Biogeochemical Transformations at Critical Interfaces Scientific Focus Area

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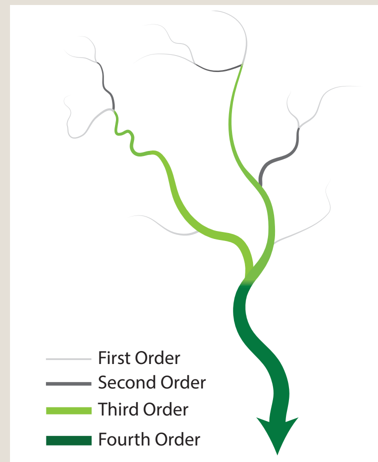
Determining the coupled hydrobiogeochemical processes controlling mercury fate and transformation in low-order freshwater stream systems

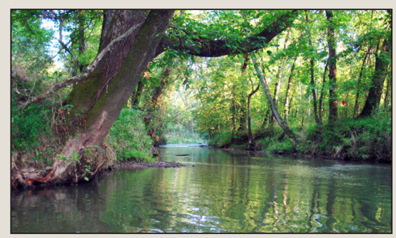
Anthropogenic releases and changing environmental conditions profoundly affect the biogeochemical cycling of trace metals such as mercury (Hg). Mercury can be methylated to form methylmercury, a neurotoxin that bioaccumulates in the food web, endangering humans and other biota. Most mercury contamination in natural environments results from atmospheric processes, but mining and industrial processes can lead to severe local pollution. On the Oak Ridge Reservation in Tennessee, for example, mercury pollution in the East Fork Poplar Creek (EFPC) watershed is caused by historical mercury use at the Y-12 National Security Complex, where large quantities of mercury were lost to the environment during the 1950s and 1960s.

To enable a predictive understanding of mercury cycling in stream systems both locally and globally, the Biogeochemical Transformations at Critical Interfaces Scientific Focus Area (SFA) led by Oak Ridge National Laboratory (ORNL) is providing foundational insight on exchange and feedback processes occurring at critical interfaces that control mercury fate and transformation. This project is supported by the Subsurface Biogeochemical Research program within the Department of Energy's (DOE) Office of Biological and Environmental Research (BER).

Influence of Low-Order Stream Systems

Low-order freshwater streams, such as EFPC, are the most frequently occurring stream type and constitute nearly 90% of total stream length in the United States. Because of their low average water velocity, these stream systems have high water-sediment contact times, which promote in-stream biogeochemical cycling. Furthermore, these streams play a prominent role at the aquatic-terrestrial interface because they represent the first aquatic environment encountered by terrestrially derived materials (solutes and particles). Developing a predictive understanding of mercury and trace metal transport and fate in these environmental systems requires deciphering complex processes (i.e., physical, chemical, and biological), deconvoluting how these processes interact with one another, and understanding the factors that control system response over broad spatiotemporal scales.





Low-Order Stream Systems. *(Top)* *Streams are ranked based on a hierarchical network of channels within a watershed. Low-order streams (i.e., first- through fourth-order streams) are located in the headwater areas and typically convey small volumes of water. Such streams play a dominant role in the flow, biogeochemistry, and water quality of downstream higher-order reaches.* ***(Bottom)*** *East Fork Poplar Creek is a third-order stream being studied as a representative use case in the Biogeochemical Transformations at Critical Interfaces SFA.*

Science Challenge and Goal

Over the past 6 years, this SFA project has made substantial progress in elucidating the mechanisms by which inorganic mercury is transformed into methylmercury at the sediment-water interface and the processes that determine net methylmercury production in contaminated sites.

During the next 9 years, the SFA seeks to address the following science challenge and goal.

- **Science Challenge:** Determine the coupled hydrobiogeochemical processes that control mercury fate and transformation in low-order freshwater stream systems.
- **Science Goal:** Process-rich predictive capability that integrates field, laboratory, and modeling studies of mercury fate and transformation dynamics across broad spatiotemporal scales in low-order streams.

Consequently, SFA project research encompasses four thrust areas including (1) ecosystem features; (2) biogeochemical mechanisms; (3) microbial community functions and geochemical influences; and (4) molecular structure, dynamics, and mechanisms.

Ecosystem Features Influencing Mercury Transformation. Research objectives are to (a) identify ecosystem compartments and hydrobiogeochemical conditions that govern net methylmercury concentration in EFPC and (b) understand the extent to which groundwater-surface water exchange drives mercury transformations in EFPC.

Biogeochemical Mechanisms Controlling Mercury Uptake and Methylation. The overarching research goal is to gain a fundamental understanding of the key geochemical and biochemical mechanisms controlling mercury sorption, uptake, and transformation at the microbe-fluid and particulate (mineral) interfaces.

Microbial Community Functions and Geochemical Influences on Mercury Transformations. Research objectives are to (a) characterize and understand the diversity and abundance of the recently discovered 2-gene cluster (*hgcAB*) that is responsible for Hg(II)-methylation and (b) elucidate the complete biogeochemical pathway for mercury methylation.

