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# Isotopic Constraints on Mercury Sources and Transformation Following Reservoir Flooding in a Subarctic Estuary

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Methylmercury (MeHg) exposure in Arctic food webs poses significant ecosystem and human health risks. While inorganic mercury (IHg) is naturally present, environmental perturbations such as reservoir creation can stimulate microbial methylation, enhancing MeHg production. However, it remains unclear whether MeHg increases following such perturbations primarily reflect newly mobilized Hg sources (e.g. river inputs) or enhanced in situ transformation within water column. Here, we use Hg stable isotopes coupled with carbon ($\delta^{13}C$) isotopes in zooplankton from a Subarctic estuarine fjord, Lake Melville (Labrador, Canada), to evaluate changes in Hg sources and transformation following upstream reservoir flooding. Zooplankton Hg isotope signatures shifted within two years of flooding, with $\delta^{202}Hg$ increasing by 0.35‰ and $\Delta^{199}Hg$ by 0.95‰. The $\Delta^{199}Hg/\delta^{202}Hg$ (2.04±0.24) and $\Delta^{199}Hg/\Delta^{201}Hg$ (1.30±0.02) slopes, used to distinguish between IHg photoreduction and MeHg photodegradation, are consistent with aqueous MeHg photodegradation and indicate that zooplankton contain varying fractions (5–72%) of photochemically processed MeHg. These isotope shifts coincide with ~2.9- and ~2.8-fold increases in total Hg and MeHg concentrations, relative to pre-flooding conditions, as well as ~1.8-fold higher reservoir water MeHg and a ~1.1°C increase in the surface-midwater temperature gradient, consistent with enhanced thermal stratification. Simultaneous negative shifts in $\delta^{13}C$ (to -28.8±0.1‰, 1SD) suggest increased incorporation of terrestrial-derived carbon, supporting enhanced methylation from newly flooded organic matter and downstream transport of bioavailable MeHg. While zooplankton $\Delta^{200}Hg$ values remained stable (0.03±0.02‰, 1SD) and comparable to lake sediment $\Delta^{200}Hg$ (0.04±0.03‰, 1SD), strong lake water column stratification that limits benthic inputs, suggest that sedimentary MeHg is unlikely to be a major source to surface zooplankton. At outer marine sites, zooplankton exhibit lower $\Delta^{199}Hg$, resulting in depleted $\Delta^{199}Hg/\delta^{202}Hg$ (0.49±0.12) and $\Delta^{199}Hg/\Delta^{201}Hg$ (1.19±0.08) slopes, along with increased $\delta^{13}C$ values (-21.5±1.7‰, 1SD), indicating isotopically distinct coastal MeHg sources with less photodegradation prior to entering the food web. Together, our results are consistent with enhanced methylation associated with newly flooded organic matter in the reservoir, reflected in downstream biota. Given multi-decadal recovery times and ongoing climate-driven changes in stratification and riverine inputs, such perturbations may delay recovery or reshape MeHg exposure pathways in Arctic ecosystems.