Mercury cycling and transformations across Louisiana estuarine and wetland gradients

By

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What We've Done To Date

Study Focus Areas:

- Hg cycling and geochemistry in sediment
- Hg speciation & Hg- DOC interactions (sed & water)
- Hg-plant interactions (emergent marsh)

5 Field Trips (2003-2006)

- Estuarine salinity Gradient:
upper rivers → Lake Pontchartrain → salt marsh sloughs
- Wetland Gradient: cyprass swamp → spartina saltmarsh
- Wetland sub-habitats: vegetated marsh, sloughs, ponds and mudflats

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Lake
Rivers
Wetlands

Tangipahoa R.

Tchefuncta R.

Baton Rouge

Bayou Lacombe

Blind R.

Rigolets

Metairie

New Orleans

Caernarvon

Jean Laffitte

Lake Junop

Lake Felicity

78 km

Data U.S. Navy
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Image USDA Farm Service Agency

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Sediment Biogeochemistry

Microbial Processes
- Hg-methylation
- MeHg-degradation
- Sulfate reduction
- Methanogenesis

Iron Chemistry
- sed. & pw Fe(II)
- sed. amorphous Fe(III)
- sed. crystalline Fe(III)

Mercury Speciation
- pw THg & MeHg
- sed. THg & MeHg
- sed. REACTIVE Hg(II)

Carbon Chemistry
- pw DOC
- pw acetate
- sed. organic content (LOI)
- sed. C/N
- sed. lignin content

Sulfur Chemistry
- pw SO_4^{2-}, HS^-
- sed. AVS (FeS)
- sed. CRS (S^0, FeS_2)

Other Chemistry
- sed. pH
- sed. Redox
- pw Cl^-

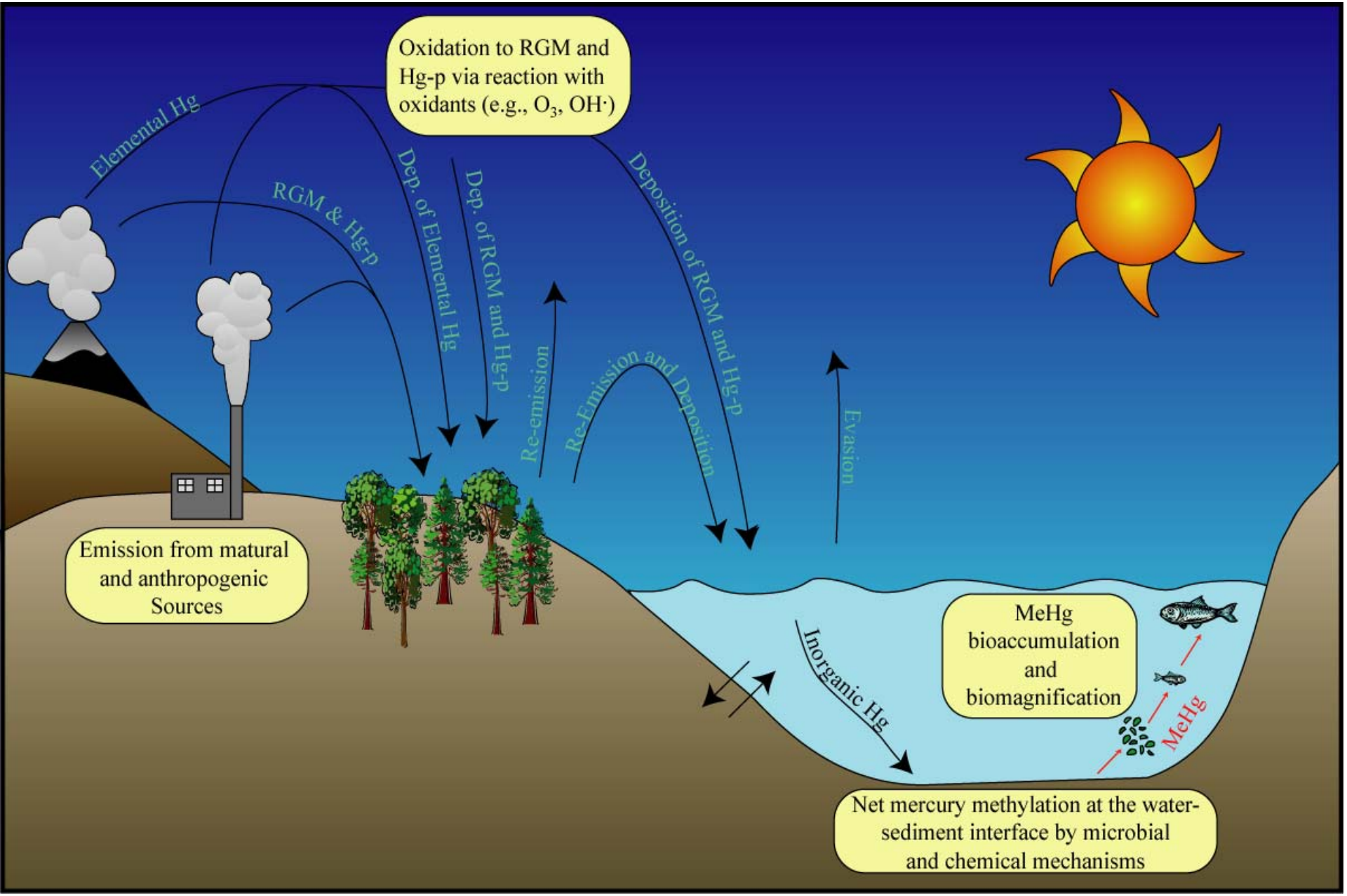
Water Chemistry & Plant-Hg Interactions

Water Chemistry

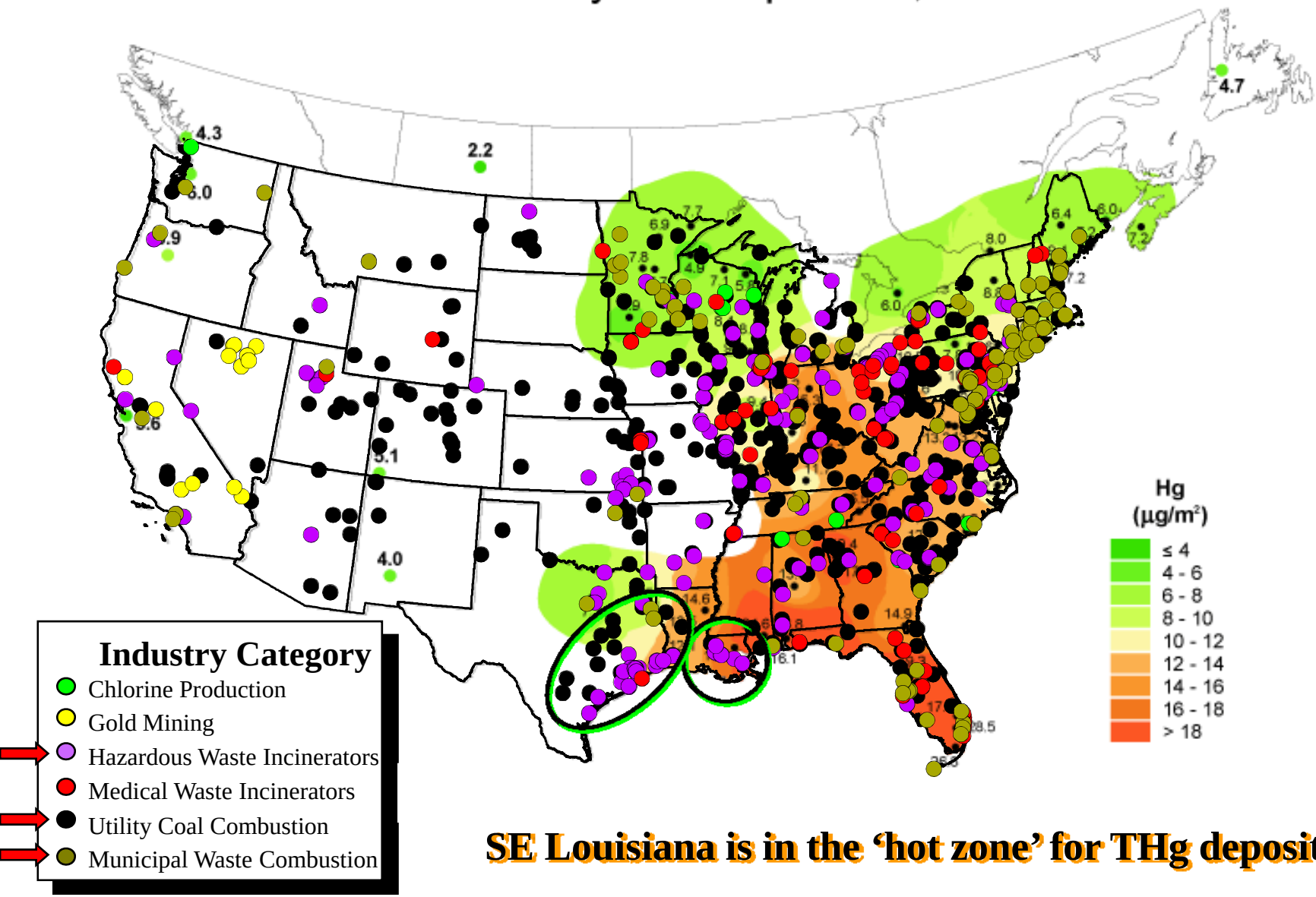
- Particulate THg & MeHg
- Dissolved THg & MeHg
- DOC
- SUVA (aromaticity)
- DOC isolates (XAD)
  - HPOA, HPON
  - TPIA, TPIN
  - LMW HPI
- pH
- major cations & anions
- redox metals (Fe, Mn)

Wetland Plant Metrics

- Above and below ground plant biomass
- Root: density & depth
- Leaf: % lignin
- Leaf and root:
  - THg & MeHg
  - %C & %N
  - ^{13}C & ^{15}N isotopes
  - K:Na ratio (stress index)

