BACKGROUND AEROSOL IN THE UNITED STATES:
NATURAL SOURCES AND TRANSBOUNDARY POLLUTION

Daniel J. Jacob and Rokjin J. Park

with support from EPRI, EPA/OAQPS

• How good are the “default estimated natural PM concentrations” proposed by EPA as 2064 endpoint for application of the Regional Haze Rule?

• To what extent does transboundary pollution compromise achievability of natural PM concentrations?
2004-2018 EMISSION REDUCTIONS FOR THE RHR ARE STRONGLY SENSITIVE TO CHOICE OF 2064 ENDPOINT

Conceptual calculation for mean western U.S. conditions, assuming linear relationship between emissions and $b_{\text{ext}}$

Desired trend in visibility

$$dv = 10 \ln \left( \frac{b_{\text{ext,natural}} + b_{\text{ext,anthrop}}}{10} \right)$$

Required % decrease of U.S. anthropogenic emissions

Phase 1

- 30%
- 48%

Year
STRATEGY FOR QUANTIFYING BACKGROUND PM CONCENTRATIONS IN UNITED STATES

Start from best *a priori* estimates of natural and anthropogenic PM sources

Simulate PM concentrations with GEOS-CHEM global model

Evaluate with observations from IMPROVE, CASTNET, other networks

Conduct sensitivity simulations

Quantify natural aerosol concentrations

Quantify transboundary pollution

Improved emission estimates
GEOS-CHEM GLOBAL CHEMICAL TRANSPORT MODEL
(http://www-as.harvard.edu/chemistry/trop/geos)

• Driven by NASA/GEOS-3 assimilated meteorological data with $2^\circ \times 2.5^\circ$ horizontal resolution, 48 levels in vertical

• Carbonaceous aerosol simulation (OC, EC) for 1998

• Coupled oxidant –sulfate-nitrate-ammonium simulation for 2001 (also 1998)
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)**
- **GLOBAL**: 22 Tg yr⁻¹
- **UNITED STATES**: 0.66 Tg yr⁻¹

Legend:
- Fossil fuel
- Biofuel
- Biomass burning
- Vegetation
BIOMASS BURNING EMISSIONS IN GEOS-CHEM:
Climatology from Duncan, Logan, et al. [JGR 2002] scaled to 1998 using TOMS and ATSR satellite data

Interannual variability derived from TOMS Aerosol Index [Duncan et al., 2002]

1998 fires from ATSR satellite data:
• Apr-May fires in Mexico
• Jul-Sep fires in U.S/Canada
SEASONAL VARIATION OF OC AND EC

Constrain OC/EC sources using simulation of monthly mean IMPROVE observations and model tracers of individual source types.

- IMPROVE
- model

model tracers (additive)

- biomass burning
- vegetation
- fossil fuel
- biofuel
LEAST-SQUARES FIT OF MODEL TO MONTHLY MEAN OC/EC OBSERVATIONS AT 45 IMPROVE SITES YIELDS OPTIMIZED A POSTERIORI SOURCES

Fossil fuel ↑15%  Biofuel ↑65%  Biomass burning ↓17%  Biogenic ↑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>Background (no U.S. anthrop. emissions, climatological fires)</td>
<td>1.3</td>
<td>1.2</td>
</tr>
<tr>
<td>Natural (no anthrop. emissions globally)</td>
<td>1.2</td>
<td>1.1</td>
</tr>
<tr>
<td>Transboundary pollution enhancements</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>
<tr>
<td>EPA default estimated natural concentrations</td>
<td>0.5</td>
<td>1.4</td>
</tr>
</tbody>
</table>

- EPA default natural concentrations for OC are OK in eastern U.S., a factor of 2-3 too low in western U.S. – quantifying fire influences is critical
- Transboundary pollution influences are relatively small
**H$_2$SO$_4$-HNO$_3$-NH$_3$-H$_2$O AEROSOL SIMULATION**

GEOS-CHEM emissions (2001)

**GLOBAL**
- Sulfur, Tg S yr$^{-1}$
  - 78
- Ammonia, Tg N yr$^{-1}$
  - 55
- NO$_x$, Tg N yr$^{-1}$
  - 43

**UNITED STATES**
- Sulfur, Tg S yr$^{-1}$
  - 8.3
- Ammonia, Tg N yr$^{-1}$
  - 2.8
- NO$_x$, Tg N yr$^{-1}$
  - 7.4
ANNUAL MEAN SULFATE (2001): GEOS-CHEM vs. IMPROVE (141 sites)

Highest concentrations in industrial Midwest (coal-fired power plants)
SULFATE AT IMPROVE, CASTNET, NADP (deposition) SITES: model vs. observed for different seasons

High correlation, no significant model bias except 25-30% negative bias in summer (we attribute it to excessive scavenging of SO₂ in convective updrafts)
ANNUAL MEAN AMMONIUM (2001):
GEOS-CHEM vs. CASTNET (79 sites)
(no ammonium data at IMPROVE sites)

Highest concentrations in upper Midwest (livestock)
ANNUAL MEAN NITRATE (2001): GEOS-CHEM vs. CASTNET (79 sites)

Highest concentrations in upper Midwest (NH$_4$NO$_3$ formation limited by ammonia availability)
AMMONIUM AND NITRATE AT CASTNET AND IMPROVE SITES: model vs. observed for different seasons

- High bias for NH$_4^+$ in fall: excessive seasonal livestock emissions
- High bias for NO$_3^-$, esp. in summer/fall, results from bias on [SO$_4^{2-}$]-2[NH$_4^+$]
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 (µg m⁻³)</th>
<th>Ammonium nitrate (µg 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>Background (no U.S. anthrop. emissions)</td>
<td>0.43</td>
<td>0.38</td>
</tr>
<tr>
<td>Natural (no anthrop. emissions globally)</td>
<td>0.11</td>
<td>0.11</td>
</tr>
<tr>
<td>Transboundary pollution enhancements</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>
<tr>
<td>EPA default estimated natural concentrations</td>
<td>0.11</td>
<td>0.23</td>
</tr>
</tbody>
</table>

- Large transboundary pollution influence prevents achievability of natural concentrations;
- Transboundary pollution influence from Asia is comparable in magnitude to that from Canada + Mexico
MODEL EVALUATION WITH ASIAN OUTFLOW OBSERVATIONS OF SULFATE FROM TRACE-P AIRCRAFT CAMPAIGN (MAR-APR 2001)

Observations from Jordan et al. [JGR 2003]
INTERCONTINENTAL TRANSPORT OF ASIAN AND NORTH AMERICAN ANTHROPOGENIC SULFATE

As determined from GEOS-CHEM 2001 sensitivity simulations with these sources shut off.
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
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 ⇒ better knowledge of \( \text{NH}_3 \) emissions is critical.

CASTNET sites

CASTNET

GEOS-CHEM

Figure 0-1