



A Geochemical Perspective on Methylmercury Exposures and Health Risks

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Why are we worried about Hg in the environment?

WELL

Should Pregnant Women Eat More Tuna?

By TARA PARKER-POPE MARCH 2, 2015 3:55 PM 73 Comments

Getty Images

Home > Consumer Reports magazine > 2014 > October > Special report: Can eating the wrong fish put you at higher risk

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Special report: Can eating the wrong fish put you at higher risk for mercury exposure?

The government wants you to eat more seafood. The key to limiting your risk is choosing the right fish.

Published: August 2014

Methylmercury is a bioaccumulative neurotoxin

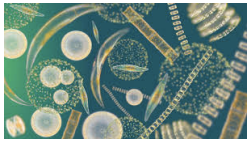
Concentrations are x10^6-10^7 water

top predators

- Neurocognitive deficits
- Impaired motor function
- Cardiovascular effects

big fish

small fish



plankton

water

mercury concentration

Global and National Strategies for Reducing Anthropogenic Hg Emissions

Minamata Convention

- First global agreement on anthropogenic Hg reductions established in 2013
- 128 Signatures, 11 Ratifications (May, 2015)

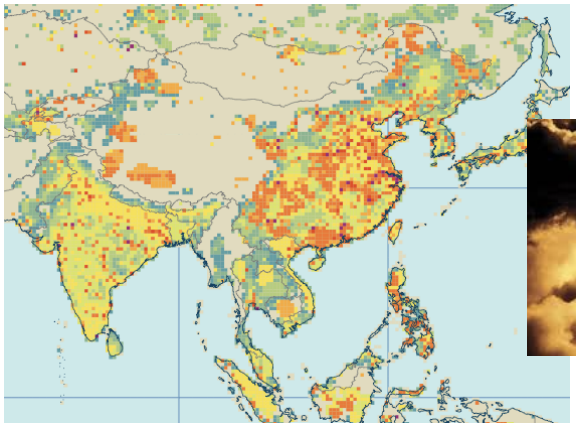
UNEP

Today's Discussion

- Global biogeochemical Hg cycle
- Anthropogenic Hg sources & trends
- Future trajectories in ocean Hg concentrations
- Use of Hg stable isotopes to better understand human exposures and risks

State-of-the-science understanding ca. 2010

Focused on present day and future



Enrichment

Atmospheric deposition increased 3x (2-5x) since 1850

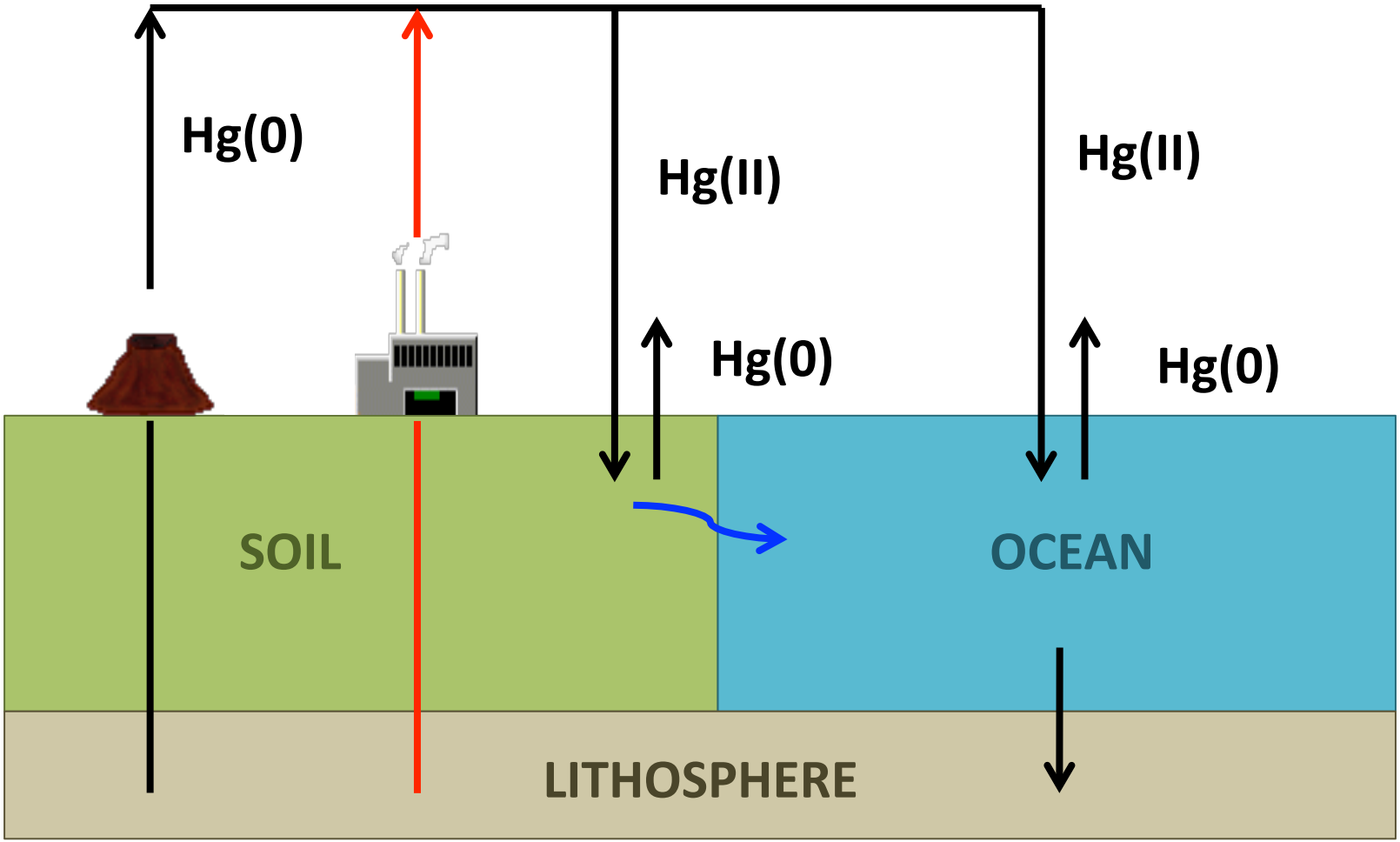
Isolated systems

atmosphere

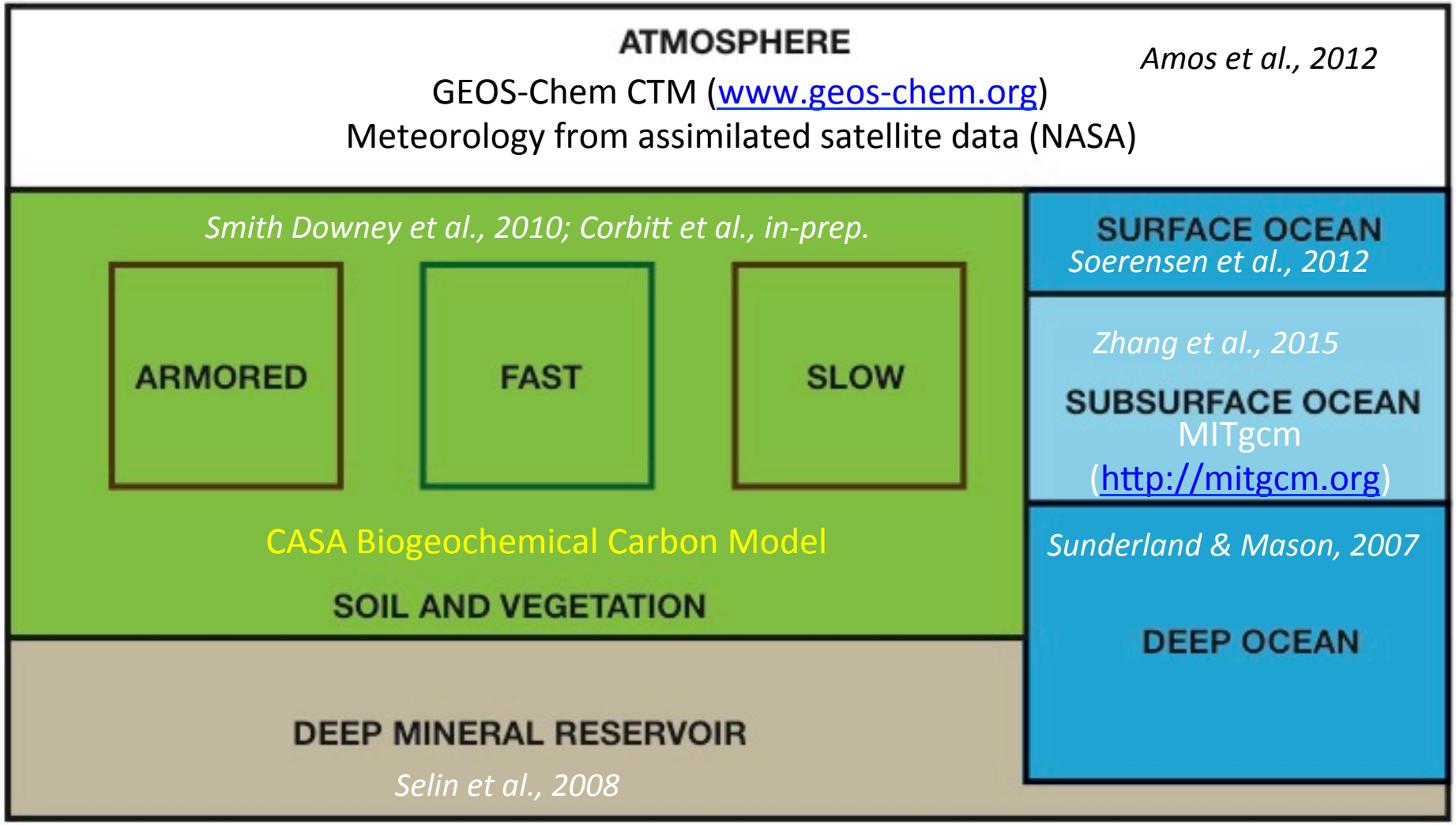
land

ocean

The global biogeochemical cycle of mercury



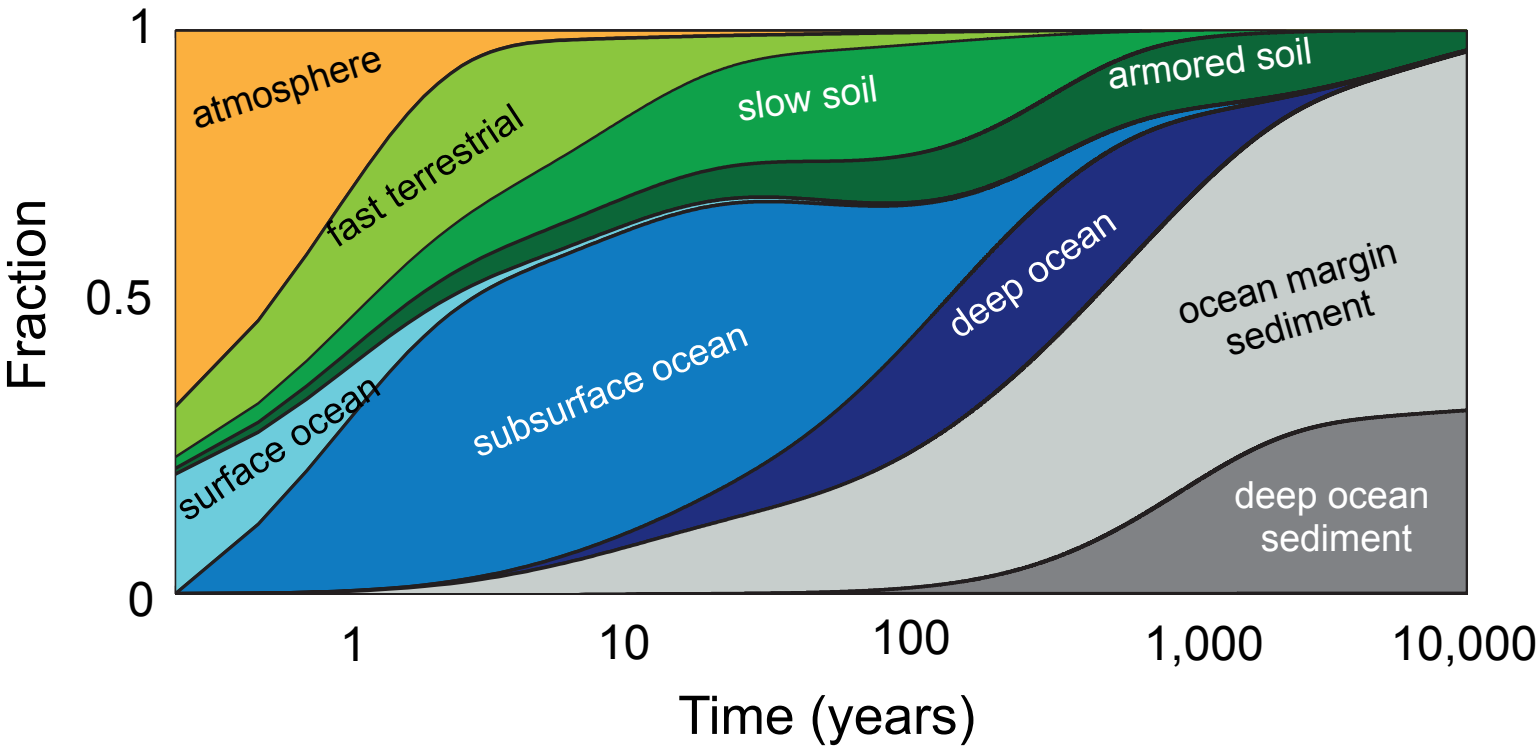
Toolbox of models for capturing global cycling



