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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 ($\delta^{15}\text{N}$), 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 $\delta^{15}\text{N}$ 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 ($\delta^{15}\text{N}$; $\delta^{13}\text{C}$ 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.

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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 predatory 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 $\delta^{15}\text{N}$ (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 $\delta^{15}\text{N}$ 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 $\delta^{15}\text{N}$. 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. $\Delta^{15}\text{N}$ 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 $\delta^{15}\text{N}$ 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 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$; 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 $\delta^{13}\text{C}$, and in the model intercepts of log Hg versus $\delta^{15}\text{N}$.

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 km², 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 $-20\text{ }^{\circ}\text{C}$.

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, SO₄], 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 $85.1 \pm 9.05\%$ ($n = 6$) for DORM-2 (dogfish muscle, certified value $4.64 \pm 0.26\text{ }\mu\text{g/g dw}$), $102 \pm 9.65\%$ ($n = 18$) for Tort-2, (certified value 0.27 ± 0.06) and $90.7 \pm 10.9\%$ ($n = 16$) 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



Fig. 1. Locations of study lakes in Ontario, Saskatchewan and Alberta, Canada.

found no differences in muscle and whole body THg concentrations in two species of small-bodied fish (Swanson and Kidd, 2010). It was also assumed that all Hg in fish was MeHg (Bloom, 1992). Though MeHg was not measured in fishes in this study, THg of lake trout, lake whitefish, yellow perch and ninespine stickleback is an average of 100 (muscle), 100 (muscle), 96 (whole body) and 93% (whole body) MeHg (Lasorsa and Allen-Gil, 1995; Wyn et al., 2009; Swanson, unpublished data).

MeHg concentrations were determined in freeze-dried, homogenized invertebrates (individuals pooled within dates to obtain adequate mass; shells or cases removed as appropriate) using GC-atomic fluorescence emission detection by Flett Research (Winnipeg, MB, Canada). Invertebrates (2–9 mg dw) were digested in 300–1000 μL volumes of 25% KOH/methanol at 75 $^{\circ}\text{C}$ for 16 h in acid-washed Teflon vessels. Aliquots

of 60–100 μL were analyzed directly for MeHg with GC-atomic fluorescence emission detection using the techniques described in Horvat et al. (1993) and Liang et al. (1994). Spiked matrix duplicates and DORM-2 reference materials were also analyzed after every 10 samples. Recoveries of the certified standard averaged $91 \pm 6\%$ ($n = 14$) and spike recoveries averaged $96 \pm 5\%$ ($n = 14$). Replicates of 10% of the samples had a deviation from the mean that averaged 1.6 ng/g or 5.9%. Replicate injections for 10% of the samples deviated from the mean on average 1.1 ng/g or 2.4%. Detection limits were about 0.5 ng MeHg/g dry tissue with 5 mg samples. Subsamples of homogenates were dried to a constant weight to determine % moistures (typically 80–90%).

Stable nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$; non-lipid extracted tissues) isotope analyses of dried and homogenized fish muscle or whole, pooled invertebrates (shells removed if present) were done as described

Table 1

Selected physical, chemical, and biological characteristics of the 14 lake trout lakes sampled in Alberta (AB), Saskatchewan (SK) and Ontario (ON), Canada (see also Table S1).

Lake	Code	Longitude ($^{\circ}\text{W}$)	Latitude ($^{\circ}\text{N}$)	Surface area (km^2)	Mean depth (m)	Max depth (m)	Algal biomass (mg/m^3)	TP ^a ($\mu\text{g}/\text{L}$)	Trophic status	TKN ^b (mg/L)	Chlor <i>a</i> ($\mu\text{g}/\text{L}$)
Cold, AB	Co	110.0	54.5	373	50	99	850	33.0	Meso-eutr	0.545	2.9
Grist, AB	Gr	110.0	55.3	25	10	NA	NA	16.0	Meso	0.383	NA
Namur, AB	Na	111.5	57.5	42	15	31	NA	14.0	Meso	0.416	NA
Athabasca, AB/SK	At	111.0	58.5	7900	26	120	1280	44.0	Eutr	0.428	3.0
Kingsmere, SK	Ki	106.0	54.0	47	21	47	1200	21.0	Meso-eutr	0.472	1.2
La Ronge, SK	La	105.0	55.3	1178	13	38	1000	8.0	Oligo	0.487	1.0
Reindeer, SK	Re	102.0	57.0	5569	17	215	350	13.0	Meso	0.213	0.8
Wollaston, SK	Wo	103.0	58.0	2062	21	97	440	12.0	Meso	0.191	0.8
Eva, ON	Ev	91.2	48.6	17	14	54	458	6.8 (4.7–8.7)	Oligo	0.247 (0.239–0.256)	2.5 (1.8–2.9)
Opeongo, ON	Op	78.2	45.4	59	15	52	226	6.0	Oligo	0.269	1.7
Paguchi, ON	Pa	91.5	49.5	25	9	30	397	16.0 (8.5–26.4)	Meso	0.331 (0.285–0.389)	2.4 (1.5–3.1)
Thunder, ON	Th	92.6	49.8	11	11	21	339	11.0 (10.1–11.5)	Meso	0.282 (0.266–0.289)	1.8 (1.5–3.6)
Sandybeach, ON	Sa	92.3	49.8	38	20	41	270	12.7 (7.0–22.8)	Meso	0.291 (0.277–0.301)	1.9 (1.2–2.7)
Simcoe, ON	Si	79.2	44.2	725	17	25	476	12.6 (8.8–15.7)	Meso	(0.458, 0.488)	(2.2, 2.3)

