

# NOAA Report to Congress: Mercury Contamination in the Great Lakes

**Briefing for Rep. Mark Kirk (R-IL)**  
**4 PM, Tuesday June 12, 2007,**  
**Room 1030, Longworth House Office Bldg**

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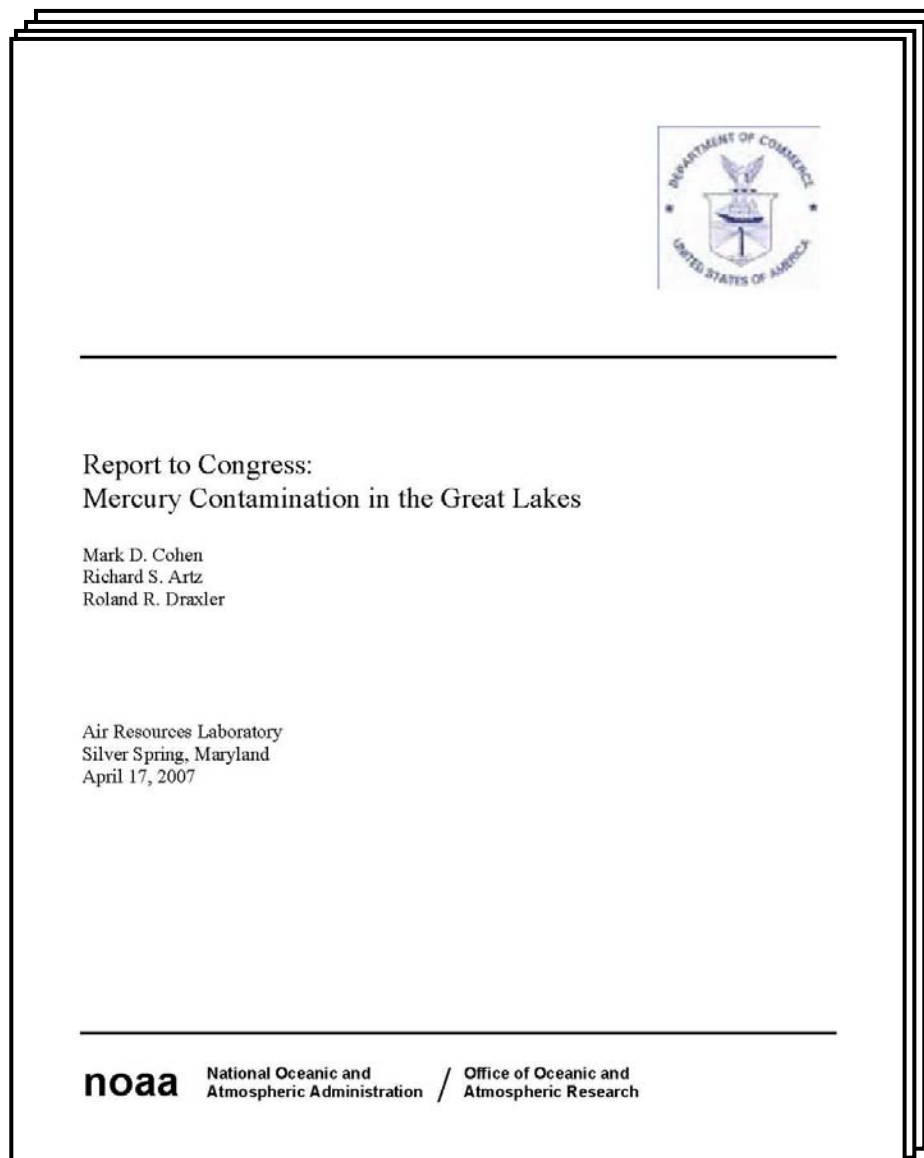


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## NOAA Report to Congress on Mercury Contamination in the Great Lakes

[http://www.arl.noaa.gov/data/web/reports/cohen/NOAA\\_GL\\_Hg.pdf](http://www.arl.noaa.gov/data/web/reports/cohen/NOAA_GL_Hg.pdf)



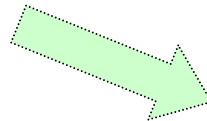
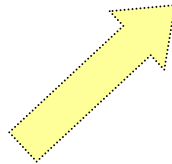
- The Conference Report accompanying the consolidated Appropriations Act, 2005 (H. Rpt. 108-792) requested that NOAA, in consultation with the EPA, report to Congress on mercury contamination in the Great Lakes, with trend and source analysis.
- Reviewed by NOAA, EPA, DOC, White House Office of Science and Technology Policy, and Office of Management and Budget (OMB);
- Preparation / review process took more than 2 years.
- Transmitted to Congress on May 14, 2007

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*This report does not attempt to report on all of the many aspects of mercury contamination in the Great Lakes.*

*It is limited to the following two primary components:*

- Analysis of the atmospheric transport and deposition of U.S. and Canadian anthropogenic mercury emissions to the Great Lakes using the NOAA HYSPLIT-Hg model;
- Illustrative literature data regarding trends in Great Lakes mercury contamination.



### **The Atmospheric Transport and Deposition of Mercury to the Great Lakes**

- A. Introduction
- B. Atmospheric Emissions
- C. Overview of Atmospheric Mercury Modeling
- D. Illustrative Modeling Results for a Single Source
- E. Overall Modeling Methodology
- F. Model Uncertainties
  - 1) Emissions
  - 2) Atmospheric Phase Behavior
  - 3) Atmospheric Chemistry
  - 4) Wet and Dry Deposition
  - 5) Meteorological Data
- G. Model Evaluation
- H. Atmospheric Modeling Results
  - 1) Lake Michigan
  - 2) Lake Superior
  - 3) Lake Huron
  - 4) Lake Erie
  - 5) Lake Ontario
  - 6) Combined Great Lakes
- I. Potential Next Steps
- J. Summary of Great Lakes Atmospheric Mercury Deposition

### **Trends in Great Lakes Mercury**

- A. Mercury Emissions
- B. Mercury Deposition
  - 1) Wet deposition measurements in the Great Lakes region
  - 2) Other mercury wet deposition measurements
  - 3) Modeled deposition to the Great Lakes from U.S. and Canadian sources
- C. Mercury Concentrations in Sediments
- D. Mercury Concentrations in Biota
- E. Summary of Great Lakes Mercury Trend Data

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# Key Findings

1. **Source-attribution information is needed for policy development**
2. **Atmospheric modeling is the only way to obtain comprehensive source-attribution information**
3. **Monitoring *alone* provides values/trends at a few locations, but it cannot answer certain key questions**
  - Why are the values what they are, e.g., source attribution?
  - What are the values in other locations? Why are there (or are there not) spatial and/or temporal trends?
  - What might happen in the future under different environment/policy conditions?
4. **However, atmospheric monitoring data are essential to evaluate and improve models**
  - Atmospheric mercury modeling and monitoring are *far* more useful together than they are apart
  - Monitoring programs need to be designed with model evaluation and improvement in mind
5. **The fate and transport of atmospheric mercury is complex**
  - Scarcity of modeling resources and monitoring\* & process\*\* data means that models haven't been adequately evaluated
    - \* *monitoring data* refers to atmospheric concentration measurements in air and precipitation
    - \*\* *process data* refers to measurements of fundamental phenomena such as chemical reactions and atmospheric deposition processes
  - Thus, while there are many uncertainties in current models, the magnitude of the uncertainties is poorly known
6. **A number of steps could be taken to characterize uncertainties and reduce them if necessary**
  - Collection of additional monitoring data and carrying out process research
  - Increased quality and frequency of emissions data and inventories
  - Comprehensive model evaluation/improvement, sensitivity analyses, and intercomparison experiments
7. **Modeling has provided preliminary, useful information about mercury deposition to the Great Lakes**
  - Detailed, source-attribution results for U.S. and Canadian anthropogenic sources
  - Of these sources, the biggest contribution is from U.S. coal-fired power plants in the Great Lakes region
  - Waste incineration emissions and deposition decreased significantly during the 1990's, but timing poorly known
8. **Further work could provide more complete information**
  - Characterize/reduce uncertainties as described above
  - Extend model to include global anthropogenic and natural sources
  - Carry out simulations of past, present, and potential future atmospheric deposition and source-attribution
9. **Trend data have been assembled for mercury in Great Lakes sediments, biota, emissions and deposition**
  - Levels tended to rise from ~1900 through the 1960's and 1970's, with a peak during WWII.
  - Reductions in the ~1970's, possibly due to the closure or changes at regional mercury-based chlor-alkali factories
  - Levels have remained relatively constant since the 1980's
  - Interpretation of trend data is complicated by a scarcity of data on historical emissions and loading rates

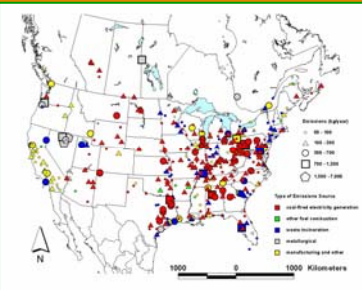


# Emissions Inventories

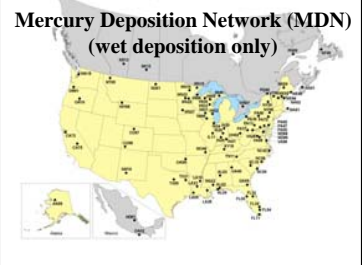


To evaluate and improve atmospheric models, emissions inventories must be:

- Accurate for each individual source (especially for large sources), including variations
- For the same time periods as measurements used for evaluation
- For all forms of mercury

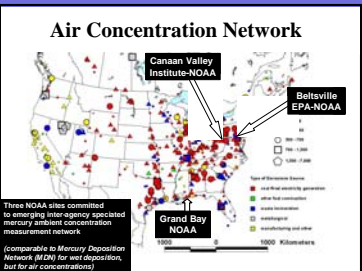


# Atmospheric Monitoring

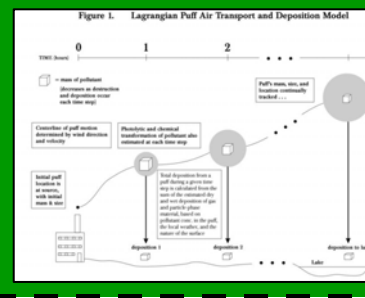
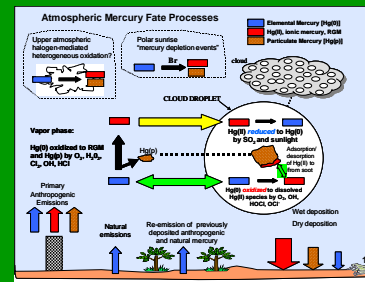


To evaluate and improve atmospheric models, atmospheric monitoring must be:

- For air concentrations (not just wet deposition)
- For all forms of mercury
- For sites impacted by sources (not just background sites)
- At elevations in the atmosphere (not just at ground level)

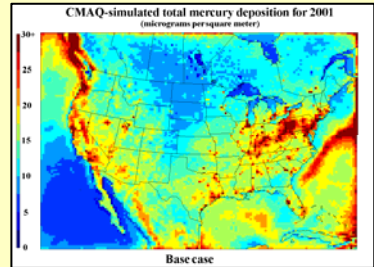


# Atmospheric Models

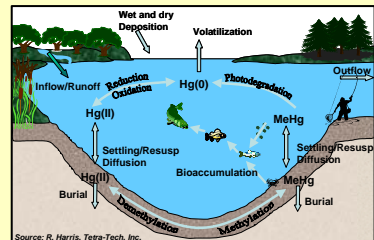


# Understanding and Decisions

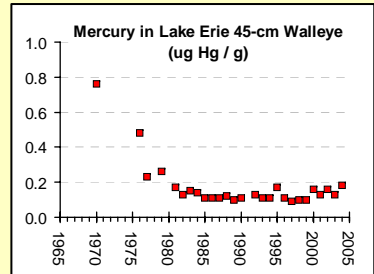
## Deposition For Entire Region



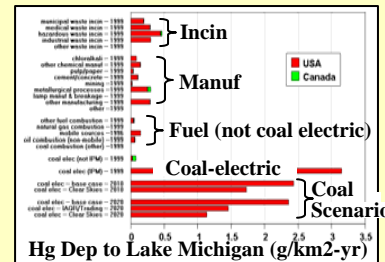
## Inputs for Ecosystem Models



## Understanding Trends

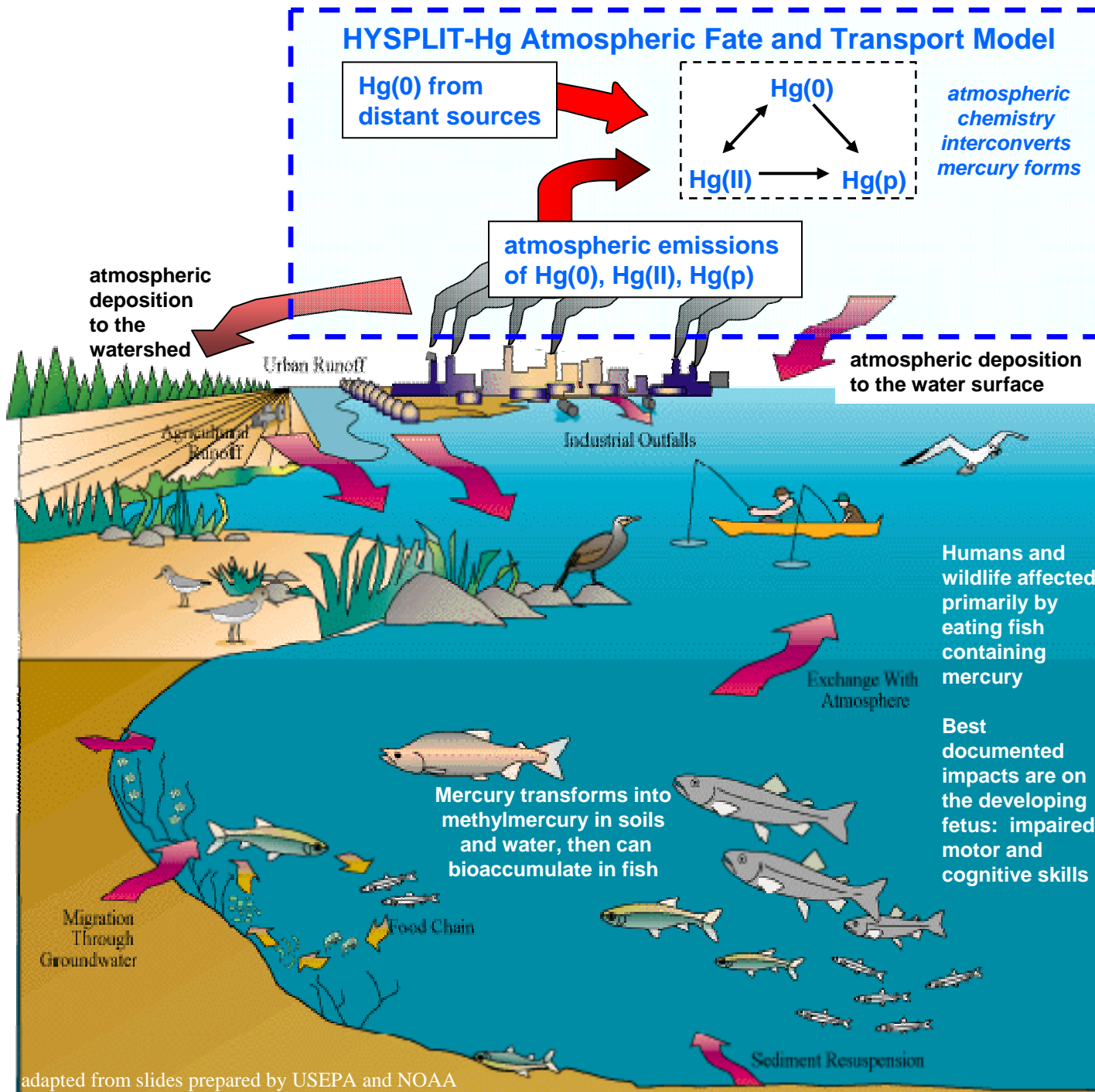


## Source-attribution; Scenarios



# Why do we need atmospheric mercury models?

- **to get *comprehensive source attribution* information**  
...we don't just want to know how much is depositing at any given location, we also want to know where it came from:
  - different source regions (local, regional, national, continental, global)
  - different jurisdictions (different states and provinces)
  - anthropogenic vs. natural emissions
  - different anthropogenic source types (power plants, waste incin., etc)
  
- **to estimate *deposition over large regions***  
...because deposition fields are highly spatially variable, and one can't measure everywhere all the time...
  
- **to estimate *dry deposition***  
... presently, dry deposition can only be estimated via models
  
- **to evaluate *potential consequences* of alternative future emissions scenarios**

