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Estimation of Speciated and Total Mercury Dry Deposition

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Contributors:

Pierrette Blanchard, David Gay, and many others

Why the study?

- **Mercury dry+wet deposition needs to be quantified** (e.g., to assess Hg mass balance, Hg effects on ecosystems, emission control policies)
- **Dry deposition can be as important as or more important than wet deposition** (e.g., results from litterfall, throughfall, modeling studies)
- **There are much larger uncertainties in dry deposition than in wet deposition** (e.g., difficulties in direct measurements, in inferential modeling)
- **Dry deposition is strongly species-dependent** (e.g., large differences in their concentrations and dry deposition velocities)

Research ideas

- **Study 1: Analysis of modeled mercury dry deposition over the Great Lakes region** **(Draft paper ready for GLAD project, 'Environmental Pollution')**
  - ✓ Evaluate the model output using speciated mercury concentration and short-term flux measurement data
  - ✓ Analyze the modeled dry deposition field
  - ✓ Generate conclusions and provide rationale for future studies
- **Study 2: Estimation of speciated and total dry deposition at rural locations (AMNet sites+Canadian sites) across eastern North America** **(Work in progress)**
- **Study 3: Development of dry and total deposition maps for eastern North America** **(To be decided later)**

Study 1: Analysis of modeled mercury dry deposition over the Great Lakes region

L. Zhang, P. Blanchard, D. Johnson, A. Dastoor,
A.Ryjkov, J.C.H. Lin, K. Vijayaraghavan, D. Gay,
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Environment Canada
Convex Logic
Lamar University
ENVIRON
Clarkson University
University of Alberta
University of Maryland
Ecosystems Research Group Ltd.
University of Michigan
Ryerson University
University of Illinois
Cornell University

Data availability

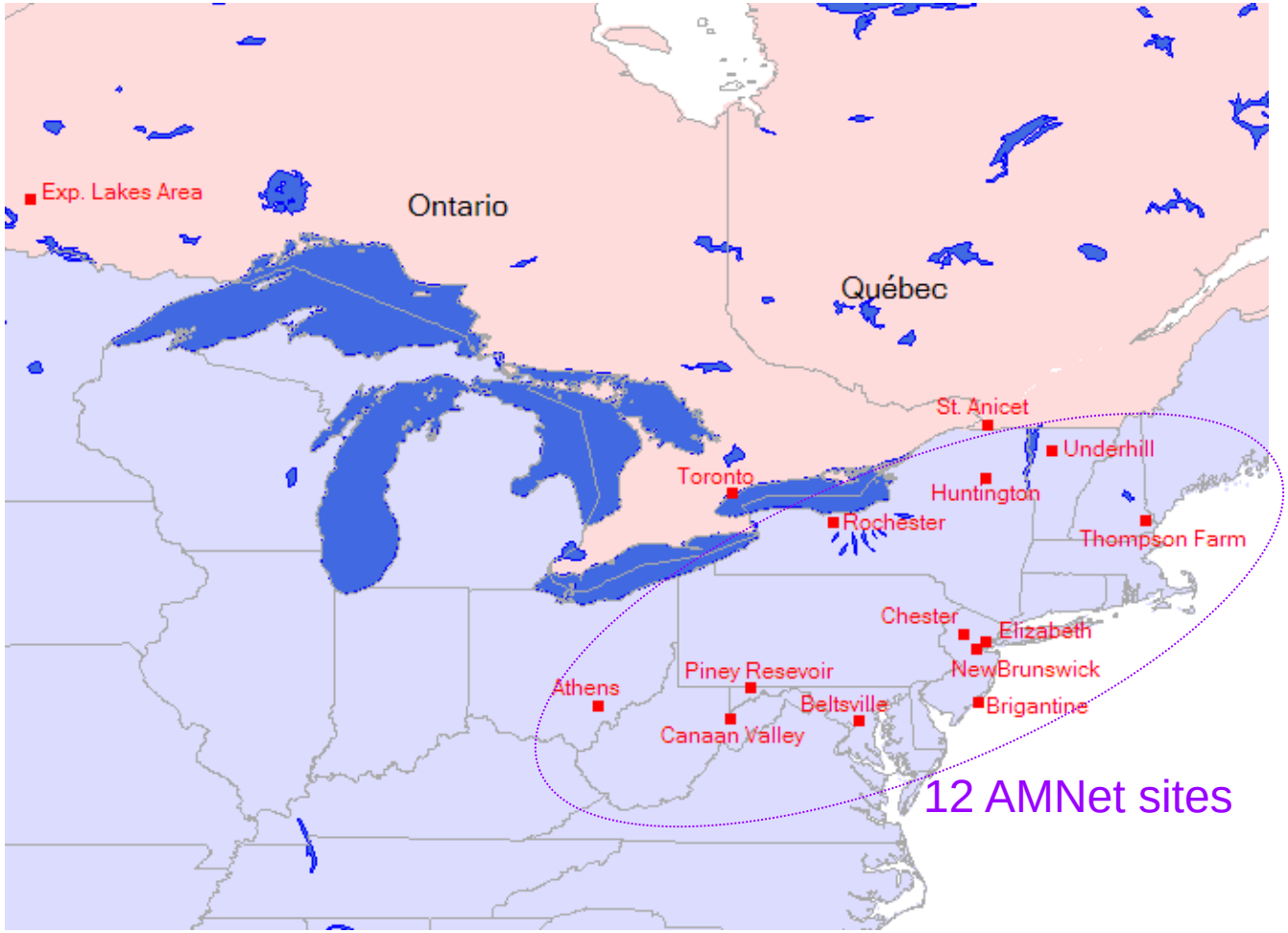
Model output: hourly speciated concentration and dry deposition

