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Biomagnification of mercury through lake trout (*Salvelinus namaycush*) food webs of lakes with different physical, chemical and biological characteristics

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HIGHLIGHTS

- Mercury biomagnifies through aquatic food webs to toxic levels in top predator fishes.
- Among-system differences in mercury transfer through food webs occur but have not been explained.
- Diverse lakes supporting lake trout were compared to understand the ecosystem processes that affect mercury biomagnification.
- Higher biomagnification of mercury was found in larger, higher nutrient lakes.
- Results show that the food web processing of mercury is related to ecosystem properties.

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ABSTRACT

Mercury (Hg) biomagnification in aquatic ecosystems remains a concern because this pollutant is known to affect the health of fish-eating wildlife and humans, and the fish themselves. The "rate" of mercury biomagnification is being assessed more frequently using stable nitrogen isotope ratios ($\delta^{15}N$), a measure of relative trophic position of biota within a food web. Within food webs and across diverse systems, log-transformed Hg concentrations are significantly and positively related to $\delta^{15}N$ and the slopes of these models vary from one study to another for reasons that are not yet understood. Here we compared the rates of Hg biomagnification in 14 lake trout lakes from three provinces in Canada to understand whether any characteristics of the ecosystems explained this among-system variability. Several fish species, zooplankton and benthic invertebrates were collected from these lakes and analyzed for total Hg (fish only), methyl Hg (invertebrates) and stable isotopes ($\delta^{15}N$; $\delta^{13}C$ to assess energy sources). Mercury biomagnification rates varied significantly across systems and were higher for food webs of larger (surface area), higher nutrient lakes. However, the slopes were not predictive of among-lake differences in Hg in the lake trout. Results indicate that among-system differences in the rates of Hg biomagnification seen in the literature may be due, in part, to differences in ecosystem characteristics although the mechanisms for this variability are not yet understood.

1. Introduction

Individual fish of the same species vary in their concentrations of mercury (Hg) within and among systems because of many well-described factors. Fishes that are slow-growing, older, and piscivorous, instead of fast-growing and insectivorous, and that live in freshwaters that promote the methylation and bioavailability of Hg tend to have the highest Hg concentrations (Wiener et al., 2003). High Hg concentrations in predaceous fishes can adversely affect the health of fish-eating wildlife and humans (Burgess and Meyer, 2008; Chan et al., 2003) in addition to risks of Hg intoxication in the fishes themselves. Decreases in survival, growth and reproduction are found in Hg-exposed fishes due to its effects on the endocrine and nervous systems (Crump and Trudeau, 2009; Kidd and Batchelar, 2011; Sandheinrich and Wiener, 2011; Weis, 2009). Increasing concern over low-exposure effects of Hg on humans and wildlife (UNEP, 2011) reinforces the need to understand the ecosystem processes affecting Hg in top predator fishes.

Methylmercury (MeHg), the organic and most abundant form of Hg in most fish tissues (>95%; Bloom, 1992), biomagnifies through food webs because it is accumulated in proteins more rapidly than

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it is excreted (Trudel and Rasmussen, 2006). Concentrations of Hg in primary through tertiary consumers are significantly related to their trophic position, which can be determined by tissue $\delta^{15}N$ (a measure of relative trophic position within a food web) (Campbell et al., 2005; Kidd et al., 1995; Wyn et al., 2009). The slope of the regression between log-transformed Hg [MeHg or total Hg (THg)] and $\delta^{15}N$ in organisms describes the average biomagnification "rate" across trophic levels within a given system (Borgå et al., 2012). Across diverse climates [arctic, temperate, tropical (Atwell et al., 1998; Campbell et al., 2005; Kidd et al., 1995, 2003; Swanson and Kidd, 2010)] and systems [oligotrophic, acidic, and eutrophic lakes (Eagles-Smith et al., 2008; Wyn et al., 2009; Rolfhus et al., 2011), reservoirs (Chumchal and Hambright, 2009), streams (Chasar et al., 2009)], biotic concentrations of Hg are consistently, positively related to $\delta^{15}N$. As such, one can start contrasting results and assessing whether ecosystem characteristics affect the biomagnification of MeHg in aquatic communities (Jardine et al., 2006; Borgå et al., 2012).

Physical, chemical and biological characteristics of systems affect Hg concentrations in aquatic biota (Munthe et al., 2007). For example, lower Hg concentrations are sometimes found in biota from more productive systems (Kidd et al., 1999; Larsson et al., 2007). This phenomenon may be because of contaminant dilution in the lowest trophic levels (dilution due to an abundance of detrital and organic particles) (Larsson et al., 1992; Pickhardt et al., 2002, 2005), and growth dilution (fast growth rates for longer lived organisms) across several trophic levels (Larsson et al., 1992; Hammar et al., 1993). In addition, the lower Hg in fishes from larger (i.e., surface area, Bodaly et al., 1993), less acidic (e.g., Wyn et al., 2009) or lower sulfate or dissolved organic carbon (DOC) lakes may be due to the effects of water temperature or chemistry on the methylation of inorganic Hg to MeHg and/or its availability to primary producers and consumers (Gilmour et al., 1992, 1998). Concentrations of Hg in fishes are tightly linked to those in their prey (e.g., Hall et al., 1997; Wyn et al., 2009) and, as such, processes at the base of the food web influence the Hg concentrations in predaceous fishes. It is also possible that the food web processing of Hg affects its concentrations in top predators; perhaps fishes from one lake are higher in Hg than fishes from a neighboring system because the rate of Hg transfer in the former lake is greater. Delta^{15}N has emerged as an important tool for addressing questions such as this one.

