Atmospheric Fate and Transport of Mercury

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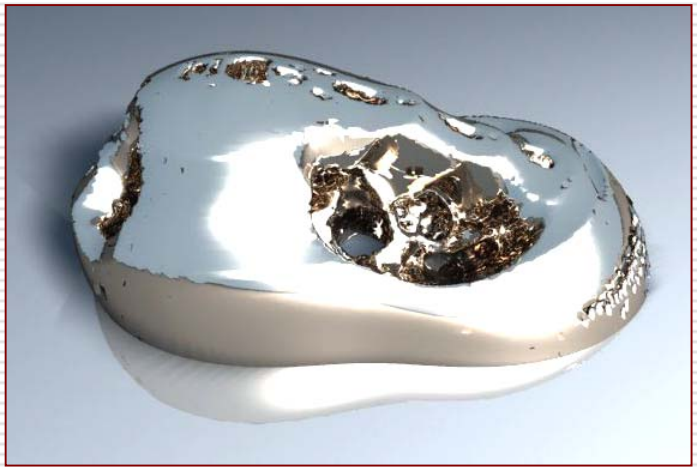
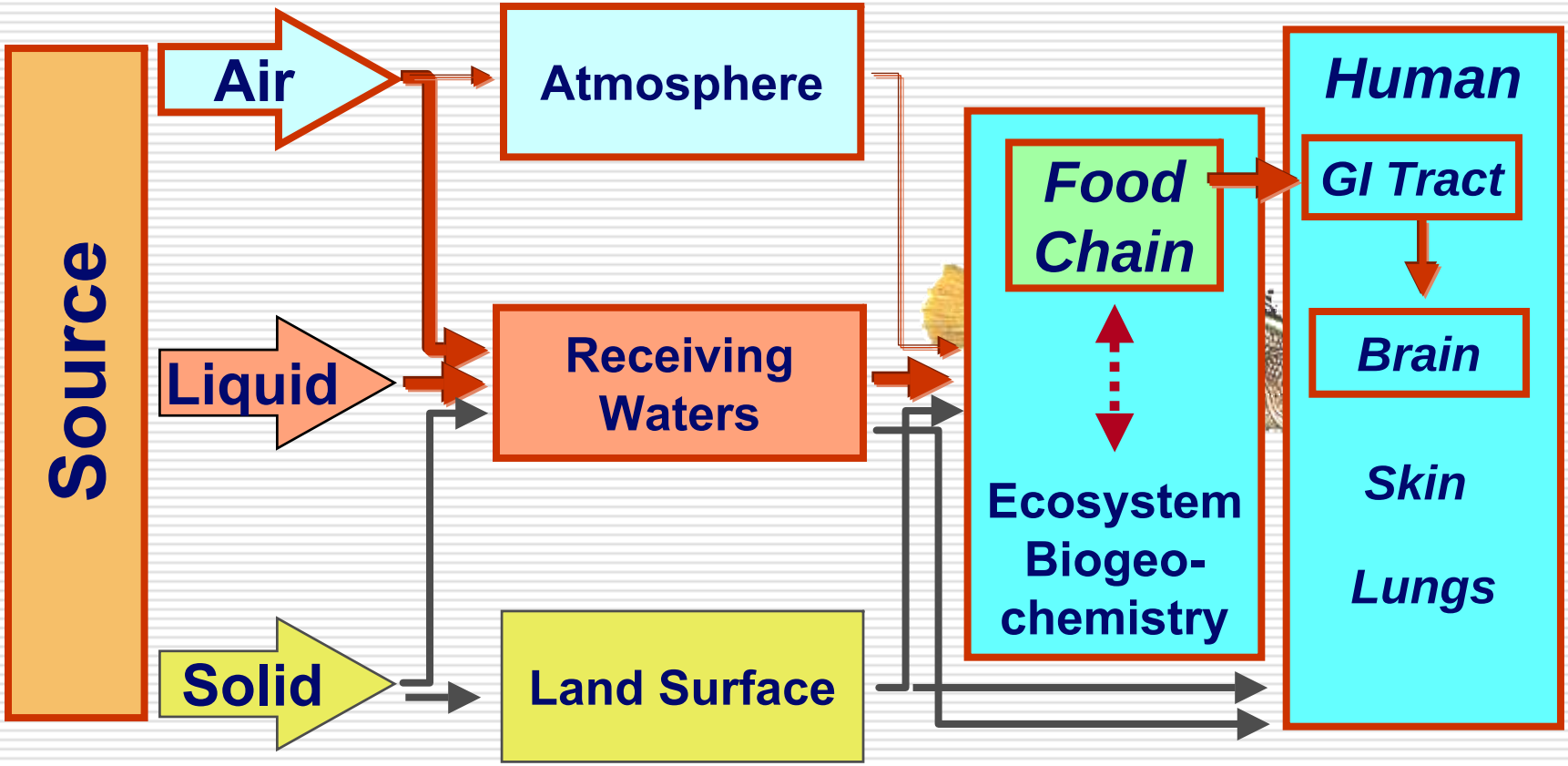
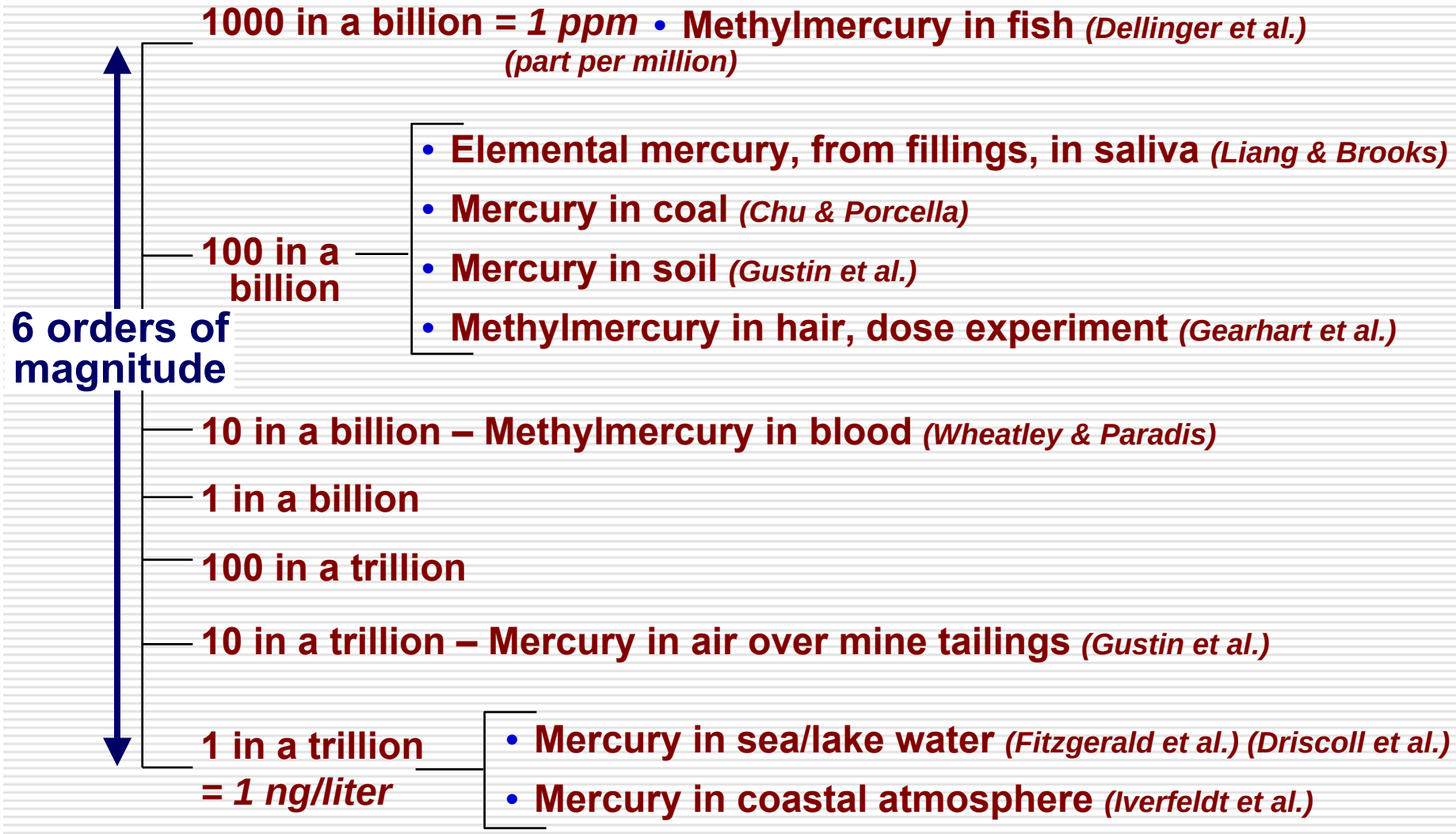
Harrisburg, Pennsylvania
October 14, 2005



Mercury Exposure Pathways in Humans

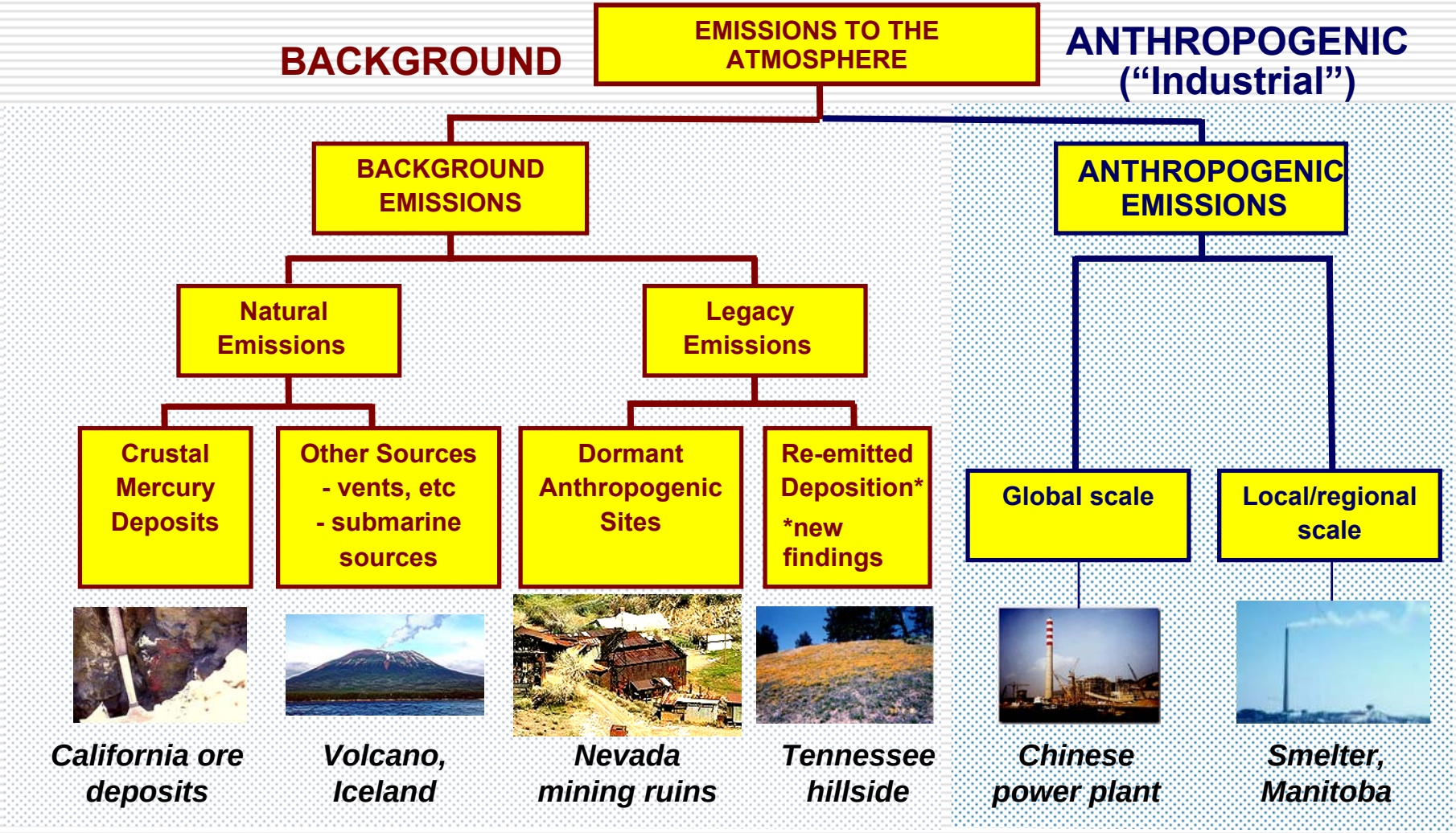


Mercury concentrations are all very small, but still vary over a million-fold range

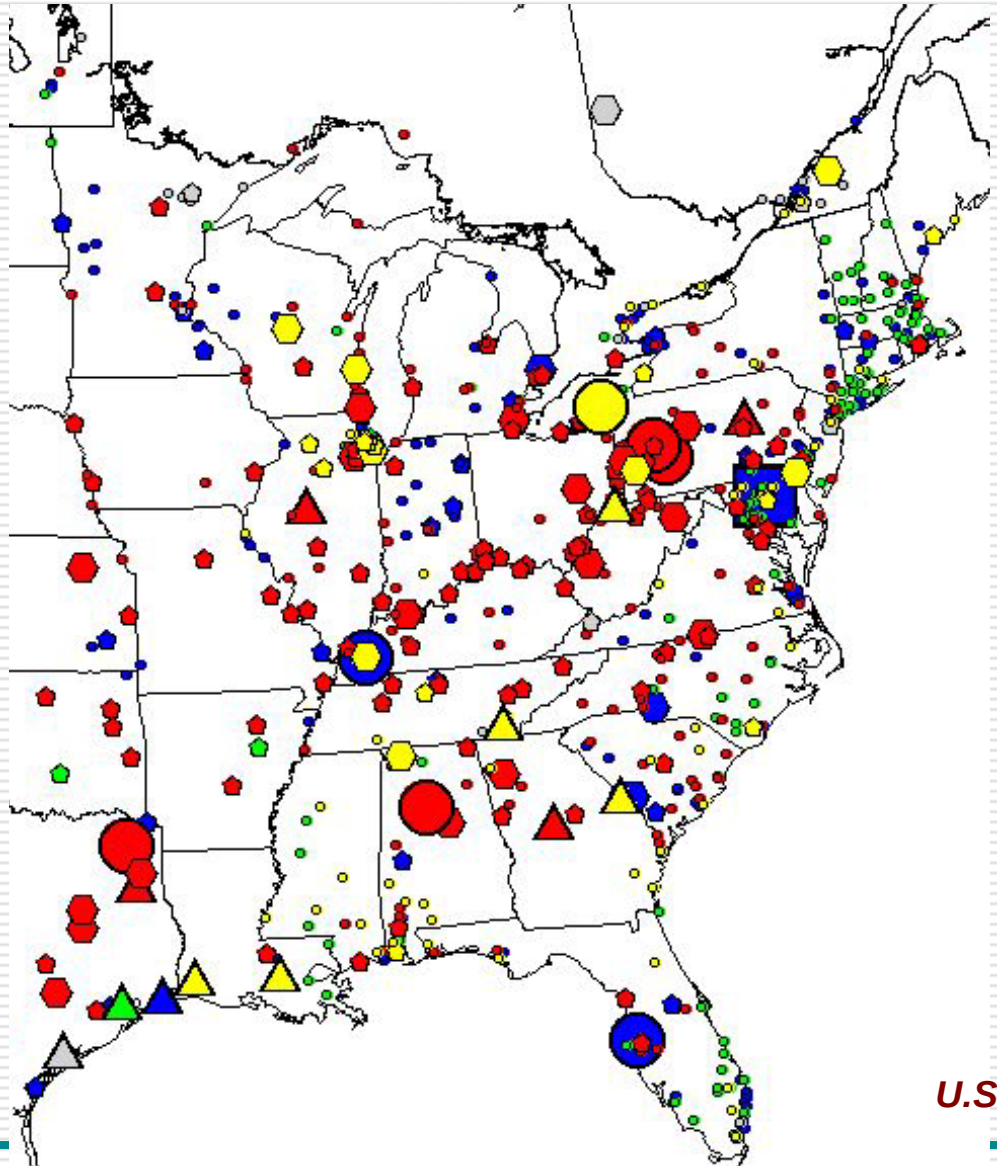


MERCURY AS A GLOBAL POLLUTANT

Mercury Is a Chemical Element with Many Sources Natural & Manmade; U.S. & Global



Mercury Sources, eastern U.S. (1999)