- ✓ CMAQ2002 (speciated deposition is not available)
  K. Vijayaraghavan of AER (now at Environ)
- ✓ CMAQ2005
  J. Lin of Lamar University
- ✓ GRAHM 2005
  A. Dastoor of Environment Canada

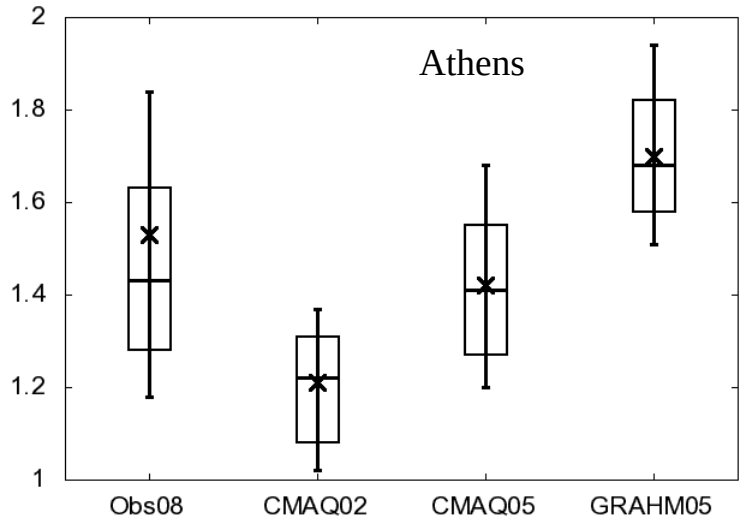
Field data:

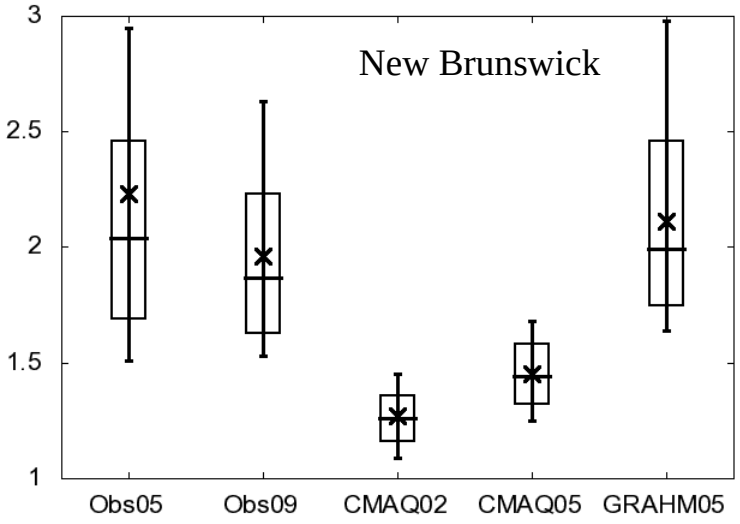
- ✓ One year speciated concentrations at 14 rural sites and one urban site (collected from 2005-2009)
- ✓ Two short-term dry flux measurements

Study domain and site locations



Measurement-model comparison: Hourly Hg^0 concentrations (ng m^{-3})





CMAQ2002:
Underestimated by 5-36% at all of the 14 rural sites

CMAQ2005
Underestimated by 5-26% at nine of the rural sites and agreed with the measurements (<5%) at the other 5 rural sites

GRAHM2005
Overestimated by 10-26% at seven of the rural sites and agreed with the measurements at the other seven rural sites (<5%).

All models underestimated Hg0 at the urban site by ~60%.

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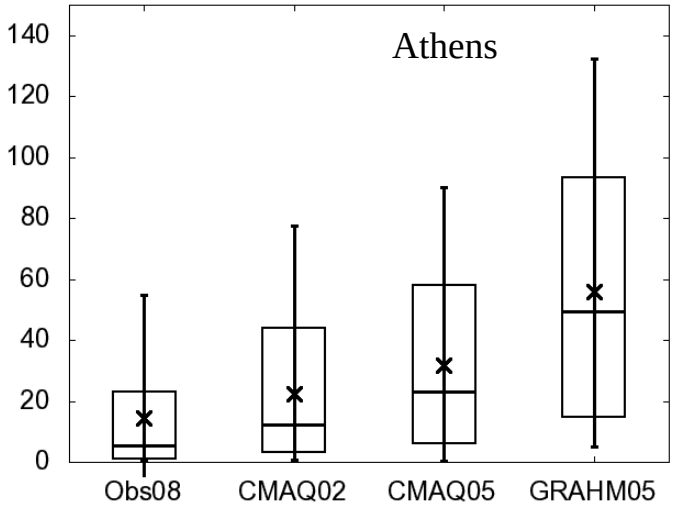
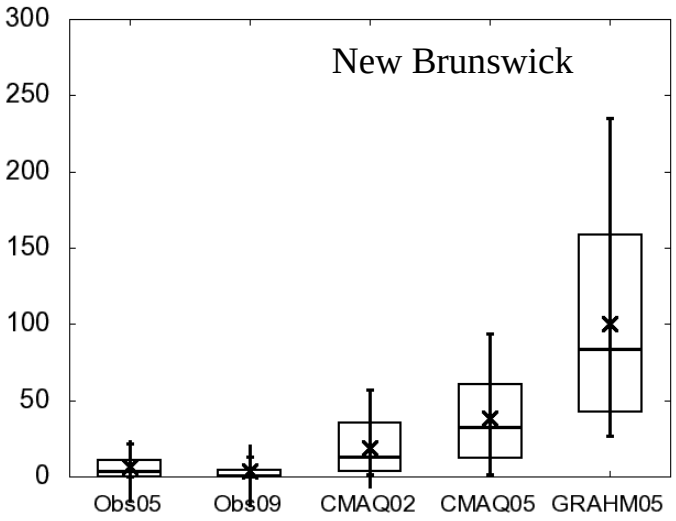
Measurement-model comparison: Hourly RGM concentrations (pg m^{-3})





