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Biogeochemical Mechanisms Affecting Mercury Species Transformation and Methylmercury Production or Degradation

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The overall goal of this project is to gain a fundamental understanding of the key biogeochemical mechanisms controlling mercury (Hg) species transformation and methylmercury (MeHg) production or degradation in the aquatic environment. One of our most notable accomplishments last year was the discovery of a previously unrecognized MeHg demethylation pathway present in methanotrophs that differs from the organomercurial lyase pathway present in Hg-resistant bacteria. We found that some methanotrophs, such as *Methylosinus trichosporium* OB3b, take up and degrade MeHg rapidly, whereas others, such as *Methylococcus capsulatus* Bath, can take up but not degrade MeHg. Demethylation by *M. trichosporium* OB3b increased with increasing MeHg concentrations, but was abolished in mutants deficient in the synthesis of methanobactin, a metal-binding compound used by some methanotrophs such as *M. trichosporium* OB3b. Our results demonstrate hitherto an unknown, yet potentially widespread biological mechanism of MeHg uptake and demethylation due to the ubiquitous presence of methanotrophs in the environment, including the contaminated East Fork Poplar Creek (EFPC) in Oak Ridge, Tennessee. These findings may open new opportunities to explore how nature detoxifies MeHg in the environment.

Further, in addition to our studies of the effects of naturally dissolved organic matter (DOM) and minerals on Hg transformations (see abstract by Zhao et al.), we demonstrated a new pathway of abiotic photochemical formation of mercury sulfide (HgS or metacinnabar) from photolysis of the complexes of Hg and DOM, resulting in rapidly decreased Hg reactivity and availability for microbial uptake and methylation. Photo-irradiation of Hg-DOM complexes resulted in rapid HgS precipitation, loss of Hg reactivity, and decrease in MeHg production by the methylating bacterium *G. sulfurreducens* PCA. Loss of Hg reactivity proceeded at a faster rate with decreasing Hg/DOM ratio due to an increase in Hg binding with thiol functional groups on DOM. Our results suggest a potentially novel pathway of abiotic photochemical formation of HgS and explain a mechanism whereby freshly deposited Hg is readily methylated but, over time, progressively becomes less available for microbial uptake and methylation.