Total Hg emissions (kg/year)

- 0 - 100
- 100 - 300
- 300 - 500
- 500 - 700
- 700 - 1000
- 1000 - 1300
- 1300 - 7000

- Coal-Fired Electricity Generation
- Waste Incineration (pre-MACT)
- Metallurgical
- Manufacturing
- Other Fuel Combustion

U.S.: 1999; Courtesy Mark Cohen, NOAA

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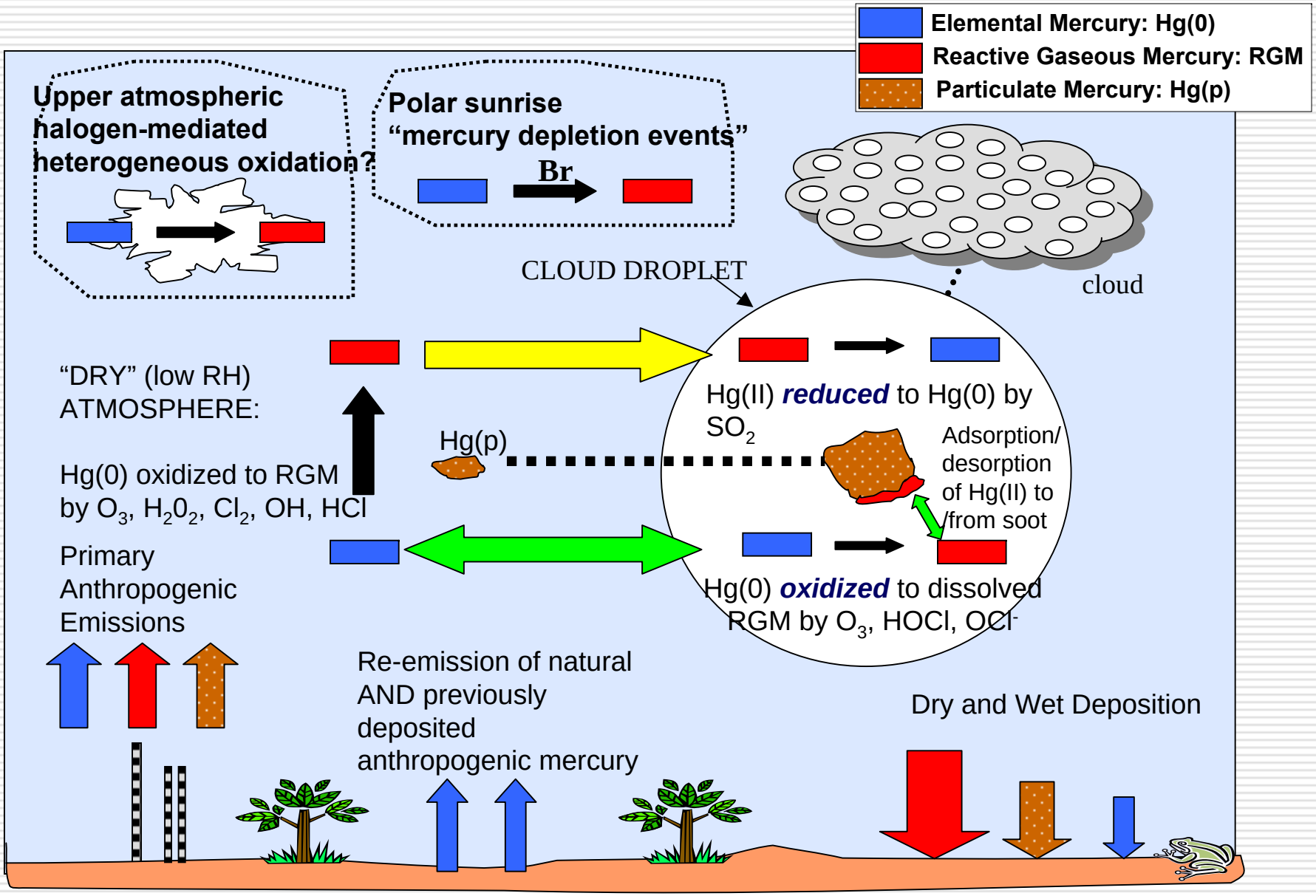
Basic Understanding: Forms of Mercury

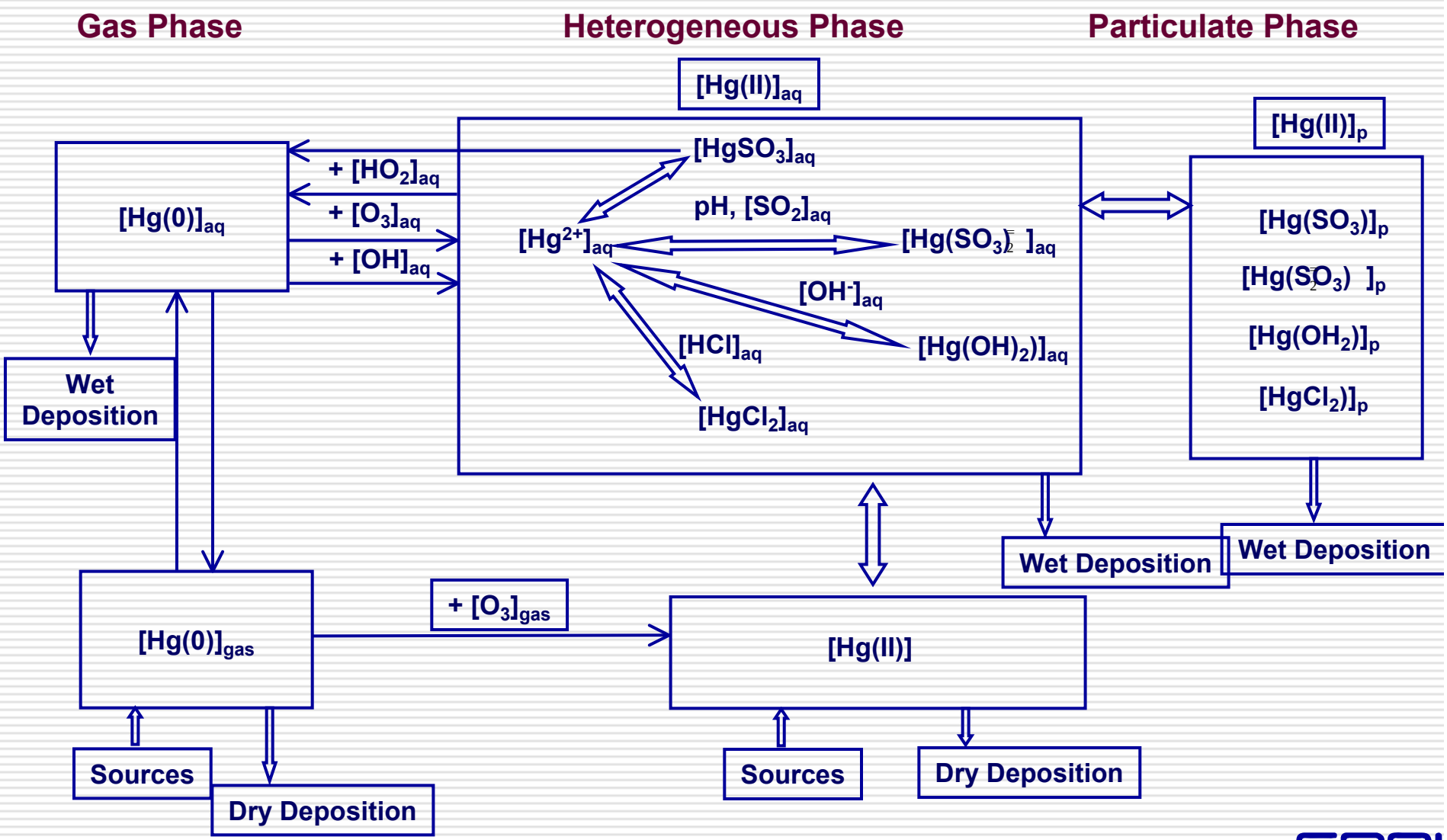
- **Inorganic mercury – 2 types or "species"**
  - **Elemental: Hg(0), silvery liquid metal; form that re-emits**
  - **Divalent: Hg(II)=ionic=oxidized=RGM; combined form in environment; 1,000,000x as water-soluble as Hg(0)**
  - **Particle-bound mercury**
    - **Extremely small fraction of emitted: 1-3% at most**
    - **NOT "particles of mercury" (aerosols of mercury) ➔ rather, Hg(0)+Hg(II) bound to solid particulates**
- **Organic mercury**
  - **Monomethylmercury: $(CH_3)Hg^+$ = MeHg; formed in aquatic systems; may wind up in fish**
  - **Dimethylmercury: $(CH_3)_2Hg$, highly toxic; reactive; rare occurrences: landfills; marine mammals?**

Atmospheric Processes for Mercury



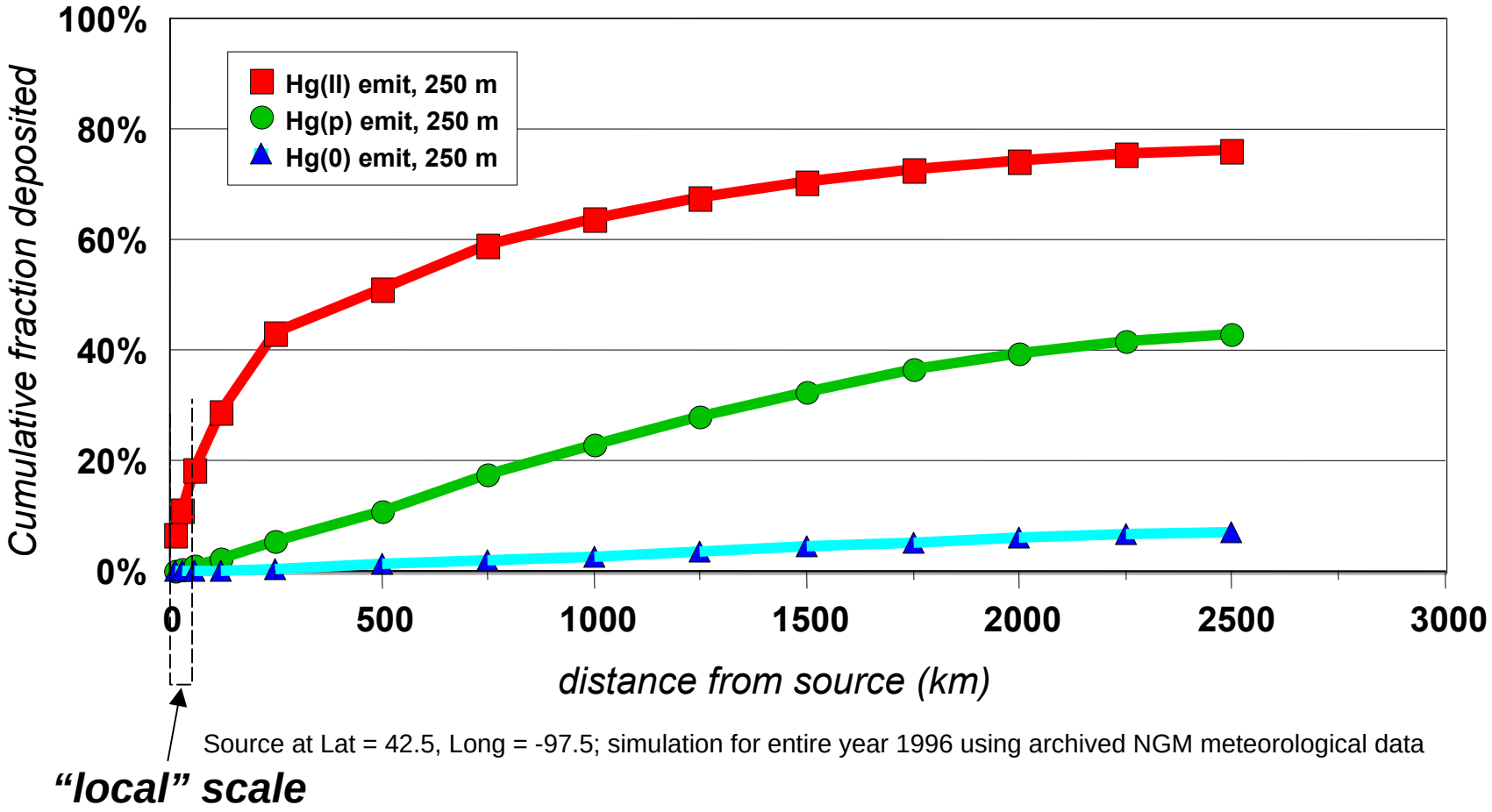
Courtesy Mark Cohen, NOAA

The Chemistry of Mercury in the Atmosphere is Complex, Still Being Understood



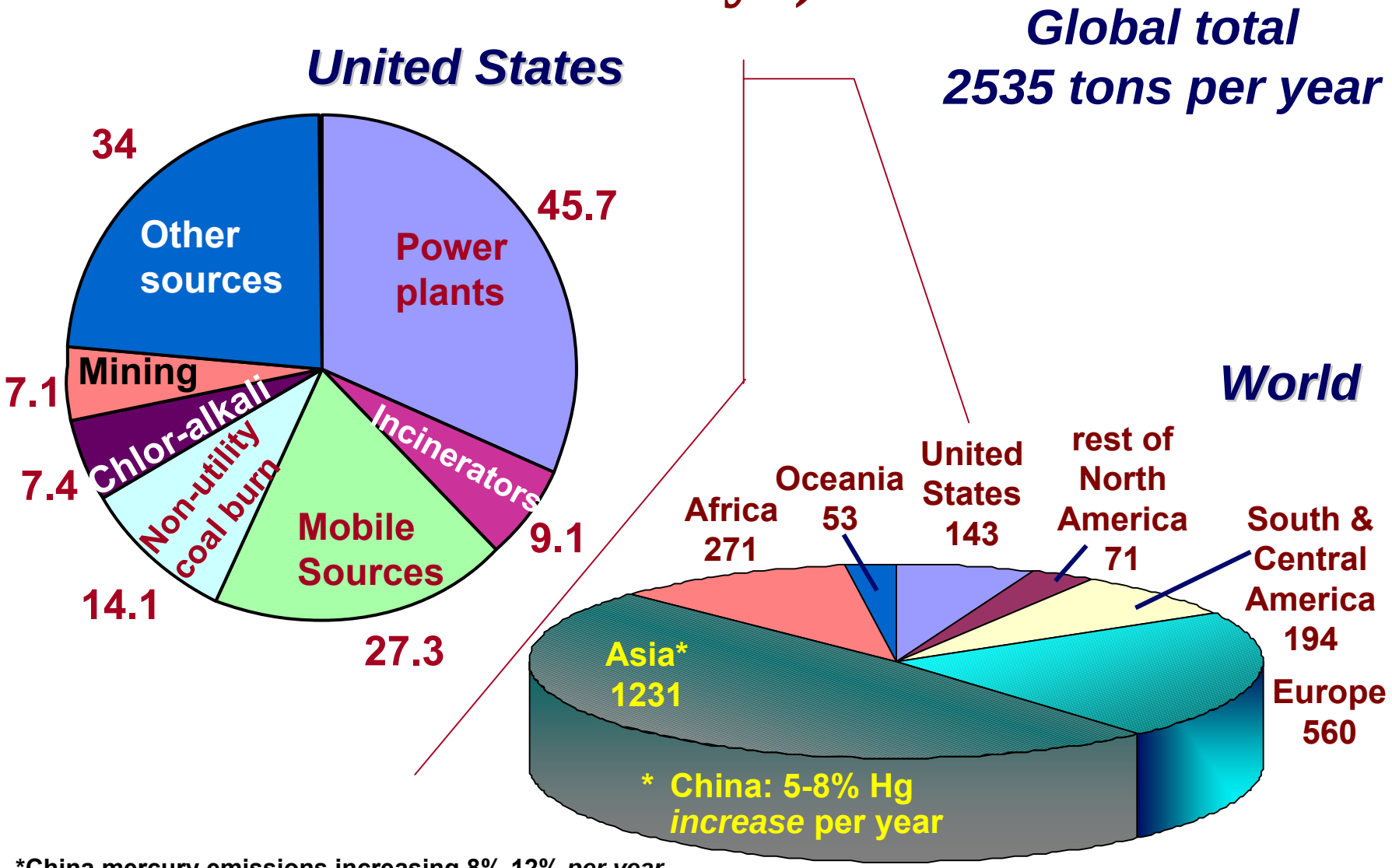
Deposition of inorganic forms of mercury vs. distance from source