CMAQ2002:
Overestimated by a factor of 2-8 at 13 sites

CMAQ2005
Overestimated by a factor of 2-27 at all of the 15 sites

GRAHM2005
Overestimated by a factor of 3.5-40 at all of the sites

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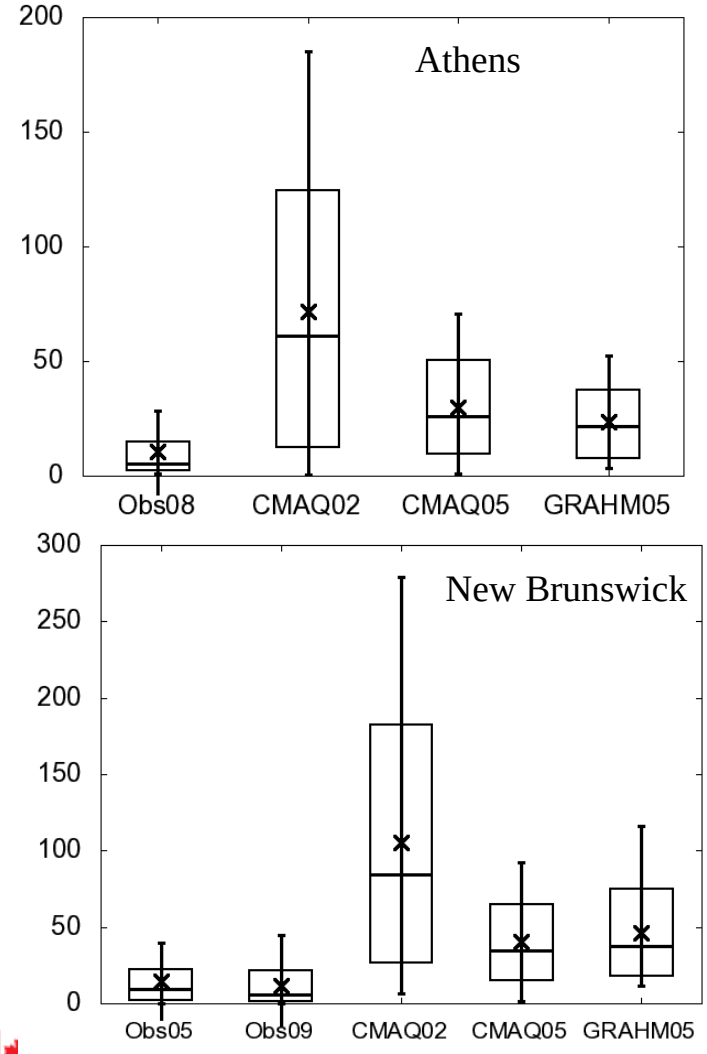
Measurement-model comparison: Hourly Hgp concentrations (pg m^{-3})