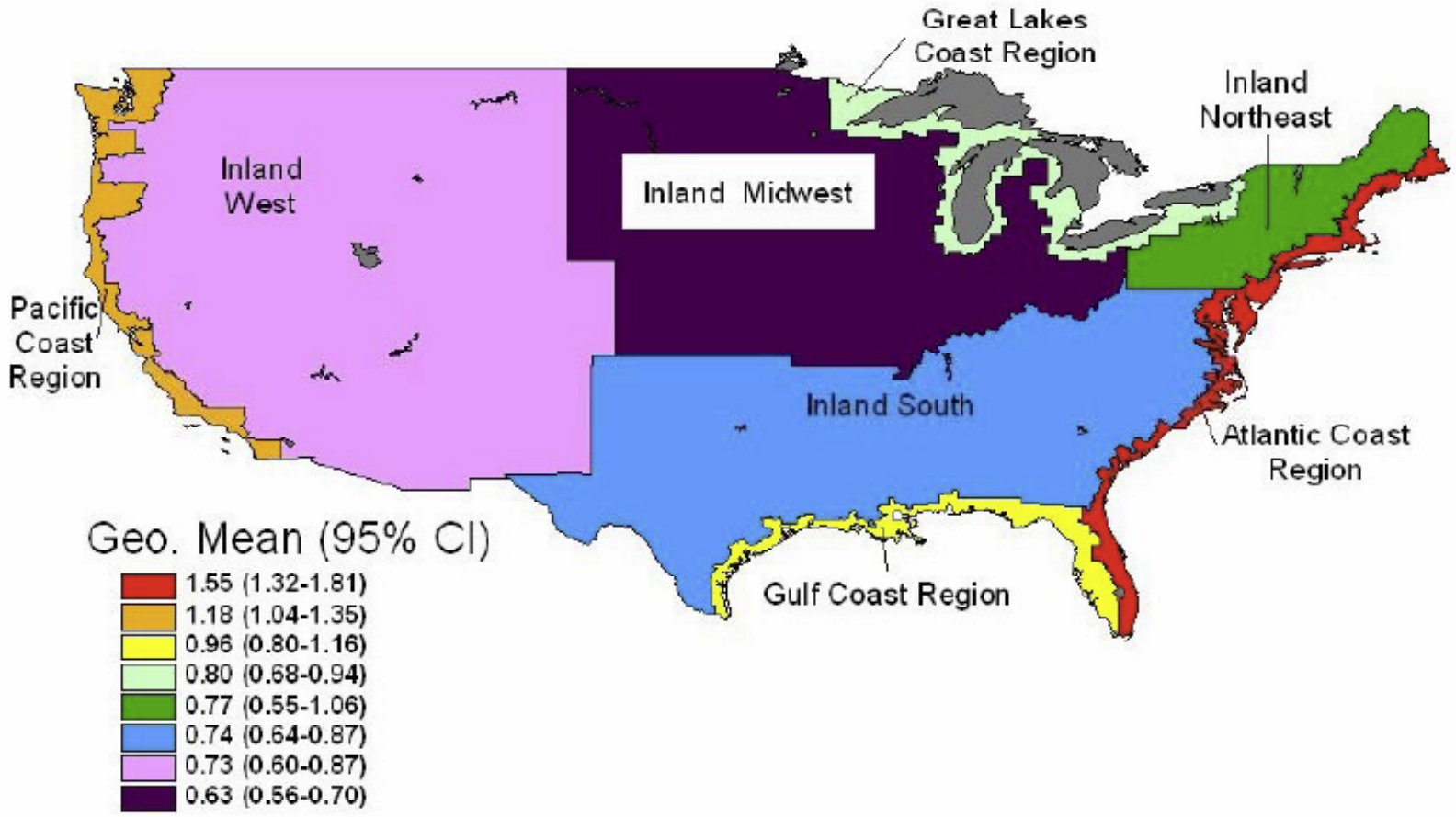

Oxidation to RGM and Hg-p via reaction with oxidants (e.g., O_3, $OH^{\cdot}$)
Elemental Hg
RGM & Hg-p
Dep. of Elemental Hg
Dep. of RGM and Hg-p
Re-emission
Re-Emission and Deposition
Deposition of RGM and Hg-p
Evasion
Emission from matural and anthropogenic Sources
Inorganic Hg
MeHg bioaccumulation and biomagnification
MeHg
Net mercury methylation at the water-sediment interface by microbial and chemical mechanisms

Source-Receptor Relations



SE Louisiana is in the 'hot zone' for THg deposition

2A. Blood total mercury concentration (µg/L)



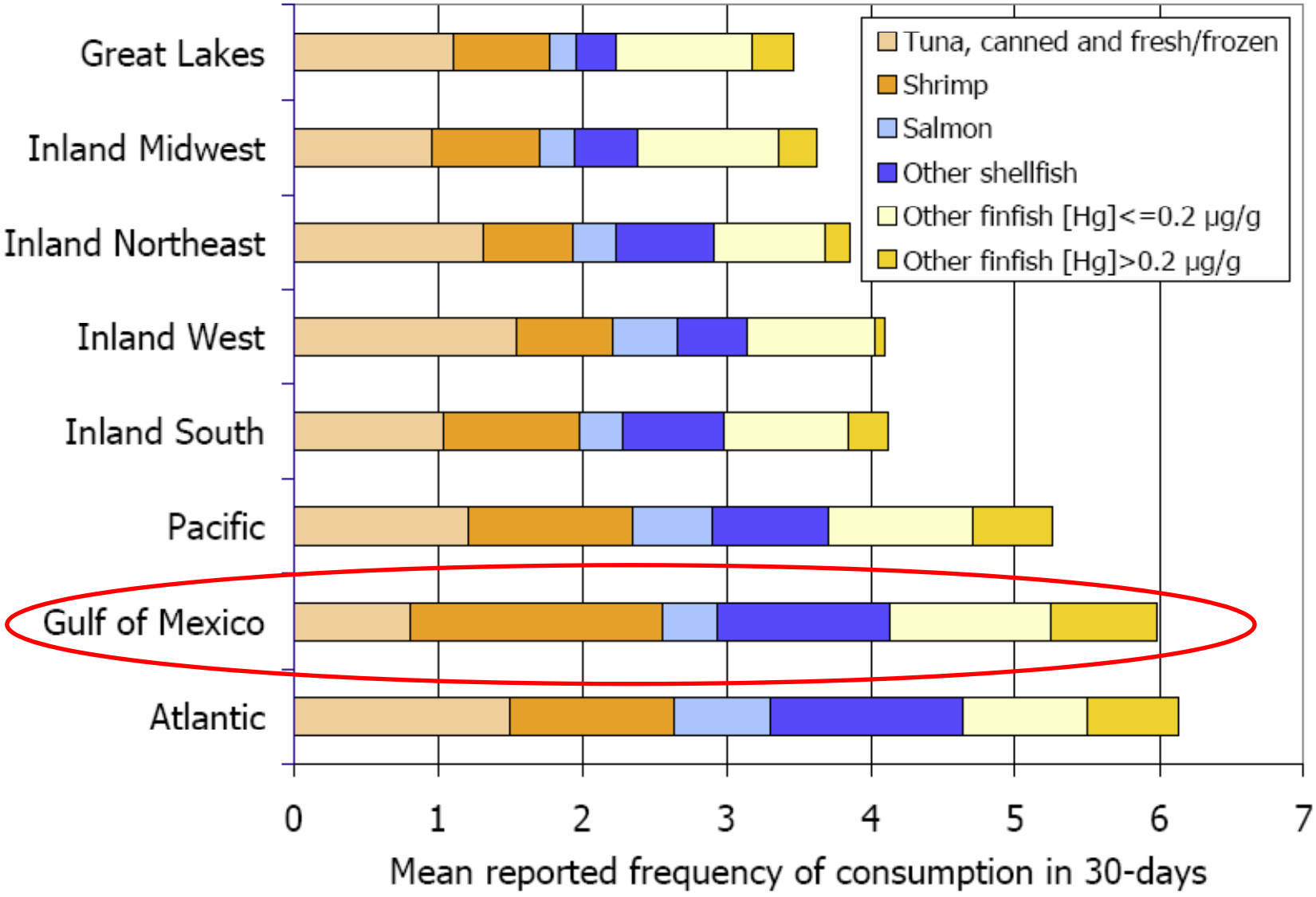


Figure 3. Species consumed and frequency of consumption by geographic residence.

Conceptual Model Mercury Cycling at the Marine-Continental Margin

Mercury emission sources

RGM sorbs to sea salt aerosols

Ocean breeze returns With elevated RGM

$HgCl_2$

Continental winds & mercury sources

$Cl_2, Br_2 \rightarrow O_3 + Hg^0$

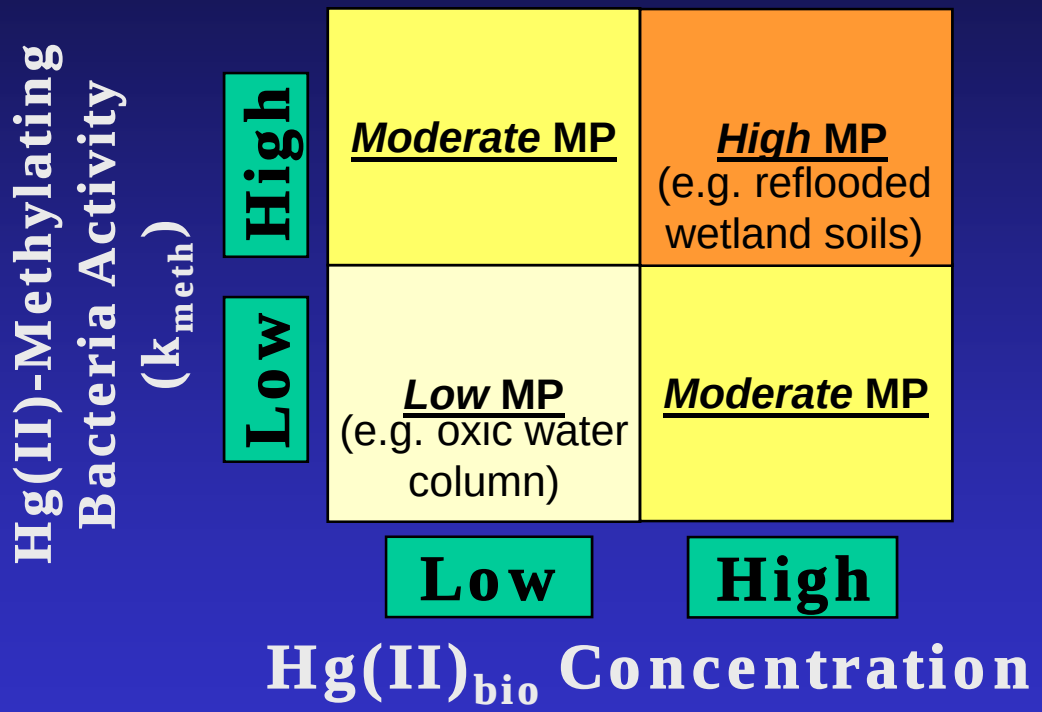
Volatilization of free halogens

Land

Ocean

What Controls MeHg Production?