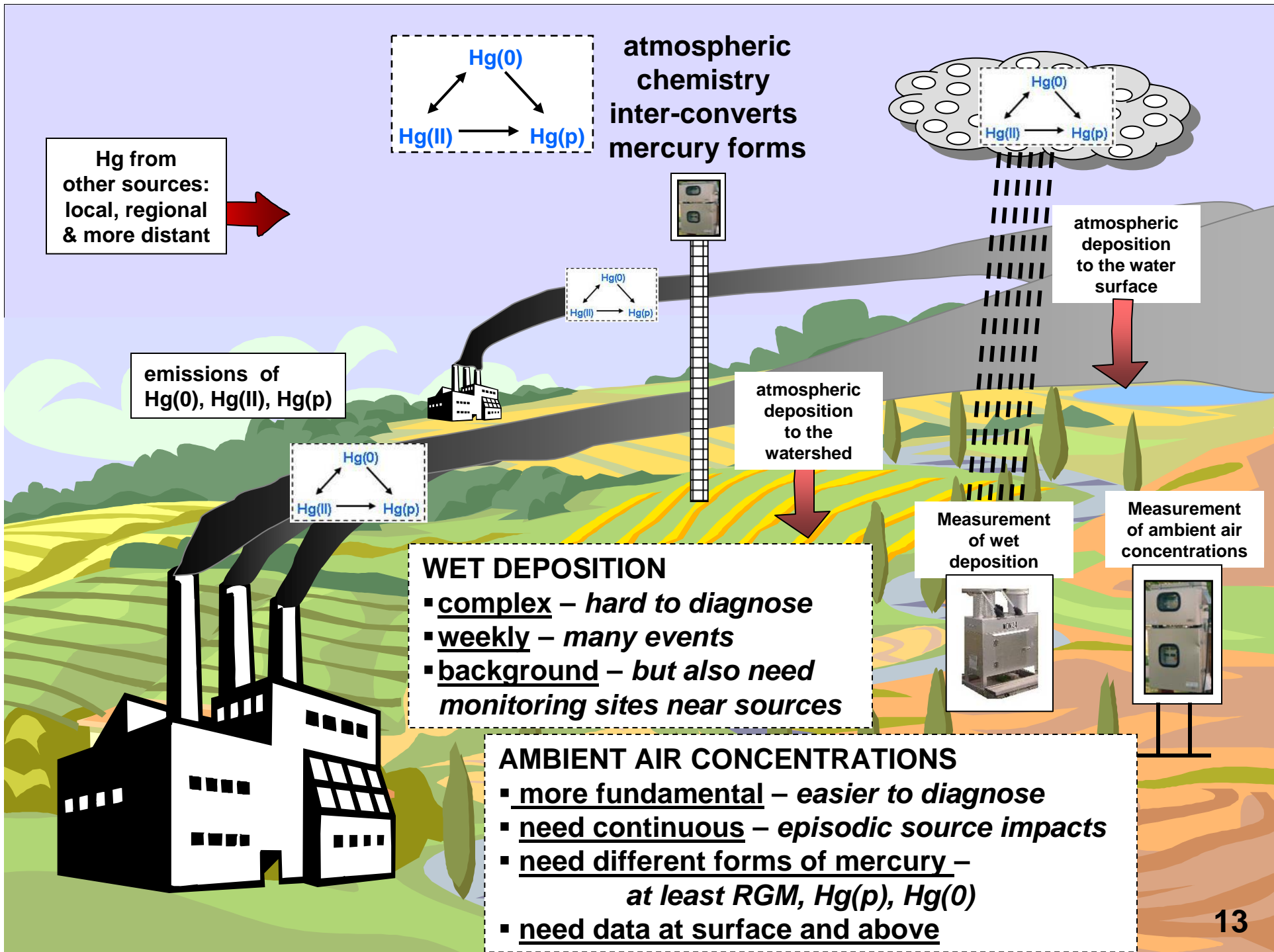


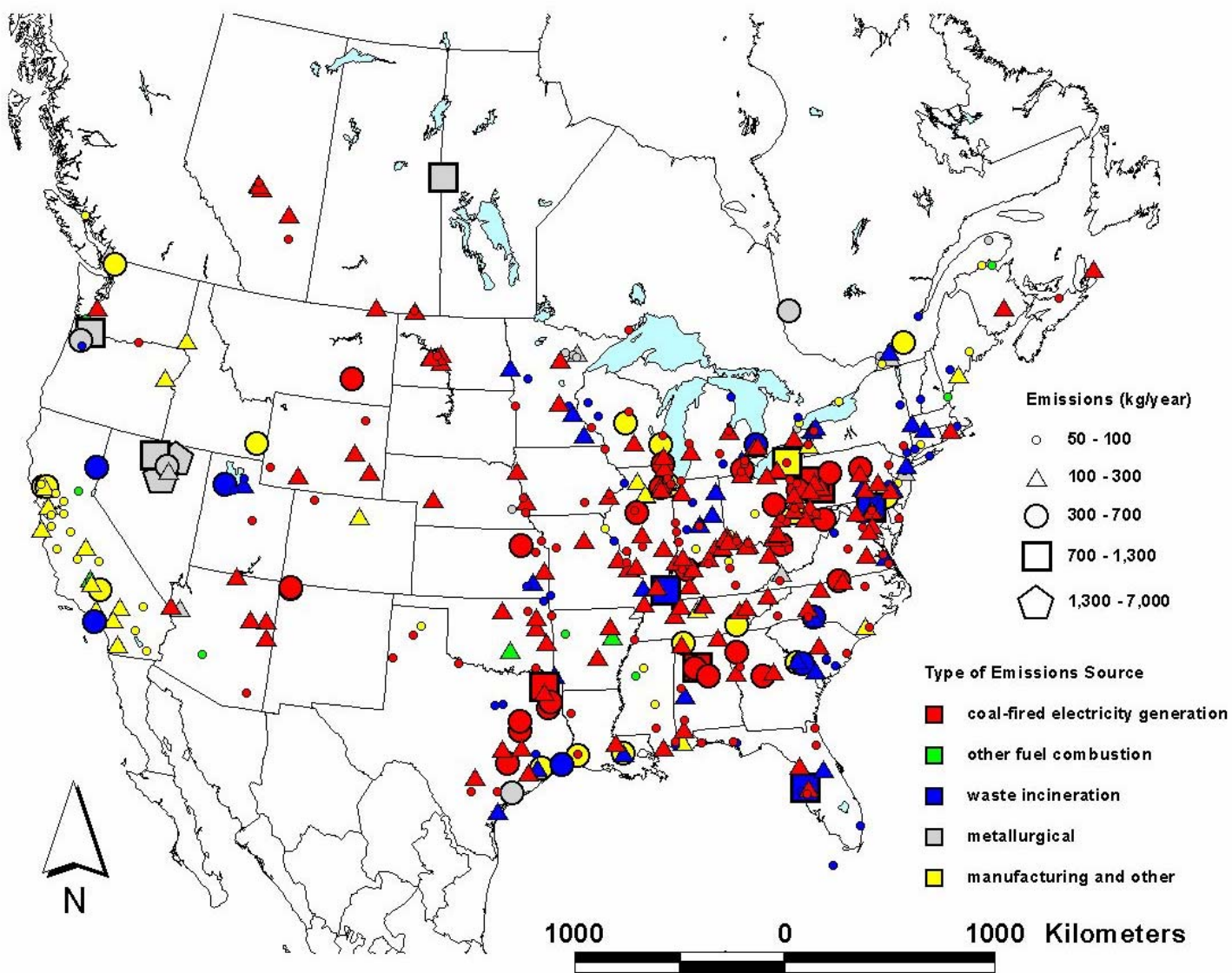
**Where does the mercury come from that is depositing to any given waterbody or watershed?**

- How much from local/regional sources?
- How much from global sources?
- Monitoring alone cannot give us the answer
- atmospheric models required, "ground-truthed" by atmospheric monitoring

**Challenges / critical data needs for model evaluation:**

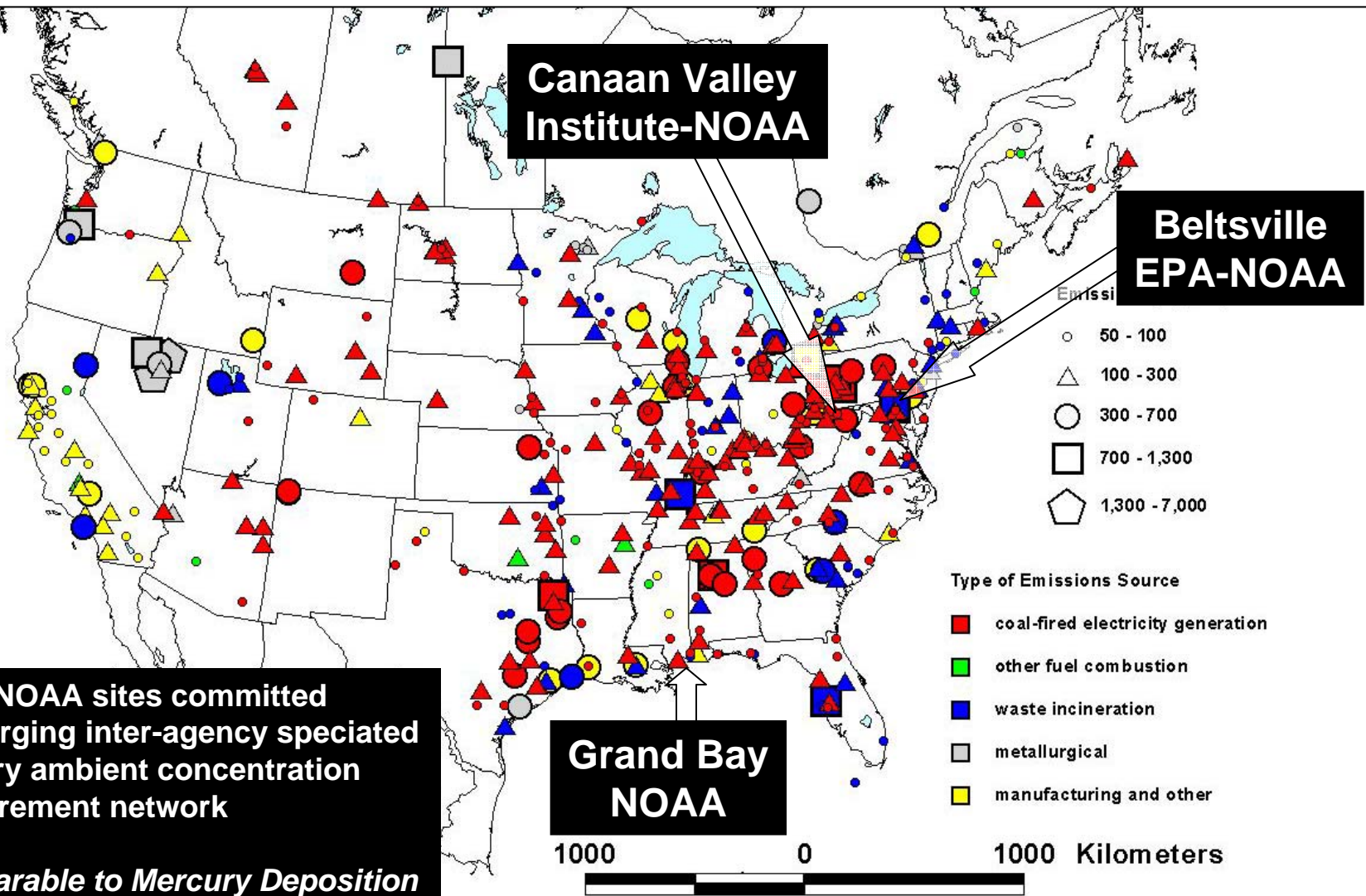
- ❑ Need wet deposition – like data collected in the existing Mercury Deposition Network (MDN) -- but also need ambient air concentrations of different forms of mercury, i.e., reactive gaseous mercury [RGM], particulate mercury [Hg(p)], and elemental mercury [Hg(0)]. Ambient air concentration data is extremely scarce.
- ❑ Need sites that are impacted by large sources as well as background sites that are not impacted by large sources. Most current measurement sites are “background” sites.
- ❑ Most current measurements are currently done at ground level. Also needed are measurements in the atmosphere above the surface (e.g., taken on aircraft, towers...)
- ❑ Unlike the wet deposition data assembled in the Mercury Deposition Network, for ambient concentration data, there are significant data availability issues for what little such data that there is.
- ❑ NOAA is playing a central role with EPA in the emerging national mercury ambient concentration measurement network under the umbrella of the National Atmospheric Deposition Program (NADP). NOAA has “donated” the first three sites for this new network. Contingent upon the cooperation of scientists and other agencies, additional sites will be added and this network will be successfully implemented.



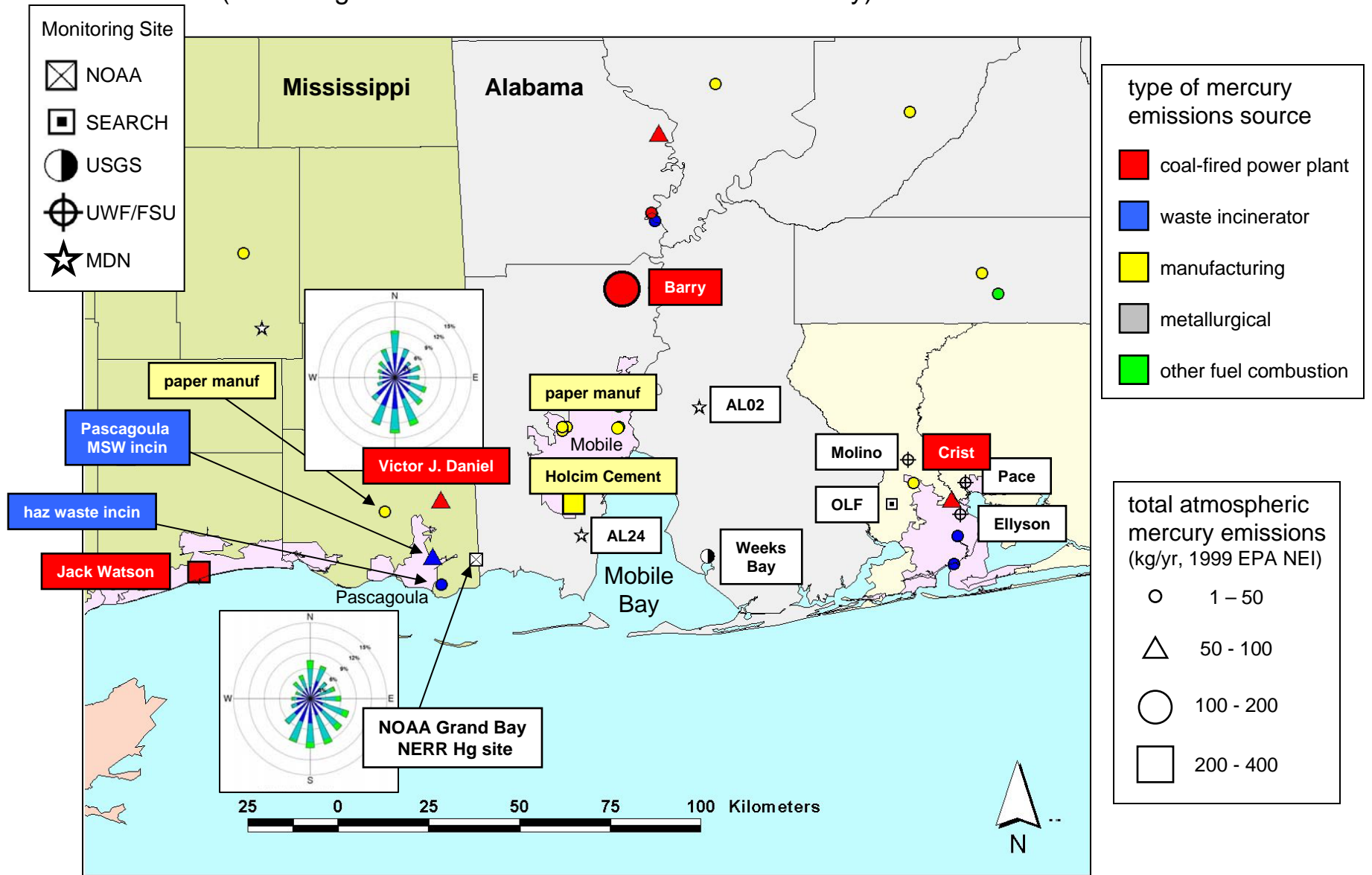


**Figure 12. Largest sources of total mercury emissions to the air in the U.S. and Canada. As discussed in the text, the data generally represent emissions for 1999-2000.**

**Largest sources of total mercury emissions to the air in the U.S. and Canada, based on the U.S. EPA 1999 National Emissions Inventory and 1995-2000 data from Environment Canada**



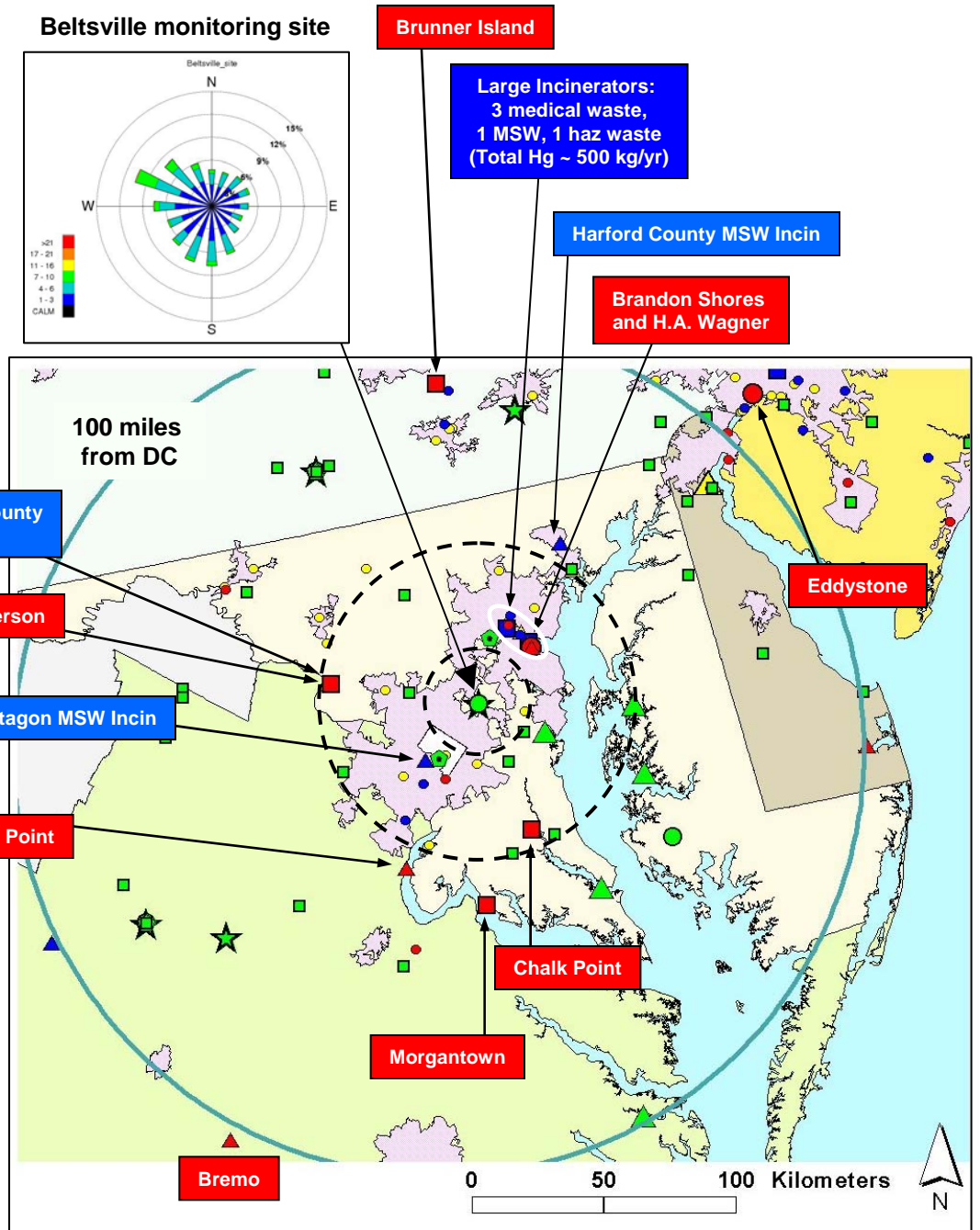
Location of the new NOAA Grand Bay NERR Atmospheric Mercury monitoring site, other atmospheric Hg monitoring sites, and major Hg point sources in the region (according to the EPA 1999 NEI emissions inventory)





Location of the new NOAA-EPA Atmospheric Mercury monitoring site at Beltsville Maryland, other atmospheric monitoring sites, and major Hg point sources in the region (according to the EPA 1999 NEI emissions inventory)

the region between the 20 km and 60 km radius circles displayed around the monitoring site might be considered the "ideal" location for sources to be investigated by the site



**Monitoring sites**

- rural AQS
- other AQS
- ★ NADP/MDN
- CASTNet
- ▲ Hg site
- ⬠ IMPROVE

**Symbol color indicates type of mercury source**

- coal
- incinerator
- metals
- manuf/other

**Symbol size and shape indicates 1999 mercury emissions, kg/yr**

- 1 - 50
- ▲ 50 - 100
- 100 - 200
- 200 - 400
- ▲ 400 - 700
- ⬠ 700 - 1000
- > 1000

- Emissions inventories are fundamental inputs for atmospheric mercury models.
- Accurate inventories are required for model evaluation and improvement, as well as for accurate simulations once the models are “perfected”

#### Inventories need to be improved

- *Inventories need to be more complete; more accurate; more transparent; uncertainties estimated.*
- *Emissions estimates needed for all forms of mercury [RGM, Hg(p), Hg(0)].*

#### Long delay before inventories released

- *2002 U.S. inventory released in 2007; till now, latest available inventory was for 1999.*
- *Can't use new measurement data to evaluate models if the inventories aren't available.*

