Modeling the Atmospheric Deposition of Mercury to the Great Lakes

(work funded through the Great Lakes Restoration Initiative)

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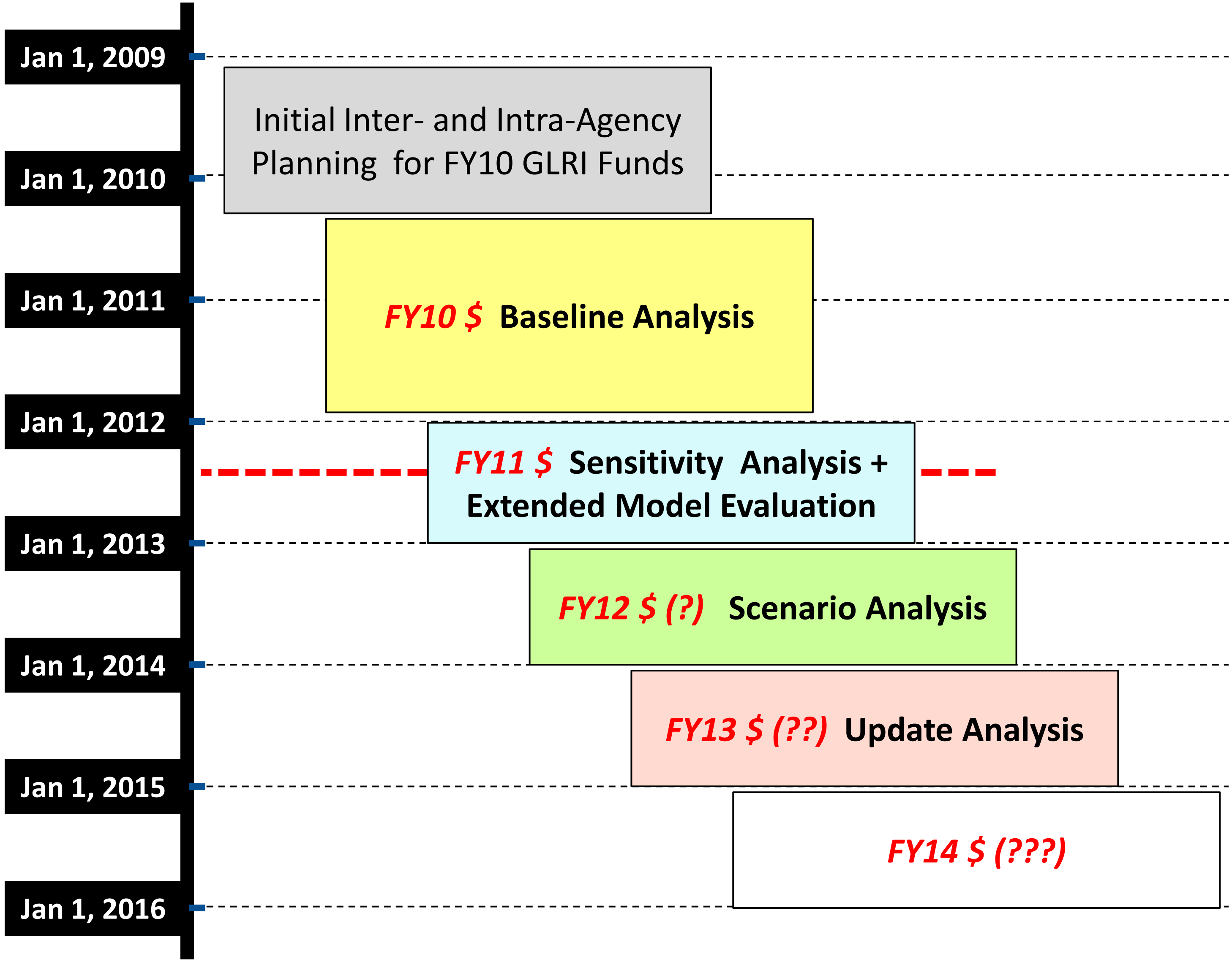
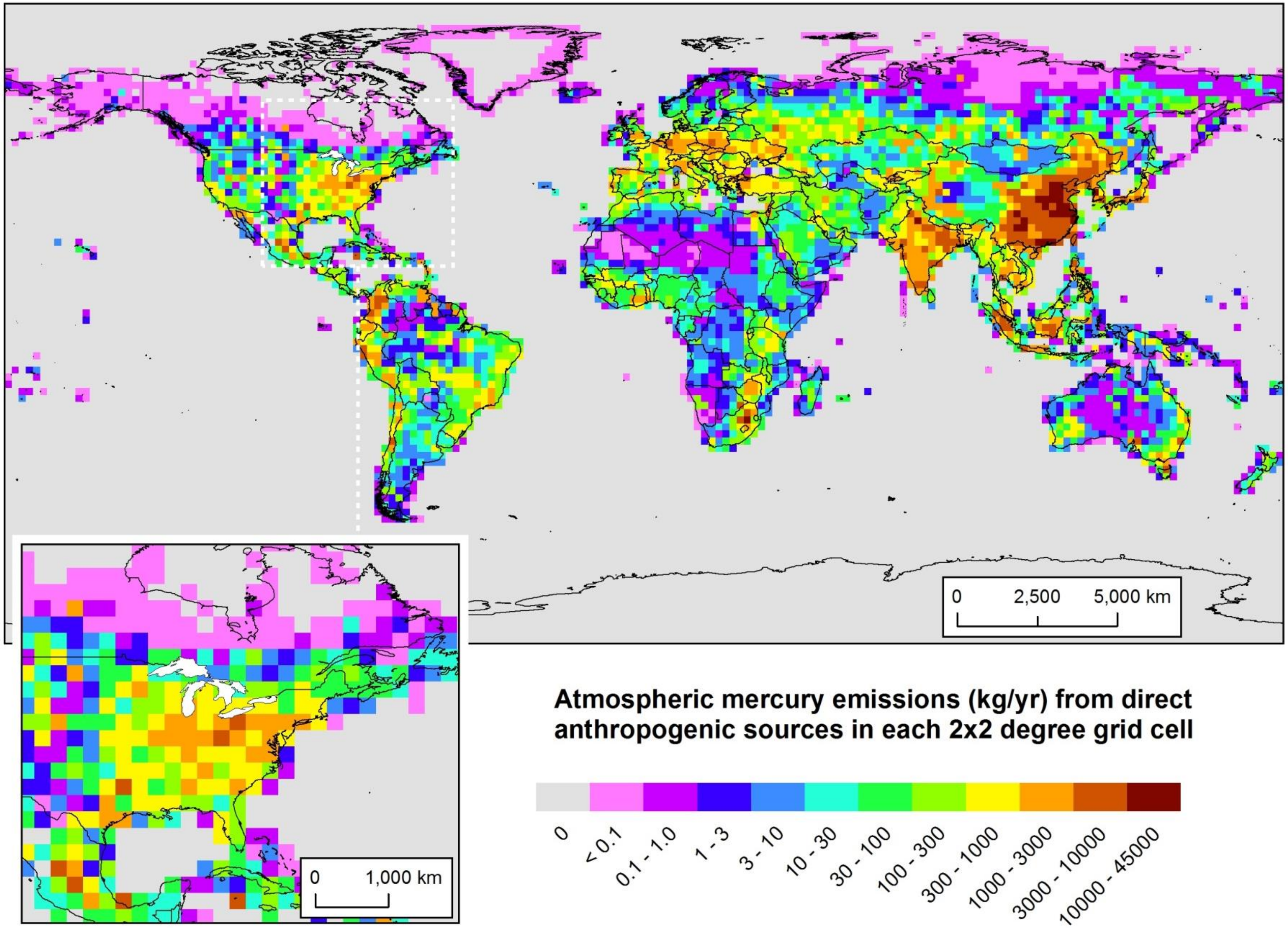
ARL's GLRI Mercury Modeling Project Timeline



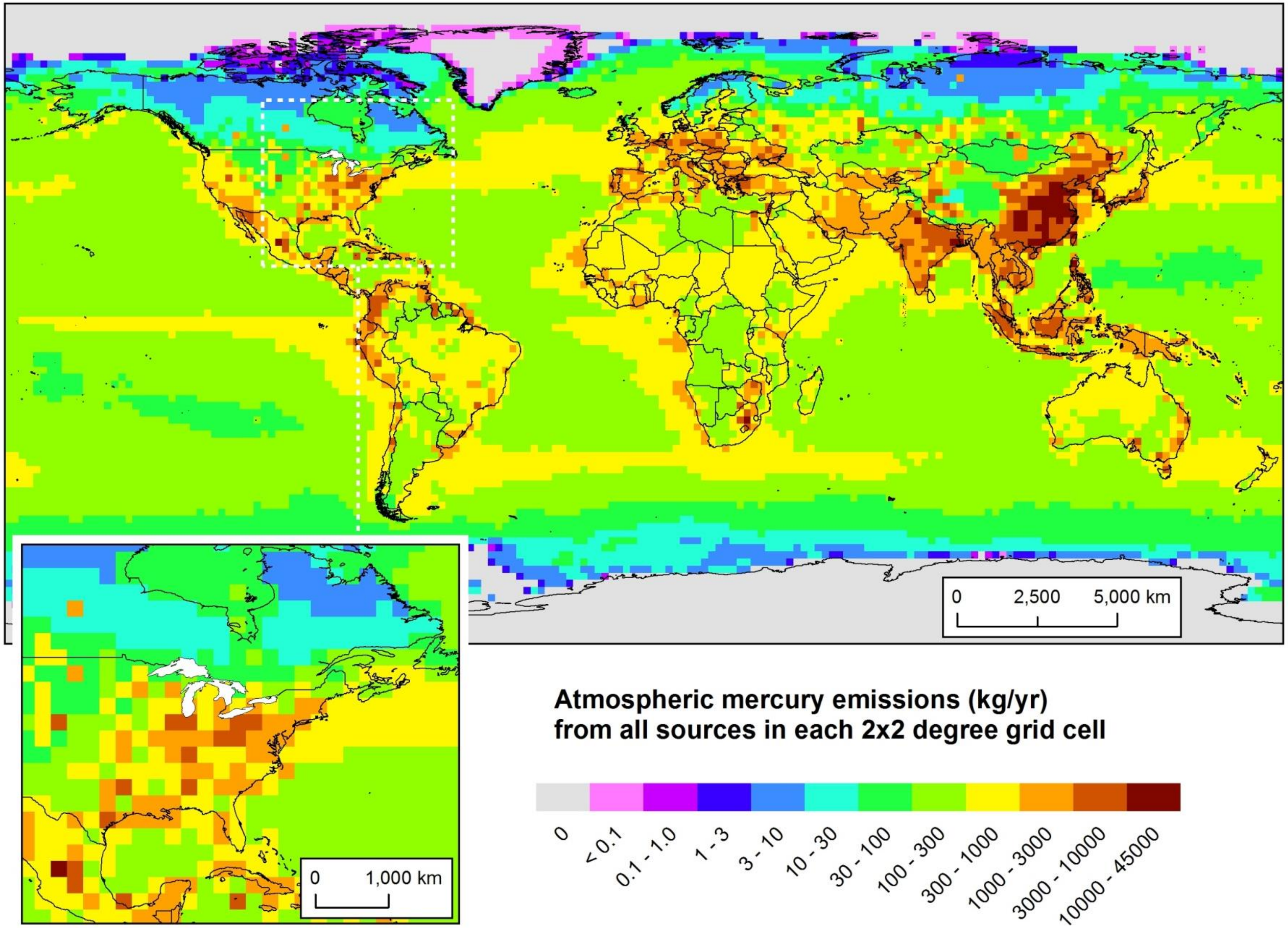
Phase 1:
Baseline Analysis

Emissions

Anthropogenic Mercury Emissions (ca. 2005)



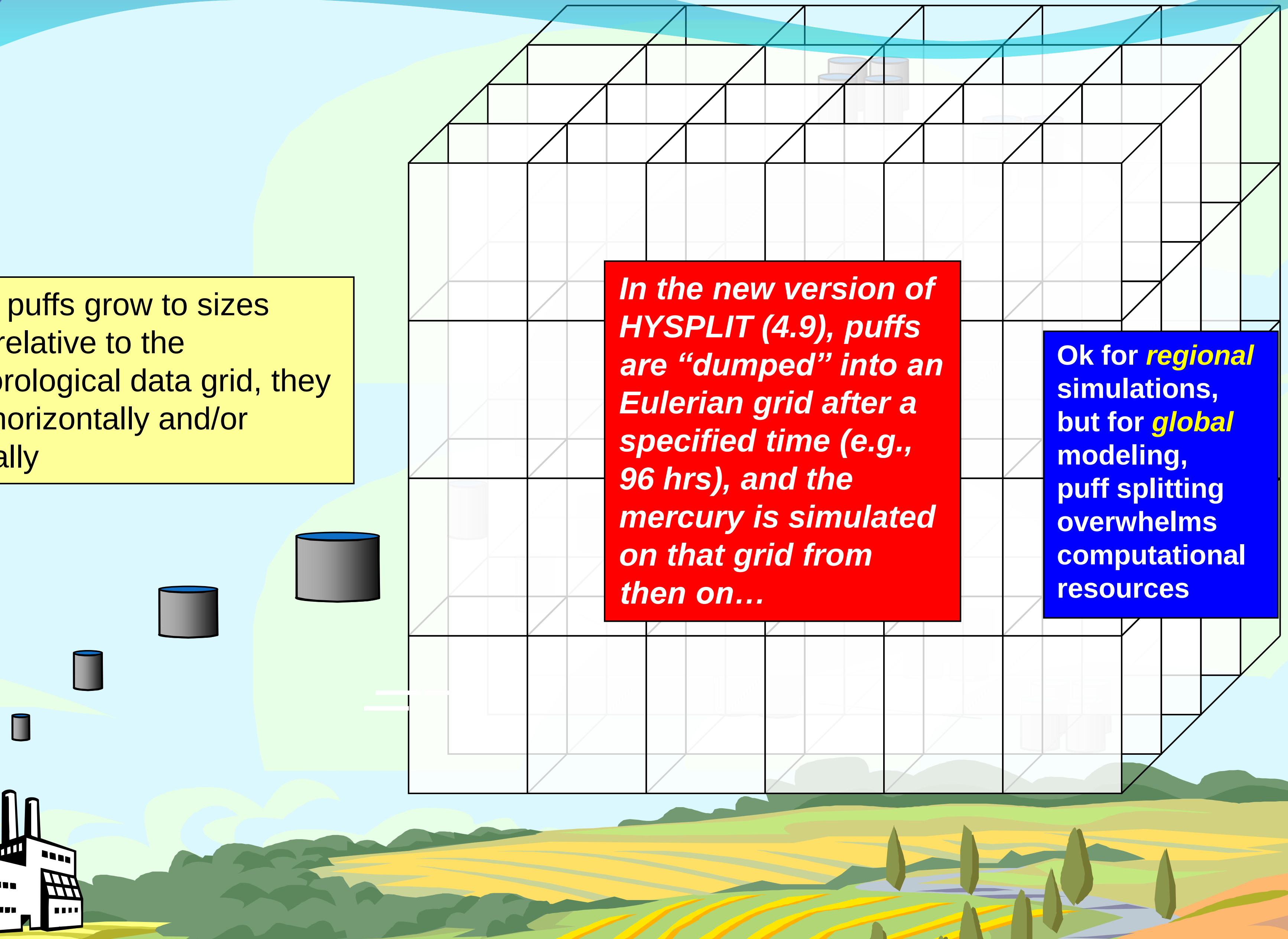
Atmospheric Mercury Emissions
(Direct Anthropogenic + Re-emit + Natural)



Methodology

Can we run HYSPLIT globally?

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



In the new version of HYSPLIT (4.9), puffs are "dumped" into an Eulerian grid after a specified time (e.g., 96 hrs), and the mercury is simulated on that grid from then on…

Ok for *regional* simulations, but for *global* modeling, puff splitting overwhelms computational resources

What year to model?

- **Mercury Emissions Inventory**
  - U.S. anthropogenic emissions inventory
  - Canadian anthropogenic emissions inventory
  - Mexican anthropogenic emissions inventory
  - Global anthropogenic emissions inventory
  - Natural emissions inventory
  - Re-emissions inventory
- **Ambient Data for Model Evaluation**
  - Wet deposition (Mercury Deposition Network)
  - "Speciated" Air Concentrations
- **Meteorological Data to drive model**
  - NCEP/NCAR Global Reanalysis (2.5 deg)
  - NCEP EDAS 40km North American Domain
  - North American Regional Reanalysis (NARR)

