

Aquatic Ecology Research and Technology Development in East Fork Poplar Creek

Teresa Mathews
Environmental Sciences Division
Oak Ridge National Laboratory
mathewstj@ornl.gov
April 10, 2024

ORNL is managed by UT-Battelle, LLC for the US Department of Energy

Key Collaborations and Partnerships

- UCOR/RSI Water Resources Restoration Program
- Y-12 CNS Compliance Organization
- Y-12 Biological Monitoring and Abatement Program
- DOE Office of Science
 - Science Focus Area at ORNL
 - Joint project with U. Michigan
- Mercury Applied Field Research Initiative (AFRI)
- UT/ORNL Carbon Fiber Tech Facility
- South River Science Team
- DuPont
- USGS
- Queens University
- James Madison University
- MSIPP – New Mexico State University
- Smithsonian Environmental Research Center
- U. Minnesota
- RT GeoSciences, Canada
- Flinders University, Australia



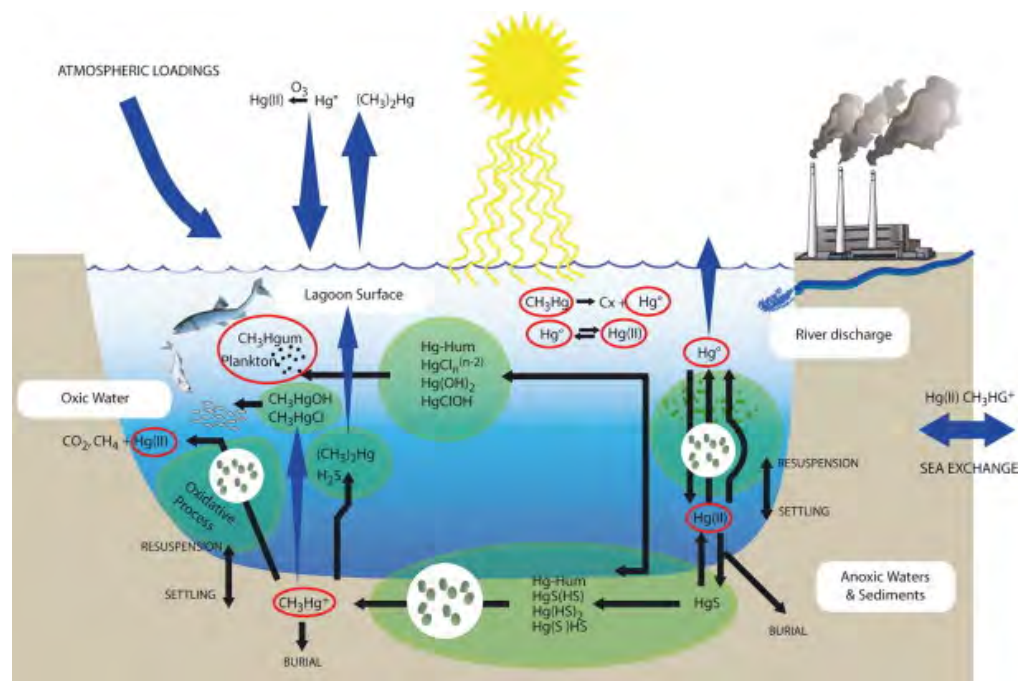
Smithsonian Environmental
Research Center



Outline

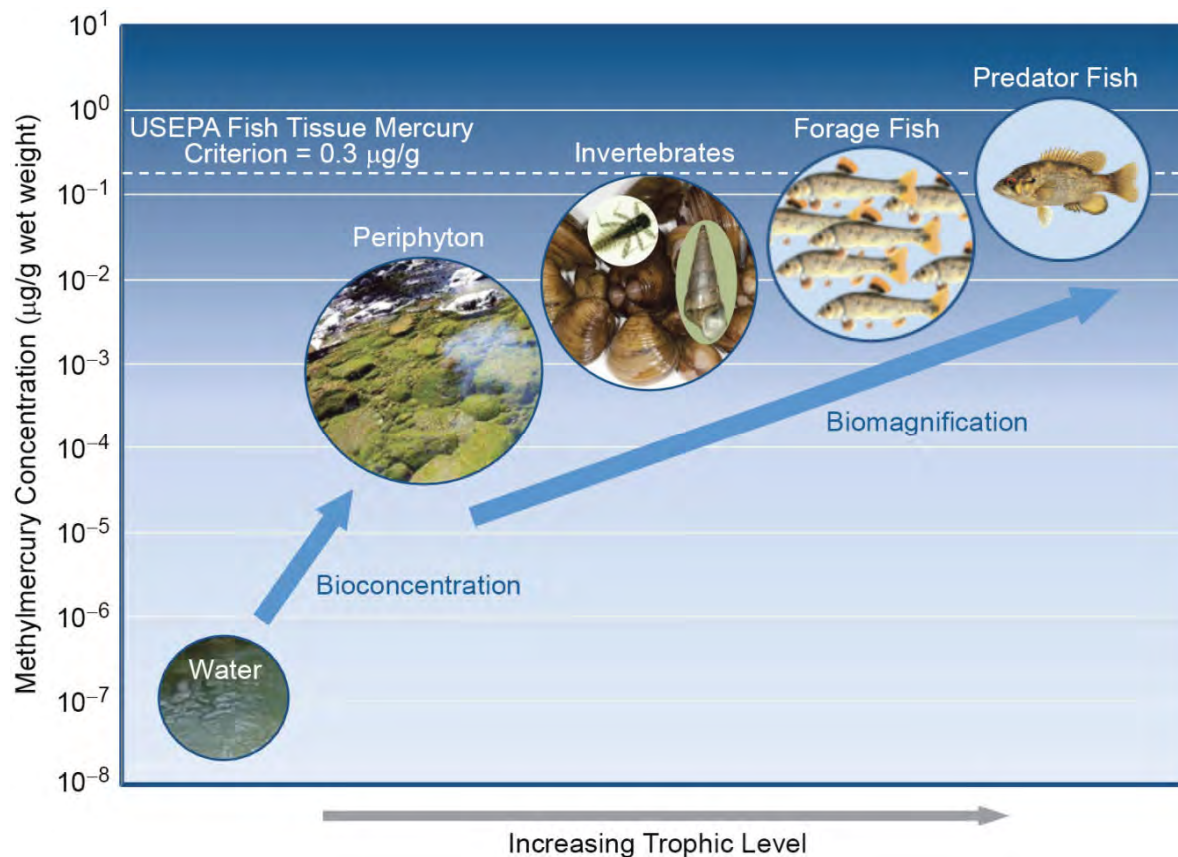
- Mercury as a global pollutant
- Mercury in Oak Ridge
- Mercury Remediation Technology Development in Oak Ridge
- Future directions

Mercury in the environment



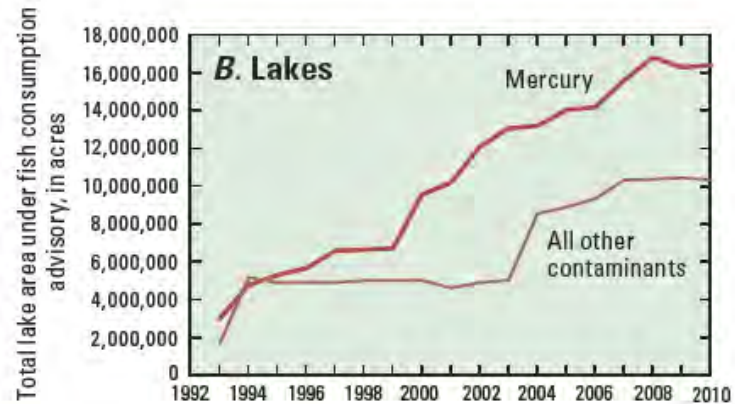
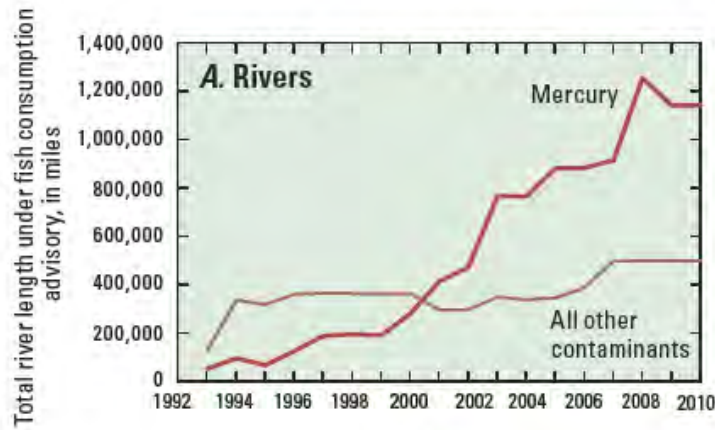
- Mercury is a global pollutant
- Mercury's cycle is complex, affecting risk but also affecting remediation
- Mercury risk and toxicity is intrinsically linked with aquatic ecosystems

Food web transfer of mercury

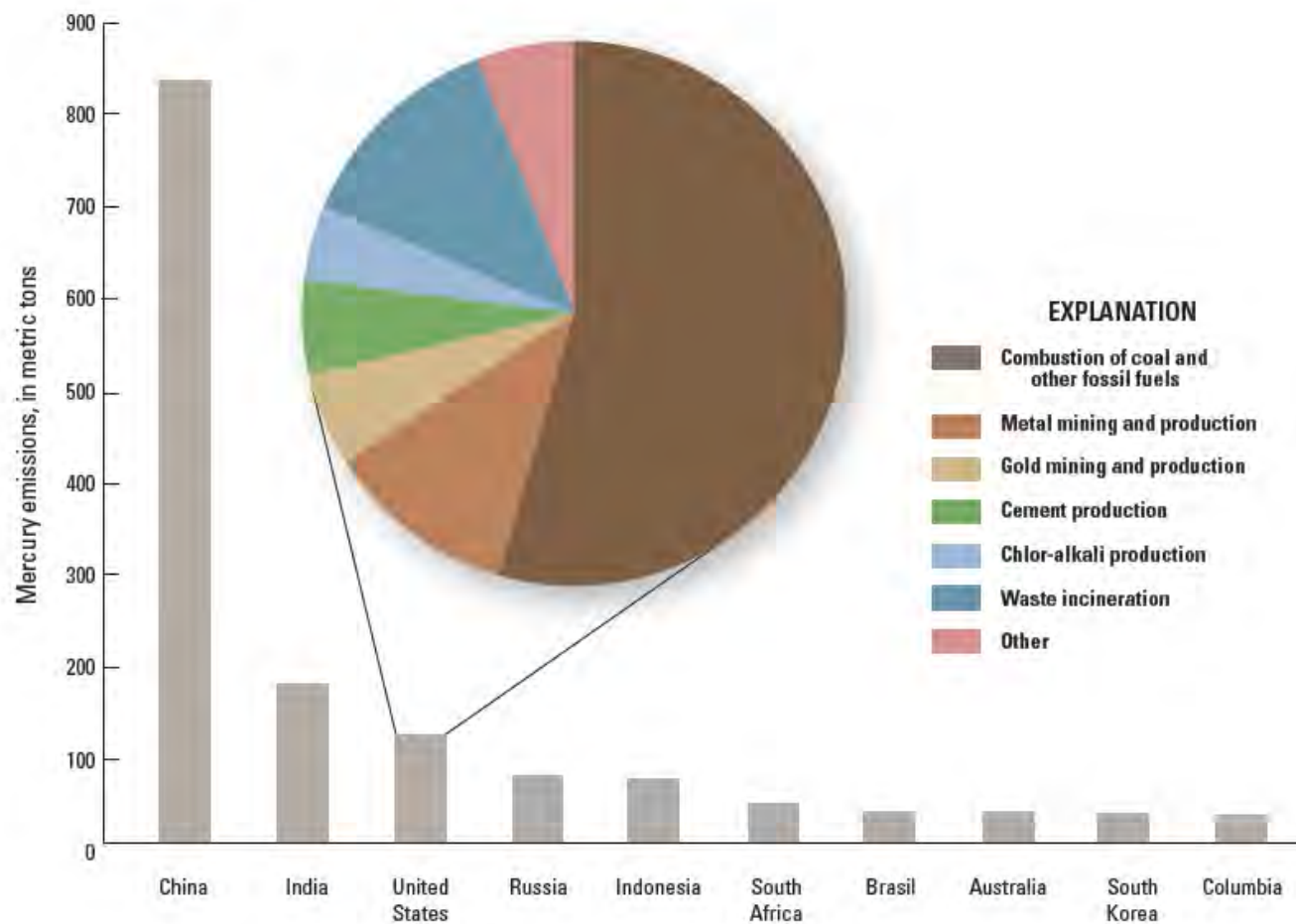


- Mercury biomagnifies as it is transferred up the food chain, leading to high Hg concentrations in fish
- This means that relatively low aqueous Hg concentrations can lead to hazardous concentrations in fish

Mercury contamination is widespread



Global mercury emissions



Mercury in Oak Ridge

- Early 20th century, Oak Ridge was an agricultural area
- Large amounts (> 10 million kg) of inorganic Hg were used for industrial processes in 50's and 60's
- Spills and releases of Hg contaminated creeks, floodplains, and downstream sediments
- Hg remediation has focused on source control (water treatment systems, sewer relining, pipe re-routing, soil removal)



Pre-industrial period steady state conditions, eastern US

Hg Air: 0.5 ng/m³

Hg deposition rate: 3.0 µg/m²/yr

Hg Water: 0.35 ng/L
Hg Fish: 0.24 µg/g

EPA aquatic and terrestrial fate, transport, and exposure model (IEM-2M Model)

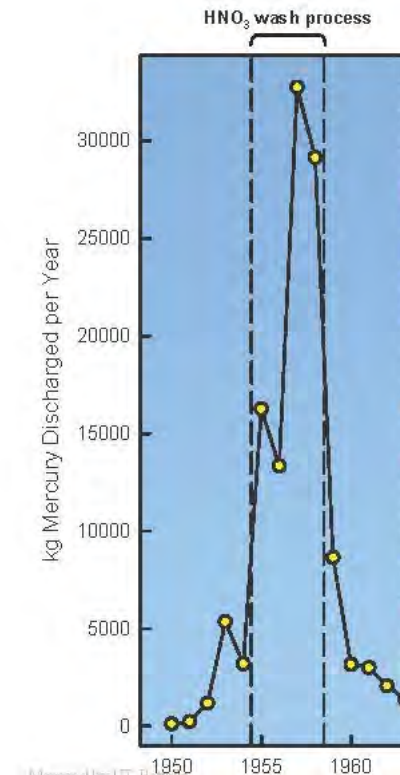
Mercury in Oak Ridge

- Early 20th century, Oak Ridge was an agricultural area
- Large amounts (> 10 million kg) of inorganic Hg were used for industrial processes in 50's and 60's
- Spills and releases of Hg contaminated creeks, floodplains, and downstream sediments
- Hg remediation has focused on source control (water treatment systems, sewer relining, pipe re-routing, soil removal)



