Atmospheric Mercury Modeling at the NOAA Air Resources Laboratory using the HYSPLIT-Hg Model

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Presentation at the
Gulf Coast Mercury Research Collaboration Meeting
Gulf Power Building, Pensacola FL, May 18-19, 2006

Outline of Presentation

- **modeling methodology**
- **some preliminary results for Mobile Bay**
  (based on this methodology)
- **model intercomparisons**
- **summary of previous work; current goals; challenges**

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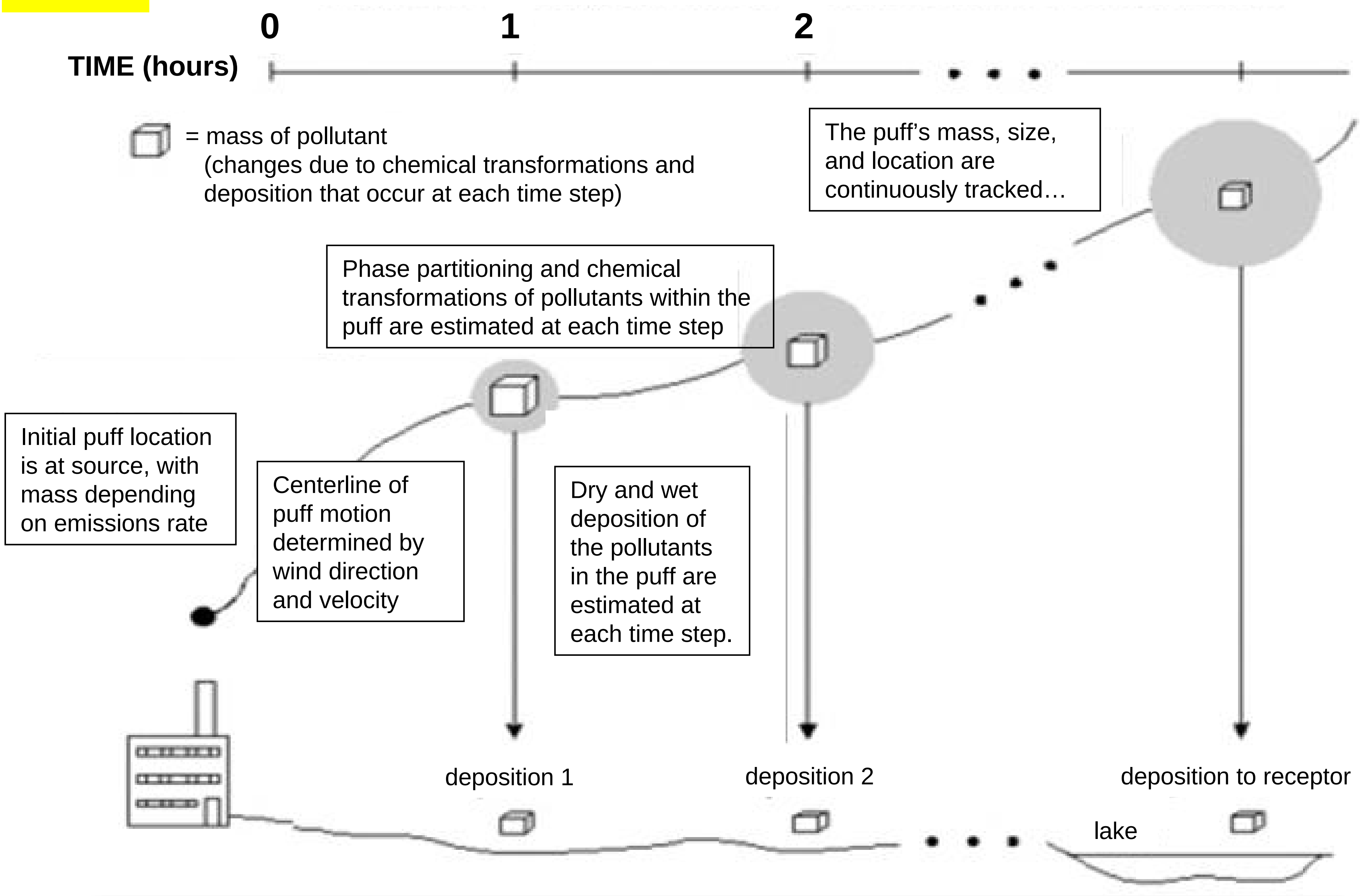
Modeling Methodology

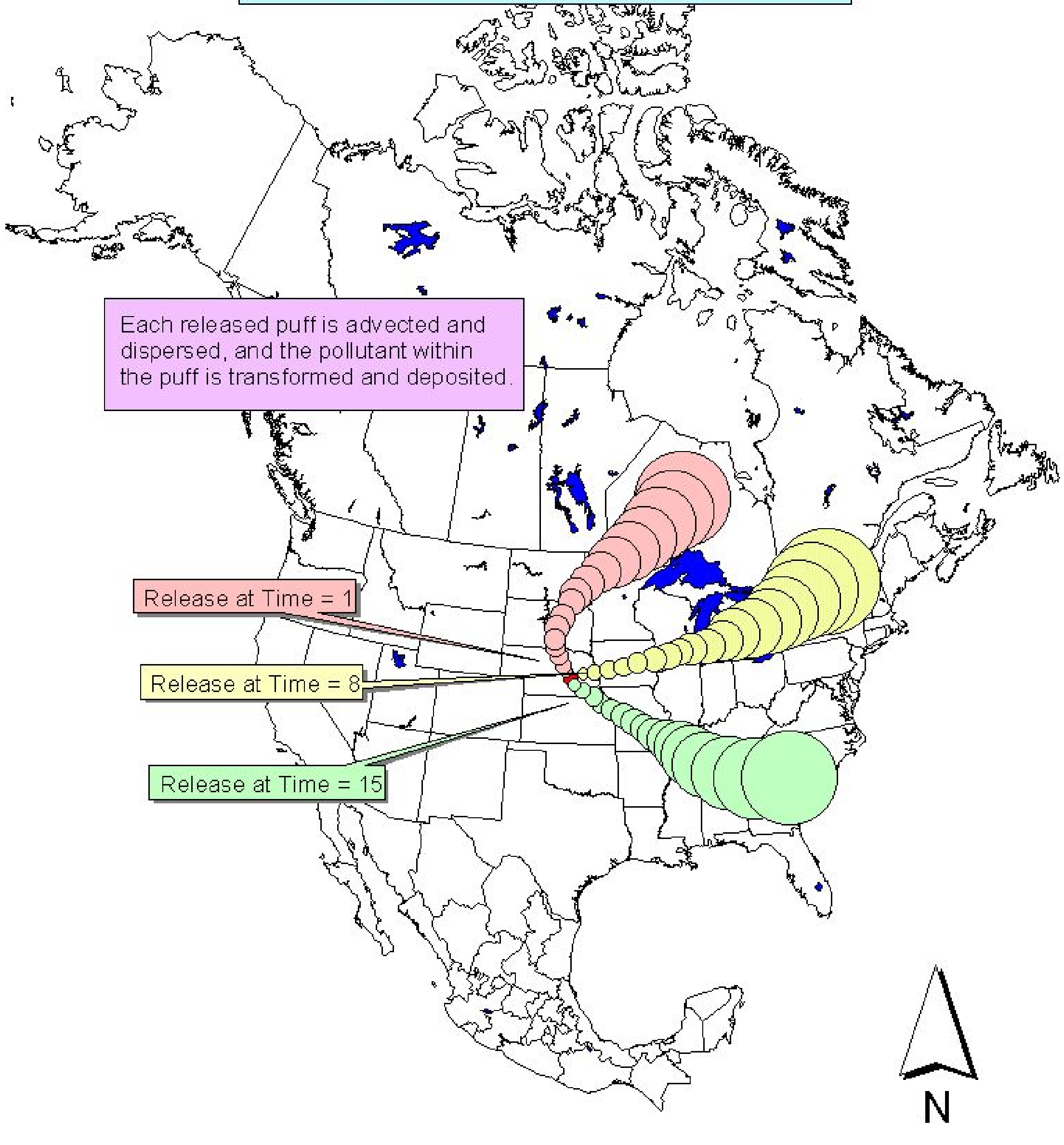
- **NOAA HYSPLIT model → HYSPLIT-Hg**

NOAA
HYSPLIT
MODEL

Lagrangian Puff Atmospheric Fate and Transport Model



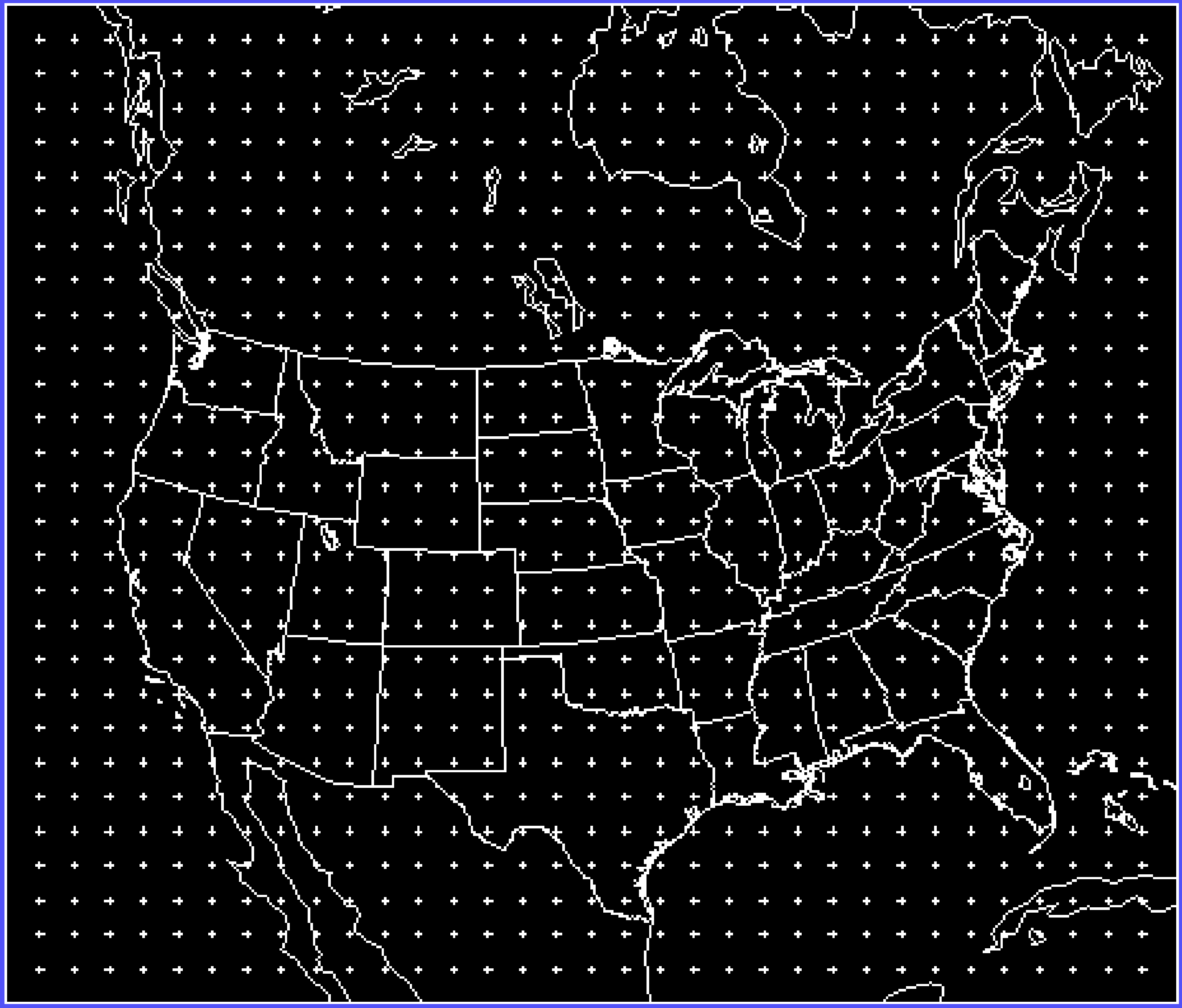
Over the entire modeling period (e.g., one year), puffs are released at periodic intervals (e.g., once every 7 hours).



Modeling Methodology

- NOAA HYSPLIT model → HYSPLIT-Hg
- **Modeling domain: North America**
  (northern half of Mexico; continental U.S.; southern half of Canada)
- **1996 meterology (180 km horizontal resolution)**

NGM Archive Grid



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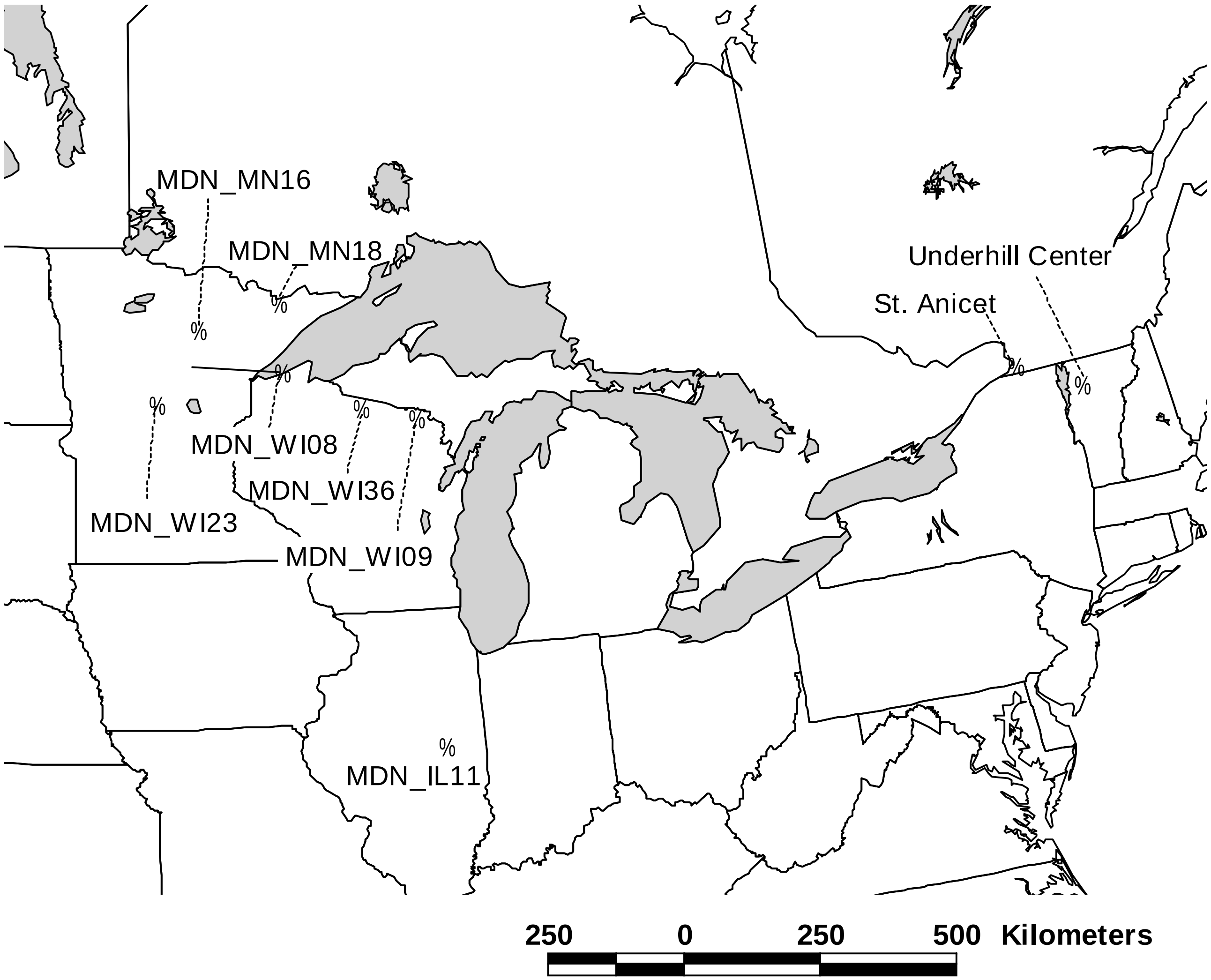
  (natural emissions, re-emissions, & global sources not included)
- **Model evaluation: 1996 emissions and 1996 monitoring data**
  (also evaluated in EMEP Hg model intercomparison project)

