Mercury Deposition in the US

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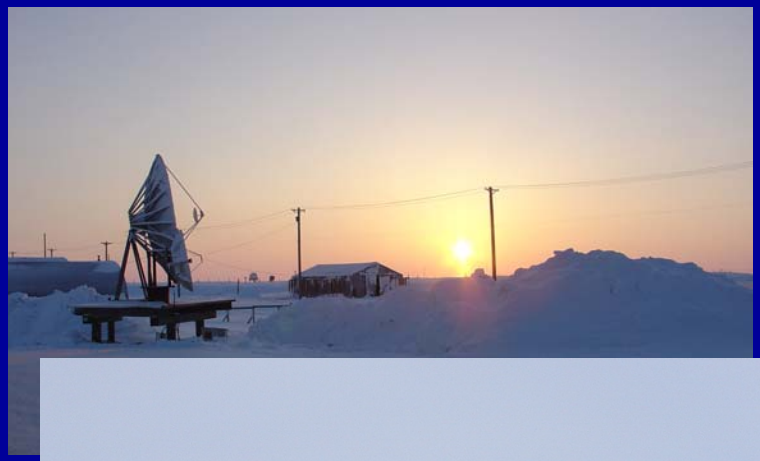
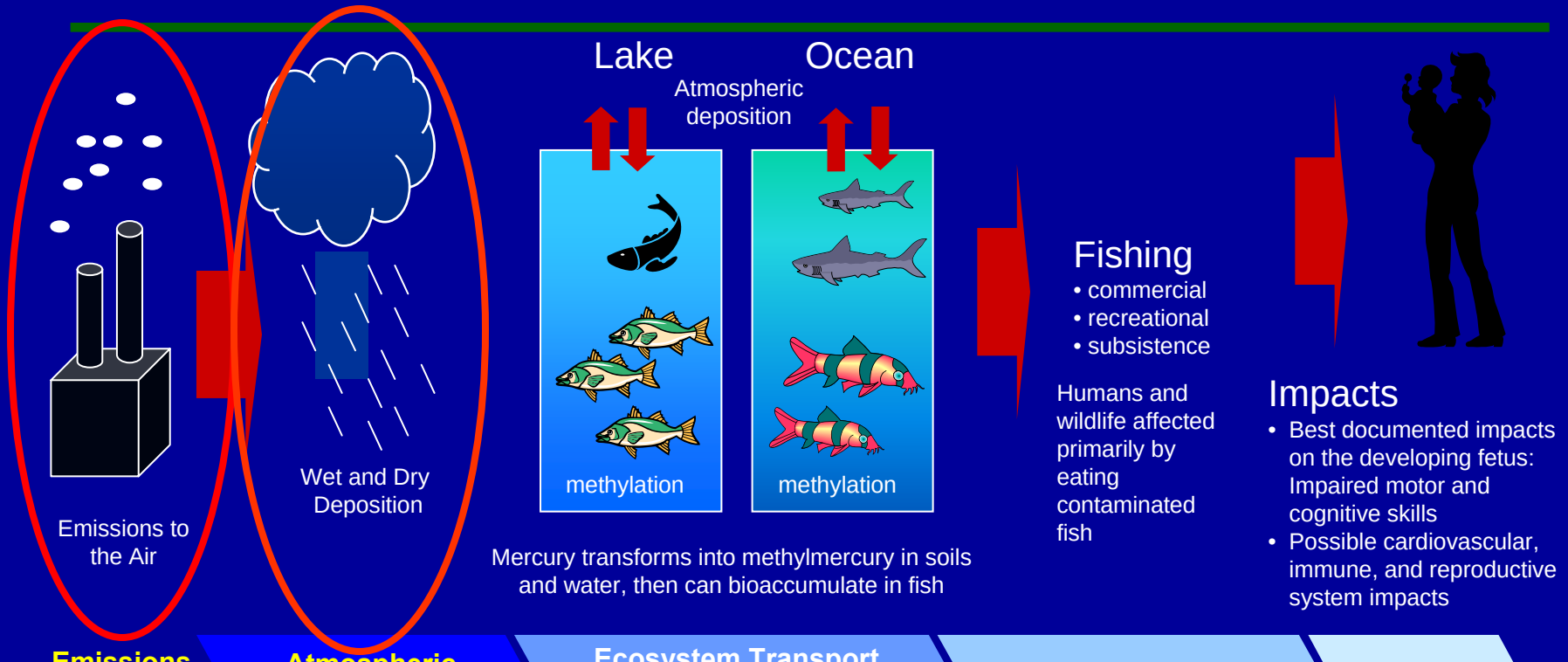
Today's Presentation

- Background
- Global Emissions Estimates for Hg
- Modeling the sources of Hg to the atmosphere
- Steubenville, OH Study
  - Levels and sources of wet deposition
  - Is this a deposition "Hot Spot"
- Dry Deposition
- What best predicts wet deposition of Hg?
- Summary

