

SIMULATING SULFUR FOR THE ACE-2 EXPERIMENT: PRELIMINARY RESULTS

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September 1999

Presented at the
Sixth Scientific Conference of the
International Global Atmospheric Chemistry Project (IGAC),
Bologna, Italy
September 13-17, 1999

ABSTRACT

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 $\sim 6\%$ of the total sulfate burden in the domain, anthropogenic emissions contributed $\sim 66\%$, 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.

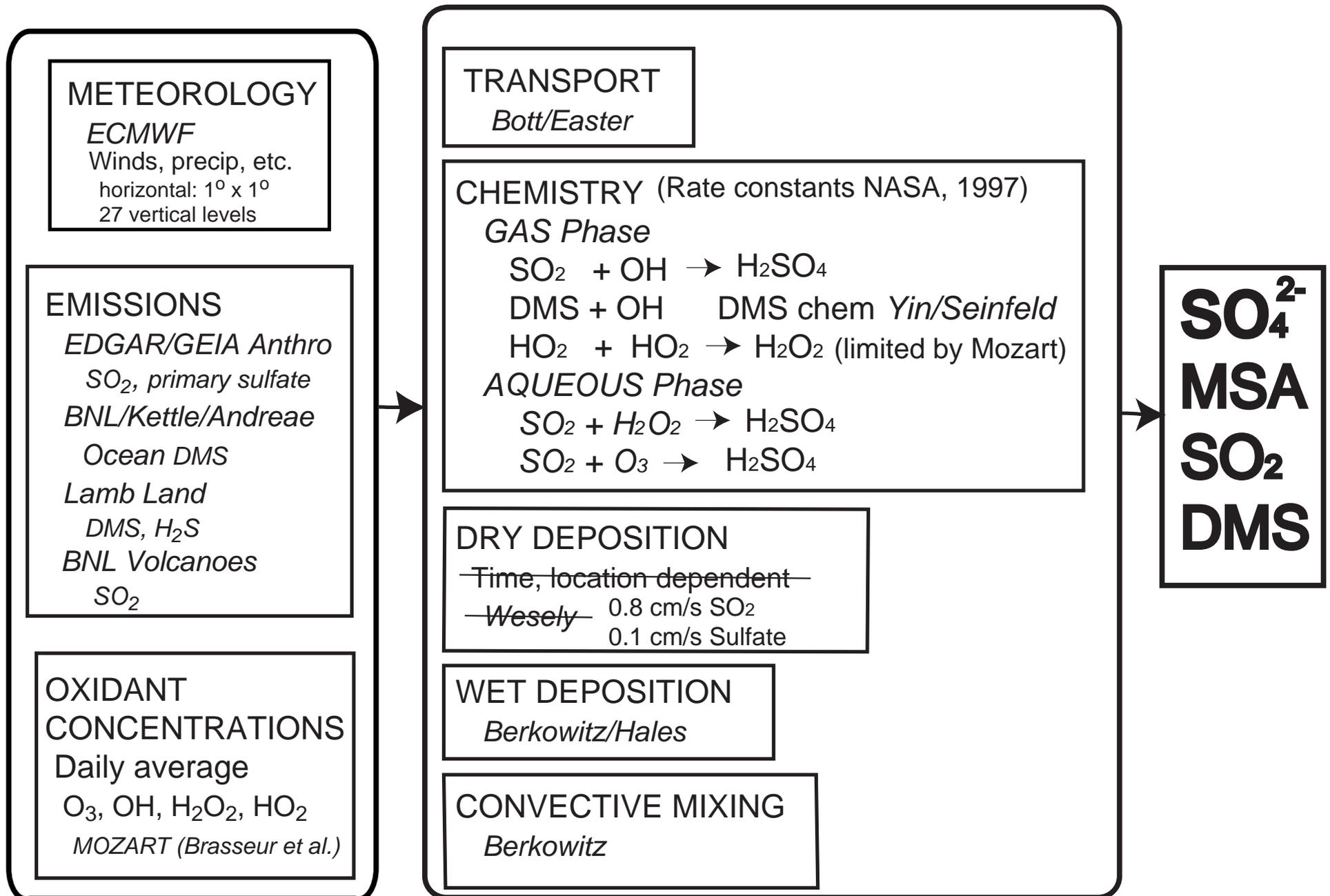
Simulating Sulfur for the ACE- 2 Experiment: Preliminary Results

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COMPONENTS OF THE TRANSPORT AND TRANSFORMATION MODEL



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 regrided using (Li , 1996) population file.**
 - **Based on GEIA 1985 inventory (Benkovitz, 1996):**
 - **Seasonal resolution: summer emissions used.**
 - **SO₂/primary sulfate speciation.**
 - **Vertical resolution (≤ 100 m, > 100 m).**

