Roles of Dissolved Organic Matter in the Speciation of Mercury and Methylmercury in a Contaminated Ecosystem in Oak Ridge, Tennessee

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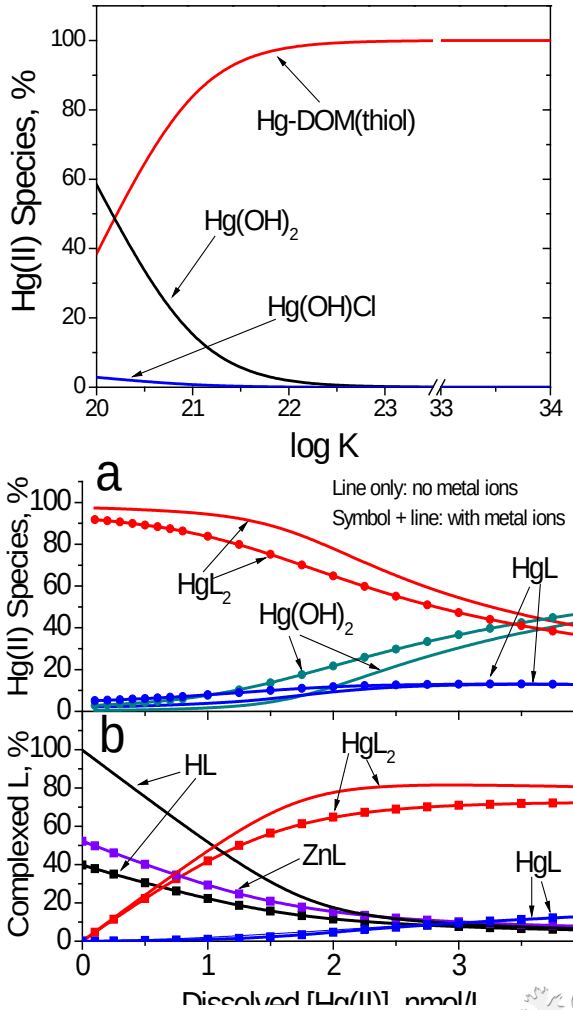
DOE/Office of Science/Biological & Environmental Research

- Mercury (Hg) presents an environmental concern owing to its transformation to the potent neurotoxin methylmercury (CH_3Hg^+). The environmental factors that control bacterial methylation of Hg are poorly understood, but we know that methylmercury is bioaccumulated and biomagnified in aquatic food webs.
- In this study, we conducted thermodynamic modeling to evaluate the dominant mercury species in the creek at the Oak Ridge Reservation.
- We show that even at low concentrations (~3 mg/L), natural dissolved organic matter strongly complexes with ionic Hg^{2+} and CH_3Hg^+.
- Coexisting metal ions may also influence Hg complexation. Results show that among various metal ions, only zinc ion, at the concentration of 0.16–0.26 nanomolar, competes with Hg^{2+} for binding with DOM, causing a decrease in the Hg-DOM complexation. However, this competition has little impact on CH_3Hg^+-DOM complexation.



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Complexation of the mercuric ion (Hg^{2+}) and methylmercury (CH_3Hg^+) with organic and inorganic ligands influences transformation and bioaccumulation of mercury in aquatic environments. In this work, aqueous speciation modeling was performed to determine the dominant species of Hg^{2+} and CH_3Hg^+ in a contaminated upper East Fork Poplar Creek in Oak Ridge, Tennessee. Using aqueous geochemical modeling, we show that natural dissolved organic matter (DOM), even at low concentrations (3 mg/L), controls the Hg speciation by forming strong Hg-DOM and CH_3Hg-DOM complexes through the reactive sulfur or thiol-like functional groups in DOM at a pH of about 7.8. In the presence of DOM, concentrations of neutral species $Hg(OH)_2$, Hg(OH)Cl, CH_3HgCl, and CH_3HgOH are negligible. Of the coexisting metal ions (including Ca^{2+}, Mg^{2+}, Zn^{2+}, Cu^{2+}, Ni^{2+}, Cd^{2+}, Pb^{2+}, Fe^{3+} and UO_2^{2+}), only Zn^{2+}, at concentrations of $1.6–2.6 \times 10^{-7}$ M, competes with Hg^{2+} for binding with DOM, causing decrease in Hg-DOM complexation but having little impact on the CH_3Hg^+-DOM complexation. This study concludes that DOM plays a dominant role in the speciation of Hg^{2+} and CH_3Hg^+ and thus potentially controls the biological uptake and methylation of Hg in this contaminated ecosystem.

Dong, W., L. Liang, S. Brooks, G. Southworth and B. Gu. 2009. Roles of dissolved organic matter in the speciation of mercury and methylmercury in a contaminated ecosystem in Oak Ridge, Tennessee. Environ. Chem. 7:94-102 (doi:10.1071/EN09091).