#### Inventories must be prepared more frequently

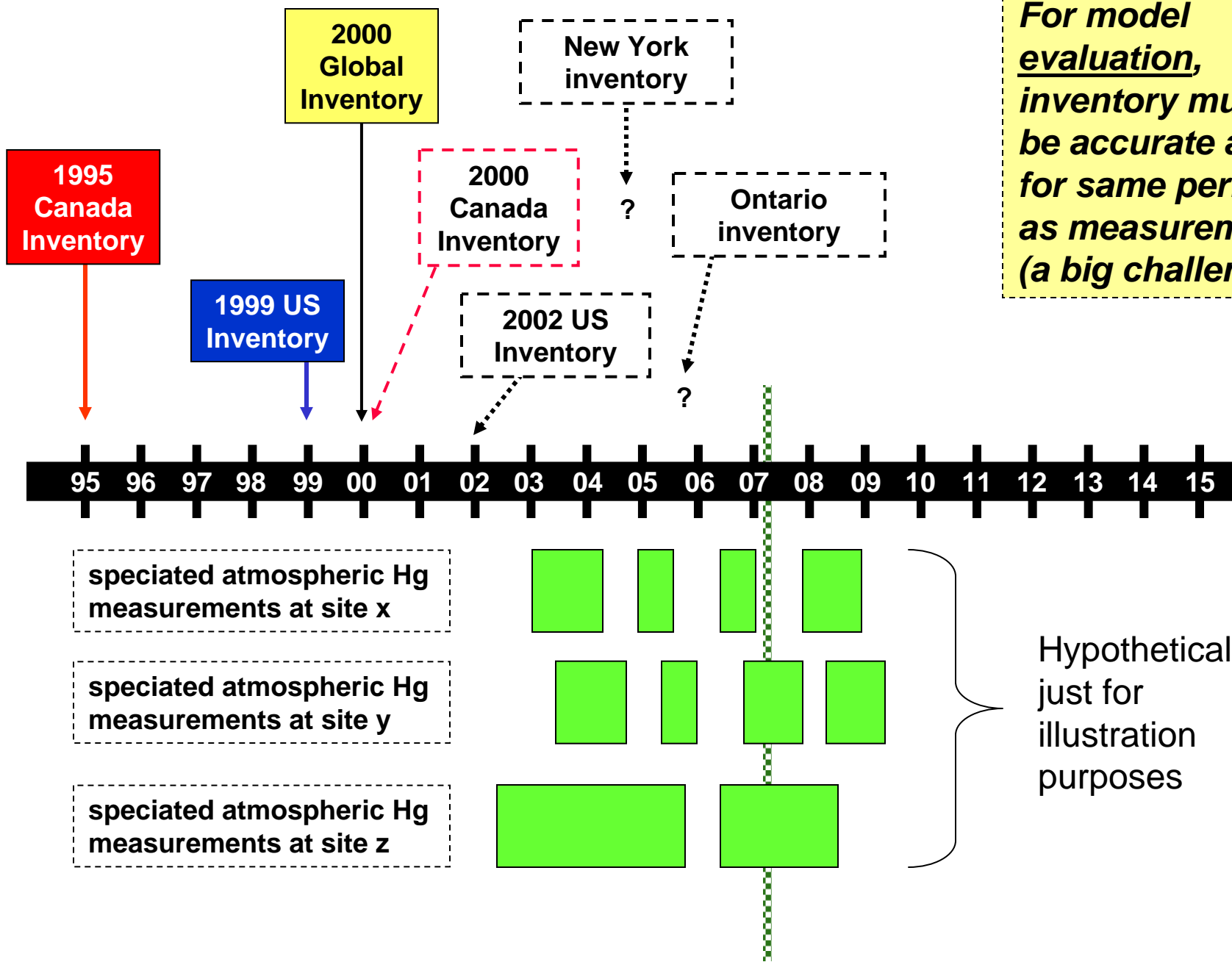
- *Currently, the only available source-by-source inventories for the U.S. are for 1999 and for 2002.*
- *Large emissions reduction between ~1990 and ~2000, but not known when reductions occurred at individual facilities. Thus, very difficult to interpret trends in monitoring data.*

#### Inventories need to include information about major “step-change” events

- *There can be abrupt “step-changes” in emissions due to shutdowns, maintenance, closures, installation of new pollution control devices, feedstock changes, and process changes, etc.*
- *Currently, the only data available in emissions inventories is an annual average. Therefore, it is difficult to interpret variations/trends in ambient measurements.*

#### Data are needed on short-term variations on time scales of minutes to hours

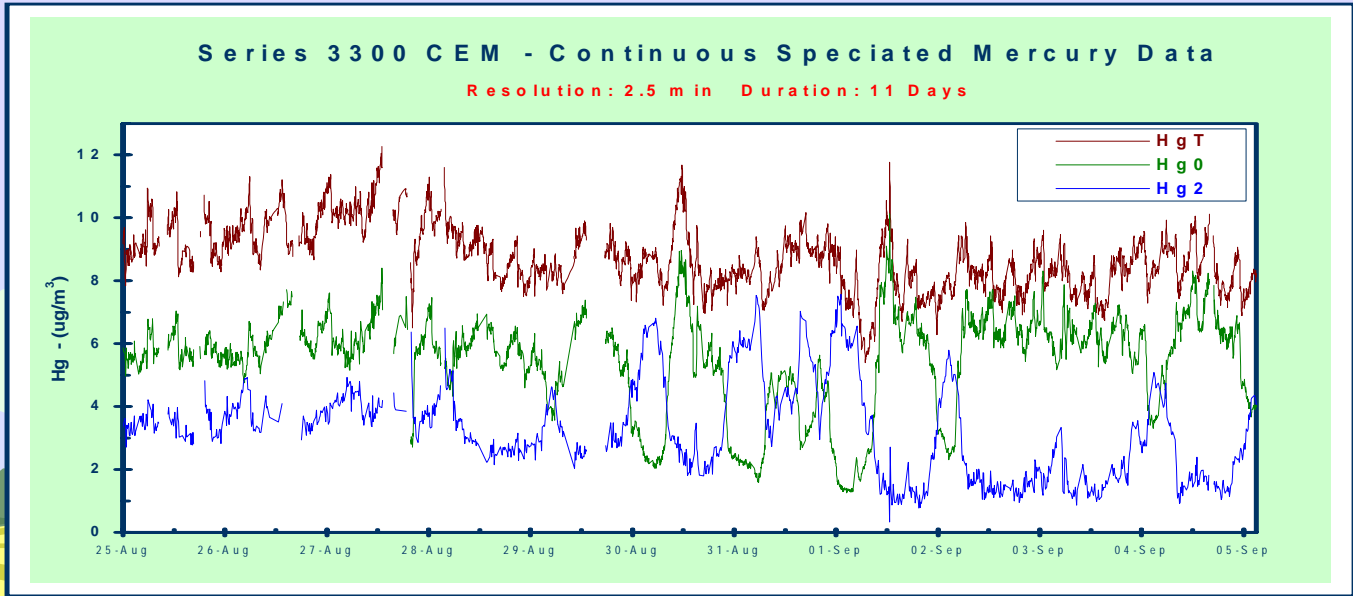
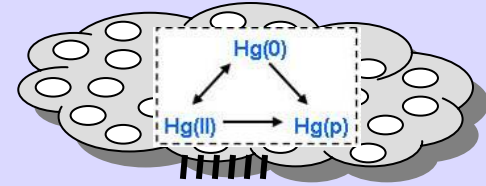
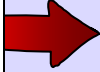
- *There are short-term variations in emissions, on scales of minutes to hours. We need to know about these short term variations to correlate emissions with measurements.*
- *Clean Air Mercury Rule requires ~weekly total-Hg measurements for coal-fired power plants. Continuous Emissions Monitors (CEM's) needed – and not just on coal-fired power plants.*
- *CEM's must measure different forms of mercury or will not be useful in developing source-receptor info.*



## Importance of time-resolved, speciated emissions measurements

- ❑ Based on CEM data collected at coal-fired power plants, it appears that there can be significant variations in emissions of Hg(2), Hg(0) and Hg(p) over time scales of minutes to hours
- ❑ Meteorological conditions – and hence source-receptor relations – can vary significantly over time scales of minutes to hours.
- ❑ If we are collecting speciated ambient concentration data downwind of sources on time scales of minutes to hours, and the source emissions are varying on the same time scales, it is critical to have data regarding the emissions variations. Without it, severe limitations on what can be learned from the ambient concentration data.
- ❑ Speciated Continuous Emissions Monitors (CEM's) are commercially available.
- ❑ CAMR does not appear to require speciated emissions data, and does not appear to require time-resolved data on the order of minutes to hours (i.e., longer term data are all that is required, e.g., on the order of ~1 week). So, we have a problem.
- ❑ For the purposes of model evaluation and improvement, and to the extent possible, it would be helpful if speciated, time-resolved CEM's could be installed at large Hg sources significantly impacting critical model-evaluation monitoring sites.

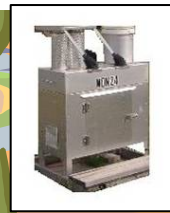
Hg from other sources: local, regional & more distant



atmospheric deposition to the water surface



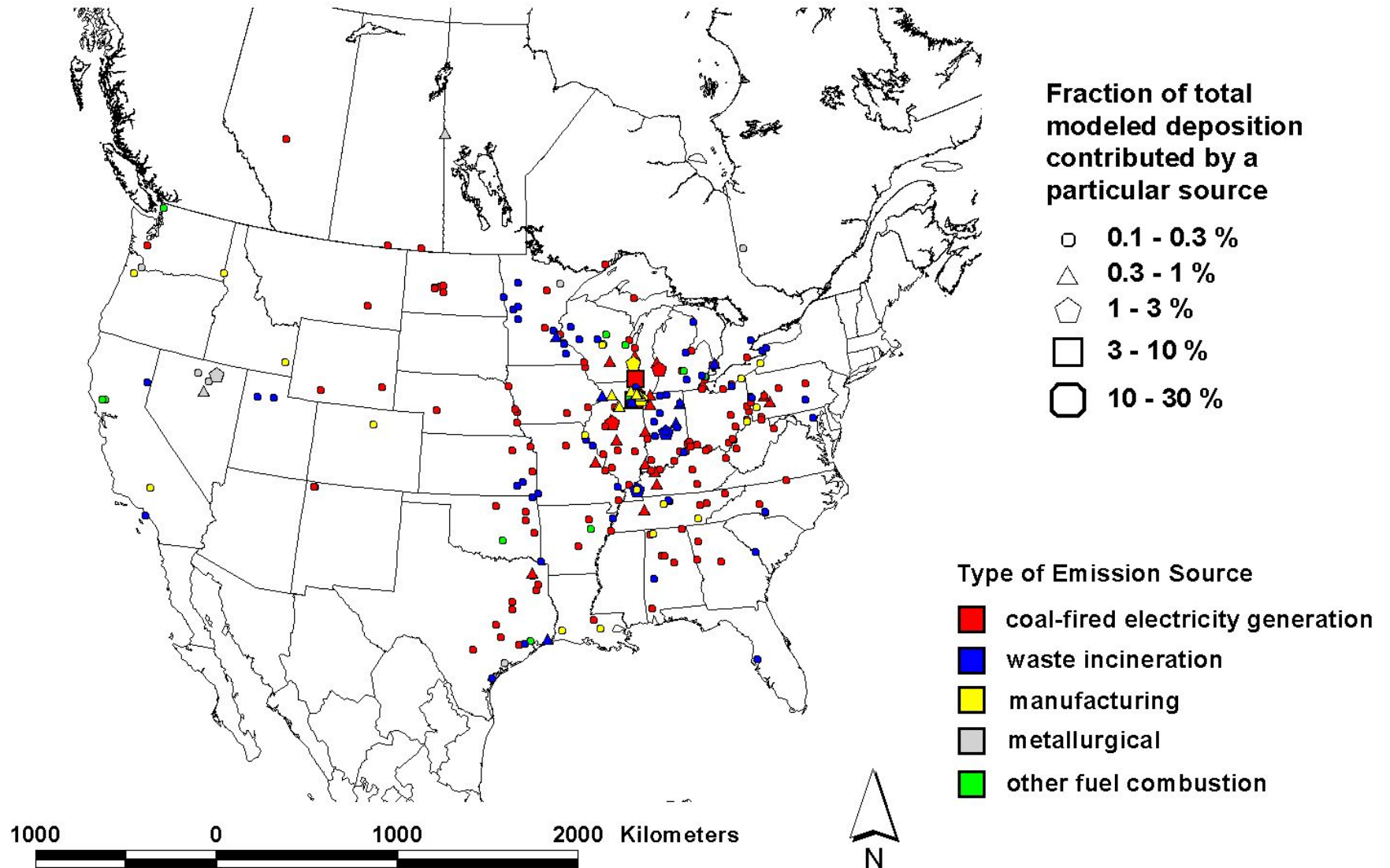
Measurement of wet deposition



Measurement of ambient air concentrations



**Largest atmospheric deposition contributors to Lake Michigan based on 1999-2000 emissions**



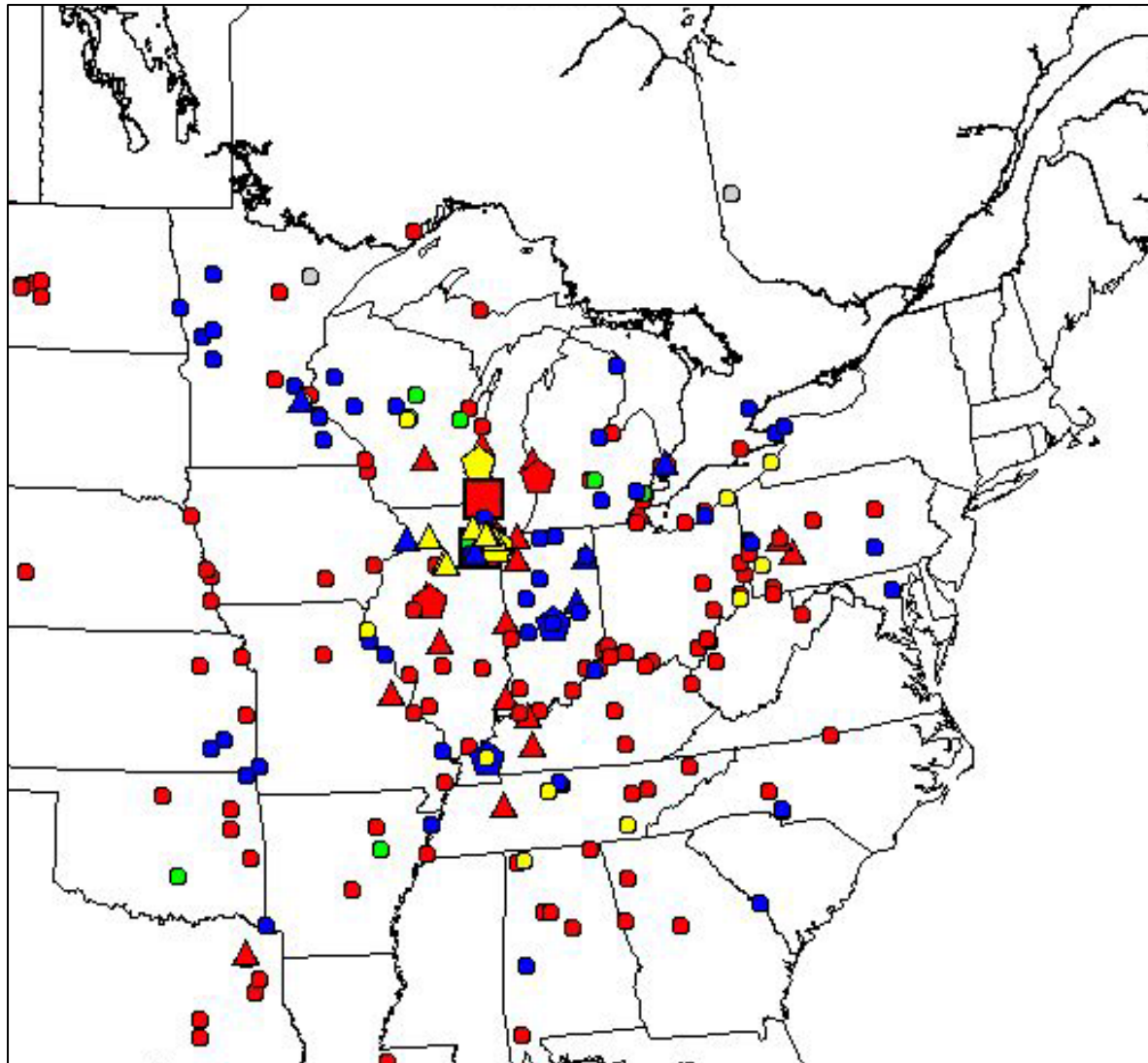
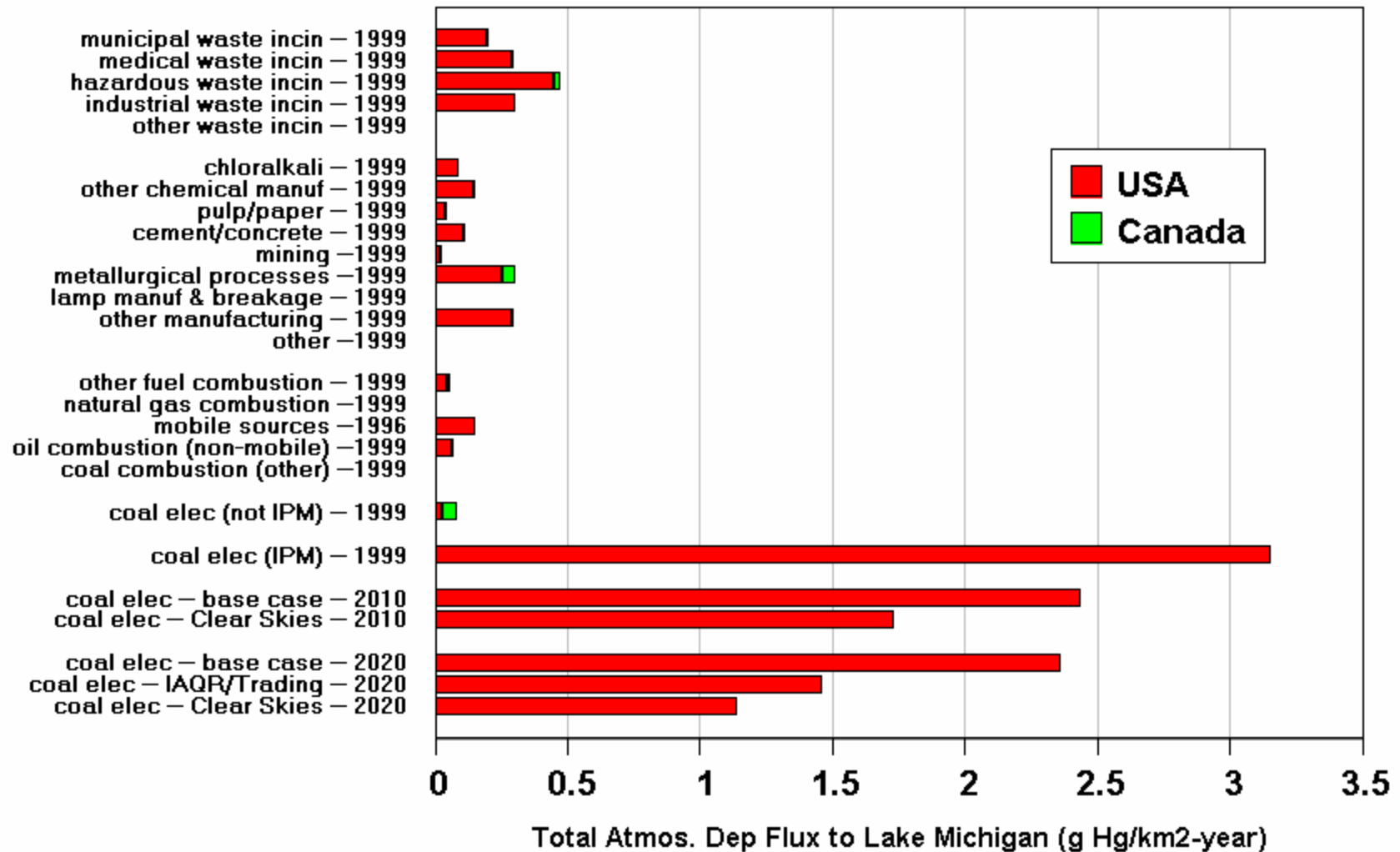


