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

Dr. Mark Cohen

NOAA Air Resources Laboratory Silver Spring, Maryland, USA mark.cohen@noaa.gov http://www.arl.noaa.gov/ss/transport/cohen.html

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



Outline of Presentation

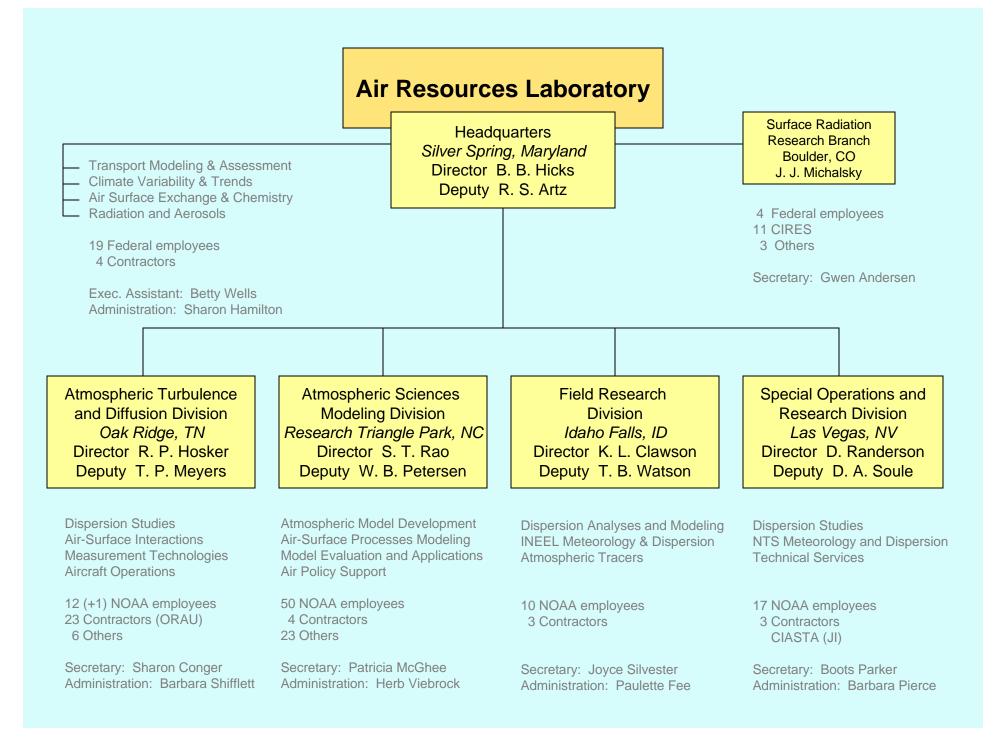
□ brief overview of Air Resources Laboratory

□ mercury problem / role of atmospheric Hg

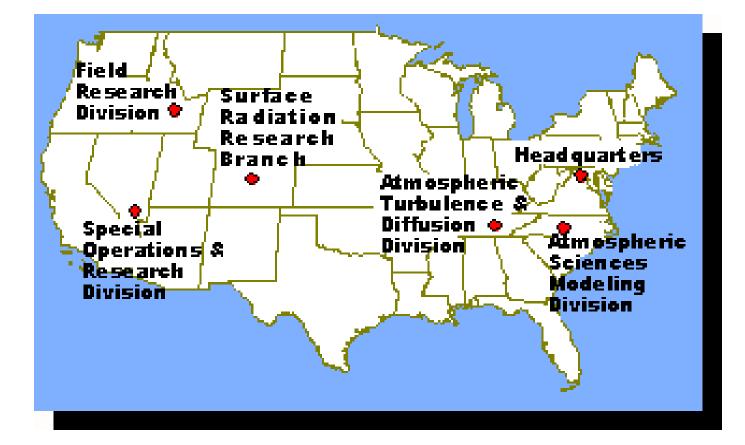
□ ARL mercury programs and collaborations

□ opportunities for collaboration within ERP

overview of the Air Resources Laboratory





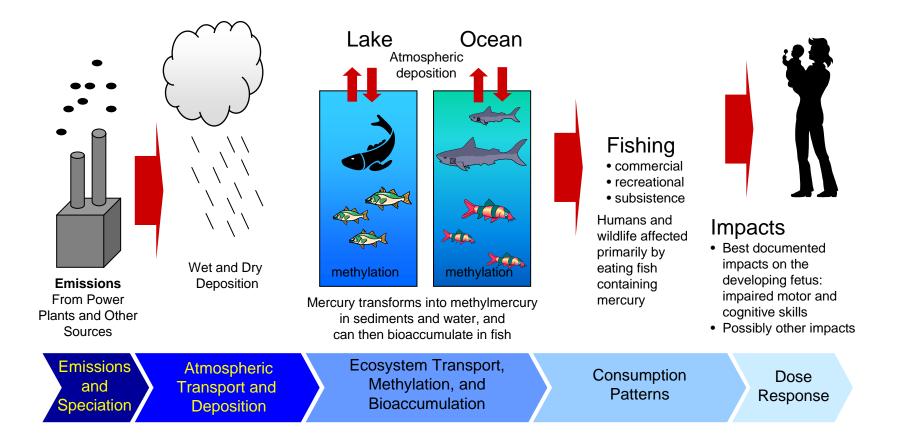


the mercury problem and the role of atmospheric mercury

The Mercury Problem

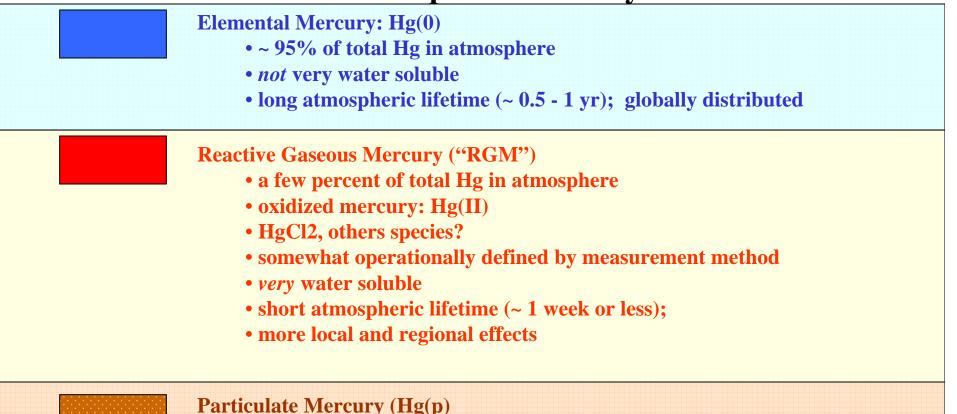
- EPA has estimated that 1 out of every 6 children born in the U.S. have already been exposed in-utero to levels of mercury that may cause problems with neurological development
- □ There are additional potential mercury-related health hazards to children, adults, and to wildlife
- □ Fish-consumption 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



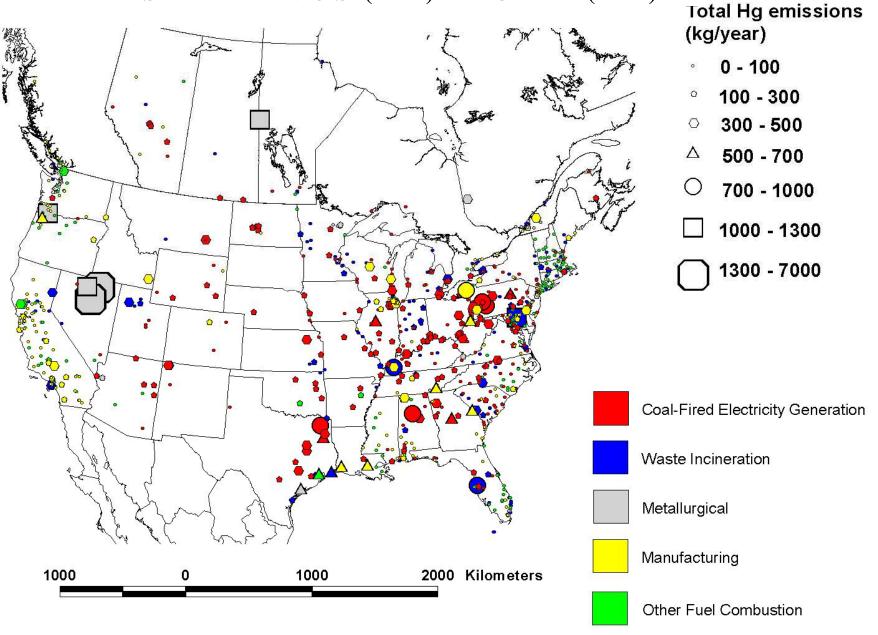
- a few percent of total Hg in atmosphere
- not pure particles of mercury...
 - (Hg compounds associated with atmospheric particulate)
- species largely unknown (in some cases, may be HgO?)
- moderate atmospheric lifetime (perhaps 1~ 2 weeks)
- local and regional effects
- bioavailability?