^a TP – total phosphorous: 4–10 $\mu\text{g}/\text{L}$ are oligotrophic (oligo), 10–20 $\mu\text{g}/\text{L}$ are mesotrophic (meso), 20–35 $\mu\text{g}/\text{L}$ are meso-eutrophic (meso-eutr) and 35–100 $\mu\text{g}/\text{L}$ are eutrophic (eutr); for all water samples, $n = 1$ unless mean and/or (range) provided, then $n = 2$ or 3; NA – not available;

^b TKN – Total Kjeldahl Nitrogen; unfiltered.

previously (Houde et al., 2008). Tissue $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ were standardized against atmospheric N_2 and Pee Dee limestone, respectively; a laboratory working standard, Pharmamedium, was run every 5 to 10 samples for both N and C analyses and precision of replicate samples was 0.05‰ ($n=52$ duplicate samples).

2.3. Data analysis

2.3.1. Standardizations and transformations

Statistical analyses were carried out using SAS® version 9.1.3 (SAS Institute Inc., 2002). When appropriate, \log_{10} transformations were used to generate normally distributed data for statistical analyses and residuals were examined to ensure that transformations were valid. Alpha was set at 0.05 for all analyses unless noted otherwise.

Delta ^{13}C ratios of all fishes were significantly related to C:N ratios (linear regression, $t=-9.77$, $P<0.0001$, $df=1,444$). Fishes with high lipid (indicated by C:N) tend to have more negative $\delta^{13}\text{C}$ ratios, and when there is a significant relationship between $\delta^{13}\text{C}$ and C:N it indicates that differences in lipid among species or lakes may confound differences in $\delta^{13}\text{C}$ (Post et al., 2007). For this reason, we adjusted fish $\delta^{13}\text{C}$ ratios using Post et al.'s (2007) equation [$\delta^{13}\text{C}_{\text{lipid}}$ ($\delta^{13}\text{C}$ adjusted for lipid) = $\delta^{13}\text{C}_{\text{raw}} - 3.32 + 0.99 \times \text{C:N}$] before conducting any further analyses.

To compare among-population differences in trophic position or the regressions of Hg versus $\delta^{15}\text{N}$ across systems, it is important to standardize the $\delta^{15}\text{N}$ of consumers to the base of the food web because nutrient inputs from human activities alter the absolute $\delta^{15}\text{N}$ of prey and their predators (Anderson and Cabana, 2005). In this study, zooplankton $\delta^{15}\text{N}$ differed significantly among lakes (ANOVA, $F=42.63$, $p<0.0001$, $df=13,29$). Baseline standardizations are typically done with a longer-lived primary consumer (clams, caddisflies; e.g., Wyn et al., 2009). Although the $\delta^{15}\text{N}$ of short-lived species can vary over time (Kidd et al., 1999), zooplankton $\delta^{15}\text{N}$ was used in this study to standardize the base of the food web because it was the only invertebrate collected in all 14 lakes. All individual $\delta^{15}\text{N}$ data for a food web were adjusted to a common baseline by subtracting the system-specific mean $\delta^{15}\text{N}$ for bulk zooplankton ($n=3/\text{lake}$) (hereafter referred to $\delta^{15}\text{N}_{\text{adj zoopl}}$).

Although some studies have converted baseline standardized $\delta^{15}\text{N}$ values to trophic positions or levels (TP or TL) by dividing these values by 3.4‰, the average discrimination factor (also called enrichment factor) in $\delta^{15}\text{N}$ from prey to predator (Borgå et al., 2012), there is ongoing debate in the literature about the relevance of an average and common value for all organisms because the discrimination factor is affected in part by the protein quality and quantity of prey (Florin et al., 2011). For this reason, we did not convert baseline-adjusted $\delta^{15}\text{N}$ to TP to calculate biomagnification rates.

Before comparing concentrations of Hg in lake trout across lakes, data were adjusted to a common total length (600 mm) using an ANCOVA model that included lake as a class variable, \log_{10} length as a continuous variable, and an interaction term. Using this model, least squares means (LSmeans) Hg concentrations were calculated for a 600 mm lake trout in each lake.

2.4. Analyses of [Hg] and Hg biomagnification

Least squares mean [Hg] for lake trout (at 600 mm total length) generated with the ANCOVA model described above were compared among lakes with a *post hoc* Tukey's test. A stepwise linear regression was then used to determine whether LSmean Hg concentrations in lake trout were related to physical, chemical, or biological variables in lakes (see Tables 1 and S1 for variables). Collinearity of variables in the regression was examined using condition indices and variance inflation factors, and only variables that were not significantly collinear were retained in the final model.

We assessed whether Hg concentrations in lake trout were related to relative reliance on pelagic and littoral food sources. To accomplish this, we used $\delta^{13}\text{C}_{\text{lipid}}$ and a 2-source mixing model (e.g., Post, 2002)

to estimate the relative reliance of individual lake trout on pelagic and benthic food sources for the 10 (of 14) lakes for which these data were available. Mean $\delta^{13}\text{C}$ for zooplankton and benthic primary consumers (pooled data for 1–3 taxa) were the pelagic and benthic end members, respectively, and we assumed no fractionation of $\delta^{13}\text{C}$ from prey to predator (Post et al., 2007). Mercury concentrations in lake trout were then related to %reliance on pelagic food sources using simple linear regression.