Cumulative fraction deposited out to different distance ranges from a hypothetical source



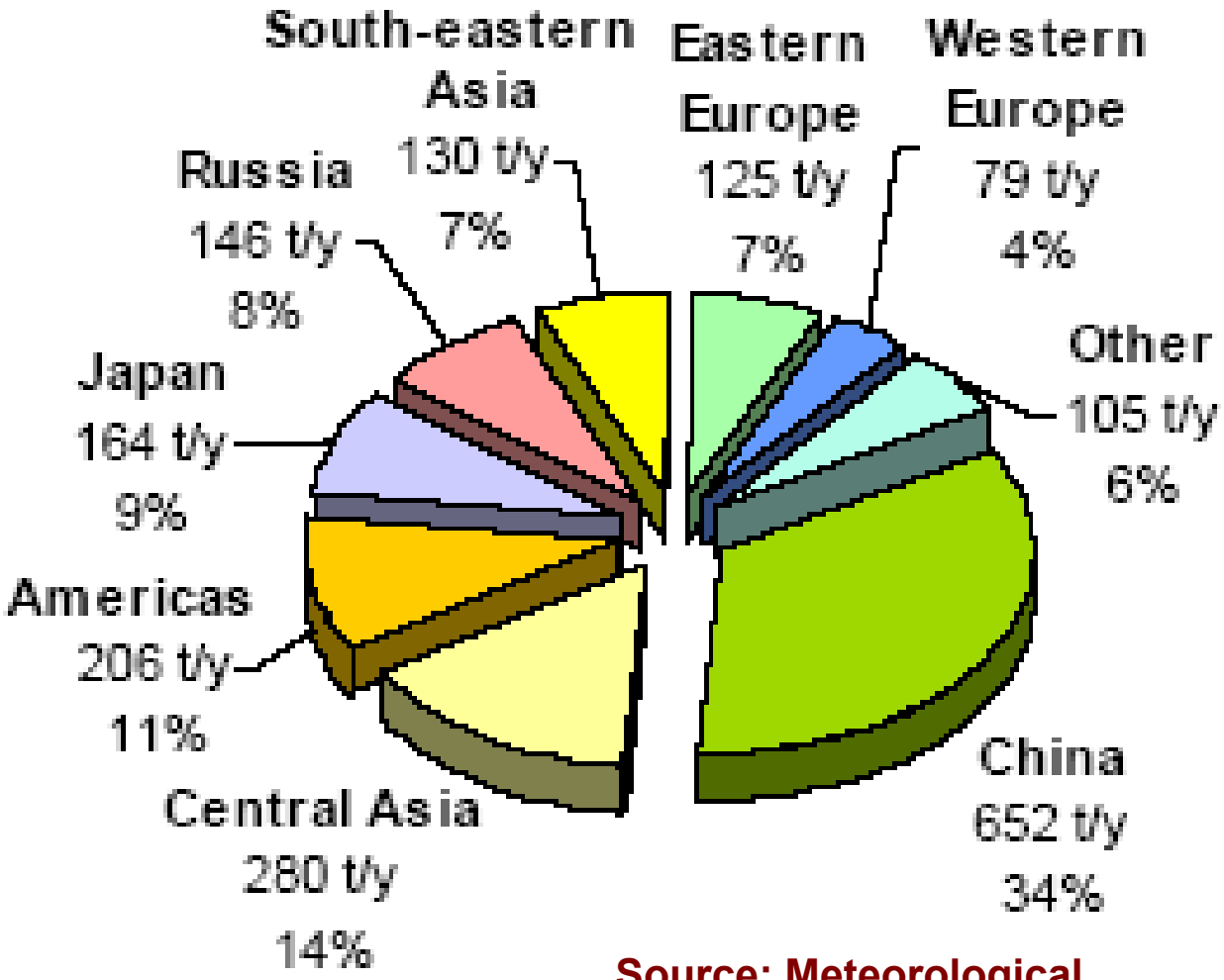
Courtesy Mark Cohen, NOAA

Anthropogenic mercury emissions (U.S. tons/yr)



*China mercury emissions increasing 8%-12% *per year*

Another Estimate: Country/Region Anthropogenic Emissions, Northern Hemisphere



Source: Meteorological Synthesizing Center-East, Moscow

Some Recent Global Mercury Balances

EMISSIONS (Mg/yr)	Bergan et al. (1999)	Mason and Sheu (2002)	Lamborg et al. (2002)	Seigneur et al. (2003) (range)
New Anthropogenic	2160	2400	-	2143
Re-emitted Anthropogenic	2000	2090	4800	2134 (1067-2670)
Natural from Land	500	810	1000	1180 (1805-878)
Natural from Oceans	1400	1300	600	954 (720-1396)
Total [re-emitted %]	6060 [50]	6600 [47]	6400	6411 [50(33-56)]
Ratio of Current/Pre-industrial	3	3.1	4	3

Seigneur et al., 2004, *Environ. Sci. Technol. 38*, 555-569

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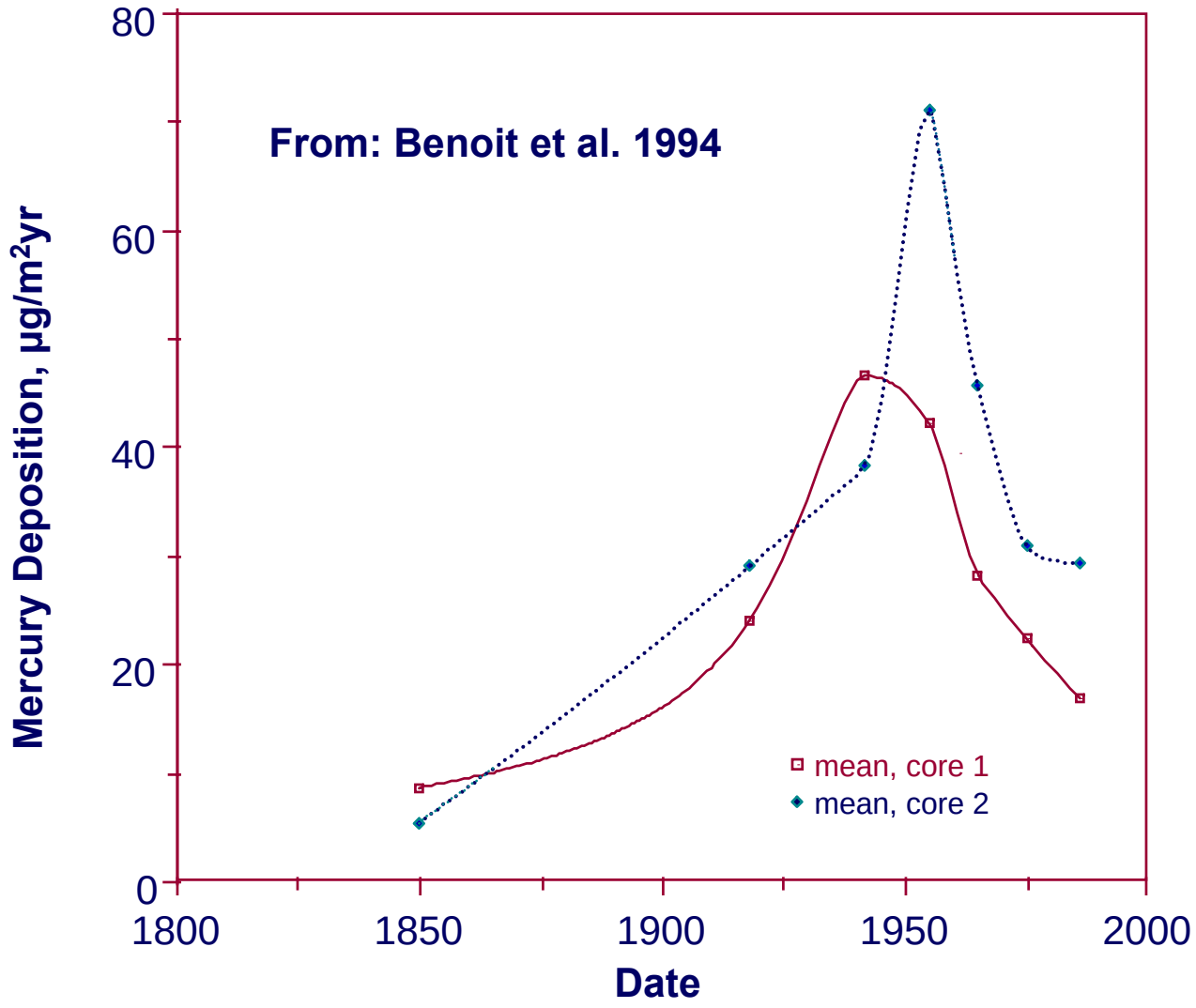
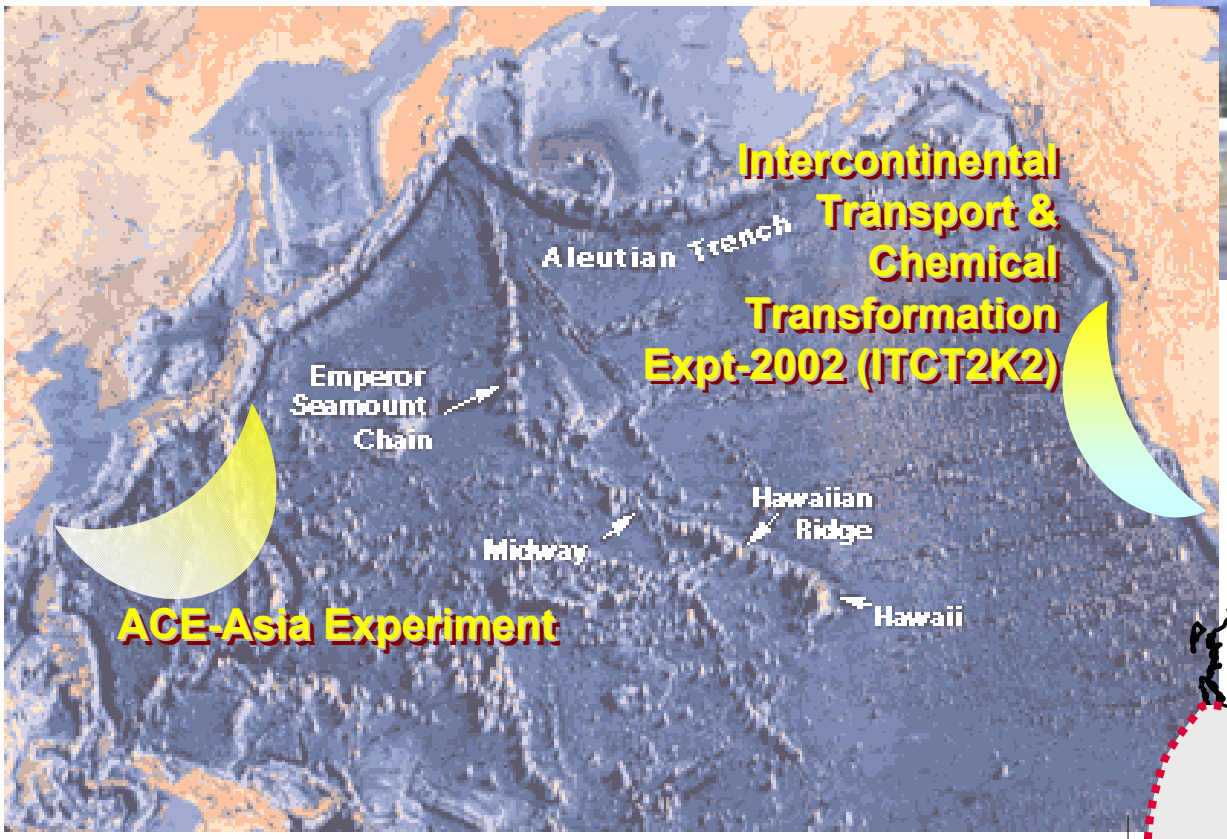
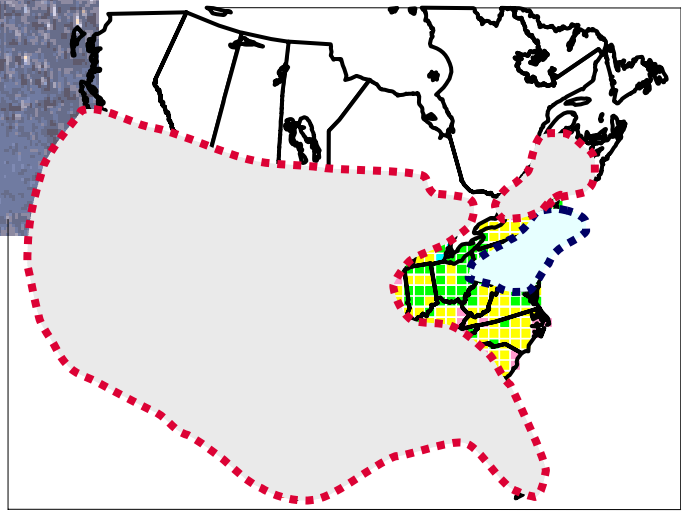
Trends in Mercury Deposition, 2 Northern Wisconsin Bogs

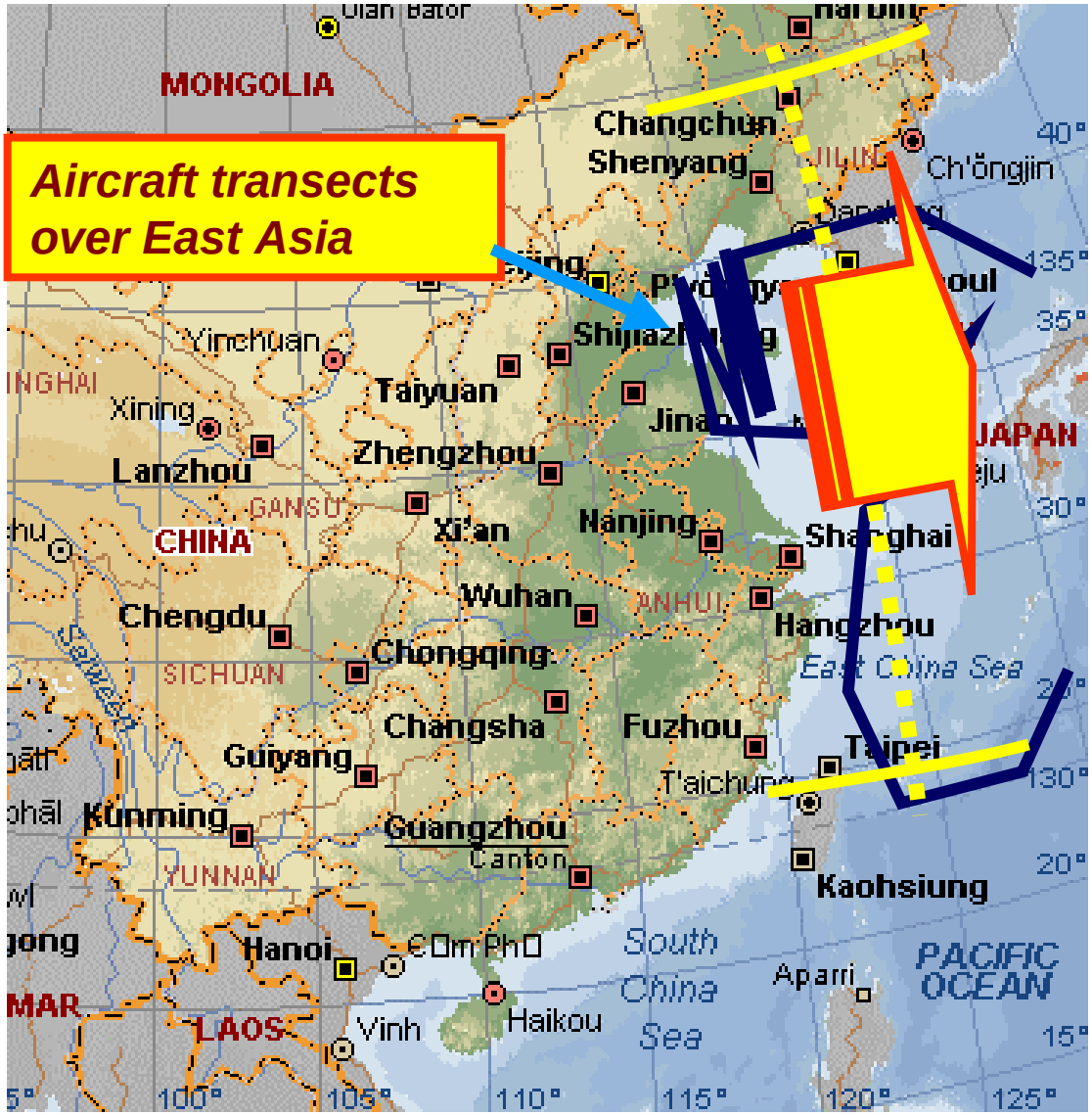


Mercury Input from Other Countries
EPRI Pacific Basin Mercury Experiments, 2001-2





Data Verification of Model Results
Mercury Emissions From Asia Toward The U.S.