CMAQ2002:
Overestimated by a factor of 4-30 at all of the 15 sites

CMAQ2005
Overestimated a factor of 2-12 at 11 sites

GRAHM2005
Overestimated by a factor of 2-9 at 10 sites

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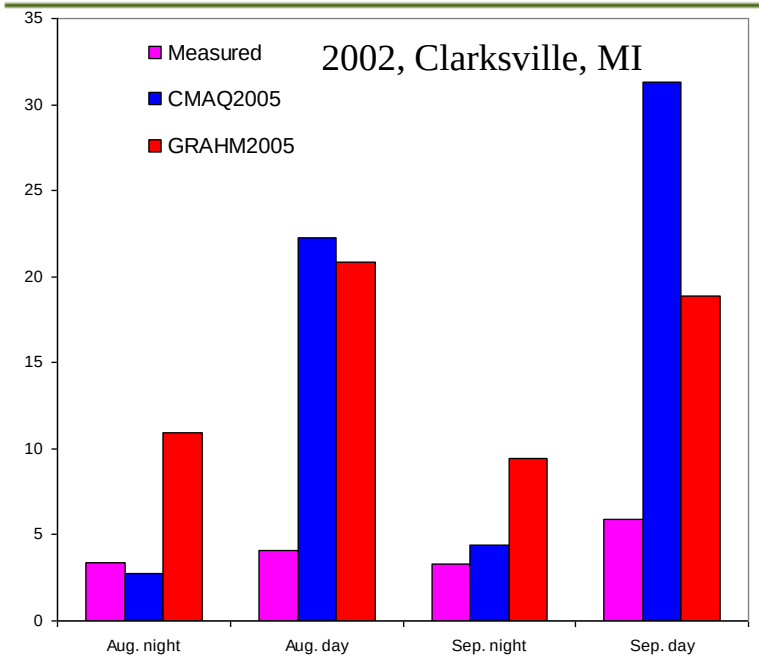
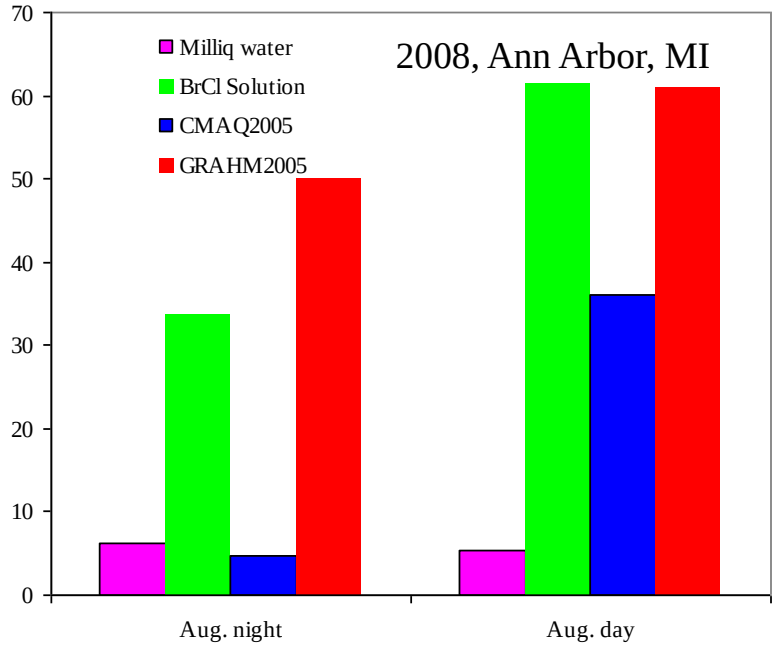
Measurement-model comparison: Short-term fluxes RGM+Hgp ($\mu g\ m^{-2}\ yr^{-1}$)



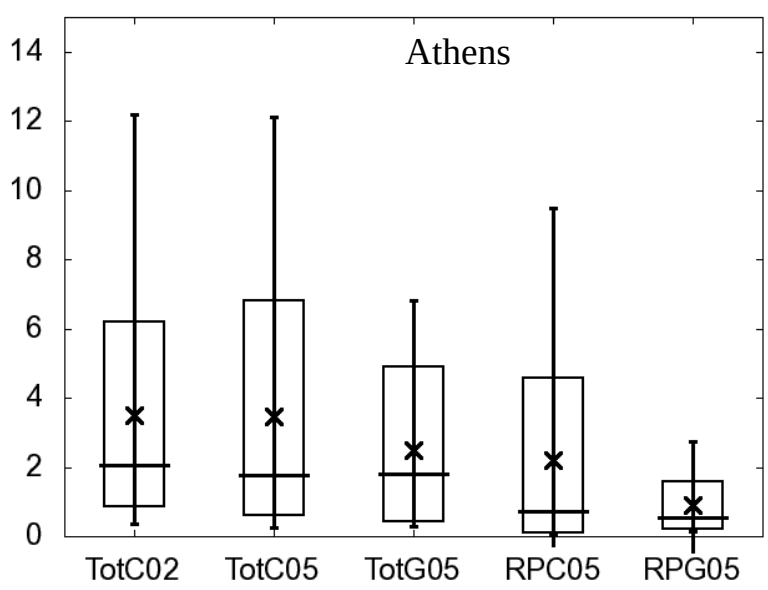


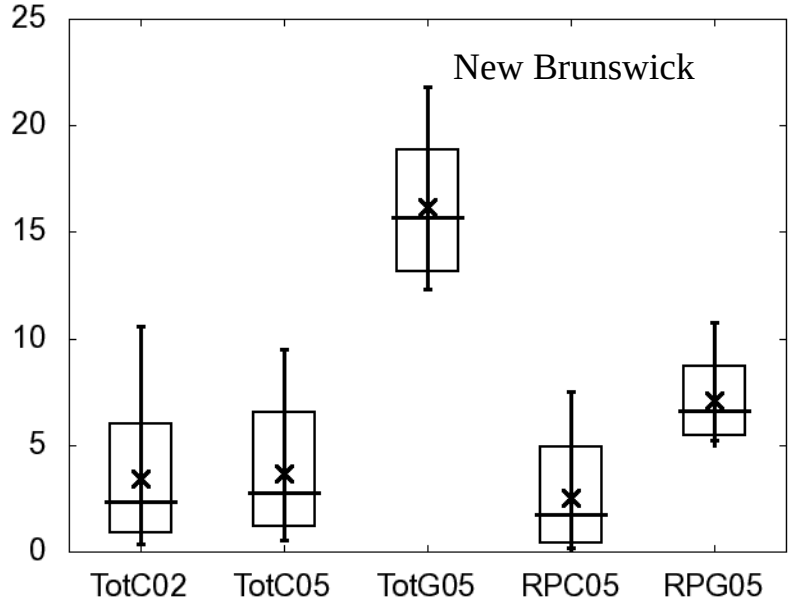
✓Large differences in measurements using different surrogated surfaces

✓Overpredicted by a factor of 2-10, mostly caused by the overprediction of concentration