Biomagnification rates of Hg were determined and compared across lakes using linear regression models of log Hg versus $\delta^{15}\text{N}$. All models used THg (ng/g dw) in fishes and MeHg (ng/g dw) in invertebrates. A previous comparison of models using THg in fishes with those using MeHg in fishes found no significant differences (Wyn et al., 2009). Three different estimates of biomagnification (i.e., slope of Hg versus $\delta^{15}\text{N}$) were generated for each lake. The first estimate was generated by regressing unadjusted log Hg concentrations versus raw $\delta^{15}\text{N}$. The second estimate was generated by regressing unadjusted log Hg concentrations versus $\delta^{15}\text{N}_{\text{adj zoopl}}$. For the third estimate, variation in Hg concentration due to fish size was removed before analysis. This was accomplished by regressing unadjusted log Hg against length within lakes, and adding lake- and species-specific mean Hg concentrations to the residuals. Log mercury concentrations adjusted for size (Hg_{size}) were then regressed against $\delta^{15}\text{N}_{\text{adj zoopl}}$ (Table 2). To determine if slopes of $\text{Hg}_{\text{size}}-\delta^{15}\text{N}_{\text{adj zoopl}}$ relationships differed significantly among lakes, an ANCOVA model was run that included Hg_{size} as the dependent variable, $\delta^{15}\text{N}_{\text{adj zoopl}}$ as a continuous independent variable, and lake as a categorical independent variable. A *post hoc* multiple comparison test (q_{crit} adjusted $\alpha=0.001$ to account for multiple comparisons) was used to determine whether the slopes of log Hg_{size} versus $\delta^{15}\text{N}_{\text{adj zoopl}}$ were significantly different among lakes. Finally, to examine whether ecosystem characteristics were related to the slopes or intercepts of these regression lines (log Hg_{size} versus $\delta^{15}\text{N}_{\text{adj zoopl}}$), stepwise regression analyses were performed to relate slopes and intercepts to lake-specific physical and chemical variables (shown in Tables 1 and S1). Intercept comparisons were made across all 14 lakes rather than within a subset of lakes with parallel slopes because the fit of the reduced model (log Hg versus lake and $\delta^{15}\text{N}$; $r^2=0.76$) was within 2 percentage points of the full ANCOVA model (log Hg versus lake, $\delta^{15}\text{N}$, lake by $\delta^{15}\text{N}$; $r^2=0.74$), suggesting that the statistically heterogeneous slopes were, in practical purposes, homogeneous (see Barrett et al., 2010).

3. Results and discussion

Oligotrophic temperate lake food webs often have very characteristic structures, with the top predators (as defined by $\delta^{15}\text{N}$) integrating carbon from both pelagic and benthic resources (as defined by $\delta^{13}\text{C}$) (Hecky and Hesselin, 1995). In the lakes studied herein, lake trout had the highest $\delta^{15}\text{N}$ values of the sampled biota (means of 9.9‰ in Opeongo to 14.1‰ in Kingsmere for raw $\delta^{15}\text{N}$) (Fig. 2; Tables S2 and S3). This species was typically about 1–4‰ higher in $\delta^{15}\text{N}$ than the next highest fish species in each lake and an average of 6.18‰ (Namur) to 9.68‰ (Sandybeach) above zooplankton (median 7.81‰). Zooplankton ranged across lakes in their mean $\delta^{15}\text{N}$ from 1.93 to 9.19‰ and varied over time within lakes by 0.6 to 4‰ in the 4 systems that were sampled multiple times; among-lake differences in mean $\delta^{15}\text{N}$ were not related to system productivity (correlations, $p>0.15$, not shown) (Table S3). The $\delta^{13}\text{C}$ values of zooplankton and benthic invertebrates were distinct at the base of the food web (separated by 5 to 9‰), and ranged within lakes over time by 2.9 to 4.6‰ for zooplankton (Table S3). Lake trout and other fishes typically had $\delta^{13}\text{C}$ that were in between these two groups of invertebrates, indicating reliance on both pelagic and benthic energy sources (Fig. 2, Tables S2 and S3). For the 10 lakes with both zooplankton and benthic invertebrate data, % pelagic carbon in lake trout diets was close to 50% (ranging from 44 to 65%) in seven systems; in three other lakes (Namur, Kingsmere and Thunder), lake trout were feeding 17, 27 and 79%, respectively, on pelagic carbon. Across lakes, measures of size

Table 2

Regressions between log Hg [THg (fish) and MeHg (inverts)(ng/g dw)] versus a) raw $\delta^{15}\text{N}$ (‰), b) after adjusting $\delta^{15}\text{N}$ to the zooplankton baseline ($\delta^{15}\text{N}_{\text{adj zoopl}}$) (‰), and c) after removing effects of fish size on Hg (Hg_{size}); see text for details] in 14 lake trout lakes in Alberta, Saskatchewan, and Ontario, Canada. All models were significant at $p < 0.0001$ ($1,24 < \text{df} < 1,65$) and had R^2 from 0.66 to 0.96 (exception Opeongo with R^2 of 0.20 to 0.26). Post-hoc pairwise differences in slope ($\alpha = 0.001$) are indicated by letters. Because $\delta^{15}\text{N}$ values were adjusted by a constant in Hg_{raw} vs. $\delta^{15}\text{N}_{\text{adj zoopl}}$, R^2 values are identical to those shown for Hg_{raw} vs. $\delta^{15}\text{N}_{\text{raw}}$.