Emissions

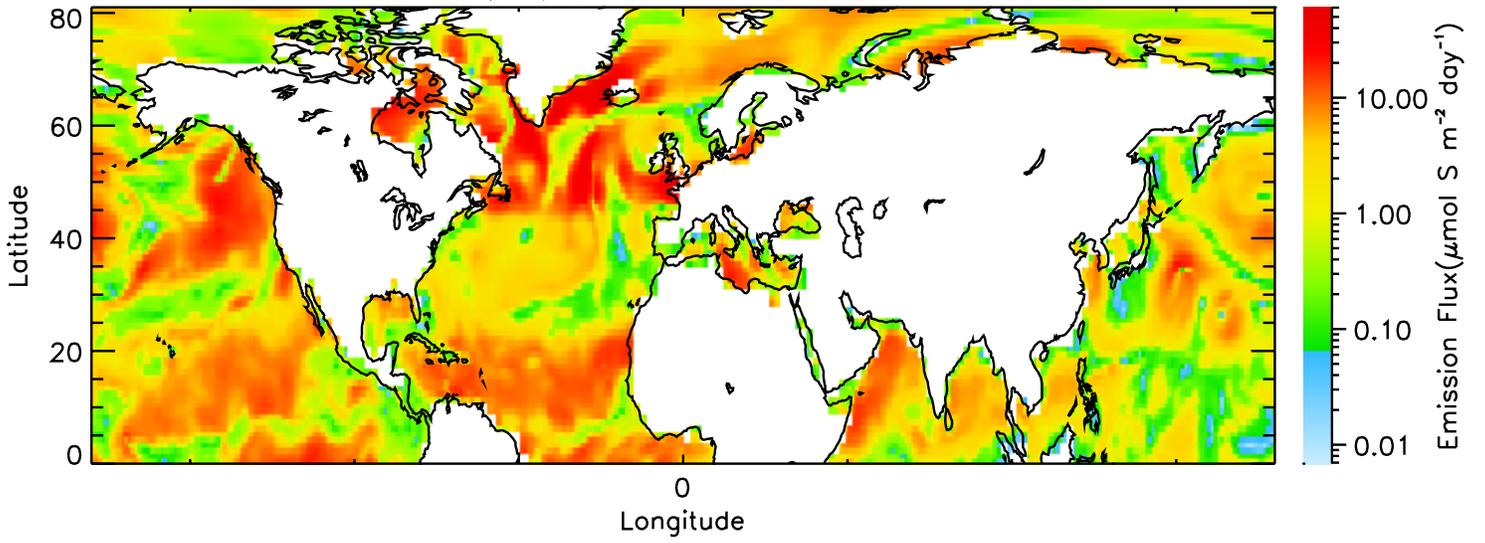
DMS

Ú Land sources: Lamb, 1992.

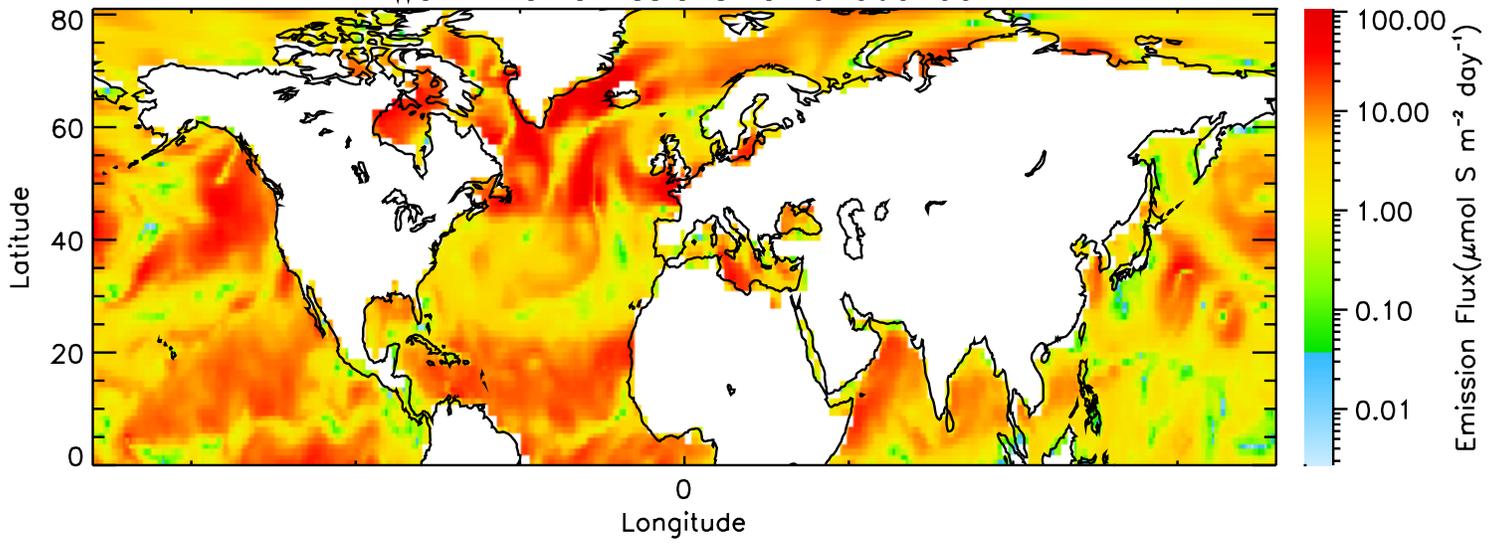
Ú Ocean

- 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.

