Adjoint inversion of Global NOx emissions with SCIAMACHY NO₂

Changsub Shim, Qinbin Li, Daven Henze, Aaron van Donkellaar, Randall Martin, Kevin Bowman, Monika Kopacz, and Annmarie Eldering

4th GEOS-Chem User’s Meeting
Apr. 08, 2009
## Global NOx emissions in GEOS-Chem

<table>
<thead>
<tr>
<th>NOx emissions</th>
<th>2005 (a priori)</th>
<th>2001 (a)</th>
<th>2005 (b)</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biofuel</td>
<td>2.02 (1995)</td>
<td>2.2</td>
<td>2.03 (2000)</td>
<td>4.5 ~ 5</td>
</tr>
<tr>
<td>Soil/fertilizer</td>
<td>5.06 (c)</td>
<td>5.77</td>
<td>5.5</td>
<td>12 ~13</td>
</tr>
<tr>
<td>Biomass Burning</td>
<td>6.7 (Climatological)</td>
<td>6.5 (d)</td>
<td>5.41 (GFEDV2)</td>
<td>10 ~16</td>
</tr>
<tr>
<td>Lightning/aircraft</td>
<td>4.2 (f)</td>
<td>4.7</td>
<td>4.5</td>
<td>~10</td>
</tr>
<tr>
<td>Total (Tg/yr)</td>
<td>42 (v6)</td>
<td>43</td>
<td>45 (v7)</td>
<td>100</td>
</tr>
</tbody>
</table>

(a): adapted from Park et al., (2004)
(b): GEOS-Chem v7-1-3.
(d): Climatological monthly biomass burning data (Duncan et al., 2003).
(e): Monthly GFEDv2 biomass burning data.
(f): Based on Wang et al., (1998)
**Adjoint Inversion**

- **Objective**
  - Inversion of NOx emissions with consideration of physiochemical feedbacks with direct computing of parameter’s sensitivity
  - Comparison with “top-down” emissions estimates (or mass balance approach) derived from satellite observations (e.g., Martin et al., 2003; 2006)

- **Advantage**
  - Can consider the chemical and physical feedbacks during optimization
    - quantifying the parameter’s sensitivity w.r.t. model predictions
  - Optimization control

\[
J(x) = (F(x) - y)^T S_\Sigma^{-1} (F(x) - y) + \gamma (x - x_a)^T S_a^{-1} (x - x_a)
\]

- **Disadvantage**
  - Still computationally expensive
    - Ex) 64 Intel Itanium2 processors (SGI architecture with LINUX)
      - 1.5 GHz clock speed with 1MB Cache + 1GB RAM
      - With parallel computing (8 CPUs)
    - Each iteration for one month time window (2°x2.5°, globally) takes 44 hours.
Data (Nov. 2005)

- SCIAMACHY NO\textsubscript{2} from Dalhousie Univ (reprocessed data), filtered cloud fraction > 40%.

- Adjoint of GEOS-Chem v6-2-5 & GEOS-4 & full chemistry (by D. Henze) with 2°x2.5° horizontal resolution

- Time window: one month (Nov. 2005)\textbf{←} a week x 4

- Emissions (NO\textsubscript{x})
  - GEIA anthropogenic NO\textsubscript{x} emission (scaled to 1998)
  - Climatological Biomass Burning(Duncan et al., 2003)
  - Biofuel emissions (Yevich et al., 2003)
  - Soil NO\textsubscript{x} (Yienger and Levy(1995) & Wang(1998))
  - Lightning NO\textsubscript{x} (Cloud Top Height; Price and Rind(1998) & Pickering (1998)): only consider the total emissions for opt.
**Observation error** = e1 + e2 + e3

* **e1**: retrieval error from SCIAMACHY
* **e2**: representation error : ~0.7 of e1 (~4.0x10^{13} molec/cm^2)
* **e3**: model transport error (from Jones et al., 2003)
  → ~0.8 of e1 (~4.5x10^{13} molec/cm^2)

Total obs. error is about factor of ~2.5 of instrumental (retrieval) error

→ Same quantity of errors were applied to mass-balance approach
Cost function from obs. Vs from a priori = ~10: 1