- EPRI aircraft measurements, joint U.S./China/ Japan/S. Korea experiment
- 660 tons mercury per year toward the U.S.
- +1 year: plumes into California from China, Asia
- Later studies: Jaffe+Prestbo, Okinawa+Oregon, 2004: >700 t/y

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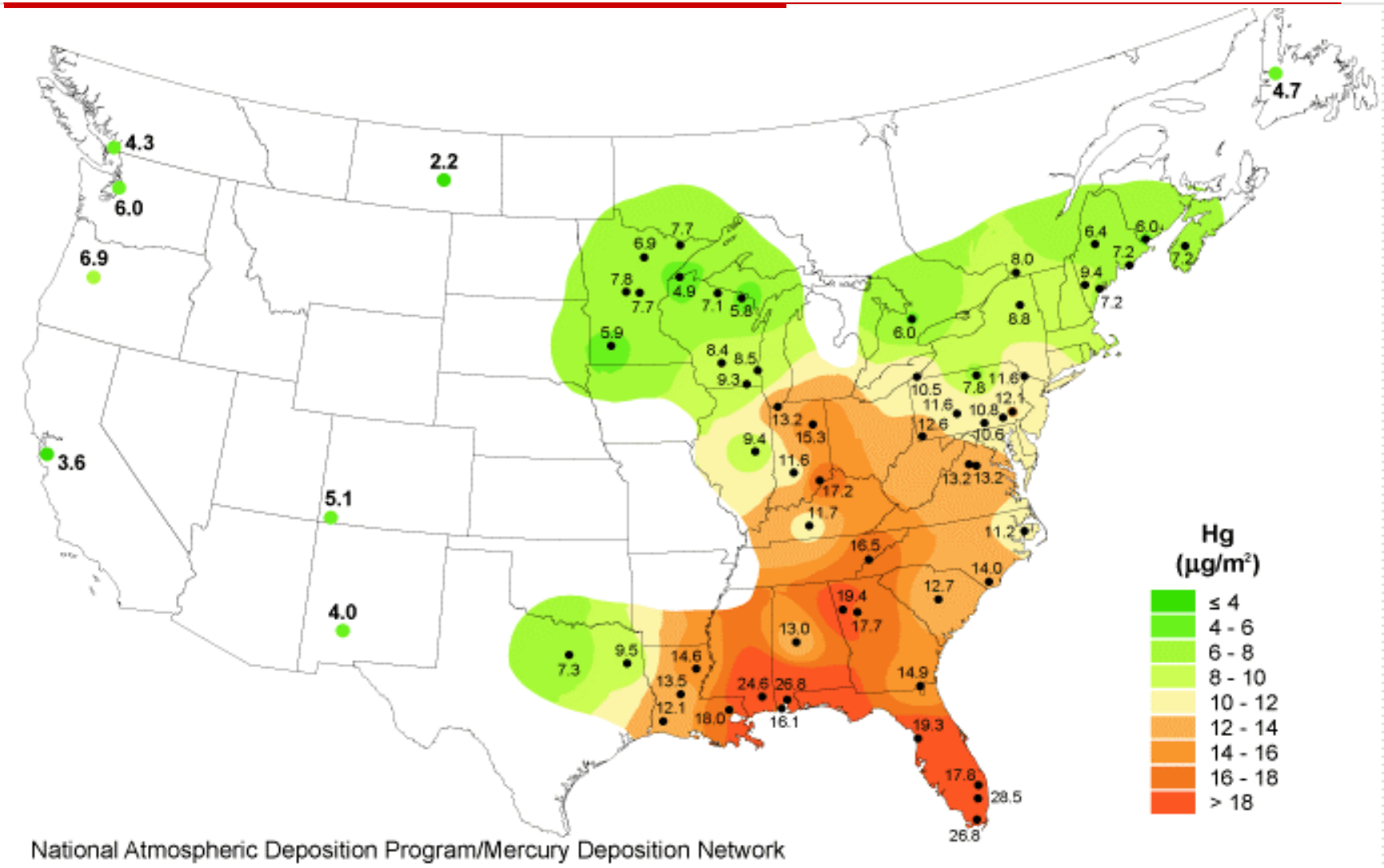
Global Mercury Balance, All Sources

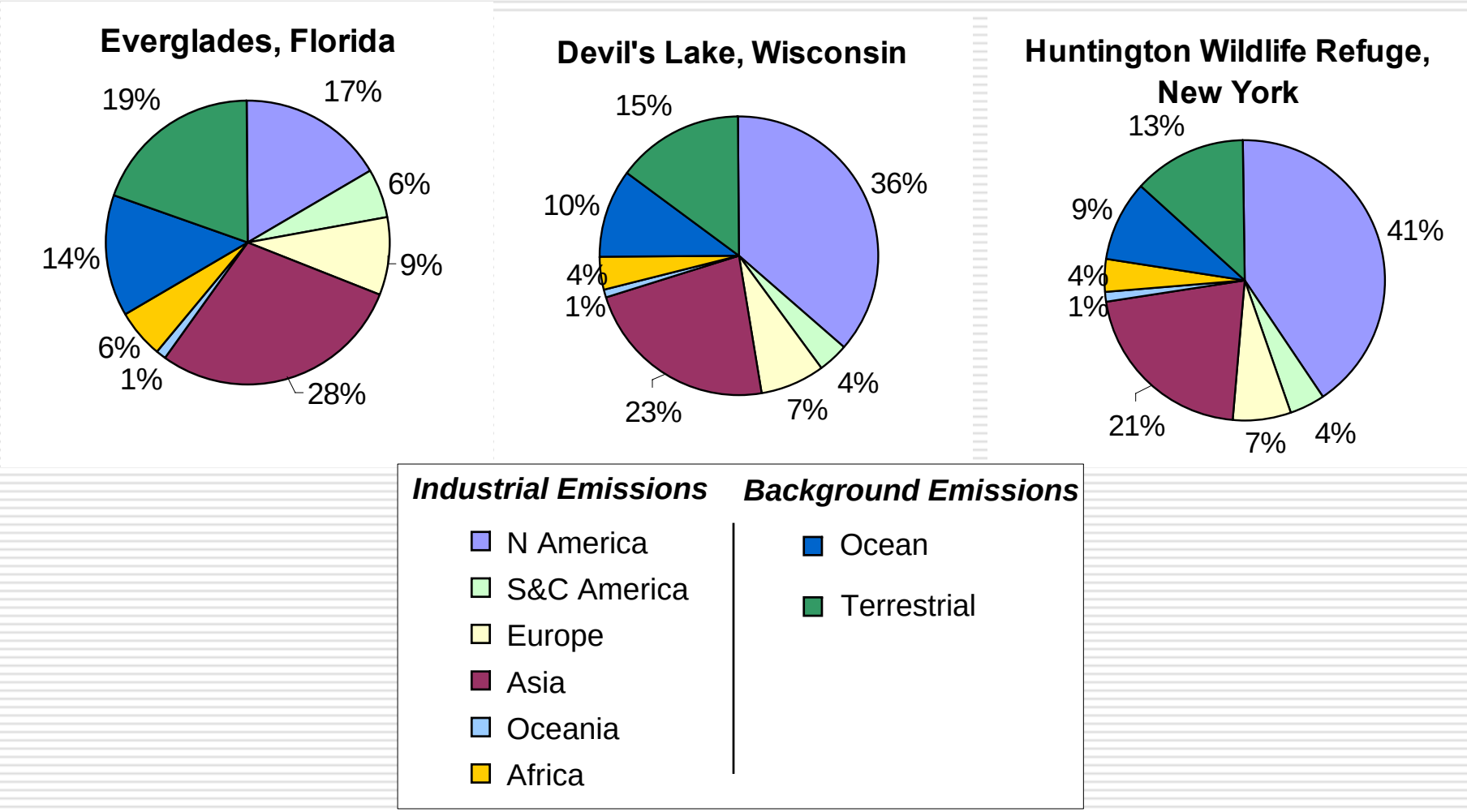
- **Former understanding**
  - **~6000-7000 Mg/y emissions**
  - **~6000+ Mg atmospheric pool**
  - **➔ average lifetime 1 year**
- **Emerging data**
  - **too many sources, not enough sinks, shorter lifetime (Lindberg et al.; Radke et al.; Slemr et al.; others)**
  - **~12,000 Mg/y sources [Increase in assessed background]**
  - **~9,000 Mg/y sinks [need more measurement]**
  - **Net lifetime: weeks to months**
- **Evidence➔ocean transects, sediment coring**
  - **emissions declined from 1960s; recent (1990s) leveling off**
  - **China mercury emissions growing 3-6% per year; India ??**
- **Re-emissions**
  - **the "grasshopper effect"**
  - **may be small (METALLICUS: 20% re-evasion; Nevada: 6%)**

What the Data Tell Us

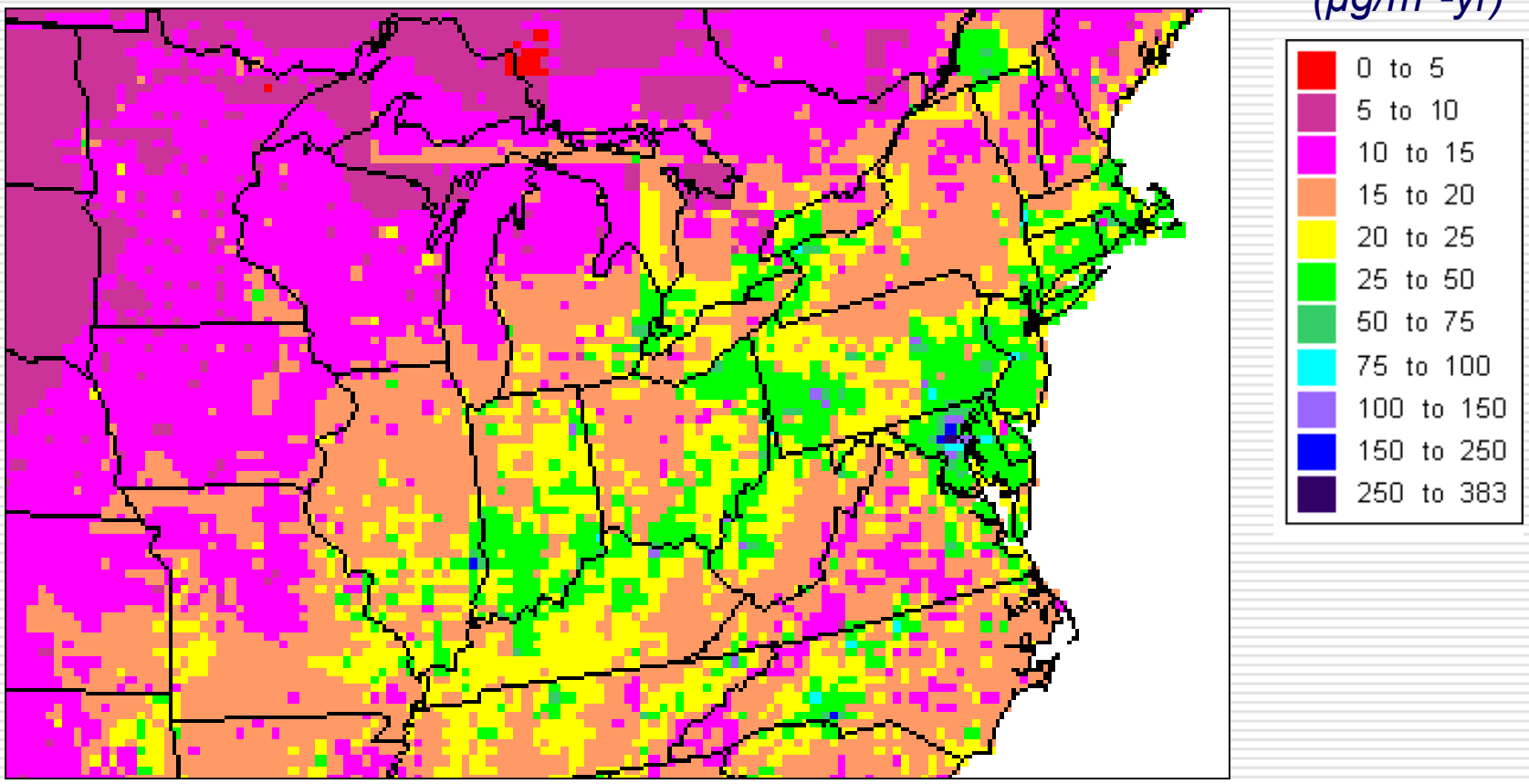
Wet Deposition, U.S. Mercury Deposition Network, 2003



Where Does U.S. Mercury Originate? Global Contributions to U.S. Hg Deposition

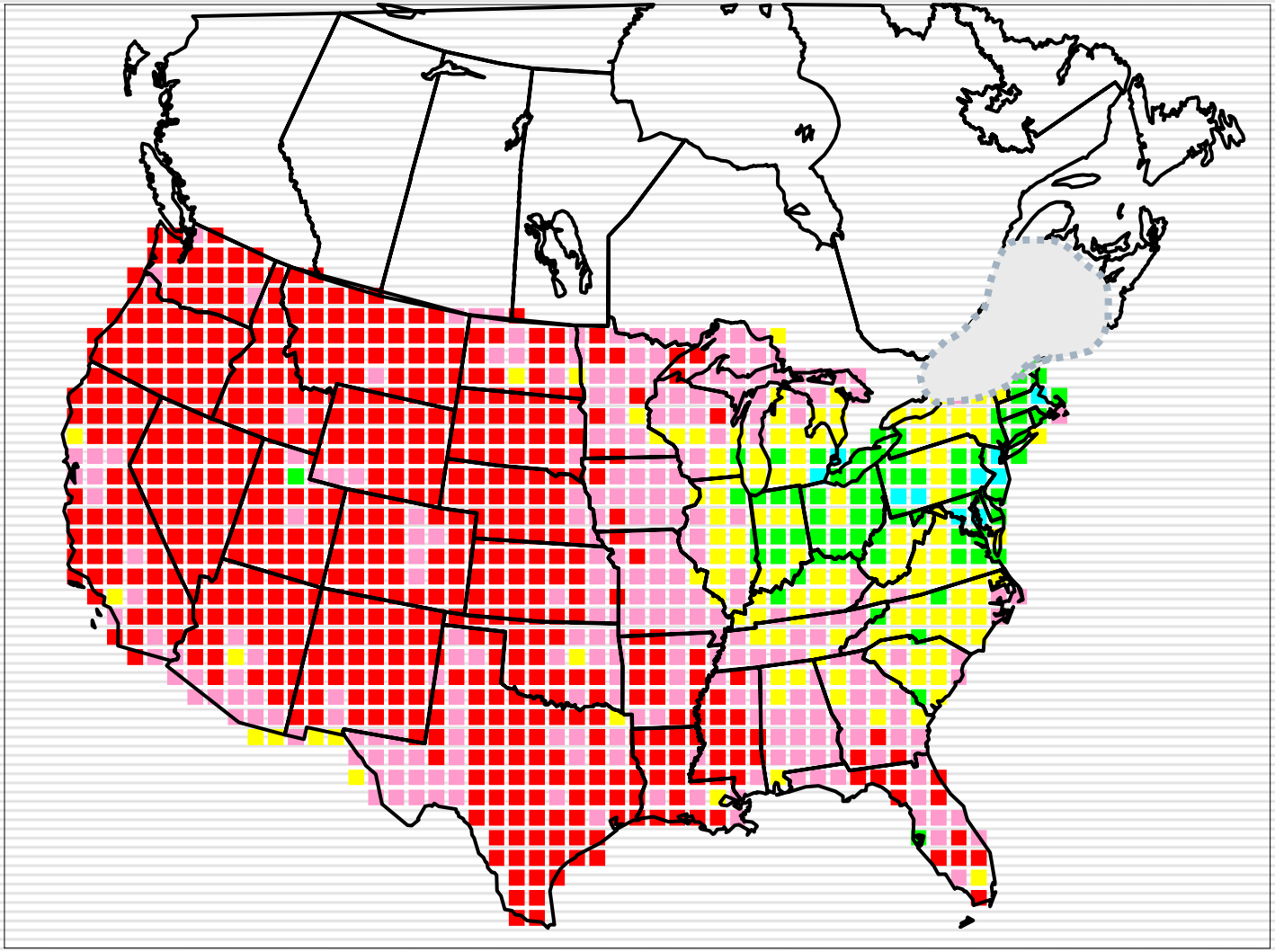


Eastern U.S. Mercury Deposition, 2004
– 20 km Fine Resolution



EPRI TEAM Model, Includes All Global and U.S. Sources

Mercury depositing in the U.S. that originates in other countries



Percent of mercury deposition that originates outside of the U.S.

