Opportunities for Mercury Collaboration between the Air Resources Laboratory and the Environmental Research Program

Dr. Mark Cohen

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meeting with Environmental Research Program February 16, 2005, Silver Spring, MD



Outline of Presentation

- ☐ brief overview of Air Resources Laboratory
- ☐ mercury problem / role of atmospheric Hg
- ☐ ARL mercury programs and collaborations
- opportunities for collaboration within ERP

overview of the Air Resources Laboratory

Air Resources Laboratory

Transport Modeling & Assessment Climate Variability & Trends Air Surface Exchange & Chemistry Radiation and Aerosols

19 Federal employees 4 Contractors

Exec. Assistant: Betty Wells Administration: Sharon Hamilton

Headquarters
Silver Spring, Maryland
Director B. B. Hicks
Deputy R. S. Artz

Surface Radiation Research Branch Boulder, CO J. J. Michalsky

4 Federal employees

11 CIRES 3 Others

Secretary: Gwen Andersen

Atmospheric Turbulence and Diffusion Division Oak Ridge, TN Director R. P. Hosker Deputy T. P. Meyers

Atmospheric Sciences
Modeling Division
Research Triangle Park, NC
Director S. T. Rao
Deputy W. B. Petersen

Field Research
Division
Idaho Falls, ID
Director K. L. Clawson
Deputy T. B. Watson

Special Operations and Research Division Las Vegas, NV Director D. Randerson Deputy D. A. Soule

Dispersion Studies Air-Surface Interactions Measurement Technologies Aircraft Operations

12 (+1) NOAA employees 23 Contractors (ORAU) 6 Others

Secretary: Sharon Conger Administration: Barbara Shifflett Atmospheric Model Development Air-Surface Processes Modeling Model Evaluation and Applications Air Policy Support

50 NOAA employees 4 Contractors 23 Others

Secretary: Patricia McGhee Administration: Herb Viebrock Dispersion Analyses and Modeling INEEL Meteorology & Dispersion Atmospheric Tracers

10 NOAA employees
3 Contractors

Secretary: Joyce Silvester Administration: Paulette Fee Dispersion Studies NTS Meteorology and Dispersion Technical Services

17 NOAA employees 3 Contractors CIASTA (JI)

Secretary: Boots Parker Administration: Barbara Pierce



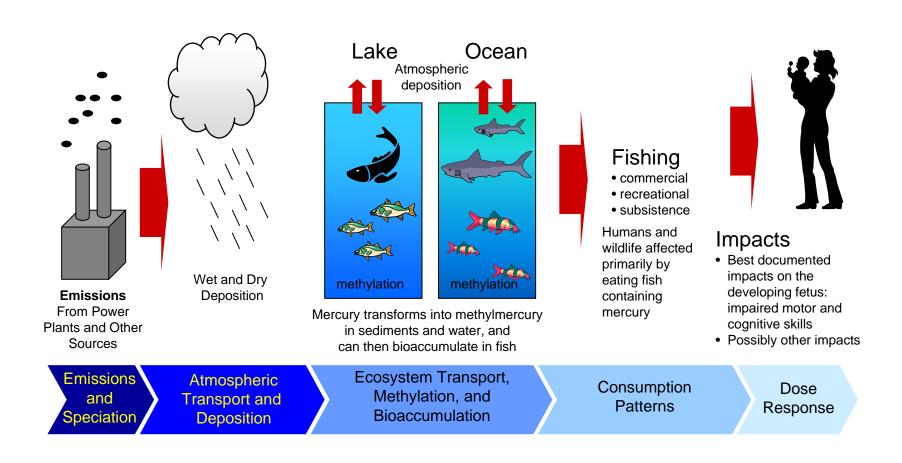


the mercury problem and the role of atmospheric mercury

The Mercury Problem

■ EPA has estimated that 1 out of every 6 children born in the U.S. have already been exposed in-utero to levels of mercury that may cause problems with neurological development ☐ There are additional potential mercury-related health hazards to children, adults, and to wildlife ☐ Fish-consumption advisories due to mercury contamination are widespread throughout U.S. rivers, lakes, and coastal areas ☐ The primary exposure route is through fish consumption ☐ Atmospheric deposition is a significant – often the most significant – pathway for mercury loading to aquatic ecosystems

Mercury Exposure Pathway



source: USEPA

Three "forms" of atmospheric mercury

Elemental Mercury: Hg(0) • ~ 95% of total Hg in atmosphere • not very water soluble • long atmospheric lifetime (~ 0.5 - 1 yr); globally distributed **Reactive Gaseous Mercury ("RGM")** • a few percent of total Hg in atmosphere • oxidized mercury: Hg(II) • HgCl2, others species? somewhat operationally defined by measurement method • very water soluble • short atmospheric lifetime (~ 1 week or less); more local and regional effects Particulate Mercury (Hg(p) • a few percent of total Hg in atmosphere not pure particles of mercury... (Hg compounds associated with atmospheric particulate) • species largely unknown (in some cases, may be HgO?) • moderate atmospheric lifetime (perhaps 1~ 2 weeks) local and regional effects • bioavailability?

Atmospheric Fate Processes for Hg

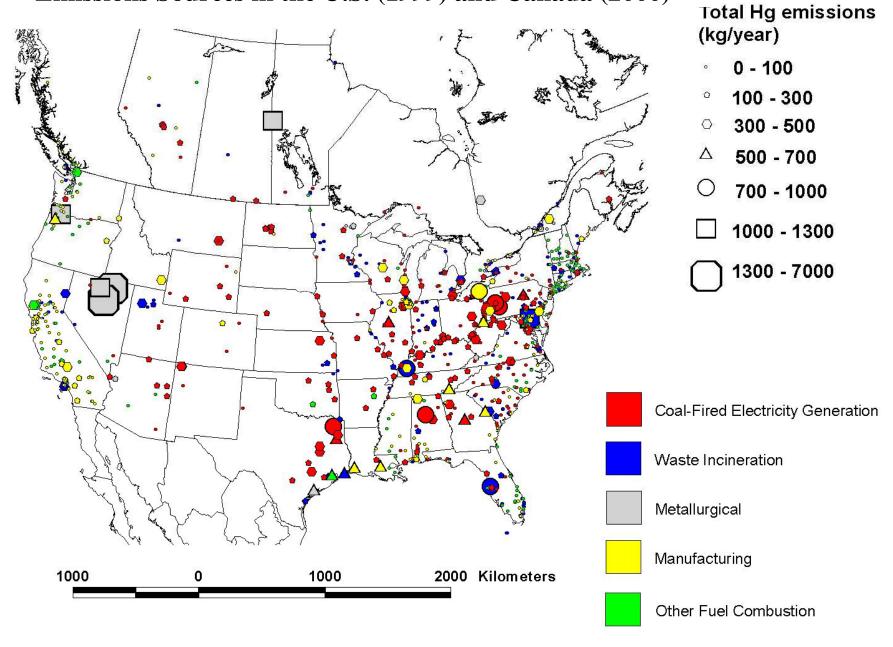
Elemental Mercury: Hg(0)
Reactive Gaseous Mercury: RGM
Particulate Mercury: Hg(p)

Upper atmospheric Polar sunrise halogen-mediated "mercury depletion events" cloud heterogeneous oxidation? **CLOUD DROPLET** "DRY" (low RH) Hg(II) reduced to Hg(0) by SO₂ ATMOSPHERE: Adsorption/ Hg(p)Hg(0) oxidized to RGM desorption of Hg(II) to by O₃, H₂O₂, Cl₂, OH, HCl /from soot **Primary** Hg(0) oxidized to dissolved Anthropogenic RGM by O₃, OH, HOCI, OCI **Emissions** Re-emission of natural Dry and Wet Deposition AND previously deposited anthropogenic mercury

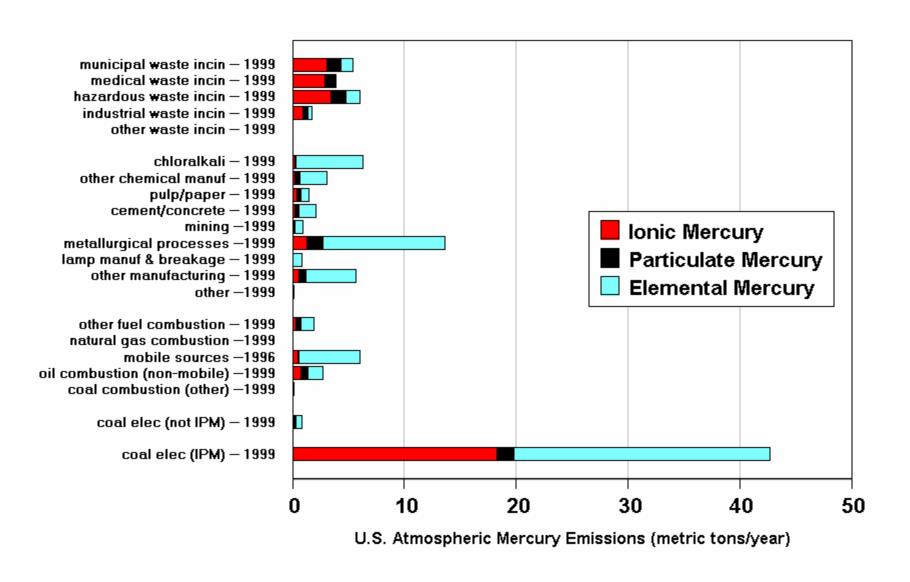
Atmospheric Chemical Reaction Scheme for Mercury

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)	
$\mathbf{Hg^0} + \mathbf{Cl_2} \rightarrow \mathbf{HgCl_2}$	4.0E-18	cm ³ /molec-sec	Calhoun and Prestbo (2001)	
$Hg^0 + OHC \rightarrow Hg(p)$	8.7E-14	cm ³ /molec-sec	Sommar et al. (2001)	
AQUEOUS PHASE REACTIONS				
$Hg^0 + O_3 \rightarrow Hg^{+2}$	4.7E+7	(molar-sec)-1	Munthe (1992)	
$Hg^0 + OHC \rightarrow Hg^{+2}$	2.0E+9	(molar-sec)-1	Lin and Pehkonen(1997)	
$HgSO_3 \rightarrow Hg^0$	$T*e^{((31.971*T)-12595.0)/T)}$ sec ⁻¹ [T = temperature (K)]		Van Loon et al. (2002)	
$\mathbf{Hg}(\mathbf{II}) + \mathbf{HO}_2\mathbb{C} \rightarrow \mathbf{Hg}^0$	~ 0	(molar-sec)-1	Gardfeldt & Jonnson (2003)	
$Hg^0 + HOCl \rightarrow Hg^{+2}$	2.1E+6	(molar-sec)-1	Lin and Pehkonen(1998)	
$Hg^0 + OCl^{-1} \rightarrow Hg^{+2}$	2.0E+6	(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)	

