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Review

The importance of bioconcentration into the pelagic food web base for methylmercury biomagnification: A meta-analysis

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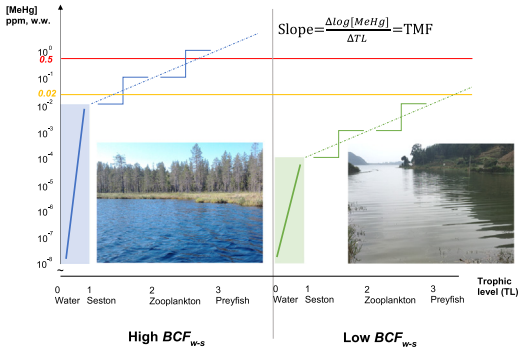
HIGHLIGHTS

- Extensive MeHg bioaccumulation literature review and meta-analysis
- MeHg bioconcentration factors at food web base predict preyfish MeHg.
- DOC and trophic status influence MeHg bioconcentration factors.
- MeHg concentration in water column does not predict MeHg in biota.
- Conceptualization of a MeHg BCF elevator leading to a BMF staircase

GRAPHICAL ABSTRACT



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abstract

Methylmercury (MeHg) transfer from water into the base of the food web (bioconcentration) and subsequent biomagnification in the aquatic food web leads to most of the MeHg in fish. But how important is bioconcentration compared to biomagnification in predicting MeHg in fish? To answer this question we reviewed articles in which MeHg concentrations in water, plankton (seston and/or zooplankton), as well as fish (planktivorous and small omnivorous fish) were reported. This yielded 32 journal articles with data from 59 aquatic ecosystems at 22 sites around the world. Although there are many case studies of particular aquatic habitats and specific geographic areas that have examined MeHg bioconcentration and biomagnification, we performed a meta-analysis of such studies. Aqueous MeHg was not a significant predictor of MeHg in fish, but MeHg in seston i.e., the base of the aquatic food web, predicted 63% of the variability in fish MeHg. The MeHg bioconcentration factors (i.e., transfer of MeHg from water to seston; BCF_{w-s}) varied from 3 to 7 orders of magnitude across sites and correlated significantly with MeHg in fish. The MeHg biomagnification factors from zooplankton to fish varied much less ($logBMF_{z-f}$, 0.75 ± 0.31), and did not significantly correlate with fish MeHg, suggesting that zooplanktivory is not as important as bioconcentration in the biomagnification of fish MeHg across the range of ecosystems represented in our meta-analysis. Partial least square (PLS) and linear regression analyses identified several environmental factors associated with increased BCF, including low dissolved organic carbon, low pH, and oligotrophy. Our study reveals the widespread importance of MeHg bioconcentration into the base of the aquatic food web for MeHg at higher trophic levels in aquatic food webs, as well as the major influences on the variability in this bioconcentration.

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Contents

1. Introduction

Methylmercury (MeHg) is known for its high degree of bioaccumulation in aquatic food webs, with MeHg concentrations in predatory fish that can be six orders of magnitude higher than in surface water (Lindqvist et al., 1991; McMeans et al., 2015). The stepwise biomagnification of MeHg from one trophic level to the next results in MeHg concentrations ([MeHg]) in aquatic biota that can pose health risks to humans as well as wildlife (Clarkson et al., 2003; Sheehan et al., 2014). Reducing environmental exposure of humans and wildlife to MeHg could save the European Union (EU) up to €8–9 billion per year (Bellanger et al., 2013). To protect human health, the World Health Organization (WHO) has set an environmental quality standard (EQS) in fish of 0.5 μg MeHg g^{-1} wet weight (w.w.) (FAO/WHO, 2015). In the European Union, the EQS for total mercury (Hg) in fish (0.02 μg Hg g^{-1} w.w.) is >20 times lower compared to the WHO EQS since the EU seeks to better protect secondary consumers in aquatic ecosystems (i.e. piscivorous birds and mammals) (EC, 2013; Vignati et al., 2013). The WHO EQS is already exceeded in >50% of Swedish lakes (Åkerblom et al., 2014) and is also often exceeded in piscivorous fish species in Northern America (Gandhi et al., 2014). Regionally, the more stringent EU EQS is often exceeded, especially in boreal freshwaters, including almost all those in Fennoscandia (Nguetseng et al., 2015).

Globally, there are large regional differences between Hg concentrations ([Hg]) in both water and fish across regions (Selin et al., 2007; Selin, 2009). Even within regions there is a considerable variability in fish [Hg], which generally does not relate well to [Hg] or MeHg concentrations [MeHg] in water (Rolfhus et al., 2011; Liu et al., 2012). An indicator to better understand the transfer of Hg from water into biota, and the subsequent biomagnification in fish, could improve the environmental policies to reduce fish [Hg]. Chen et al. (2014) have already shown the significance of using water column particulate MeHg to predict fish MeHg through an investigation of 10 estuary sites in Northeastern USA. This would be valuable since the environmental response to policies such as Hg emission reductions can be slow, often on a timescale of decades to centuries (KumamotoPrefecture, 1998; Meili et al., 2003; Kindaichi and Matsuyama, 2005; Danielsson et al., 2011), and fraught with uncertainties.

