Toward Understanding the Biomolecular Mechanism of HgcA

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Our team combines experimental and computational approaches to understand the molecular-scale biophysical processes involved in mercury (Hg) transformations in the environment. Previously, we identified a two-gene cluster required for mercury methylation in anaerobic bacteria. The genes encode a corrinoid protein, HgcA, and a ferredoxin, HgcB, consistent with roles as a methyl carrier and electron donor, respectively. A strictly conserved Cys residue in HgcA was predicted to be a ligand to Co(III) in the corrinoid cofactor and to play a key role in methyl transfer to Hg substrates. More recently, the importance of this residue has been confirmed through in vivo Hg methylation experiments. Our current focus is on characterizing HgcA and elucidating its biochemical mechanism. The effects of Cys coordination on the redox properties of the corrinoid cofactor in HgcA are not well understood. Here, we have used density functional theory (DFT) to compute standard reduction potentials of a selection of corrinoids. Our calculations are in good agreement with experimentally measured values, and reveals new insight into the redox properties of the HgcA. This work will contribute to elucidating the mechanism of Hg methylation involving the HgcAB system in anaerobic microorganisms.