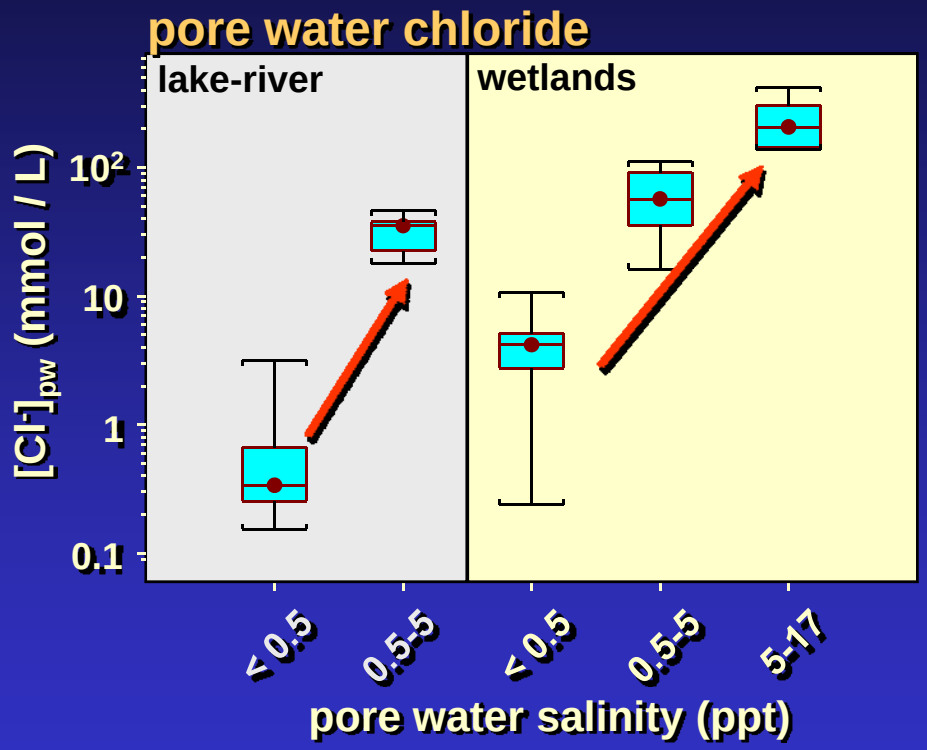
MeHg Production = k_{meth} x $Hg(II)_{bio}$

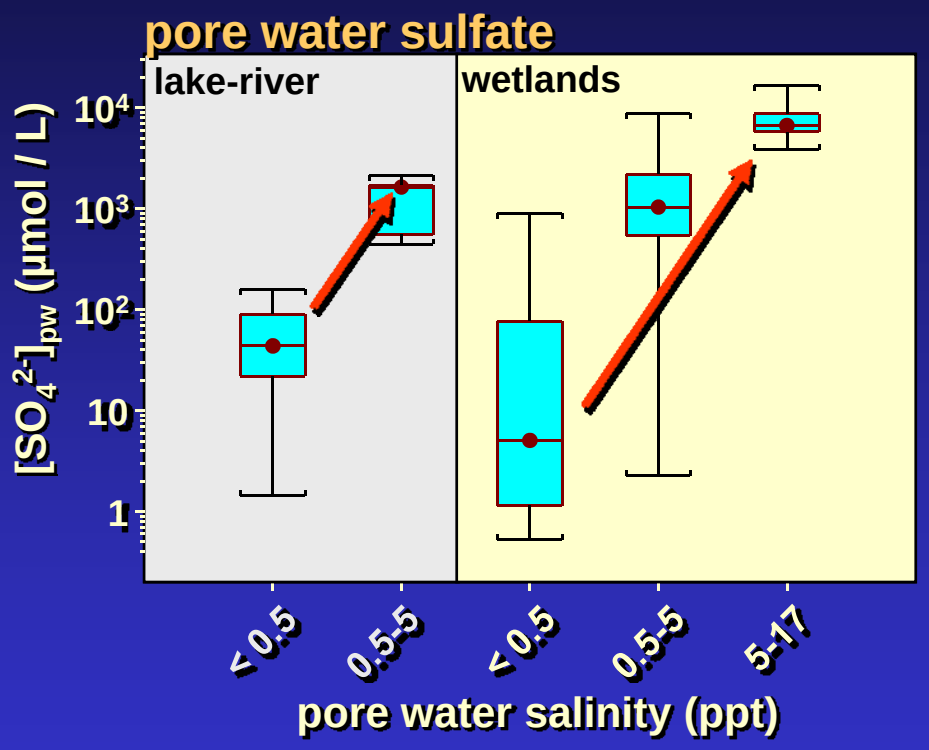


- $^{203}Hg(II)$ Incubations: method for assessing (k_{meth})
- Reactive mercury ($Hg(II)_R$): surrogate for $Hg(II)_{bio}$

Sediment: All Data (2003-2006)

Habitat Type and Salinity Range

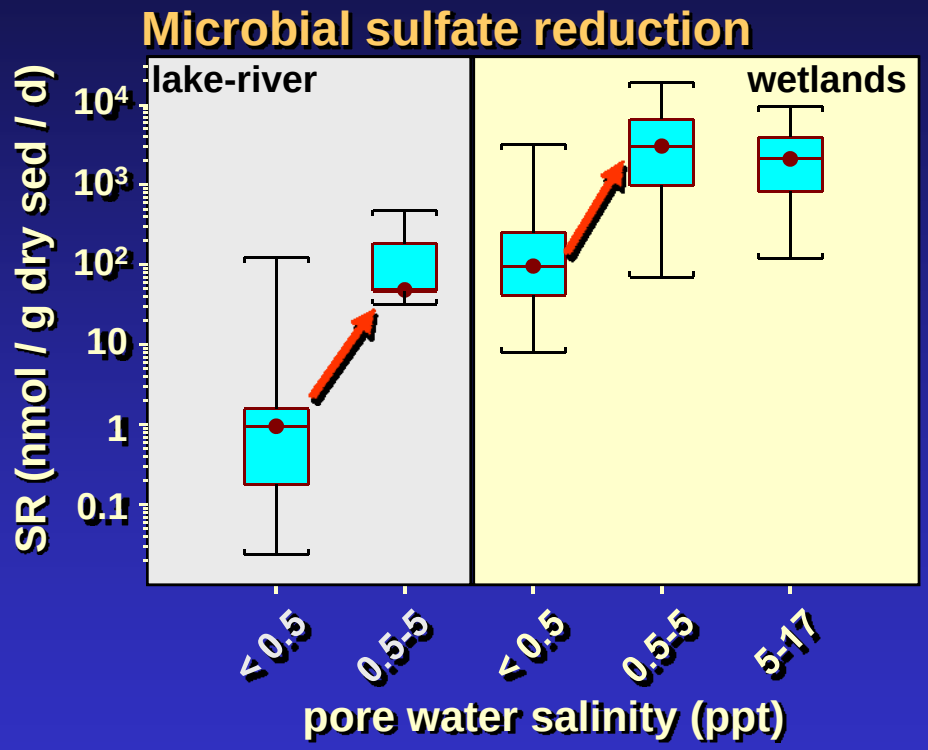


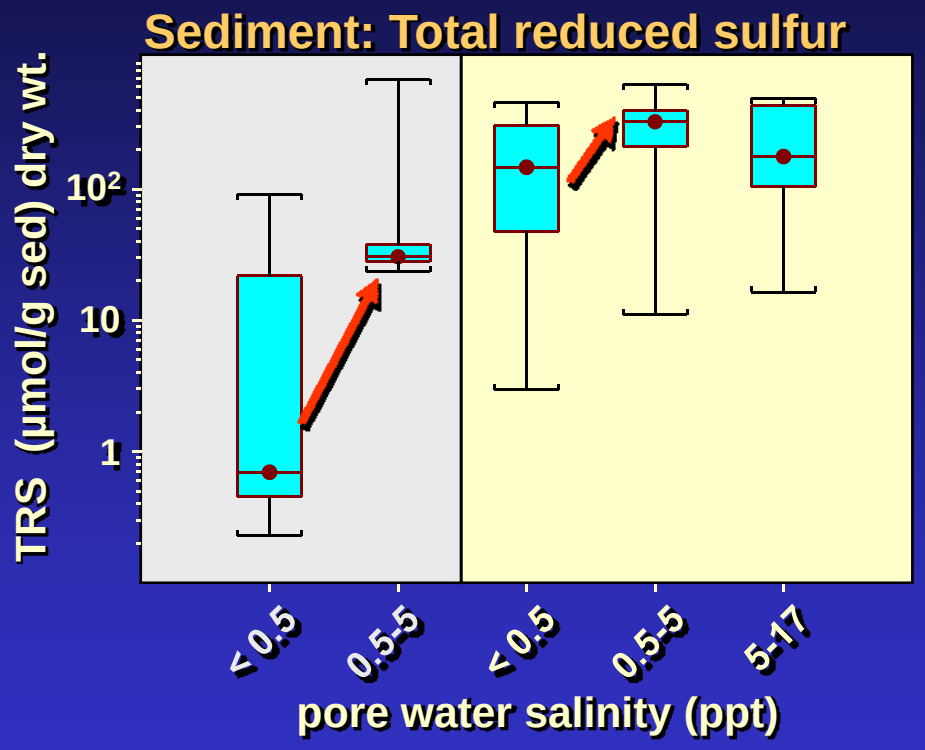


- pore water salinity = $[Cl^-]$ + $[SO_4^{2-}]$ (mg/L = ppt)
- [Sulfate]: drives microbial sulfate reduction (SR)
- SR: a key microbial process in MeHg formation