While Hg in fish poses a threat to consumers regardless of its chemical speciation, MeHg is the Hg species that bioaccumulates most efficiently (Clarkson et al., 2003). Many studies that analyzed [MeHg] in aquatic biota with a focus on MeHg bioaccumulation in piscivorous wildlife and its relation to Hg exposure have shown that [MeHg] increases steadily with each trophic level (Mason et al., 1995; Munthe et al., 2007; Clayden et al., 2013). A comprehensive review by Lavoie et al. (2013) on [Hg] and [MeHg] along the aquatic food chains across the globe has put much of the existing data in perspective. They found that biomagnification patterns above the base of the food web were relatively similar with some consistent differences between regions. Lavoie's review, however, used longer-lived benthic mussels/snails as the trophic baseline. Thus, the review did not address the entry of Hg into the base of the aquatic food web, i.e., the step from aqueous [MeHg] to organisms at the lowest trophic level. Using seston (i.e., suspended particles, mostly composed of small algae and bacteria) as the base of the pelagic food web, a study of the western Great Lakes region found that dissolved organic matter (DOM) limited MeHg uptake from water by seston, zooplankton, and fish, yielding lower bioconcentration factors (BCF, defined as the ratio between the MeHg concentrations in biota and water), while the trophic magnification factor (TMF) from the base of the food web upwards remained stable across different food webs (Rolfhus et al., 2011). This finding is an indication of the complexity of Hg bioconcentration from water into the biota due to the influence of different environmental factors, but also the importance of the variability of bioconcentration into the base of the pelagic food web for the biomagnification to higher trophic levels.

Correlations between catchment characteristics and fish [Hg] have also been utilized to examine the influence of environmental factors on [Hg] in aquatic biota. Kidd et al. (2011) have argued that physical and chemical characteristics, e.g. DOC and pH, reflect Hg bioavailability in the environment that may be a critical influence on Hg biomagnification. For example, low pH surface water (pH < 6) with higher [Hg] is associated with higher [Hg] in generally oligotrophic freshwater ecosystems (Le Faucheur et al., 2014). On the other hand, eutrophic aquatic ecosystems are associated with increased plankton biomass and consequently lower [Hg] in the aquatic biota (per unit biomass), which is described as "biodilution" (Pickhardt et al., 2002; Karimi et al., 2007). Some very eutrophic aquatic ecosystems in China and Eastern Europe have relatively high aqueous [MeHg] (>1 ng L^{-1}), but rather little [MeHg] in biota (<0.2 μg g^{-1} w.w.) (Farkas et al., 2000; Nguyen et al., 2005; Suchanek et al., 2008; Liu et al., 2012). This stands in contrast to higher [MeHg] in the biota of oligotrophic boreal ecosystems (>0.5 μg g^{-1} w.w.) despite aqueous [MeHg] that are often <1 ng L^{-1} or even <0.1 ng L^{-1} (Meili, 1991; Watras and Bloom, 1992; Clayden et al., 2013). Some of these differences may derive from varying MeHg photodemethylation (PD) activity in freshwater lakes as a function of color (Lehnherr and St. Louis, 2009). Nevertheless, the magnitude of the discrepancy between aqueous [MeHg] and fish [MeHg] across regional and global scales, but also within regions, points to the need for a better understanding of what controls the transfer of MeHg from water into the base of pelagic food webs. We are specifically interested in how the basal MeHg transfer influences the overall distribution of MeHg in aquatic ecosystems (i.e., lakes, reservoirs, and estuaries).

Mason et al. (2012) and Le Faucheur et al. (2014) have summarized experimental approaches to studying MeHg bioconcentration from water into biota. They found that the transfer of bioavailable MeHg fractions from water into seston is a critical step for subsequent MeHg bioaccumulation in aquatic food webs, and that MeHg accumulates more in seston than inorganic Hg. These experimental studies, together with Driscoll et al. (2007), also showed that the variation in trophic transfer is lower than the variation in BCF from water to planktivorous or small omnivorous fish. Mackay et al. (2013) stated from a mathematical point of view that BCF is critical to chemical concentrations in food webs and can be determined in laboratory tests. This also suggests that the large degree of variation in the [MeHg] of aquatic biota may stem from variation in the degree of bioconcentration into the base of the food web. To test this hypothesis against field observations, it is necessary to distinguish the movement of MeHg from water into the base of the food webs (bioconcentration) from the trophic transfer further up the food web (biomagnification). To date, however, there is no literature meta-analysis on MeHg bioconcentration from water through the base of the pelagic food web into fish which could complement the review by Lavoie et al. (2013) on MeHg bioaccumulation further up in food webs.

