A latitudinal investigation of aquatic ecosystem sensitivity to methylmercury bioaccumulation in the eastern Arctic

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Overview of the project

The main objective of this project is to investigate how climate affects methylmercury (MeHg) bioaccumulation in Arctic freshwater food webs. Recent evidence indicates that inorganic mercury (Hg) loadings to Arctic lakes decline with latitude (Muir et al. 2009); however, MeHg concentrations in benthic invertebrates and fish do not similarly decline along this gradient in Hg loading (Gantner et al. 2010, van der Velden et al. 2013). These observations suggest that regional environmental factors may play an important role in ecosystem sensitivity to Hg bioaccumulation in the Canadian Arctic.

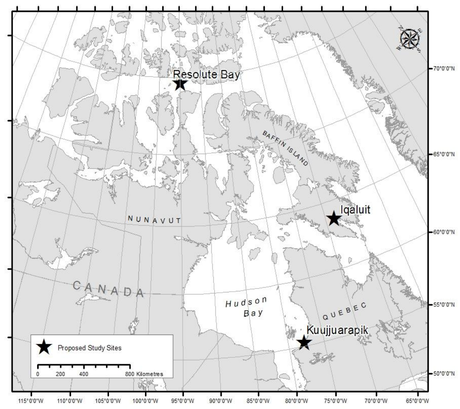

Figure 1. Study locations during the three year project (2012-2015) that fall within representative Canadian Arctic ecosystems along a north-south transect spanning approximately 20° latitude. Kuujjuaraapik, Iqaluit and Resolute are located in taiga, tundra and polar desert environments, respectively.

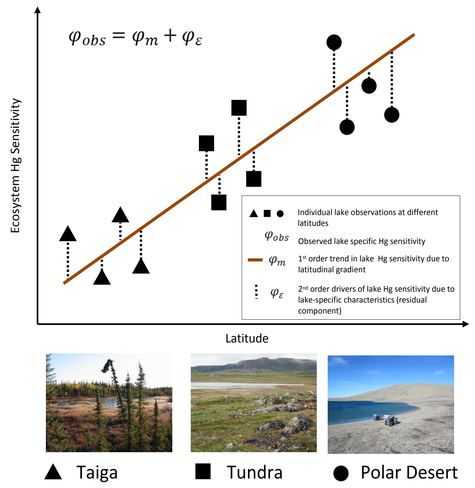

Figure 2. Conceptual framework for characterizing first- and second-order drivers of aquatic ecosystem Hg sensitivity along a latitudinal gradient. We hypothesize that ecosystem Hg sensitivity increases with latitude corresponding to gradients in MeHg bioavailability and organism growth rates. Mercury sensitivity can be quantified as the amount of MeHg in biota relative to Hg loading to the ecosystem.

Year 1: Sub-Arctic taiga

Table 1. Characteristics of the study lakes investigated near Kuujjuaraapik in northern Quebec (July 2012).

Water body	Lake area (km^2)	Mean depth (m)	DOC (mg/L)	SPC (µS/cm)	pH	Fish present?
Site 1	0.08	0.8	6.0	165	8.1	no
Site 2 (Christmas Lake)	0.11	1.3	4.2	102	7.9	yes
Site 3	0.01	0.5	4.1	165	8.0	yes
Site 4 (Kachishayoot Lake)	0.29	2.6	4.0	43	7.1	yes
Site 5	0.05	1.4	7.6	56	7.4	no
Site 6	0.08	0.7	4.4	19	6.6	no
Site 7	0.39	1.2	5.8	46	7.3	yes
Site 8	0.10	1.0	3.9	31	6.6	yes

Lake chemistry and watershed characteristics

The eight water bodies sampled are shallow, first- or second-order lakes with short residence times ranging from approximately ten days to two years. Watershed to lake area ratios range from 2 to 72, implying substantial differences in terrestrial-aquatic influences across the eight study sites. This is corroborated by the strong, positive relationships between several key water quality parameters (including specific conductivity [SpC], pH, THg and MeHg) and watershed area to lake area ratios (Fig. 3a-3d). In contrast to these findings, organic matter characteristics within the lakes (including DOC concentration, UV absorbance and Specific UV absorbance) were unrelated to watershed to lake area ratio. Moreover, none of the water quality variables tested were associated with landcover proportions extracted from Spot 5 satellite imagery.