Elemental Mercury: Hg(0) **Atmospheric Fate Processes for Hg Reactive Gaseous Mercury: RGM** Particulate Mercury: Hg(p) Upper atmospheric Polar sunrise halogen-mediated "mercury depletion events" cloud heterogeneous oxidation? Br CLOUD DROPLET "DRY" (low RH) Hg(II) reduced to Hg(0) by SO_2 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

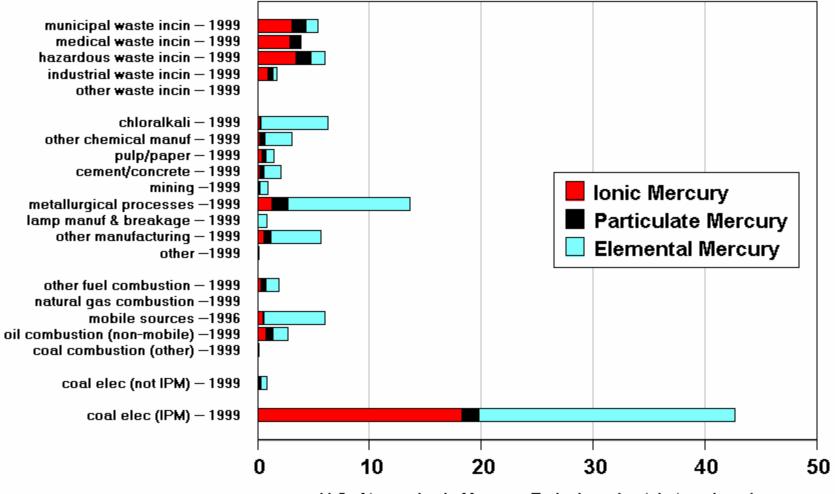
Atmospheric Chemical Reaction Scheme for Mercury

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)	
$Hg^0 + Cl_2 \rightarrow HgCl_2$	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		Van Loon et al. (2002)	
	[T = temperature (K)]			
$Hg(II) + HO_2C \rightarrow Hg^0$	~ 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)

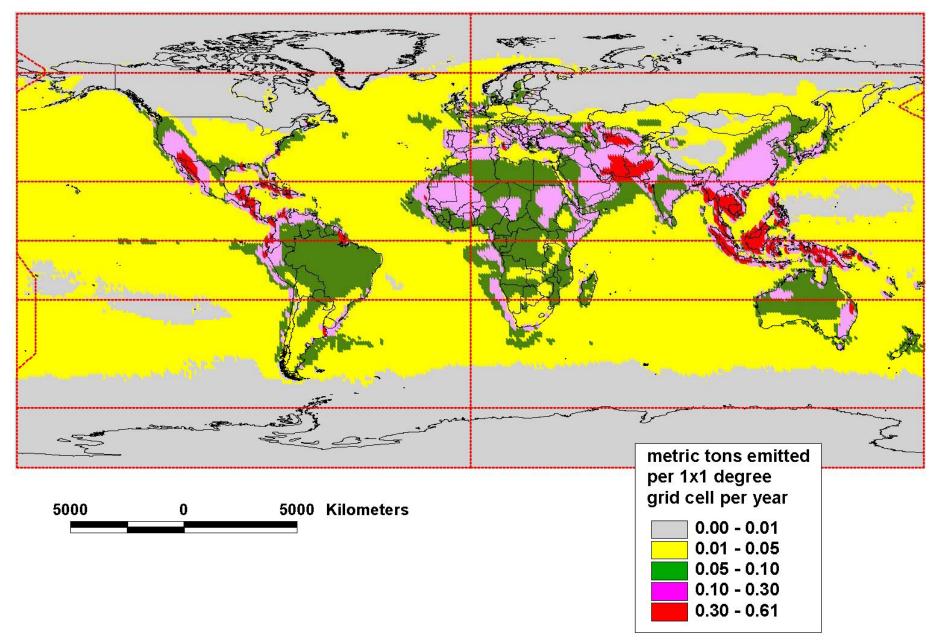


Estimated 1999 U.S. Atmospheric Anthropogenic Mercury Emissions

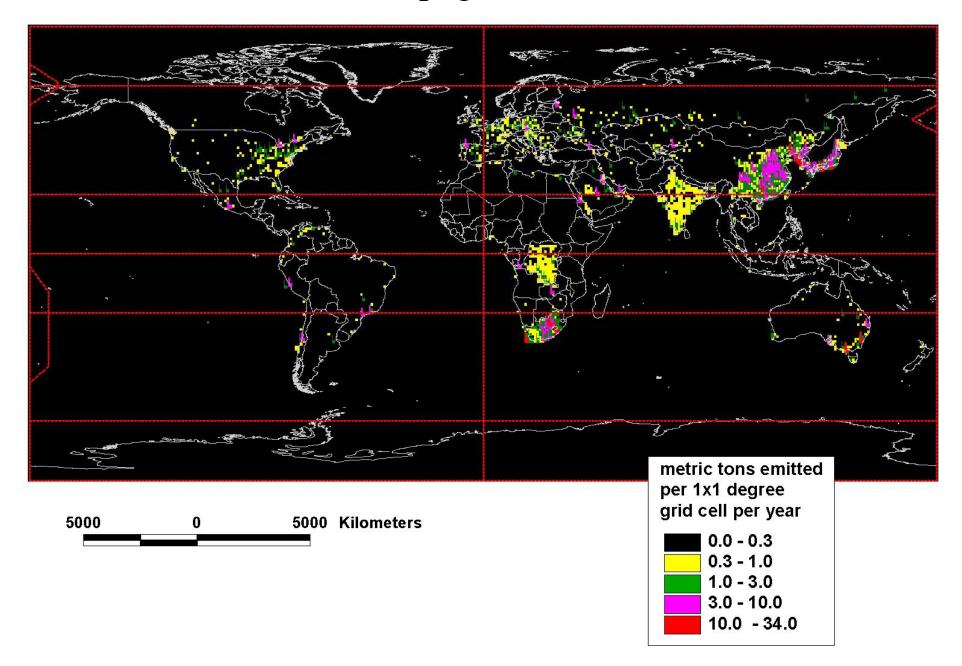


U.S. Atmospheric Mercury Emissions (metric tons/year)

estimated natural emissions (1999)

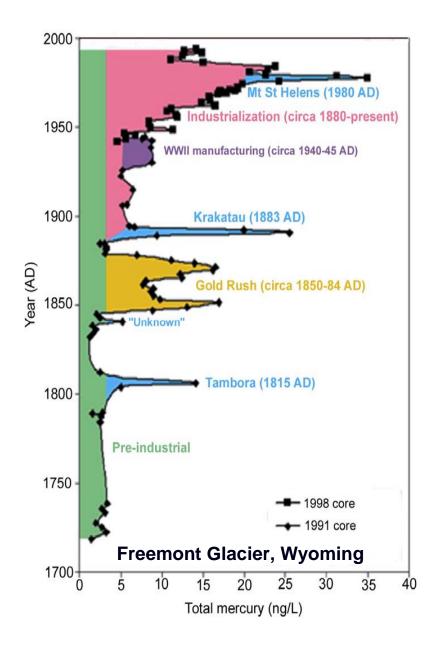


estimated anthropogenic emissions (1995)



Natural vs. anthropogenic mercury?

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



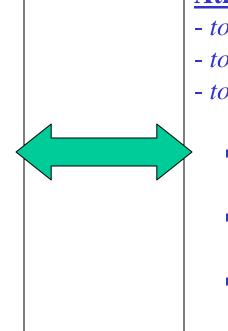
source: USGS, Shuster et al., 2002

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

U Within NOAA

D 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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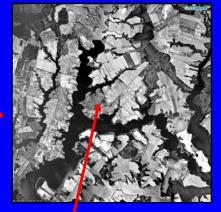
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Summer 2004 Chesapeake Bay Atmospheric Mercury Measurement Sites



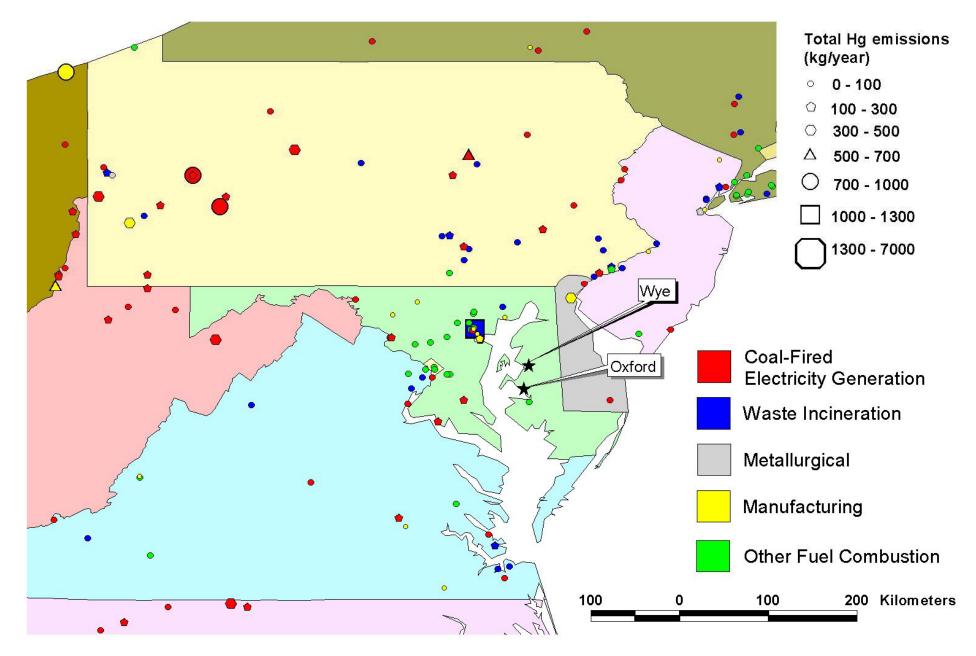


Wye Research and Education Center (38.9131EN, 76.1525EW)



Cooperative Oxford Lab (38.678EN, 76.173EW)