Now cost function reached ~ 0.40 of initial value after 7th iteration

<table>
<thead>
<tr>
<th>Iteration</th>
<th>Norm of grad.</th>
<th>Cost func. ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.405D+03</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>1.857D+03</td>
<td>0.90</td>
</tr>
<tr>
<td>3</td>
<td>7.530D+02</td>
<td>0.73</td>
</tr>
<tr>
<td>4</td>
<td>3.180D+02</td>
<td>0.62</td>
</tr>
<tr>
<td>5</td>
<td>1.485D+02</td>
<td>0.52</td>
</tr>
<tr>
<td>6</td>
<td>1.112D+03</td>
<td>0.46</td>
</tr>
<tr>
<td>7</td>
<td>1.948D+02</td>
<td>0.403</td>
</tr>
<tr>
<td>8</td>
<td>8.180D+01</td>
<td>0.397</td>
</tr>
</tbody>
</table>

Cost Function

\[ J(x) = (F(x) - y)^{\top} S_{\Sigma}^{-1} (F(x) - y) + \gamma (x - x_a)^{\top} S_{a}^{-1} (x - x_a) \]

Gradient

\[ \nabla_x J(x) = 2 \nabla_x F^{\top} S_{\Sigma}^{-1} (F(x) - y) + 2 \gamma S_{a}^{-1} (x - x_a) \]
Inversion Results (A priori vs A posteriori)
Unit: $10^{15}$ molecules/cm$^2$  Nov. 2005
Inversion Results: NOx emissions ratio (a posteriori / a priori)
- Large reductions in N. Ame, Europe, and India
- Moderate reductions in Africa and S. Ame
- China shows mixed features

Unit: Gg N/ month
Global NOx emissions are lower by ~28% (Nov 2005): Annual proj. (42 vs 31 Tg N/yr)
N.Ame (~48%) and Europe (~30%) have significant reduction in industrial NOx emissions (2005 vs 1998)
A posteriori BB emissions are well matched with GFEDv2 (global total)
Comparison with Mass Balance emissions estimate

Based on Martin et al., (2003)

\[
\ln E = \frac{\ln E_t \left( \ln \varepsilon_a \right)^3 + \ln E_a \left( \ln \varepsilon_t \right)^3}{\left( \ln \varepsilon_a \right)^3 + \left( \ln \varepsilon_t \right)^3}
\]

- Some over-reduction over N. Ame and Europe
- less reduction over S. Ame, Africa, and India
- Less increase over China and Australia
- No latitudinal correlations (Lifetime?)
<table>
<thead>
<tr>
<th>Region</th>
<th>a Priori</th>
<th>Mass-Balance</th>
<th>Adjoint</th>
</tr>
</thead>
<tbody>
<tr>
<td>N. Ame.</td>
<td>682</td>
<td>424 (-38%)</td>
<td>364 (-47%)</td>
</tr>
<tr>
<td>Europe</td>
<td>490</td>
<td>410 (-16%)</td>
<td>357 (-27%)</td>
</tr>
<tr>
<td>E. Asia</td>
<td>535</td>
<td>687 (+28%)</td>
<td>481 (-10%)</td>
</tr>
<tr>
<td>India</td>
<td>202</td>
<td>126 (-38%)</td>
<td>130 (-36%)</td>
</tr>
<tr>
<td>S. Ame.</td>
<td>391</td>
<td>249 (-36%)</td>
<td>286 (-27%)</td>
</tr>
<tr>
<td>Africa</td>
<td>652</td>
<td>458 (-30%)</td>
<td>451 (-31%)</td>
</tr>
<tr>
<td>Aus.</td>
<td>169</td>
<td>237 (+40%)</td>
<td>166 (-2%)</td>
</tr>
<tr>
<td>Global</td>
<td>3456</td>
<td>2877 (-19%)</td>
<td>2507 (-28%)</td>
</tr>
</tbody>
</table>
SCIAMACHY NO$_2$ columns (Nov 2005) from Dalhousie Univ

Previous version

Reprocessed (currently used)

Previous - reprocessed

Reprocessed SCIAMACHY
• Improved in solar measurements, and cloud fields for signal fitting and AMF calculation

(10$^{15}$ Molecules/cm$^2$)
Conclusions

- According to the adjoint inversion, the N.Ame and European anthropogenic NOx emissions are greatly reduced by 48% and 30% (1998 vs 2005)
- Large increase in Chinese industrial NOx emissions is evident by SCIAMACHY, but the adjoint inversion does not represent enough enhancements
- Natural a priori NOx emissions overestimated (Nov. 2005) and a posteriori biomass burning emission is closer to GFEDv2 in global total

More ..

- Comparison with recent emissions inventory for specific regions (EPA, EMEP, Streets, etc..) will be continued.