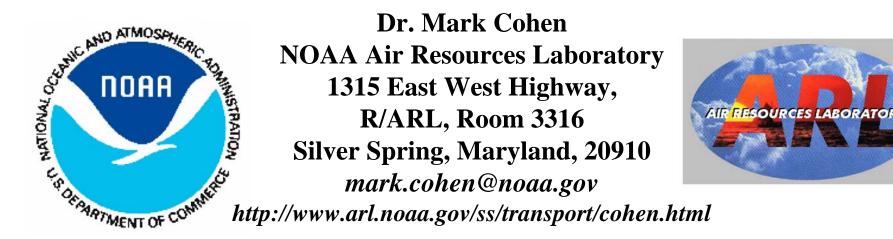
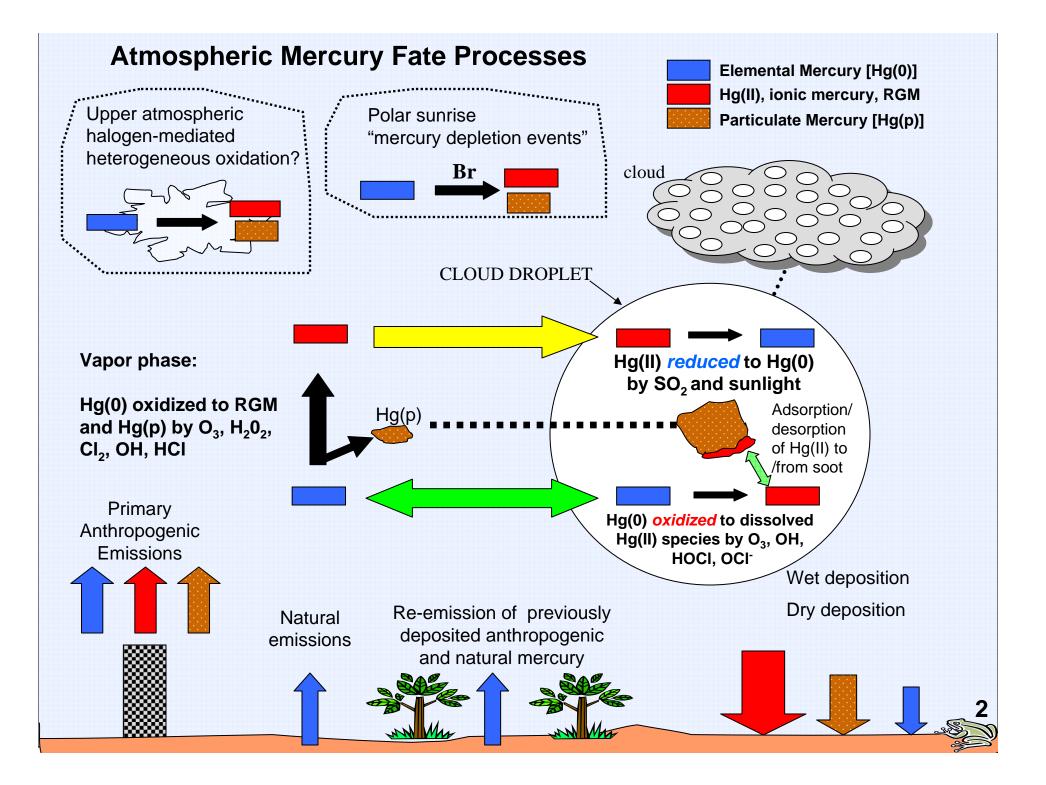
Local and Regional Deposition Impacts of Atmospheric Mercury Emissions



Presentation at Mercury Rule Workgroup Meeting PA Department of Environmental Protection Harrisburg, PA, November 18, 2005



- 1. Depends on amount emitted
- 2. Very close in, depends on stack height
- 3. Depends on form of mercury emitted
- 4. Depends on distance & direction from source
- 5. Very Episodic

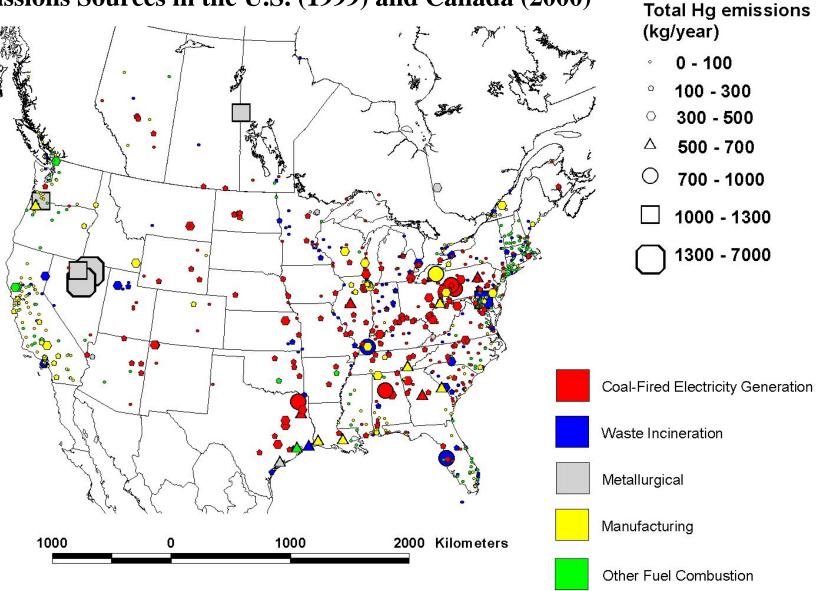
- 6. Depends on chemistry in plume
- 7. Measurement-based evidence:
 - examples
 - advantages
 - limitations
- 8. Modeling-based evidence:
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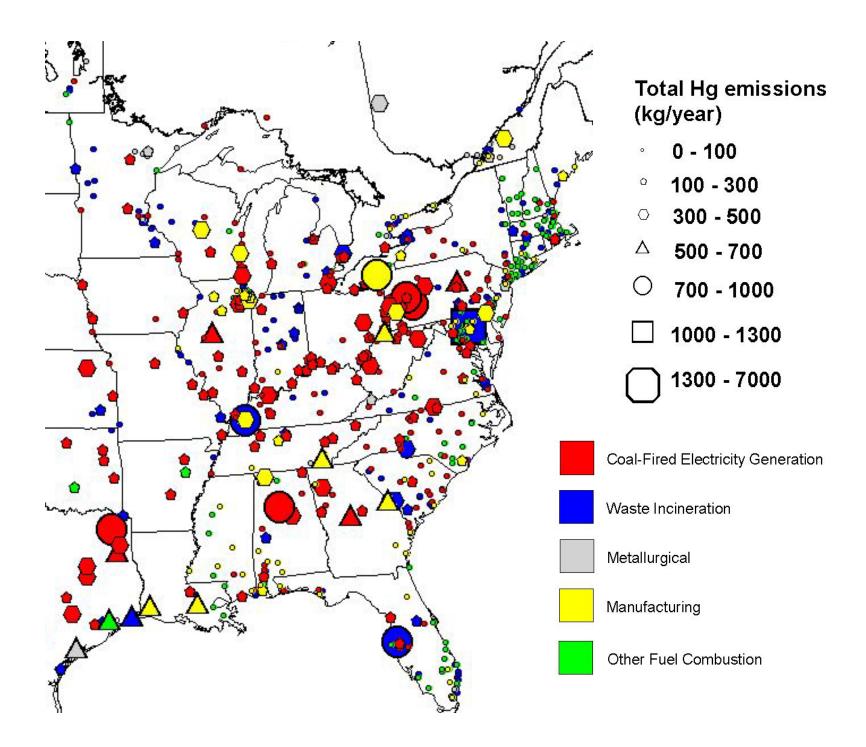
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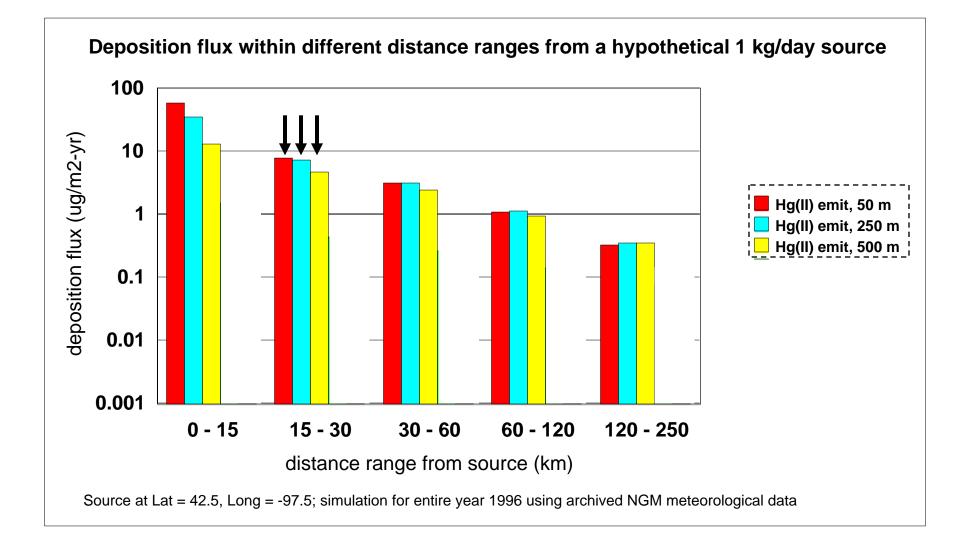
Geographic Distribution of Largest Anthropogenic Mercury Emissions Sources in the U.S. (1999) and Canada (2000)





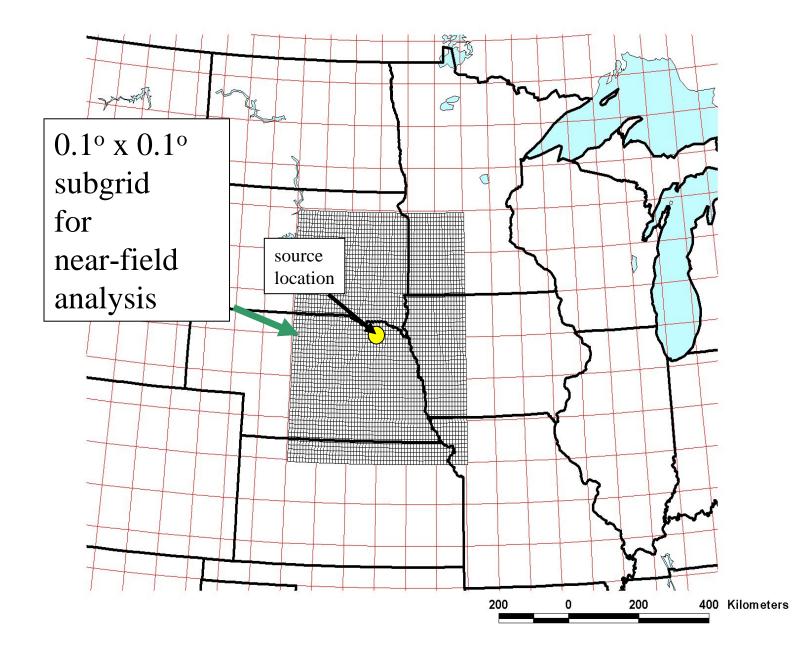
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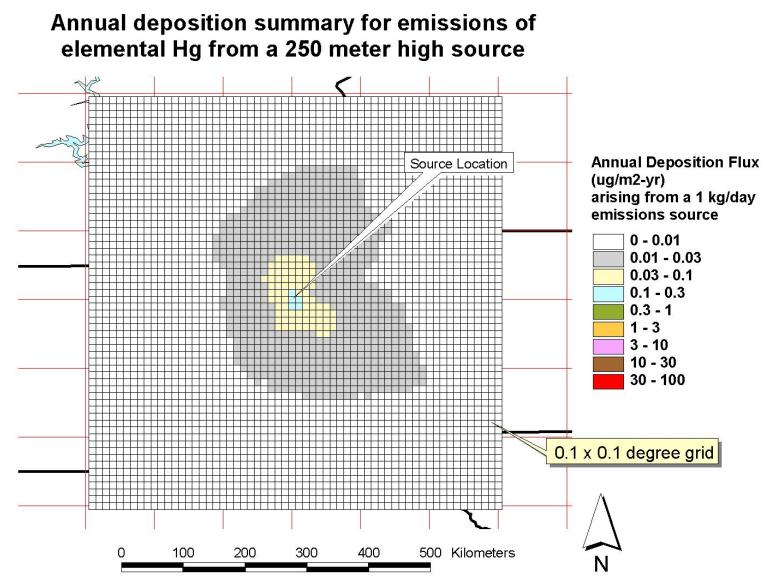
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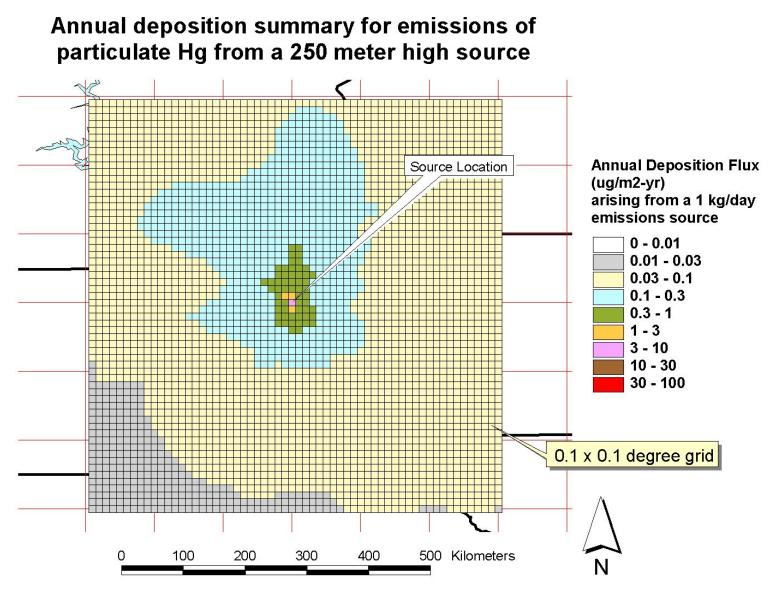
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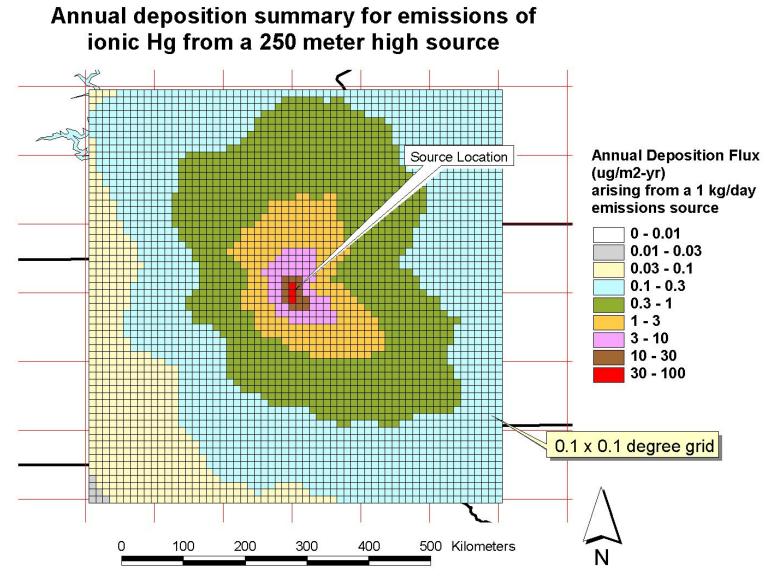