What have I done since the last time I spoke at the Endicott House Symposium



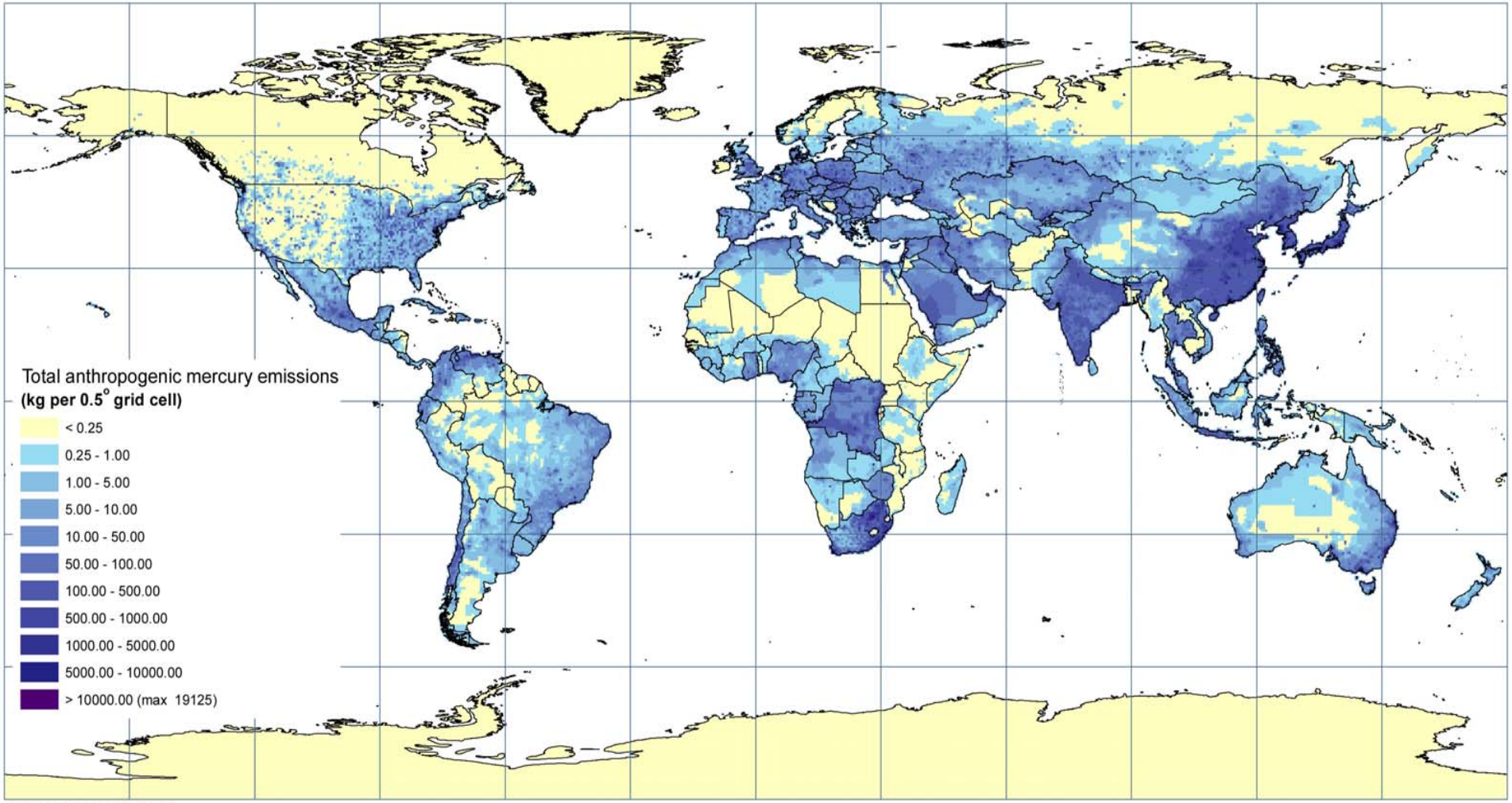
Mercury Emissions Contribute to Exposure to Mercury



- **The primary pathway of human exposure to mercury in the U.S. is through eating contaminated fish.**
- **Power plants emit approximately 48 tons of mercury and are the largest source of mercury emissions in the U.S. (approximately 41%).**

Spatially Distributed Inventories of Global Anthropogenic Emissions of Mercury to the Atmosphere, 2000

Total Hg, point sources + distributed sources, 0.5° grid



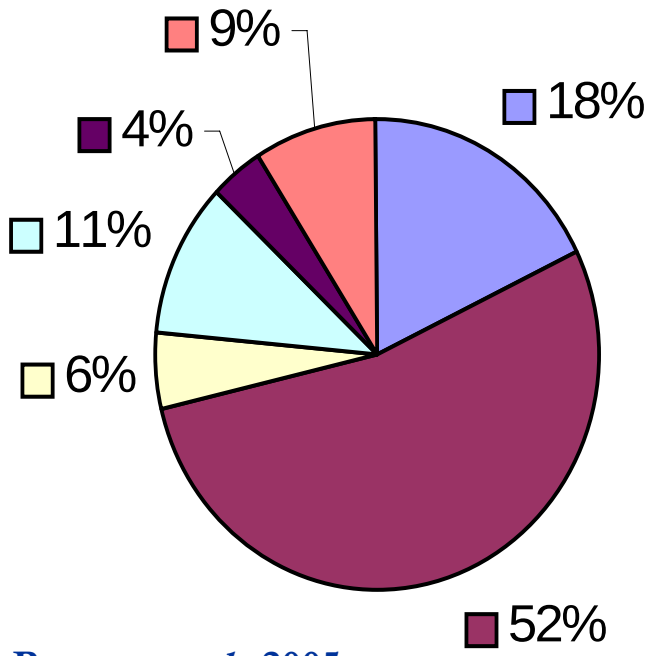
unprojected (geographic)

citation:
Pacyna, J., S. Wilson and F. Steenhuisen. 2005.
Spatially Distributed Inventories of Global Anthropogenic
Emissions of Mercury to the Atmosphere.
(www.amap.no/Resources/HgEmissions/HgInventoryMain.html)