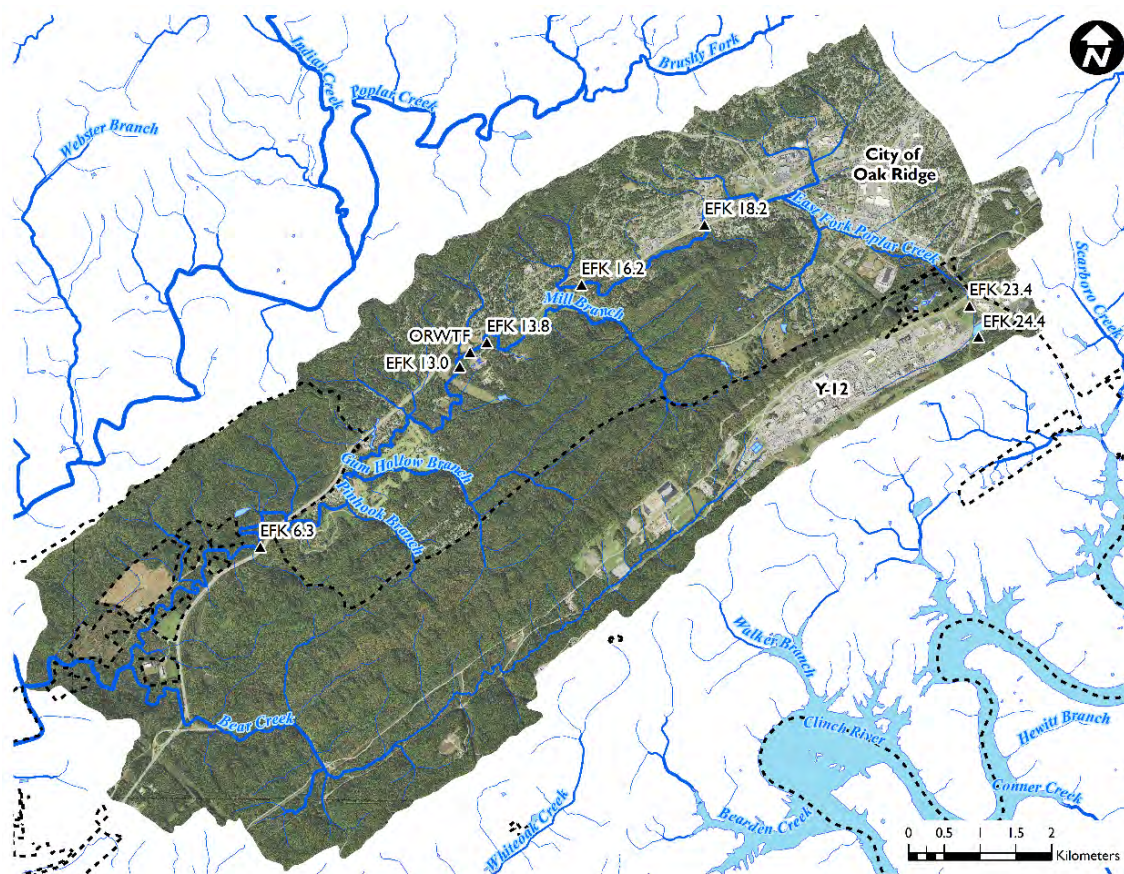
Mercury in Oak Ridge

- Early 20th century, Oak Ridge was an agricultural area
- Large amounts (> 10 million kg) of inorganic Hg were used for industrial processes in 50's and 60's
- Spills and releases of Hg contaminated creeks, floodplains, and downstream sediments
- Hg remediation has focused on source control (water treatment systems, sewer relining, pipe re-routing, soil removal)



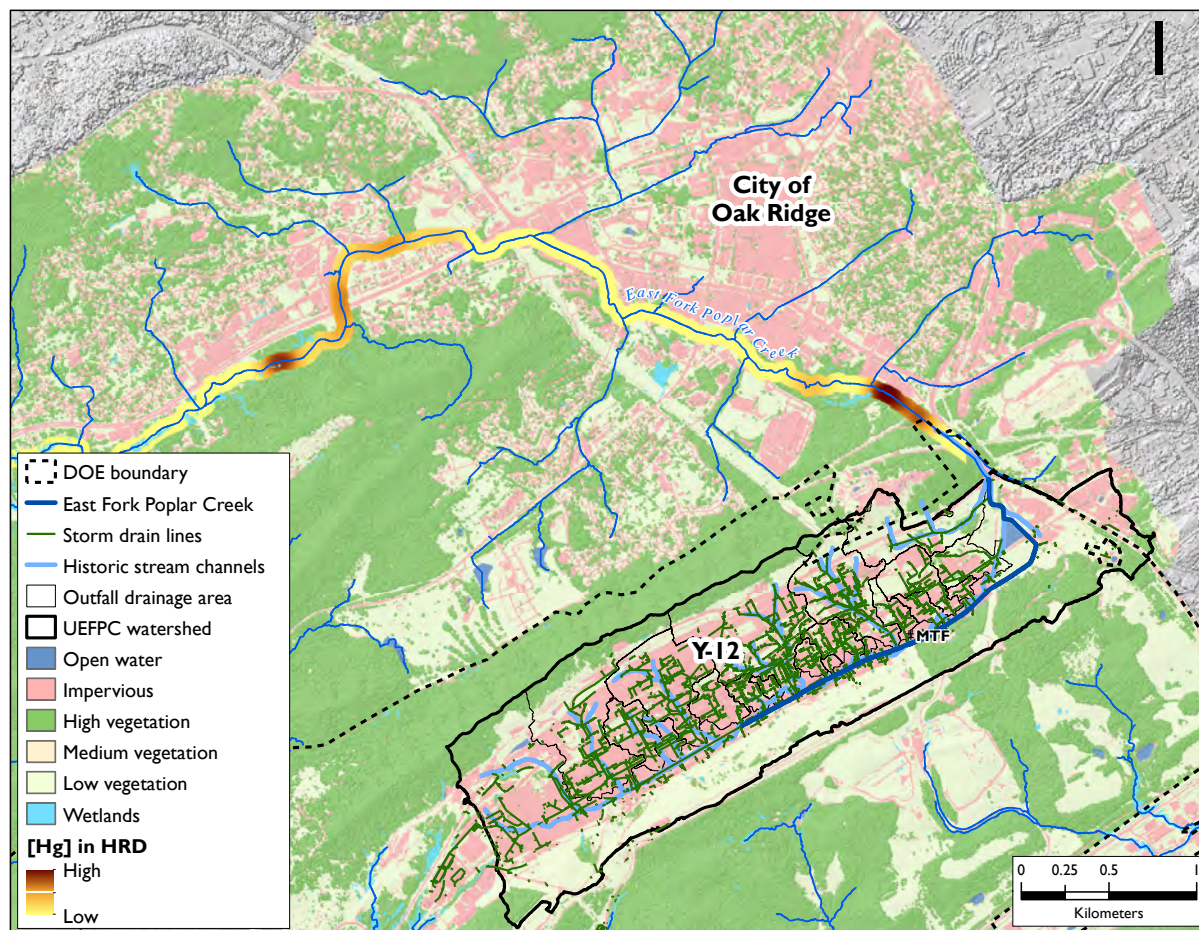
Brooks and Southworth 2011. History of mercury use and environmental contamination at the Oak Ridge Y-12 Plant. *Environmental Pollution* 159; 219-228.

East Fork Poplar Creek (EFPC)



- EFPC is a 25 km stream originating in Y-12 NSC
- Mercury use in the 1950's-60's resulted in contaminated soils, water, and fish
- Actions taken over the past four decades have significantly improved water quality in the stream, but issues still remain

East Fork Poplar Creek (EFPC)

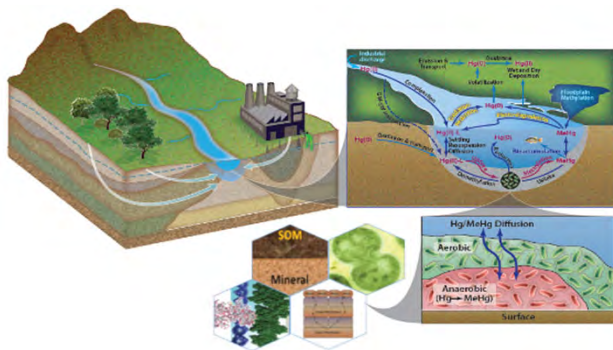


- EFPC is a 25 km stream originating in Y-12 NSC
- Mercury use in the 1950's-60's resulted in contaminated soils, water, and fish
- Actions taken over the past four decades have significantly improved water quality in the stream, but issues still remain

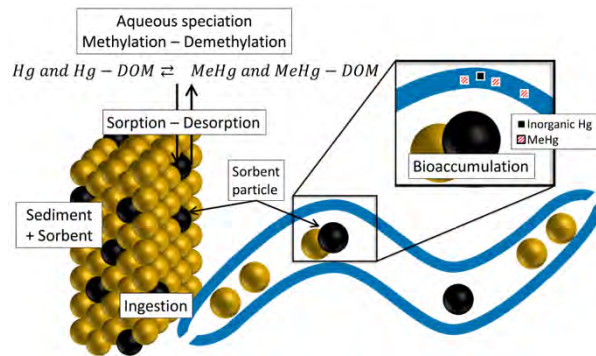
DOE Environmental Mercury Research Spans Technology Pipeline



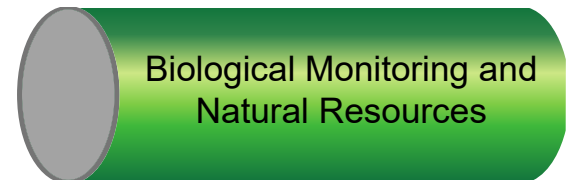
DOE SC



OFFICE OF ENVIRONMENTAL MANAGEMENT OREM

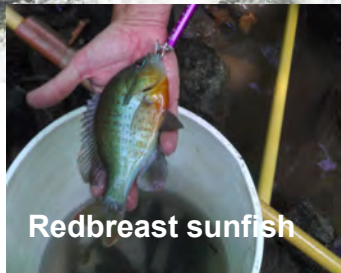
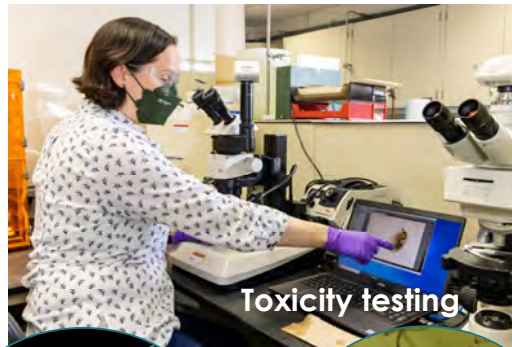


OFFICE OF ENVIRONMENTAL MANAGEMENT Y-12 Compliance



Long term biological monitoring on the Oak Ridge Reservation

- Objectives of the biological monitoring program are to:
 - Ensure DOE activities are protective of aquatic life
 - Assess ecological impacts, identify causes, and evaluate effectiveness of pollution abatement actions
 - Monitor ecological recovery



