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SIMULATING SULFUR FOR THE ACE-2 EXPERIMENT: PRELIMINARY RESULTS

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The Global Chemistry Model driven by Observation-derived meteorology (GChM-O, Benkovitz and Schwartz, 1997; Benkovitz et al., 1994) has been modified to include the production of H_2O_2 from HO_2, aqueous phase sulfur chemistry in non-precipitating clouds, use of daily averaged oxidant concentrations from the MOZART model (Brasseur, 1998), and dynamic estimation of the cloud water pH. Resolution has been enhanced to $1^\circ \times 1^\circ$ in the horizontal and 27 η coordinate levels in the vertical (surface to ~100 hPa). The model uses the time and location dependent meteorological data from ECMWF. The model has been applied to simulate mixing ratios of sulfate, SO_2, DMS, and MSA for the Northern Hemisphere to 81˚N during the ACE-2 experimental period. Eighty four percent of the sulfur emissions in the modeling domain are contributed by anthropogenic sources, 12% by biogenic sources, and 4% by volcanos. Primary sulfate emissions are only 3% of the sulfur emissions; other statistics for the sulfur emissions are also presented. Analysis of the results for one specific day, June 26, 1997 at 0 UT, show that biogenic emissions contributed ~6% of the total sulfate burden in the domain, anthropogenic emissions contributed ~66%, and volcano emissions contributed 28%. The contributions to the sulfate column burden of the anthropogenic emissions by main source regions (generically called North America, Europe, and Asia) and by formation process (primary, gas-phase, aqueous-phase) are also presented. Vertical profiles of sulfate concentrations at two ACE-2 land sites and the largest contributor at each level demonstrate the difference between a site within an area of heavily anthropogenic emissions and a site away from the direct influence of such emissions.

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Simulating Sulfur for the ACE- 2 Experiment: Preliminary Results

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COMPONENTS OF THE TRANSPORT AND TRANSFORMATION MODEL

METEOROLOGY
ECMWF
Winds, precip, etc.
horizontal: 1° x 1°
27 vertical levels

EMISSIONS
EDGAR/GEIA Anthro
SO_2, primary sulfate
BNL/Kettle/Andreae
Ocean DMS
Lamb Land
DMS, H_2S
BNL Volcanoes
SO_2

OXIDANT CONCENTRATIONS
Daily average
O_3, OH, H_2O_2, HO_2
MOZART (Brasseur et al.)

→

TRANSPORT
Bott/Easter

CHEMISTRY (Rate constants NASA, 1997)
GAS Phase
$SO_2 + OH \rightarrow H_2SO_4$
DMS + OH DMS chem *Yin/Seinfeld*
$HO_2 + HO_2 \rightarrow H_2O_2$ (limited by Mozart)
AQUEOUS Phase
$SO_2 + H_2O_2 \rightarrow H_2SO_4$
$SO_2 + O_3 \rightarrow H_2SO_4$

DRY DEPOSITION
~~Time, location dependent~~
~~*Wesely*~~ 0.8 cm/s SO_2
0.1 cm/s Sulfate

WET DEPOSITION
Berkowitz/Hales

CONVECTIVE MIXING
Berkowitz

→

SO_4^{2-}
MSA
SO_2
DMS

Simulation of the ACE-2 Experiment

★ Geographic domain:
- Model: Northern Hemisphere from 0° to 81°N.
- Experiment: 25°W to 8°W, 23°N to 44°N.

★ Time period:
- Model: June 1 to July 25, 1997.
- Experiment: June 16 to July 25, 1997.

Emissions

Anthropogenic

★ EDGAR Version 2 with the following changes:
- Population-based regridded using (Li , 1996) population file.
- Based on GEIA 1985 inventory (Benkovitz, 1996):
  - Seasonal resolution: summer emissions used.
  - SO_2/primary sulfate speciation.
  - Vertical resolution ($\leq$ 100 m, > 100 m).