S. Wilson (AMAP), F. Steenhuisen (Arctic Centre, RuG), J. Pacyna (NILU)

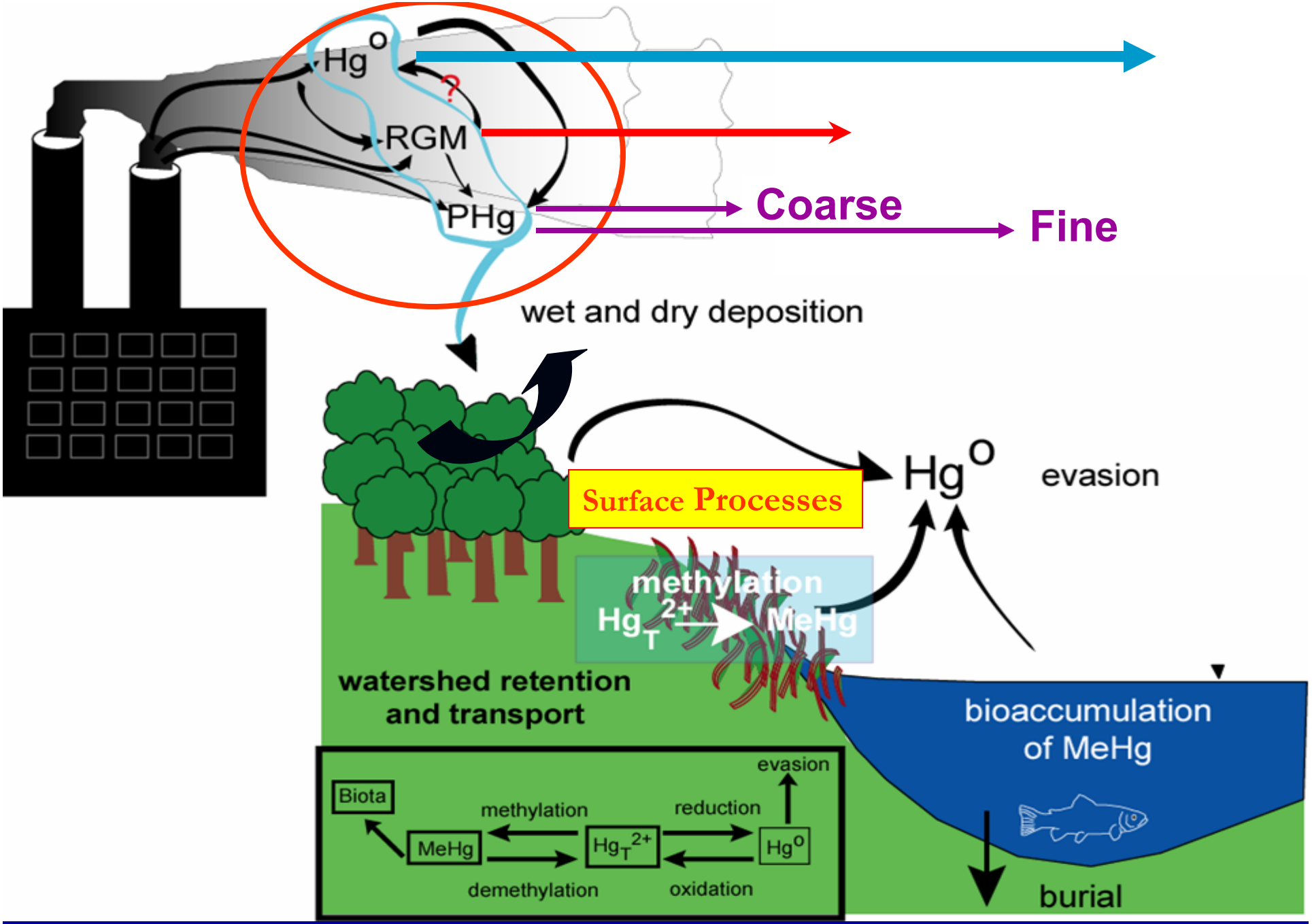
Worldwide Emissions

Total Hg Emissions = 2300 tonnes



Pacyna *et al.* 2005

Speciation of mercury emissions the most important factor!
Form of mercury emitted varies greatly by source type.



Modified from cartoon by Charles Driscoll, Syracuse University

Source Apportionment Modeling

- Relates sources and environmental concentrations
- Approaches to source apportionment

  Deterministic modeling (e.g., CMAQ)
  - Requires emission inventory, chemistry, and meteorology
  - Models emission source impacts on predicted concentrations

  Receptor modeling
  - Requires comprehensive environmental measurements
  - Statistically identifies sources impacting measured concentrations
- Approaches are independent and complementary

Mercury Modeling Limitations

Current Deterministic Models

- Global speciated emission inventories
  - Data improving but incomplete
- Incomplete understanding of Hg kinetics
- Precipitation prediction often problematic
- Wet and dry deposition parameterizations
  - Insufficient handling of air-surface coupling, e.g. Hg^0 uptake by vegetation.

Multivariate Receptor Modeling

- Identify major “factors” by statistical analysis of an element measurement matrix
- Relate “factors” to source types using tracer compounds
- Example tracers
  - Coal Combustion – S, Se; Oil Combustion – Ni, V
- Requires many samples (150 +)