Fully coupled biogeochemical box model: Amos *et al*., 2013

Perturbation analysis reveals timescales of couplings between global reservoirs

Fate of a unit pulse of Hg to the atmosphere



Amos et al. (2013), *GBC*; Amos et al. (2014), *ES&T*

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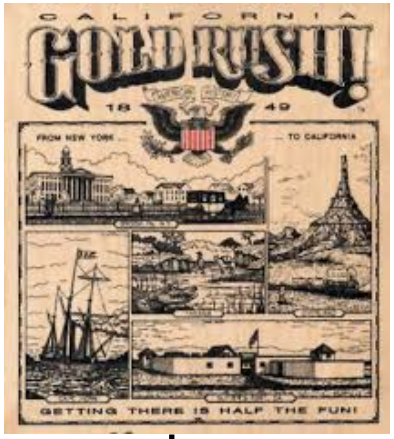
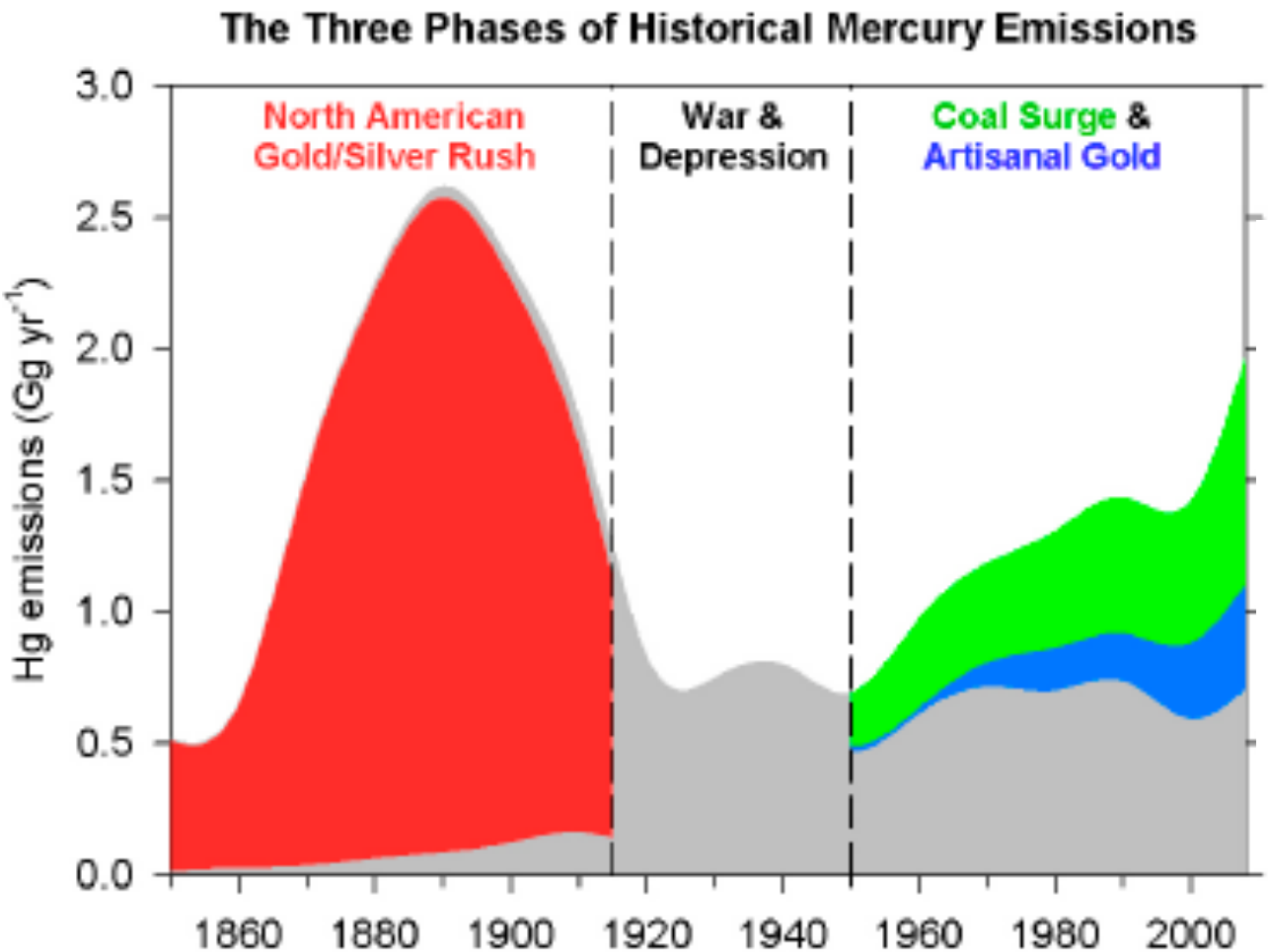
We've been using mercury since antiquity



2000 B.C.

Today

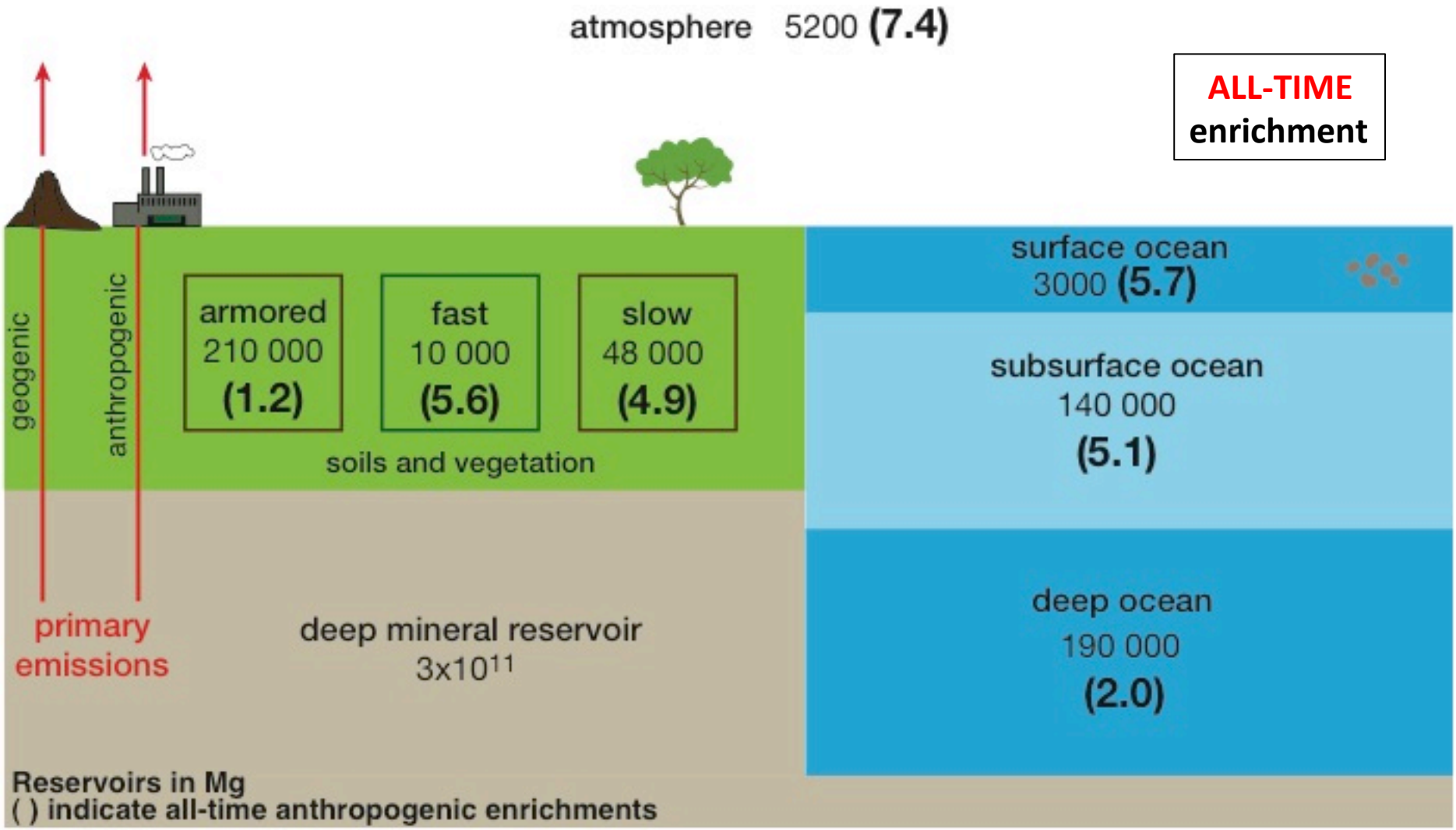
All-Time Historical Atmospheric Hg Releases



- 350 Gg total release
- 61% 1850-present
- 39% prior to 1850

Streets et al. , 2011

All biogeochemical reservoirs have experienced anthropogenic enrichment

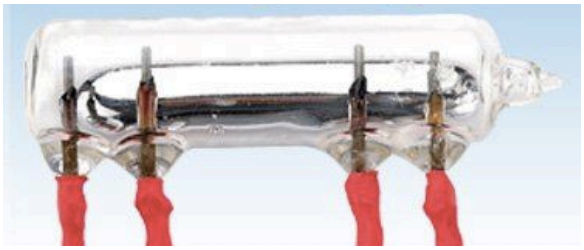


Amos et al., 2013

Mercury has lots of useful properties

Forms amalgams with silver and gold

Anti-fouling agent

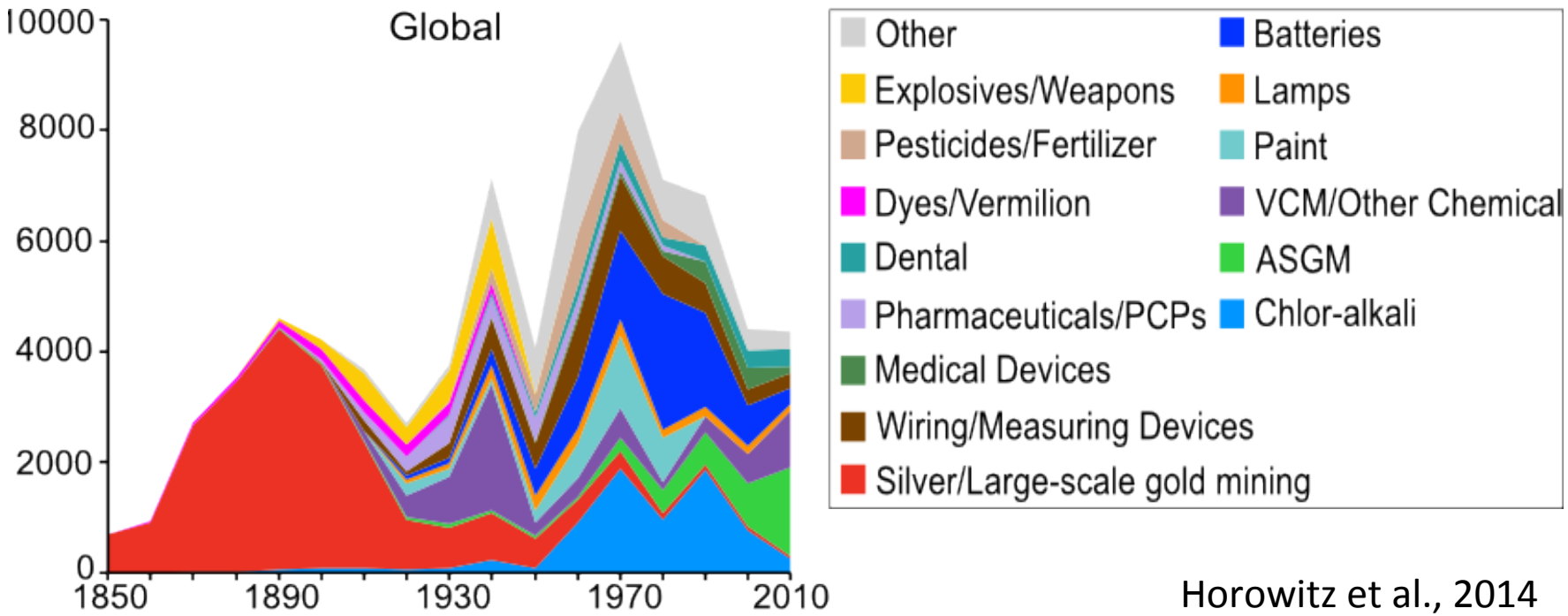


Good conductor

Mercury in products
Horowitz et al. (2014)

