Global Biogeochemical Cycling of Mercury and Implications for Human Exposure

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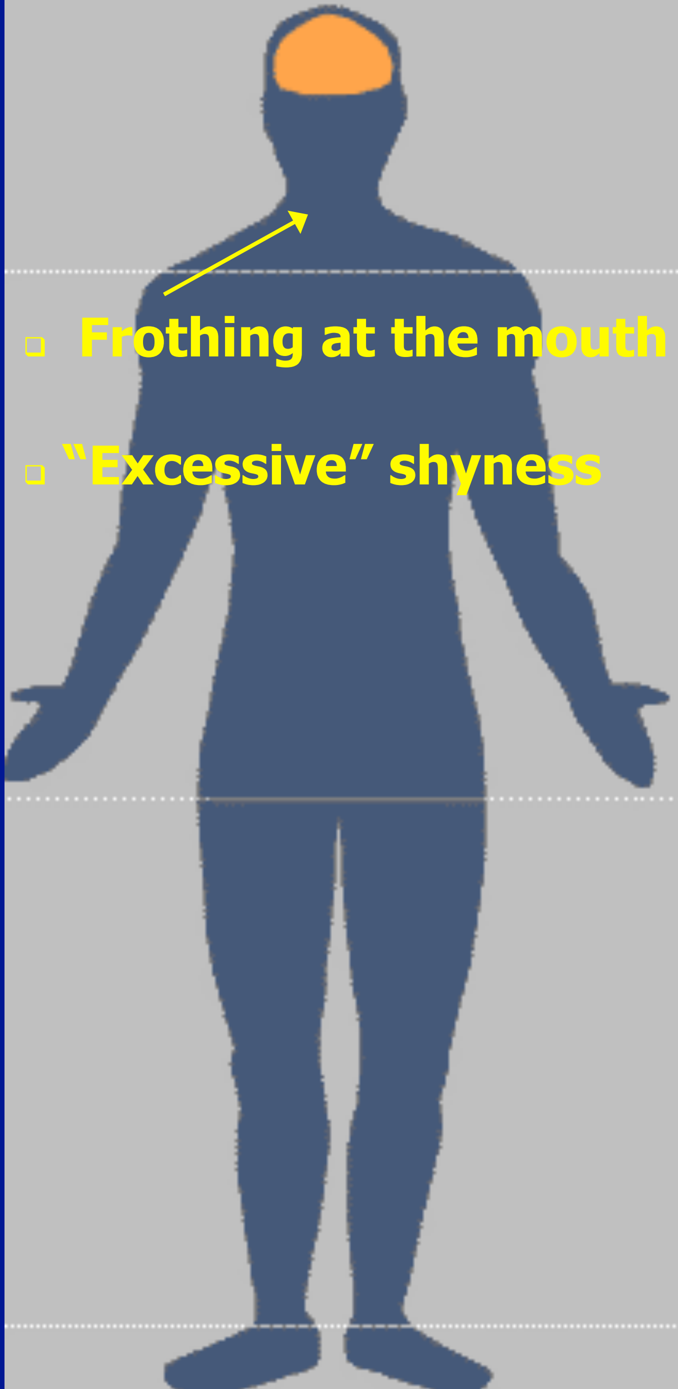
Why are we worried about Hg in the environment?

Mercury in Fish Predicted To Soar

Pollution From Asian Power Plants Threatens Children's Health

WASHINGTON - May 6 - A landmark study by scientists from the U.S. Geological Survey (USGS) <http://www.eenews.net/public/25/10743/features/documents/2009/05/01/document_gw_03.pdf> and universities in the U.S. and Australia has, for the first time, documented how escalating mercury-laden air emissions, chiefly from coal-fired electrical power plants in Asia, are being transformed into methylmercury, a potent neurotoxin that is increasingly polluting the North Pacific Ocean and contaminating tuna, swordfish and other popular seafood.



- Frothing at the mouth
- "Excessive" shyness

- Deteriorates nervous system
- Impairs hearing, speech, vision and gait
- Causes involuntary muscle movements
- Corrodes skin and mucous membranes
- Causes chewing and swallowing to become difficult

Source: *USGS Fact Sheet*

Negative effects on neurodevelopment in children focus of most guidelines

EPA's RfD (2000)

Children IQ Deficits

- From fetal exposures above MeHg RfD
- From any fetal MeHg exposures

Adult Cardiovascular Effects

- Male consumers of non-fatty freshwater fish with high MeHg
- Male fish consumers
- All fish consumers

Decreasing Credibility →

Source: Rice and Hammitt, 2005

Discussion by EPA on use of cardiovascular health outcomes in regulatory assessments

Evaluation of the Cardiovascular Effects of Methylmercury Exposures: Current Evidence Supports Development of a Dose-Response Function for Regulatory Benefits Analysis

Henry A. Roman, Tyra L. Walsh, Brent A. Coull, Eric Dewailly, Eliseo Guallar, Dale Hattis, Koenraad Mariën, Joel Schwartz, Alan H. Stern, Jyrki K. Virtanen, and Glenn Rice

Environmental Health Perspectives, 2011, 119(5): 607-614.
doi: 10.1289/ehp.1003012 (available at http://dx.doi.org/)

Adult toxicity can occur in rare cases

IMAX®

Sushi 2x /day for 10 years

“After a personal experience with mercury poisoning, Richard Gelfond ’82 (IMAX CEO) established a new fund in the amount of $1 million to improve understanding of environmental cycling and public health effects of mercury.”

http://www.stonybrook.edu/research/news/RN/resnew100512.shtml

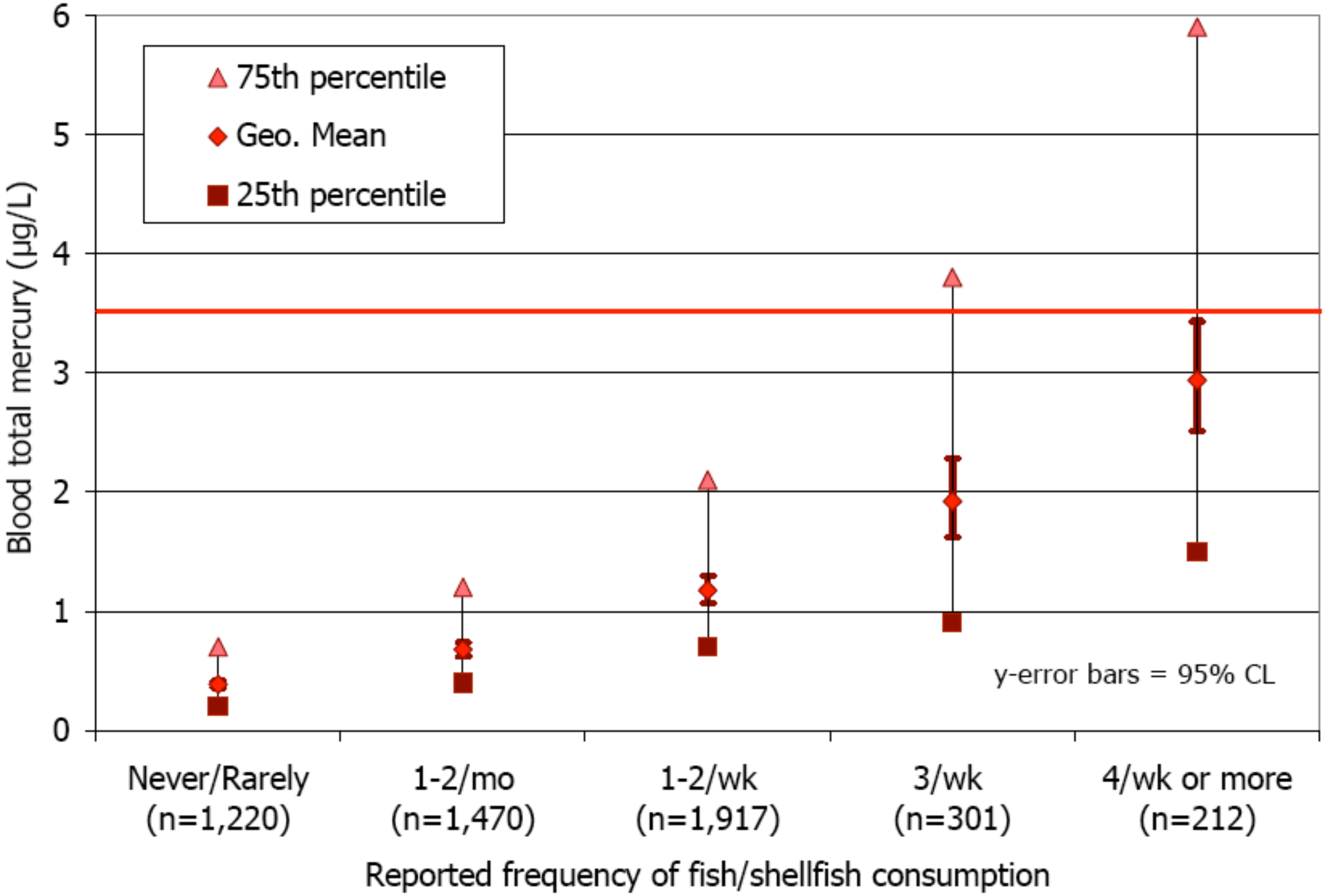
Methylmercury (MeHg) is more toxic than inorganic Hg (Hg^{II} and Hg^0)

- Easily absorbed through gastrointestinal system
  - Food type makes no difference in absorption
  - 95% of ingested MeHg is absorbed
  - Distributed throughout the body
  - Easily passes the placenta and blood-brain barrier
  - Half-life ~50-70 days

- Biomarkers of exposure
  - Blood
  - Hair
  - Nails

Methylmercury (MeHg) exposure is from fish consumption

U.S. Women of Childbearing Age (16-49), NHANES 2004 data



Mahaffey et al., 2009, EHP

2008 U.S. Fish Advisories

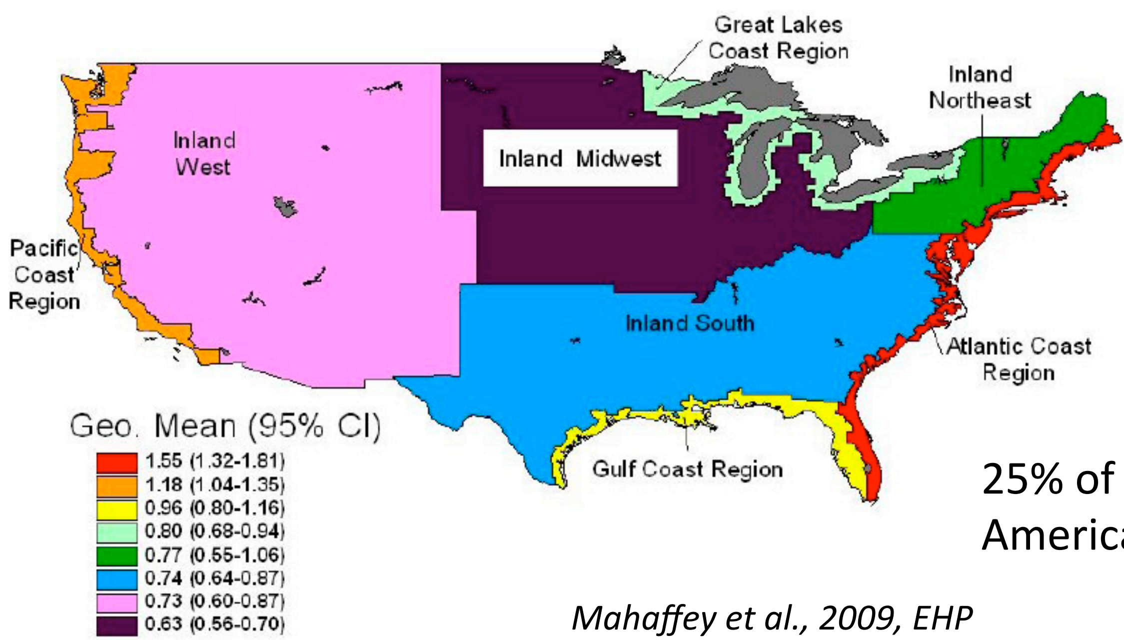
Chemical	No. of Advisories	Lakes under Advisory
Mercury	3361	16,808,032
PCBs	1025	6,049,506
Dioxins	123	35,400
DDT	76	876,520
Chlordane	67	842,913

Source: U.S. EPA Office of Water

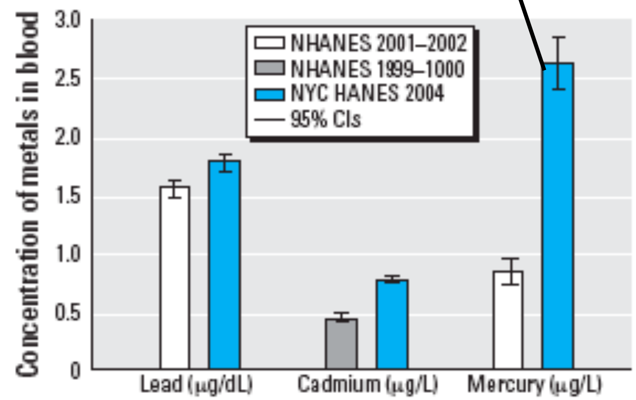
Geographic Variability in Hg Exposures

NHANES Blood Hg in US Women 16-49

~6% > (EPA’s RfD) (1999-2002) => 316,588 - 637,233 children



Women in NYC



25% of New Yorkers and 50% of Asian Americans in NY exceed safety standards

Mahaffey et al., 2009, EHP

McKelvey et al., 2007, EHP

$$\text{Hg Exposure} = \Sigma(\text{Meal Frequency} \times \text{Meal Size} \times \text{Hg in Fish})$$