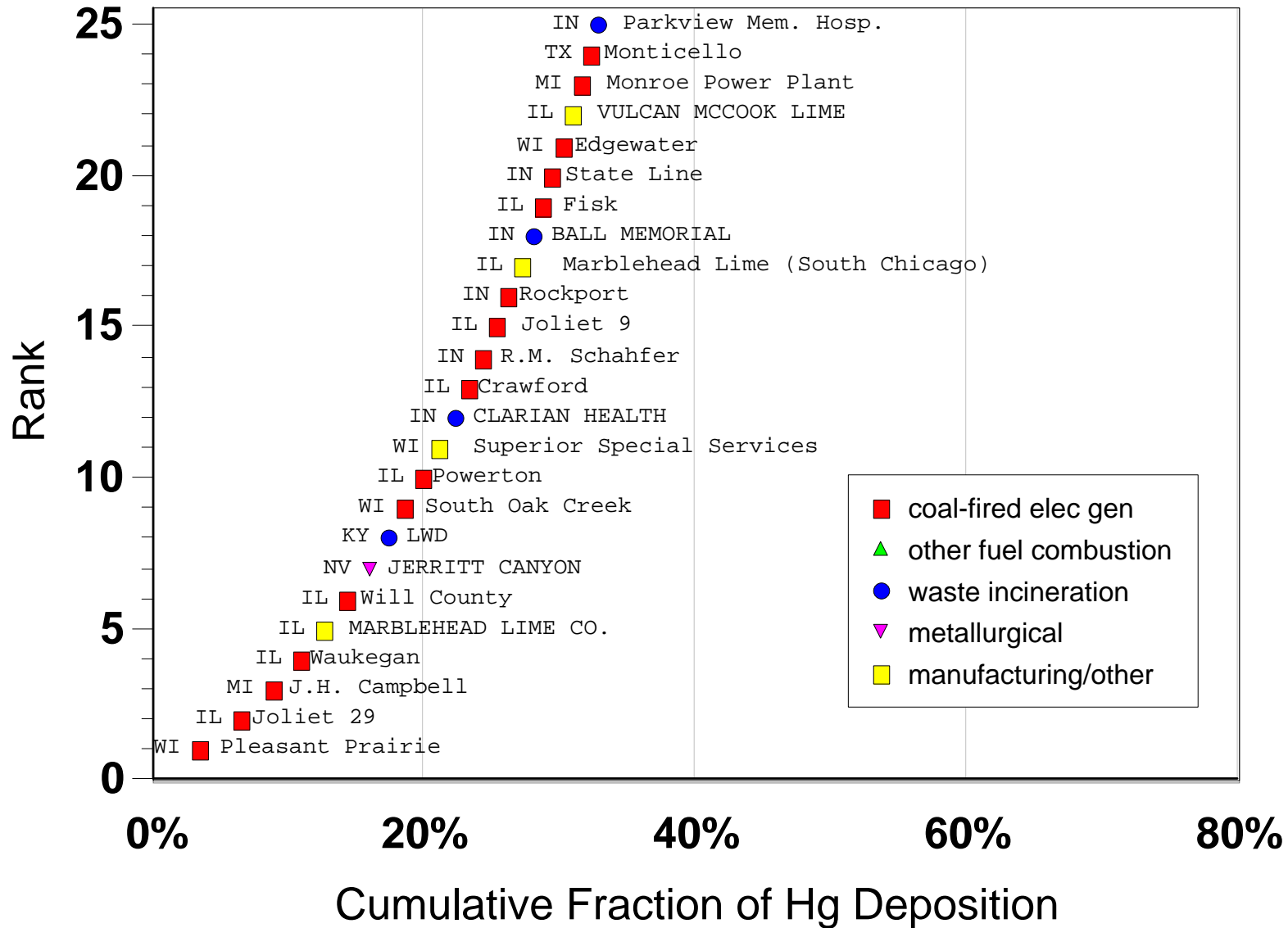
Figure 44. Largest modeled contributors to Lake Michigan (close-up).  
(same legend as previous slide)

# Atmospheric Deposition Flux to Lake Michigan from Anthropogenic Mercury Emissions Sources in the U.S. and Canada

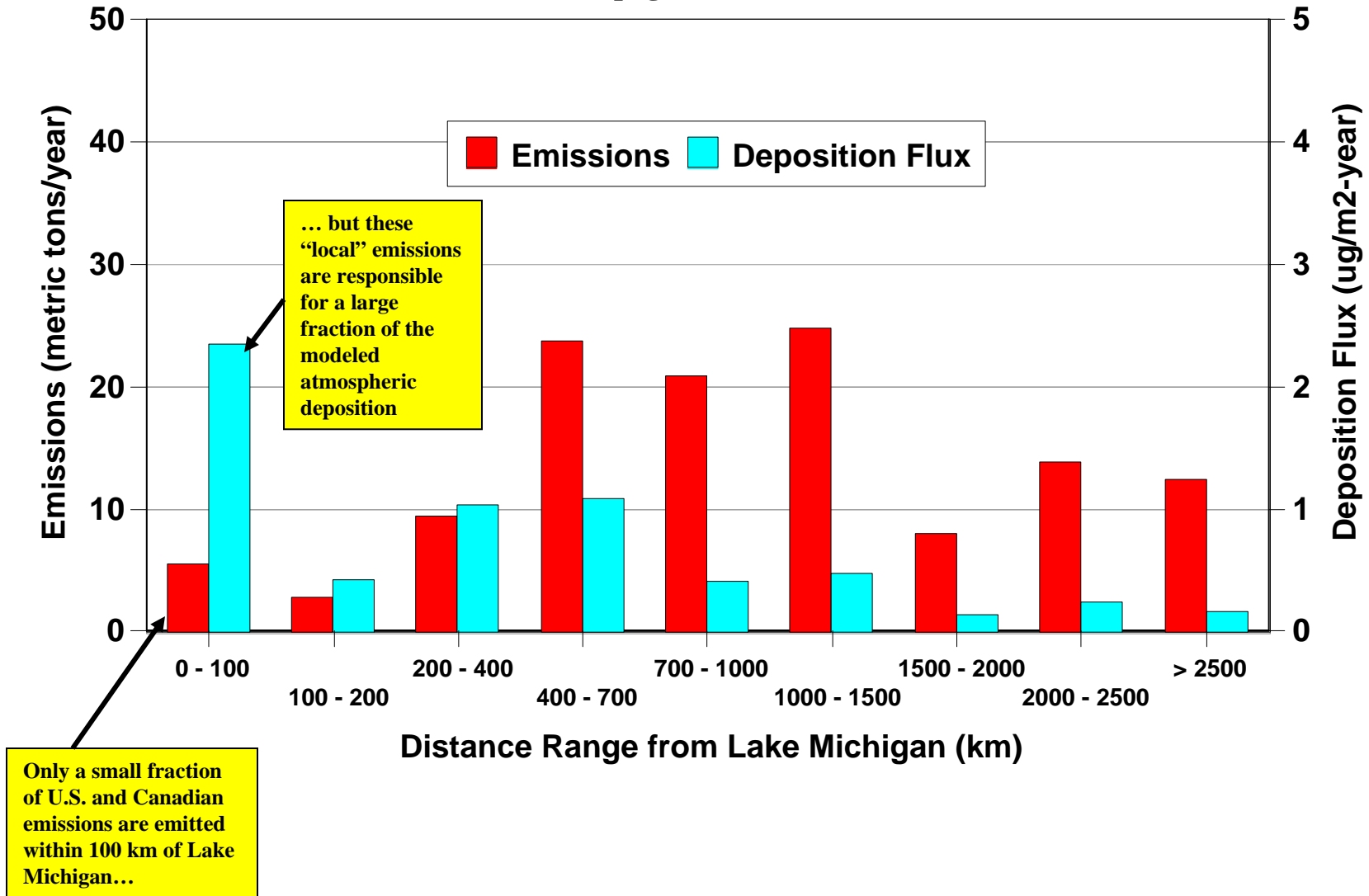




## Top 25 modeled sources of atmospheric mercury to Lake Michigan (based on 1999 anthropogenic emissions in the U.S. and Canada)



## Emissions and deposition to Lake Michigan arising from different distance ranges (based on 1999 anthropogenic emissions in the U.S. and Canada)



**Next Steps:** As resources permit, steps could be taken to refine/extend mercury modeling, e.g.:

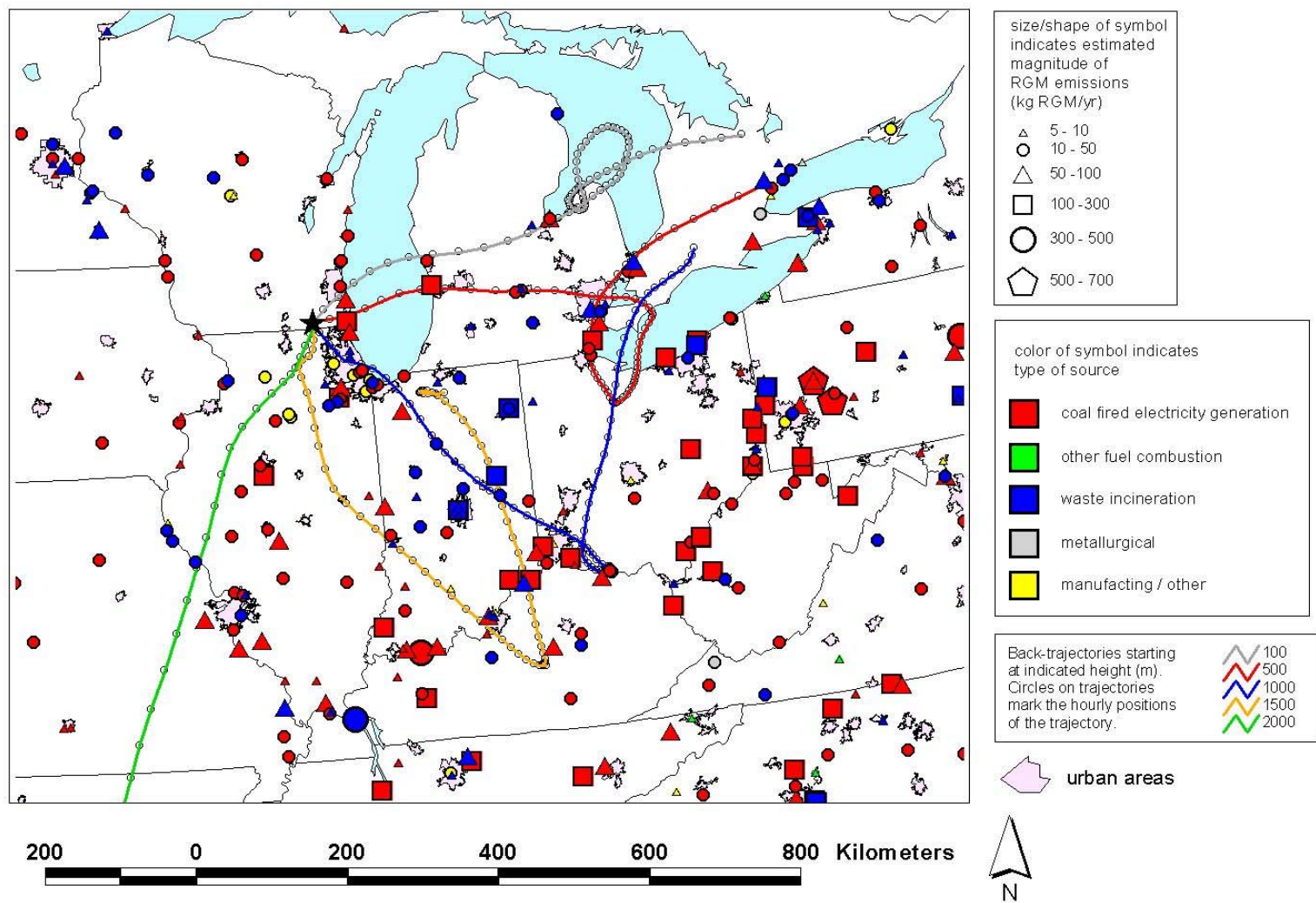
1. Extension of the model from North American domain to simulate impacts of global sources
2. Inclusion of the impact of natural emissions and re-emissions of anthropogenic mercury.
3. The use of more detailed meteorological data (e.g., described on finer spatial scales).
4. Development of a system for incorporating observed precipitation data into the model.
5. Further evaluation of the model against wet deposition data and ambient concentration data for elemental, ionic, and particulate mercury.
6. Process-related measurements of atmospheric chemistry, phase-partitioning behavior, and atmospheric deposition to evaluate and refine model algorithms.
7. Sensitivity tests -- investigating the influence of uncertainties in model inputs and model algorithms -- to help determine which uncertainties are the most critical for model improvement.
8. Linkage of the atmospheric model to other models to form a multi-media mercury modeling system to track mercury from emissions to ecosystem loading to food chain bioaccumulation to human exposure.
9. Use of updated emissions inventories as inputs to the model.
10. Estimation of the time-course of atmospheric loading to the Great Lakes by running the model over long periods using a continuous record of historical emissions.
11. Estimation of the impacts of potential future emissions scenarios.
12. Participation in additional model comparison studies.

## Relative Importance of Anthropogenic vs. Natural Sources?

- Many studies have shown increased amounts of bio-available mercury in ecosystems due to anthropogenic activities (~2x – 5x, sometimes more), but a large number of factors influence the relative increases, e.g., proximity to sources, relative proportions of different forms of mercury emitted from sources, particular biogeochemistry of the ecosystem, etc.

## Relative Importance of Global vs. Domestic Sources?

- NOAA HYSPLIT-Hg work to date has not yet attempted to explicitly answer this question.
  - New work could be done to address this issue.
  - It is noted, however, that in many cases, much of the deposition in U.S. regions with significant sources appears like it can be accounted for by consideration of U.S sources alone.
- A few estimates have been made using other models. Results to date suggest that:
  - There is no “one” answer – the relative importance varies from location to location.
  - In regions with significant sources, the relative importance of global sources appears to be diminished
  - The answer also obviously varies depending on the time period
  - Like with analysis of national sources, the global modeling to date is limited by a number of uncertainties (emissions inventories, atmospheric chemistry, deposition processes) and the evaluation of the models is significantly limited by a lack of observations. Thus, the significance of the uncertainties is not well known.



**Figure 40. WI99, 72-hour back trajectories for high deposition event during the week of 4/18/00 to 4/25/00.**

Butler, T., Likens, G., Cohen, M., Vermeylen, F. (2007). Mercury in the Environment and Patterns of Mercury Deposition from the NADP/MDN Mercury Deposition Network. Final Report to USEPA.

## Summary of Great Lakes Region Trend Information for Atmospheric Mercury Emissions and Deposition

### *Trends in Great Lakes region atmospheric mercury emissions:*

- Data are scarce and uncertain, but it appears that they rose significantly from ~1880 until ~1945, were approximately level from 1945-1970, and decreased between 1970-1980.

### *Trends in U.S. atmospheric mercury emissions from the early 1990's to ~2001:*

- Significant decrease in emissions from municipal and medical waste incinerators, but exact timing of changes at individual facilities poorly characterized.
- Emissions from coal-fired electricity generation and other source categories were relatively constant.

### *Trends in Canadian atmospheric mercury emissions:*

- From 1990-2000, Canadian emissions are reported to have decreased by ~75 percent, largely due to process changes at metal smelting facilities.

### *Trends in mercury wet deposition at monitoring sites in the Great Lakes region:*

- Five long-term Mercury Deposition Network sites, with data beginning in 1996.
- For this report, data for 1996-2003 examined.
- Possible decrease between 2000 and 2001, and this *may* have been related to decreases in regional mercury emissions from waste incinerators.
- There were only moderate changes in estimated ionic mercury emissions in the vicinity of these sites between 1995-1996 and 1999-2001, but the precise timing of these changes is not known. Thus, it is difficult to determine if the trends in precipitation mercury concentrations are related to these reductions.

### *Trends in mercury deposition to the Great Lakes:*

- Trends in model-estimated deposition to the Great Lakes decreased significantly between 1995-1996 and 1999-2001, primarily due to decreases in emissions from U.S. municipal and medical waste incinerators.
- In both periods, the model results suggest that U.S. sources contributed much more to Great Lakes atmospheric mercury deposition than Canadian sources.

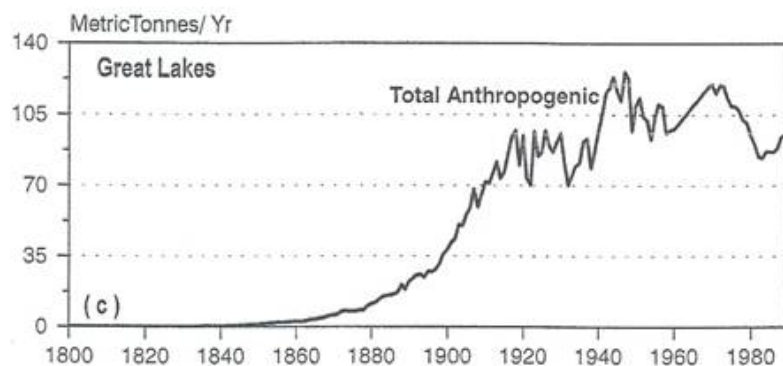
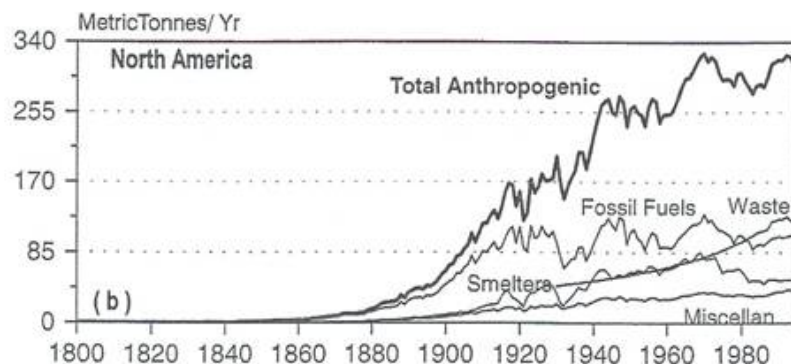
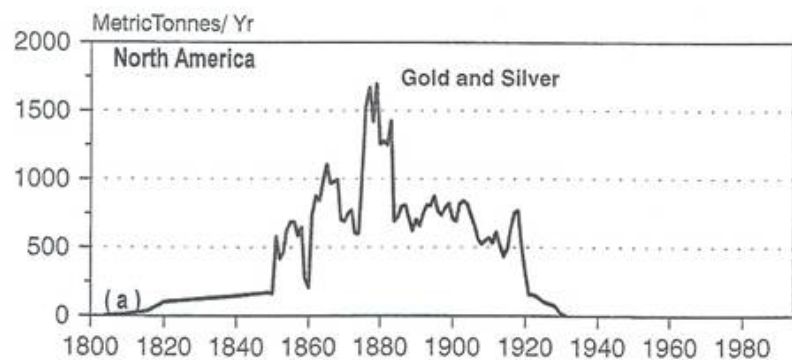
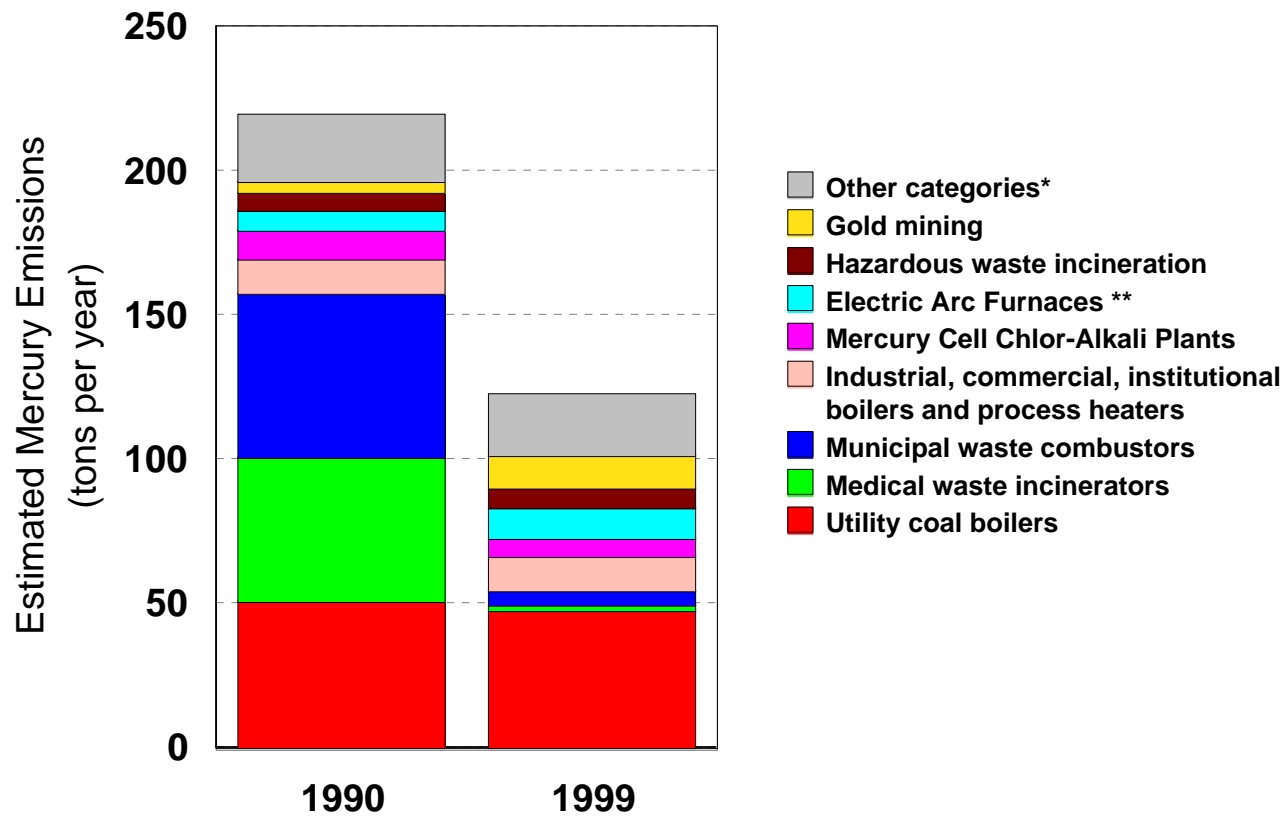


Figure 75. Mercury emissions in North America and the Great Lakes region (1800-1990).