Liss-Mery emissions for 97060100

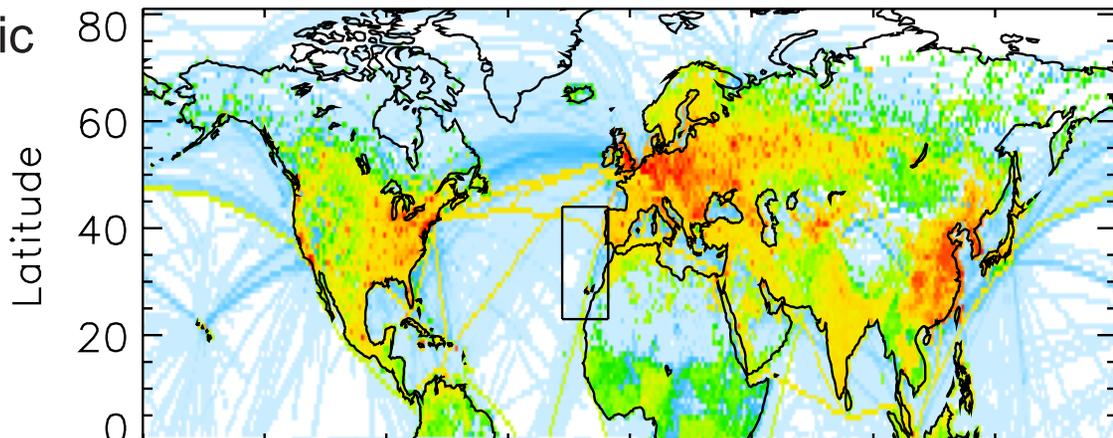


Wannikhof emissions for 97060100

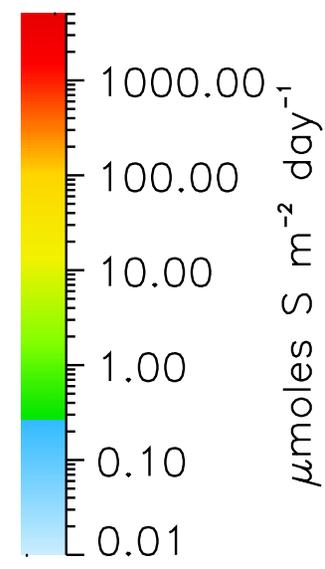
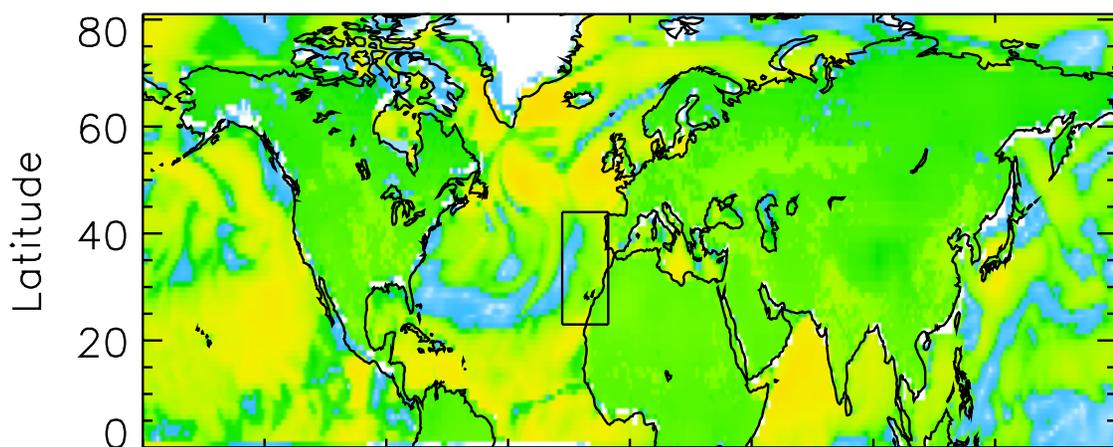


Emissions Flux

Anthropogenic



DMS
Jun 28, 1997



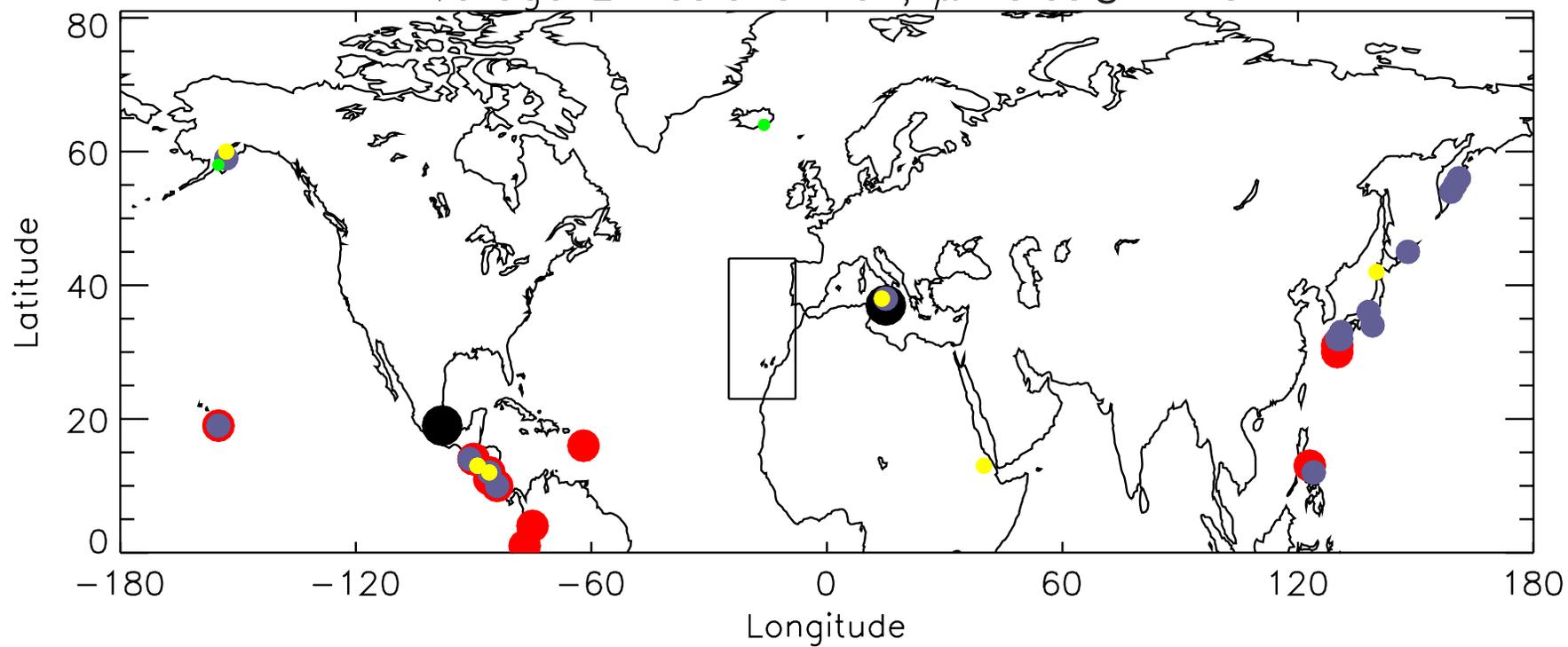
Longitude

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).

