Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.19247

Reconsideration of marine bivalves as mercury biomonitoring tool: a national isotopic survey along the coast of South Korea.

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Coastal sediments act as a reservoir for inorganic and organic contaminants. Among them, mercury (Hg), a toxic and bioaccumulative heavy metal, is efficiently transferred from sediment to marine biota and ultimately to humans via fisheries consumption. Marine bivalves have been recognized as an effective bioindicator for sediment pollution as their suspension or deposit feeding behaviors are physically and chemically tied to the sediment. Due to their broad geographic distribution, bivalve-based monitoring programs, known as the 'Mussel Watch', had been developed around the world including South Korea. However, due to multiple natural and anthropogenic Hg sources, absence of significant Hg relationships between bivalves and sediment have been reported in coastal regions. To investigate bivalve Hg sources and efficiency as coastal sediment Hg bioindicators, paired sediment and bivalves (blue mussel, *Mytilus edulis*, and Pacific oyster, *Magallana gigas*) were sampled in 50 coastal sites in South Korea and measured for total Hg (THg) concentration and Hg isotope ratios. Sediment THg varied from 0.7 to 195.7 ng·g^{-1}, except for two highly contaminated sites at Pohang (600.1 ng·g^{-1}) and Onsan (1417.9 ng·g^{-1}). Bivalve THg ranged between 21.1 and 225.2 ng·g^{-1} and was not correlated with sediment THg (linear regression, R^2=0.01, p>0.05). Only a handful of studies have compared Hg isotope ratios between bivalves and sediment and observed isotopic differences (mainly δ^{202}Hg values) were either attributed to biogeochemical processes occurring in the different fractions of the sediment prior to exposure or to the preferential accumulation of dissolved Hg from the water column by bivalves [1,2]. Based on these identified shifts, Hg isotope ratios are used to identify biogeochemical and/or ecological factors dictating the extent of sediment Hg bioaccumulation to bivalves. Finally, we further plan to quantify carbon, nitrogen, and sulfur isotopes to assess the importance of species feeding behavior and dietary sources on Hg bioaccumulation.

[1] Kwon et al., Mercury isotope study of sources and exposure pathways of methylmercury in estuarine food webs in the Northeastern US. Environ.Sci.Technol. **2014**, 48, 10089-10097.

[2] Li et al., Environmental origins of methylmercury accumulated in subarctic estuarine fish indicated by mercury stable isotopes. Environ.Sci.Technol. **2016**, 50, 11559-11568.

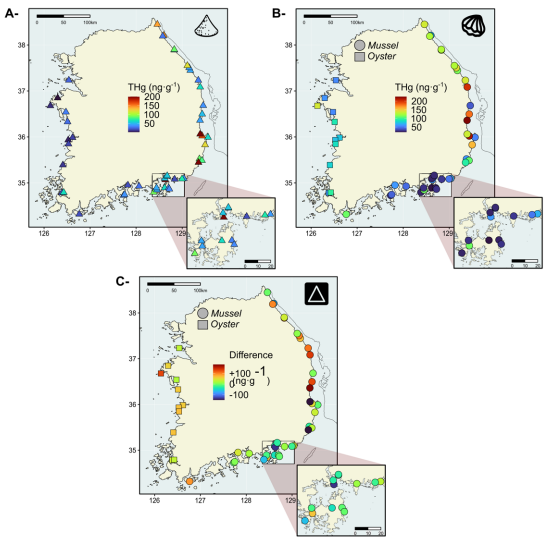

Figure 1-Map of THg concentration (ng·g^{-1}) in sediment (A) and bivalve (B) including mussel (circles) and oyster (squares) sampled along the coast of South Korea. Difference between bivalve and sediment THg is also shown for each site (C). The boundary of the continental shelf is represented by the 200 m bathymetric line in black.

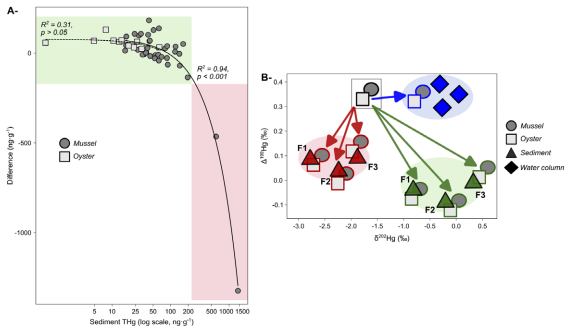

Figure 2-(A) Difference between bivalve and sediment THg as a function of sediment THg. Linear regressions were separately applied for mussels and oysters with associated R^2 and p-value shown in the Figure. (B) Hg isotopic shifts observed in bivalves exposed to Hg from the sediment (in red and green) or to dissolved Hg from the water column (in blue)[1,2] (please refer to the Abstract for references). Isotopic variations between sediment ellipses reflect differences in Hg contamination sources (red *vs* green) while variations inside a similar Hg sediment pool reflect different biogeochemical fractions of the sediment (here F1, F2 and F3) preferentially accumulated by bivalves.