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Environmental, geographic and trophic influences on methylmercury concentrations in macroinvertebrates from lakes and wetlands across Canada

Meredith G. Clayden · Karen A. Kidd · John Chételat · Britt D. Hall · Edenise Garcia

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Abstract Macroinvertebrates are a key vector in the transfer of methylmercury (MeHg) to fish. However, the factors that affect MeHg concentrations and bioaccumulation in these organisms are not as well understood as for fish, and studies on a broad geographic scale are lacking. In this study, we gathered published and unpublished MeHg and carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope data for freshwater macroinvertebrates from 119 lakes and wetlands across seven Canadian provinces, along with selected physical, chemical and biological characteristics of these systems. Overall, water pH was the most important determinant of MeHg concentrations in both predatory and non-predatory invertebrates [$R^2_{adj} = 0.32$, $p < 0.001$; multivariate canonical redundancy analysis (RDA)]. The location of lakes explained additional variation in invertebrate MeHg (partial $R^2 = 0.08$ and 0.06 for latitude and longitude, respectively; RDA), with higher concentrations in more easterly and southerly regions. Both invertebrate foraging behaviour and trophic position (indicated by functional feeding groups and $\delta^{15}N$ values, respectively) also predicted MeHg concentrations in the organisms. Collectively, results indicate that in addition to their feeding ecology, invertebrates accumulate more MeHg in acidic systems where the supply of MeHg to the food web is typically high. MeHg concentrations in macroinvertebrates may also be influenced by larger-scale geographic differences in atmospheric mercury deposition among regions.

Keywords Macroinvertebrates · Methylmercury · Stable isotopes · Lakes · Wetlands

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M. G. Clayden · K. A. Kidd (✉)
Biology Department, Canadian Rivers Institute, University of New Brunswick, 100 Tucker Park Road, Saint John, NB E2L 4L5, Canada
e-mail: kiddk@unb.ca

J. Chételat
National Wildlife Research Centre, Environment Canada, 1125 Colonel By Drive, Ottawa, ON K1A 0H3, Canada

B. D. Hall
Department of Biology, University of Regina, 3737 Wascana Parkway, Regina, SK S4S 0A2, Canada

E. Garcia
Nature Conservancy, 280 Avenida Nazaré, Bairro de Nazaré, Belém, PA 66.035-170, Brazil

Introduction

Fish are exposed to methylmercury (MeHg), the form of mercury (Hg) that bioaccumulates in organisms and is of greatest concern to fish-eating humans and wildlife, primarily through their diet (Hall et al. 1997; Hrenchuk et al. 2012). Littoral macroinvertebrates are important prey for forage and juvenile predatory fishes, and therefore the factors that control MeHg bioaccumulation in macroinvertebrates are also, by association, determinants of fish Hg concentrations. Within primary or secondary consumer groups, macroinvertebrates vary in MeHg concentrations by up to 30-fold from one lake or stream to another (Chételat et al. 2011; Jardine et al. 2013), and some of this variability explains among-site differences in fish Hg (e.g. Wyn et al. 2009). While the factors influencing either fish MeHg concentrations (e.g. pH, organic matter, food web length; Spry and Wiener 1991; Driscoll et al. 1995; Kidd et al. 1999) or rates of net methylation in systems (for a review see Ullrich et al. 2001) are relatively well-studied, only a few studies have examined factors affecting MeHg concentrations in macroinvertebrates (e.g. Chételat et al. 2011; Haro et al. 2013).

Available studies on aquatic macroinvertebrates have shown correlations between MeHg concentrations in individual taxa and water chemistry parameters, but they have been limited to a regional scale (e.g. Rennie et al. 2005). For example, MeHg concentrations in aquatic invertebrates are related to water pH and dissolved organic carbon (DOC), as well as the overall system productivity (i.e. lake trophic status; e.g. Watras et al. 1998; Allen et al. 2005; Tsui and Finlay 2011); however these relationships are inconsistent across taxa. More specifically, Rennie et al. (2005) found that MeHg concentrations in predatory invertebrates in boreal lakes and reservoirs increased with decreasing pH and increasing DOC, whereas collector-shredders had higher MeHg concentrations only in lakes with higher DOC concentrations. Other parameters examined (year and month of collection, lake temperature, redox potential, and surface water ammonium and phosphate and sediment MeHg concentrations) were not explanatory. Allen et al. (2005) found that MeHg concentrations in predatory invertebrates (*Aeshna* and *Cordulia* spp.) in boreal lakes were negatively correlated with aqueous pH and total phosphorus and cation (K^{+}, Ca^{2+}, Mg^{2+}) concentrations. Omnivorous taxa in these lakes exhibited similar negative relationships with cations, but not pH or total phosphorus. Neither taxonomic group showed relationships between their MeHg and aqueous DOC concentrations, speculated to result from different proportions of autochthonous and allochthonous carbon among sites (Allen et al. 2005). Although these regional studies included relatively large numbers of lakes ($n = 12$–23), studies of macroinvertebrate MeHg concentrations across a broader scale are lacking, especially when compared to such studies for fish (e.g. Kamman et al. 2005; Depew et al. 2013). Thus, the extent to which the findings of these regional studies apply on a larger geographic scale is not known.

Within a system, MeHg concentrations also vary across macroinvertebrate taxa and are generally higher in predators than primary consumers (Cremona et al. 2008; Haro et al. 2013), suggesting that dietary habits also affect MeHg in these lower trophic levels (Jardine et al. 2013; Riva-Murray et al. 2011, 2013). Stable isotope analysis (SIA) is a widely used technique for understanding the structure of, and nutrient transfer through, food webs, and is also applied to understanding sources and fate of contaminants in aquatic biota (Jardine et al. 2006). In particular, stable isotope ratios of nitrogen (expressed as $\delta^{15}N$) fractionate from one trophic level to the next. Although diet quality can affect this fractionation (Florin et al. 2011; Bunn et al. 2013), $\delta^{15}N$ adjusted for differences in basal $\delta^{15}N$ is a good proxy for the relative trophic level of organisms. In aquatic systems, $\log_{10}$Hg concentrations in biota are positively related to their $\delta^{15}N$ and the slopes of these models represent the average rate of food web transfer of Hg (Kidd et al. 2012a).