This brief overview indicates the need for a better understanding of the factors which mediates the transfer of bioavailable MeHg from water into the base of the aquatic food web, and eventually fish. For that purpose, we conducted a literature meta-analysis on MeHg in aquatic ecosystems where a comparison was made between MeHg bioconcentration and the subsequent MeHg biomagnification to fish. We compared the increase of [MeHg] from water into the base of the food web, operationally defined as seston, with the increase from the base of the food web up into planktivorous fish. We assessed the magnitude of these two transfer steps using metrics of bioconcentration from water to seston (bioconcentration factors, $BCF_{w\text{-}s}$) and biomagnification from seston into zooplankton, then to planktivorous fish (biomagnification factors, $BMF_{s\text{-}z}$ and $BMF_{z\text{-}f}$). Finally, we sought to identify environmental factors related to the variability in MeHg bioconcentration and biomagnification.

2. Materials and methods

2.1. Published data and site characteristics

The literature selection identified publications in Web of Science™ and PubMed (pubmed.gov) that contained MeHg concentrations ([MeHg]) in water, plankton (seston and/or zooplankton), as well as planktivorous fish from pelagic food webs in lakes, reservoirs, and estuaries, since 1975. In total, 8511 studies were screened by searching for matches with "mercury/Hg" and "accumulation" in all fields (includes Title/Keywords/Abstracts). From these, 217 studies were checked through the full-text to identify their study methods and results. The detailed selection process is presented in the Supporting Information (SI), Table S1. Eventually, 32 papers from the 1970s to 2015 ended up matching the selection criteria. These papers reported data from 59 aquatic ecosystems at a total of 22 sites (SI Fig. S1 and Table S2). The spatial expanse of the studies ranged from the tropics in the southern hemisphere to the boreal region, with most of them coming from temperate latitudes (SI Table S3).

The [MeHg] in water, seston and/or zooplankton, and fish (measured in muscle tissues from low-trophic-level fishes that are mostly planktivorous) were then extracted from the publications, either the main text or the supplementary information. Data were digitalized using WebPlotDigitizer (Rohatgi, 2016), if only figures were available for data extraction. The seston <50 μm, consisting primarily of algae and bacteria, is the size fraction most susceptible to grazing. However, the plankton size fraction <50 μm may not be the preferred food for planktivorous fish due to size-selective feeding habits, by which the largest prey size is preferably ingested, i.e., >50 μm (Brooks and Dodson, 1965). In the compilation of different literature sources selected by the meta-analysis, seston was operationally defined, based on what is reported in each publication, as either the smallest particulates filtered out by glass microfiber filters, the known primary producers within aquatic food webs with a size range from 0.45 to 200 μm, or the smallest plankton size fraction reported. The criterion used for defining seston from each specific publication is noted in SI Table S2. Bioconcentration factors between [MeHg] in water and seston were calculated. The [MeHg] in zooplankton and fish were used to calculate biomagnification factors, since this MeHg represents not only direct dietary food uptake from seston but also that derived from both facilitated transport and passive transfer through cell membranes (Mason et al., 1996). It was not always possible to differentiate between herbivorous and predatory zooplankton, thus it is possible that some zooplankton also ingested other consumers. The [MeHg] in planktivorous and small omnivorous fish (i.e., preyfish) were extracted to define a consistent trophic level for "fish" across the sites. Although we focused on lower trophic-level pelagic fish, benthivorous and occasionally piscivorous feeding could not be entirely ruled out. It was assumed that reported [MeHg] in aquatic food web compartments were at an equilibrium state.

Information and measurements on catchment characteristics, including trophic status, acidity, and biome were also extracted from the site descriptions in the published papers (SI 3, Tables S2 and S3).

2.2. Data handling and statistical analysis

The BCF is defined as the enrichment of MeHg from water into biota (Gobas and Morrison, 2000). The enrichment of MeHg from water to the base of the food web as defined by seston was designated as $BCF_{w\text{-}s}$. This was calculated as the ratio between [MeHg] in seston ($[MeHg]_{seston}$, ng MeHg g^{-1} dry weight (d.w.)) and [MeHg] in water ($[MeHg]_{water}$, ng MeHg mL^{-1}) (Eq. (1)).

$$BCF_{w-s} = \frac{[MeHg]_{seston}}{[MeHg]_{water}} \quad (mL\ g^{-1}) \tag{1}$$

Methylmercury dry weight concentrations in zooplankton and preyfish were also used to calculate BCF, even though we recognize that [MeHg] in these biota include food uptake from seston as well as uptake directly from water (Mason et al., 1996). The BCF from water to zooplankton and preyfish were termed as $BCF_{w\text{-}z}$ and $BCF_{w\text{-}f}$, respectively (Eqs. (2) and (3)).