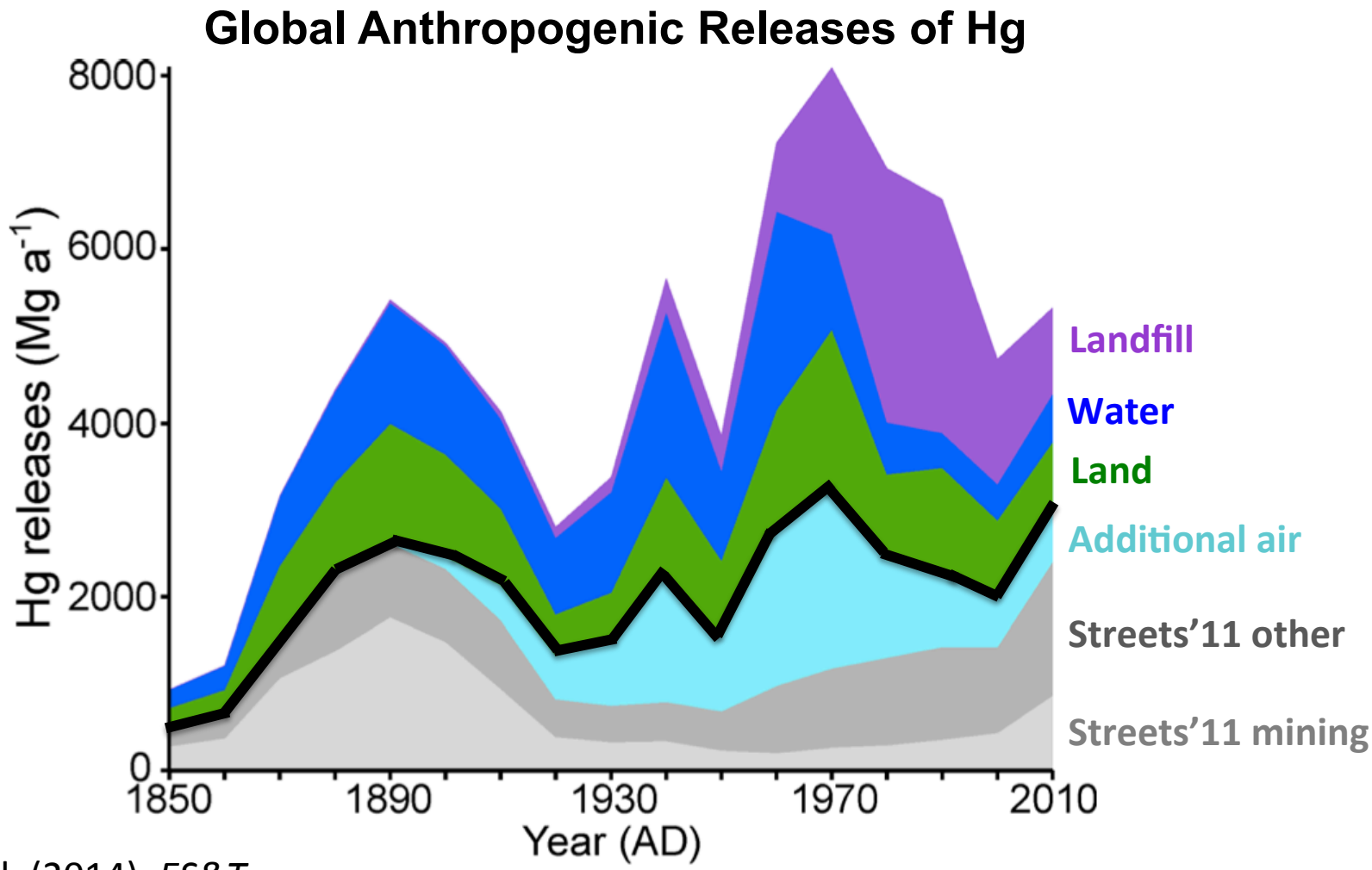
Global Consumption of Hg in Commercial Applications is well-constrained

Hg consumption (Mg yr^{-1})



Horowitz et al., 2014

Emissions from China and artisanal gold mining drive global trajectory in recent decades

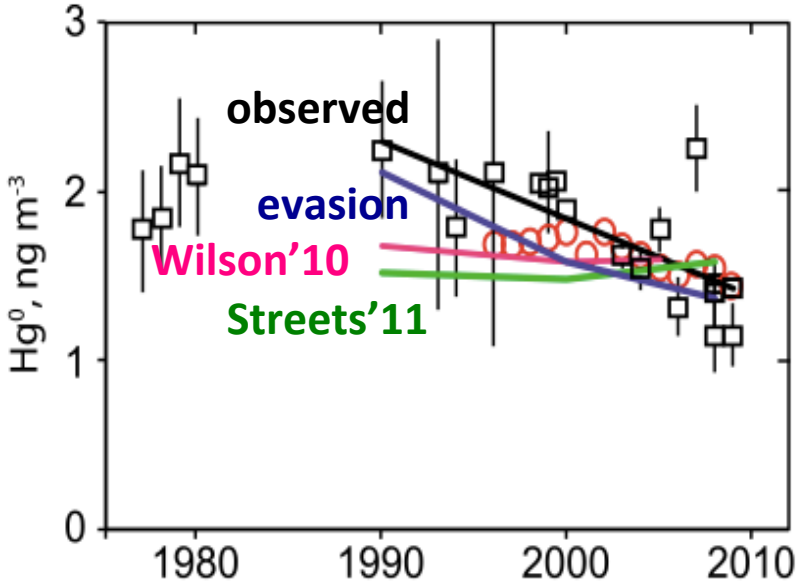


Horowitz et al. (2014), *ES&T*

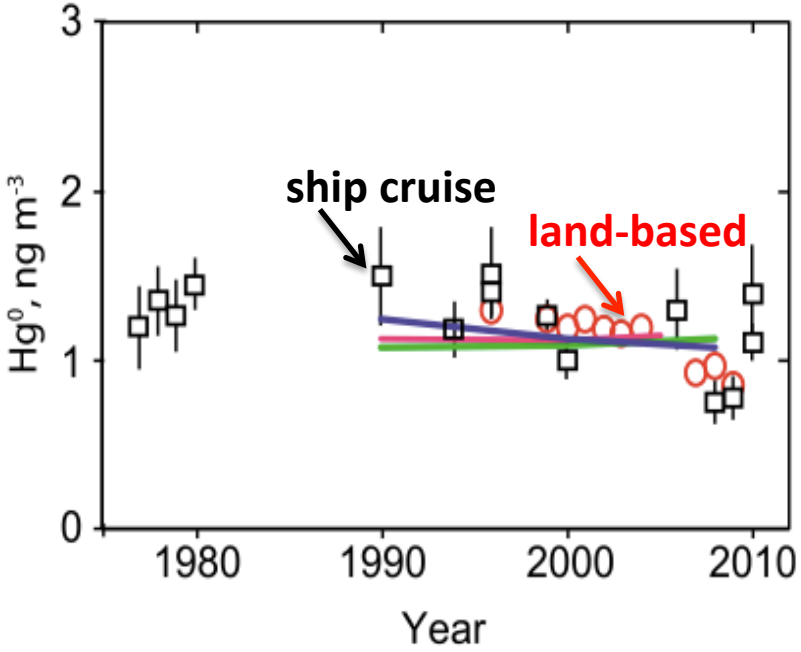
Explaining measured trends has challenged our understanding of Hg cycling and emissions

Marine Boundary Layer

North Atlantic

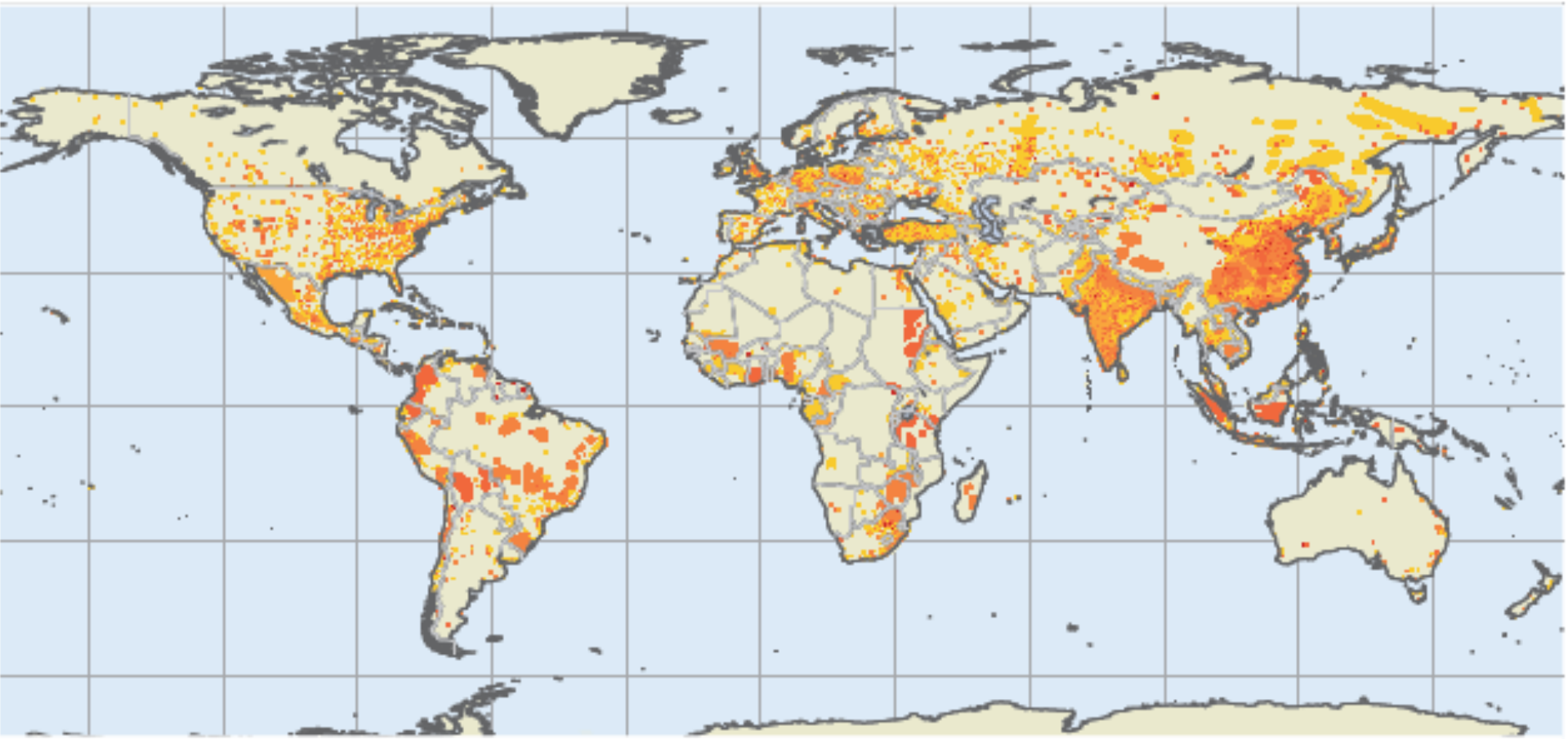


South Atlantic



Soeresen et al. (2012); Ebinghaus et al. (2011); Slemr et al. (2011)

