

Role of geochemistry and non-mercury methylators in supporting mercury methylation in aquatic sediments

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Aquatic sediments are hotspots for the microbial production of methylmercury (MeHg), a bioaccumulative neurotoxin that can cross the blood-brain barrier and impair central nervous system functioning. Although it is known that MeHg is produced by mercury (Hg)-methylators under certain geochemical conditions, the influence of the larger microbial community on the production of MeHg is unclear. Sediments from four sites with historical Hg contamination were collected to examine the distribution of putative Hg-methylating and putative non-Hg-methylating microbes using 16S rRNA sequencing and quantitative PCR for the Hg-methylation gene *hgcA* and functional genes of known methylators. Both salinity and organic matter influenced the distribution of Hg-methylators; sulfate-reducing bacteria (e.g., *Desulfosarcinaceae*) dominated marine sediments while in freshwater sediments there was a greater diversity of Hg-methylators, including *Methanogens*, iron reducing bacteria, and *Nitrospiraceae*. Random forest, network, and Spearman correlation statistical analyses indicated that non-Hg-methylators (e.g., *Simkaniaceae*, *Rhizobiaceae*) were strongly associated with MeHg concentration and Hg-methylators across all sediments, though specific non-Hg-methylator keystone families, drivers of microbial community structure and functioning, varied by site. Functional analysis of the 16s rRNA data suggested that non-Hg methylators play an important role in the cycling of carbon and sulfur, for example, in chemoheterotrophy, thiosulfate respiration, and sulfur respiration. Non-Hg-methylators could therefore play a role in Hg-methylation by influencing microbial activity and Hg speciation. Our results demonstrated that sulfate-reducing bacteria are not necessarily the dominant Hg-methylators in all sediment environments and that the potential roles of non-Hg-methylators in Hg methylation need to be further explored. Ultimately, this data could be utilized in the development of microbial predictors for MeHg production and mitigation strategies in sediments.