Ohio Mercury Measurement and Receptor Modeling Study

Applied both positive matrix factorization (PMF) and UNMIX models

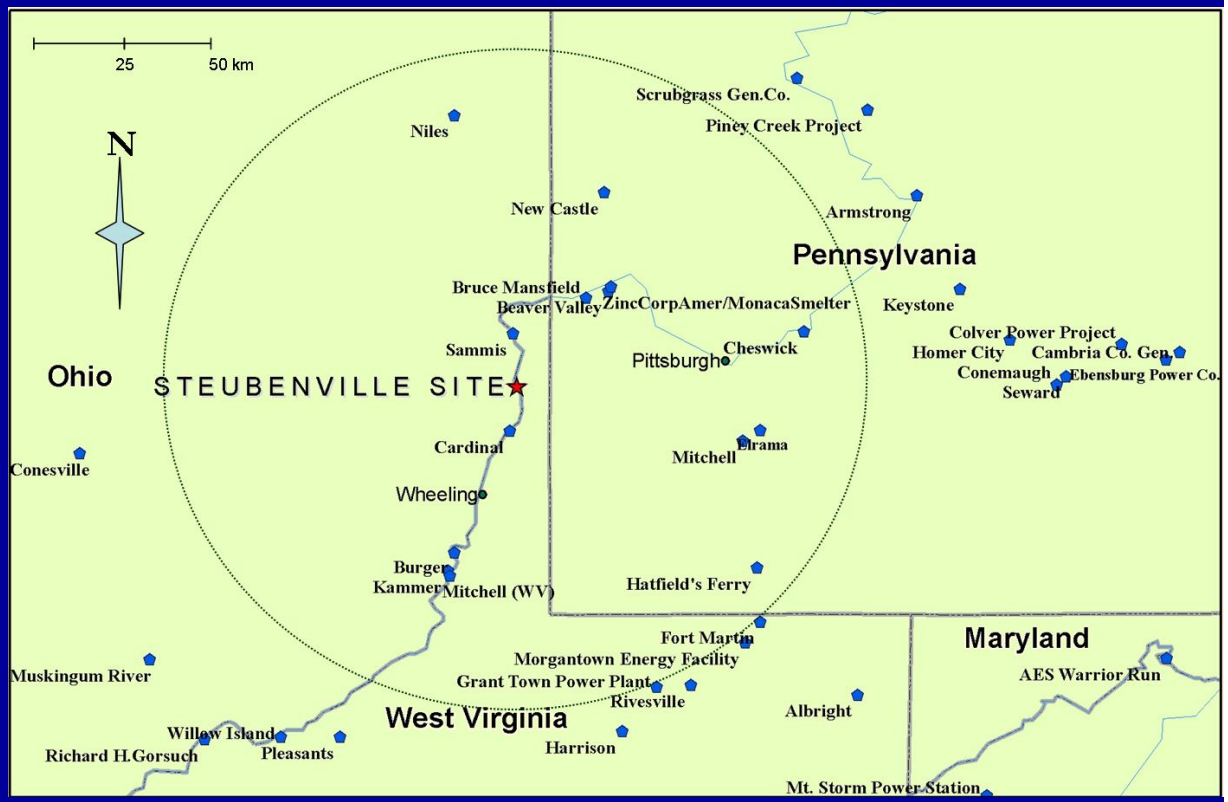
- Estimated source contributors to measured event wet Hg deposition

Uncertainty Analysis

- Both PMF & Unmix incorporate Bootstrap Uncertainty Analysis and provide the total uncertainty with the mean:
  - Fits the 5^{th} & 95^{th} Percentile in distribution of profiles.

Primary Objective of Study

Determine the impact of local/regional coal combustion sources on Hg deposition in the ORV.



Ohio Mercury Study

- Cooperative Research with EPA ORD
- Sate-of-the-art measurement/analyses
  - Aerosols - Integrated and Continuous
  - Wet Deposition - Daily Event
  - Criteria Gases - Continuous
  - Meteorology - Continuous
- Receptor Modeling
  - UNMIX and PMF
  - Hybrid Modeling (Regional Transport)