Atmospheric mercury emissions from (a) gold and silver mining in North America; (b) modern anthropogenic sources in North America; and (c) modern anthropogenic sources in the Great Lakes region (including the 8 Great Lakes U.S. states and the province of Ontario). [a reproduction of Figure 2 from Pirrone et al. (1998).]

Pirrone, N., Allegrini, I., Keeler, G., Nriagu, J., Rossman, R., Robbins, J. (1998). Historical atmospheric mercury emissions and depositions in North America compared to mercury accumulations in sedimentary records. *Atmos. Environ.* 32(5): 929-940.

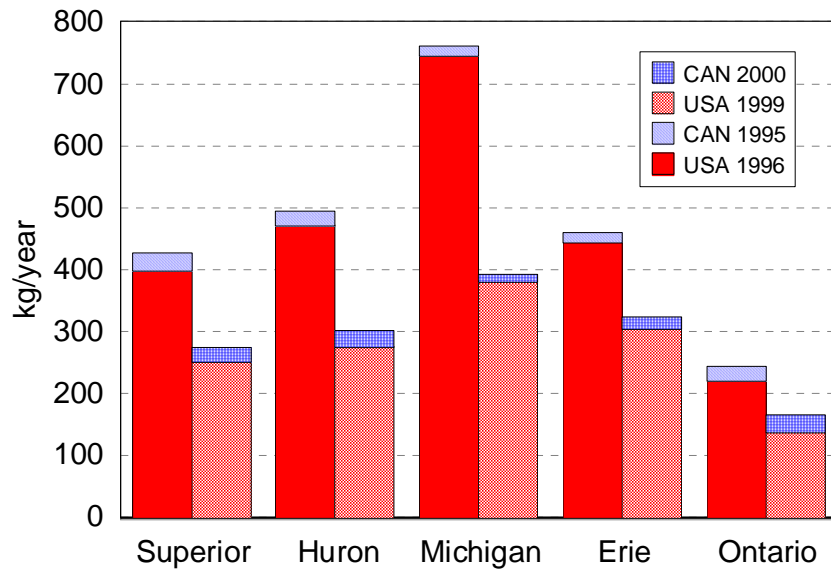


\* Data for Lime Manufacturing are not available for 1990.

\*\* Data for Electric Arc Furnaces are not available for 1999. The 2002 estimate (10.5 tons) is shown here.

Figure 76. Mercury emissions trend data from the U.S. EPA.

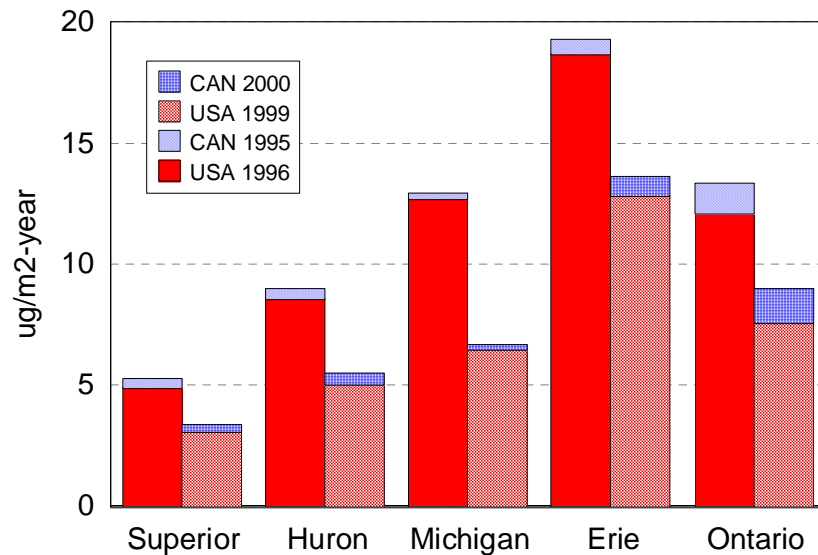




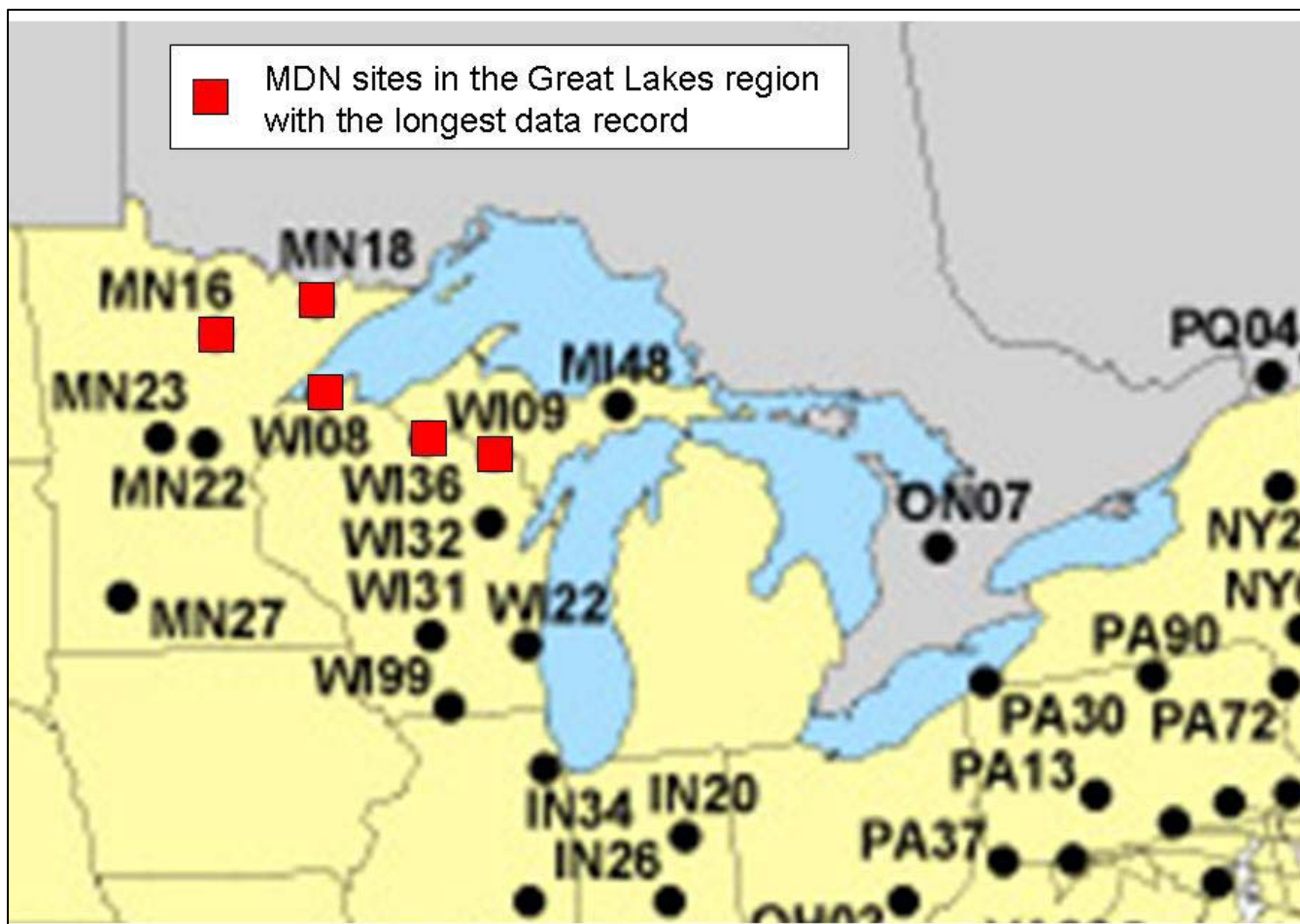
**Modeled mercury deposition (kg/year) to the Great Lakes (1995-1996 vs. 1999-2000), arising from anthropogenic mercury air emissions sources in the U.S. and Canada**

**Model results for atmospheric deposition show that:**

- **U.S. contributes much more than Canada**
- **Significant decrease between 1996 and 1999 (primarily due to decreased emissions from waste incineration)**

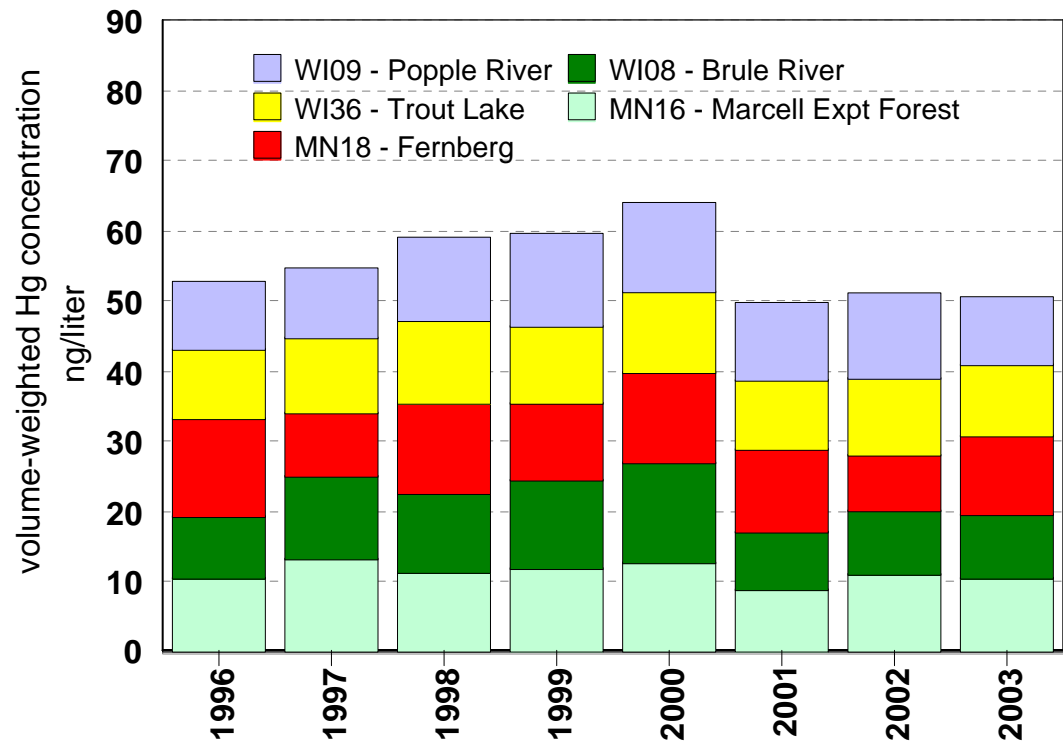
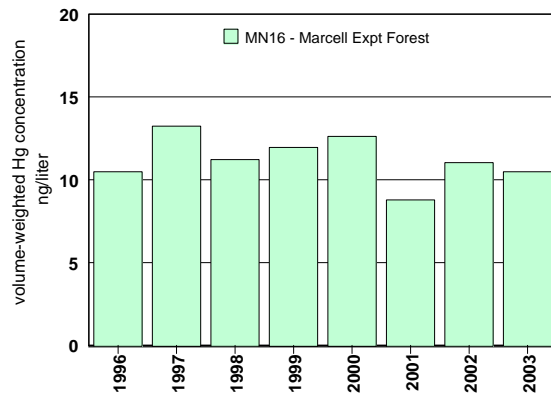
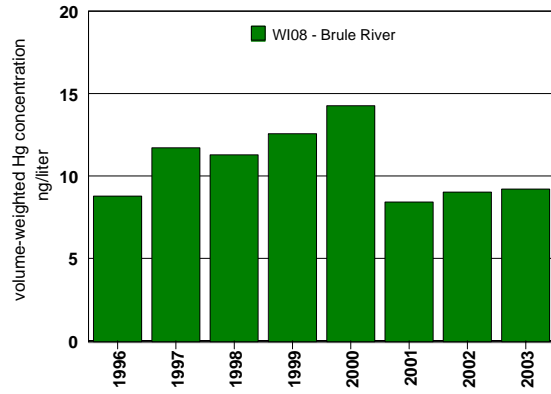
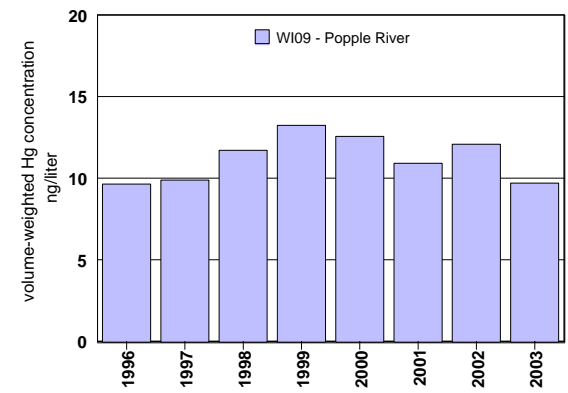
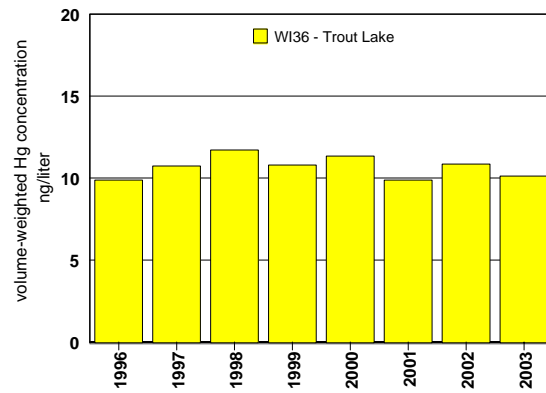
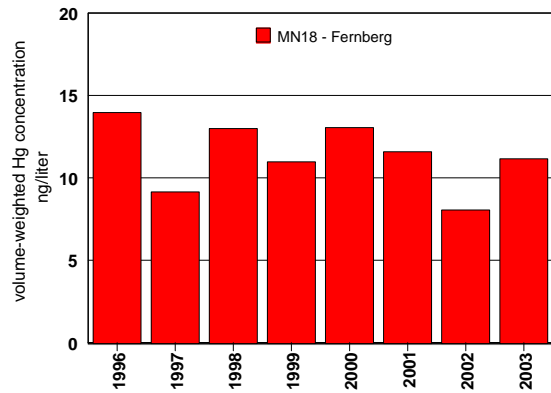


**Modeled mercury flux (ug/m<sup>2</sup>-yr) to the Great Lakes (1995-1996 vs. 1999-2000), arising from anthropogenic mercury air emissions sources in the U.S. and Canada**



**Figure 77. Great Lakes MDN sites with the longest measurement record.**

**Figures 78-83. Mercury concentration in precipitation at long-term MDN sites in the Great Lakes region.**



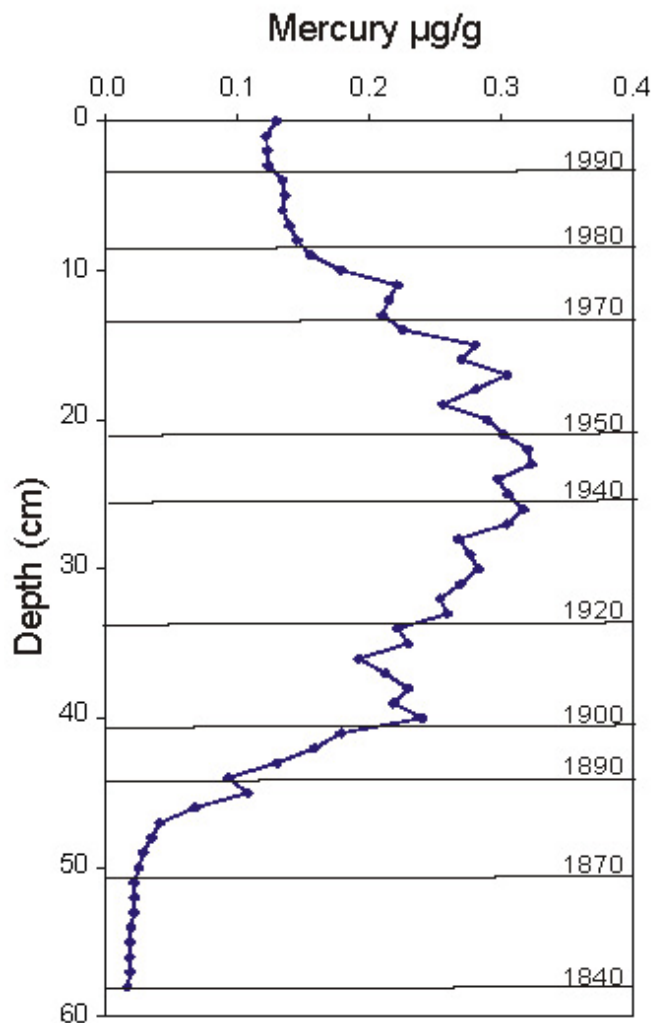
## Summary of Great Lakes Region Trend Information for Sediments and Biota

### *Trends in mercury in Great Lakes sediments:*

- Examples of sediment mercury trend data were found for each of the Great Lakes except for Lake Huron.
- The data typically show a 1940-1960 peak in sediment mercury, and in some cases there are also secondary peaks in the 1970's.
- Since the 1970's sediment mercury concentrations appear to have generally been decreasing in the Great Lakes.

### *Trends in mercury levels in Great Lakes biota:*

- Data on mercury levels in Great Lakes fish and Herring Gull eggs are generally available starting in the 1970's, while data on levels in mussels are available beginning in 1992.
- While there are variations among species and among lakes, the data generally seem to show a reduction from 1970 to the mid-1980's, with little change since the mid-1980's.
- This is most likely due to the significant reduction that occurred in the 1970's in effluent discharges to the Great Lakes (and their tributaries) from a number of sources (e.g., chlor-alkali plants).



**Figure 95. Trend in sediment mercury in Lake Michigan. Profile of total mercury (ug/g dry weight) levels in a core sample from Lake Michigan.**

Source: Marvin, C.; Painter, S., and Rossmann, R. (2004). Spatial and temporal patterns in mercury contamination in sediments of the Laurentian Great Lakes. *Env. Research* 95(3):351-362.