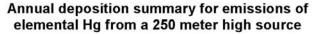
Hypothetical emissions source at lat = 42.5, long = -97.5; simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

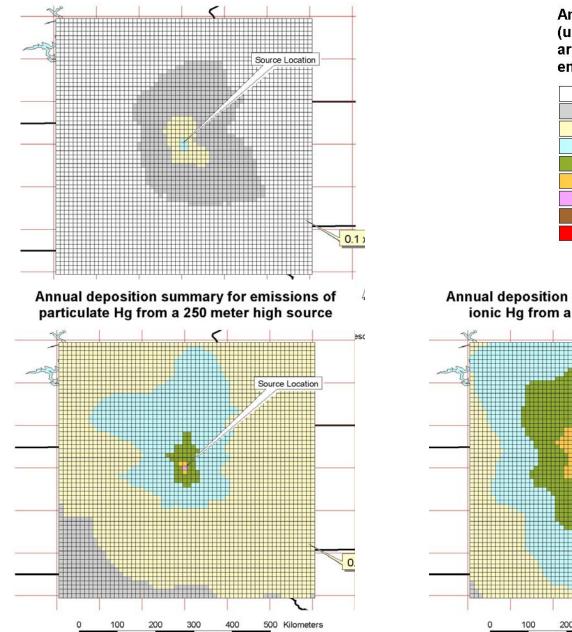


Hypothetical emissions source at lat = 42.5, long = -97.5; simulation for entire year 1996 using archived NGM meteorology (180 km resolution)



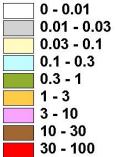
Hypothetical emissions source at lat = 42.5, long = -97.5; simulation for entire year 1996 using archived NGM meteorology (180 km resolution)



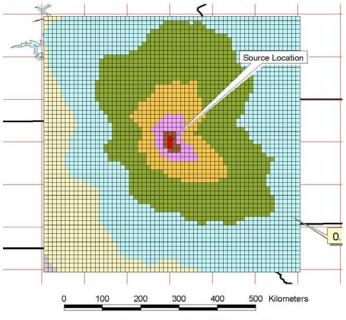


Hypothetical emissions source at lat = 42.5, long = -97.5; simulation for entire year 1996 using archived NGM meteorology (180 km $\scriptstyle\rm I$

Annual Deposition Flux (ug/m2-yr) arising from a 1 kg/day emissions source

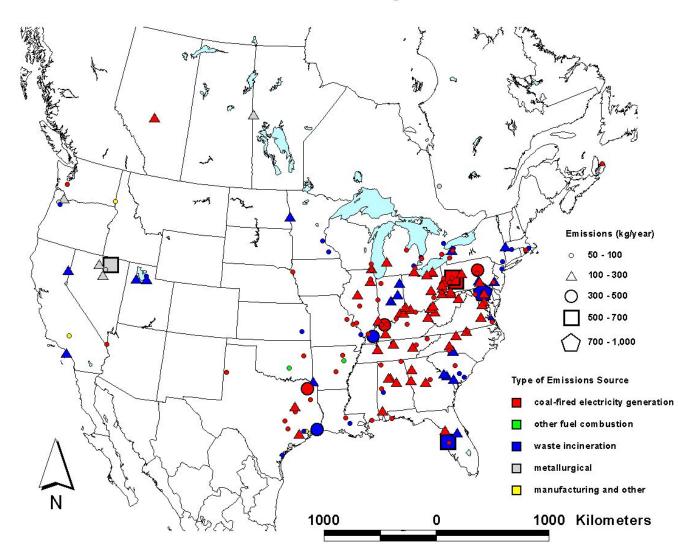


Annual deposition summary for emissions of ionic Hg from a 250 meter high source

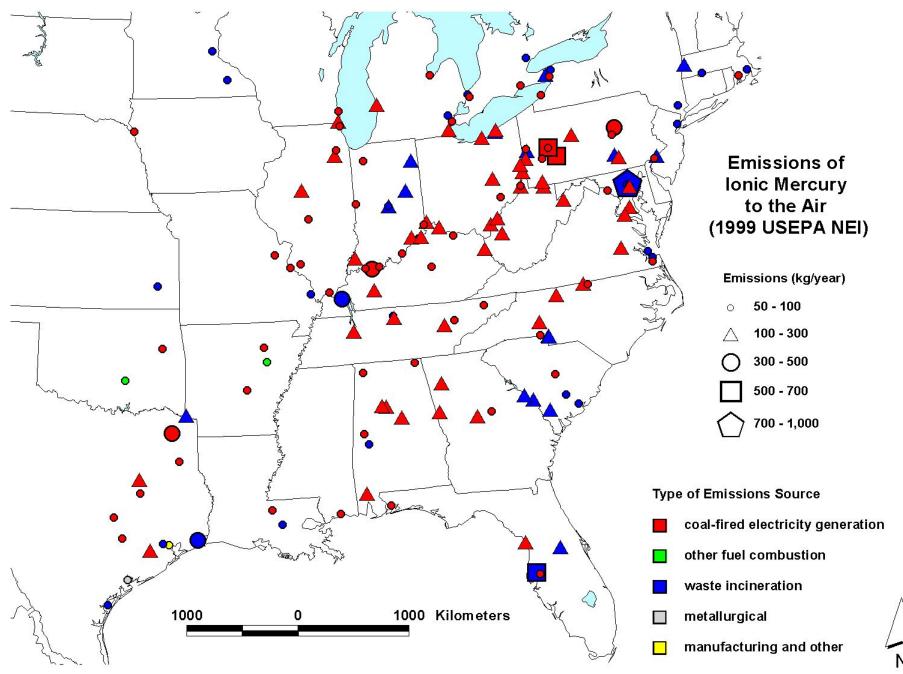


Hypothetical emissions source at lat = 42.5, long = -97.5; simulation for entire year 1996 using archived NGM meteorology (180 km r

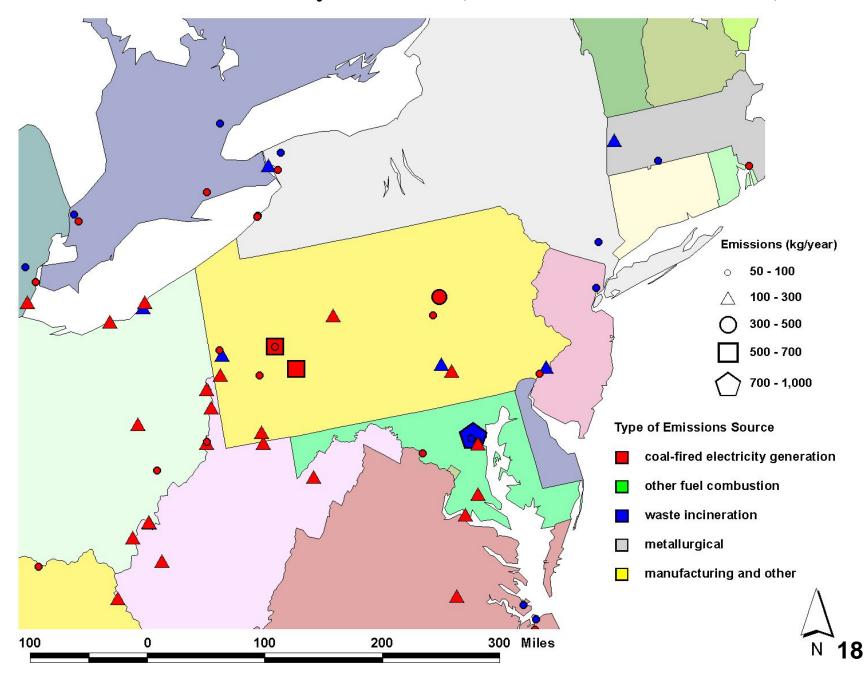
So where is RGM emitted?



Emissions of Ionic Mercury to the Air



N 17

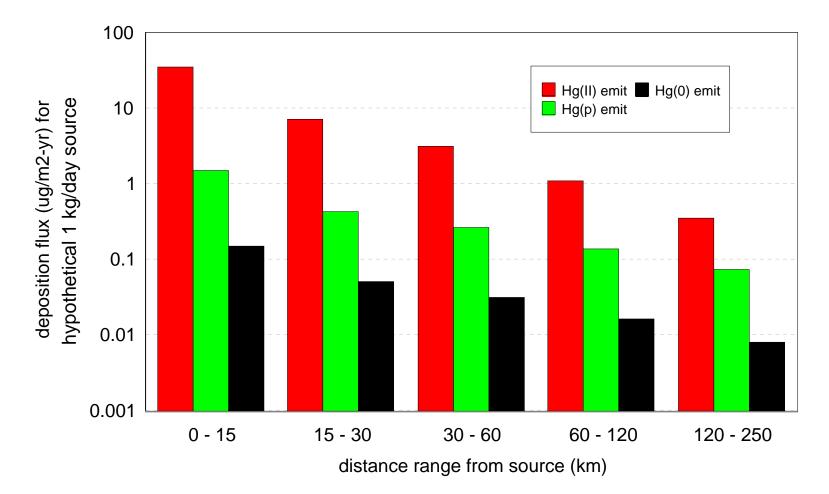


Reactive Gaseous Mercury Emissions (based on USEPA 1999 NEI)

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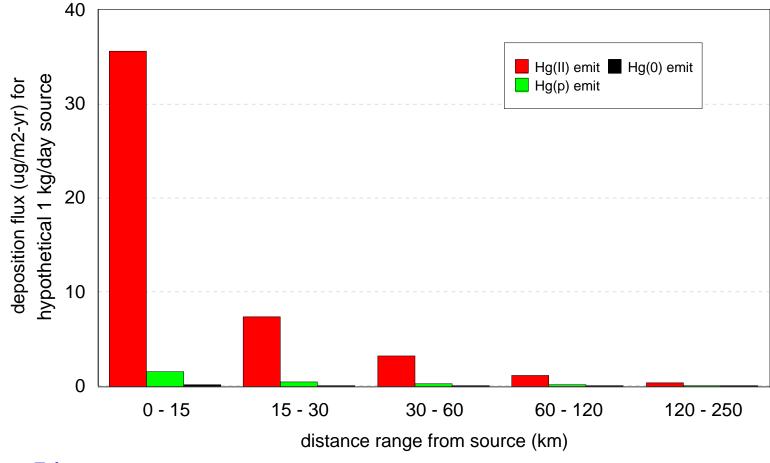
Why is emissions speciation information critical?



Logarithmic

NOTE: distance results averaged over all directions – Some directions will have higher fluxes, some will have lower 20

Why is emissions speciation information critical?

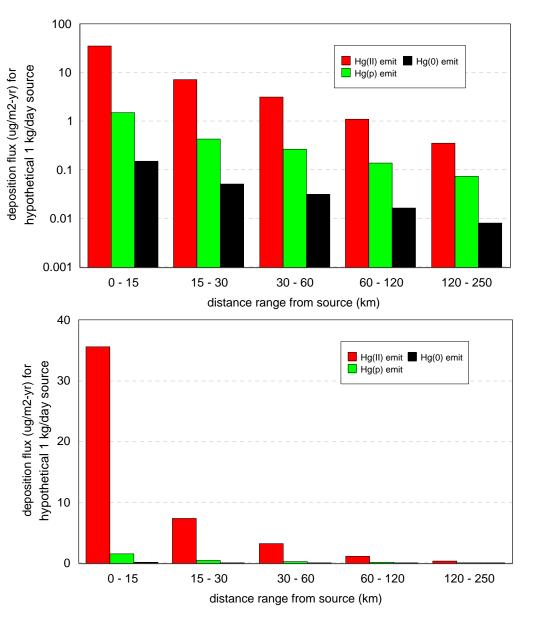


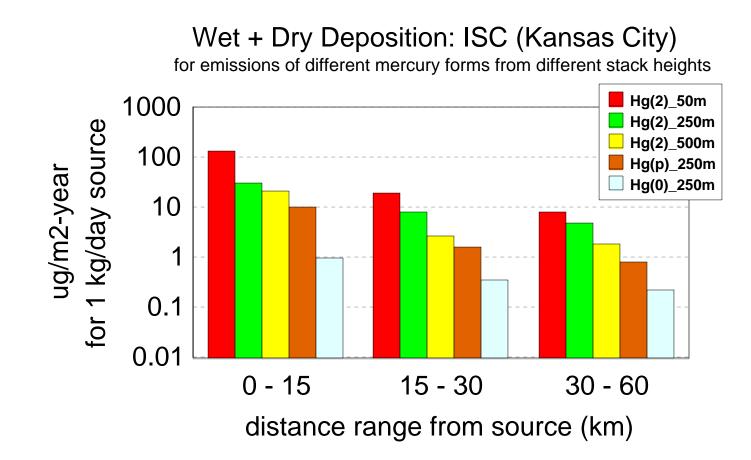
Linear

Why is emissions speciation information critical?