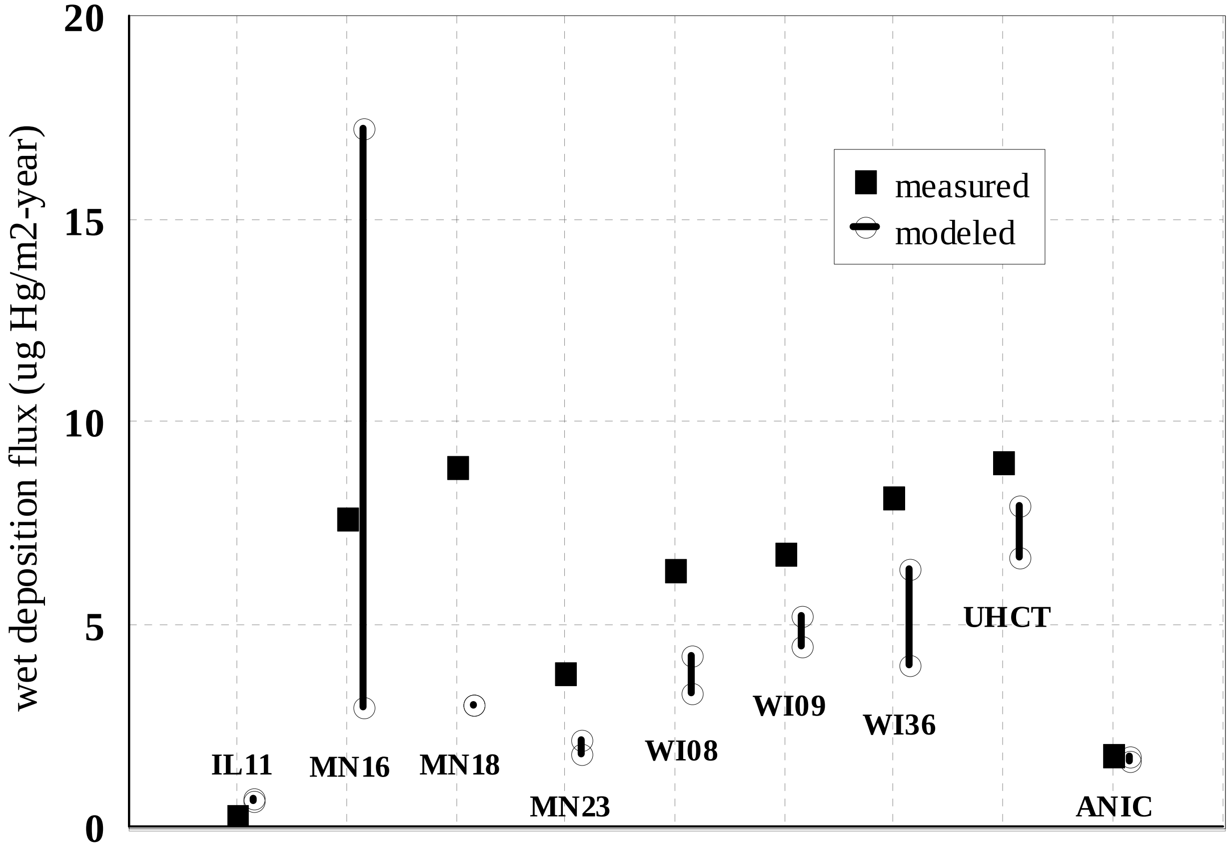
Figure 7. Model evaluation sites for wet deposition fluxes within 250 km of any Great Lake with available data for 1996.

(Cohen et al., 2004, Environmental Research 95: 247-265)

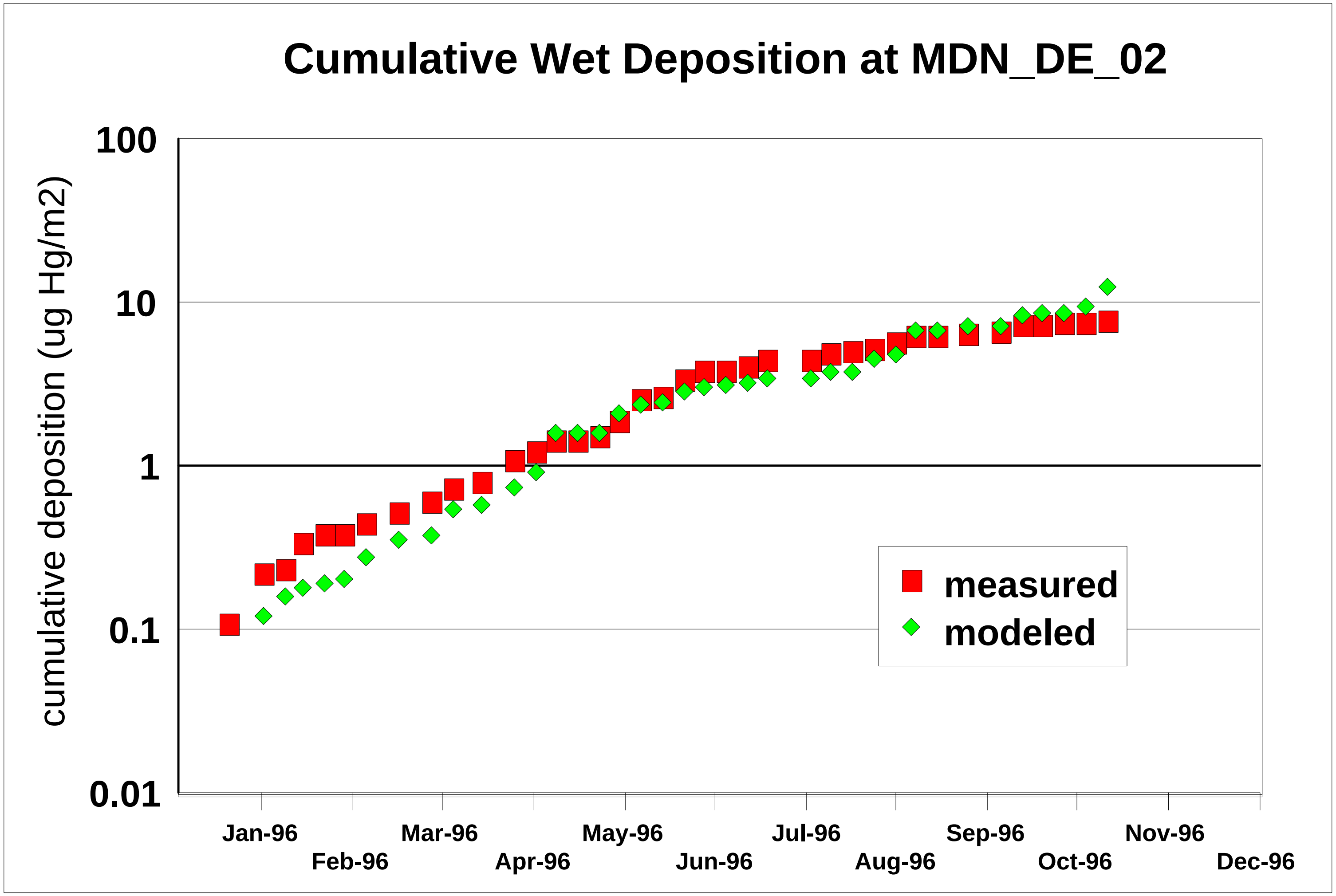


(Cohen et al., 2004, Environmental Research 95: 247-265)

Figure 8. Comparison of annual model-estimated wet deposition fluxes with measured values at sites within 250 km of the Great Lakes during 1996. The range of modeled estimates shown for each site represents the difference in estimated deposition in using the NGM-forecast *model* precipitation and the *actual* precipitation at the site. (Cohen et al., 2004, Environmental Research 95: 247-265)



Modeled vs. Measured Wet Deposition at Mercury Deposition Network Site DE_02 during 1996



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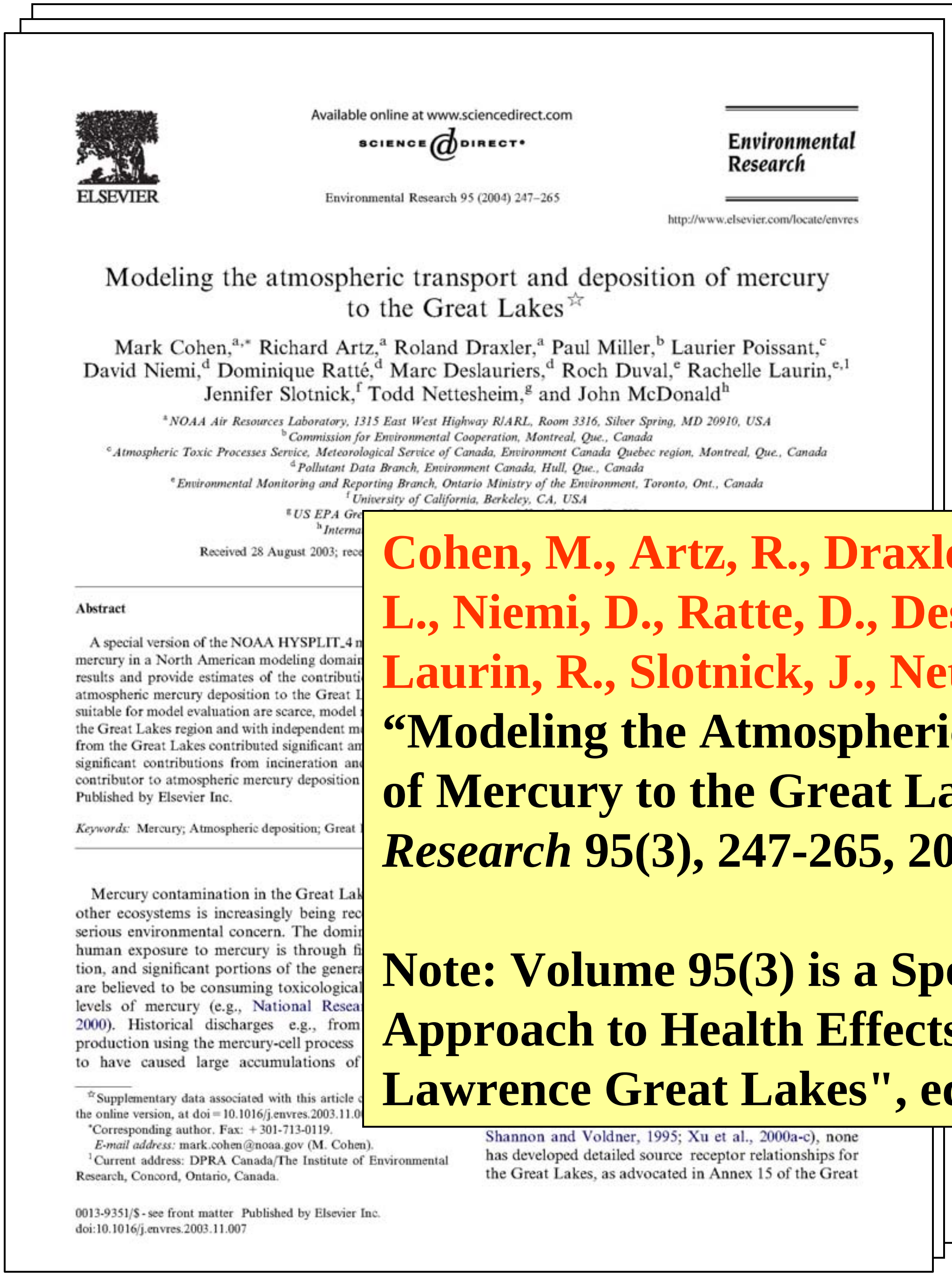
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Environmental Research 95 (2004) 247–265

Environmental Research

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Modeling the atmospheric transport and deposition of mercury to the Great Lakes☆

Mark Cohen,[a,*] Richard Artz,[a] Roland Draxler,[a] Paul Miller,[b] Laurier Poissant,[c] David Niemi,[d] Dominique Ratté,[d] Marc Deslauriers,[d] Roch Duval,[e] Rachelle Laurin,[e,1] Jennifer Slotnick,[f] Todd Nettesheim,[g] and John McDonald[h]

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[h] Interna

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Abstract

A special version of the NOAA HYSPLIT_4 m
mercury in a North American modeling domain
results and provide estimates of the contributi
atmospheric mercury deposition to the Great L
suitable for model evaluation are scarce, model
the Great Lakes region and with independent m
from the Great Lakes contributed significant am
significant contributions from incineration and
contributor to atmospheric mercury deposition
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Keywords: Mercury; Atmospheric deposition; Great

Mercury contamination in the Great Lak
other ecosystems is increasingly being rec
serious environmental concern. The domi
human exposure to mercury is through fi
tion, and significant portions of the genera
are believed to be consuming toxicological
levels of mercury (e.g., National Resea
2000). Historical discharges e.g., from
production using the mercury-cell process
to have caused large accumulations of

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

☆Supplementary data associated with this article c
the online version, at doi=10.1016/j.envres.2003.11.0

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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 O. Carpenter.

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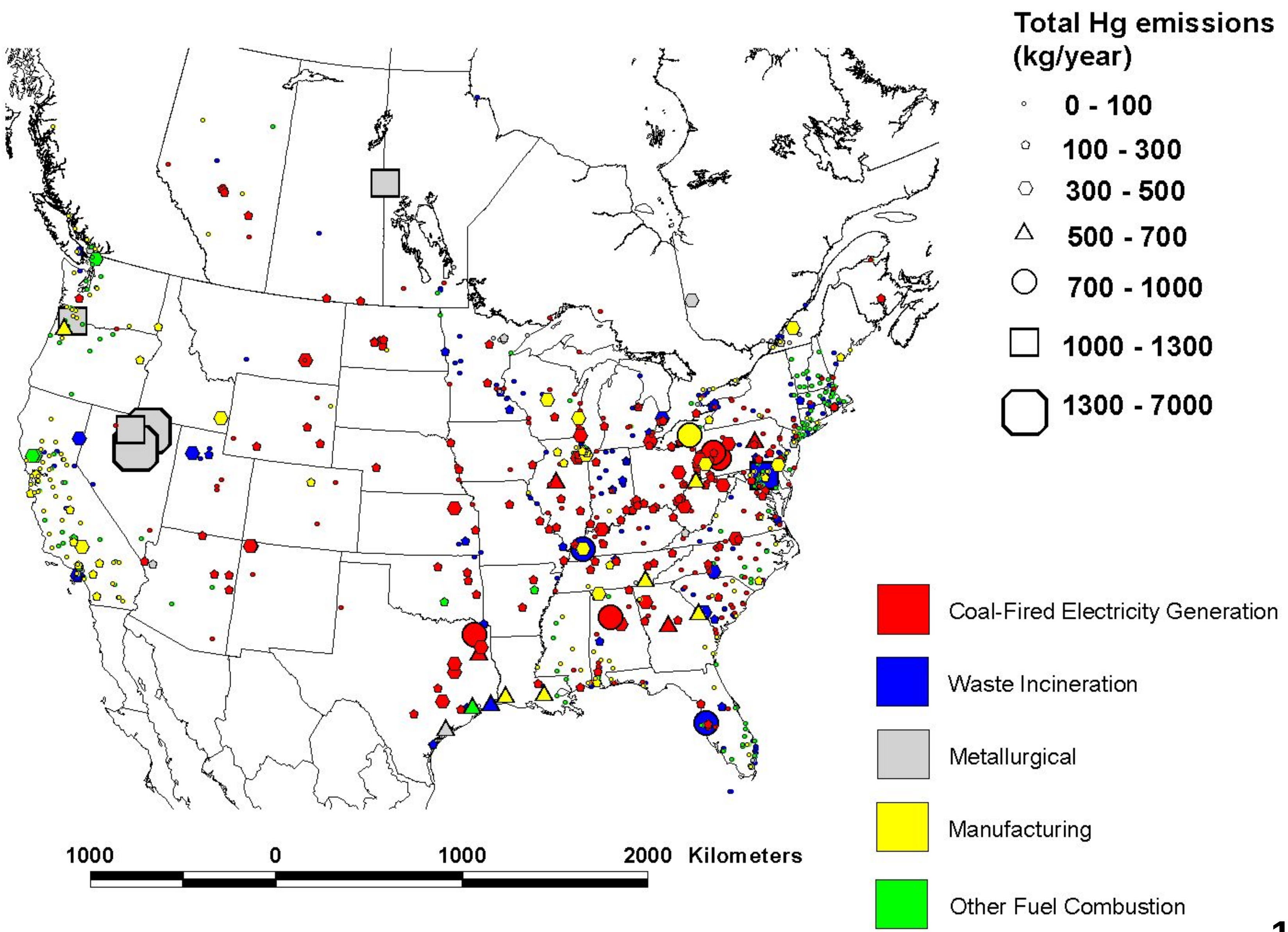
- **Model evaluation: 1996 emissions and 1996 monitoring data**
  (also evaluated in EMEP Hg model intercomparison project)
- **1st set of results – Cohen et al. 2004**
- **2nd set of results (examples shown today) –**
  - ***1996 meteorology***
  - ***1999 U.S. EPA National Emissions Inventory***
  - ***2000 emissions data from Environment Canada***