$$BCF_{w-z} = \frac{[MeHg]_{zooplankton}}{[MeHg]_{water}} \quad (mL\ g^{-1}) \tag{2}$$

$$BCF_{w-f} = \frac{[MeHg]_{fish}}{[MeHg]_{water}} \quad (mL\ g^{-1}) \tag{3}$$

The accumulation of MeHg between adjacent trophic levels was defined as a dimensionless biomagnification factor (BMF) (Gobas and Morrison, 2000), representing trophic transfer of MeHg. $BMF_{s\text{-}z}$ stands for the bioaccumulation from primary producers (seston) to pelagic primary consumers (zooplankton) (Eq. (4)). Similarly, the BMF from the primary consumers (zooplankton) to pelagic secondary consumers (preyfish), termed $BMF_{z\text{-}f}$, was calculated using preyfish MeHg ($[MeHg]_{fish}$, ng MeHg g^{-1} d.w.) versus zooplankton MeHg ($[MeHg]_{zooplankton}$, ng MeHg g^{-1} d.w.) (Eq. (5)):

$$BMF_{s-z} = \frac{[MeHg]_{zooplankton}}{[MeHg]_{seston}} = \frac{BCF_{w-z}}{BCF_{w-s}} \tag{4}$$

$$BMF_{z-f} = \frac{[MeHg]_{fish}}{[MeHg]_{zooplankton}} = \frac{BCF_{w-f}}{BCF_{w-z}} \tag{5}$$

BCF and BMF calculations for each aquatic ecosystem were made using measured [MeHg] from the publications (median values if more than one measurement was made in an aquatic ecosystem).

The statistical analyses of the data were performed using the software JMP 10 (© SAS Institute Inc.) and SIMCA 14 (MKS Umetrics). The environmental factors (i.e., pH and dissolved organic carbon (DOC)) and log-transformed [MeHg] from the reviewed aquatic ecosystems all passed the Shapiro-Wilk Goodness-of-Fit test for data normality (Shapiro and Wilk, 1965). Regression analyses were used to test how well aqueous MeHg predicted MeHg in aquatic biota. Linear regressions were used to examine how the $BCF_{w\text{-}s}$ and $BCF_{w\text{-}z}$ related to fish MeHg. The level of significance for all tests was set at $p < 0.05$. We also used Partial Least Square (PLS) analysis to assess the relative importance of different environmental factors in explaining the variation of MeHg transfer in biota. Unlike multiple linear regression, all testing in PLS is made simultaneously, hence no correction is needed. The details of PLS models and results are listed in the Supplementary Information (SI).

3. Results

3.1. Study sites and aquatic ecosystems

Freshwater studies dominated the selected publications on MeHg in aquatic ecosystems. The most famous and the earliest one however, was conducted in an estuary site at Minamata Bay (SI Table S2). Among all 32 papers investigated in this meta-analysis, 28 reported [MeHg] at 21 sites in the northern hemisphere with only 3 studies focused on a tropical freshwater lake in Papua New Guinea (Site 9). Freshwater sites located in Asia and southern Europe were characterized by strong anthropogenic influences, eutrophication and neutral to alkaline water (Site 11, 17, 18, 19, and 21, SI Table S3). This contrasts with often more pristine and oligotrophic conditions, as well as acidic to circumneutral waters in Scandinavian and North American boreal sites (Site 2, 3, 5, 14, 16, and 20, SI Table S3). We have not treated the Great Lakes region as one site, given the geographical and ecological differences across the Great Lakes. We have also not taken riverine sites into the final list, although they were considered in the literature screening. One reason for this is that fish in rivers/streams can get parts of their diet, as well as MeHg, from terrestrial prey, including terrestrial insects, but also spiders and even rodents (Di Prinzio et al., 2015; Brett et al., 2017). Another concern is the mobility of lotic fish which means that they can get MeHg from other sources and waters besides those where the fish are caught. Thus, the dietary trajectories of MeHg in lotic fish are not restricted to the sampling locality. No studies from the Brazilian Amazon matched our publication selection standards so far. For instance, Roulet et al. (2000) investigated $[MeHg]_{water}$, $[MeHg]_{seston}$, and $[MeHg]_{zooplankton}$ in an Amazonian river, but no $[MeHg]_{fish}$ was reported. In parallel investigations on aqueous and zooplankton [MeHg], no MeHg data on fish from low trophic levels were found (Kehrig et al., 2009; Kasper et al., 2014).

