Modeling the Atmospheric Transport and Deposition of Mercury to the Great Lakes

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- Great Lakes Restoration Initiative
**Mercury in Great Lakes Fish**

<table>
<thead>
<tr>
<th>Fish species</th>
<th>Mean overall fish mercury (ppm, ww)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rainbow smelt</td>
<td>0.01</td>
</tr>
<tr>
<td>Alewife</td>
<td>0.01</td>
</tr>
<tr>
<td>Whitefish</td>
<td>0.01</td>
</tr>
<tr>
<td>Brown trout</td>
<td>0.01</td>
</tr>
<tr>
<td>Coho salmon</td>
<td>0.01</td>
</tr>
<tr>
<td>Lake sturgeon</td>
<td>0.01</td>
</tr>
<tr>
<td>Rainbow trout</td>
<td>0.01</td>
</tr>
<tr>
<td>Yellow perch</td>
<td>0.01</td>
</tr>
<tr>
<td>Chinook salmon</td>
<td>0.01</td>
</tr>
<tr>
<td>American eel</td>
<td>0.01</td>
</tr>
<tr>
<td>Common carp</td>
<td>0.01</td>
</tr>
<tr>
<td>Lake trout</td>
<td>0.01</td>
</tr>
<tr>
<td>Smallmouth bass</td>
<td>0.01</td>
</tr>
<tr>
<td>Walleye</td>
<td>0.01</td>
</tr>
<tr>
<td>Northern pike</td>
<td>0.01</td>
</tr>
<tr>
<td>Largemouth bass</td>
<td>0.01</td>
</tr>
</tbody>
</table>

0.05 ppm level recommended by the Great Lakes Fish Advisory Workgroup (2007)

NOAA’s HYSPLIT atmospheric transport and dispersion modeling system
http://journals.ametsoc.org/doi/abs/10.1175/BAMS-D-14-00110.1

HYSPLIT can do more than just back-trajectories

Transport and dispersion based on local or rawinsonde data

Transport and dispersion based on gridded meteorological data
To simulate the global transport of mercury, puffs are transferred to an Eulerian grid after a specified time downwind (~3 weeks), and the mercury is simulated on that grid from then on.

When puffs grow to sizes large relative to the meteorological data grid, they split, horizontally and/or vertically.

This is how we model the local & regional impacts.

But for global modeling, puff splitting overwhelms computational resources.

Puffs of pollutant are emitted and dispersed downwind.

Atmospheric chemistry and deposition simulated for each puff.
Today's talk will just present Eulerian-only results, for 2005

These Eulerian-only simulations were much faster (a few weeks) than Lagrangian-only and hybrid Lagrangian-Eulerian simulations (3-6 months)

It's important to fail fast!
1/e Half-Lives for Fate Processes within HYSPLIT-Hg

Starting with Uniform Mixing Ratio Throughout the Atmosphere

1/e Half-Life (years)

Hg(p)
- Hg(p) dry dep only
- Hg(p) wet and dry dep
- Hg(p) wet dep only (wetr=40K)
- Hg(p) wet dep only (wetr=80K)

Hg(II)
- Hg(II) hv reduction
- Hg(II) SO2 reduction
- Hg(II) reduction
- Hg(II) dry dep only
- Hg(II) wet dep only

Hg(0)
- Hg(0) wet dep only
- Hg(0) all aq phase oxid33 reactions
- Hg(0) other gas phase oxid33 reactions
- Hg(0) gas-phase o3 oxid 6.0E-21
- Hg(0) gas-phase o3 oxid 1.0E-20
- Hg(0) dry dep only
- Hg(0) gas-phase oh oxid 2.9E-14

Hg(0) oxidation rate 67% – no reduction
Hg(0) oxidation base case – no reduction

Hg(0) oxidation base case – no reduction
- 0.84 year = 10 months

Hg(II) dry dep only
- 0.2 year = 2.4 months

Hg(II) wet dep only
- 0.048 year = 2.5 weeks

Hg(p)
- 0.74

Hg(II)
- 2.9

Hg(0)
- 828
- 213
- 7.1
- 5.6
- 3.5
- 3.5
- 1.4
- 1.4
- 0.84 year = 10 months
Study Year was 2005

Emissions Flux (ng/m2-day)
2000.gbl2p5_emit_joined_to_grid
Hgtot_ngm2
Emissions Inventories with Net Ocean and Terrestrial Hg(0) Fluxes

- **anthrop Hg(tot)**
- **anthrop Hg(2) + Hg(p)**
- **anthrop Hg(p)**
- **anthrop Hg(2)**
- **anthrop Hg(0)**
- **total net ocean + terrestrial Hg(0)**
- **total net terrestrial Hg(0)**
- **total net ocean Hg(0)**

**GEOS-Chem**
Grid is too coarse to expect good agreement with measurements, but...

2.5° x 2.5° grid

Type of Emissions Source
- coal-fired power plants
- other fuel combustion
- waste incineration
- metallurgical
- manufacturing & other

Emissions (kg/yr)
- 5–10
- 10–50
- 50–100
- 100–300
- 300–500
- 500–1000
- 1000–3000
Which inventory components show the right seasonal variation?
modeled vs. measured Hg(0) concentrations during 2005, for measurement sites in the Great Lakes region and sites outside the region
Hg(II) + Hg(p) at Reno (Desert Research Inst.)

