

## **Fish muscle and liver mercury isotope ratios reveal anthropogenically influenced estuaries**

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Multiple anthropogenic mercury sources exist along the west coast of South Korea. Given that much fisheries products originate from the west coast, understanding the sources and processes governing ecosystem mercury exposure is key to mitigating human health impacts. We measured total mercury concentration (THg) and mercury isotope ratios in the sediment and fish (e.g., marbled flounder, flathead grey mullet) collected from seven locations along the western estuaries of South Korea. The  $\delta^{202}\text{Hg}$  (-0.73 to -0.43‰) and  $\Delta^{199}\text{Hg}$  (0.00 to 0.15‰) of the sediment were similar to estuarine sediments in the San Francisco Bay and in the northeastern U.S. ( $\delta^{202}\text{Hg}$  = -0.99 to -0.30‰,  $\Delta^{199}\text{Hg}$  = -0.04 to 0.19‰; Gehrke et al., 2011; Kwon et al., 2014). Using the mercury isotope ratios of the fish muscle composed mainly of MeHg, we estimated  $\delta^{202}\text{Hg}$  reflecting methylmercury (MeHg) prior to photo-degradation. The comparison between the estimated MeHg  $\delta^{202}\text{Hg}$  and the sediment  $\delta^{202}\text{Hg}$  at each estuarine site revealed small differences (0.10 to 0.35‰), suggesting that MeHg produced from the sediment is the dominant source to the estuarine fish. Despite the low THg in the sediment (7.5 to 27 ng/g), we found elevated fish muscle THg (23 to 442 ng/g) and liver to muscle THg ratio (0.5 to 17). The fish liver to muscle THg ratio has been used to quantify the degree of inorganic mercury (IHg) exposure and sites impacted by anthropogenic activities. The liver to muscle THg ratios were negatively correlated with the magnitude of  $\delta^{202}\text{Hg}$  differences between the muscle and liver ( $r^2 = 0.43$ ,  $p < 0.05$ ). This suggests that the tissues of fish inhabiting sites impacted by anthropogenic activities are overwhelmed by anthropogenic mercury sources. The large  $\delta^{202}\text{Hg}$  difference between the muscle and liver at sites where ecosystem mercury exposure is low and liver to muscle THg ratios are low indicates that internal MeHg demethylation is unlikely to explain the  $\delta^{202}\text{Hg}$  difference. Our study suggests that THg and mercury isotope ratios in the muscle and liver of fish can help identify sites impacted by on-going anthropogenic activities.