Geographic Distribution of Largest Anthropogenic Mercury Emissions Sources in the U.S. (1999) and Canada (2000)



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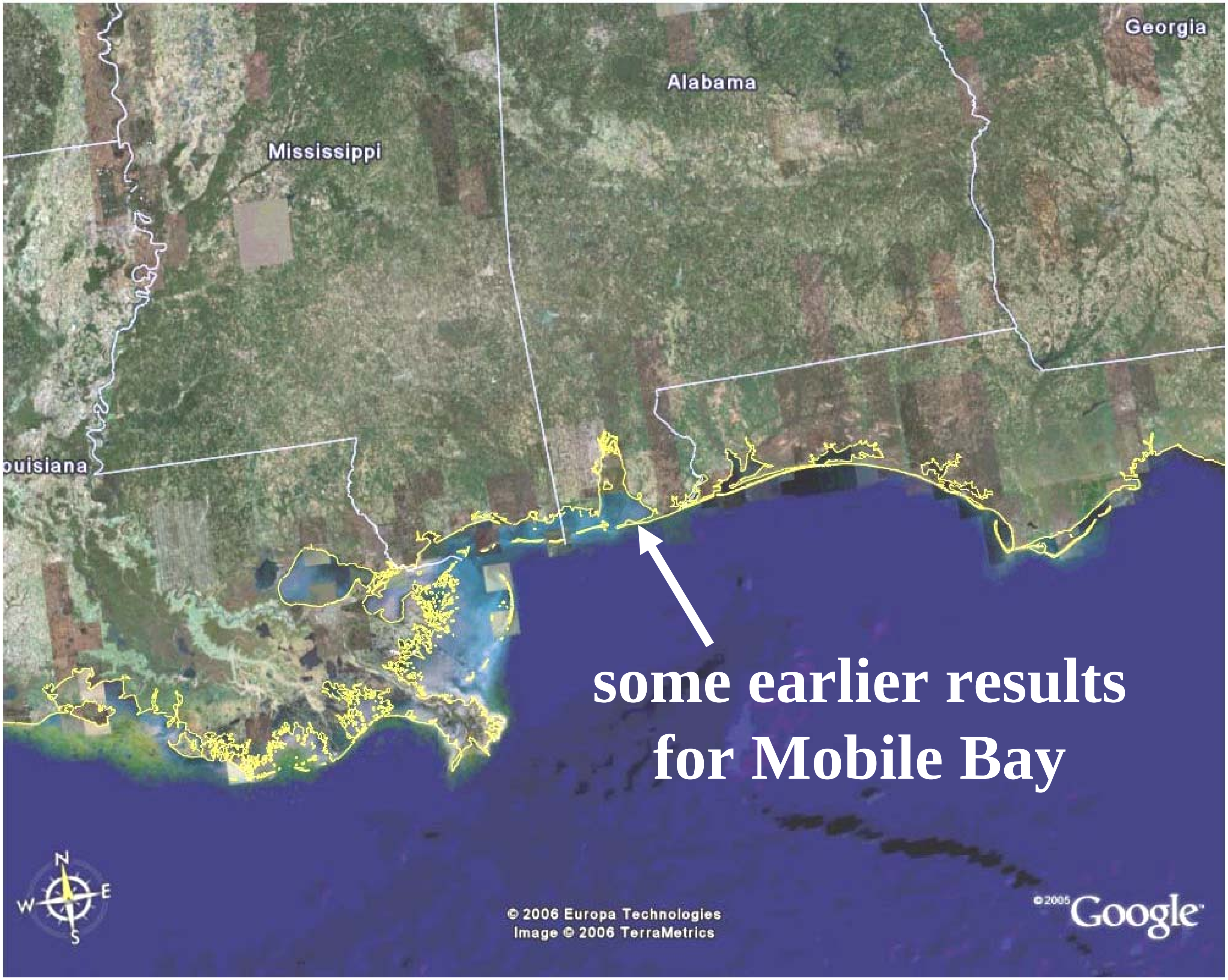
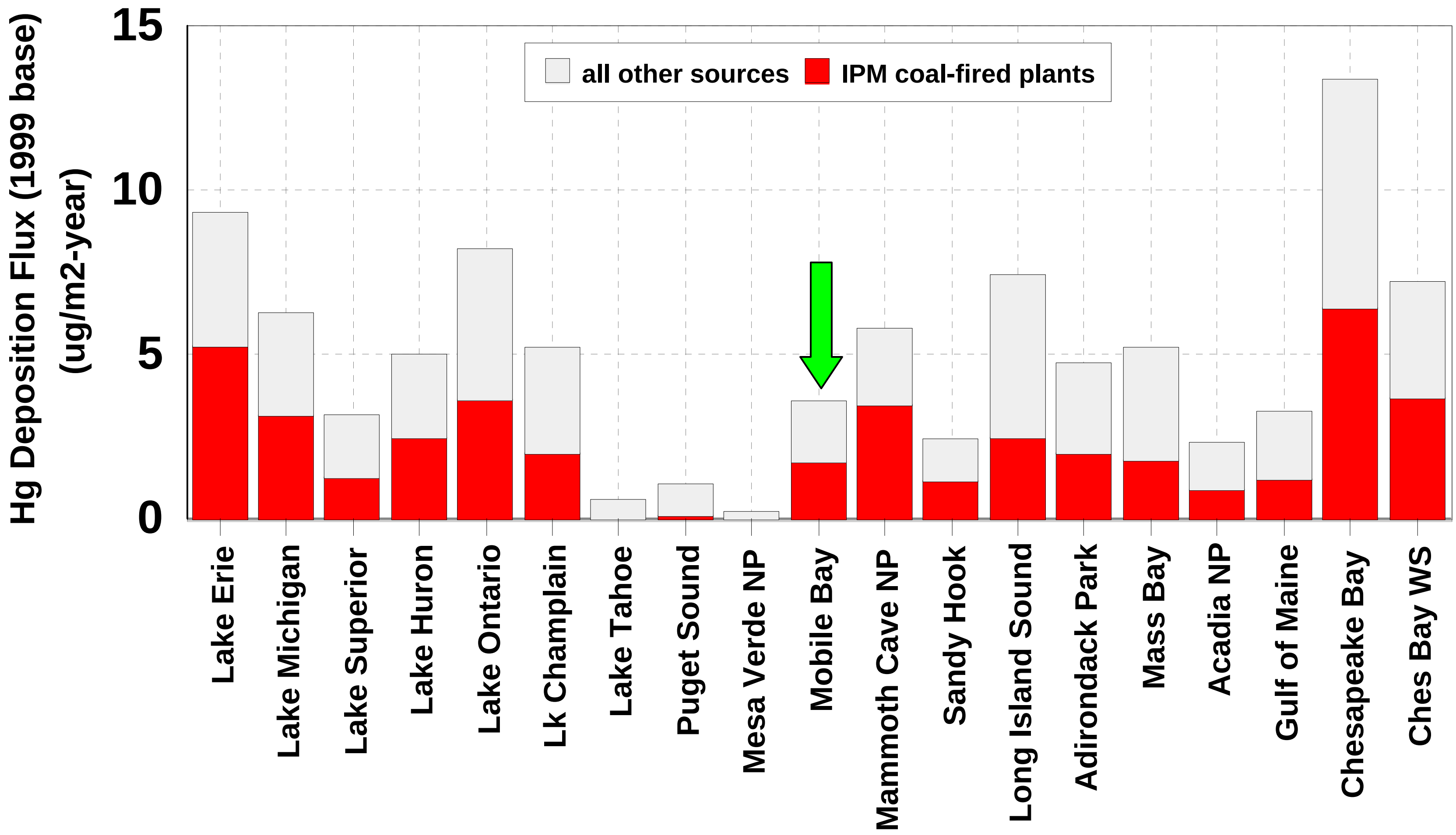
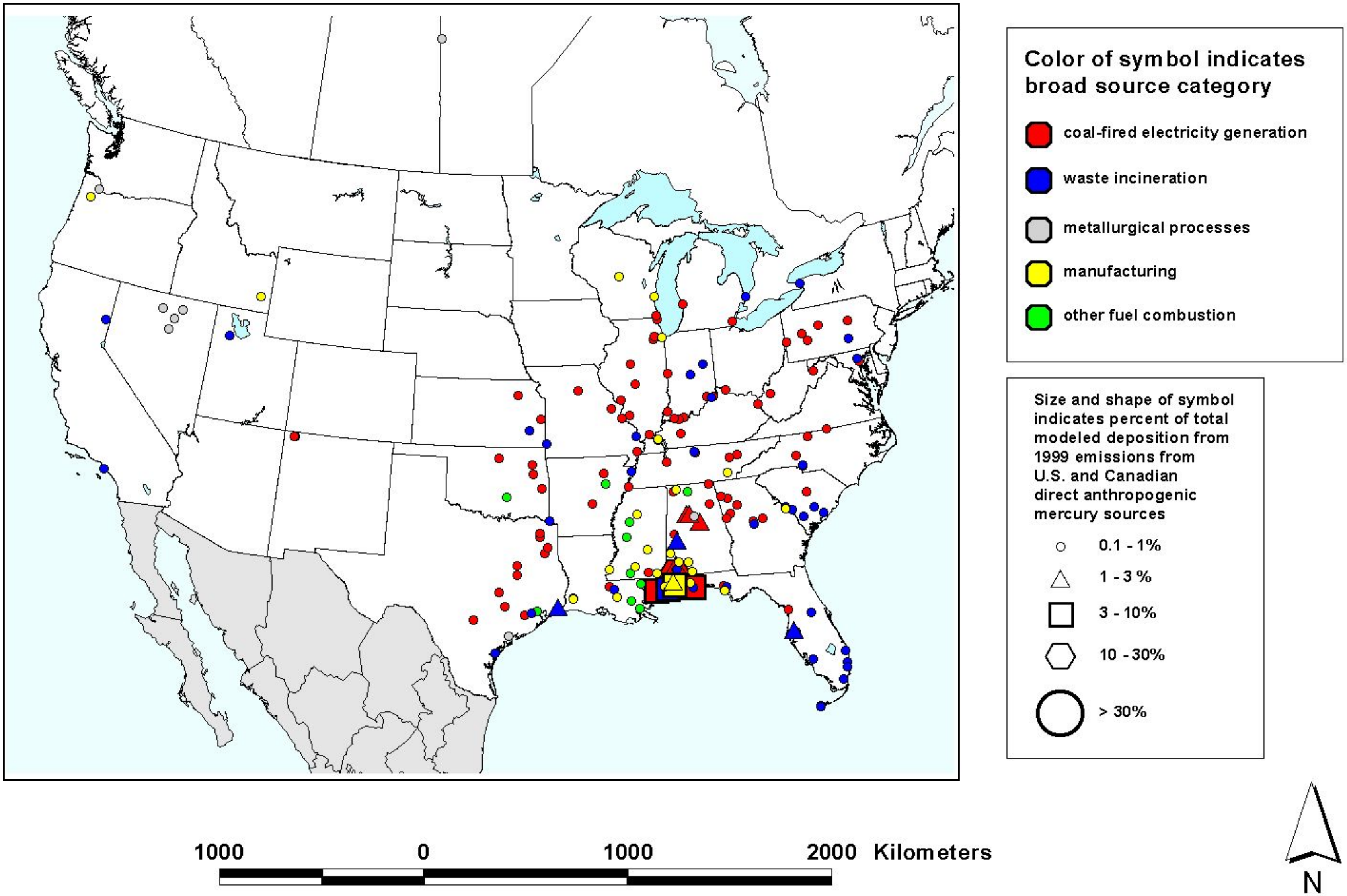

Georgia
Alabama
Mississippi
ouisiana
some earlier results
for Mobile Bay
© 2006 Europa Technologies
Image © 2006 TerraMetrics
© 2005 Google

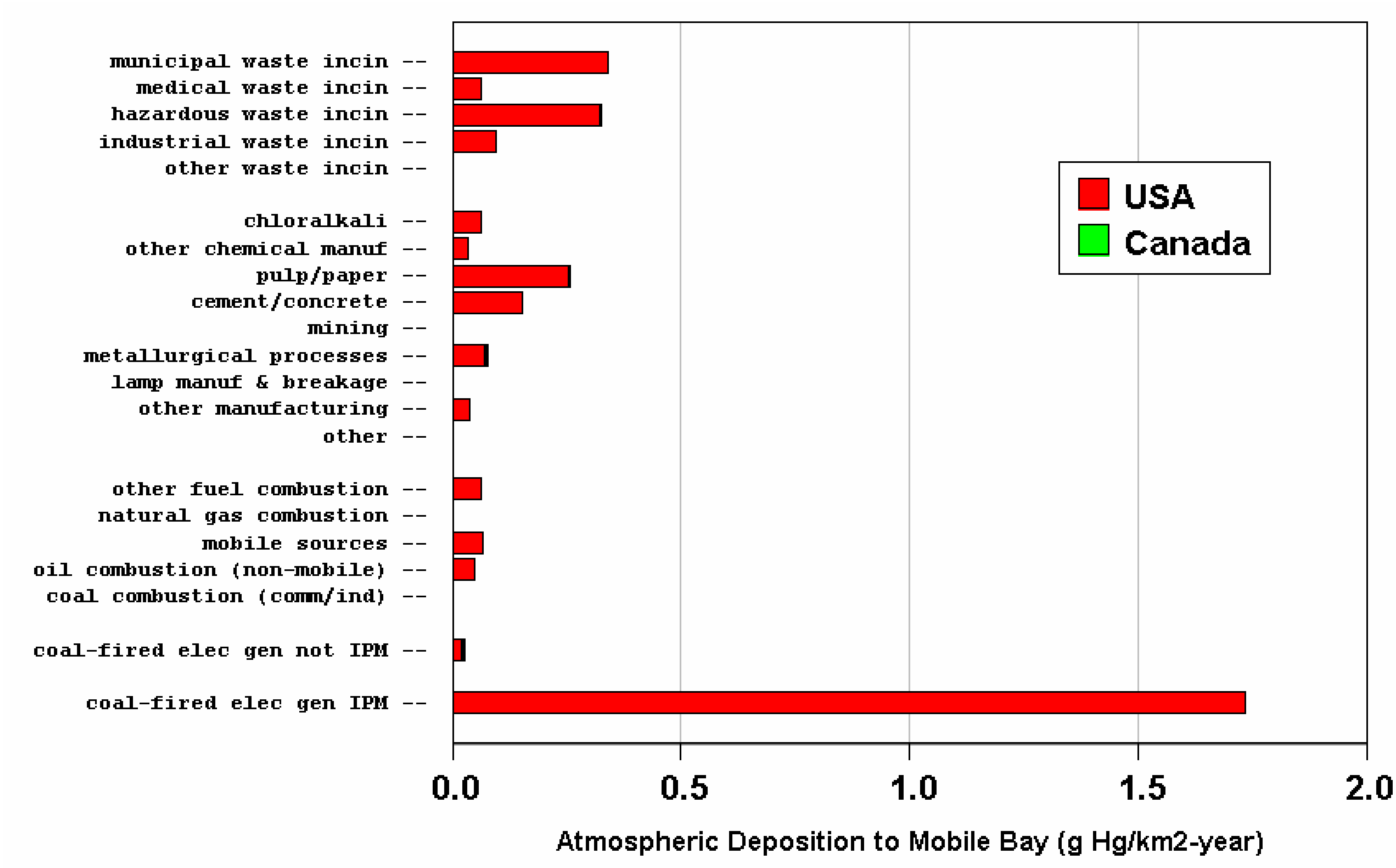
Total modeled mercury deposition at selected receptors arising from from 1999 direct anthropogenic emissions sources in the United States and Canada (IPM coal fired plants are large coal-fired plants in the U.S. only)



Largest Modeled Individual Sources Contributing Mercury Deposition Directly to Mobile Bay (national view)

- *1996 meteorology (NGM)*
- *1999 U.S. emissions (EPA NEI)*
- *2000 Canadian emissions (Envr. Canada)*
- *no sources other than U.S. & Can. anthropogenic emissions*
- *total modeled deposition to Mobile Bay ~ 3.5 g Hg/km^2-year*