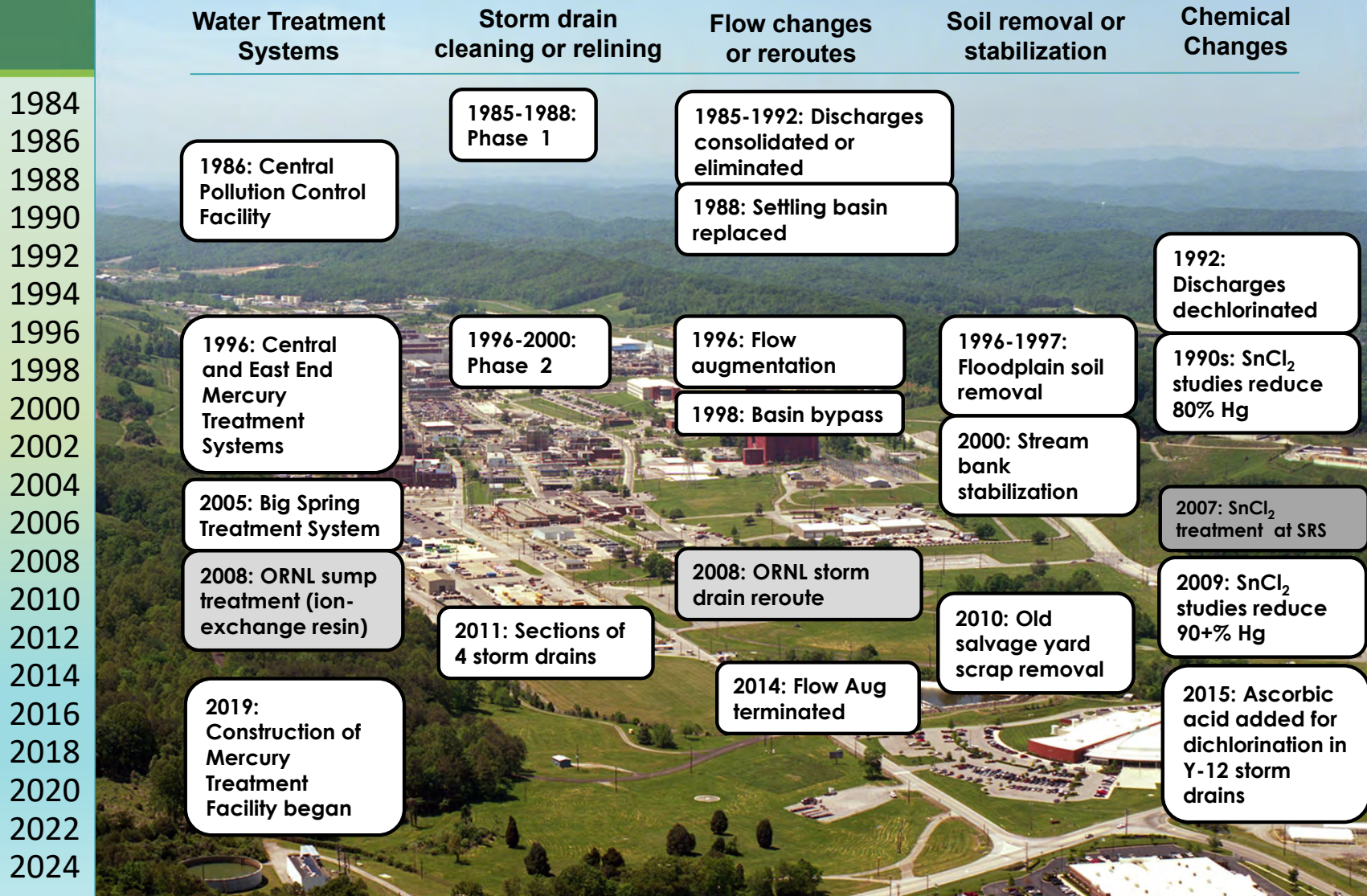
Ceriodaphnia dubia



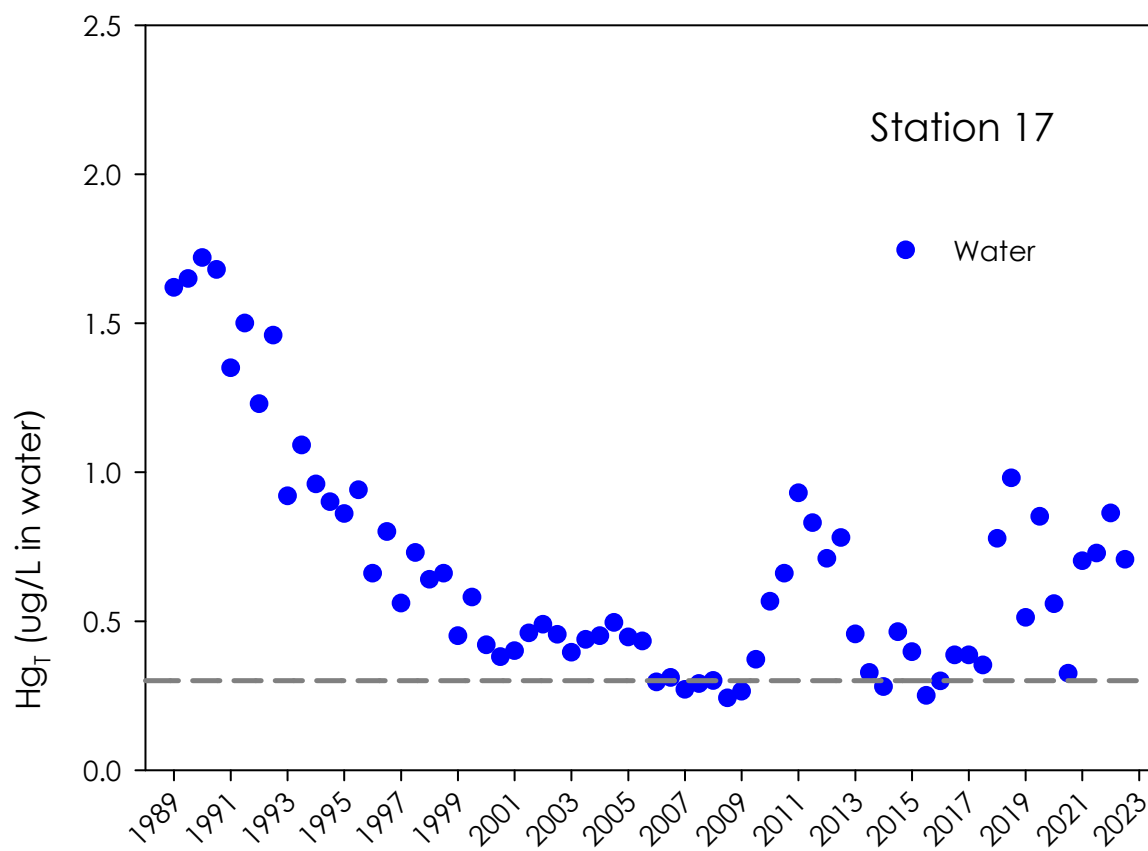
Fathead minnow



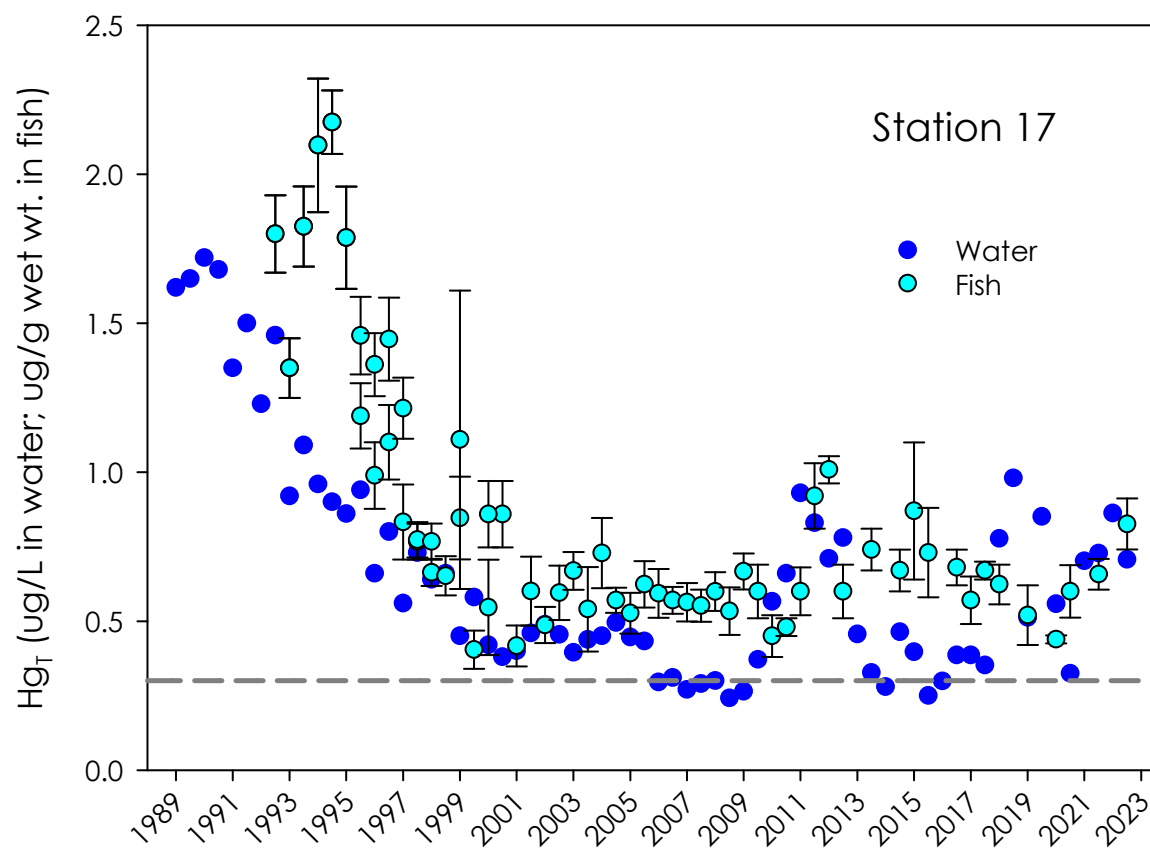
Timeline of actions taken, mostly at Y-12



In response to treatment, aqueous mercury has decreased

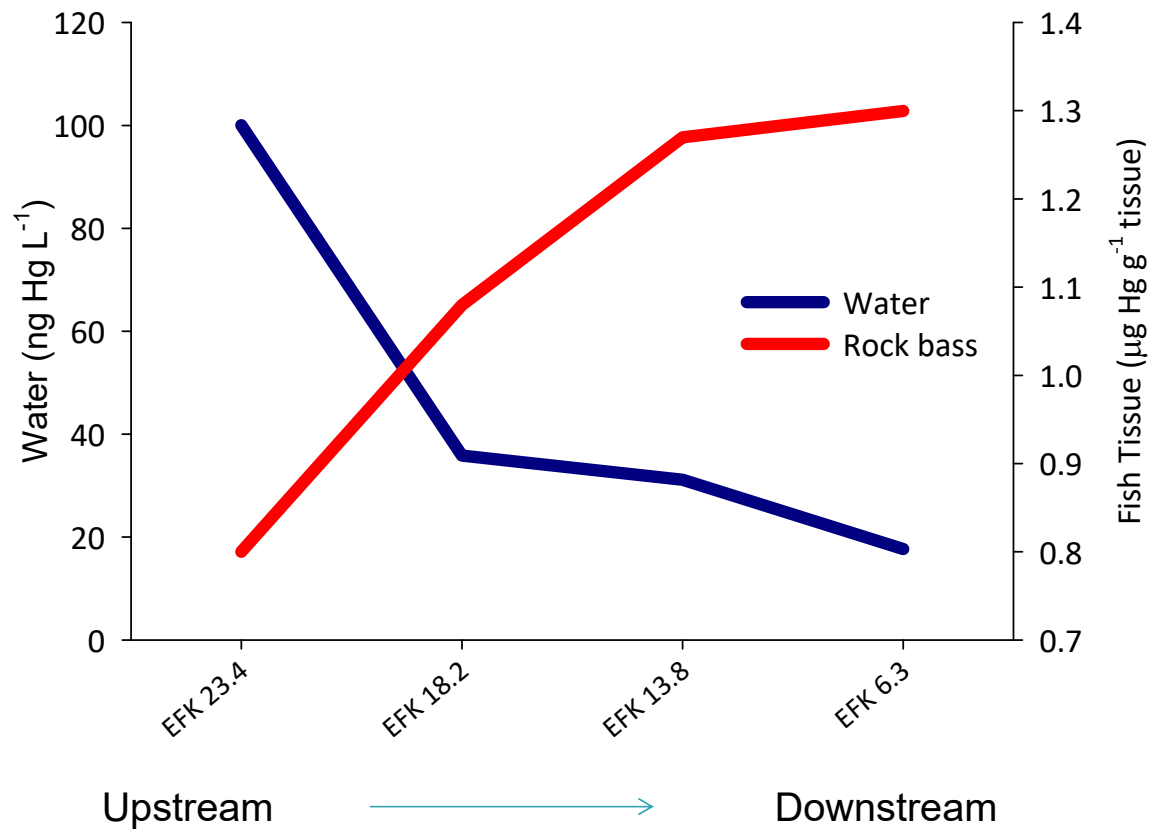


In response to treatment, aqueous mercury has decreased
but mercury in fish remains elevated

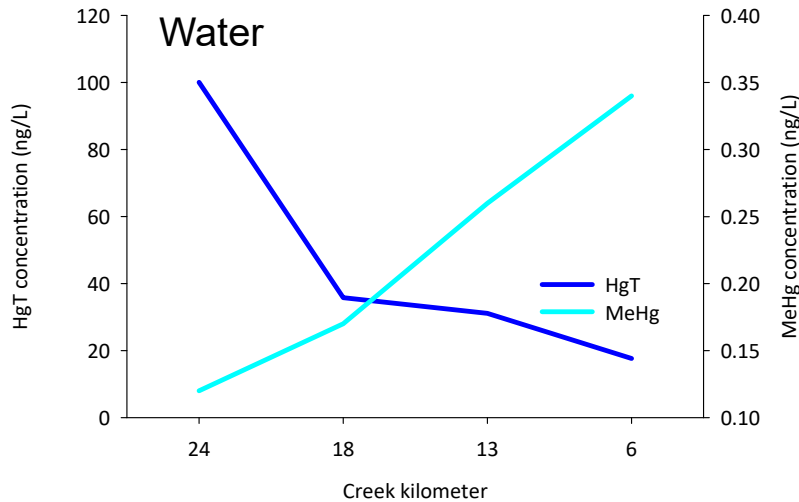


Spatial trends in mercury concentrations

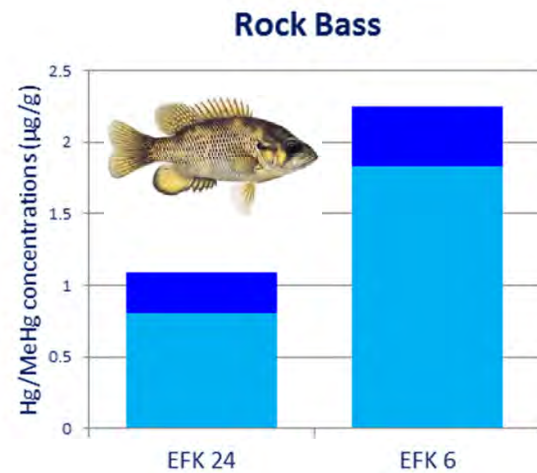
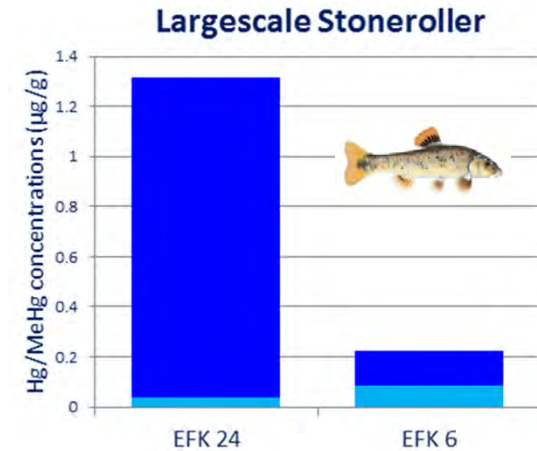
Mercury concentrations in water decrease further away from source, but fish tissue concentrations increase



Mercury methylation is controlling fish concentrations



- While HgT concentrations decrease with further distance downstream, MeHg concentrations increase
- Hg in lower trophic level fish reflect aqueous HgT concentrations, while Hg in higher trophic level fish reflects aqueous MeHg concentrations
- Upper trophic level fish have higher MeHg concentrations downstream compared to upstream

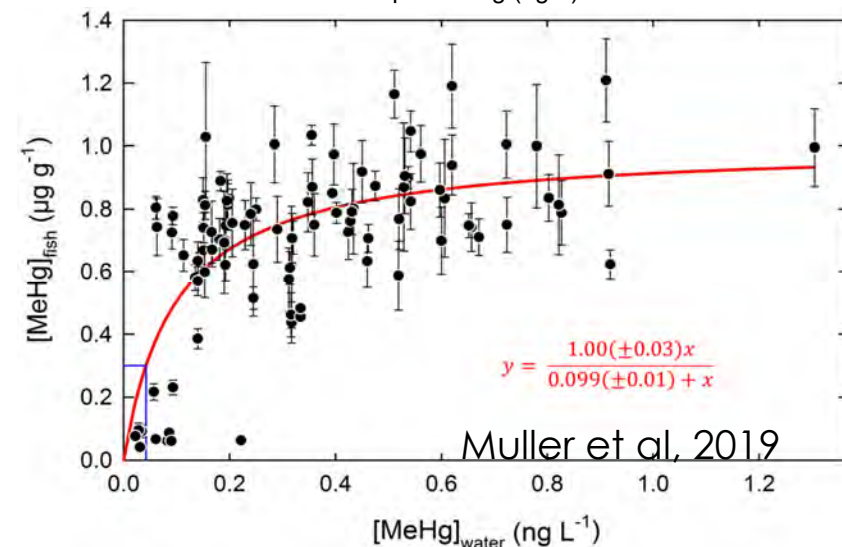
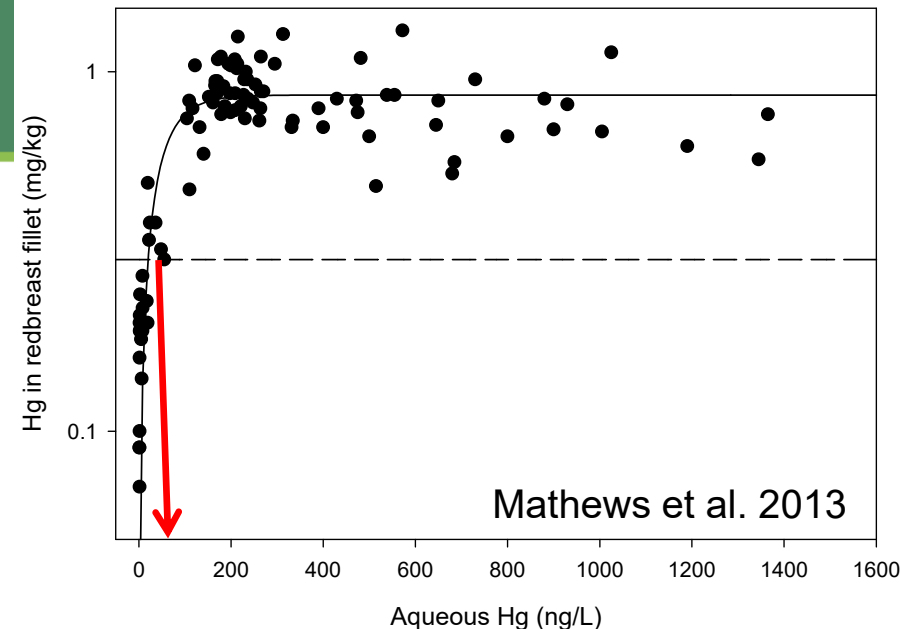


What is an appropriate remediation target?