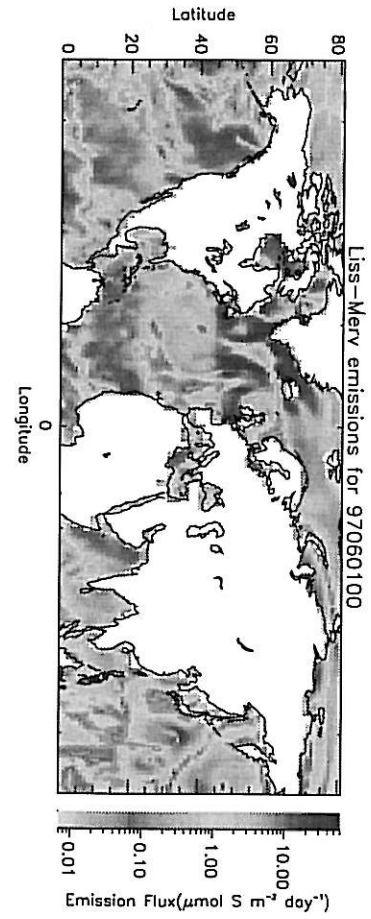
Emissions

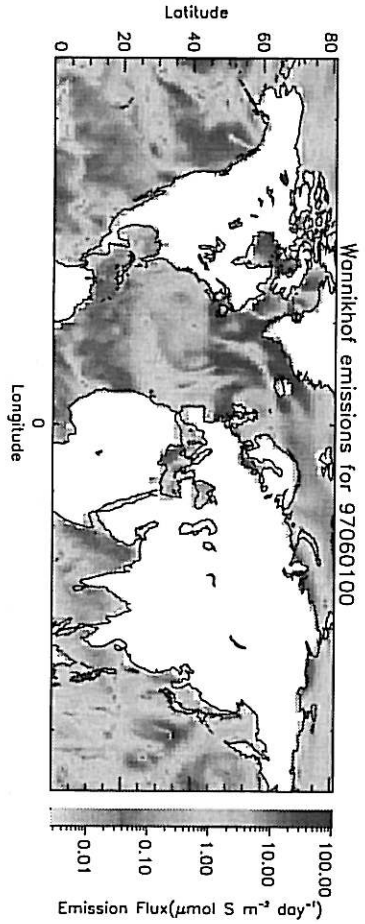
DMS

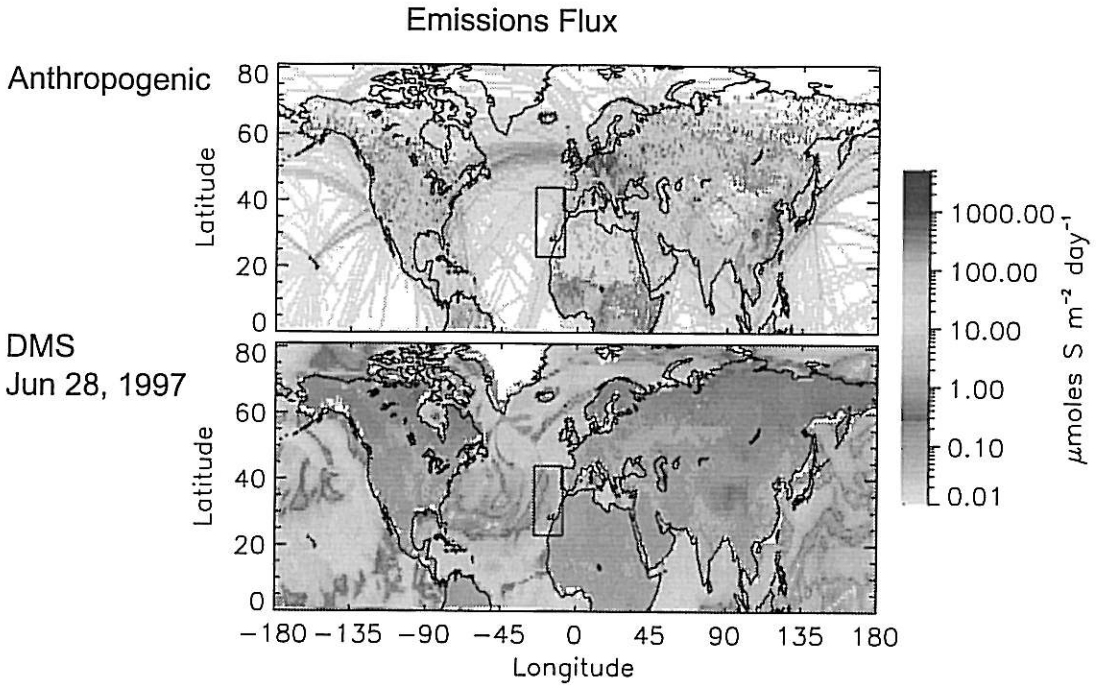
Land sources: Lamb, 1992.