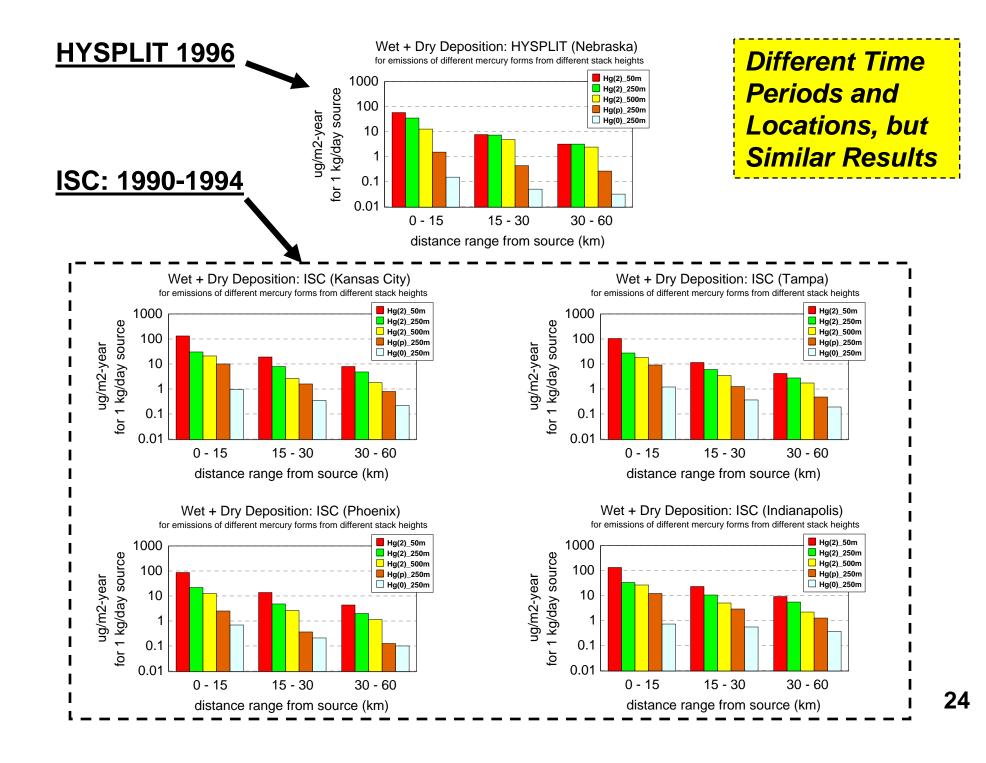
Logarithmic

Linear





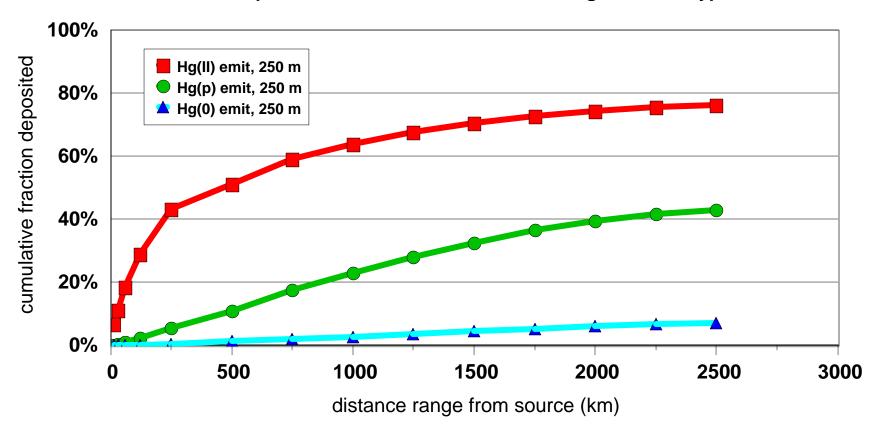
Calculated from data used to produce Appendix A of USEPA (2005): Clean Air Mercury Rule (CAMR) Technical Support Document: Methodology Used to Generate Deposition, Fish Tissue Methylmercury Concentrations, and Exposure for Determining Effectiveness of Utility Emissions Controls: Analysis of Mercury from Electricity Generating Units



The <u>fraction deposited</u> and the <u>deposition flux</u> are both important, but they have very different meanings... The fraction deposited nearby can be relatively "small",

But the area is also small, and the relative deposition flux can be very large...

Cumulative Fraction Deposited Out to Different Distance Ranges from a Hypothetical Source

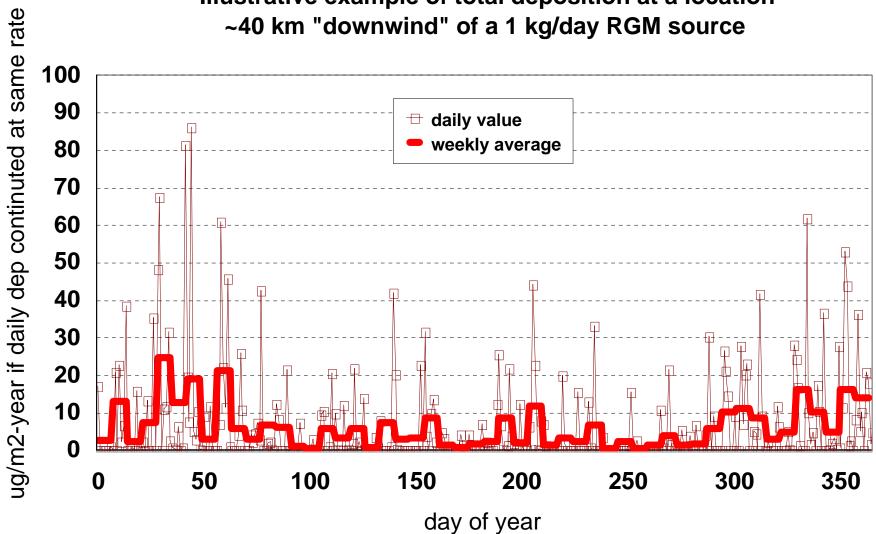


Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data

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Illustrative example of total deposition at a location

27

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- If significant reduction of RGM to Hg(0) is occuring in power-plant plumes, then it would have a big impact on local/regional deposition
- No known chemical reaction is capable of causing significant reduction of RGM in plumes –

e.g. measured rates of SO₂ reduction can't explain some of the claimed reduction rates

□ Very hard to measure

- □ Aircraft
- □ Static Plume Dilution Chambers (SPDC)
- **Ground-based measurements**

Interconversion of Emitted Atmospheric Mercury Species in Coal-Fired Power Plant Plumes

E. Prestbo and P. Swartzendruber (Frontier Geosciences) L. Levin (EPRI) D. Laudal, R. Schulz and G. Dunham (EERC), W. Aljoe (U.S. Department of Energy) J. Jansen and L. Monroe (Southern Company) R. Valente (TVA) D. Michaud (WE Energies)

> Ail Quality / Conference September 2005

SPDC - Hg Species Interconversion Results for all Study Sites 1995-2003

				Hg(II) to Hg ⁰ conversion as a percent of total	Mass	
Power Plant	Year	Coal	PCD	mercury	Balance	n
Presque Ilse, MI*	1995	W-SB	ESP 1 Unit	33% ± 6.8%	96% ± 14%	10
Dickerson, MD*	1998	E-SB	Scrubber 2 Units ESP Scrubber	41% ± 7.9%	128% ± 22%	8
Dickerson, MD	1998	MW	and-Carbon Inj.	-23% ± 12%	84% ± 15%	7
EERC Pilot Plant	2000	E-B	ESP	23% ± 6.0%	86% ± 19%	8
EERC Pilot Plant	2000	E-B	Baghouse	11% ± 8.0%	75% ± 9.2%	6
Bowen, GA	2002	E-B	ESP	9.4% ± 2.9%	109% ± 6.6%	6
Pleasant Prairie, WI	2003	PRB	ESP/SCR	3.2% ± 3.7%	90% ± 22%	3
*value may be biased	high du	ie to bias i	n fluegas spe	ciation measurmen	ts	

Eric Prestbo Ph.D. (ericp@frontiergeosciences.com)

Frontier GeoSciences Inc.

Hg(II) to Hg ⁰ conversion as a						
Power Plant	Method	percent of total mercury	Mass Balance	n		
Bowen, GA	2002	9.4% ± 2.9%	109% ± 6.6%	6		
"	Aircraft	13%		4		
Pleasant Prairie, Wl	SPDC	3.1% ± 3.7%	90% ± 22%	3		
	Aircraft - 0km	15% ± 6.5%	123% ± 24%	5		
	Aircraft - 8km	23% ± 2.6%	198% ± 120%	4		
	Aircraft - 16km	24% ± 5.3%	152% ± 62%	3		

Eric Prestbo Ph.D. (ericp@frontiergeosciences.com)

Frontier GeoSciences Inc.

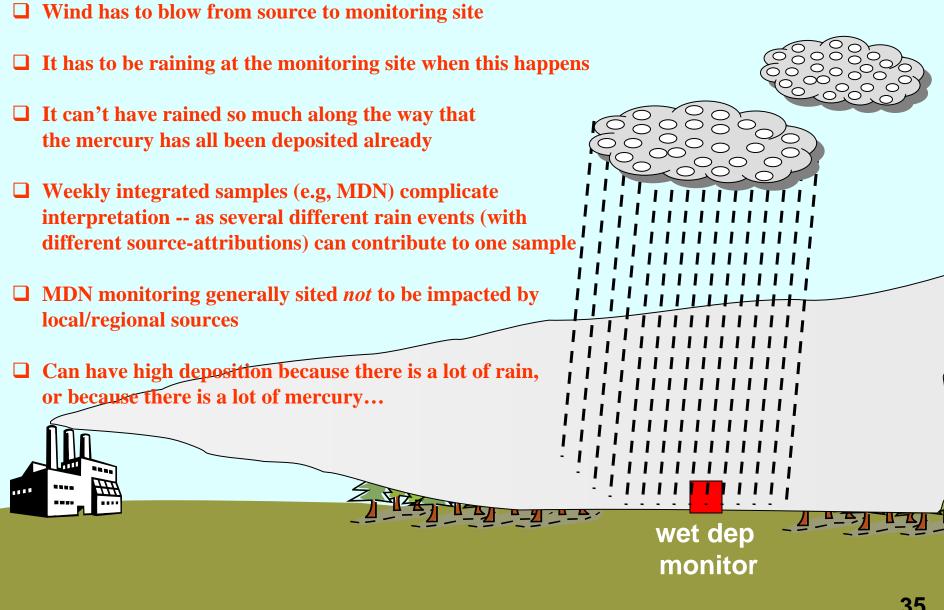
How important is RGM reduction in power-plant plumes?

- Most current state-of-the-science models including the EPA CMAQ model used to generate analyses for the CAIR/CAMR rulemaking process – do not include processes that lead to significant reduction in plumes
- Recent measurement results show less reduction
- Significant uncertainties e.g., mass balance errors comparable to measured effects...
- Current status inconclusive... but weight of evidence suggest that while some reduction may be occurring, it may be only a relatively small amount

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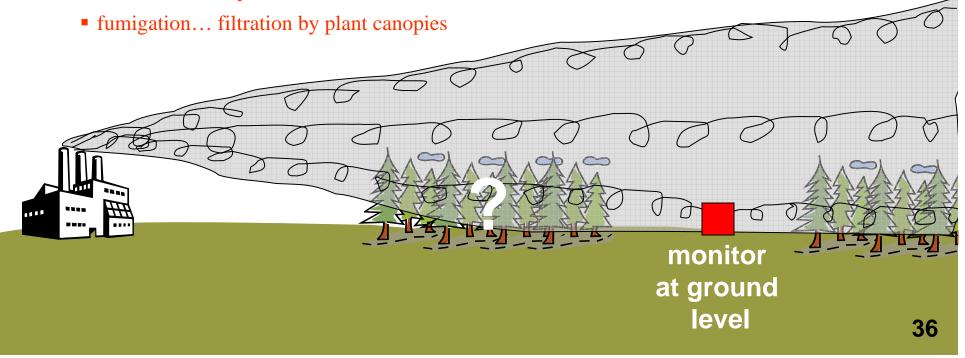
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Challenges of using wet deposition data to assess local and regional deposition impacts...



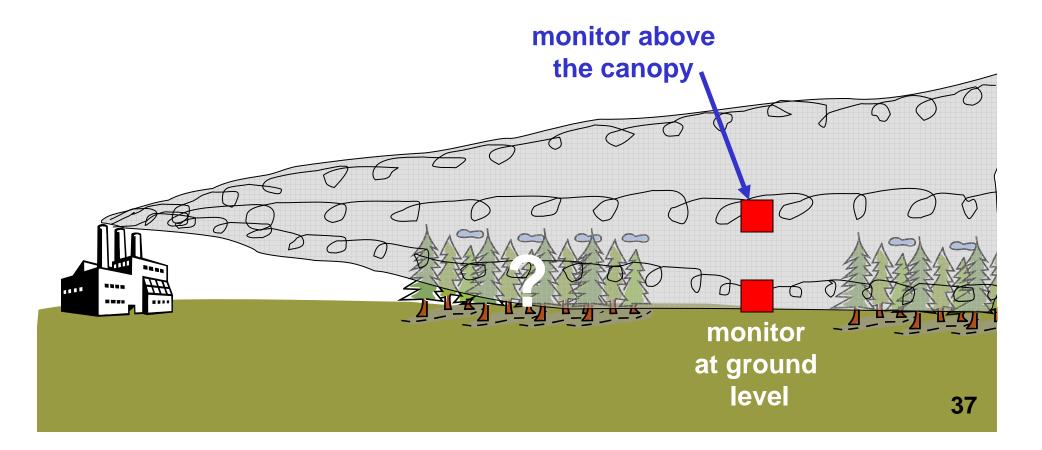
<u>Challenges of using air concentration data</u> <u>to assess local and regional deposition impacts...</u>

- Need speciated data (Hg0, Hg(p), RGM)
- Relatively expensive and time-consuming
- Still have problem of having the plume hit the site, but can measure continuously... and the plume hit and rain doesn't have to occur at the same time (as with wet dep monitors) ...
- Results from ground-level monitors can be hard to interpret
 - rapid dry deposition ... large vertical gradients ... measuring right where things are changing very rapidly ... don't want the whole analysis to depend on whether the sampler was at an elevation of 10 meters or 2 meters...



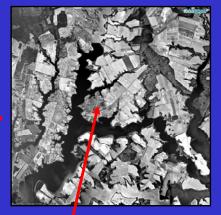
Observations of "depleted" RGM at ground-based stations downwind of power plants – sometimes thought to be evidence of RGM reduction to Hg0 -- might be strongly influenced by RGM dry deposition...

would be better to have a monitor far above the canopy...



