Assessing Contributions to Mercury Pollution in Northeastern US Using CMAQ-PPTM Mercury Tagging

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BACKGROUND

- The Northeast Center for Atmospheric Science and Policy (NCASP) has been established collaboratively by the Northeast States Center for a Clean-Air Future (NESCCAF) and the University of New Hampshire's AIRMAP Program with the objectives of improving understanding of the science behind current pressing atmospheric policy issues as well as developing regionally-appropriate analytical tools for supporting policy development.
- Currently NCASP's major focus is on mercury issue in the region. Extensive scientific data show that mercury is pervasive in freshwater fish in the Northeast at levels that pose plausible health risks to fish consumption people and wildlife.
- The goal of this NCAPS project is to develop and evaluate a state-of-science air quality modeling tool for assessing the mechanisms and contributing sources which lead to mercury pollution in the Northeastern U.S., and thus to support development of effective mercury control strategies for the region.

MODELING DESCRIPTION

- CMAQV4.5.1 mercury version with the Particle and Precursor Tagging Methodology (PPTM) developed by ICF International for U.S. EPA (Braverman et al., 2006; SAI, 2006) is adopted to be the air quality model. CB-IV gas phase chemical mechanism and mercury version of AQCHEM aqueous chemical mechanism are used. With CCTM_PPTM-Hg, mercury species in the emissions and initial and boundary condition can be tagged by source regions, or/and by source categories, and tracked throughout the CMAQ simulation.
- The modeling domain has 12km horizontal resolution with 162X144 grid cells and 21 vertical layers with first 3 layers in the lowest 100 meters. It covers the Northeastern U.S. and high emission regions in central and southeastern U.S. as shown in Figure 1. CMAQ-Hg run is conducted by UNH on the coarse 36km domain to provide boundary condition for the 12km domain.
- Meteorology is developed by UNH using the regional climate modeling system (RCMS) including MM5, then processed with MCIP3.2. Dry deposition velocity of Gaseous Elemental Mercury (GEM) and Reactive Gaseous Mercury (RGM) are also computed in MCIP.
- Modeling period covers a 4-month summer period from Jun. 2 to Sep.27 of 2004 because of the availability of ambient mercury measurements for model evaluation. The first 10 days is considered as spin-up to eliminate impact of initial concentration.
- UNH AIRMAP Program has continuous hourly measurements of total atmospheric mercury (GEM+RGM) for this period at Thompson Farm site. In addition, the mercury deposition network (MDN) continues to collect weekly wet deposition data at 43 sites within the 12km domain during this period. The location of monitoring sites is shown in Figure 2.
- Total mercury emission distribution is shown in Figure 3. Emission inventory (EI) of mercury is based on a regional Hg EI developed by NESCCAF for the 2002/2003 time period. The mercury inventory outside of the NESCCAF region relies on the U.S. EPA's Clean Air Mercury Rule (CAMR) inventory and the 2000 Canadian mercury inventory from Environment Canada. The recent version of year 2002 Regional Planning Organization (RPO) EI and the updated (i.e. 2002/2000) Canadian EI are used for the criteria air pollutants. Emission scenario is processed with SMOKE v2.3.

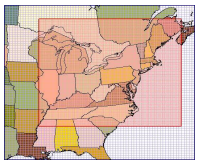


Figure 1. Modeling domains in this study

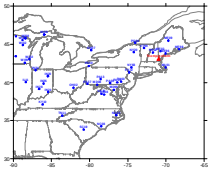


Figure 2. MDN sites (blue circle) and Thompson Farm site (red triangle) in 12km domain

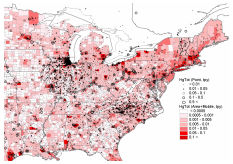


Figure 3. Total Hg emission distribution

Mercury Tags

- Tag1: GEM (Hg0) from Keystone facility
- Tag2: RGM (HgII) from Keystone facility
- Tag3: PHg (particulate Hg) from Keystone facility
- Tag4: All coal burning EGUs within in 200km radius of Steubenville measurement site
- Tag5: All non-coal burning EGUs within in 200km radius of Steubenville measurement site
- Tag6: All other Hg emission
- Tag7: Hg boundary condition

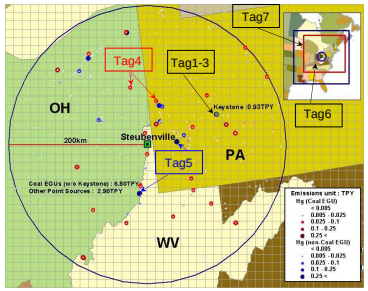


Figure 4. Illustration of Hg emission tag1-7. The total Hg emission within the circle is 10.6 TPY which is ~10% of total US mercury point source emissions.