- Non linear response; threshold concentration?
- MeHg is controlling fish concentrations but Hg methylation is difficult to predict and control



Parks et al., (2013) Science.
“The genetic basis for bacterial mercury methylation”

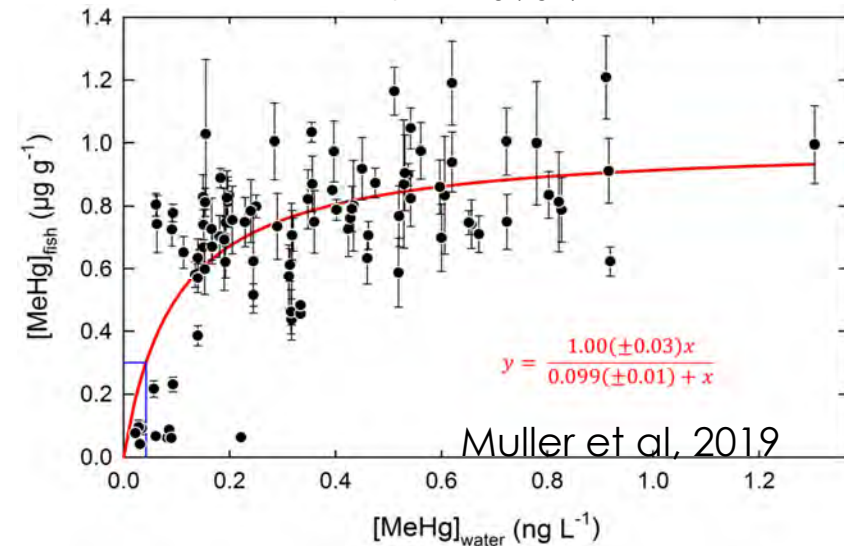
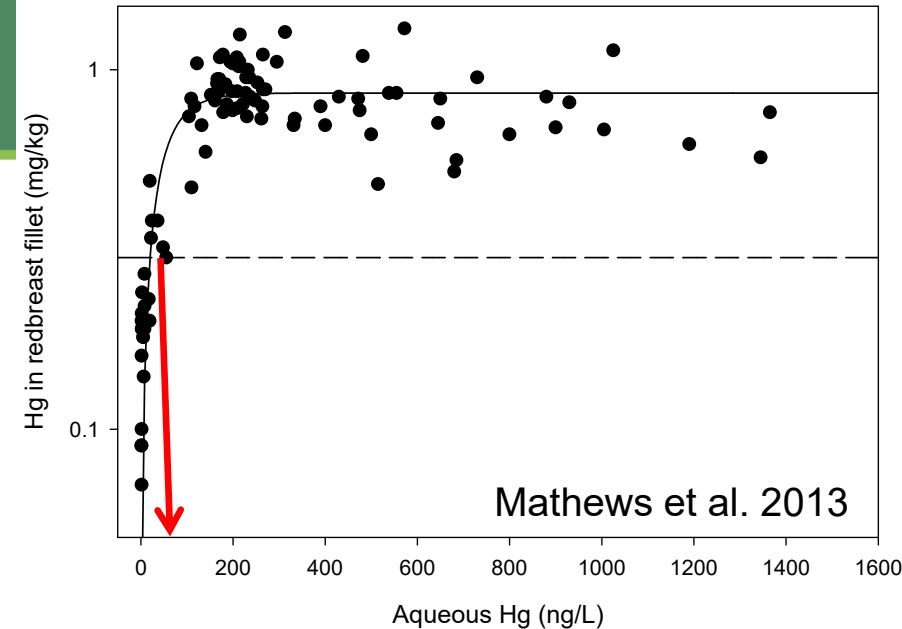


What is an appropriate remediation target?

- Non linear response; threshold concentration?
- MeHg is controlling fish concentrations but Hg methylation is difficult to predict and control

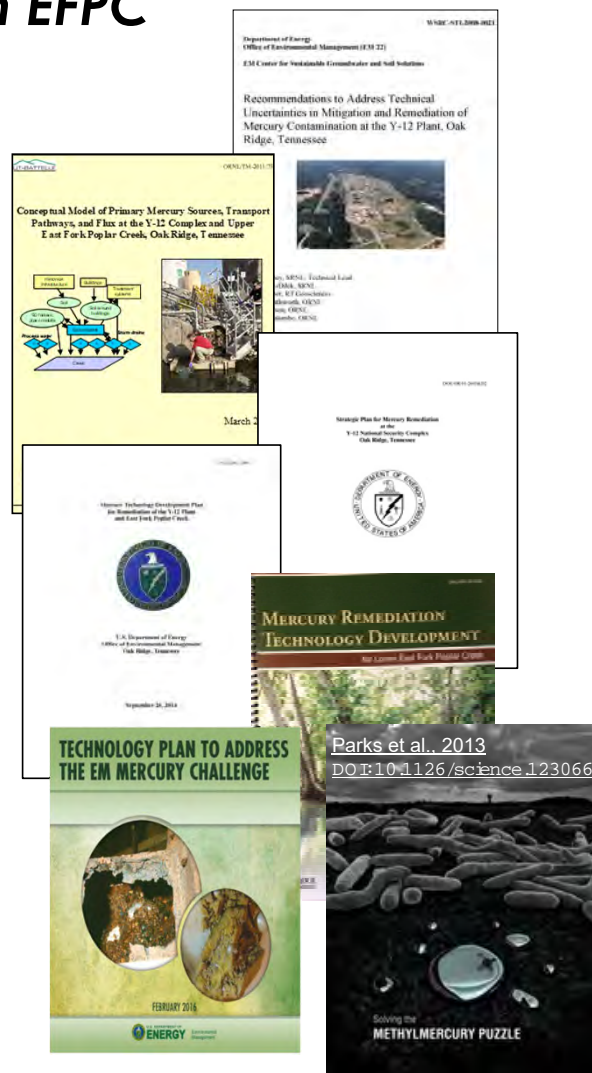
Path forward:

1. Understand relationship between Hg and MeHg in stream
2. Understand factors controlling MeHg production in stream systems
3. Understand the ecological interactions that lead to high Hg concentrations in fish



The Need for Technology Development in EFPC

- Regulatory pressure to address Hg stream exceedances (water concentration, flux, fish)
- Increased understanding of the importance of downstream sources and environmental factors controlling Hg
- Complex system, uncertain remedial options
- A watershed-scale “systems” approach
- Developed based on extensive literature review of the state of mercury remediation science and technology development as well as on-site data
- A desire to avoid large-scale removal of downstream soils and sediments; enhance natural resources
- Leverages ORNL’s basic science capabilities



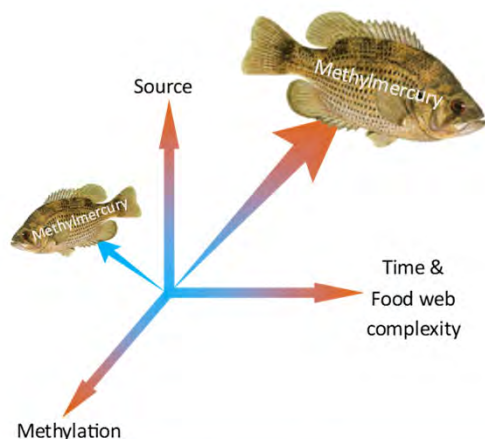
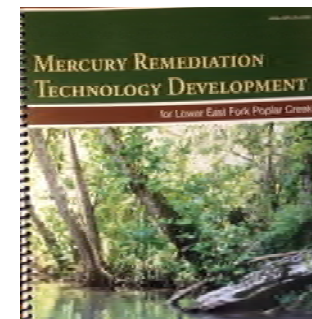
Technology Development (TD) Strategy

- Detailed field characterization early in program transitioning to more targeted field study to address key research questions
- Increased use of quantitative modeling coupled with field and lab data to inform future remedial prioritization and decision-making
- Develop new mercury remediation technologies and approaches to decrease Hg flux, water concentration, and fish concentrations in EFPC
- Primarily bench scale TD studies early in program
- Greater emphasis on increasing scale of testing

FY2023
report
Available



The EFPC TD strategy focuses on developing technologies to decrease mercury in fish



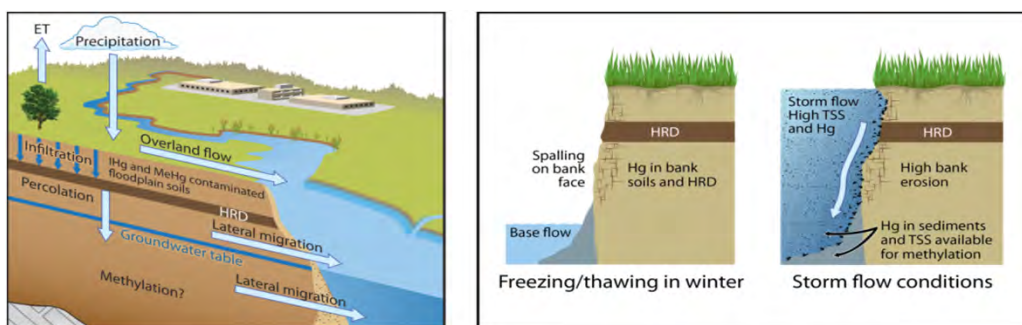
Three key factors determine the level of mercury contamination in fish—the amount of inorganic mercury available to an ecosystem, the conversion of inorganic mercury to methylmercury, and the bioaccumulation of methylmercury through the food web.

-USGS Circular 1395 (2014)

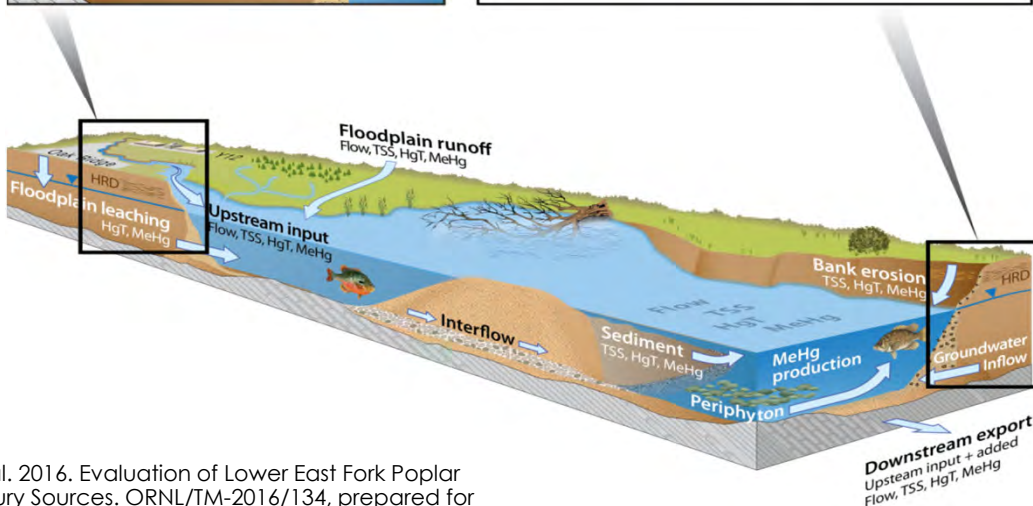
	Tasks	Goals
1	Soil and ground-water source control	Decrease mercury source inputs, flux
2	Water chemistry and sediment	Decrease mercury concentration and methylation
3	Ecological manipulation	Decrease mercury bioaccumulation
4	Aquatic ecology lab upgrade	Improve technology readiness by increasing scale of testing
5	Watershed modeling	Decision support to forecast outcomes of management and remediation decisions

Task 1: Reducing Hg flux

While much of upper EFPC flux occurs under baseflow conditions, the majority of Hg flux from Lower EFPC occurs during storm flow conditions

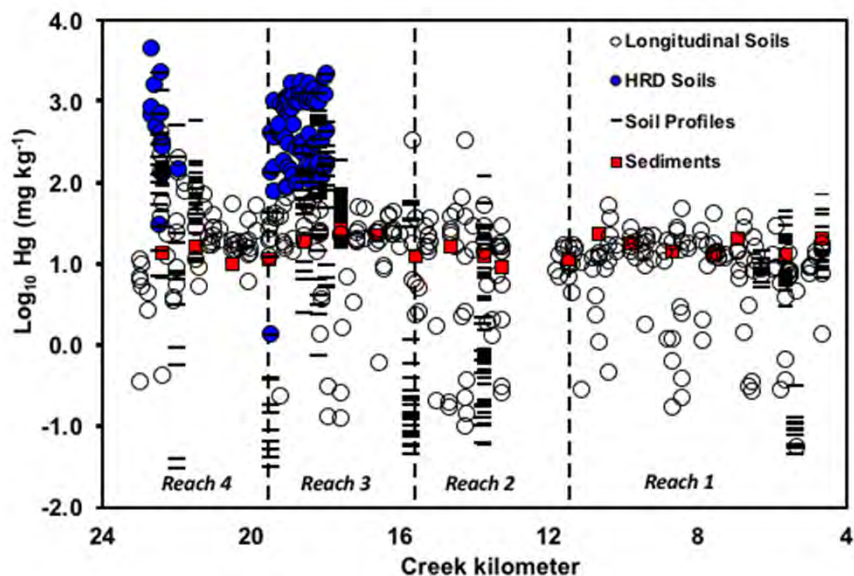


ET Evapotranspiration
 TSS total suspended solid
 HgT total mercury
 MeHg methylmercury
 HRD Historical release deposits



Watson, et al. 2016. Evaluation of Lower East Fork Poplar Creek Mercury Sources. ORNL/TM-2016/134, prepared for the US Department of Energy by Oak Ridge National Laboratory, Oak Ridge, Tenn.

