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Stable Isotopes of Mercury Bioavailable Fractions from Sediment to Bivalves

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Contamination of sediment by mercury (Hg) in aquatic environment is of significant ecosystem and public health concern given the toxicity and bioaccumulative nature of Hg. Many prior studies have reported absence of significant Hg relationship between sediment and benthic organisms, suggesting the presence of multiple chemical species of Hg with varying bioavailability within the sediment. Here, we collected sediment samples from three Hg contaminated sites (Hyeongsan; 17 mg/kg, Bonghwa; 4.0 mg/kg, Yeocheon; 13 mg/kg) across South Korea and characterized the isotopic compositions in three sequentially extracted fractions (F1-F2; weak acid-soluble, F3; organo-chelated) including the bulk sediment. We then deployed Asian clams (*Corbicula fluminea*) grown from unpolluted location (sediment; 0.018 mg Hg/kg) to the Hg contaminated sites to identify the chemical species of Hg that is preferentially transferred to Asian clams from the sediment. Across all Hg contaminated sites, the organo-chelated Hg fractions (F3) displayed significantly lower δ^{202}Hg values (0.47-0.70 ‰ lower than bulk) relative to the bulk sediment and F1-F2 fractions. Negligible δ^{199}Hg differences were observed among the sequentially extracted fractions and the bulk sediment. After 28-days of deployment, the Asian clams (initial δ^{202}Hg; -0.62 ‰, δ^{199}Hg; 0.35 ‰) from all Hg contaminated site exhibited measurable isotopic shift towards the F1-F2 fractions. Using the initial Asian clam and F1-F2 fractions as end-members, the binary mixing model suggests that 57-62% of sediment Hg is transferred to the Asian clams within a 28-day period. Overall, our study suggests that weak acid-soluble forms of Hg (F1-F2) is more bioavailable relative to the organo-chelated forms in bivalves. Our results are consistent with prior studies that showed significant Hg relationships between bivalves and porewater, subjected to Hg diffusion from sediment. Our study demonstrates that Hg stable isotopes can be used as a powerful tracer for assessing the bioavailable fractions of Hg in sediments.