NATURAL AND TRANSBOUNDARY INFLUENCES ON PARTICULATE MATTER IN THE UNITED STATES: IMPLICATIONS FOR THE EPA REGIONAL HAZE RULE

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ACCESS VII, September 5, 2003
EPA REGIONAL HAZE RULE

- Federal class I areas (including national parks, other wilderness areas) to return to “natural visibility” conditions by 2064
- State Implementation Plans to be submitted by 2007 for linear improvement in visibility over the 2004-2018 period

These defaults are not traceable to actual data in the literature. What do they mean in terms of actual natural vs. anthropogenic influences?
OBJECTIVES and APPROACH

- Determine present-day natural PM concentrations in the U.S., compare to EPA “default” values.
- Determine the contributions from foreign anthropogenic emissions (Canada, Mexico, Asia) to visibility degradation in the U.S.
- Determine the pervasiveness of natural fires and dust events, particularly from outside the U.S, in decreasing natural visibility in the U.S.

**Improved emission estimates**

- Start from best *a priori* estimates of natural and anthropogenic PM sources
- Conduct global PM simulation with GEOS-CHEM chemical tracer model on 2°x2.5° horizontal resolution
- Evaluate with aerosol data from IMPROVE, CASTNET, other networks
- Conduct sensitivity simulations
- Assessment of EPA “default estimated natural PM concentrations”
- Assessment of transboundary pollution influences
CARBONACEOUS AEROSOL SIMULATION

Best a priori sources (1998)

ORGANIC CARBON (OC)

GLOBAL

130 Tg yr⁻¹

UNITED STATES

2.7 Tg yr⁻¹

ELEMENTAL CARBON (EC)

22 Tg yr⁻¹

0.66 Tg yr⁻¹

- Fossil fuel
- Biofuel
- Biomass burning
- Vegetation
ANNUAL MEAN OC AND EC (1998): GEOS-CHEM vs. IMPROVE (45 sites)

- High OC in southeast U.S.: vegetation
- High OC in Mexico, Canada: fires
LEAST-SQUARES FIT OF MODEL TO OBSERVATIONS GENERATES OPTIMIZED A POSTERIORI SOURCES

Fossil fuel \(\uparrow 15\%\)  Biofuel \(\uparrow 65\%\)  Biomass burning \(\downarrow 17\%\)  Biogenic \(\uparrow 11\%\)
CARBONACEOUS AEROSOL IN THE U.S.: contributions from natural sources and transboundary pollution

Annual regional means from GEOS-CHEM standard and sensitivity simulations

<table>
<thead>
<tr>
<th></th>
<th>OC (µg m⁻³ as OMC)</th>
<th>EC (µg m⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>West</td>
<td>East</td>
</tr>
<tr>
<td>Baseline (1998)</td>
<td>2.0</td>
<td>3.2</td>
</tr>
<tr>
<td>Zero anthropogenic emissions in U.S.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>– GEOS-CHEM (w/ climatological fires)</td>
<td>1.3</td>
<td>1.2</td>
</tr>
<tr>
<td>– EPA default values</td>
<td>0.5</td>
<td>1.4</td>
</tr>
<tr>
<td>Contributions from transboundary anthropogenic sources</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Canada and Mexico</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Asia</td>
<td>0.013</td>
<td>0.007</td>
</tr>
</tbody>
</table>

- We find that EPA default natural concentrations are too low by factors of 2-3 except for OC in eastern U.S. – quantifying fire influences is critical
- Transboundary pollution influences are relatively small except for EC from Canada/Mexico
**H₂SO₄-HNO₃-NH₃-H₂O AEROSOL SIMULATION**

**GEOS-CHEM emissions (2001)**

### GLOBAL
- **Sulfur, Tg S yr⁻¹**
  - Industrial: 78%
  - Volcanoes: 4%
  - Biomass burning: 13%
  - Oceans: 5%
- **Ammonia, Tg N yr⁻¹**
  - Livestock: 55%
  - Fertilizer: 23%
  - Humans: 6%
  - Industry: 5%
  - Biofuels: 2%
  - Soils/vegetation: 4%
  - Oceans: 6%
  - Biomass burning: 1%
- **NOₓ, Tg N yr⁻¹**
  - Fossil fuel: 43%
  - Fertilizer: 12%
  - Biofuel: 9%
  - Lightning: 3%
  - Soils: 11%
  - Biomass burning: 4%