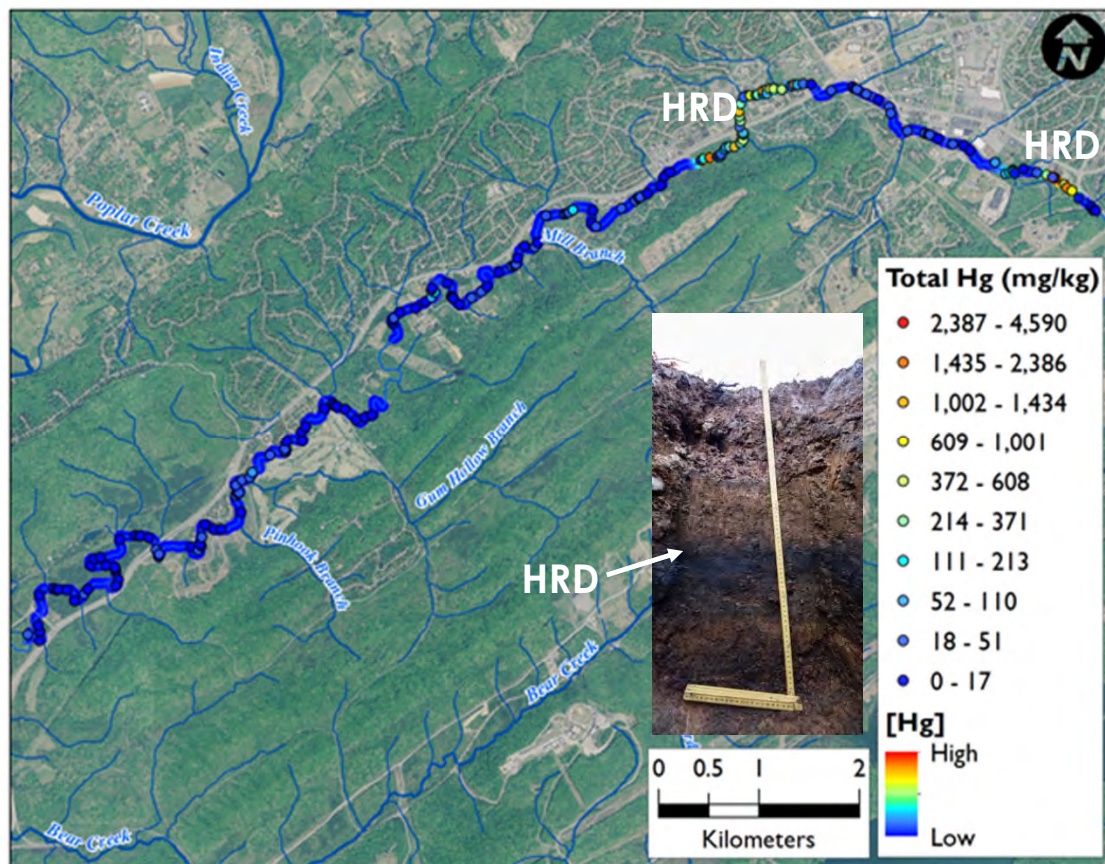
Soil surveys prioritized mercury hotspots



- HRD (historical release deposits) are highest (note log scale)
- Soil Hg >> Sediment Hg

HgT (mg/kg): n = 778, mean = 126, median = 18, minimum = 0.03, maximum = 5,000

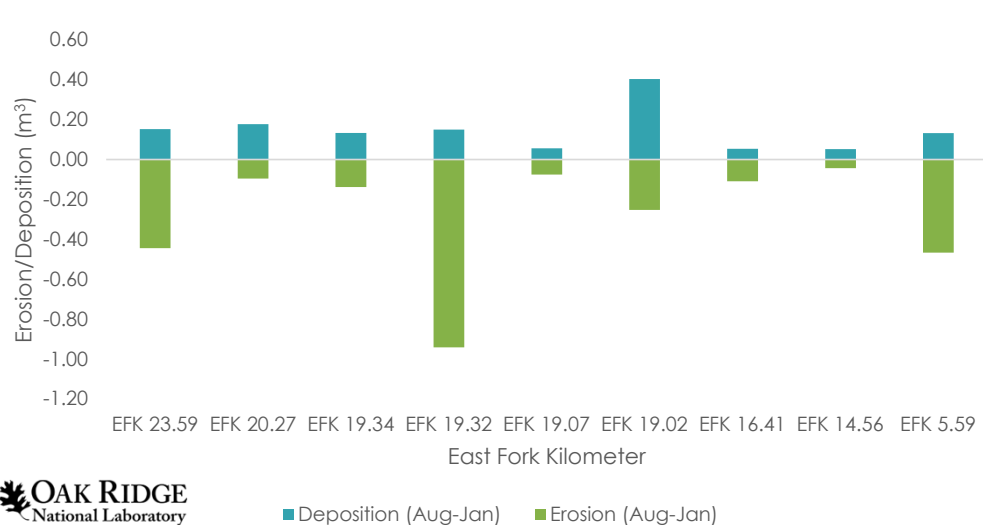
MeHg (ng/g): n = 252, mean = 5, median = 3, minimum = 0.01, maximum = 60



Dickson, et al. (2019) Source relationships between streambank soils and streambed sediments in a mercury-contaminated stream. *Journal of Soils & Sediments*
<https://doi.org/10.1007/s11368-018-2183-0>

Erosion measurements using Lidar, erosion pins

- Erosion is a key factor contributing to Hg release into the stream, and MeHg production in the stream
- Continue erosion pin measurements (since 2013) as they present a long-term record (despite being low-tech, and not amenable to calculating a mass eroded)
- New high-resolution, high-precision lidar measurements at 9 sites (since 2020), add additional sites (2023)

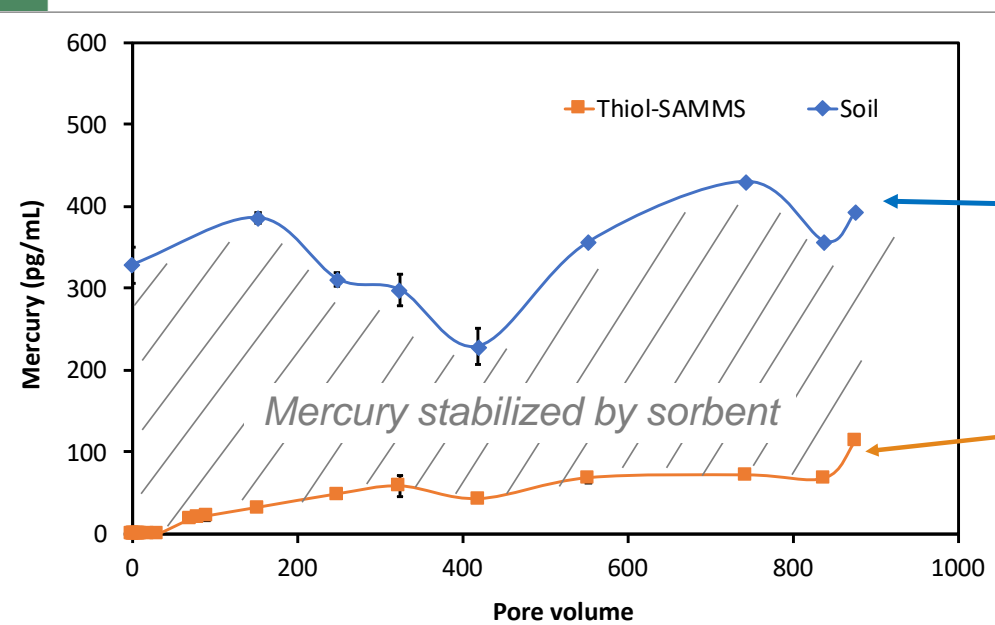


Sites	Hg total (mg/kg)	Mass of eroded soil (kg)	Mass of eroded Hg (kg)
EFK 23.59	1070	464	0.49599
EFK 20.27	35.9	0.00	0.00000
EFK 19.34	763	8.48	0.00648
EFK 19.32	739	1140	0.84279
EFK 19.07	429	25.7	0.01095
EFK 19.02	582	0.00	0.00000
EFK 16.41	9.05	88.9	0.00081
EFK 14.56	10.9	0.00	0.00000
EFK 5.597	8.10	504	0.00408



Column study to evaluate sorbent uptake of Hg

Thiol-SAMMS captures ~ 86 % of Hg

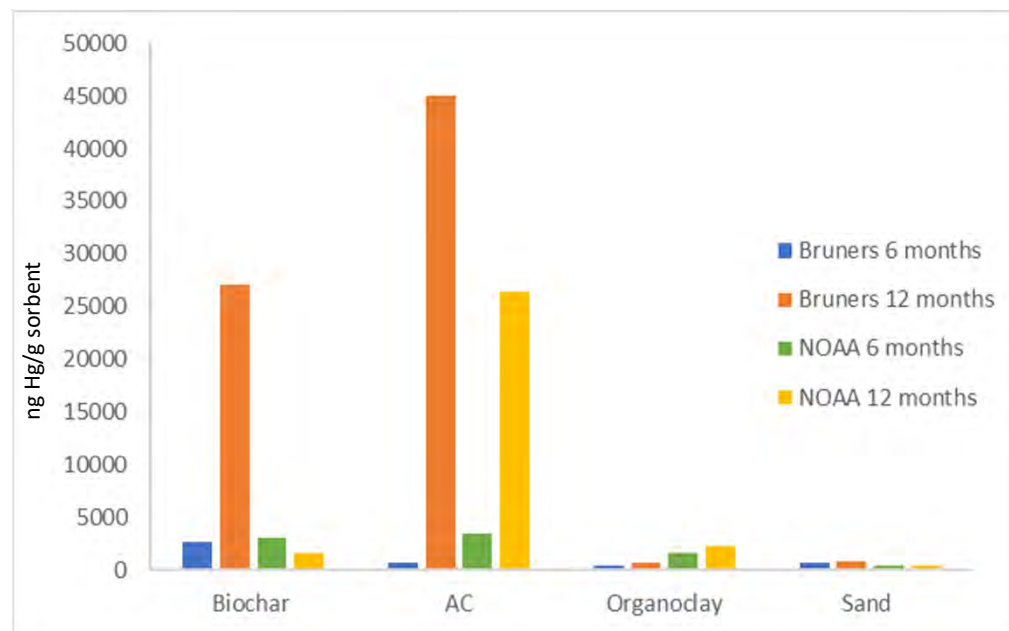


Material	Hg sorbed (%)
Sand (control)	15.4
Sand (control)	7.5
ThiolSAMMS®	84.7
ThiolSAMMS®	88.8
SediMite™	61.6
SediMite™	62.0
Organoclay™ PM199	70.7
Organoclay™ PM199	81.3
Biochar	84.9
Biochar	52.1

- Most sorbents were effective, but none have 100 % uptake

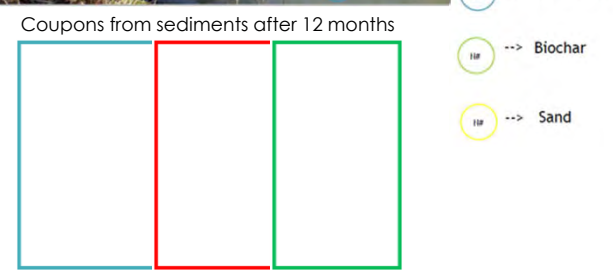
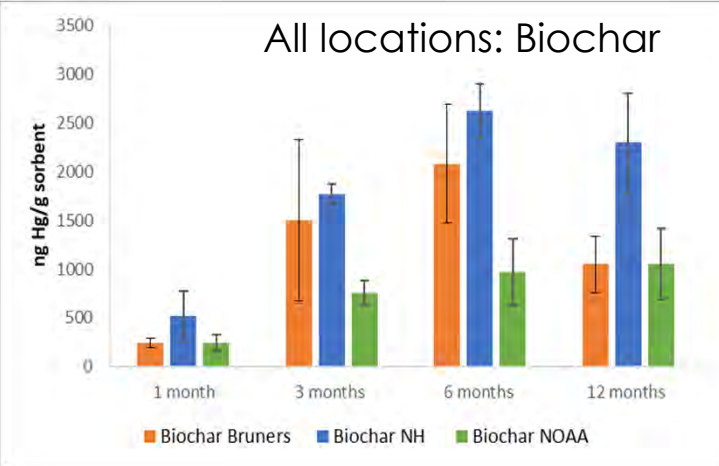
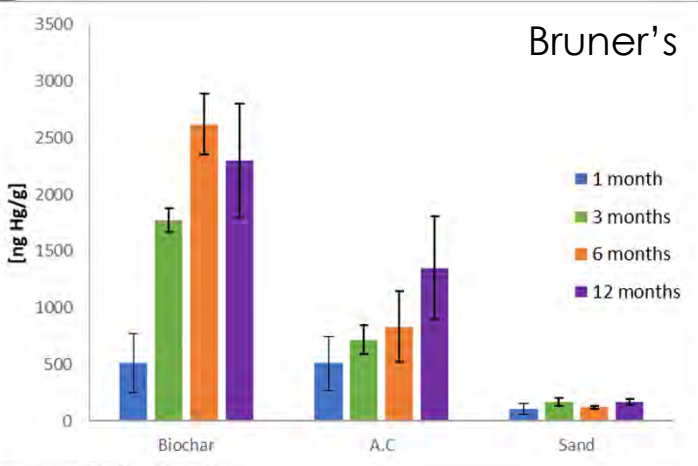
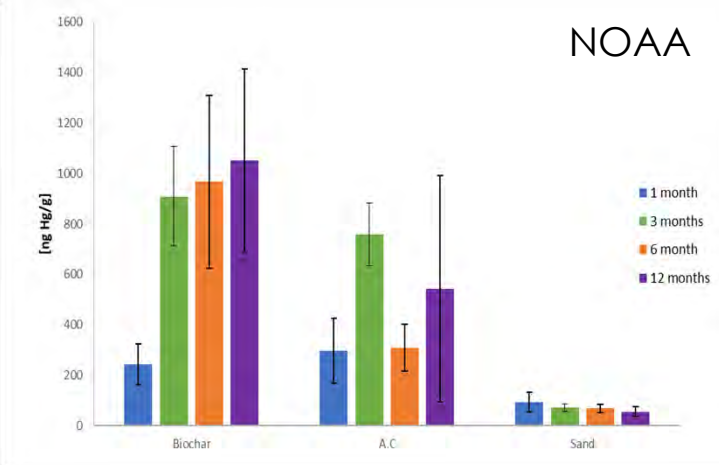
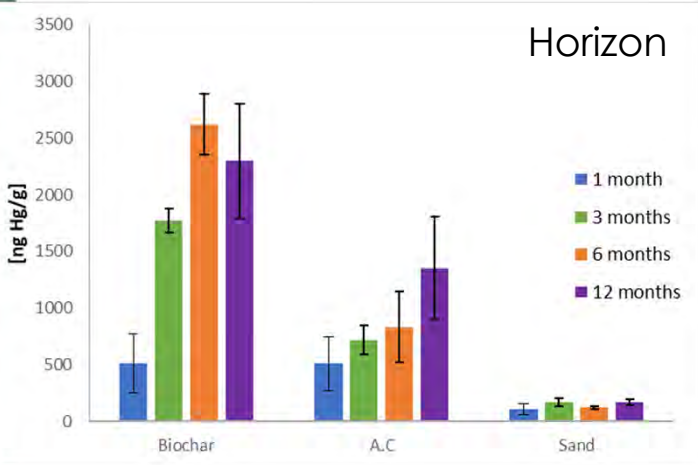
Field sorbent coupon study – Soil