In contrast, because carbon isotope ratios (expressed as $\delta^{13}C$) are not strongly fractionated (<1 ‰) during the trophic transfer of this element, ultimate sources of energy (e.g. pelagic carbon vs littoral benthic carbon in lakes) to food web organisms can be estimated using SIA (Jardine et al. 2006). Although less common in the literature, sources of dietary carbon have also been shown to be important in explaining MeHg variation in macroinvertebrates. More specifically, within sites primary consumers with the lowest $\delta^{13}C$ had the highest MeHg concentrations (Riva-Murray et al. 2013) and among sites macroinvertebrates with more reliance on autochthonous carbon had MeHg concentrations that were affected more by stream water chemistry than those with greater reliance on terrestrial carbon (Jardine et al. 2012). Therefore, within- and among-site differences in macroinvertebrate MeHg concentrations may be explained in part by dietary habits using $\delta^{13}C$ (Cremona et al. 2009). Through a review and analysis of MeHg concentrations in aquatic macroinvertebrates from a range of studies, the objectives of this paper are:

1. To examine and compare concentrations of MeHg within and across functional feeding groups (FFGs) of macroinvertebrates from lakes and wetlands in Canada.
2. To assess the relative importance of physical, chemical and biological characteristics of ecosystems that affect MeHg concentrations in invertebrates of these lakes and wetlands.
3. To determine the biomagnification of MeHg from primary to secondary invertebrate consumers in these systems (using both FFGs and $\delta^{15}N$ of organisms).

Methods

Data sources

Concentrations of MeHg and stable isotope data ($\delta^{13}C$ and $\delta^{15}N$) for freshwater macroinvertebrates from a total of 107 lakes and 12 prairie wetland ponds (119 sites in total) across seven Canadian provinces were obtained from published and unpublished sources, along with available physical and chemical site characteristics. A list of data sources and the characteristics of the systems are provided in Table S1 of the Supplementary Information (SI) file that accompanies this manuscript. The systems ranged in size from 0.9 to 790,000 ha and mean depth from 1 to 50 m.

Table 1 Physical and chemical characteristics (mean ± standard deviation, range and number of sites, n) of study lakes and wetlands

	Mean ± SD	Range	*n*
Latitude (decimal degrees)	49.492 ± 3.540	44.286–59.367	121
Longitude (decimal degrees)	84.443 ± 16.554	65.210–115.907	121
Surface area (ha)	15,351 ± 94,395	0.9–790,000	108
Catchment area (km^2)	37.1 ± 156.8	0.3–1,086.8	66
Mean depth (m)	6 ± 7	1–50	89
pH	6.7 ± 0.9	4.3–9.0	109
MeHg (ng L^{-1})	0.737 ± 1.733	0.04–9.70	32
THg (ng L^{-1})	2.730 ± 1.529	0.44–7.23	32
Specific conductivity (µS cm^{-1})	300 ± 640	12–2,957	61
Chlorophyll-a (µg L^{-1})	3.1 ± 3.9	0.2–22.9	79
Calcium (mg L^{-1})	4.5 ± 7.5	0.3–34.4	78
Sulfate (mg L^{-1})	54.8 ± 205.0	0.5–1,353.0	91
Organic carbon (mg L^{-1})	10.0 ± 10.4	2.0–59.2	120
Total nitrogen (mg L^{-1})	0.275 ± 0.095	0.140–0.545	66
Total phosphorus (mg L^{-1})	0.013 ± 0.009	0.005–0.044	90

With the exception of New Brunswick and Nova Scotia lakes, DOC concentrations were available for a majority of sites in the dataset. Only total organic carbon (TOC) concentrations were available for the lakes in New Brunswick and Nova Scotia; however, TOC in these lakes is believed to be mainly DOC (Clair et al. 2007, 2008). Henceforth, TOC and DOC concentrations from all study sites are referred to as OC. Sulfate and OC ranged from 0.46 to 1,353 to 2.0–59.2 mg L^{-1}, respectively, while pH ranged from acidic (4.3) to alkaline (9.0) across the systems (Table 1). Arctic sites were excluded from this review because MeHg data were often limited to one macroinvertebrate taxon and supporting system characteristics were not available. Each lake or wetland was also assigned to a disturbance category (disturbed or undisturbed) to account for potential anthropogenic influences. They were considered disturbed if either logging or forest burning occurred in the catchment, or there was a local point source of pollution. This information was obtained from previous disturbance indices made by the authors of the original studies (e.g. Chételat et al. 2011), or assigned based on information that was gathered for this study.

The level of taxonomic detail available for different invertebrate groups and systems varied from order to species, so each individual invertebrate sample was assigned to one of five FFGs: scrapers, shredders, filter-collectors, gatherer-collectors or predators (Merritt and Cummins 1996; Barbour et al. 1999). Although this is a coarse way of classifying invertebrates, the use of FFGs facilitated broader comparisons since the distribution of different taxa varied among sites and regions (Table S2). Furthermore, FFGs have generally been found to aptly categorize freshwater macroinvertebrates and the relative amounts of contaminants they accumulate (Cremona et al. 2008). FFGs are also a useful way of grouping these organisms to obtain enough biomass to analyze for Hg and stable isotopes. Shredders, gatherer-collectors and predators were the most common FFGs collected ($n = 66$–97 sites), whereas data for scrapers ($n = 35$) and filter-collectors ($n = 10$) were less common. Approximately 8 % of samples were identified to the species level, whereas 17, 40 and 35 % of samples were identified to the genus, family and order levels, respectively. All MeHg concentrations are reported in µg g^{-1} on a dry weight basis.

Data transformations

All data were examined for normality and homogeneity of variance. MeHg concentrations in invertebrates, as well as all lake and wetland physical and chemical characteristics except pH, were log-transformed. Water sulfate concentrations were measured as the concentration of total aqueous sulfate (SO_4^{2-}) in some studies and sulfate-sulfur in others (S-SO_4); thus, S-SO_4 concentrations were converted to SO_4^{2-} concentrations according to the stoichiometric ratios of these two species. All studies that included analysis of total Hg (THg) or MeHg concentrations in lake water reported unfiltered concentrations, so no adjustments were needed for these data.

Although it has been recommended to use the mean $\delta^{15}N$ ratio of a primary consumer group (such as scrapers and shredders) as a baseline (Anderson and Cabana 2007), these FFGs were not present in all sites. Therefore, to correct for among-site differences in basal $\delta^{15}N$, the taxonomic group of primary consumers with the lowest $\delta^{15}N$ value in each system was used to adjust the $\delta^{15}N$ ($\delta^{15}N_{adj}$) for all other taxa within that system (Cabana and Rasmussen 1996). For some sites, data were only available for one group (Table S2), so $\delta^{15}N$ data could not be adjusted for these systems and were not included in subsequent analyses.

Data analysis

Multivariate canonical redundancy analysis (RDA) was performed in R statistical package (R Development Core Team, http://www.R-project.org) on a sub-set of sites to identify which environmental characteristics best explained mean MeHg concentrations in invertebrates. Because this analysis cannot handle missing data, invertebrate FFGs were grouped into predatory and non-predatory categories, and seven characteristics (latitude, longitude, surface area, mean depth, pH, aqueous organic carbon content and total phosphorus) were chosen to maximize the number of sites that could be included ($n = 62$ lakes). A second RDA was run on a subset of lakes ($n = 43$) where $\delta^{15}N_{adj}$ values of predatory and non-predatory invertebrates were available to test for the additional influence of trophic position (within each invertebrate category) on the analysis. A forward selection RDA procedure (a multivariate extension of forward stepwise regression) was used to identify environmental variables that explained a statistically significant ($p < 0.05$) portion of the variation in invertebrate MeHg concentrations.