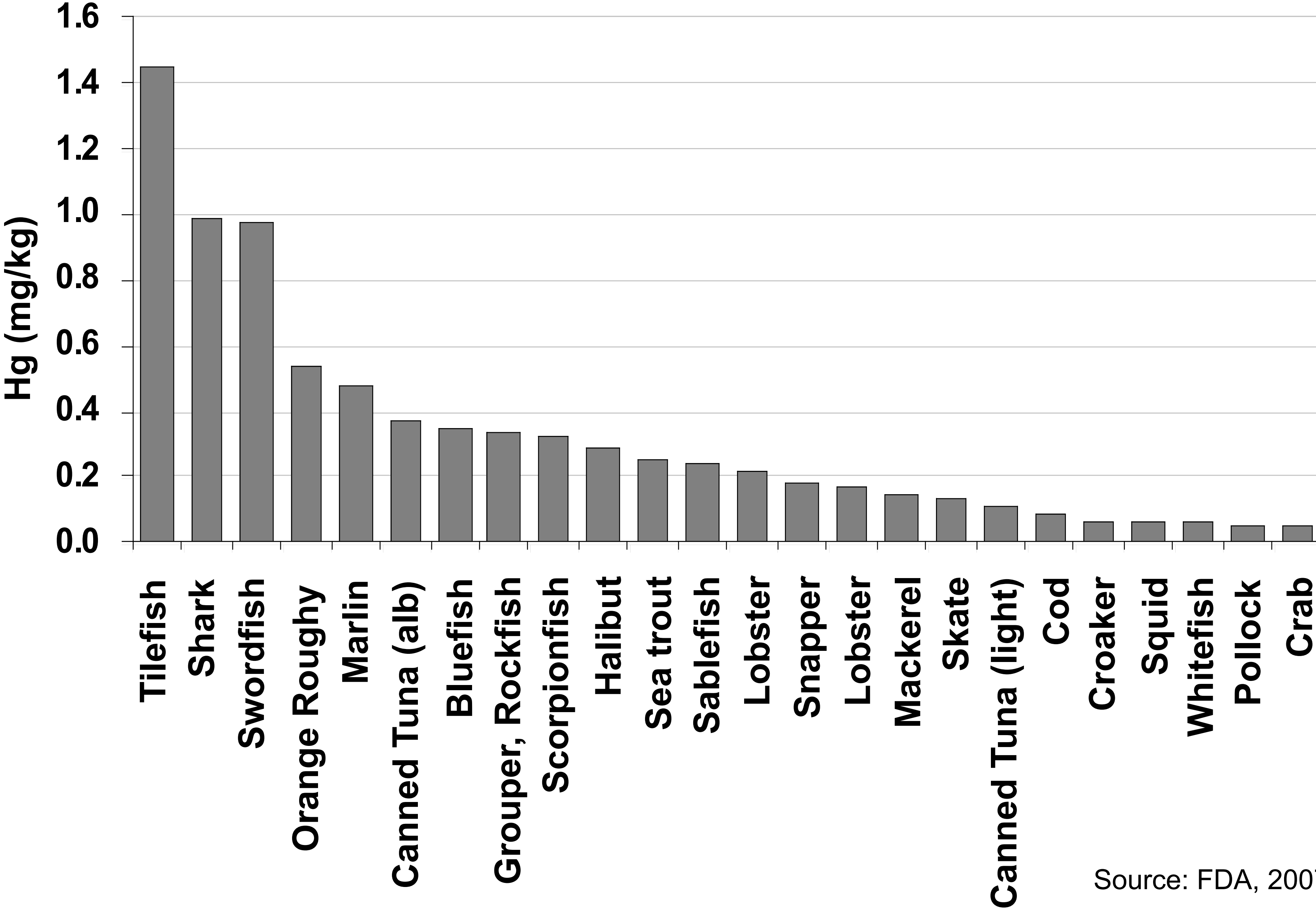
Uncertainty Dietary Survey Data

-Length/type of survey
-Demographic groups represented
-Meal frequency recall
-Meal sizes
-Species selection

Variability Physical Environment

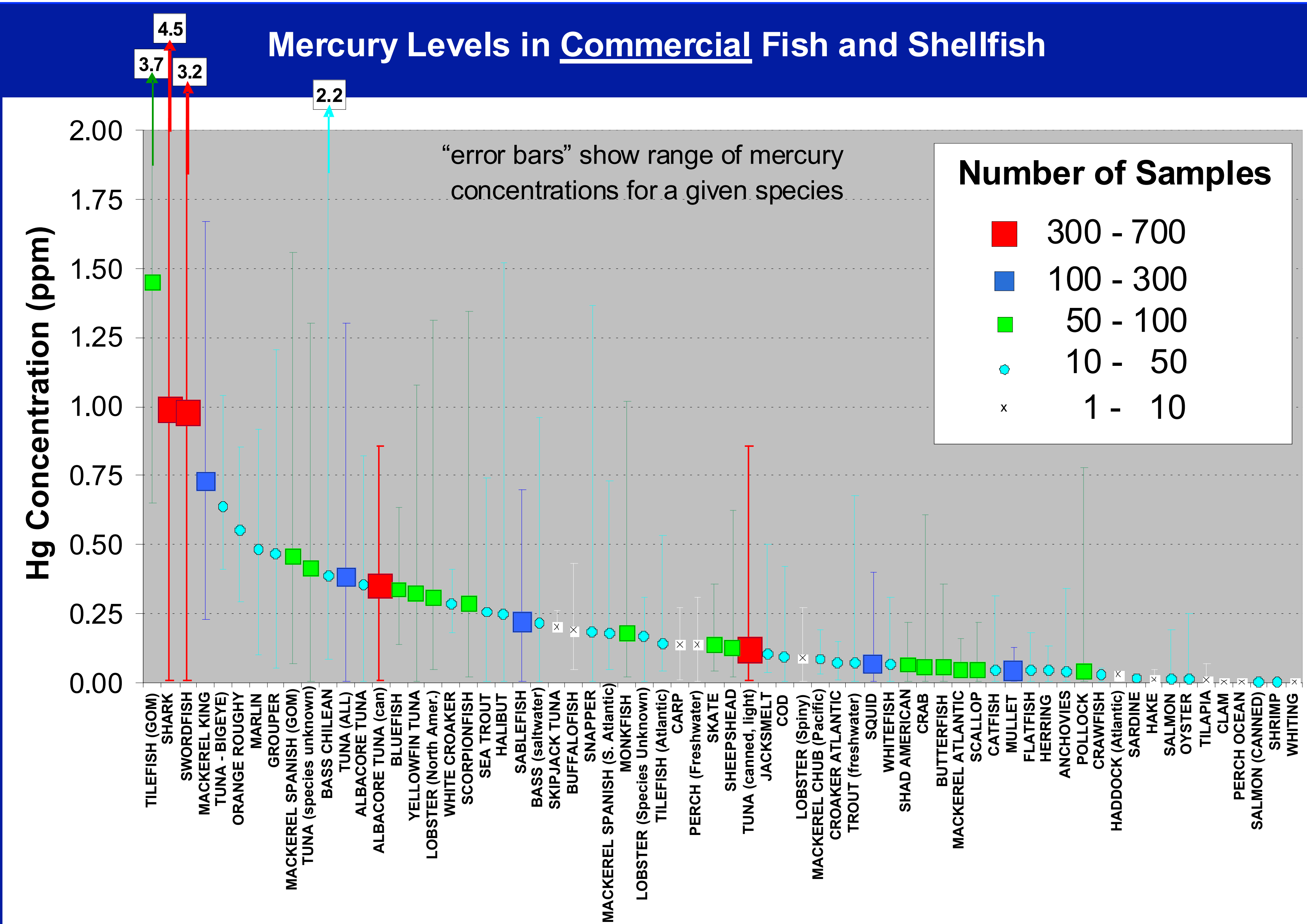
-Stochasticity (within species)
-Biological differences (between species)
-Anthropogenic impacts (geographic)
-Bioavailability (geographic)

FDA Reported National Hg Values



Source: FDA, 2007

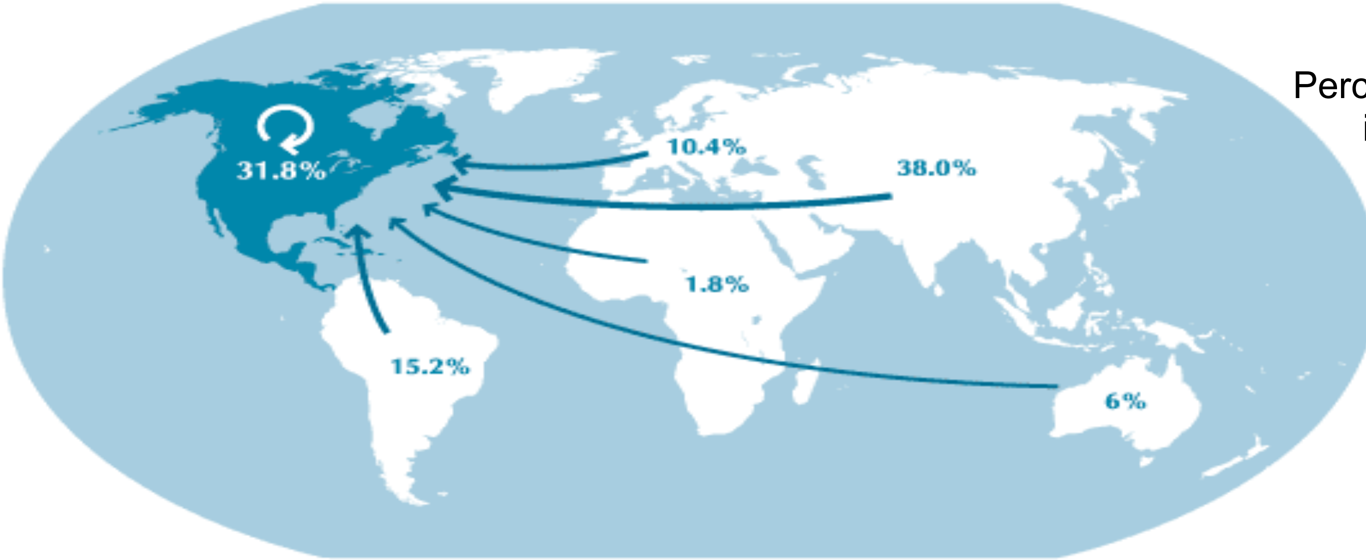
Mercury Levels in Commercial Fish and Shellfish



Fish concentration data from NOAA and FDA. Downloaded Sept 2008 from the EPA-FDA fish-mercury website: *http://www.cfsan.fda.gov/~frf/sea-mehg.html*

Where Do Fish Come From?

North America import flows



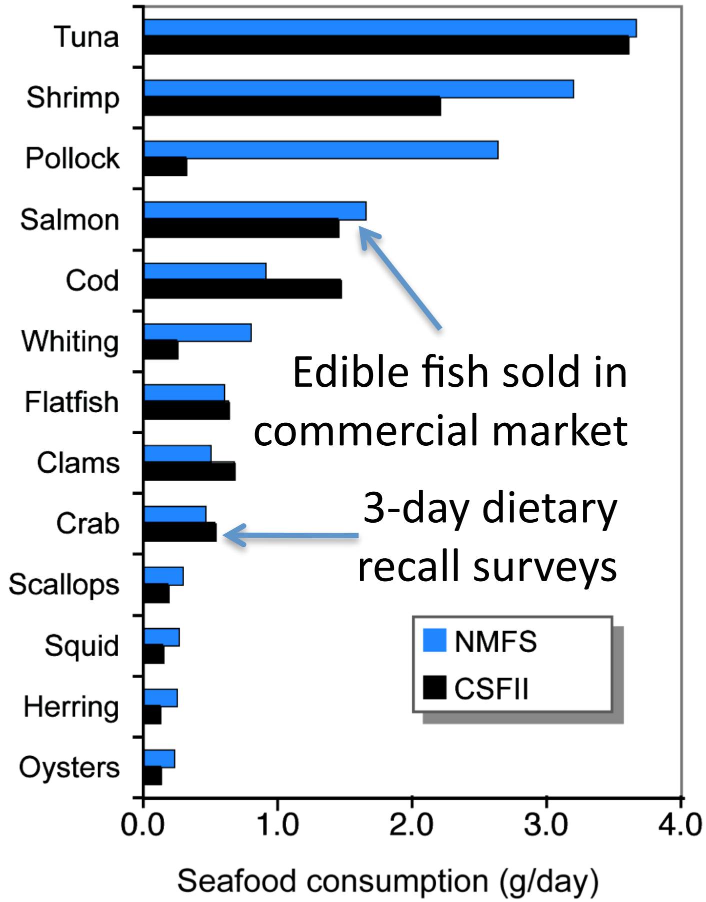
Percent of total imports

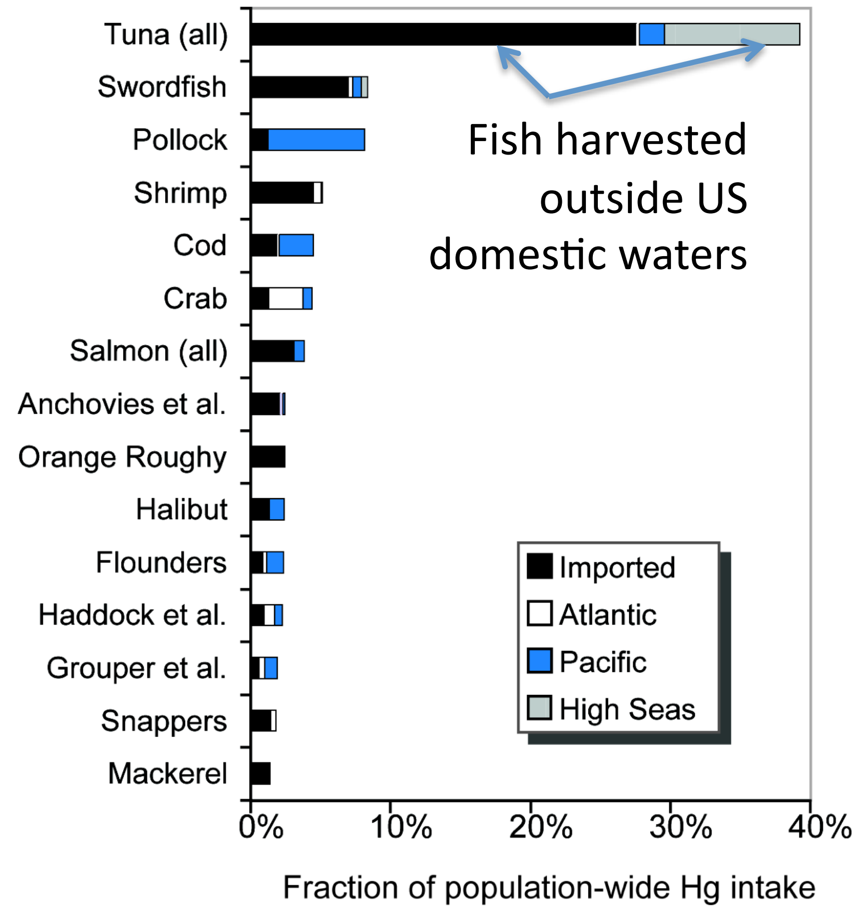
Notes: Average import flows for 1998-2000. Source: FAO State of World Fisheries and Aquaculture, 2002

- Commercial marine fish consumption is a major dietary source of Hg exposure in the U.S.