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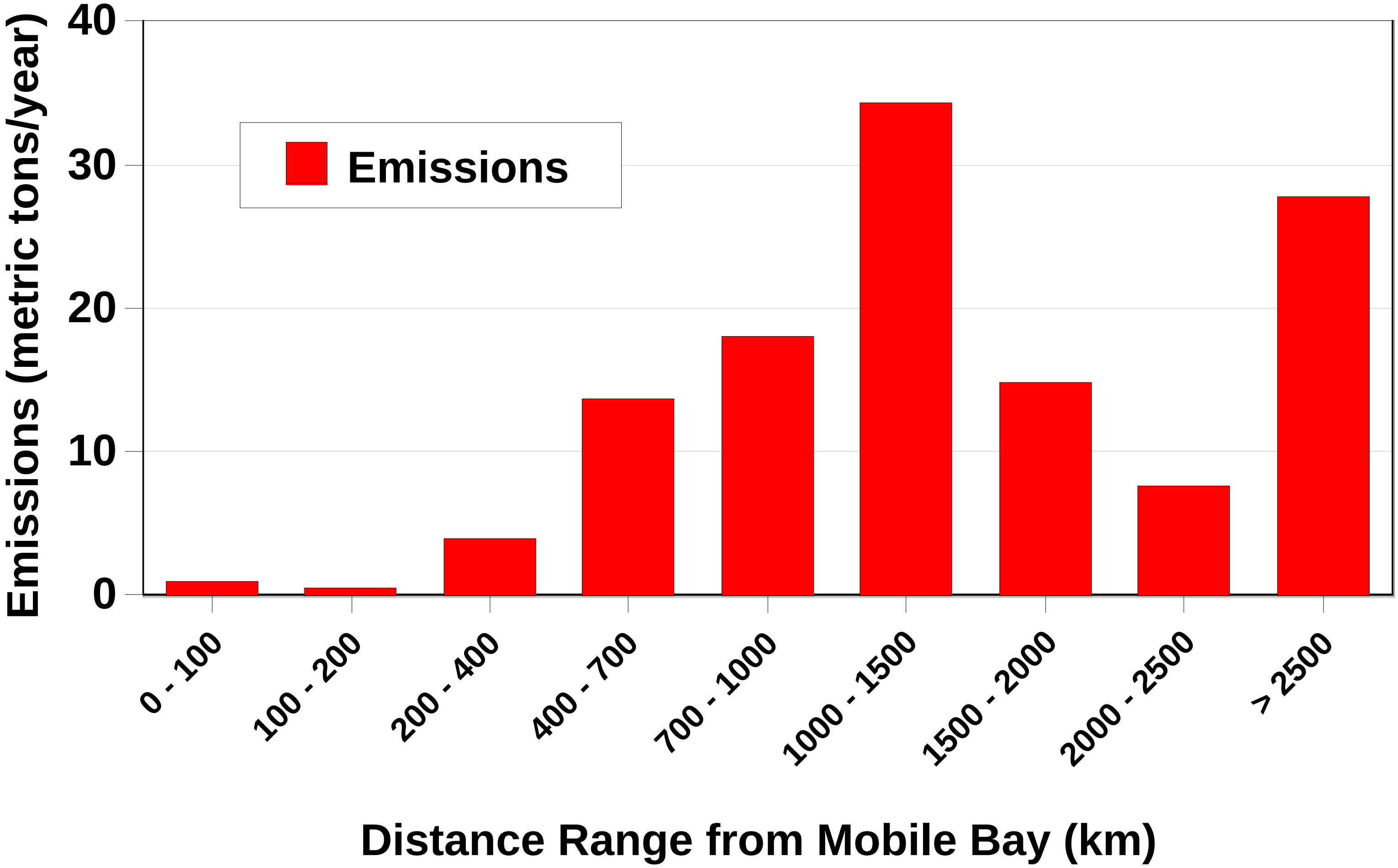
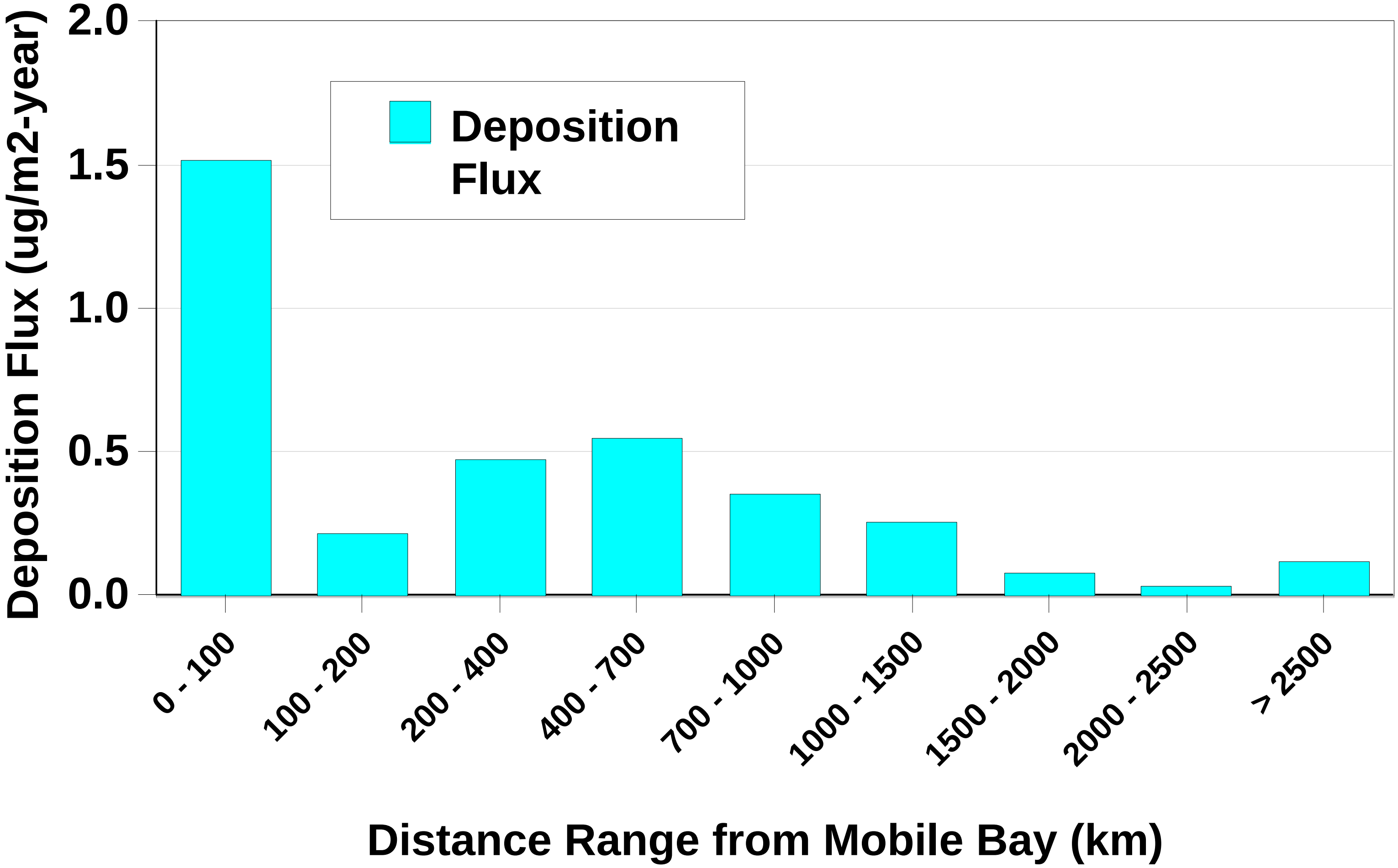
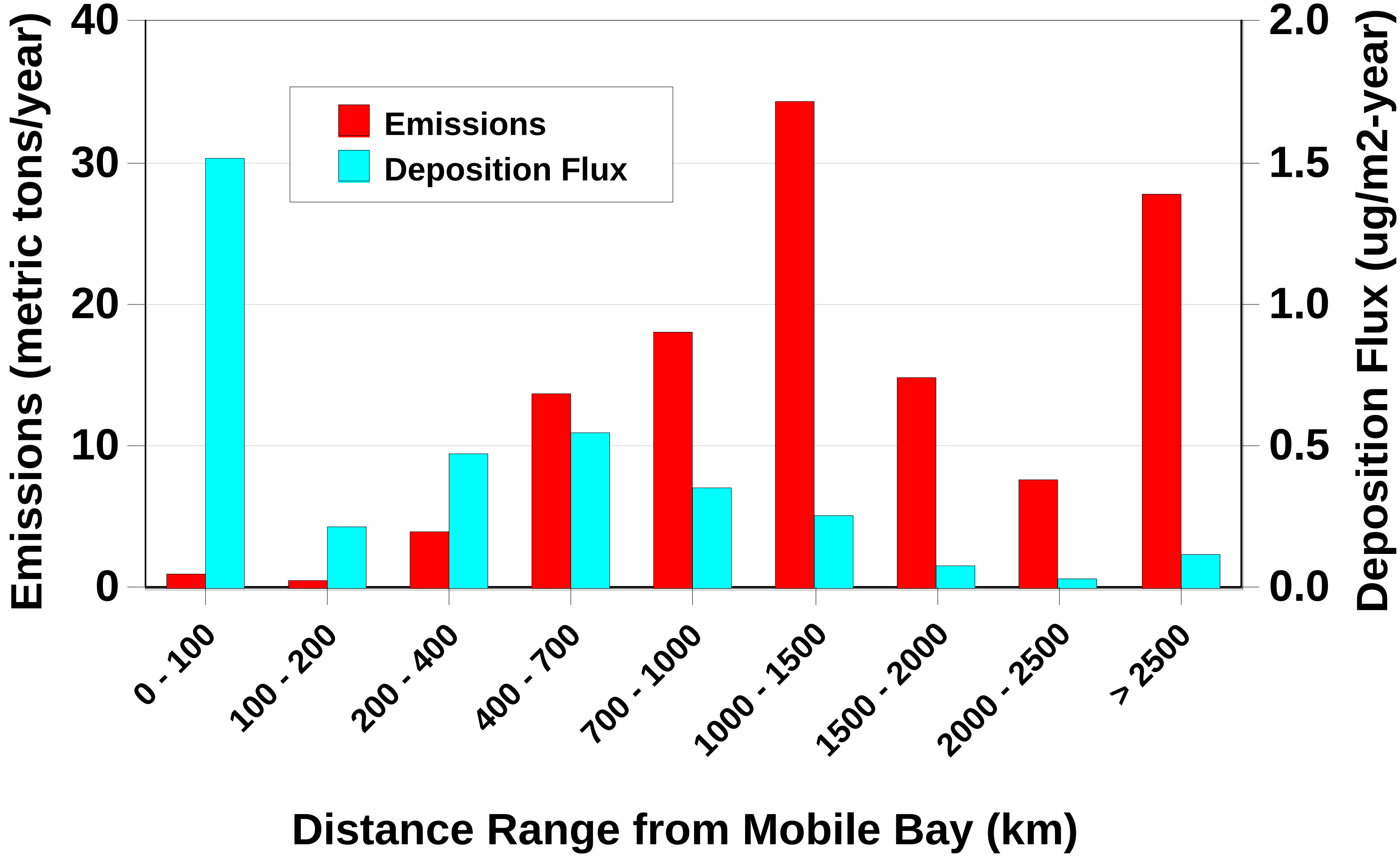


Emissions (metric tons/year)
40
30
20
10
0
Emissions
0 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
> 2500
Distance Range from Mobile Bay (km)


Deposition Flux (ug/m2-year)
2.0
1.5
1.0
0.5
0.0
Deposition Flux
0 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
> 2500
Distance Range from Mobile Bay (km)



Emissions (metric tons/year)
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Distance Range from Mobile Bay (km)

Largest Modeled Individual Sources Contributing Mercury Deposition Directly to Mobile Bay (large regional view)

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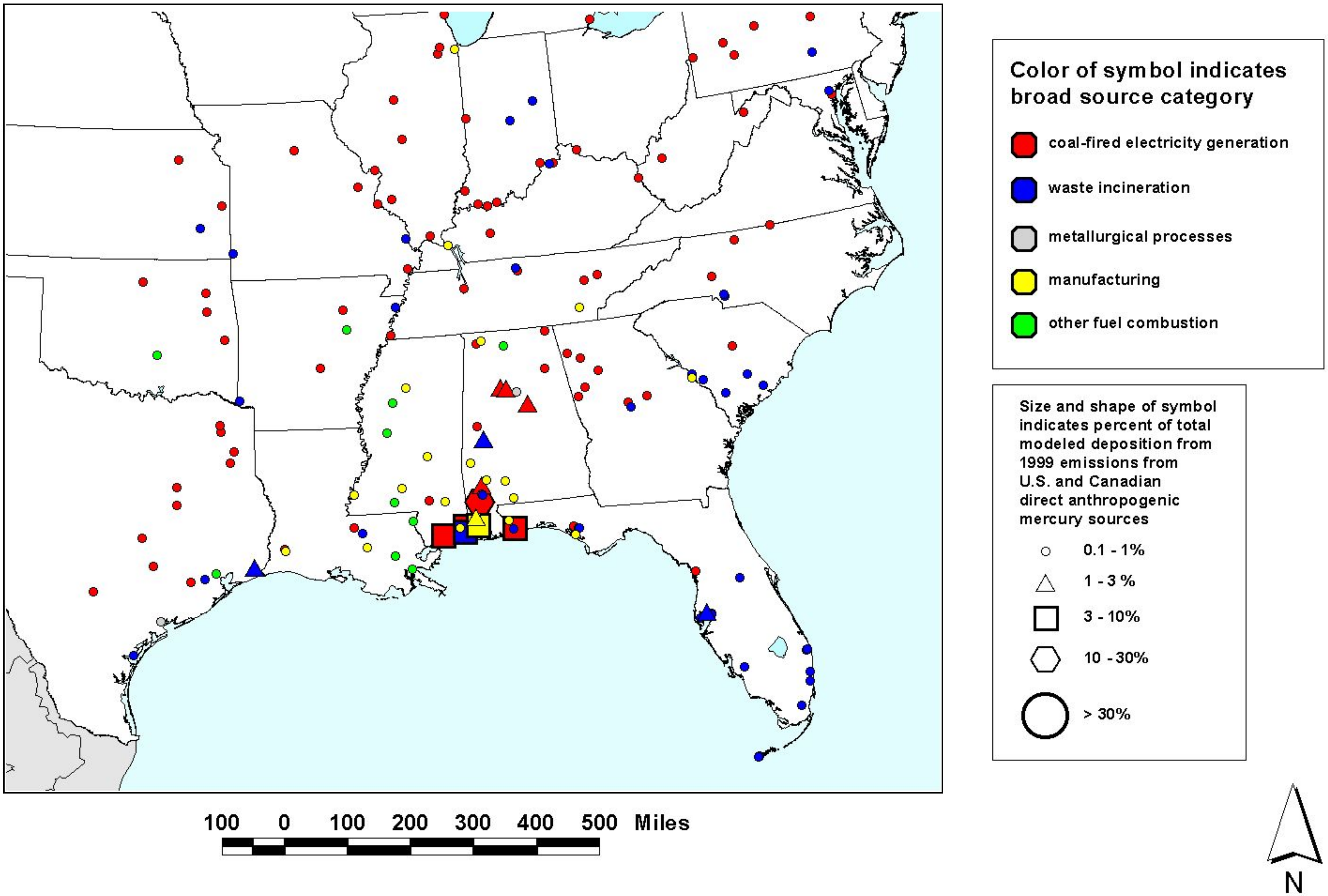

