Effect of organic ligands and cell metabolism on Hg(II) sorption and coordination to *E. coli*

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Introduction

Organic ligands are ubiquitous in aquatic environments and promote Hg(II) solubility by competing with binding sites at particulate surfaces [1]. Once Hg(II) is bound to a bacterial surface (*i.e.*, membrane), it can be internalized through an energy-dependent process, leading to Hg(II) accumulation and enhanced biosorption. The object of this study was to determine the impact of cell metabolism on Hg(II) biosorption in the presence of competing ligands with high Hg(II) affinities (*e.g.,* cysteine and EDTA). In controlled lab studies, 50 nM and 500 nM total Hg(II) with and without organic ligands was exposed to metabolizing and non-metabolizing *E. coli* for 3 hours prior to measuring surface adsorbed and intracellular Hg(II). Hg L_{III}-edge XANES and EXAFS spectra of Hg(II) bound to cells under conditions used in sorption experiments were collected to directly link sorption results with Hg(II) coordination environment.

Main Results

For non-metabolizing cells, high concentrations (0.1 mM and 1 mM) of cysteine and EDTA decreased the extent of Hg(II) biosorption compared to a control with no organic ligand. Using a previously documented membrane wash protocol to measure intracellular Hg [2,3], we determine all Hg(II) is located at the membrane of non-metabolizing cells regardless of the presence of organic ligand. In metabolizing cells, however, both EDTA and cysteine enhance the sorption of Hg(II). Most notably, nearly 100% of Hg(II) remaining in solution after the exposure period of metabolizing cells to 0.1 mM cysteine is sorbed to the cells, compared ~ 70% sorbed Hg(II) in the absence of organic ligand. Our membrane wash results for metabolizing cells suggest that all Hg(II) in the presence of 0.1 mM cysteine is located inside the cytoplasm, compared to a majority bound to the membrane in its absence. XANES and EXAFS spectra of Hg(II) bound to non-metabolizing cells for all experimental conditions as well as metabolizing cells without organic ligand are identical and reveal Hg(II) is bound to 2–4 thiol groups, on average. However, Hg(II) bound to metabolizing cells in the presence of cysteine appears to be solely 4-fold coordinated to thiols.

Significance

We show that organic ligands with high Hg(II) affinities promote Hg(II) sorption and internalization by metabolizing bacteria, which is linked to 4-fold Hg-thiol coordination in cells. Hence, organic ligands have promising applications for enhanced Hg(II) biosorption in microbial remediation.

[1] Ravichandran (2004) *Chemosphere* **55**, 319-331. [2] Schaefer & Morel (2009) *Nat Geosci* **2**, 123-126. [3] Thomas, Tong & Gaillard (2014) *Metallomics* **6**, 2213-2222.