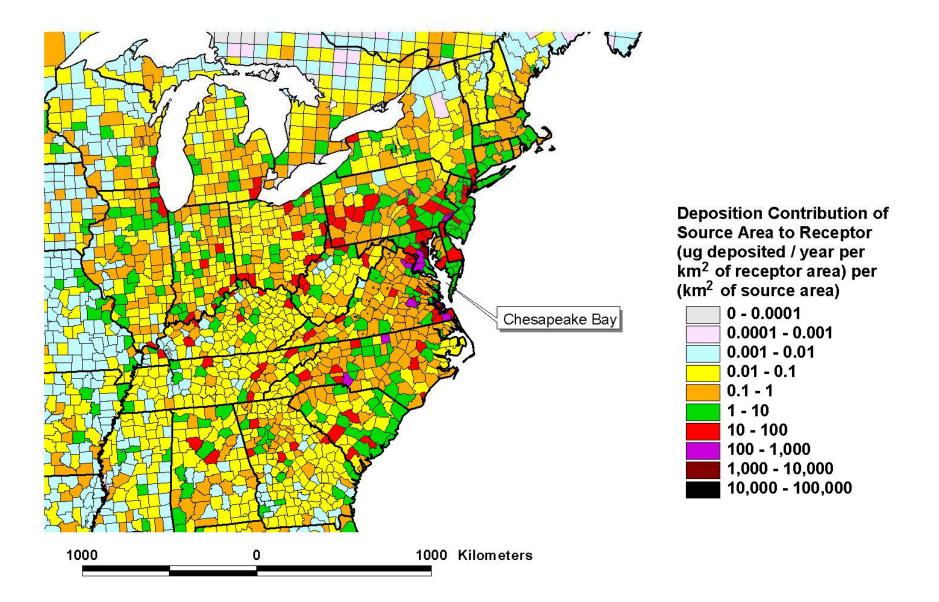
regional emissions (1999) and sampling sites for summer 2004 Ches Bay Hg study

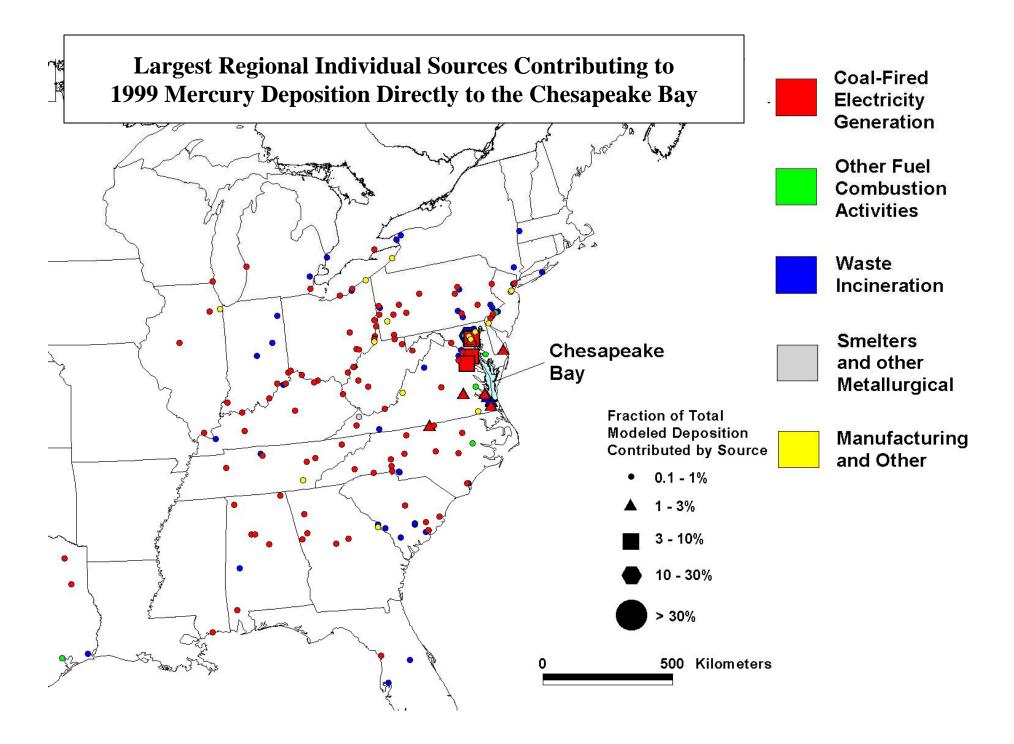


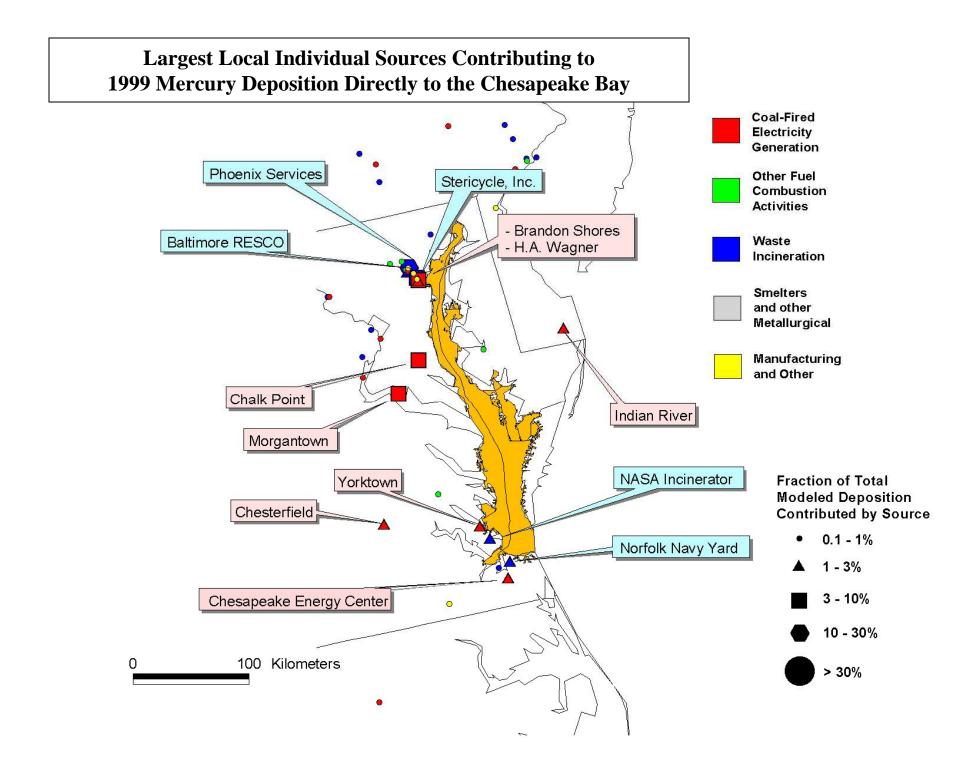
Summer 2004 Chesapeake Bay Atmospheric Hg Study (June – August 2004)

	Oxford	Wye
Event-based precipitation samples analyzed for Hg	\checkmark	✓
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)

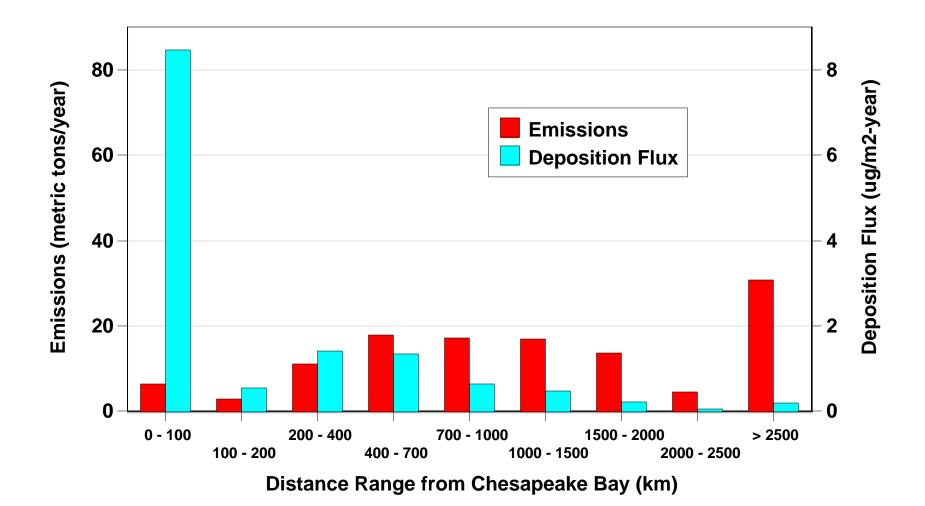
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (regional close-up)



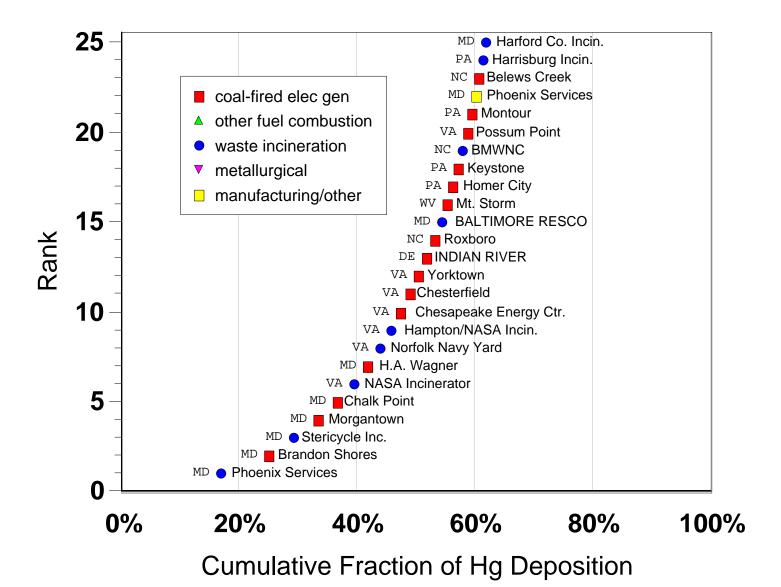




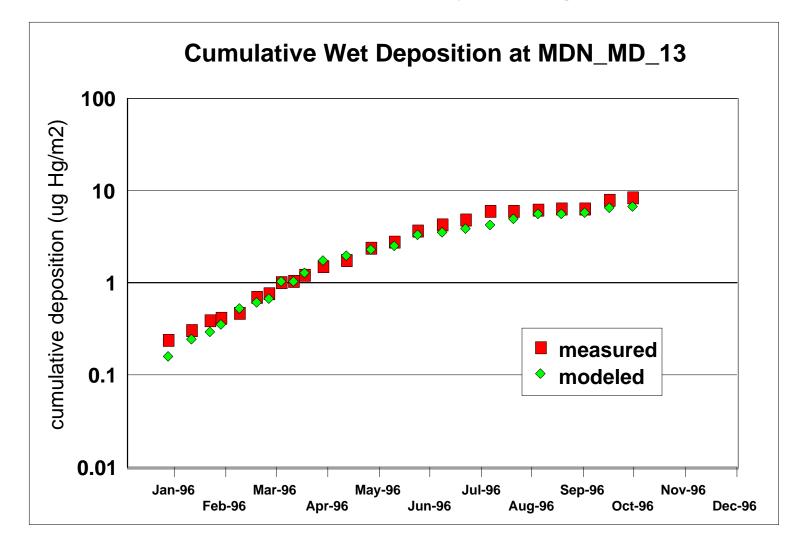
Emissions and Direct Deposition Contributions from Different Distance Ranges Away From the Chesapeake Bay