Ordinary least squares regression analyses were used to test for relationships between MeHg concentrations in individual FFGs and site characteristics. Significance values of these regressions were Holm corrected to account for the family-wise error rate (Holm 1979). This analysis was conducted in addition to the RDA because of the uneven sample sizes for FFGs and measured environmental variables. Stepwise multiple linear regression modeling ($\alpha = 0.10$ to enter, 0.15 to remove) was also used to test the relative importance of environmental characteristics, trophic position and dietary habits (see below) as predictors of macroinvertebrate MeHg concentrations. In order to incorporate FFGs into these regression analyses, each FFG was assigned a dummy variable. FFGs were not highly correlated with $\delta^{15}N_{adj}$ ($r = 0.432$). Because RDA cannot handle missing data, this multiple regression approach allowed us to analyze a larger number of samples and variables, and provided complementary information to the RDA. A version of Akaike's Information Criterion (AIC) adjusted for small sample size (AIC_c) was used to identify the most likely regression model in each case (Burnham and Anderson 2002). This modeling was done with and without prairie wetland pond sites to investigate the influence of these unique systems (see "Results and discussion" section) on the overall patterns of MeHg in invertebrates. MeHg biomagnification factors (BMFs) were calculated as the ratio of the site-mean concentration of MeHg in predatory invertebrates to that of each non-predatory FFG, as well as to the mean MeHg concentration of all non-predatory invertebrates in that system. Again, stepwise multiple regression analysis using AIC_c values was conducted on the BMFs to determine whether they were influenced by physical or chemical characteristics of the sites. The prairie wetland ponds did not factor into these analyses of BMFs because they did not have the associated physical data available (e.g. surface area, due to their ephemeral nature; Bates and Hall 2012). MeHg biomagnification was also calculated as the slope of the linear regression of log-MeHg concentrations in macroinvertebrates versus their $\delta^{15}N_{adj}$; these slopes were compared among groups of sites using analysis of covariance (ANCOVA; $\alpha = 0.05$).

Results and discussion

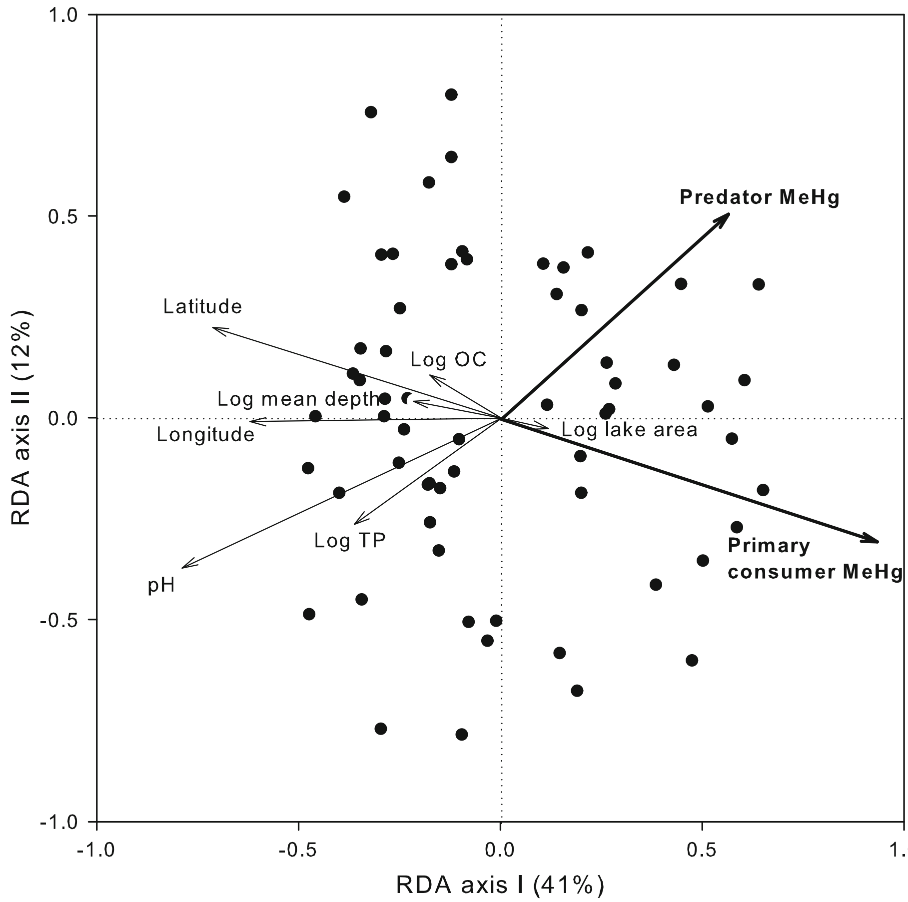
MeHg concentrations in macroinvertebrates

Concentrations of MeHg in macroinvertebrates ranged over four orders of magnitude from 0.0002 to 1.342 $\mu g\ g^{-1}$ among sites and FFGs. However, the majority of MeHg concentrations fell within a more narrow range of 0.013–0.242 $\mu g\ g^{-1}$ (10th and 90th percentiles, respectively). Median MeHg concentration was highest in predators (0.151 $\mu g\ g^{-1}$), followed by filter-collectors (0.081 $\mu g\ g^{-1}$), and was lowest in scrapers (0.046 $\mu g\ g^{-1}$), gatherer-collectors (0.045 $\mu g\ g^{-1}$), and shredders (0.031 $\mu g\ g^{-1}$). Within systems, predatory invertebrates were 0.7- to 28-fold higher (median 3.9-fold) in MeHg concentrations than the non-predatory taxa, and consistent with the general trend of higher concentrations in predatory invertebrates compared to omnivorous or herbivorous taxa in freshwater lakes and wetlands (Chételat et al. 2011; Bates and Hall 2012), streams (Mason et al. 2000; Jardine et al. 2013; Riva-Murray et al. 2011; Tsui et al. 2009), and reservoirs (Tremblay and Lucotte 1997; Hall et al. 1998). Indeed, data from the current study support the general premise that MeHg concentrations in aquatic macroinvertebrates are affected by their dietary habits, and this is examined in more detail below.