Global atmospheric Hg emissions from anthropogenic sources in 2010

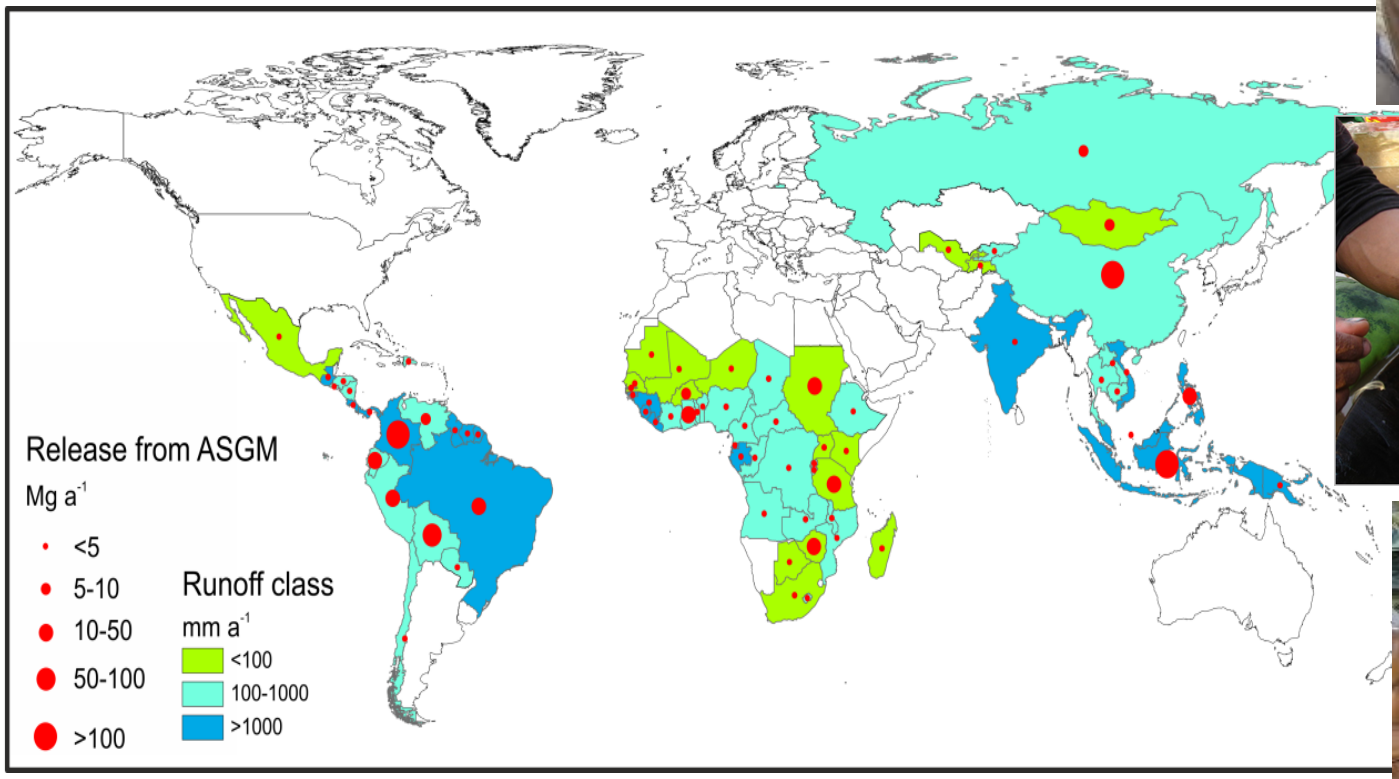


Mercury emission 2010, g/km²

0 2 5 10 100 1000

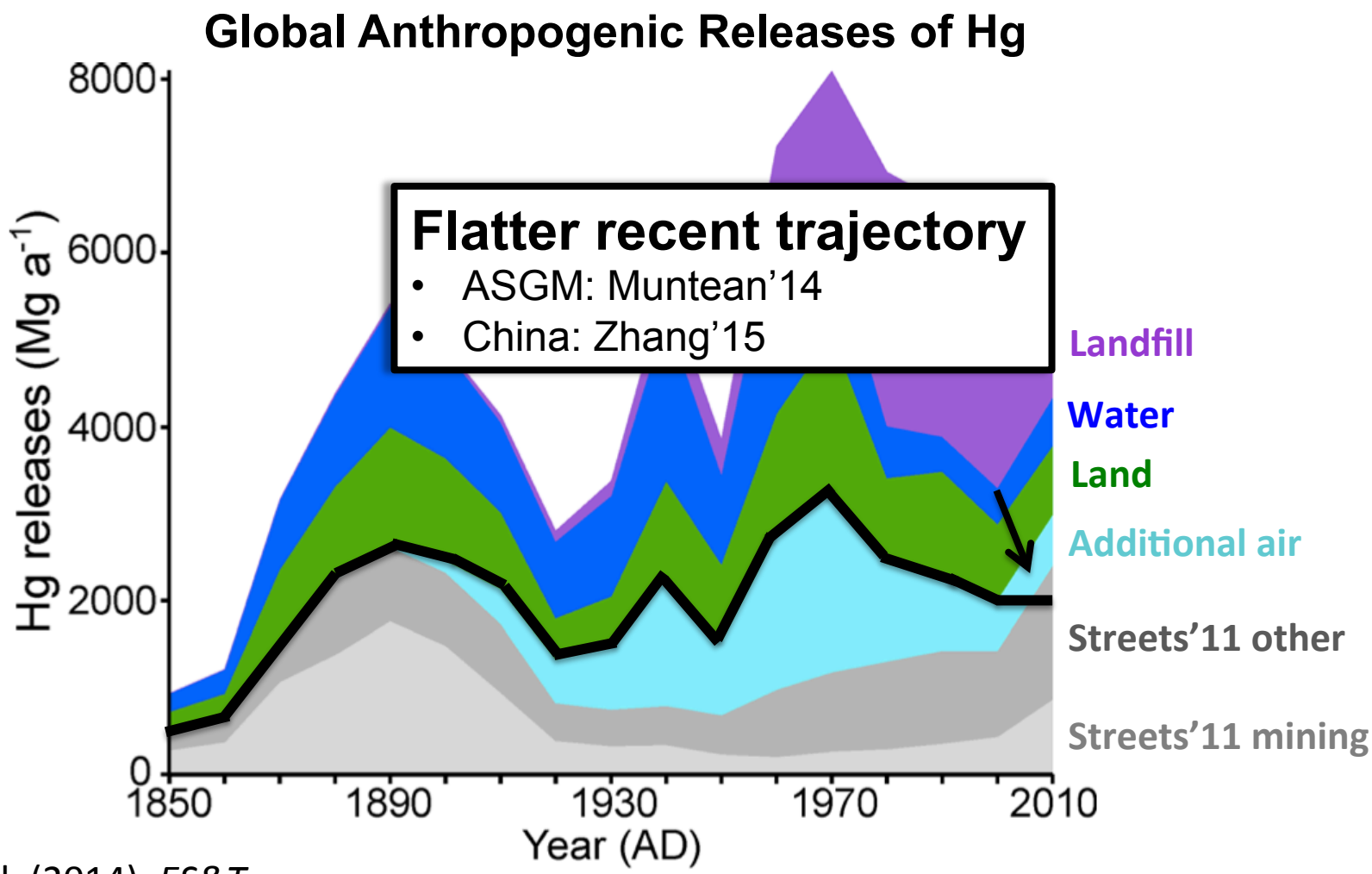
Source: UNEP, 2013

Artisanal and small-scale gold-mining now the largest global Hg source



Kocman et al., 2015

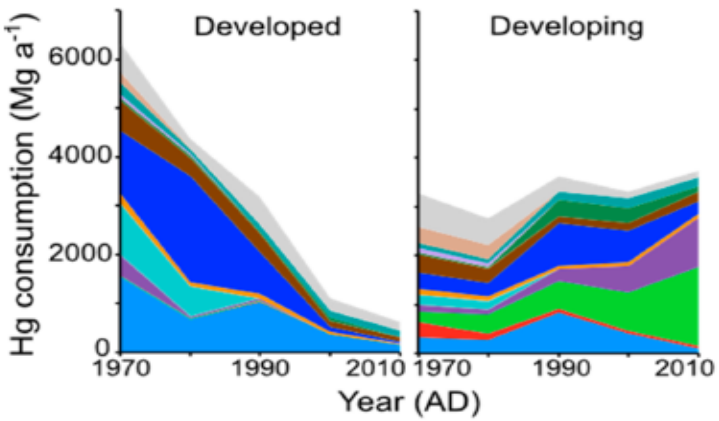
Emissions from China and artisanal gold mining drive global trajectory in recent decades



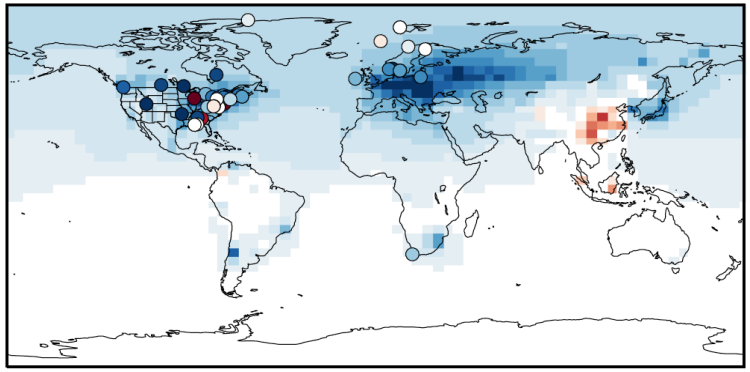
Horowitz et al. (2014), *ES&T*

Decreased Hg use in products and co-benefits from SO_2 control explain Hg trends

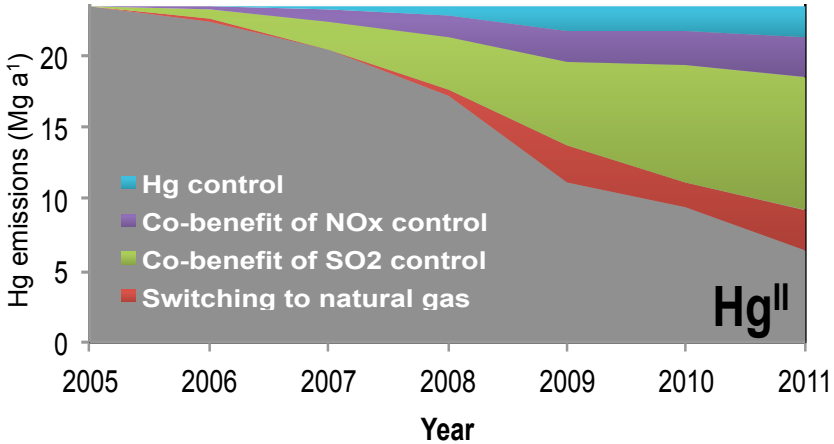
Horowitz'14 inventory with products



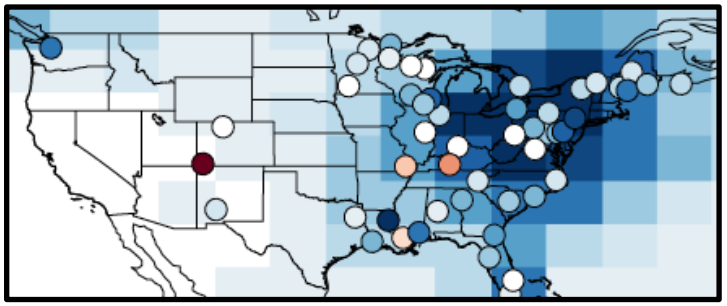
Atmospheric Hg^0 trend, 1990-2010



US utilities Hg^{II} emissions



US wet deposition trend, 1990-2010



Zhang et al. (in prep.)