Summer 2004 NOAA ARL Hg Measurement Sites

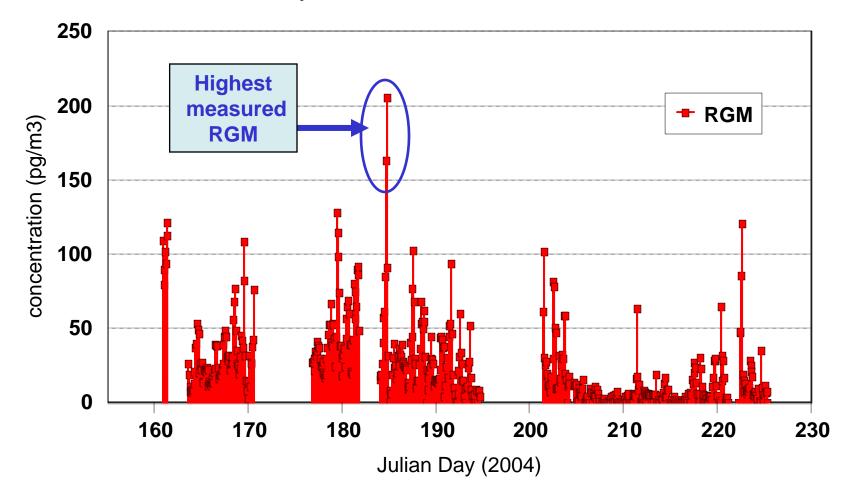




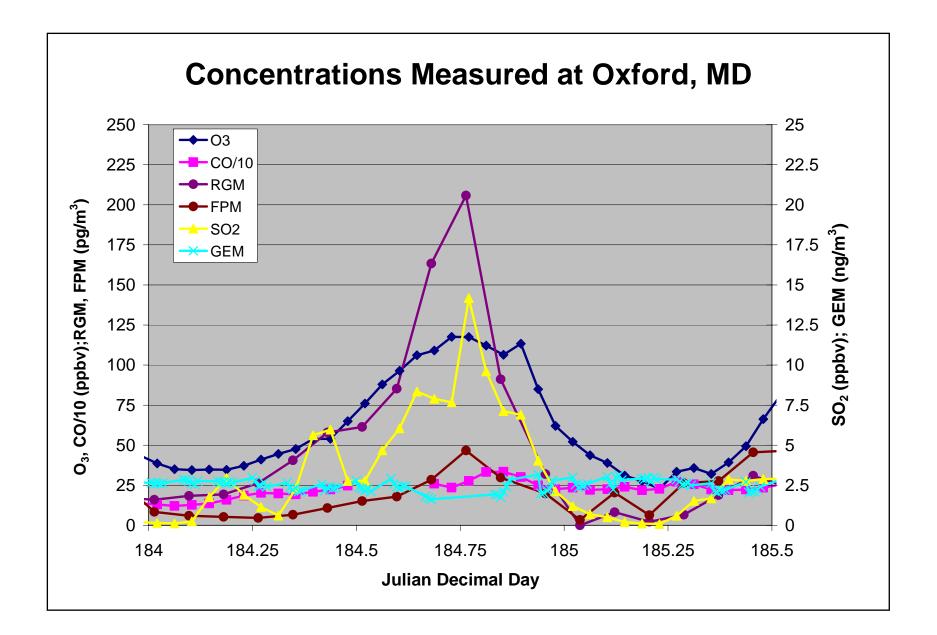
Wye Research and Education Center (38.9131EN, 76.1525EW)



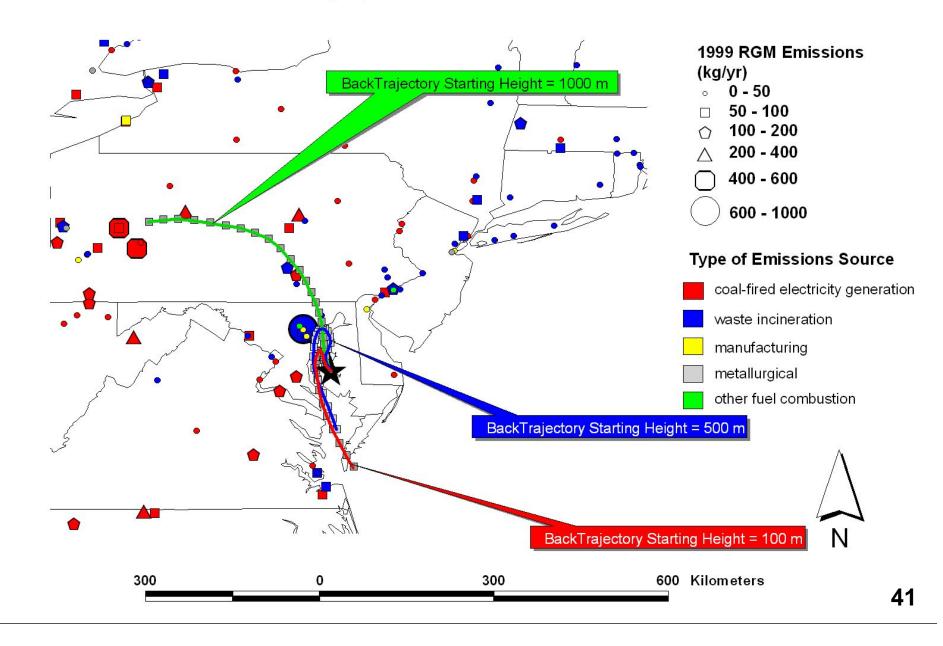
Cooperative Oxford Lab (38.678EN, 76.173EW)



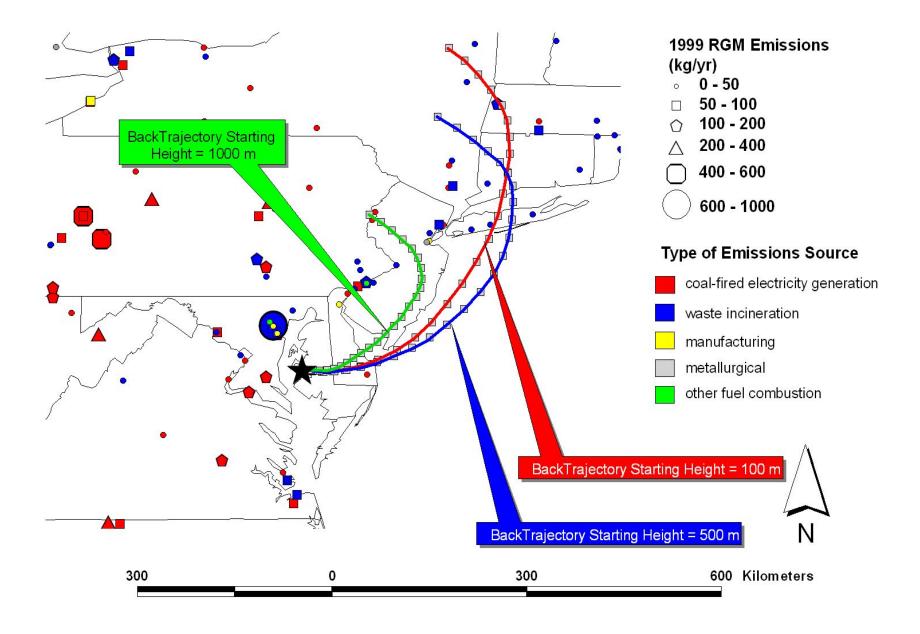
Measured Atmospheric Concentrations at Oxford MD, Summer 2004



Oxford July 2, 2004 Peak Concentration in RGM





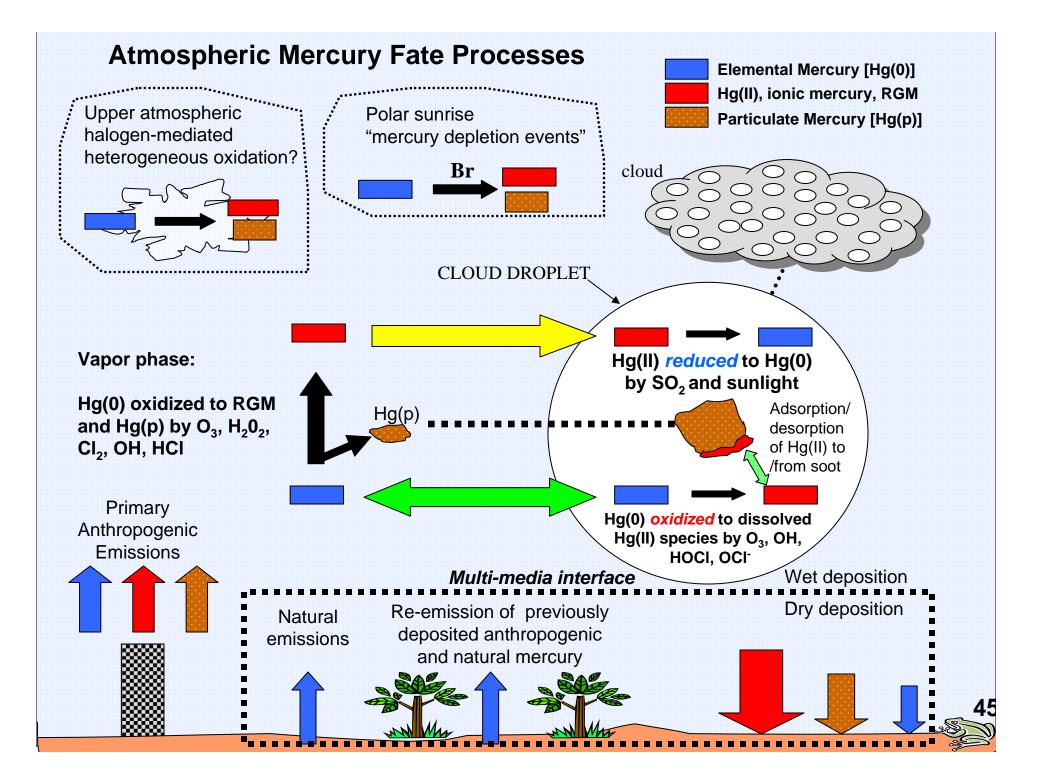


Measurements tell you the "exact" answer (ignoring measurement uncertainties for the moment) but it is usually very difficult to figure out what that answer is telling you, e.g., regarding source-attribution for measured quantities

What are the local and regional deposition impacts of atmospheric mercury emissions?

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So how good are current models, and how do they compare with one another?

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

EMEP/TFMM Workshop on the Review of the MSC-E Models on HMs and POPs Oct 13-14, 2005 Hotel Mir, Moscow Russia Summary presented by Mark Cohen, NOAA Air Resources Laboratory, Silver Spring, MD, USA

EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury											
Intro-	Stage I	Stage I Stage II				Stage III					
duction	Chemistry	Hg^{0}	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	sions			
			Par	ticipa	ants						
D.	Syrakov	••••			Bul	garia	.NIMH				
A.	Dastoor,	D. Davi	gnon	•••••	Car	nada	MSC-C	Can			
J.	Christens	en	• • • • • • • • • •	•••••	DenmarkNERI						
G.	Petersen	, R. Ebir	nghaus		GermanyGKSS						
J. 1	Pacyna				Noi	way	. NILU				
J. 1	Munthe, I	l. Wängl	berg		Sweden IVL						
R.	Bullock			USAEPA		.EPA					
Μ	. Cohen, l	R. Artz,	R. Draxl	USANOAA							
C. Seigneur, K. Lohman						A	AER/E	PRI			
A.	Ryabosh	apko, I.	ovEM	EP	MSC-E						

EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury									
Intro- duction	Stage I	Stage II			Stage III			Conclu-	
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	sions	

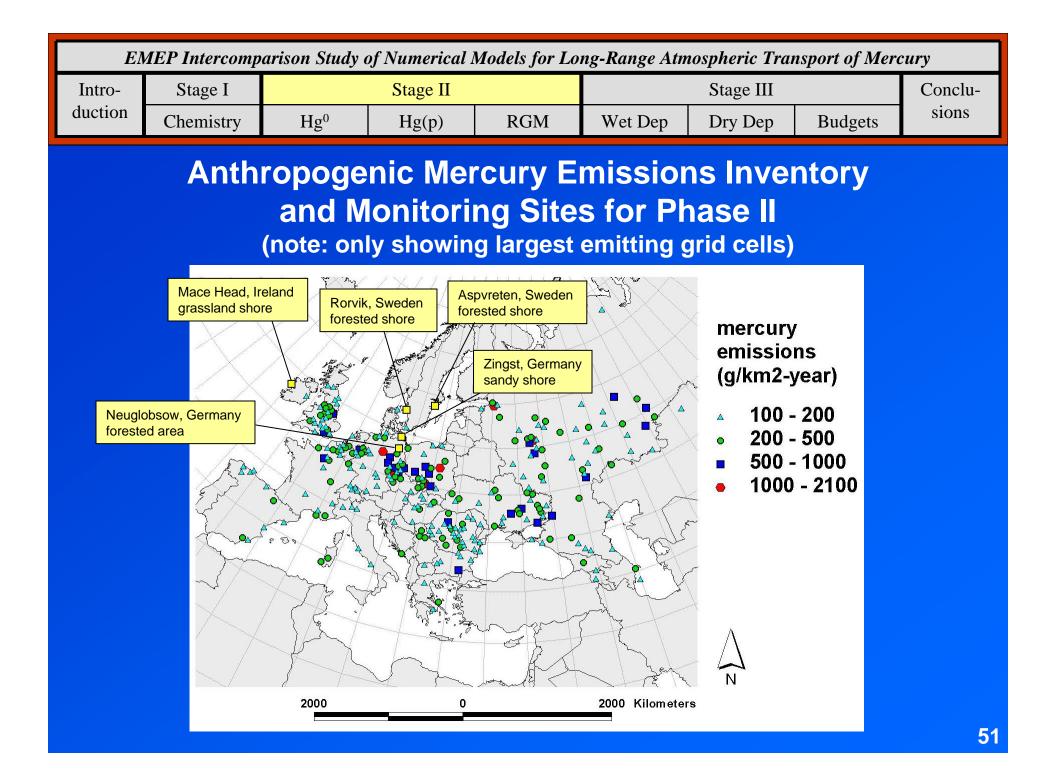
Intercomparison Conducted in 3 Stages

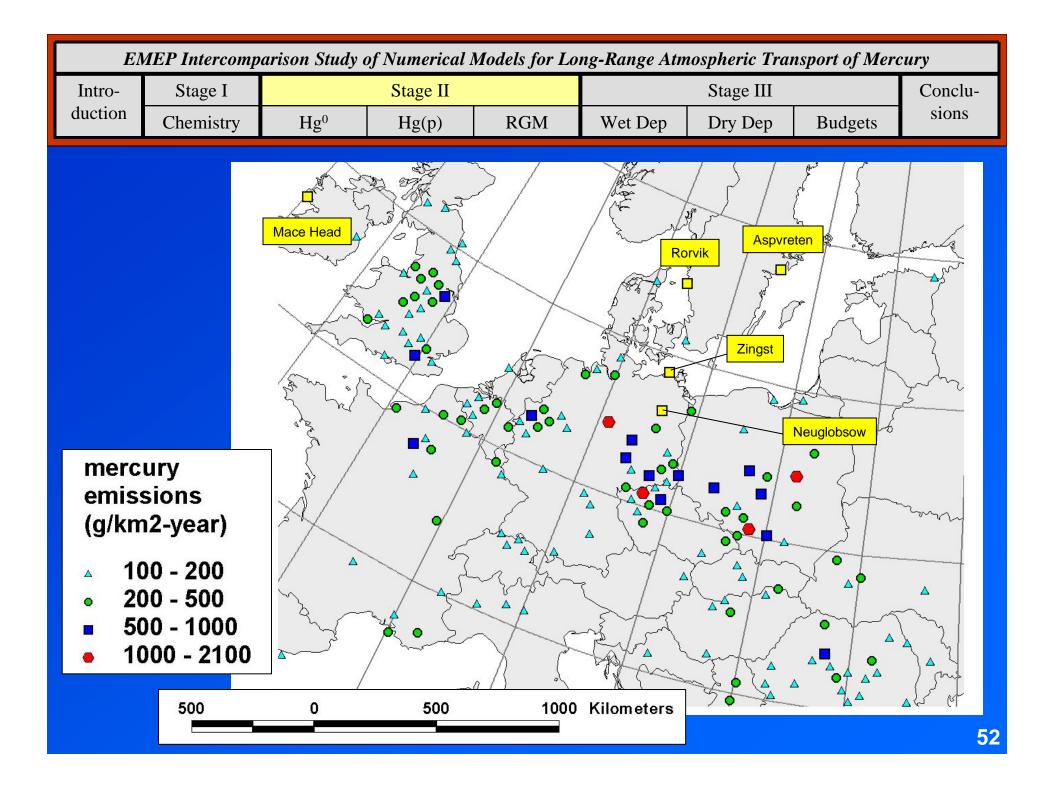
I. Comparison of chemical schemes for a cloud environment