Ocean

- **DMS ocean concentrations from Kettle et al. krigged to 1E resolution.**
  - Four regions: Atlantic, Pacific, and Indian Oceans.
  - Measurements for June, July used.
  - Coastal and estuarine measurements not used.
  - Kriging confined to each ocean basin; results merged, boundaries smoothed.
  - Iterative 16x16 window average for missing data.
- **ACE-2 measurements krigged and replace defaults.**
- **6-hr ECMWF winds at 10 m.**
- **Liss & Merlivat and Wanninkhof methodologies used.**
- **Liss & Merlivat results used.**







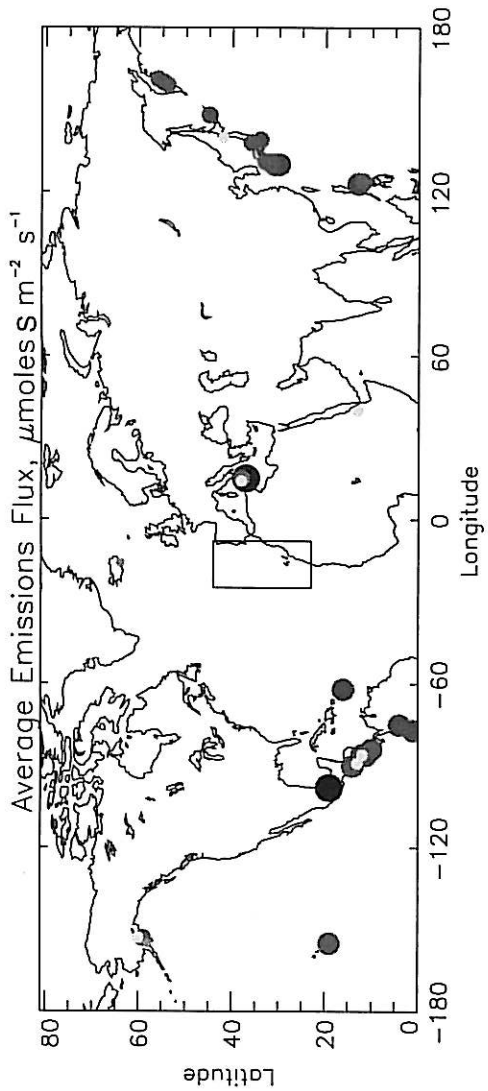
Emissions

Volcanos

★ Emissions for simulation period whenever possible.

★ Active volcanos from Smithsonian Volcano Activity Reports.

★ Emissions estimated using:
- Measurements and interpolation.
- Min/max emissions with random function.
- 25-yr average degassing (Andres, 1997).



Emissions, Percent by Source Type

Source Type	Model Domain 0° to 360° Equator to 81°N	ACE-2 Domain 25°W to 8°W 23°N to 44°N
Anthropogenic	84.5	79.8
Biogenic	11.7	20.2
Volcanos	3.8	0

Anthropogenic Sulfur Emissions

Percent SO_2 vs Primary Sulfate

SO_2	Sulfate
96.8	3.2

Percent by Release Height

Release Height	% SO_2	% Sulfate
Surface to 100 m	55.1	61.4
100 to 260 m	44.8	38.6
Above 260 m	< 1.0	0