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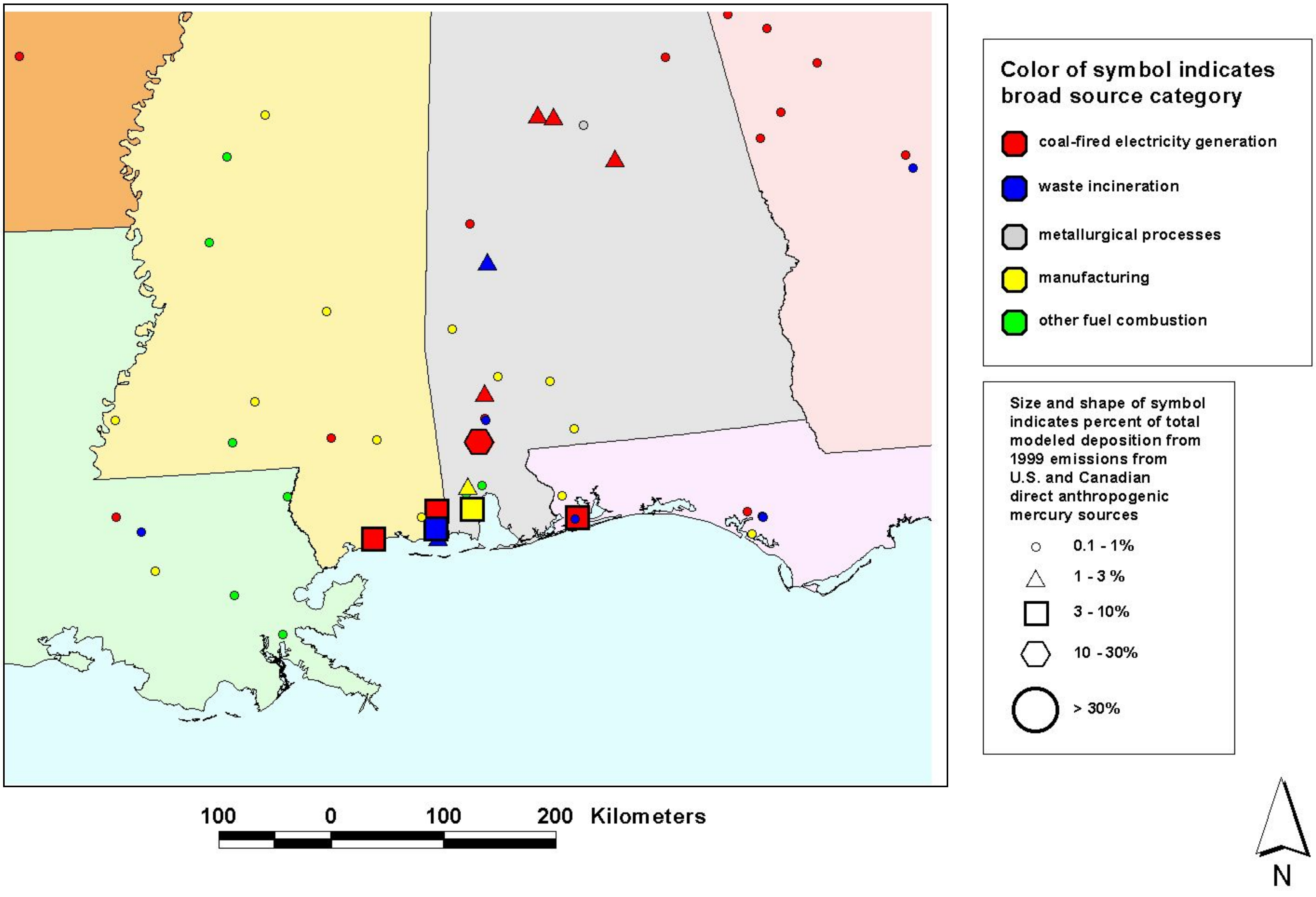

Largest Modeled Individual Sources Contributing Mercury Deposition Directly to Mobile Bay (local view)

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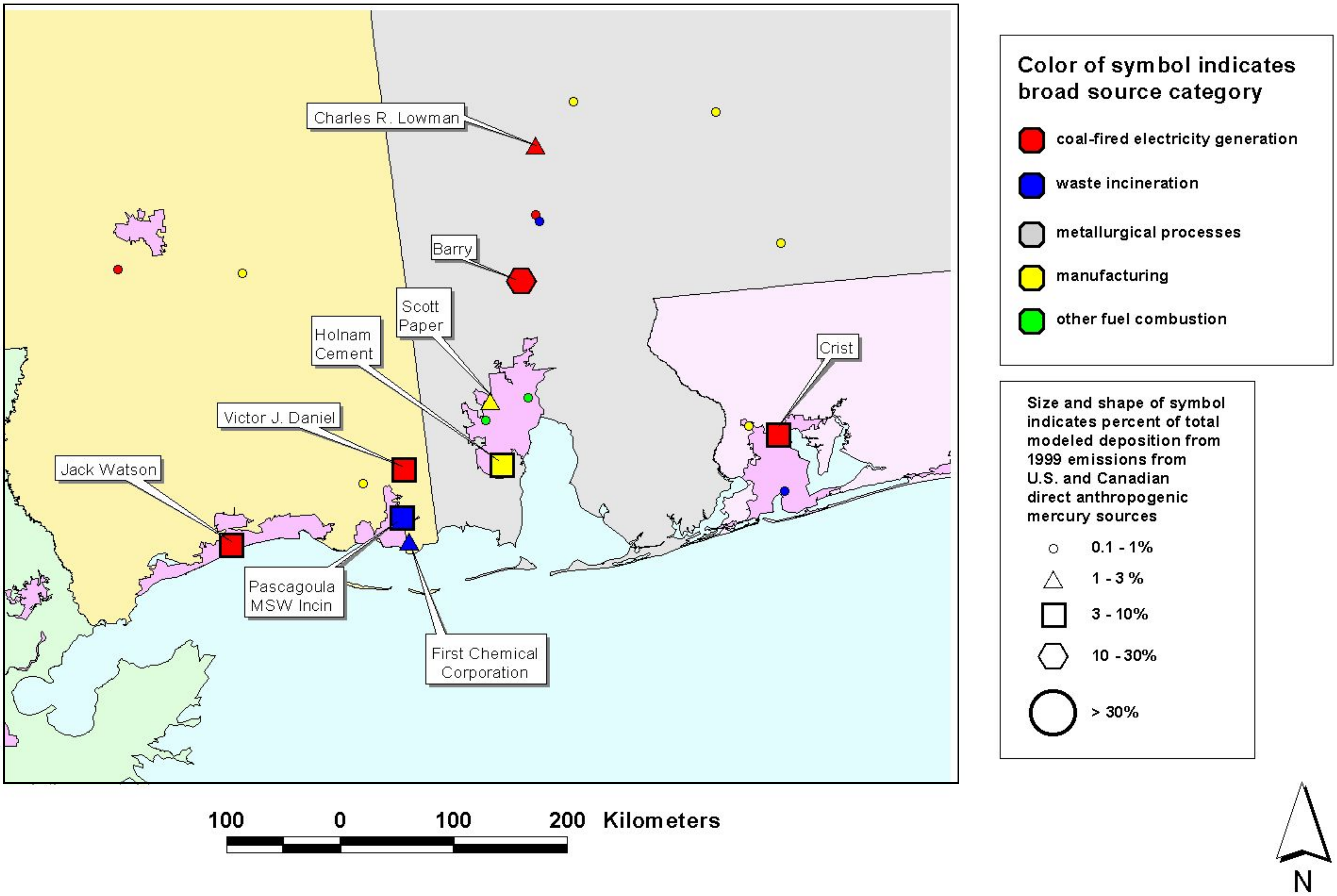
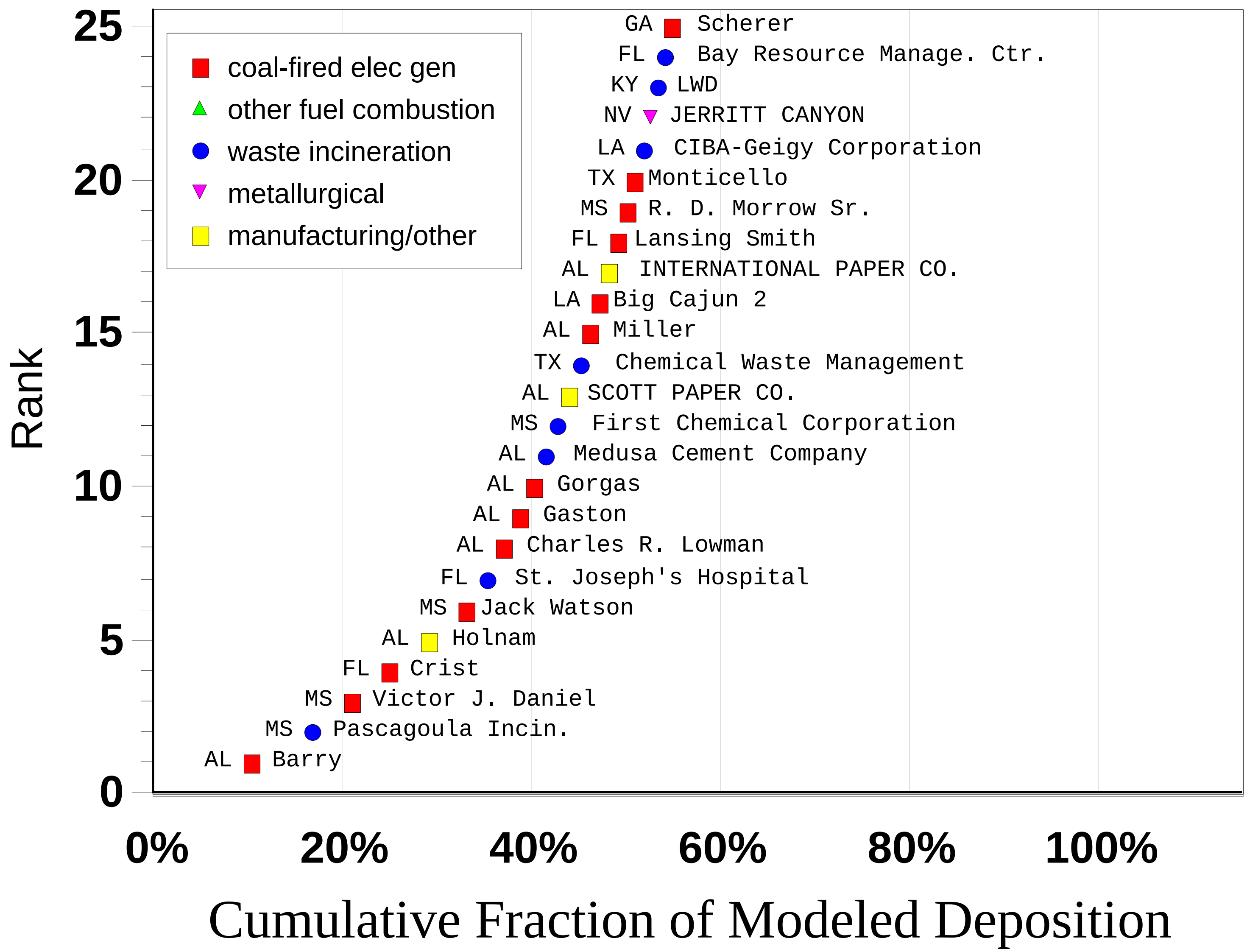
- *1999 U.S. emissions (EPA NEI)*
- *2000 Canadian emissions (Envr. Canada)*
- *no sources other than U.S. & Can. anthropogenic emissions*
- *total modeled deposition to Mobile Bay ~ 3.5 g Hg/km^2-year*



Top 25 Modeled Contributors to 1999 Hg Deposition Directly to Mobile Bay, considering anthropogenic direct emission sources in the United States and Canada



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- ❑ **model intercomparisons**
- ❑ summary of previous work; current goals; challenges

Model Intercomparisons

- EMEP MSC-East (~7 models)
- HYSPLIT-Hg vs. ISC
- HYSPLIT-Hg vs. CMAQ-Hg

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Participants

D. Syrakov ………………………………. **Bulgaria**….**NIMH**

A. Dastoor, D. Davignon ………………. **Canada**...... **MSC-Can**

J. Christensen …………………………. **Denmark**…**NERI**

G. Petersen, R. Ebinghaus ……………..... **Germany**…**GKSS**

J. Pacyna ………………………………. **Norway**…..**NILU**

J. Munthe, I. Wängberg ……………….. **Sweden**….. **IVL**

R. Bullock ………………………………**USA**………**EPA**

M. Cohen, R. Artz, R. Draxler …………**USA**………**NOAA**

C. Seigneur, K. Lohman ………………..**USA**……... **AER/EPRI**

A. Ryaboshapko, I. Ilyin, O.Travnikov…**EMEP**……**MSC-E**

Intercomparison Conducted in 3 Stages

I. Comparison of chemical schemes for a cloud environment

II. Air Concentrations in Short Term Episodes

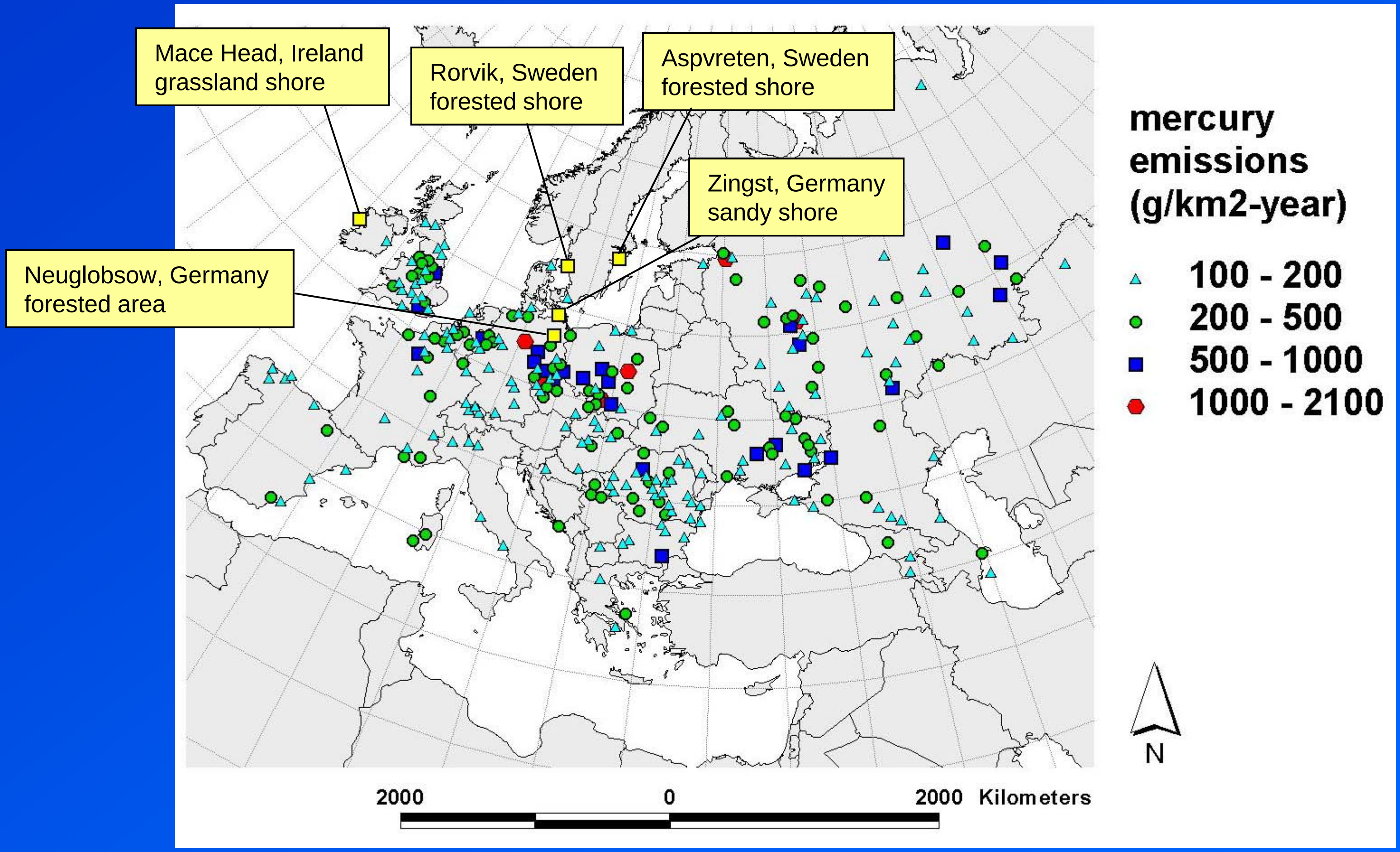
III. Long-Term Deposition and Source-Receptor Budgets