Where do fish people eat come from?

>90% of U.S. population-wide Hg exposure from consumption of estuarine & marine fish (Sunderland, 2007)





Majority of U.S. Population-Wide Hg Intake is from Harvests in Open Ocean Regions

~200 kg MeHg per year consumed in fish and shellfish

Harvest Location	% MeHg Intake
Fresh & Farmed	14.9%
Nearshore Marine	7.9%
North Atlantic	6.5%
Atlantic	14.7%
North Pacific	29.5%
Pacific/Indian	25.4%
Mediterranean	1.0%
Southern Ocean	0.1%
TOTAL	**100.0%**

77% offshore catch

Data Sources: Sunderland, 2007, NMFS 2000-2006, UNFAO 2000-2006

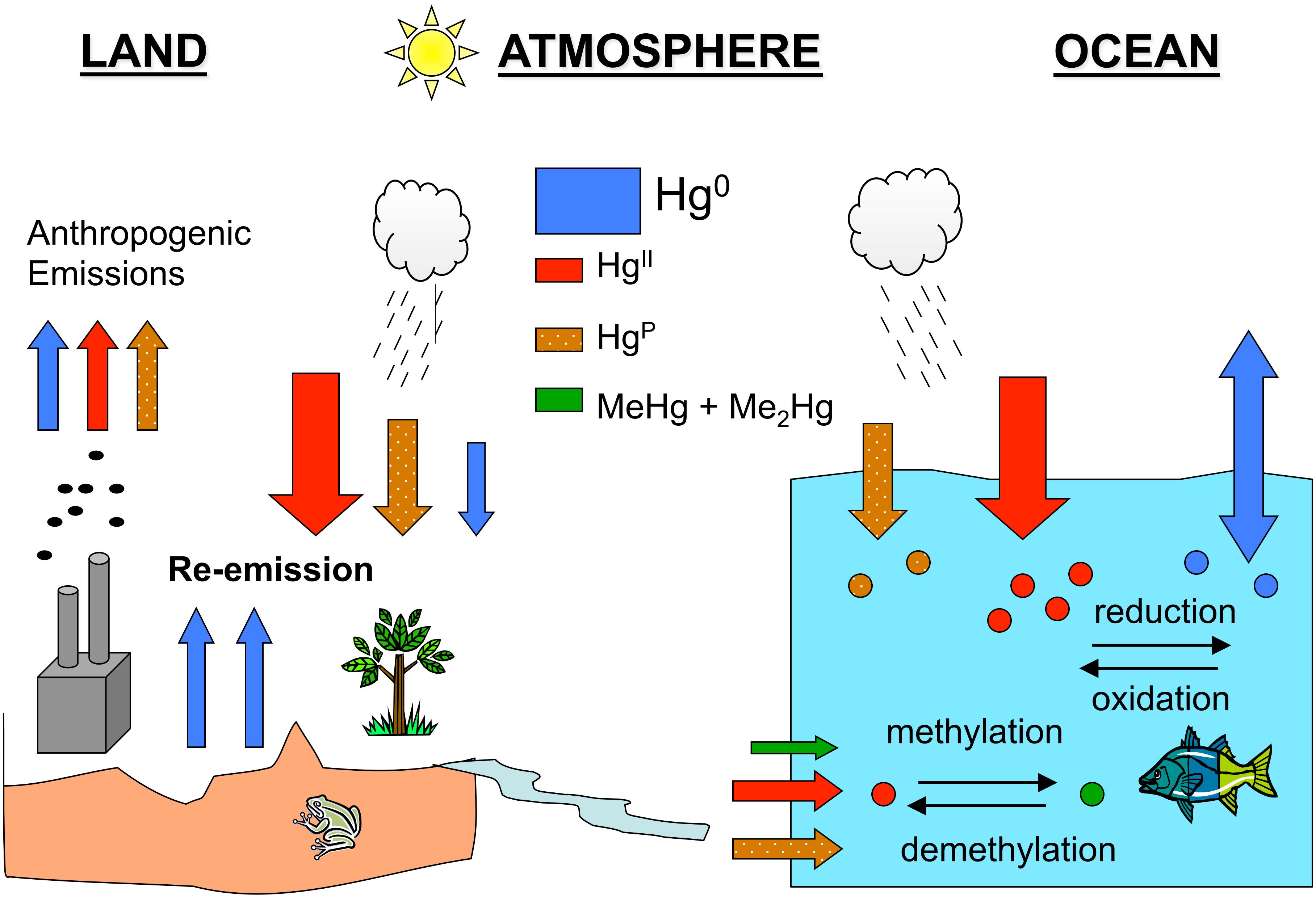
Interventions that can reduce human exposure to mercury:

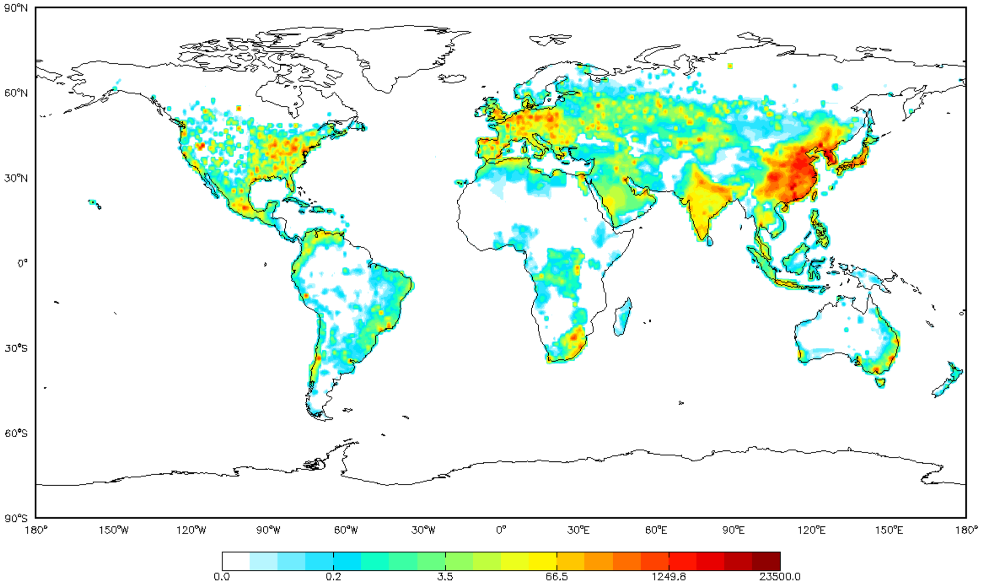
- Reduce levels in the environment (long-term)

- Dietary advisories on fish species selection and consumption rates (short-term)

Relevant Regulations in the US

- <u>*Clean Air Act*</u>
  - Emissions controls on anthropogenic sources
  - Effectiveness monitoring
- <u>*Clean Water Act*</u>
  - Fish Tissue Criterion to protect human health
  - (0.3 ppm tissue residue derived from RfD)
  - Load reductions of mercury from point and non-point sources

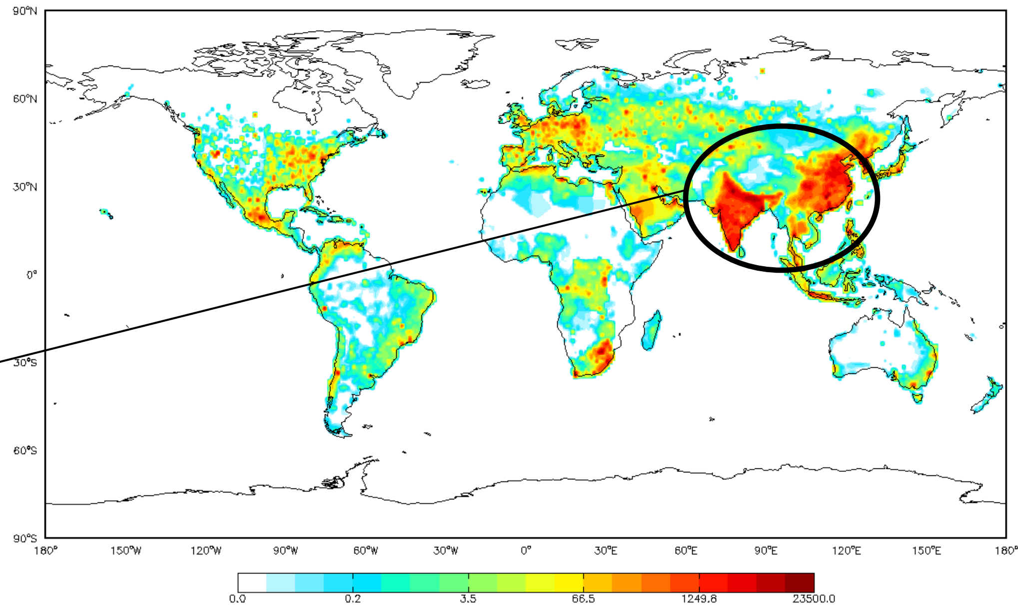

LAND
ATMOSPHERE
OCEAN
Hg^0
Hg^{II}
Hg^P
$MeHg + Me_2Hg$
Anthropogenic Emissions
Re-emission
reduction
oxidation
methylation
demethylation



2006 Anthropogenic Emissions

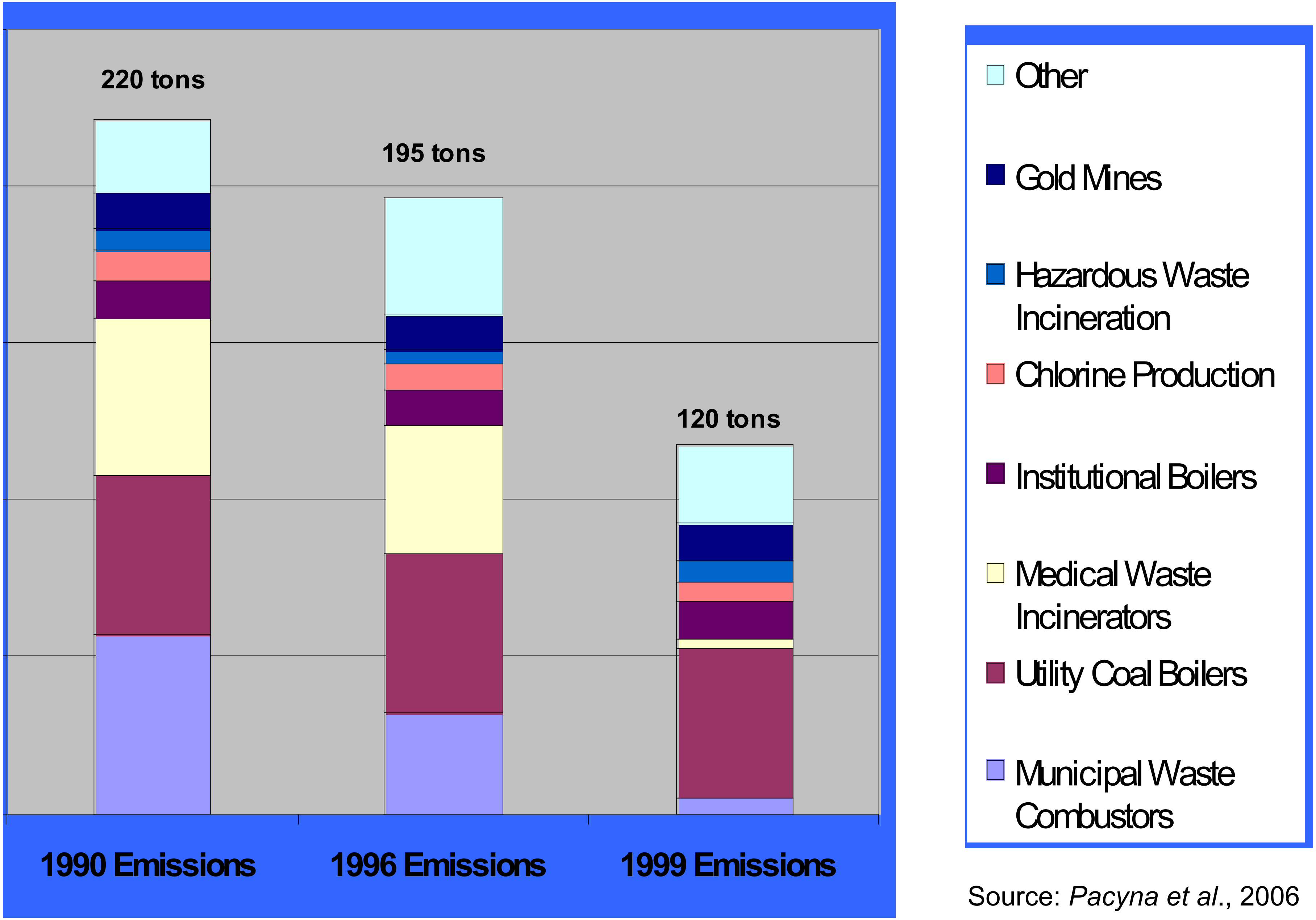
2050 Anthropogenic Emissions (A1B)

