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ABSTRACT TITLE: Oxidation of Hg(0) to Hg(II) by Anaerobic Bacteria: Its Effect on Hg Stable Isotope Fractionation and Methylmercury Production

ABSTRACT: Background: The methylation of inorganic Hg is known to be mediated by microorganisms under anoxic conditions. Previous studies have elucidated the chemical species of mercuric Hg [Hg(II)] that are bioavailable to methylating microbes. The reduced form of mercury, elemental Hg [Hg(0)], is generally considered chemically inert and unavailable for biologic uptake by anaerobic bacteria. However, the oxidation and subsequent methylation of Hg(0) by anaerobic bacteria have never been tested. In this project, we are conducting laboratory experiments to examine the anaerobic oxidation of Hg(0) and the effect of microbial Hg(0) oxidation on Hg stable isotope fractionation and MeHg production.

Hypotheses:
1: In anoxic environments, anaerobic bacteria catalyze the oxidization of Hg(0) to Hg(II).
2: Microbial oxidation of Hg(0) to Hg(II) imparts a mass-dependent fractionation of Hg stable isotopes.
3: Anaerobic Hg-methylating bacteria produce MeHg when provided with Hg(0) as the sole Hg source.

Results: We examined the Hg(0) oxidation activity of the obligate anaerobic bacteria Desulfovibrio desulfuricans ND132 (Deltaproteobacterium) and Geothrix fermentans H5 (Acidobacterium), and the facultative anaerobic bacteria Shewanella oneidensis MR-1 (Gammaproteobacterium) and Ralstonia metallidurans AE104 (Betaproteobacterium). Anoxic cultures were exposed to Hg(0) in the dark, and samples were collected and analyzed for the formation of non-purgeable Hg. We found that all four bacterial strains produced non-purgeable Hg from dissolved Hg(0) under anoxic conditions. Derivatization of the non-purgeable Hg in the cell suspensions to diethylmercury and analysis of Hg(0)-reacted bacterial cells using X-ray absorption near edge structure (XANES) spectroscopy demonstrated that cell-associated Hg was dominantly in the oxidized Hg(II) form.

The fractionation of Hg stable isotopes by microbial Hg(0) oxidation was investigated using multi-collector inductively coupled mass spectrometry. Oxidation of Hg(0) to Hg(II) by D. desulfuricans ND132 resulted in mass-dependent fractionation of Hg stable isotopes.

The production of methylmercury by D. desulfuricans ND132 was determined by distillation and ethylation-gas chromatography. When exposed to a constant source of Hg(0), D. desulfuricans ND132 produced up to 118 μg/L of methylmercury after 36 h of incubation. A major fraction of the methylated Hg was exported out of the cell and released into the culture medium. These results indicate that anaerobic Hg-methylating bacteria can catalyze the oxidization of Hg(0) to Hg(II), and produce MeHg when provided with Hg(0) as the sole Hg source.