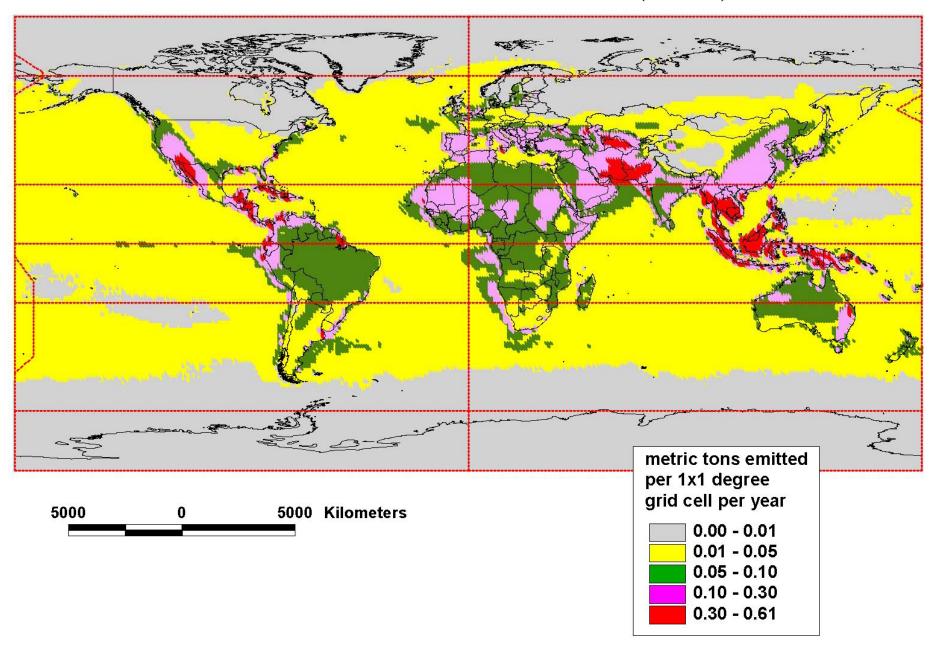
Geographic Distribution of Largest Anthropogenic Mercury Emissions Sources in the U.S. (1999) and Canada (2000)



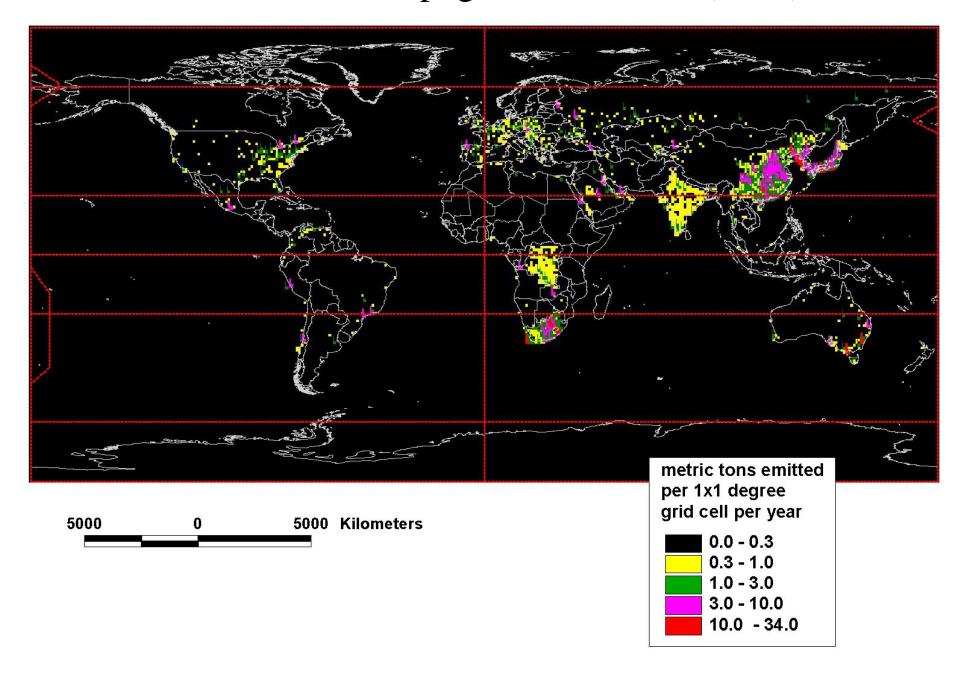
Estimated 1999 U.S. Atmospheric Anthropogenic Mercury Emissions



estimated natural emissions (1999)

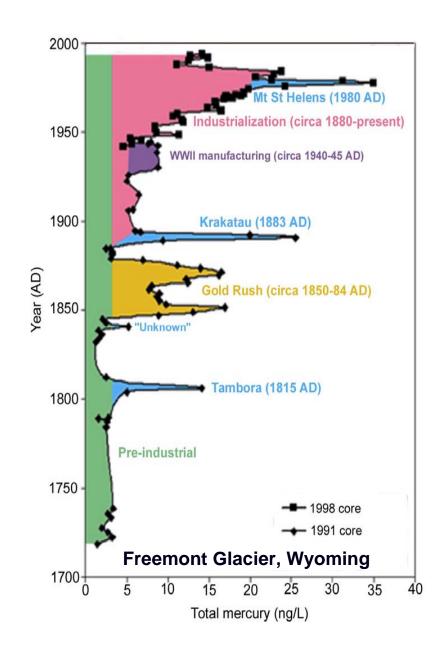


estimated anthropogenic emissions (1995)



Natural vs. anthropogenic mercury?

Studies show that anthropogenic activities have typically increased bioavailable Hg concentrations in ecosystems by a factor of 3-10



source: USGS, Shuster et al., 2002

Air Resources Laboratory: Atmospheric Mercury Research and Collaborations

ARL Mercury Research

Atmospheric measurements

- process understanding,
- study spatial/temporal trends
- develop & evaluate models
 - ground-level speciated air concentrations
 - upper-air speciated air conc. using aircraft
 - wet and dry deposition
 - surface exchange

Atmospheric modeling

- to interpret measurements,
- to get source-receptor data,
- to predict future impacts
 - back-trajectory modeling using HYSPLIT
 - HYSPLIT-Hg atmos. fate and transport model
 - CMAQ-Hg atmospheric fate and transport model

ARL Mercury Collaborations

- ☐ Within NOAA
- ☐ External to NOAA

Examples of Mercury Collaborations within NOAA

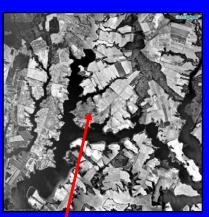
- ☐ NOAA Chesapeake Bay Office
 - Maggie Kerchner, Bob Wood
 - ongoing measurement and modeling study FY 2004-2005
 - proposal in the works for continued collaboration
- □ NOAA Arctic Program Office
 - with CMDL, ORNL, EPA, others
 - measurements and modeling at Barrow Alaska
- **and discussions for potential projects with:**
 - Lake Champlain Sea Grant
 - MS/AL Sea Grant (Gulf of Mexico)
 - NCCOS (Jawed Hameedi, David Whitall)

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Summer 2004 Chesapeake Bay Atmospheric Mercury Measurement Sites



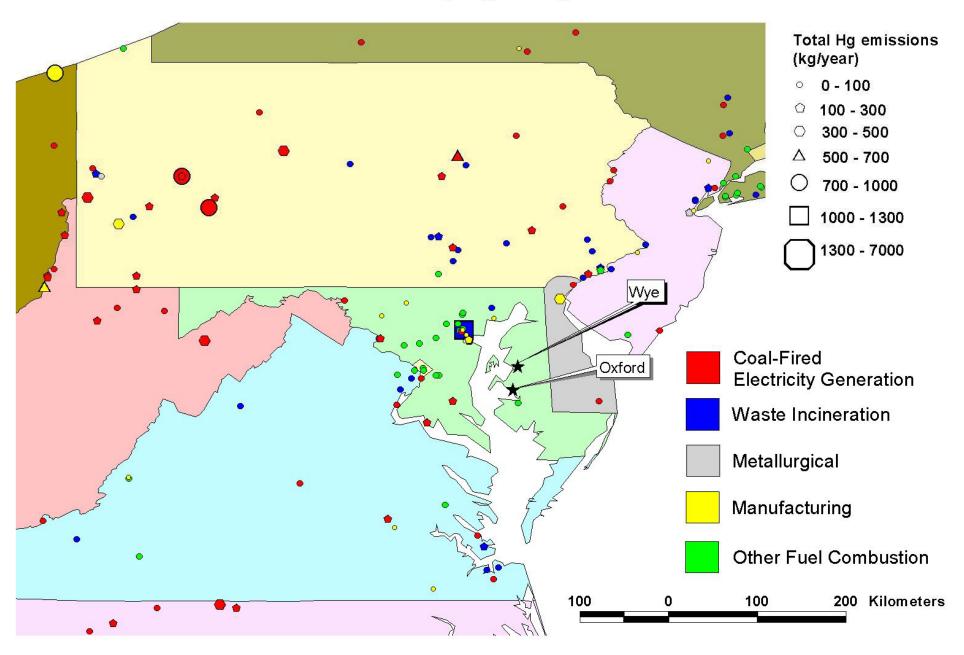


Wye Research and Education Center (38.9131EN, 76.1525EW)



Cooperative Oxford Lab (38.678EN, 76.173EW)

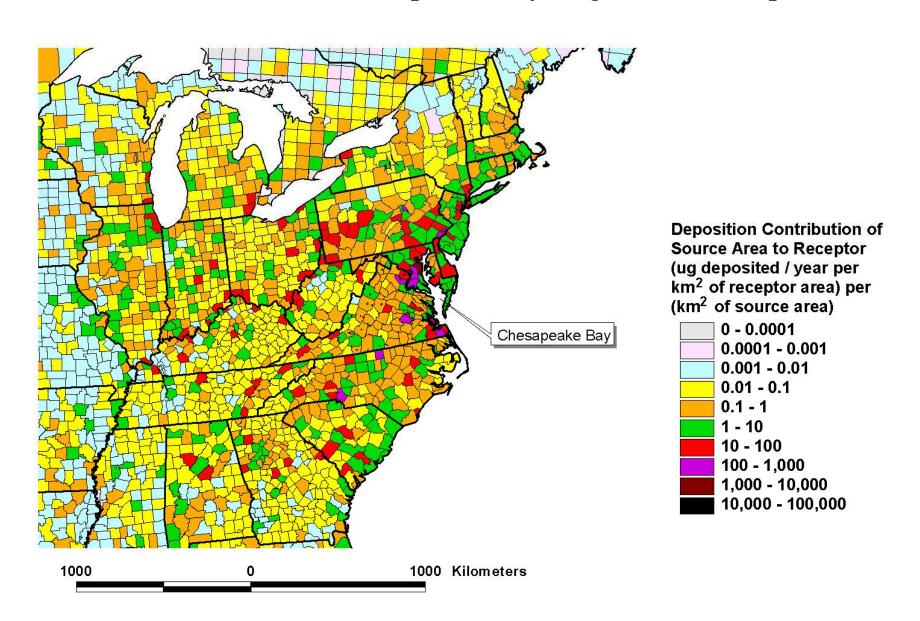
regional emissions (1999) and sampling sites for summer 2004 Ches Bay Hg study