Sediment: All Data (2003-2006)

Habitat Type and Salinity Range

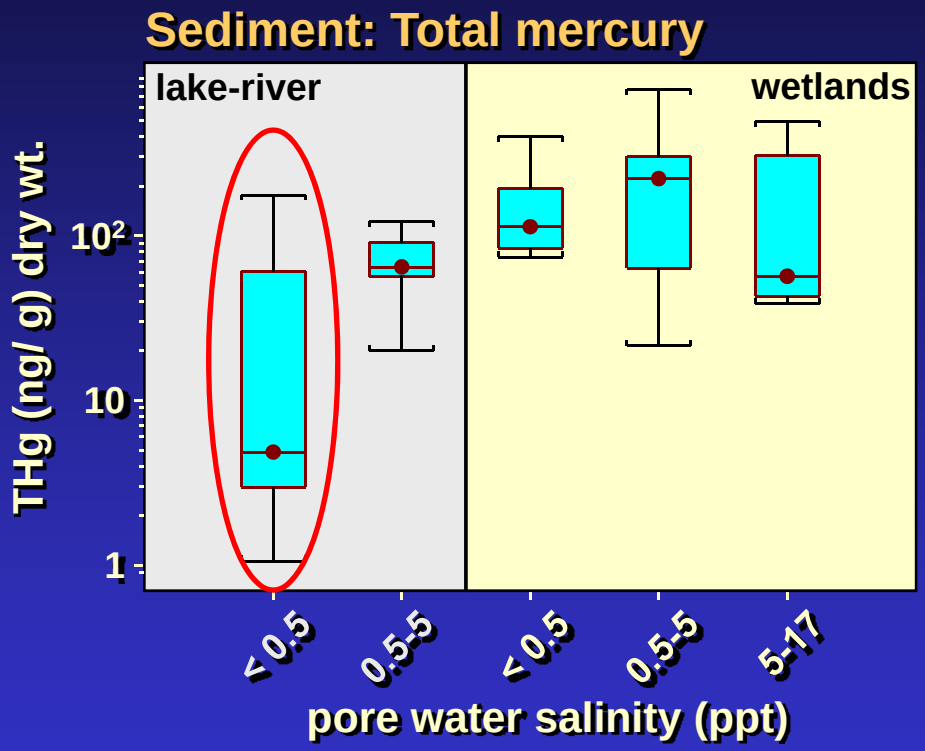


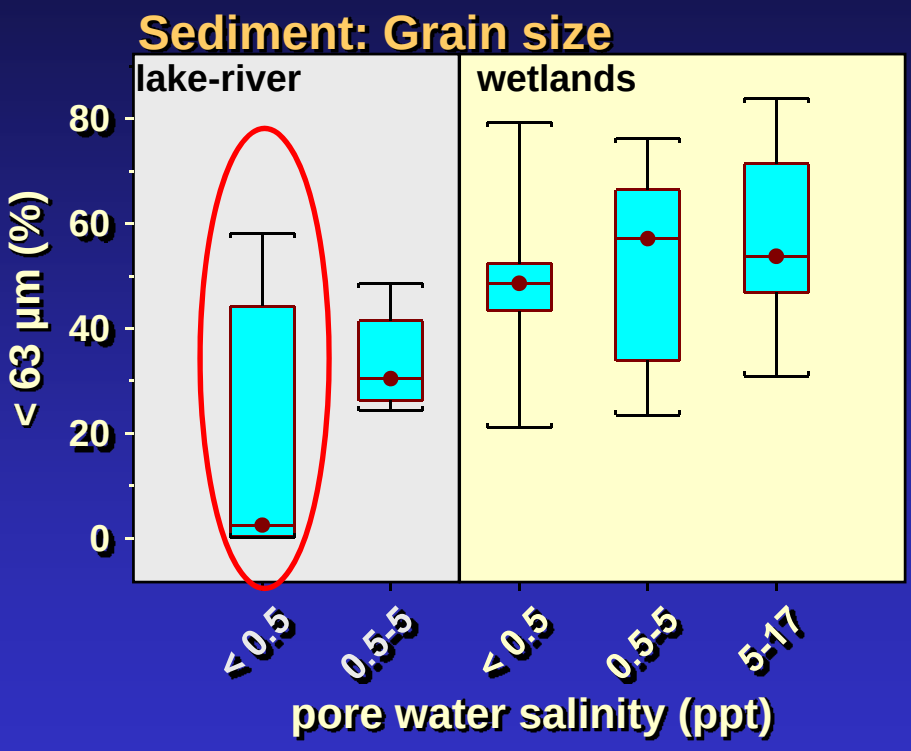


- SR an TRS are generally higher in wetland sites
- both increase from fresh (< 0.5 ppt) to low brackish (0.5-5 ppt)

Sediment: All Data (2003-2006)

Habitat Type and Salinity Range

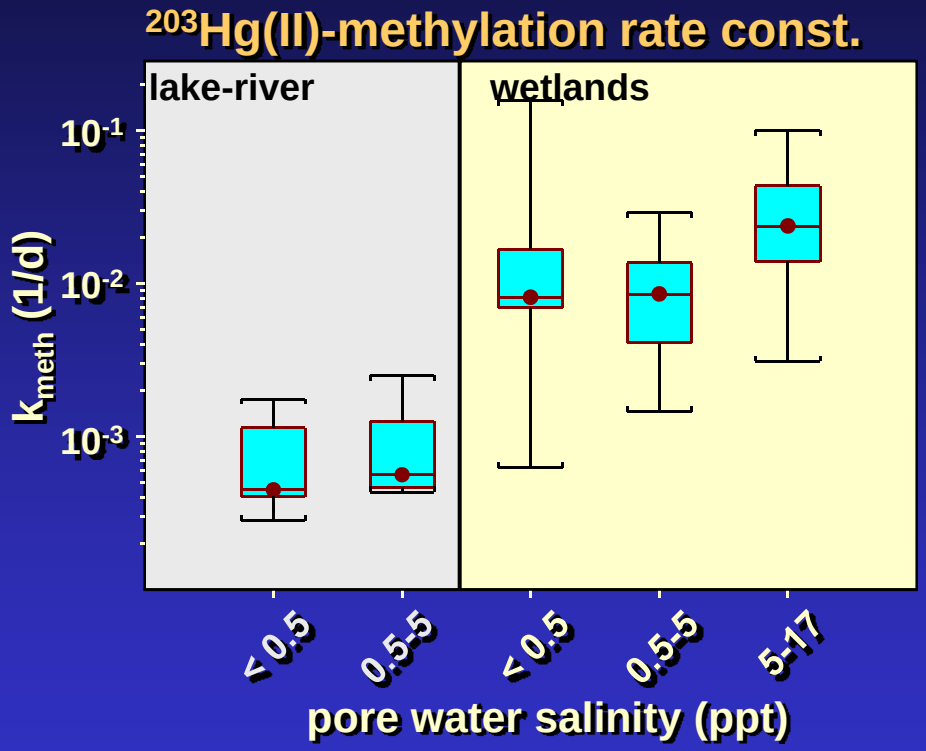


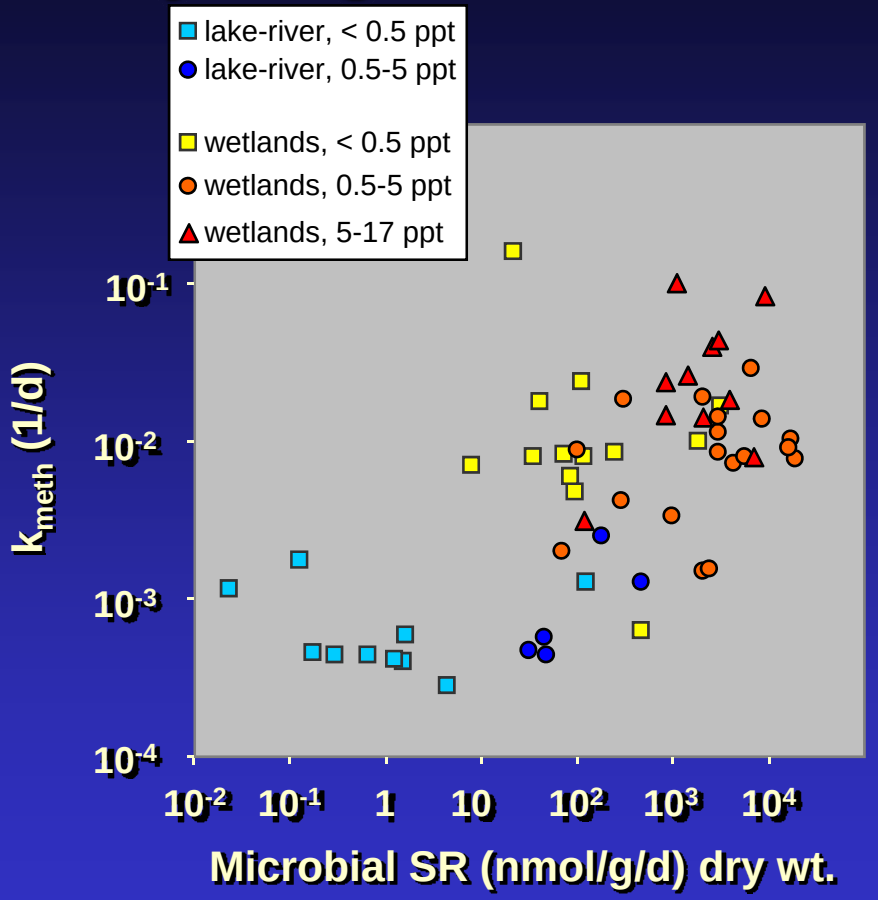


- THg: lowest in upper freshwater rivers, highest in depositional areas
- tracts sediment grain size distribution