- 0 to 20%
- 20 to 40%
- 40 to 60%
- 60 to 80%
- 80 to 100%

EPRI TEAM regional model, global chemical model

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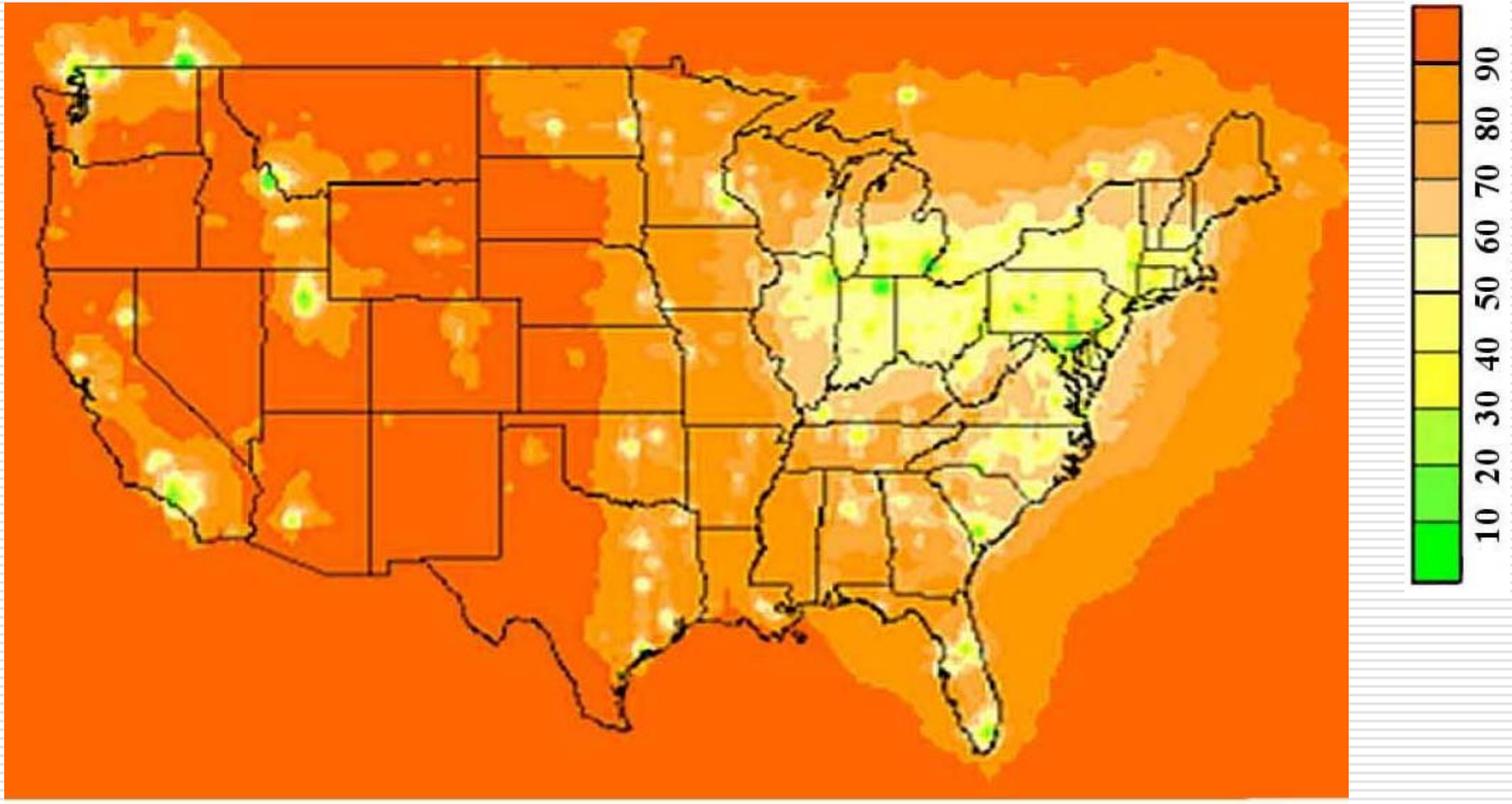
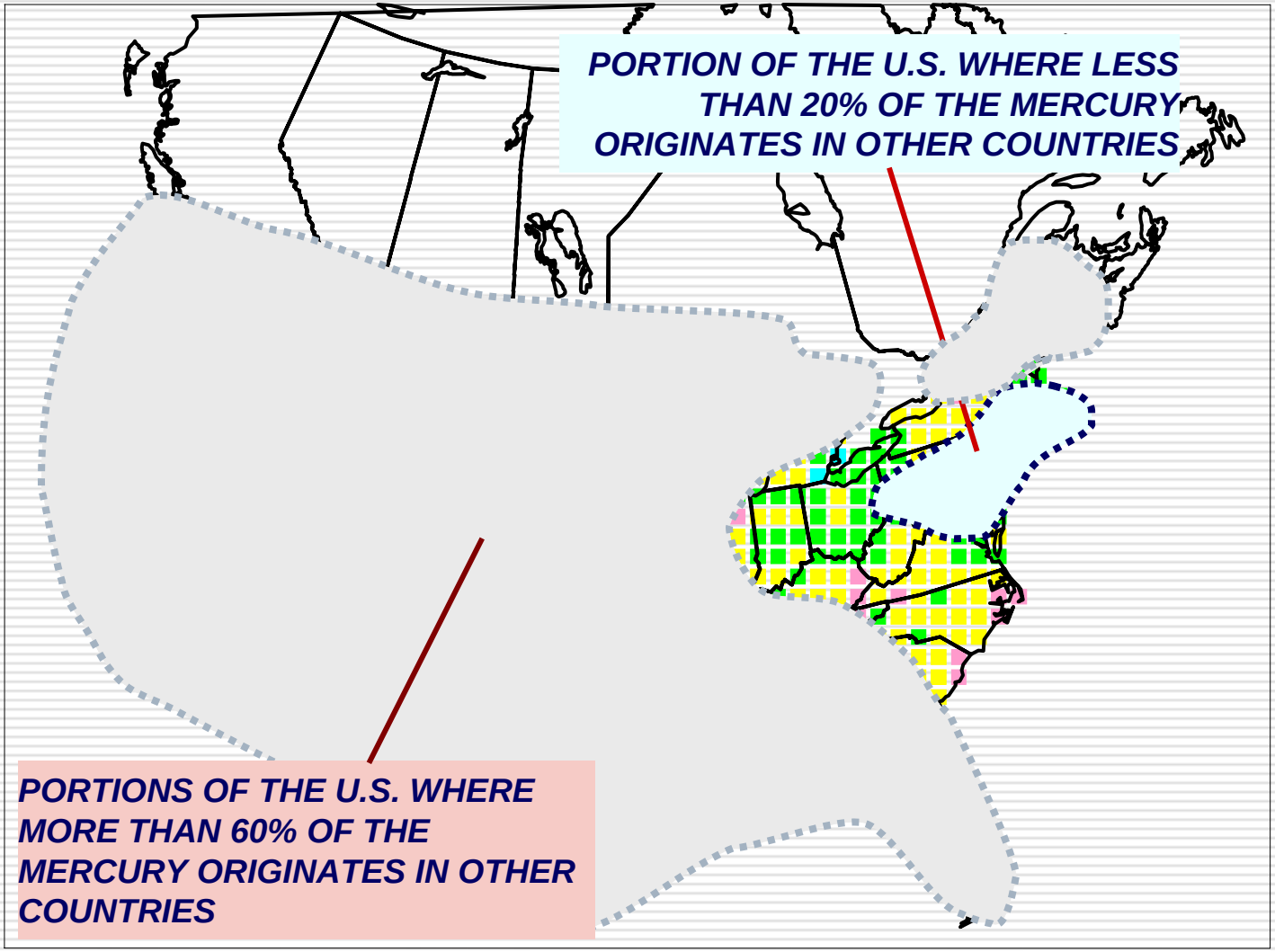
EPA Results: U.S. Deposition from Non-U.S. Sources

Percent deposition from non-US/Canada sources



EPA REMSAD Mercury Modeling (36-km grid)

Is there a mercury management "floor"?



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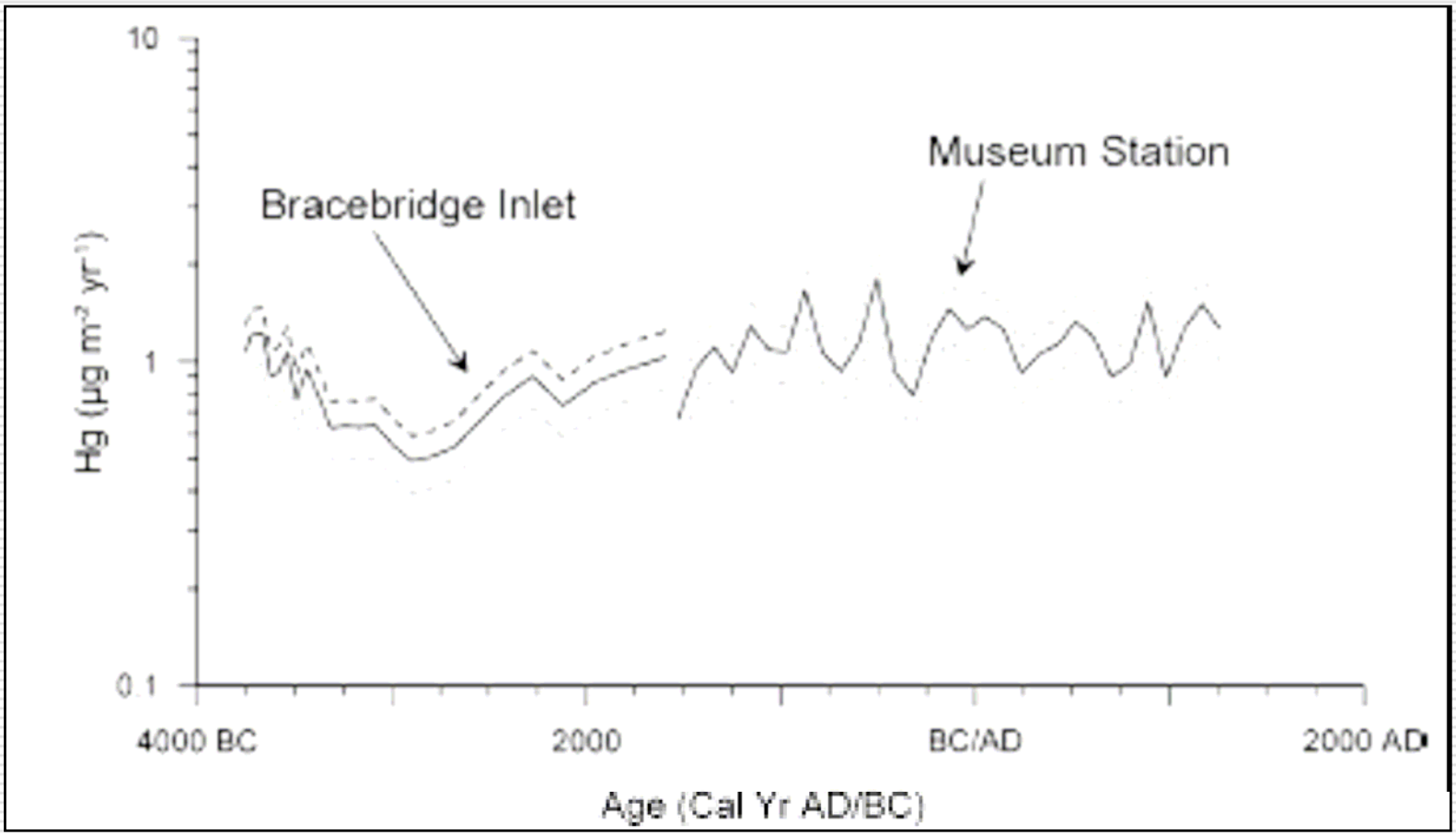
- 60 to 80%
- 80 to 100%

EPRI TEAM regional model, global chemical model

WHAT WAS MERCURY DEPOSITION BEFORE THE RISE OF INDUSTRY?

What is “natural” mercury deposition?



Givelet et al., 2004, *Environ. Sci. Technol., 38*, 4964-4972

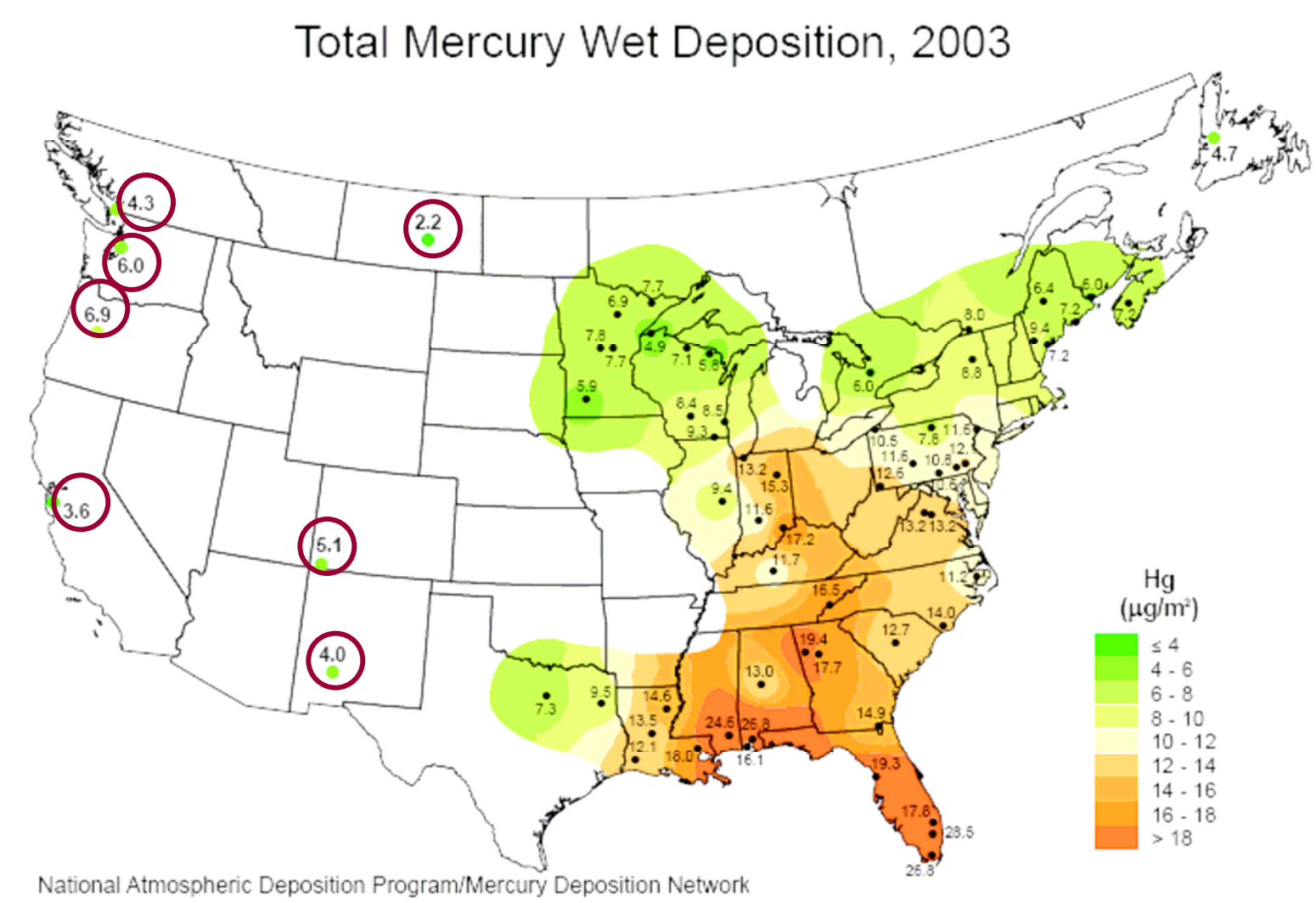
Some Pre-industrial Mercury Deposition Values

site	sample medium	episode	dates	deposition ($\mu g/m^2$-yr)
UFG*	ice	Gold Rush	1850-1878	4.84
UFG	ice	Tambora	1815	8.60
UFG	ice	preindustrial	1719-1847	0.78
Minnesota	lake sed	–	<1850	3.7
Minnesota	lake sed	–	<1750	2
Arctic	lake sed	–	<1850	5
New York	lake sed	–	<1850	7.6