Today's Discussion

- Global biogeochemical Hg cycle
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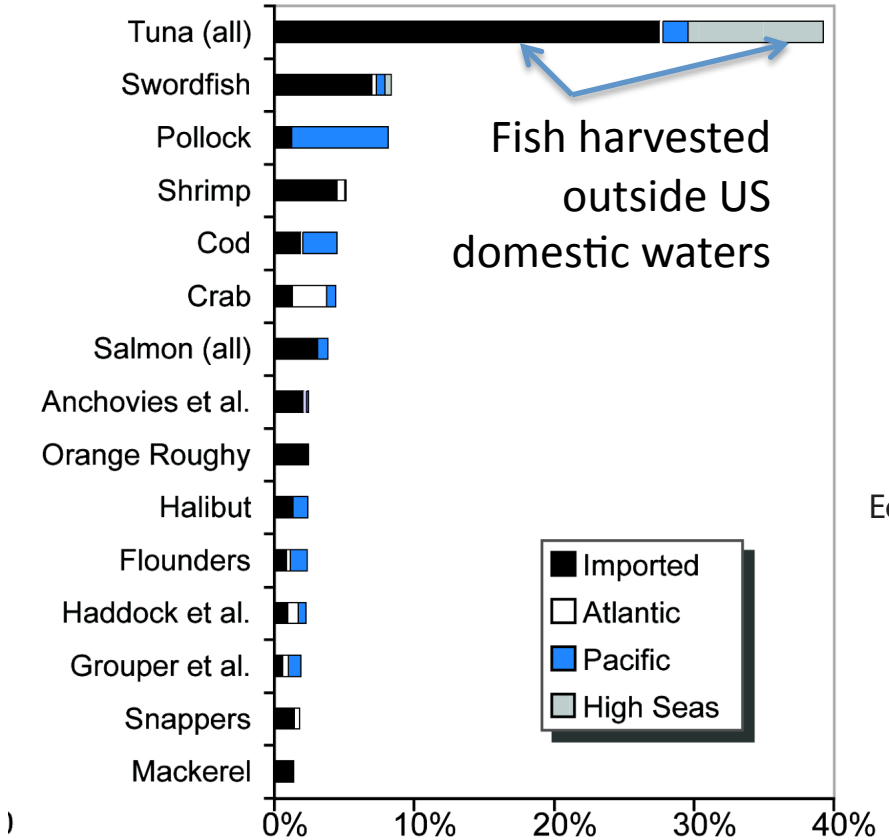
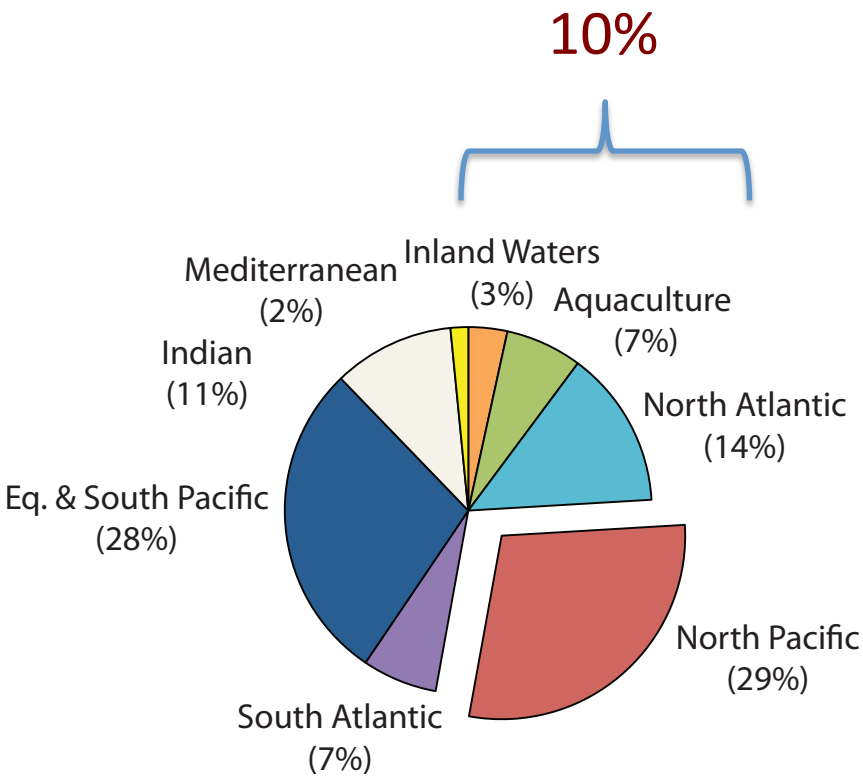
More than 90% of U.S. population-wide MeHg exposure is from consumption of marine fish

Fraction of U.S. Population-Wide Hg Intake (%)

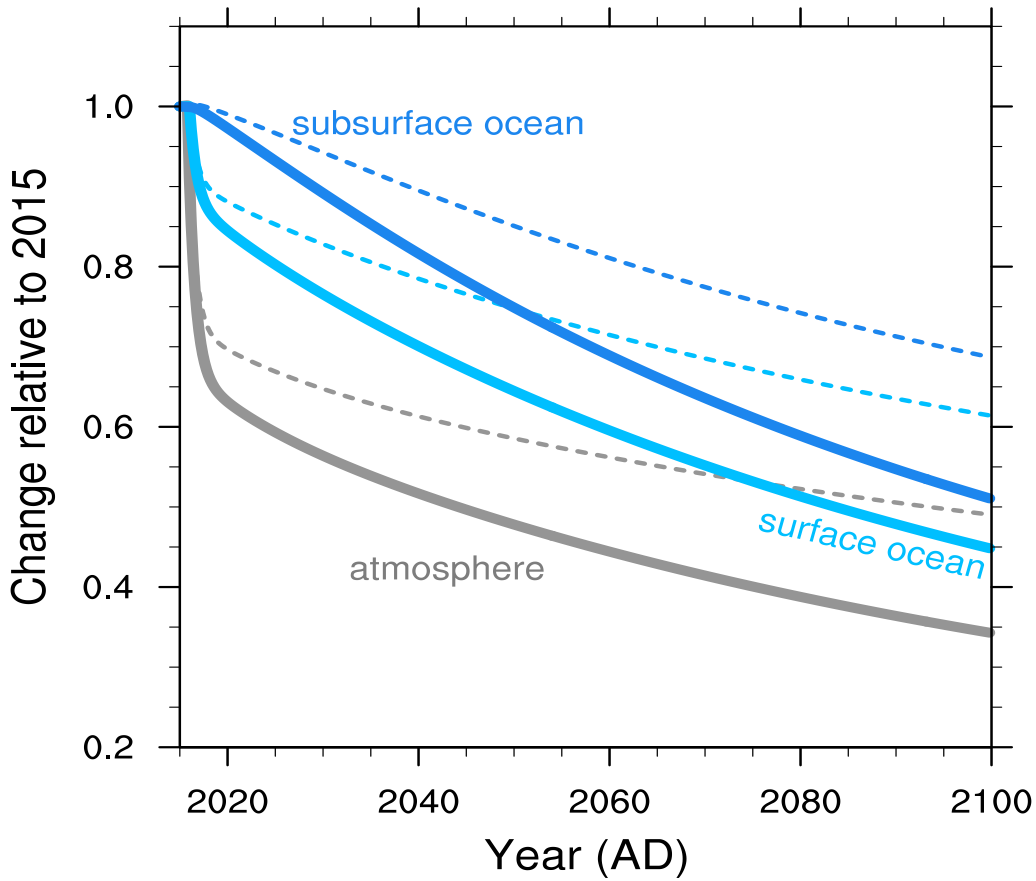




Sunderland, 2007

Oceans respond rapidly to reductions in anthropogenic emissions

Magnitude of decline sensitive to past emissions and coastal burial



Illustrative response to zero-out of anthropogenic emissions globally in 2015.

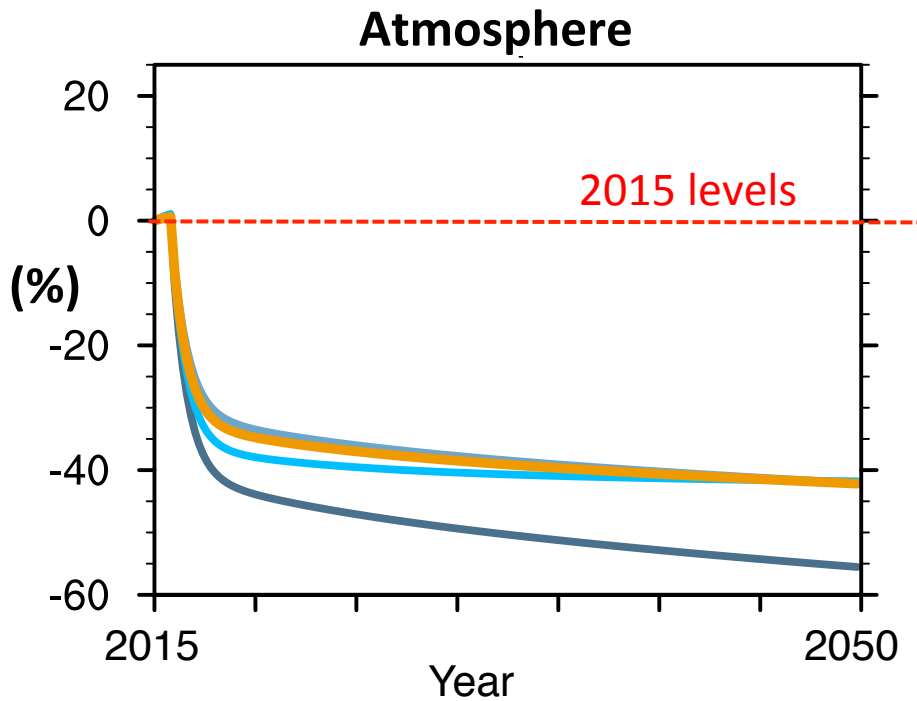
Dashed lines = model without coastal sediment sink

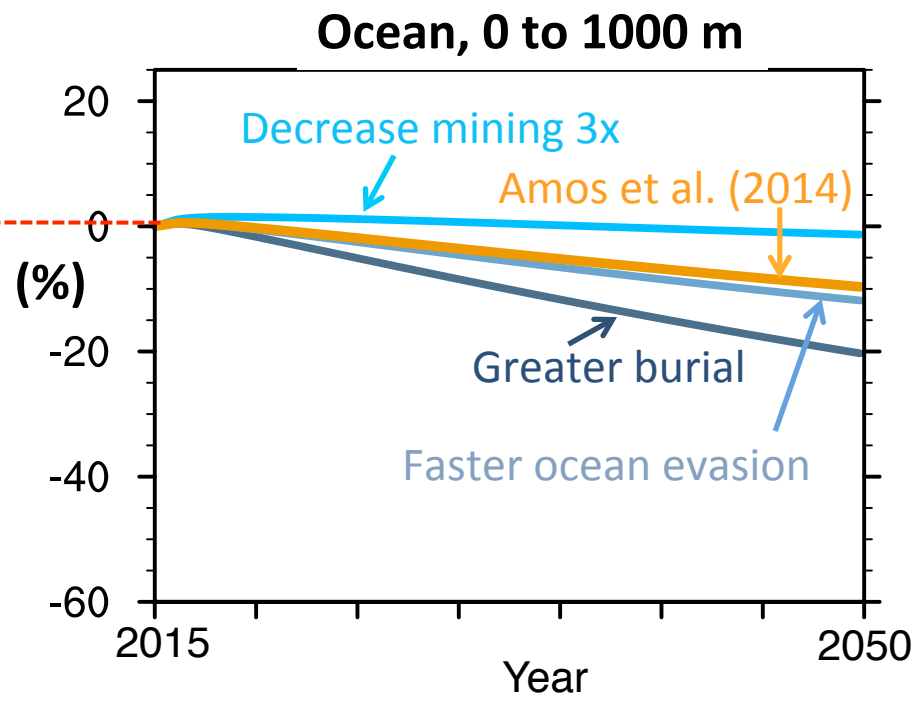
Solid lines = updated fully coupled geochemical model

Amos et al. (2014)

The need for aggressive emission reductions to stabilize ocean Hg is robust to uncertainty

Modeled response to terminating anthropogenic emissions
(normalized to 2015)