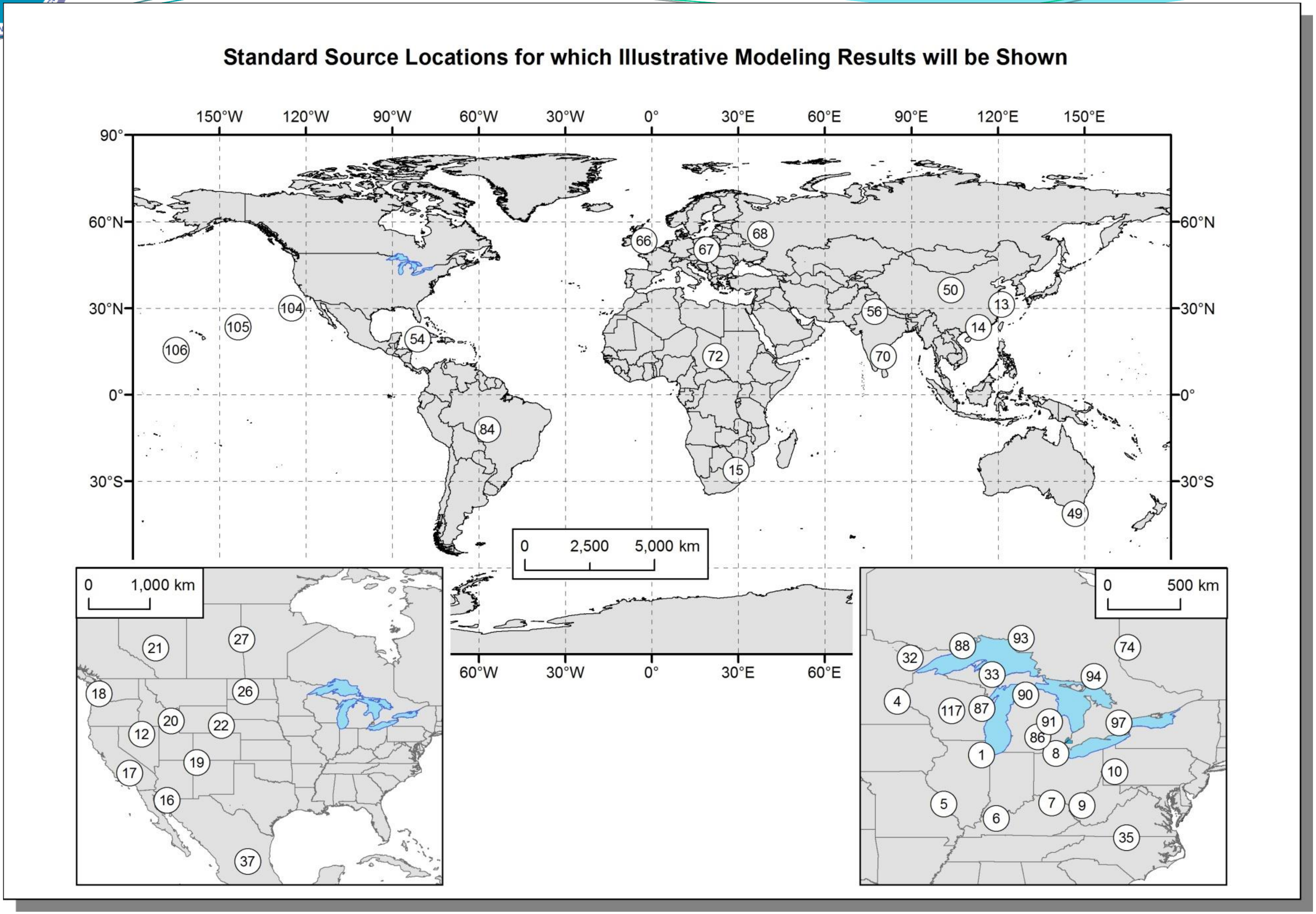
Dataset Available for 2005

Need all of these datasets for the same year

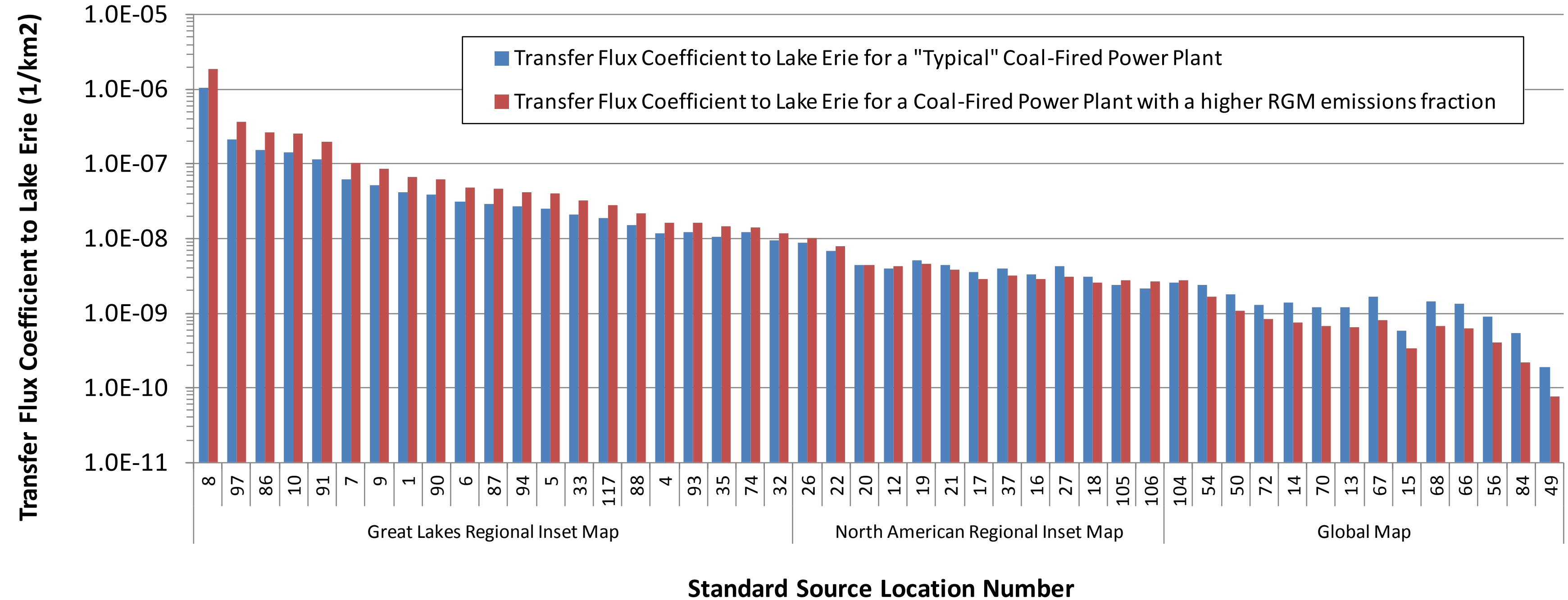
2005 chosen for baseline analysis

Illustrative Results for Single Sources

Standard Source Locations for Illustrative Modeling Results



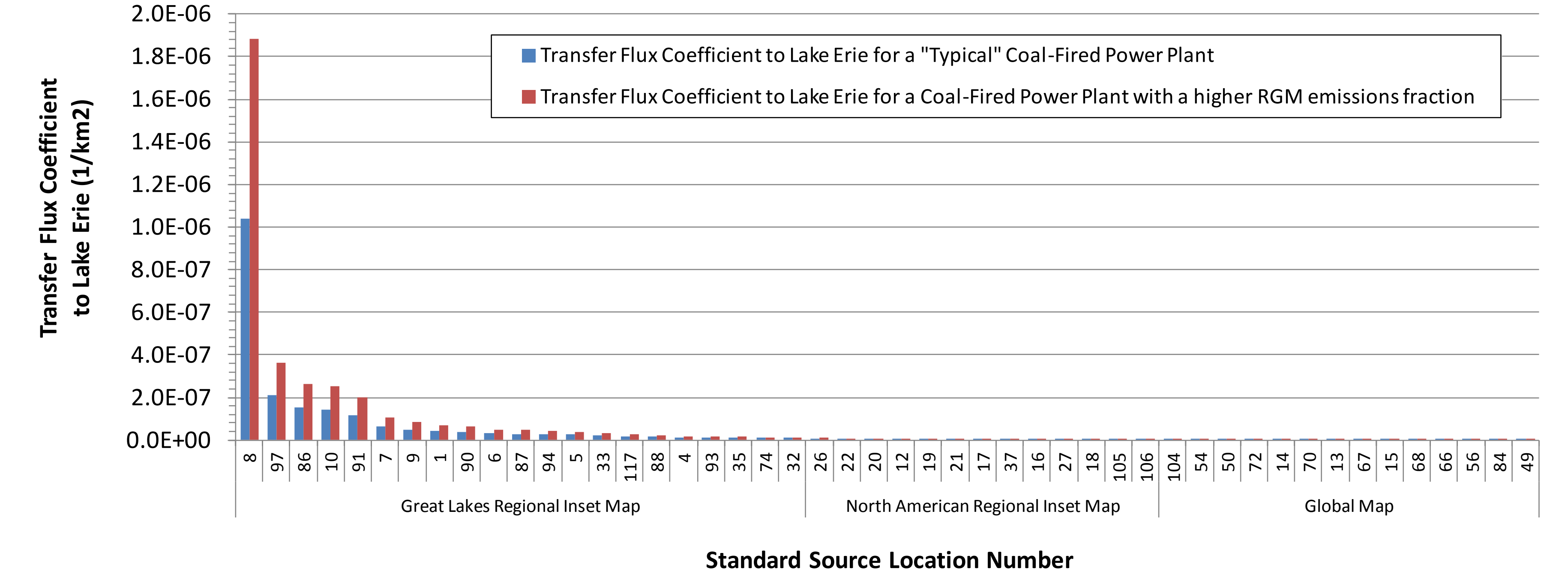
Lake Erie Transfer Flux Coefficients for two kinds of Generic Coal-Fired Power Plants (logarithmic scale)



The "Transfer Flux Coefficient" is calculated as the atmospheric deposition flux to a given receptor (in this case, Lake Erie) in units of g/km2-yr, divided by the total emissions from the source, in units of g/yr.

With this transfer flux coefficient, if one knows the emissions of the source in the given location, then the atmospheric deposition flux impact of the source on the receptor can be estimated, by simply multiplying the emissions by the transfer flux coefficient.

Lake Erie Transfer Flux Coefficients for two kinds of Generic Coal-FIred Power Plants (linear scale)



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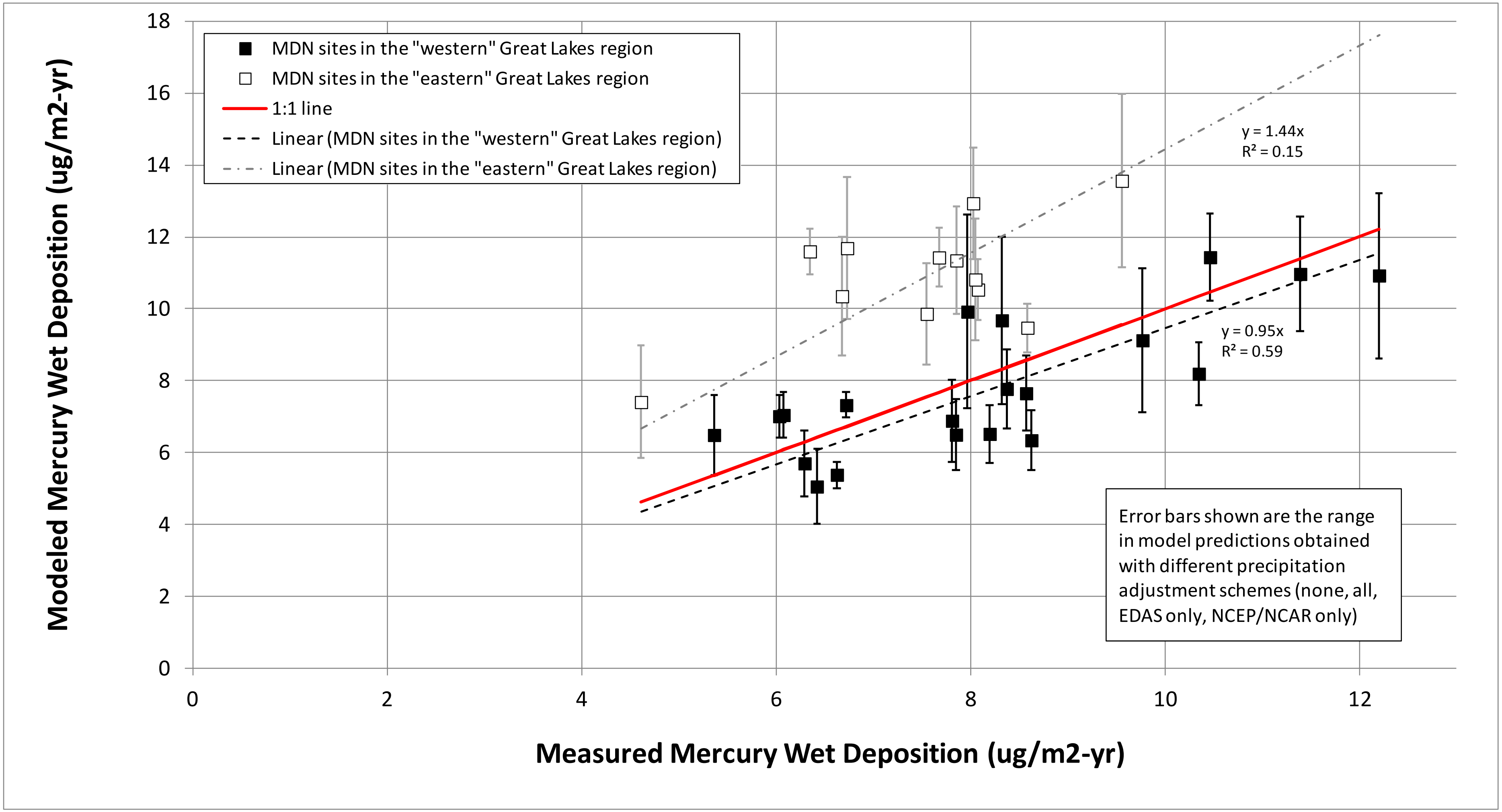
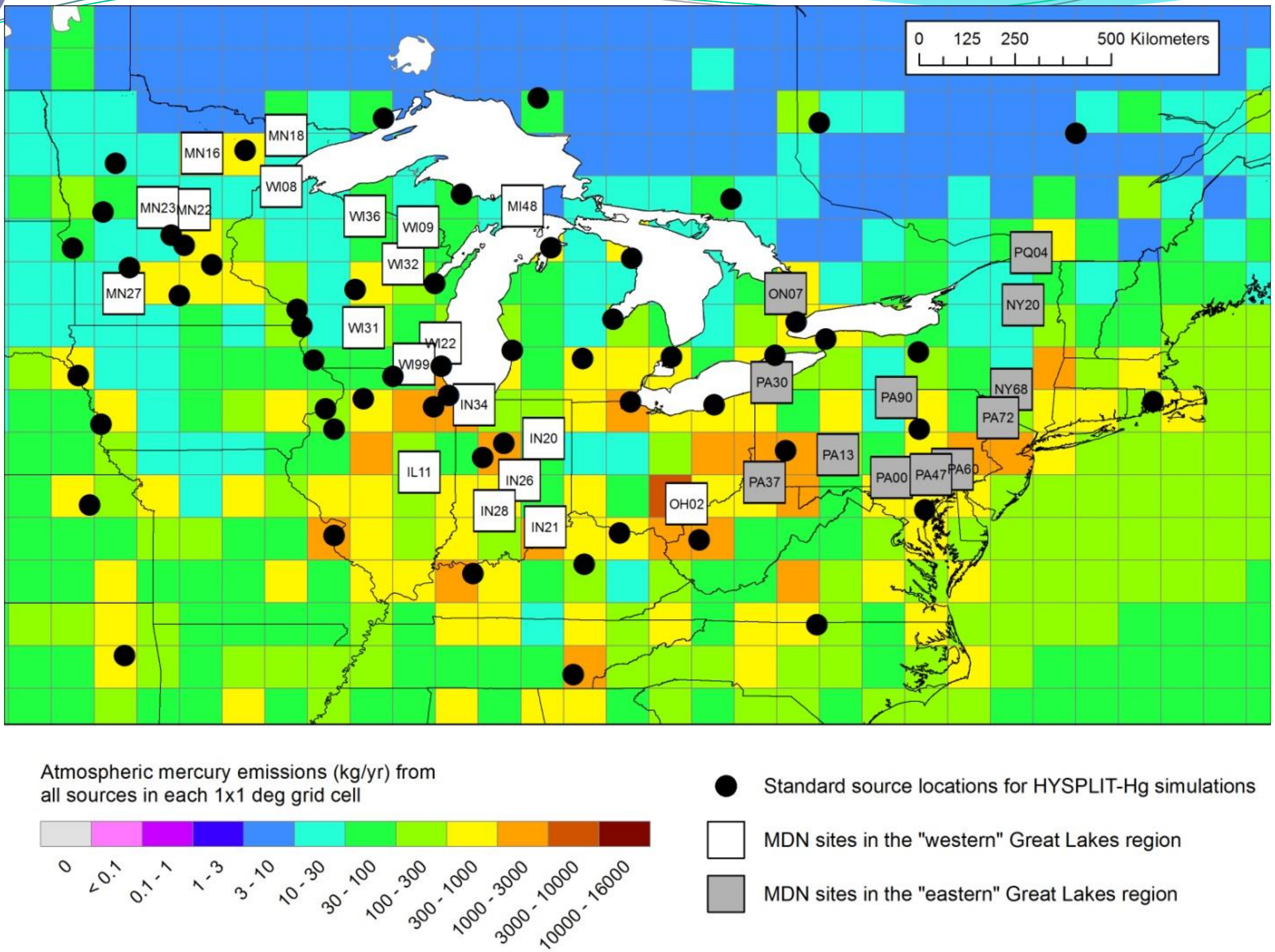
Model Evaluation

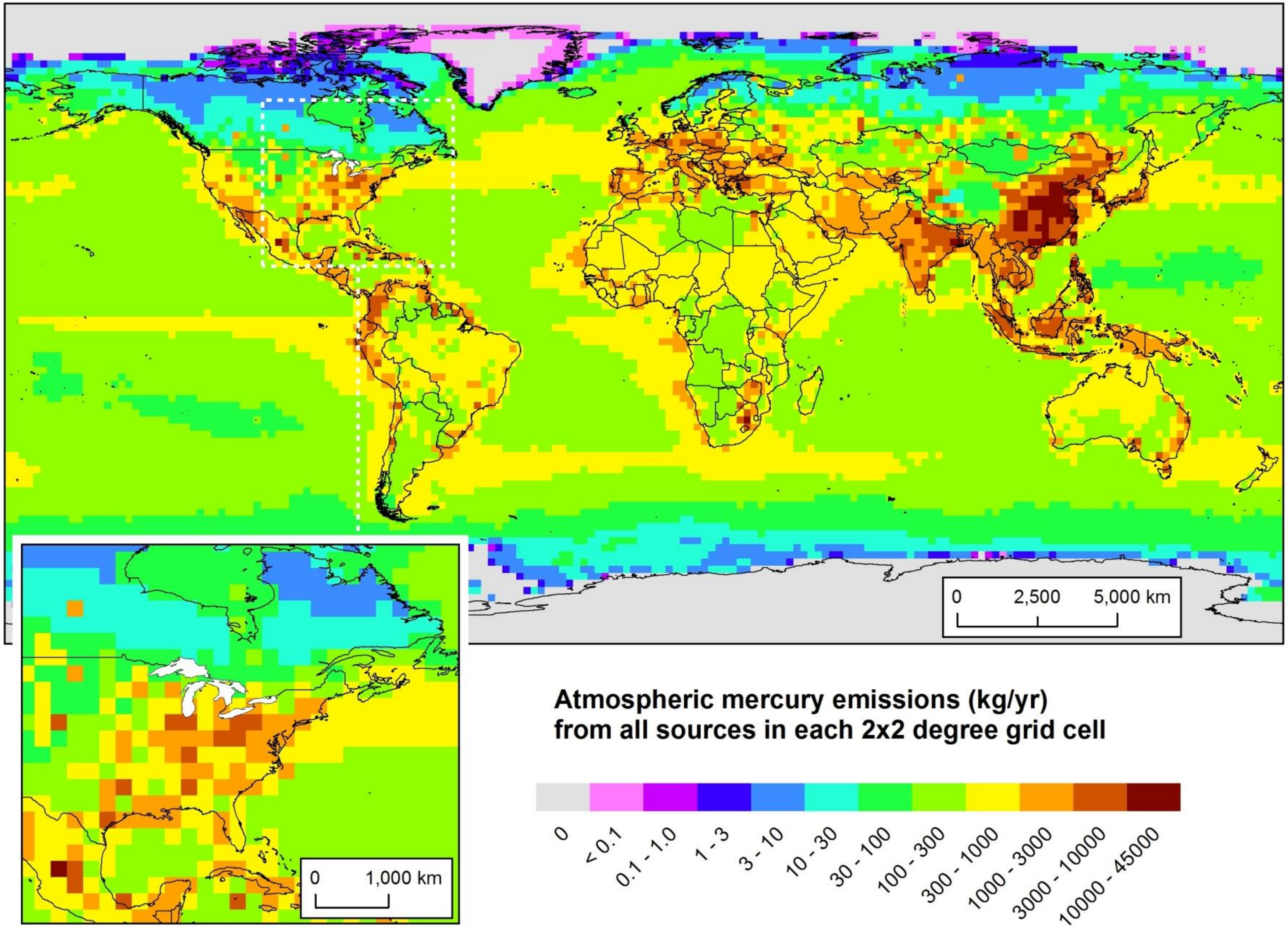
Modeled vs. Measured Wet Deposition of Mercury at Sites in the Great Lakes Region



Standard source locations, MDN sites, and mercury emissions in the Great Lakes region

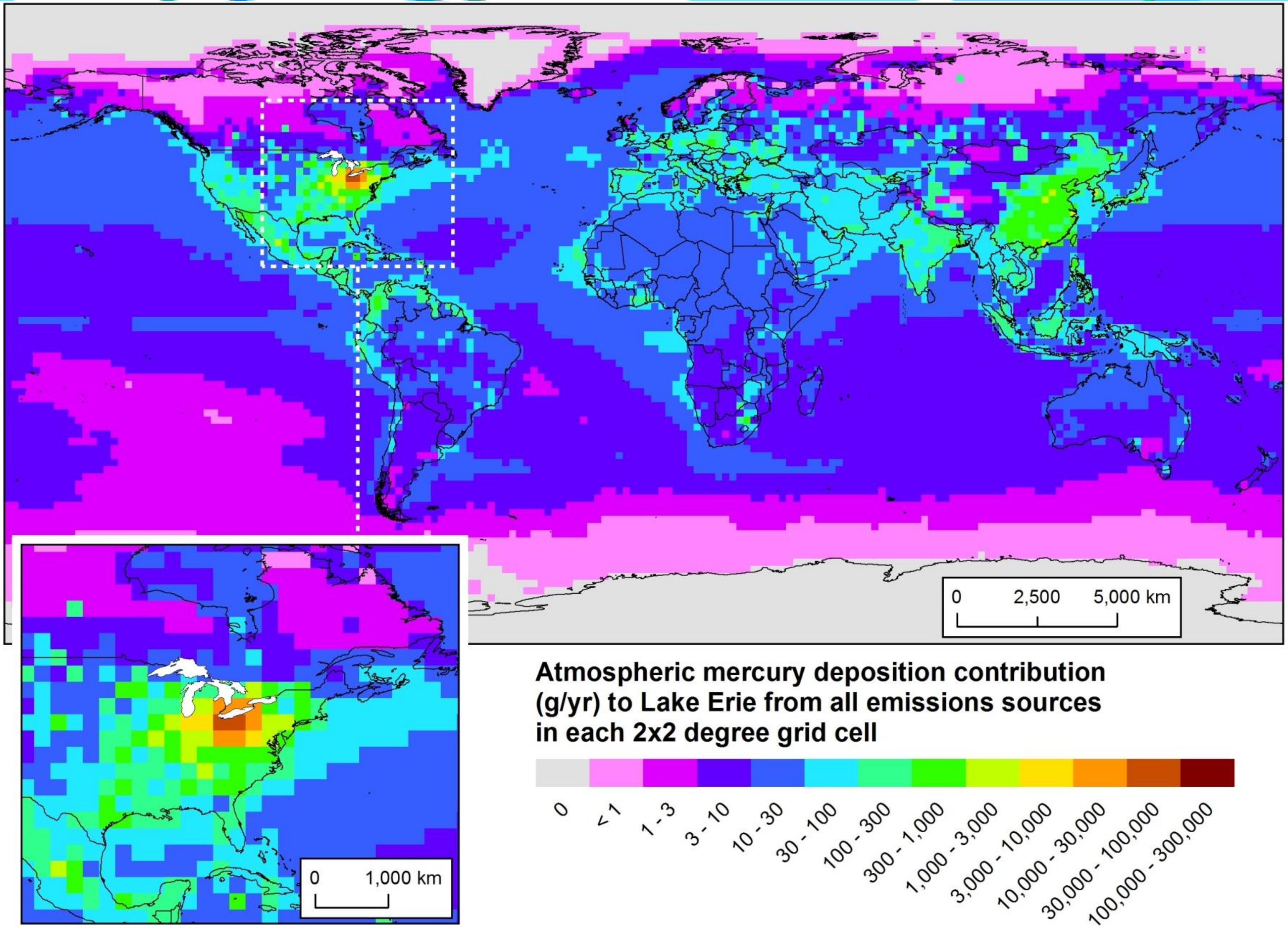


Overall Results for the Great Lakes



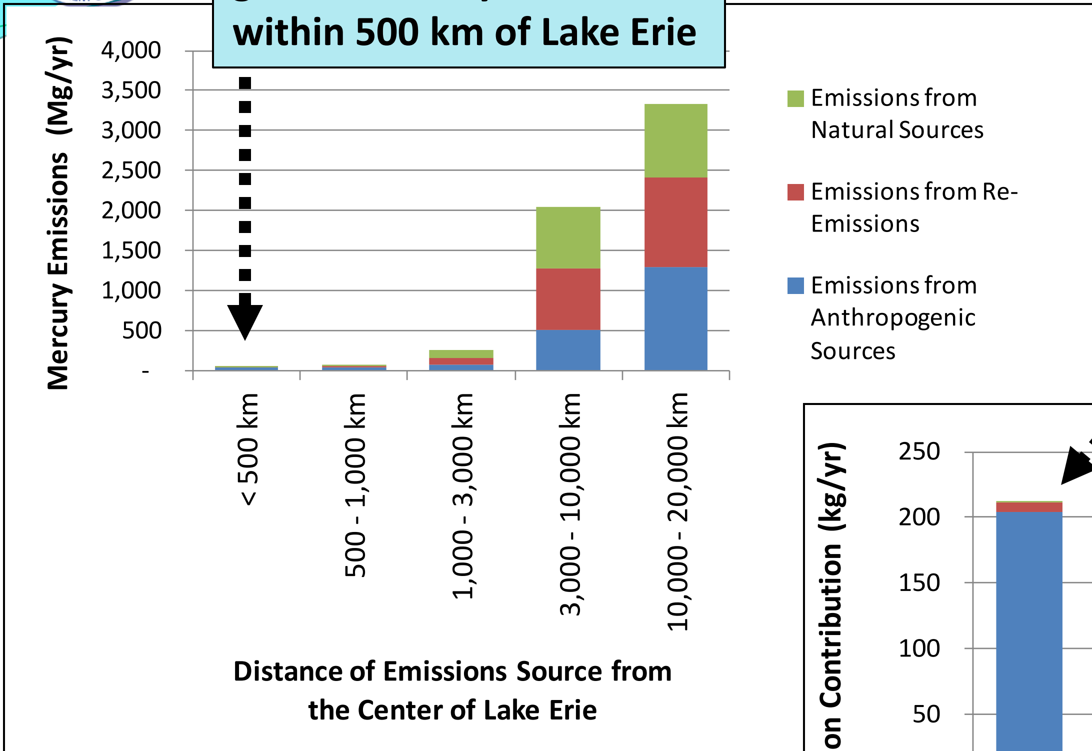
Geographical Distribution of 2005 Atmospheric Mercury Emissions (Natural + Re-emit + Direct Anthropogenic)