Growth of emissions in East Asia and India



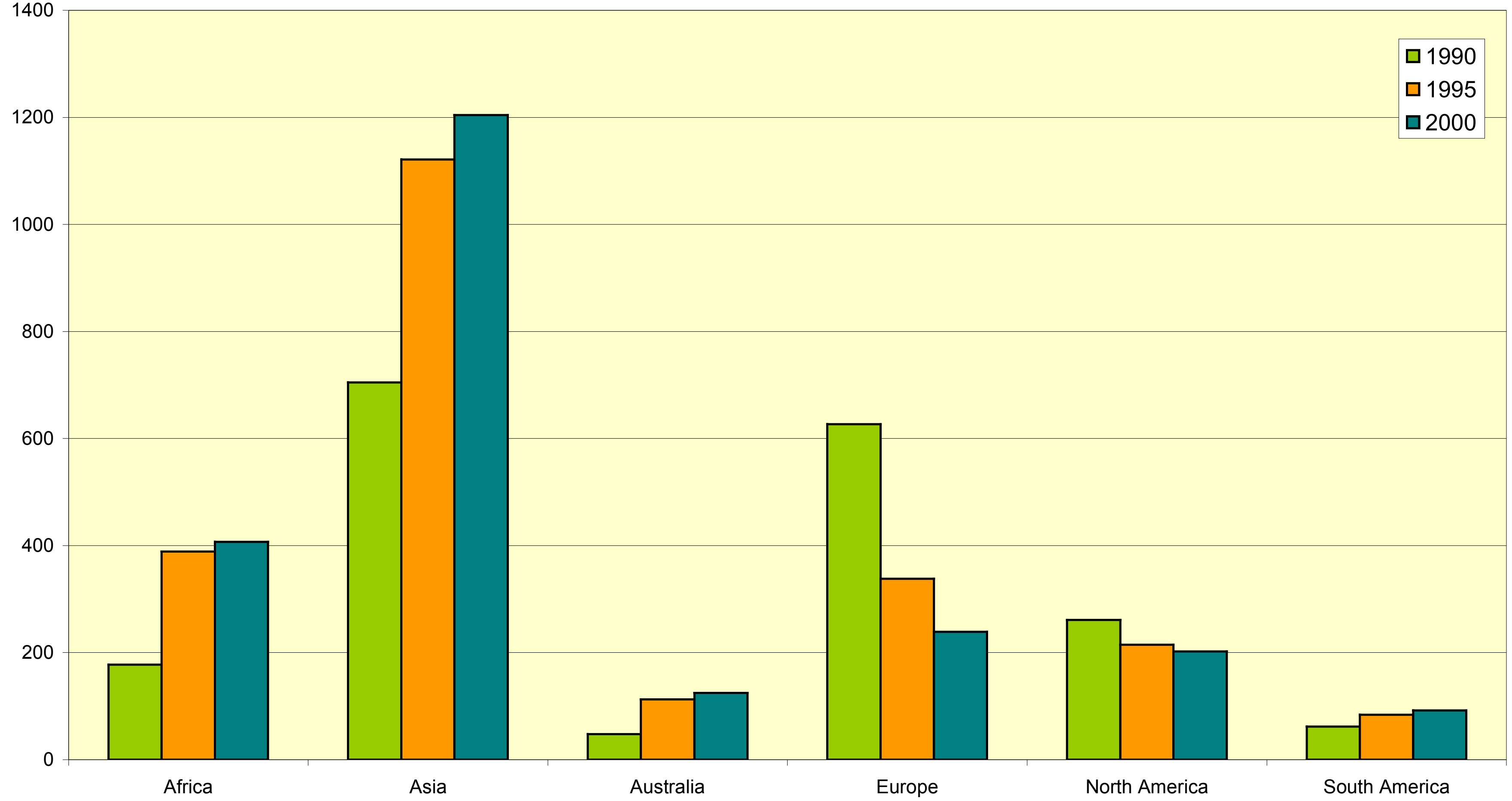
Corbitt et al., 2011

U.S. Mercury Emissions Have Dropped 45% Since 1990



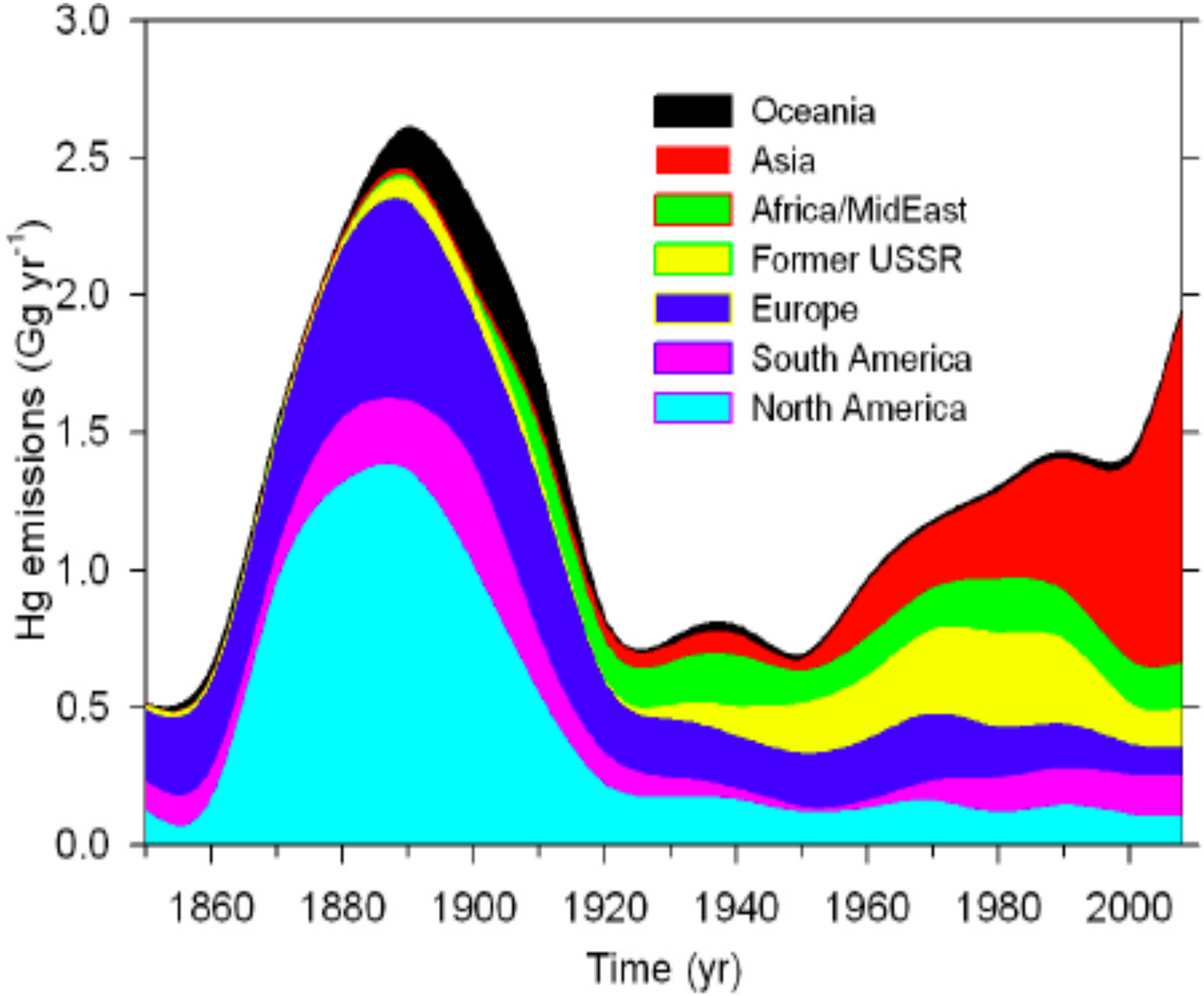
Source: *Pacyna et al*., 2006

Global anthropogenic Hg emissions 1990-2000 (tonnes per year)

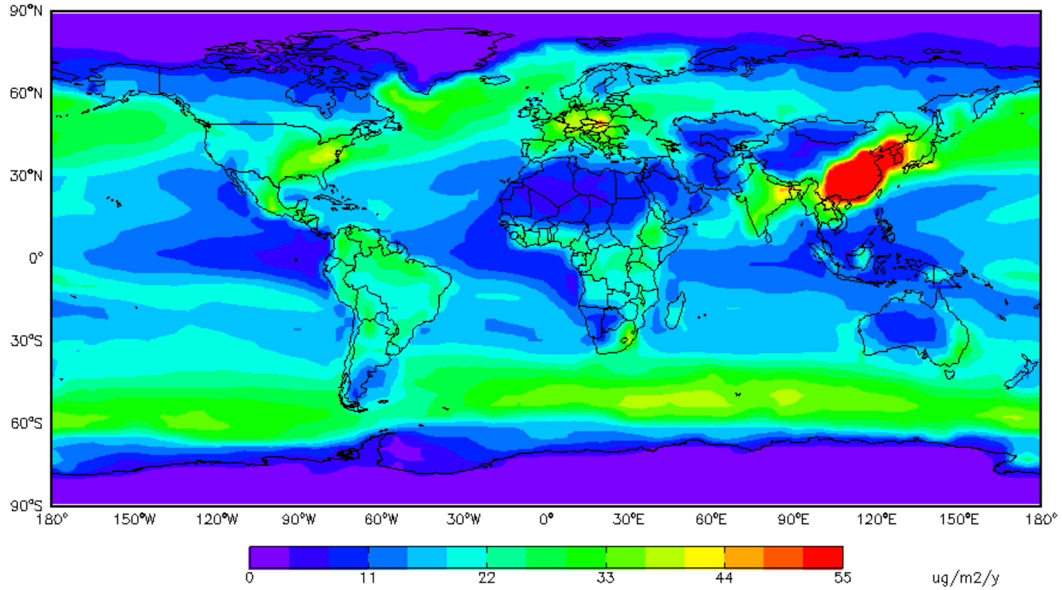


Source: Pacyna et al., 2006

Historical Hg Emissions by Region

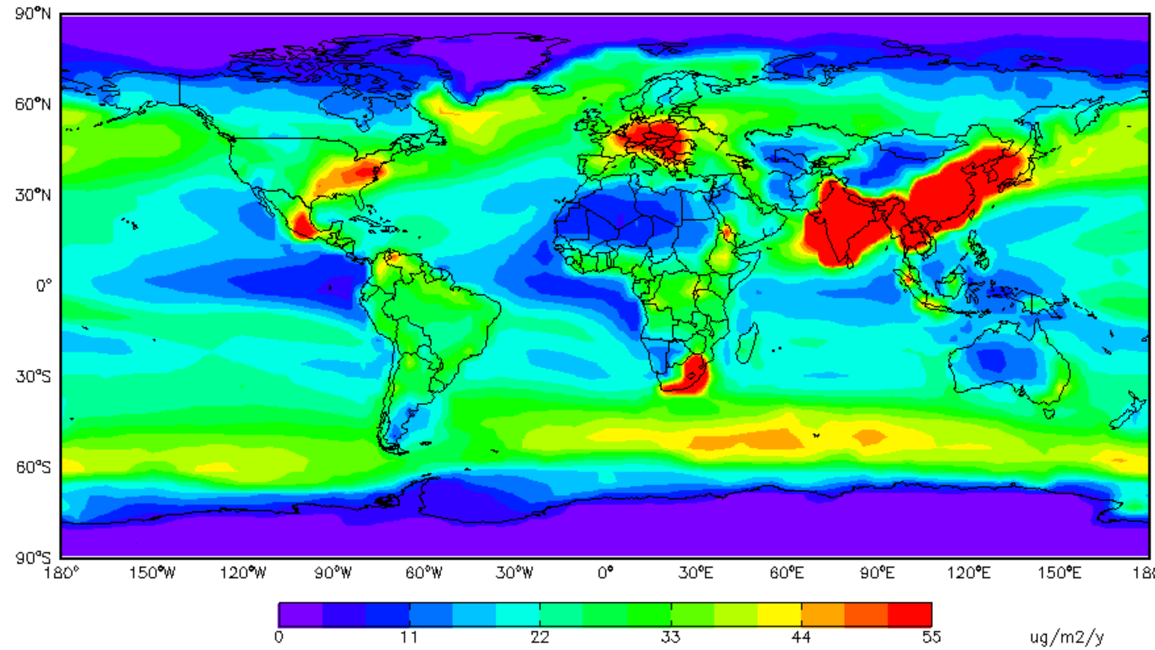


Streets et al. (2011, in review)