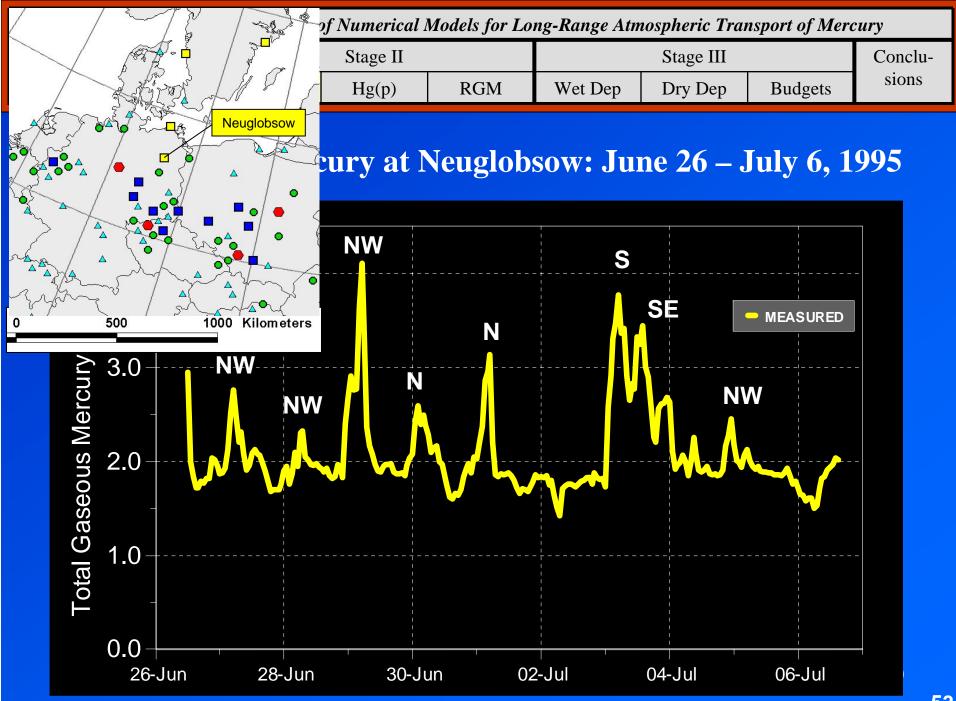
II. Air Concentrations in Short Term Episodes

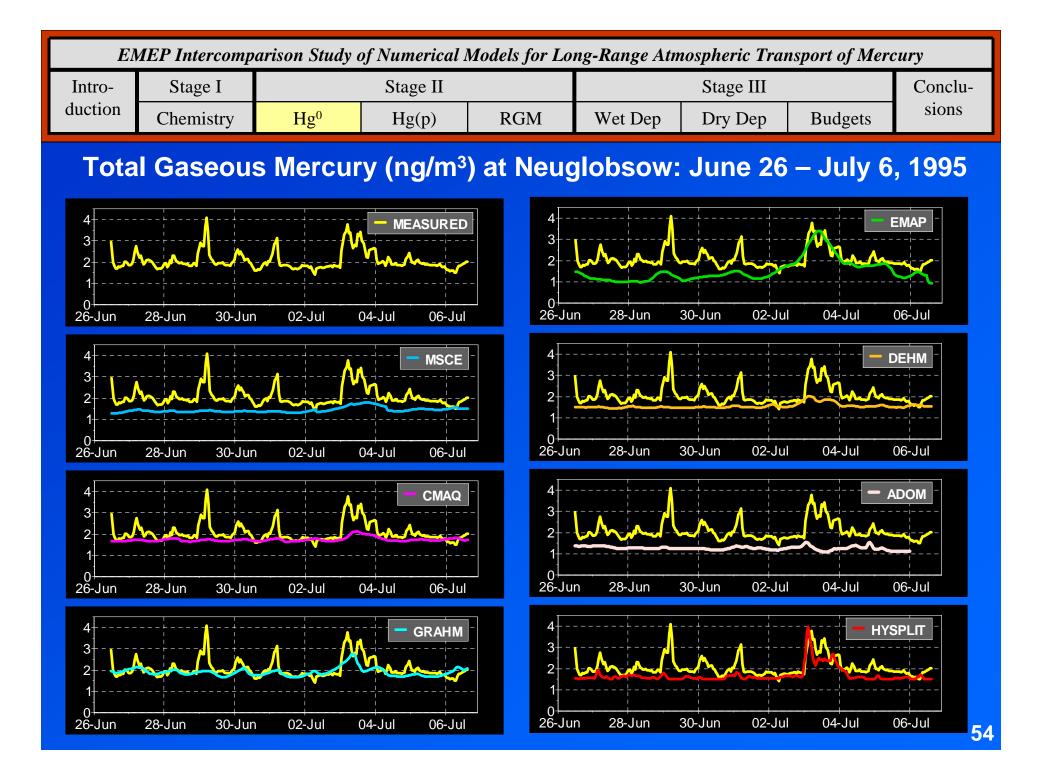
III. Long-Term Deposition and Source-Receptor Budgets

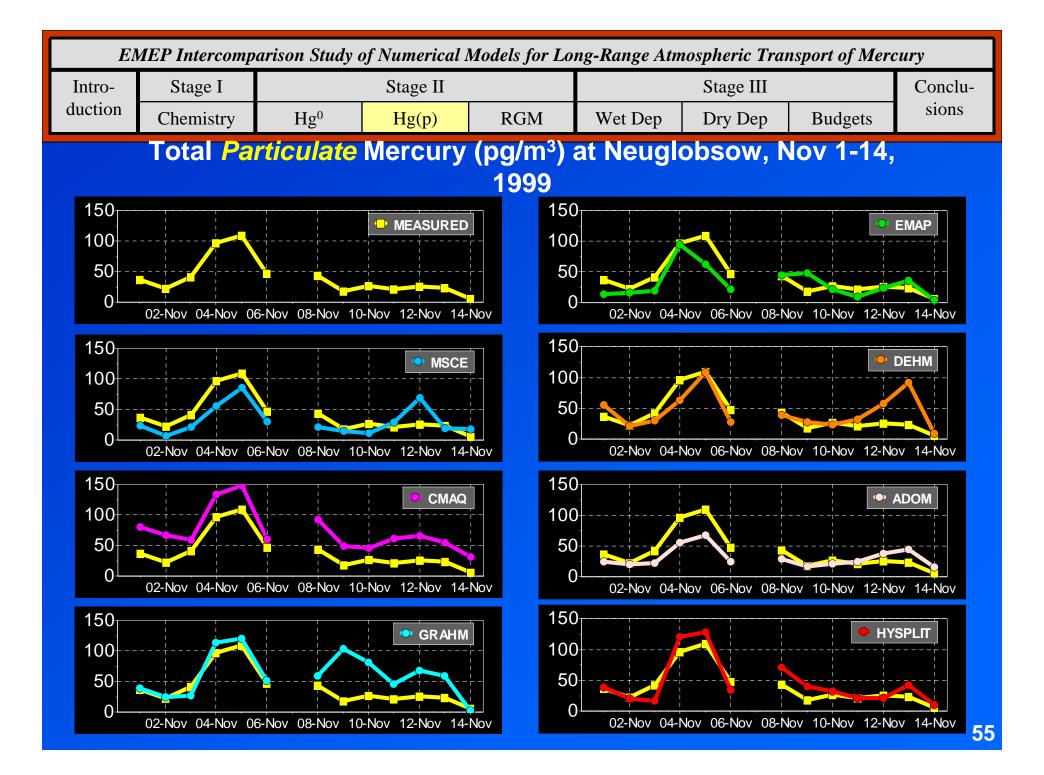
EN	EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury										
Intro-	Stage	I Stage II Stage III								onclu-	
duction	Chemis	try Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budge	ets	S	ions	
		Dar	ticin	ating	Mod						
		Fa	ucip	aung							
Model A	Model Acronym <i>Model Name</i> and Institution							St	age		
							Ι		Π	ш	
	CAM	Chemistry of Atmos. Mercury model, Environmental Institute, Sweden									
	MCM	Mercury Chemist	Mercury Chemistry Model, Atmos. & Environmental Research, USA								
	CMAQ	Community Multi	Community Multi-Scale Air Quality model, US EPA								
	ADOM	Acid Deposition a	Acid Deposition and Oxidants Model, GKSS Research Center, Germany								
Μ	SCE-HM	MSC-E heavy met	MSC-E heavy metal regional model, EMEP MSC-E								
	GRAHM	Global/Regional A	Global/Regional Atmospheric Heavy Metal model, Environment Canada								
	EMAP	Eulerian Model for Air Pollution, Bulgarian Meteo-service									
	DEHM	Danish Eulerian	Danish Eulerian Hemispheric Model, National Environmental Institute								
H	HYSPLIT	Hybrid Single Par	Hybrid Single Particle Lagrangian Integrated Trajectory model, US NOAA								
MSCE-I	HM-Hem	MSC-E heavy met	tal hemispheric	c model, EME	P MSC-E						

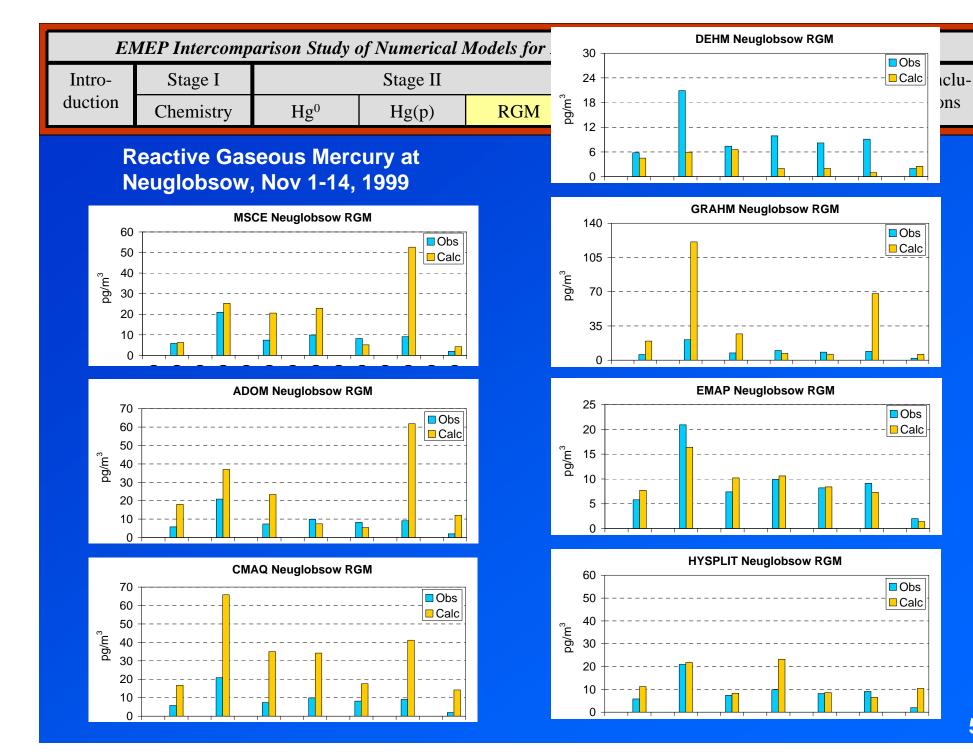








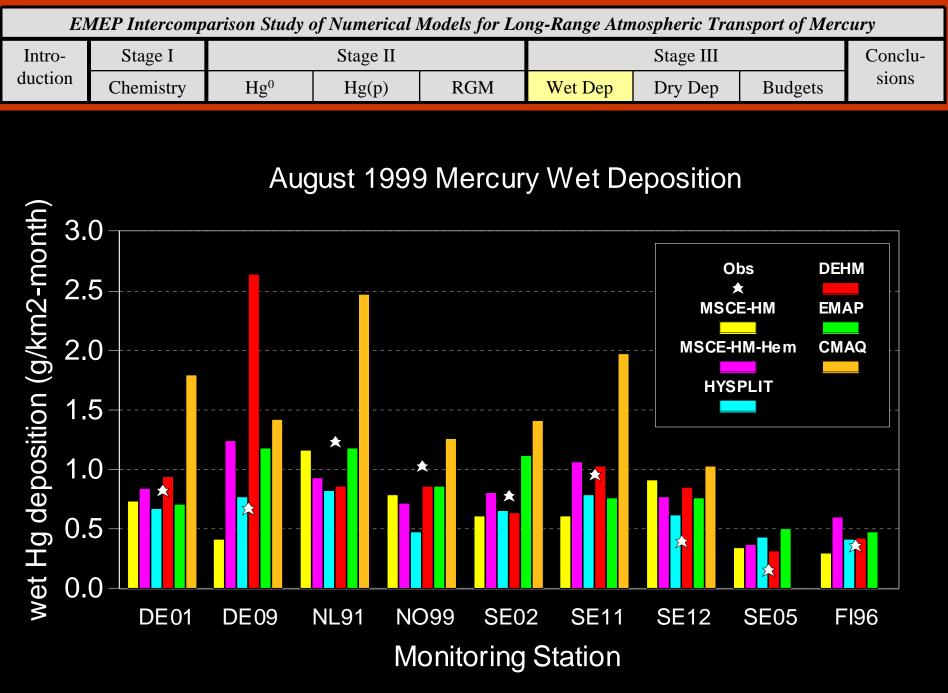




EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury									
Intro- duction	Stage I	Stage II				Conclu-			
	Chemistry	Hg^0	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	sions	

Stage II Publications:

- 2003 Ryaboshapko, A., Artz, R., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Davignon, D., Draxler, R., Ebinghaus, R., Ilyin, I., Munthe, J., Petersen, G., Syrakov, D. Intercomparison Study of Numerical Models for Long Range Atmospheric Transport of Mercury. Stage II. Comparisons of Modeling Results with Observations Obtained During Short Term Measuring Campaigns. Meteorological Synthesizing Centre East, Moscow, Russia.
- 2005 Ryaboshapko, A., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Syrakov, D., Artz, R., Davignon, D., Draxler, R., and Munthe, J. *Intercomparison Study of Atmospheric Mercury Models. Phase II. Comparison of Models with Short-Term Measurements*. Submitted to <u>Atmospheric Environment</u>.



EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury									
Intro-	Stage I	Stage II			Stage III			Conclu-	
duction	Chemistry	Hg^{0}	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	sions	

Stage III Publication:

2005 Ryaboshapko, A., Artz, R., Bullock, R., Christensen, J., Cohen, M., Draxler, R., Ilyin, I., Munthe, J., Pacyna, J., Petersen, G., Syrakov, D., Travnikov, O. *Intercomparison Study of Numerical Models for Long Range Atmospheric Transport of Mercury.* Stage III. Comparison of Modelling Results with Long-Term Observations and Comparison of Calculated Items of Regional Balances. Meteorological Synthesizing Centre – East, Moscow, Russia.

EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury									
Intro-	Stage I	Stage II			Stage III			Conclu-	
duction	Chemistry	Hg^{0}	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	sions	

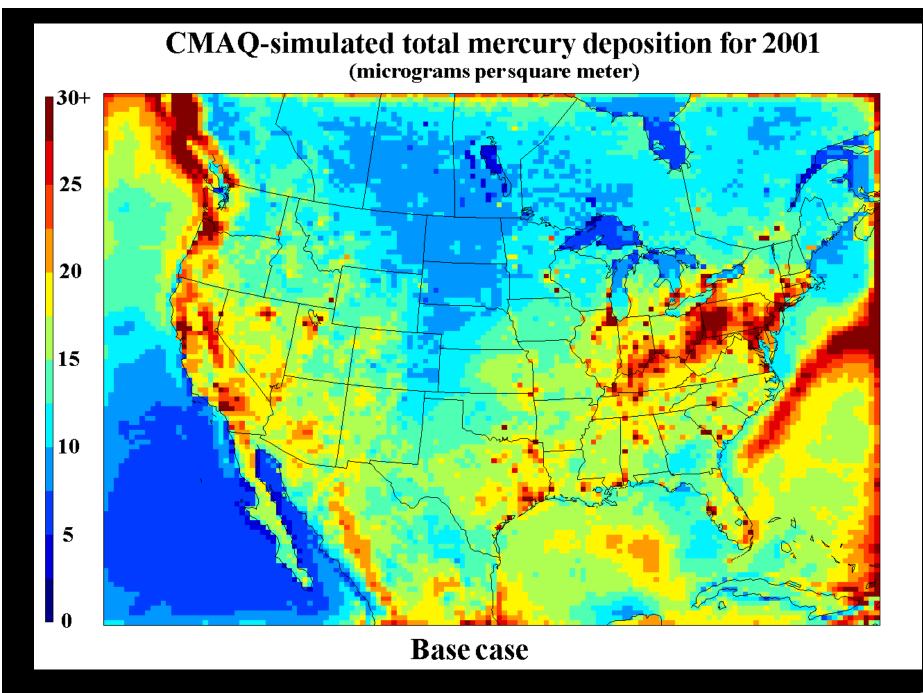
Conclusions: Uncertainties in Mercury Modeling

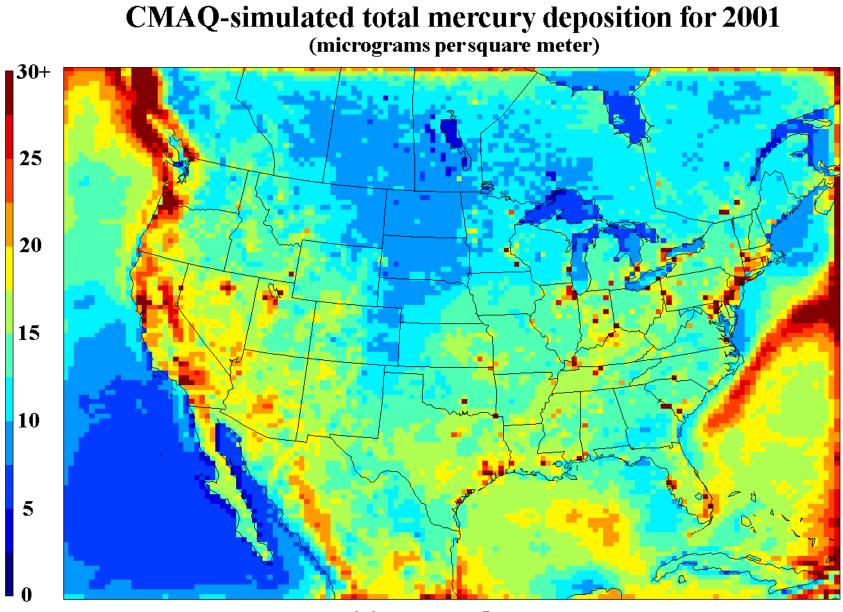
- Elemental Hg in air
- Particulate Hg in air
- Oxidized gaseous Hg in air
- Total Hg in precipitation
- Wet deposition
- Dry deposition
- Balances for countries

- factor of 1.2
- factor of 1.5
- factor of 5
- factor of 1.5
- factor of 2.0
- factor of 2.5
- factor of 2

Models give you a lot of information about why a given concentration or deposition occurs, and gives you information over broad areas, but due to uncertainties – in emissions, meteorology, chemistry, and deposition processes – current models cannot generally give you the exact answer...

Some CMAQ results, used in the development of the CAMR rule, courtesy of Russ Bullock, EPA





Utility Zero Out

Possible underestimation of local and/or regional impacts in CMAQ-Hg modeling done in support of CAMR:

 36 km grid too coarse to capture local impacts – they are artificially diluted

USEPA (2005). Clean Air Mercury Rule (CAMR) Technical Support Document: *Methodology Used to Generate Deposition, Fish Tissue Methylmercury Concentrations, and Exposure for Determining Effectiveness of Utility Emissions Controls:* Analysis of Mercury from Electricity Generating Units, page 4

 inclusion of hydroperoxyl radical (HO2•) chemical reaction reducing RGM back to elemental mercury – most models no longer include this reaction since strong evidence exists that it does not occur in the atmosphere

Gardfeldt, K. and M. Jonnson (2003). Is bimolecular reduction of Hg(II)complexes possible in aqueous systems of environmental importance? *J. Phys. Chem. A*, **107** (22): 4478-4482. **Possible overestimation of global impacts in CMAQ-Hg modeling done in support of CAMR**:

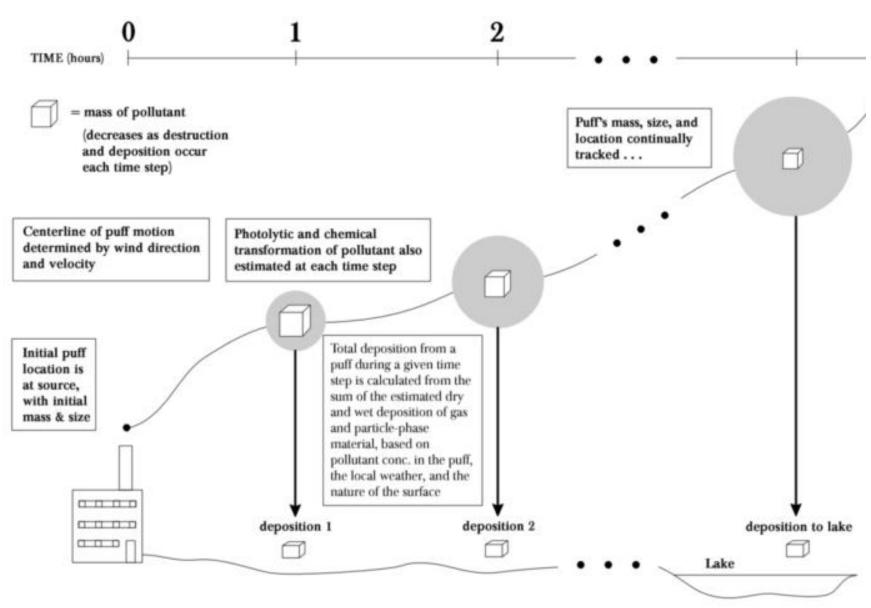
- Strong influence of boundary conditions; appears that RGM may have been specified too high on the boundary; *perhaps* (?) due to an inconsistency in physics/chemistry between global model (GEOS-Chem) providing boundary conditions and that of CMAQ-Hg?
- Two reactions included in CMAQ oxidizing elemental Hg to RGM may have been significantly overestimated (O₃ and OH)

Calvert, J., and S. Lindberg (2005). Mechanisms of mercury removal by O3 and OH in the atmosphere. Atmospheric Environment 39: 3355-3367.

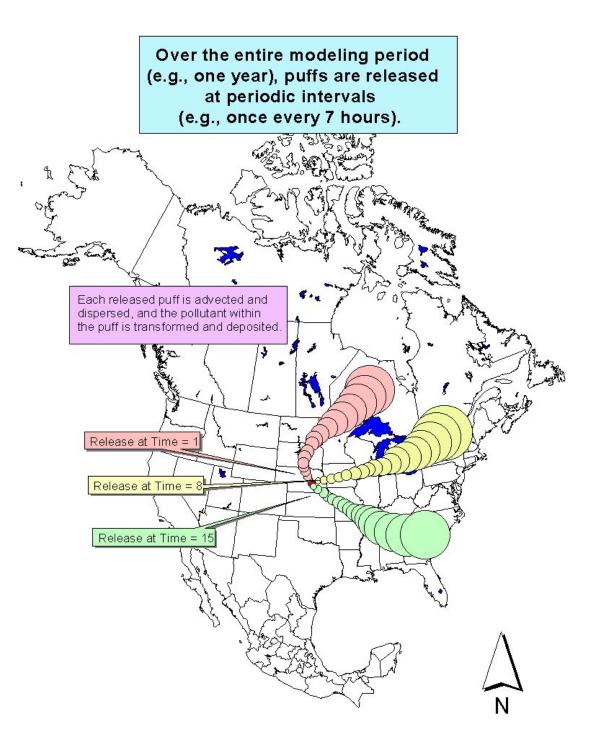
Some HYSPLIT-Hg results, for impacts of U.S. and Canadian anthropogenic sources on selected receptors

NOAA HYSPLIT MODEL

Lagrangian Puff Air Transport and Deposition Model



68





Abstract

A special vension of mercupy in a North Arresults and provide esatmospheric mercury suitable for model eval the Great Lakes region from the Great Lakes significant contribution contribution to atmosp Published by Elsevier

Reports Mercury, At-

Mercury contamis other ecosystems is serious environment human exposure to tion, and significant are believed to be o levels of mercury 2000. Historical o production using the to have caused in

*Supplementary data the online vention, at doi "Corresponding author

E-real address: mark coherol(nona.gov (M. Cohero). ¹Current address: IPPRA Canada/The Institute of Environmental Research, Concord, Ontario, Canada

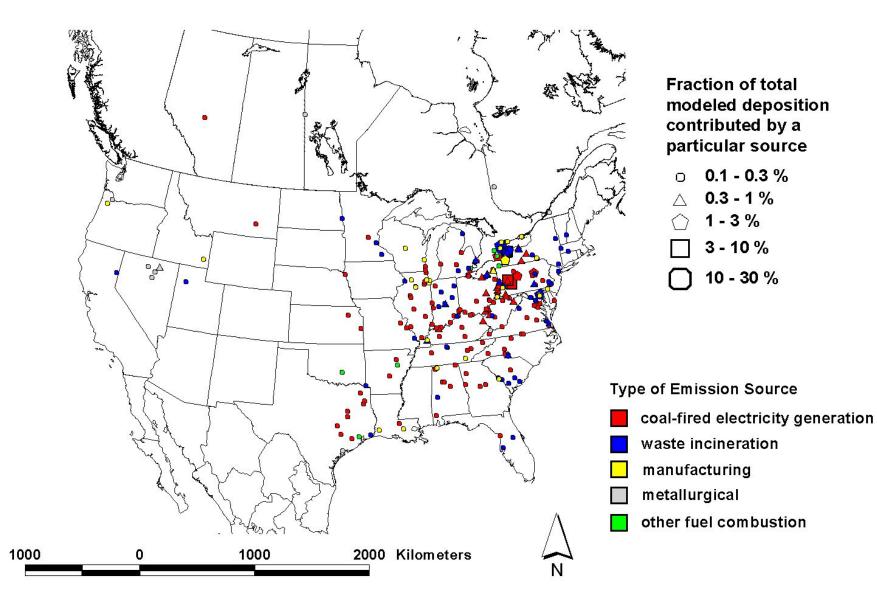
0013-9351/3- a se fr out matter Published by Elsevier Inc. doi:10.1016/j.envres.200311.007

Cohen, M., Artz, R., Draxler, R., Miller, P., Poissant, L., Niemi, D., Ratte, D., Deslauriers, M., Duval, R., Laurin, R., Slotnick, J., Nettesheim, T., McDonald, J. "Modeling the Atmospheric Transport and Deposition of Mercury to the Great Lakes." *Environmental Research* **95**(3), 247-265, 2004.

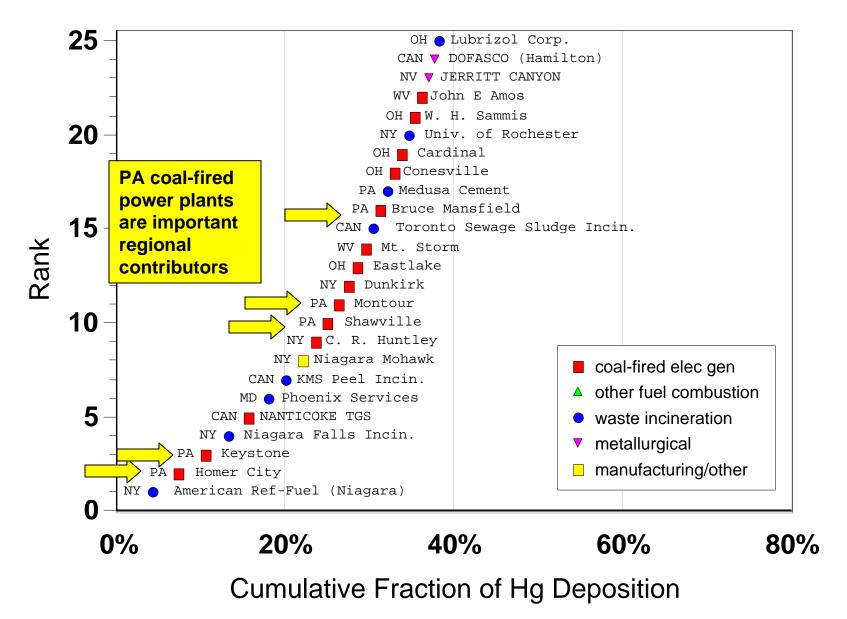
Note: Volume 95(3) is a Special Issue: "An Ecosystem Approach to Health Effects of Mercury in the St. Lawrence Great Lakes", edited by David O. Carpenter.