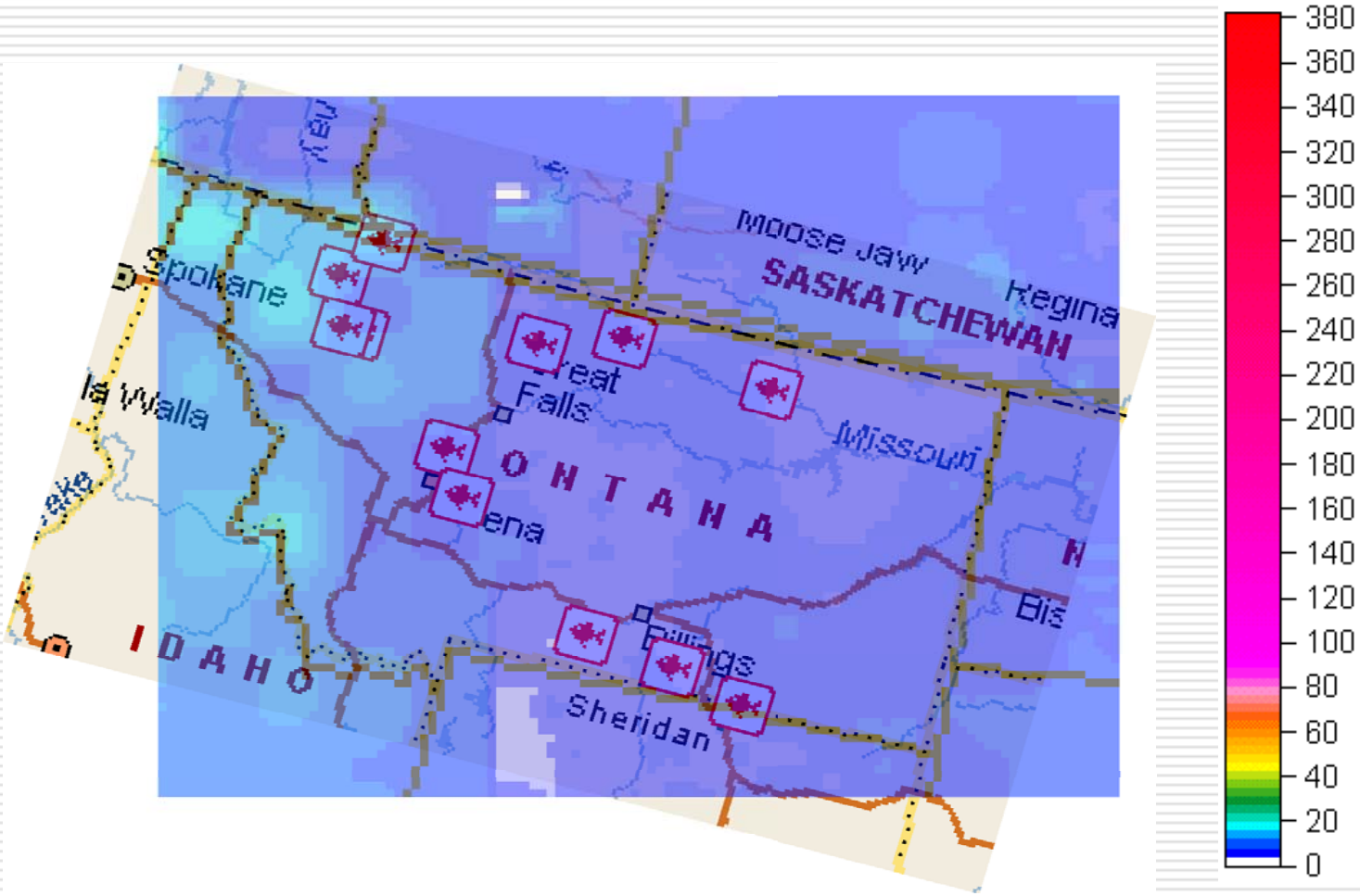
*Upper Fremont Glacier, Wind River mountains, Wyoming, U.S.

Shuster et al., 2002, *Environ. Sci. Technol.*

U.S. wet deposition network data, 2003



...yet "background"-like mercury deposition can lead to fish mercury exceedances



Overlay map: 2004 Hg_{TOT} deposition <10 μg/m^2-y
Background map: (most) Montana waters with 2001 fish levels > 0.3 ppm Hg_{TOT}

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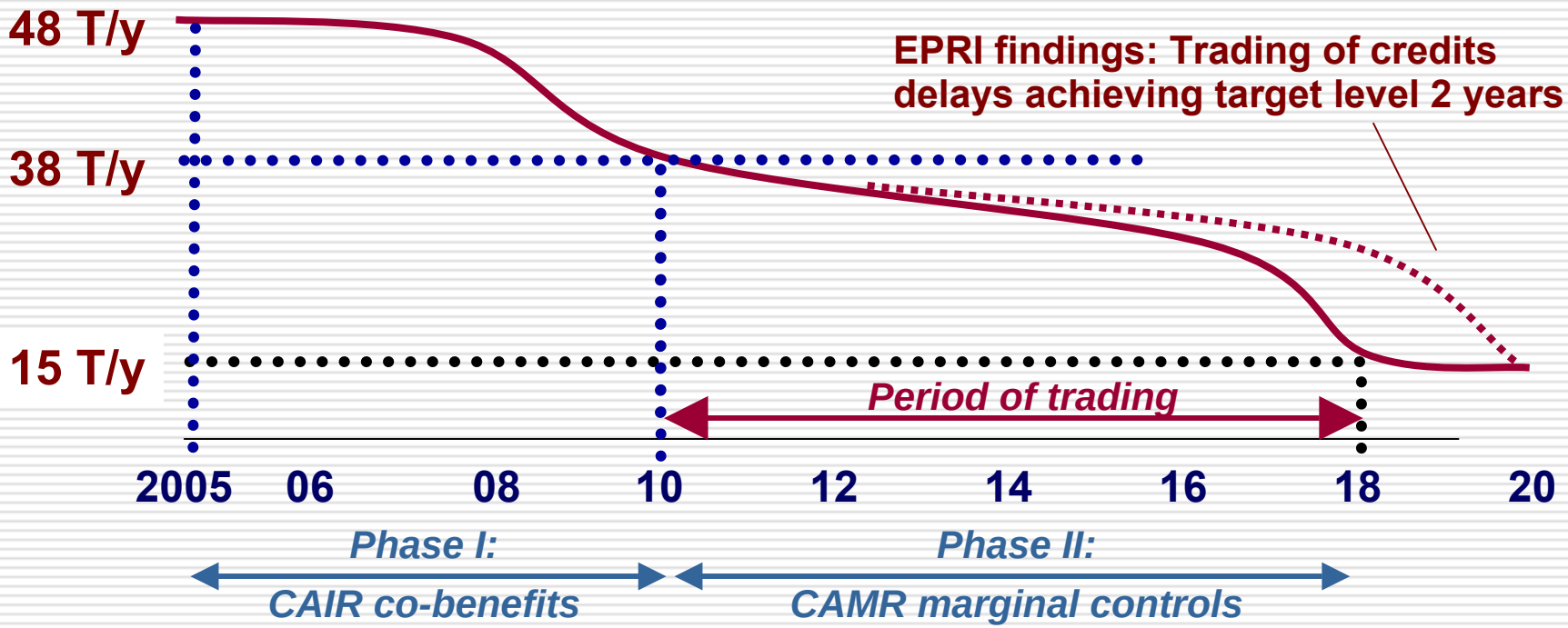
UTILITY MERCURY REGULATION

The March 15 Clean Air Mercury Rule (CAMR): Cap & Trade



Cap-&-trade: 70% emissions cut by 2018

CAIR: Clean Air Interstate Rule (midwestern/eastern utility SOx, NOx, PM)
CAMR: Clean Air Mercury Rule (all coal plants, national-state Hg caps)

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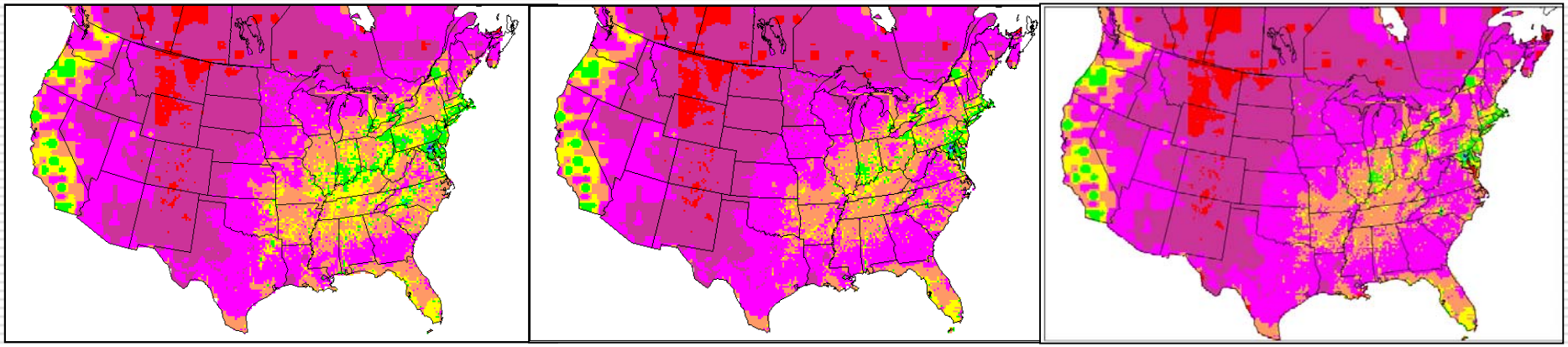
Deposition of mercury, current + 2 scenarios



Mercury deposition, 2004

Mercury deposition, 2020, CAIR+CAMR

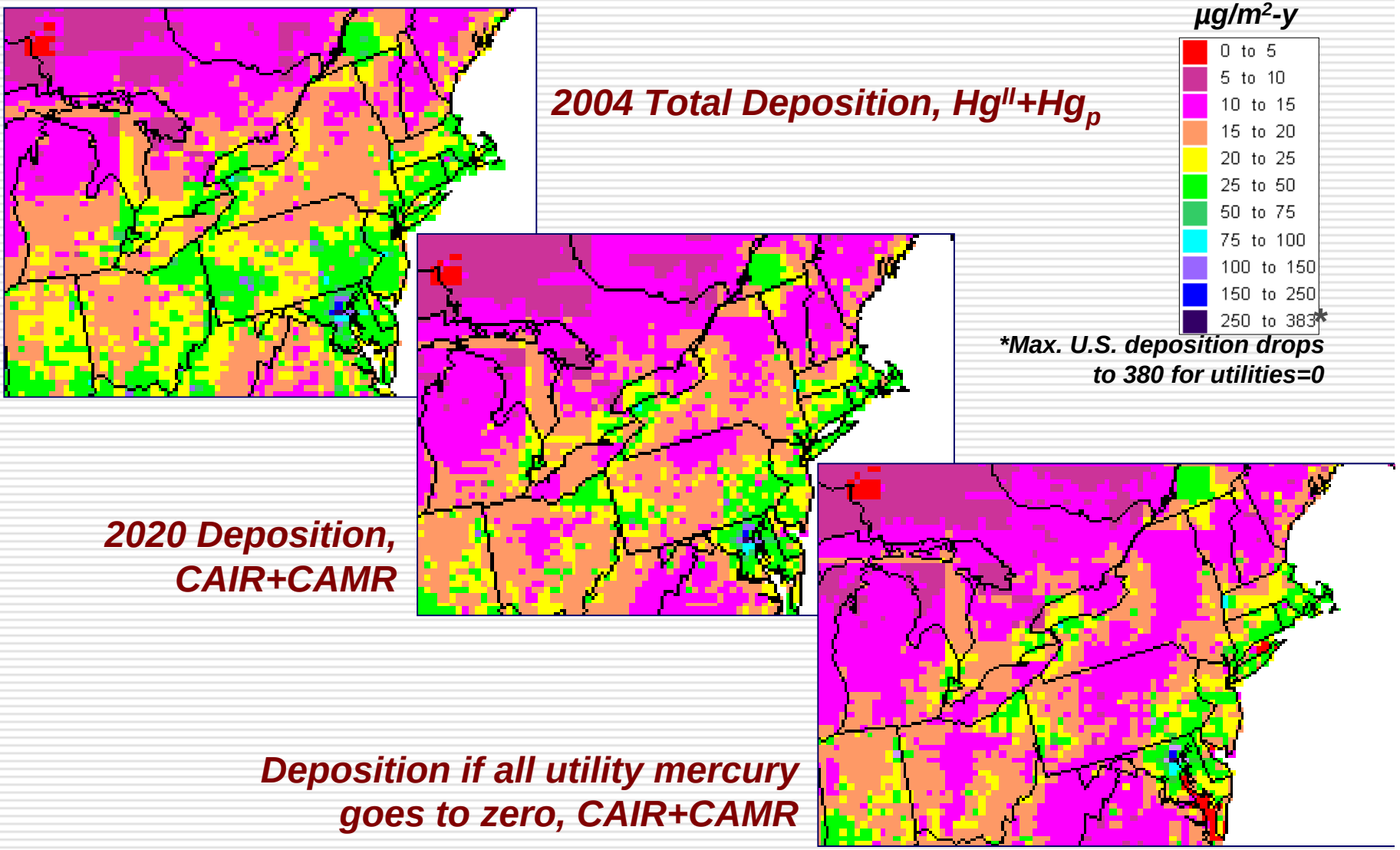
Mercury deposition, IF utility mercury is forced to 0