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



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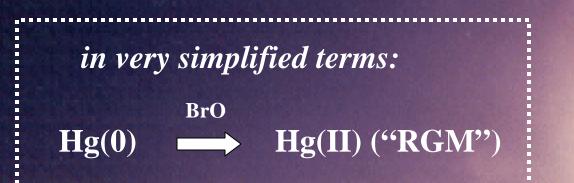
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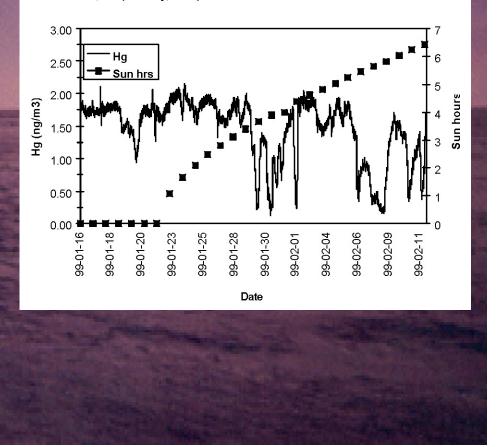


Steve Brooks, NOAA ATDD, at Barrow Alaska



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

Fig. 1. First Confirmation of Mercury Depletion Events in the U. S. Arctic, Barrow, AK (January, 1999).



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

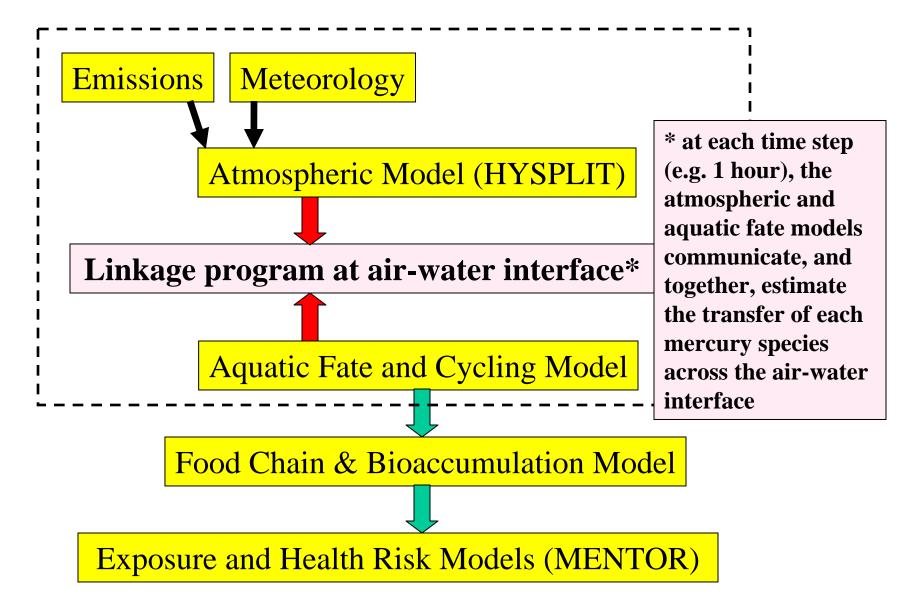




Environmental and Occupational Health Sciences Institute (EOHSI)

SEPA United States Environmental Protection Agency Principal Investigators: Dr. Mark Cohen, NOAA Dr. Panos Georgopoulos, EOHSI Dr. John Johnston, USEPA Dr. Elsie Sunderland, USEPA

Multi-Media Hg Modeling System



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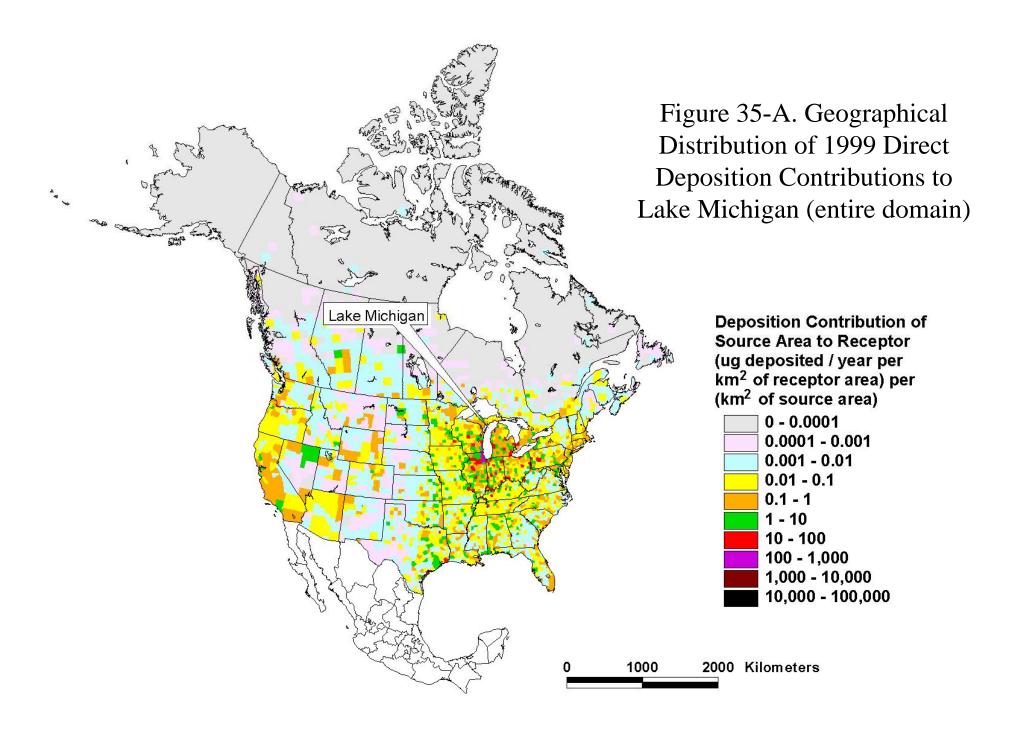


Figure 35-B. Geographical Distribution of 1999 Direct Deposition Contributions to Lake Michigan (regional view)