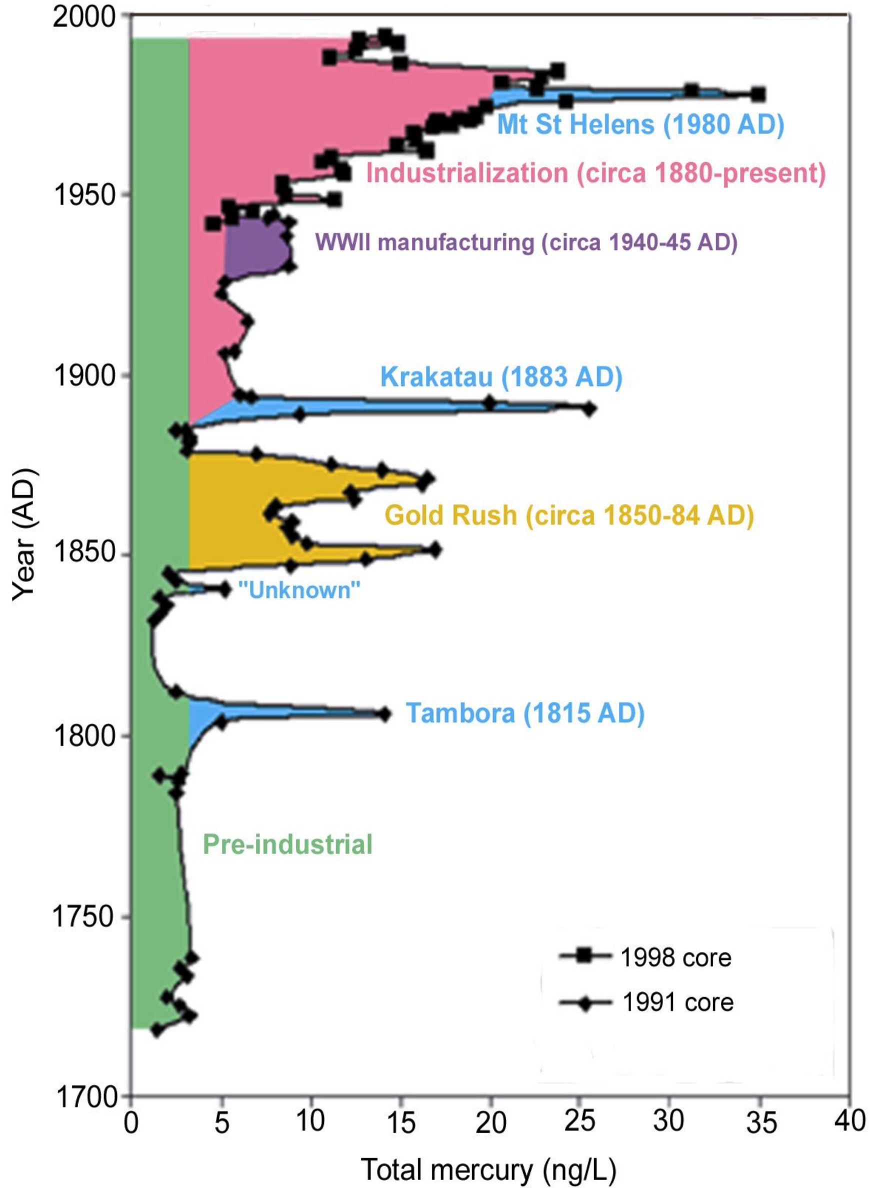
2006 Wet + Dry Hg Deposition

2050 A1B Wet + Dry Hg Deposition



Corbitt et al., 2011

Historic Hg Deposition from Natural and Human Sources from Ice Core Records

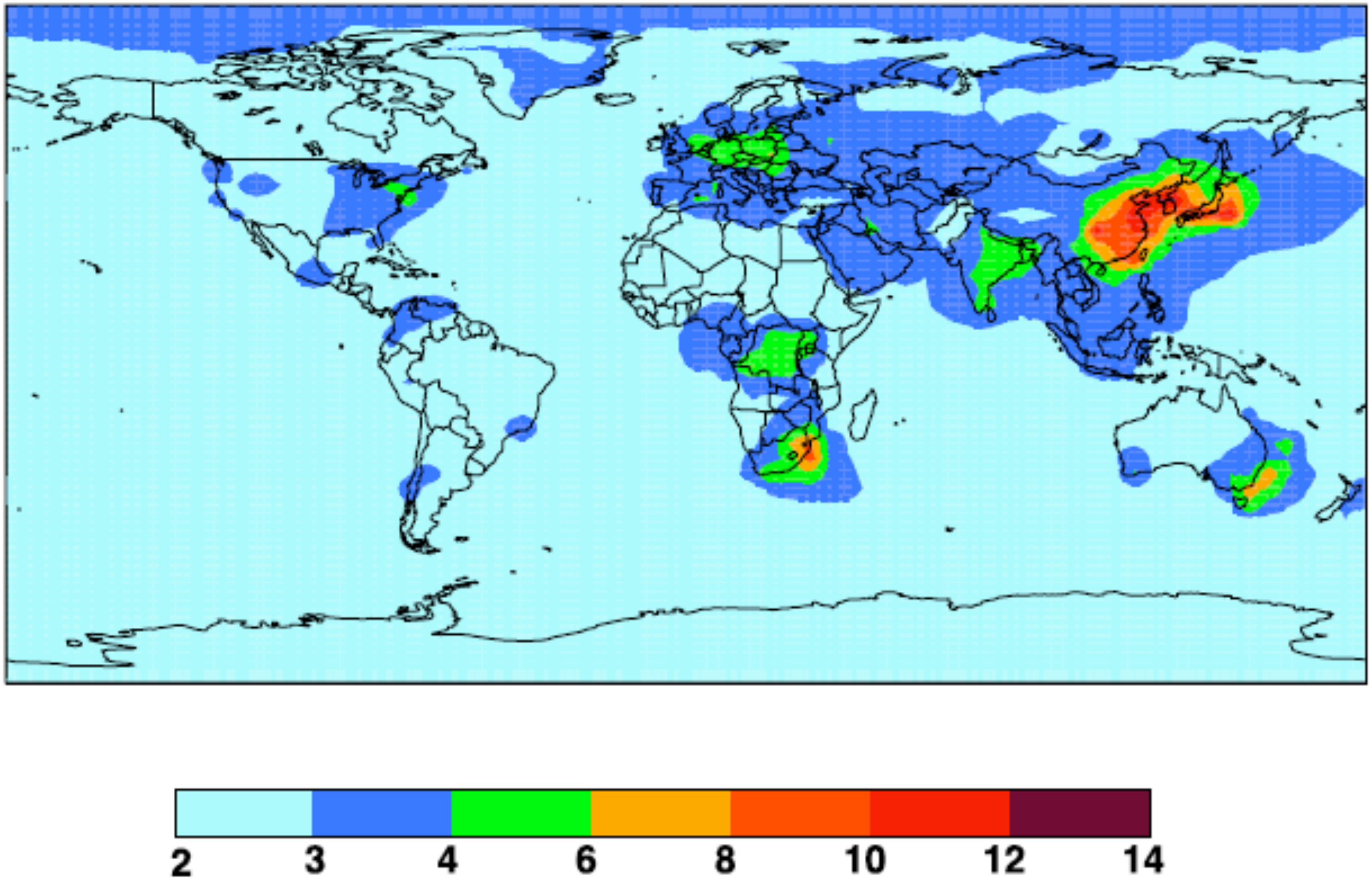


Lessons from the Fremont Glacier:

- **270-year record**
- **Large changes in mercury deposition**
- **Regional-to-global scale impacts from varying Hg sources.**
- **70% of Hg accumulation over the past 100 years resulting from man's activities**
- **Distinct decline last 10 years**

Source: Shuster et al., 2002

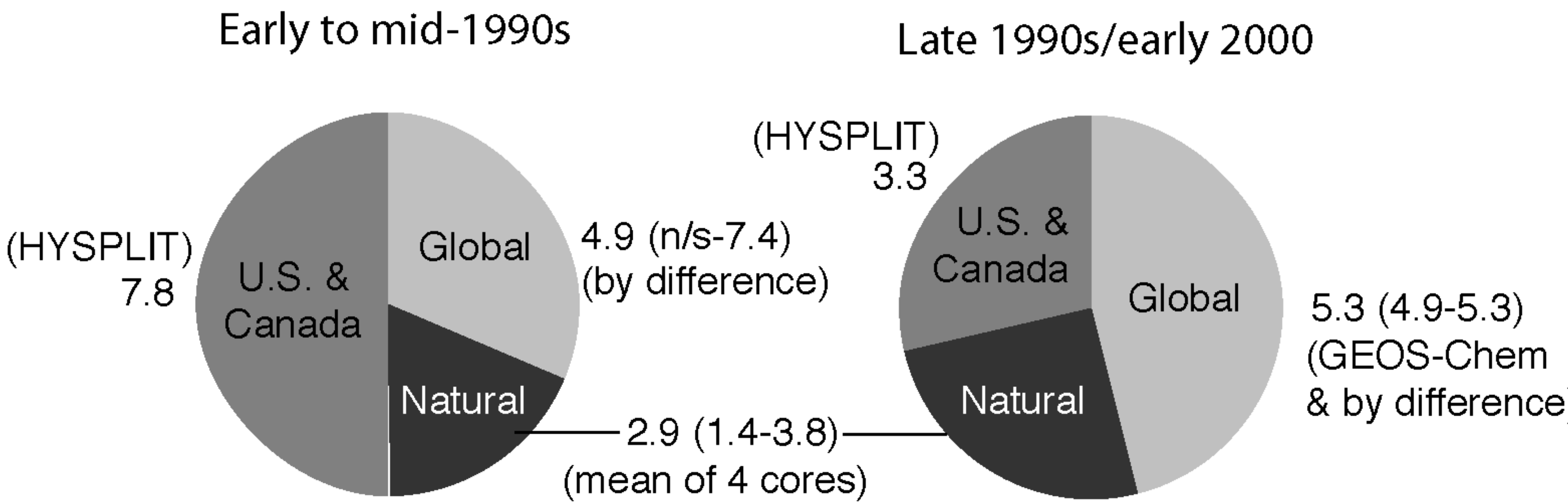
Anthropogenic Enrichment of Mercury Deposition



Source: *Selin et al., 2008, Global Biogeochemical Cycles*

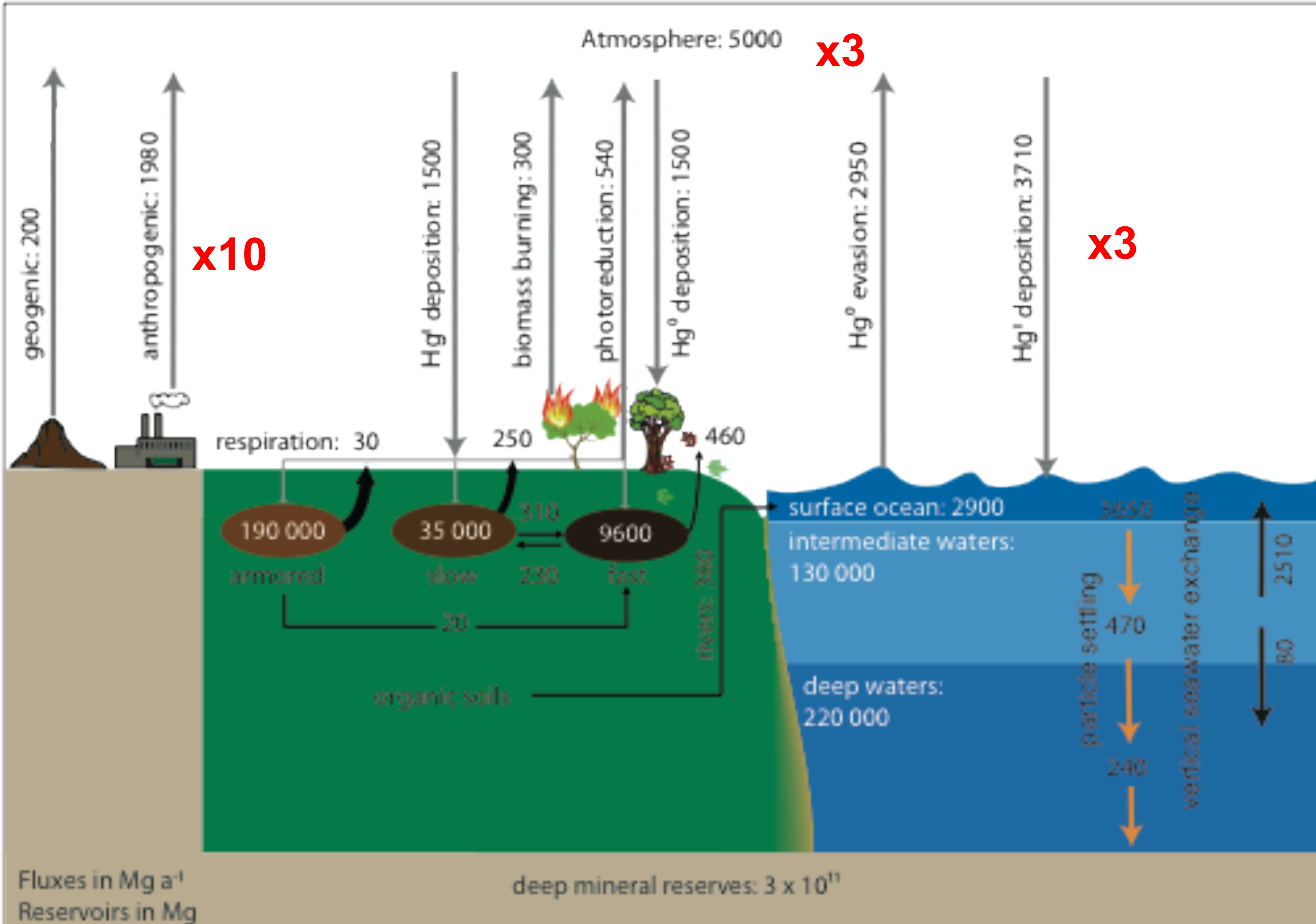
There are both local and global contributions to Hg inputs to ecosystems

