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Biogeochemical Transformations at Critical Interfaces in a Mercury-Perturbed Watershed Scientific Focus Area

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Freshwater resources supplied by headwater streams and their surrounding watersheds are being threatened by severe pollution from anthropogenic releases of nutrients and trace metals (e.g., mercury [Hg]). Preserving these assets for future use requires developing a deeper understanding of watershed structure and function. Research findings during Phase I of the Critical Interfaces Scientific Focus Area (SFA) project have led to the realization that transient storage zones (TSZs)—and more specifically, metabolically active transient storage zones (MATSZs)—are hot spots for biogeochemical transformations that can exert a controlling influence on downstream water quality. TSZs are surface and subsurface locations (e.g., hyporheic zones) that delay the downstream flow of water in comparison with the main channel. Over the next three years (Phase II), the project aims *to determine the fundamental mechanisms of and environmental controls on Hg biogeochemical transformations in MATSZs in low-order streams*. A key component of this renewal is to parameterize our biogeochemical modeling framework for predicting Hg transformations in East Fork Poplar Creek (EFPC).

Over the past seven months, the SFA team has (1) initiated a planning effort to conduct a series of reach-scale conservative and reactive tracer experiments in EFPC to estimate the volume of TSZs in support of the field-scale model development activity, (2) gained new insights into the effect of light exposure and seasonal changes on Hg transformations in periphyton biofilms, (3) refined our previously developed kinetic model that accounts for changes in Hg bioavailability, and (4) identified a novel Hg stable isotope fractionation mechanism. Additionally, the SFA team is developing new techniques for isolating novel Hg methylators from EFPC microbial communities and gaining new insights into the function of HgcAB proteins using a range of techniques. These include monitoring the growth and metabolite profiles from mutant strains of *Desulfovibrio desulfuricans* ND132 (our model organism), demonstrating that ND132 cell lysates can irreversibly convert mercuric Hg to methylmercury, and predicting the structure of the HgcAB complex using metagenomic sequence analyses. Collectively, these efforts are allowing us to gain a deeper understanding of Hg transformations in EFPC. Although Hg and the EFPC watershed serve as representative use cases, the information generated, and the integrated multiscale approach pioneered by ORNL, will yield a transformational paradigm for integrating an understanding of biogeochemical processes operating in MATSZs into a framework that enables more accurate predictions of water quality at the scale of individual stream reaches and small watershed catchments.