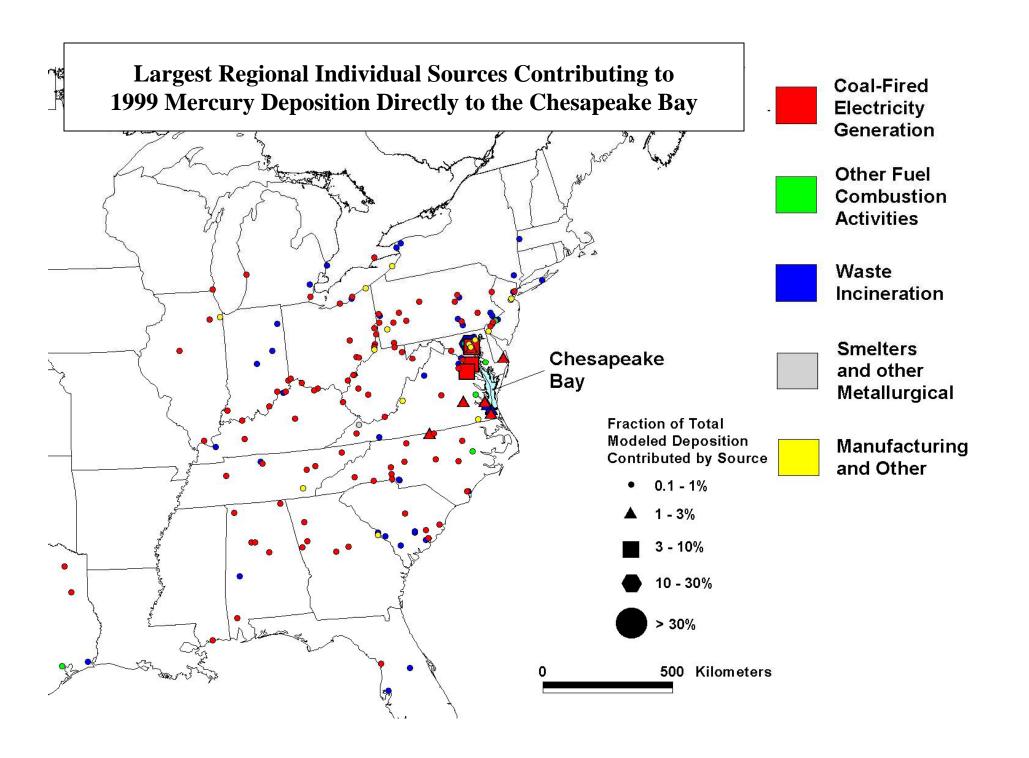


Summer 2004 Chesapeake Bay Atmospheric Hg Study (June – August 2004)

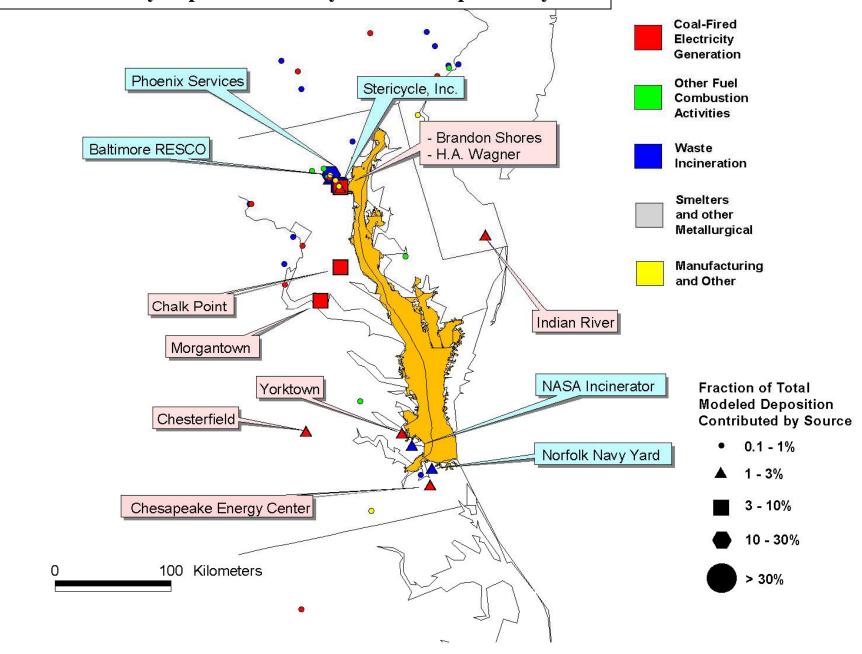
	Oxford	Wye
Event-based precipitation samples analyzed for Hg	✓	✓
Speciated Hg concentrations in ambient air (RGM, Hg(p), Hg ⁰)	✓	✓
Ambient concentration of ozone and sulfur dioxide	√ (continuous)	(weekly via AirMON Dry)
Ambient concentration of carbon monoxide	✓	
Meteorology	✓	(via NADP/NTN site)
Major ions in precipitation		(via NADP/NTN site)

Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (regional close-up)

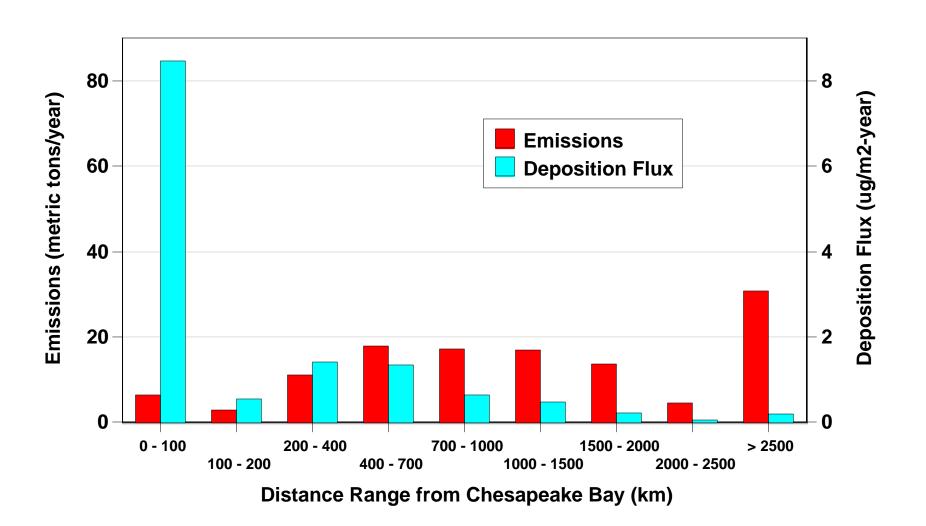




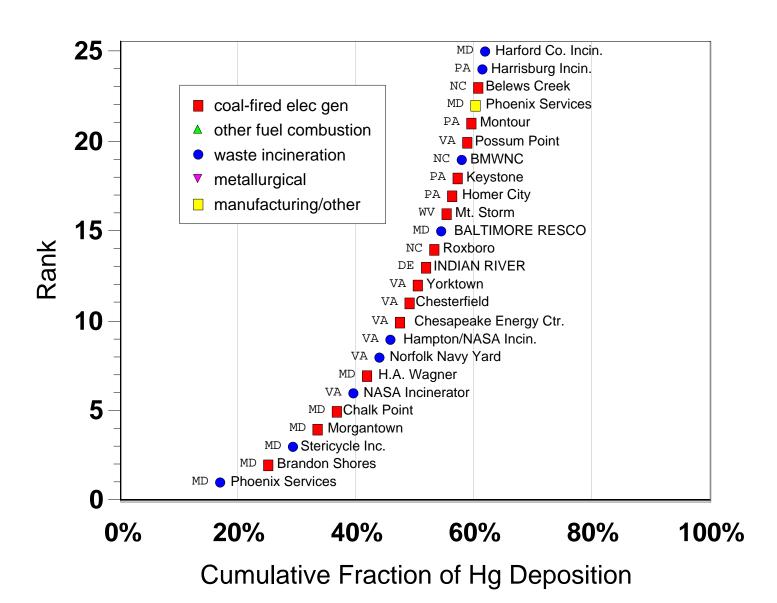
Largest Local Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay



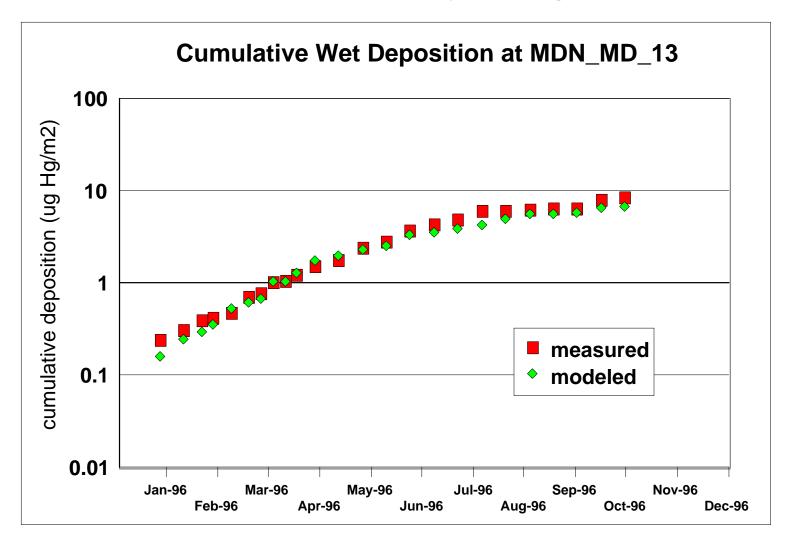
Emissions and Direct Deposition Contributions from Different Distance Ranges Away From the Chesapeake Bay



