

## Methylmercury in food webs in northern midcontinental lakes: Bioaccumulation, adverse effects, and trends

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This presentation illustrates key aspects of the environmental mercury (Hg) problem, with emphasis on mid-continental lakes (USA). Atmospheric deposition is the dominant source of Hg in many of these waters, and much of the deposited Hg is from anthropogenic sources. Nearly all of the mercury in fish is methylmercury (MeHg), a toxic compound that bioaccumulates and can biomagnify to high concentrations in organisms atop aquatic food webs. The microbial methylation of inorganic Hg(II) strongly influences the concentration of MeHg in food webs; thus, concentrations of Hg in prey and piscivorous fishes can vary several fold among nearby lakes, in response to variations in ecosystem factors that influence the microbial production and abundance of MeHg. Concentrations of MeHg in predatory fish from many lakes exceed the USEPA fish-tissue criterion of 0.3 mg/kg wet weight, established to protect the health of humans who consume non-commercial fish. Consumption of fish is the primary pathway for exposure of wildlife and humans to MeHg, and documented consequences of MeHg contamination of fish include (1) direct adverse effects on the health of fish-eating humans and wildlife; (2) diminished nutritional, socioeconomic, cultural, and recreational benefits of fishery resources; and (3) adverse socio-cultural effects in some communities that had fished for subsistence. Northern pike with high MeHg concentrations exhibit depressed concentrations of sex hormones essential for development of gonads and gametes, a finding in agreement with recent experimental results. Rates of accumulation of total Hg in lacustrine sediments have recently declined, and data from Minnesota and Ontario lakes suggest concomitant decreases in concentrations of MeHg in game fish.

## Microbial transformations in the mercury geochemical cycle

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Microorganisms play a critical role in the geochemical cycling of mercury by carrying out oxidation and reduction reactions and by transforming mercury between the organic and inorganic forms, thus affecting the accumulation of methylmercury (MeHg) in the environment.

Sulfate reducing bacteria (SRB) in anoxic sediments methylate mercury. However, the biochemical pathways of methylation are not fully understood and recent new data suggest that more than one pathway may be implicated in mercury methylation by SRB.

Methylmercury may be degraded either oxidatively to carbon dioxide and, most likely, inorganic mercury (Hg(II)), or reductively to methane and elemental mercury (Hg(0)), by bacteria that produce the enzymes organomercury lyase, which splits the carbon mercury bond, and mercuric reductase, which reduces the resulting Hg(II) to Hg(0). These enzymes are encoded by mercury resistance (*mer*) operons in mercury resistant prokaryotes in diverse environments. Because *mer* operon functions depend on induction by nM concentrations of Hg(II), we hypothesized that MeHg degradation and Hg(II) reduction by *mer*-specified functions affect mercury speciation in highly contaminated environments while other processes take place in less impacted environments. This hypothesis was accepted by demonstrating *mer* induced reductive demethylation in an industrially contaminated site in New Jersey, USA, while in pristine sites in New Jersey impacted by atmospheric deposition of mercury MeHg was degraded oxidatively [Schaefer et al., 2004] and Hg(II) was reduced photochemically or as a result of phytoplankton activities at the Experimental Lakes Area, Ontario, Canada [Poulain et al., 2004]. The demonstration that inducible reductive MeHg degradation is stimulated in contaminated environments provides the first plausible explanation for the commonly observed trend of low MeHg accumulation in highly contaminated, relative to more pristine, aquatic environments.

Finally, Hg(0) may be oxidized to Hg(II) by enzymes that lessen oxidative stress in microorganisms. The significance of this process in production of Hg(II), the substrate for methylation, in the mercury geochemical cycle is at present unknown.

### References

- Schaefer, J.K. et al., (2004) *Env. Sci. Technol.* **38**, 4304-4311  
Poulain, A. et al., (2004) *Limnol. Oceanog.* **49**, 2265-2275