Application of Regional Models for Assessing Source-Receptor Relationships for a Global Pollutant: Capabilities and Concerns for Atmospheric Mercury

Russell Bullock

National Oceanic and Atmospheric Administration (NOAA)
Atmospheric Sciences Modeling Division
(in partnership with the U.S. Environmental Protection Agency)

UNEP Global Partnership on Atmospheric Mercury Transport and Fate Research
&
Task Force on Hemispheric Transport of Air Pollution of the UNECE-LRTAP Convention

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Regional Modeling of a Global Pollutant

- Even if we understand all of what is happening inside the model domain (certainly questionable for mercury), we also need to know what is happening outside, or at least at the boundaries.
- Limited-area modeling of the atmosphere has always required some definition of conditions at the model boundaries.
- Weather models only need physical conditions (e.g., wind, temperature, pressure, humidity) but air pollution modeling also needs air concentration data for the pollutants in question and all of their reactants.
Defining Boundary Conditions

• In the early days of atmospheric Hg modeling, typical “background” values would usually suffice for all levels and all lateral sides of the modeling domain.

• For early Hg modeling, we just used 1.6 ng/m$^3$ for Hg$^0$ and 40 ppb for ozone (and most of us remembered to scale the Hg$^0$ values to atmospheric density aloft).

• Atmospheric Hg was almost entirely Gaseous Elemental Mercury (GEM), a nearly inert gas with a very long atmospheric lifetime. But sometimes a small fraction of aerosol Hg could be detected or a small fraction of gaseous Hg that was unusually reactive.

• Boundary concentrations for these odd forms of Hg were almost always set to near zero.
Long Range Transport of Air Pollutants

• But acid deposition was found to have a significant component from intercontinental transport.

• The same was found for ozone and for various other pollutants.

• The “good old days” of regional Hg modeling were about to end!
Intercontinental Transport

• GEM was known to travel VERY long distances in a year since jet-stream winds can carry air around the world in a week. But “RGM” was so reactive that its long range transport was generally discounted.

• “Reactive Gaseous Mercury” was observed in industrial emissions, but its production from GEM oxidation was thought to be minor.

• The first observations of “disappearing GEM” in the arctic were believed to be some sort of measurement error due to harsh conditions.
What you don’t know can’t hurt ya.
It’s what you know for sure that just ain’t so that’ll kill ya!

- Yogi Berra
The U.S. EPA’s Clean Air Mercury Rule

• On March 15, 2005, the U.S. EPA issued the Clean Air Mercury Rule (CAMR) to permanently cap and reduce mercury emissions from coal-fired electric generating units (EGUs).

• The CAMR builds on the U.S. EPA’s Clean Air Interstate Rule (CAIR) targeting $\text{SO}_2$ and $\text{NO}_x$. When fully implemented, these rules will reduce EGU emissions of mercury from 48 tons per year to 15 tons per year.

• The Community Multiscale Air Quality (CMAQ) model was used to support the development of the CAIR and the CAMR.
CMAQ Mercury Model Description

• 3-D Eulerian-type model
• Multi-scale (urban to continental)
• For this study:
  ▪ 36-km horizontal grid (112 x 148)
  ▪ 14 vertical layers (hi-res in PBL)
• Initial condition and boundary condition (IC/BC) data provided by GEOS-Chem global-scale model simulation
Simulated Total Hg Deposition for the CAMR Analysis
(micrograms per square meter)
CMAQ-Simulated Total Hg Deposition in 2020
(micrograms per square meter)
Percent Reduction in Hg Deposition from the 2001 Base Case
North American Mercury Model Intercomparison Study (NAMMIS)

Motivation

- Various modeling studies have been conducted to estimate the sources responsible for observed Hg deposition in the United States and other nations. These studies have sometimes come to rather different conclusions.

- An earlier Hg model inter-comparison study was conducted by the Meteorological Synthesizing Centre – East (MSC-E). This first study provided a valuable measurement of variation in modeling results for Europe.