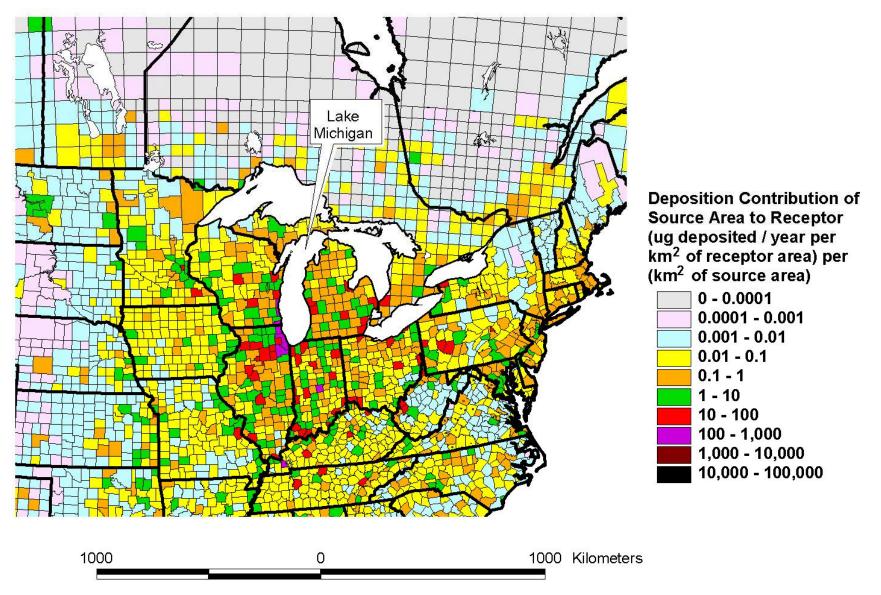
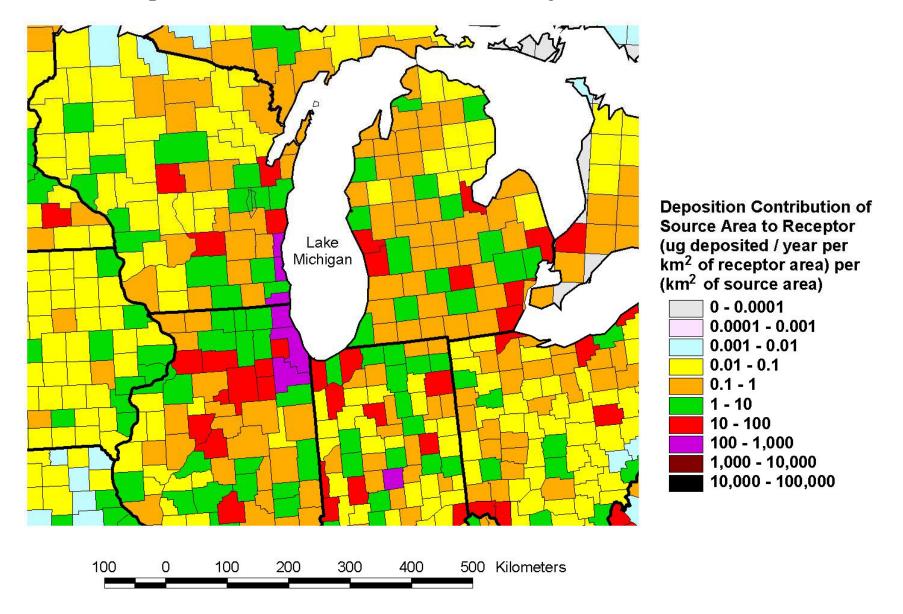
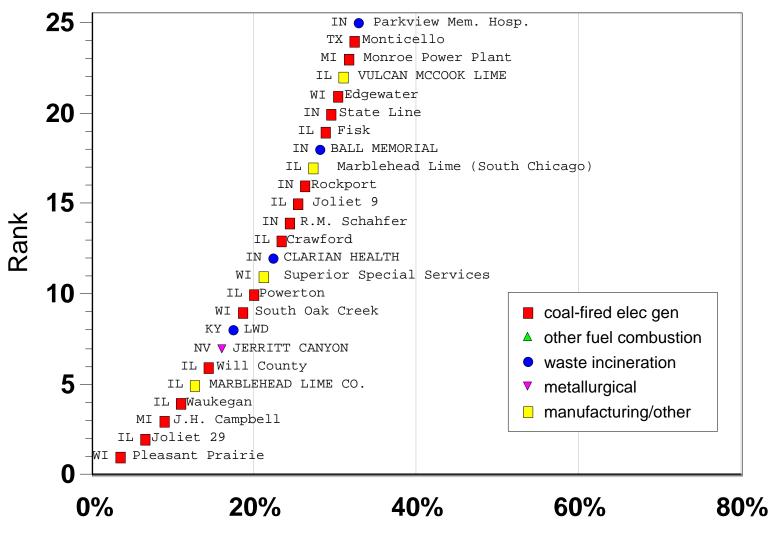


Figure 35-C. Geographical Distribution of 1999 Direct Deposition Contributions to Lake Michigan (more local view)







Cumulative Fraction of Hg Deposition



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Environmental Research 95 (2004) 247-265

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

Modeling the atmospheric transport and deposition of mercury to the Great Lakes $\stackrel{\text{\tiny{$\stackrel{\leftrightarrow}{$}$}}}{\to}$

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

^aNOAA Air Resources Laboratory, 1315 East West Highway R/ARL, Room 3316, Silver Spring, MD 20910, USA ^bCommission for Environmental Cooperation, Montreal, Que., Canada ^cAtmospheric Toxic Processes Service, Meteorological Service of Canada, Environment Canada Quebec region, Montreal, Que., Canada ^dPollutant Data Branch, Environment Canada, Hull, Que., Canada ^eEnvironmental Monitoring and Reporting Branch, Ontario Ministry of the Environment, Toronto, Ont., Canada ^fUniversity of California, Berkeley, CA, USA ^gUS EPA Great Lakes National Program Office, Chicago, IL, USA ^hInternational 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

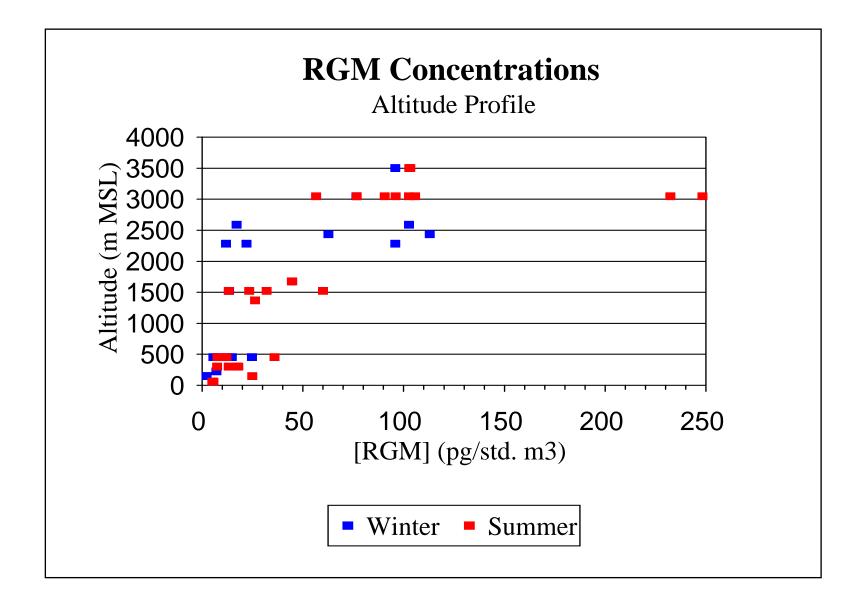
measurements include:

- atmospheric parameters
- trace gases $(O_3, SO_2, NO_x, etc.)$
- speciated mercury



N48RF

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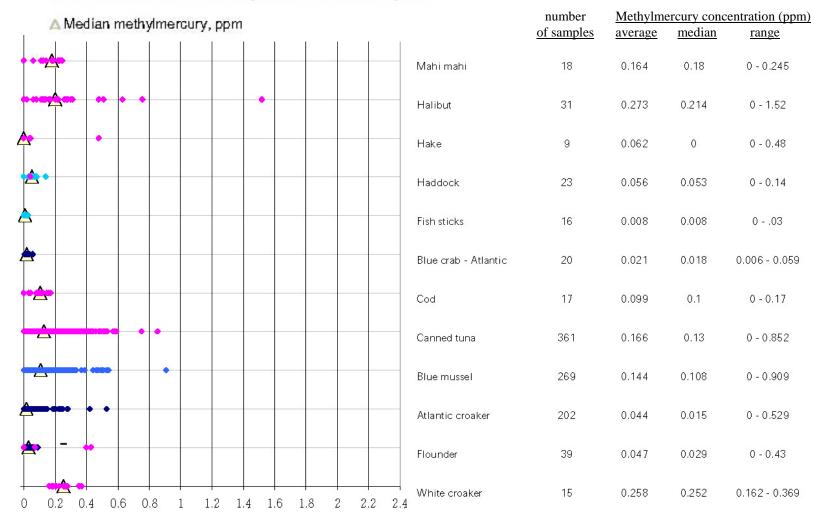
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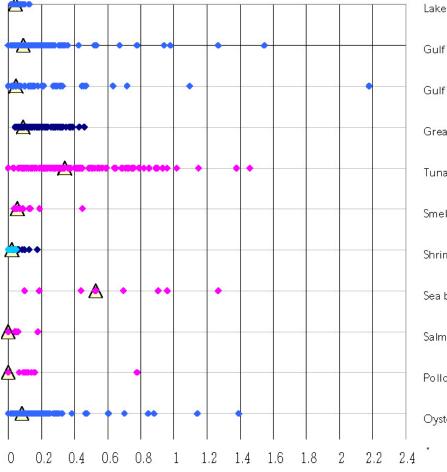
- FDA seafood surveillance program
- FDA Total Diet Study
- NOAA Gulfchem relational database
- EPA Environmental Monitoring and Assessment Program



Distribution of methyl-mercury concentrations in different fish species

source: Environmental Working Group

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- 🛆 Median methylmercury, ppm

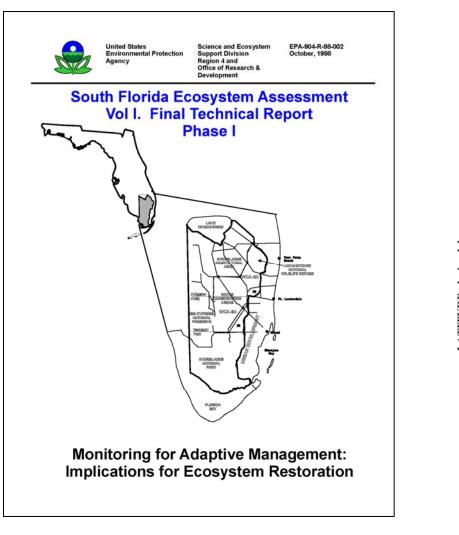


Distribution of methyl-mercury concentrations in different fish species

	number of samples	Methylmercury concentration (ppm) average median range		
Lake Whitefish	88	0.051	0.045	.018126
Gulf Coast Oysters	396	0.123	0.09	0 - 1.55
Gulf Coast Crabs	47	0.228	0.047	1 - 2.18
Great Lakes Salmon	88	0.173	0.09	0.05 - 0.43
Tuna steaks	122	0.417	0.34	0 - 1.46
Smelt	16	0.097	0.054	0.036 - 0.45
Shrimp	59	0.033	0.023	1 - 0.177
Sea bass	10	0.606	0.529	0.1 - 1.27
Salmon	51	0.008	0	0 - 0.18
Pollock	32	0.063	0	0 - 0.78
Oyster	396	0.111	0.083	0 - 1.392

source: Environmental Working Group

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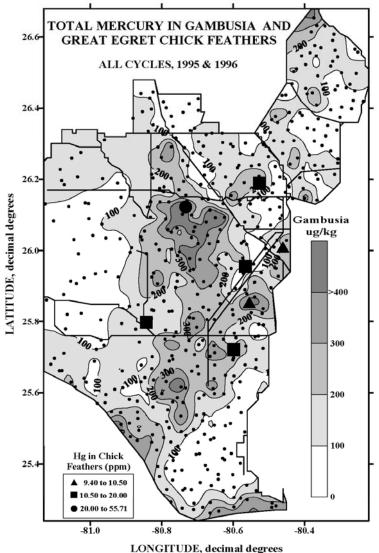