✓CMAQ2005 nighttime fluxes close to measurements (due to the low deposition velocity used in the model)

Model-Model comparison:
Total fluxes and RGM+Hgp fluxes ($\mu g\ m^{-2}\ yr^{-1}$)

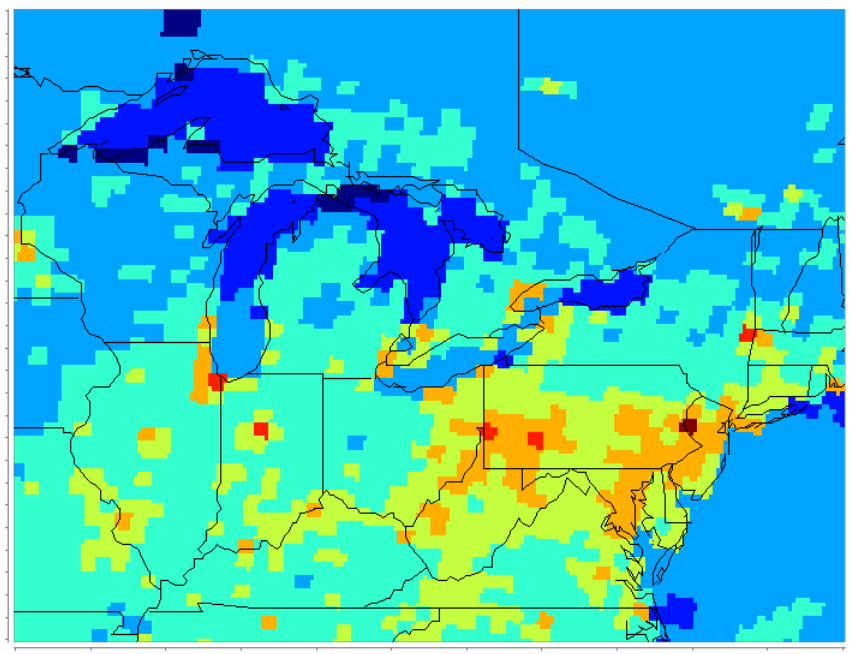


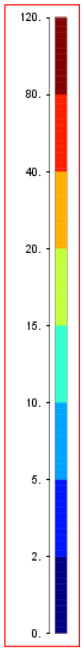


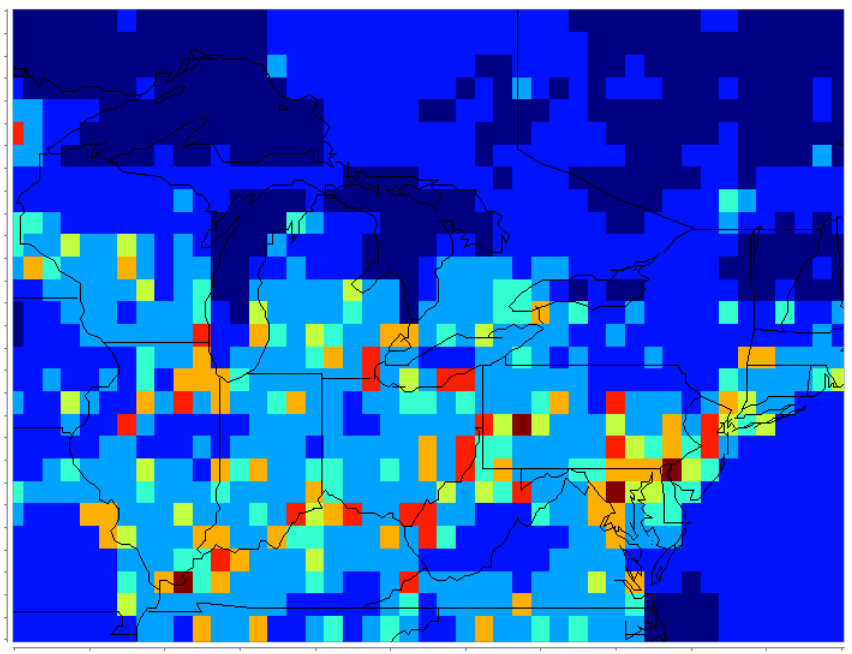
✓Typically 20-100% differences in total and RGM+Hgp deposition (for sites not close to point sources)

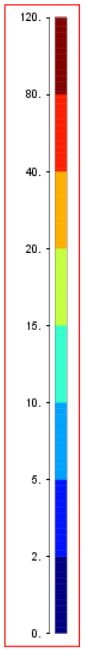
✓Can be larger than a factor of 2 at locations close to point sources

Modeled annual RGM+Hgp dry deposition ($\mu g\ m^{-2}\ yr^{-1}$)