Steubenville PMF Results

2003 & 2004

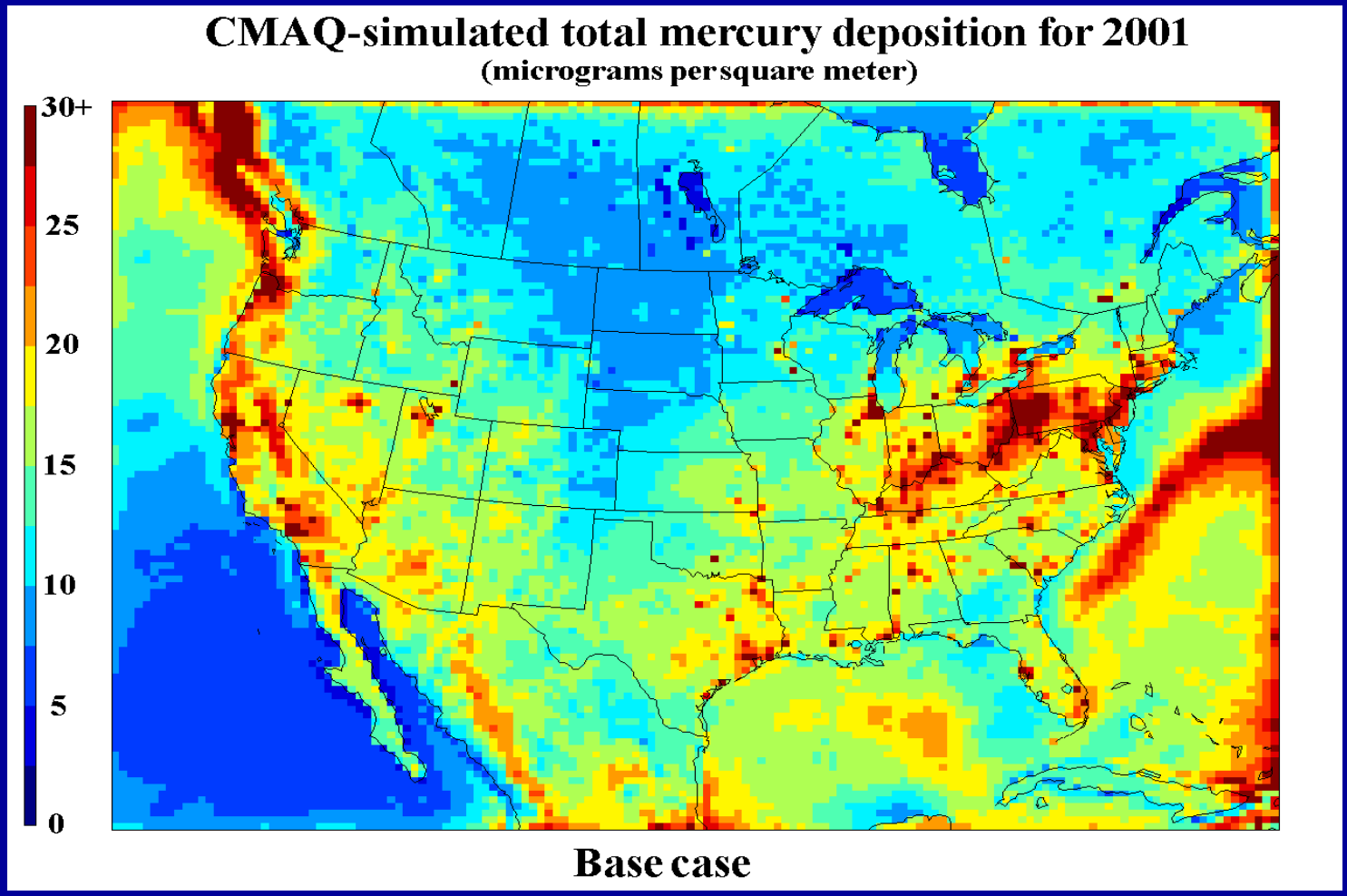
Analyte	Source 1 Iron/Steel Production	Source 2 Oil & Incineration	Source 3 Crustal	Source 4 Coal Combustion	Source 5 Phosphorous	Source 6 Molybdenum
Mg	187	*	558	*	101	*
Al	51	80	355	37	*	52
P	7.8	*	*	*	63.8	*
S	*	*	642	11299	197	*
Cl	267	20480	*	584	*	771
V	2.9	1.1	*	*	*	*
Cr	2.5	*	*	*	*	*
Mn	54.4	*	34.1	*	15.4	*
Fe	344	102	17	37	27	*
Ni	*	3.19	*	*	0.68	*
Cu	1.8	14.0	*	18.4	2.7	7.0
Zn	4.0	44.1	6.1	10.7	5.3	15.6
As	*	0.81	0.10	0.49	0.05	0.27
Se	*	0.97	*	1.73	*	1.30
Sr	0.48	3.30	5.64	0.95	1.61	*
Mo	*	*	*	*	*	4.02
Cd	0.09	0.27	*	0.31	0.02	0.23
La	*	0.13	0.63	*	*	0.04
Ce	0.02	*	1.23	*	*	*
Hg	0.01	*	*	0.15	< 0.01	*
Pb	1.10	6.59	0.59	3.62	0.36	1.13
NO_3	*	8639	1501	4532	314	*
% Hg	6	*	*	73	2	*

Source Apportionment Results
Steubenville, Ohio

	Measured	PMF Estimated CFUB* Contribution	UNMIX Estimated CFUB* Contribution
2003	13.5	Mean = 9.1 (5-95% CIΩ) = (6.4 – 14.7)	Mean = 9.9 (5-95% CIΩ) = (5.9 – 15.1)
2004	19.7	Mean = 13.1 (5-95% CIΩ) = (9.3 – 21.4)	Mean = 15.5 (5-95% CIΩ) = (9.1 – 23.1)

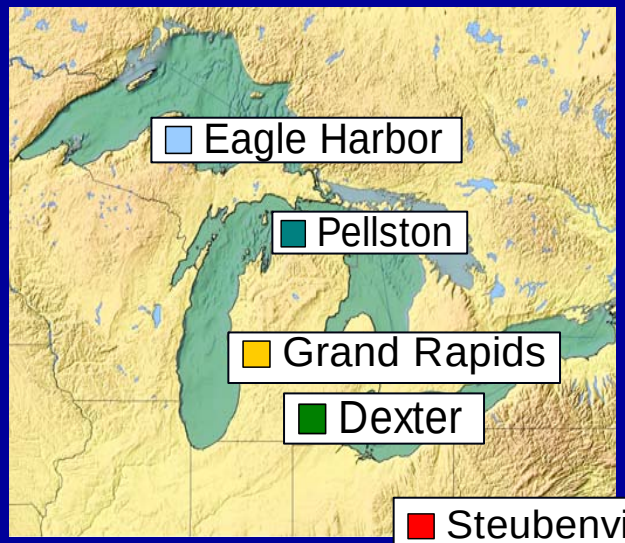
*Coal-fired Utility Boiler
Ω Confidence Interval