Predictors of MeHg concentrations in macroinvertebrates

An analysis of the dataset showed that environmental characteristics, spatial (geographic) factors and trophic position all played a role in explaining the variation in MeHg concentrations in macroinvertebrates across systems. Canonical RDA was conducted on a subset of lakes ($n = 62$) to identify environmental variables associated with site-mean MeHg concentrations in predatory and non-predatory (pooled across FFG) invertebrates (Fig. 1). A forward selection procedure identified water pH as the best environmental predictor ($R^2_{adj} = 0.32$, $p < 0.001$), with higher MeHg concentrations found in invertebrates from acidic systems (Fig. 1). The location of sites (latitude and longitude) explained an additional 15 % of the MeHg variation in predatory and primary consumer invertebrates (cumulative $R^2_{adj} = 0.47$, $p = 0.002$ for both variables). Macroinvertebrate MeHg concentrations declined at higher latitudes and more westerly longitudes. Other site characteristics, specifically mean depth, surface area, and water

Fig. 1 Canonical RDA correlation triplot for associations of environmental variables (site surface area and water pH, OC and TP) and spatial variables (latitude, longitude) with MeHg concentrations in predatory and primary consumer invertebrates. The response variables (invertebrate MeHg concentrations) are bold vectors and the explanatory variables are regular vectors. Points are fitted size scores for a subset of 62 lakes



concentrations of TP and OC, were not significant explanatory variables ($p \geq 0.086$). A second RDA was run on a smaller number of lakes ($n = 43$) for which $\delta^{15}N_{adj}$ data were also available, and the explanatory variables latitude and pH remained highly significant ($p \leq 0.001$) after including minor variation in trophic position of predatory and non-predatory invertebrates. Longitude was no longer a significant variable likely because all the lakes in the reduced RDA were located in eastern Canada. This second RDA suggests that the main differences in trophic position were adequately captured by the separation of invertebrates into predatory and non-predatory categories, and that the absence of $\delta^{15}N_{adj}$ data in the larger RDA with 62 lakes did not markedly affect the observed relationships of invertebrate MeHg concentrations with environmental and geographical variables.

Previous studies have shown that MeHg concentrations in macroinvertebrates are related to several chemical characteristics of systems, and results from this study support these relationships across a broader scale. Using all available water chemistry data on a larger set of data than was used for RDA, water pH and calcium or specific conductivity were found to best explain MeHg concentrations in the four FFGs (ordinary least squares regressions; Table S3; Figs. 2, S1). Specific conductivity explained the most variation in scraper ($R^2 = 0.532$) and gatherer-collector ($R^2 = 0.389$) MeHg concentrations, while calcium or latitude were the best individual predictors of predator ($R^2 = 0.391$) and shredder ($R^2 = 0.327$) MeHg concentrations, respectively (Table S3). Water pH was a highly significant and negative predictor variable within all four FFGs ($R^2 = 0.301{-}0.525$, $p \leq 0.002$; Table S3; Fig. 2), as has been found within predatory and collector-shredder macroinvertebrates in lakes across a smaller geographic region (Rennie et al. 2005) and for some stream taxa (Mason et al. 2000; Harding et al. 2006), likely because of its positive effects on Hg methylation and MeHg availability (Ullrich et al. 2001; Kelly et al. 2003). The negative influence of specific conductivity and calcium concentration (see Table S3) on macroinvertebrate MeHg concentrations likely reflects, at least in part, the association of these water quality measures with water pH because acidic lakes tend to have lower base cation concentrations. However, additional processes may be driving the trends, such as reduced Hg bioavailability in lakes with higher calcium concentrations (Daguené et al. 2012). It is not clear why there were differences in the chemical variables that were most strongly related to the different FFGs, but all of

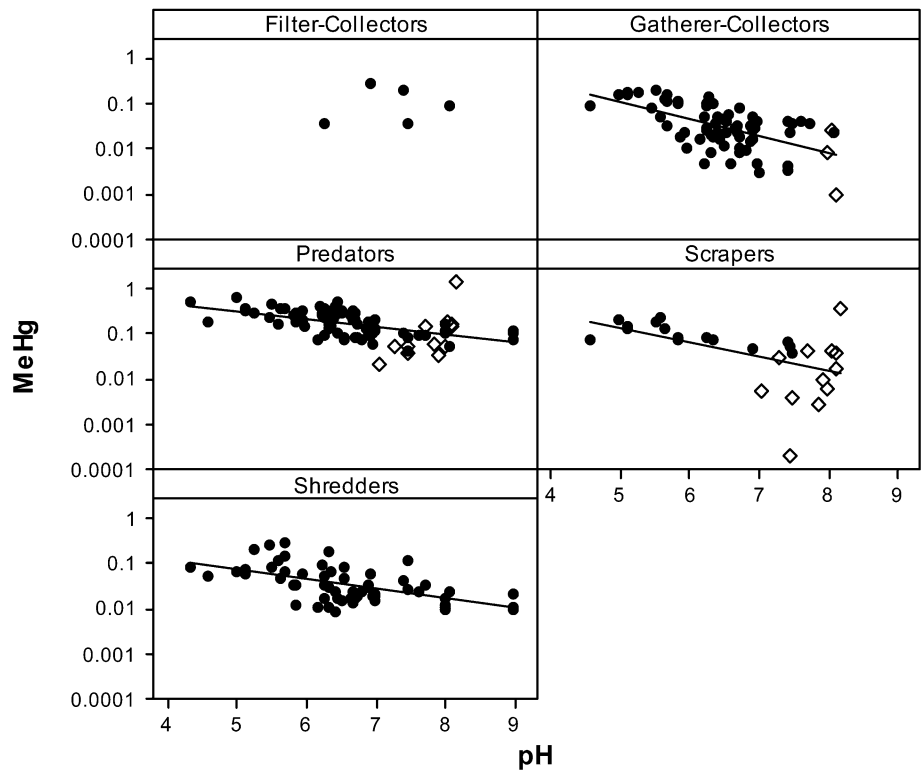


Fig. 2 Linear regressions of site-mean MeHg concentrations (μg g^{-1} dry weight) in filter-collector ($R^2 = 0.040$, $p = 0.74$, $n = 5$ sites), gatherer-collector ($R^2 = 0.301$, $p < 0.001$, $n = 66$ sites), predator ($R^2 = 0.391$, $p < 0.001$, $n = 78$ sites), scraper ($R^2 = 0.525$, $p = 0.002$, $n = 15$ sites) and shredder ($R^2 = 0.317$, $p < 0.001$, $n = 58$ sites) macroinvertebrates from lakes and wetlands in seven Canadian provinces (lakes represented by *filled circle*, wetlands represented by *open diamond*) versus water pH. Wetlands were not included in regression analyses

these relationships may arise from the covariation of pH, calcium and specific conductivity as well as differences among sites in the water chemistry variables that were measured.