Sediment: All Data (2003-2006)

Habitat Type and Salinity Range

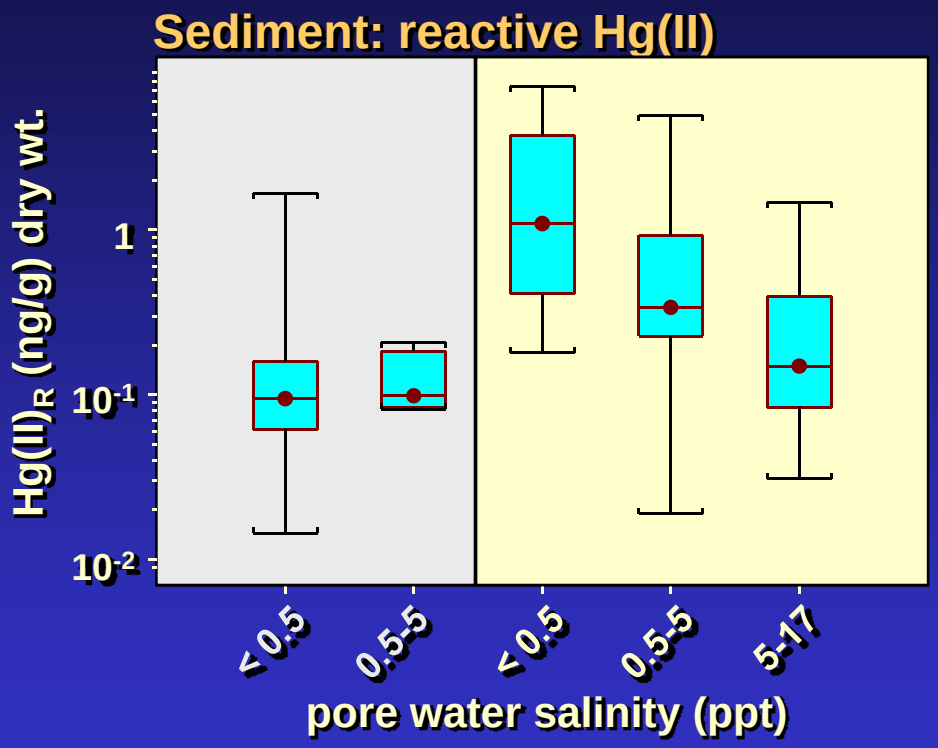


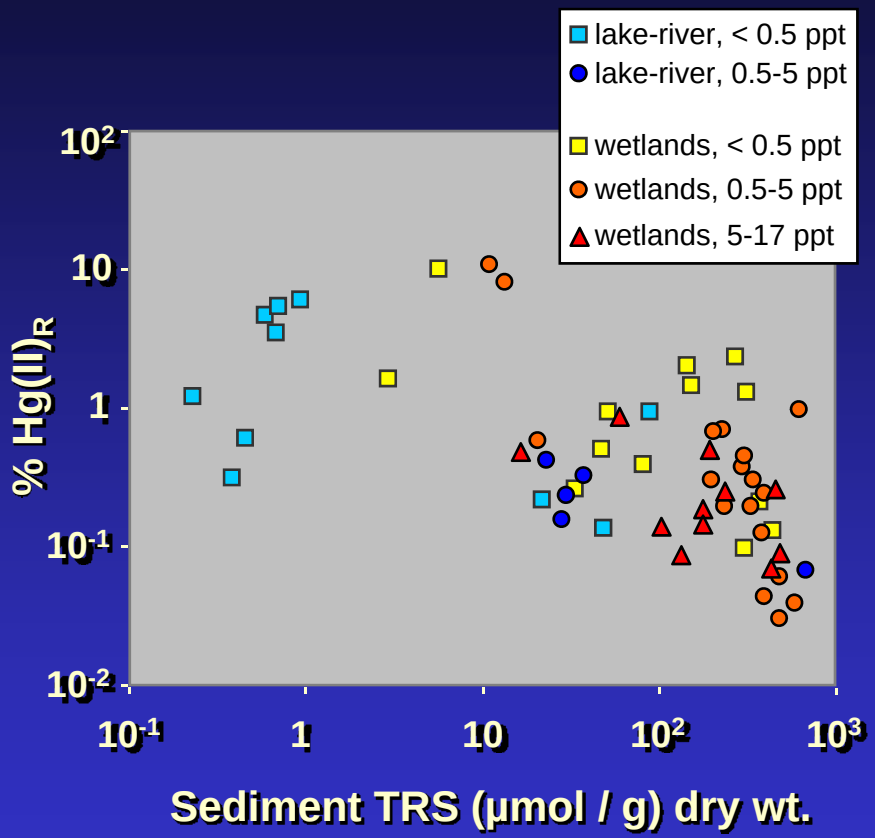


- Microbial Hg(II)-methylation activity (k_{meth}) highest in wetlands
- Increases with microbial sulfate reduction

Sediment: All Data (2003-2006)

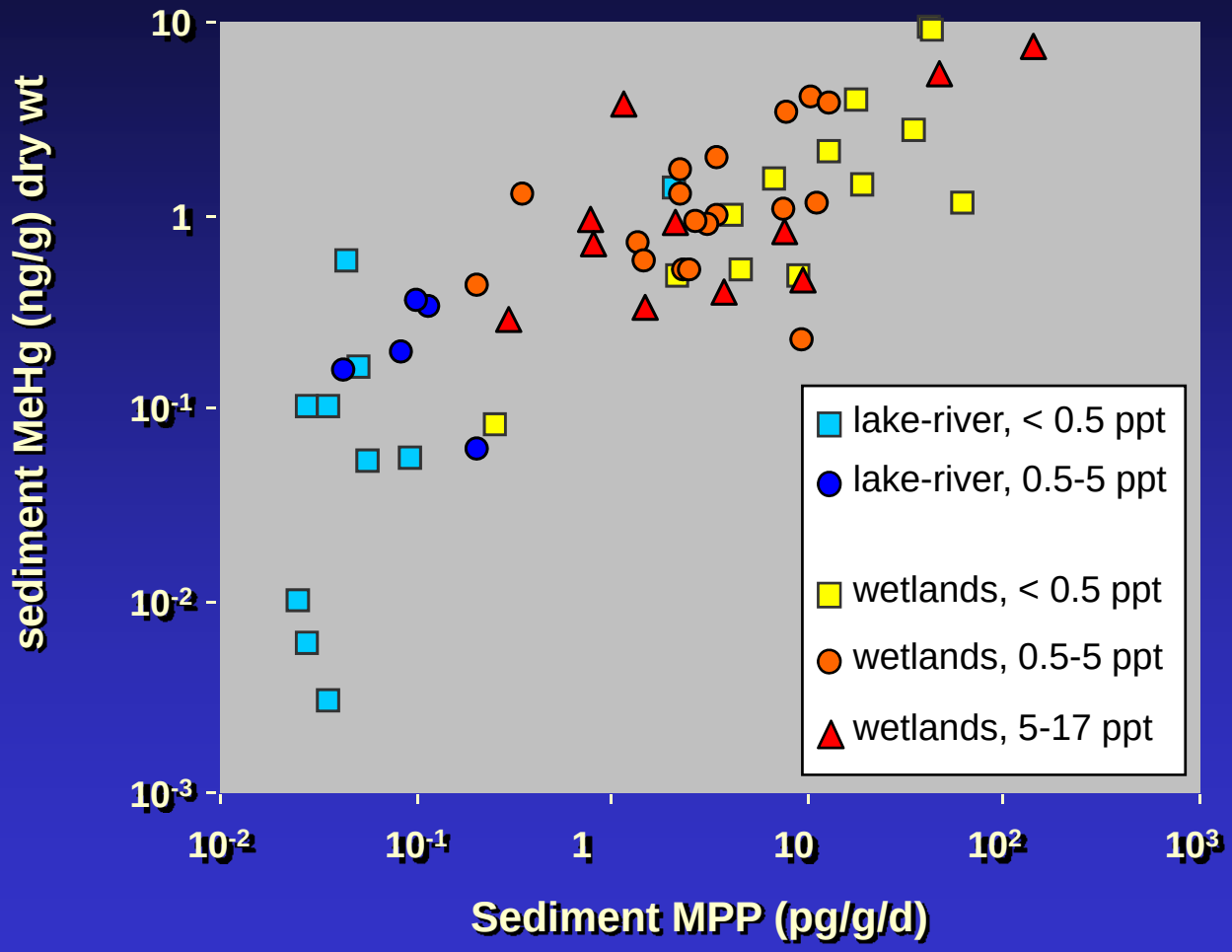
Habitat Type and Salinity Range





- Microbially available 'reactive' Hg(II): highest in wetlands
- $\%Hg(II)_R$ decreases with with increasing TRS concentration