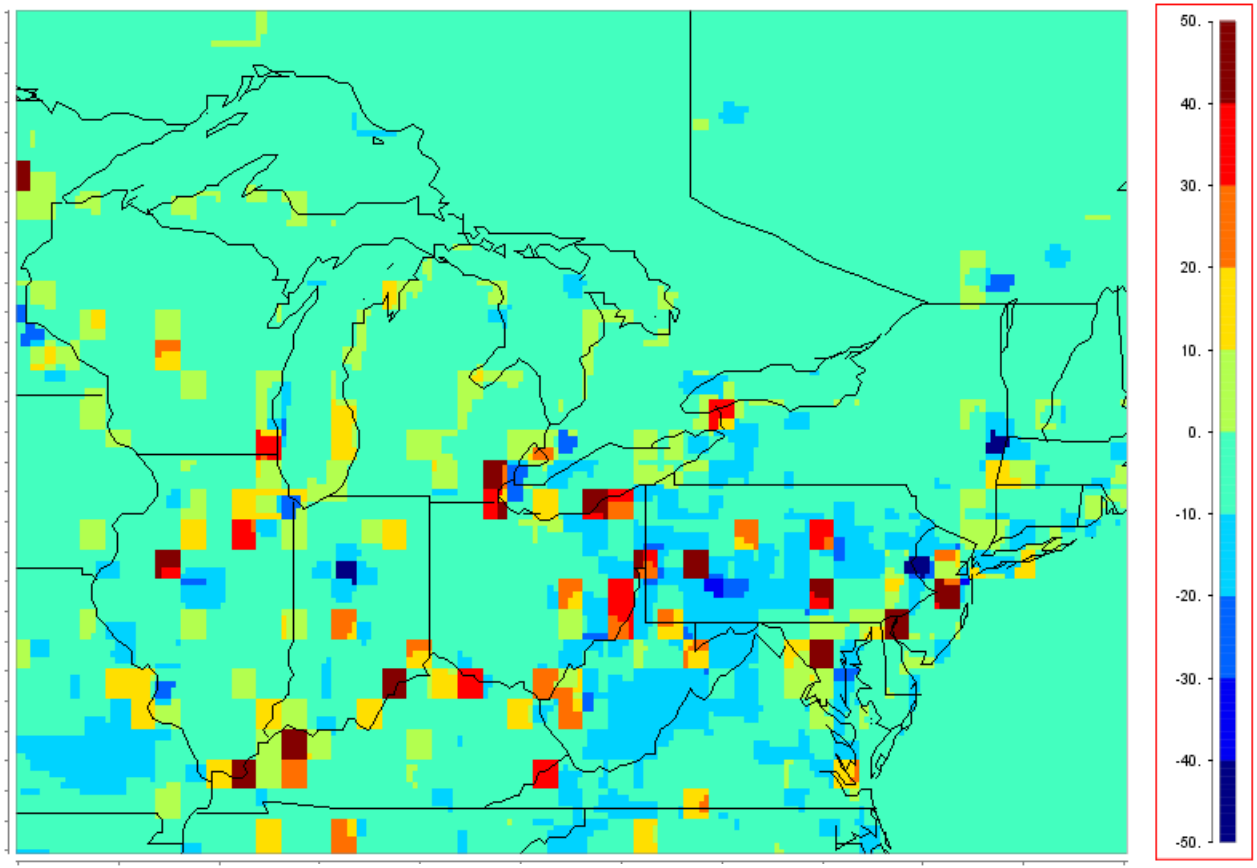


CMAQ2005

GRAHM2005

- ✓10-40 $\mu g\ m^{-2}$ from CMAQ2005 and 5-40 $\mu g\ m^{-2}$ from GRAHM2005 south of the border
- ✓CMAQ2005 shows a clear gradient with the highest deposition in Pennsylvania and its surrounding areas, but not GRAHM2005
- ✓GRAHM2005 has more hot spots (> 40 $\mu g\ m^{-2}$) than CMAQ2005
- ✓Lower than 15 $\mu g\ m^{-2}$ from CMAQ2005 and lower than 5 $\mu g\ m^{-2}$ from GRAHM2005 north of the border
- ✓Lower than 5 $\mu g\ m^{-2}$ water surface

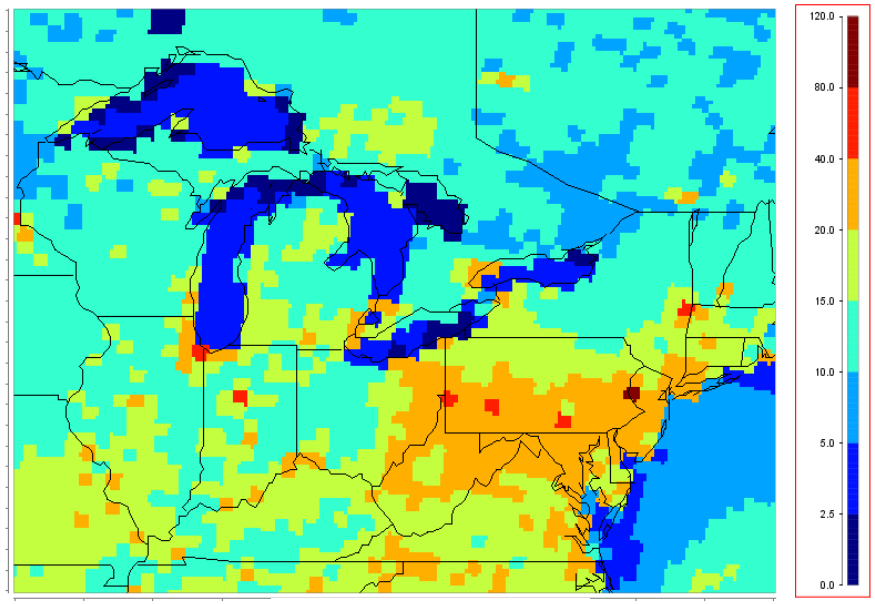
Differences between two models in annual RGM+Hgp dry deposition ($\mu g\ m^{-2}\ yr^{-1}$)