### UNITED STATES
- **Sulfur, Tg S yr⁻¹**
  - Industrial: 8.3%
- **Ammonia, Tg N yr⁻¹**
  - Livestock: 2.8%
- **NOₓ, Tg N yr⁻¹**
  - Fossil fuel: 7.4%
ANNUAL MEAN SULFATE (2001): GEOS-CHEM vs. IMPROVE (141 sites)

Highest concentrations in industrial Midwest (coal-fired power plants)

High correlation for different seasons ($R^2 = 0.79 - 0.94$)
No significant model bias (Slope = 0.71 - 1.02) except in summer (=0.71) (excessive scavenging of $SO_2$ in convective updrafts?)
ANNUAL MEAN AMMONIUM AND NITRATE (2001): GEOS-CHEM vs. CASTNET (79 sites)

(no ammonium data at IMPROVE sites)

Highest concentrations in upper Midwest

High correlation for different seasons ($R^2 = 0.82-0.90$)
High bias for $\text{NH}_4^+$ in fall: error in seasonal variation of livestock emissions

$R^2 = 0.36-0.66$ except for summer
High bias for $\text{NO}_3^-$; esp. in summer/fall, results from bias on $[\text{SO}_4^{2-}]-2[\text{NH}_4^+]$
SEASONAL VARIATION OF AEROSOL ACIDITY

- Aerosol is mostly neutralized (highest acidity in eastern U.S. in summer); aerosol phase is therefore a major issue for visibility assessment.

- Except in the upper Midwest, \( \text{NH}_4\text{NO}_3 \) formation is limited by \( \text{NH}_3 \) rather than by \( \text{HNO}_3 \) availability \( \Rightarrow \) better knowledge of \( \text{NH}_3 \) emissions is critical.
INTERCONTINENTAL TRANSPORT OF ASIAN AND NORTH AMERICAN ANTHROPOGENIC SULFATE

As determined from GEOS-CHEM 2001 sensitivity simulations with these sources shut off
SULFATE-NITRATE-AMMONIUM AEROSOL IN THE U.S.: contributions from natural sources and transboundary pollution

Annual regional means from GEOS-CHEM standard and sensitivity simulations

<table>
<thead>
<tr>
<th></th>
<th>Ammonium sulfate ($\mu$g m^{-3})</th>
<th>Ammonium nitrate ($\mu$g m^{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>West</td>
<td>East</td>
</tr>
<tr>
<td>Baseline (2001)</td>
<td>1.52</td>
<td>4.11</td>
</tr>
<tr>
<td>Zero anthropogenic emissions in U.S.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>– GEOS-CHEM</td>
<td>0.43</td>
<td>0.38</td>
</tr>
<tr>
<td>– EPA default values</td>
<td>0.11</td>
<td>0.23</td>
</tr>
<tr>
<td>Contributions from transboundary anthropogenic sources</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Canada and Mexico</td>
<td>0.15</td>
<td>0.14</td>
</tr>
<tr>
<td>Asia</td>
<td>0.13</td>
<td>0.12</td>
</tr>
</tbody>
</table>

- Achievability of EPA default estimates is compromised by transboundary pollution influences
- Transboundary pollution influence from Asia is comparable in magnitude to that from Canada + Mexico
**CONSEQUENCES FOR 2004-2018 IMPLEMENTATION OF EPA REGIONAL HAZE RULE**

Visibility (deciviews: \( dv = 10 \ln(\frac{b_{ext}}{10}) \))

<table>
<thead>
<tr>
<th></th>
<th>WEST</th>
<th>EAST</th>
</tr>
</thead>
<tbody>
<tr>
<td>Baseline ( dv ) (current conditions)</td>
<td>15.6</td>
<td>22</td>
</tr>
<tr>
<td>Estimated natural ( dv )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- EPA default</td>
<td>4.7</td>
<td>7.4</td>
</tr>
<tr>
<td>- This work</td>
<td>8.8</td>
<td>8.8</td>
</tr>
<tr>
<td>Required improvement in ( dv ), 2004-2018</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- using EPA default</td>
<td>2.5</td>
<td>3.4</td>
</tr>
<tr>
<td>- using this work</td>
<td>1.6</td>
<td>3.1</td>
</tr>
<tr>
<td>Required % improvement in ( b_{ext} ), 2004-2018:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- using EPA default</td>
<td>21%</td>
<td>29%</td>
</tr>
<tr>
<td>- using this work</td>
<td>13%</td>
<td>27%</td>
</tr>
</tbody>
</table>

Illustrative calculation assuming \( f(RH) = 2 \) (west) or 3 (east), adopting EPA default values for dust, and using mean eastern/western U.S. PM concentrations.