Amos et al. (2015), *ES&T*

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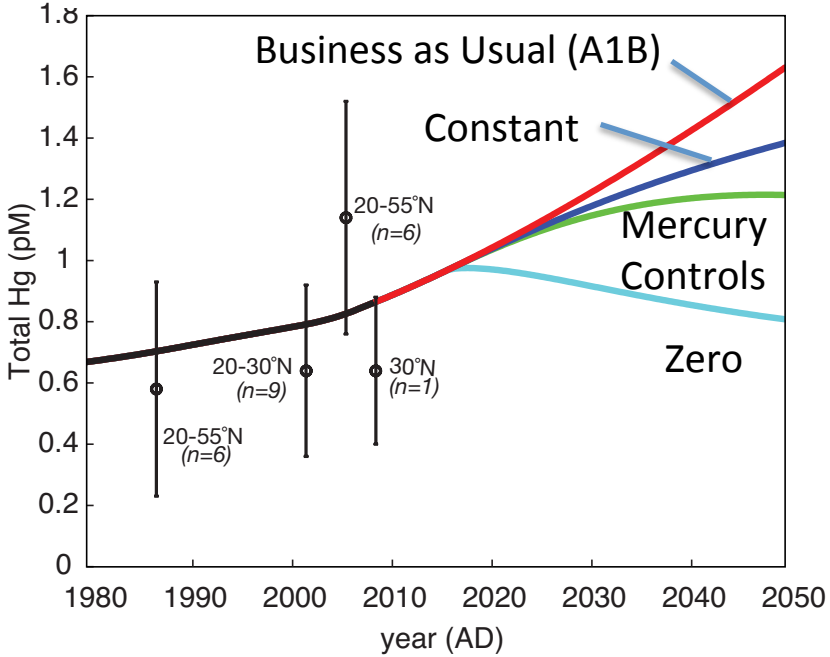
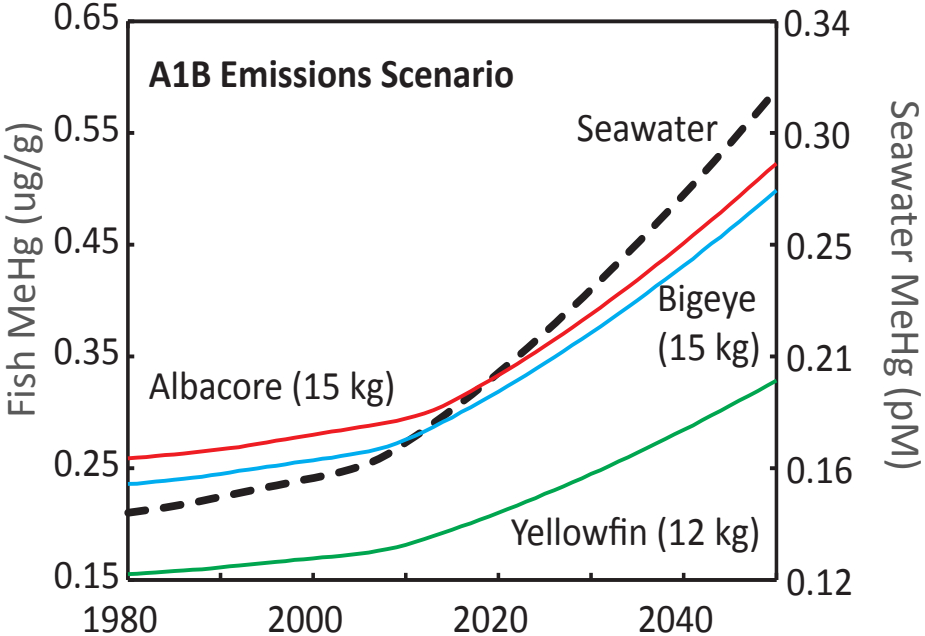
Modeled North Pacific seawater and fish concentrations based on anthropogenic emissions

- Range of future concentrations dependent on anthropogenic emissions choices
- Fish MeHg levels track water column concentrations with lag << 10 years

Total Hg North Pacific Seawater (pM)



North Pacific Tuna MeHg (ug/g)



Sunderland et al., 2013

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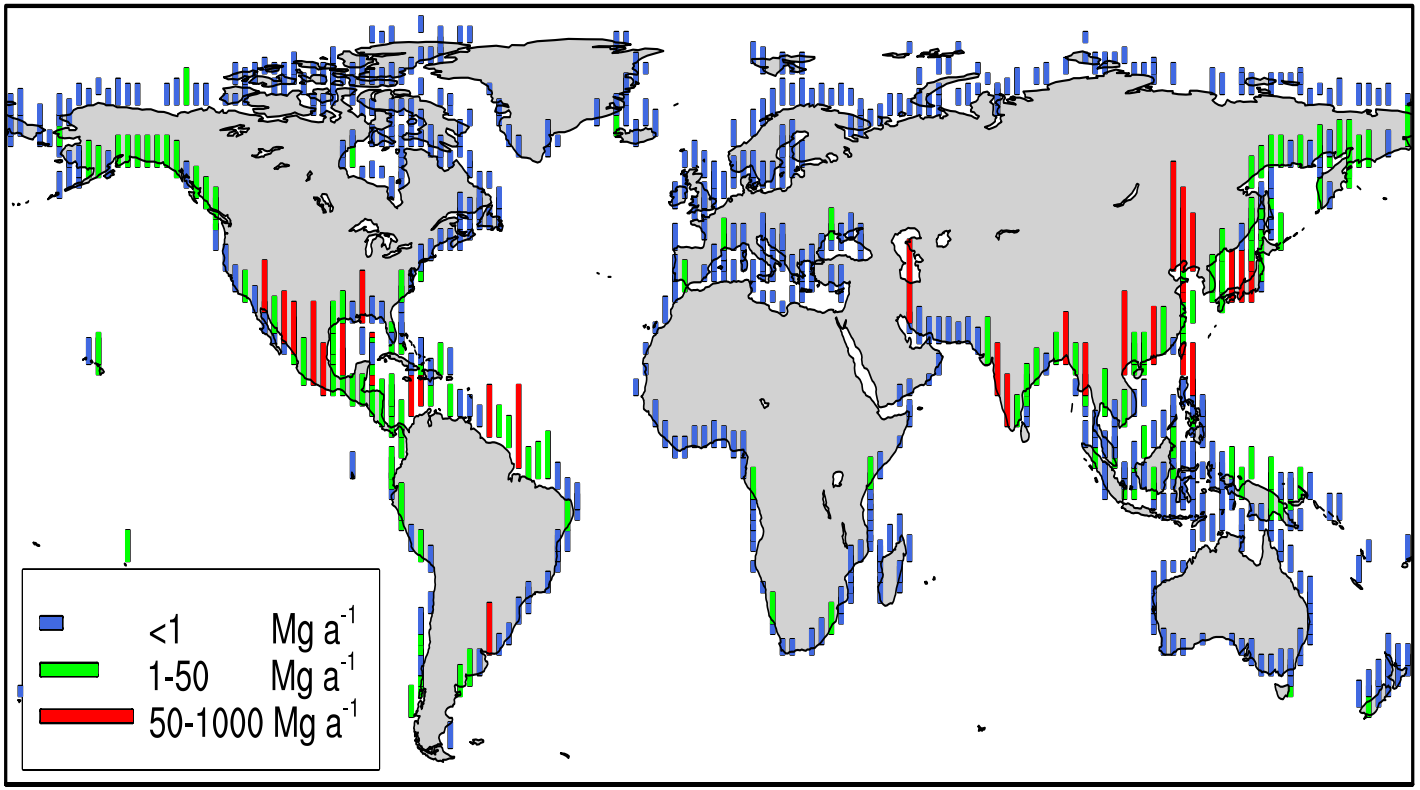
New Global Inventory for Hg Discharges from Rivers

New Inventory: Rivers: 5400 ± 2700 Mg y^{-1}

Anthropogenic emissions to the atmosphere~ 2000 Mg y^{-1}



Amos et al. (2014)

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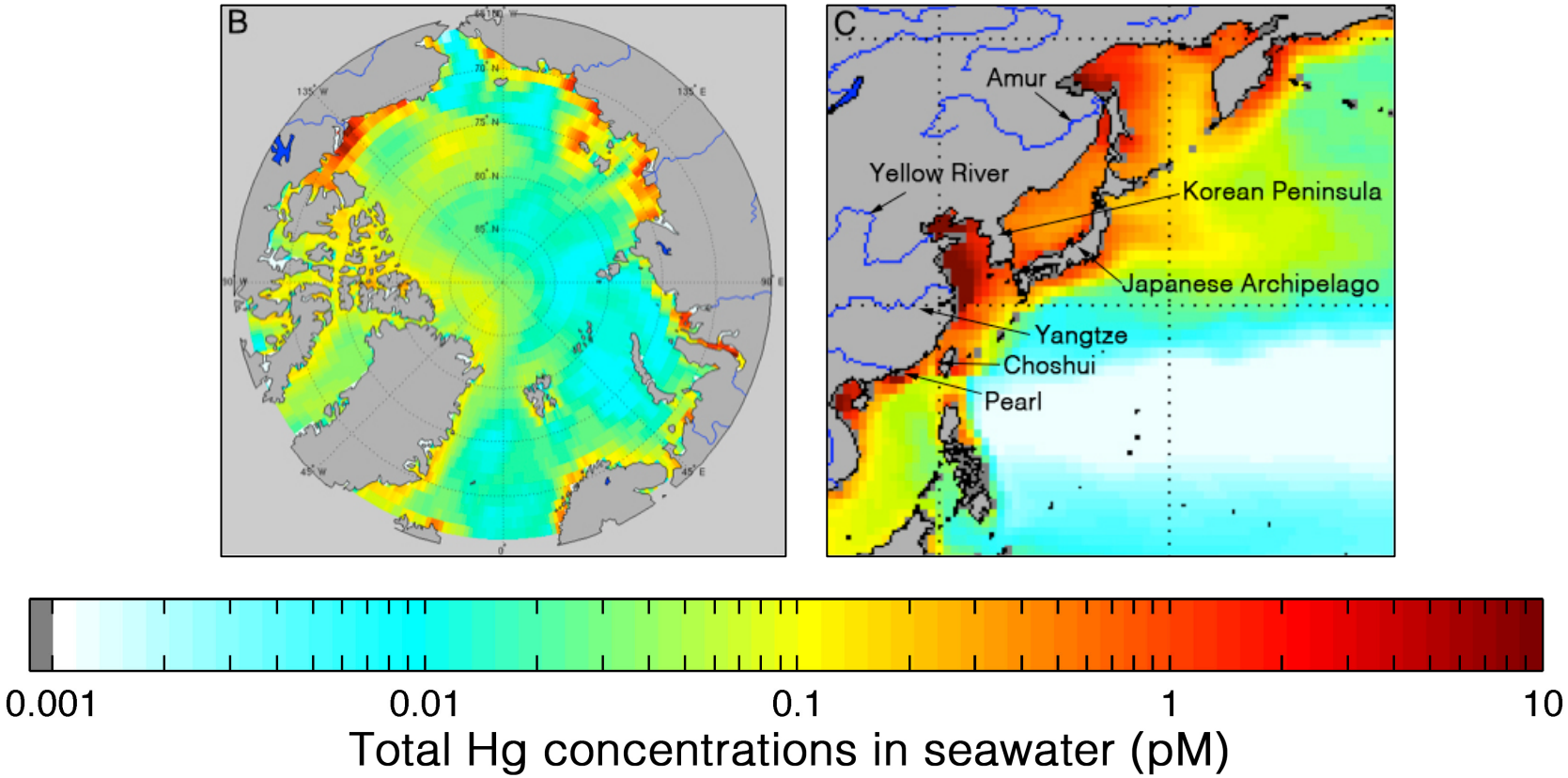
High concentrations of bioaccumulative contaminants pose risks for marine fisheries

Modeled contributions of global rivers discharges to seawater mercury concentrations



Source: Zhang et al., 2015 (in-review)