Mercury Modeling Performance Evaluation

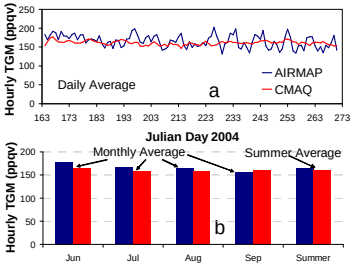


Figure 5. Temporal distribution of hourly TGM on AIRMAP measurement and CMAQ prediction at Thompson Farm site. a) daily average; b) monthly average and summer average

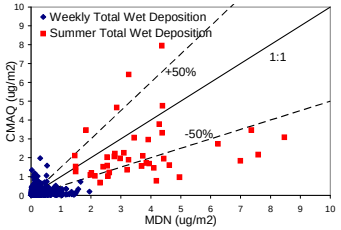
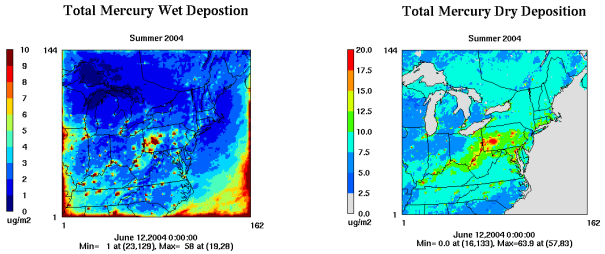


Figure 6. Comparison of MDN measurement and CMAQ prediction on weekly total Hg wet deposition and summer total Hg wet deposition.

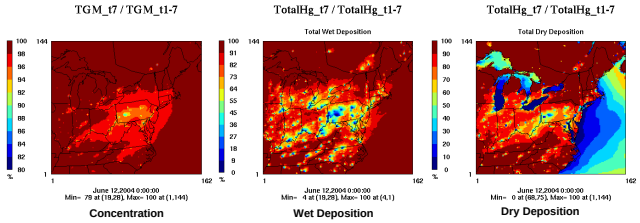
- Comparison of hourly TGM from AIRMAP measurement and CMAQ prediction at Thompson Farm site (Figure 5) shows a general agreement of them on long term average hourly value. The bias of mean hourly TGM varies from -0.8 to 1.7ppq for each of the 4 month and the whole summer modeling period with a summer mean of 165ppqv hourly TGM observation.
- The hourly TGM at rural site Thompson Farm exhibits strong daily oscillation ranging ~100ppqv. The model fails to capture such significant diurnal variation on daily basis.
- Comparison of weekly total of total mercury wet deposition between MDN measurement and CMAQ at MDN sites (Figure 6) also shows better agreement at longer term average.

Spatial Pattern of Mercury Deposition



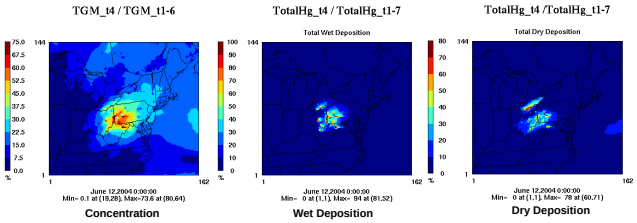
- Maximum wet deposition and dry deposition are observed at PA and OH area where large amount of anthropogenic Hg sources exist

Contribution from Boundary Condition



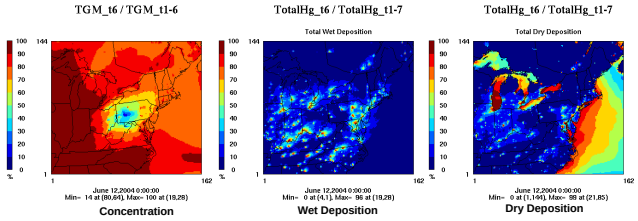
- Mercury from boundary condition dominates in ambient TGM concentration, as well as wet deposition and dry deposition most of domain including Northeastern U.S..
- Mercury from boundary condition contribute little to dry deposition over the water because of no dry deposition for GEM.

Contribution from Coal Burning EGUs in the Circle



- Mercury from all coal burning EGUs in 200km radius of Steubenville becomes major component in local and region ambient TGM concentration without boundary Hg.
- Mercury from coal burning EGUs contributes >90% in total Hg wet deposition, and >70% in total Hg dry deposition in region, even with boundary Hg included.

Contribution from Emission Sources outside the Circle



- While not considering boundary Hg, Mercury from emission sources out the Steubenville circle dominants ambient TGM concentration within area.
- Mercury from outside the circle only become significant contributor to wet deposition in spots where large Hg point sources locate, and to dry deposition over coast water.

Summary

- This study has established a modeling tool with CMAQ_PPTM-Hg model with updated EI to assess mercury impact over northeastern U.S. and to identify pollution sources for supporting effective control strategies policymaking.
- The model can characterize fate of Hg within a long term perspective.
- In general Hg across the boundary contributes most to Hg deposition in Northeastern U.S. with exception at area with large Hg point sources.
- The modeling shows strong local or near the source impact on mercury deposition. The >90% contribution on wet deposition for Hg from coal burning EGUs near Steubenville is consistent with receptor-based modeling results (G. Keeler et al., 2006).

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