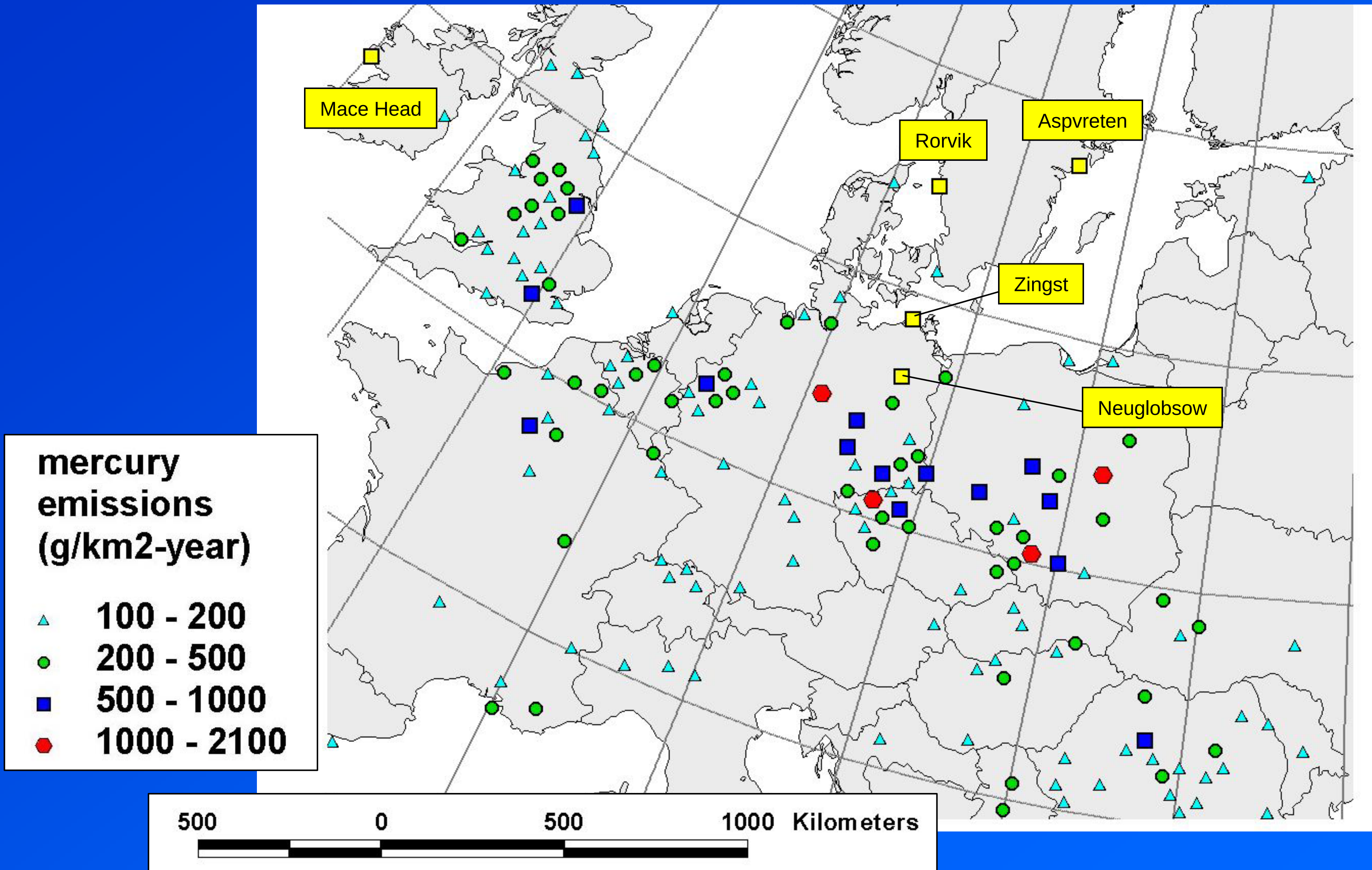
Participating Models

Model Acronym	*Model Name* and Institution	Stage		
		I	II	III
CAM	*Chemistry of Atmos. Mercury model*, Environmental Institute, Sweden			
MCM	*Mercury Chemistry Model*, Atmos. & Environmental Research, USA			
CMAQ	*Community Multi-Scale Air Quality model*, US EPA			
ADOM	*Acid Deposition and Oxidants Model*, GKSS Research Center, Germany			
MSCE-HM	*MSC-E heavy metal regional model*, EMEP MSC-E			
GRAHM	*Global/Regional Atmospheric Heavy Metal model*, Environment Canada			
EMAP	*Eulerian Model for Air Pollution*, Bulgarian Meteo-service			
DEHM	*Danish Eulerian Hemispheric Model*, National Environmental Institute			
HYSPLIT	*Hybrid Single Particle Lagrangian Integrated Trajectory model*, US NOAA			
MSCE-HM-Hem	*MSC-E heavy metal hemispheric model*, EMEP MSC-E			

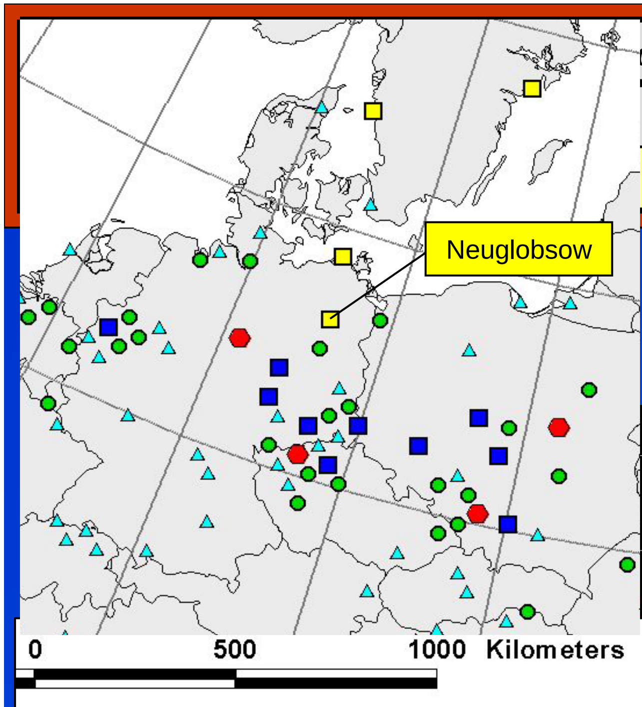
Anthropogenic Mercury Emissions Inventory and Monitoring Sites for Phase II

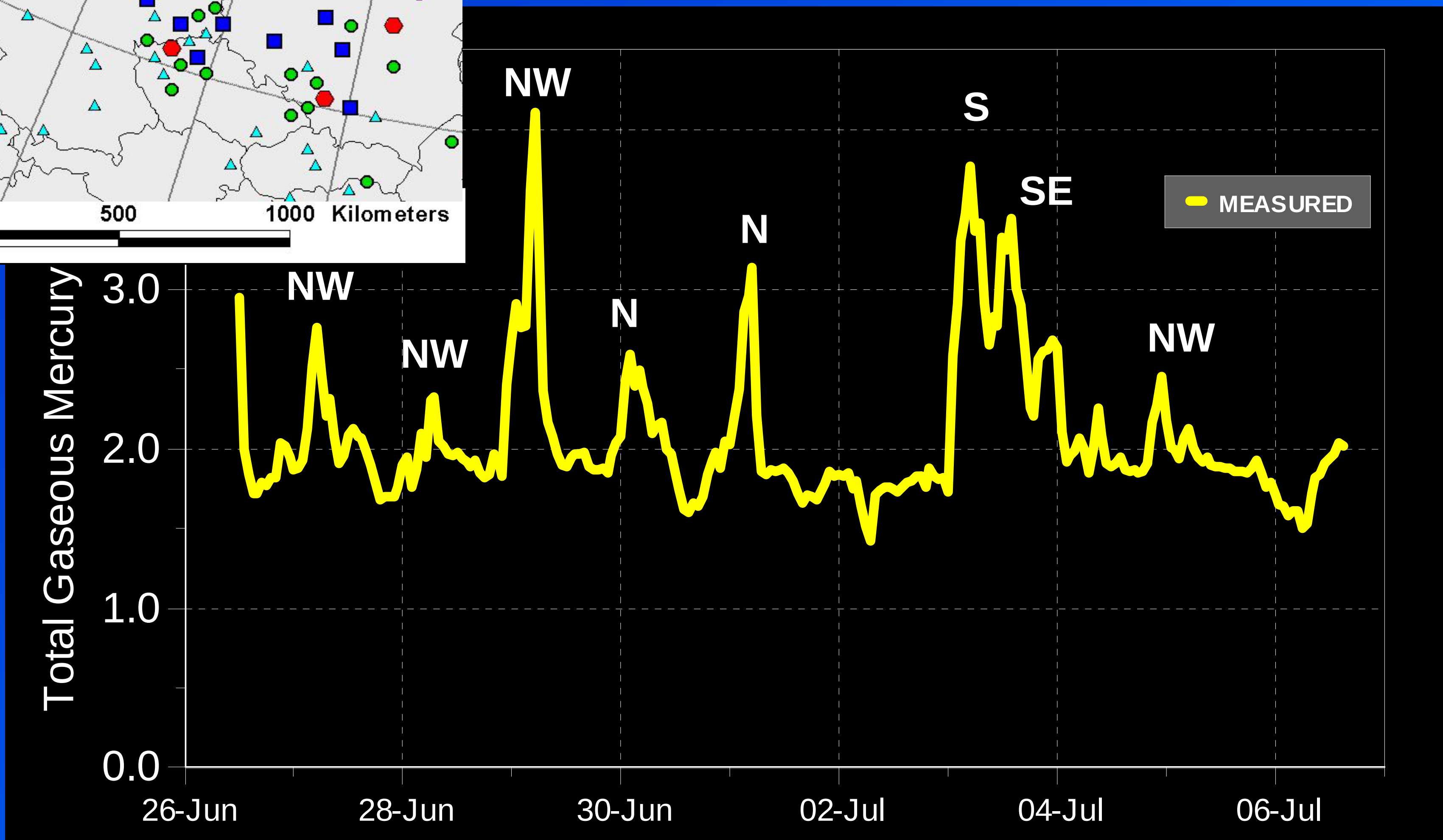
(note: only showing largest emitting grid cells)



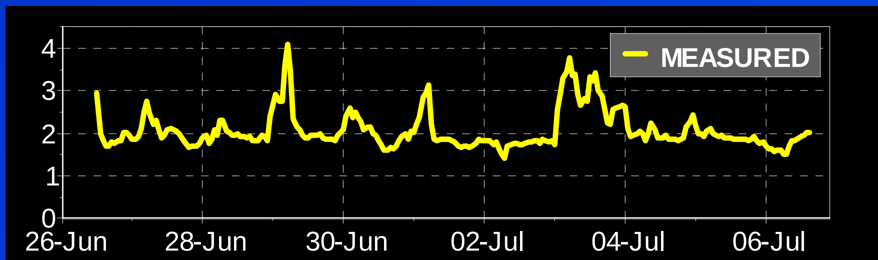

Mace Head
Rorvik
Aspvreten
Zingst
Neuglobsow
mercury emissions (g/km2-year)
100 - 200
200 - 500
500 - 1000
1000 - 2100
500
0
500
1000
Kilometers

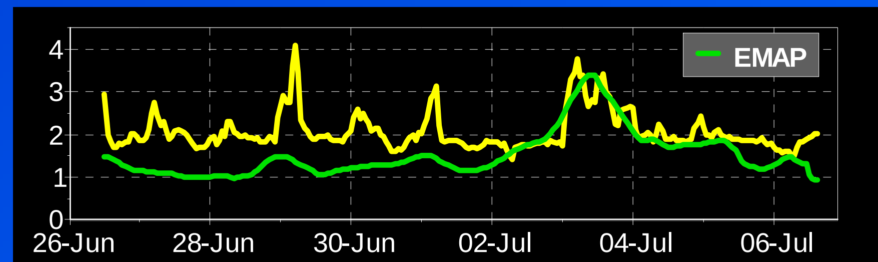
...rcury at Neuglobsow: June 26 – July 6, 1995

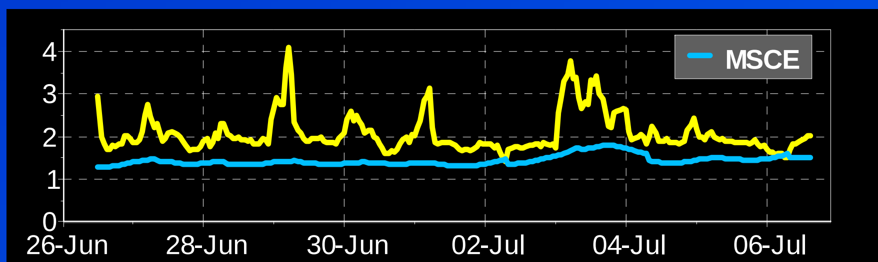


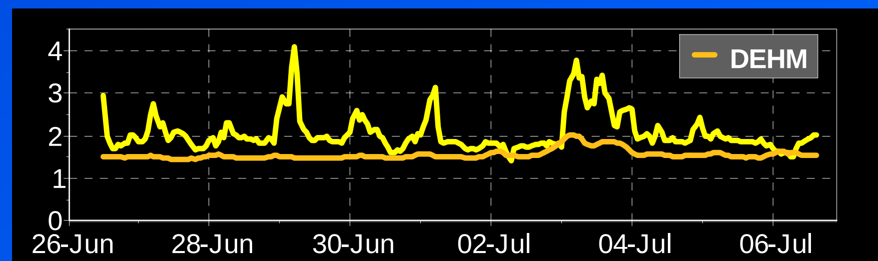


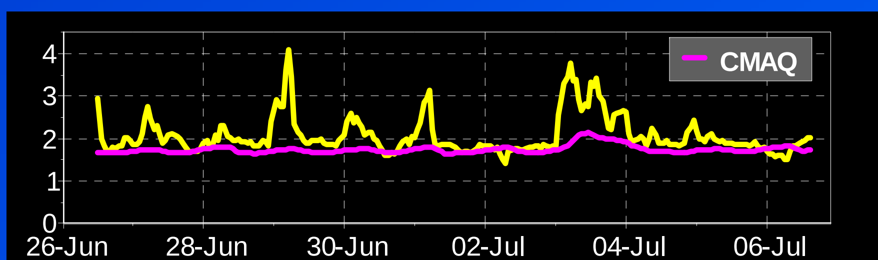
Total Gaseous Mercury (ng/m^3) at Neuglobsow: June 26 – July 6, 1995