Here's where the mercury came from that was deposited to Lake Erie (~2005)

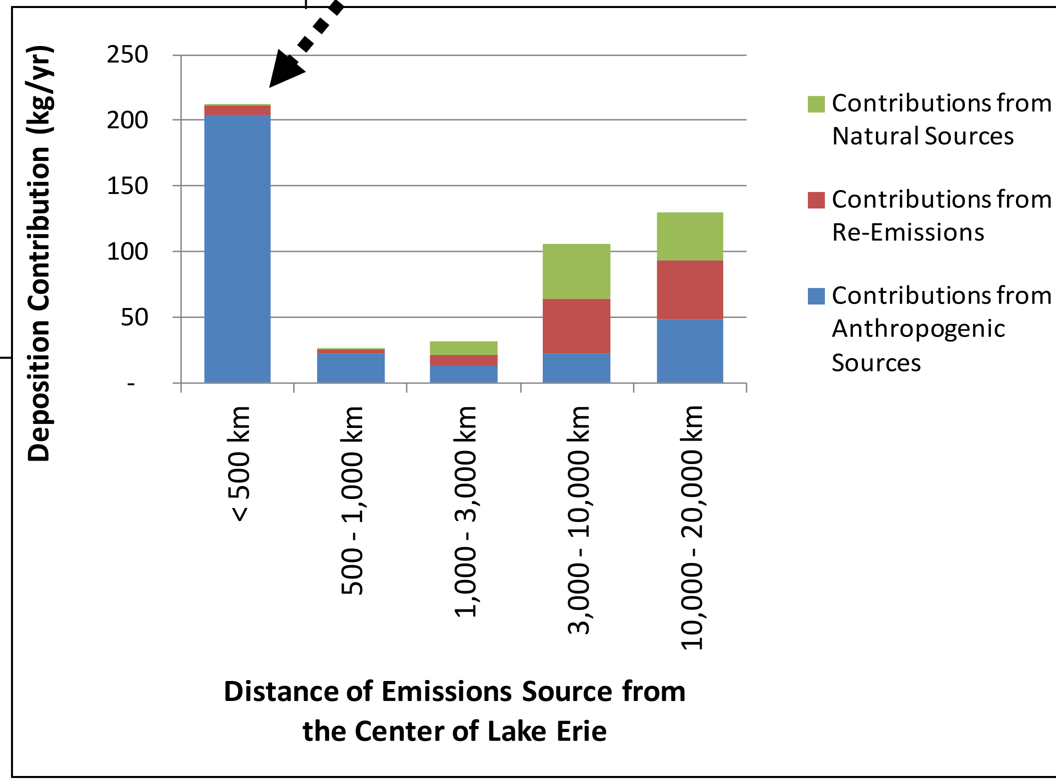


Geographical Distribution of 2005 Atmospheric Mercury Deposition Contributions to Lake Erie

A tiny fraction of 2005 global mercury emissions within 500 km of Lake Erie

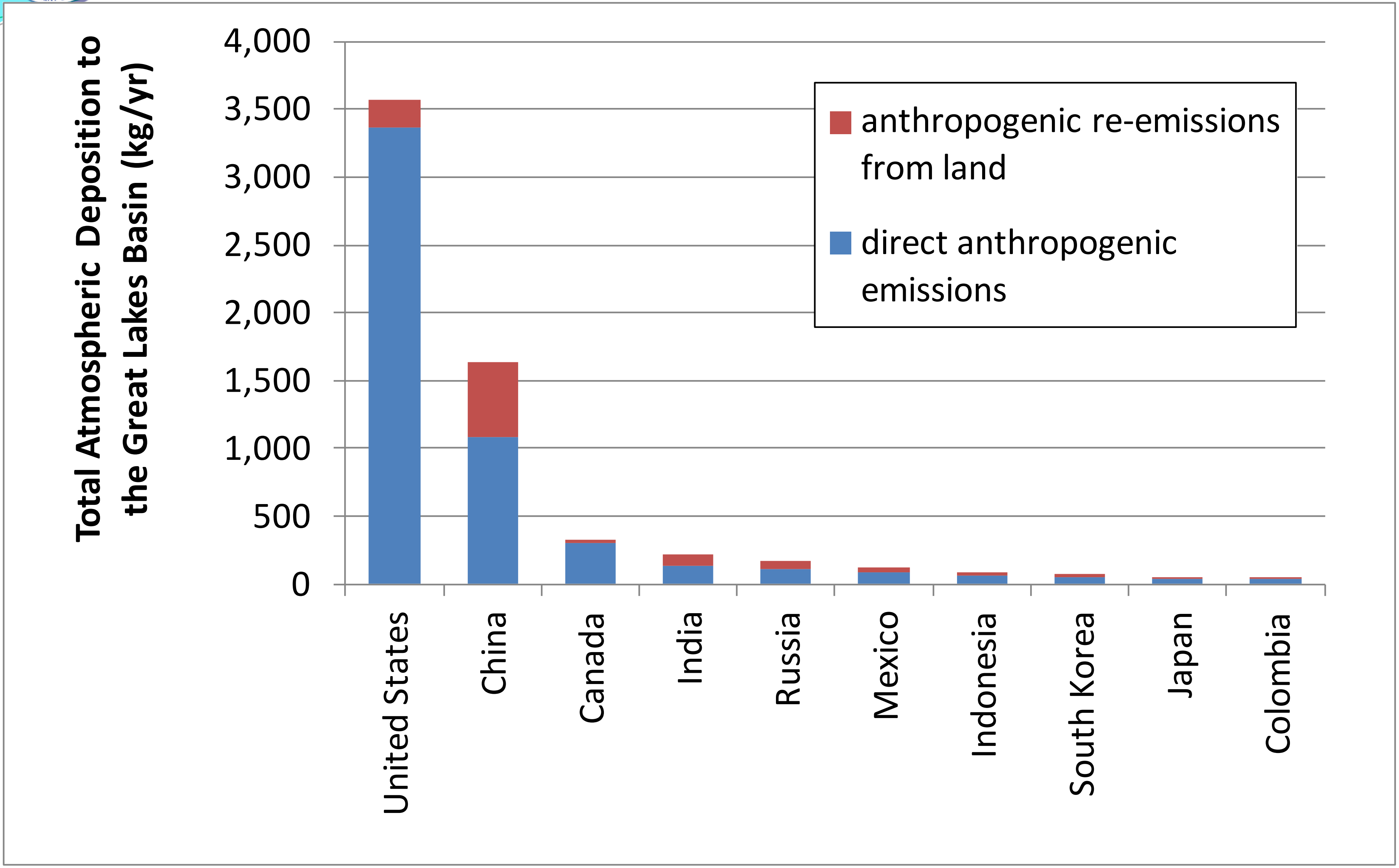


Modeling results show that these “regional” emissions are responsible for a large fraction of the modeled 2005 atmospheric deposition

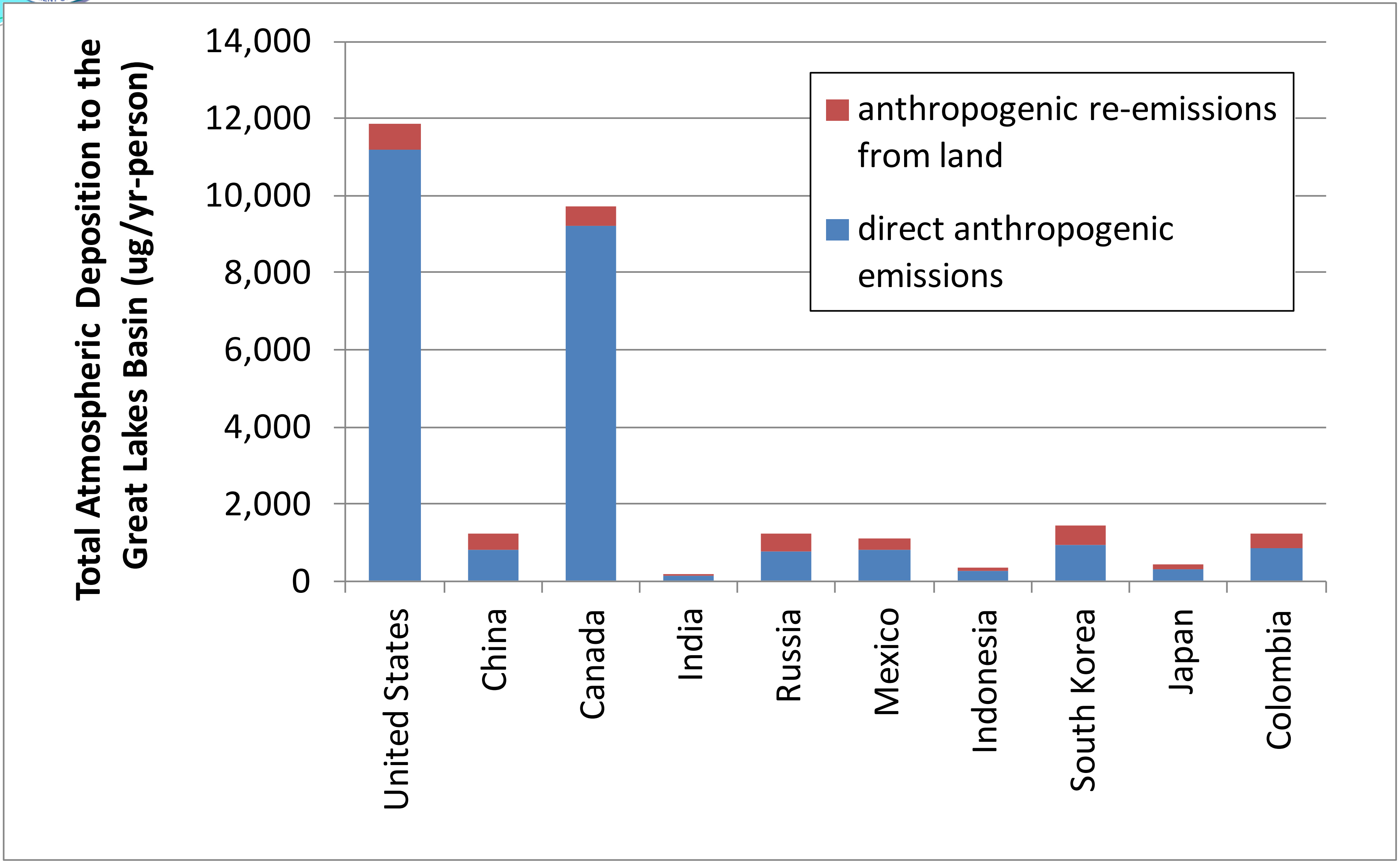


Important policy implications!

Model-estimated 2005 deposition to the Great Lakes Basin from countries with the highest modeled contribution from direct and re-emitted anthropogenic sources

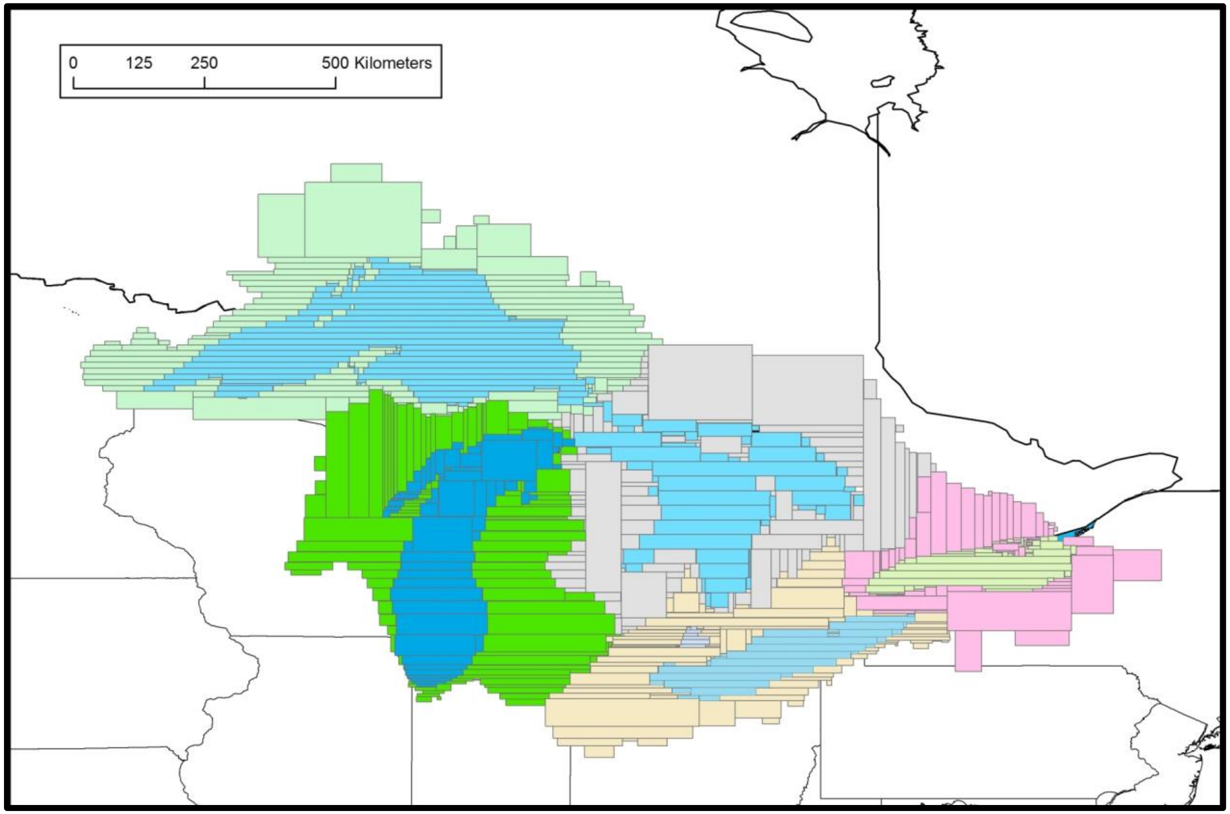


Model-estimated *per capita* 2005 deposition to the Great Lakes Basin from countries with the highest modeled contribution from direct & re-emitted anthropogenic sources



Some Key Features of this Analysis

- **Deposition explicitly modeled to actual lake/watershed areas**
  - As opposed to the usual practice of ascribing portions of gridded deposition to these areas in a post-processing step



Some Key Features of this Analysis

- **Deposition explicitly modeled to actual lake/watershed areas**
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- **Combination of Lagrangian & Eulerian modeling**
  - allows accurate and computationally efficient estimates of the fate and transport of atmospheric mercury over all relevant length scales – from "local" to global.

- **Detailed source-attribution information is created**
  - deposition contribution to each Great Lakes and watersheds from each source in the emissions inventories used is estimated individually
  - The level of source discrimination is only limited by the detail in the emissions inventories
  - Source-type breakdowns not possible in this 1st phase for global sources, because the global emissions inventory available did not have source-type breakdowns for each grid square

Some Key Findings of this Analysis

- **“Single Source” results illustrate source-receptor relationships**
  - For example, a “typical” coal-fired power plant near Lake Erie may contribute on the order of 100x the mercury – for the same emissions – as a comparable facility in China.

- **Regional, national, & global mercury emissions are all important contributors to mercury deposition in the Great Lakes Basin**
  - For Lakes Erie and Ontario, the U.S. contribution is at its most significant
  - For Lakes Huron and Superior, the U.S. contribution is less significant.
  - Local & regional sources have a much greater atmospheric deposition contributions than their emissions, as a fraction of total global mercury emissions, would suggest.

Some Key Findings of this Analysis (…continued)

- **Reasonable agreement with measurements**
  - Despite numerous uncertainties in model input data and other modeling aspects
  - Comparison at sites where significant computational resources were expended – corresponding to regions that were the most important for estimating deposition to the Great Lakes and their watersheds – showed good consistency between model predictions and measured quantities.
  - For a smaller subset of sites generally downwind of the Great Lakes (in regions not expected to contribute most significantly to Great Lakes atmospheric deposition), less computational resources were expended, and the comparison showed moderate, but understandable, discrepancies.

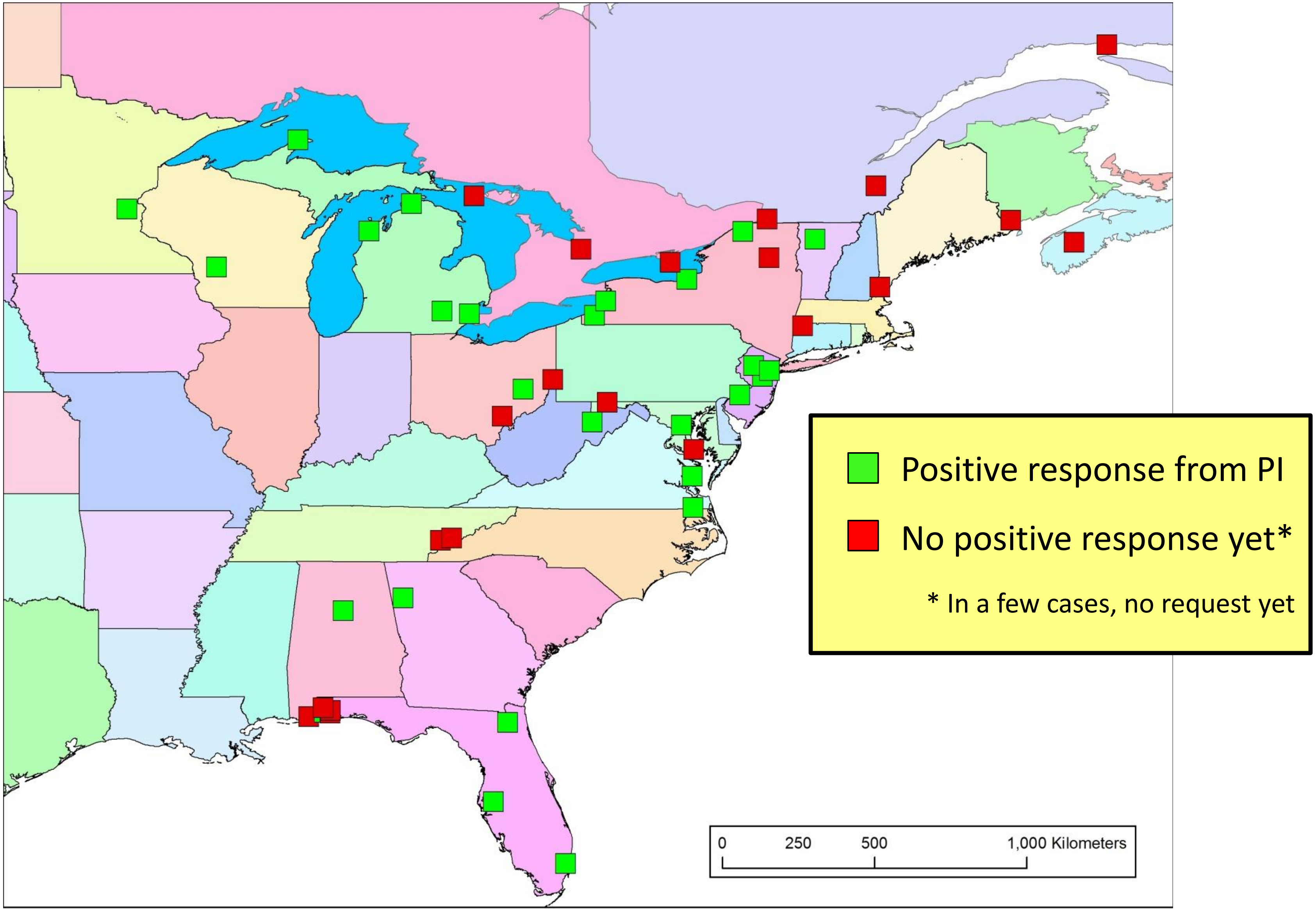
Modeling Atmospheric Mercury Deposition to the Great Lakes. Final Report for work conducted with FY2010 funding from the Great Lakes Restoration Initiative. December 16, 2011. Mark Cohen, Roland Draxler, Richard Artz. NOAA Air Resources Laboratory, Silver Spring, MD, USA. 160 pages.

http://www.arl.noaa.gov/documents/reports/GLRI_FY2010_Atmospheric_Mercury_Final_Report_2011_Dec_16.pdf

http://www.arl.noaa.gov/documents/reports/Figures_Tables_GLRI_NOAA_Atmos_Mercury_Report_Dec_16_2011.pptx

Phase 2: Sensitivity Analysis And Extended Model Evaluation

Sites That May Have Ambient Mercury Concentration Measurement Data for 2005 for Model Evaluation



Sensitivity Analysis (in progress)

- **Meteorological Data**
  - NARR instead of EDAS40km for North America

- **Standard Source Locations and Interpolation Methodologies**
  - Adding pts in Eastern Great Lakes region to see if results improve
  - Numerical experiments with different interpolation methods
  - Numerical experiments with subsets of std pts – do we need them all?

- **Model Parameters**
  - Simulation (time step, release elevation …)
  - Dispersion (number of puffs, freq. of splitting, “conage”, …)
  - Deposition (wet and dry deposition algorithms, …)
  - Phase partitioning (gas-droplet, gas-particle, …)
  - Chemistry (reaction rates, reactant concentrations, …)

Sensitivity Analysis (in progress)

- **Investigating Impact of Variations on:**
  - model evaluation results?
  - model-estimated deposition to the GL?
  - Model-estimated source attribution

- **Tactical Considerations**
  - Cannot do “Full” Phase-1 analysis for more than a few cases
  - Screening on a small subset of source locations
  - “Full Analysis” may be possible on reduced subset of std pts

Phase 3: Scenarios

What Scenarios Should be Used?