The $[MeHg]_{water}$ in all aquatic ecosystems ranged from 0.02 to 1.94 ng MeHg L^{-1}. Out of the 59 ecosystems analyzed, 39 identified both seston and zooplankton. The $[MeHg]_{seston}$ varied from 1.7 to 410 ng MeHg g^{-1} d.w, while the $[MeHg]_{zooplankton}$ were much higher and varied from 2.7 to 2600 ng MeHg g^{-1} d.w. The highest [MeHg] in seston and zooplankton were both recorded from site 21 in the Babeni Reservoir, Romania, sampled with 65 μm and 200 μm plankton nets, respectively (Bravo et al., 2014). The [MeHg] in preyfish ranged from 24.1 to 3400 ng MeHg g^{-1} d.w. (SI Table S2). Most of the aquatic ecosystems had $[MeHg]_{fish}$ that were below the WHO EQS (0.5 μg MeHg g^{-1} w.w. equivalent to 2500 ng g^{-1} d.w. assuming 80% water content in fish, for planktivorous or small omnivorous fishes) (USEPA, 2012). However, in some pristine boreal lakes (Site 2) and contaminated sites (Sites 21 and 22), $[MeHg]_{fish}$ were well above this limit (Lindqvist et al., 1991; Bravo et al., 2010; Dominik et al., 2014).

3.2. Relationships between MeHg in aqueous and particulate sources

We found no correlation between $\log[MeHg]_{water}$ and $\log[MeHg]_{fish}$ ($R^2 < 0.01$, $p > 0.9$). Additionally, $\log[MeHg]_{water}$ could not predict $\log[MeHg]_{seston}$ ($R^2 = 0.01$, $p > 0.5$) or $\log[MeHg]_{zooplankton}$ ($R^2 < 0.01$, $p > 0.5$), unless one high $[MeHg]_{water}$ (site 16, $[MeHg]_{water} > 1$ ng L^{-1}) was removed from the regression. Both $\log[MeHg]_{seston}$ ($R^2 = 0.63$, $p < 0.0001$) and $\log[MeHg]_{zooplankton}$ ($R^2 = 0.72$, $p < 0.0001$) strongly correlated with $\log[MeHg]_{fish}$, but $\log[MeHg]_{zooplankton}$ was a better predictor (Fig. 1). This is consistent with previously published works reporting the importance of plankton as a major food and MeHg exposure source to planktivorous fish regardless of site differences (Hirota et al., 1979; Kasper et al., 2014; Lehnherr, 2014).

3.3. MeHg transfer from water to biota and within trophic levels

The mean bioconcentration of MeHg from water to seston ($\log BCF_{w\text{-}s}$) (5.32 ± 0.73, Mean ± S.D.), or zooplankton ($\log BCF_{w\text{-}z}$) (5.82 ± 0.69), were very similar (*t*-test two-sample unequal variance, $p < 0.003$) and in both cases over six orders of magnitude greater than the biomagnification of MeHg from zooplankton to preyfish, $\log BMF_{z\text{-}f}$ (0.75 ± 0.31) (Fig. 2). The $\log BCF_{w\text{-}f}$ (6.60 ± 0.69) and the range of BCF values were close to what have been reported from the experimental work by Mason et al. (1995) as well as in a more recent field investigation of the Western Great Lakes Region (Rolfhus et al., 2011). The degree of MeHg bioconcentration into the base of the pelagic food web, as represented by $BCF_{w\text{-}s}$, is thus much greater, but also more variable between sites, when compared with $BMF_{z\text{-}f}$, the transfer of MeHg from plankton to preyfish at a low trophic position in the food web. The two MeHg trophic transfer values ($\log BMF_{s\text{-}z}$ and $\log BMF_{z\text{-}f}$) are also similar (*t*-test two-sample unequal variance, $p < 0.0001$) (Fig. 2).

The $BCF_{w\text{-}s}$ was not correlated to $BMF_{z\text{-}f}$ (pairwise: $r = -0.08$, $p = 0.68$). The $BCF_{w\text{-}s}$ was, however, a significant predictor of $[MeHg]_{fish}$ ($R^2 = 0.27$, $p = 0.002$), as well as $BCF_{w\text{-}z}$ ($R^2 = 0.82$, $p < 0.001$) and $BCF_{w\text{-}f}$ ($R^2 = 0.78$, $p < 0.001$) (Fig. 3). The significance of the correlation remains regardless of whether values from heavily contaminated sites, e.g., site 1, 21, and 22, are excluded or not. Trophic transfer of MeHg, either from seston to zooplankton ($BMF_{s\text{-}z}$), or from zooplankton to fish ($BMF_{z\text{-}f}$), could not predict $[MeHg]_{fish}$ ($R^2 < 0.03$, $p > 0.2$).

3.4. Effect of environmental factors on MeHg transfer

We looked at how predictor variables can influence MeHg transfer from water to biota in a PLS model that predicts BCF ($\log BCF_{w\text{-}s}$, $\log BCF_{w\text{-}z}$, $\log BCF_{w\text{-}f}$) (SI Fig. S2). With two predictive components, the model explained ($R^2Y = 62.6\%$) and predicted ($Q^2 = 42.7\%$) much of the variability (SI Table S4). The correlations of the environmental factors (DOC, pH, biome region and trophic status) with BCFs are stronger in freshwater ecosystems than estuaries, likely due to the smaller number of estuary studies included (SI Fig. S2a). VIP scores of highly influential variables (VIP scores > 1.0) are listed in Table 1.