Lake	Hg_{raw} vs. $\delta^{15}\text{N}_{\text{raw}}$		R^2	Hg_{raw} vs. $\delta^{15}\text{N}_{\text{adj zoopl}}$		Hg_{size} vs. $\delta^{15}\text{N}_{\text{adj zoopl}}$		R^2	Pairwise differences
	Slope (95% CI)	Int (95% CI)		Slope (95% CI)	Int (95% CI)	Slope (95% CI)	Int (95% CI)		
Opeongo, ON	0.11 (0.05–0.16)	1.68 (1.23–2.14)	0.26	0.11 (0.05–0.16)	2.00 (1.69–2.30)	0.12 (0.05–0.19)	1.92 (1.51–2.32)	0.20	ab
Sandybeach, ON	0.14 (0.12–0.16)	1.36 (1.21–1.52)	0.79	0.14 (0.12–0.16)	1.80 (1.70–1.91)	0.14 (0.12–0.16)	1.79 (1.68–1.90)	0.78	b
Eva, ON	0.15 (0.12–0.18)	1.50 (1.28–1.72)	0.66	0.15 (0.12–0.18)	1.79 (1.62–1.97)	0.15 (0.11–0.18)	1.82 (1.61–2.02)	0.56	b
Thunder, ON	0.16 (0.13–0.19)	1.29 (1.02–1.56)	0.76	0.16 (0.13–0.19)	1.97 (1.81–2.14)	0.16 (0.13–0.20)	1.95 (1.78–2.13)	0.74	ab
Grist, AB	0.17 (0.15–0.20)	0.739 (0.52–0.96)	0.76	0.17 (0.15–0.20)	1.45 (1.32–1.59)	0.18 (0.15–0.21)	1.43 (1.27–1.59)	0.69	ab
Paguchi, ON	0.18 (0.16–0.21)	0.943 (0.75–1.14)	0.82	0.18 (0.16–0.21)	1.58 (1.45–1.71)	0.19 (0.17–0.21)	1.55 (1.42–1.69)	0.81	ab
Namur, AB	0.18 (0.14–0.23)	0.30 (–0.19–0.79)	0.68	0.18 (0.14–0.23)	1.62 (1.41–1.83)	0.18 (0.14–0.22)	1.62 (1.42–1.82)	0.69	ab
Wollaston, SK	0.19 (0.15–0.22)	0.301 (–0.08–0.68)	0.82	0.19 (0.15–0.22)	1.31 (1.10–1.52)	0.18 (0.15–0.22)	1.32 (1.12–1.53)	0.83	ab
Cold, AB	0.20 (0.18–0.22)	0.295 (0.11–0.48)	0.96	0.20 (0.18–0.22)	1.01 (0.87–1.14)	0.20 (0.18–0.22)	1.02 (0.88–1.15)	0.96	ab
Kingsmere, SK	0.20 (0.16–0.24)	0.045 (–0.46–0.55)	0.80	0.20 (0.16–0.24)	1.38 (1.12–1.64)	0.18 (0.16–0.24)	1.38 (1.13–1.64)	0.81	ab
La Ronge, SK	0.21 (0.16–0.26)	0.123 (–0.43–0.67)	0.66	0.21 (0.16–0.26)	1.41 (1.14–1.69)	0.20 (0.15–0.25)	1.47 (1.19–1.74)	0.63	ab
Athabasca, SK	0.21 (0.18–0.24)	0.500 (0.23–0.76)	0.91	0.21 (0.18–0.24)	1.06 (0.87–1.25)	0.23 (0.19–0.28)	0.94 (0.66–1.22)	0.85	a
Simcoe, ON	0.21 (0.17–0.25)	–0.750 (–1.33– –0.17)	0.77	0.21 (0.17–0.25)	1.16 (0.94–1.39)	0.23 (0.20–0.26)	1.06 (0.89–1.22)	0.88	ab
Reindeer, SK	0.23 (0.20–0.26)	0.350 (0.053–0.648)	0.85	0.23 (0.20–0.26)	1.08 (0.87–1.29)	0.23 (0.19–0.26)	1.09 (0.82–1.35)	0.78	a

or productivity (log surface area, mean depth, log TP) did not predict pelagic feeding by lake trout (linear regression, $p > 0.42$, $\text{df} = 1,8$).

In spite of this variability in dietary habits across populations, % pelagic carbon in lake trout diets and THg concentrations were not related (dw data pooled across lakes; linear regression, $p = 0.53$, $\text{df} = 1,168$). In contrast, Guildford et al.'s (2008) study on many of these same lakes and lake trout found higher ΣPCBs in fish that fed more on pelagic carbon. Within three of the lakes (Kingsmere, Thunder, Reindeer) herein, individual lake trout that relied less on pelagic carbon and more on benthic carbon had higher Hg ($p = 0.002$ – 0.07 , $\text{df} = 1,13$ – 18); within one lake (Opeongo) the relationship was the opposite ($p = 0.003$, $\text{df} = 1,12$). These contrasting “effects” of carbon source on fish Hg have also been found in other systems. In tropical Lake Malawi, fishes linked to benthic carbon had lower Hg than those at a similar trophic level but relying on pelagic carbon (Kidd et al., 2003). In contrast, within eutrophic Clear Lake in California fish species that relied more on benthic carbon (littoral and profundal) had higher concentrations of Hg than fishes supported by pelagic production (Eagles-Smith et al., 2008).

