

Coupled interactions between mercury (Hg), organic ligands, and microorganisms on Hg reduction, oxidation, and methylation

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Mercury (Hg) sorption and redox transformation affect Hg speciation and thus control the rates of Hg uptake and methylation by certain anaerobic bacteria such as *Geobacter bemidjensis*, *Geobacter sulfurreducens* PCA, and its *c*-cytochrome deletion mutant, $\Delta omcBESTZ$. Hg sorption, reduction, oxidation, and methylation are found to occur simultaneously on these bacterial cells under dark, anaerobic conditions. Cells are able to reduce Hg(II) at relatively low cell biomass:Hg ratios, but the reduction becomes inhibited and Hg oxidation commences at increasing cell:Hg ratios due to an increased surface complexation between Hg(II) and cell sulfhydryl (-SH) functional groups. Complexing ligands, such as cysteine and naturally dissolved organic matter (DOM), are found to compete with cells for Hg binding and decrease Hg sorption and methylation initially. Over time, cells appear to overcome the competition and resume uptake of Hg, leading to enhanced methylation even at a cysteine concentration of 1 mM. In contrast, cysteine is found to inhibit methylmercury (MeHg) production by the $\Delta omcBESTZ$ mutant, regardless the cysteine concentration and the reaction time. Furthermore, we studied the factors affecting MeHg export, sorption and distribution in cells, on cell surfaces, and in solution by the *G. bemidjensis* Bem and *G. sulfurreducens* PCA strains. We found that thiols, such as cysteine, can greatly facilitate desorption and export of MeHg. These results highlight complex interactions among Hg/MeHg, complexing ligands, and bacterial cells that are likely important in controlling Hg cycling in anoxic water and sediments.