Top 25 Contributors to 1999 Hg Deposition Directly to the Chesapeake Bay



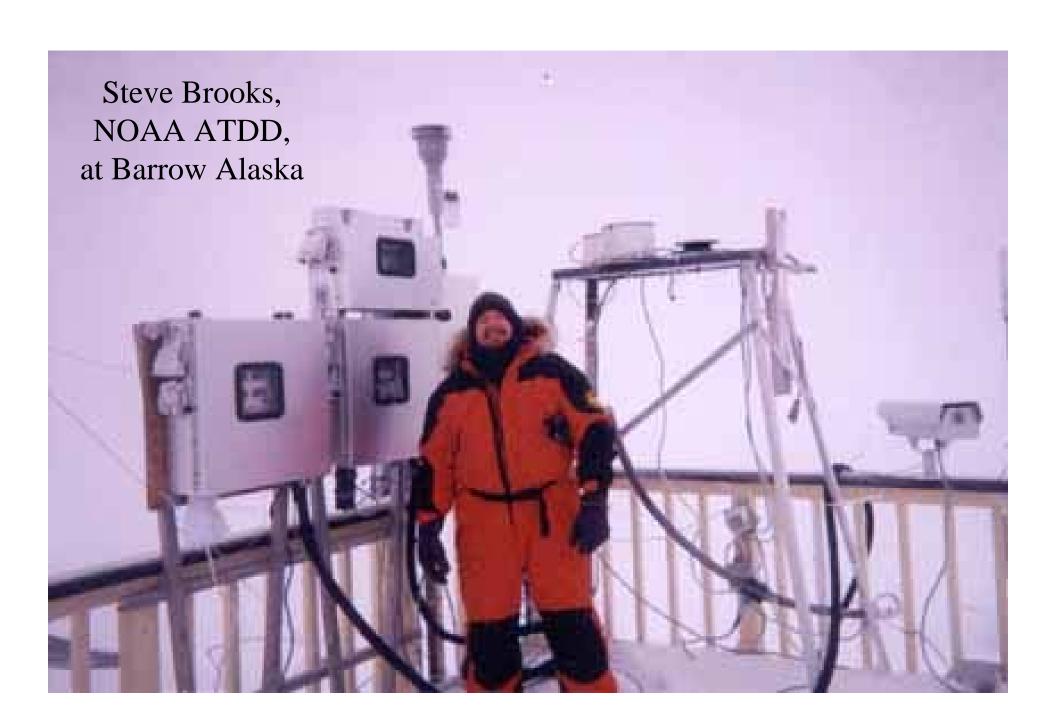
Modeled vs. Measured Wet Deposition at Mercury Deposition Network Site MD_13 (Wye) during 1996



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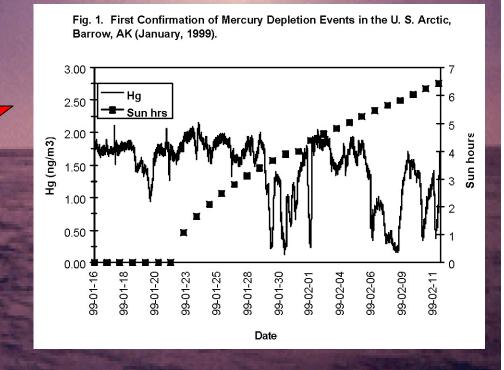




in very simplified terms:

 $Hg(0) \longrightarrow Hg(II) ("RGM")$

ARL Scientists played a key role in discovering and explaining the Mercury Depletion Events occurring in the Arctic at Polar Sunrise



Examples of External Mercury Collaborations -- Modeling

- ☐ Environmental Protection Agency
 - National Exposure Research Laboratory
 - Clean Air Markets Division
 - Great Lakes National Program Office
- ☐ International Joint Commission (Great Lakes)
 - Int'l Air Quality Advisory Board
- ☐ Commission for Environmental Cooperation (NAFTA)
 - Atmospheric Hg deposition to the Gulf of Maine
 - Hg deposition impacts of future energy generation scenarios
- ☐ EMEP LRTAP Protocol (Europe)
 - Mercury model intercomparison (HYSPLIT-Hg, CMAQ-Hg)
- ☐ Environment Canada
 - Emissions inventories

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MULTI-MEDIA MERCURY MODELING PROJECT

Update for the International Air Quality Planning Board

January 26, 2005





Environmental and Occupational Health Sciences Institute (EOHSI)

I had bloom or bloom. No recommend in the design

Principal Investigators:

Dr. Mark Cohen, NOAA

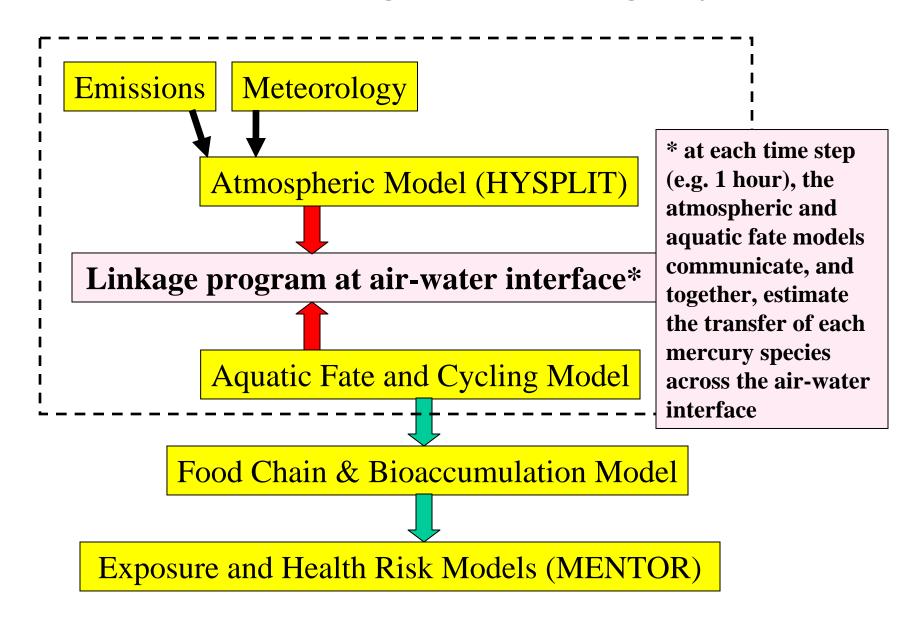
Dr. Panos Georgopoulos, EOHSI

Dr. John Johnston, USEPA

Dr. Elsie Sunderland, USEPA



Multi-Media Hg Modeling System



External Mercury Collaborations -- Modeling

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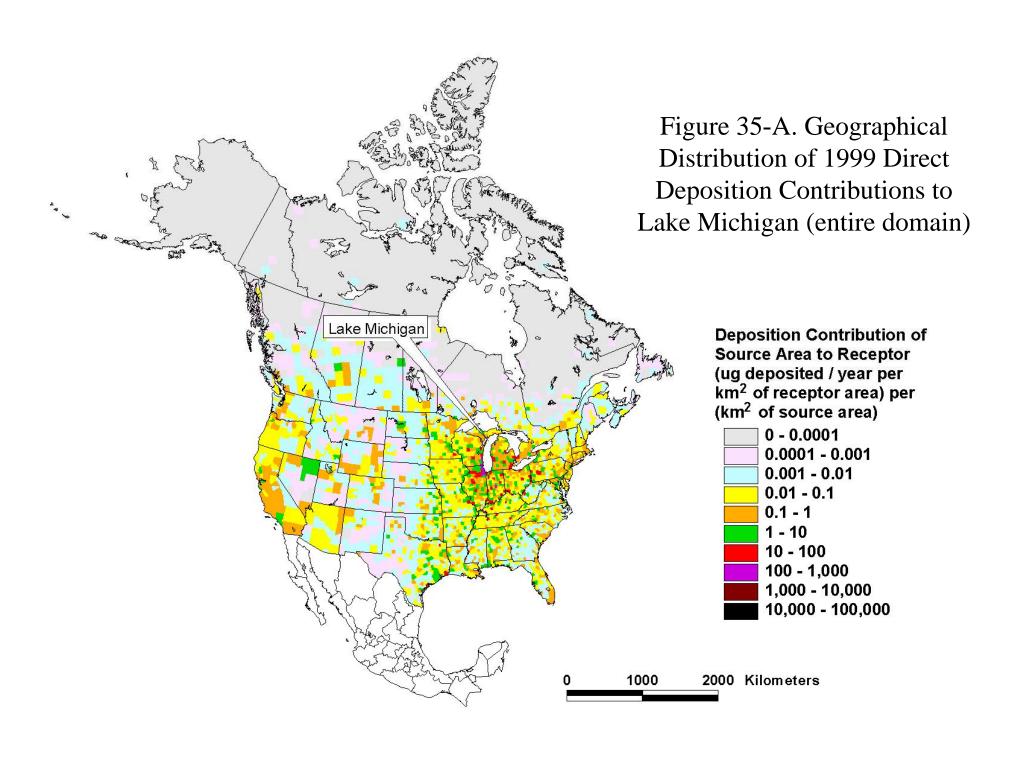


Figure 35-B. Geographical Distribution of 1999 Direct Deposition Contributions to Lake Michigan (regional view)