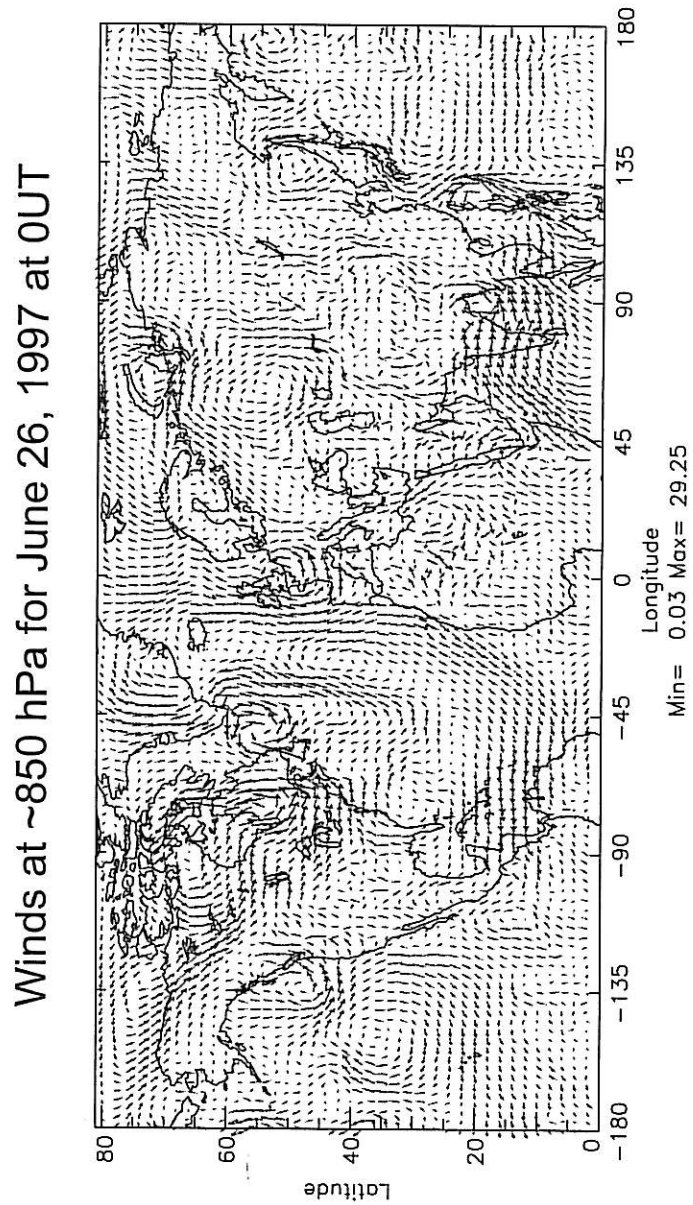
Emissions from Volcanos

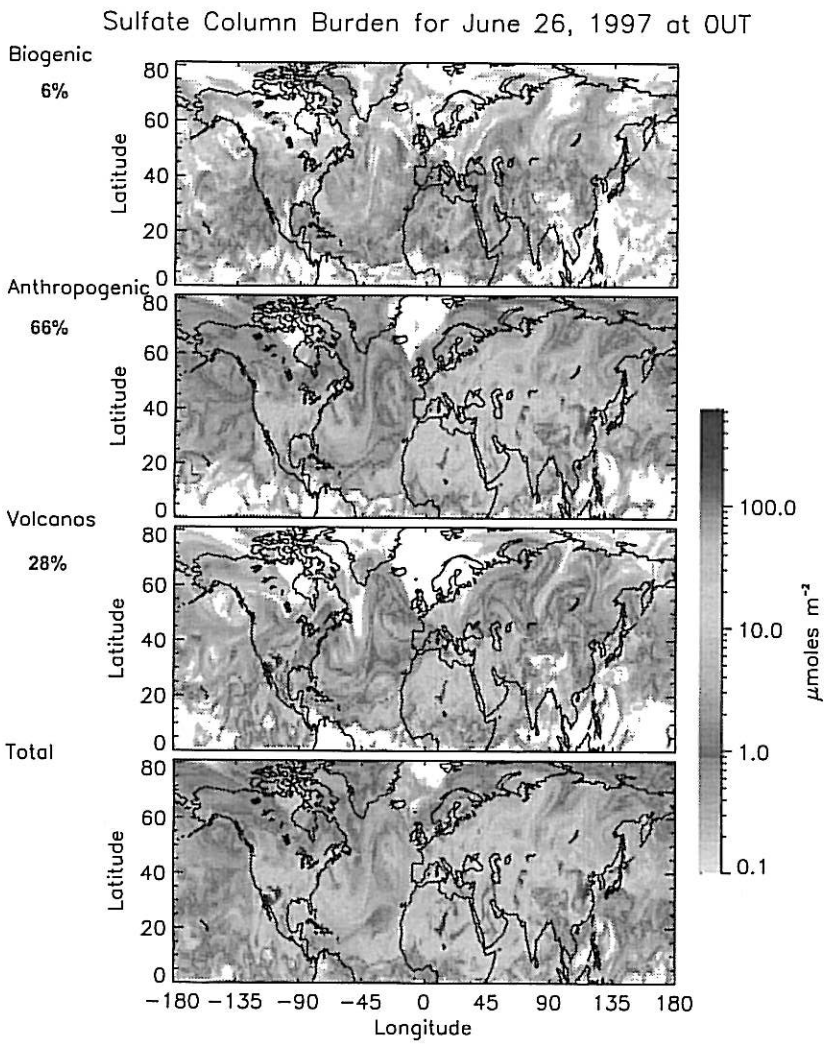
Percent by Release Height

Release Height	% Emissions
