Anaerobic Mercury Methylation and Demethylation by *Geobacter bemidjiensis* Bem, and factors affecting methylmercury export and distribution

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Microbial methylation and demethylation are two competing processes controlling the net production and bioaccumulation of neurotoxic methylmercury (MeHg) in natural aquatic ecosystems. Although mercury (Hg) methylation by anaerobic microorganisms and demethylation by aerobic Hg-resistant bacteria have both been extensively studied, little attention has been given to MeHg degradation by anaerobic bacteria, particularly the iron-reducing bacterium *Geobacter bemidjiensis* Bem. We report, for the first time, that the strain *G. bemidjiensis* Bem can both methylate inorganic Hg and degrade MeHg concurrently under anoxic conditions. Results suggest that *G. bemidjiensis* cells utilize a reductive demethylation pathway to degrade MeHg, with elemental Hg(0) as the major reaction product, possibly due to the presence of homologs encoding both organo-mercurial lyase (MerB) and mercuric reductase (MerA) in this organism. Similarly as observed with the *G. sulfurreducens* PCA strain, *G. bemidjiensis* Bem cells can mediate multiple reactions including Hg/MeHg sorption, Hg reduction and oxidation, resulting in both time and concentration dependent Hg species transformations. Moderate concentrations (10–500 μM) of Hg-binding ligands such as cysteine enhance Hg(II) methylation but inhibit MeHg degradation. These findings indicate a cycle of methylation and demethylation among anaerobic bacteria and suggest that *mer*-mediated demethylation may play a role in the net balance of MeHg production in anoxic water and sediments.

Additionally, we studied the factors affecting MeHg export, sorption and distribution in cells, on cell surfaces, and in solution by known methylators including *G. bemidjiensis* Bem, *G. sulfurreducens* PCA, and *D. desulfuricans* ND132. We found that thiols, such as cysteine, can greatly facilitate desorption and export of MeHg, particularly by the PCA and Bem cells. In thiol-free assays, only a small percentage of the synthesized MeHg was found in solution, while most of the MeHg was associated with PCA or Bem cells, of which about 65-75% was sorbed on the cell surface and ~10-20% remained inside the cells. In comparison, ND132 cells were much more effective in excreting MeHg, with about 80% MeHg found in solution, leaving only a small percentage of the MeHg either sorbed on or remained inside the cells. These results indicate that MeHg export is bacteria-specific and is influenced by thiols, implicating important roles of complexing ligands, such as natural organic matter, in MeHg production and mobilization in the environment.