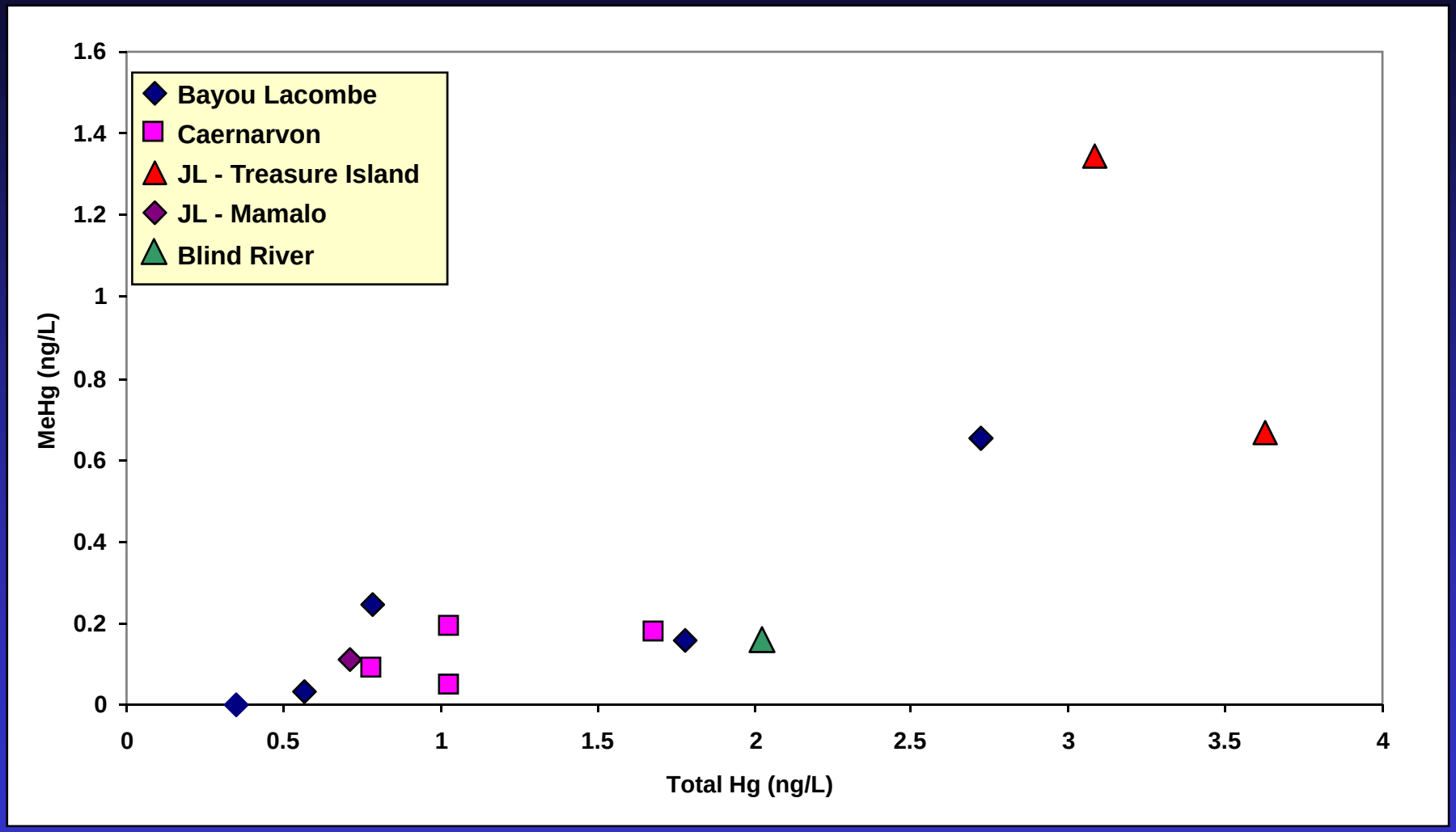
MPP = k_{meth} x $Hg(II)_R$: Predicts sediment MeHg concentration



- Much better than k_{meth} or $Hg(II)_R$ individually (not shown)

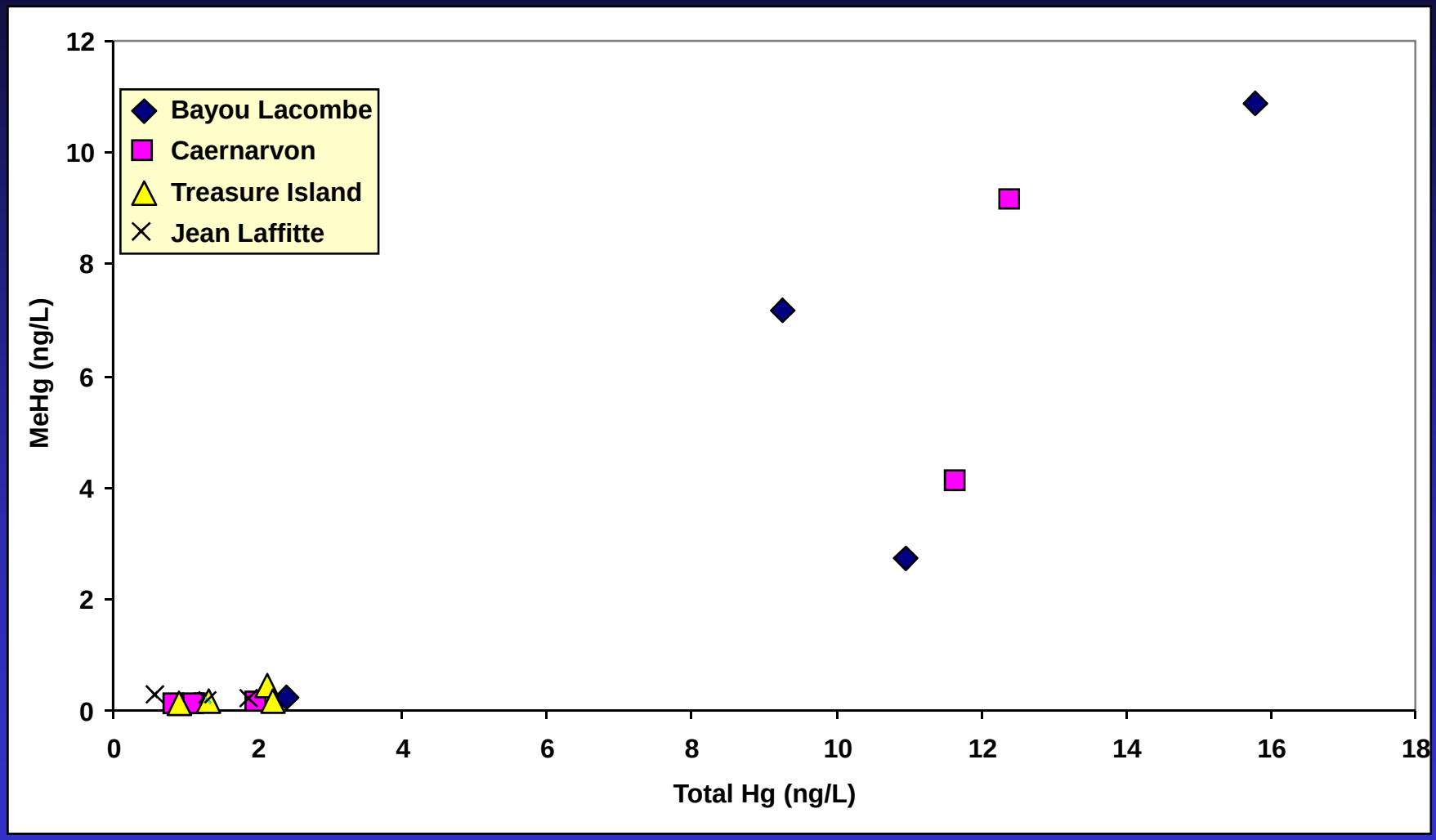
Surface Water: THg and MeHg

Sept. 2006 data



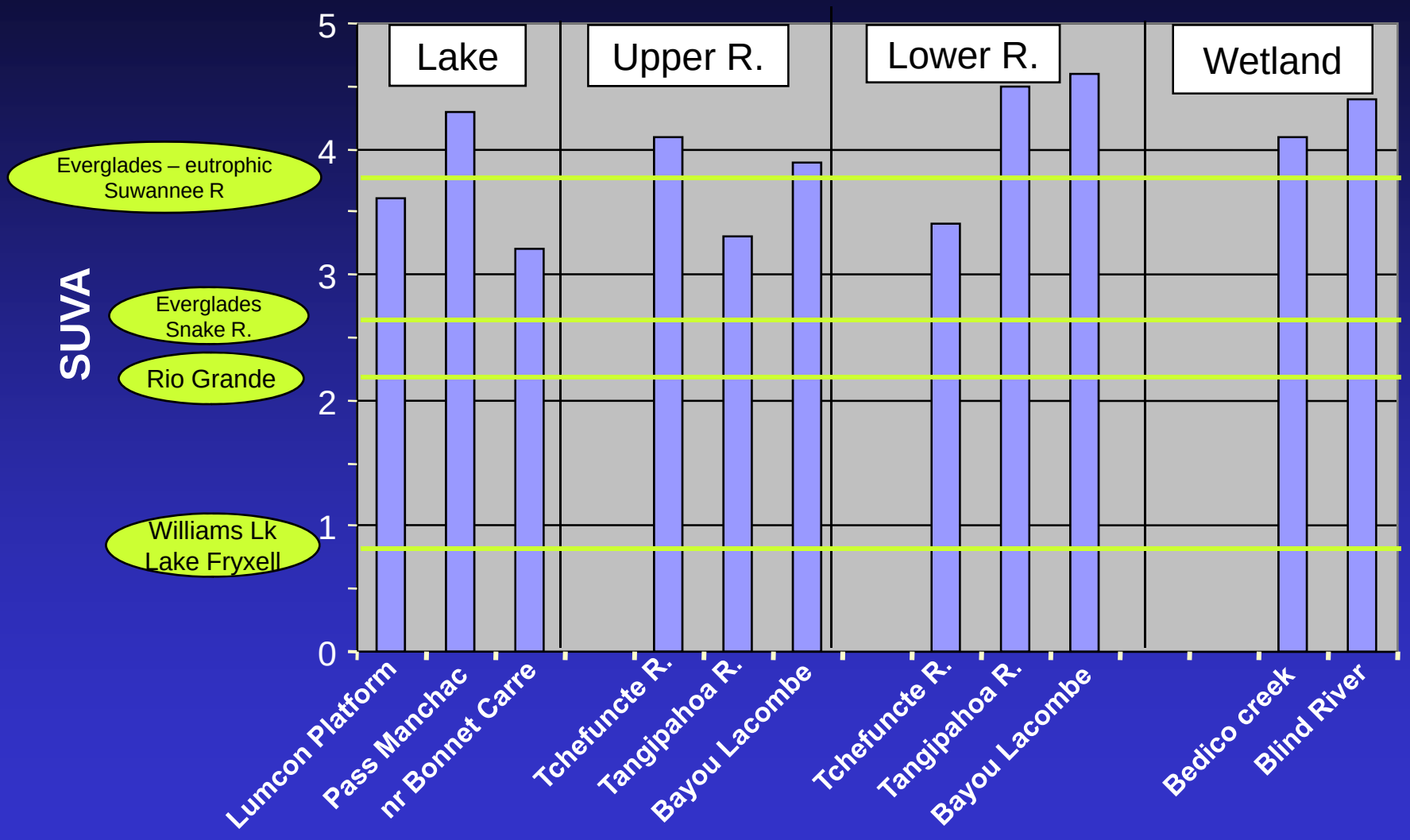
Sediment Pore water: THg and MeHg

Sept. 2006 data



S.E. Louisiana: August ’03

Overlying Water DOC is very Aromatic Stuff!

