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From land to the coast: submarine groundwater discharge (SGD) as an overlooked source of dissolved mercury and its implication for methylmercury formation

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Submarine groundwater discharge (SGD) is a complex hydrogeological pathway that delivers groundwater to the coast, acting as a vector for nutrients, microorganisms and contaminants. SGD may represent an overlooked source of neurotoxic mercury (Hg) and methylmercury (MeHg) to the coastal ocean, becoming relevant for the global oceanic Hg budget and for the coastal biogeochemical cycles.

Hence, we aimed to quantify the SGD-driven Hg fluxes to the Mar Menor, a large Mediterranean hypersaline coastal lagoon subject to polluted SGD, and to elucidate the role of nutrient-enriched SGD in microbial Hg cycling. First, our results revealed that porewaters were enriched in total dissolved Hg (5.13 ± 4.37 pM) and MeHg (0.206 ± 0.053 pM) compared to the lagoon (1.17 ± 0.09 pM of THg and 0.028 ± 0.012 pM of MeHg). These porewaters were distinguished by higher concentrations of nutrients such as ammonium, nitrate, and phosphate, as well as terrestrial fresh humic-like fluorescent dissolved organic matter (FDOM) characterized by lower A/C ratios. Second, the Hg input through SGD to the Mar Menor lagoon (4300 mmol yr^{-1}) was similar to atmospheric deposition (4500 mmol yr^{-1}) and one order of magnitude greater than stream-driven input (60 mmol yr^{-1}). Finally, the low oxygen concentrations (0.46-8.7 mg O_2 L^{-1}) and oxidation-reduction potential (ORP; -89.4 ± 21.4 mV) measured in porewaters were characteristic of iron and, specially, sulphate reduction conditions. These porewaters together with the lagoon shore waters exhibited high concentration of labile dissolved organic matter and higher MeHg concentrations, suggesting biological MeHg formation. This was supported by experimental evidence of microbial Hg methylation in brackish shoreline porewaters using enriched Hg stable isotopes. On-going metagenomic analyses will help elucidate the diversity of the microorganisms involved in the Hg methylation/demethylation pathways and confirm the role of iron and sulfate reducing bacteria in this process.

The combined geochemical and biological approach allowed to identify SGD as a relevant source of dissolved Hg to the coastal ocean which is then methylated into MeHg by active marine benthic microorganisms. Hence, this MeHg becomes bioavailable for biomagnification in coastal food webs, threatening the health of ecosystems.