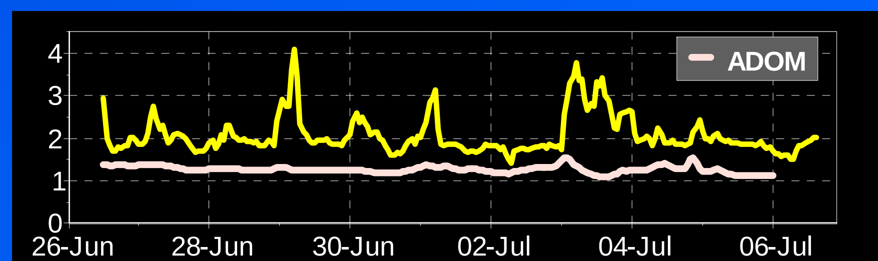


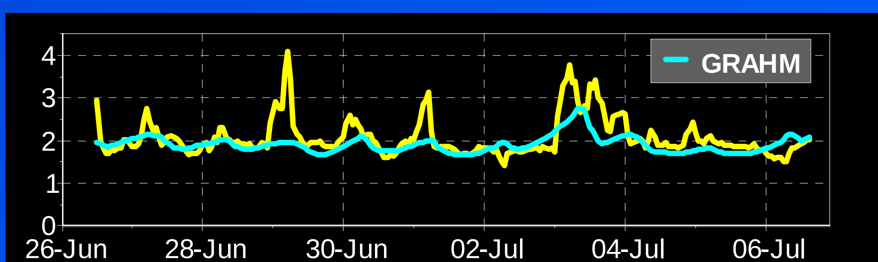


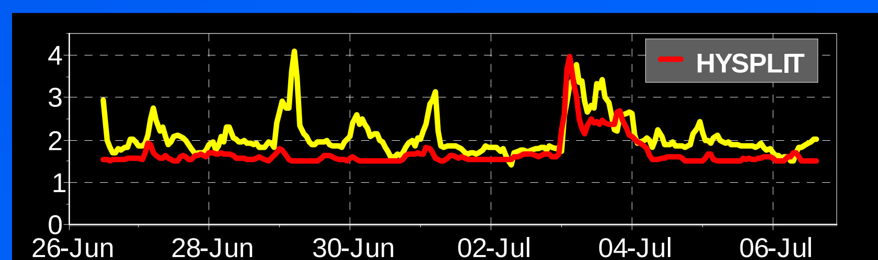




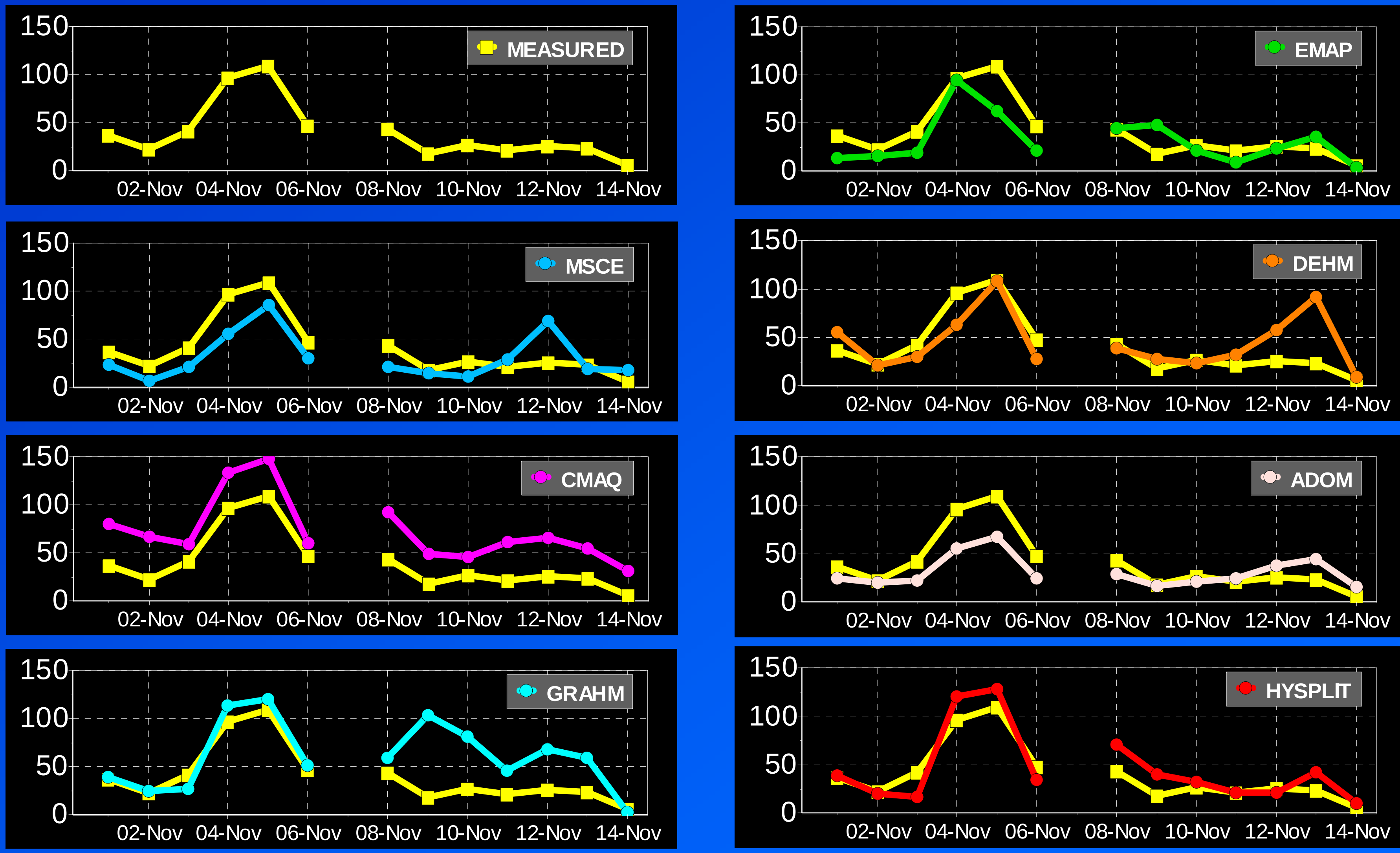








Total *Particulate* Mercury (pg/m³) at Neuglobsow, Nov 1-14, 1999



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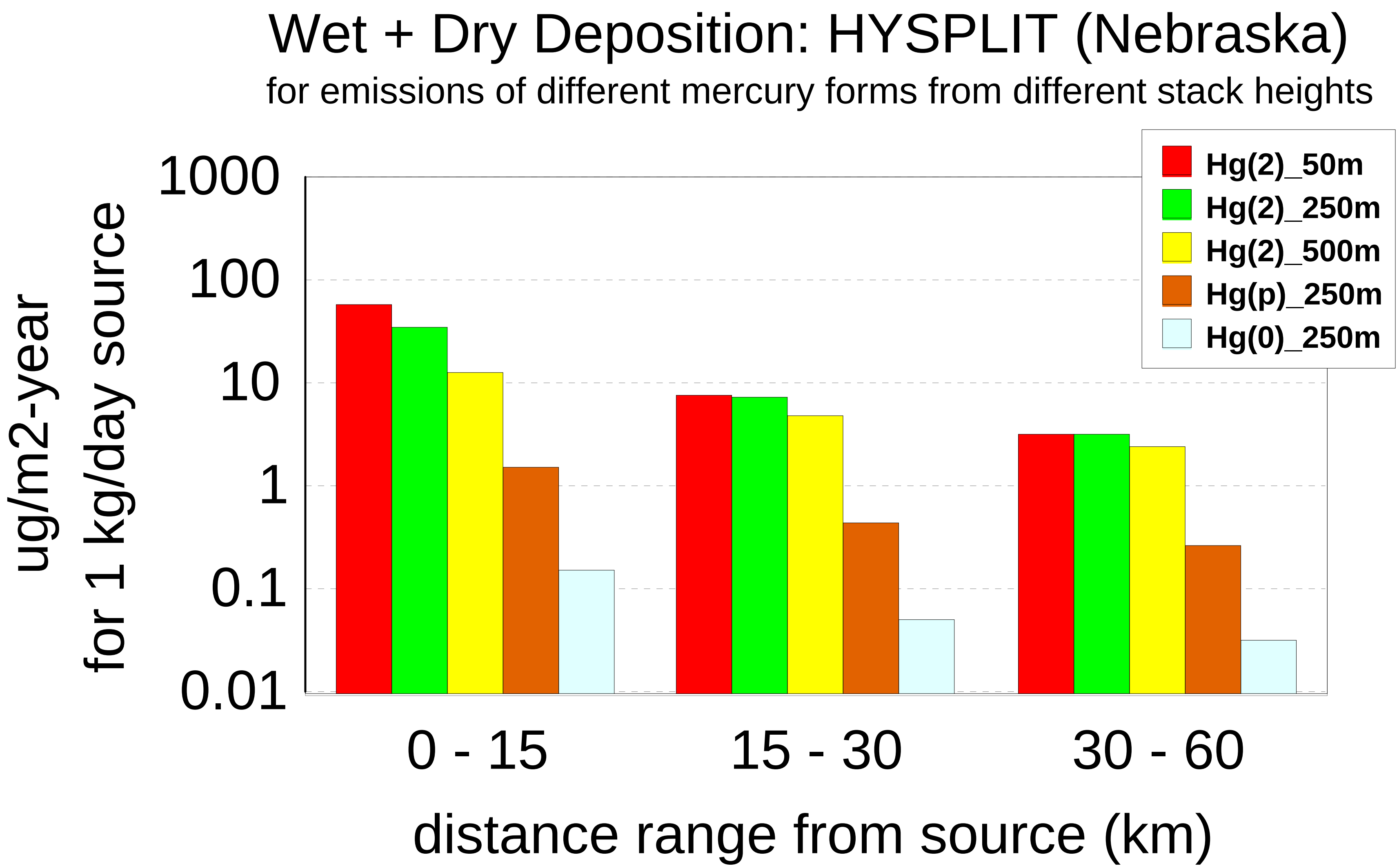
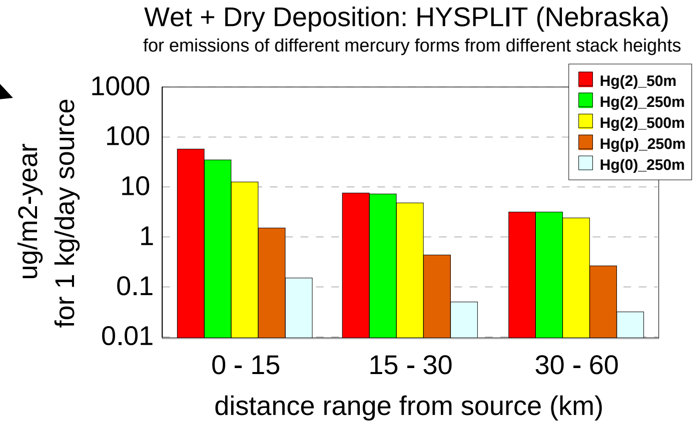


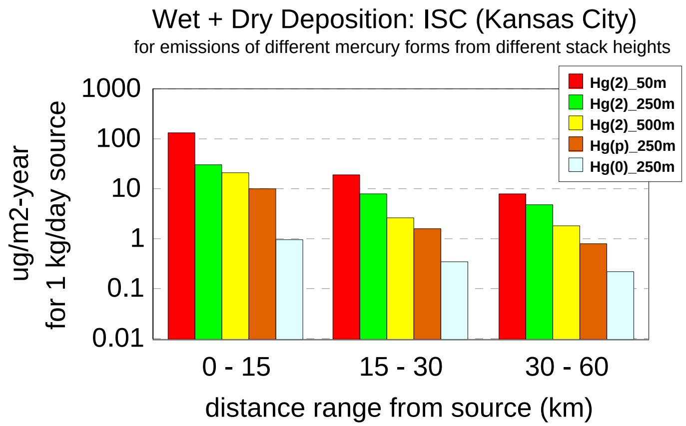
Wet + Dry Deposition: HYSPLIT (Nebraska)
for emissions of different mercury forms from different stack heights
ug/m2-year
for 1 kg/day source
1000
100
10
1
0.1
0.01
Hg(2)_50m
Hg(2)_250m
Hg(2)_500m
Hg(p)_250m
Hg(0)_250m
0 - 15
15 - 30
30 - 60
distance range from source (km)

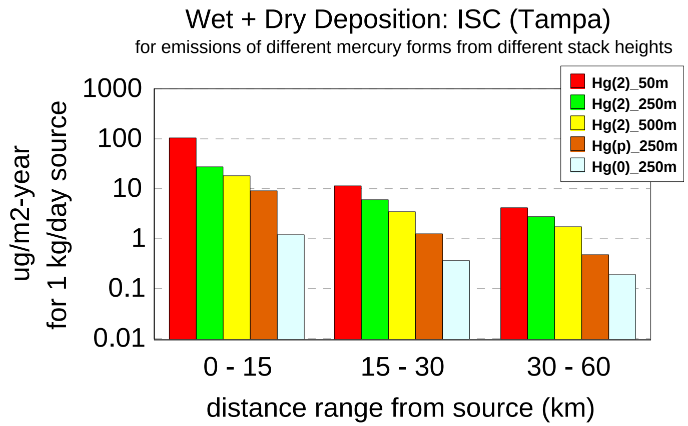
HYSPLIT 1996

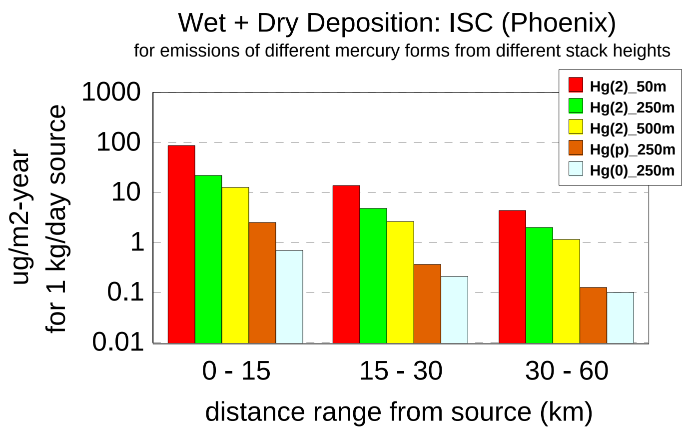


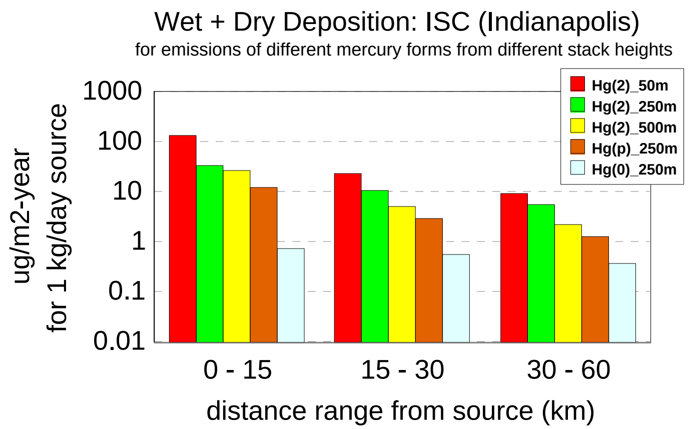
Different Time Periods and Locations, but Similar Results

ISC: 1990-1994









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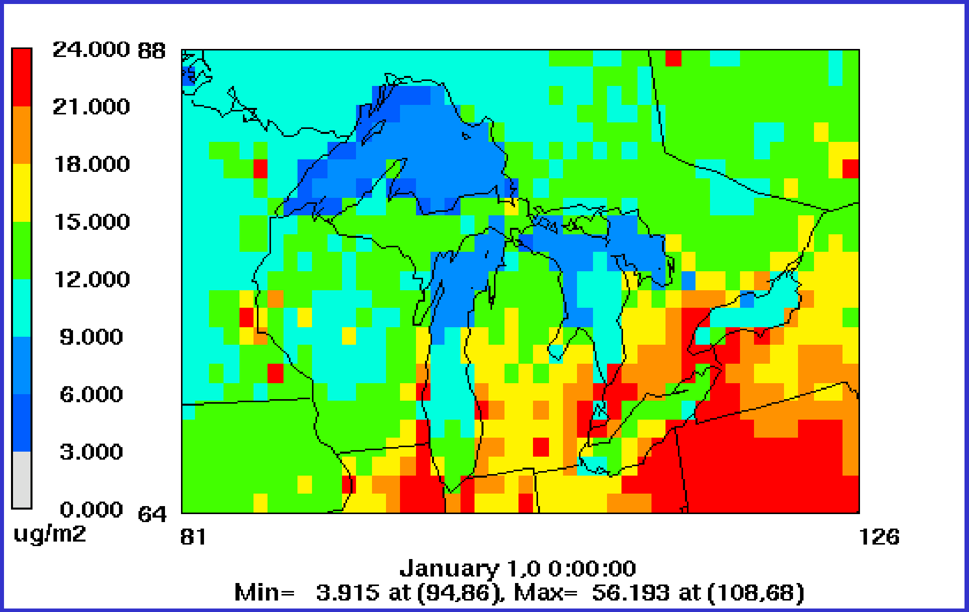
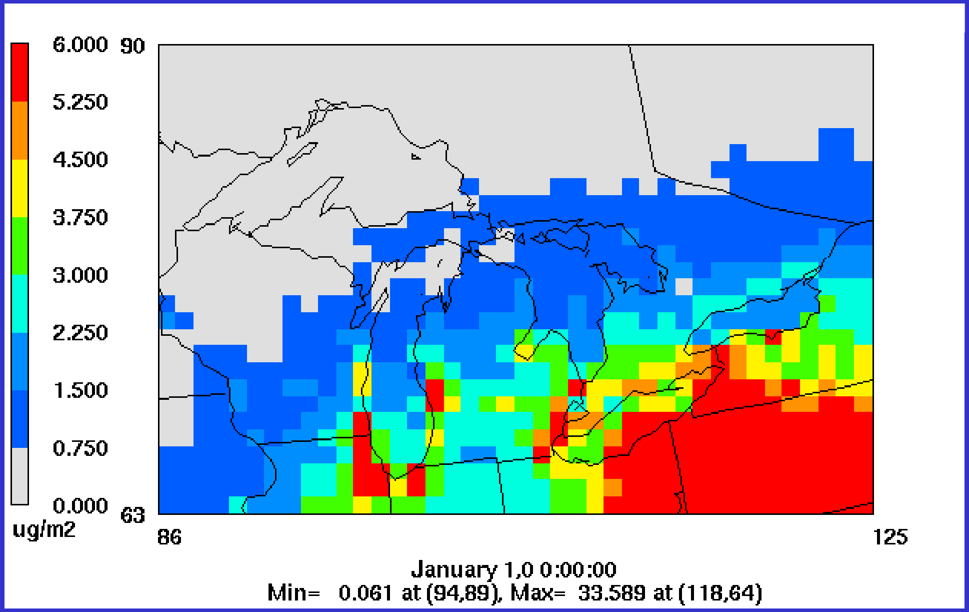
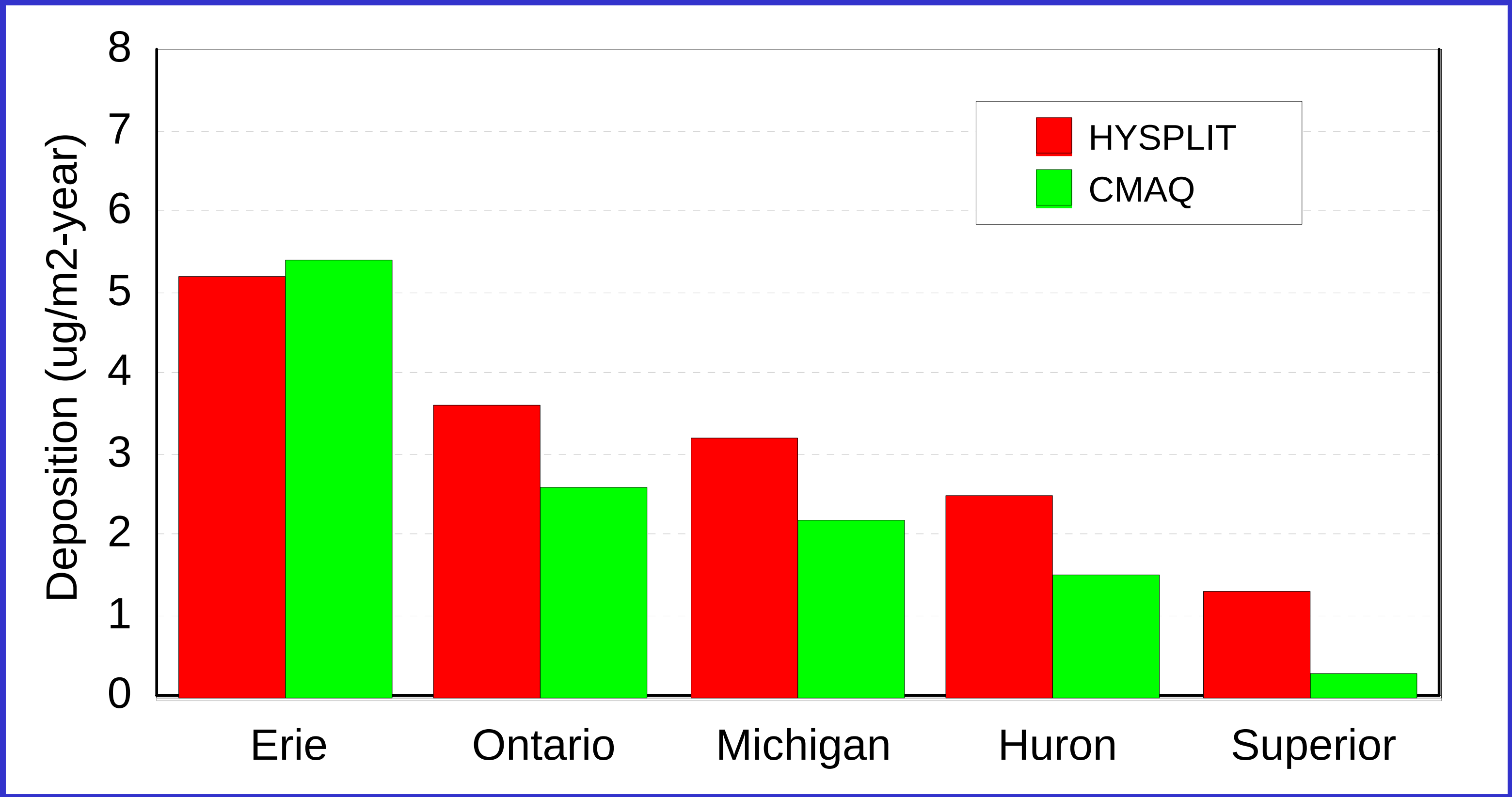
CMAQ-Hg results from EPA analysis performed for the Clean Air Mercury Rule



