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Biomolecular Processes Contributing to Hg Transformations at Critical Interfaces

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Mercury (Hg) methylation is an enzyme-catalyzed process associated with the activity of anaerobic bacteria and archaea carrying the *hgcAB* gene cluster. We determined the kinetics of enzymatic Hg methylation in cell lysates of the model Hg-methylating bacterium *Desulfovibrio desulfuricans* ND132 and demonstrated that the activity of the proteins HgcA and HgcB irreversibly converts mercuric Hg (Hg(II)) to methylmercury (MeHg). The chemical speciation of the Hg(II) substrate is an important factor to consider because of the high affinity of Hg(II) for biological thiols with stability constants of up to 45 for Hg(II)-bis-thiolate complexes, which may limit the availability of Hg(II). We investigated the impact of thiol levels on MeHg formation by HgcA and HgcB and found that methylation activity is not impaired at high thiol/Hg(II) ratios. Furthermore, we explored the roles of cellular metabolites in Hg methylation activity in ND132 cell lysates. We identified a dependence between S-adenosyl methionine levels and Hg methylation rates. The results provide new insights into the function of HgcA and HgcB and their interdependence with cellular metabolism.

Methanotrophs expressing the copper-chelating compound and chalkophore methanobactin can bind and demethylate MeHg, potentially limiting the net production of MeHg in the environment. Initial studies investigated the prevalence of methanotrophs and Hg-resistant bacteria in East Fork Poplar Creek (EFPC) by analyzing metagenomic data for biomarkers of methanotrophs and the prevalence and diversity of Mbn gene clusters, which are essential for the expression of methanobactins. Furthermore, we analyzed methanobactin precursor sequences (*mbnA*) and identified a set of methanobactins potentially present in EFPC. The overall goal is to assess the role of methanotrophic-mediated Hg detoxification among the broader microbial community in EFPC.