Microbial cell surface interactions and biogeochemical controls on mercury (Hg) redox transformation and methylation

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Methylmercury (MeHg) is produced primarily by certain anaerobic bacteria, such as G. sulfurreducens PCA and has received extensive interest due to its trophic transfer and bioaccumulation in biota. Although the genetic basis of bacterial methylation of mercury (Hg) is recently discovered, the biogeochemical processes controlling of Hg bioavailability and uptake are not fully understood. We previously showed that PCA cells can reduce, adsorb, and methylate mercuric species, Hg(II), but it remains unclear how the reduction and adsorption processes affect Hg methylation and whether this organism can also oxidize dissolved elemental Hg(0), thus affecting Hg speciation and methylation through redox transformations. Here, we employ kinetic experiments to gain insight into how microbes affect Hg redox transformation and whether cell-Hg association may influence Hg uptake and methylation. We show that PCA cells are capable of not only reducing Hg(II) but also oxidizing dissolved elemental Hg(0) under anaerobic conditions. The kinetics of this transformation is determined by the cell biomass (or cell thiols) to Hg ratio. We further examined the correlations between Hg-cell interactions (e.g., adsorption, reduction and oxidation) and methylation at given cell to Hg ratios. At a low cell to Hg ratio, Hg(II) reduction was dominant; whereas at a high cell to Hg ratio, oxidation of Hg(0) to Hg(II) became a dominant process. Mercury methylation was positively correlated to cell adsorption ($r = 0.96$) but negatively correlated to Hg reduction ($r = -0.87$). The inverse correlation between reduction and methylation was verified using a PCA mutant, ΔomcBESTZ, which is deficient in five outer membrane c-cytochrome genes but possesses two times higher cell-SH than that of the wide type. Similarly, using the methylation deficient mutant, ΔhgcAB, with a lower abundance of cellular thiols than the wild type, we observe increased Hg reduction but decreased adsorption and intracellular uptake of Hg by the ΔhgcAB cells. Our research highlights the need to consider complex Hg-cell interactions and/or biogeochemical controls on Hg speciation, uptake and methylation in natural environments.