Average Emissions Flux, $\mu\text{moles S m}^{-2} \text{s}^{-1}$



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 SO₂ vs Primary Sulfate

SO₂	Sulfate
96.8	3.2

Percent by Release Height

Release Height	% SO₂	% 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

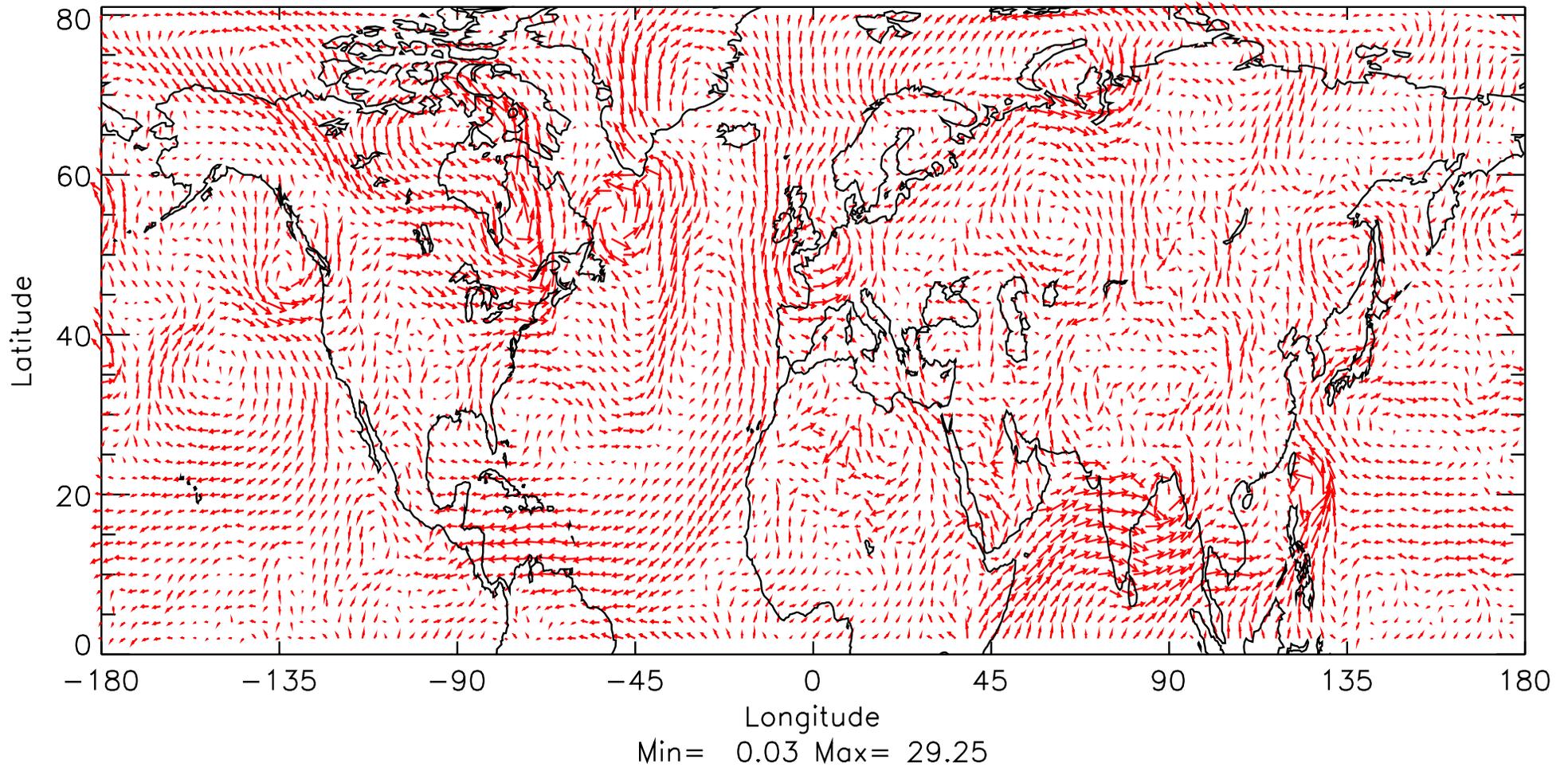
Percent by Release Height

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

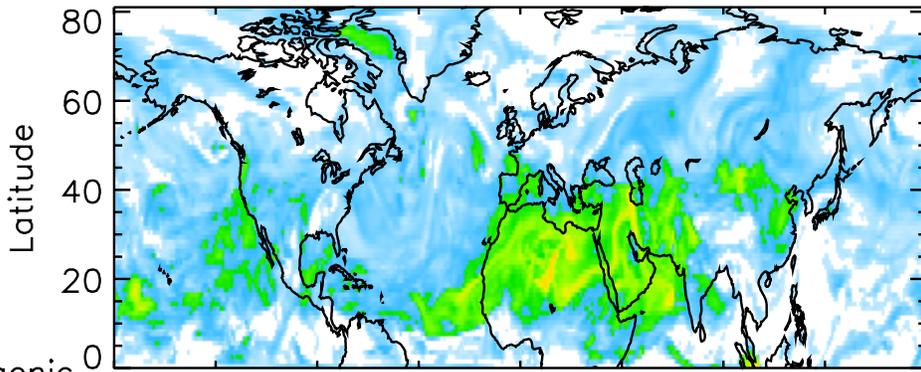
Winds at ~850 hPa for June 26, 1997 at 0UT