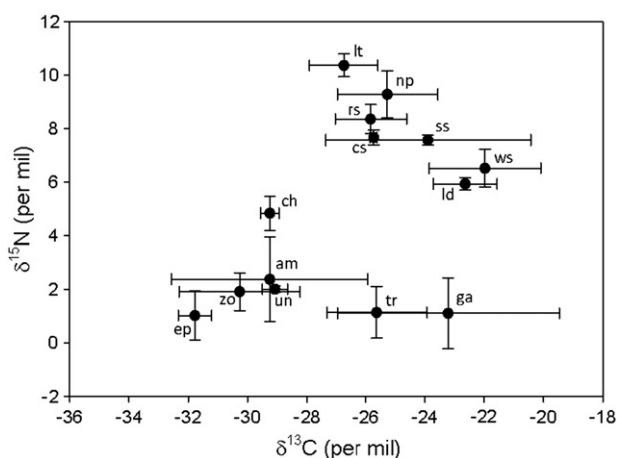


Fig. 2. Mean (\pm SD) $\delta^{15}\text{N}$ versus $\delta^{13}\text{C}$ (‰) of biota from Eva Lake, ON, Canada. (Codes are: am – Amphipoda, ch – chironomids, cs – common shiner, ep – Ephemeroptera, ga – Gastropoda, ld – longnose dace, lt – lake trout, un – Unionidae (mussels), np – northern pike, ns – ninespine stickleback, rs – rainbow smelt, ss – spottail shiner, tr – Trichoptera, ws – white sucker, zo – zooplankton).

Lake-to-lake variability in fish Hg concentrations can often be partly explained by differences in water chemistry or other system characteristics (Munthe et al., 2007). As examples, higher Hg is found in fishes from smaller lakes (Bodaly et al., 1993; Evans et al., 2005), and those with lower productivity (Chen et al., 2005; Kidd et al., 1999) or pH (Wyn et al., 2010). In this study, mean THg in lake trout ranged by ~2–3 fold among lakes from 131 to 407 ng/g ww (Table S2), were similar to concentrations reported previously for this species at temperate latitudes (e.g., Bodaly et al., 1993), and differed across lakes after standardization to a common size (least squares mean Hg at 600 mm length, dw; ANCOVA, $F = 2.40$, $p = 0.0051$, $\text{df} = 13, 198$). Stepwise linear regression analyses revealed that least squares mean Hg concentrations were negatively predicted by only Ca^{2+} after collinear variables were removed ($t = -2.64$, $p = 0.021$, $\text{df} = 1,12$). Lab studies have shown that higher concentrations of cations like Ca^{2+} tend to decrease uptake of metals such as Hg (e.g., Rodgers and Beamish, 1983). While most Hg in fishes is derived from the diet (Hall et al., 1997), direct uptake across the gills also occurs (Kidd and Batchelar, 2011). Calcium can compete with Hg for uptake sites and alter gill permeability and electric charge so as to inhibit Hg uptake (Rodgers and Beamish, 1983). The lack of other significant predictor variables for lake trout Hg concentrations was surprising given that several physical and chemical characteristics of systems are known to affect Hg in fishes (Munthe et al., 2007). For example, DOC was the best predictor of Hg in lake trout in Ontario although lake size and other water chemistry variables were also significant correlates (McMurtry et al., 1989).

As for lake trout, concentrations of MeHg in invertebrates varied between taxa within lakes and within taxa from one lake to another, possibly due to differences in the methylation rates and availability of Hg to the base of the food webs. Mean MeHg concentrations in zooplankton ranged from 2.9 to 52.6 ng/g dw across all of the lakes, by up to 3 fold in lakes sampled several times over the season (data shown in Table S3), and were comparable to concentrations in zooplankton from remote, unimpacted sites (Westcott and Kalff, 1996; Table S3). Predaceous, benthic invertebrates (damselflies, dragonflies) were consistently higher in MeHg than the herbivorous organisms (snails, caddiflies, chironomids) from the same lake and habitat, and support results from previous studies showing that dietary habits of lower-trophic-level organisms are important determinants of their Hg concentrations (e.g., Hall et al., 1998; Wyn et al., 2009; Chételat et al., 2011).

It is clear that $\delta^{15}\text{N}$ can be used across diverse systems to quantify the rate of Hg biomagnification through aquatic ecosystems (Atwell et

al., 1998; Kidd et al., 2003; Campbell et al., 2005; Chasar et al., 2009). In these temperate lake trout lakes, simple linear regressions of log Hg versus $\delta^{15}\text{N}$ (raw) were significant ($p < 0.05$, $df > 1,24$) for all 14 food webs (examples shown in Fig. 3), and $\delta^{15}\text{N}$ explained from 66 to 96% (except Opeongo, $r^2 = 0.26$) of the variability in Hg concentrations (Table 2). The rates of Hg biomagnification determined in this study using raw Hg data (0.12 in Opeongo to 0.23 in Reindeer, Simcoe, Athabasca) were within the range of those for other lakes (Kidd et al., 1995; Wyn et al., 2009; Rolffhus et al., 2011). However, some of the among-study variability may be due to different experimental designs (e.g. no lower-trophic-level sampling as in Kidd et al., 1995; different mesh sizes for zooplankton as in Rolffhus et al., 2011) and, as such, it is not clear how much is due to variable biomagnification of Hg.

In this study similar sampling and chemical analyses were used across the 14 lakes to facilitate among-system comparisons in the rates of Hg biomagnification. Significant differences in log Hg_{size} versus $\delta^{15}\text{N}_{\text{adj zoop}}$ regressions were found among lakes supporting lake trout (Table 2), suggesting that rates of Hg biomagnification vary across temperate systems. Although THg concentrations in lake trout were also significantly different across lakes (see above; Table S2), regression slopes of Hg versus $\delta^{15}\text{N}$ were not correlated to mean size-adjusted THg (dw) in the trout ($p = 0.28$, $n = 14$), indicating that Hg in lake trout was not directly predicted by the rate of biomagnification through the supporting food web.