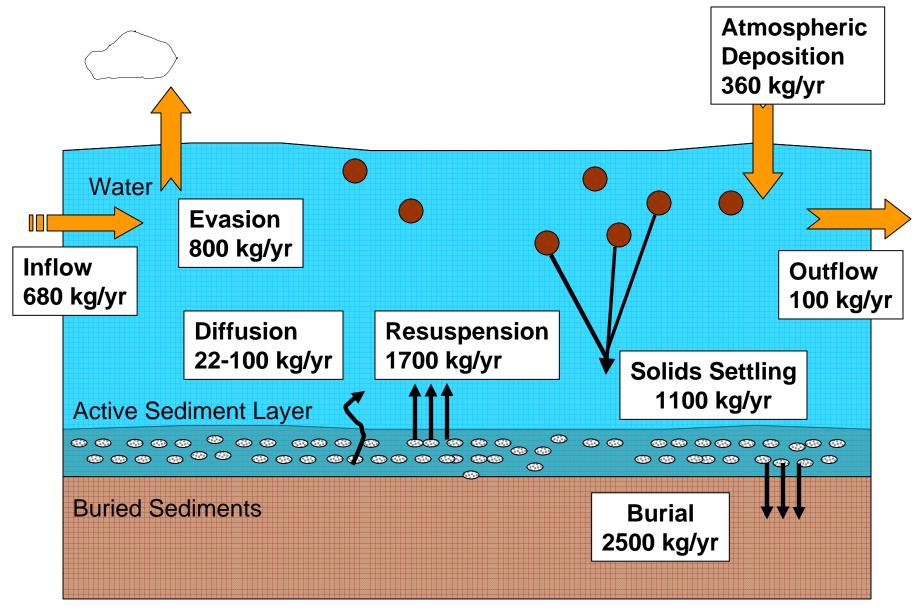
Figure 8.55 Hg concentrations in Great Egret chick feathers and mosquitofish indicate spatial distribution of Hg bioaccumulation.

http://www.epa.gov/region4/sesd/reports/epa904r98002.html

8-71

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Total Mercury Fluxes Lake Ontario

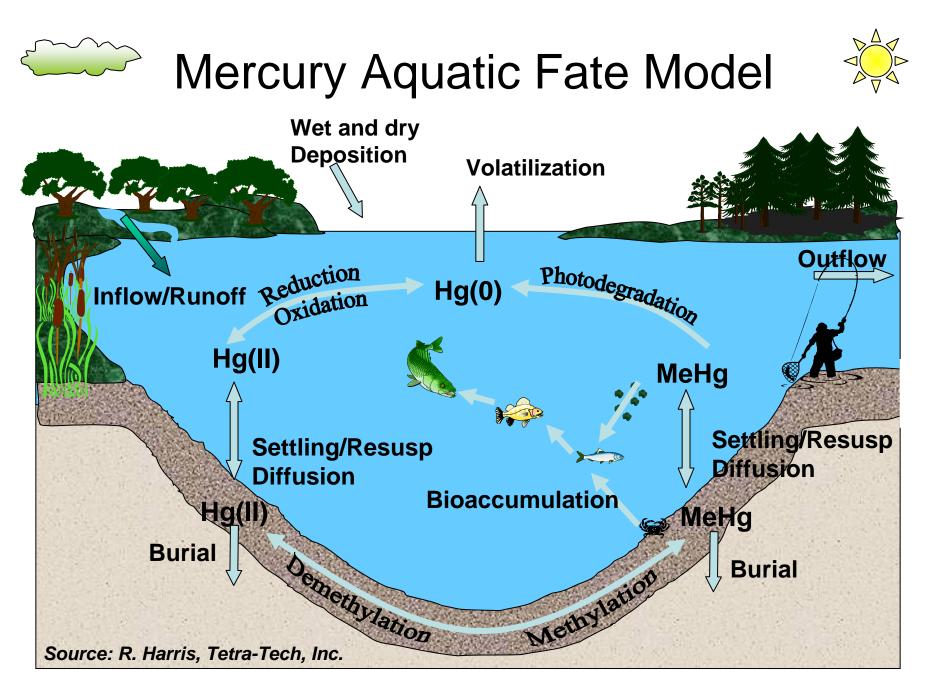


slide courtesy of Elsie Sunderland, USEPA

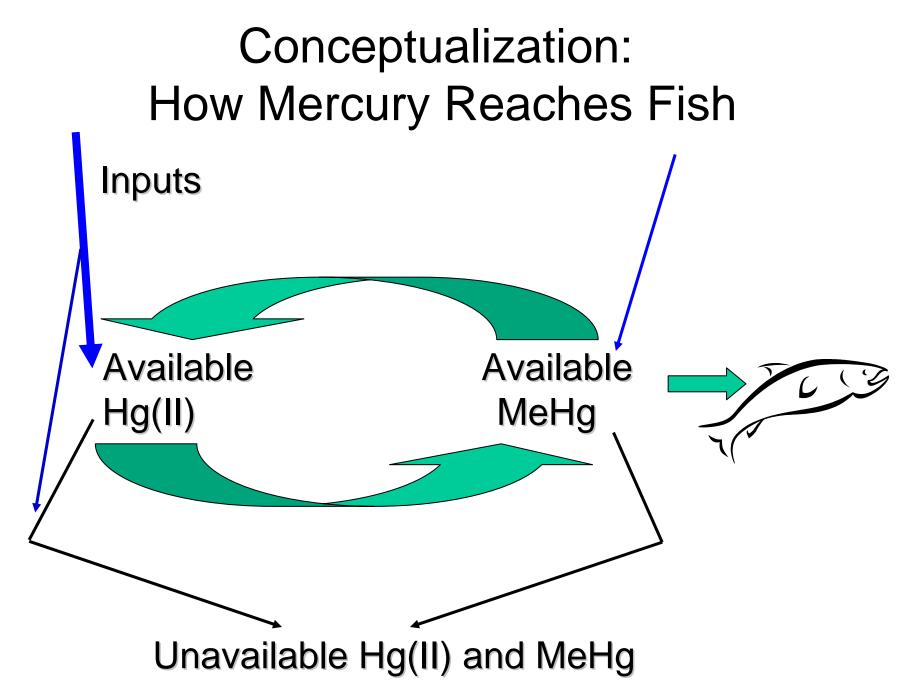
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slide courtesy of Elsie Sunderland, USEPA



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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)

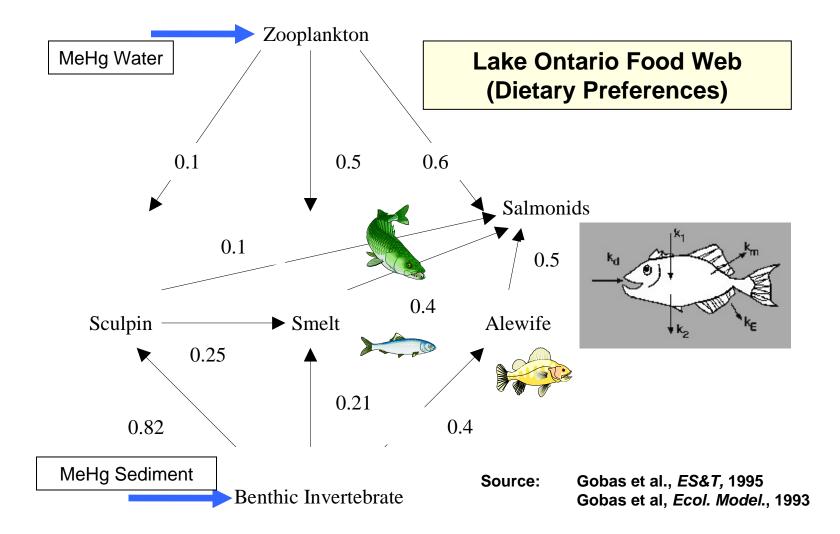
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Bioaccumulation Model Framework



slide courtesy of Elsie Sunderland, USEPA

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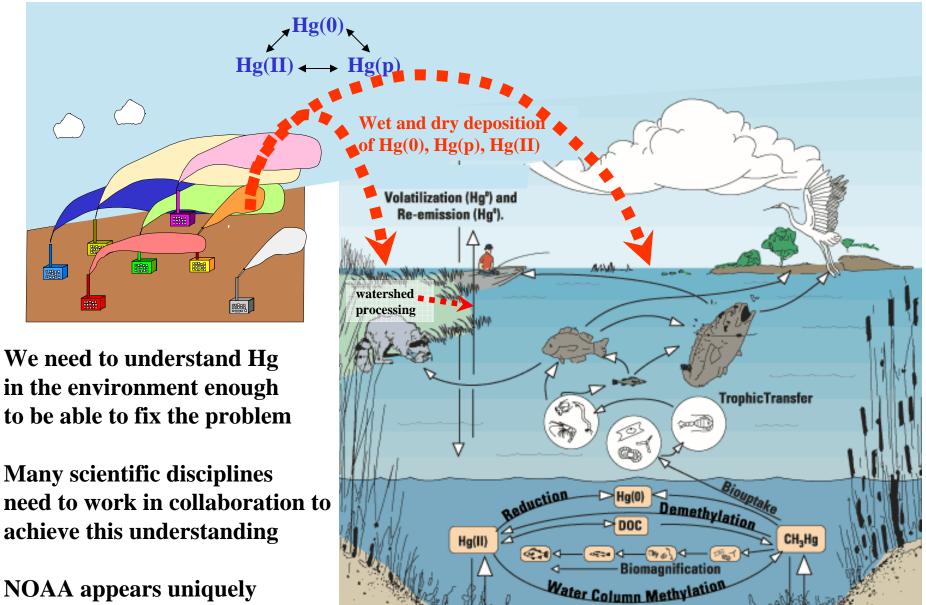
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<u>potential Air Resources Laboratory contributions</u> <u>to collaborative mercury work within NOAA:</u>

- □ 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



Hg(II)

Exchange with Sediments

Methylation in Sediment

CH₃Hg

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

THANKS !