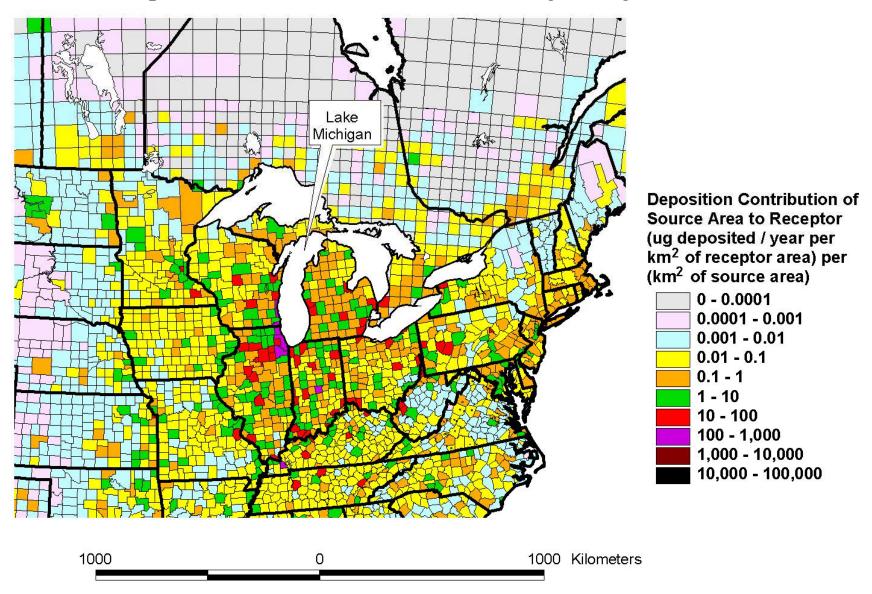
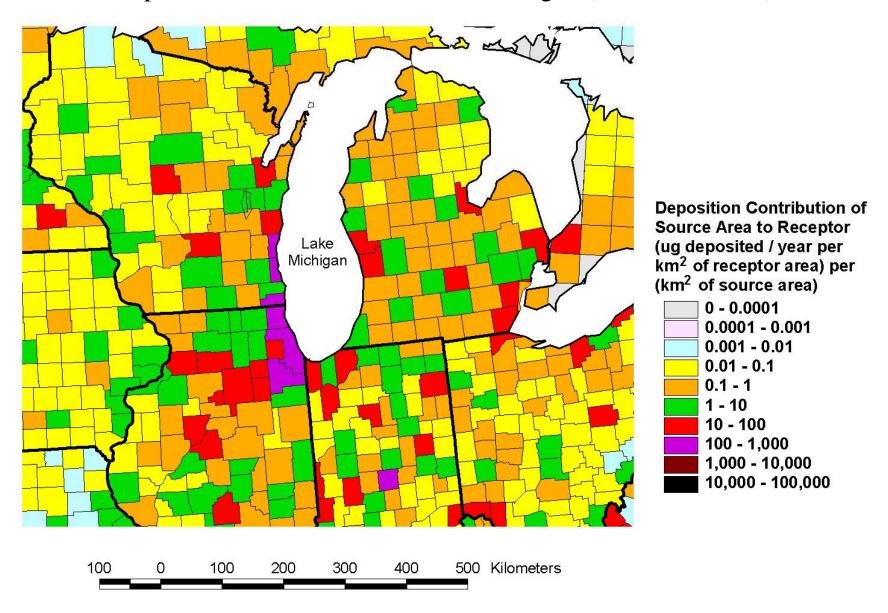
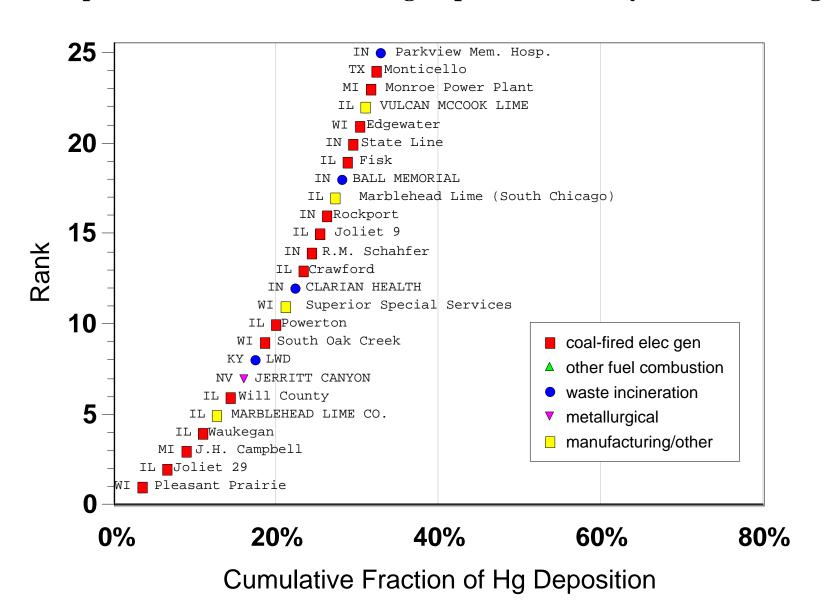


Figure 35-C. Geographical Distribution of 1999 Direct Deposition Contributions to Lake Michigan (more local view)



Top 25 Contributors to 1999 Hg Deposition Directly to Lake Michigan





Available online at www.sciencedirect.com



Environmental Research

Environmental Research 95 (2004) 247-265

http://www.elsevier.com/locate/envres

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

a NOAA Air Resources Laboratory, 1315 East West Highway R/ARL, Room 3316, Silver Spring, MD 20910, USA
b Commission for Environmental Cooperation, Montreal, Que., Canada
c Atmospheric Toxic Processes Service, Meteorological Service of Canada, Environment Canada Quebec region, Montreal, Que., Canada
d Pollutant Data Branch, Environment Canada, Hull, Que., Canada
e Environmental Monitoring and Reporting Branch, Ontario Ministry of the Environment, Toronto, Ont., Canada
f University of California, Berkeley, CA, USA
g US EPA Great Lakes National Program Office, Chicago, IL, USA
h International Joint Commission, Windsor, Ont., Canada

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

Examples of External Mercury Collaborations -- Monitoring

- ☐ Environmental Protection Agency
 - National Exposure Research Laboratory aircraft Hg measurements
- ☐ International Measurement Intercomparison Campaigns
 - Arctic (polar sunrise phenomena)
 - Antarctic
- ☐ University of Alabama (Sea Grant)
 - measurements in the Gulf of Mexico region ship-based and landbased measurements.

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DeHavilland DHC-6 Twin Otter

measurements include:

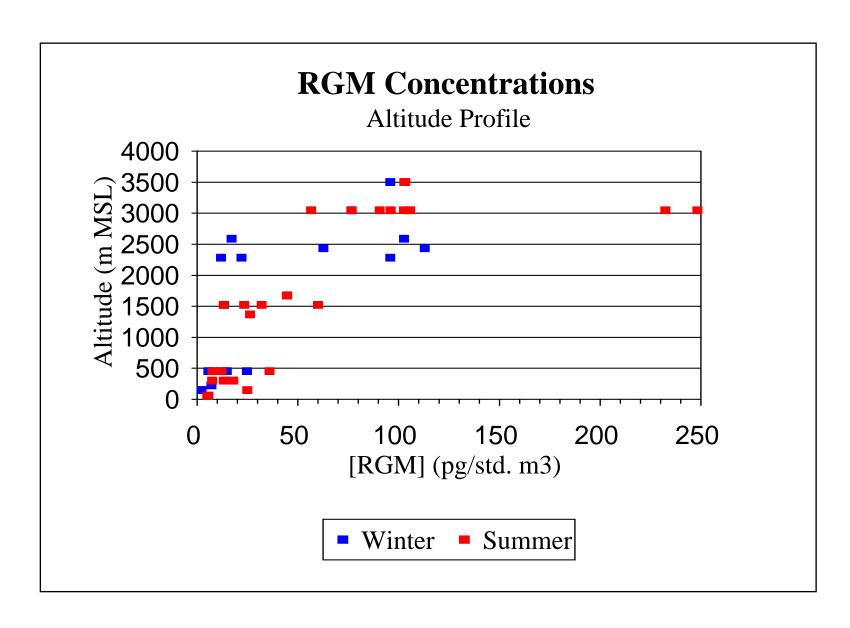
• atmospheric parameters

• trace gases $(O_3, SO_2, NO_x, etc.)$

• speciated mercury



Unexpectedly high concentrations of Reactive Gaseous Mercury (RGM) were found in the upper atmosphere!



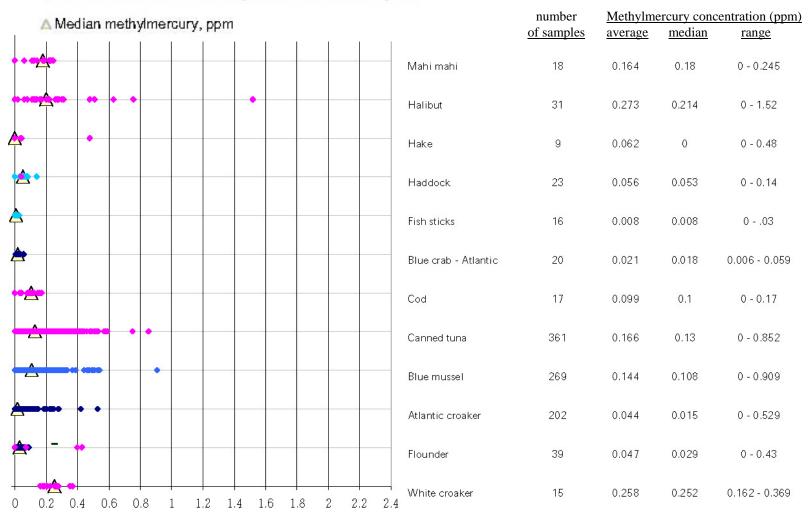
Collaboration Possibilities within ERP

Establish and update status of mercury contamination in NOAA trust resources Gulf of Mexico, Chesapeake Bay, Upper Atlantic, Great Lakes, Lake Champlain
 □ Characterize and understand reasons for spatial and temporal trends □ Construct mass balance of mercury (relative loading from air, tributaries, etc.) □ Quantify past, present, and future sources of mercury contamination □ Understand fate and cycling of mercury, e.g., sedimentation, methylation □ Understand watershed processing □ Understand the food web and mercury bioaccumulation □ Understand effects on wildlife (e.g., fish-eating birds)
What are the major uncertainties? How can we reduce these uncertainties?
With the above, provide information for risk communication and sound science to support decision-making at the local, regional, national, and international level

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- FDA seafood surveillance program.
- FDA Total Diet Study
- NOAA Gulfchem relational database
- EPA Environmental Monitoring and Assessment Program

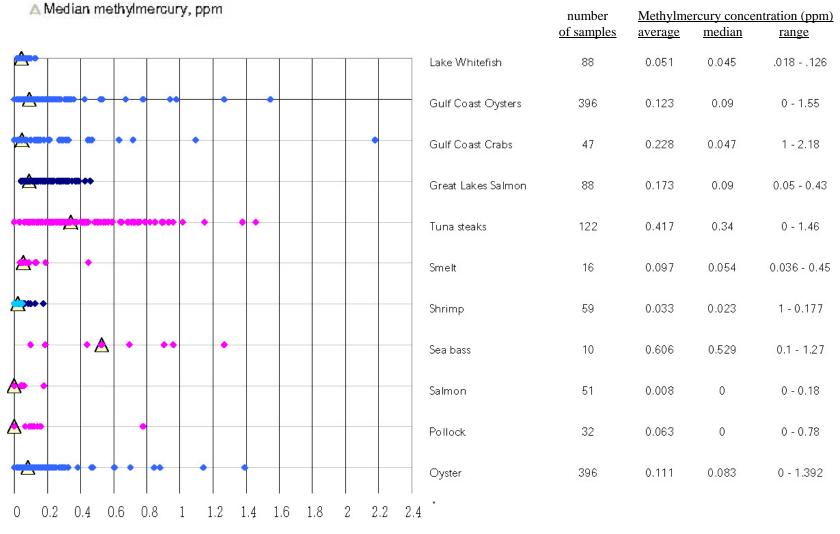
Distribution of methyl-mercury concentrations in different fish species



