Bacterial biouptake and cell-mediated ligand-exchange of Hg(II) complexed with organic ligands

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Introduction

The bacterial biouptake of Hg(II) is an integral process in the bacterial production of methylmercury (MeHg) – a powerful neurotoxin that poses a threat to human health [1]. The formation of Hg(II) complexes with thiol-containing organic ligands in solution has been shown to control bacterial Hg(II) biouptake [2,3]. Yet, the influence of other organic ligands on Hg(II) biouptake is not well documented. Here, a model gram-negative bacterium *(E. coli)* with a chromosomally inserted *mer-lux* fusion was used to probe the biouptake of Hg(II) strongly complexed with synthetic and biogenic organic ligands. Additionally, Hg L_{III}-edge XANES spectroscopy was used to assess the binding environment of Hg(II) associated with the bioreporter cells under conditions relevant to biouptake experiments.

Main Results

Contrary to our expectations, we observed that all synthetic ligands studied (EDTA, EDDS, DTPA and NTA) facilitated Hg(II) biouptake. The biouptake of Hg(II) complexed with biogenic ligands (cysteine, penicillamine, and glutathione) was highly dependent on ligand concentration and type. Consistent with Hg(II) methylation experiments [3], Hg(II) biouptake was greatly enhanced in the presence of micromolar concentrations of cysteine and generally inhibited in the presence of penicillamine and glutathione. Bioreporter exposure to increasing concentrations of Hg(II) fully complexed with EDTA, DTPA, EDDS and cysteine shows the extent of uptake is dose-dependent until a plateau is reached. XANES spectra of Hg(II) associated with the bioreporter indicate that a ligand-exchange occurs between Hg(II) complexed with synthetic ligands and membrane bound thiol moieties.

Significance

This study demonstrates that facilitated biouptake of Hg(II) complexed with organic ligands is not limited to biogenic ligands but occurs with synthetic ligands as well. The ability of thiols at the cell membrane to outcompete aqueous ligands with high Hg(II) affinities and bind Hg(II) has implications for Hg(II) biouptake and fate in the environment.

[1] Hsu-Kim *et al* (2013) *Environ Sci Technol* **47**, 2441-2456. [2] Schaefer *et al* (2011) *PNAS USA* **108**, 8714-8719. [3] Schaefer & Morel (2009) *Nat Geosci* **2**, 123-126.