Biomolecular processes contributing to Hg transformations at critical interfaces

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Project Abstract: A complex, yet finite set of geochemical and biomolecular processes control mercury (Hg) transformations and net methylmercury (MeHg) production in the environment. A robust predictive understanding of Hg biogeochemistry requires knowledge about the underlying molecular mechanisms. The production of MeHg in anaerobic bacteria and archaea is mediated by the *hgcAB* gene cluster. Proteomics and immunoblot data suggest that the abundance of the proteins HgcA and HgcB in cells of the model Hg-methylating bacterium *Desulfovibrio desulfuricans* ND132 is extremely low. Thus, in order to characterize the function of HgcA and HgcB, we have coexpressed HgcAB in a *E. coli* expression host. Hg methylation assays with cell lysates demonstrate that the heterologously co-expressed HgcAB complex is able to convert added mercuric (Hg[II]) mercury to MeHg. In addition, to facilitate purification of the HgcAB complex for spectroscopic and structural characterization, we expressed a His_6-tagged HgcAB construct in *E. coli*, which also showed methylation activity in cell lysates. Interestingly, the methylation activity of all constructs is substantially enhanced after adding cell lysates from deletion mutants (Δ*hgcAB*) of *Desulfovibrio desulfuricans* ND132 and *Geobacter sulfurreducens* PCA suggesting that there are other cellular components contribute to the mercury methylation activity of HgcAB. Currently, we are evaluating cellular metabolites potentially involved in enhancing Hg methylation activity and have identified a positive correlation between the levels of exogenously added S-adenosyl methionine and Hg methylation rates. The present results establish the foundation for biochemical, spectroscopic and structural characterization of the HgcAB complex, which will provide essential information about the role of HgcA and HgcB in Hg(II) methylation and its integration with cellular metabolism.