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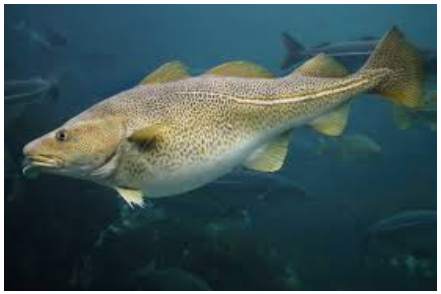
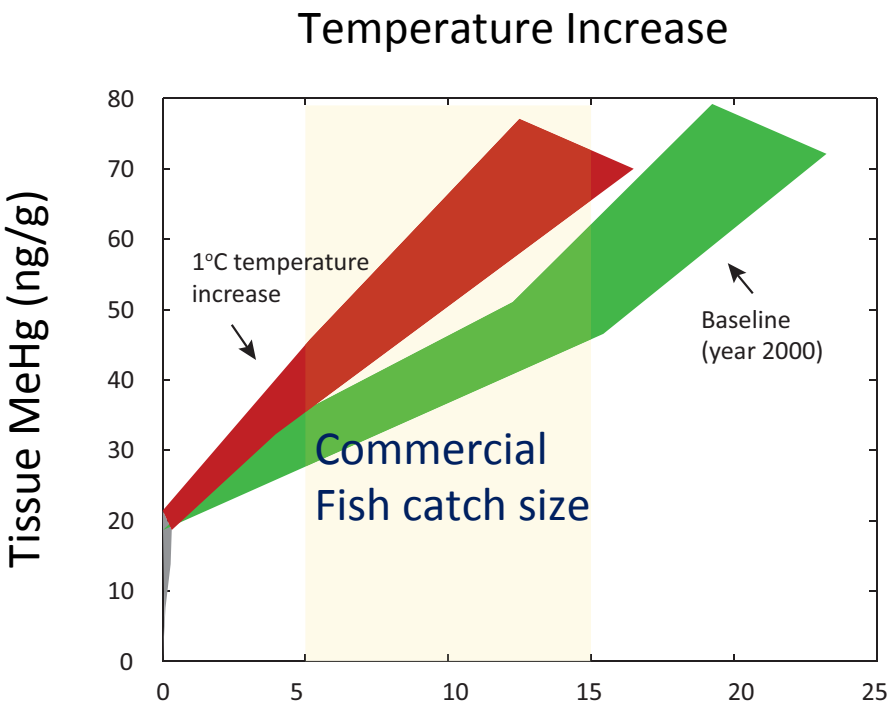
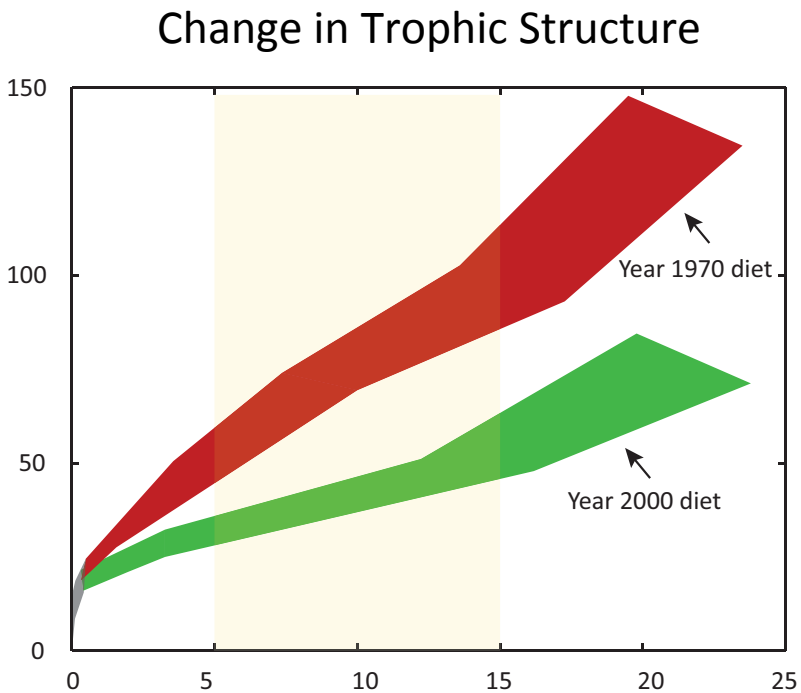
We have developed a generic model for MeHg bioaccumulation in marine ecosystems

Example: Atlantic Cod (*Gadus morhua*) in the Gulf of Maine







Wet weight (kg)

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The Form of Hg Determines its Health Impact

- **Inorganic Hg** includes:
- *Quicksilver*
- 0.01% absorption

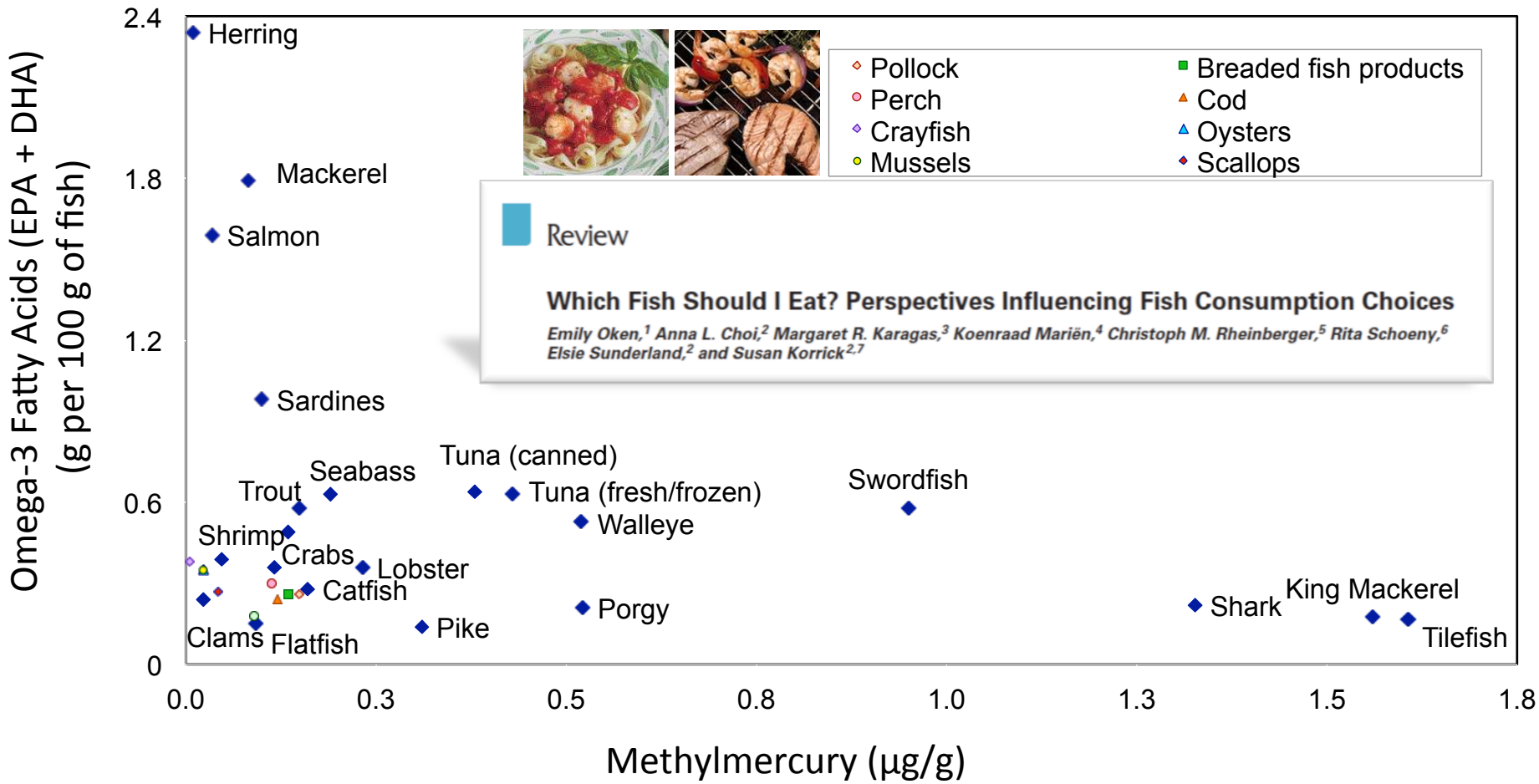
- *Hg^0 (inhalation)*
- *~70% absorption*

- *Hg^{II}*
- *Avg of 7% absorption (range 1-17%)*

- **MeHg** (CH_3Hg)

- >90% absorption efficiency
- Primarily a central nervous system toxin
- Half-life of 50-70 days
- Chelation not effective

Ongoing debate about benefits and risks of seafood



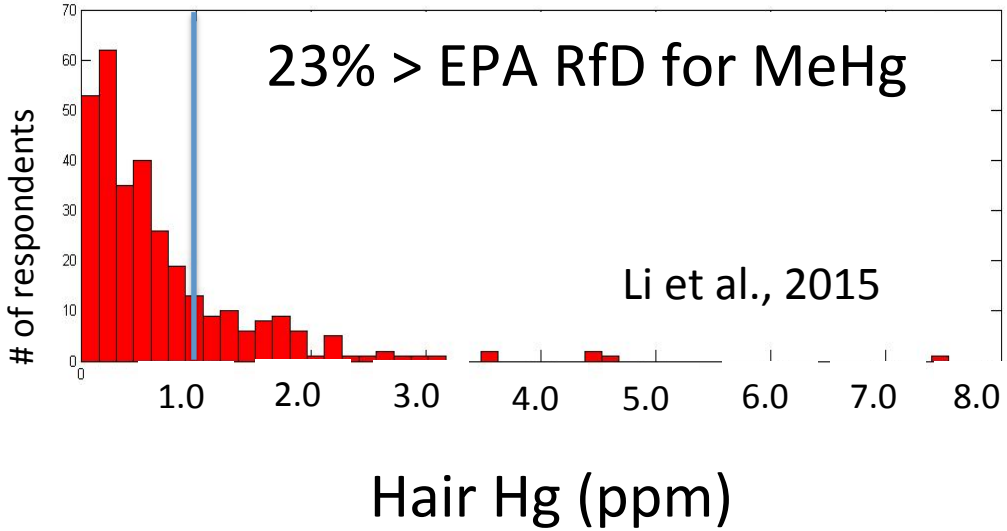
Source: Mahaffey et al., 2008

MeHg Exposure =

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

Individual Preferences **Environmental Exposure**

U.S. individuals consuming > 3 fish meals per week



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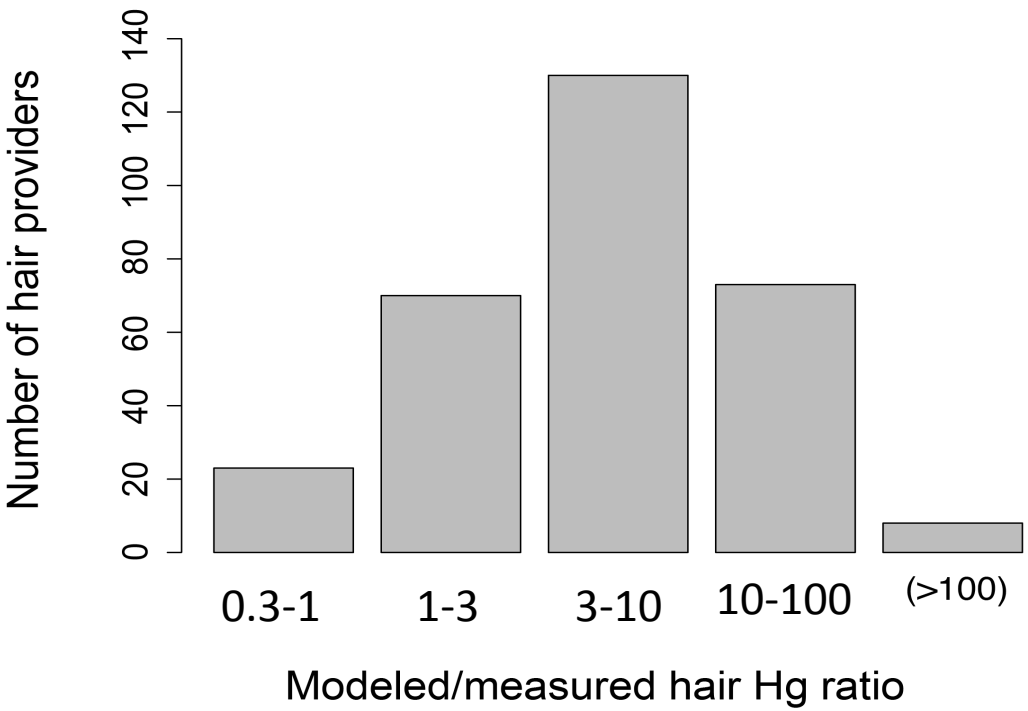
Many individuals have much less MeHg in their hair than predicted based on their diet

Individuals consuming > 3 fish meals per week

1 = perfect agreement model/measured
<1 = underpredict biomarker concentration
>1 = overpredict biomarker concentration



- Correction applied for species specific over-reporting
- Data quality verified for all surveys
- Individuals with low hair Hg are a mix of ethinicities, genders, and ages from diverse regions in the US
- This pattern has been observed across many studies