Data were grouped by biome regions in both PLS and linear regression models (Fig. 4, SI Fig. S2b). DOC had the highest predictive power in the PLS, and DOC also stood out in the linear statistical models as a very important factor for predicting BCF (Fig. 4), even though trophic status, pH and/or latitude were also important predictors. DOC best predicted $\log BCF_{w\text{-}s}$ ($R^2 = 0.45$, $p < 0.001$), followed by $\log BCF_{w\text{-}f}$ ($R^2 = 0.41$, $p < 0.001$) and $\log BCF_{w\text{-}z}$ ($R^2 = 0.31$, $p < 0.001$) (Fig. 4).

As for DOC, the pH is also negatively correlated with MeHg $BCF_{w\text{-}s}$. Accordingly, an increase in pH results in lower MeHg BCF ($\log BCF_{w\text{-}s}$: $R^2 = 0.31$, $p < 0.002$). By contrast, MeHg BMF was not significantly affected by pH (pH and $\log BMF_{s\text{-}z}$: $R^2 = 0.05$, $p = 0.23$; $\log BMF_{z\text{-}f}$: $R^2 = 0.01$, $p = 0.5$), biome region/latitude ($\log BMF_{z\text{-}f}$ in boreal versus temperate region ANOVA: $F(1,47) = 0.21$, $p = 0.65$), or trophic status (classified into oligotrophic and eutrophic ecosystems) ($\log BMF_{s\text{-}z}$ ANOVA: $F(1,25) = 1.35$, $p = 0.26$; $\log BMF_{z\text{-}f}$ ANOVA: $F(1,44) = 0.44$, $p = 0.51$).