500 to 2,000 m	30.4
2,000 to 4,000 m	25.7
Above 4,000 m	44.0

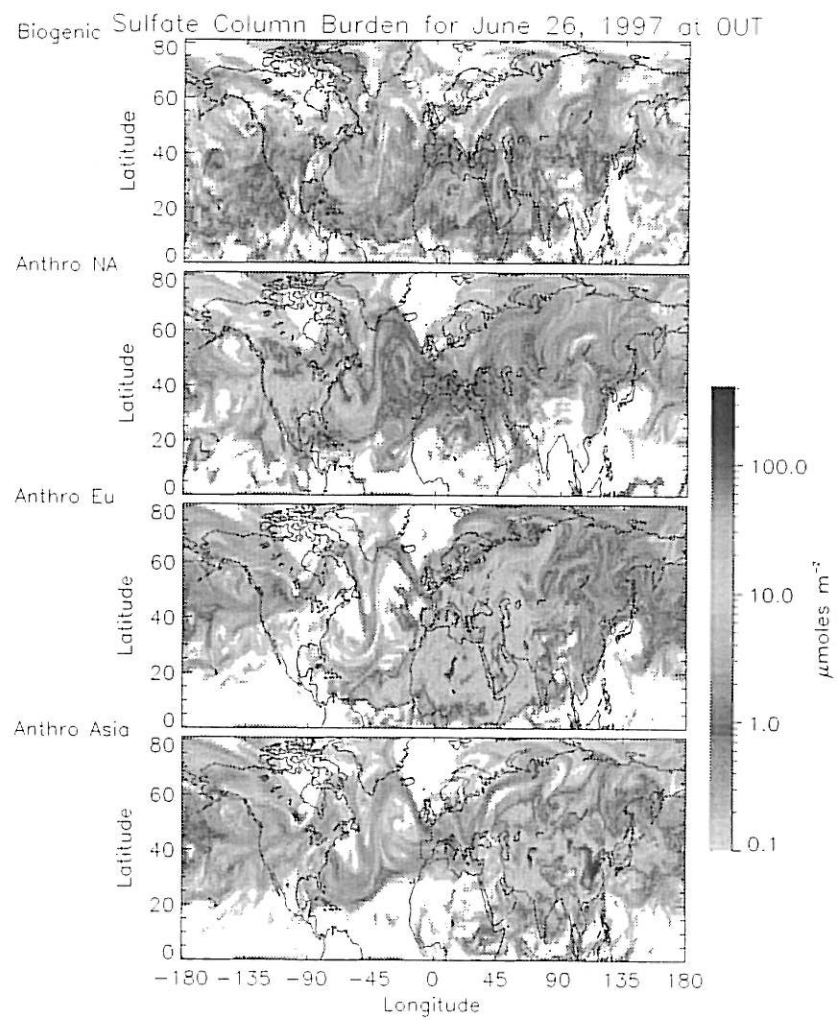
Percent By Method of Estimating Emissions