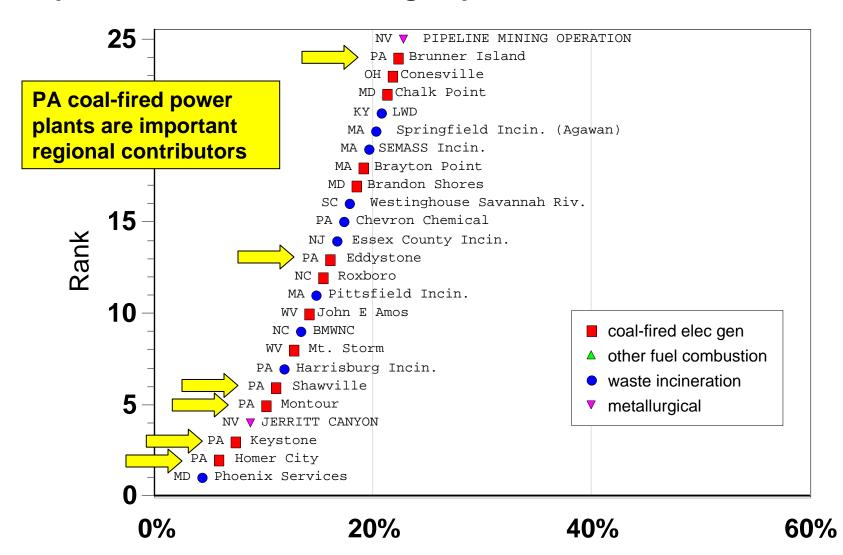
> has developed detailed source-receptor relationships for the Great Lakes, as advocated in Annex 15 of the Great

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



Top 25 Contributors to 1999 Hg Deposition Directly to Lake Ontario





Top 25 Contributors to 1999 Hg Deposition to Acadia National Park

- The HYSPLIT modeling results presented here have only considered the impacts from anthropogenic sources in the United States and Canada
- □ the model is currently being extended to a global domain... but results are not yet available
- However, even if every source in the world was modeled, it is highly likely that these local and regional sources would still be the top contributing sources to local/regional receptors...
- It is unlikely that a coal-fired power plant in China, for example, could contribute as much to one of these receptors as a comparable facility in the U.S.

Concluding Observations

- **Coal-fired power plants emit large amounts of mercury**
- Local and regional impacts depend on a number of factors (e.g., relative proportions of the different forms emitted)
- Challenges in using monitoring approaches to assess impacts
- local/regional impacts are highly episodic and spatially variable
- measurements to date can't unambiguously assess such impacts
- definitive field experiments have not yet been carried out
- Challenges in using modeling analyses to assess impacts
- significant uncertainties in emissions, meteorology, and fate processes
- adequate data for model evaluation and improvement not yet available
- However, limited model evaluations are encouraging and suggest that models are generating reasonable results

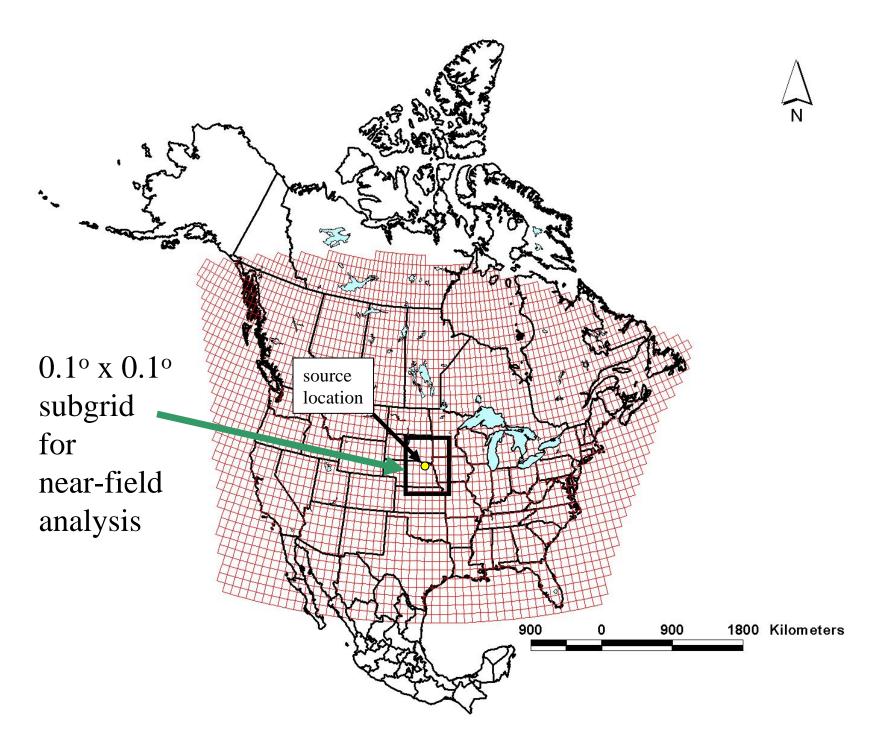


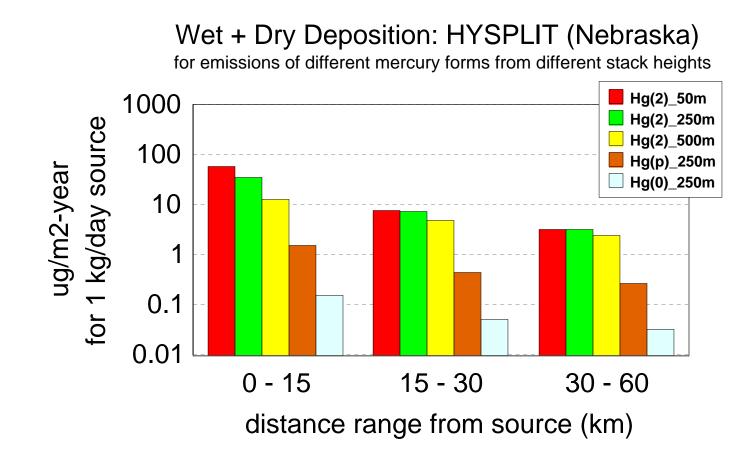
Modeling tends to show significant local/regional impacts

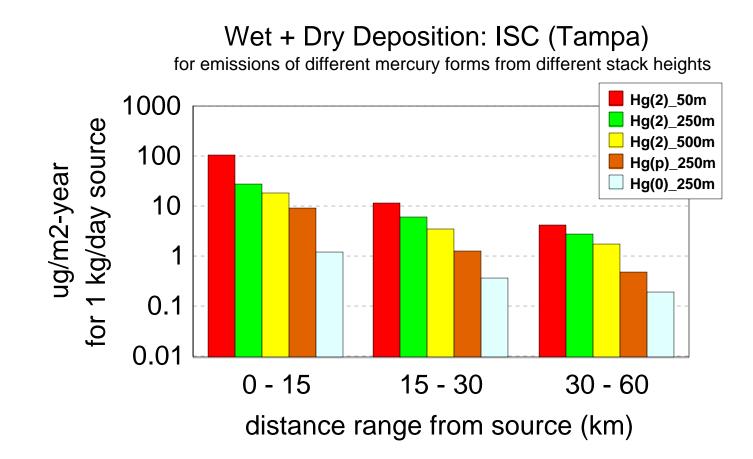
Emissions trading will result in winners and losers...

Thanks!

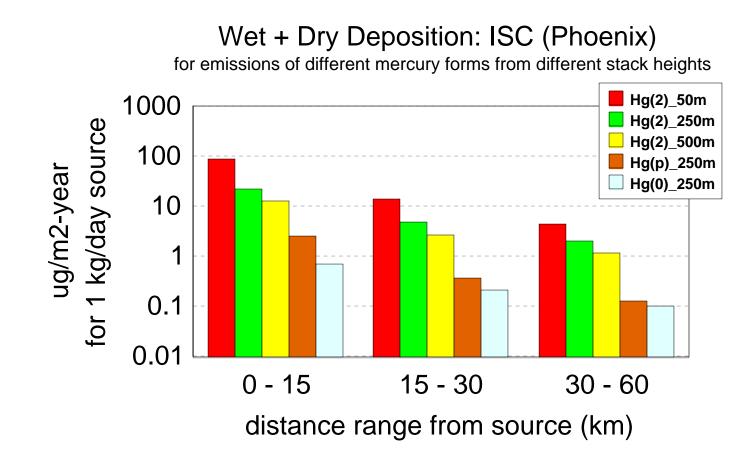
Extra Slides



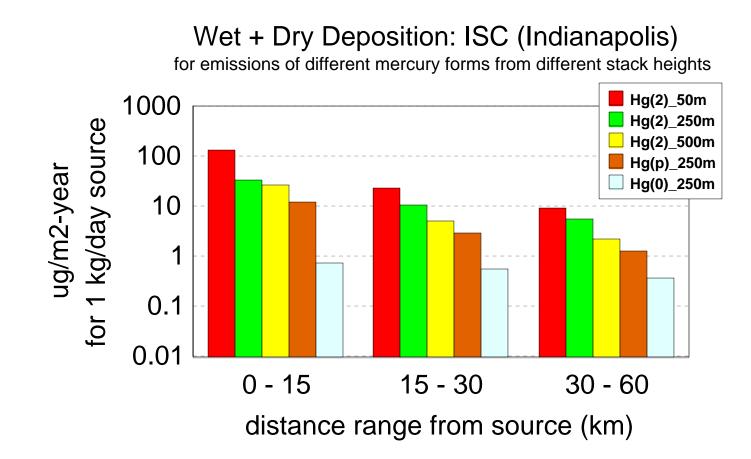




Calculated from data used to produce Appendix A of USEPA (2005): Clean Air Mercury Rule (CAMR) Technical Support Document: Methodology Used to Generate Deposition, Fish Tissue Methylmercury Concentrations, and Exposure for Determining Effectiveness of Utility Emissions Controls: Analysis of Mercury from Electricity Generating Units

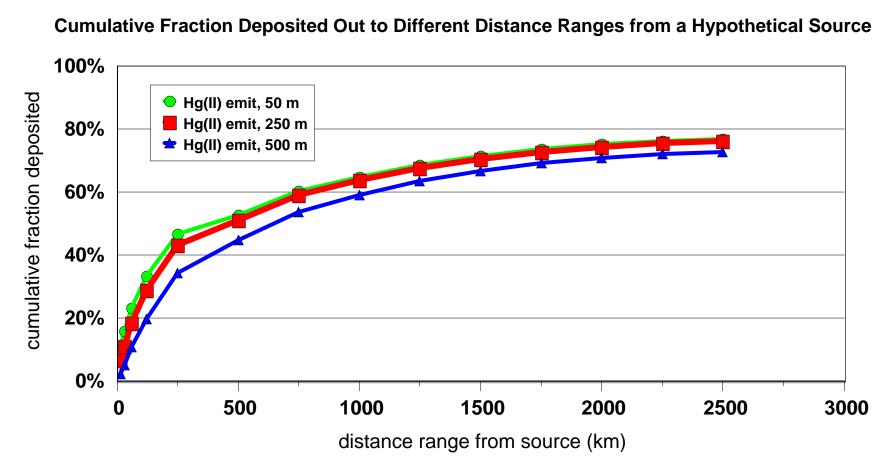


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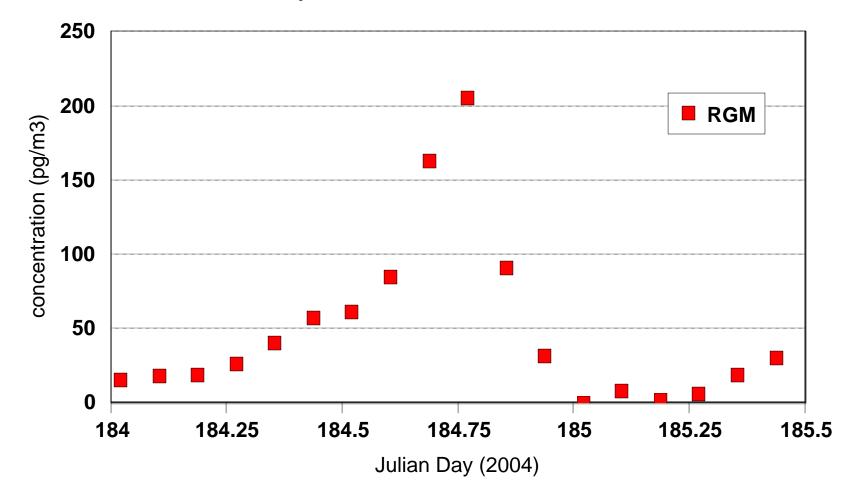


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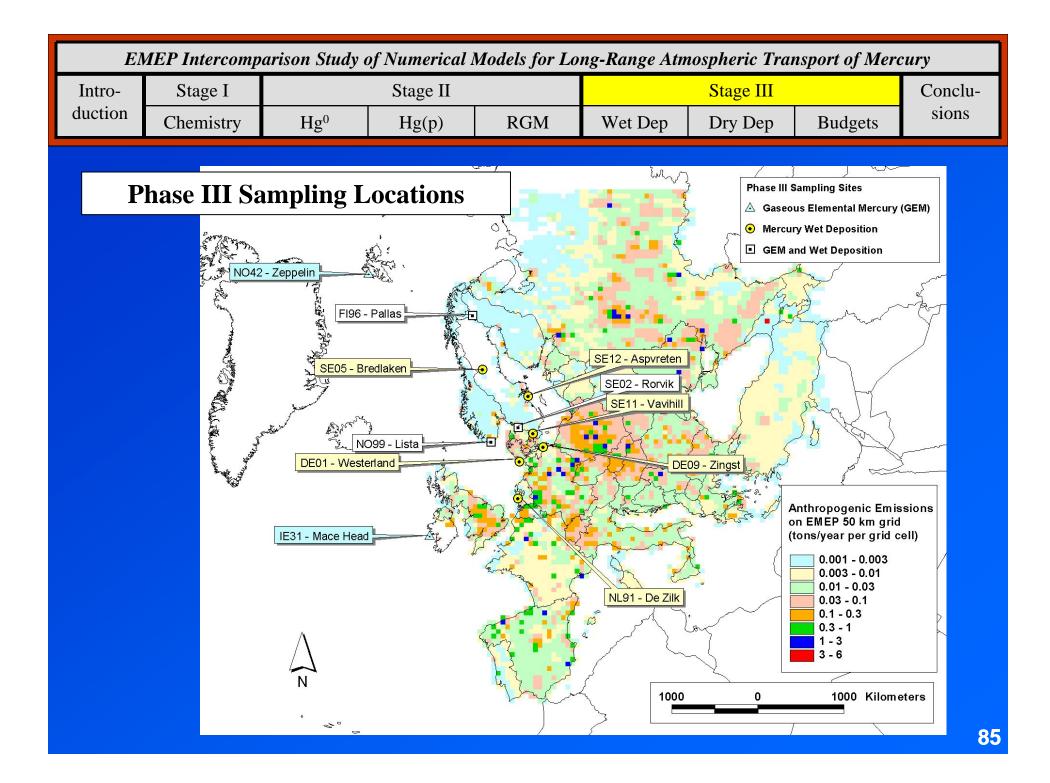
ionic Hg emitted from different source heights