Bay of Fundy/Gulf of Maine



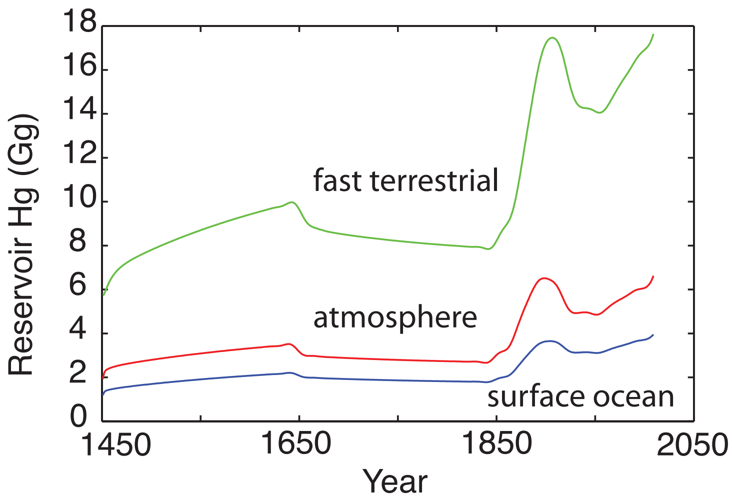
Source: *Sunderland et al.*, 2008, *Environmental Pollution*

PRESENT GLOBAL HG BUDGET



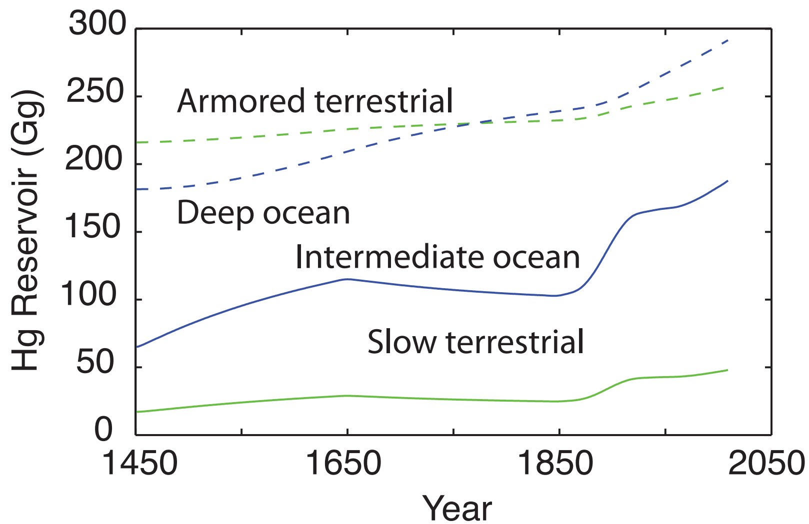
Anthropogenic Hg Accumulation 1450-present

Surface Reservoirs



Fast reservoirs closely track atmospheric Hg < 1yr to steady state

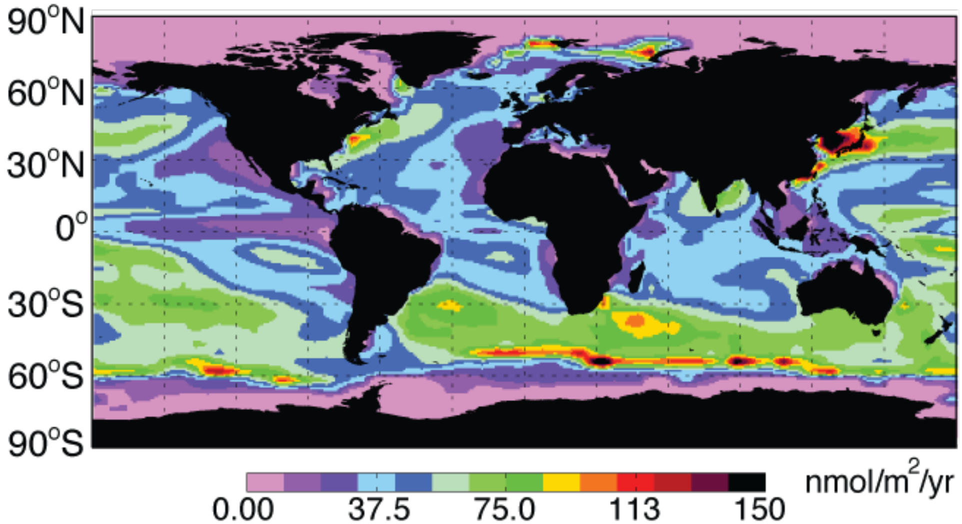
Slow Reservoirs



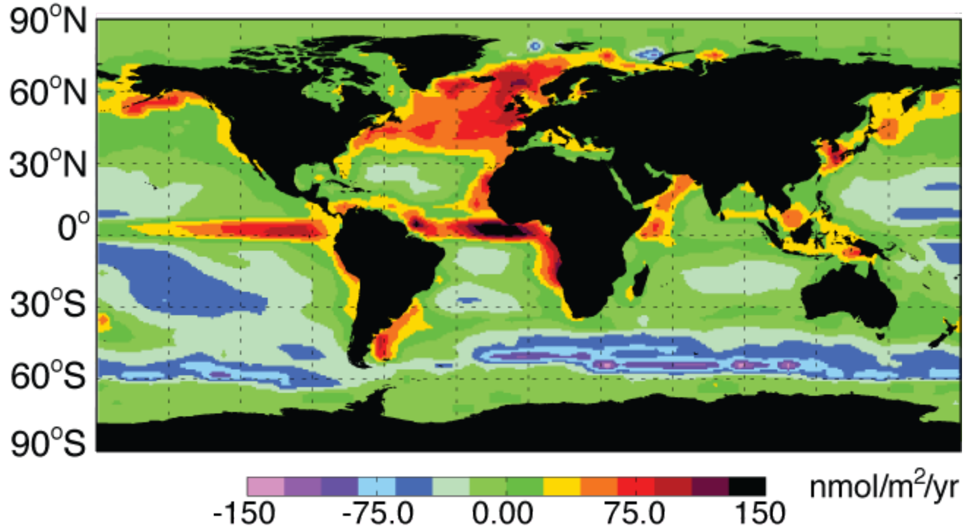
Intermediate and slow reservoirs still accumulating Hg.

Most anthropogenic Hg accumulating in deep ocean.