Locations		Sorbents	Time	Height
Soil	NOAA	Sand	6 month	Above HRD
		Biochar		HRD
	Bruner	Organoclay	12 months	HRD
		Activated Carbon (AC)		Below HRD
Creek	NOAA	Sand	1 month	-
	Bruner	Biochar	3 months	-
	New Horizon	Organoclay	6 months	-
		Activated Carbon (AC)	12 months	-
Total amount of coupons: 384				



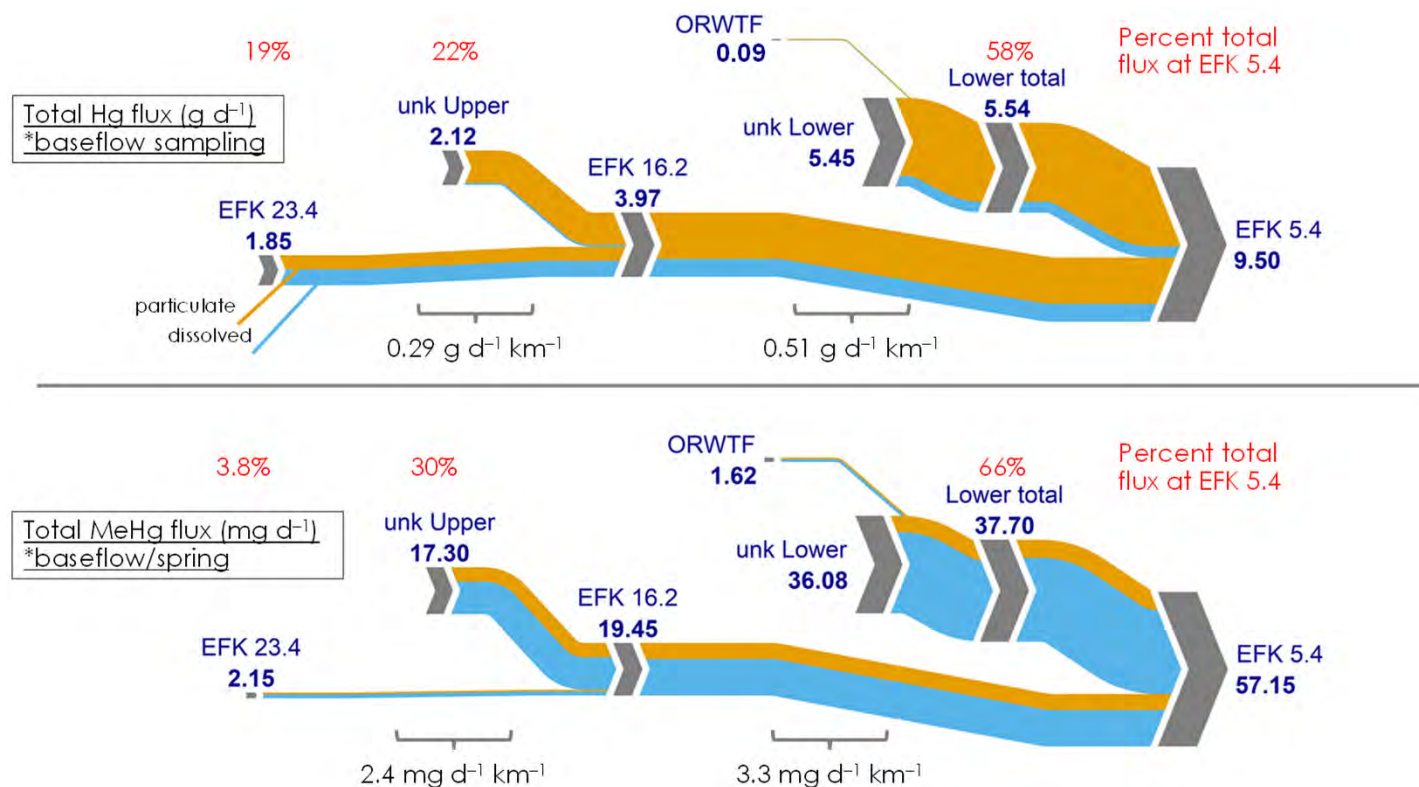
- Biochar and activated carbon are most effective
- Usually more sorption after 12 months

Sorbent coupons – Creek sediments



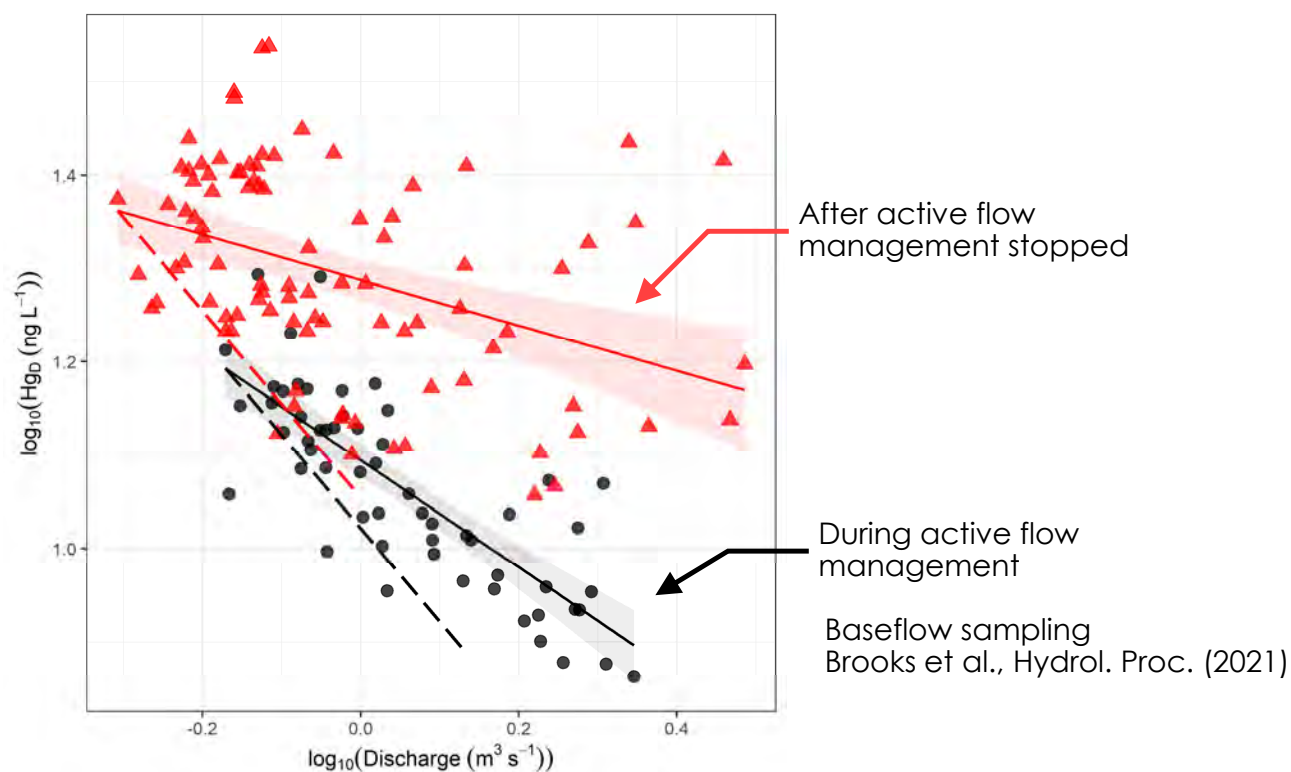
- Sorbed Hg increased over the entire duration
- Biochar may reach equilibrium after 12 months
- Biochar sorbed more Hg than AC and Organoclay PM199
- Sorbent performance depends on location

Task 2: Reducing Hg concentration and methylation

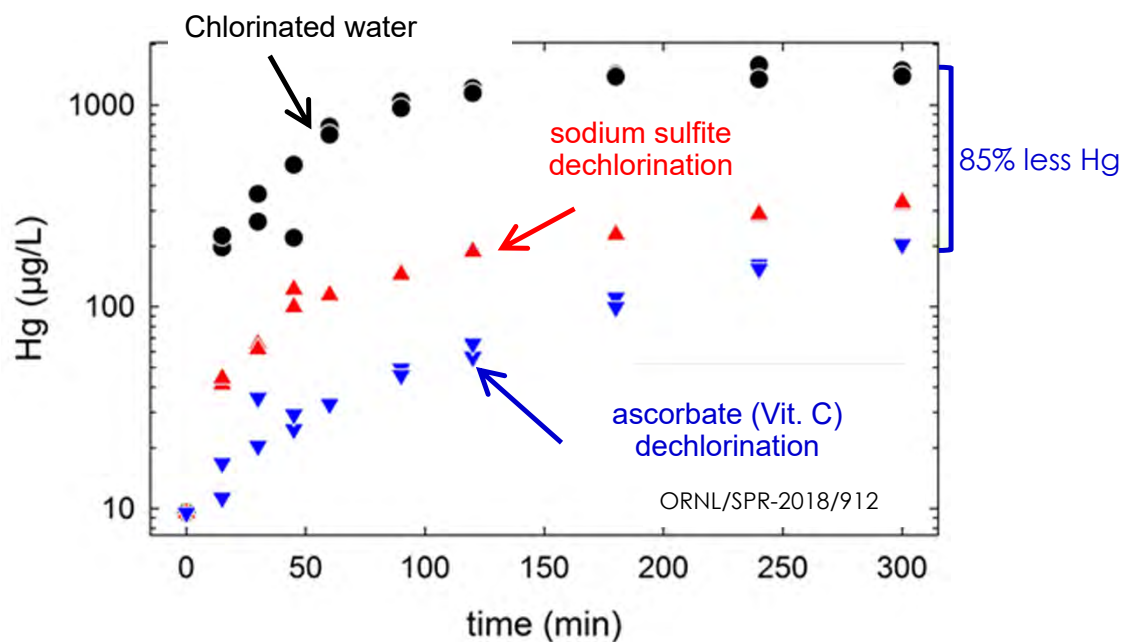
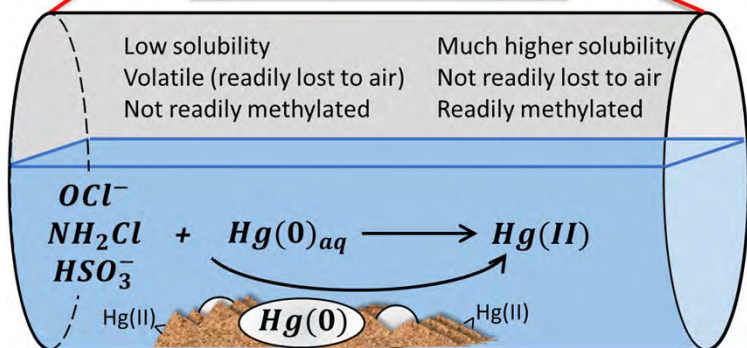


Diffuse legacy sources outside of Y-12 contribute ~80% of Hg flux at EFK 5.4 (baseflow)

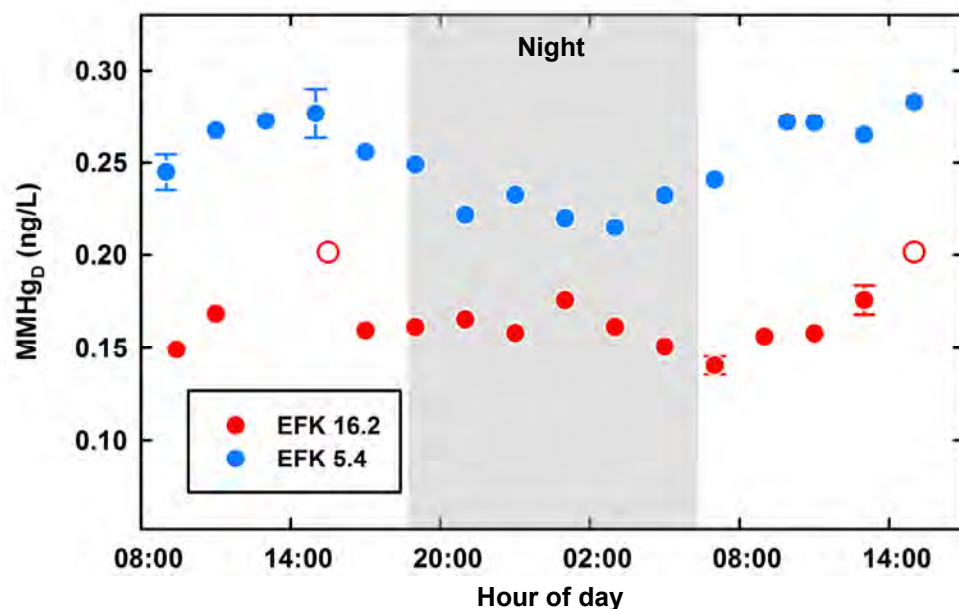
Under a similar range of flows, dissolved Hg concentrations have been higher at EFK 5.4 since active flow management stopped



Alternate dechlorination chemicals markedly decrease Hg(0) dissolution



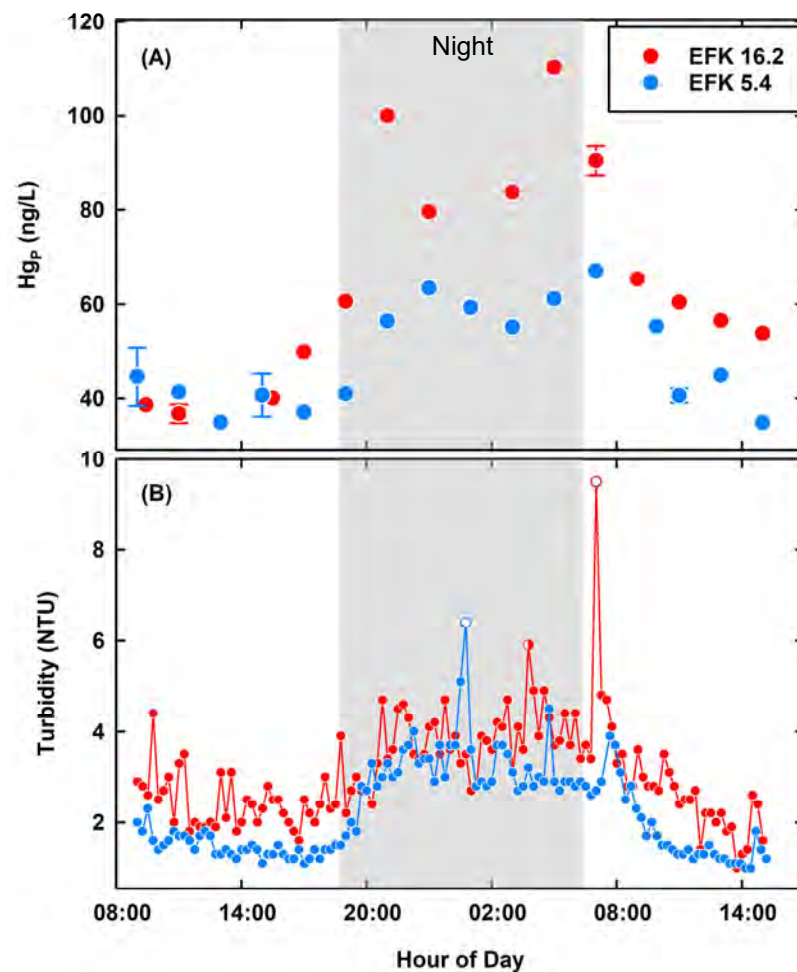
Dissolved MeHg concentration increases during the day