Growth rates and size of some fishes can affect their Hg, with higher concentrations in slower-growing and older individuals (Trudel and Rasmussen, 2006). Because studies of Hg biomagnification are typically dominated by fish samples with fewer lower-trophic-level biota (Borgå et al., 2012), comparisons between lakes with different fish sizes (and growth rates) could confound interpretations of among-system differences in the slopes of log Hg versus $\delta^{15}\text{N}$. When effects of size on Hg concentrations in lake trout were removed prior to generating the regressions of log Hg_{size} vs. $\delta^{15}\text{N}_{\text{adj zoop}}$, there were few changes in the slopes and overlap in the 95% CI for all regression slopes within each lake (Table 2). In a previous study on coastal Arctic food webs, removal of the size-related variance changed the magnitude of the slope in some but not all systems, and removal of age-related variance acted to decrease slopes in lakes with slow-growing fish (Swanson and Kidd, 2010). Unfortunately, we were unable to examine and compare fish growth rates across lakes (age data were not available). However, across 35 populations of lake trout in Quebec growth rate was not a significant predictor of Hg, although it was for walleye and northern pike (Lavigne et al., 2010).

A number of factors could potentially affect the slopes of regressions between contaminants and $\delta^{15}\text{N}$, including physical or chemical characteristics of the systems, or characteristics of the organisms within the food web. As an example, if there is lower trophic transfer efficiency of Hg (or higher food conversion efficiency) from prey to predator in eutrophic systems, one would expect lower rates of transfer (slopes of the log Hg versus $\delta^{15}\text{N}$) of this contaminant through these food webs when compared to less productive lakes when all else is equal. Across all 14 lakes, slopes of log $\text{Hg}_{\text{size}} - \delta^{15}\text{N}_{\text{adj zoop}}$ regressions were best predicted by log lake surface area (positive) and log TP concentration (positive) (stepwise multiple regression, $F = 10.76$, $p = 0.003$, $df = 2,11$; Table 3 and Fig. 4). This suggests that greater food web biomagnification of Hg occurs in larger, more productive systems. However, the majority of these lakes were mesotrophic (Table 1). Studies on lakes that represent a broader and more balanced range of trophic status would be valuable for understanding whether the rates of Hg biomagnification are consistently higher in more productive systems.

Effects of lake characteristics on the biomagnification of contaminants have been found for some persistent organic pollutants (POPs). In a study that was done concurrently with this one on a larger number of lakes (14 of 17 were the same), the biomagnification rates [calculated as trophic magnification factors (TMFs), the antilog of log [POP] versus TL (calculated directly from $\delta^{15}\text{N}$ using discrimination factors)] were

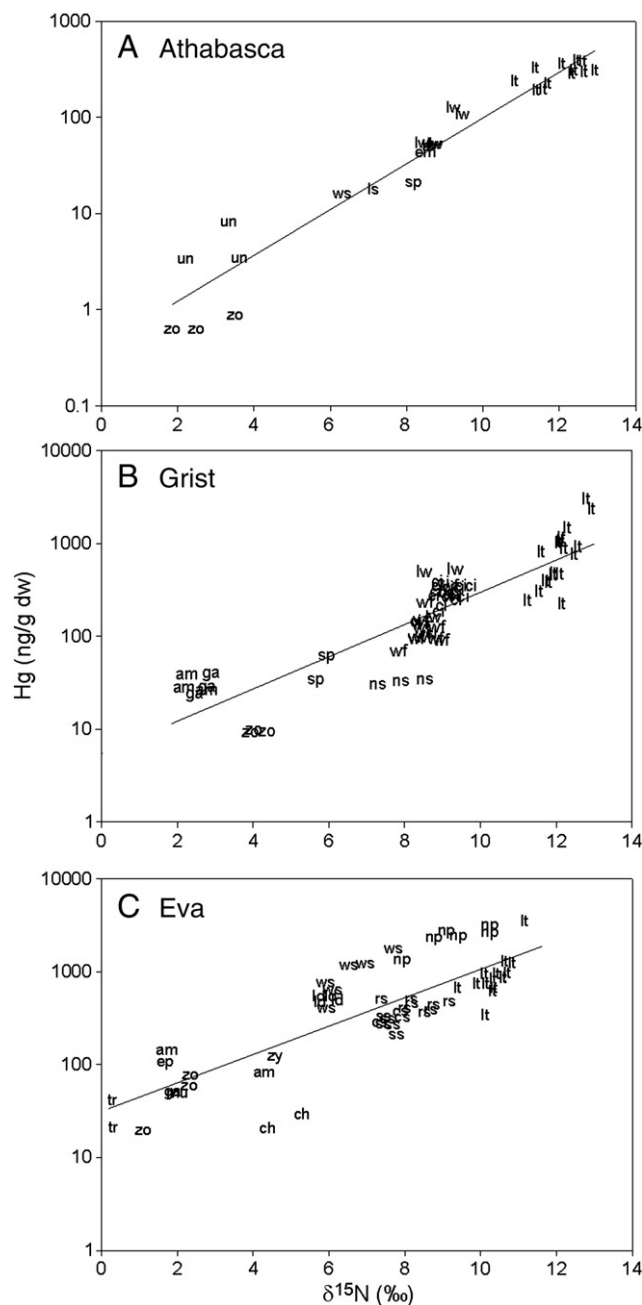


Fig. 3. Regressions of log-Hg ($\mu\text{g/g dw}$) versus $\delta^{15}\text{N}_{\text{raw}}$ (%) for fishes and invertebrates collected from 3 of the 14 lakes in Alberta, Saskatchewan and Ontario, Canada. A) – Athabasca Lake, $\log \text{Hg} = 0.23 (0.02) \delta^{15}\text{N}_{\text{raw}} + 0.32 (0.185)$; B) – Grist Lake, $\log \text{Hg} = 0.18 (0.0146) \delta^{15}\text{N}_{\text{raw}} + 0.699 (0.135)$; C) – Eva Lake, $\log \text{Hg} = 0.15 (0.1695) \delta^{15}\text{N}_{\text{raw}} + 1.53 (0.133)$ (note different scales on Y axes). (see Fig. 2 legend for some abbreviations, also: ch – Chironomidae, ci – cisco, em – emerald shiner, ga – Gastropoda, lw – lake whitefish, wf – whitefish spp., zy – Zygoptera.). Values in parentheses are standard errors.