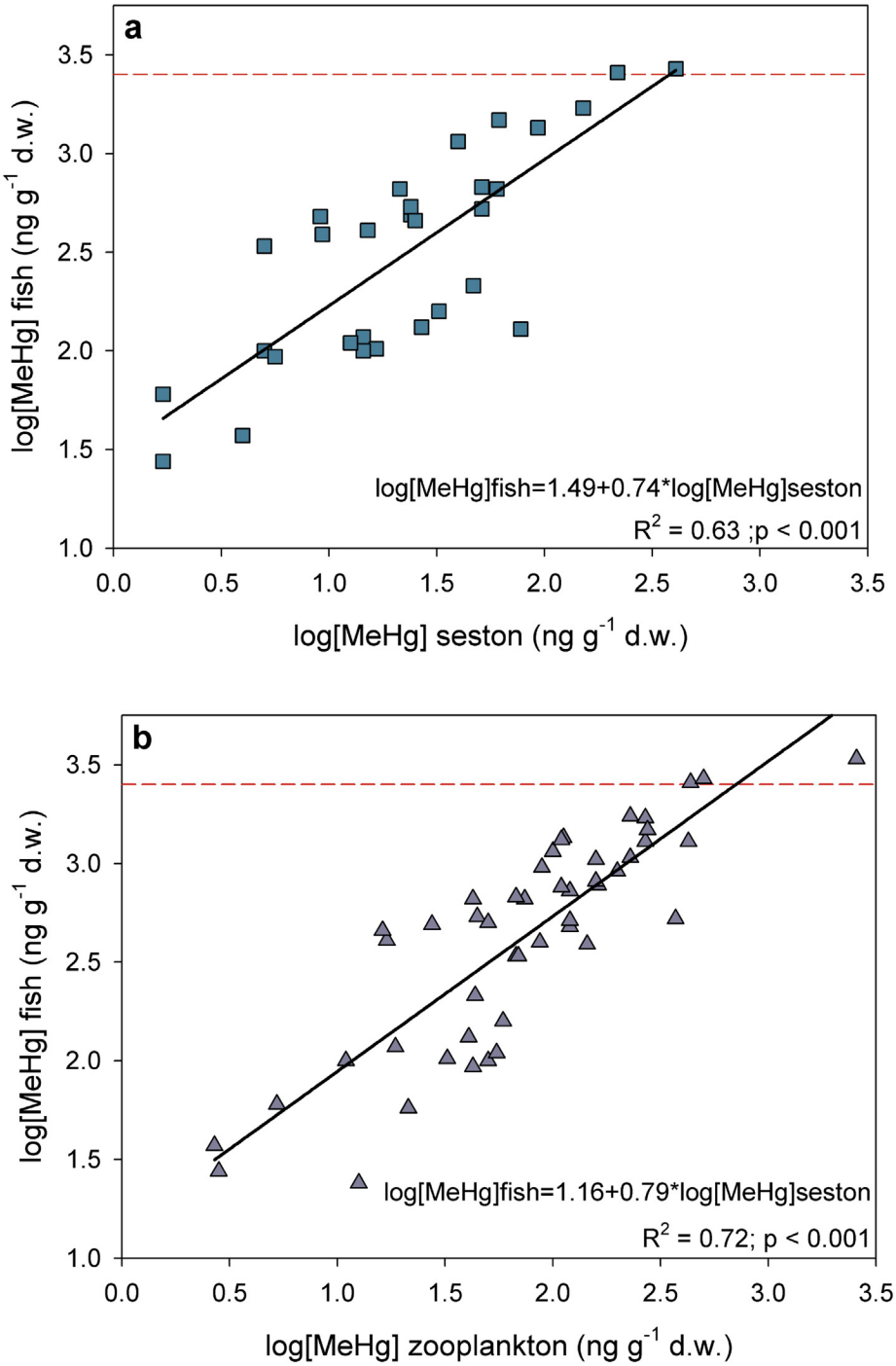

Fig. 1. The relationship between $[MeHg]_{seston}$ (ng g^{-1} d.w.) and preyfish [MeHg] (ng g^{-1} d.w.) for all of the aquatic ecosystems from the different study sites where seston was identified (aquatic ecosystems n = 40, (a), upper panel), as well as between $[MeHg]_{zooplankton}$ (ng g^{-1} d.w.) and preyfish [MeHg] (ng g^{-1} d.w.) where zooplankton was identified (aquatic ecosystems n = 58, (b), lower panel). Dashed line indicates WHO EQS of 0.5 μg MeHg g^{-1} w.w., assuming 100% MeHg.

MeHg is also more efficiently transferred from water to the food web base in oligotrophic waters than eutrophic waters ($logBCF_{w\text{-}s}$ in oligotrophic waters significantly higher than in eutrophic waters ANOVA: $F(1,29) = 5.35$, $p = 0.028$).

4. Discussion

4.1. Comparison of MeHg basal transfer with other studies

We reviewed studies covering a wide range of environmental conditions with regard to geographic and nutrient conditions from which several key commonalities became apparent. $BCF_{w\text{-}s}$ values are similar within geographically similar catchments (i.e. temperate or boreal waters). The findings are largely consistent with the regionally comprehensive summary study of the Western Great Lakes Region by Rolfhus et al. (2011). We also found that aqueous MeHg bioconcentration into seston ($BCF_{w\text{-}s}$) and zooplankton ($BCF_{w\text{-}z}$) were higher in high latitude, boreal waters. Moreover, MeHg tends to be more efficiently transferred along the food chain at higher latitudes. This is similar to what Lavoie et al. (2013) found for MeHg transfer between trophic levels, even though the regional difference between BCF that we found is larger than the regional difference in BMF. The effect of latitude on BCF and

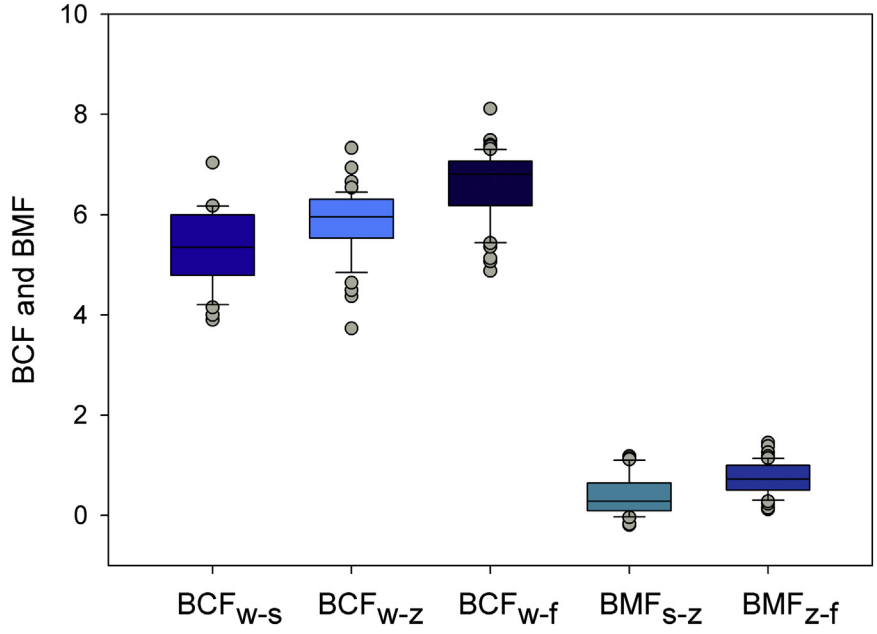

Fig. 2. Bioconcentration factors (BCF, mL g^{-1}) and biomagnification factors (BMF) (log-transformed mean ± standard deviation) representing, from left to right, BCF of MeHg transfer from water to seston ($logBCF_{w-s}$, 5.32 ± 0.73), from water to zooplankton ($logBCF_{w-z}$, 5.82 ± 0.69), and water to preyfish ($logBCF_{w-f}$, 6.60 ± 0.69), then MeHg BMF within the food web from seston to zooplankton ($logBMF_{s-z}$, 0.38 ± 0.38), and finally the MeHg trophic transfer from zooplankton to preyfish ($logBMF_{z-f}$, 0.75 ± 0.31).