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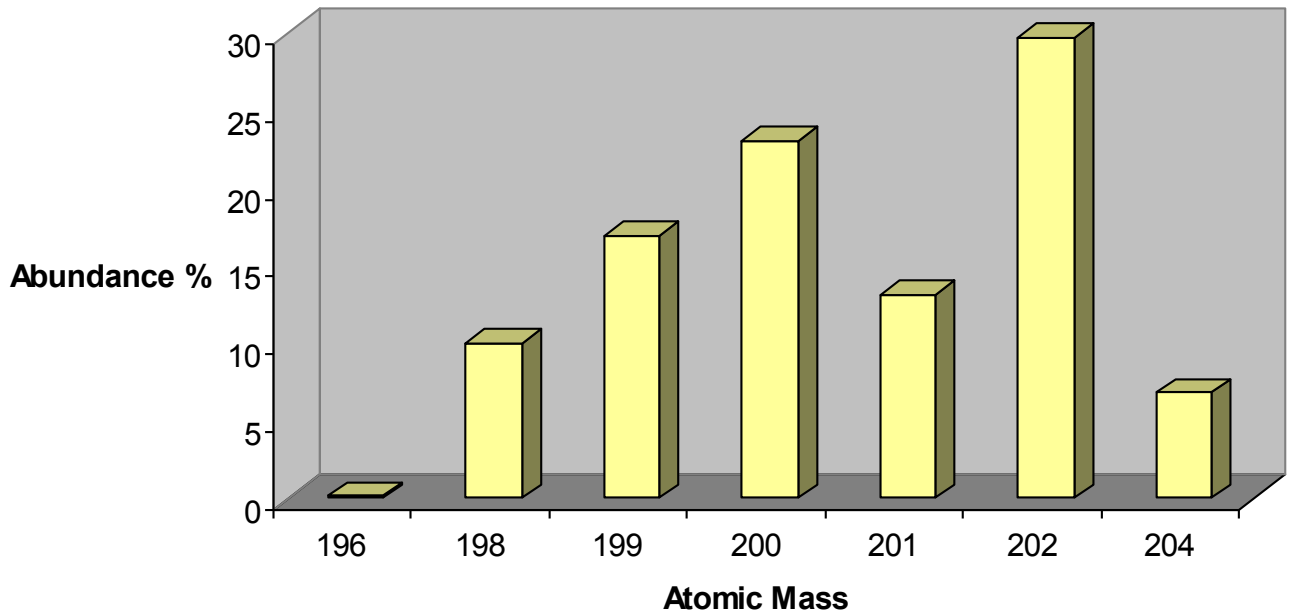
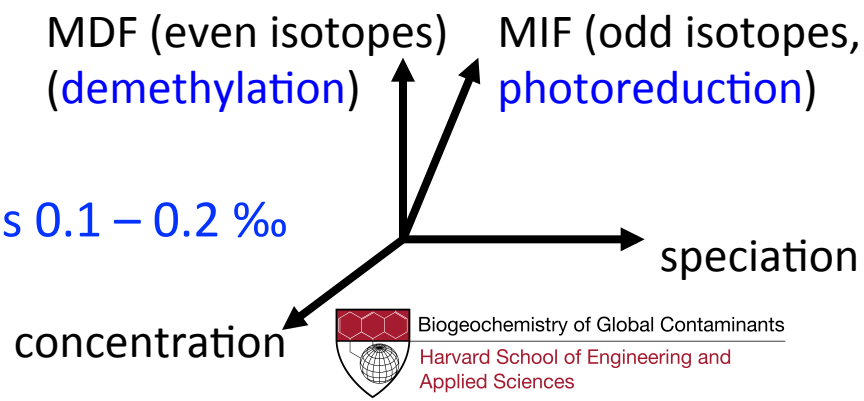
Hg stable isotopes provide new information on environmental processes and Hg exposure sources



Example: ^{202}Hg abundance in nature varies from 29.6 to 29.7 %



Typical measurement precision for δ^{202}Hg is 0.1 – 0.2 ‰

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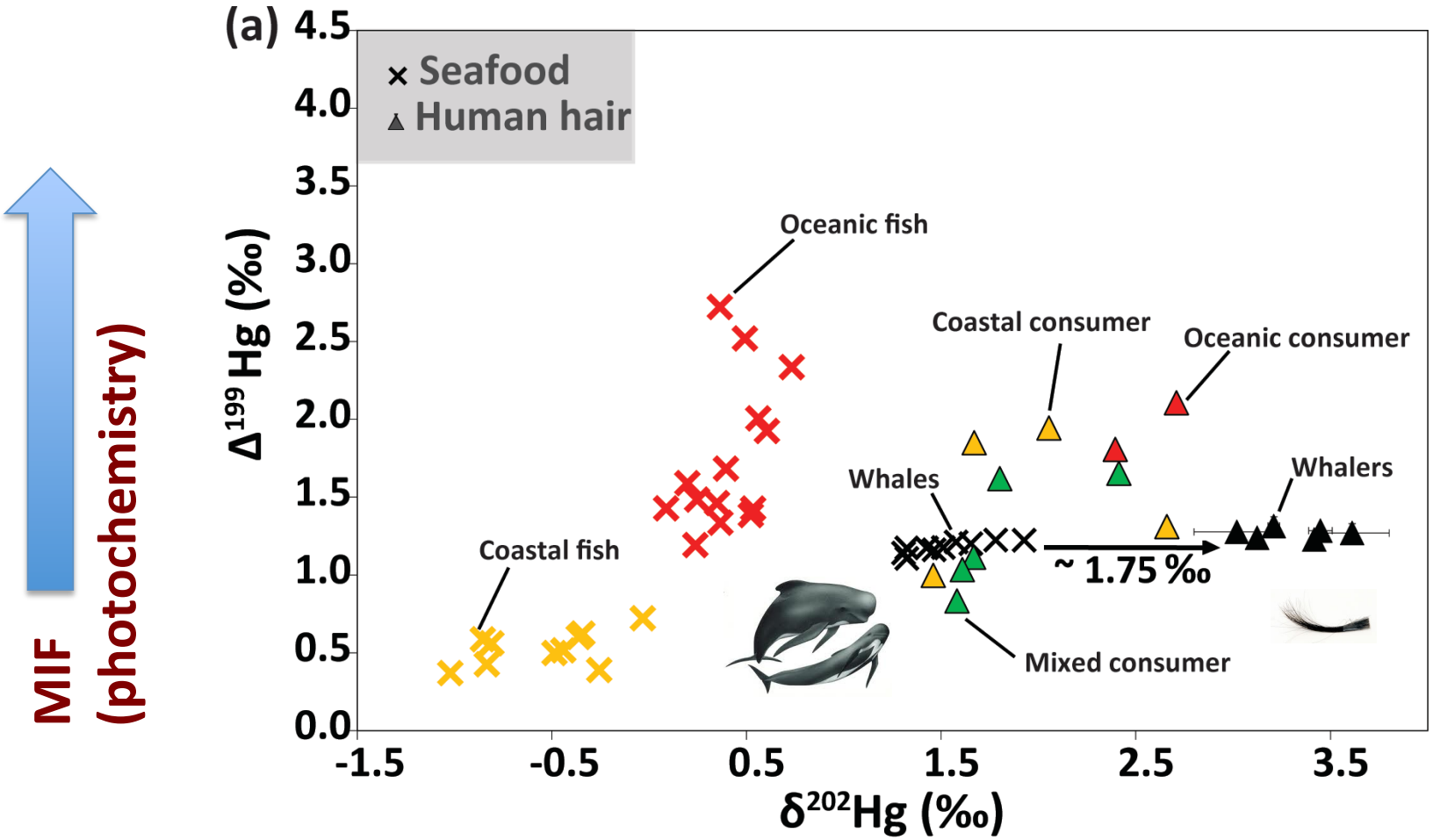
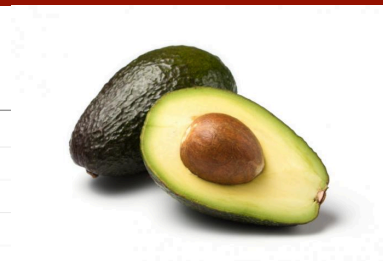
You are what you eat (no MIF change) plus 2‰ MDF

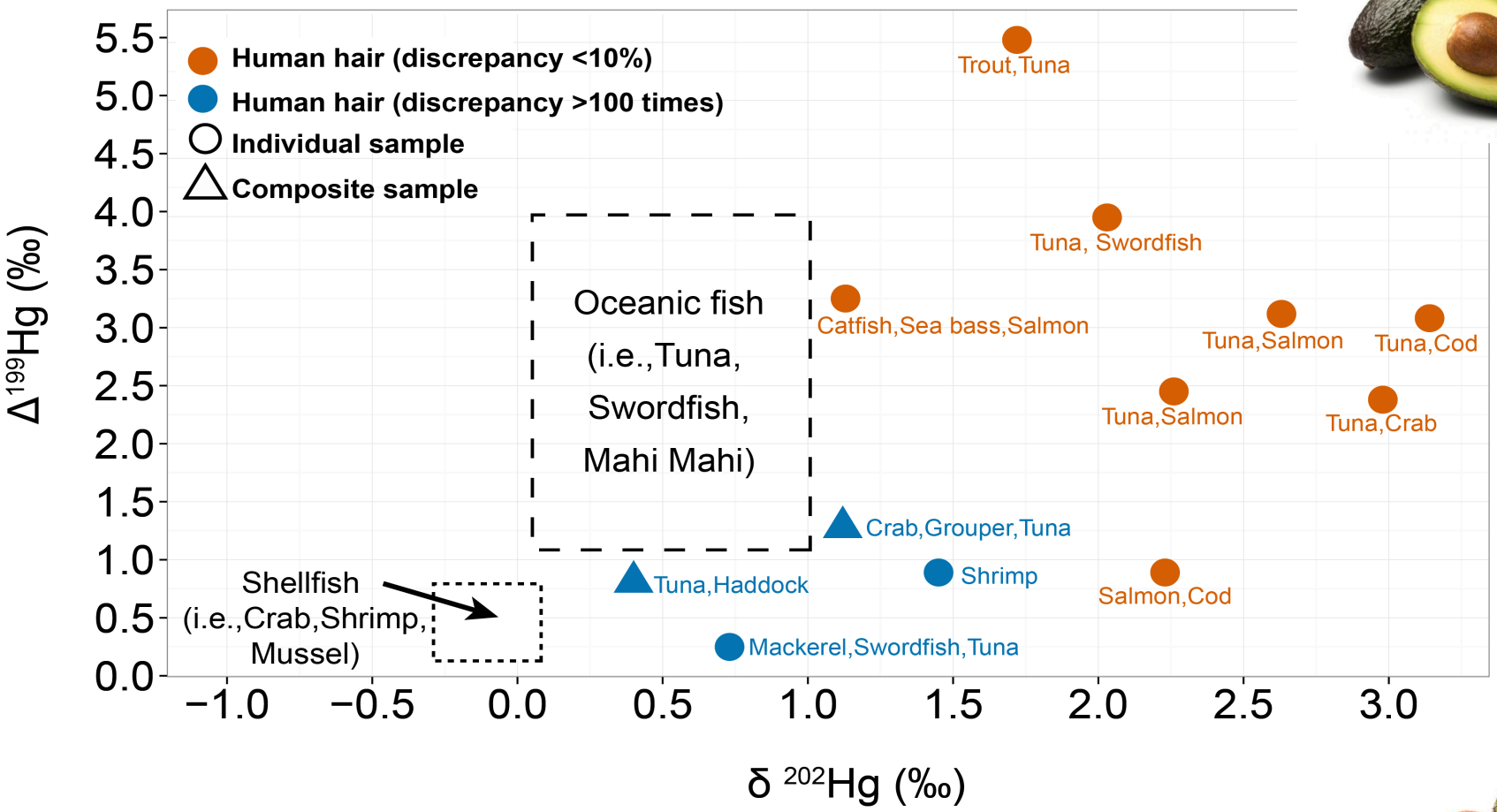


MDF (demethylation)

Li et al. (2014)

Demethylation does not explain discrepancy between intake and biomarker Hg levels





Li et al., 2015, in prep.

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Summary

- Human activity has substantially enriched Hg in surface reservoirs
- Timescales of coupling of the ocean, atmosphere and organic soils range from a few months to millennia
- Substantial enrichment of the subsurface ocean in Hg from historic activity
- Short term dietary interventions may be possible for altering the assimilation efficiency of MeHg allowing consumption of fish and traditional foods with minimal risk