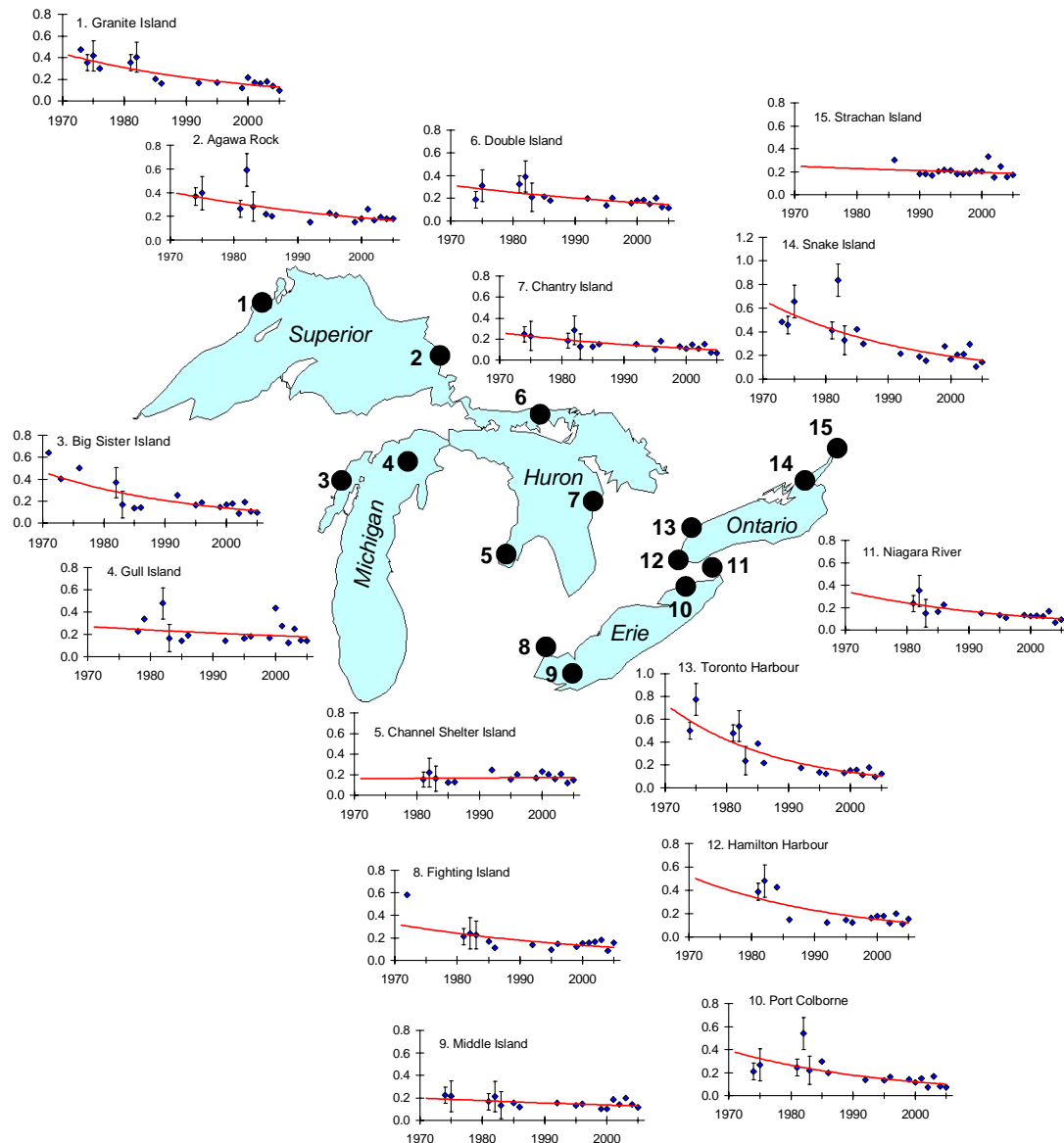
# Figure 106. Trends in Herring Gull Egg Hg concentrations.

Source of data – Canadian Wildlife Service. Total mercury concentrations in eggs from colonies in the Great Lakes region expressed in units of ug Hg/g (wet weight).

From 1971 – 1985, analysis was generally conducted on individual eggs (~10) from a given colony, and the standard deviation in concentrations is shown on the graphs.

From 1986 to the present, analysis was generally conducted on a composite sample for a given colony.

The trend lines shown are for illustration purposes only; they were created by fitting the data to a function of the form  $y = cx^b$ .

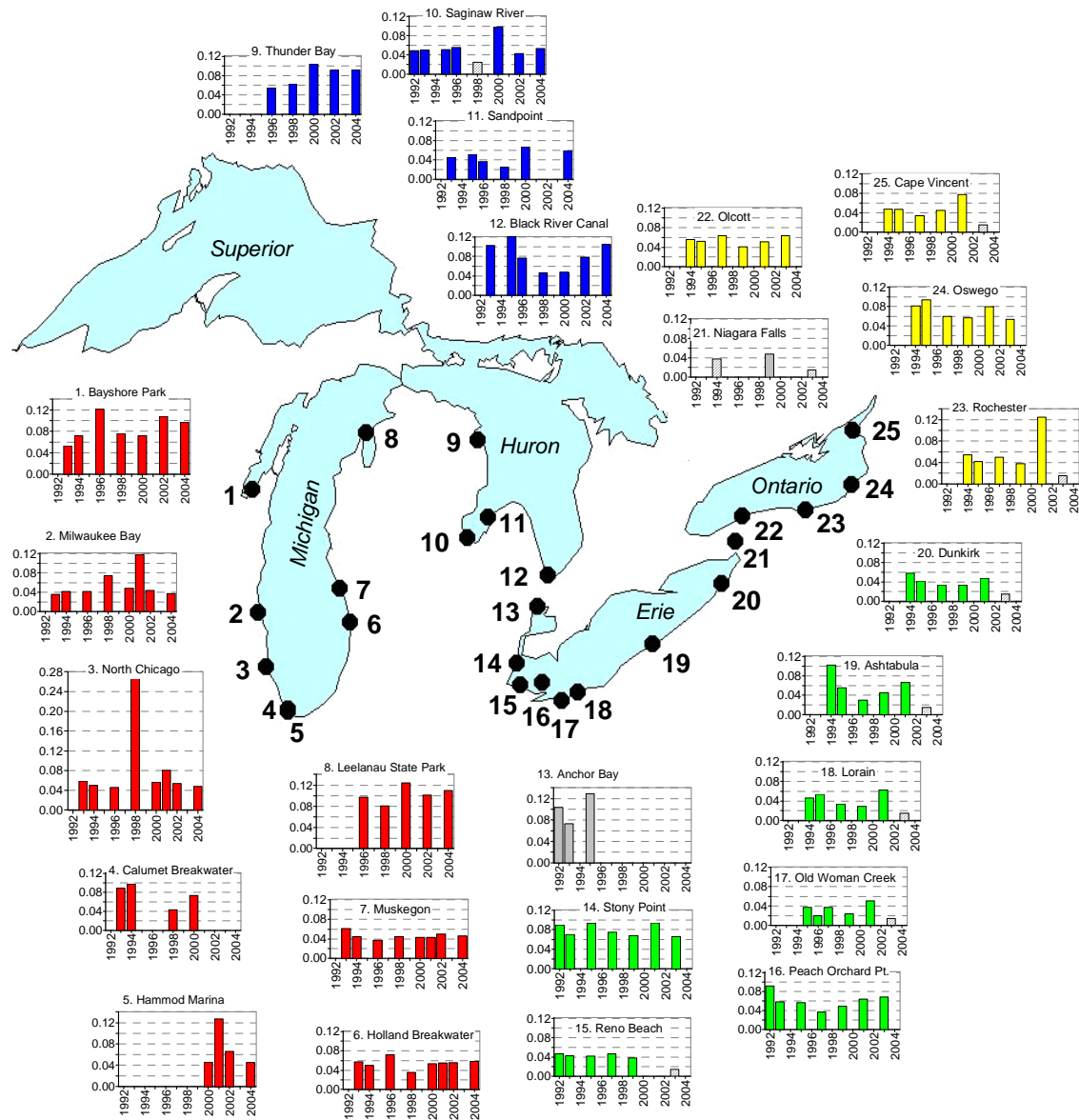


**Figure 107. Mercury concentration in Great Lakes region mussels (1992-2004). Total mercury in mussels (ug/g, on a dry weight basis).**

In a few cases (e.g. for several sites in 2003), mercury concentrations were below the detection limit. In these cases the concentrations are shown with a white cross-hatched bar at a value of one-half the detection limit; in reality, the mercury concentration could have been anywhere between zero and the detection limit.

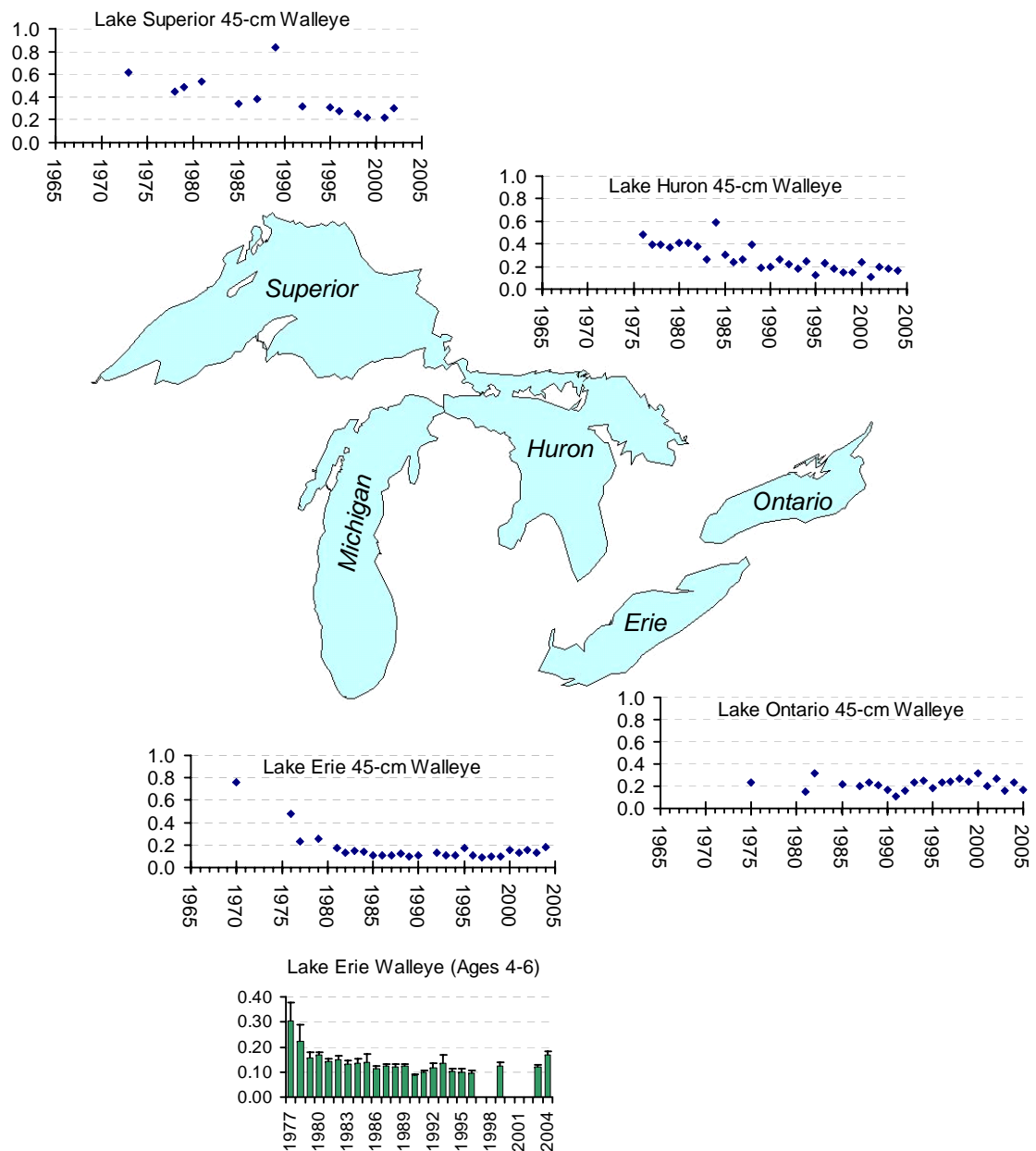
Source of data: NOAA Center for Coastal Monitoring and Assessment (CCMA) (2006) and "Monitoring Data - Mussel Watch" website:

[http://www8.nos.noaa.gov/cit/nsandt/download/mw\\_monitoring.aspx](http://www8.nos.noaa.gov/cit/nsandt/download/mw_monitoring.aspx)



**Figure 101. Mercury concentration trends Great Lakes Walleye. Total mercury concentrations (ppm or ug Hg/g).**

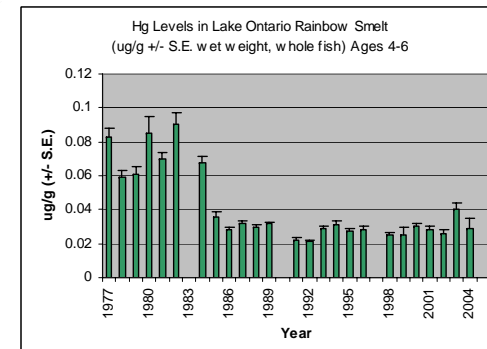
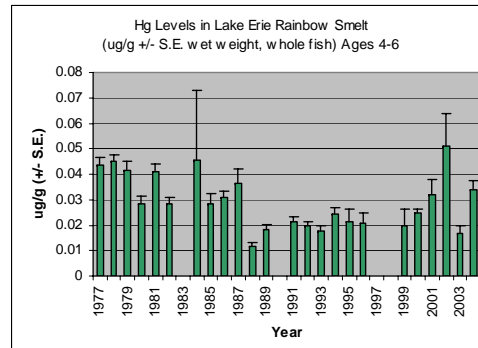
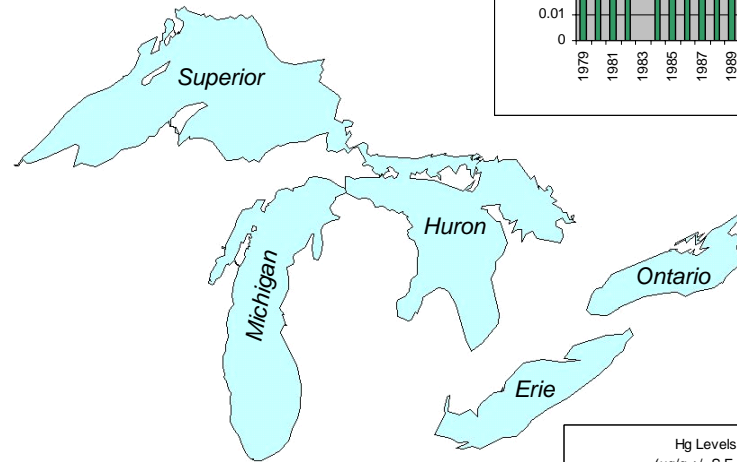
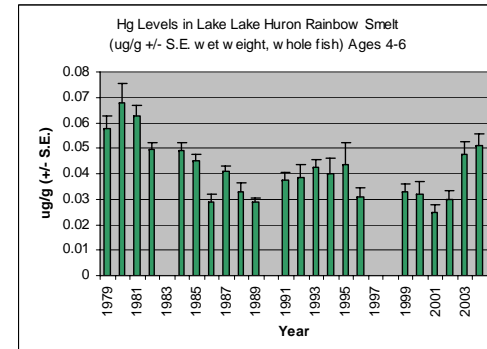
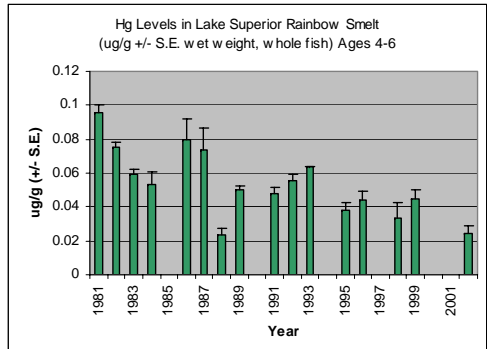
**Sources of data: Ontario Ministry of the Environment (2006b), for 45-cm Walleye data, and Environment Canada (2006), for data on Lake Erie Walleye ages 4-6.**

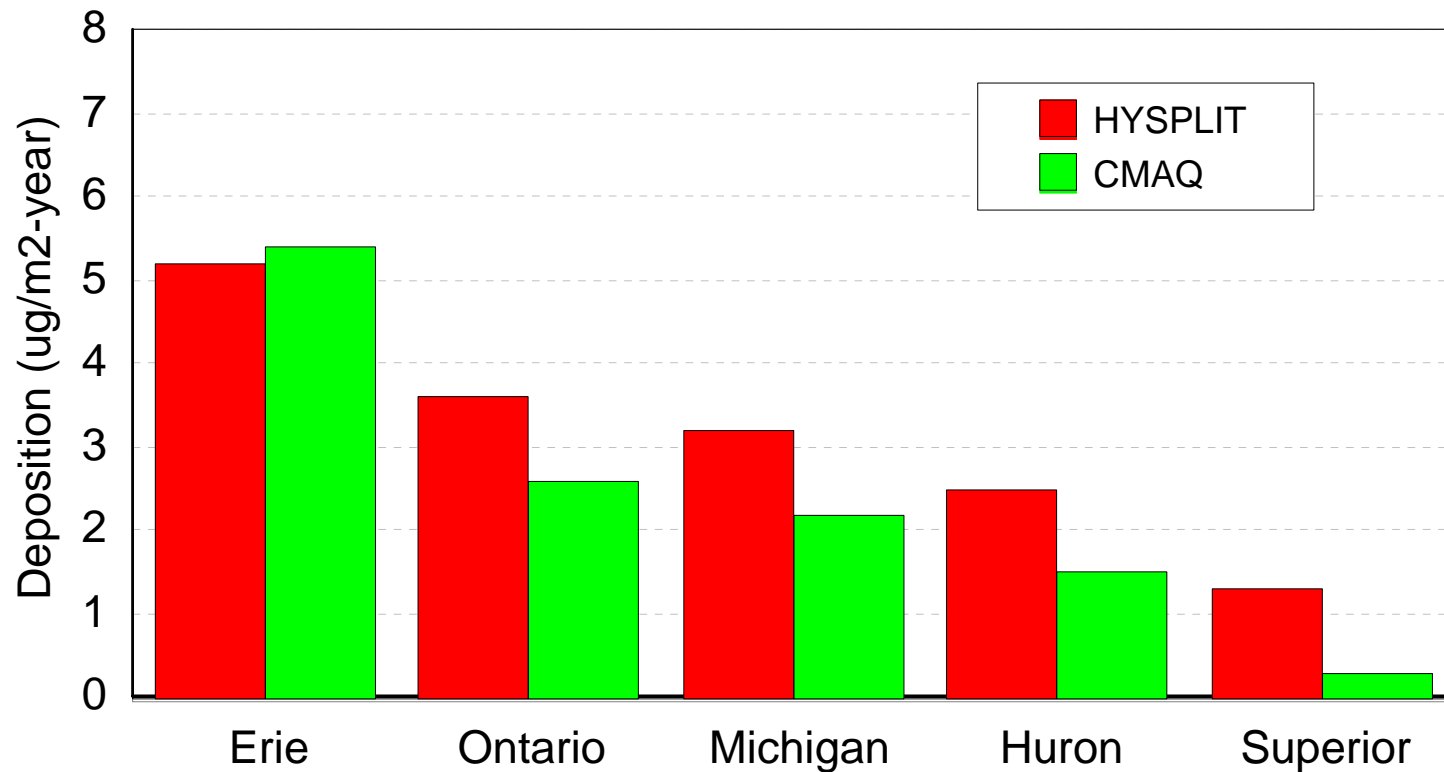




**Figure 102. Total mercury levels in Great Lakes Rainbow Smelt, 1977-2004.**

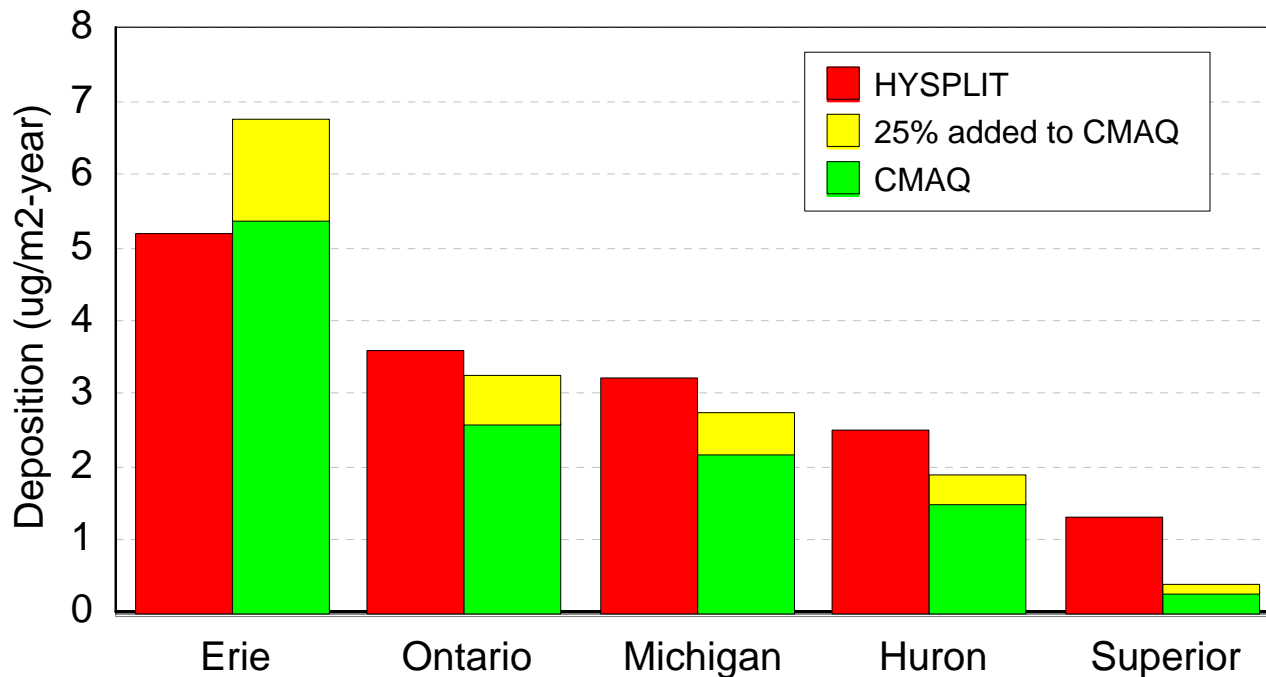
**Source of data: Environment Canada (2006). Note that the scales for the lakes are different.**





**Model-estimated U.S. utility atmospheric mercury deposition contribution to the Great Lakes: HYSPLIT-Hg (1996 meteorology, 1999 emissions) vs. CMAQ-Hg (2001 meteorology, 2001 emissions).**

*Note: Uncertainty estimates for these results could be developed in future work*



- ❑ Model-estimated U.S. utility atmospheric mercury deposition contribution to the Great Lakes: HYSPLIT-Hg (1996 meteorology, 1999 emissions) vs. CMAQ-Hg (2001 meteorology, 2001 emissions).
- ❑ This figure also shows an added component of the CMAQ-Hg estimates -- corresponding to 25% of the CMAQ-Hg results -- in an attempt to adjust the CMAQ-Hg results to account for the deposition underprediction found in the CMAQ-Hg model evaluation.