Although the physical characteristics of lakes (e.g. size, mean depth, latitude) explain some variability in fish Hg (Bodaly et al. 1993) and in the trophic transfer of Hg through aquatic food webs (Kidd et al. 2012b), comparable information is not available for macroinvertebrates. Similar to the above-mentioned RDA results of the current study, latitude and longitude were significant individual variables that explained 5-45 % of the variation in MeHg concentrations of each FFG (Table S3; Figs. 3, 4). Of the two, latitude was a better predictor of MeHg concentrations in shredders ($R^2 = 0.327$) and predators ($R^2 = 0.211$), whereas longitude explained more of the variability in scrapers ($R^2 = 0.446$) and gatherer-collectors ($R^2 = 0.210$; Table S3). Though it is not clear why the relative importance of latitude and longitude differed among FFGs, the results did indicate that geographic location was an important determinant of MeHg concentrations across FFGs. This spatial influence may be due to geographic variation in ecosystem sensitivity to Hg (Munthe et al. 2007) and/or to patterns of atmospheric Hg deposition. In Canada, atmospheric Hg deposition is greatest in the eastern provinces of Ontario, Quebec and the Maritimes and declines at higher latitudes (Dastoor and Larocque 2004; Muir et al. 2009), consistent with the general spatial trends in macroinvertebrate MeHg concentrations observed in this study.

Water OC concentrations were not a significant predictor of MeHg concentrations in any of the FFGs in this study. This finding contrasts with other studies that have found a positive relationship between MeHg bioaccumulation and aqueous DOC for predatory macroinvertebrates from lakes (Rennie et al. 2005) and in macroinvertebrates from streams (shredders and predators, Riva-Murray et al. 2011; Diptera, Harding et al. 2006; filter feeders, Tsui and Finlay 2011). In another study, all FFGs had higher MeHg concentrations in streams with higher catchment areas, and DOC and nutrient concentrations also increased with catchment area (Tsui et al. 2009). In the current study, differences in OC quality and type among ecosystems may have masked effects, as this has been shown to affect the cycling of MeHg in other systems (Hall et al. 2008; Riscassi and Scanlon 2011).

MeHg bioaccumulation in macroinvertebrates and zooplankton tends to increase with higher water concentrations of MeHg (Watras et al. 1998, Chételat et al. 2011). In this review, concentrations of MeHg in water were negatively related to MeHg concentrations in scrapers and gatherer-collectors when lakes and wetlands were analyzed together ($R^2 = 0.350$ and 0.354, $p \leq 0.004$, respectively). However, when the prairie wetland ponds were removed from the regressions, these relationships were not significant

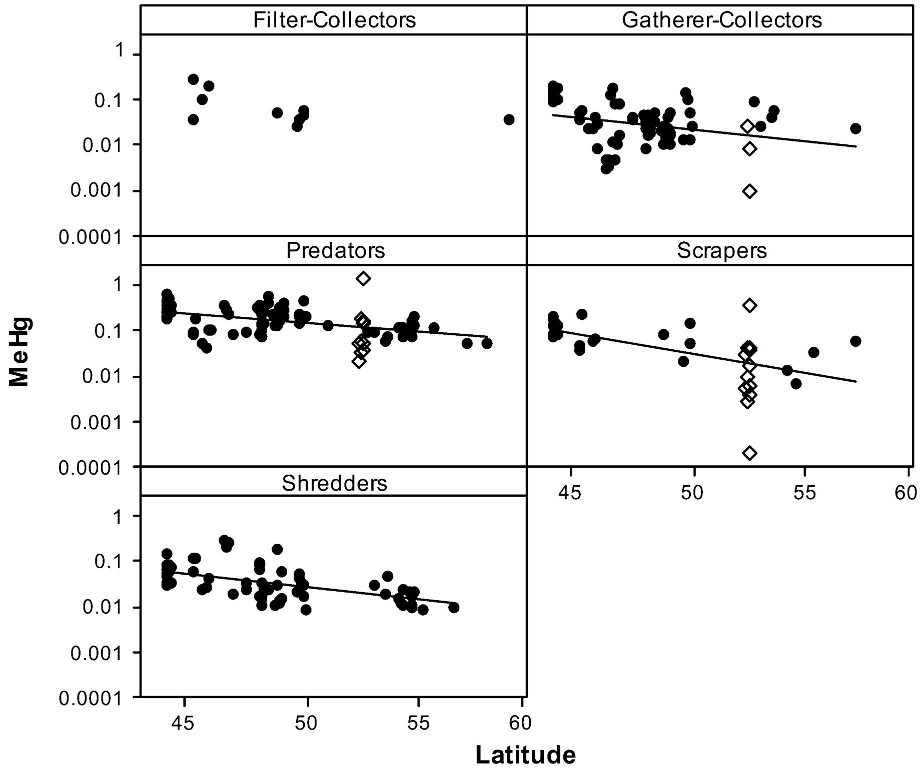


Fig. 3 Linear regressions of site-mean MeHg concentrations (μg g^{-1} dry weight) in filter-collector ($R^2 = 0.297$, $p = 0.103$, $n = 10$ sites), gatherer-collector ($R^2 = 0.049$, $p = 0.060$, $n = 73$ sites), predator ($R^2 = 0.211$, $p < 0.001$, $n = 85$ sites), scraper ($R^2 = 0.394$, $p = 0.001$, $n = 23$ sites) and shredder ($R^2 = 0.327$, $p < 0.001$, $n = 64$ sites) macroinvertebrates from lakes and wetlands in seven Canadian provinces (lakes represented by *filled circle*, wetlands represented by *open diamond*) versus latitude. Wetlands were not included in regression analyses