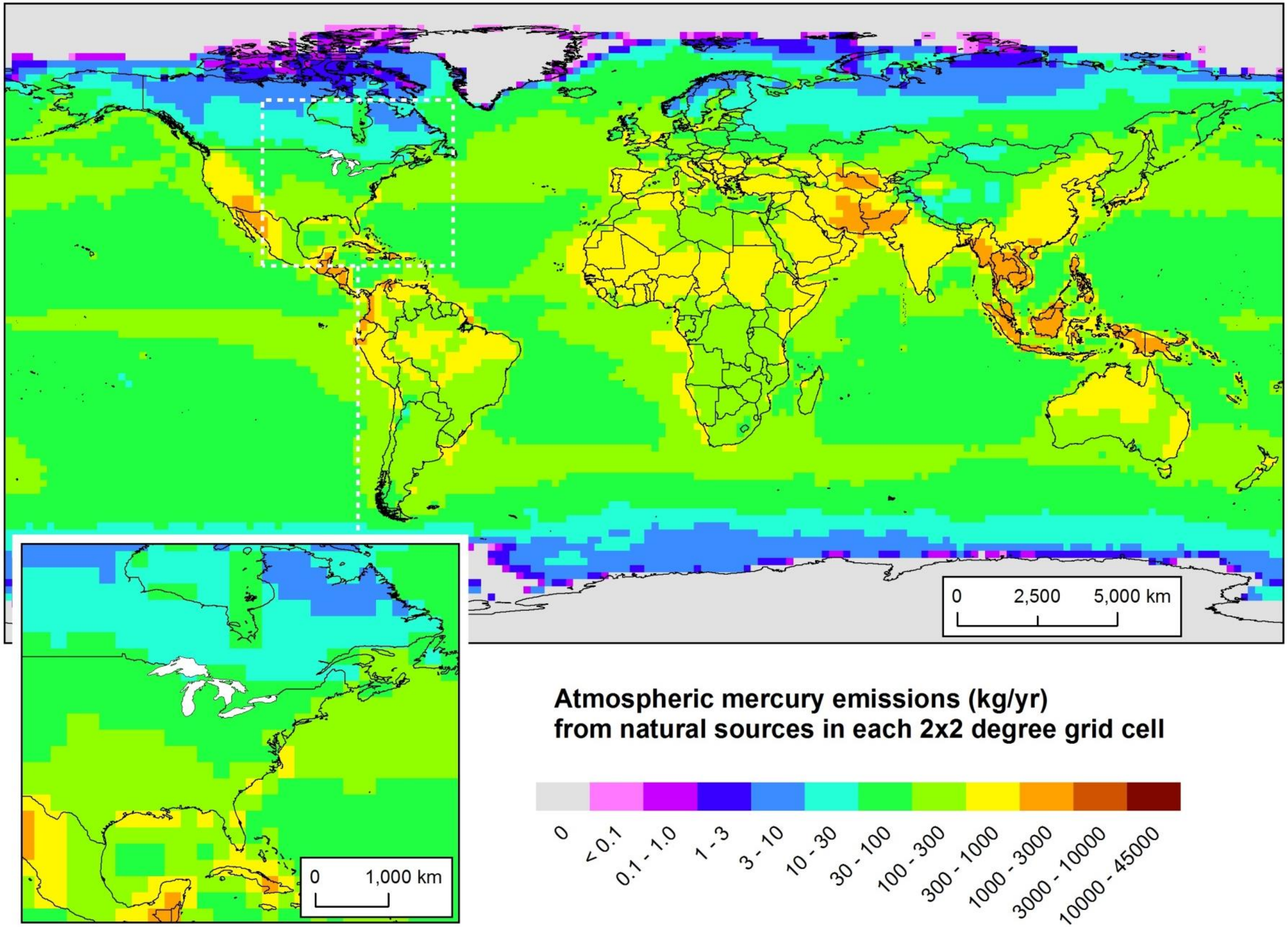
- **How to generate / solicit scenarios?**
- **Significant role for IAQAB?**


Thanks!
Questions? Suggestions?

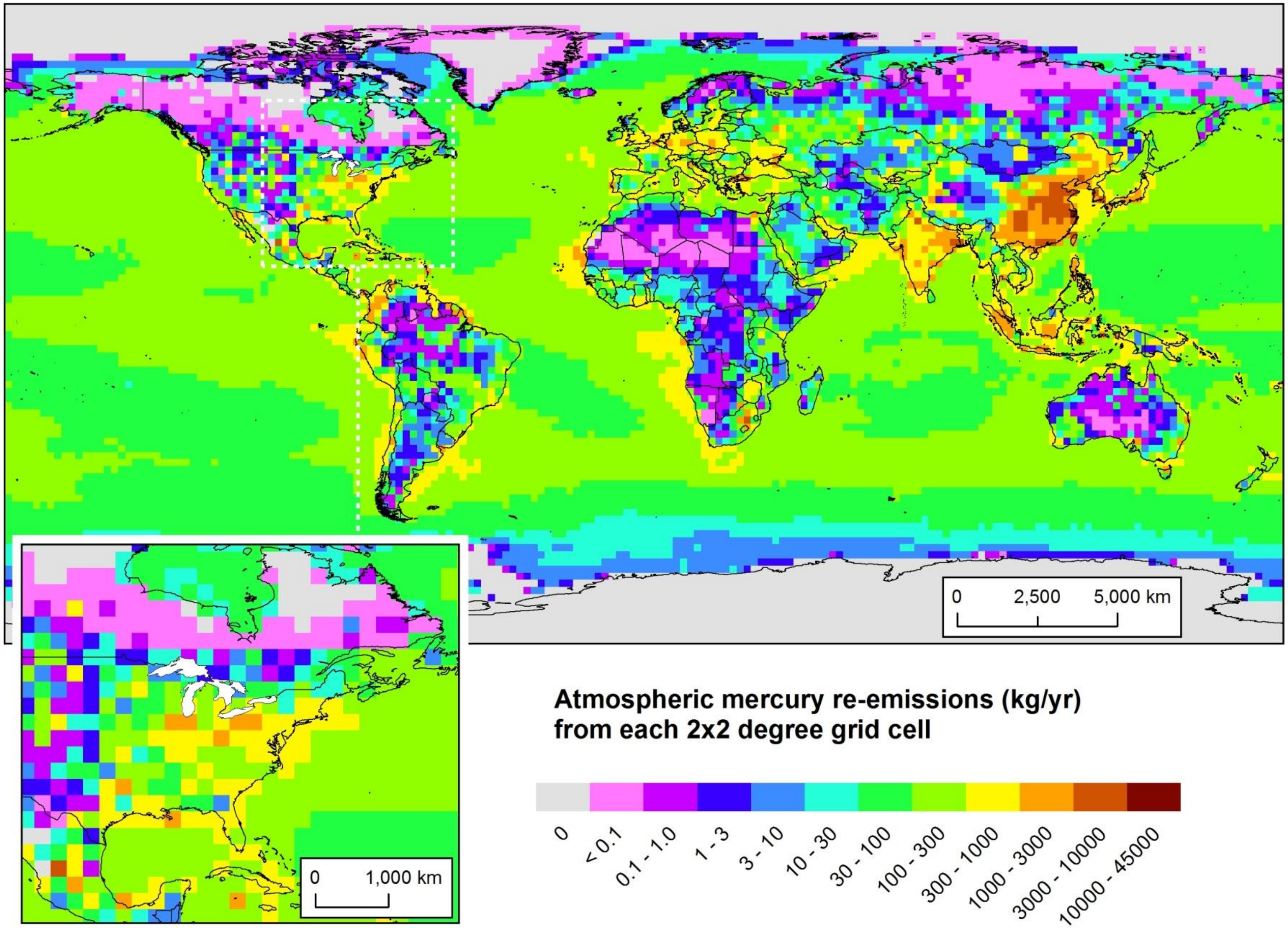
Extra Slides

Emissions

Natural mercury emissions

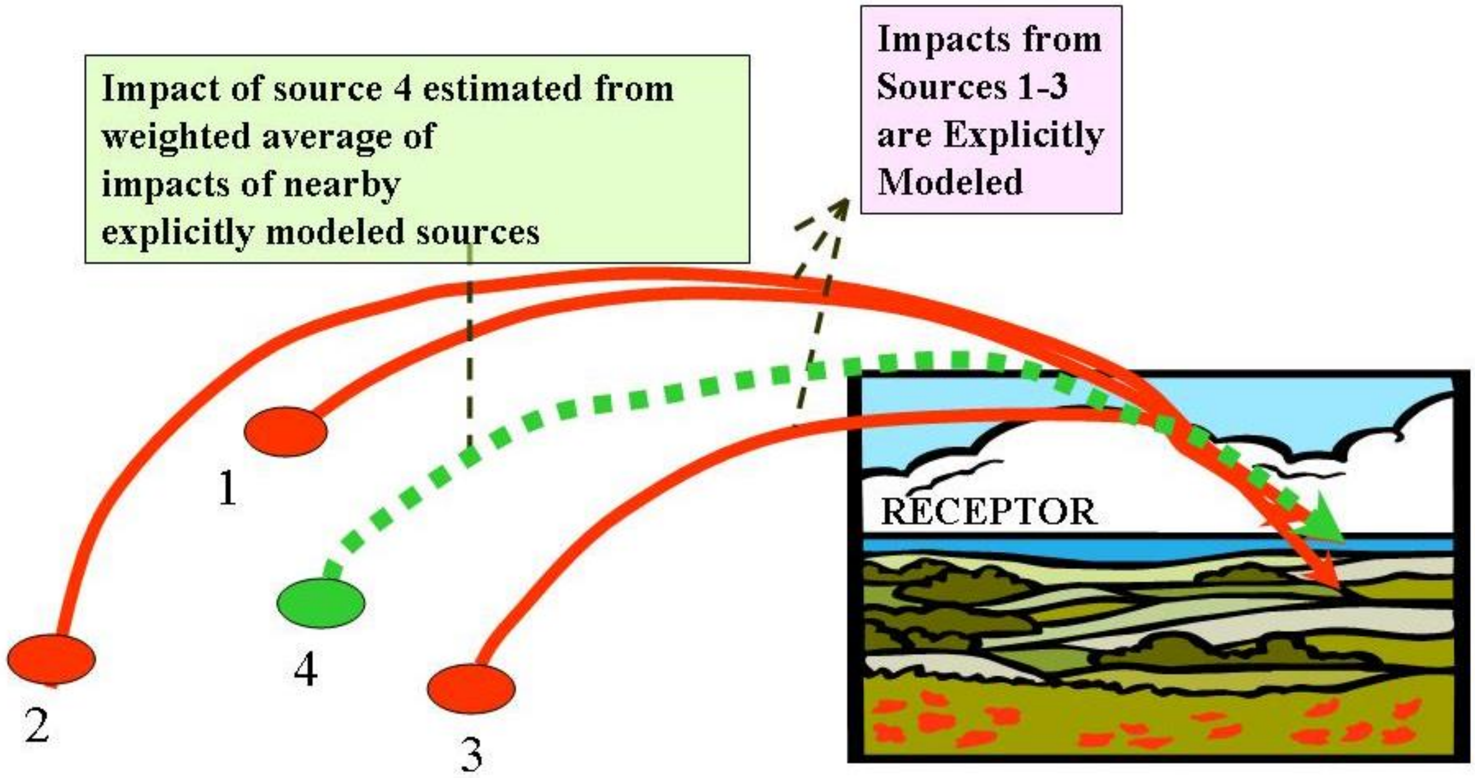


Atmospheric re-emissions of previously deposited mercury from anthropogenic sources

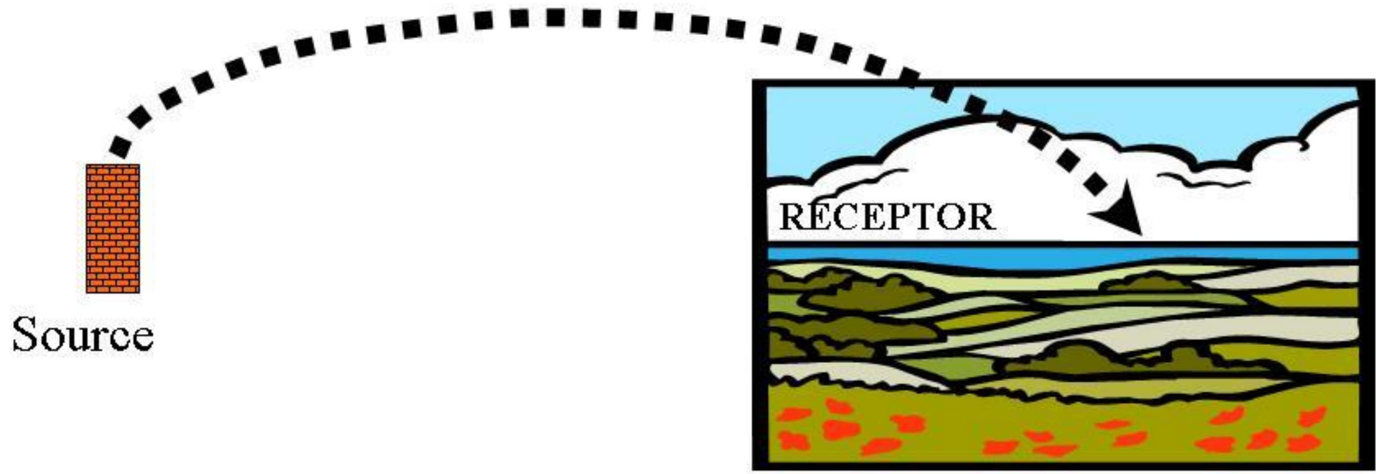


Methodology

Spatial Interpolation



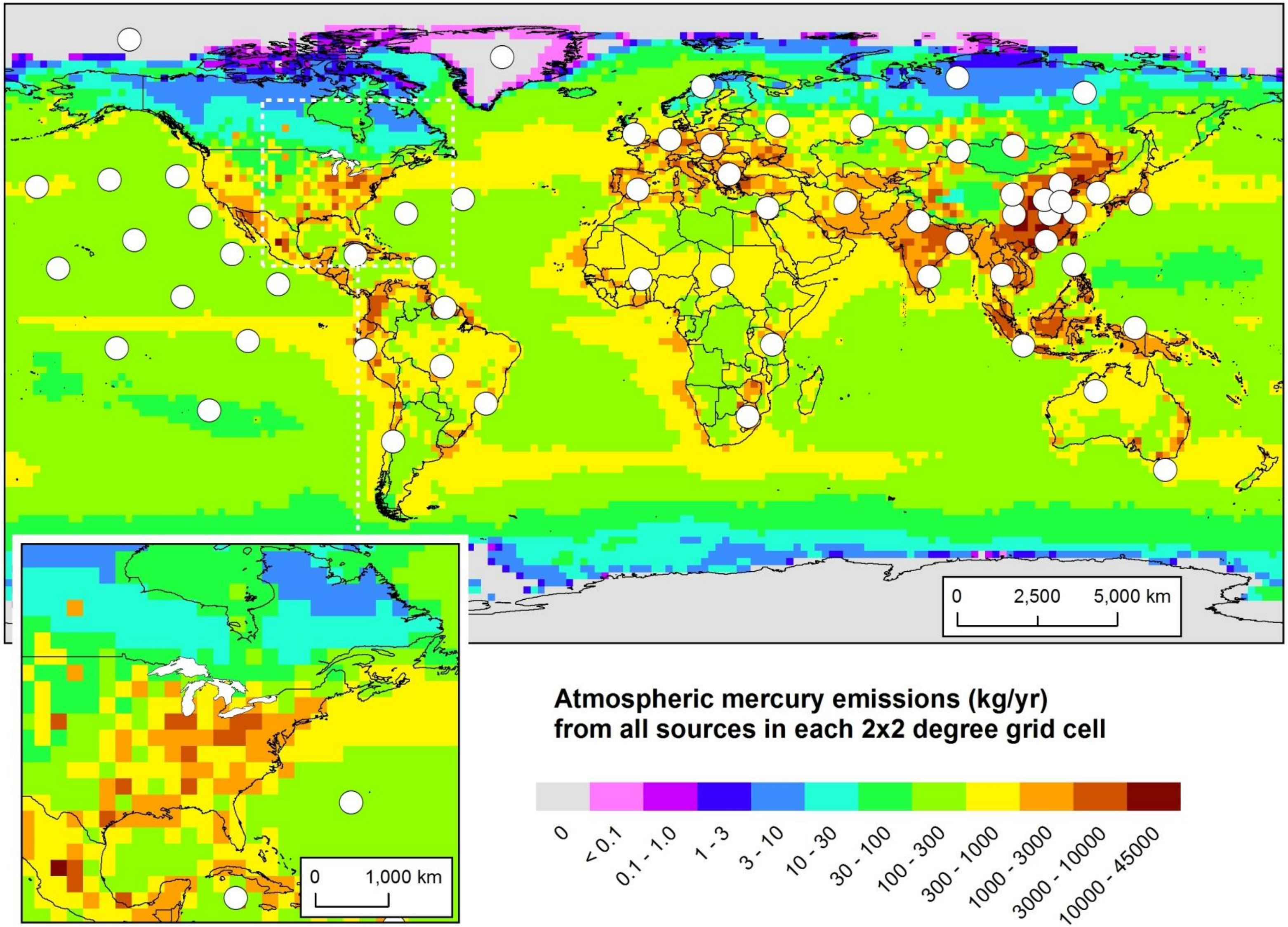
Chemical Interpolation



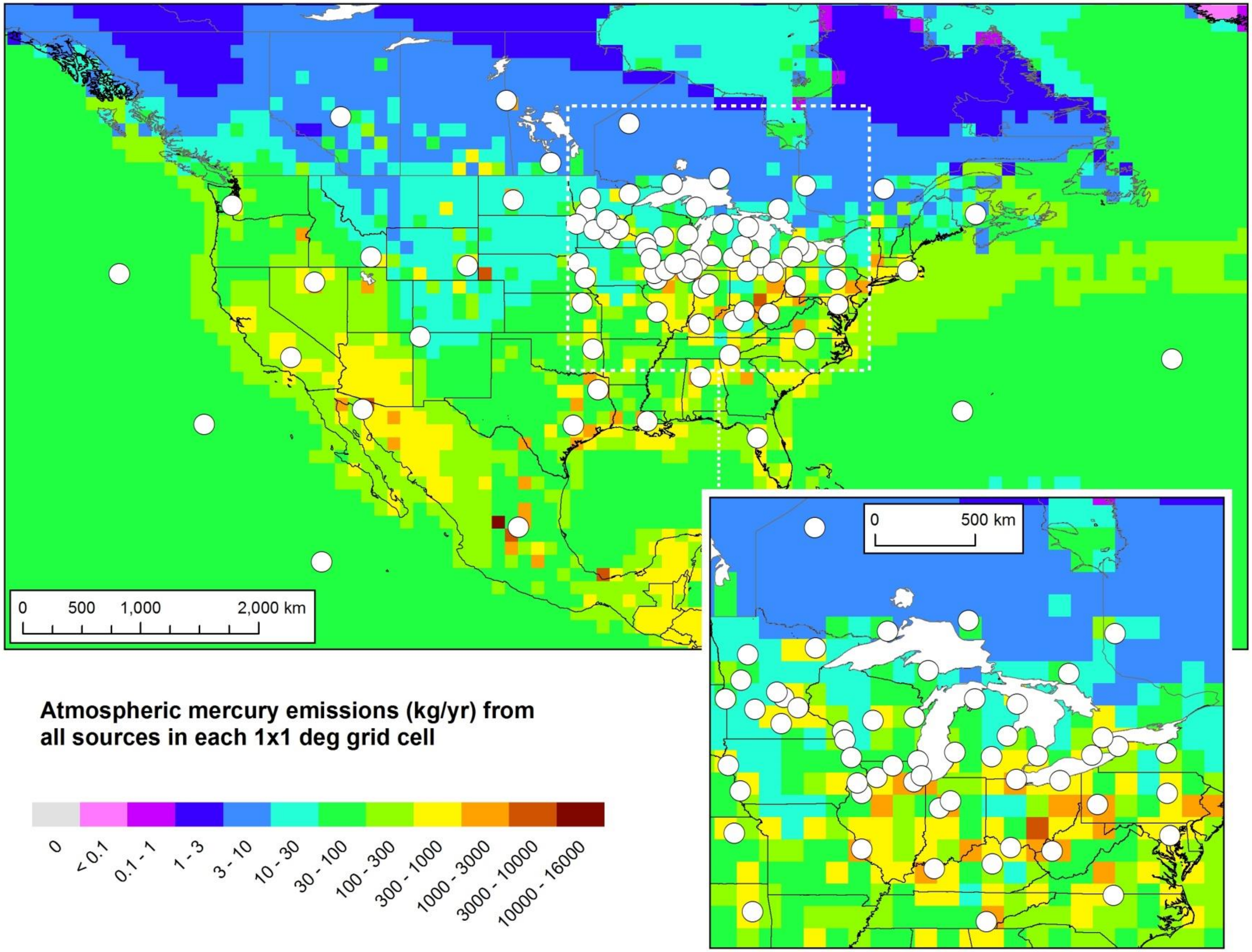
Impact of Source Emitting 30% Hg(0) 50% Hg(II) 20% Hg(p)	=	0.3 x Impact of Source Emitting Pure Hg(0) + 0.5 x Impact of Source Emitting Pure Hg(II) + 0.2 x Impact of Source Emitting Pure Hg(p)

Impact of Source Emitting 30% Hg(0), 50% Hg(II), 20% Hg(p) = 0.3 x [Impact of Source Emitting Pure Hg(0)] + 0.5 x [Impact of Source Emitting Pure Hg(II)] + 0.2 x [Impact of Source Emitting Pure Hg(p)]

