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Complex Biogeochemical Mechanisms Controlling Mercury Species Transformation and Methylmercury Production in the Environment

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The overall goal of this research is to understand complex biogeochemical processes and interactions between mercury (Hg) and microbes, naturally dissolved organic matter (DOM), particulate organic matter, and minerals in influencing Hg species transformation and availability for microbial uptake and methylation. Using *D. desulfuricans* ND132 as a model methylating organism, we first examined the dynamics of concurrent Hg sorption, uptake, and methylation to resolve whether cells take up Hg passively or actively. We found that, in the absence of thiols, >60% of the added Hg was taken up passively in 48 h by both live and heat-killed cells, as well as cells treated with the proton gradient uncoupler, carbonyl cyanide *m*-chlorophenyl hydrazone (CCCP). Heat treatment or CCCP treatment halted Hg methylation, but it did not stop cellular Hg uptake. Similarly, treatment with CCCP impaired the ability of spheroplasts to methylate Hg but did not stop Hg uptake. Our results indicate that cellular Hg uptake is primarily controlled by Hg speciation and ligand exchange and is independent of active metabolic processes. We also investigated Hg sorption-desorption characteristics on three organo-coated hematite particles and a Hg-contaminated sediment and evaluated the potential of particulate-bound Hg for microbial methylation. Mercury was found to rapidly adsorb onto particulates; however, the presence of Hg-binding ligands—such as low-molecular-weight thiols and humic acids—resulted in up to 60% of Hg desorption from the Hg-laden hematite particulates, but <6% from the sediment.

Importantly, the particulate-bound Hg was bioavailable for uptake and methylation by ND132 cells, and the methylation rate was 4–10 times higher than the desorption rate of Hg. These results suggest that direct contacts and interactions between particulate-adsorbed Hg and cells likely caused rapid exchange and uptake of Hg by ND132 cells. This observation questions the common notion that sorbed or particulate-bound Hg is unavailable for microbial uptake and suggests important roles for particulates as significant sources of Hg for methylation in the environment. We further examined Hg isotope fractionation during abiotic dark oxidation of dissolved elemental Hg(0) in the presence of thiol compounds and DOM. We observed equilibrium mass-dependent fractionation with enrichment of heavier isotopes in oxidized Hg and a small negative mass-independent fractionation owing to nuclear volume effects. These findings provided additional experimental constraints on interpreting Hg isotope signatures, with important implications for the use of Hg isotope fractionation as a tracer of Hg biogeochemical cycles in nature.