A strong statistical relationship between MeHg concentrations in lakes and watershed to lake area ratios implies that watershed processes may either enhance the supply of MeHg to the study lakes, or indirectly enhance net-MeHg production within them. There is a particularly strong relationship between MeHg concentrations and SpC within the lakes (Fig. 3e). This relationship might reflect the influence of watershed runoff or groundwater inputs and associated supply of electron donors such as sulphate, on MeHg production in lake sediments or the water column. Within this sub-Arctic landscape, watershed morphometry is a strong predictor of aquatic physico-chemistry and the accumulation of MeHg in lake water.

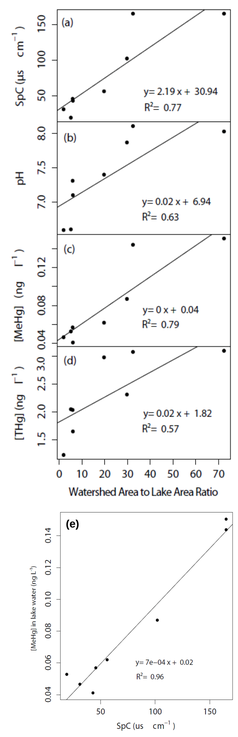

Figure 3. (a-d) Relationships between lake water chemistry and watershed to lake area ratios. (e) Relationship between specific conductivity and MeHg concentration in water.

Food web transfer of methylmercury

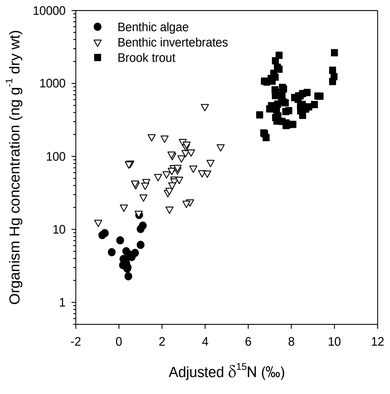

Figure 4. Relationship between adjusted $\delta^{15}N$ ratio and Hg concentration in benthic algae, benthic invertebrates and fish in the lakes.

Concentrations of MeHg in aquatic biota ranged from 2−16 ng g^{-1} dry wt in benthic algae and 17−482 ng g^{-1} dry wt in benthic invertebrates (Fig. 4). Brook trout was the dominant fish species in the study lakes and muscle THg levels ranged from 181−2,642 ng g^{-1} dry wt. Organism Hg concentrations increased as a function of trophic position, as estimated by their adjusted $\delta^{15}N$ ratio (r^2 = 0.81, p < 0.001, n = 122).

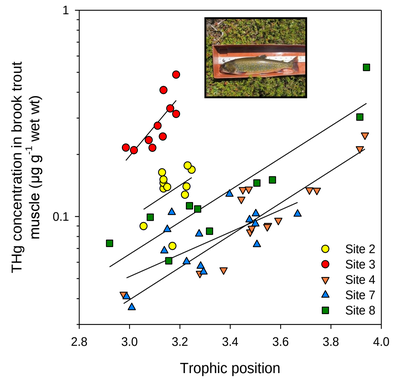

Figure 5. Relationship between trophic position and THg concentration in brook trout muscle from 5 lakes.

The THg concentrations in brook trout varied both within each site and among sites (Fig. 5). In general, the THg concentration of individual fish at each site increased with trophic position, age and total length. After correcting for trophic position, significant differences in average THg concentrations of brook trout were found among sites (ANCOVA, Site p < 0.001, n = 61). Higher concentrations of water MeHg and bioavailable MeHg were measured at sites 2 and 3 where higher brook trout THg levels were also found, indicating site differences were related to MeHg supply to the food web.

