Chemical and physical speciation of mercury in Offatts Bayou: A seasonally anoxic bayou in Galveston Bay

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Limnol. Oceanogr., 52(4), 2007, 1380-1392 | DOI: 10.4319/lo.2007.52.4.1380

ABSTRACT: A chemical equilibrium model was used to predict the solution speciation of dissolved mercury (Hg) in the stratified water column of Offatts Bayou, a subestuary in Galveston Bay, Texas, which undergoes seasonal anoxia in bottom waters. Chemical equilibrium modeling was conducted using conditional stability constants and concentrations of Hg-complexing organic ligands experimentally determined by competitive ligand equilibration methods. Dissolved Hg complexation was dominated by interactions with sulfide and dissolved organic matter (DOM) ($HOHgHS^0$, HOHgHS(DOM), $HgSHS^-$, and HgS_2^{2-} at all depths. Sulfide and glutathione competed for methylmercury (MeHg) complexation in oxic layers; in anoxic waters, sulfide complexation dominated MeHg speciation. The particle-water distribution coefficient (K_d) of Hg decreased in the anoxic layer of the water column, where the dissolved sulfide concentration increased, providing evidence that sulfide complexation influences the solubility of Hg. The solubility of MeHg was elevated in the anoxic as compared to the oxic layers, and this distributional feature was coincident with a change in the solution speciation of dissolved MeHg from glutathione/sulfide complexation in the oxic layers to a predominantly sulfide complexation in the anoxic layers. Maximum enrichment of Hg, MeHg, and iron (Fe) in suspended particulate matter was observed in the lower layer of the pycnocline, most likely resulting from formation of insoluble Fe oxide, which scavenged dissolved Hg-sulfide and MeHg-sulfide species. The concomitant decrease in dissolved inorganic Hg, Fe, and sulfide in the anoxic layers is suggested to result from scavenging of inorganic Hg by FeS, which is in accordance with the Hg speciation model. Overall, Hg cycling in the water column of Offatts Bayou was associated with sulfide and DOM complexation, Fe dissolution/precipitation, water column production of MeHg, and/or efflux of MeHg from anoxic sediment.