*Note: Uncertainty estimates for these results could be developed in future work*

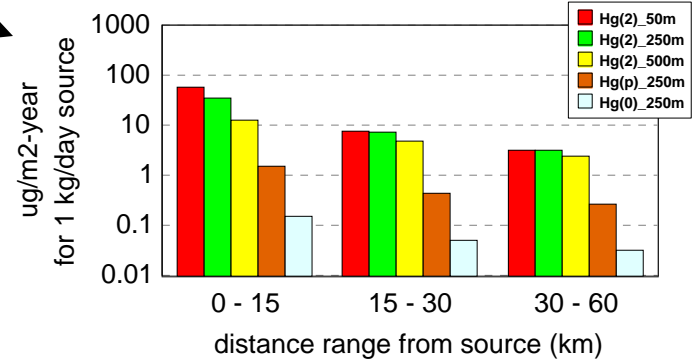
# HYSPLIT 1996



# ISC: 1990-1994

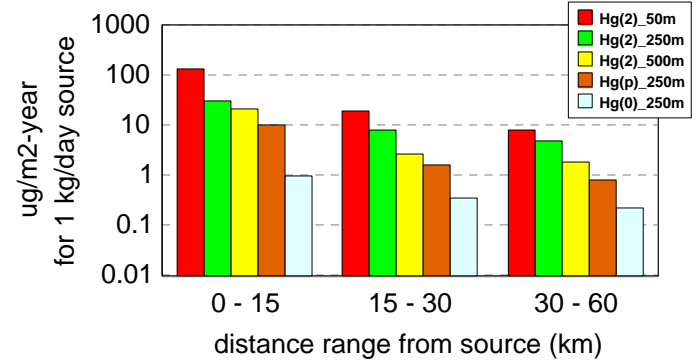


Wet + Dry Deposition: HYSPLIT (Nebraska)  
for emissions of different mercury forms from different stack heights

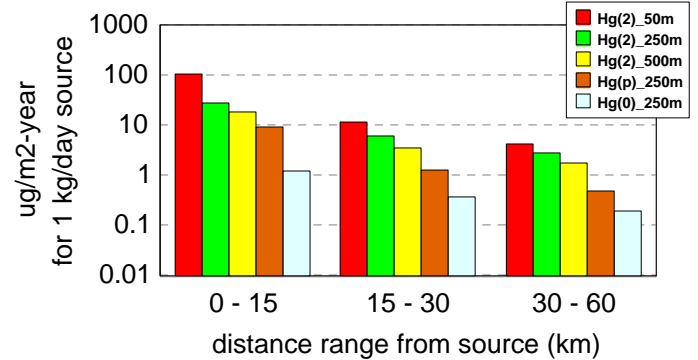


**Different Time Periods and Locations, but Similar Results**

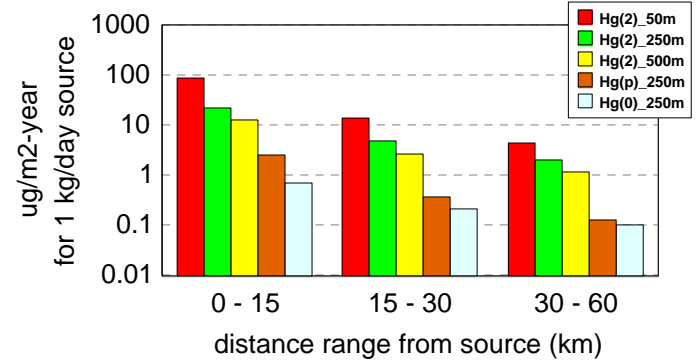
Wet + Dry Deposition: ISC (Kansas City)  
for emissions of different mercury forms from different stack heights



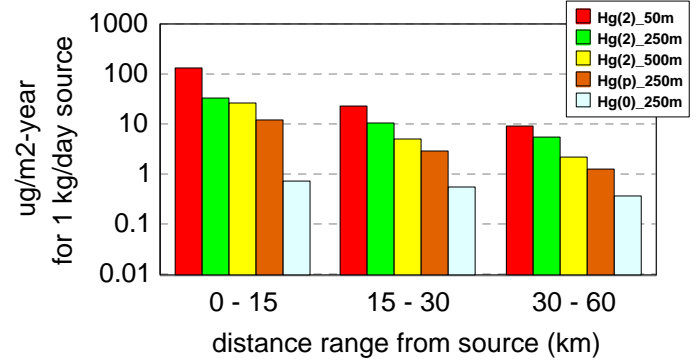
Wet + Dry Deposition: ISC (Tampa)  
for emissions of different mercury forms from different stack heights



Wet + Dry Deposition: ISC (Phoenix)  
for emissions of different mercury forms from different stack heights



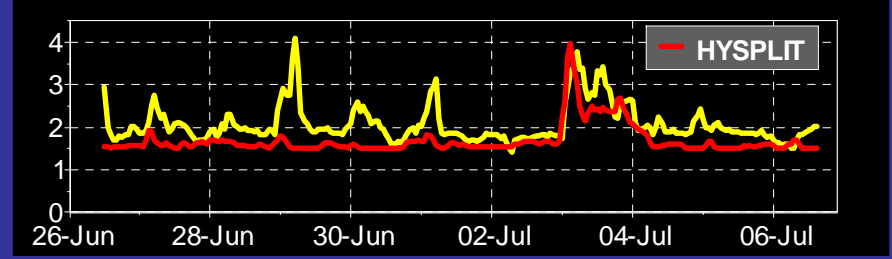
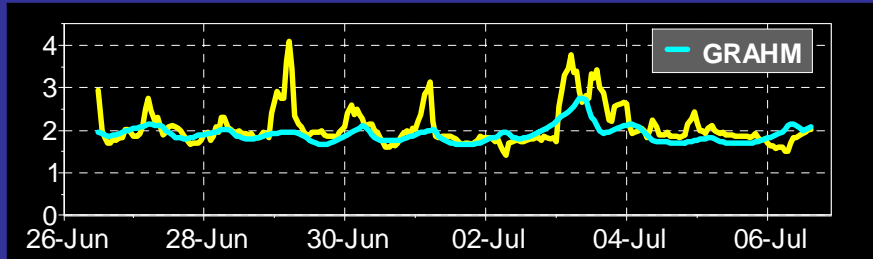
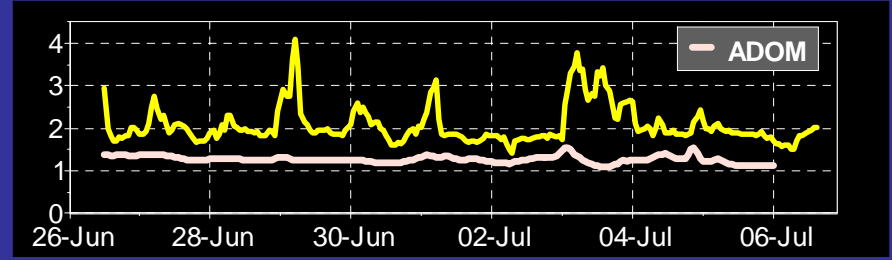
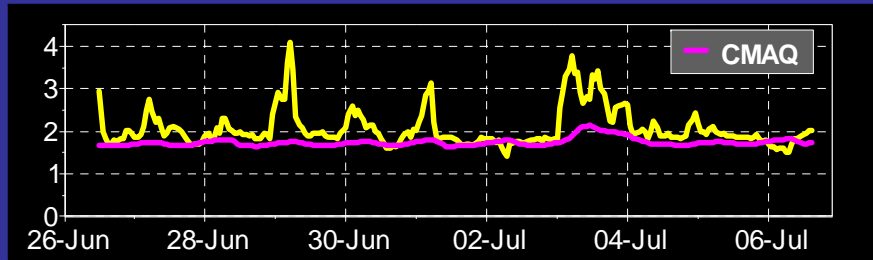
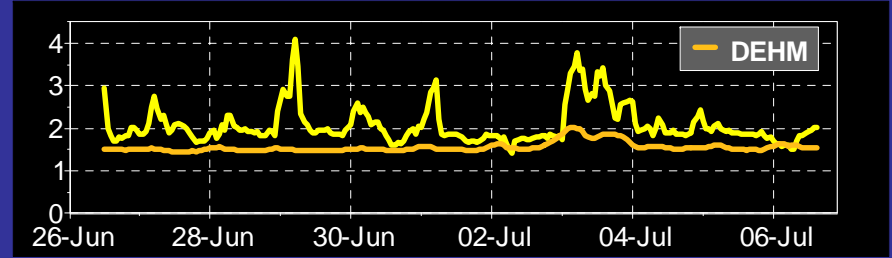
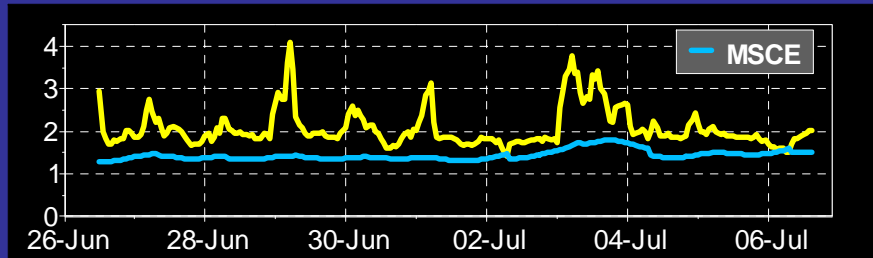
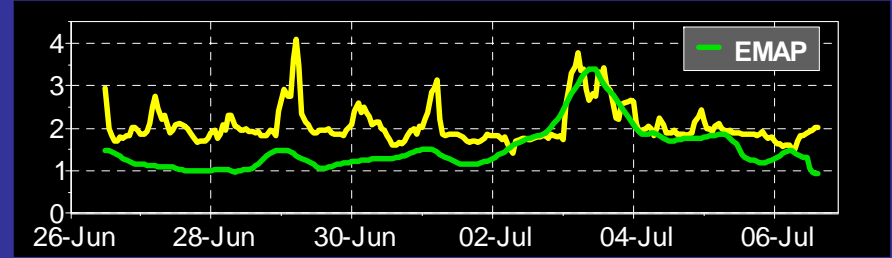
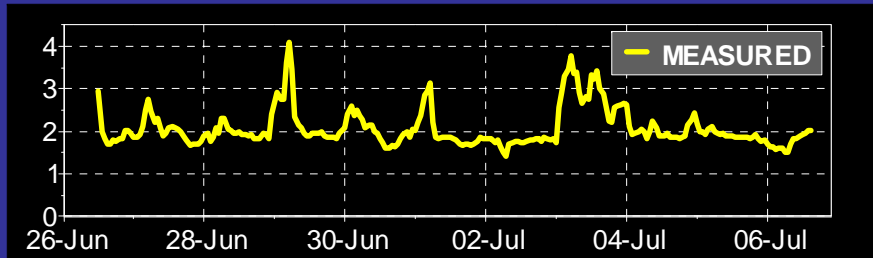
Wet + Dry Deposition: ISC (Indianapolis)  
for emissions of different mercury forms from different stack heights



*EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury*

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg <sup>0</sup>	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

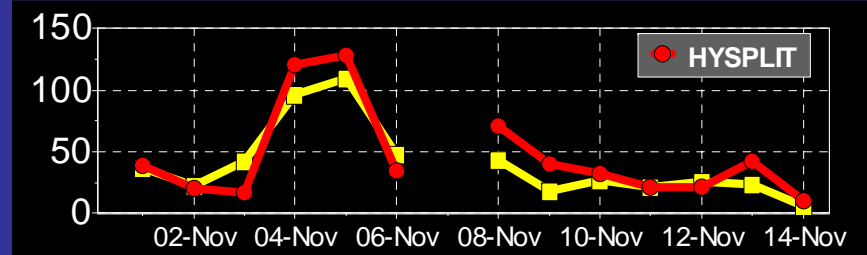
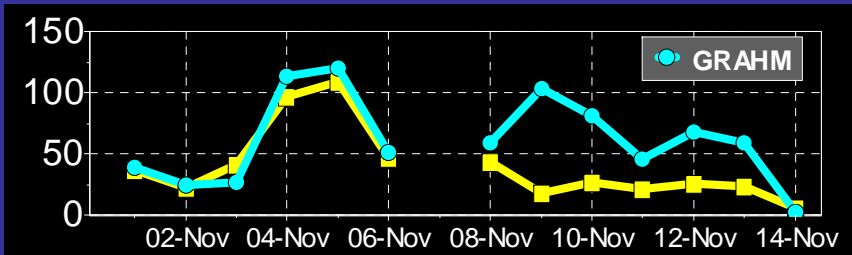
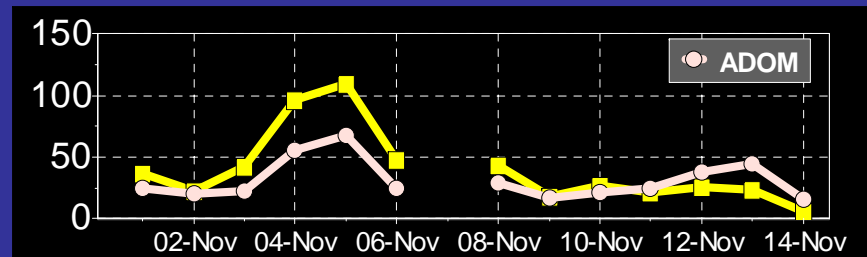
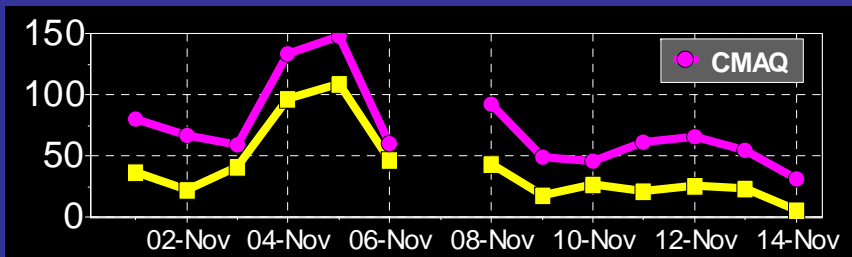
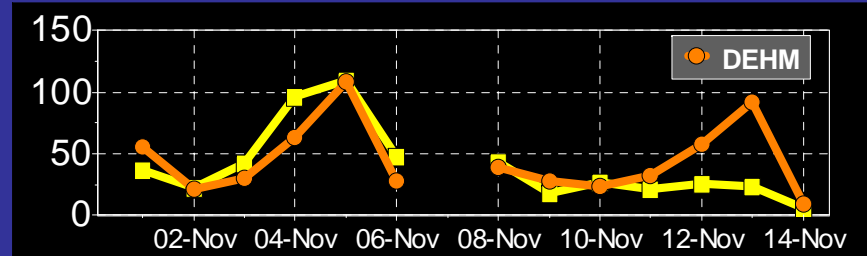
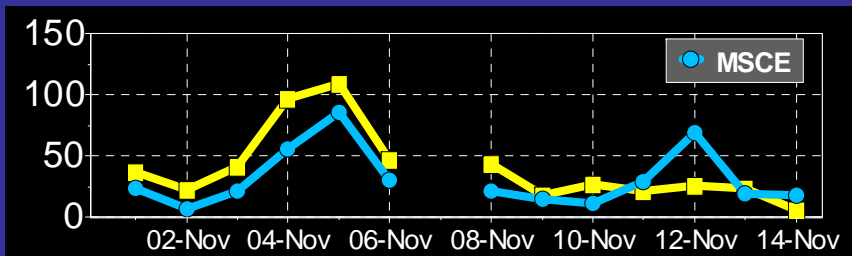
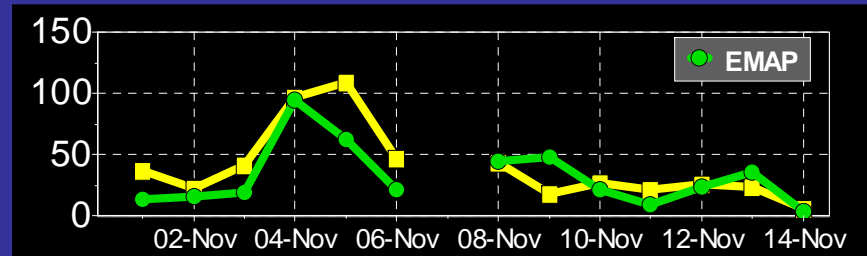
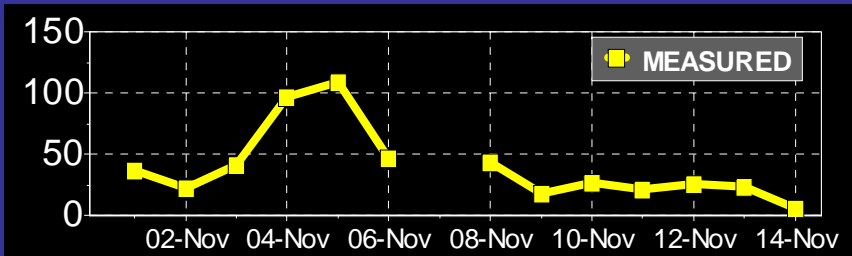
**Total Gaseous Mercury (ng/m<sup>3</sup>) at Neuglobsow: June 26 – July 6, 1995**



*EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury*

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg <sup>0</sup>	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