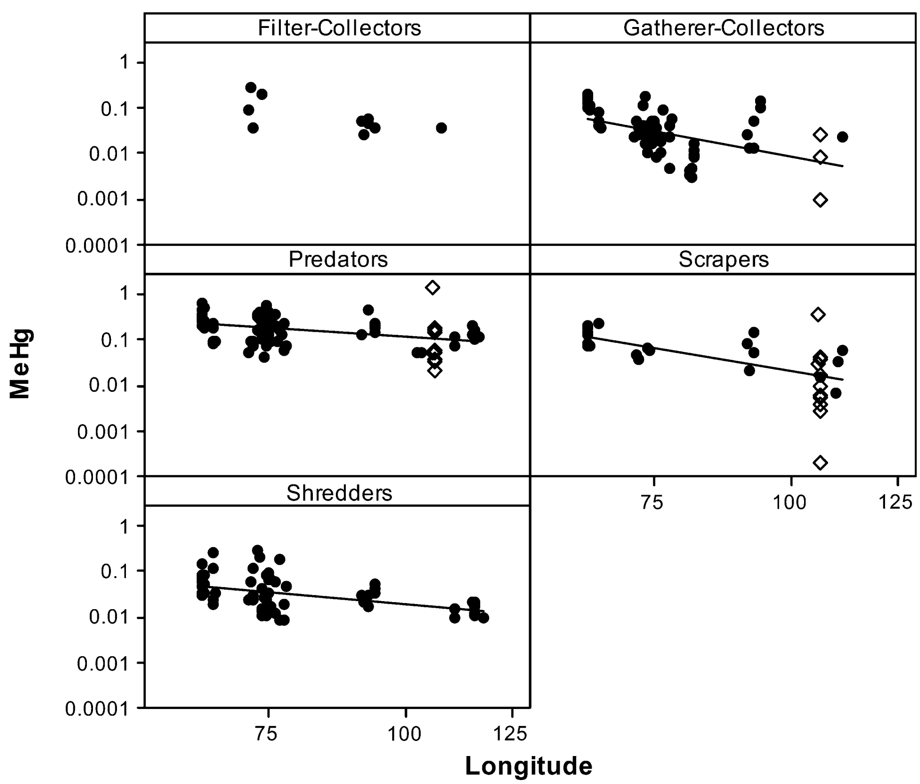


Fig. 4 Linear regressions of site-mean MeHg concentrations (μg g^{-1} dry weight) in filter-collector ($R^2 = 0.455$, $p = 0.032$, $n = 10$ sites), gatherer-collector ($R^2 = 0.210$, $p < 0.001$, $n = 72$ sites), predator ($R^2 = 0.113$, $p = 0.002$, $n = 85$ sites), scraper ($R^2 = 0.446$, $p < 0.001$, $n = 23$ sites) and shredder ($R^2 = 0.230$, $p < 0.001$, $n = 64$ sites) macroinvertebrates from lakes and wetlands in seven Canadian provinces (lakes represented by *filled circle*, wetlands represented by *open diamond*) versus longitude. Wetlands were not included in regression analyses

(Table S3). Thus, the negative relationships found in the current study were driven by elevated water MeHg and low invertebrate MeHg concentrations measured at some prairie wetland sites. Prairie wetland ponds tend to have high particulate loads, due in part to shallow depths, loose sediments, emergent plants, and constant mixing by wind (de la Cruz 1979 as cited in Murkin 1989), and, as a result, the majority of MeHg in the water column may be bound to particles and not in the dissolved phase. For example, in 12 prairie wetlands sampled for dissolved and particulate Hg there was a large range in the proportion of THg and MeHg in the dissolved phase (21–95 and 42–99 % for dissolved THg and MeHg, respectively; Hall, B.D., unpublished data). In wetlands where the majority of MeHg is bound to particles, MeHg may be less bioavailable to some macroinvertebrates, depending on their feeding behaviour.

Greater algal production in lakes can reduce MeHg uptake into food webs through biodilution (Pickhardt et al. 2002). In the current study, lake trophic status, estimated by water chlorophyll *a*, was negatively related to MeHg concentrations in all FFGs (though not significantly for scrapers; Table S3). On the other hand, MeHg in scrapers was better predicted by total nitrogen and phosphorus (TN and TP). These negative relationships support the premise that organisms in less productive systems (and often more acidic systems, e.g. Wyn et al. 2009) accumulate more Hg.

Past studies have also shown that foraging strategies (as indicated by different δ^{13}C) affect MeHg concentrations in both primary and secondary consumers. Both Jardine et al. (2012) and Riva-Murray et al. (2013) found that differences in carbon sources explained some variability in MeHg concentrations in macroinvertebrates. In the latter study, primary consumer FFGs that had lower δ^{13}C had higher MeHg concentrations when compared to organisms with higher δ^{13}C values at the same site. MeHg concentrations in macroinvertebrates that relied more on autochthonous carbon were better predicted by stream water pH than those that fed more on terrestrial inputs (Jardine et al. 2012). In the current study, δ^{13}C was a significant and positive predictor of invertebrate MeHg across FFGs when data from the wetland sites were excluded (multiple linear regressions, Table 2). However, within FFGs this relationship was only significant for shredders based on individual linear regression analyses (Fig S2). The δ^{13}C of littoral versus pelagic food sources in lakes are influenced by the availability and source of inorganic carbon, and algae from more productive lakes are typically higher in δ^{13}C than those from less productive systems (Hecky and Hesslein 1995). The lack of data on potential food sources (e.g. algae, detrital material) for the macroinvertebrates in the current study means it is not possible to attribute the positive relationship between MeHg concentrations and δ^{13}C to specific habitat use. Furthermore, δ^{13}C explained only a small proportion of additional variability in invertebrate MeHg compared to other variables ($R^2 = 0.566$ and 0.573, models 4 and 5, Table 2).

Primary consumers vary in their δ^{15}N across sites because human activities (agriculture, wastewaters) can increase both the quantity and δ^{15}N values of nitrogen supporting aquatic ecosystems. In this review, δ^{15}N values were highly variable within taxa across systems (e.g. −2.4 to 3.7 ‰ in Ephemeroptera and 1.5–9.2 ‰ in Odonata), likely reflecting spatial differences in nitrogen inputs and cycling as well as differences in dietary habits. Interestingly, within each FFG raw δ^{15}N was positively and significantly related to the pH of the systems ($R^2 = 0.33$, $p < 0.001$; Fig. S3a), but this relationship did not remain after baseline adjustment of δ^{15}N ($p = 0.43$; Fig. S3b). The lower δ^{15}N in macroinvertebrates from lower pH systems suggests that acidity affects the cycling of inorganic nitrogen isotopes, but the mechanisms behind this are not known. Past laboratory studies found that the δ^{15}N of predatory macroinvertebrates (terrestrial and aquatic) is ~0.5–4.6 ‰ higher than that of their prey (McCutchan et al. 2003). From our field data, the median absolute difference in δ^{15}N between predators and all non-predatory taxa was 2.12 ‰, and was similar between predators and gatherer-collectors or scrapers and slightly higher for comparisons using shredders at 2.01, 2.05 and 2.97 ‰, respectively (Fig. 5). These data suggest that the trophic enrichment in δ^{15}N to aquatic macroinvertebrates is lower than the 3.4 ‰ value often applied to aquatic systems, and more comparable to the 2.0 ‰ value reported for riverine invertebrates (Bunn et al. 2013).