Sulfate Column Burden for June 26, 1997 at OUT

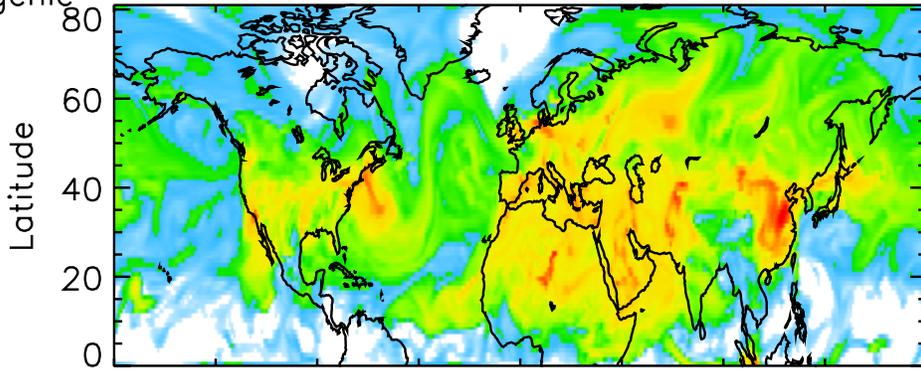
Biogenic

6%



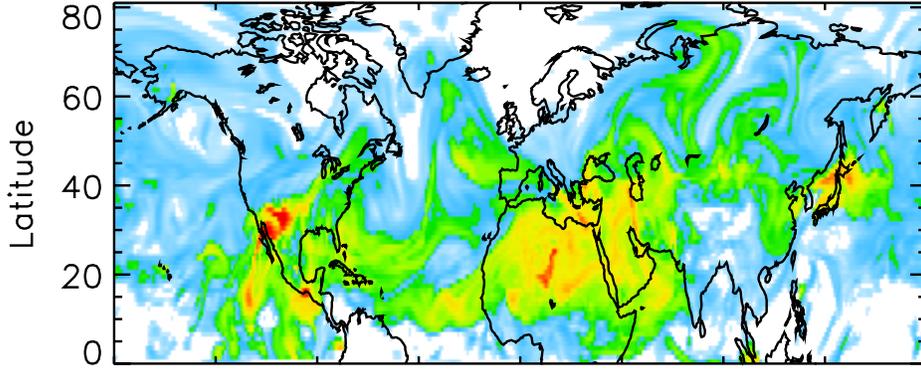
Anthropogenic

66%

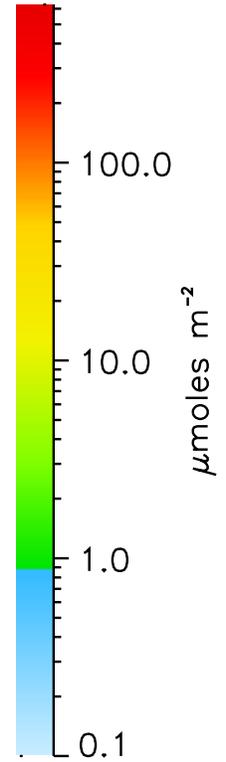
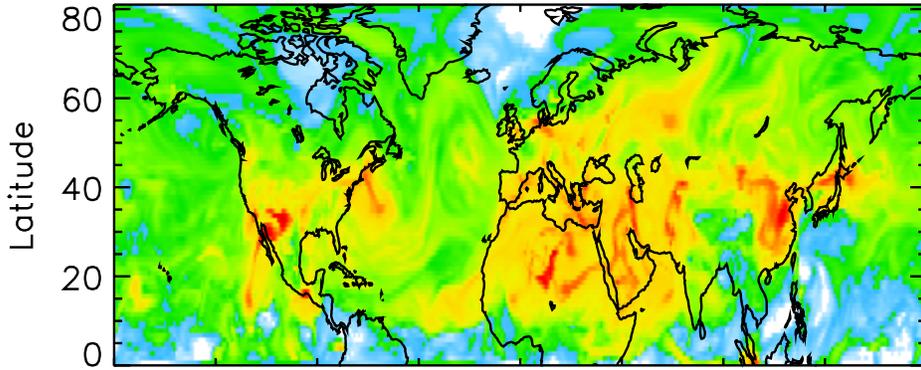