Broader spatial comparisons of Hg in lake biota are invaluable in understanding the ecosystem characteristics that promote Hg bioaccumulation in fishes and biomagnification through their supporting food webs. However, most of the previous studies examining Hg versus $\delta^{15}N$ relationships were conducted on one or a few food webs within a smaller region (e.g. Wyn et al., 2009); only recently have larger scale comparisons been done to understand whether trophic transfer of Hg is consistent across systems with variable biotic communities and/or physical/chemical properties (Gantner et al., 2010; Rolfhus et al., 2011). While variability in the rate of Hg biomagnification exists across food webs, the drivers that underlie these differences are not completely understood. In this study, we examined Hg biomagnification through 14 lake food webs across three provinces in Canada; these lakes were also part of a larger study examining the trophic transfer of persistent organic pollutants (POPs) (Houde et al., 2008) and factors affecting concentrations of POPs in lake trout (Guildford et al., 2008). We assessed food web structure using $\delta^{15}N$ and $\delta^{13}C$; the latter is used to distinguish reliance of consumers on benthic or pelagic carbon because of the often unique signatures of benthic and pelagic primary producers and the conserved signals from prey to consumer (Hecky and Hesslein, 1995). Our objective was to understand whether Hg biomagnifies at a similar rate through food webs supporting lake trout regardless of the inherent characteristics of the system. In addition, we assessed factors related to among-system differences in Hg concentrations in lake trout (*Salvelinus namaycush* W.), including their reliance on benthic versus pelagic carbon using $\delta^{13}C$, and in the model intercepts of log Hg versus $\delta^{15}N$.

2. Materials and methods

2.1. Field collections

Lake trout (n = 14 to 20/lake) and other fishes (n = 1 to 20/species/lake) were collected from 14 lakes between 1998 and 2001 from Ontario, Saskatchewan and Alberta (Canada; Fig. 1). These lakes are found within two main geological regions — the Canadian Shield for all of lakes in Ontario and Reindeer and Wollaston Lakes in Saskatchewan, and the Western Plain for the remaining systems. These lakes were chosen because they have important subsistence or sport fisheries, and were in relatively undeveloped watersheds (exception is Lake Simcoe). In addition, they ranged in surface area from 11 to 7900 km^2, in mean depth from 9 to 50 m, and from oligotrophic to eutrophic (Tables 1 and S1). Collections of fishes varied from lake to lake (2 to 8 species/lake) and depended on the species present and the success of the capture gear (see Table S2 for total numbers, species and general dietary habits). Fishes were collected in the spring or fall using trap and gill nets, minnow traps and angling, but were sampled in only one year. Fresh weights and total lengths were taken from all fishes. Small-bodied fishes were frozen whole for further processing in the lab whereas larger fishes were subsampled on site for dorsal muscle. All tissues were kept frozen at −20 °C.

Invertebrates were collected from all lakes in the same years as the fish sampling. In four lakes (Sandybeach, Eva, Paguchi and Thunder), zooplankton and littoral invertebrates were collected 3 times (June, July and August) during the open water season and samples were kept separate by date; in the remaining 10 lakes, lower-trophic-level sampling was done in the summer months (typically mid-June through mid-August), a time with less seasonal variability for stable isotopes (Kidd et al., 1999), with several independent replicates collected during one trip. Cold and Simcoe Lakes were sampled in September. Bulk plankton were collected from all lakes by towing a 156 μm mesh net through the water column; in some lakes these samples may include some larger algal taxa but visual inspections of the samples were made to ensure that they consisted mostly of zooplankton. Herbivorous/detritivorous and predaceous macroinvertebrates were sampled from the littoral zone of 10 of the 14 lakes (no benthic invertebrates were collected from Athabasca, Simcoe, La Ronge and Opeongo; only dragonflies were collected from Wollaston and Reindeer), live sorted into major taxa, and then frozen along with the zooplankton (Table S3). Water samples were collected and analyzed for a number of parameters [Total Phosphorous (TP), Total Kjeldahl Nitrogen (TKN), Mg, K, Cl, Ca, Al, Fe, Mn, Dissolved Organic Carbon (DOC), Na, SO_4], including chlorophyll *a* and algal biomass as described in Houde et al. (2008). Based on TP concentrations, three lakes were oligotrophic, one was eutrophic, two were meso-eutrophic, and the remainder was mesotrophic (Table 1). Water and sediments were not collected for MeHg analyses.

2.2. Lab analyses

Total mercury (THg) concentrations in wet, homogenized individual fish muscle or whole bodies were determined using cold-vapor atomic absorption spectrometry. Within each species, fish were selected from those captured to represent a range of sizes. Certified standard reference materials were analyzed with each run and recoveries were 85.1 ± 9.05% (n = 6) for DORM-2 (dogfish muscle, certified value 4.64 ± 0.26 ug/g dw), 102 ± 9.65% (n = 18) for Tort-2, (certified value 0.27 ± 0.06) and 90.7 ± 10.9% (n = 16) for DOLT-2 (dogfish liver; certified value 2.14 ± 0.28, National Research Council of Canada). Replicate injections (n = 12) deviated from the mean an average of 2.6%. The detection limit for THg analyses was 10 ng/g with a wet sample mass of 0.25 g. A subsample from each fish was dried to a constant weight to determine % moisture and this was used to convert THg concentration from wet to dry weight for some statistical analyses. THg was not standardized to either muscle or whole body because a previous study