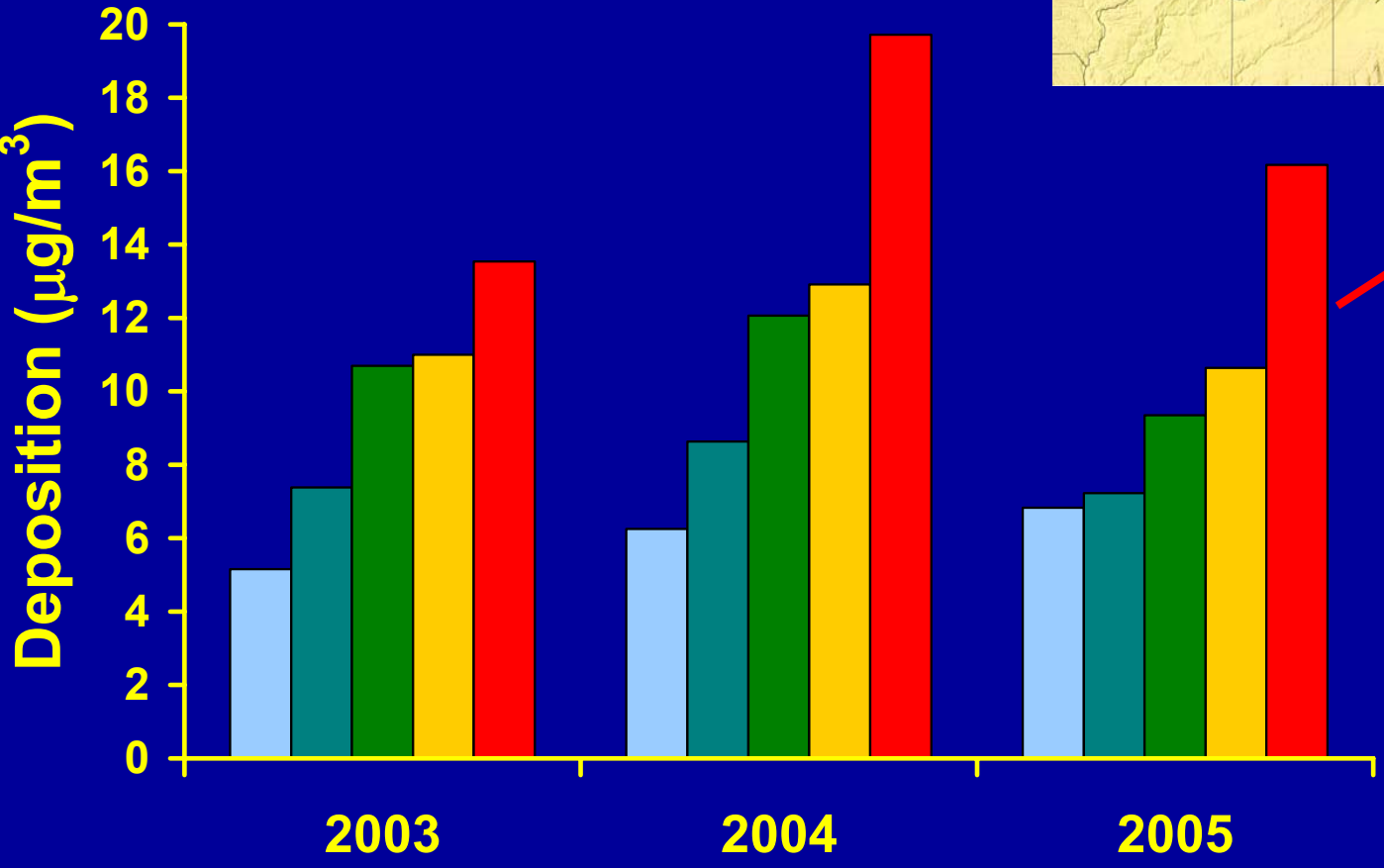
Modeling Used for Clean Air Mercury Rule



CMAQ Simulations performed by CSC for OAQPS (6FEB04)

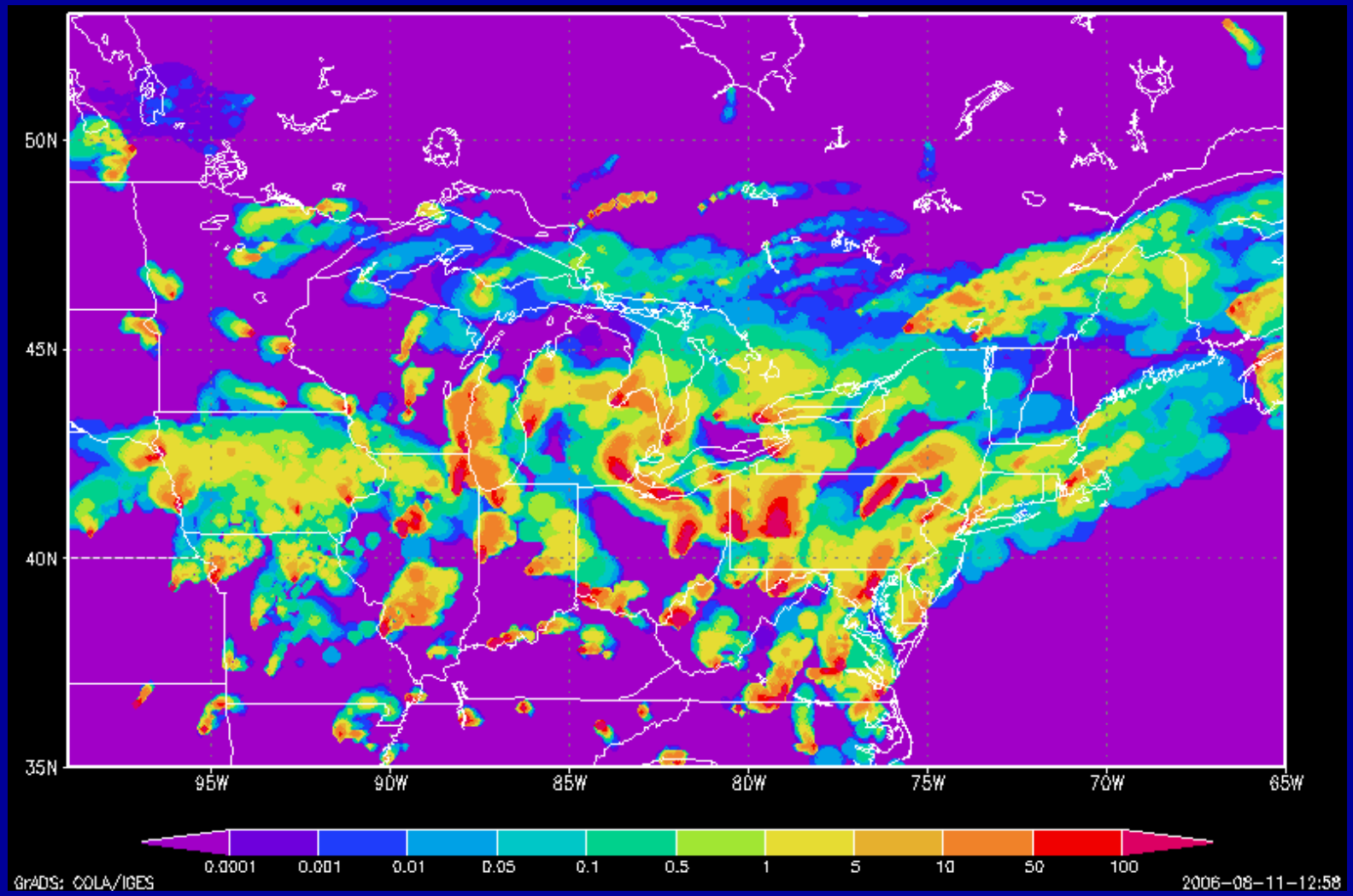
2003 – 2005 Great Lakes Deposition Comparison