Volcanos

28%

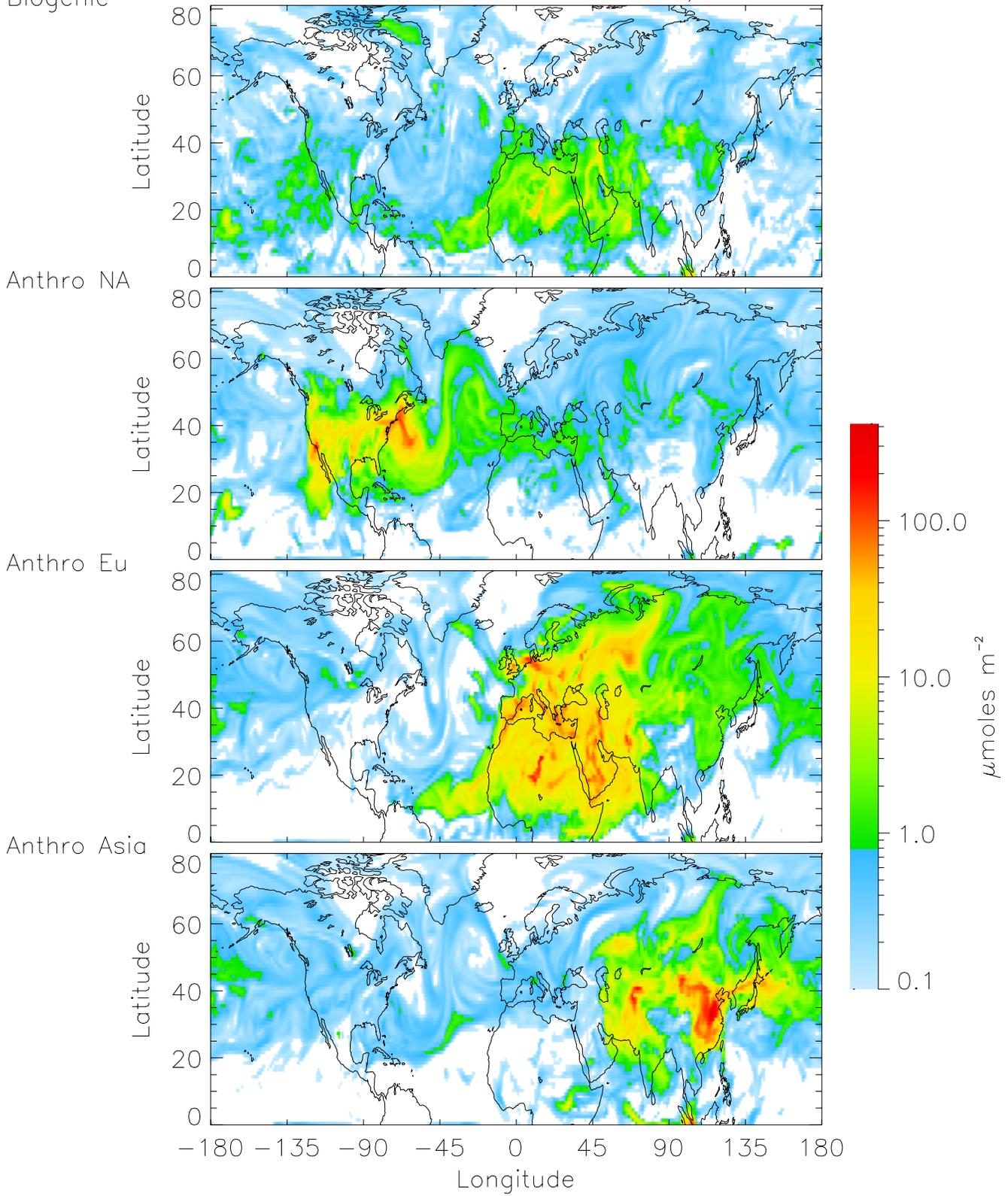


Total

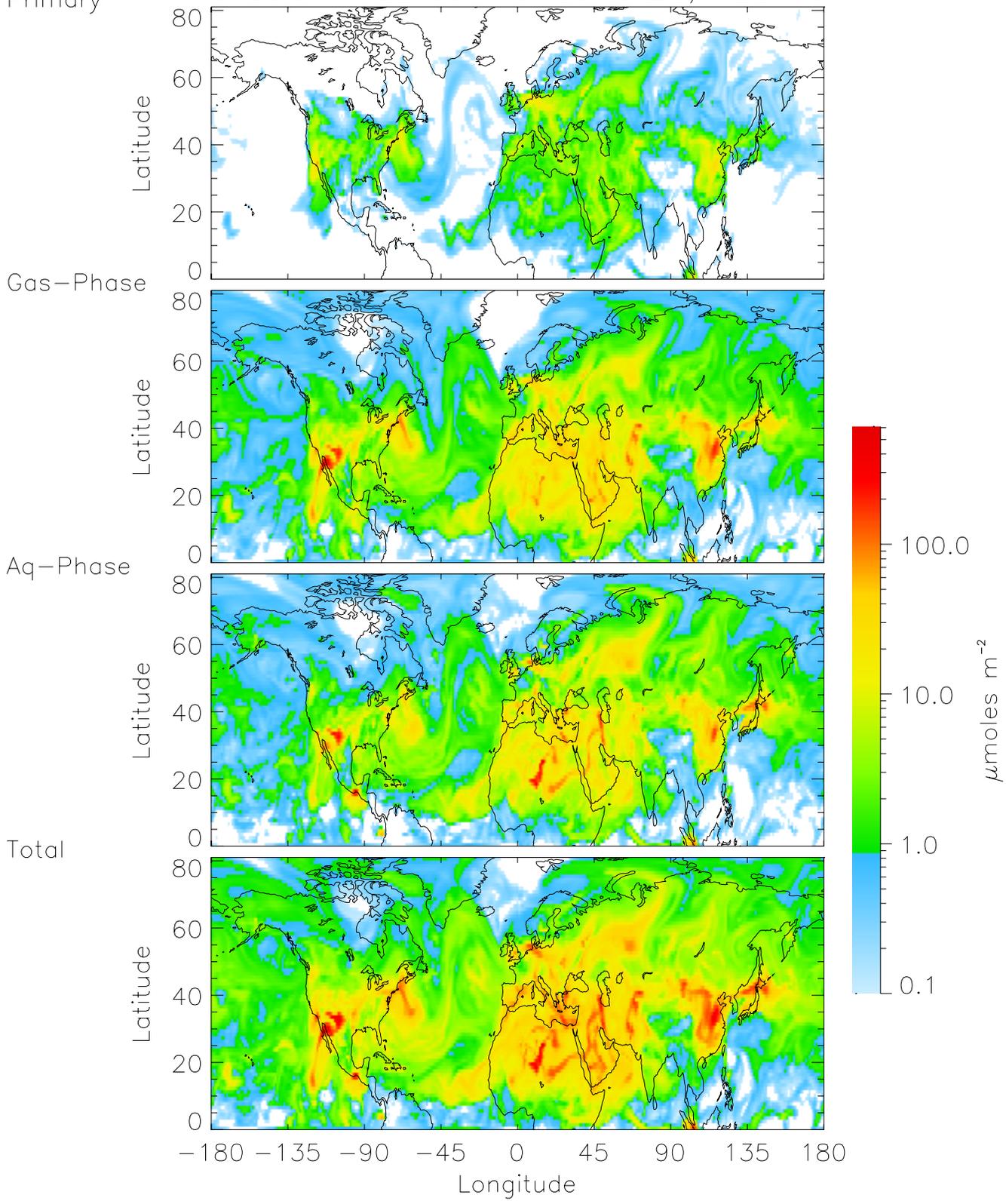


**Contribution
to Total Sulfate
Burden**

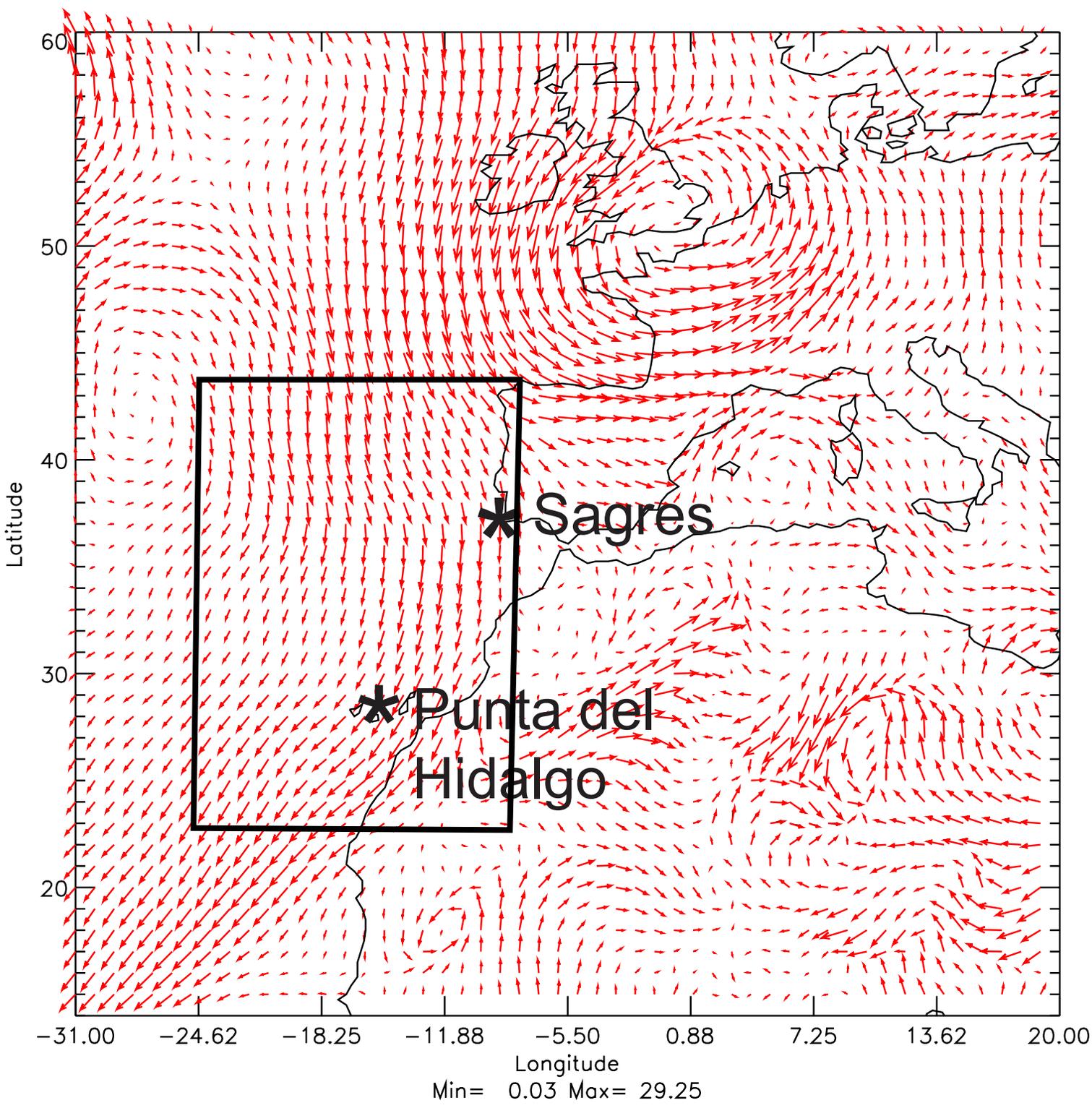