**Total *Particulate* Mercury (pg/m<sup>3</sup>) at Neuglobsow, Nov 1-14, 1999**

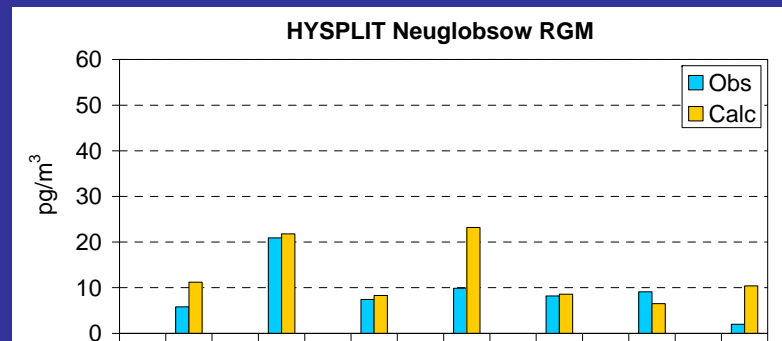
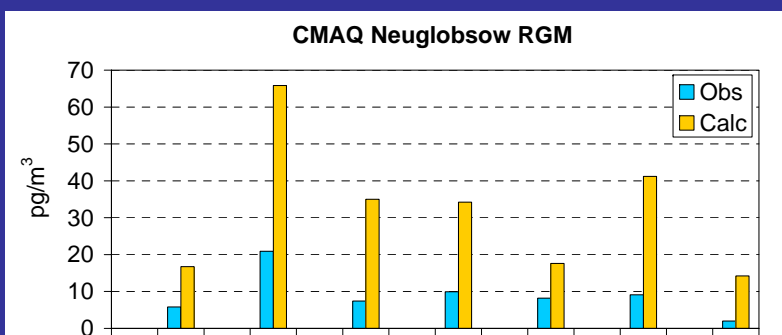
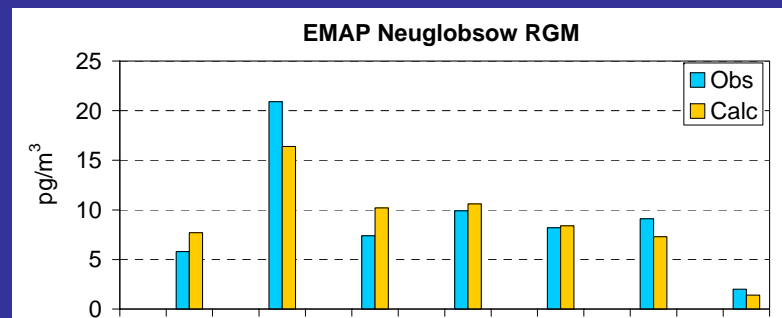
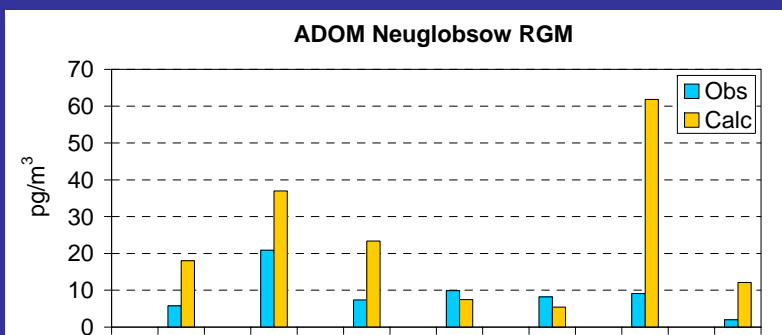
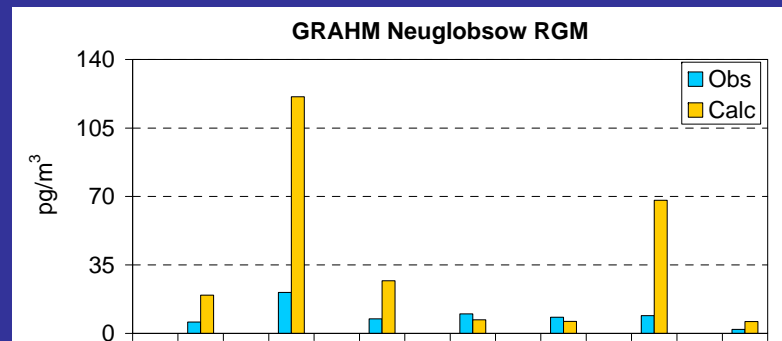
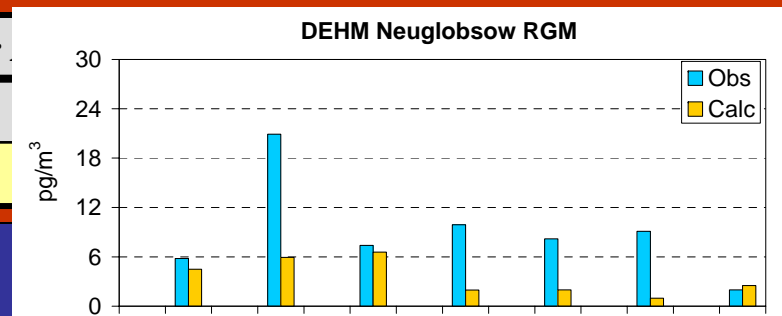
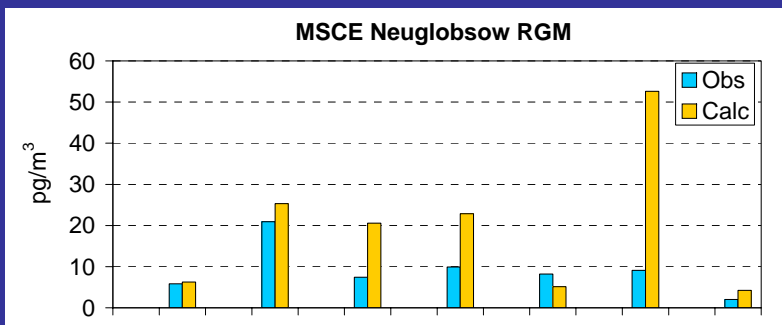


*EMEP Intercomparison Study of Numerical Models for*

Intro- duction	Stage I	Stage II		
	Chemistry	Hg <sup>0</sup>	Hg(p)	RGM

conclu-  
sions

**Reactive Gaseous Mercury at Neuglobsow, Nov 1-14, 1999**





## Intercomparison study of atmospheric mercury models: 1. Comparison of models with short-term measurements

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Received 21 June 2006; received in revised form 29 December 2006; accepted 10 January 2007

Available online 26 February 2007

### Abstract

Five regional scale models with a horizontal domain covering the European continent and its surrounding seas, one hemispheric and one global scale model participated in an atmospheric mercury modelling intercomparison study. Model-predicted concentrations in ambient air were compared against mercury species observed at four monitoring stations in Central and Northern Europe and a station on the Irish west coast. The modelled concentrations of total particulate mercury (TPM) were generally consistent with the measurements at all sites. The models exhibited significant ability to simulate concentrations of gaseous elemental mercury (GEM), but some of the short-duration peaks at the Central European stations could not be consistently reproduced. Possible reasons for these discrepancies include (1) errors in the anthropogenic emissions inventory utilized; (2) coarse spatial resolution of the models; and (3) uncertainty of natural and re-emitted mercury sources. The largest discrepancies between measurements and modelled concentrations were found for reactive gaseous mercury (RGM). For these models, the uncertainty in predicting short-term (two-week episode) variations of mercury species in air can be characterized by the following overall statistics: 90% of the results for TGM are within a factor of 1.35 of the measurements; for TPM, 90% are within a factor of 2.5; and for RGM, 90% are within a factor of 10.  
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**Keywords:** Mercury species; Atmospheric concentrations; Atmospheric transport; Numerical modelling; Model intercomparison

### 1. Introduction

The Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) was established by the

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doi:10.1016/j.scitotenv.2007.01.072

Ryaboshapko, A., *et al.*  
(2007). Intercomparison  
study of atmospheric  
mercury models: 1.  
Comparison of models  
with short-term  
measurements. *Science  
of the Total Environment*  
**376**: 228–240.




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Science of the Total Environment xx (2007) xxx–xxx

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 Science of the Total Environment  
An International Journal for Research, Reviews, and the Environment and its Relationship with Human Health  
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## Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets

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Ashu Dastoor<sup>e</sup>, Ilia Ilyin<sup>a</sup>, Gerhard Petersen<sup>f</sup>, Dimiter Syrakov<sup>g</sup>, Oleg Travnikov<sup>a,\*</sup>,  
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Received 21 June 2006; received in revised form 29 December 2006; accepted 10 January 2007

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**Abstract**

Five regional scale models with a horizontal domain covering the European continent and its surrounding seas, two hemispheric and one global scale model participated in the atmospheric Hg modelling intercomparison study. The models were compared between each other and with available measurements from 11 monitoring stations of the EMEP measurement network. Because only a very limited number of long-term measurement records of Hg were available, significant attention was given to the intercomparison of modelling results. Monthly and annually averaged values of Hg concentrations and depositions as well as items of the Hg deposition budgets for individual European countries were compared. The models demonstrated good agreement (within  $\pm 20\%$ ) between annual modelled and observed values of gaseous elemental Hg. Modelled values of Hg wet deposition in Western and Central Europe agreed with the observations within  $\pm 45\%$ . The probability to predict wet depositions within a factor of 2 with regard to measurements was 50–70% for all the models. The scattering of modelling results for dry depositions of Hg was more significant (up to  $\pm 50\%$  at the annual scale and even higher for monthly data). Contribution of dry deposition to the total Hg deposition was estimated at 20–30% with elevated dry deposition fluxes during summer time. The participating models agree in their predictions of transboundary pollution for individual countries within  $\pm 60\%$  at the monthly scale and within  $\pm 30\%$  at the annual scale. For the cases investigated, all the models predict that the major part of national anthropogenic Hg emissions is transported outside the country territory.  
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**Keywords:** Atmospheric mercury; Numerical modelling; Model intercomparison; Transport and deposition; Transboundary pollution; Uncertainty

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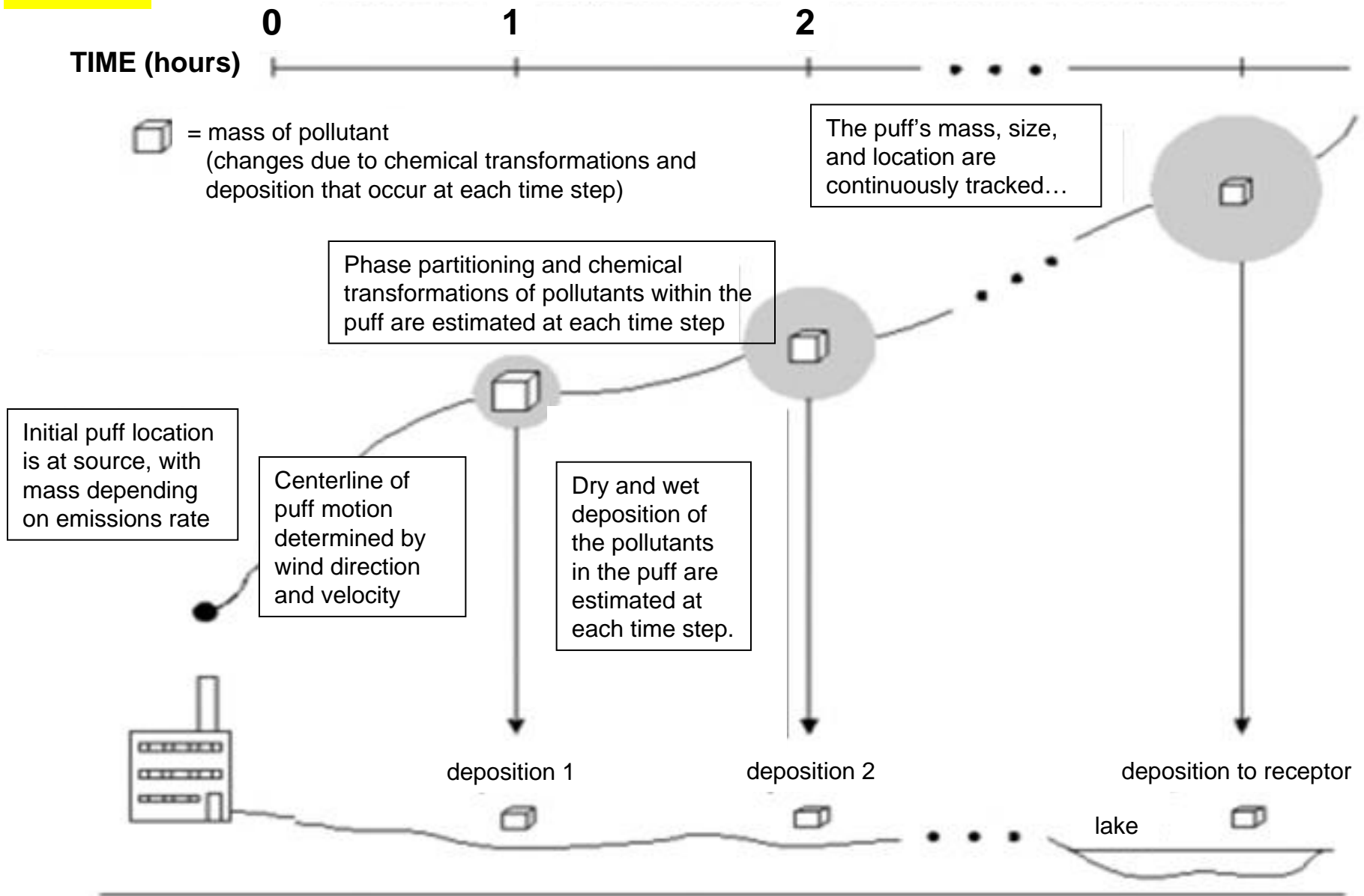
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 doi:10.1016/j.scitotenv.2007.01.071

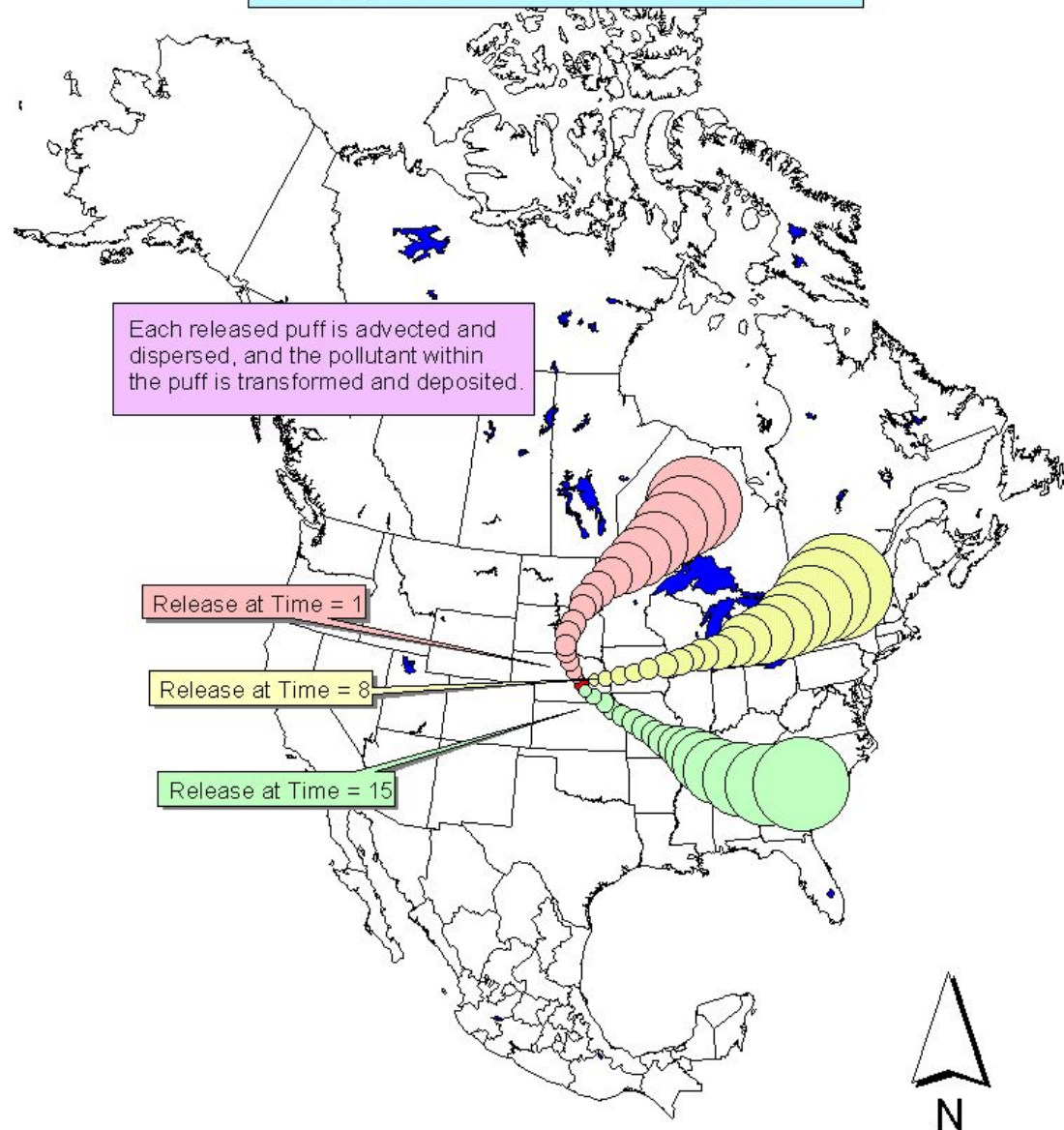
Please cite this article as: Ryaboshapko A et al. Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets. *Sci Total Environ* (2007), doi:10.1016/j.scitotenv.2007.01.071

Ryaboshapko, A., *et al.* (2007). Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets. *Science of the Total Environment* **377**: 319-333.

# Lagrangian Puff Atmospheric Fate and Transport Model



Over the entire modeling period (e.g., one year), puffs are released at periodic intervals (e.g., once every 7 hours).





## Modeling the atmospheric transport and deposition of mercury to the Great Lakes<sup>☆</sup>

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Received 28 August 2003; received in revised form 12 November 2003; accepted 19 November 2003

### Abstract

A special version of the NOAA HYSPLIT4 model has been developed and used to estimate the atmospheric fate and transport of mercury in a North American modeling domain. Spatial and chemical interpolation procedures were used to expand the modeling results and provide estimates of the contribution of each source in a 1996 anthropogenic US/Canadian emissions inventory to atmospheric mercury deposition to the Great Lakes. While there are uncertainties in the emissions inventories and ambient data suitable for model evaluation are scarce, model results were found to be reasonably consistent with wet deposition measurements in the Great Lakes region and with independent measurement-based estimates of deposition to Lake Michigan. Sources up to 2000 km from the Great Lakes contributed significant amounts of mercury through atmospheric transport and deposition. While there were significant contributions from incineration and metallurgical sources, coal combustion was generally found to be the largest contributor to atmospheric mercury deposition to the Great Lakes.

Published by Elsevier Inc.

**Keywords:** Mercury; Atmospheric deposition; Great Lakes; Source-receptor modeling; Emissions

Mercury contamination in the Great Lakes and many other ecosystems is increasingly being recognized as a serious environmental concern. The dominant route of human exposure to mercury is through fish consumption, and significant portions of the general population are believed to be consuming toxicologically significant levels of mercury (e.g., National Research Council, 2000). Historical discharges—e.g., from chlor-alkali production using the mercury-cell process—are believed to have caused large accumulations of mercury in

sediments in Lake Erie and Lake Ontario (Marvin et al., 2003). As these discharges have been substantially reduced, atmospheric deposition is now believed to be a more significant loading pathway for these lakes. Mass balance calculations for Lake Michigan (Mason and Sullivan, 1997) and Lake Superior (Dolan et al., 1993) indicate that atmospheric deposition accounts for approximately 75% of the overall mercury loading to these lakes.

While there have been several mercury modeling efforts in North America (Ballock et al., 1998; Ballock and Behne, 2002; Dvonch et al., 1998; Lin et al., 2001; Pai et al., 1997; Seigneur et al., 2000, 2001, 2003a, b; Shannon and Voldner, 1995; Xu et al., 2000a-c), none has developed detailed source-receptor relationships for the Great Lakes, as advocated in Annex 15 of the Great

<sup>☆</sup>Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.envres.2003.11.007

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Cohen et al (2004). “Modeling the Atmospheric Transport and Deposition of Mercury to the Great Lakes.” *Environmental Research* 95(3), 247-265.

Note: Volume 95(3) is a Special Issue: "An Ecosystem Approach to Health Effects of Mercury in the St. Lawrence Great Lakes", edited by David Carpenter.