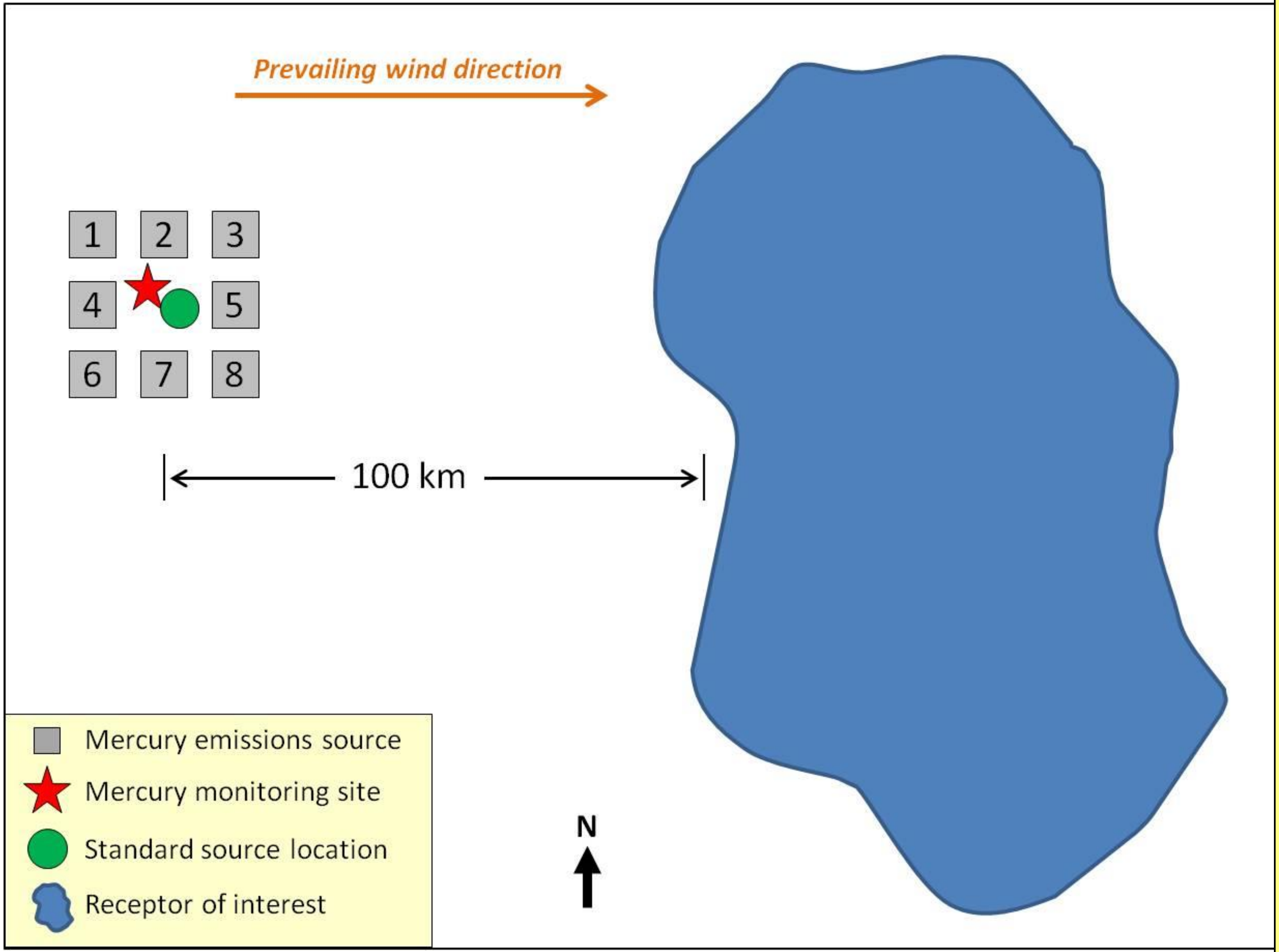
Standard Points Outside of North America



Standard Points in North America



Getting good ground-truthing results harder than estimating deposition to the Great Lakes



One Standard Source Location (green dot) would do a decent job of estimating deposition to the receptor, for all of the hypothetical, "actual" source locations shown (numbered boxes)

But the same Standard Source Location would be completely inadequate to estimate deposition and concentrations at the monitoring site (red star)

Illustrative Results for Single Sources

In order to conveniently compare different model results, a "transfer flux coefficient" X will be used, defined as the following:

$$X = \frac{deposition\ flux\ rate}{emissions\ rate} = \frac{\frac{grams\ Hg\ deposited\ per\ year}{km^2\ of\ receptor\ area}}{grams\ Hg\ emitted\ per\ year\ from\ the\ source}\ [=]\ \frac{1}{km^2}$$

$$deposition\ flux\ rate = \frac{grams\ Hg\ deposited\ per\ year}{km^2\ of\ receptor\ area}\ \left(\frac{g}{km^2 yr}\right)$$

$$= transfer\ flux\ coefficient\ \left(\frac{1}{km^2}\right) * source\ mercury\ emissions\ \left(\frac{g}{yr}\right)$$

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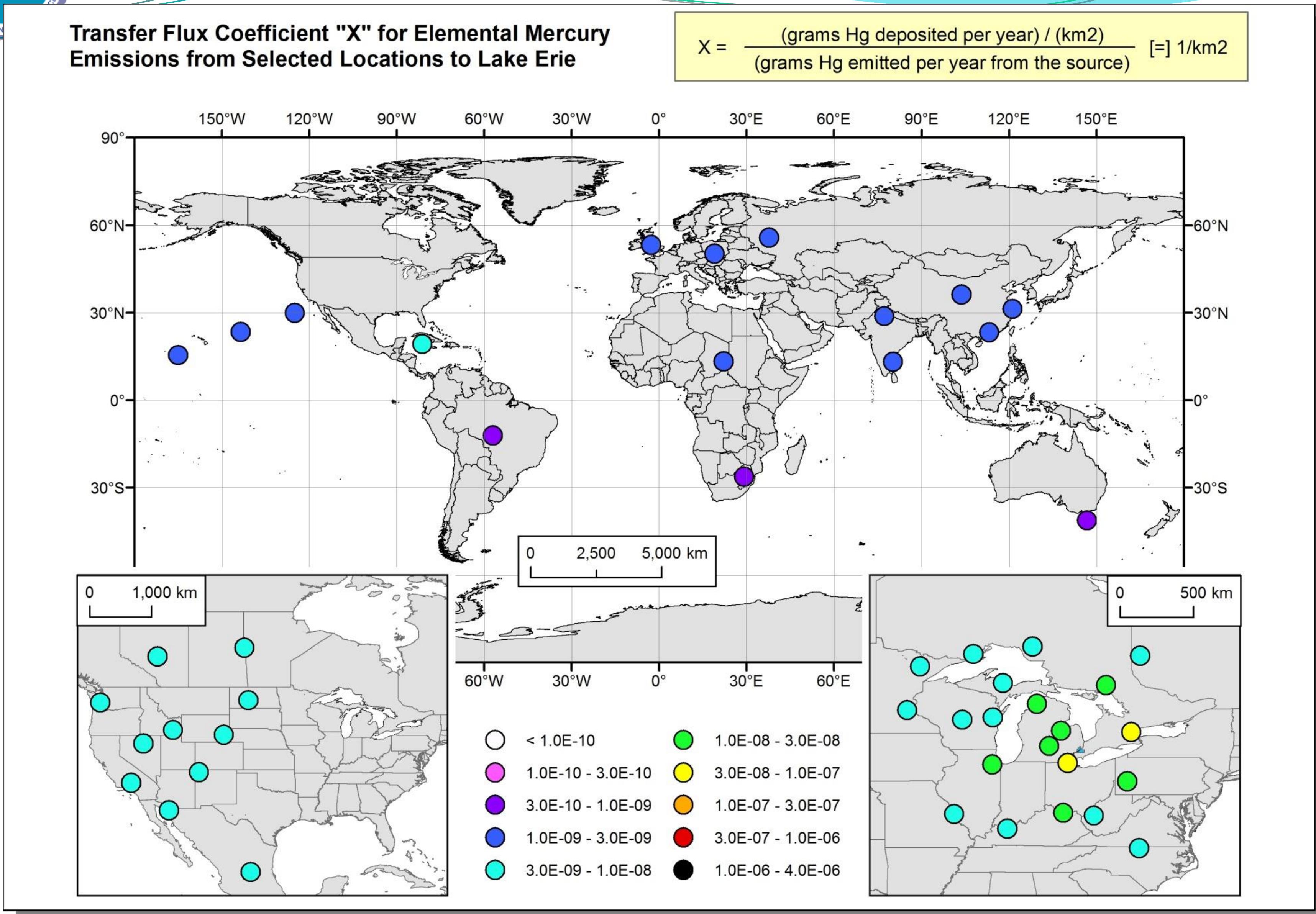
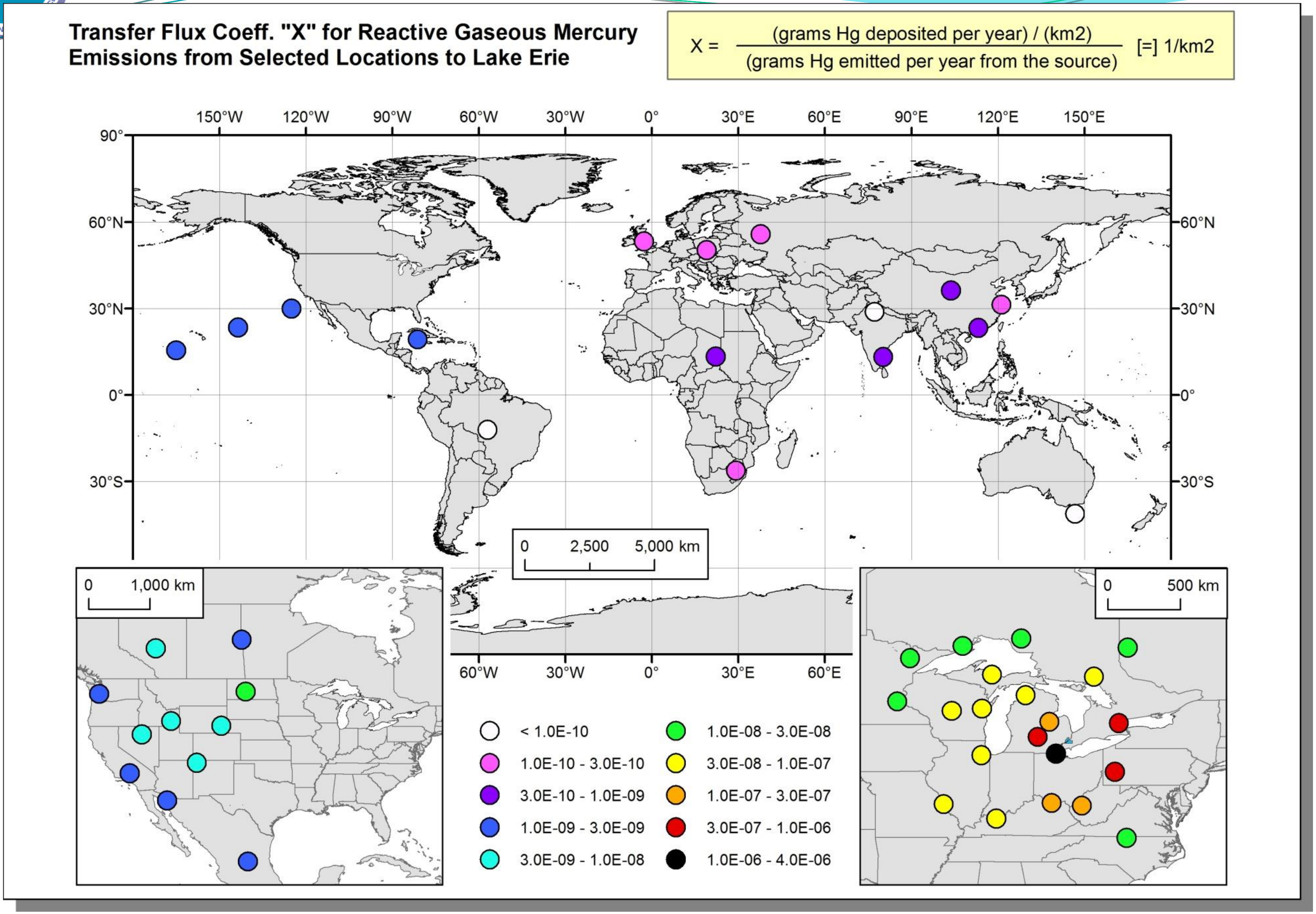
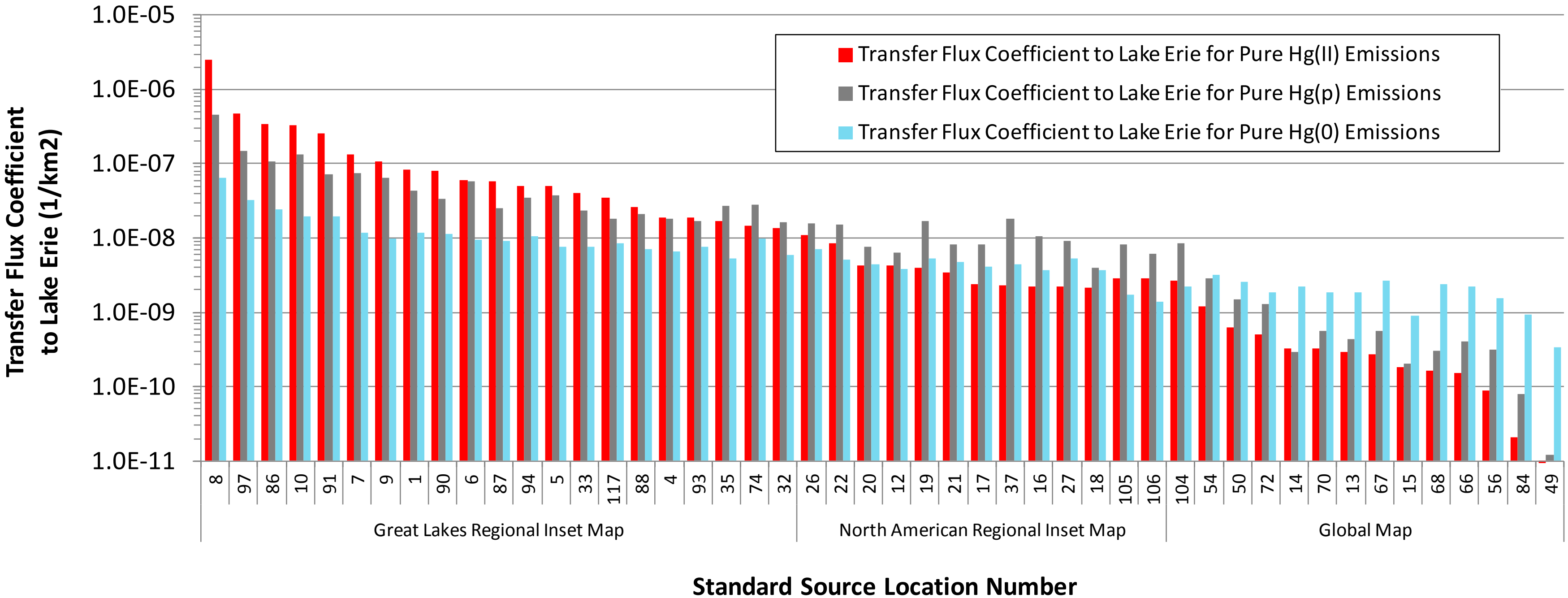
Transfer Flux Coefficients For Pure Elemental Mercury Emissions at an Illustrative Subset of Standard Source Locations, for Deposition Flux Contributions to Lake Erie



Transfer Flux Coefficients For Pure Reactive Gaseous Mercury Emissions at an Illustrative Subset of Standard Source Locations, for Deposition Flux Contributions to Lake Erie



Transfer Flux Coefficients For Hg(0), Hg(II), and Hg(p) to Lake Erie (logarithmic scale)



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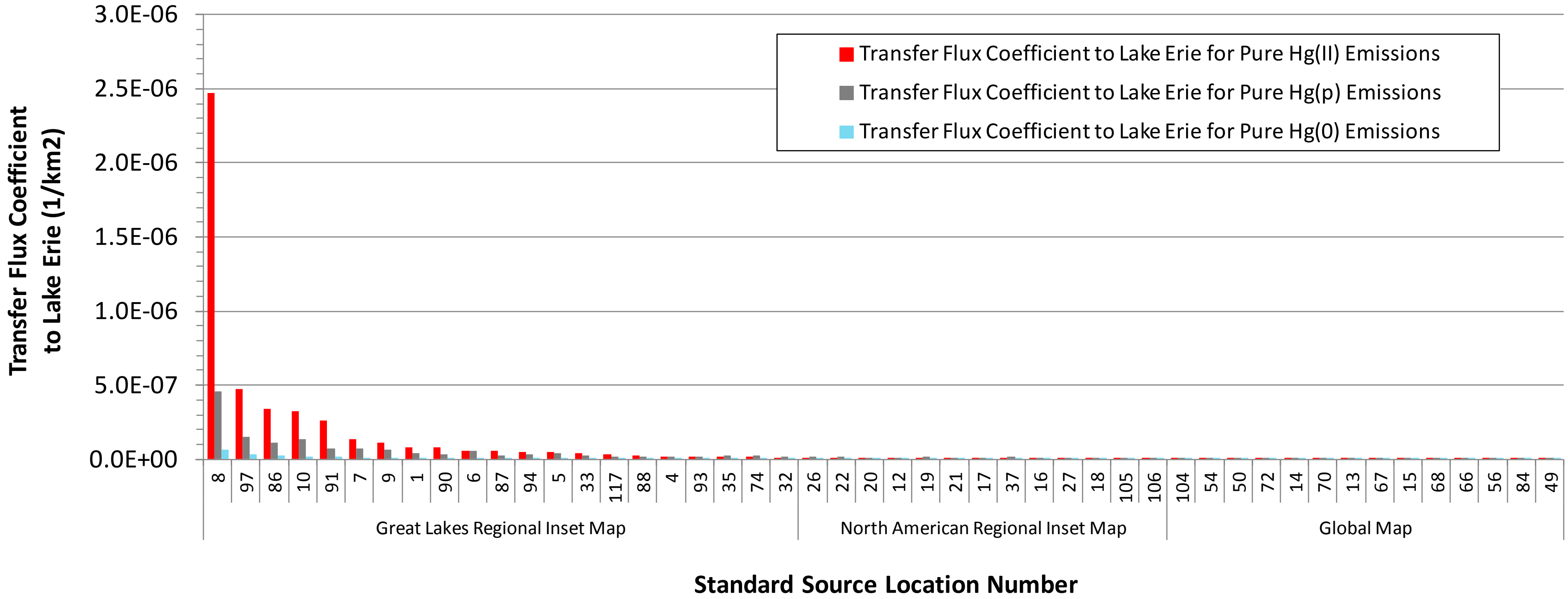
Transfer Flux Coefficients For Hg(0), Hg(II), and Hg(p) to Lake Erie (linear scale)



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Model Evaluation

Figure 55. Mercury Deposition Network Sites in the Great Lakes Region Considered in an Initial Model Evaluation Analysis