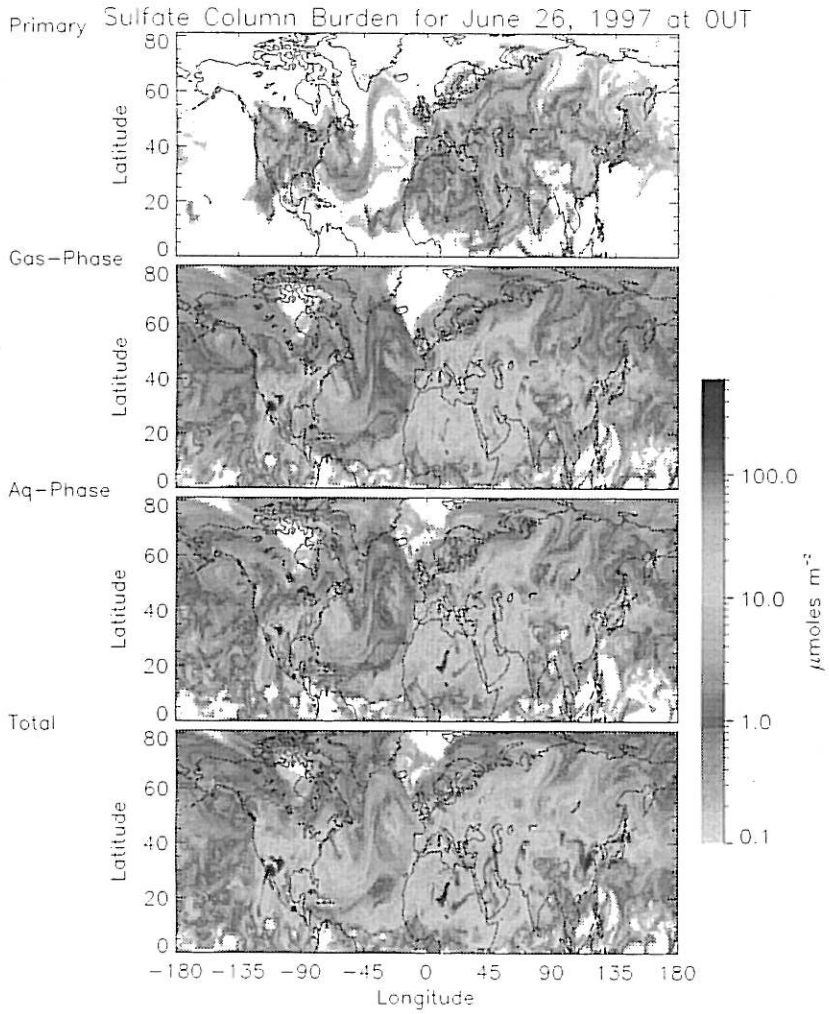
Methodology	Emissions (%)
Measurements	59.0
Temporal Average	37.7
Range of emissions	3.4