Ocean concentrations reflect both atmospheric deposition and historical inputs



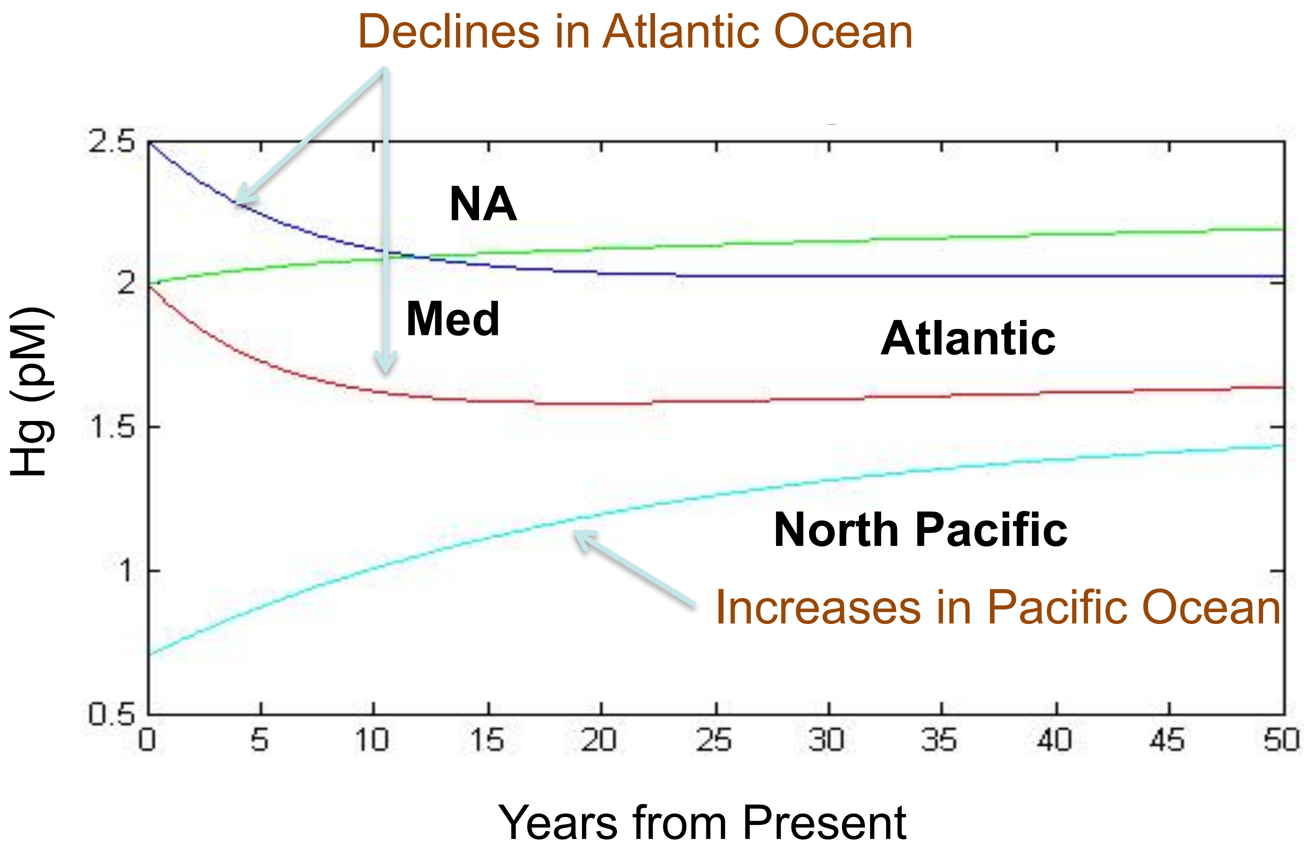
Atmospheric Deposition
60% Globally



Subsurface Ocean: Entrainment + Ekman Pumping
40% Globally

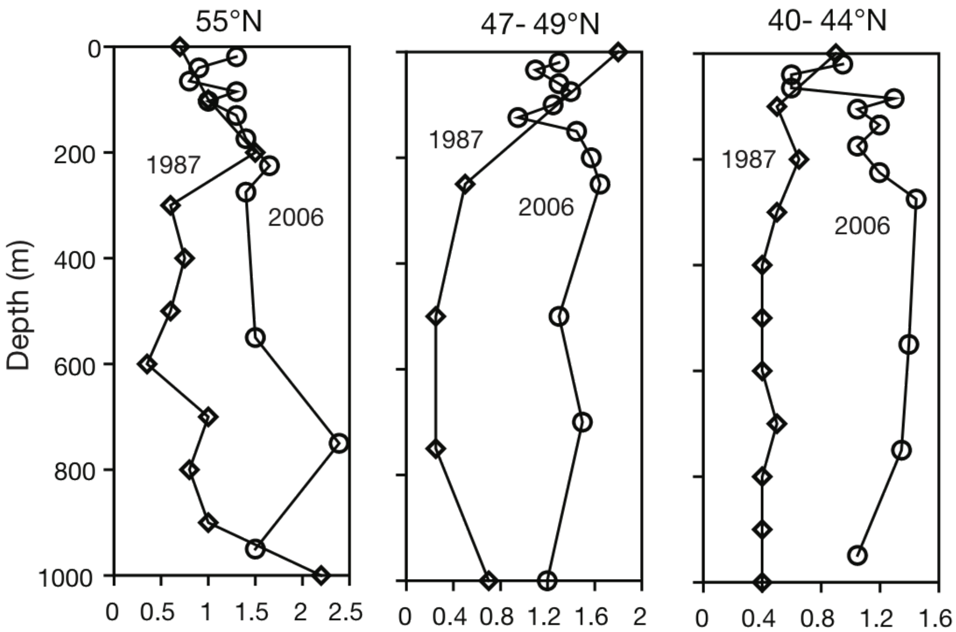
Modeled 50 yr Seawater Hg Trends

No Change in Present-Day Hg emissions



Modeled trends are consistent with limited observations for Atlantic & Pacific

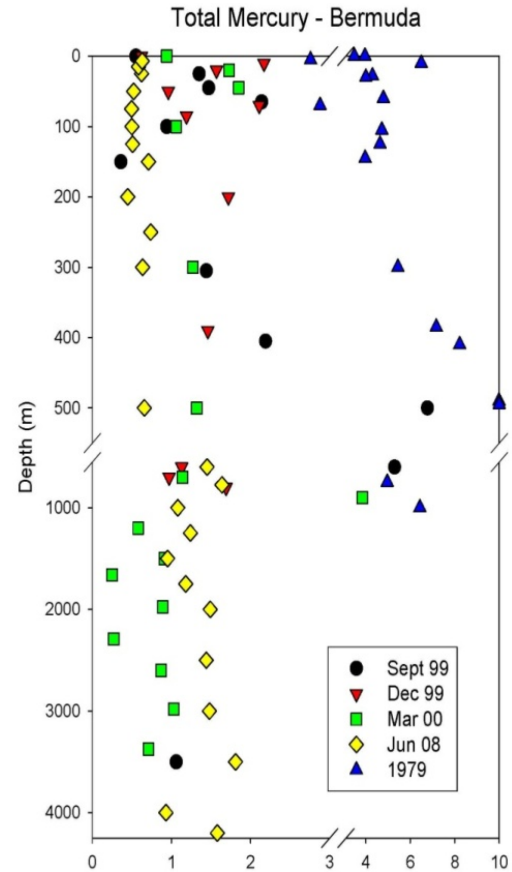
North Pacific Ocean



Hg (pM) Seawater

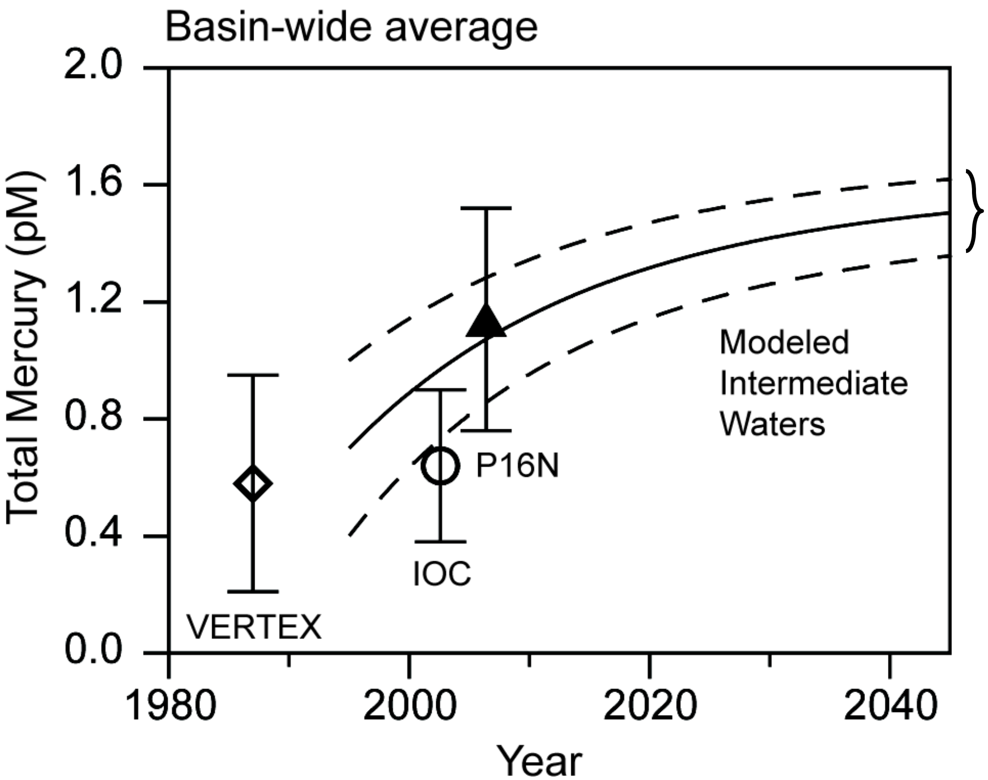
Sunderland et al., 2009

Atlantic Ocean



Mason, pers. comm.

Temporal Trends in Hg – North Pacific Ocean



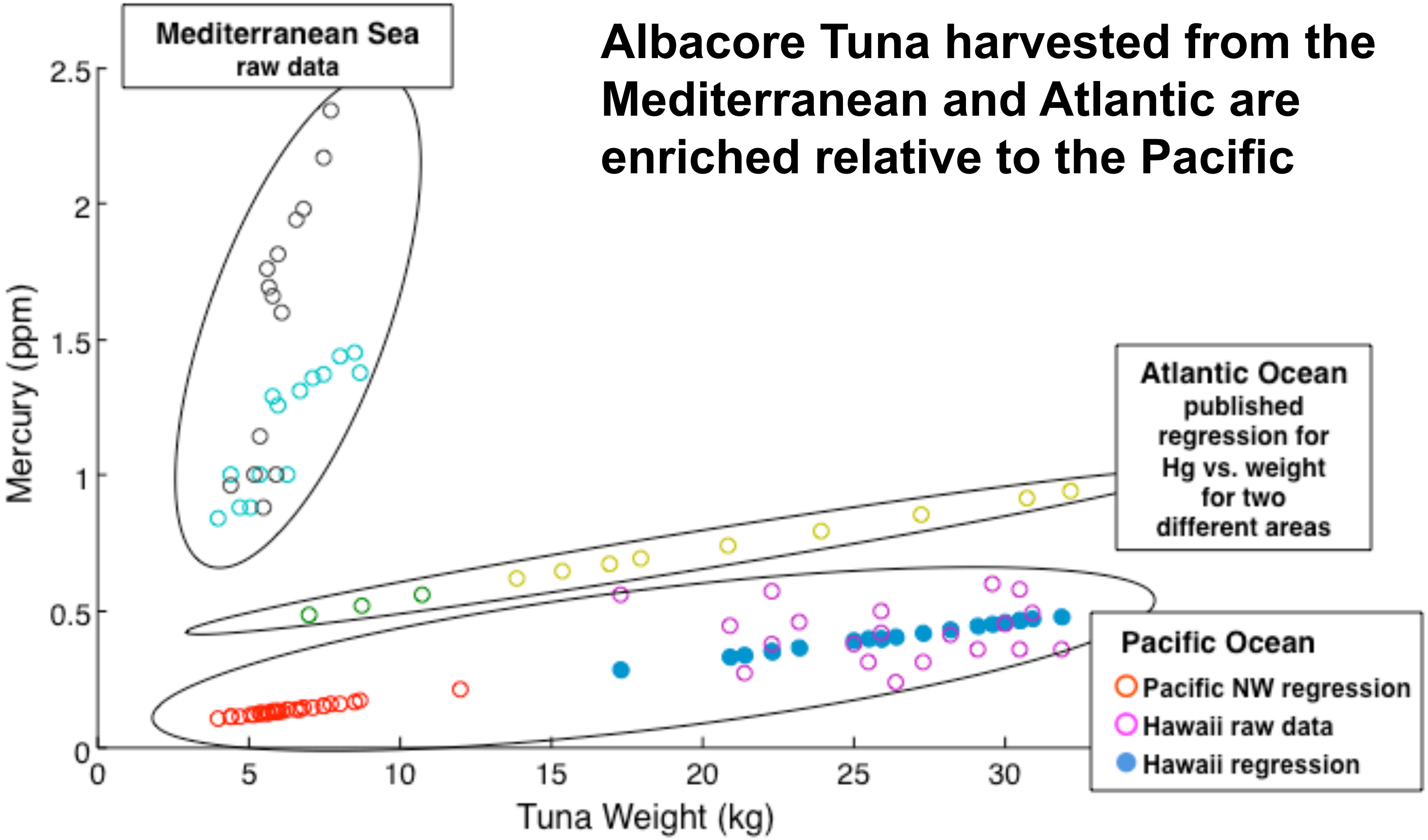
95% Confidence Interval for model results based on empirically constrained fluxes

At present atmospheric Hg deposition rates, North Pacific seawater Hg may double relative to ca. 1995 by 2050

Source: Sunderland et al., 2009, Global Biogeochem. Cy.

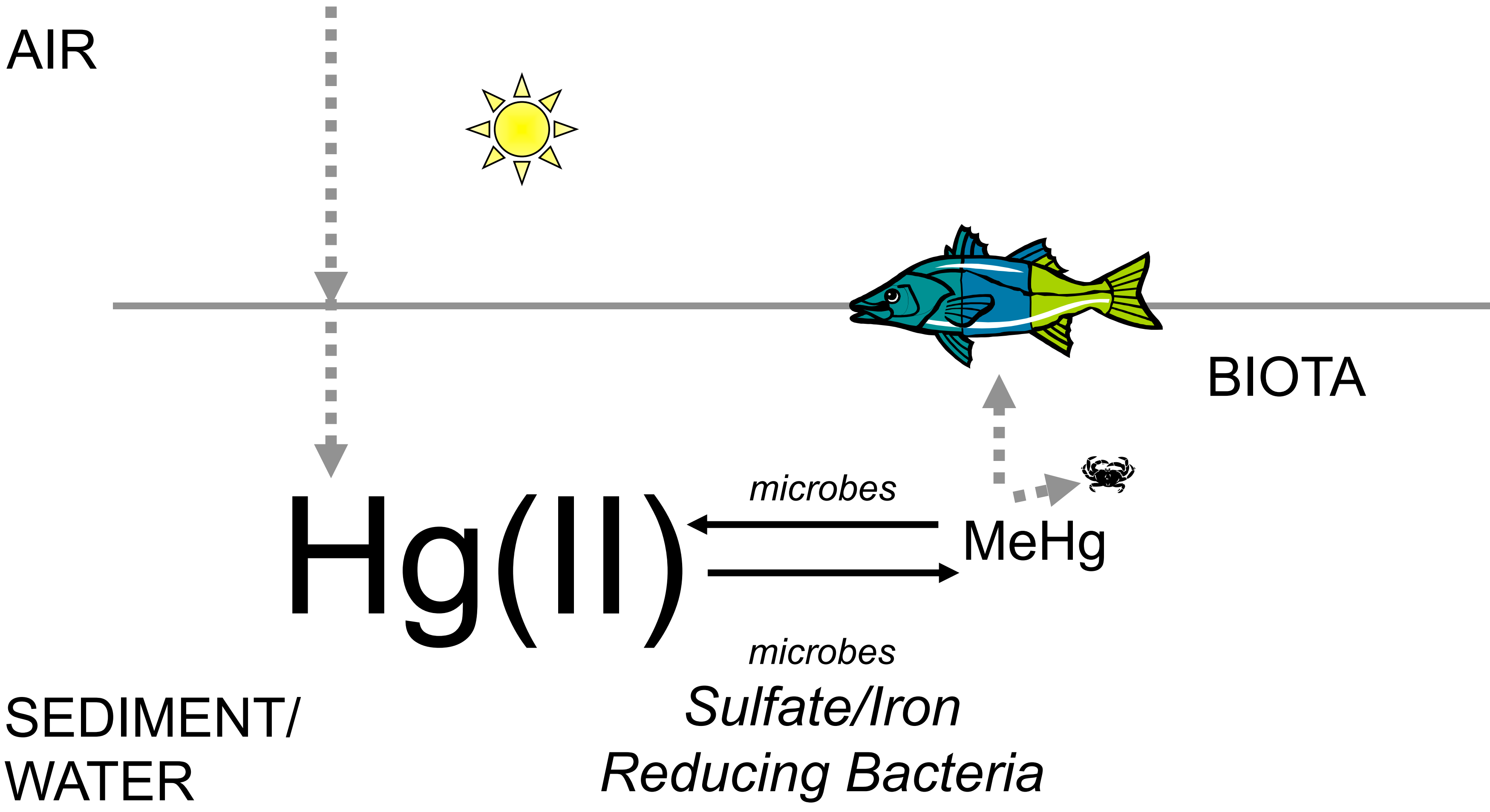
Link between Hg in Seawater and Fish

Albacore Tuna harvested from the Mediterranean and Atlantic are enriched relative to the Pacific



Tuna Hg data from the following sources: http://www.atuna.com, Brooks, 2004 (Hawaii); Storelli et al., 2002 (Ionian Sea); Storelli & Marcotrigiano, 2004 (Adriatic Sea); Morrisey et al., 2004 (Pacific NW, USA). Atlantic: Anderson and Depledge, (1997).