Dry Deposition of Hg

August 10-11, 2006



Comparison of USEPA CMAQ Results and Measured Mercury Wet Deposition at Steubenville

	Hg Deposition ($mg\ m^{-2}y^{-1}$)	CFUB Contribution (%)
CMAQ 2001	13.6 (modeled)	43
PMF/UNMIX 2003-2004	16.5 (measured)	72

CMAQ Simulations performed by CSC for EPA (6FEB04)

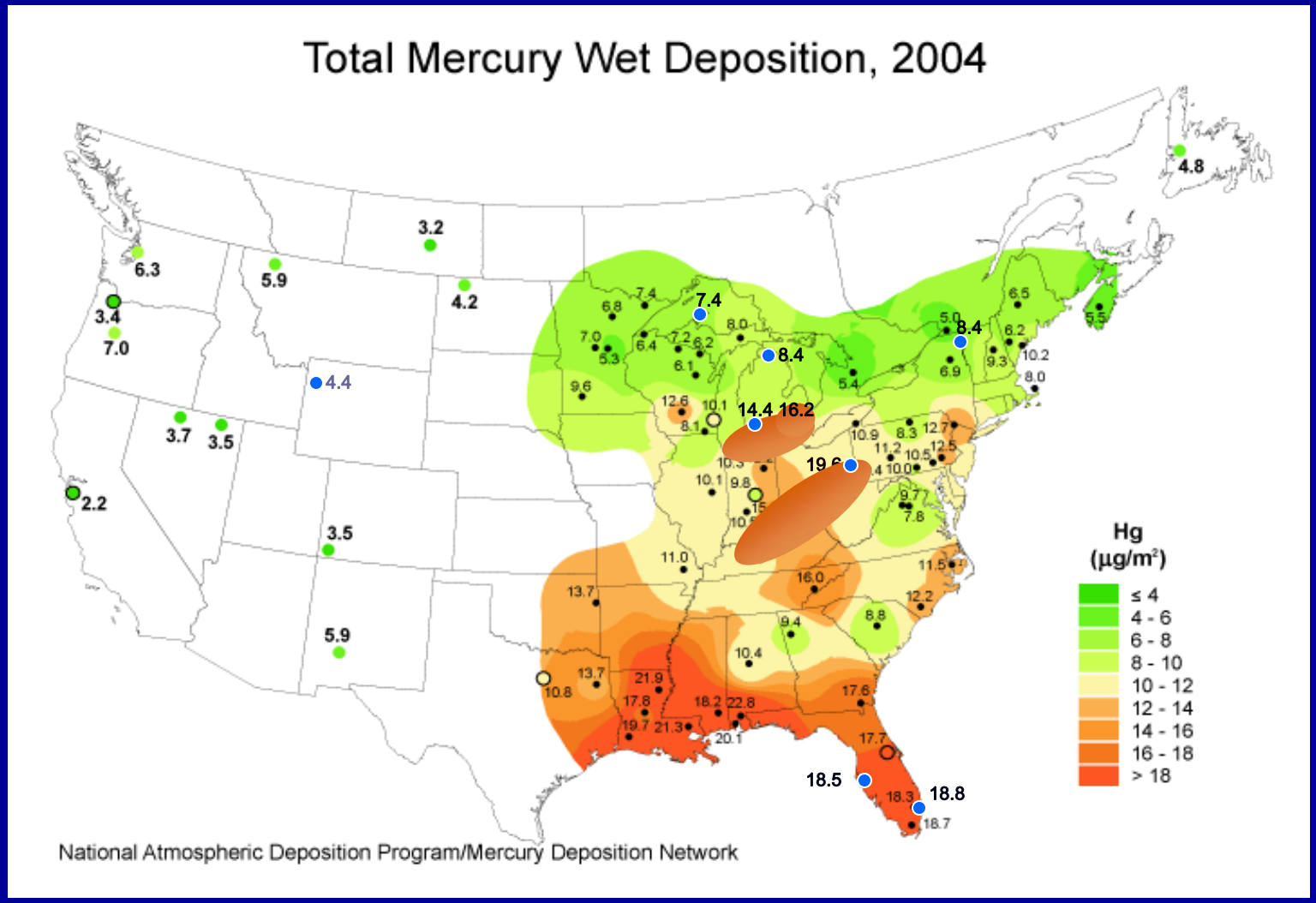
CMAQ Modeled versus UMAQL Measured Hg Wet Deposition