ADDITIONAL BACKGROUND SLIDES



Hg <u>Headquarters Division -- HQ</u> (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)

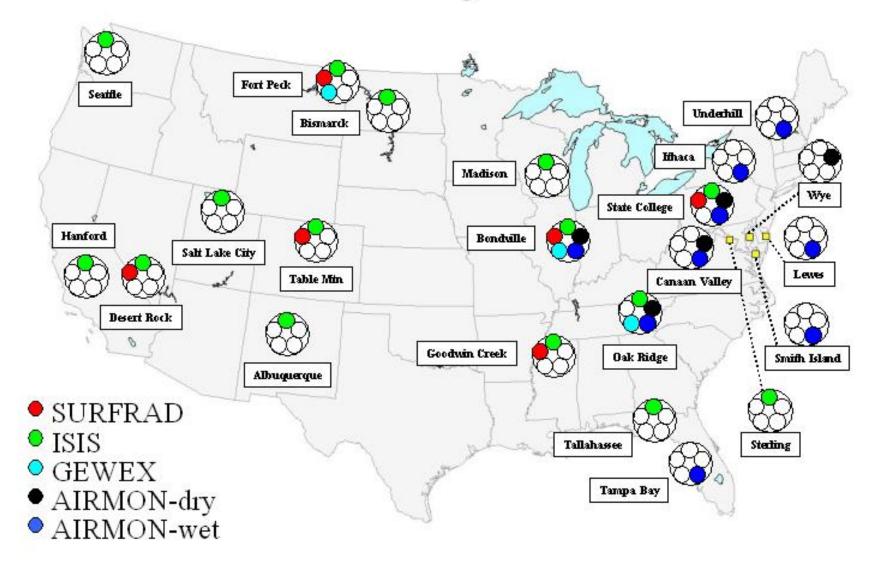
field atmospheric tracer testing facility, for developing transport and diffusion models; FRD's mesonet and modeling expertise supports the Idaho National Laboratory.

<u>Special Operations and Research Division – SORD</u> (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)

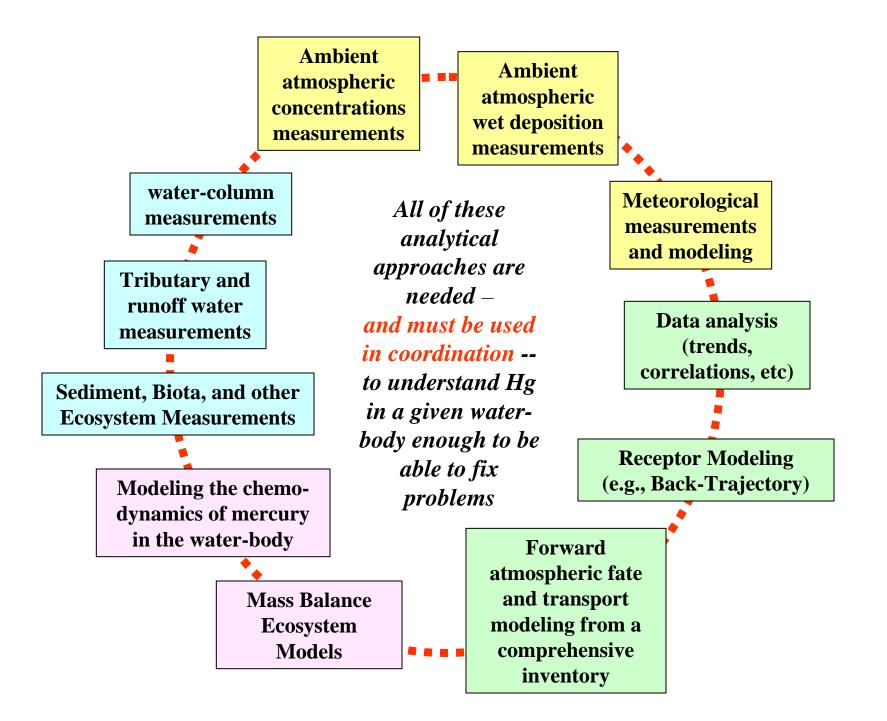
provides basic data on radiation fields, for the next generation of atmospheric transport and dispersion models, by climate assessments, and by evaluations of climate models.

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 Fluorescer	nce 0.1	90 s
H_2O_2	Liquid Phase Fluorescer	nce 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

onvention on Long-Range Transboundary Air Pollution

emep :

Co-operative programme for monitoring ind evaluation of the long-range ransmittation of air pollutants in Europe 1/2003 June 2003

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

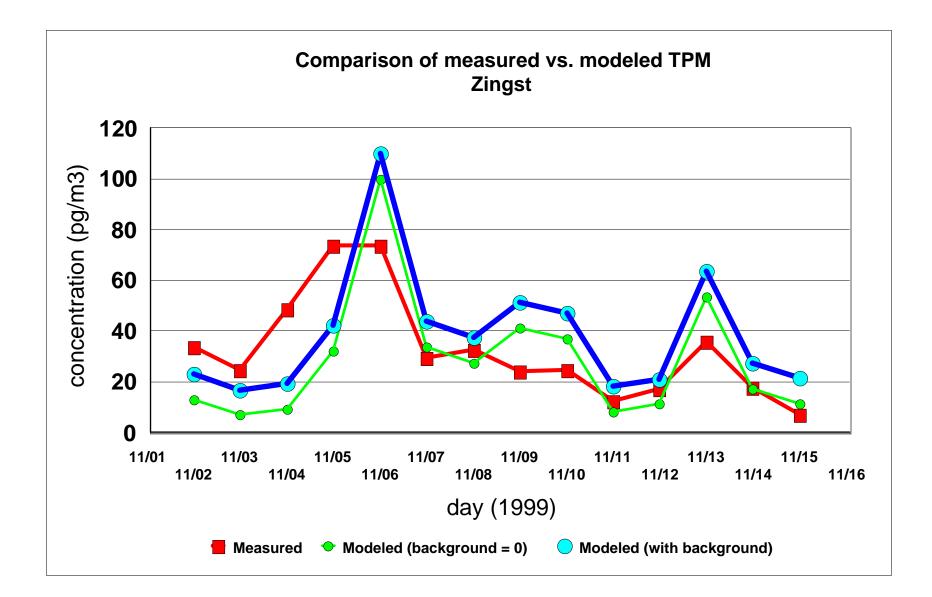
Stage II. Comparison of modeling results with observations obtained during short-term measuring campaigns

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. Sytakov



msc-e

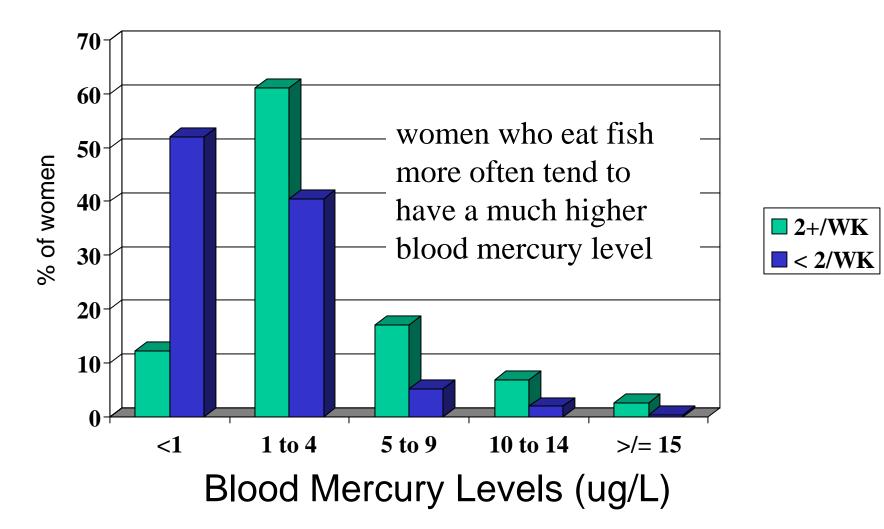


<u>Estimated Number of Newborns with</u> <u>*In Utero* Methylmercury Exposures >/= RfD</u>