Multiple regression models were used to understand the combination of variables that predict MeHg concentrations across all macroinvertebrate taxa. Before including site characteristics, $\delta^{15}N_{adj}$ explained 20 % of the variation in

Table 2 Results of stepwise multiple regression modeling ($\alpha = 0.10$ to enter, 0.15 to remove) of individual log-MeHg concentrations in macroinvertebrates with $\delta^{15}N_{adj}$, site pH, surface area, disturbance, latitude, longitude, δ^{13}C and FFG, with the most likely models indicated in bold-type[a]

Regression equation	R^2_{adj}	AIC_c[b]
Lakes only ($n = 239$ samples)		
0.559*FFG	0.310	−448.388
0.549*FFG − 0.412*pH	0.478	−514.495
0.536*FFG − 0.289*pH − 0.281*Lat	0.541	−543.850
0.423*FFG − 0.309*pH − 0.225*Lat + 0.208*$\delta^{15}N_{adj}$	0.566	−556.587
0.417*FFG − 0.332*pH − 0.215*Lat + 0.213*$\delta^{15}N_{adj}$ + 0.092*δ^{13}C	**0.573**	**−559.065**
Lakes and wetlands ($n = 306$ samples)		
0.455*$\delta^{15}N_{adj}$	0.204	−436.061
0.419*$\delta^{15}N_{adj}$ − 0.290*pH	0.285	−467.852
0.246*$\delta^{15}N_{adj}$ − 0.379*pH + 0.383*FFG	**0.397**	**−518.759**

[a] Disturbance, longitude and surface area did not meet criteria for entry ($\alpha = 0.10$) in lakes-only modeling; additionally, δ^{13}C and latitude did not meet criteria for entry in lakes and wetlands modeling

[b] $p < 0.001$ for all models; model likelihood determined from an AIC_c adjusted for small sample size (see "Methods" section for details)

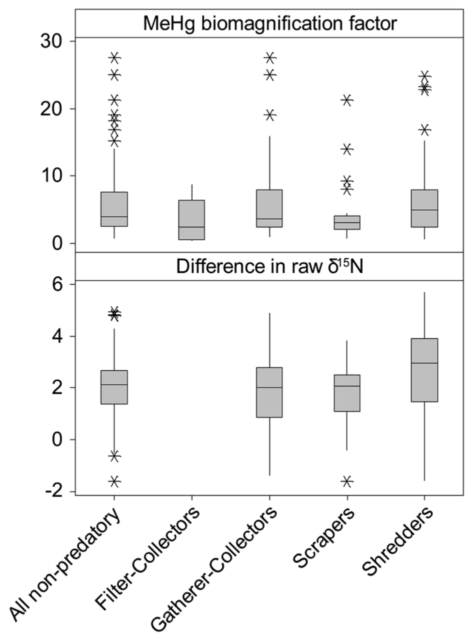


Fig. 5 Boxplots of MeHg BMF (*top panel*) and differences in raw δ^{15}N (‰; *bottom panel*) between predatory and non-predatory FFGs of freshwater invertebrates (mean δ^{15}N predator—mean δ^{15}N non-predatory FFG) from 78 lakes and wetlands across Canada. Two outliers from Saskatchewan were removed from the MeHg BMF plot (gatherer-collectors and all non-predatory groups, with BMFs of 161 and 190, respectively)

macroinvertebrate MeHg concentrations when lakes and prairie wetlands were treated together (model 6, Table 2). However, the best regression model overall (based on AIC$_c$) included FFG, water pH, latitude, $\delta^{15}N_{adj}$, and δ^{13}C as independent variables (model 5, $R^2_{adj} = 0.573$; Table 2). Other environmental characteristics, specifically surface area, longitude, and disturbance category, were not significant variables ($p > 0.10$). The importance of both $\delta^{15}N_{adj}$ and FFG in the model suggests that differences in MeHg bioaccumulation among FFGs were related to invertebrate feeding ecology in addition to trophic position. Scrapers, shredders, gatherer-collectors and filter-collectors occupy various micro-habitats in aquatic ecosystems and may consume different basal resources (i.e. algae, bacteria, and detritus) that originate from distinct sources (benthic substrates, water column, terrestrial watershed). These ecological differences among FFGs may also help to explain why different chemical and physical variables predicted MeHg concentrations in the different groups, as discussed above. The significant effect of FFG in the model is consistent with other studies that have shown that foraging strategies and habitat influence MeHg bioaccumulation in aquatic primary consumers (Chételat et al. 2011; Riva-Murray et al. 2011).

Past studies have shown that MeHg concentrations in aquatic macroinvertebrates can also vary with watershed characteristics. For example, MeHg concentrations in taxa from systems with higher proportions of wetland in their catchments have higher MeHg concentrations than those with less wetland area (Allen et al. 2005; Chasar et al. 2009). Allen et al. (2005) also found that invertebrates from lakes with greater percent conifer coverage had higher MeHg concentrations. Concentrations of MeHg in invertebrates from prairie systems were higher in ponds surrounded by perennial grasses compared to annual cultivated crops (Bates and Hall 2012). In the current study, our analysis of watershed characteristics was limited to a disturbance index assigned to each lake (see "Methods" section). Its insignificance in the regression models is likely due to the coarse nature of the index (presence/absence) because it did not reflect the degree of disturbance in these watersheds.

MeHg biomagnification

In the current study, median BMFs of MeHg were 3.92, 2.42, 3.57, 3.00, and 4.87 for comparisons of predatory invertebrates with all non-predatory invertebrates, filter-collectors, gatherer-collectors, scrapers or shredders, respectively (Fig. 5). These BMFs overlapped with those for streams in New Brunswick, Canada (mean of 2.5 for BMFs of predators to primary consumers; Jardine et al. 2013), a floodplain lake in Bolivia (1.7–6 for predatory macroinvertebrates to primary consumers; Molina et al. 2010) and streams in Maryland, U.S. (predatory invertebrates to potential food sources; BMFs of 2–5; Mason et al. 2000). When multiple regression analyses were done in the current study, BMFs using all non-predatory invertebrates as prey were best predicted by a combination of latitude, longitude and pH, with higher BMFs in systems at greater latitudes and longitudes and with lower pH ($R^2 = 0.50$; Table 3). Similar models were found for BMFs calculated using collector-gatherers ($R^2 = 0.62$, latitude, longitude) and shredders ($R^2 = 0.23$, latitude, longitude, pH), although less variability for BMFs was explained for the latter FFG. For analyses using scrapers, site depth and surface area best explained the variability in BMFs ($R^2 = 0.55$). Although higher bioaccumulation or bioconcentration factors (BAFs, BCFs; ratios of Hg concentrations in water and organisms) have been associated with ecosystem characteristics such as OC (e.g. Watras et al. 1998; Rolfhus et al. 2011), few studies have examined the variables affecting BMFs from primary to secondary consumers; those that have, have shown no relationship with pH (Jardine et al. 2013).