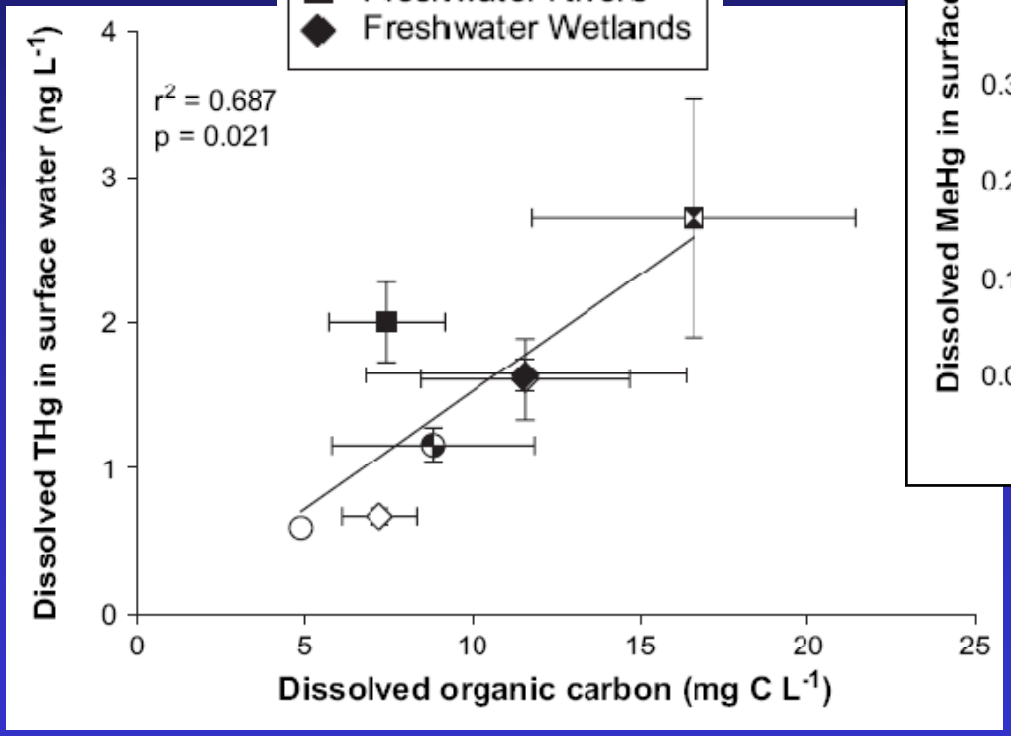


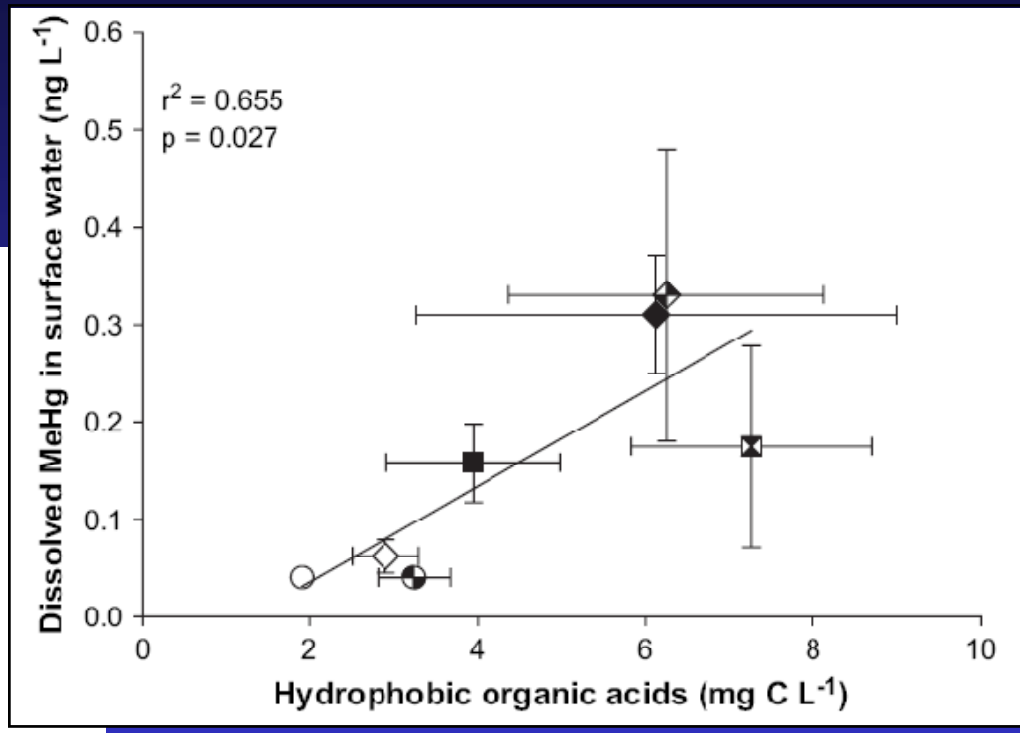
- SUVA = measure of DOC ‘aromaticity’

Surface Water: THg and MeHg

2003-2005 data

Hall et al. 2008, Environ. Pollut., vol. 154, p. 124-134

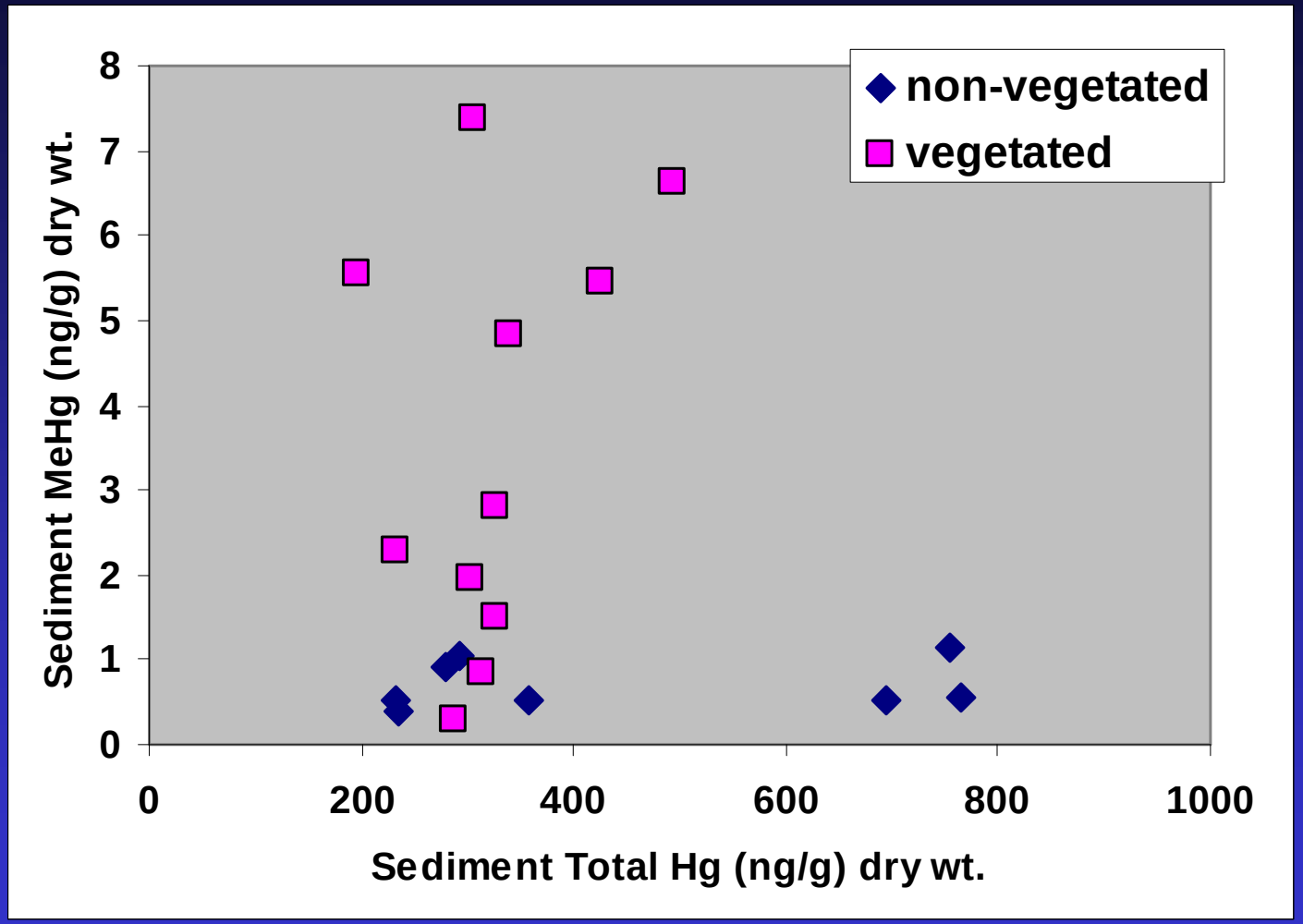




MeHg is NOT correlated with DOC, but with HPOA (aromatic DOC)

