SIMULATING SULFUR FOR THE ACE-2 EXPERIMENT: PRELIMINARY RESULTS

C. M. Benkovitz, S. E. Schwartz, M. A. Mubaraki, and M. A. Miller
Environmental Chemistry Division
Brookhaven National Laboratory
Upton, NY 11973

Timothy S. Bates
Pacific Marine Environmental Laboratory
NOAA
Seattle, WA

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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$_2$O$_2$ 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° $\times$ 1° in the horizontal and 27 $\eta$ 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.
Simulating Sulfur for the ACE-2 Experiment: Preliminary Results

Carmen M. Benkovitz, Stephen E. Schwartz, M. Altaf Mubarak, Mark A. Miller
Brookhaven National Laboratory
Upton, NY, USA

Timothy S. Bates
Pacific Marine Environmental Laboratory, NOAA
Seattle, WA, USA

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COMPONENTS OF THE TRANSPORT AND TRANSFORMATION MODEL

METEOROLOGY
ECMWF
Winds, precip, etc.
horizontal: $1^\circ \times 1^\circ$
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
$SO_2 + OH \rightarrow H_2SO_4$
$DMS + OH \rightarrow$ 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
$0.8 \text{ cm/s } SO_2$
$0.1 \text{ cm/s } Sulfate$

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

DMS


/ 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

<table>
<thead>
<tr>
<th>Source Type</th>
<th>Model Domain</th>
<th>ACE-2 Domain</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0° to 360°</td>
<td>25°W to 8°W</td>
</tr>
<tr>
<td></td>
<td>Equator to 81°N</td>
<td>23°N to 44°N</td>
</tr>
<tr>
<td>Anthropogenic</td>
<td>84.5</td>
<td>79.8</td>
</tr>
<tr>
<td>Biogenic</td>
<td>11.7</td>
<td>20.2</td>
</tr>
<tr>
<td>Volcanos</td>
<td>3.8</td>
<td>0</td>
</tr>
</tbody>
</table>

Anthropogenic Sulfur Emissions

Percent SO₂ vs Primary Sulfate

<table>
<thead>
<tr>
<th>SO₂</th>
<th>Sulfate</th>
</tr>
</thead>
<tbody>
<tr>
<td>96.8</td>
<td>3.2</td>
</tr>
</tbody>
</table>

Percent by Release Height

<table>
<thead>
<tr>
<th>Release Height</th>
<th>% SO₂</th>
<th>% Sulfate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface to 100 m</td>
<td>55.1</td>
<td>61.4</td>
</tr>
<tr>
<td>100 to 260 m</td>
<td>44.8</td>
<td>38.6</td>
</tr>
<tr>
<td>Above 260 m</td>
<td>&lt; 1.0</td>
<td>0</td>
</tr>
</tbody>
</table>
Emissions from Volcanos

Percent by Release Height

<table>
<thead>
<tr>
<th>Release Height</th>
<th>% Emissions</th>
</tr>
</thead>
<tbody>
<tr>
<td>500 to 2,000 m</td>
<td>30.4</td>
</tr>
<tr>
<td>2,000 to 4,000 m</td>
<td>25.7</td>
</tr>
<tr>
<td>Above 4,000 m</td>
<td>44.0</td>
</tr>
</tbody>
</table>

Percent By Method of Estimating Emissions

<table>
<thead>
<tr>
<th>Methodology</th>
<th>Emissions (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurements</td>
<td>59.0</td>
</tr>
<tr>
<td>Temporal Average</td>
<td>37.7</td>
</tr>
<tr>
<td>Range of emissions</td>
<td>3.4</td>
</tr>
</tbody>
</table>
Winds at ~850 hPa for June 26, 1997 at 0UT
Sulfate Column Burden for June 26, 1997 at 0UT

Contribution to Total Sulfate Burden

Biogenic: 6%
Anthropogenic: 66%
Volcanos: 28%
Total:
Winds at ~850 hPa for June 26, 1997 at 0UT
Largest contributor at each level:

* Biogenic
* Anthropogenic
* Volcanos

June 26, 1997 at 0UT
June 26, 1997 at 0UT

Punta del Hidalgo

Largest contributor at each level
* Biogenic
* Anthropogenic
* Volcanos

Height, km

nmoles S m⁻³
★ 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?