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Methylmercury Uptake and Degradation by Methanotrophs: A Hitherto Unknown, but Potentially Important Environmental Process

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The overall goal of this project is to fully characterize a hitherto unknown mechanism of methylmercury degradation performed by aerobic methanotrophs. Previous work in our laboratories has shown that aerobic methanotrophs, through the production of a novel metal chelator called methanobactin, can bind inorganic mercury (Hg[II]), and by doing so, substantially reduce Hg[II] toxicity. We have also shown that methanobactin can bind a much more toxic form of mercury, the neurotoxin methylmercury (MeHg). Recently, the PIs have found that methanotrophs expressing methanobactin are also able to demethylate significant amounts of MeHg. Unlike the canonical organomercurial lyase in Hg-resistant bacteria, methanotrophs take up and degrade MeHg at environmentally relevant pH and Hg concentrations (i.e., picomolar to nanomolar), suggesting that methanotrophs likely play a critical role in controlling net MeHg production and toxicity *in situ*. However, although methanobactin is necessary for methanotrophic-mediated MeHg degradation, it is not sufficient as purified methanobactin binds, but does not degrade MeHg. That is, methanobactin appears to serve as “delivery” mechanism to enable demethylation of MeHg by an as yet unknown process. Given that methanotrophs are ubiquitous, there exists a critical gap in our understanding of mercury cycling as methanotrophs likely play an important, yet poorly characterized role in controlling MeHg concentrations and by extension, MeHg bioaccumulation. To address this fundamental gap, we will characterize: (1) the products of methanotrophic-mediated MeHg demethylation using ¹³C-labeled MeHg; (2) the mechanism of methanotrophic-mediated MeHg degradation using a suite of selective inhibitors as well as construction of mutants where genes potentially involved in MeHg degradation are knocked out, and (3) the role of methanotrophs in MeHg degradation at the mercury contaminated East Fork Poplar Creek (EFPC) site in Oak Ridge, TN.

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