source: Environmental Working Group

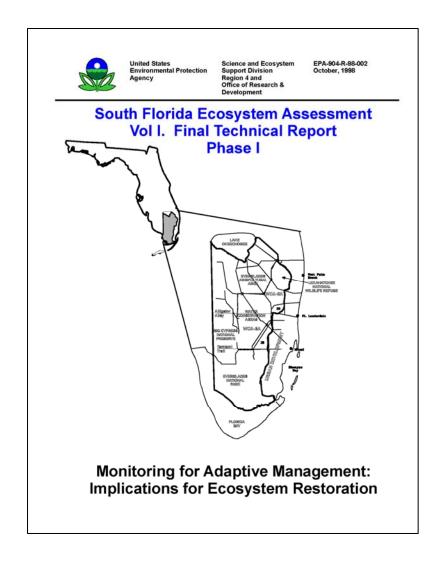
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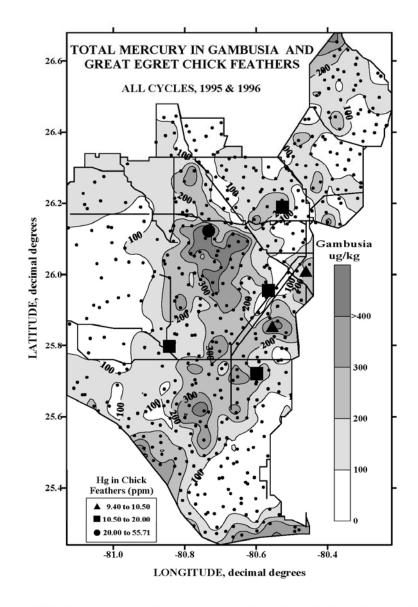
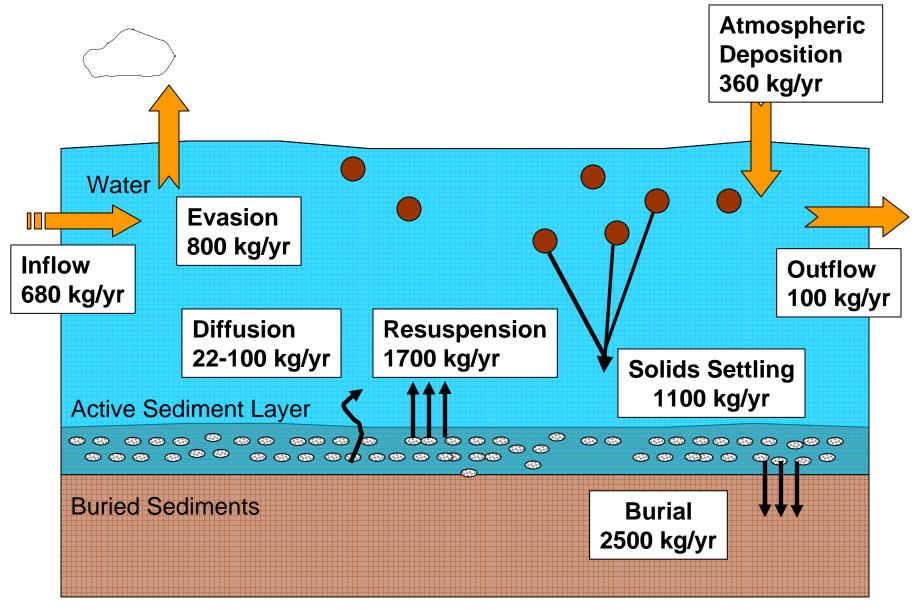


Figure 8.55 Hg concentrations in Great Egret chick feathers and mosquitofish indicate spatial distribution of Hg bioaccumulation.

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Total Mercury Fluxes Lake Ontario



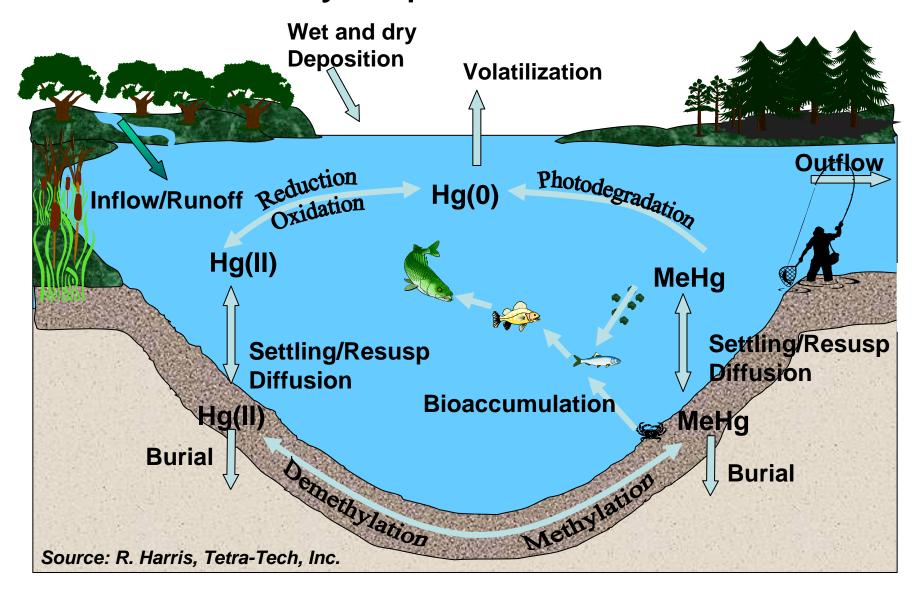
slide courtesy of Elsie Sunderland, USEPA

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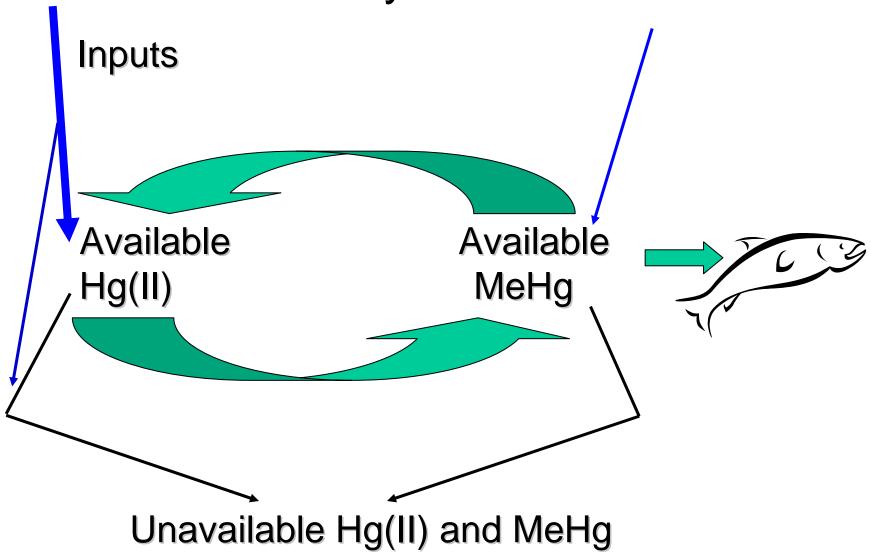


Mercury Aquatic Fate Model





Conceptualization: How Mercury Reaches Fish



slide courtesy of Elsie Sunderland, USEPA

Summary: Factors Affecting Methylation

• Hg Bioavailability in Different Ecosystems

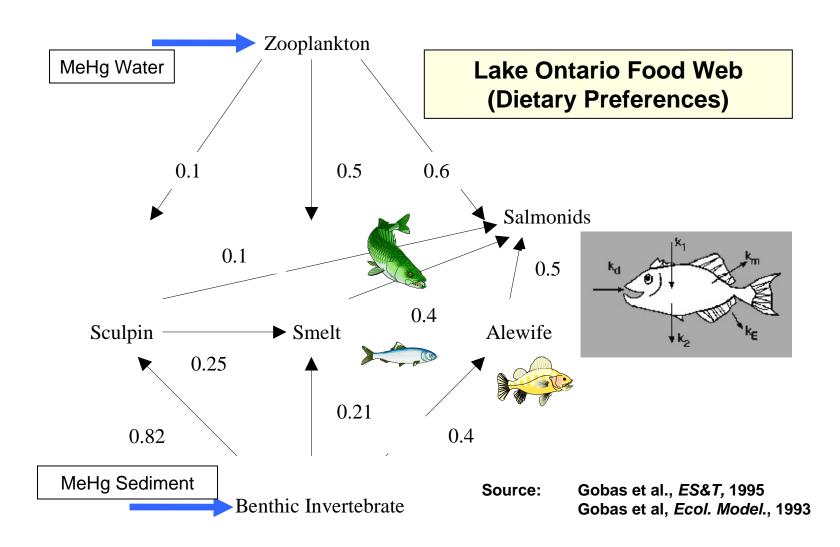
- Sulfide
- Chemistry (dissolved organic carbon, pH)
- Amount and types of solids in water and sediments

Microbial Activity Producing Methylmercury

- Temperature
- Suitable environments (oxygen, resuspension)
- Sulfate
- Organic matter
- Other biological processes that affect chemistry (sulfide oxidizers; iron reducers)

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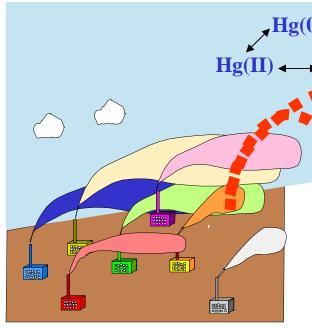
Bioaccumulation Model Framework



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<u>potential Air Resources Laboratory contributions</u> <u>to collaborative mercury work within NOAA:</u>

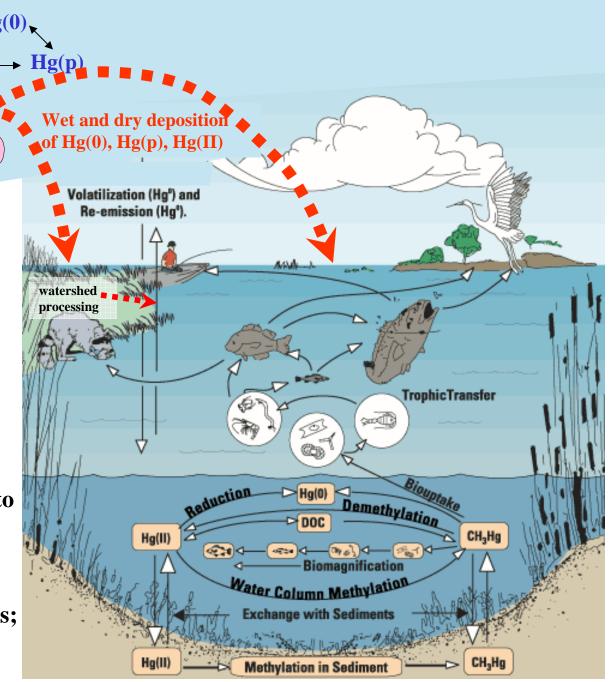
Further process-related monitoring and model development to increase understanding of atmospheric transport and deposition of mercury
Monitoring and modeling used together to estimate the atmospheric deposition of different forms of mercury to a give waterbody or watershed
Monitoring and modeling used together to estimate the relative contributions of different source regions and source types to the atmospheric deposition of mercury to a given waterbody or watershed
Modeling to estimate past and possible future atmospheric loadings to a given waterbody or watershed.
Monitoring and modeling to help understand and characterize re-emissions processes at the air-water interface and the air-watershed interface



We need to understand Hg in the environment enough to be able to fix the problem

Many scientific disciplines need to work in collaboration to achieve this understanding

NOAA appears uniquely qualified to tackle Hg problems; ARL would like to help



THANKS!

