Roles of Dissolved Organic Matter in Aqueous Speciation of Hg(II) and CH_3Hg^+ in East Fork Poplar Creek (EFPC), Oak Ridge, Tennessee

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The aqueous speciation of mercury [Hg(II)] and methylmercury (CH_3Hg^+) in contaminated East Fork Poplar Creek (EFPC) at Oak Ridge, Tennessee, is not well understood despite recognition of the important roles played by dissolved organic matter (DOM) in controlling the cycling, transport and bioavailability of Hg in the system. We determined Hg species distributions in the EFPC based on the water chemistry and a selected database of reaction constants of Hg(II) and CH_3Hg^+ with various organic and inorganic ligands, using the "PHREEQC" geochemical speciation computer code. Results show that DOM affects both Hg(II) and CH_3Hg^+ speciation by forming strong Hg(II)-DOM and CH_3Hg-DOM complexes through reactive sulfur groups or thiol-like functional groups in EFPC surface waters under conditions of pH = 7.5 – 8.1 and DOM ≈ 3 mg/L. The neutral complexes of $Hg(OH)_2$ and Hg(OH)Cl, and CH_3HgCl and CH_3HgOH were found to be of secondary importance in EFPC surface water. The competitive effects of co-existing metal ions (i.e., Ca^{2+}, Mg^{2+}, Zn^{2+}, Cu^{2+}, Ni^{2+}, Cd^{2+}, Pb^{2+}, Fe^{3+} and UO_2^{2+}) on the Hg(II)-DOM and CH_3Hg-DOM complexation in the EFPC surface water were evaluated using analogous model organic compounds (i.e., cysteine and glutathione) in the lack of reliable binding constants of these metal ions with thiol-like functional groups in DOM. Results show that only Zn^{2+} (present at relatively high concentrations in EFPC) competes with Hg(II) for the binding sites (thiol) in DOM and therefore contributes to the decreased distribution of Hg(II)-DOM. The effects of other metal ions on the distribution of Hg(II)-DOM and CH_3Hg-DOM complexes are estimated to be minimal.