µg/m²-y

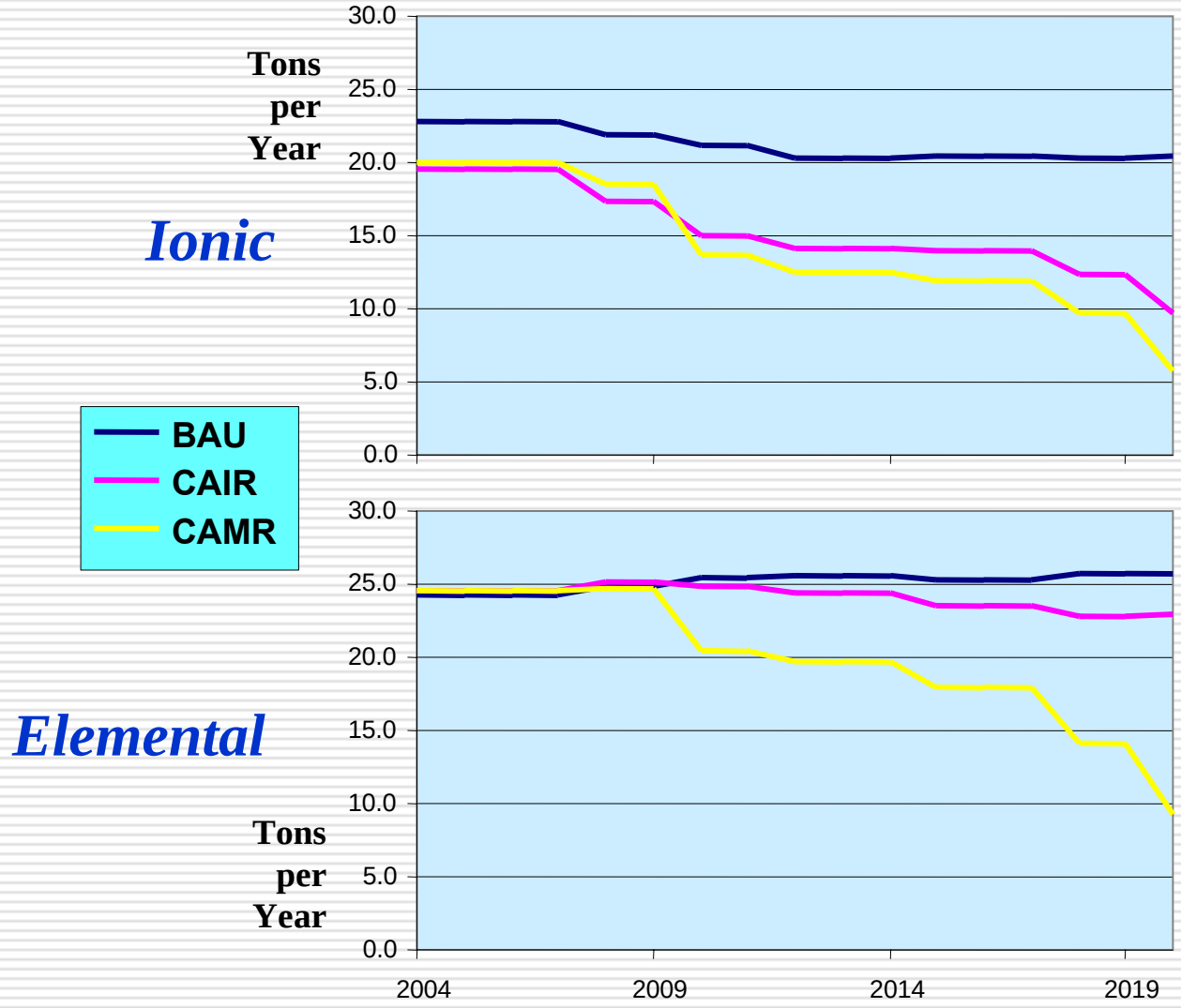
- 0 to 5
- 5 to 10
- 10 to 15
- 15 to 20
- 20 to 25
- 25 to 50
- 50 to 75
- 75 to 100
- 100 to 150
- 150 to 250
- 250 to 383

- Most U.S. mercury originates in other countries, but with regional variations (kept constant w/time)
- Most deposition changes are due to CAIR, not CAMR (easier, earlier capture of ionic mercury)
- Exposure to U.S. women will decline only slightly, even if utilities are “zeroed out”

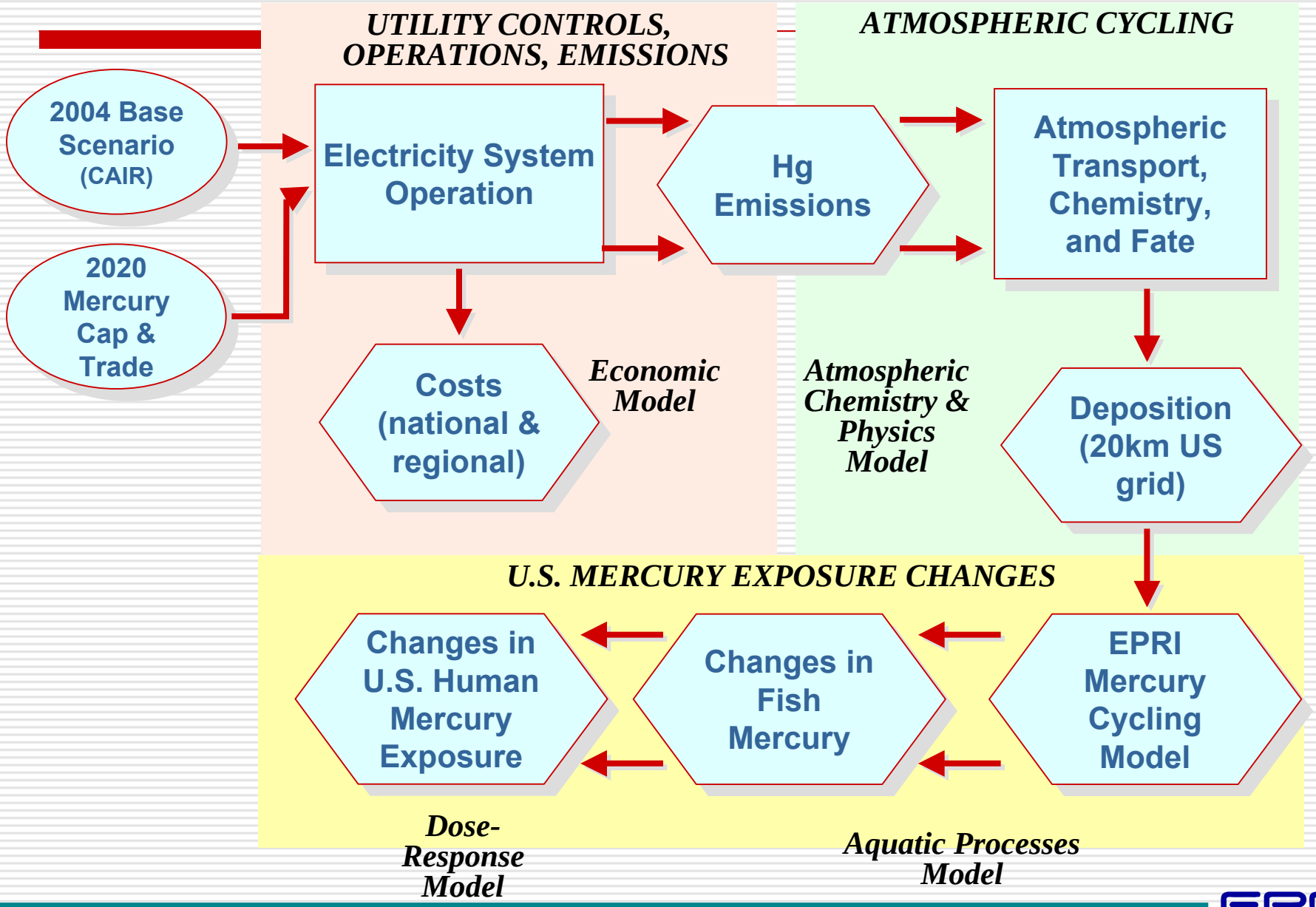
Focus on Pennsylvania



U.S. Mercury Emissions by Species Under CAIR, CAIR+CAMR



Integrated Modeling Approach



EPRI+EPA Findings: Mercury Exposures Drop Slightly, CAIR+CAMR

% Exposure Drop by State, 2020 CAMR

EPRI emissions and regional air models; U.S. EPA fish data by state; EPRI Mercury Cycling Model

Cuts in Utility Emissions, Deposition, and Costs

- ¾ of all mercury emissions leave the U.S. ➔ large cuts in emissions ➔ much smaller cuts in deposition

↑ Emissions ↓ Deposition	↑ Hg Total Utility Mercury EMISSIONS, (U.S.) tons/yr	↑ Hg % DROP in Utility Mercury EMISSIONS from 2004	Hg ↓ Total U.S. Mercury DEPOSITION, from ALL MERCURY SOURCES	Hg ↓ % DROP in Mercury DEPOSITION from Base Case	$ TOTAL COST (Net Present Value) to Reach Target
CURRENT CONDITIONS (2004 Base Case)	46.6 tons/yr	–	164 tons/yr	–	
2020 MACT SCENARIO (initial proposal)	30.2 tons/yr	- 35%	156 tons/yr	- 5%	$10 billion
2020 CAP & TRADE SCENARIO	14.9 tons/yr	- 68%	153 tons/yr	- 7%	$2 billion

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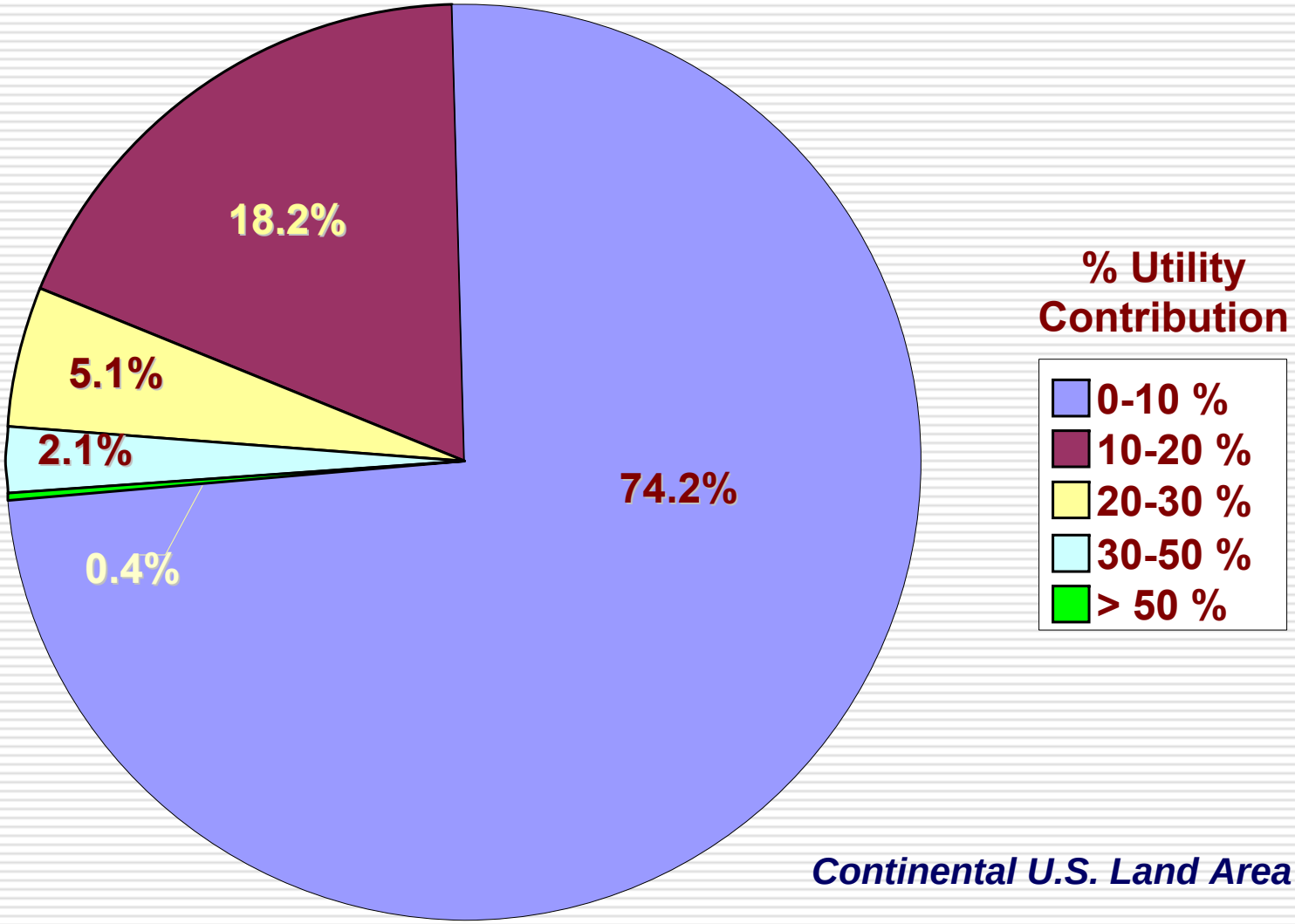
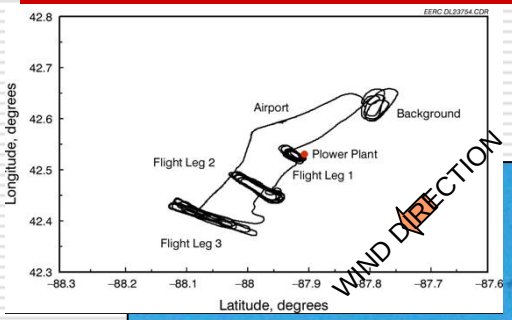
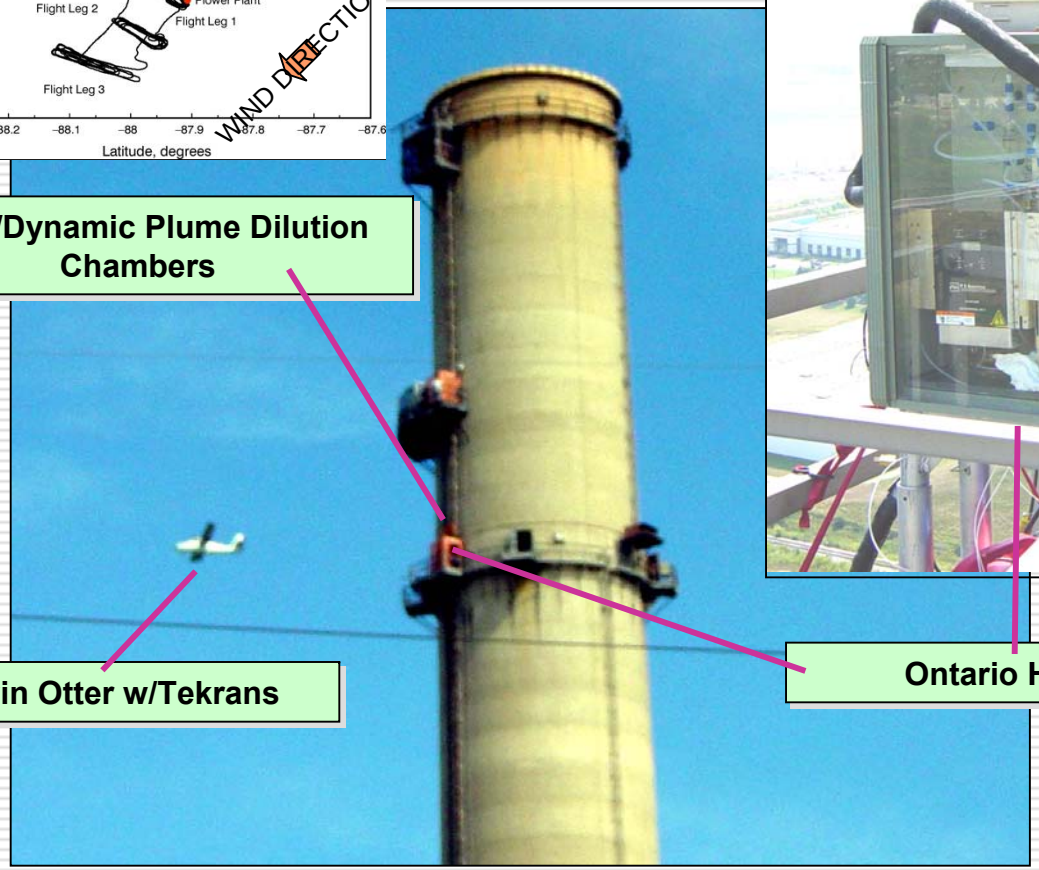
Mercury “hot spots”? (locations where utility mercury dominates the deposition)



Continental U.S. Land Area

Do Power Plant Plumes Enhance the Reduction of Ionic to Elemental Mercury?





Static/Dynamic Plume Dilution Chambers

Twin Otter w/Tekrans

Ontario Hydro/CEMs

WHERE DOES THE RESEARCH GO NEXT?