ADDITIONAL BACKGROUND SLIDES



Hg Headquarters Division -- HQ (Silver Spring, MD)

development of improved transport and dispersion models; making ARL products operational through direct interaction with NCEP.

- **Hg** <u>Atmospheric Sciences Modeling Division ASMD</u> (Research Triangle Park, NC) development of improved air quality models, for both assessment and forecasting, through direct interaction with the EPA and other federal partners.
- Hg <u>Atmospheric Turbulence and Diffusion Division ATDD</u> (Oak Ridge, TN) improving descriptions of atmospheric dispersion and deposition in models, emphasizing complex situations, and on developing improved instrumentation.

Field Research Division - FRD (Idaho Falls, ID)

field atmospheric tracer testing facility, for developing transport and diffusion models; FRD's mesonet and modeling expertise supports the Idaho National Laboratory.

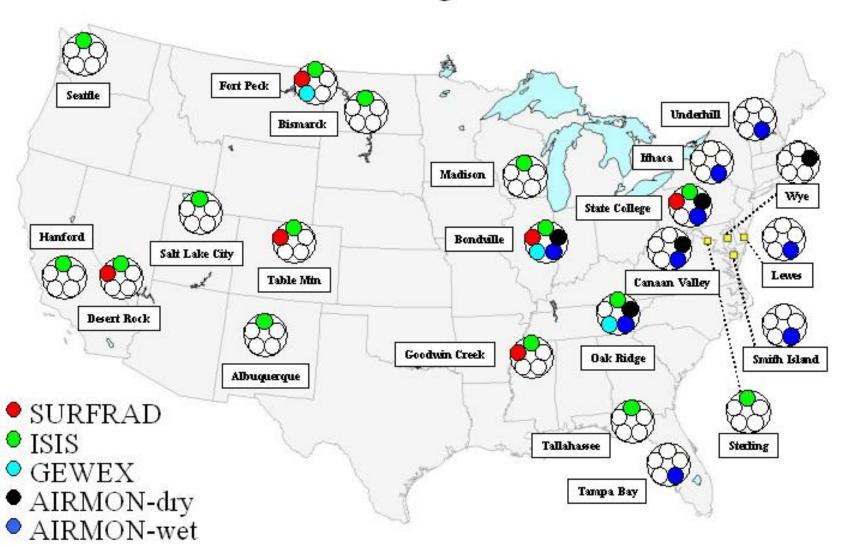
Special Operations and Research Division – SORD (Las Vegas, NV)

models atmospheric transport, dispersion, and deposition over complex terrain; studies the effects of airborne particles on atmospheric radiation and opacity; and provides dispersion guidance to DOE managers of the Nevada Test Site.

Surface Radiation Research Branch - SRRB (Boulder, CO)

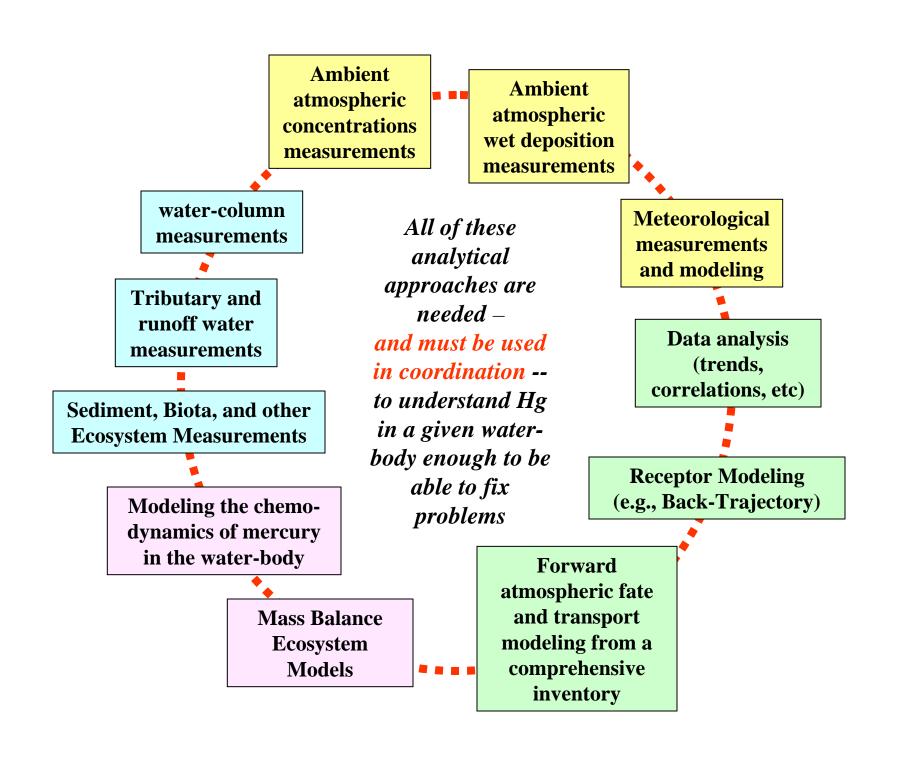
provides basic data on radiation fields, for the next generation of atmospheric transport and dispersion models, by climate assessments, and by evaluations of climate models.

NOAA Air Resources Laboratory Research and Monitoring Sites



Aircraft Chemical Instrumentation (in addition to mercury)

Species	Method	LOD (ppbv)	response
O_3	UV Absorption	2	10 s
CO	NDIR/GFC	30	15 s
SO_2	Pulsed Fluorescence	0.4	15 s
NO	Ozone CL	0.02	1 s
NO_2	Photolysis/CL	0.06	2 s
NO_{Y}	Molybdenum/CL	0.06	1 s
CN	Optical Counts	$< 100 \text{ cm}^{-3}$	0.2 s
HNO_3	Converter Difference	0.25	5 s
Major Ions	Filter Pack	N/A	N/A
Aerosol size	Optical Counts	0.3 to 10 um	1s
CH_2O	Liquid Phase Fluorescen	ce 0.1	90 s
H_2O_2	Liquid Phase Fluorescen	ce 0.02	90 s
PAN	Fast GC/Luminol	0.01	30 s
NMHCs	Grab Samples/GCFID	varies	30 s



Nevertheless,
many models
seem to be
performing
reasonably well,
i.e., are able to
explain a lot of
what we see

Convention on Long-Range Transboundary Air Pullution

emep

Co-operative programme for monthies, and evaluation of the long-range transmission of air pollutants in Europe 1/2003 June 2003

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

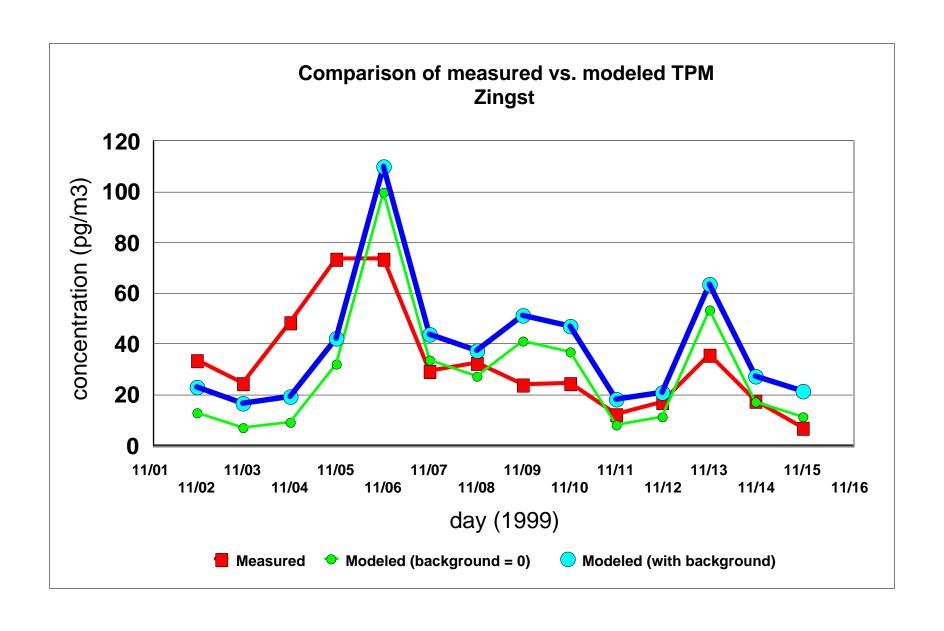
Stage II. Comparison of modeling results with observations obtained during short-term measuring campaigns

Technical Report 1/2003

A. Ryaboshapko, R. Artz, R. Bullock, J. Christensen, M. Cohen, A. Dastoor, D. Davignon, R. Draxler, R. Ebinghaus, I. Ilyin, J. Munthe, G. Petersen, D. Syrakov