All 2001 Results

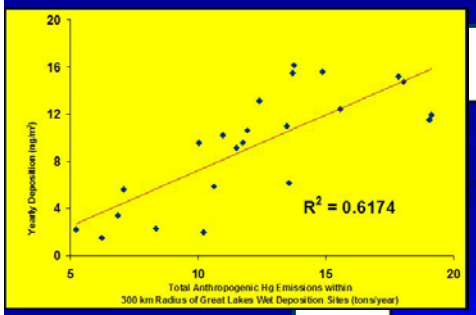
Site	CMAQ Wet Deposition ($\mu g\ m^{-2}$)	Measured ($\mu g\ m^{-2}$)
Dexter, MI	8.3	12.5
Pellston, MI	4.6	10.5
Eagle Harbor, MI	4.7	7.7
Underhill, VT	4.4	8.6

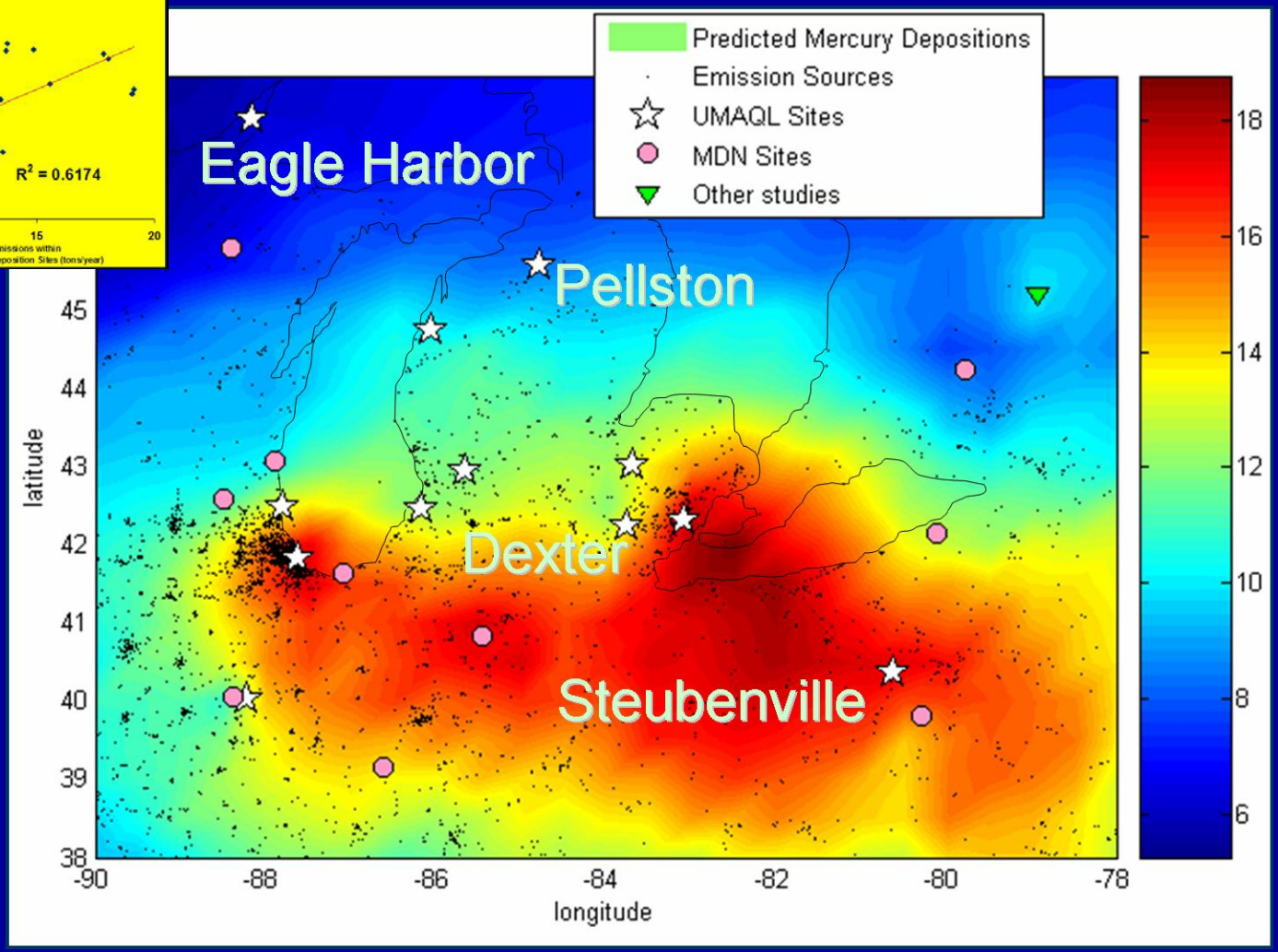
CMAQ Results provided by Russ Bullock., USEPA

UMAQL EVENT SAMPLING SITES



What Best Predicts Hg Deposition?





Summary

- **Hg wet deposition at Steubenville**
  - **~ 80% attributable to local/regional anthropogenic sources**
  - **~ 70% is attributable to coal combustion**
  - **~ 20% from reemission/global background**
- **A significant portion of total Hg wet deposition is driven by a few local coal combustion dominated precipitation events**
  - **In 2004, >8% of Hg wet deposition occurred during 1 event and understanding the source(s) contributing important.**
- **Dry deposition even more local in origin and due to the bi-directional flux of Hg these measurements must be done on a high frequency.**

Acknowledgements

- Funding and other support provided by:
  - National Science Foundation
  - Michigan Great Lakes Protection Fund
  - EPA Great Lakes National Program Office
  - EPA Region V
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- Collaborators at MI DEQ including Joy Taylor-Morgan