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

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Mercury (Hg) is a persistent environmental pollutant that bioaccumulates in the form of neurotoxic methylmercury affecting human health. Its redox cycling in anoxic environments critically links to the bioavailability of Hg for uptake and methylation, but factors that affect this process are poorly understood. We studied the kinetics of Hg reduction, oxidation and species transformation mediated by both abiotic and biological processes under dark, anaerobic conditions. Both reduction and surface complexation of Hg(II) were found to occur simultaneously on *G. sulfurreducens* PCA cells. Reduction of Hg(II) to elemental Hg(0) initially followed a pseudo-first order kinetics with a half-life of < 2 h in a phosphate buffer (pH 7.4). However, cell surface complexation of Hg(II) was found to compete and inhibit Hg(II) reduction by cells, and this inhibitory effect is attributed to strong binding of Hg(II) via the sulfhydryl functional groups on cell surfaces, similarly as observed for reactions between Hg and natural organic matter (NOM) and thiolate compounds. NOM simultaneously reduces and oxidizes Hg due to the presence of both reducing moieties (such as reduced quinones) and thiol-containing functional groups in NOM. Using several strains of bacteria within the genera *Desulfovibrio* and *Geobacter* spp., we also showed that certain bacteria can methylate Hg via anaerobic Hg(0) oxidation. *Desulfovibrio desulfuricans* ND132 was able to both oxidize and methylate Hg(0) in washed cell assays, with an observed methylation rate constant of up to 1.3×10^{-3} h^{-1}. *Desulfovibrio alaskensis* G20 can oxidize Hg(0) but not methylate mercury. *Geobacter sulfurreducens* PCA showed little Hg(0) oxidation and methylation activity under the same experimental conditions, but amendment with cysteine led to substantially enhanced oxidation and subsequent methylation. Our findings highlight several distinct cell surface interactions, reduction, oxidation, and complexation, under which microbial Hg(II) uptake and methylation occur. These observations could therefore have important implications for geochemical cycling of Hg and methylmercury formation in the natural environment. Our observations also demonstrate that contrary to expectation, reduction of Hg(II) to Hg(0) does not necessarily prevent Hg uptake and microbial methylation.