Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.19276

Mercury (Hg) Species Transformations in Shelf to Marginal Waters of the Mediterranean Sea – Experimental Evidence for In-situ Biotic Dimethyl-Hg (DMHg) Formation

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Important efforts have been made to understand the main factors controlling methylated Hg (MeHg = MMHg + DMHg) concentrations in seawater, presumably linked to in-situ microbial methylation of inorganic Hg. The cycling of dimethylmercury (DMHg) in seawater is less understood, but it appears to be connected to the formation and decomposition of monomethyl-Hg (MMHg). Insufficient observational data on DMHg in seawater is impeding the progress of understanding its cycling, primarily due to the challenges posed by analyzing sub-pM concentrations that cannot be preserved for long. Few studies have successfully quantified DMHg formation rates. The western Mediterranean Sea is known for its high methylation potential and high MeHg concentrations. Two stations (shelf, marginal) were chosen to investigate (a-)biotic sources and sinks of MeHg species during two campaigns (shelf: fall, marginal: late spring). Focusing on important biogeochemically defined depths: the surface mixed layer (SML), deep chlorophyll max (DCM), minimum oxygen zone (OMZ), and deep waters (DW). We combined Hg speciation measurements, incubation experiments with species-specific enriched stable isotopic tracers, and microbial diversity analysis (16S rDNA sequencing).

THg, MMHg, DMHg, and dissolved gaseous mercury (DGM) concentrations increased with depth at both stations, reaching maximum concentrations of approximately 1.10 pM, 0.05 pM, 0.45 pM, and 0.40 pM, respectively. DMHg dominates the DGM and MeHg pool below the photic zone in marginal waters. In surface waters, photochemical demethylation was comparable for both stations (range: 0.85-1.33 d^{-1}, n=6) and dark processes were considerably slower (<DL – 0.07 d^{-1}, n= 6). Methylation of inorganic Hg was not detected. Net formation of DMHg was observed from added MMHg at the OMZ (~500m, 6.62 ± 0.44 d^{-1}, n=3) and DW of the marginal station (~800m, 5.48 ± 0.24 d^{-1}, n= 3). The formation of enriched DMHg was not observed at the maximum chlorophyll depth or in filtered controls.

Biologically-mediated processes appear to have contributed to the formation of DMHg, whereby the origin of MMHg requires further investigation. Finally, we combine our results with 1D Modelling and information on the microbial community taxonomic composition to better constrain sources and investigate potential transformation pathways of MeHg in the water column.