Although the slope of the relationship between Hg concentrations and $\delta^{15}N_{adj}$ in organisms is often used as a measure of Hg trophic transfer (or Hg biomagnification rate) within individual food webs (Jardine et al. 2006), small sample sizes of invertebrates and ranges of trophic levels within sites prevented the comparison of these

Table 3 Results of stepwise multiple regression modeling ($\alpha = 0.10$ to enter, 0.15 to remove) of MeHg BMFs for predatory to non-predatory FFGs of macroinvertebrates with site pH, mean depth, surface area (SA), disturbance (Dist), latitude (Lat) and longitude (Long) with the most likely models indicated in bold-type

Model	Expression	R^2_{adj}	p	n	AIC_c
All non-predatory					
1	0.390*Lat	0.135	0.004	53	−112.454
2	1.815*Lat + 1.545*Long	0.485	0.000	53	−138.724
3	**1.890*Lat + 1.559*Long − 0.187*pH**	**0.508**	**0.000**	**53**	**−139.829**
Gatherer-collectors					
1	0.393*Lat	0.135	0.006	47	−110.367
2	**1.982*Lat + 1.735*Long**	**0.622**	**0.000**	**47**	**−148.009**
Scrapers					
1	−0.605*Depth	0.326	0.008	18	−68.869
2	**−0.976*Depth + 0.612*SA**	**0.550**	**0.001**	**18**	**−74.385**
Shredders					
1	0.299*Lat	0.069	0.041	47	−97.545
2	1.334*Lat + 1.080*Long	0.147	0.011	47	−100.412
3	**1.528*Lat + 1.136*Long − 0.351*pH**	**0.237**	**0.002**	**47**	**−104.374**

Variables that are not listed in the models did not meet criteria for entry ($\alpha = 0.10$)

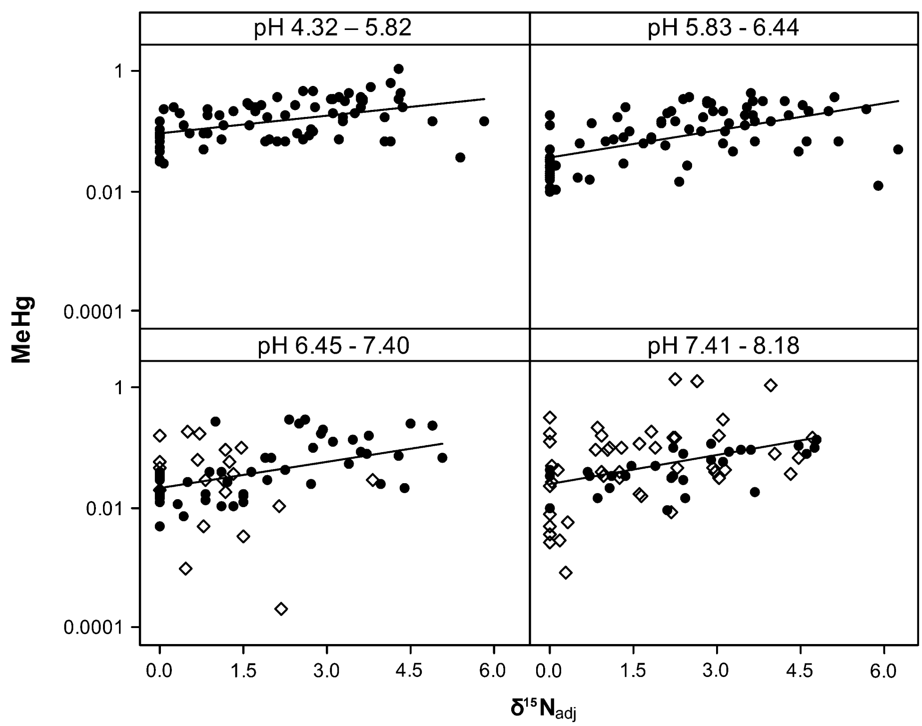


Fig. 6 Linear regressions of log MeHg concentrations (μg g^{-1} dry weight) versus $\delta^{15}N_{adj}$ (‰) in invertebrates from sites (lakes represented by *filled circle*, wetlands represented by *open diamond*) across Canada in order of increasing pH. **a** pH 4.32–5.82, $R^2 = 0.278$, $p < 0.001$, slope = 0.115, $n = 75$, **b** pH 5.83–6.44, $R^2 = 0.412$, $p < 0.001$, slope = 0.171, $n = 78$, **c** pH 6.45–7.40, $R^2 = 0.204$, $p < 0.001$, slope = 0.145, $n = 72$, **d** pH 7.41–8.18, $R^2 = 0.170$, $p < 0.001$, slope = 0.166, $n = 75$

relationships herein. However, when sites were binned by pH, the slopes of the relationships between MeHg and $\delta^{15}N_{adj}$ were significant and ranged from 0.115 to 0.166 (Fig. 6), similar to estimates of Hg biomagnification across a range of other ecosystem types (Kidd et al. 2012a). The slopes in the current study did not differ significantly for different pH ranges (ANCOVA $p = 0.507$). Even though the range in $\delta^{15}N_{adj}$ was not large (typically 4.5 to 6 ‰), MeHg concentrations in all macroinvertebrates were positively related to their $\delta^{15}N_{adj}$, although the variability explained by relative trophic position was lower (R^2 of 0.170 to 0.412, Fig. 6) than is often seen when these relationships are examined through individual food webs and when higher trophic-level organisms such as fish are included (e.g. Clayden et al. 2013).

Conclusions

Aquatic invertebrates are key sources of MeHg to fish (Kidd et al. 2012a) and terrestrial biota (Sarica et al. 2004). As an example of the latter, songbirds feeding on spiders that consume aquatic emerging insects had high concentrations of MeHg, suggesting that birds with links to aquatic systems may be at greater risk for MeHg toxicity (Rimmer et al. 2005). Although the importance of invertebrates in the trophic transfer of MeHg is recognized, studies examining factors that control or predict MeHg in invertebrates across broader scales are few.

Our work identified several variables that predict MeHg concentrations in invertebrates from a large range of Canadian aquatic systems. RDA indicated that pH and geographical location (site-mean MeHg in primary and secondary consumers increased from west to east and from north to south) were the most important predictors of MeHg concentrations in both predatory and non-predatory invertebrates, whereas multiple linear regression analysis indicated that the combination of pH, geography, FFGs and trophic position were important. Thus, data from the current study show that both MeHg supply to food webs (as indicated by environmental predictors such as pH) and bioaccumulation processes (as indicated by differences in MeHg in FFGs and in BMFs) influence the concentration of MeHg in aquatic invertebrates. Whereas previous regional-scale studies have shown that MeHg in these organisms depends on a number of these individual factors, the broad geographic scope and range of ecosystem characteristics data in this study reinforce that environmental (e.g. physical and chemical variables), trophic, ecological (e.g. foraging behaviour) and geographic factors are all important to consider when examining MeHg accumulation in macroinvertebrates. Together, these variables control the trophic transfer of MeHg to higher level consumers (e.g. fish, birds) in aquatic and terrestrial ecosystems.

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Conflict of interest The authors declare that they have no conflict of interest.

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