determined for ΣPCB , ΣDDT , and 13 individual chlorinated pesticides (or their transformation products) (Houde et al., 2008). Some POPs had higher TMFs in lakes at greater latitudes or longitudes [ΣPCB , hexachlorobenzene (HCB), α -hexachlorocyclohexane (α -HCH), lindane, *trans*-nonachlor (longitude only)] or with greater mean depths (CB52, CB153) or surface areas (HCB). In addition, TMFs were also significantly, positively correlated with DOC (cis-Chlordane) or TP (ΣPCB , CB99, HCB, *trans*-nonachlor, α -HCH) for some POPs. The best individual predictor of TMFs in this study depended on the POP. For example, TP was the best correlate for TMFs of ΣPCB whereas log surface area was the best correlate for TMFs of HCB. It is interesting that some of the same physical and

Table 3

Significant predictor variables for slopes and intercepts of $\log Hg_{size} - \delta^{15}N_{adj\ zoop}$ regressions (N = 14 lakes). Slopes and intercepts were related to lake physical and chemical variables with stepwise multiple regression. Slopes were significantly and positively related to surface area and total phosphorus concentration. Intercepts were significantly and negatively related to surface area and total phosphorus concentration, and significantly and positively related to aluminum concentration.

	$Hg_{size} - \delta^{15}N_{adj\ zoop}$ slope			$Hg_{size} - \delta^{15}N_{adj\ zoop}$ intercept		
	t	P	Partial R ²	t	P	Partial R ²
Lake surface area (km ²)	3.04	0.011	0.46	-7.96	<0.0001	0.63
Total phosphorus concentration (mg/L)	2.26	0.045	0.32	-6.48	<0.0001	0.21
Aluminum (mg/L)	.	.	.	3.67	0.0043	0.09

chemical characteristics of lakes affect the biomagnification rates of both POPs and Hg.

Concentrations of Hg in fishes are related to processes at the base of the food web such as its bioavailability, methylation and uptake into lower-trophic-level organisms (Munthe et al., 2007). Greater Hg in fishes is found in systems with higher Hg concentrations in invertebrates and this has been observed in streams, lakes and reservoirs (Chasar et al., 2009; Chumchal and Hambricht, 2009; Wyn et al., 2009). Although it has not been examined previously, the intercept of the Hg- $\delta^{15}N$ regression (adjusted for differences in basal $\delta^{15}N$) may represent inputs of this pollutant to the base of the food web. For these lake trout lakes, intercepts of all $\log Hg_{size}$ versus $\delta^{15}N_{adj\ zoop}$ relationships were positively related to MeHg concentration in zooplankton (Fig. 5A). However, although the latter analysis suggests that the intercepts represent inputs of MeHg to the