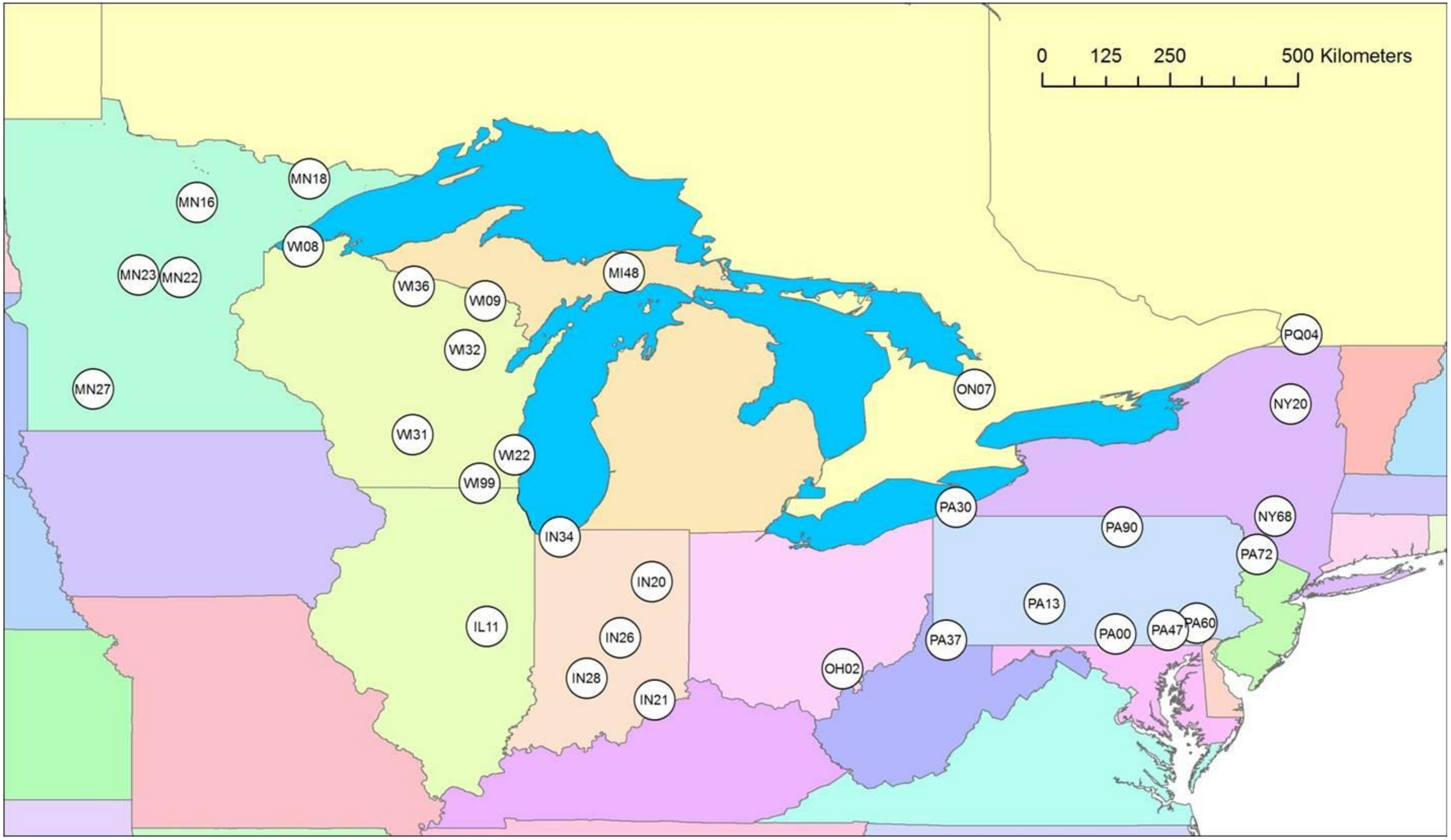
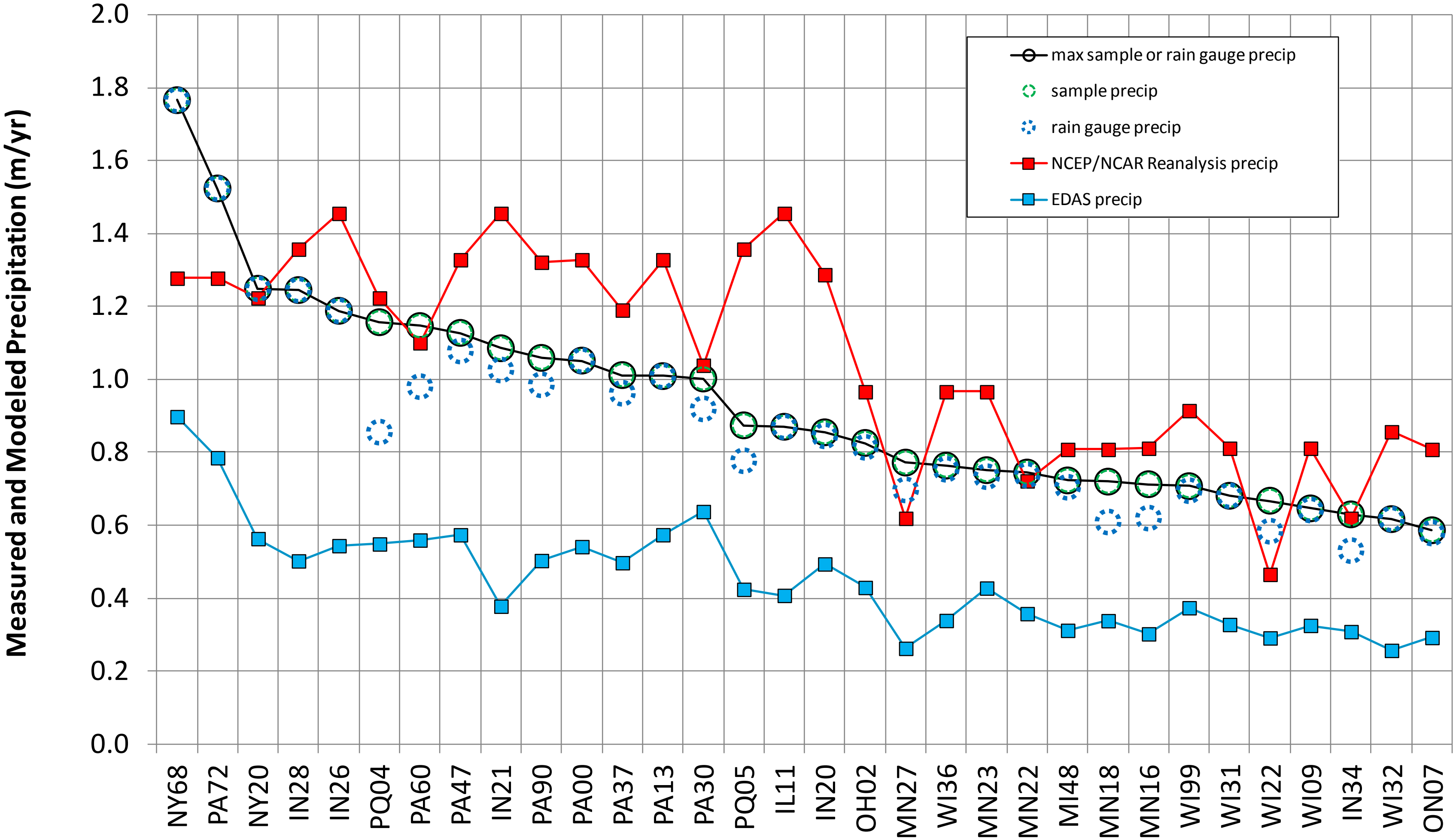
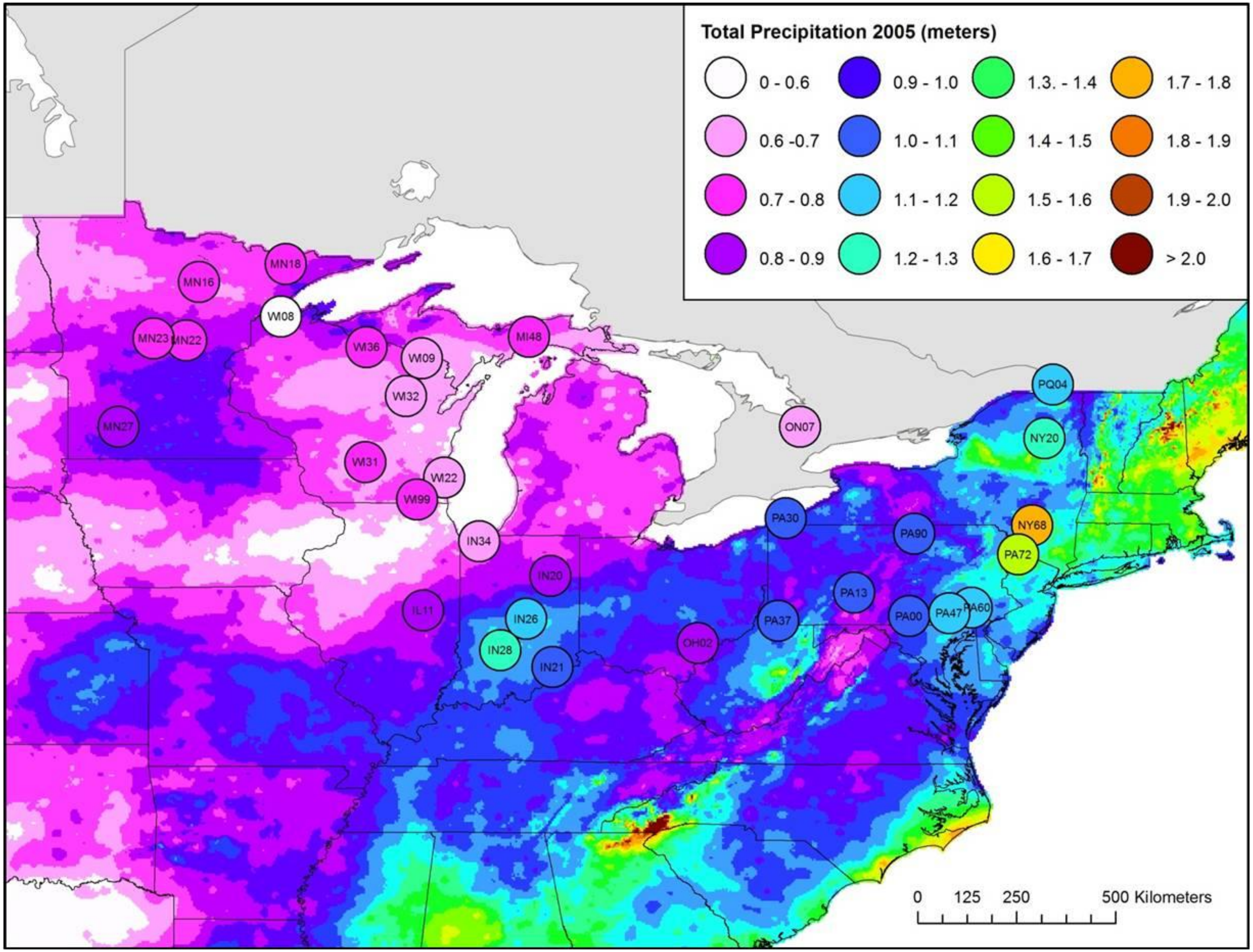


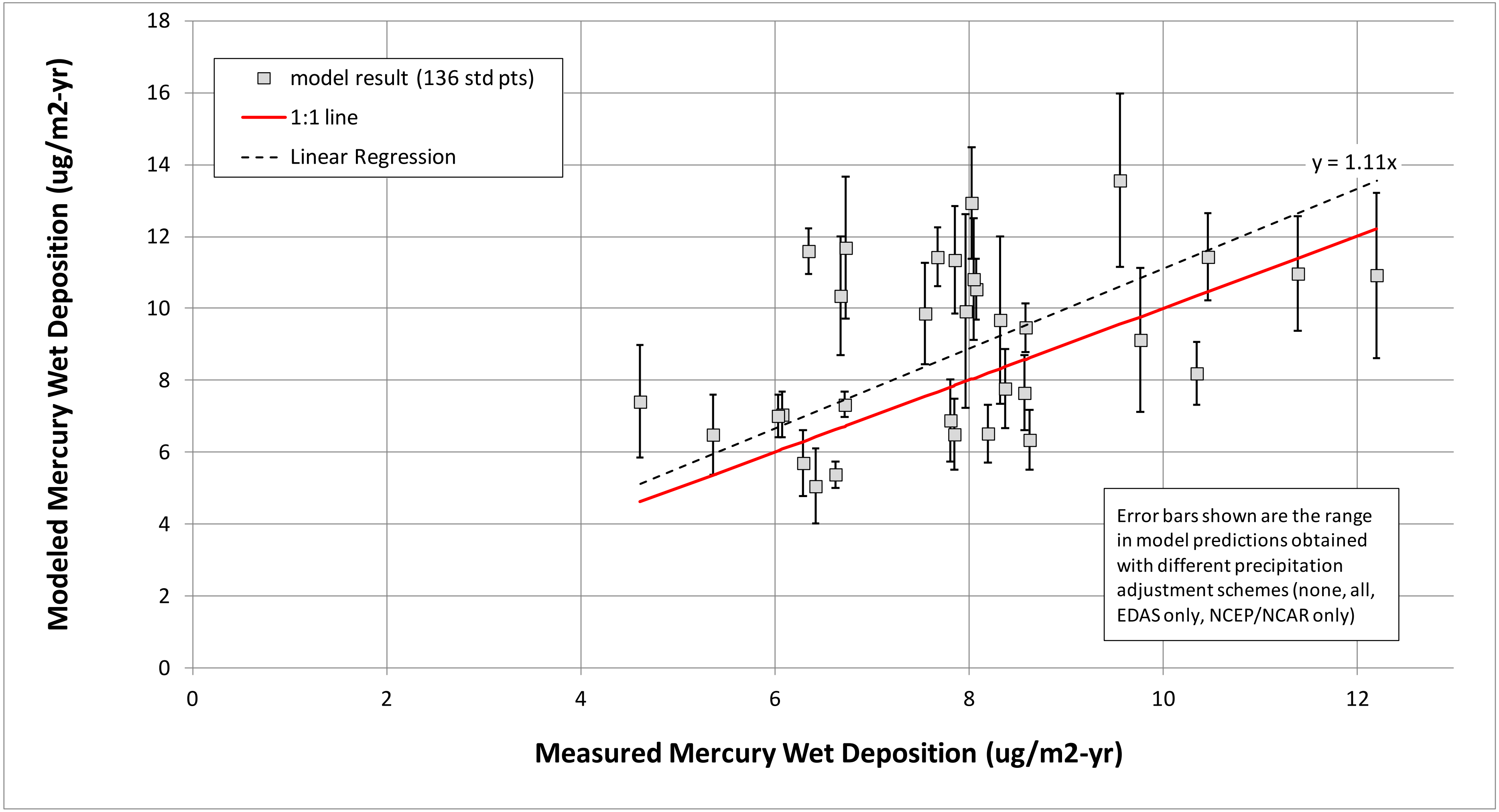
Figure 56. Comparison of Total 2005 Precipitation Measured at each of the Great-Lakes Region MDN Sites with the Precipitation in the Meteorological Datasets Used as Inputs to this Modeling Study



Comparison of 2005 precipitation total as measured at MDN sites in the Great Lakes region (circles) with precipitation totals assembled by the PRISM Climate Group, Oregon State University



Modeled vs. Measured Wet Deposition of Mercury at Sites in the Great Lakes Region



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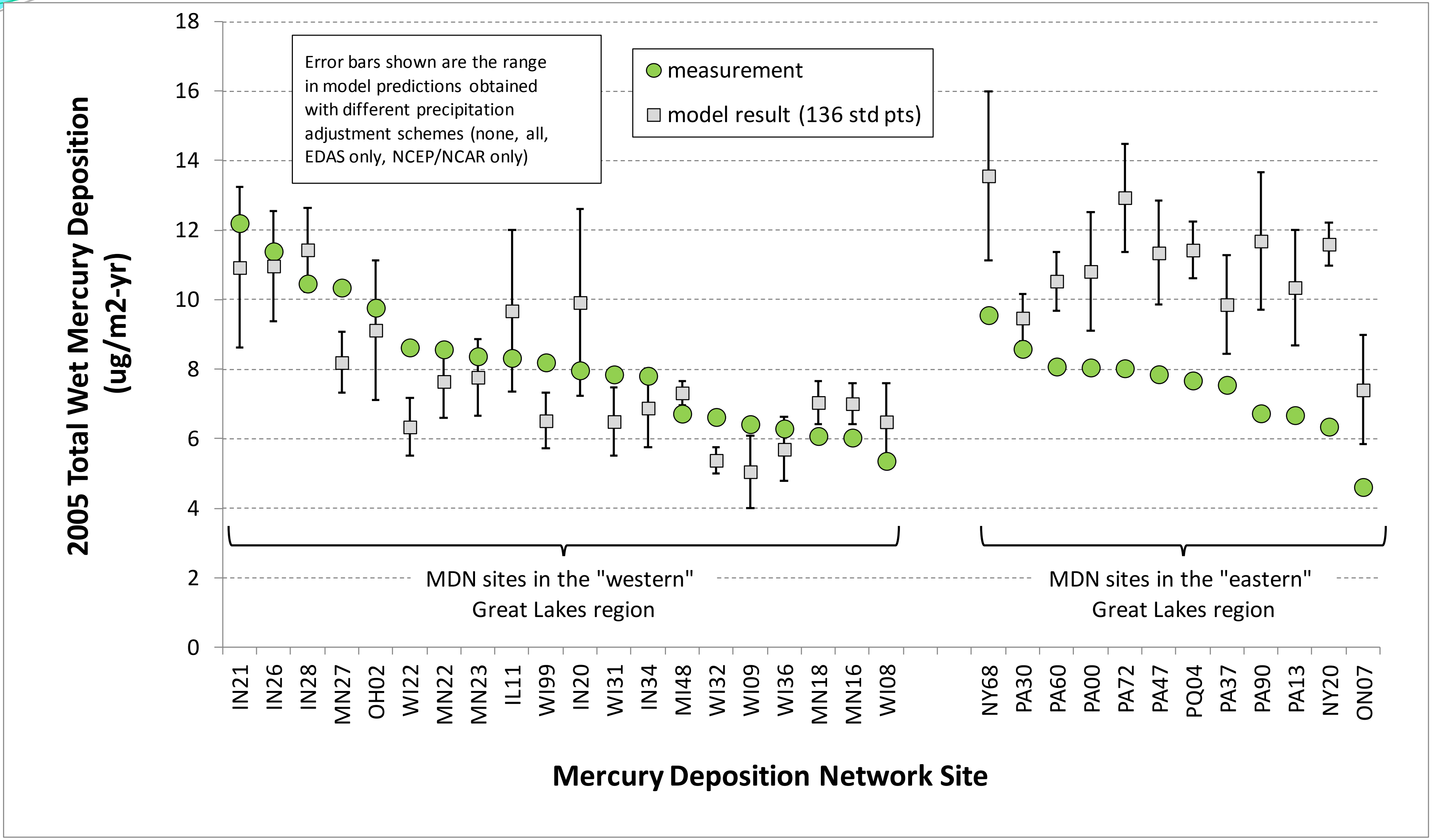
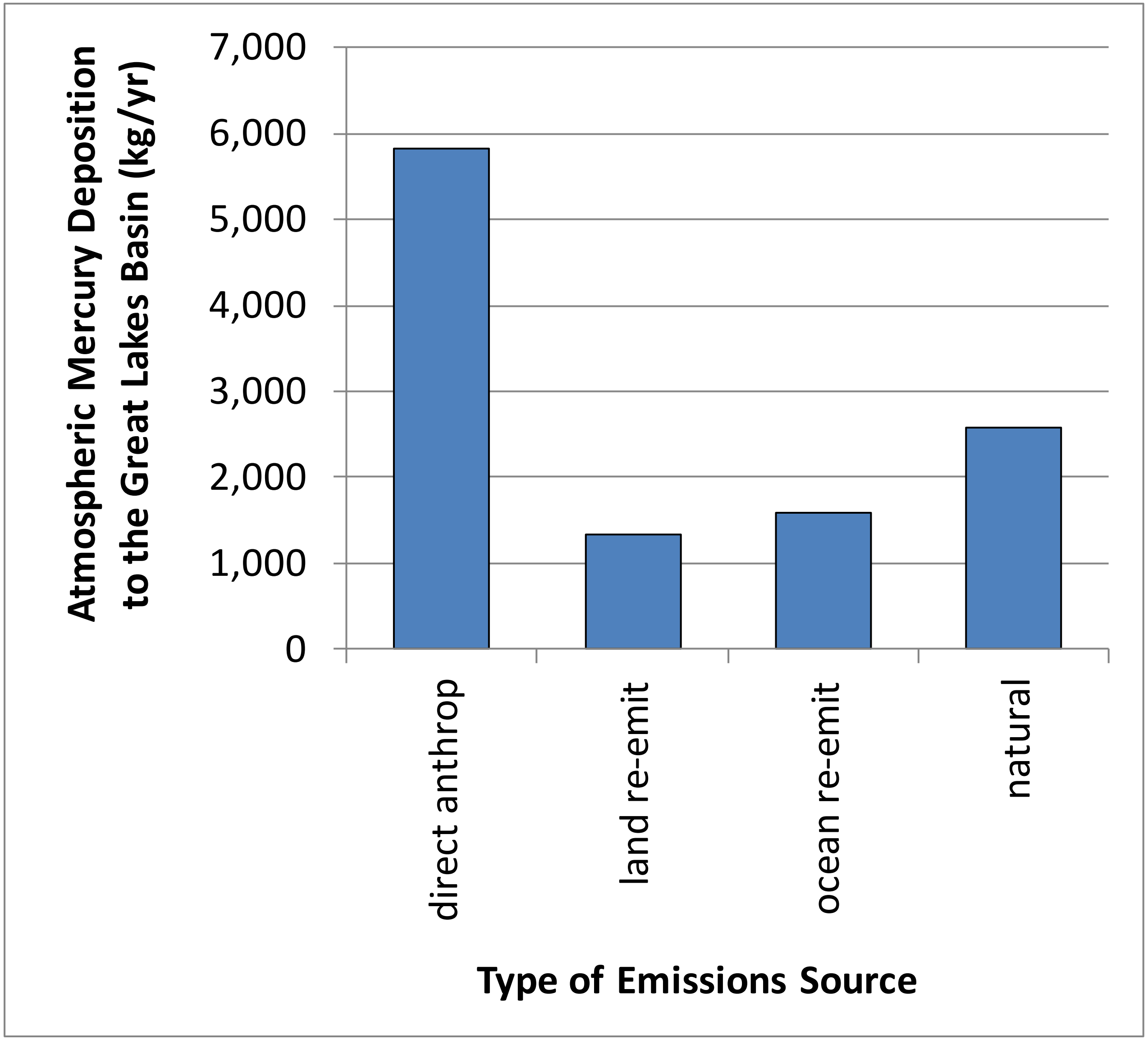
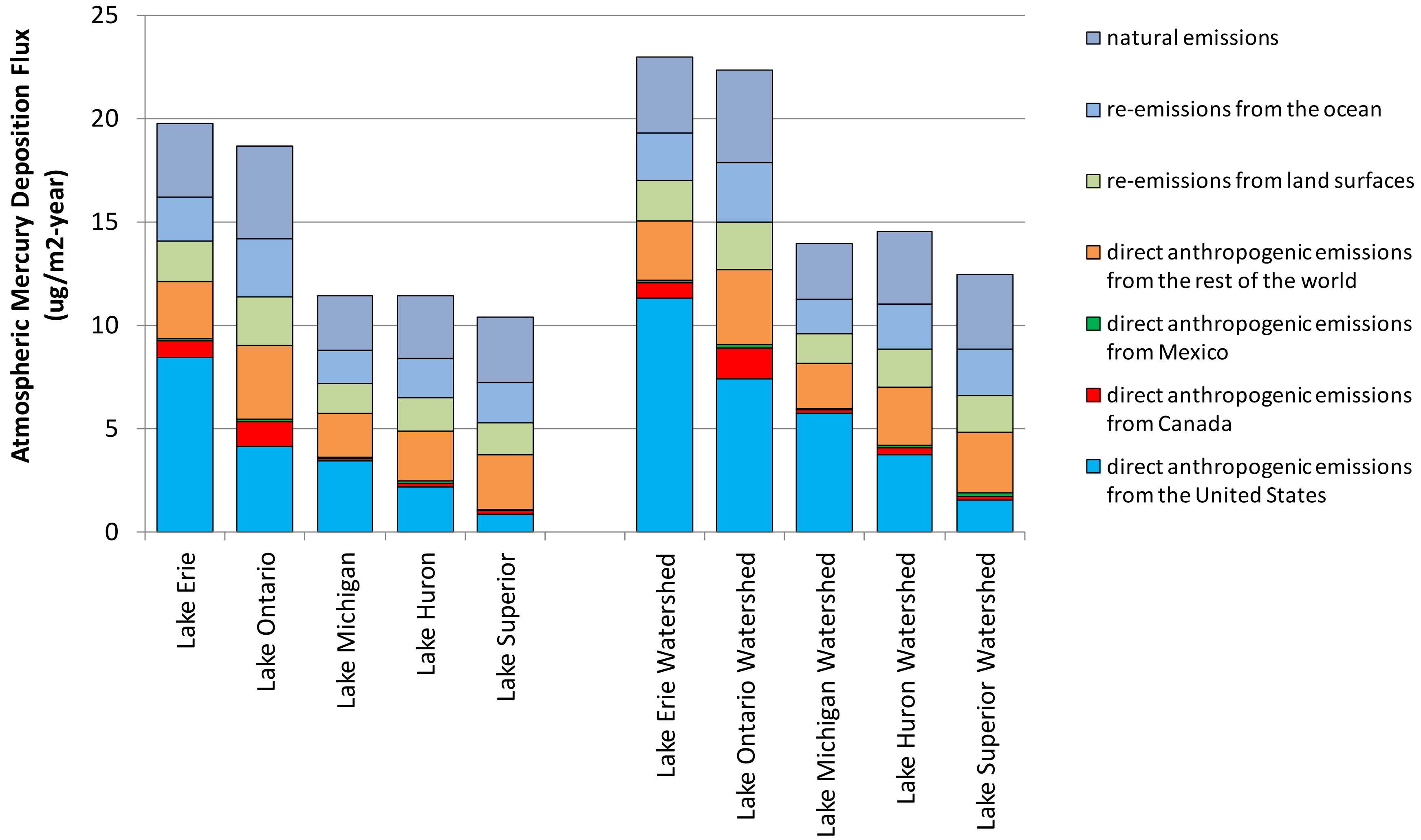