Biogenic Sulfate Column Burden for June 26, 1997 at OUT



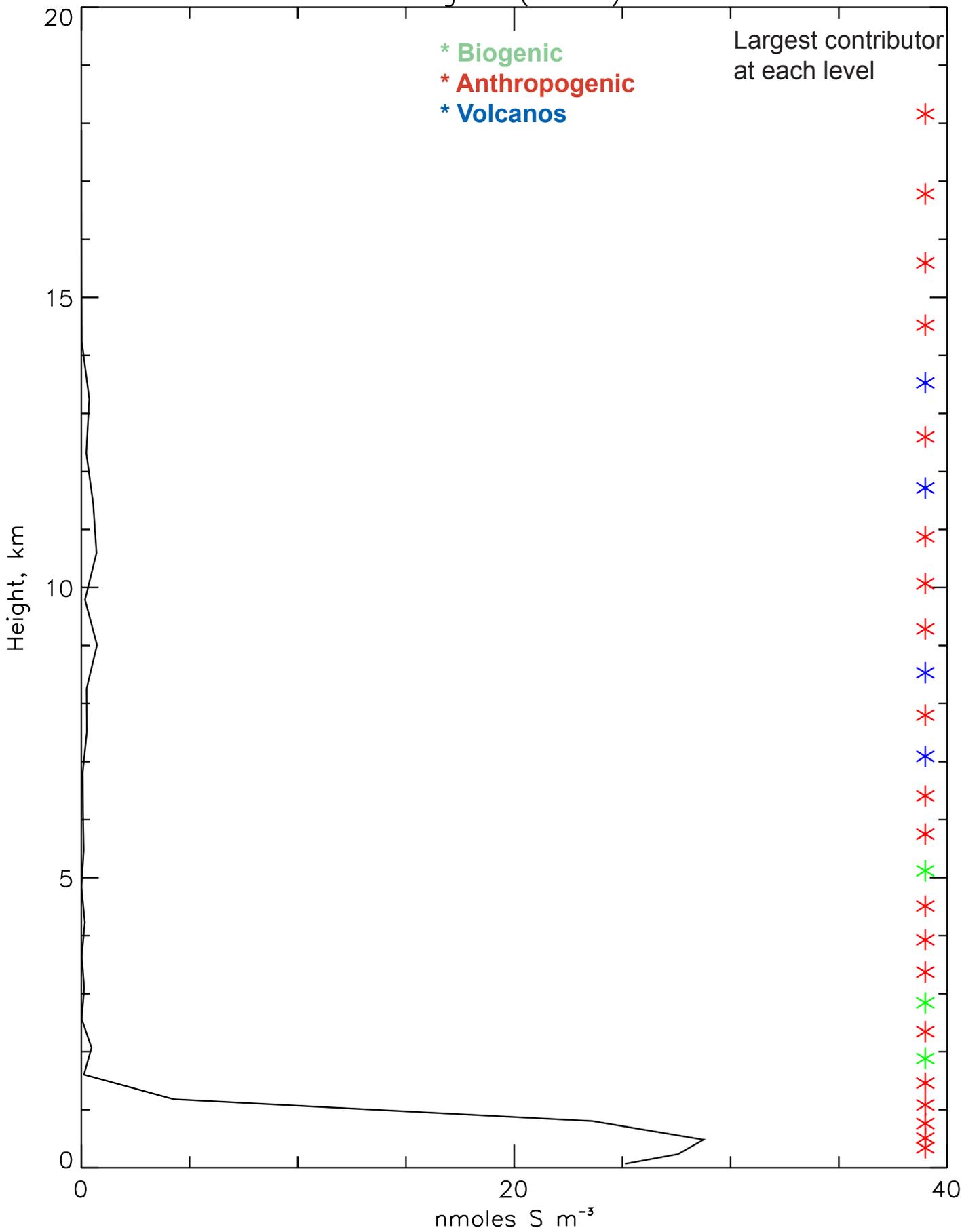
Primary Sulfate Column Burden for June 26, 1997 at OUT