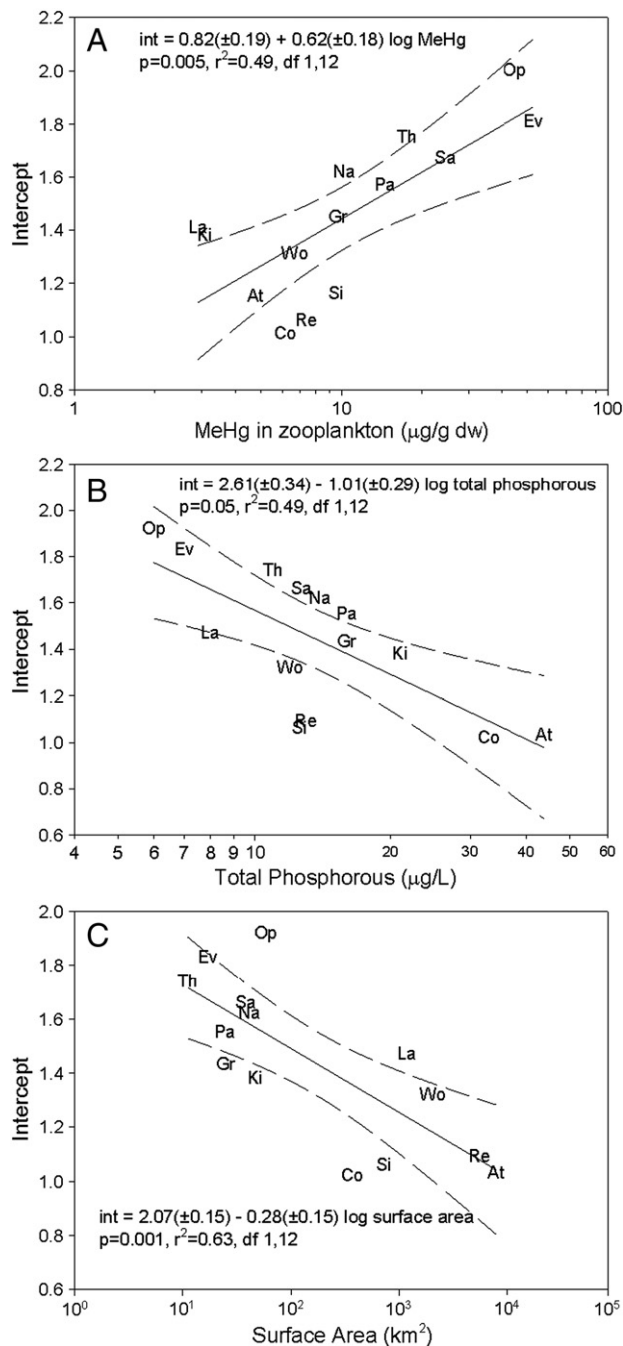


Fig. 5. Intercepts of individual lake regressions [$\log Hg_{size}$ (THg for fishes, MeHg for invertebrates; dw) versus $\delta^{15}N_{adj\ zoop}$] versus A) MeHg in zooplankton (ng/g dw), B) log TP ($\mu\text{g/L}$), and C) log lake surface area (km^2) for 14 lake trout food webs (see Table 1 for lake codes; dashed lines are 95% CI).

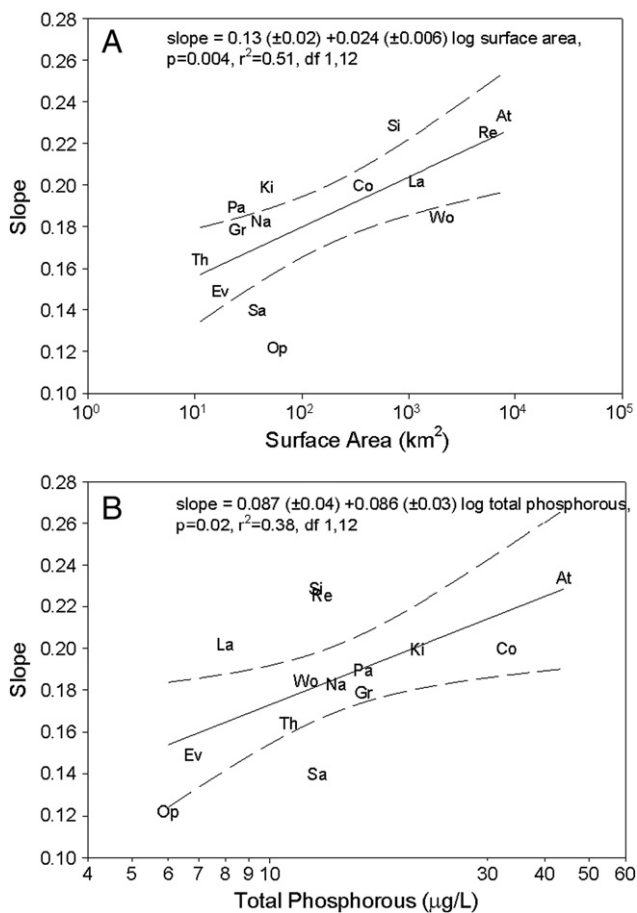


Fig. 4. Slopes of individual lake regressions [$\log Hg_{size}$ (THg for fishes, MeHg for invertebrates; dw) versus $\delta^{15}N_{adj\ zoop}$] versus A) log lake surface area (km^2), and B) log total phosphorous (TP) ($\mu\text{g/L}$) (see Table 1 for lake codes; dashed lines are 95% CI).

base of the food web, among-system variability in slopes also affects the intercepts (lakes with higher slopes have lower intercepts) and warrants consideration. Further, if intercepts of Hg versus $\delta^{15}N$ relationships are to be compared among studies, care must be taken to ensure that data are adjusted for baseline (or not) in a similar fashion, as this will significantly alter intercept estimates (e.g., Table 2).

Assuming that the intercepts do indeed represent inputs of MeHg to the lower trophic levels, we examined factors that may explain their among-lake variability. For all 14 lakes, regression intercepts were best predicted by a model that included log TP (positive), log surface area (positive), and log Al (negative) (stepwise multiple regression, $F = 44.63$, $p < 0.001$ $df = 3, 10$; Table 3 and Fig. 5 B,C). The

effect of TP on the regression intercepts is consistent with other studies that have found lower concentrations of MeHg in organisms from more productive systems (Chen et al., 2005). Similarly, warmer systems are known to have greater Hg methylation rates (Ramlal et al., 1993) and this would explain the higher intercepts in smaller, shallower lakes in this study. The inverse relationship between the intercepts and AI is not possible to explain but may be linked to changes in inorganic Hg or MeHg bioavailability through competitive ligand binding. It is possible that other factors such as pH or watershed characteristics would also explain some of the among-lake differences in the intercepts but these endpoints were not available.

In summary, this study examined the biomagnification of Hg through lake trout food webs across lakes that were geographically remote and that differed in their physical and chemical characteristics. The food web transfer of Hg up to lake trout was similar to what has been observed in other studies and higher in lakes with a greater surface area and TP concentration. However, Hg concentrations in lake trout were unrelated to the rates of Hg transfer (regression slopes) across trophic levels. The mechanisms behind the different rates of food web transfer of Hg are not known, but may be due to among-lake differences in energy transfer efficiencies. While several studies have shown that $\delta^{15}\text{N}$ is a strong predictor of Hg concentrations in food webs, the significance of the regression intercepts is less certain. Here we showed that the intercepts are related to physical (surface area) and chemical (TP, AI) variables that we know or suspect from other studies affect either the methylation or uptake of MeHg into organisms at the base of the food web. Overall, broader scale comparisons such as this one allow us to examine ecosystem variables that affect the movement of Hg through aquatic food webs and can be used to refine models that predict risk to fish consumers and to the fishes themselves.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2012.08.057>.

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