msc-e

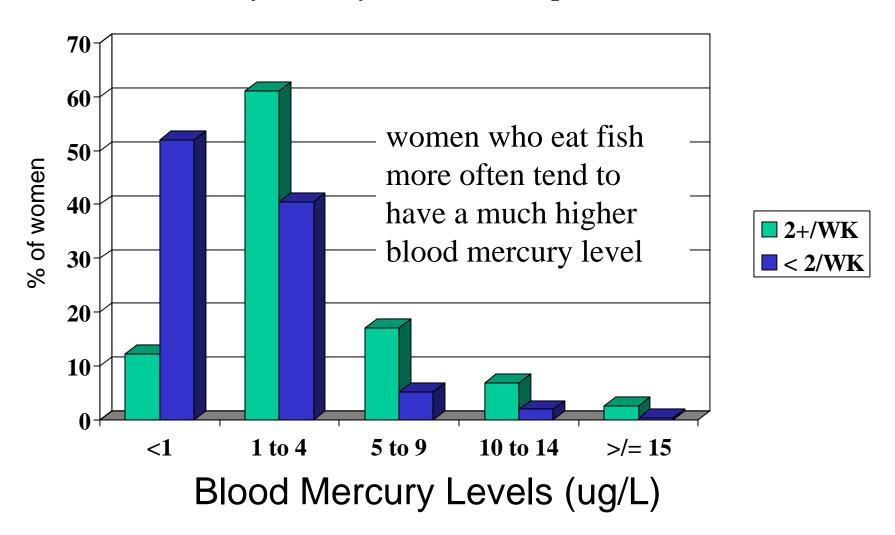


Estimated Number of Newborns with *In Utero* Methylmercury Exposures >/= RfD

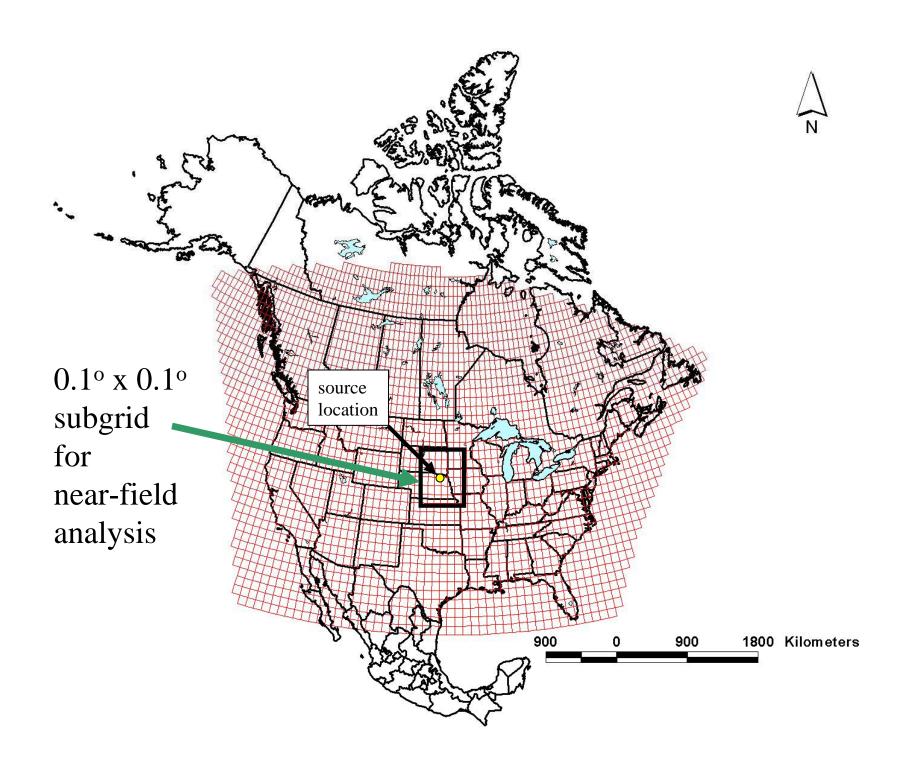
- Number of US births in 2000: 4,058,814 (National Vital Statistics Reports).
- 1:1 ratio of cord to maternal blood [Hg], i.e., 5.8 cord to 5.8 maternal, 7.8% of women had total blood [Hg] >/= 5.8, ~ 300,000 newborns each year > 5.8 ug/L (Mahaffey et al., 2003).
- 1.7: 1 ratio of cord to maternal blood [Hg], i.e. 5.8 cord to ~3.5 maternal, 15.7% of women had total blood [Hg] >/= 3.5 ug/L, ~630,000 newborns each years >/= 5.8 ug/L cord blood.

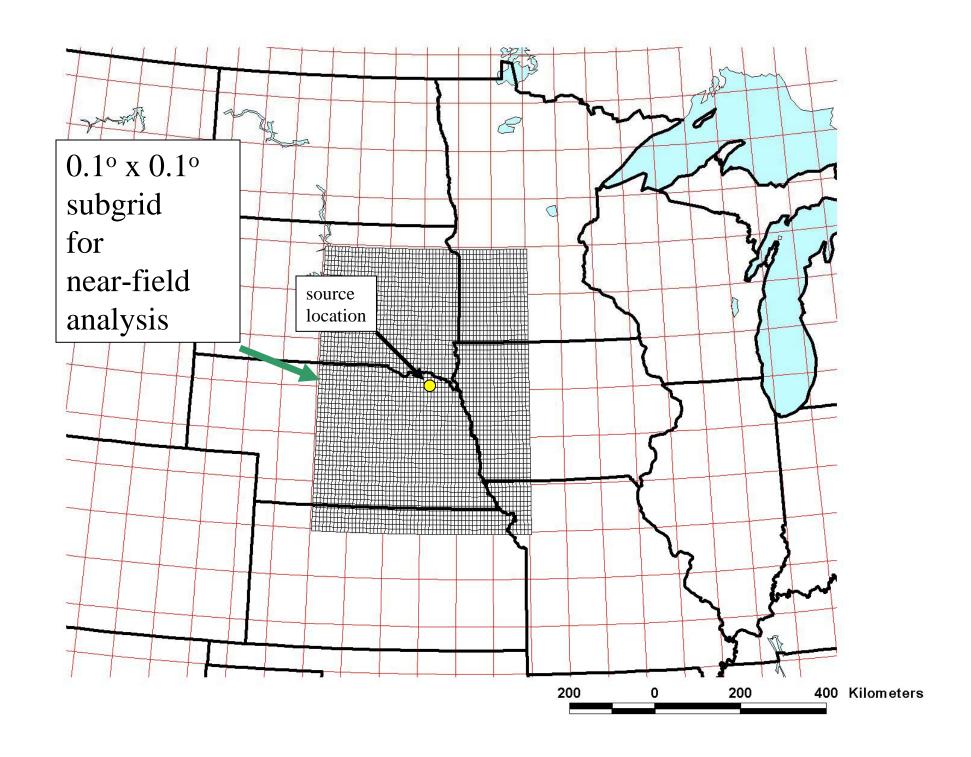
source: Kate Mahaffey, USEPA

Total Mercury Levels in Women, Aged 16-49 by Weekly Fish Consumption Levels

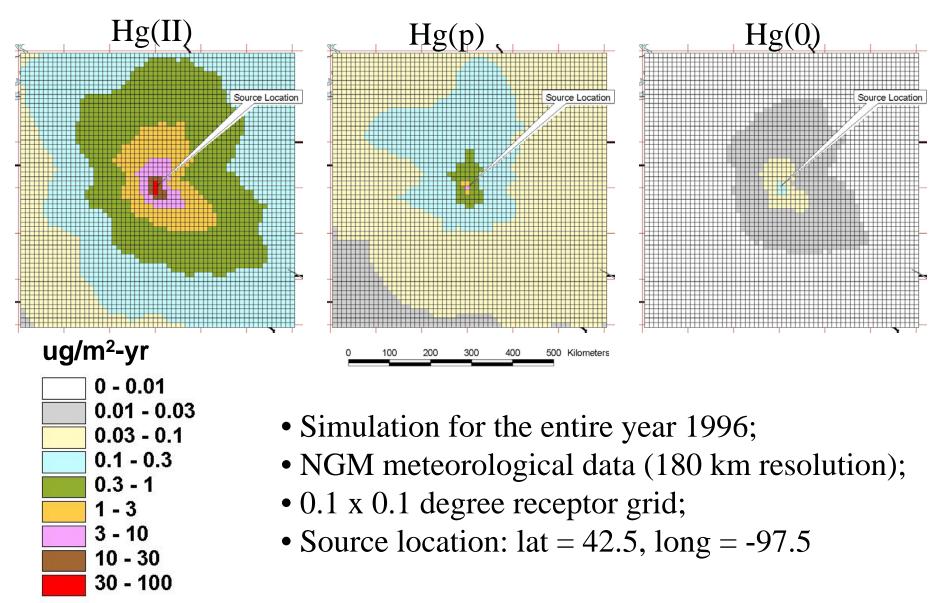


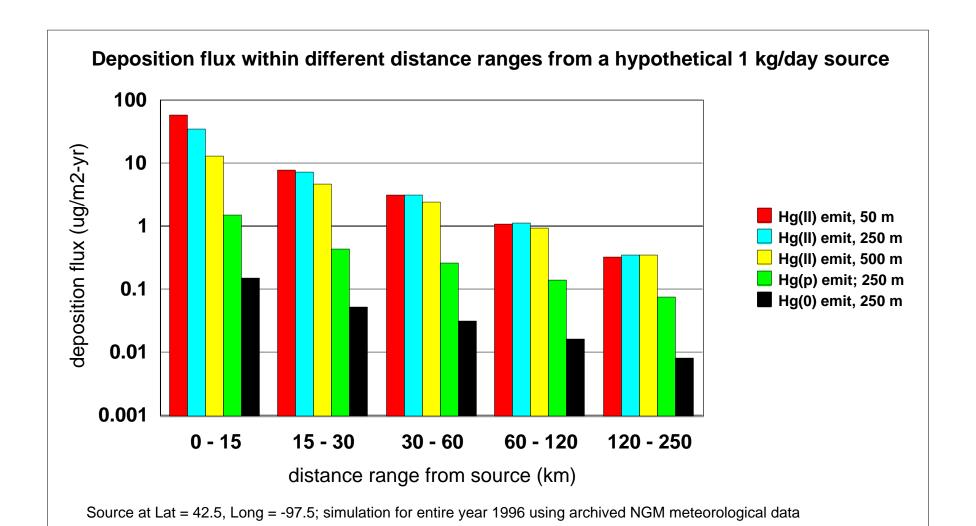
source: Kate Mahaffey, USEPA



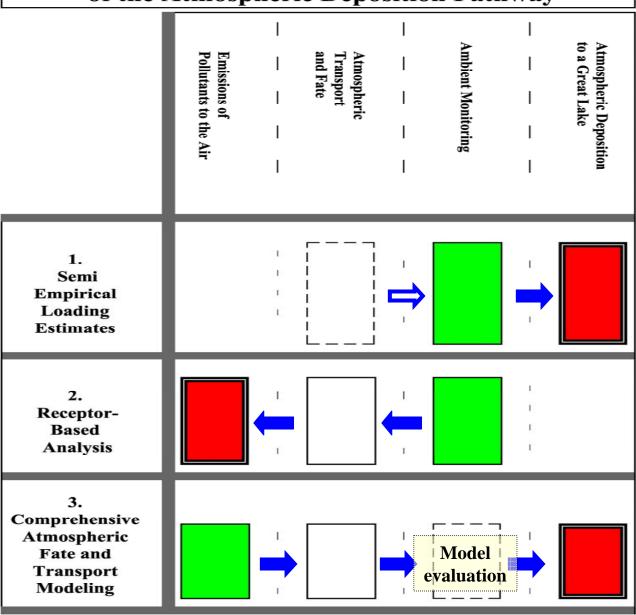


Annual deposition flux arising from a hypothetical 1 kg/day 250 meter high source of different forms of mercury





Methodological Approaches for Analysis of the Atmospheric Deposition Pathway



TIME (hours) = mass of pollutant Puff's mass, size, and (decreases as destruction and location continually 0 deposition occur each time step) tracked... Centerline of puff motion Photolytic and chemical determined by wind transformation of pollutant also direction and velocity estimated at each time step Initial puff Total deposition from a puff during location is at source, with a given time atep is calculated from the initial mass sum of the estimat-& size ed dry and wet. deposition of gas and particle-phase material, based on pollutant conc. in the puff, the local weather, and the nature of the aurface. O II II D ____ deposition 1 deposition 2 deposition to lake Lake

Figure 1. Lagrangian Puff Air Transport and Deposition Model

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