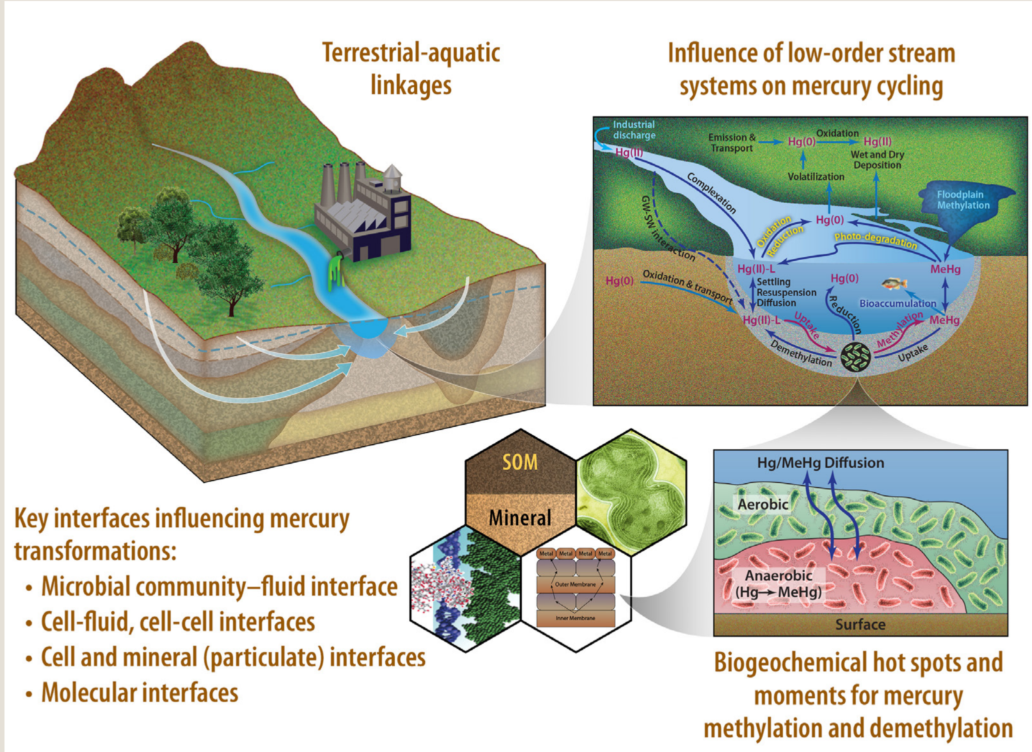
Molecular Structure, Dynamics, and Mechanisms of Mercury Transport and Transformations. The research goal is to investigate structures, reactions, energetics, and dynamics to understand at the molecular scale how mercury is transformed and transported by biological macromolecules and abiotic species encountered in natural and contaminated environments.

Scientific Impact

By developing a deeper understanding of mercury speciation and fluxes in low-order stream systems, this system science project is providing new knowledge and tools required to predict net methylmercury production both locally and across similar stream systems globally. In particular, this foundational information is important for addressing one of the most pressing environmental challenges facing the United Nations Environment Program (a global increase in mercury-contaminated sites) and DOE (remediation of mercury contamination on the Oak Ridge Reservation).

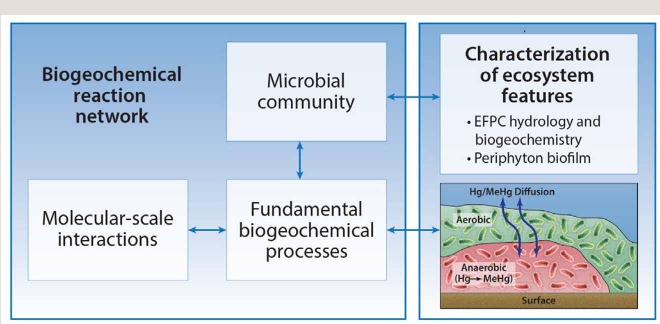
Additionally, while mercury and EFPC serve as representative use cases, the integrated, multiscale approach being pioneered by this SFA will establish a transformational paradigm for, and can be extended to, the understanding of biogeochemical processes that affect fate, toxicity, and fluxes of other trace metals, including radionuclides, in complex, heterogeneous, and multiscale environmental systems.

For more information about the Biogeochemical Transformations at Critical Interfaces SFA, go to **www.esd.ornl.gov/programs/rsfa/**.



Key Biogeochemical Interfaces. *Interfaces are common boundaries between two or more system compartments or phases where steep gradients develop and govern the fate and transformation of material crossing those gradients. Knowledge of exchange and feedback processes at these critical interfaces is required for developing a predictive understanding of mercury transformation in environmental systems.*

Surface Water Sampling. *Researchers collect surface water samples **(right)** for study in the laboratory **(inset)**.*



Mercury Transformation and Migration Model. *Schematic representation of the role each SFA research thrust area plays in the model framework development process to enable the assessment of natural and anthropogenic forcings on environmental mercury cycling.*

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