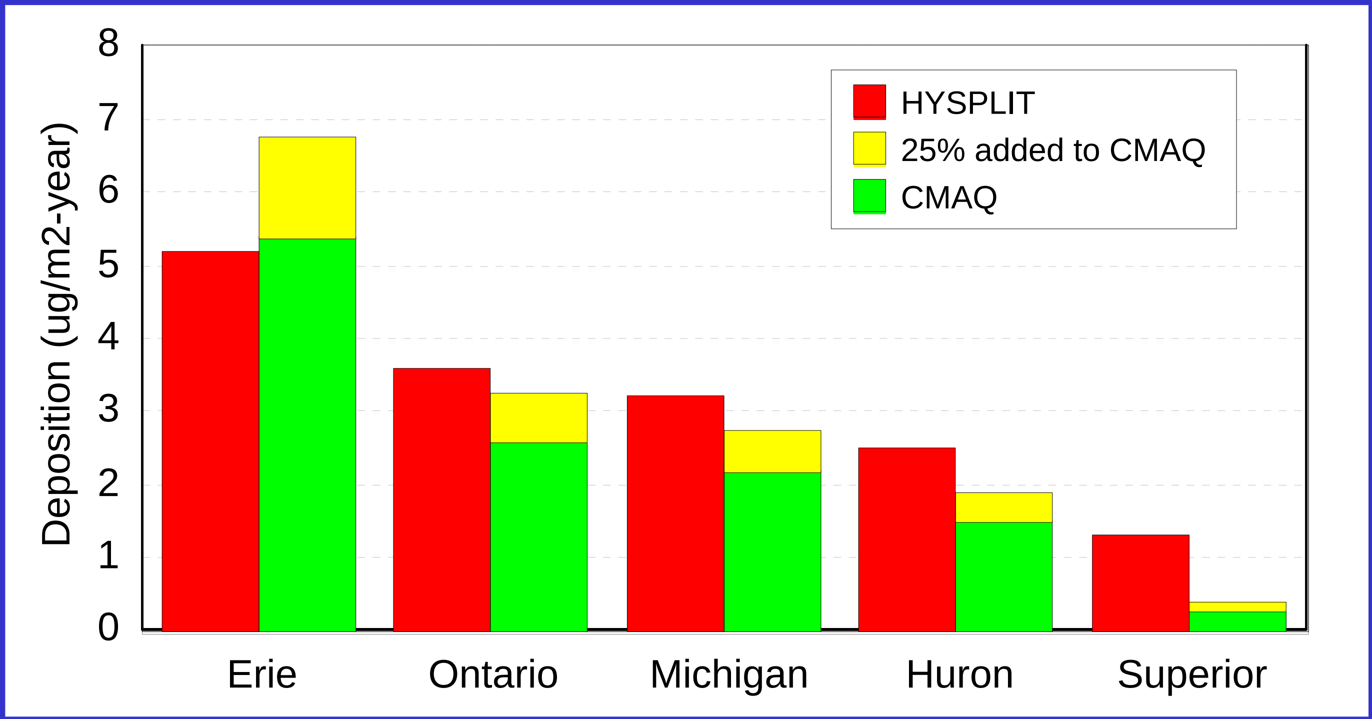
Modeled Mercury Deposition in the Great Lakes Region from all sources during 2001



Modeled Mercury Deposition in the Great Lakes Region attributable to U.S. coal-fired power plants during 2001



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).



- 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 30% 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.

Outline of Presentation

- modeling methodology
- some preliminary results for Mobile Bay
  (based on this methodology)
- model intercomparisons
- **summary of previous work; current goals; challenges**

Emissions Inventories	
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Current Objectives	• 2002 U.S. NEI • 2002 Canada • Global – 2000 (Pacyna-NILU) • Natural sources • Re-emitted anthropogenic
Challenges and Notes	• Speciation? • Short-term variations (e.g. hourly) [CEM's?] • Longer-term variations (e.g., maintenance)? • Mobile sources • Harmonization of source-categories • Emissions inventories currently only become available many years after the fact; how can we evaluate models using current monitoring data?

Meteorological Data	
Previous Work	• **For U.S./Canadian modeling, 1996 data from NOAA Nested Grid Model (NGM), 180 km**

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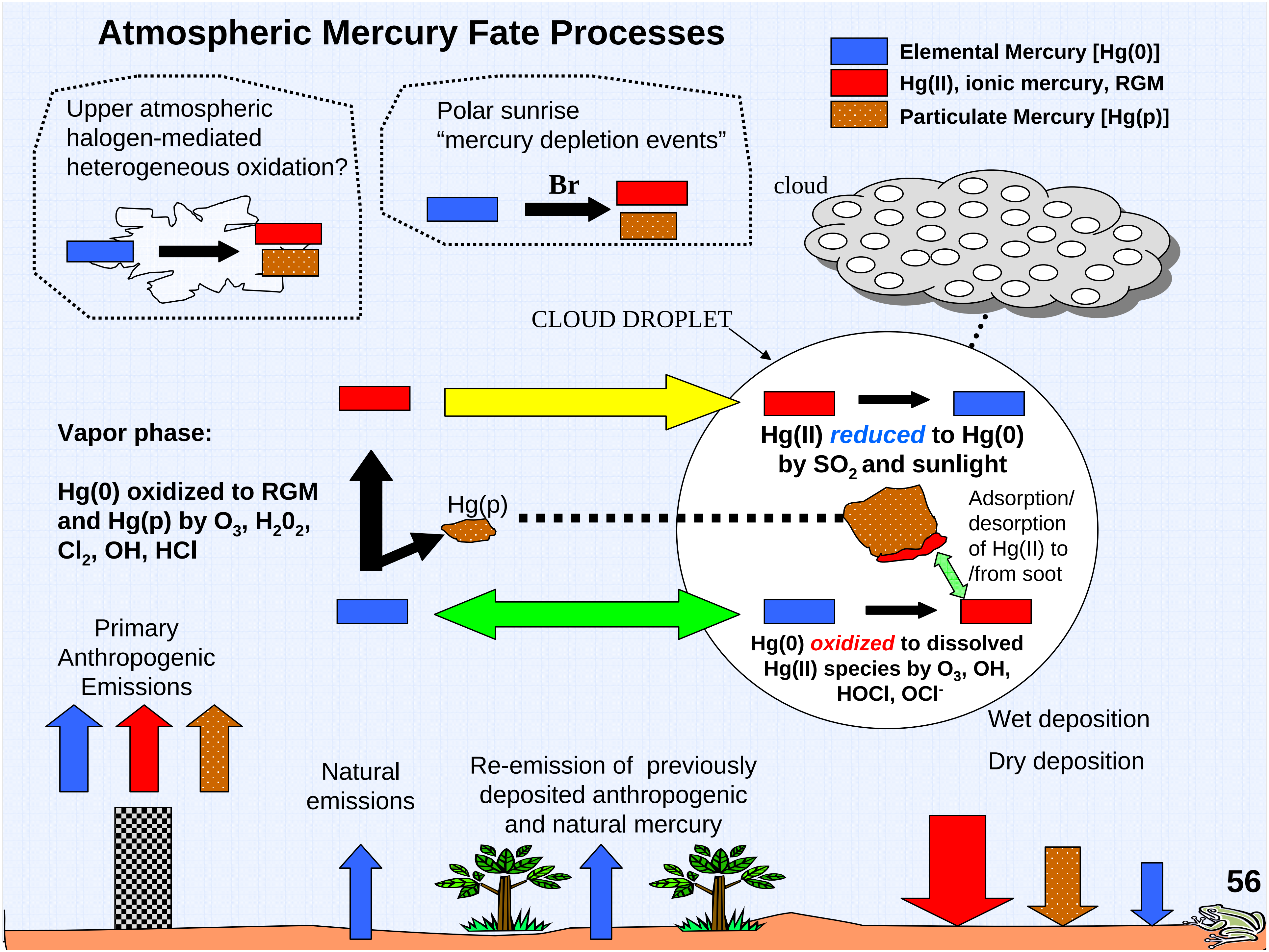
Challenges and Notes	• Forecast vs. Analysis • Data assimilation • Precipitation?? • Difficult to archive NOAA analysis datasets • Need finer-resolution datasets, especially for near-field analysis and model evaluation • We have conversion filters (e.g., for MM5), but these data are not readily available • What is the best way to archive and share data?

Atmospheric Chemistry and Physics	
Previous Work	• **Typical chemical mechanism** • **Prescribed fields for reactive trace gases (e.g., O_3, OH, SO_2) and other necessary constituents (e.g., soot) based on modeled, measured, and/or empirical relationships**

Atmospheric Chemical Reaction Scheme for Mercury

Reaction	**Rate**	**Units**	**Reference**
GAS PHASE REACTIONS			
$Hg^0 + O_3 \rightarrow Hg(p)$	3.0E-20	cm^3/molec-sec	Hall (1995)
$Hg^0 + HCl \rightarrow HgCl_2$	1.0E-19	cm^3/molec-sec	Hall and Bloom (1993)
$Hg^0 + H_2O_2 \rightarrow Hg(p)$	8.5E-19	cm^3/molec-sec	Tokos et al. (1998) (upper limit based on experiments)
$Hg^0 + Cl_2 \rightarrow HgCl_2$	4.0E-18	cm^3/molec-sec	Calhoun and Prestbo (2001)
$Hg^0 + OH\bullet \rightarrow Hg(p)$	8.7E-14	cm^3/molec-sec	Sommar et al. (2001)
AQUEOUS PHASE REACTIONS			
$Hg^0 + O_3 \rightarrow Hg^{+2}$	4.7E+7	$(\text{molar-sec})^{-1}$	Munthe (1992)
$Hg^0 + OH\bullet \rightarrow Hg^{+2}$	2.0E+9	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1997)
$HgSO_3 \rightarrow Hg^0$	$T*e^{((31.971*T)-12595.0)/T)}$ [T = temperature (K)]	sec^{-1}	Van Loon et al. (2002)
$Hg(II) + HO_2\bullet \rightarrow Hg^0$	~ 0	$(\text{molar-sec})^{-1}$	Gardfeldt & Jonnson (2003)
$Hg^0 + HOCl \rightarrow Hg^{+2}$	2.1E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$Hg^0 + OCl^{-1} \rightarrow Hg^{+2}$	2.0E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$Hg(II) \leftrightarrow Hg(II)_{(soot)}$	9.0E+2	liters/gram; t = 1/hour	eqlbrm: Seigneur et al. (1998) rate: Bullock & Brehme (2002).
$Hg^{+2} + h< \rightarrow Hg^0$	6.0E-7	$(sec)^{-1}$ (maximum)	Xiao et al. (1994); Bullock and Brehme (2002)


Atmospheric Mercury Fate Processes
Elemental Mercury [Hg(0)]
Hg(II), ionic mercury, RGM
Particulate Mercury [Hg(p)]
Upper atmospheric halogen-mediated heterogeneous oxidation?
Polar sunrise "mercury depletion events"
Br
cloud
CLOUD DROPLET
Vapor phase:
Hg(0) oxidized to RGM and Hg(p) by O3, H202, Cl2, OH, HCl
Hg(p)
Hg(II) reduced to Hg(0) by SO2 and sunlight
Adsorption/ desorption of Hg(II) to /from soot
Hg(0) oxidized to dissolved Hg(II) species by O3, OH, HOCl, OCl-
Primary Anthropogenic Emissions
Natural emissions
Re-emission of previously deposited anthropogenic and natural mercury
Wet deposition
Dry deposition
56

Atmospheric Chemistry and Physics	
Previous Work	• Typical chemical mechanism • Prescribed fields for reactive trace gases (e.g., O_3, OH, SO_2) and other necessary constituents (e.g., soot) based on modeled, measured, and/or empirical relationships
Current Objectives	• **Include new information on chemistry, e.g., bromine reactions, etc.** • **Sensitivity analyses** • **Use gridded chemical output from full-chemistry atmospheric model (e.g., CMAQ)** • **Option - run HYSPLIT in Eulerian mode for chemistry; conduct one-atmosphere simulation**

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Challenges and Notes	• **What is RGM?** • **What is Hg(p)?** • **What is solubility of Hg(p)?** • **Fate of dissolved Hg(II) when droplet dries out?** • **What reactions don't we know about yet?** • **What are rates of reactions?**

Model Evaluation	
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Current Objectives	• Attempt to utilize all available 2002-2005 speciated ambient concentrations and wet deposition data from U.S. and other regions
Challenges and Notes	• Comprehensive evaluation has not been possible due to large gaps in availability of monitoring and process-related data • Need data for upper atmosphere as well as surface • Need data for both source-impacted and background sites • Use of recent monitoring data with EPA 2002 inventory? • Time-resolved monitoring data vs. non-time-resolved emissions? • Hard to diagnose differences between models & measurements • Can we find better ways to share data for model evaluation (and other purposes)? To this end, discussion is beginning on national, cooperative, ambient Hg monitoring network

Thanks!

For more information on this modeling research:

http://www.arl.noaa.gov/ss/transport/cohen.html