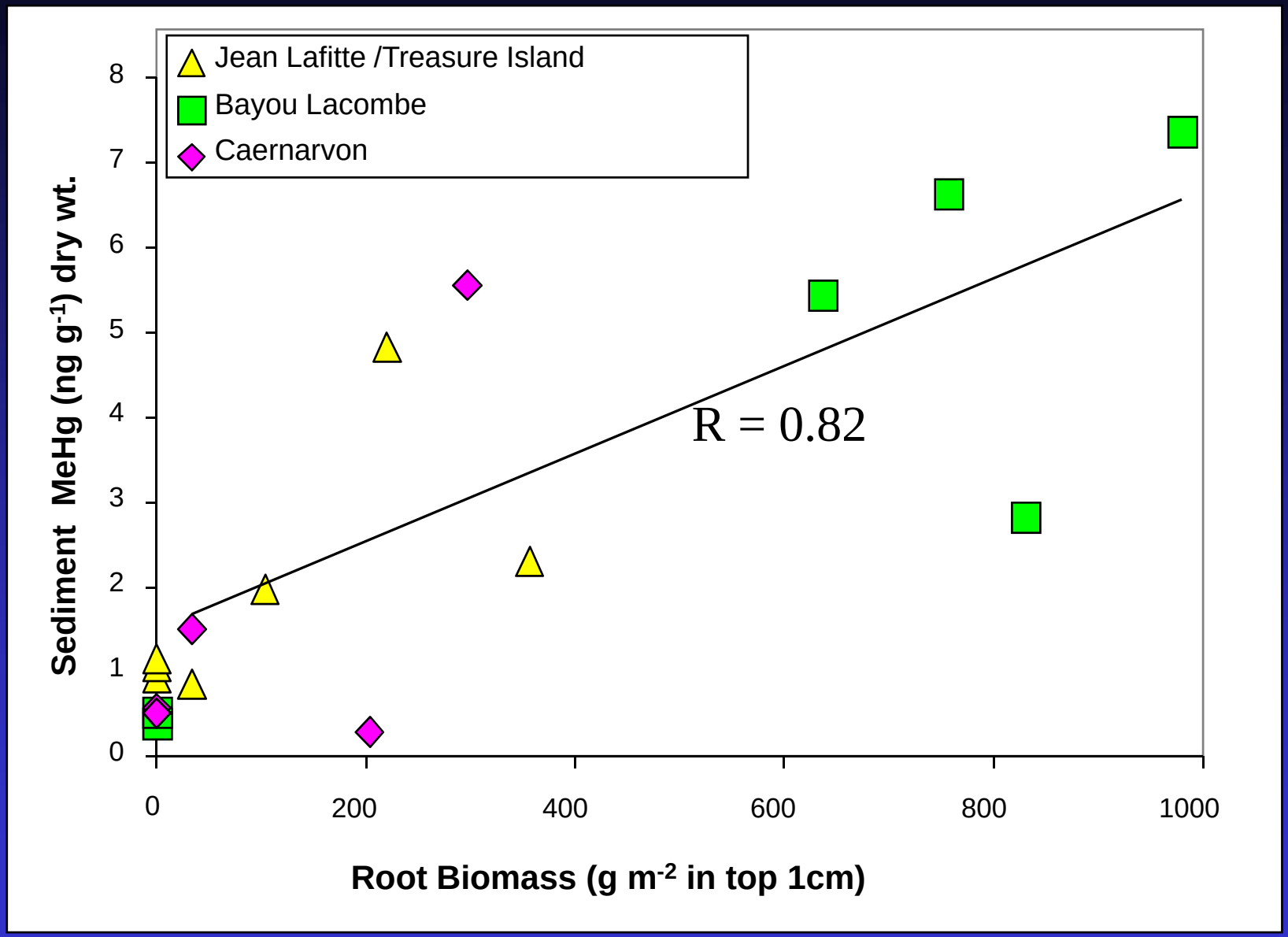
- **THg is correlated with DOC**

Wetlands: vegetated sites have higher sediment MeHg concentrations



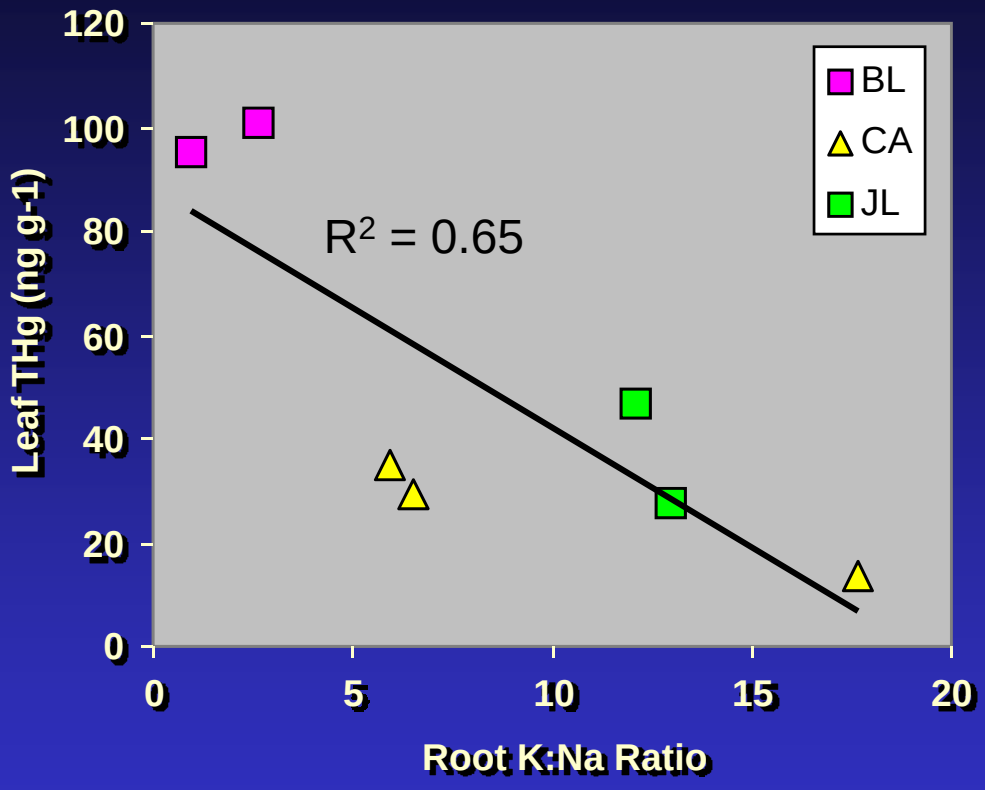
- Sept. 2006 data only

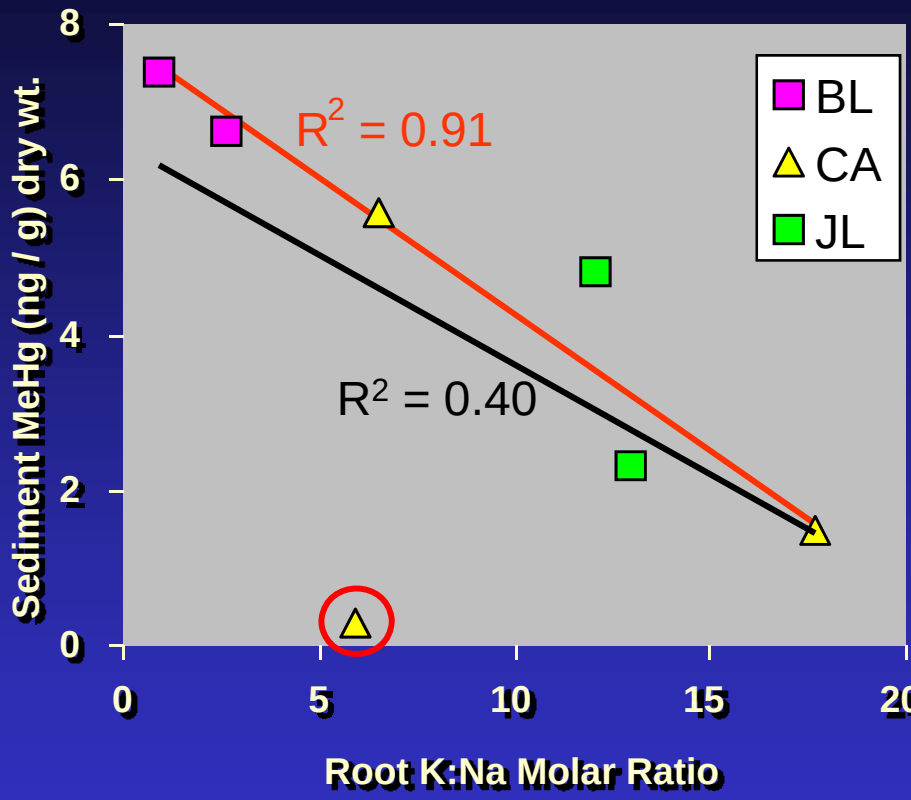
Root Biomass correlated with sediment [MeHg]



- Sept. 2006 data only

K:Na Ratio: Plant Stress Indicator





- Low ratio = stressed conditions
- Potential (cheap) indicator of wetland MeHg production ‘hot spots’?

Take Home Message

- The Gulf coast region is a 'hot spot' for Hg deposition and elevated Hg blood levels in humans
- May be linked to diet and/or regional enhancement of MeHg in the local food chain
- MeHg production is a function of BOTH microbial activity (k_{meth}) and Hg(II) availability ($Hg(II)_R$)
  - k_{meth} increases with microbial SR (highest in saltmarshes)
  - $Hg(II)_R$ highest in freshwater wetlands; decreases with TRS
- Habitat type is a key factor controlling MeHg production, with wetlands as 'hot spots'
- Surface water MeHg transport is associated with aromatic DOC
- Emergent wetland plant stress index appears associated with regional MeHg production potential

Future Directions / Collaborative Opportunities

- More detailed synoptic freshwater → marine transects
- Explore further the use of wetland plant type, stress indicators and seasonal life cycles (e.g. senescence) as a predictor of MeHg 'hot spots' → extrapolate with GIS mapping
- Add biota components to current Hg investigations
- Examine fundamental relationships between Hg and C cycling (large geographic relevance)
- Further explore coastal regions as zone of enhanced reactive Hg(II) deposition
- Managed vs non-managed wetlands