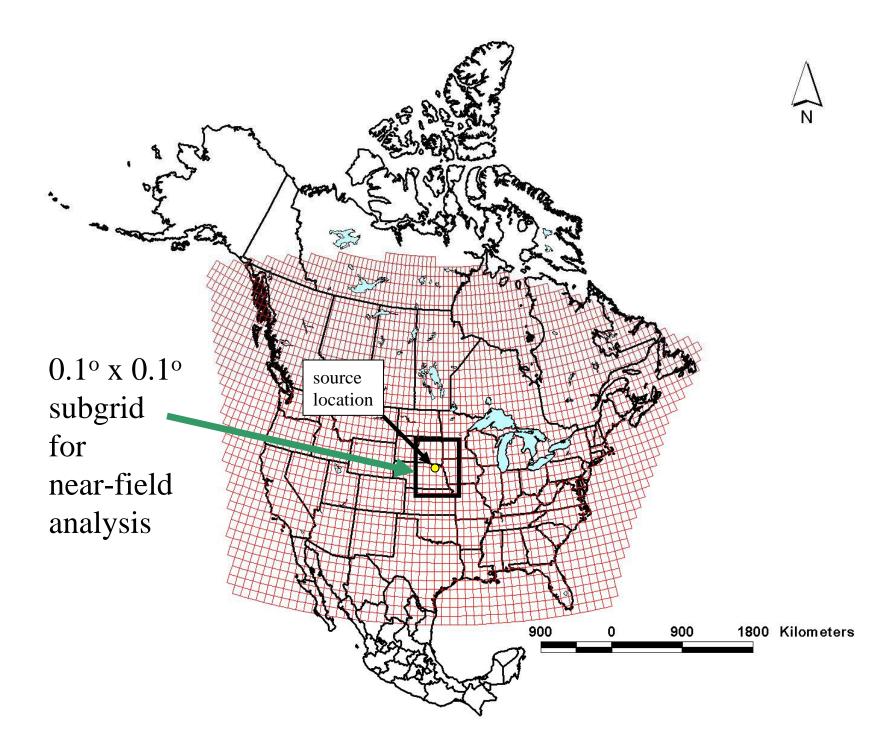
- Number of US births in 2000: 4,058,814 (*National Vital Statistics Reports*).
- 1: 1 ratio of cord to maternal blood [Hg], i.e., 5.8 cord to 5.8 maternal, 7.8% of women had total blood [Hg] >/= 5.8, ~ 300,000 newborns each year > 5.8 ug/L (Mahaffey et al., 2003).
- 1.7: 1 ratio of cord to maternal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

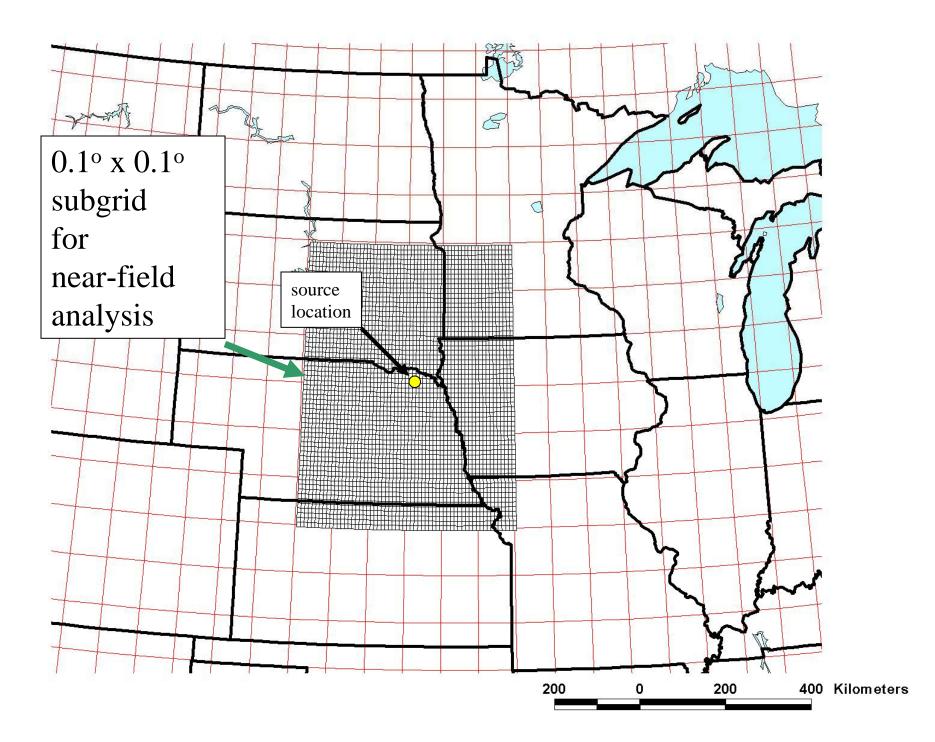
source: Kate Mahaffey, USEPA

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

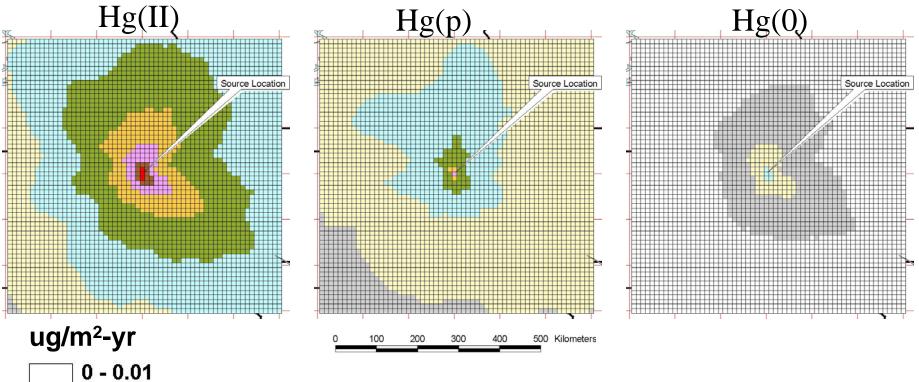


source: Kate Mahaffey, USEPA

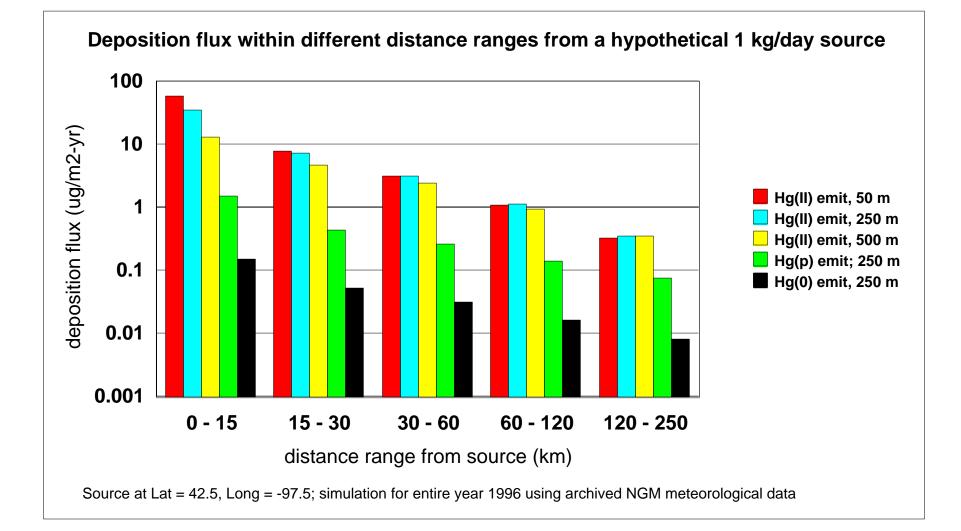


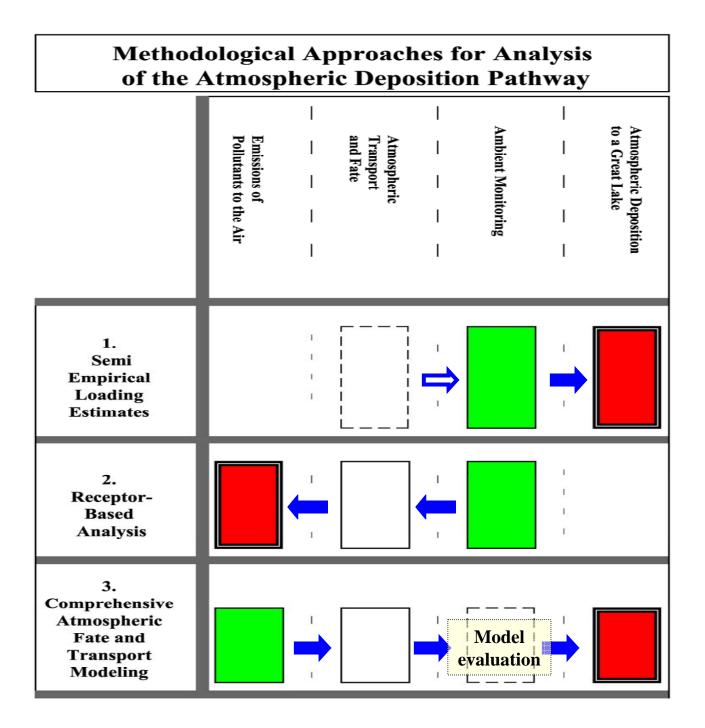


Annual deposition flux arising from a hypothetical 1 kg/day 250 meter high source of different forms of mercury



- 0 0.01 0.01 - 0.03 0.03 - 0.1 0.1 - 0.3 0.3 - 1 1 - 3 3 - 10 10 - 30 30 - 100
- Simulation for the entire year 1996;
- NGM meteorological data (180 km resolution);
- 0.1 x 0.1 degree receptor grid;
- Source location: lat = 42.5, long = -97.5





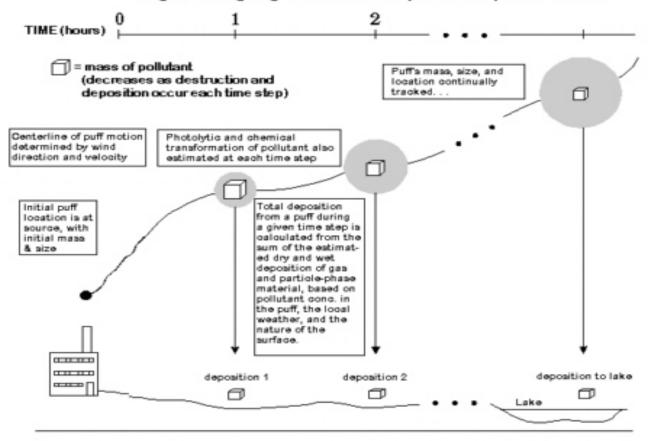


Figure 1. Lagrangian Puff Air Transport and Deposition Model

