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Speciation of Mercury on Carbon-Based Sorbents and Iron-Bearing Swelling Clays under Reduced Conditions using HERFD-XANES

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The mobility and transformation of mercury (Hg) in soils and aquatic systems are strongly influenced by its speciation^[1], and controlled by complex interactions with organic matter, sulfidic species, pH and redox conditions. Sorbents, either engineered materials or natural clay minerals can reduce Hg mobility through sorption processes^[2]. However, relative stabilities of sequestered Hg in reducing environments remain poorly characterized. In this study, aqueous reduction using ascorbic acid (AA, $E^{\circ} = +0.06 \text{ V}$) and stannous chloride (SnCl₂, $E^{\circ} = -0.14 \text{ V}$) was used to assess the relative binding strengths of Hg[3] bound to carbon-based sorbents: granular activated carbon (GAC) and two commercially available Hg remediation sorbents- MerSorb® (SORB) and MercLok® (LOK). Experiments were also conducted with Fe-bearing swelling 2:1 clays: bentonite (BEN), nontronite (NAu-1, 30% Fe(III)), and smectite (SWy-1, 3% Fe(III), 0.3% Fe(III)). After addition of AA or SnCl₂, purgeable elemental Hg (Hg⁰) and dissolved Hg were measured at 0 and 24 hours. The fraction of Hg⁰ released from carbon-based sorbents was <1% of total Hg from AA treatments to 2% of total Hg for SnCl₂ treatments. The fraction of Hg⁰ ranged from 7% to 12% of total Hg in experiments with clay samples. Solid-phase Hg speciation after 24 hour AA reduction was analyzed using high-energy-resolution fluorescence detection XANES (HERFD XANES) at the SSRL beamline 15-2. Speciation analysis revealed Hg(II) as the dominant species in GAC (92%), LOK (82%) and BEN (75%) while Hg⁰ was the dominant species in NAu-1 (75%), and Hg(II) and Hg⁰ were present in SWy-1 (53% and 47% respectively). In SORB Hg(I) was the dominant species (72%). These findings suggest that ligands and functional groups in carbon sorbents significantly influence Hg reduction behavior and binding strengths. These insights improve our understanding of Hg cycling under reducing conditions and have implications for the design of Hg remediation strategies.

References

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