Zooplankton dynamics

Zooplankton were sampled from the eight lakes and an additional eight ponds near Kuujjuaraapik (Fig. 6). Relatively elevated average concentrations of MeHg were measured, ranging from 56−415 ng g^{-1} dry wt in bulk zooplankton samples, from 93−675 ng g^{-1} dry wt in sorted *Daphnia*, and from 68−281 ng g^{-1} dry wt in sorted *Chaoborus*. Most samples fell within the range of 57−279 ng g^{-1} dry wt, with the exception of one highly contaminated rock pond (415 ng g^{-1} dry wt for bulk zooplankton, 675 ng g^{-1} dry wt for *Daphnia*). The concentration of MeHg in the zooplankton was not correlated to the concentrations of inorganic or organic Hg in the water column at each site. Other physicochemical and biological variables, such as nutrient stoichiometry and trophic interactions, are likely responsible for variation between sites. Our preliminary results show that samples of sorted *Daphnia* have consistently higher concentrations of MeHg than bulk zooplankton at each site, confirming that *Daphnia* play an important role in the bioaccumulation of MeHg. *Daphnia* have been previously identified as key vectors for MeHg transfer within food chains (Chételat and Amyot 2009).

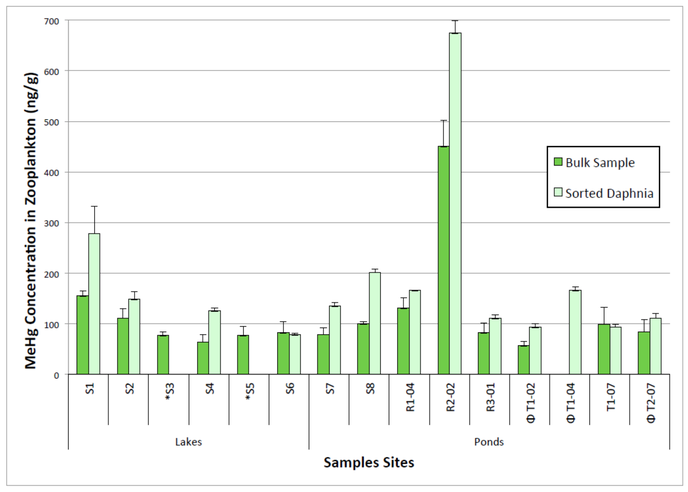

Figure 6. MeHg concentrations in zooplankton from 8 lakes and 8 ponds in Kuujjuaraapik; results shown are for bulk sample (dark green) and sorted samples of *Daphnia* (pale green), * shows lakes with less than 5% *Daphnia*, Φ shows ponds with more than 85% *Daphnia* (% for relative species abundance).

Methylmercury bioavailability in sediment

Diffusive gradient in thin film (DGT) samplers measure the fraction of MeHg in sediment porewater or overlying water that is dissolved and labile (i.e. not bound to large molecules), and therefore is considered most likely bioavailable. DGT measurements of bioavailable MeHg were taken at the sediment-water interface and at depths of 0-6 cm and 6-12 cm in the sediment. Considerable variation in bioavailable MeHg was observed among sites (Fig. 7). Sites with higher bioavailable MeHg concentrations reflect sediment with greater potential to produce MeHg, which subsequently diffuses upwards into the overlying water. Sediment MeHg concentrations measured by DGTs were positively correlated with MeHg concentrations in benthic chironomids (r^2 = 0.71, p = 0.005, n = 8) and site-averaged THg concentrations in brook trout (corrected for trophic position) (r^2 = 0.83, p = 0.021, n = 5). These findings suggest that the DGT samplers are a useful tool to predict MeHg exposure for aquatic biota in sub-Arctic lakes.

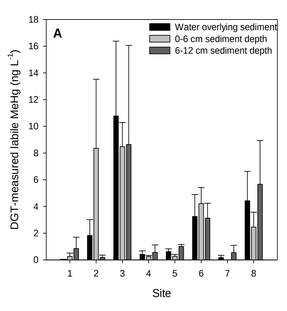

Figure 7. DGT sampler measurements of labile MeHg concentrations in sediment and overlying water.

Summary

We conducted a detailed characterization of mercury cycling in sub-Arctic lakes near Kuujjuaraapik. Our preliminary results show that watershed processes, MeHg bioavailability to food webs, invertebrate taxonomy, and trophic transfer contribute to MeHg bioaccumulation in the sub-Arctic lakes. The information on mercury cycling will be compared to higher latitude sites over the next two years to identify climate influences on MeHg bioaccumulation in aquatic food webs of the eastern Arctic.

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