- Most other water bodies in the world have lower MeHg concentration during the day due to photodemethylation
- In EFPC canopy cover minimizes photodemethylation
- Hg-methylating photosynthetic biofilms generate MeHg during day
- This diel pattern weakens and disappears through autumn into winter

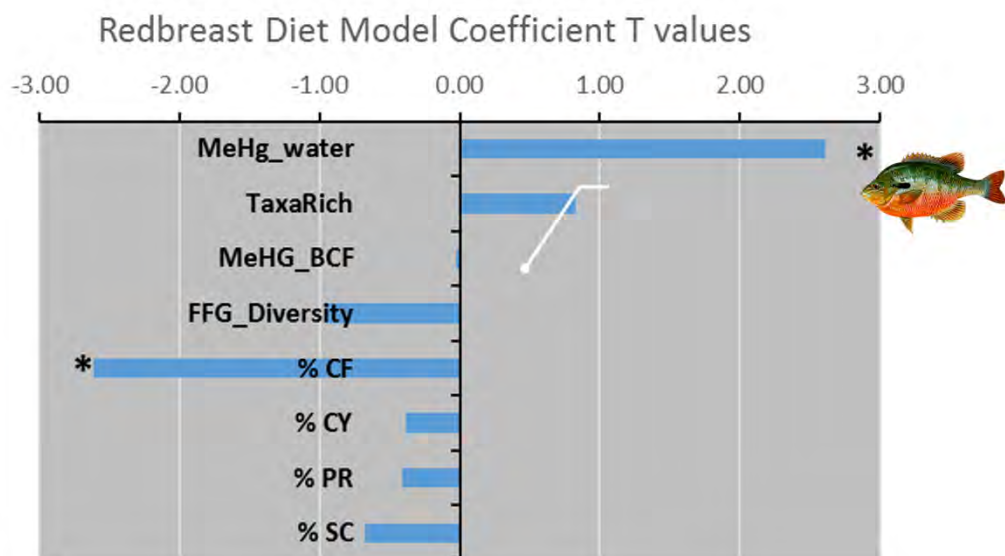
Particulate Hg and MeHg increase at night

- Particulate Hg and MeHg concentration increases overnight coincident with increased turbidity and TSS
- Likely due to diel patterns in biotic activity that resuspend creek sediments
- Pattern dampens through autumn and disappears in winter, emerging the following spring

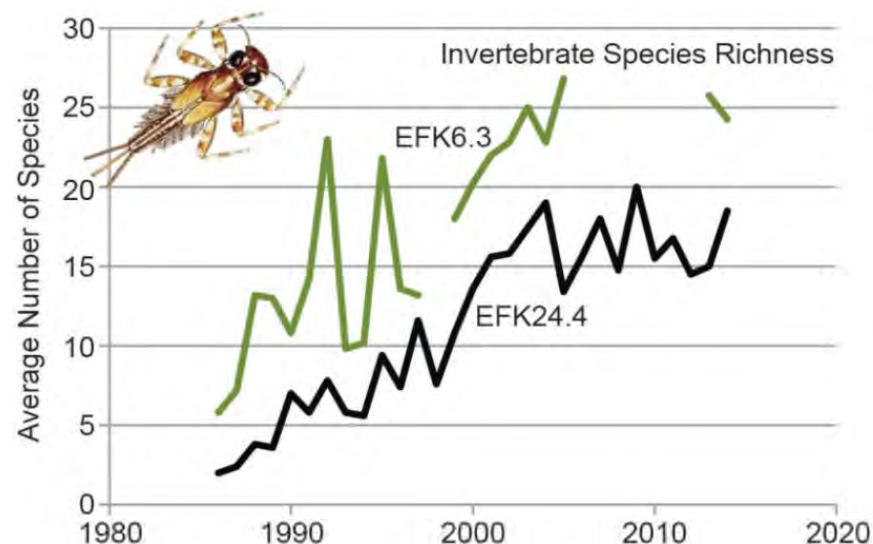


Task 3: Decrease mercury bioaccumulation

Factors controlling MeHg bioaccumulation in fish



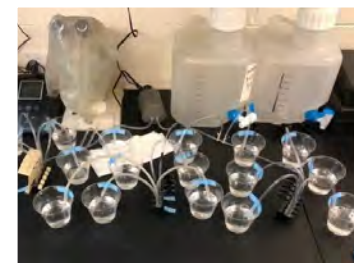
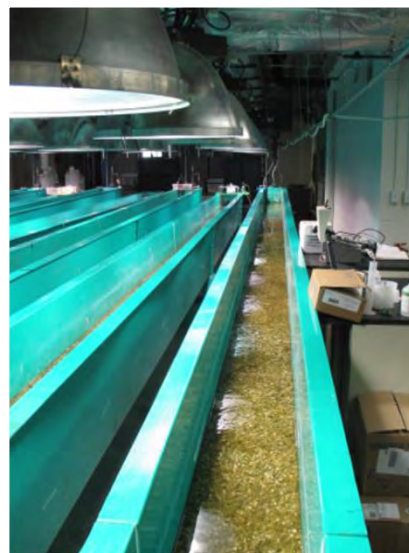
Previous work done in ORNL highlighted that **MeHg in fish tissues (redbreast)** is **negatively correlated** to the **occurrence of filter-feeders bivalves**



- We examined how the community structure has changed over time
- We examined how certain niche groups and aqueous MeHg concentrations correlate to MeHg in fish

Bivalve testing

- Mussels are highly effective in removing particles from water
- Mussels high in total Hg, **low in MeHg**
- Evaluating species filtration rates under different environmental conditions to examine the effects of light, temperature, and particulate load
- Collaborating with Tennessee Wildlife Resources Agency's Cumberland Water Research Center to culture native mussels for testing



Filtration capacity of native mussel species through clearance rates assessment



The Asian clam (*Corbicula fluminea*) used as **reference**, is the only bivalve species currently living in EFPC

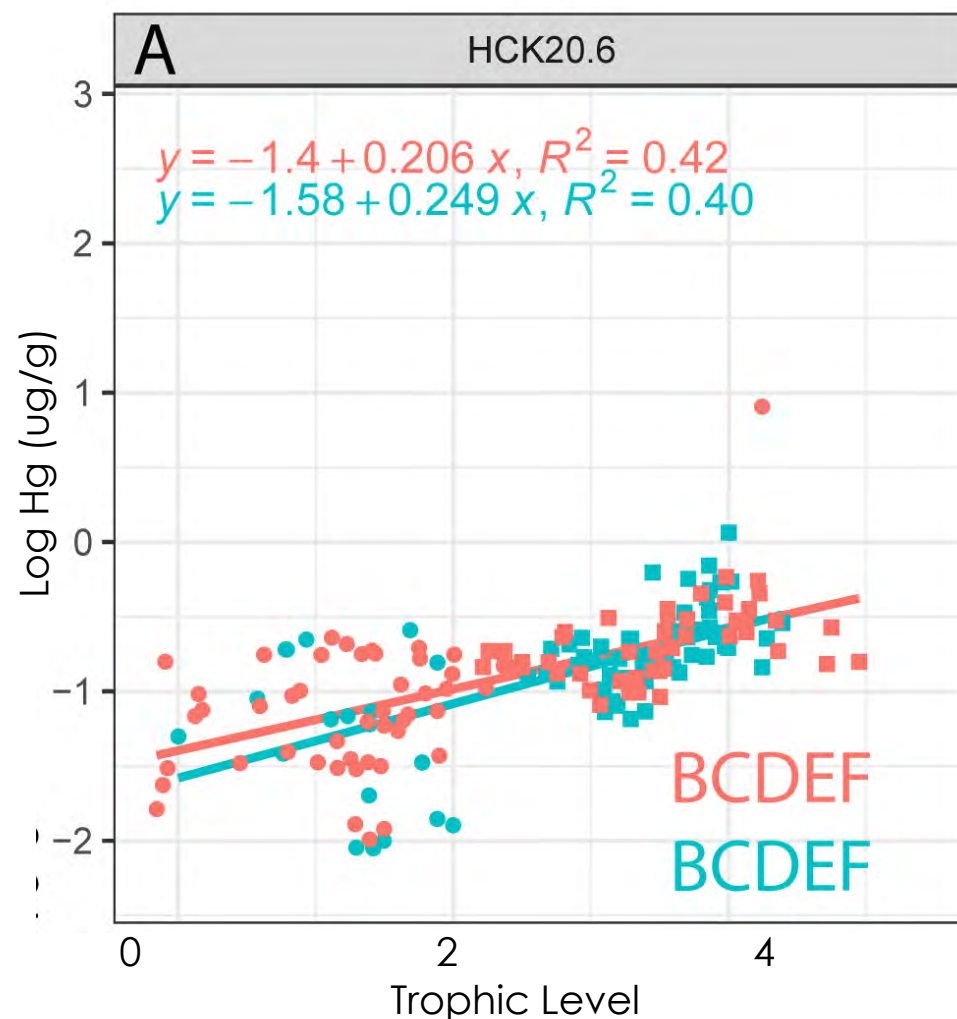


Several native mussel species (Unionidae) studied:

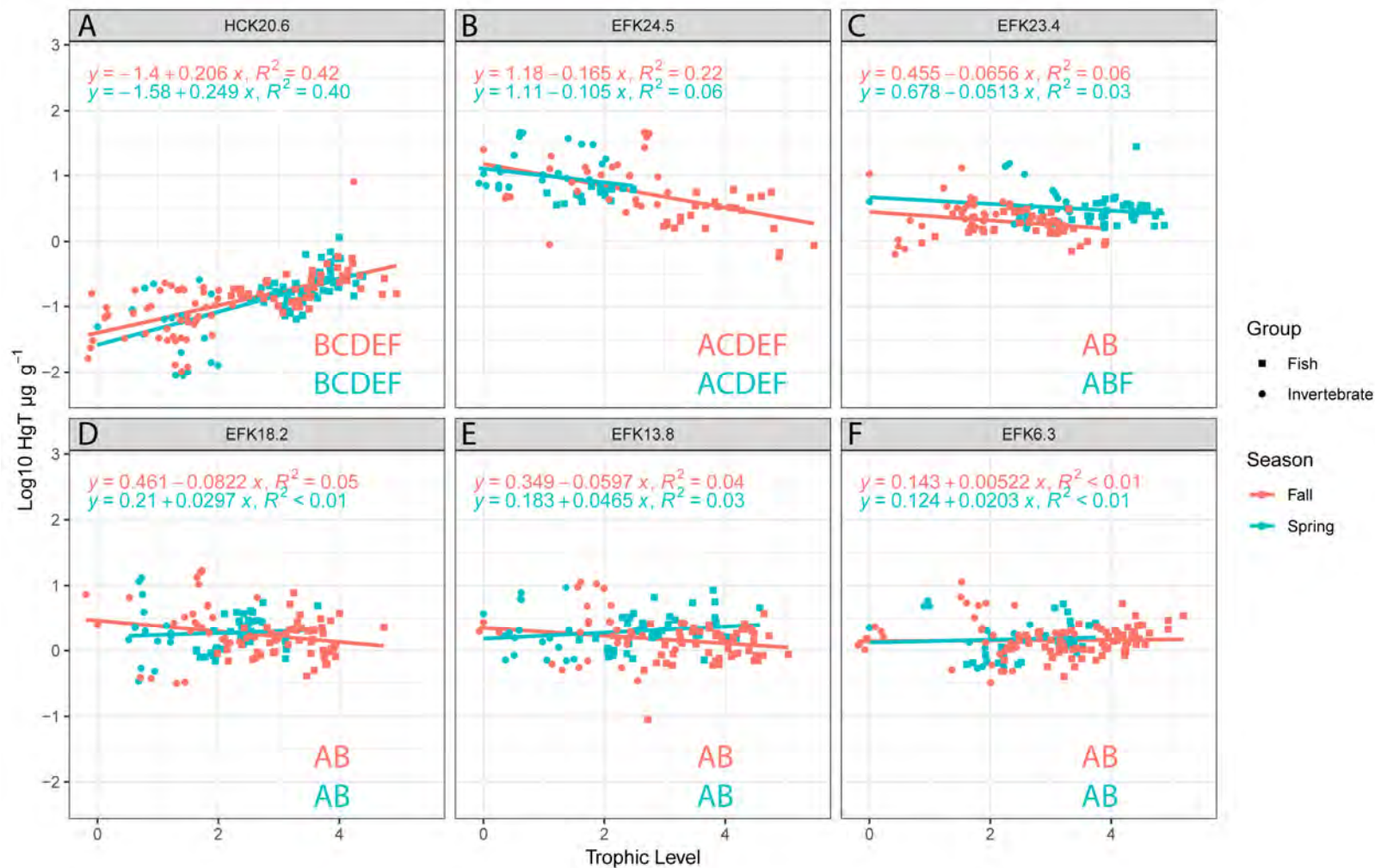
- *Lampsilis ovata* (presented here)
- *Utterbackia imbecillis*
- *Villosa iris*