Graph showing the concentration of Hg(II) + Hg(p) over time from January 1, 2005, to December 1, 2005. The graph includes different categories such as reemission, land, biomass, anthropogenic, geogenic, and ocean measurements. The concentrations are measured in parts per billion per cubic meter (pg/m³).
modeled vs. measured Hg(II) and Hg(p) concentrations during 2005

Model estimated total for all non-elemental mercury is higher than measurements:

Are the measurements too low or is the model too high?
modeled vs. measured 2005 Hg wet deposition for Mercury Deposition Network (MDN) sites in different regions
2005 Modeled Atmospheric Mercury Deposition (ug/m²-yr)

Erie  Ontario  Michigan  Huron  Superior  GL Avg

- OTHER
- RUSSIA
- MEXICO
- INDIA
- CANADA
- CHINA
- USA
- geogenic
- biomass
- re-emission
- land
- ocean
Base-case results compared with recent CMAQ modeling:
<table>
<thead>
<tr>
<th></th>
<th>Base Case</th>
<th>Variation 1</th>
<th>Variation 2</th>
<th>Variation 3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>4-D-iii</td>
<td>5-O-i</td>
<td>6-V-i</td>
</tr>
<tr>
<td>Oxidation Reaction rates</td>
<td>default</td>
<td>60% less</td>
<td>default</td>
<td>default</td>
</tr>
<tr>
<td>Prompt Hg(II) Conversion</td>
<td>0%</td>
<td>0%</td>
<td>50%</td>
<td>67%</td>
</tr>
<tr>
<td>Anthropogenic Emit (Mg/yr)</td>
<td>1930</td>
<td>1930</td>
<td>1930</td>
<td>1930</td>
</tr>
<tr>
<td>Land + Reemissions + Ocean</td>
<td>4350</td>
<td>2300</td>
<td>4350</td>
<td>4350</td>
</tr>
<tr>
<td>{Net Hg(0) emissions} (Mg/yr)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Particle Fraction of Hg(0)</td>
<td>10%</td>
<td>25%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>oxidation</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WETR (wet deposition parameter)</td>
<td>40,000</td>
<td>80,000</td>
<td>40,000</td>
<td>40,000</td>
</tr>
<tr>
<td>Great Lakes Hg(0) bias</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
</tr>
<tr>
<td>Great Lakes Hg wet dep bias</td>
<td>+2%</td>
<td>-8%</td>
<td>-10%</td>
<td>-14%</td>
</tr>
</tbody>
</table>
Mercury Emissions (Mg/yr)

- **anthrop Hg(tot)**
- **anthrop Hg(2)**
- **anthrop Hg(p)**
- **anthrop Hg(0)**

- **total net ocean + terrestrial Hg(0)**
- **total net terrestrial Hg(0)**
- **total net ocean Hg(0)**
2005 Modeled Atmospheric Mercury Deposition (ug/m²-yr)

**Base Case**

- Erie
- Ontario
- Michigan
- Huron
- Superior
- GL Avg

**1: 60% oxid; ~50% L+R+O net Hg0**

- OTHER
- RUSSIA
- MEXICO
- INDIA
- CANADA
- CHINA
- USA

**2: Plume HgII convert 50%**

- OTHER
- RUSSIA
- MEXICO
- INDIA
- CANADA
- CHINA
- USA

**3: Plume HgII convert 67%**

- OTHER
- RUSSIA
- MEXICO
- INDIA
- CANADA
- CHINA
- USA
- HYSPLIT-Hg Base Case gives reasonable results for 2005
  - Several variations also considered, which can also give reasonable results for 2005

- Source Attribution results for the different cases are understandably somewhat different
  - However, policy implications of the different modeling results may not be that different

- Next steps:
  - Modeling for more recent years, including detailed model evaluation at three NOAA/EPA monitoring sites (within AMNet):
    - Beltsville MD; Grand Bay MS; and Mauna Loa HI
  - + Nested grid + explicit sub-surface terrestrial & ocean layers
Thanks!

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