Overall Results for the Great Lakes

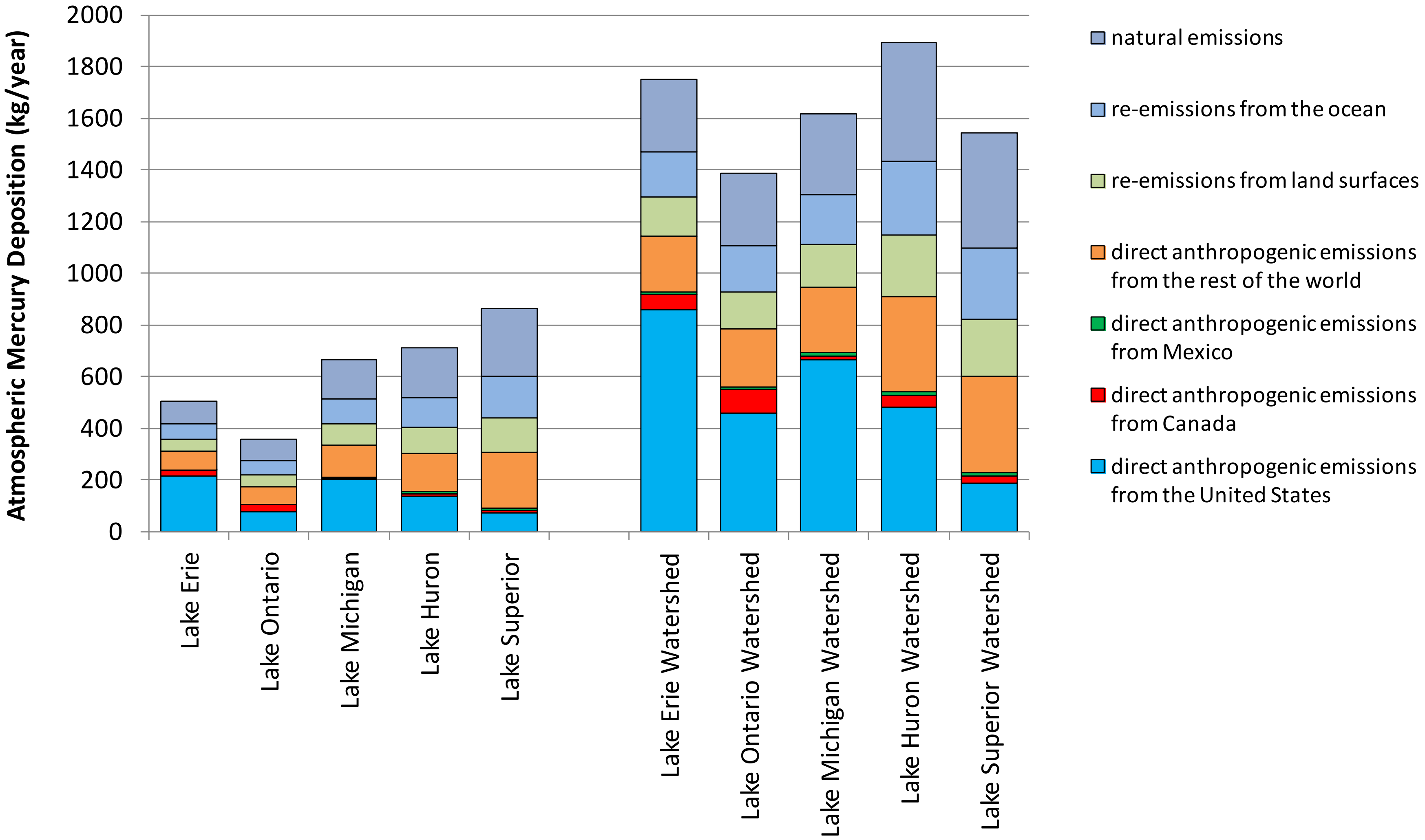
Overall model estimates of mercury deposition to the Great Lakes Basin



Model-estimated 2005 deposition fluxes (ug/m2-year) to the Great Lakes and Great Lakes Watersheds



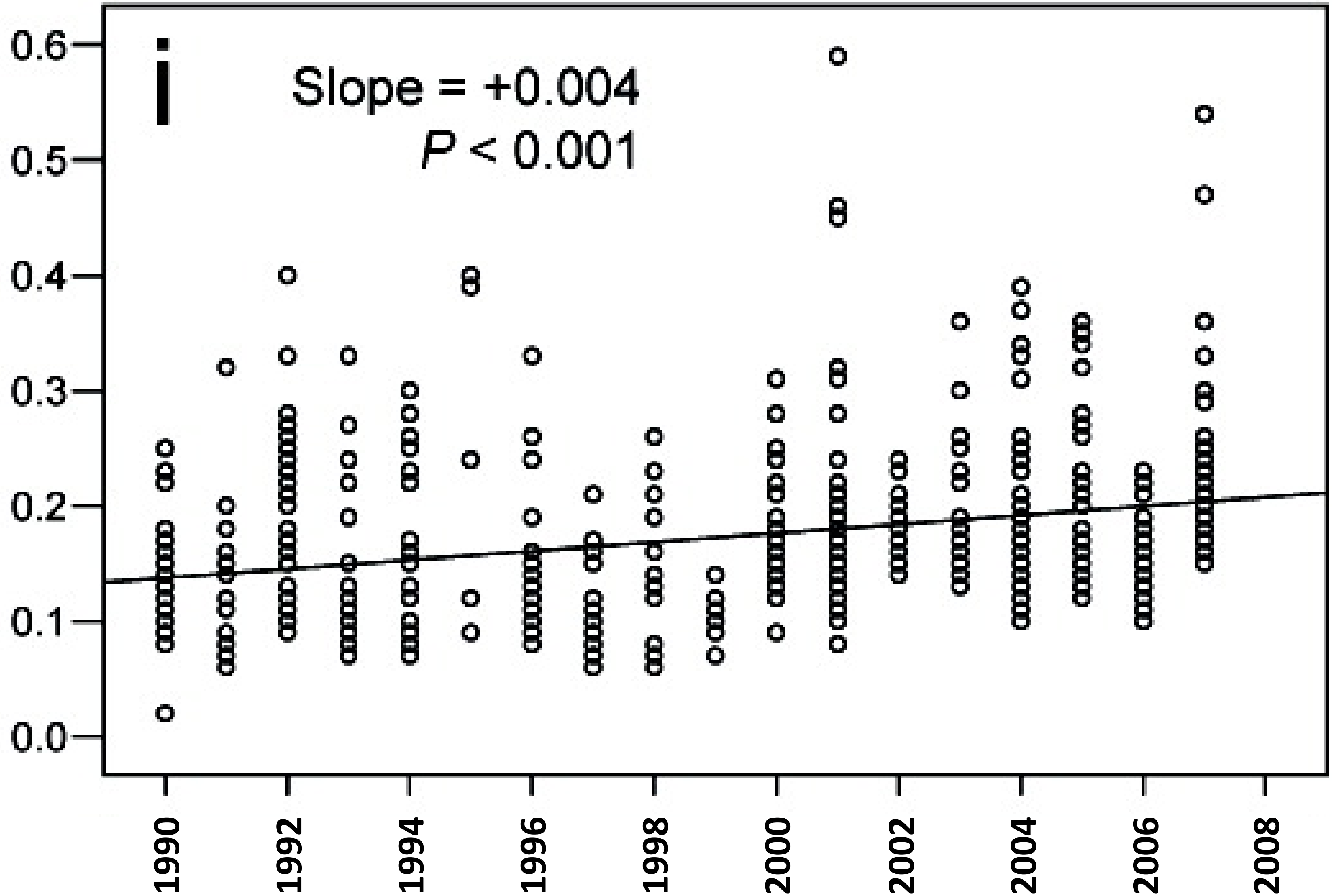
Model-estimated 2005 deposition amounts (kg/year) to the Great Lakes and Great Lakes Watersheds

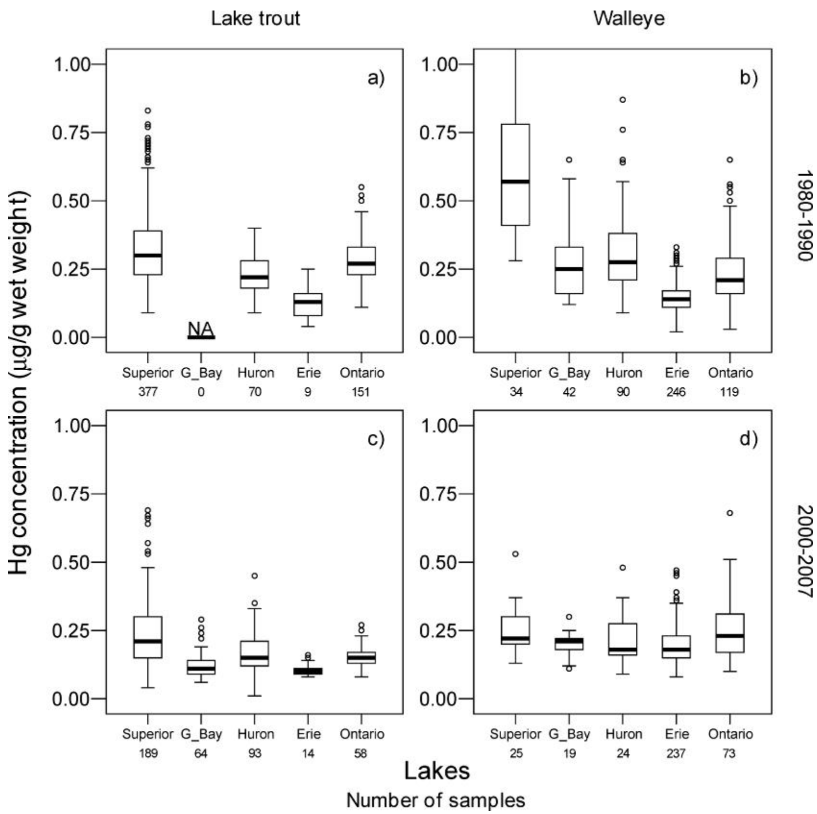


Time Trends of Mercury Concentrations in GL Fish

Satyendra P. Bhavsar; Sarah B. Gewurtz; Daryl J. McGoldrick; Michael J. Keir; Sean M. Backus;
Environ. Sci. Technol. **2010,** 44, 3273-3279.
DOI: 10.1021/es903874x

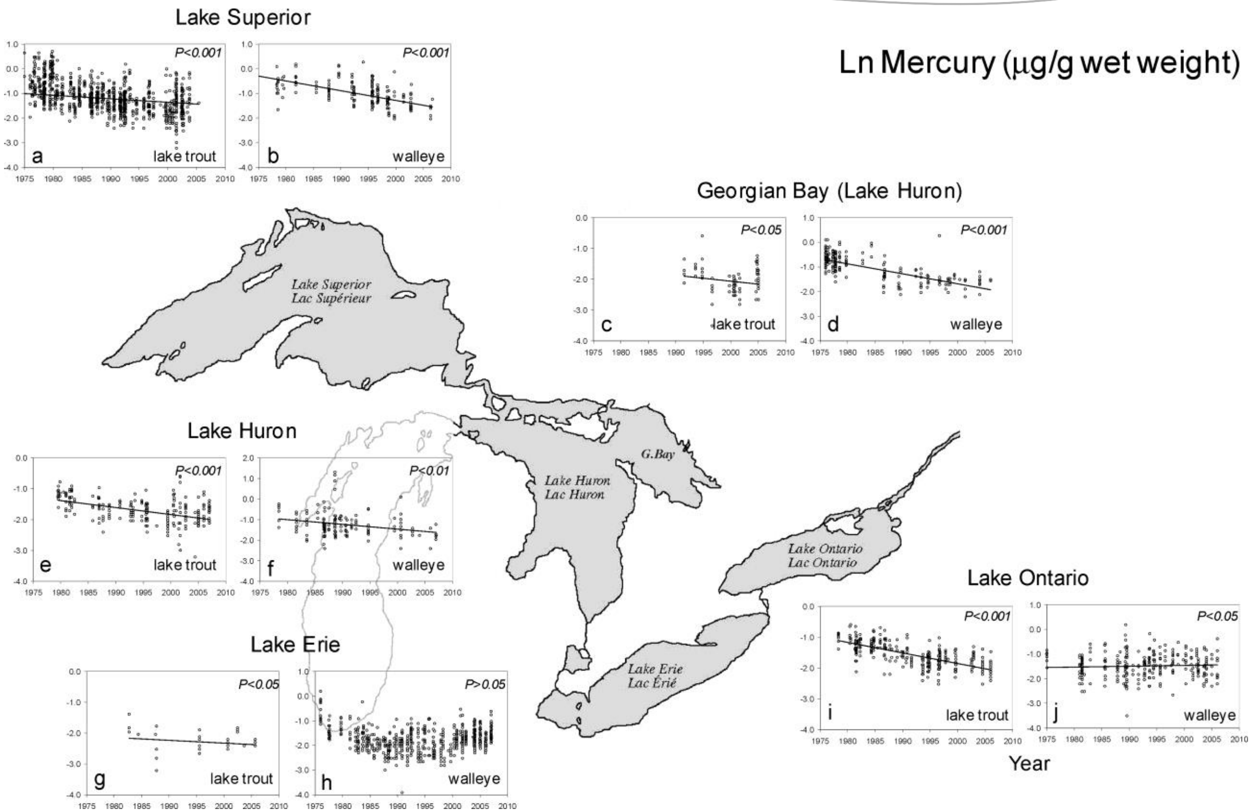
Temporal trends of mercury in OMOE skinless fillets of Lake Erie 45–55 cm walleye collected between 1990–2007 (from Bhavsar et al., 2010)





Measured Hg concentrations (µg/g wet weight) in OMOE skinless fillets. (a, c) 55−65 cm lake trout and (b, d) 45−55 cm walleye collected from Lakes Superior, Georgian Bay, Huron, Erie and Ontario during (a, b) 1980−1990 and (c, d) 2000−2007. Box plots: the line within the box indicates median, the box indicates 25th and 75th quartile values, the whiskers indicate the upper and lower values not classified as statistical outliers or extremes. The outliers (open circles) and extremes (not shown) were values more than 1.5 and 3 times 25th−75th interquartile range away from the closest end of the box, respectively. The numbers below lake names are for sample sizes (*N*). NA indicates not available.

Published in: Satyendra P. Bhavsar; Sarah B. Gewurtz; Daryl J. McGoldrick; Michael J. Keir; Sean M. Backus; *Environ. Sci. Technol.* **2010,** 44, 3273-3279.
DOI: 10.1021/es903874x



Temporal trends of mercury (natural log-transformed, µg/g ww) in OMOE skinless fillets. (a, c, e, g, i) 55−65 cm lake trout, and (b, d, f, h, j) 45−55 cm walleye collected from Lakes Superior (a-b), Georgian Bay (c,d), Huron (e,f), Erie (g,h), and Ontario (i,j) between 1973 and 2007. *P*-value is for statistical significance.

Published in: Satyendra P. Bhavsar; Sarah B. Gewurtz; Daryl J. McGoldrick; Michael J. Keir; Sean M. Backus; *Environ. Sci. Technol.* **2010,** 44, 3273-3279.
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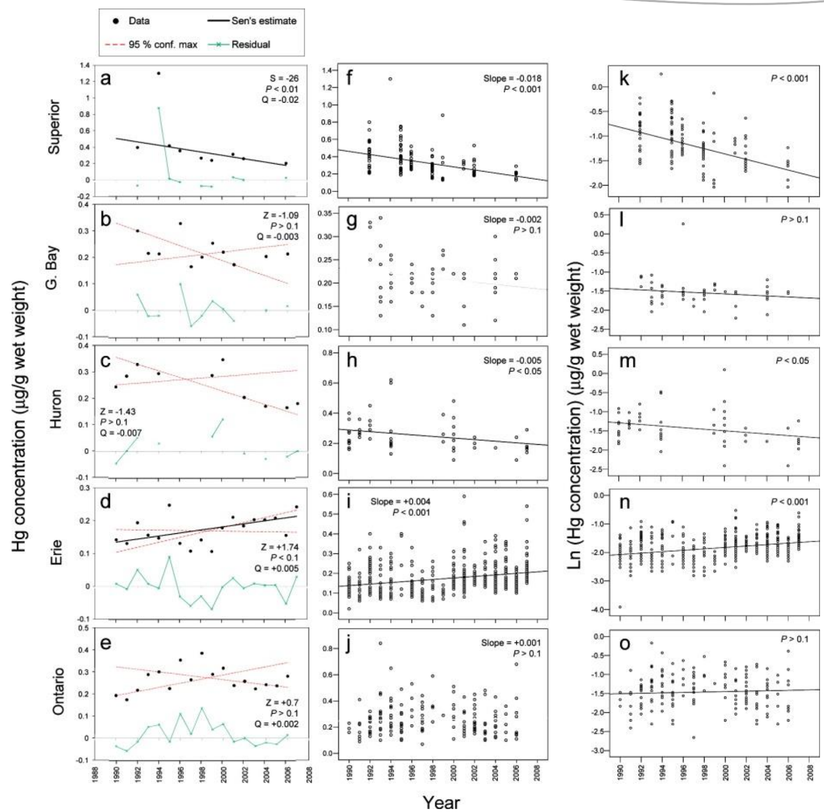

Temporal trends of mercury in OMOE skinless fillets of 45−55 cm walleye collected between 1990−2007. (a−e) Man-Kendall test, (f−j) linear regression on regular concentrations, and (k−o) linear regression on natural log-transformed concentrations. *P*-value is for statistical significance. Sen's estimate line is shown only for statistically significant ($P < 0.05$) upward/downward trend.

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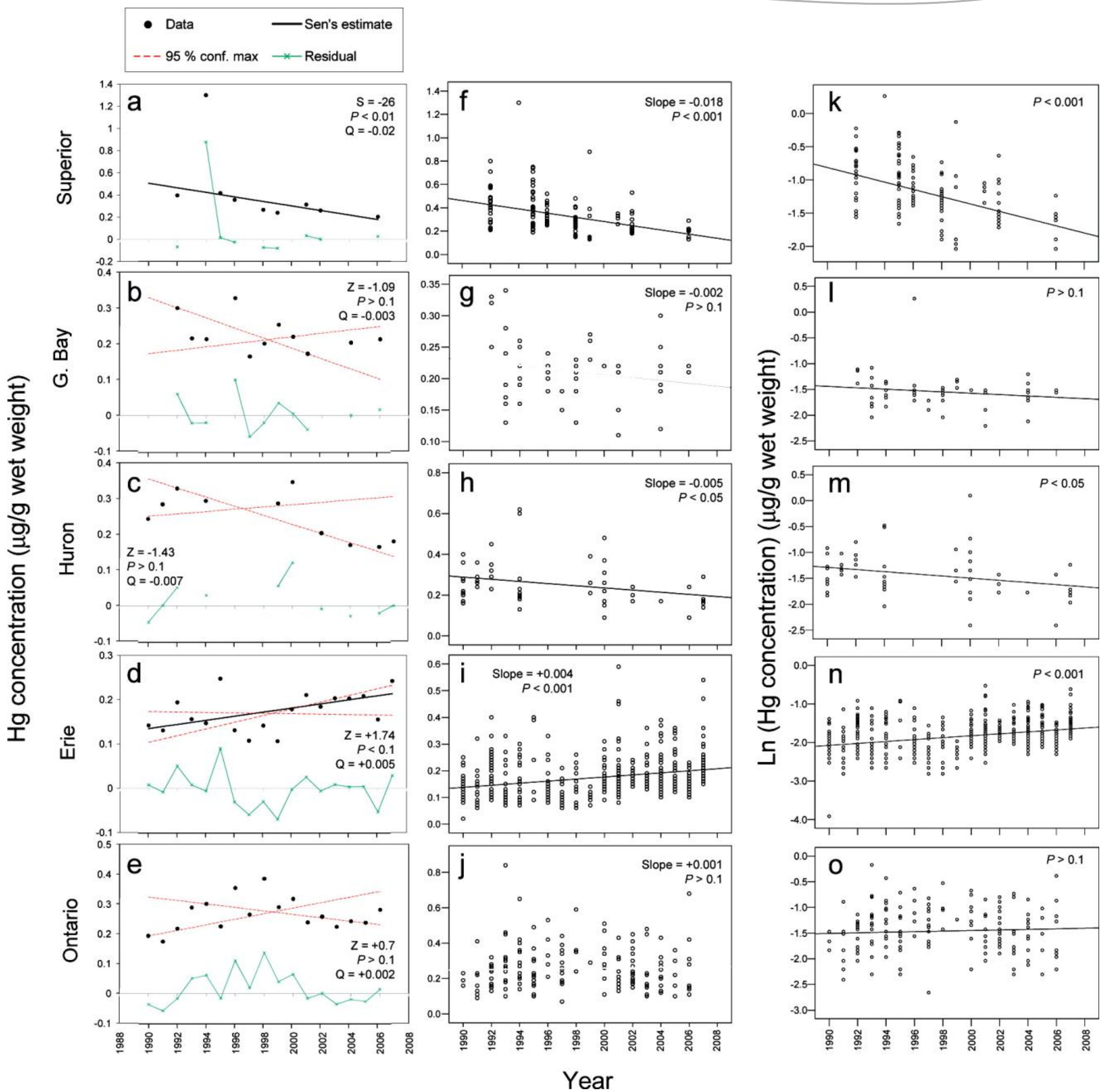
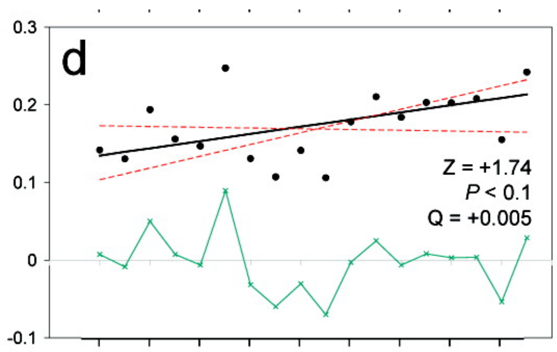
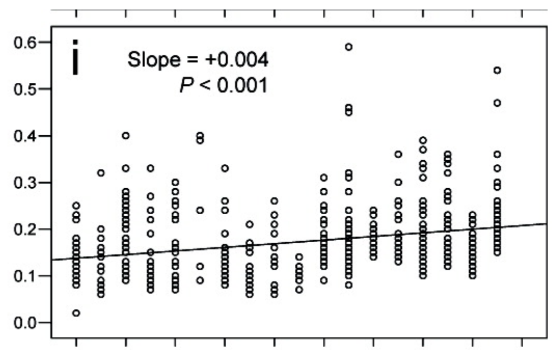
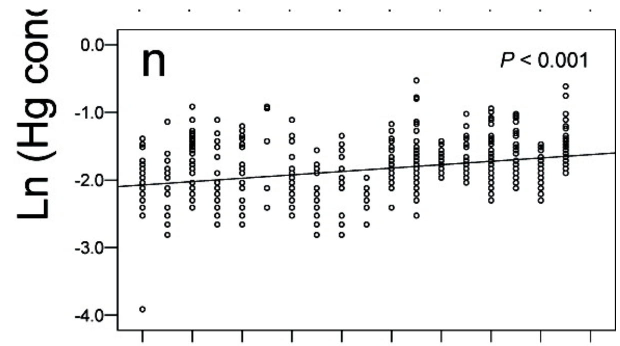
DOI: 10.1021/es903874x