RESEARCH NEEDS FOR PUBLIC POLICY SUPPORT

- **Monitoring progress following regulation**
  - Basis: increased protection of public health
  - Metrics?
    - Lower deposition ("smoothing the peaks")
    - Drops in fish levels of mercury
    - Measurable declines in blood mercury, most-sensitive women of childbearing age But…
- **How well we see the signal of progress?**
  - Will local improvement be masked by global growth?
  - Where do we look? Most-sensitive indicators… Locations of highest deposition… Locations with elevated fish Hg
  - How do we look? Improvements in sampling strategy, analysis
  - When do we look? Rapid response of deposition? Slow response of fish and water bodies?
  - The puzzle of the NHANES data

The *METALLICUS* Project

Mercury Experiment To Assess Lake Loading In Canada and the United States

^{199}Hg
^{201}Hg
^{203}Hg

IVL, Sweden
U.S. Geological Survey
U.S. Department of Energy
Canada Lakes and Oceans
University of Montreal
McGill University
Trent University
Smithsonian Institution
University of Connecticut
University of Wisconsin
Oak Ridge National Laboratory
Electric Power Research Institute
U.S. Environmental Protection Agency
Ontario Ministry of the Environment

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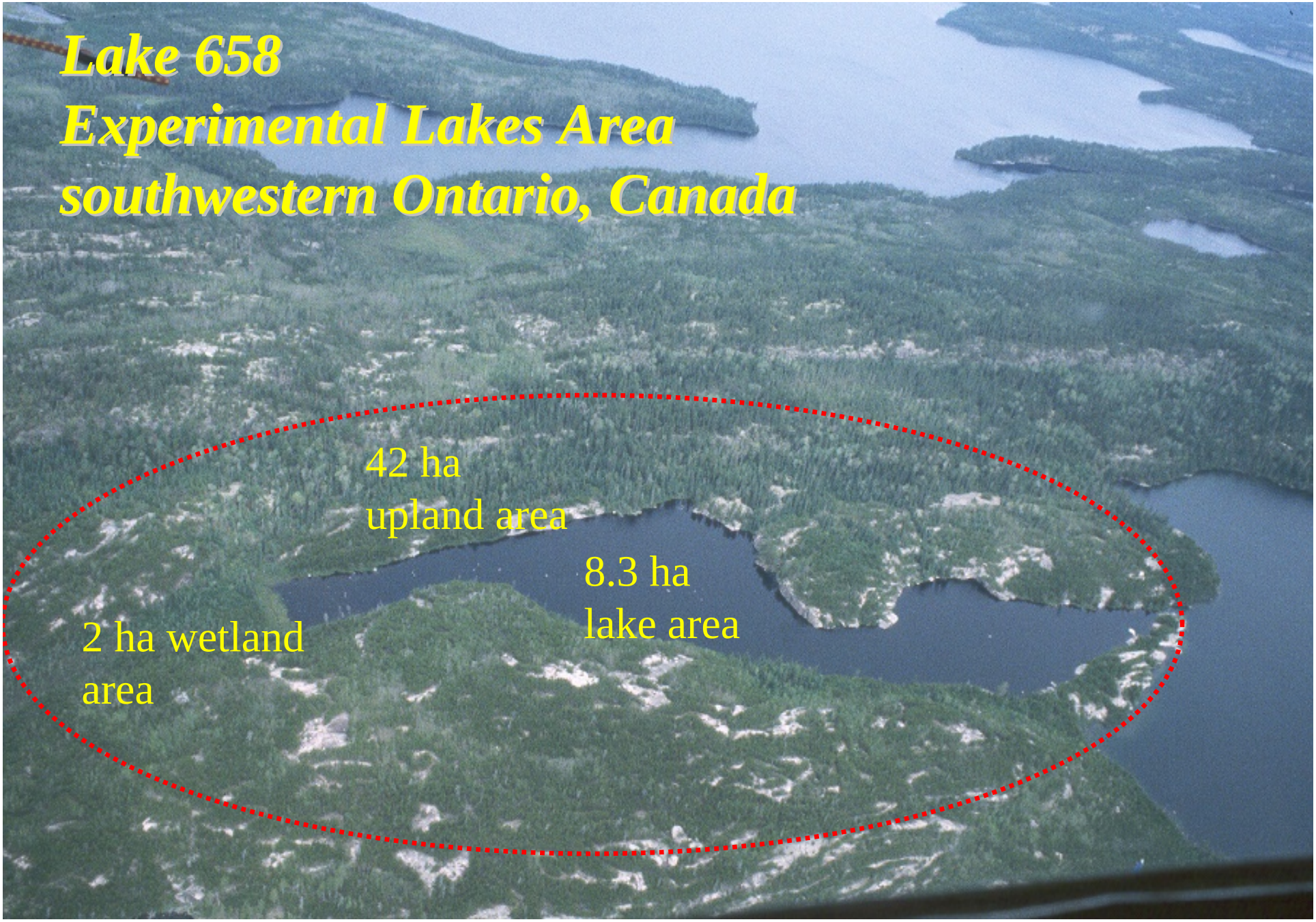
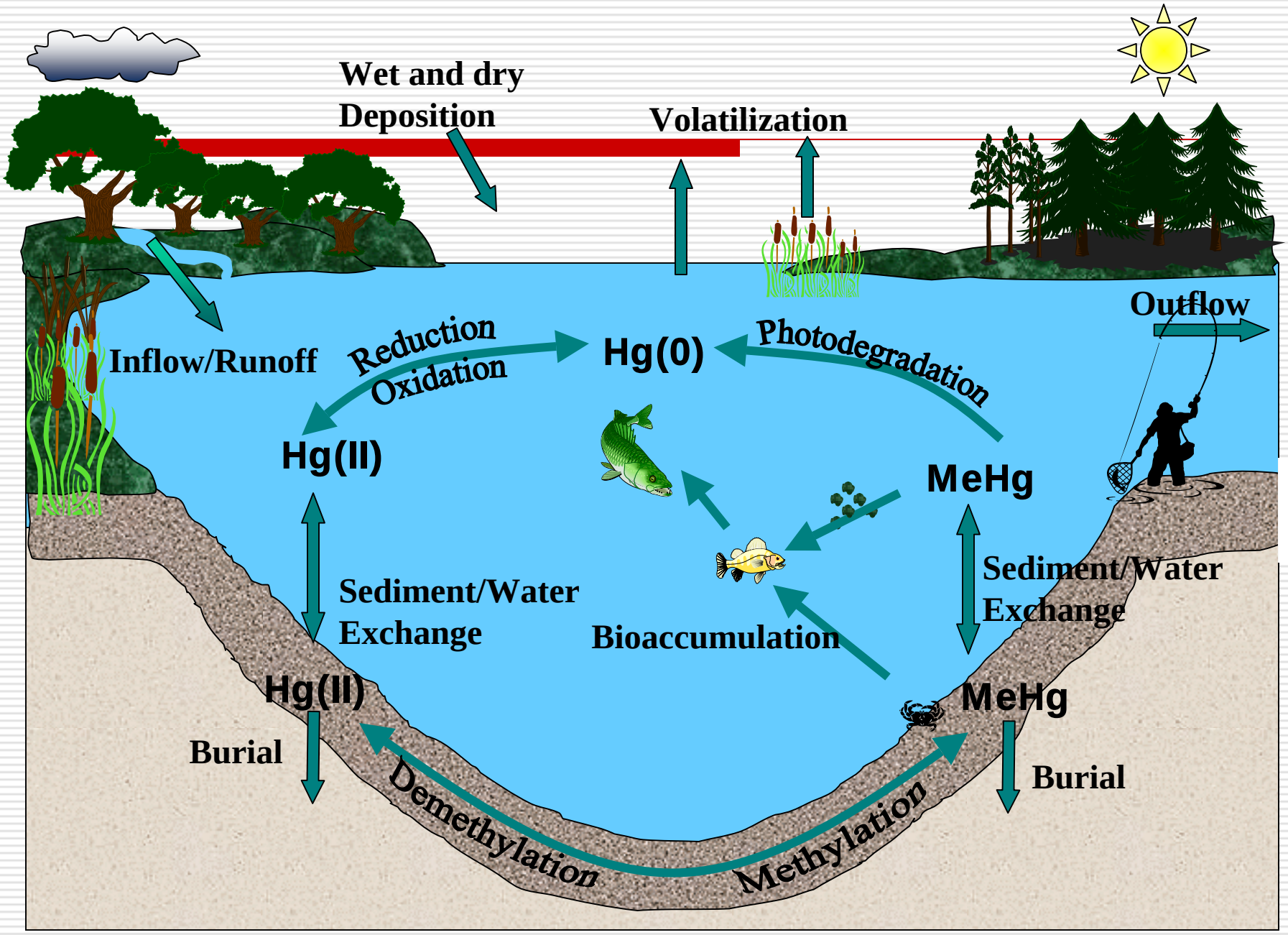
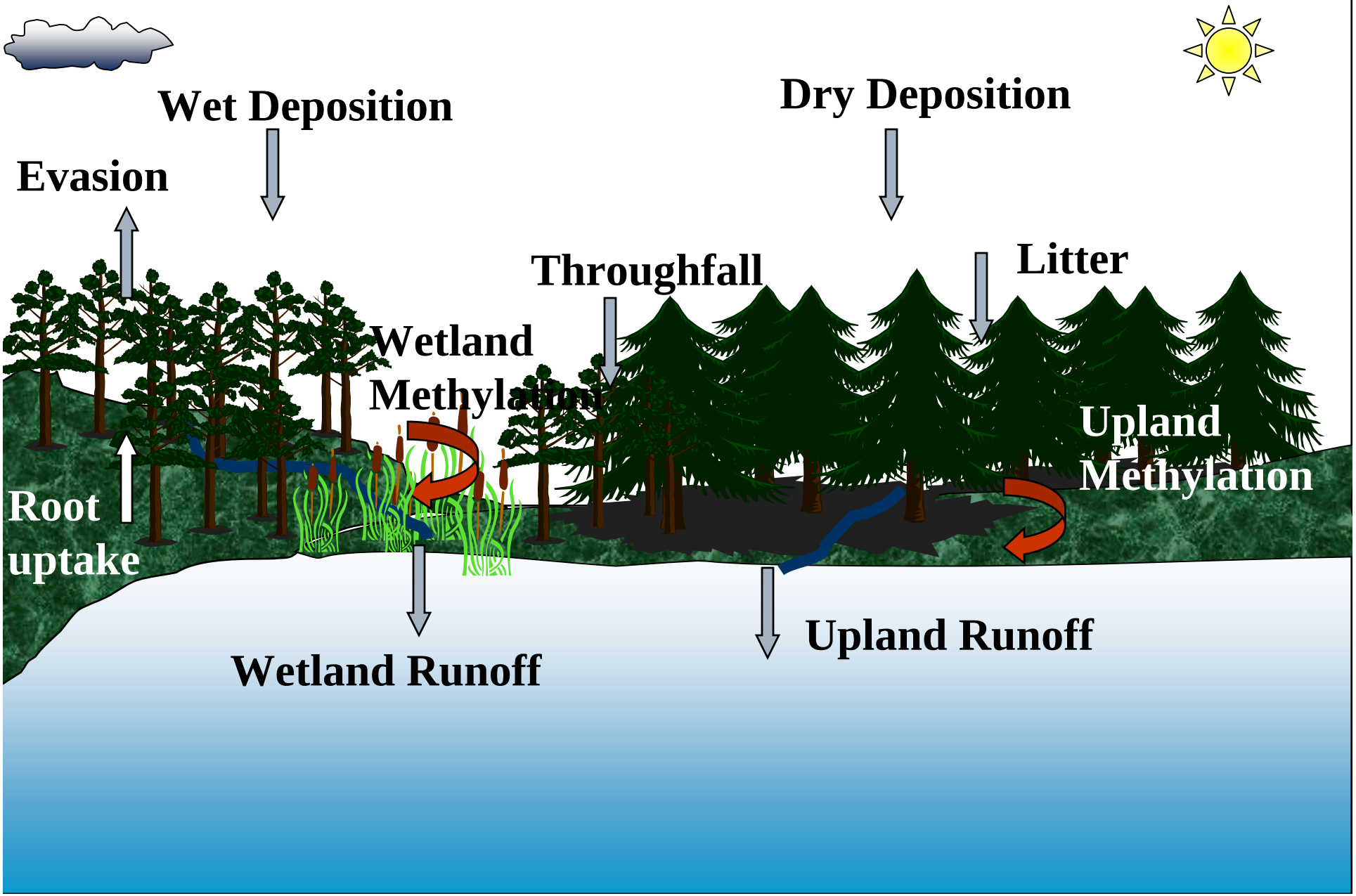
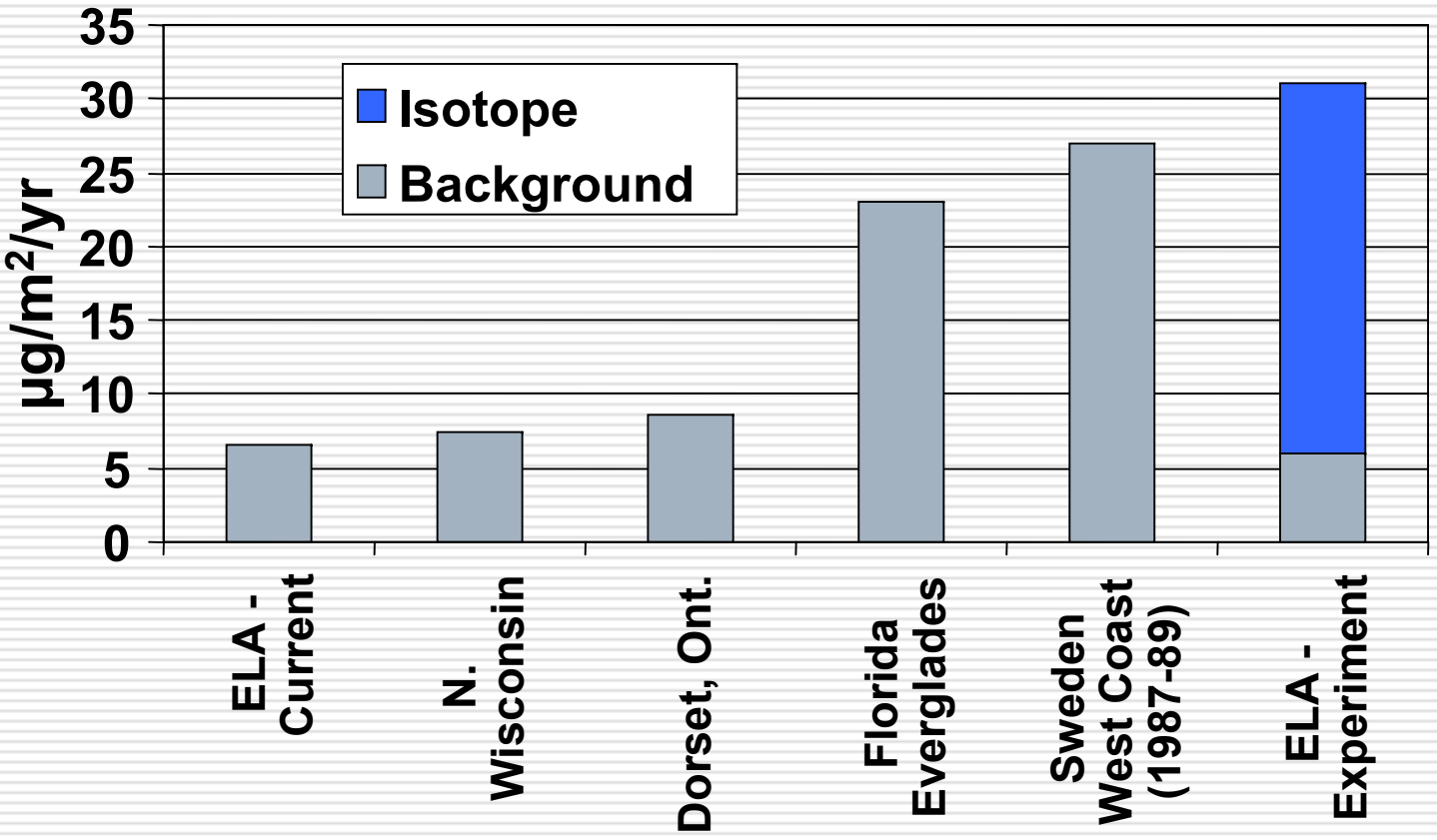

Lake 658
Experimental Lakes Area
southwestern Ontario, Canada
42 ha
upland area
8.3 ha
lake area
2 ha wetland
area


Wet and dry
Deposition
Volatilization
Outflow
Inflow/Runoff
Reduction
Oxidation
Hg(0)
Photodegradation
Hg(II)
MeHg
Sediment/Water
Exchange
Bioaccumulation
Sediment/Water
Exchange
Hg(II)
MeHg
Burial
Demethylation
Methylation
Burial

METALLICUS in the Terrestrial System



Spiking the background*



**with permission of the provincial and national governments and the generous supply of mercury stable non-radioactive isotopes by the government of Russia*

Basic Issues

- MERCURY HEALTH EFFECTS: Reducing uncertainties: tests, confounders, group differences; adult-onset effects
- MERCURY SOURCES: Natural sources; international inputs
- MERCURY DYNAMICS: Similar depositions across the U.S.; mercury re-emissions to atmosphere
- RISK ASSESSMENT: Small changes in random risk; monitoring changes over time
- DUE DILIGENCE: “Early sentinels” of mercury changes; long trends from small changes

QUESTIONS?

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