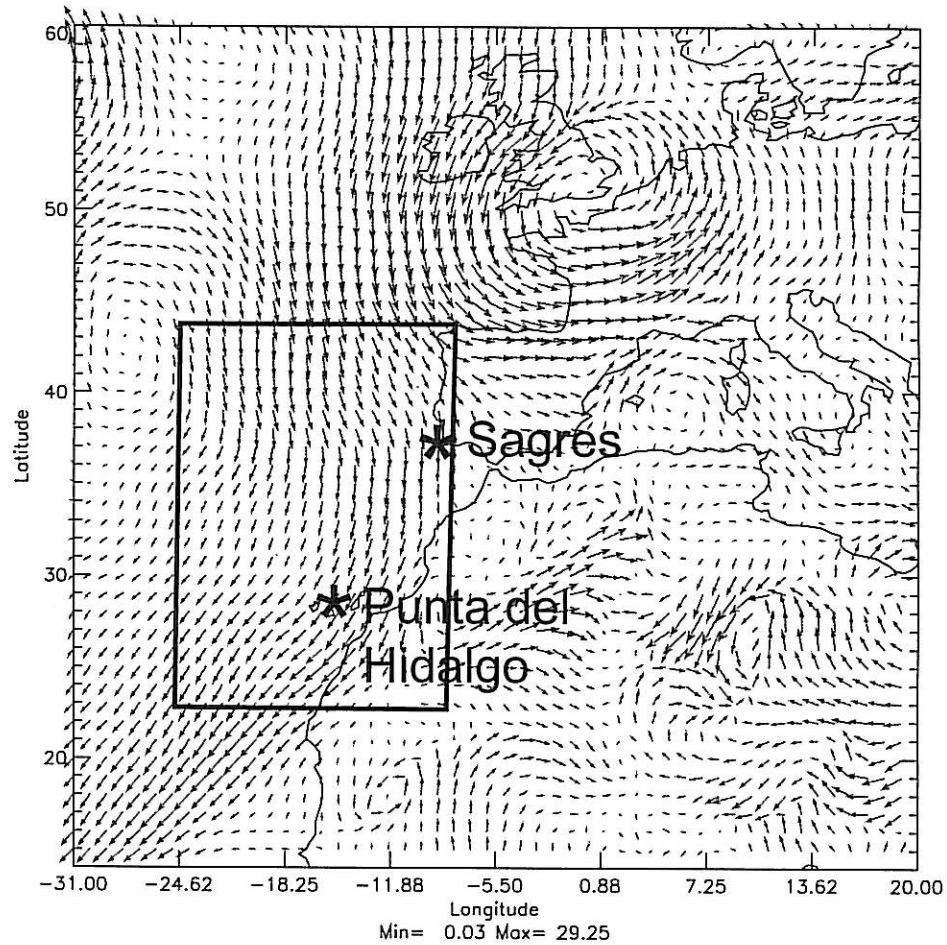


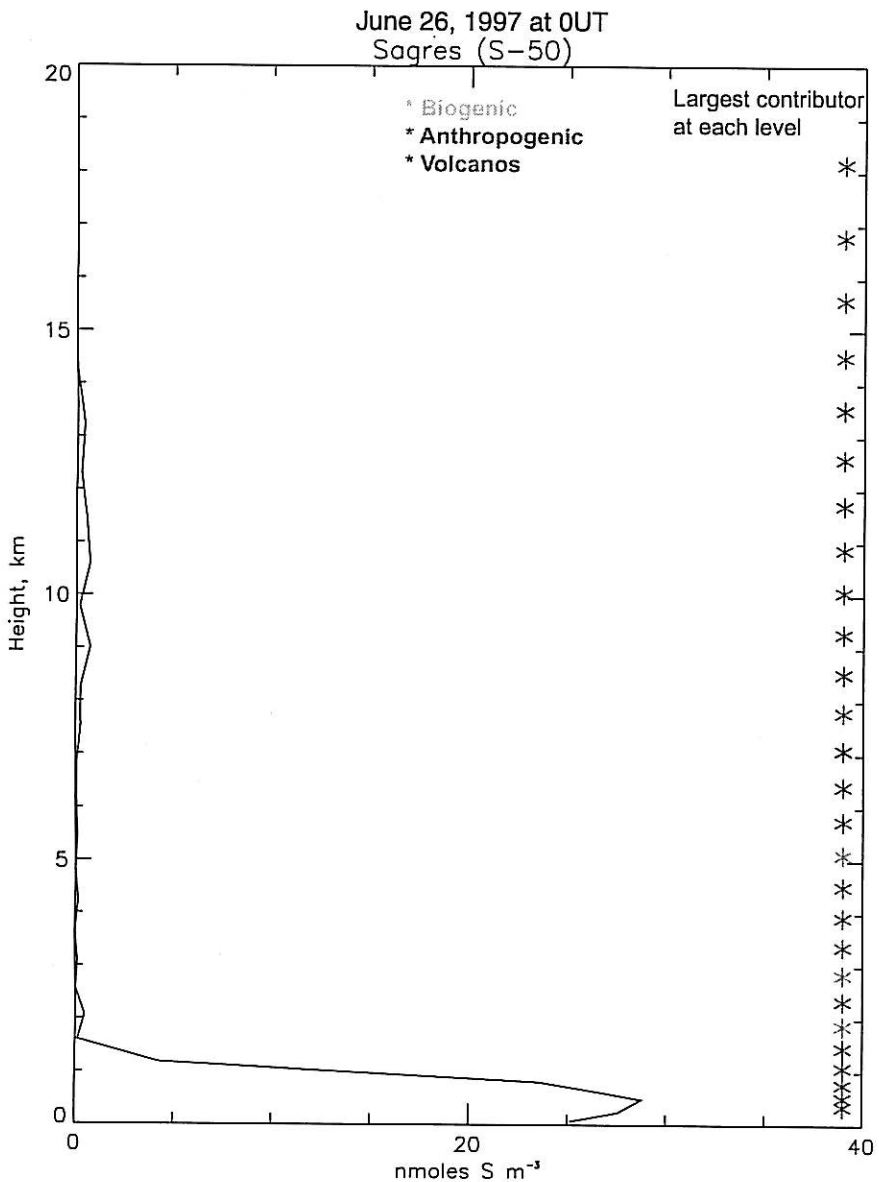
Contribution to Total Sulfate Burden

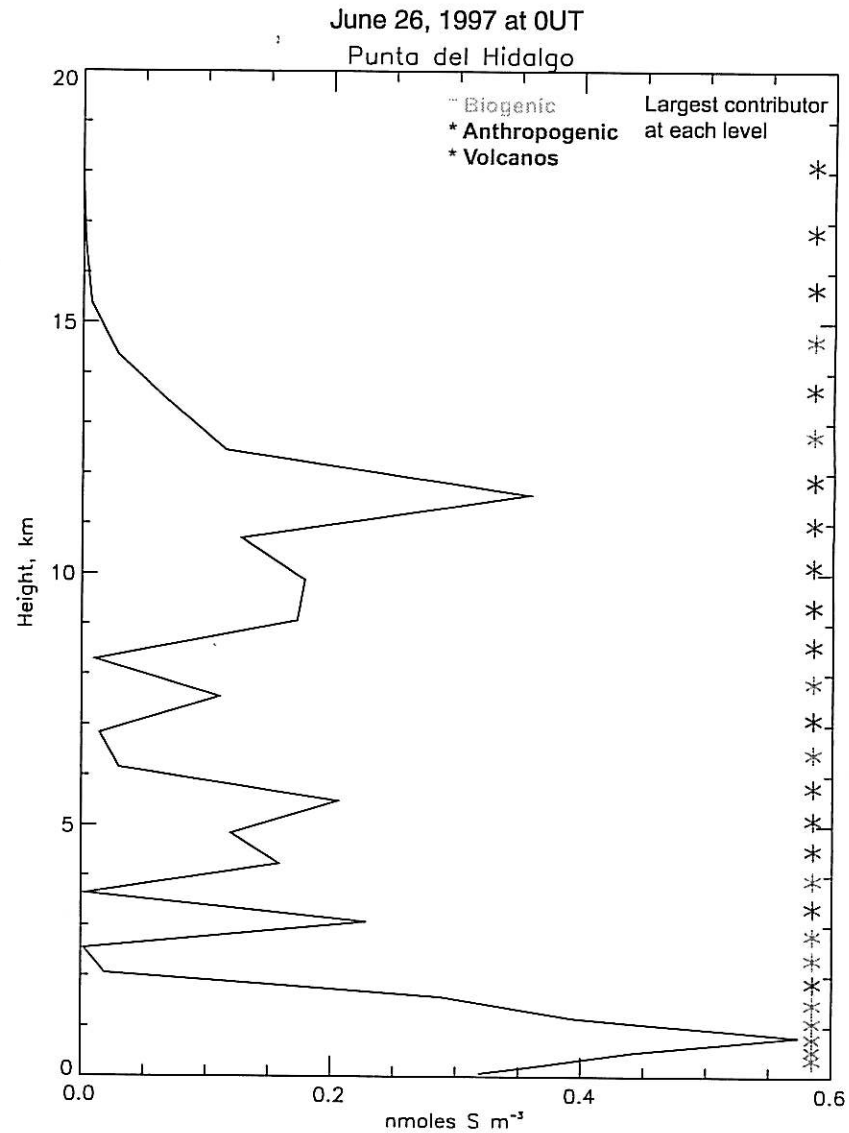




Winds at ~850 hPa for June 26, 1997 at 0UT




June 26, 1997 at 0UT
Sagres (S-50)
* Biogenic
* Anthropogenic
* Volcanos
Largest contributor at each level
Height, km
nmoles S m⁻³
0
5
10
15
20
0
20
40


June 26, 1997 at 0UT
Punta del Hidalgo
* Biogenic
* Anthropogenic
* Volcanos
Largest contributor at each level
Height, km
nmoles S m⁻³
0
5
10
15
20
0.0
0.2
0.4
0.6

Work in Progress

- ★ Parallelize computer code.
- ★ Merge into current model
  - Implementation of the Quadrature Method of Moments to represent aerosol microphysical properties.
- ★ Develop sea salt and dust emissions.