Data
Sen's estimate
95 % conf. max
Residual
Hg concentration (µg/g wet weight)
Ln (Hg concentration) (µg/g wet weight)
Superior
G. Bay
Huron
Erie
Ontario
a
S = -26
P < 0.01
Q = -0.02
b
Z = -1.09
P > 0.1
Q = -0.003
c
Z = -1.43
P > 0.1
Q = -0.007
d
Z = +1.74
P < 0.1
Q = +0.005
e
Z = +0.7
P > 0.1
Q = +0.002
f
Slope = -0.018
P < 0.001
g
Slope = -0.002
P > 0.1
h
Slope = -0.005
P < 0.05
i
Slope = +0.004
P < 0.001
j
Slope = +0.001
P > 0.1
k
P < 0.001
l
P > 0.1
m
P < 0.05
n
P < 0.001
o
P > 0.1
Year

Lake Erie

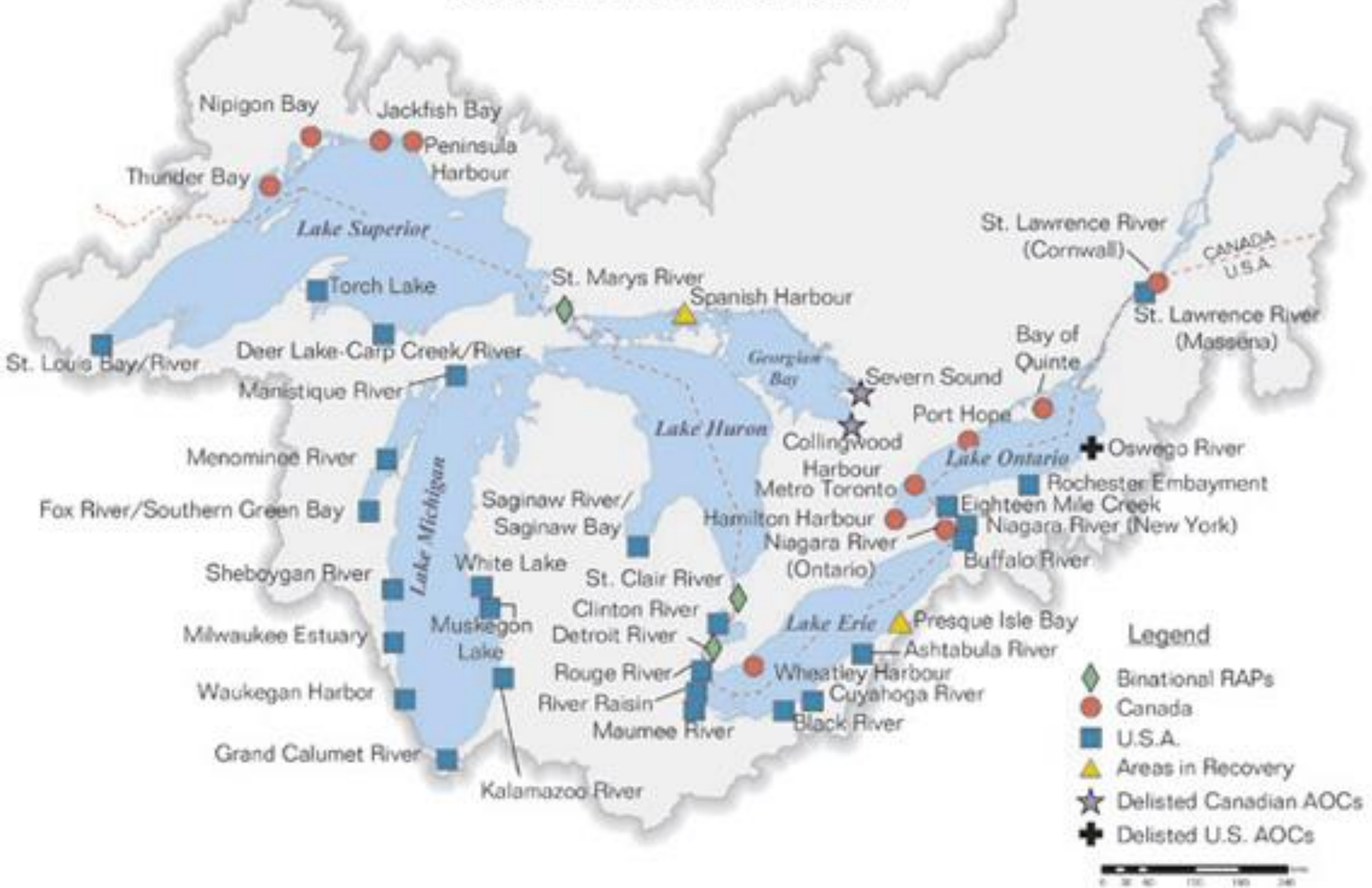

d
0.3
0.2
0.1
0
-0.1
Z = +1.74
P < 0.1
Q = +0.005


i
Slope = +0.004
P < 0.001
0.6
0.5
0.4
0.3
0.2
0.1
0.0

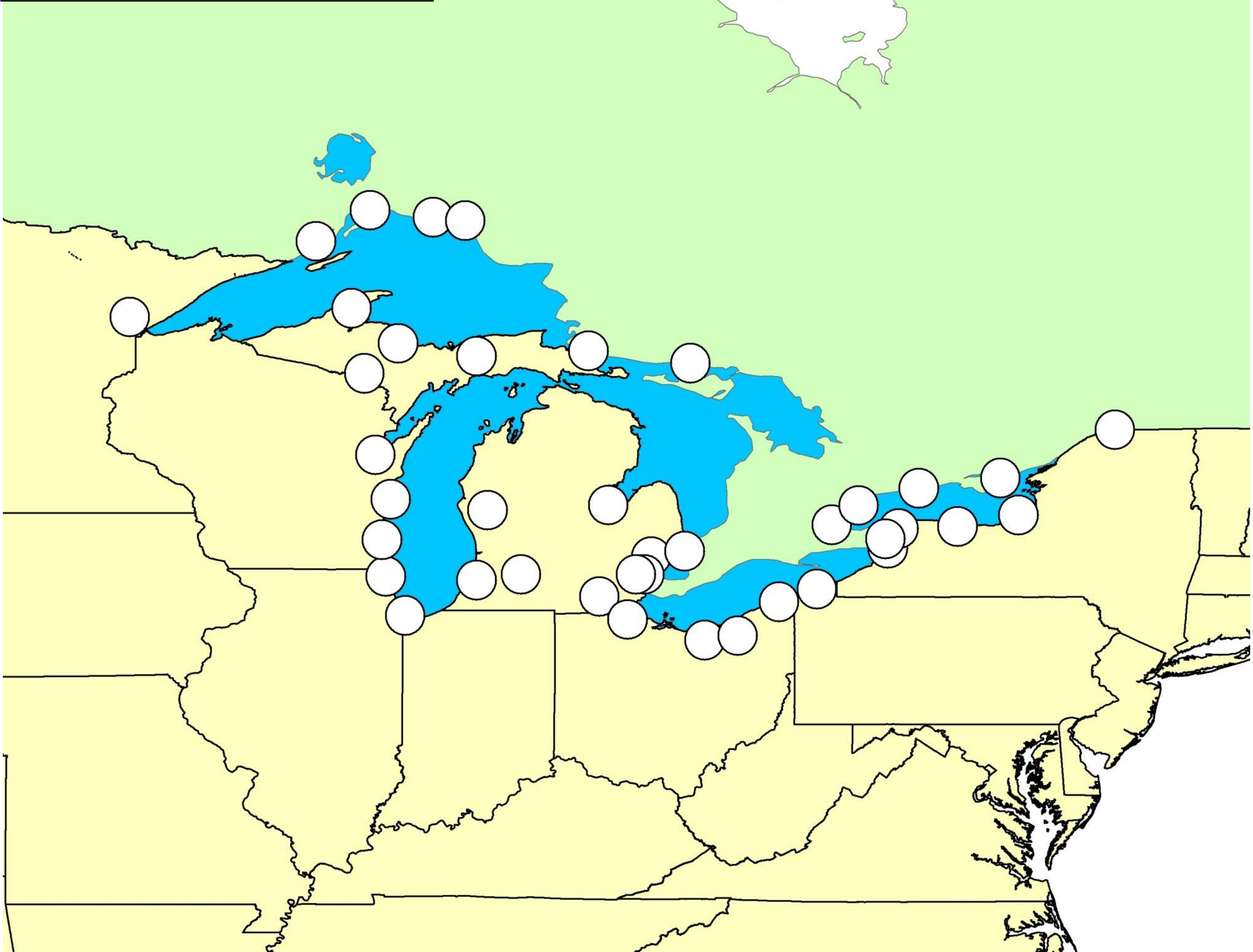

n
P < 0.001
0.0
-1.0
-2.0
-3.0
-4.0

Areas of Concern

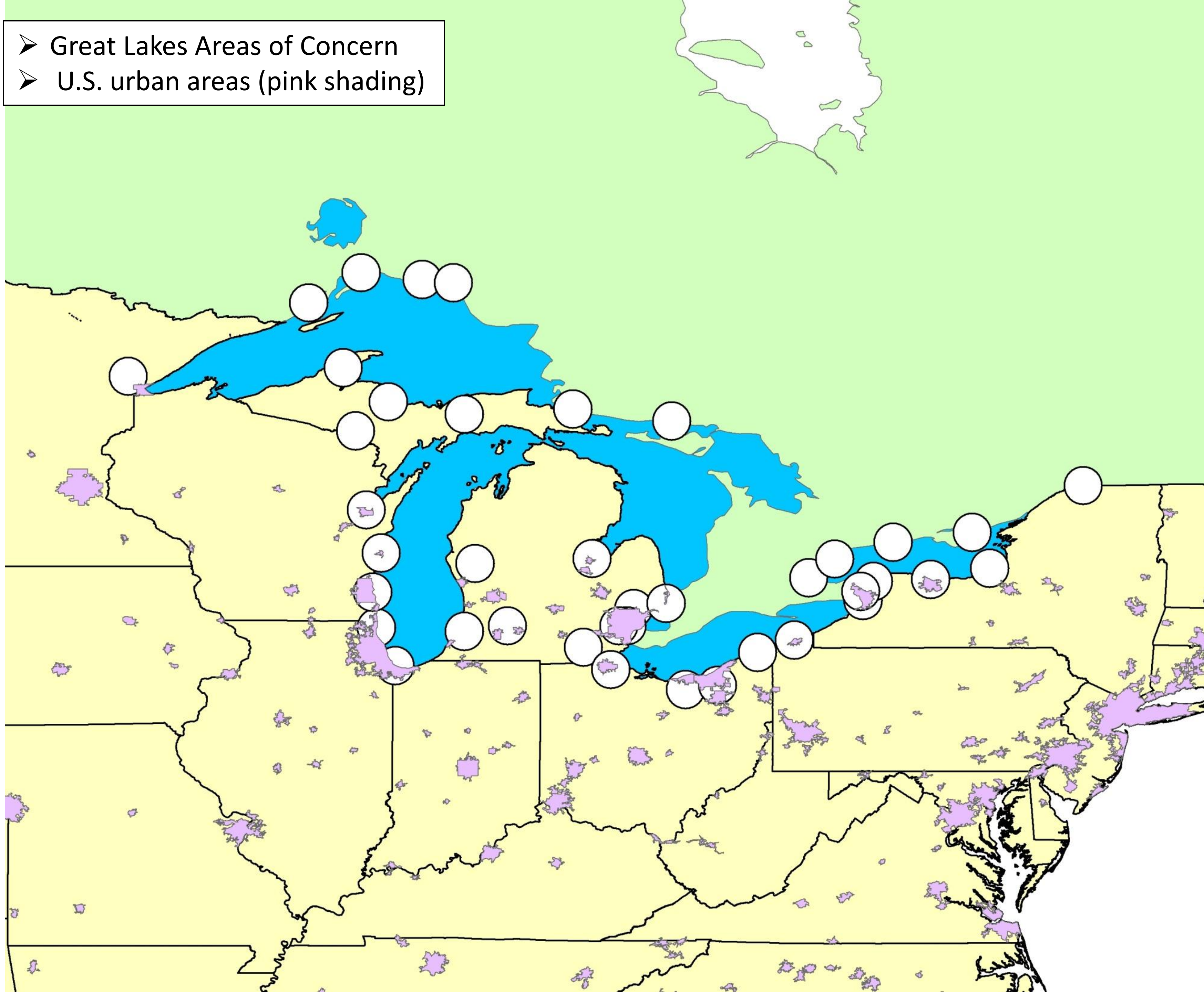
Areas of Concern in the Great Lakes - St. Lawrence River Basin

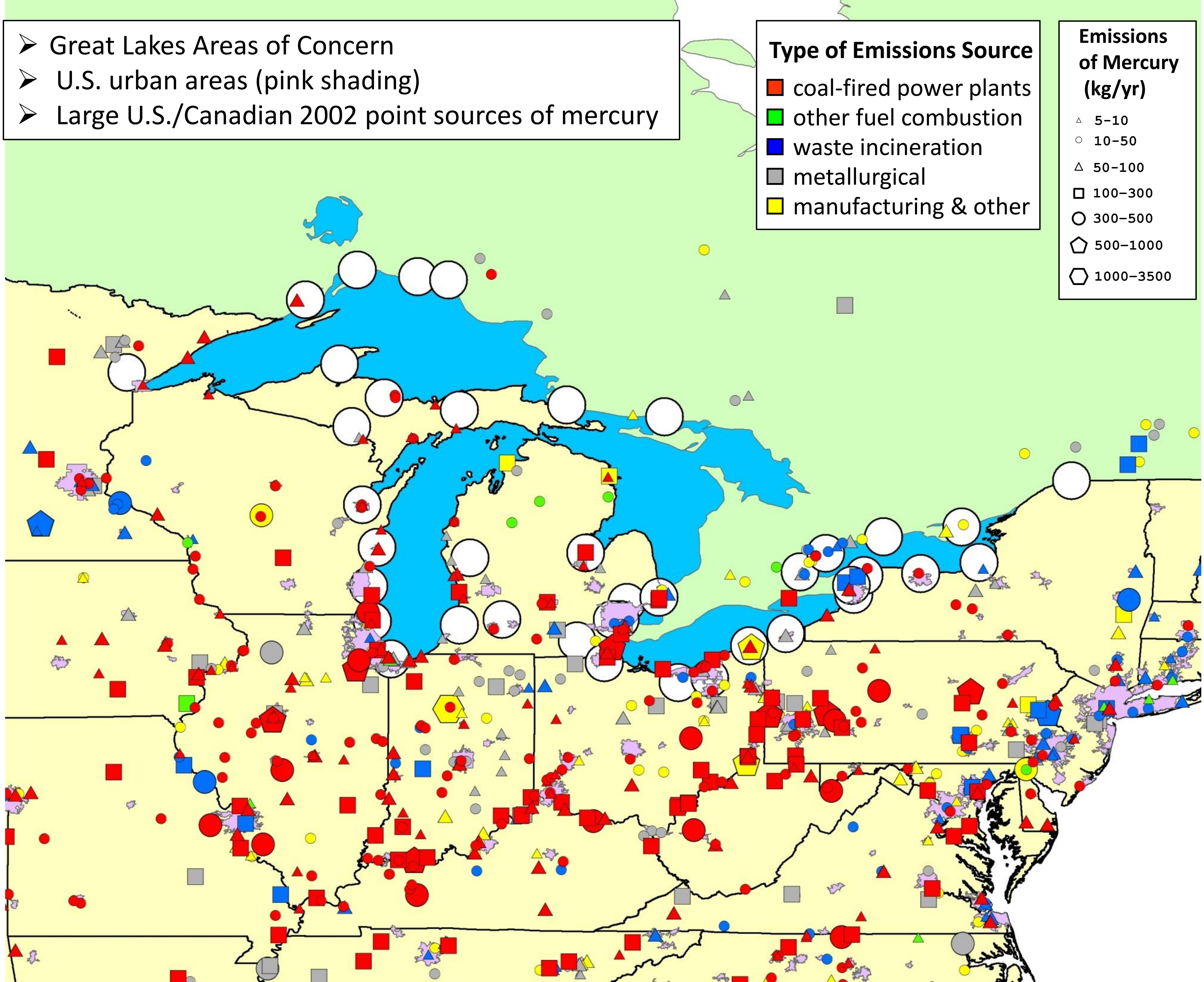


- Great Lakes Areas of Concern

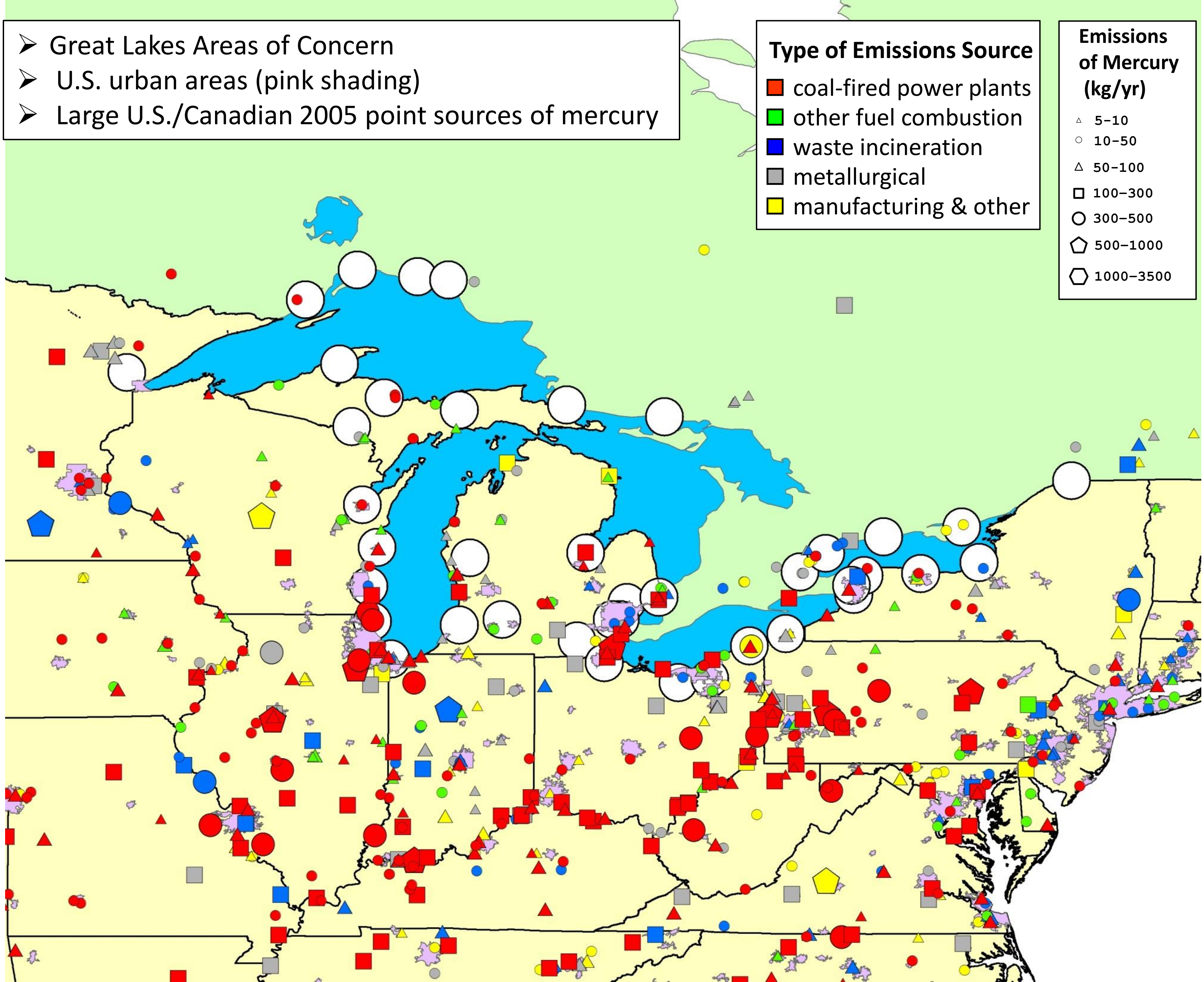


- Great Lakes Areas of Concern
- U.S. urban areas (pink shading)





Great Lakes Areas of Concern
U.S. urban areas (pink shading)
Large U.S./Canadian 2002 point sources of mercury
Type of Emissions Source
coal-fired power plants
other fuel combustion
waste incineration
metallurgical
manufacturing & other
Emissions of Mercury (kg/yr)
5-10
10-50
50-100
100-300
300-500
500-1000
1000-3500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