Predicting Methylmercury Production Kinetics in Sediment with a Transient Availability Model

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Project Abstract: East Fork Poplar Creek (EFPC) in Oak Ridge, TN, USA is contaminated with high concentrations of mercury (Hg). In the EFPC ecosystem, anaerobic microorganisms containing the *hgcAB* gene cluster transform Hg to monomethylmercury (MMHg), a neurotoxin that bioaccumulates in the food web. To improve understanding of watershed controls on Hg cycling, our research is designed to: (1) identify ecosystem compartments and hydro-biogeochemical conditions that govern net MMHg concentration, and (2) understand the extent to which groundwater–surface water exchange drives Hg transformations in EFPC. Our work develops a kinetic model for net MMHg production in EFPC sediment that accounts for competing processes that may reduce Hg availability for methylation and MMHg availability for demethylation. This transient availability model combines kinetic expressions for multisite sorption of Hg and MMHg, Hg(II) reduction/Hg(0) oxidation, and methylation/demethylation kinetics.

We conducted experiments in two EFPC sediment types: silty and sandy. The silty sediment has a longer water residence time and is relatively anoxic, carbon-rich, and more metabolically active compared to the sandy sediment. Despite these differences in sediment characteristics, sequencing of the *hgcAB*+ organisms in the two sediments revealed similar diversity distributions, with *Deltaproteobacteria* dominating. Microcosm experiments using stable Hg and MMHg isotopes were used to determine Hg and MMHg sorption rates to the sediments and to track methylation and demethylation in sediment slurry microcosms. We found a much higher MMHg production potential in the silty sediment compared to the sandy sediment. Demethylation rates were similar between the two sediments. Trends in Hg adsorption to the sediments were broadly similar, with moderately more and faster sorption to the silty sediment. MMHg sorption was very different between sediments with 85% of MMHg sorbing to the solid phase after 30h compared to only 24% in the sandy sediment.

Overall, results indicate that silty sediment would have greater overall MMHg production in EFPC due to the high MMHg production potential and long water residence time. However, in the sandy sediment, a large proportion of any MMHg that is produced could be readily delivered to the water column due to lower MMHg sorption rates. The similarity in methylator diversity in the sediments indicates that differences in MMHg production are not due to different methylating microbial communities but could be due to differences in *hgcAB*