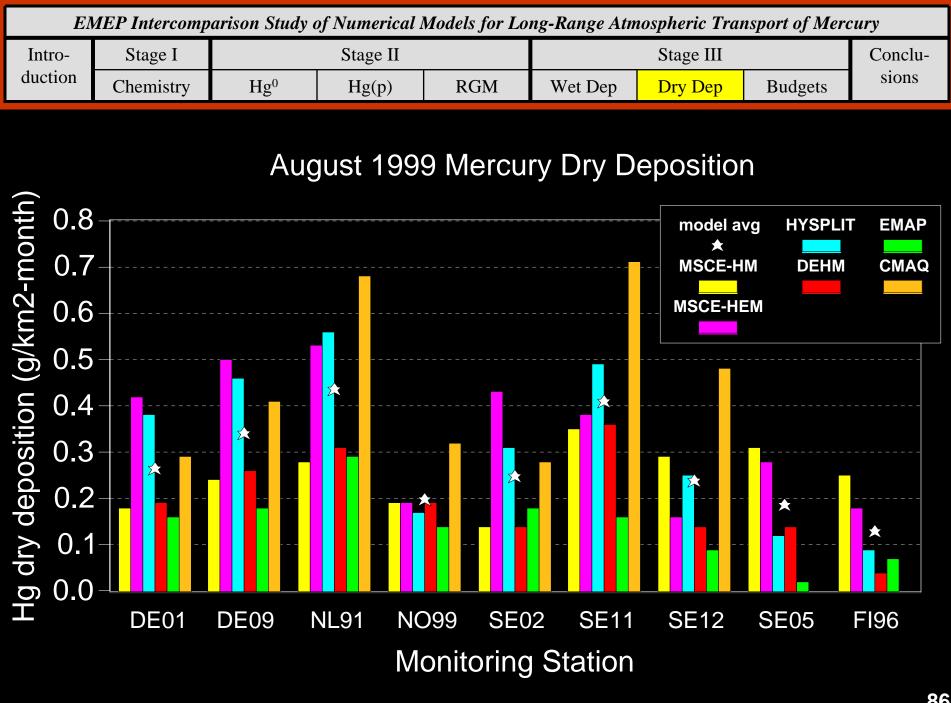


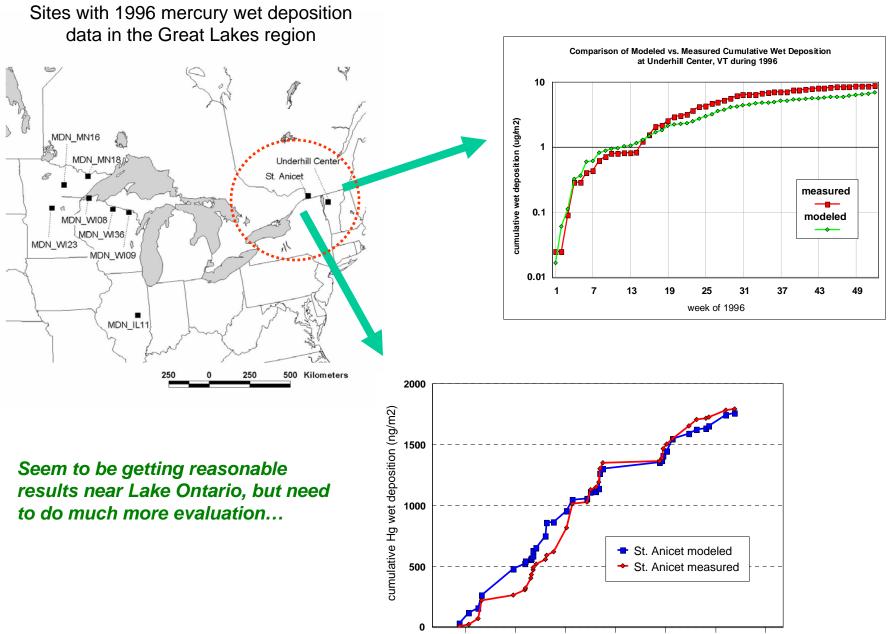
Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data



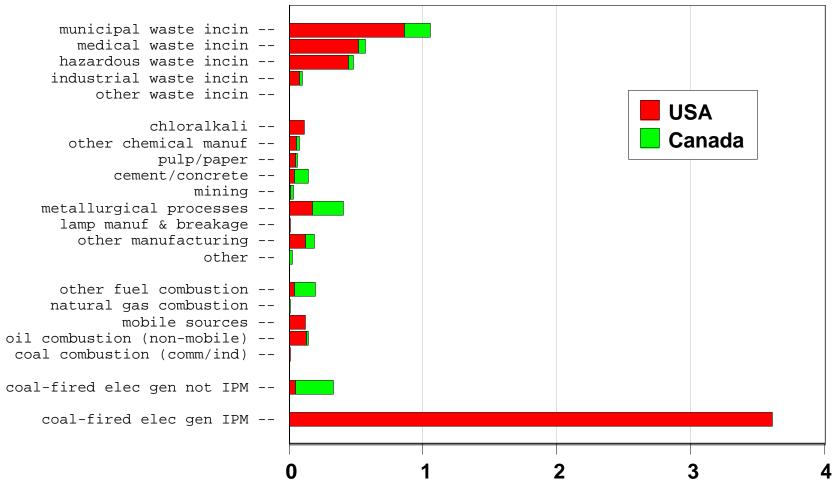
Measured Atmospheric Concentrations at Oxford MD, Summer 2004



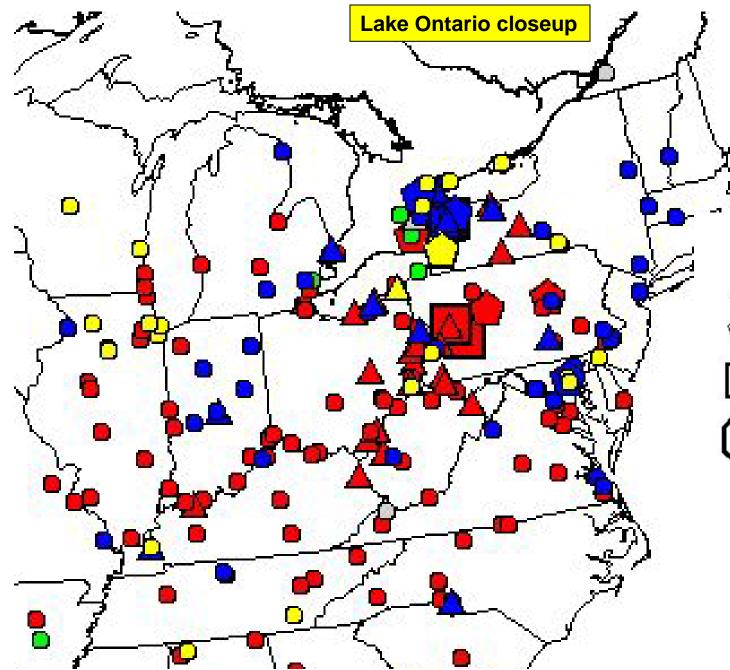




05-Feb-96 05-Apr-96 04-Jun-96 03-Aug-96 02-Oct-96 01-Dec-96 30-Jan-97



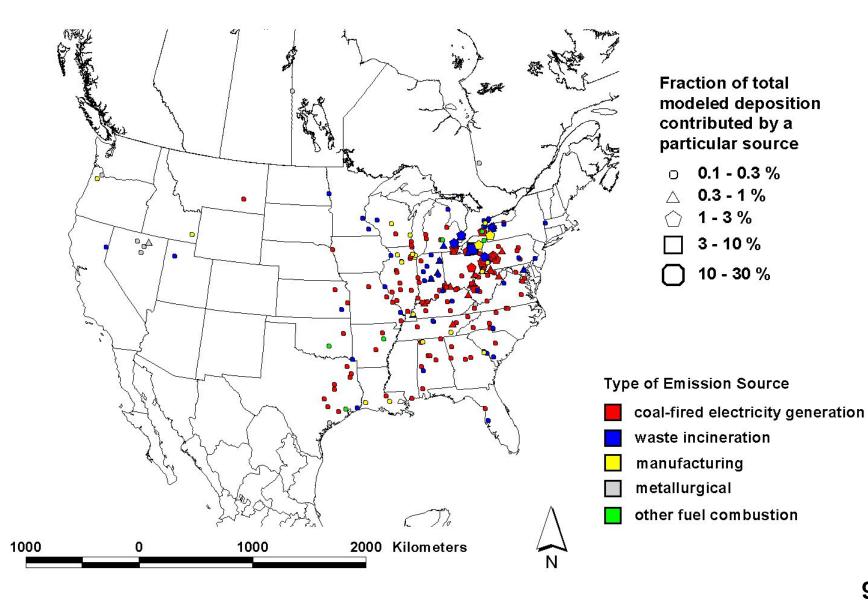
Total Atmos. Dep Flux to Lake Ontario (g Hg/km2-year)



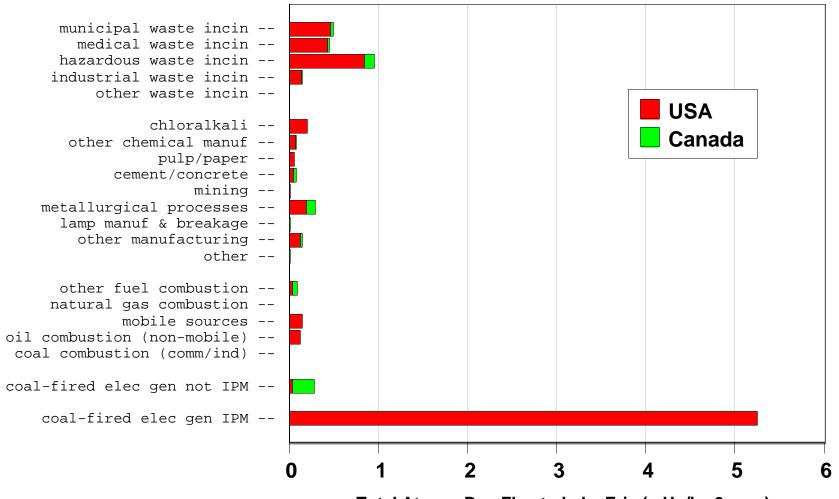
Fraction of total Modeled deposition Contributed by a Particular source

○ 0.1 - 0.3 %
△ 0.3 - 1 %
○ 1 - 3 %
□ 3 - 10 %
○ 10 - 30 %

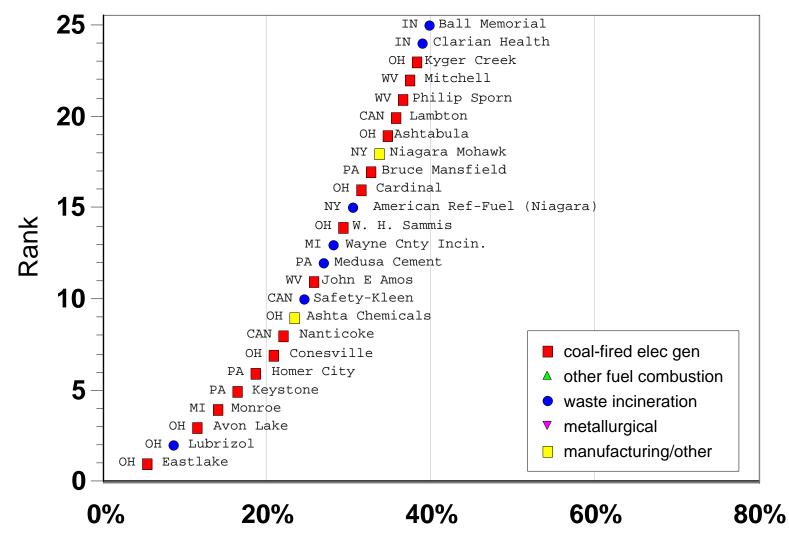
Type of Emission Source coal-fired electricity generation waste incineration manufacturing metallurgical other fuel combustion Largest atmospheric deposition contributors to Lake Erie based on 1999-2000 emissions



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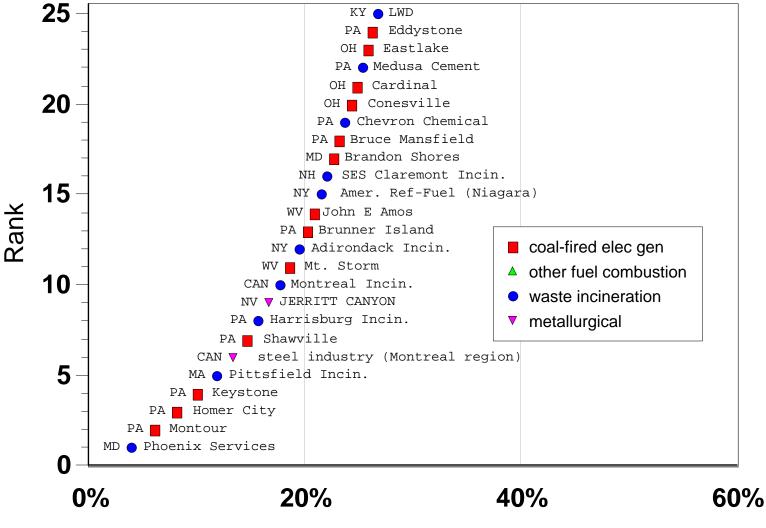


Total Atmos. Dep Flux to Lake Erie (g Hg/km2-year)



Top 25 Contributors to 1999 Hg Deposition Directly to Lake Erie

Top 25 Contributors to 1999 Hg Deposition Directly to Lake Champlain



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