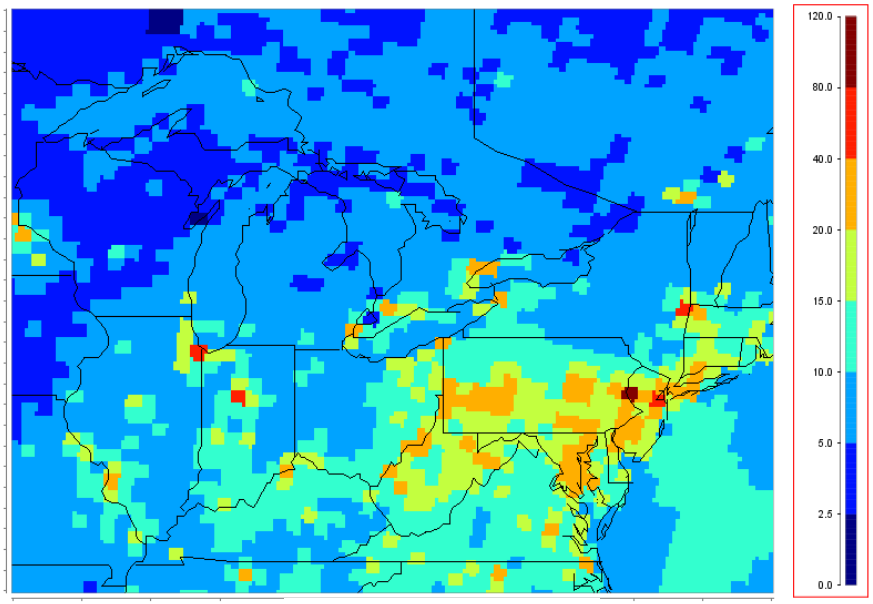
GRAHM2005-CMAQ2005

Differences can be larger than a factor of 2 in many places

Seasonal pattern in annual RGM+Hgp dry deposition from CMAQ2005



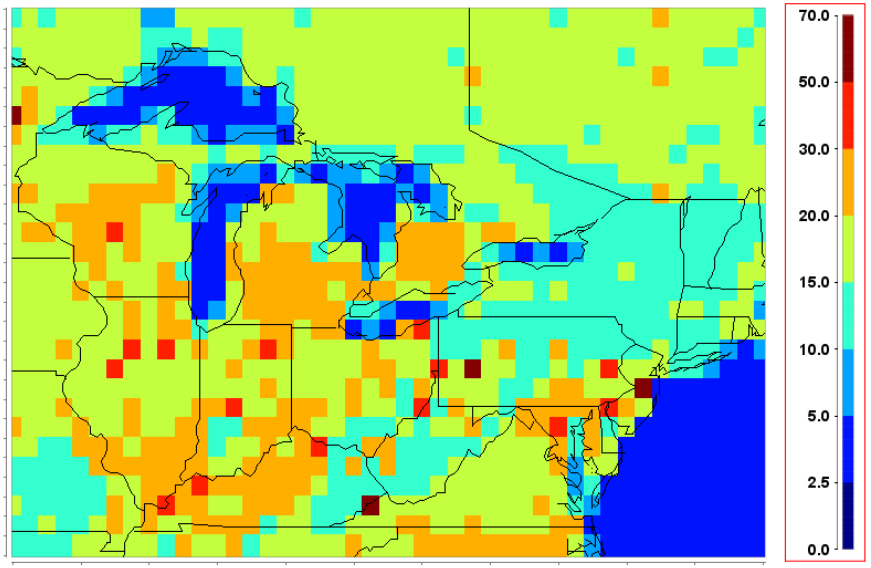
Mar-Apr-May



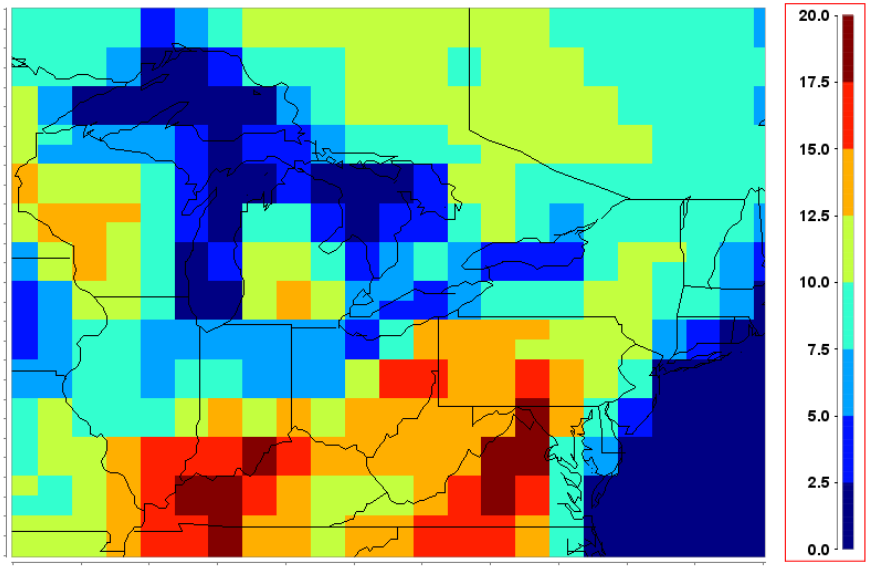
Dec-Jan-Feb

CMAQ2005: Spring>Summer>Fall>winter

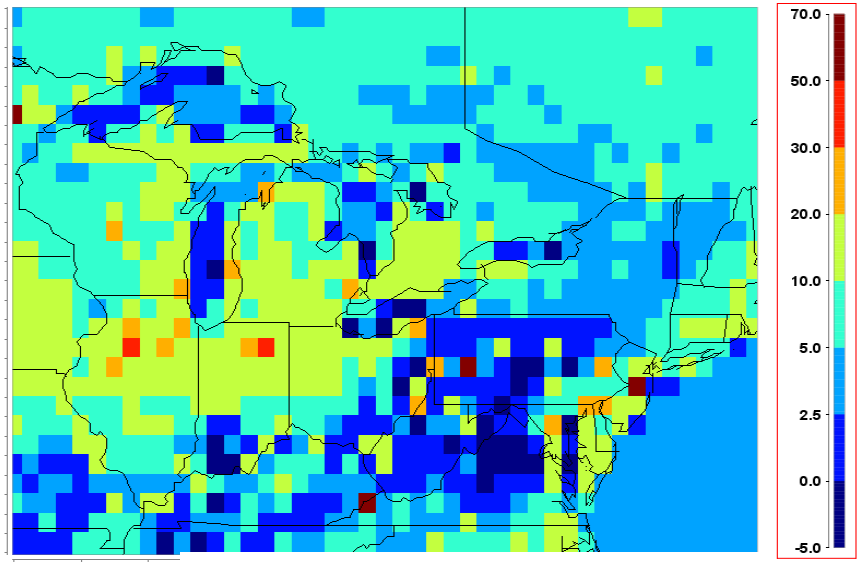
Modeled Hg0 deposition/reemission from GRAHM2005



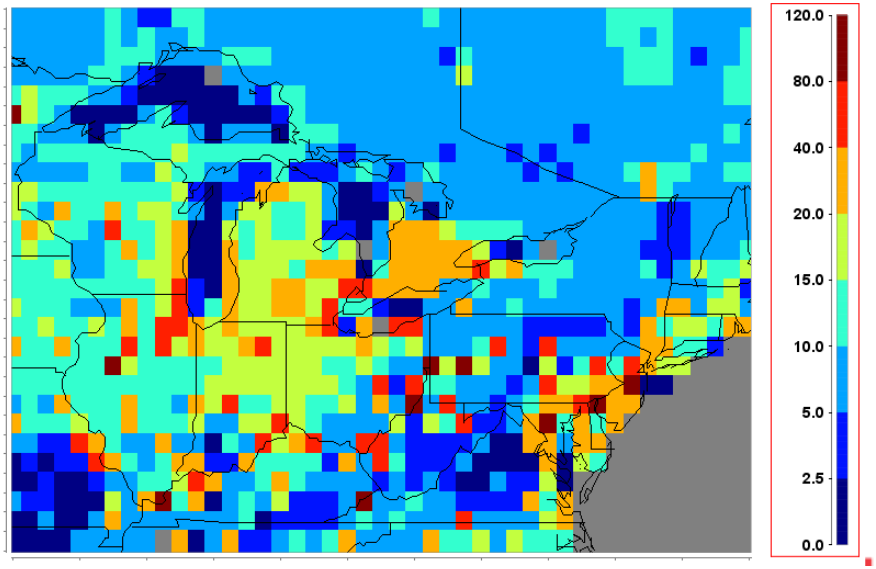
Hg0 deposition **µg m^{-2} yr^{-1}**



Hg0 reemission **µg m^{-2} yr^{-1}**



Hg0 (deposition – reemission)



(RGM+Hgp+Hg0) deposition – Hg0 (natural+reemission)

Conclusions from study 1

- ✓ Models seem to produce reasonable Hg0 concentrations, but not for RGM and Hgp
- ✓ Differences in modeled dry deposition can be as large as a factor of 2 between different models. The confidence in the modeled deposition is very low
- ✓ If using monitored Hgp and RGM concentrations to estimate the mercury dry deposition, one can expect much lower values than the modeled values presented here
- ✓ Hg0 deposition can be as important as or more important than RGM+Hgp (at places with low soil Hg contents)

Planned work for study 2

Estimation of speciated and total mercury dry deposition at rural locations across eastern North America. L. Zhang, P. Blanchard, D. Gay, M.R. Risch, RE.K. Miller, T. Holsen, J. Graydon, M.S. Castro, et al.

➢Objective: To get a relatively good estimation of mercury dry deposition at multiple (15+) locations across eastern North America

➢Approach:

- ✓ Inferential modeling
  - -Speciated concentration data (all AMNet sites in eastern USA and two Canadian sites)
  - -Dry deposition models (Zhang et al., 2001, 2003)
  - -Surface layer meteorological data (15 km by 15 km): archived from Canadian weather forecast model (3-hourly).
  - -Deposition and reemission of Hg0 will be considered.
- ✓ compare the estimated dry deposition with litterfall measurements
- ✓ Quantify the relative importance of dry and wet deposition

Study 3?

Objective:

- ✓ To develop a method of generating dry and total deposition maps for eastern North America

Rationale:

- ✓ Modeled deposition maps have large uncertainties
- ✓ Not enough dry deposition monitoring sites to generate maps
- ✓ More wet deposition monitoring sites
- ✓ Can wet deposition be linked with surface air concentration? (e.g., washout ratio concept, but complicated by more than one Hg species and in-cloud removal)