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

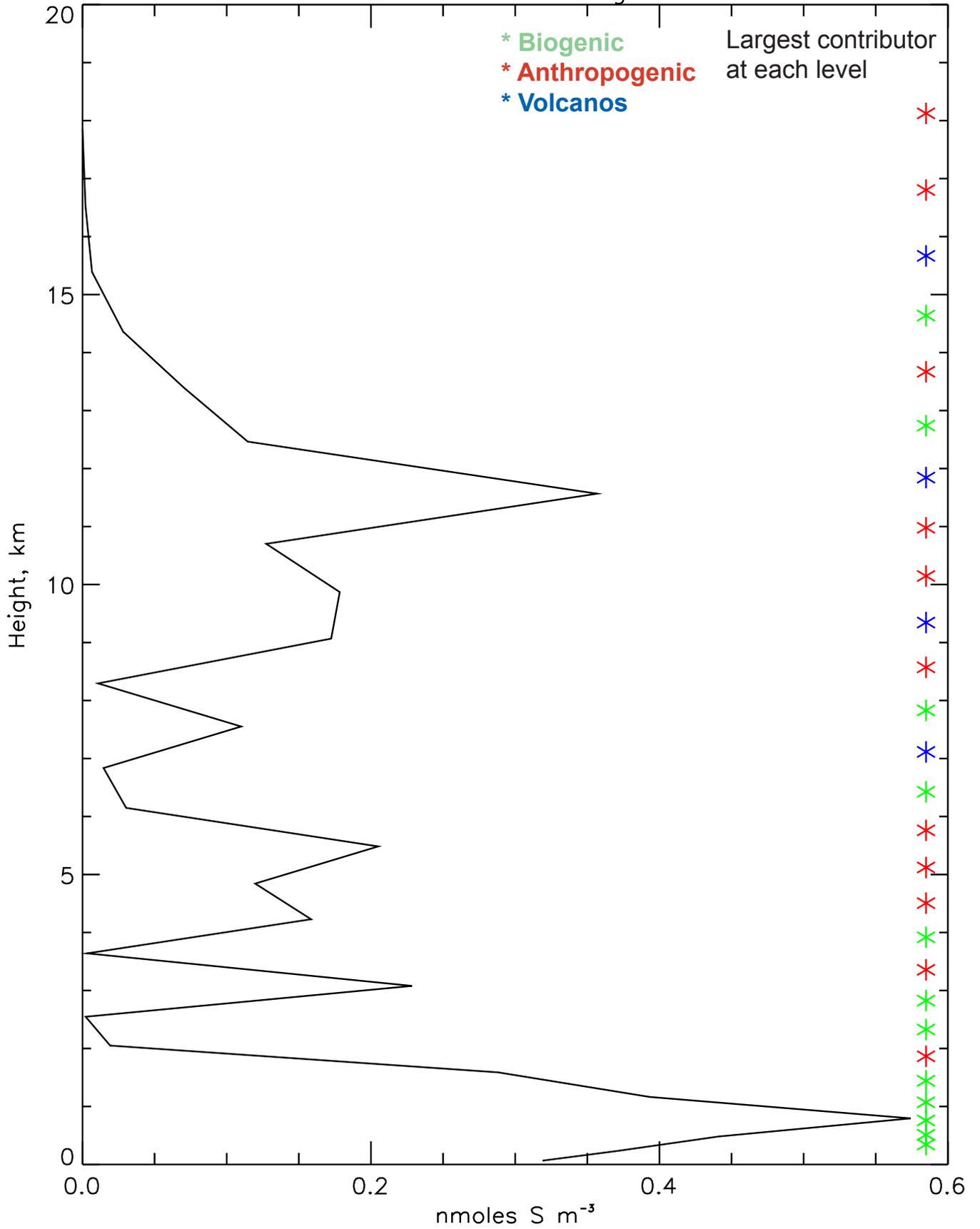


June 26, 1997 at OUT
Sagres (S-50)



June 26, 1997 at OUT

Punta del Hidalgo



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.**

Future Work

★ Add additional aerosol species

- Size-resolved anthropogenic emissions of primary particles.
- Particle emissions from volcanos.
- Organics?