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Large-scale distribution and pathways of methylmercury in North American Arctic margin sediments

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Methylmercury (MeHg) is the main Hg species that biomagnifies in aquatic food webs and coastal marine sediments constitute a reservoir where inorganic mercury is converted to MeHg by anaerobic organisms, among which sulfate-reducing bacteria have been recognized as highly efficient actors. Up until now, however, there is very few data on the occurrence of MeHg and on Hg methylating pathways in Arctic margin sediments.

Sediment box-cores (40 cm long) were collected along a transect extending from the Bering and Chukchi Seas to the western Beaufort Sea through the Canadian Archipelago and into Baffin Bay were analyzed for total Hg and MeHg as well as for reduced inorganic S, which is a product of sulfate reduction, and for Mn and Fe oxyhydroxides. In most of the cores from the Beaufort Sea, Canadian Archipelago and Baffin Bay, sediments are characterized by high levels of Mn and Fe oxyhydroxides down to several cm depth below the sediment-water interface and low levels and inventories of reduced inorganic S and MeHg. In contrast, sediments from the Bering and Chukchi Seas contain negligible amount of Mn and Fe oxyhydroxides, even in surface sediments, but high levels of reduced inorganic S and MeHg. In these highly productive regions of the North American Arctic Margin (NAAM), reduced inorganic S is already detectable in surface sediments (0-0.5 cm), increases with depth in the sediments and reach concentrations at the bottom of the cores ($\sim$40 cm) that are about 10 times higher than those measured in the sediments of the Beaufort Sea, Canadian Archipelago and Baffin Bay. Likewise, MeHg concentrations increase with depth in the sediments of the Bering and Chukchi Seas to values significantly higher (1-2 ng of Hg per g of sediments; 2-4% of total Hg) than those measured in the sediments of the other regions of the NAAM (<0.2 ng/g; 0.3% of total Hg).

Our results show 1) that the anaerobic oxidation of organic C is mainly driven by the reduction of sulfate in Bering and Chukchi shelves sediments and by that of Mn and Fe oxyhydroxydes in the sediments of the others regions of the NAAM, 2) that significant accumulation of MeHg in NAAM sediments only occurs in the Bering and Chukchi Seas, and 3) that sulfate-reducing bacteria are the key microorganisms responsible for the conversion of inorganic Hg to MeHg in the sediments from these two Arctic Ocean marginal seas.