1.04
1.02
1.00
0.98
0.96
0.94
1.40
1.30
1.20
1.10
1.00
0.90
0.80
0.70
(b)
(d)