Food web transfer of mercury

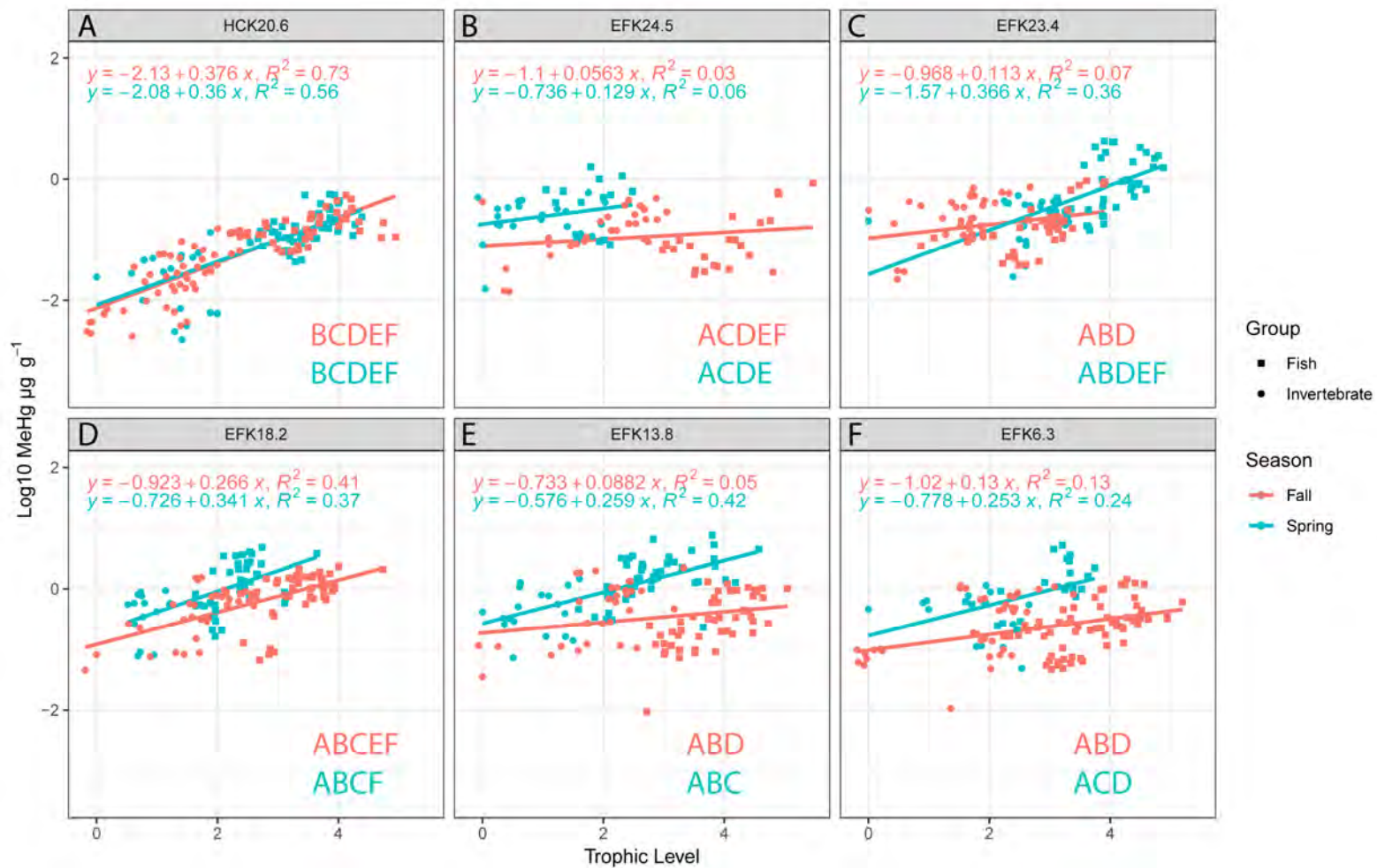
- Historical biological monitoring data for total Hg- water and fish
- Sampled algae invertebrates and fish throughout the food web
- Measured Hg, MeHg, stable isotope of N
- Slope of the line between Hg and trophic level is a measure of bioaccumulation efficiency



Food web transfer of mercury



Food web transfer of mercury



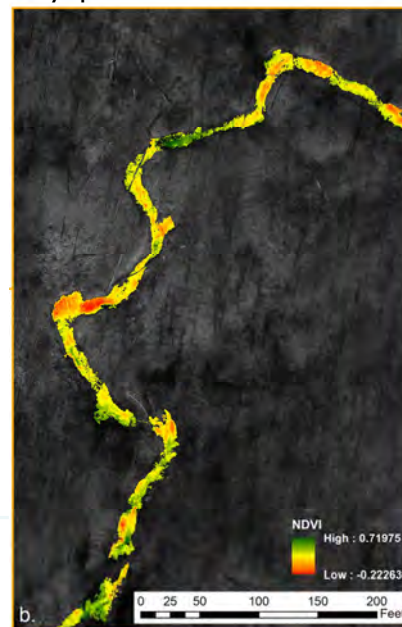
What controls mercury methylation?

Mapping periphyton with drones to understand methylmercury production

- The base of the food web in stream ecosystems
- An entryway for contaminants into food webs
- Habitat for aquatic microbes that can methylate mercury
- A complex community of algae, bacteria, and detritus

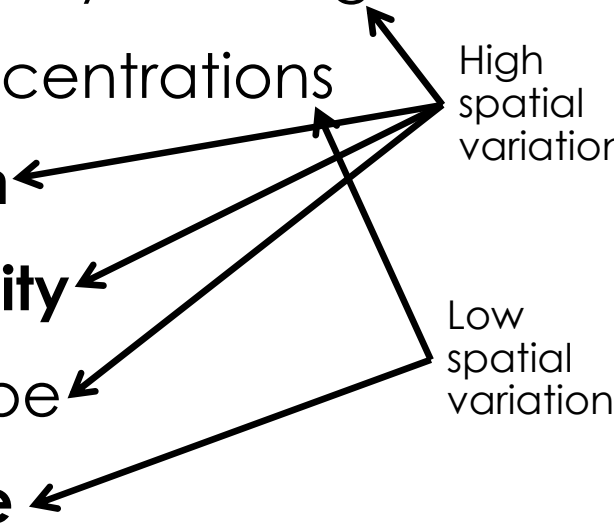
Need for improved understanding of relationships and spatial patterns

- Periphyton distribution and abundance
- Periphyton community composition
- Correlations with MeHg production



Closeup of normalized difference vegetation index for section of stream (higher values indicate healthier vegetation)

What are the factors controlling periphyton distribution and abundance, (that thus affect MeHg production)?

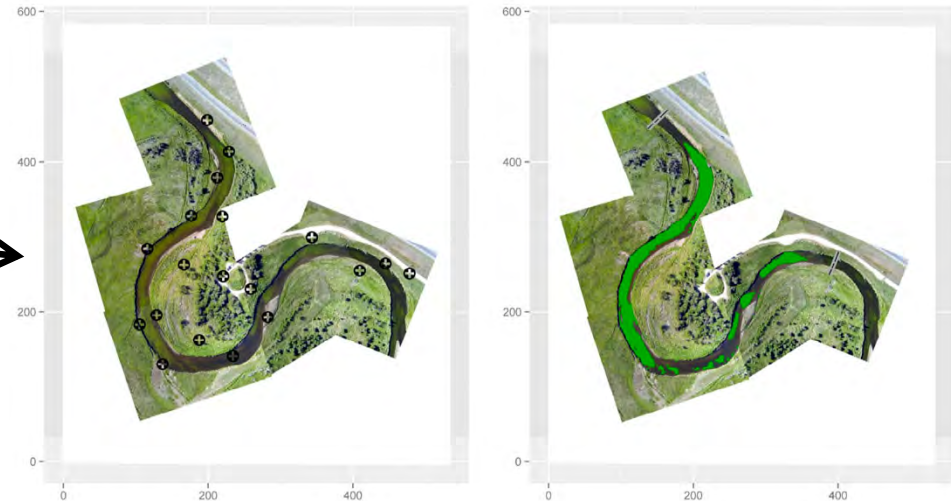
- **Light** availability/shading
 - **Nutrient** concentrations
 - Water **depth**
 - Water **velocity**
 - **Substrate** type
 - **Temperature**
- High spatial variation
- Low spatial variation
- 



Surveying East Fork Poplar Creek

How do we currently measure periphyton?

- In streams and rivers with **open canopy, remote sensing** at scale is viable option.



Clark Fork River. From Flynn and Chapra, 2014.

- In **small forested streams** time-consuming direct measurements required
 - Scraping rocks
 - In-situ fluorescence
 - ^Both approaches have limited spatial coverage



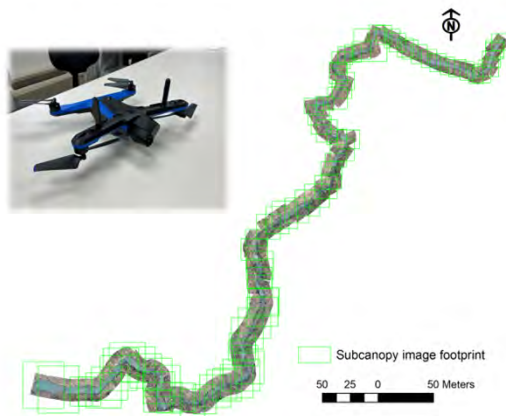
In small forested streams, how can we **scale up** our understanding of periphyton distribution?

- Drones and sensors

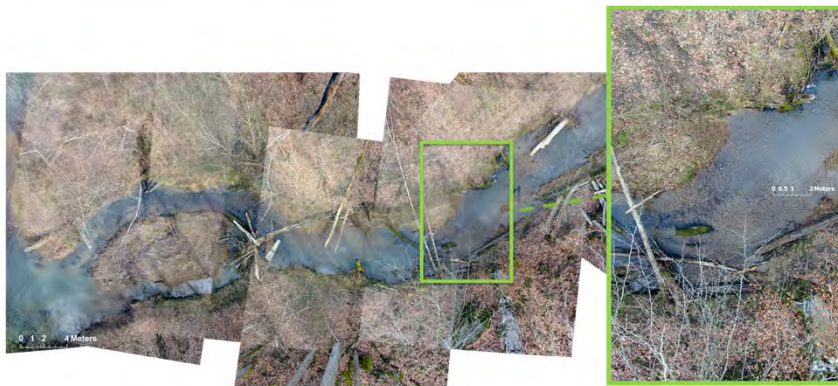
- Topo-Bathy lidar
 - Terrestrial and riverbed elevations, vegetation structure
- Terrestrial lidar
 - Terrestrial and water surface elevations, vegetation structure
- Hyperspectral/multispectral
 - minerals, vegetation, man-made materials
- True color RGB
 - Things discernible to naked eye



Habitat Mapping

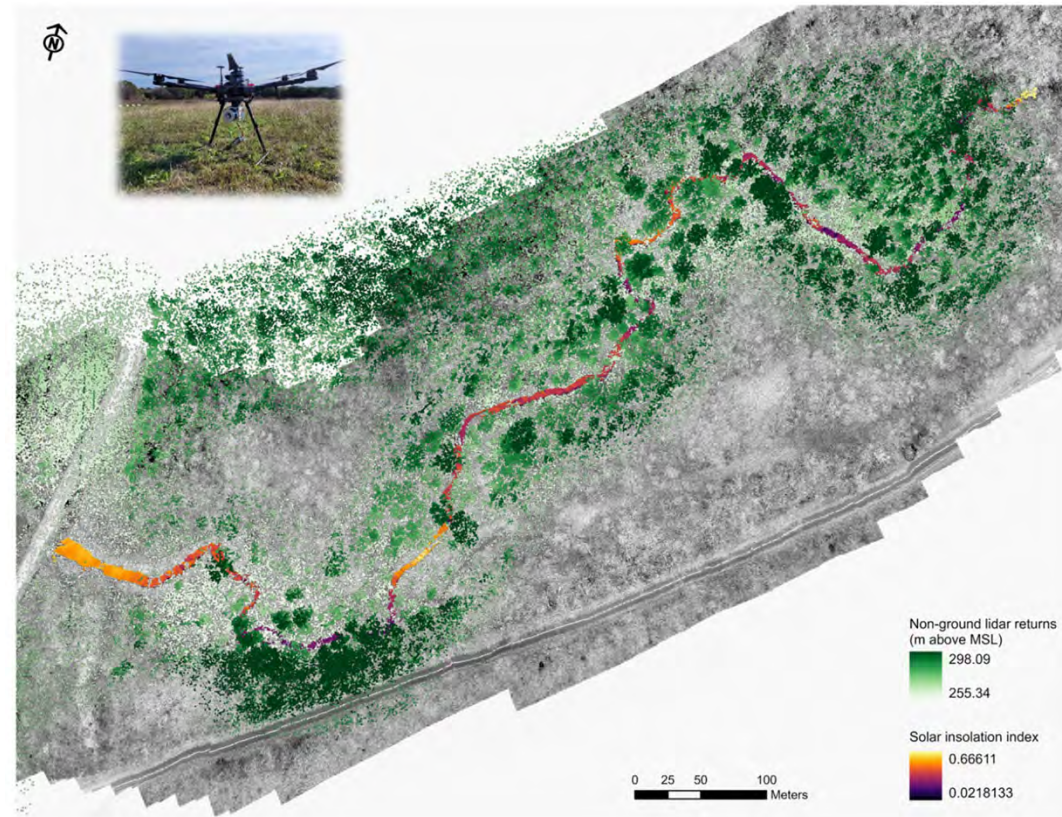


Skydio 2+ sub-canopy flight at Bear Creek on March 1, 2023. Eighty six 0.3cm subcanopy images were georeferenced to a 2cm resolution above canopy image collected on the same day. Skydio 2+ shown in inset.



Closeup of ~75m section of Bear Creek subcanopy imagery collected on March 1, 2023. The spatial resolution of 0.3cm achieved by flying 10 feet above the stream channel allows for mapping instream features with exceptional detail.

Light Penetration Mapping



Non-ground lidar point cloud returns and solar insolation index. Data collected on March 1, 2023 in Bear Creek. Non-ground returns shown in green, with darker green indicating taller trees. Solar insolation index shows estimate of solar exposure throughout stream channel, with higher values indicating higher exposure. Phoenix Lidar onboard the DJI M600 shown in inset.

Conclusions

- Mercury concentrations in fish and water have decreased but remain above guidelines
- Given the complexity of the mercury sources in EFPC, it is likely that a watershed approach and different remediation strategies will be necessary
- Factors other than mercury concentrations (e.g. flow, nutrients, land use change, climate change) may drive mercury dynamics and risk
- Our understanding of flow and flux temporally and spatially has improved significantly, allowing for predictive management tools

Potential future strategies for mitigating Hg in EFPC?

Decrease Hg sources

- MTF will decrease Hg flux and downstream erosion
- Develop bank stabilization and sorbent solutions for high Hg streambanks



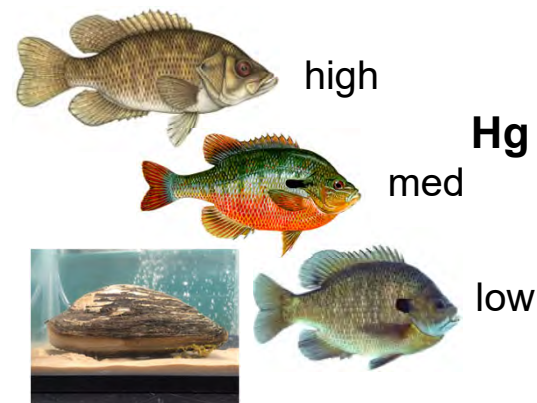
Develop watershed scale recommendations that can impact surface water variables

- What “knobs” need to be turned to decrease Hg methylation?
- Decrease flashy flows, modify nutrients, algae, light, habitat?



Modify the food chain to decrease Hg risks while improving natural quality

- Reintroduce native mussels to decrease particle-associated Hg
- Fish or periphyton management actions



Surface Hydrology

Hydraulics

Bank erosion & Sediment Trans.

Floodplain & GW Hydrology

Periphyton Biomass & Methylation

Fish & Invert Biomass, Biodynamics

Fish & Invert Functional Roles

Acknowledgments

- Mark Peterson
- Louise Stevenson
- Chris DeRolph
- Paul Matson
- Trent Jett
- Michael Jones
- Nikki Jones
- Sujith Surendran Nair
- Natalie Griffiths
- Peijia Ku
- Jordan Jatko
- Mark Lueders
- Micah Bowman
- Ryan McManamay
- Support from Y-12 Compliance
 - Jim Stinnett
 - Kris Kinder
 - Kim Hanzelka
 - Stacey Loveless
- DOE Environmental Management and UCOR
 - Brian Henry
 - Elizabeth Phillips
 - Alex Schenk
 - Bo Buechley
 - John Mojka
 - Brian Shaw

Questions?