- The NAMMIS was a follow-on effort to apply atmospheric Hg models in a more tightly constrained testing environment where all models use exactly the same input data and the focus of the study is on North America.
Each regional-scale model used the same horizontal modeling domain, the same meteorology and emissions data, and the same boundary conditions.

Three sets of initial condition / boundary condition (IC/BC) data are used which are based on simulations of three separate global-scale models.
North American Mercury Model Intercomparison Study (NAMMIS)

Global Models for IC/BC development
- Chemical Transport Model (CTM) developed and applied by Atmospheric and Environmental Research, Inc.
- GEOS-Chem model developed and applied by Harvard University.
- Global/Regional Atmospheric Heavy Metals (GRAHM) model developed and applied by Environment Canada.

Regional Models that are the focus of the study
- Community Multi-scale Air Quality (CMAQ) model developed and applied by NOAA and U.S. EPA.
- Regional Modeling System for Aerosols and Deposition (REMSAD) developed and applied by ICF International.
- Trace Element Analysis Model (TEAM) developed and applied by Atmospheric and Environmental Research, Inc.
NAMMIS Regional Modeling Domain
(same as for CAMR)
Our first bit of insight was gained from looking at the boundary conditions provided from the three global models.
Annual Average Western Boundary Values
*(global models certainly differ – which is correct?)*

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<th>Hg⁰</th>
<th>RGM</th>
<th>PHg</th>
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February 2001 Average Lateral Boundary Values
February 2001 Average Lateral Boundary Values
February 2001 Average Lateral Boundary Values

Height (m)

HgP conc. (ppm: atoms of Hg per molecule of air)
Regional Model Simulations and Results Analysis

• CMAQ, REMSAD and TEAM were each applied for all of 2001 using all three IC/BC data sets.
• Air concentration fields for Hg species and wet and dry deposition patterns for total-Hg have been compared among these three models.
• Wet deposition from these three models have been compared against observations from the Mercury Deposition Network.
Annual Avg. Air Concentrations of Hg\(^0\) (ng/m\(^3\))
(surface layer)
Annual Avg. Air Concentrations of RGM (pg/m³) (surface layer)
Annual Avg. Air Concentrations of $Hg_P$ (pg/m$^3$) (surface layer)
Observed Hg Wet Deposition in 2001 from the Mercury Deposition Network (MDN)
Regional Modeling Results Comparison to Observed Total Hg Wet Deposition for 2001
Regional Modeling Results Comparison to Observed Total Hg Wet Deposition for 2001

$R^2$ Correlation Factors
Boundary Conditions and Modeling Accuracy

- Observed model sensitivity to changes in Hg species concentrations at the boundary shows that global-scale transports are important to regional-scale assessments.

- Additional analyses of the CMAQ results have been performed to better quantify the effects of uncertainies regarding global-scale transport of “imported” Hg.
Wet Deposition Patterns from Three Test Cases and Total Precipitation Pattern

CTM

GEOS-Chem

GRAHM

Total Precip
Wet Deposition of “Imported” Mercury

• The CMAQ simulations were repeated for all three IC/BC test cases, this time with all natural and anthropogenic mercury emissions set to zero.
CTM zero-emission results
GEOS-Chem zero-emission results
GRAHAM zero-emission results
Findings / Conclusions

- Spatial patterns for wet deposition of “imported” Hg are largely the same for each of the three boundary condition cases, but magnitudes vary.

- Global models differ in their concentrations of Hg species at the lateral boundaries of the study domain.

- Observations of Hg⁰, RGM and particulate Hg are needed at all levels of the troposphere, especially along the western boundary.
Further Comments or Questions?

E-mail: bullock.russell@epa.gov
       o.russell.bullock@noaa.gov

Phone: (919) 541-1349

CAMR web site: http://www.epa.gov/air/mercuryrule/

Two journal articles on the NAMMIS submitted to JGR.