The Form or "Species" of Mercury is Important

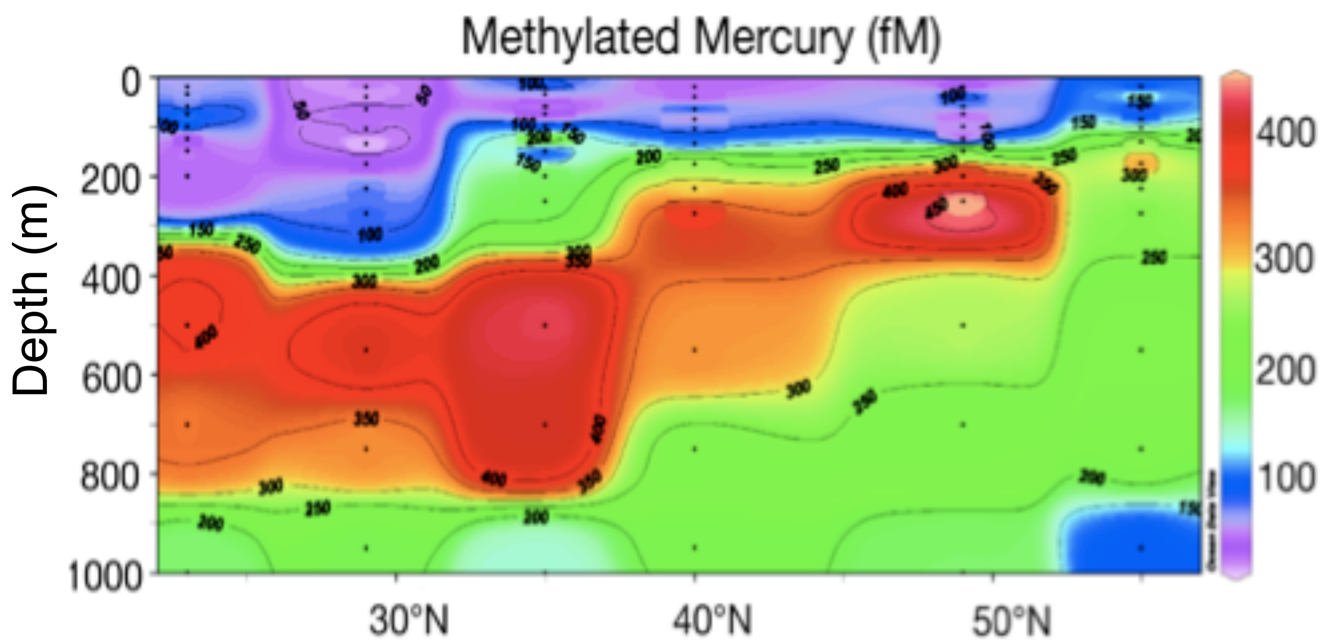


Biological Hg Burdens Driven by MeHg

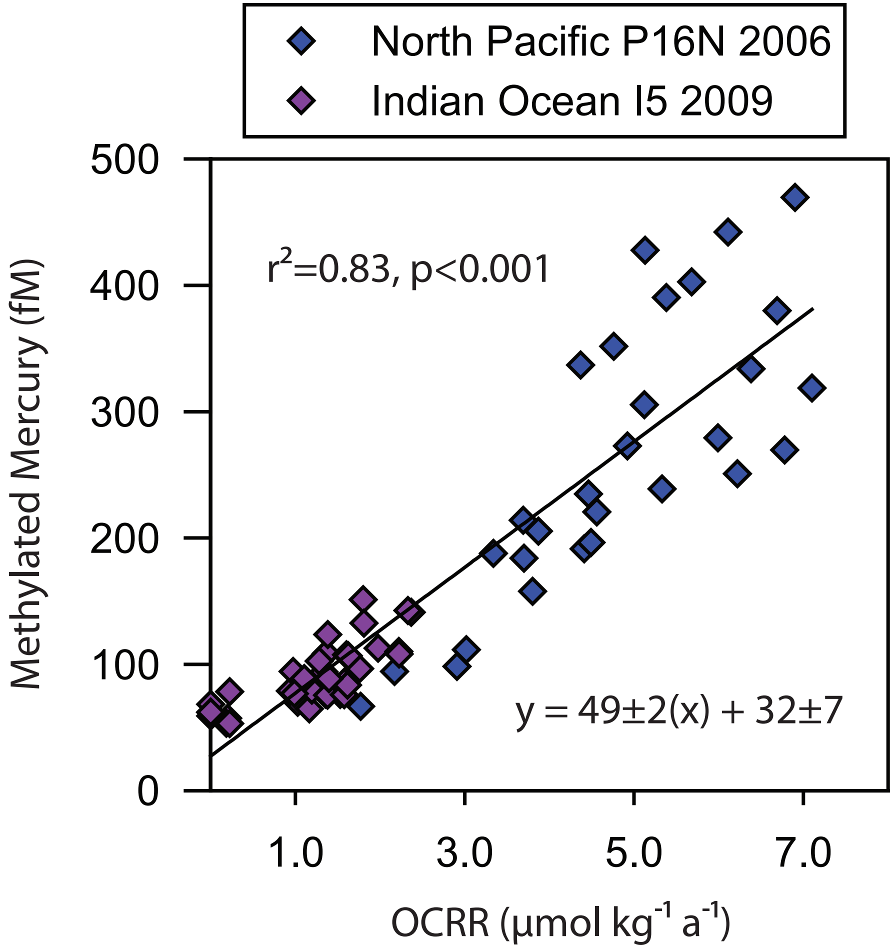
Phytoplankton | Zooplankton | Planktivorous fish | Piscivorous fish

Bf ~ 10^4 | Bf ~ 10^5 | Bf ~ 10^6 | Bf = 10^6–10^7

North Pacific Ocean



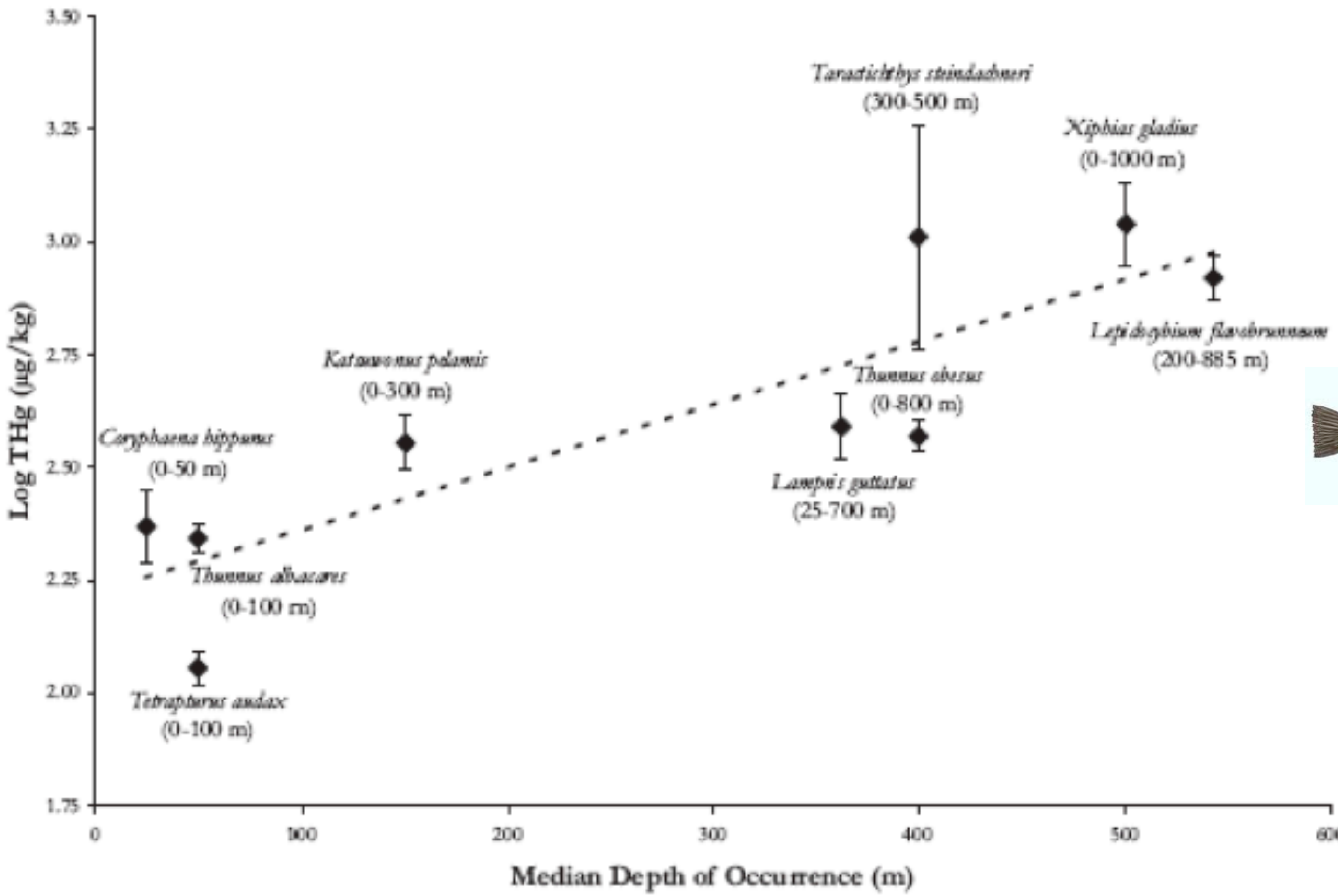
Sunderland et al., 2009

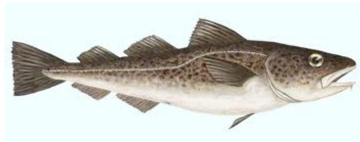


Highly significant relationship observed between methylated Hg and organic carbon remineralization rates (OCRR) in intermediate ocean waters

OCRR = ΔC_{org}/AGE = ($R_{C:O}$*AOU)/AGE (*Feely et al.*, 2004)

Fish Hg levels are correlated with predominant feeding & swimming depths





Choy et al., 2009, PNAS

Conclusions

1. Models of the physical environment are essential for accurately characterizing exposures for risk analysis

2. Risk management must consider timescales of potential changes in exposures (e.g., advisories vs. emissions reductions)

3. Additional data needed on harvesting regions and types of fish consumed by high-risk groups

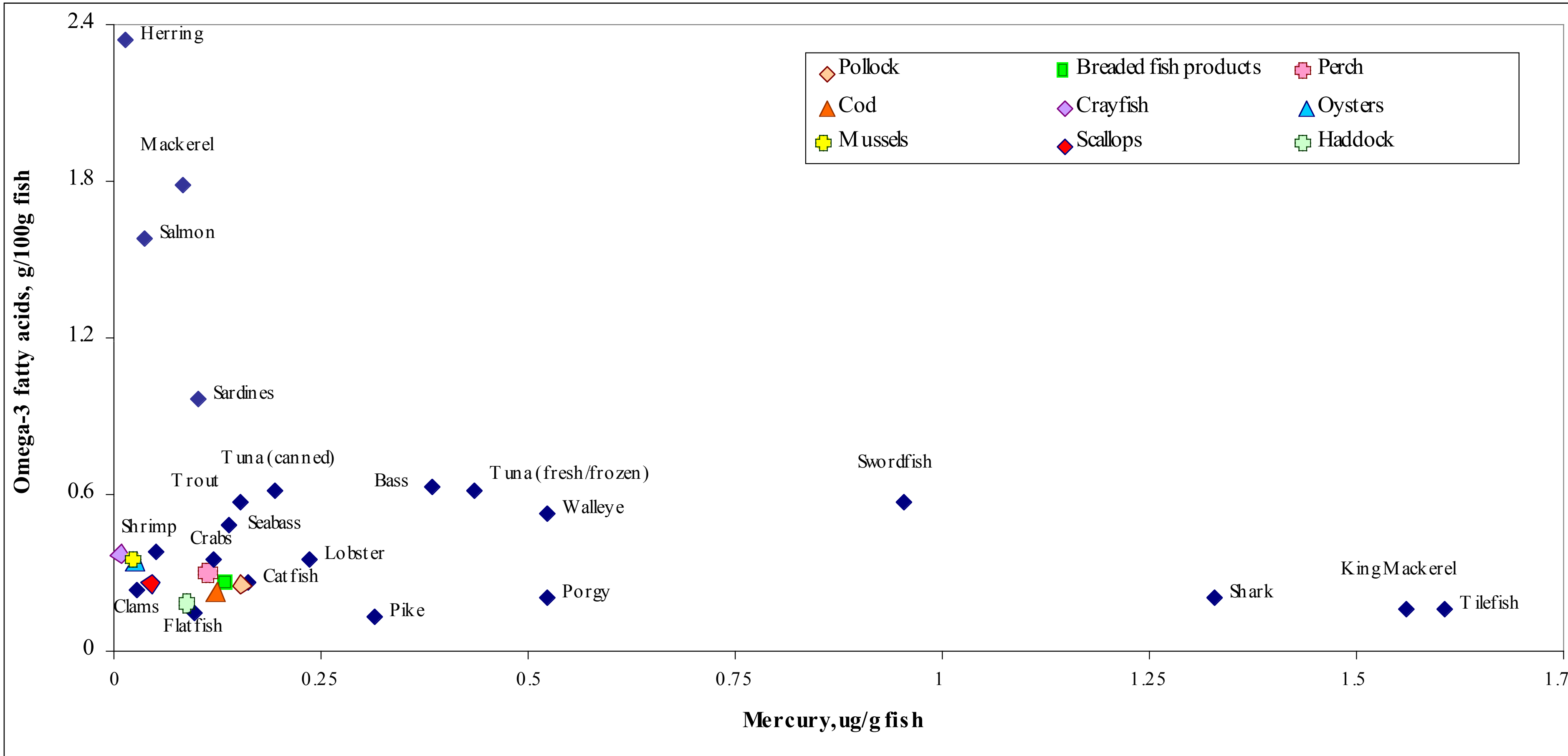
Lead Article

Balancing the benefits of n-3 polyunsaturated fatty acids and the risks of methylmercury exposure from fish consumption

Kathryn R Mahaffey, Elsie M Sunderland, Hing Man Chan, Anna L Choi, Philippe Grandjean, Koenraad Mariën, Emily Oken, Mineshi Sakamoto, Rita Schoeny, Pál Weihe, Chong-Huai Yan, and Akira Yasutake

Fish and shellfish are widely available foods that provide important nutrients, particularly n-3 polyunsaturated fatty acids (n-3 PUFAs), to many populations globally. These nutrients, especially docosahexaenoic acid, confer benefits to brain and visual system development in infants and reduce risks of certain forms of heart disease in adults. However, fish and shellfish can also be a major source of methylmercury (MeHg), a known neurotoxicant that is particularly harmful to fetal brain development. This review documents the latest knowledge on the risks and benefits of seafood consumption for perinatal development of infants. It is possible to choose fish species that are both high in n-3 PUFAs and low in MeHg. A framework for providing dietary advice for women of childbearing age on how to maximize the dietary intake of n-3 PUFAs while minimizing MeHg exposures is suggested.

Concentrations of Mercury and Omega-3 Fatty Acids (EPA + DHA) in Selected Fish and Shellfish Species



Source: Mahaffey et al., 2008, Environmental Research