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ABSTRACT TITLE: Dissolution Potential and Bioavailability of Nanoscale Mercury Sulfides

ABSTRACT: The production of methylmercury (MeHg) in the environment depends, in part, on the speciation of inorganic mercury and the portion that is available for uptake into methylating microorganisms. In benthic sediments and other anaerobic settings, inorganic mercury Hg(II) is typically coordinated to reduced sulfur-containing ligands including inorganic sulfide and natural organic matter. This presentation will discuss research to characterize and identify the forms of Hg(II) that are bioavailable to methylating bacteria. We performed laboratory experiments to demonstrate that the combination of Hg(II), sulfide, and dissolved organic matter (DOM) in solution results in a heterogeneous mixture of dissolved Hg(II) complexes, amorphous Hg-S-DOM nanoparticles, and crystalline HgS particles. The composition of this mixture evolves over time and is not necessarily predicted by equilibrium speciation models. Moreover, in bacterial cultures of sulfate reducers, the production of methylmercury depended on the form of mercury added. Net production of MeHg was greatest in cultures receiving dissolved Hg, while cultures receiving Hg-S-DOM nanoparticles generated 20-40% less MeHg in most cases, depending on the productivity of methylating microbes. This result was in stark contrast to the minimal production of methylmercury in all cultures amended with microcrystalline metacinnabar HgS. The production of MeHg in the nanoparticle exposures also depended on the age of the nanoparticle stock solutions. Small angle X-ray scattering and X-ray absorption spectroscopy was used to characterize the structure of the nanoparticles as they changed in their stock solution. The results indicated that organic matter was altering the rate of crystal ripening and agglomeration of the nanoparticles. These changes resulted in nanoparticles with a lower dissolution potential when added to bacterial culture media in the methylation experiments. These experimental results point to the need of a kinetics-based reaction model that links inorganic mercury speciation (including nanoscale species) and methylation potential. Overall we anticipate that this approach may enable the consideration of nanoscale geochemistry for predictions of Hg reactivity and bioavailability in the environment.