BMF may be explained by several factors including: (1) temperature: slower growth of individuals relative to food consumptions in colder climates at higher latitudes which facilitates MeHg bioaccumulation (Simoneau et al., 2005; Baumann et al., 2017); (2) biodiversity: boreal aquatic ecosystems have lower species richness and productivity compared to temperate ones (Schindler, 1990). This type of simple pelagic food web, particularly in oligotrophic ecosystems, could support more efficient Hg biomagnification, assuming that fluctuations in MeHg concentrations at the base of a simpler food web would more readily trigger fluctuations of MeHg trophic transfer, which is different from biomass dilution (Lavoie et al., 2013); (3) more efficient microbial activity in Hg methylation within aquatic ecosystems in colder regions, specifically polar regions (Barkay et al., 2011).

In contrast to previous studies (Paterson et al., 1998; Watras et al., 1998; Hammerschmidt et al., 2013), we did not find that aqueous [MeHg] could predict MeHg concentrations in either plankton or fish with statistical significance. However, removing one site with high aqueous [MeHg] (Site 16, aqueous [MeHg] > 1 ng L^{-1}) enabled significant prediction of seston MeHg from aqueous [MeHg] ($R^2 = 0.22$, $p < 0.01$). Yet there remained a lack of statistical significance between aqueous [MeHg] and zooplankton [MeHg] or fish [MeHg]. Direct uptake of MeHg from water to fish under extreme MeHg pollution is one possible cause (Fujiki et al., 1976). We consider it more likely, though, that aqueous MeHg bioavailability to the food web base varies from place to place under the influence of environmental variables (pH, DOC, trophic status, etc.), and consequently aqueous [MeHg] exposure alone is not a strong predictor of biological MeHg concentrations.

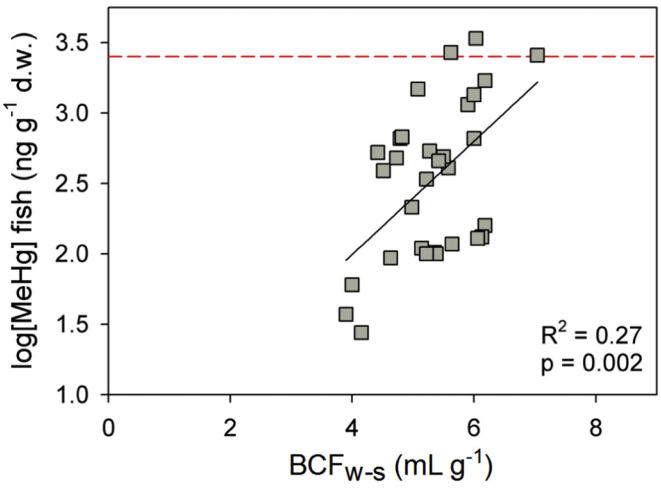

Fig. 3. Relationship between $logBCF_{w-s}$ and preyfish [MeHg] (ng g^{-1} d.w.), where seston [MeHg] (ng g^{-1} d.w.) was identified (aquatic ecosystems n = 40). Red dashed line indicates WHO EQS of 0.5 μg MeHg g^{-1} w.w. assuming 100% MeHg. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Our meta-analysis underlines the importance of MeHg transfer from water into the base of the pelagic food web (as represented by BCF_{w-s}) relative to the subsequent MeHg bioaccumulation from seston to zooplankton and then on to preyfish (as represented by BMF_{z-f}). A comparison of MeHg BCF_{w-s} with BMF_{z-f} showed that the former is the major control on the [MeHg] in preyfish. This is in accordance with Mackay et al. (2013) who used the mathematical relationships between BCF, BMF, and TMF to predict that BCF (without dietary input of a chemical) can be regarded as the fundamental determinant of bioaccumulation and biomagnification in aquatic food webs. Furthermore, $[MeHg]_{fish}$ correlated significantly with, and could be explained by BCF_{w-s}, while BMF_{z-f} did not show any predictive power. Further biomagnification along trophic levels will increase MeHg concentrations, but more steadily and predictably with increasing food web lengths as shown by Cabana et al. (1994), Da Silva et al. (2005), and Kidd et al. (2011). Wyn et al. (2009) have also stressed the prominence of water-seston MeHg uptake in food webs over trophic factors in low-pH lakes of

Table 1
Highly influential predictor variables (VIP scores > 1.0) of BCF from our PLS analysis of the PLS BCF.

Variables	VIP ± SE	Coefficient
DOC	1.85 ± 1.37	−0.36
pH	1.52 ± 0.63	−0.23
Eutrophic to hypereutrophic	1.47 ± 1.22	−0.25
Boreal	1.24 ± 0.56	0.14
Temperate	1.24 ± 0.56	−0.14

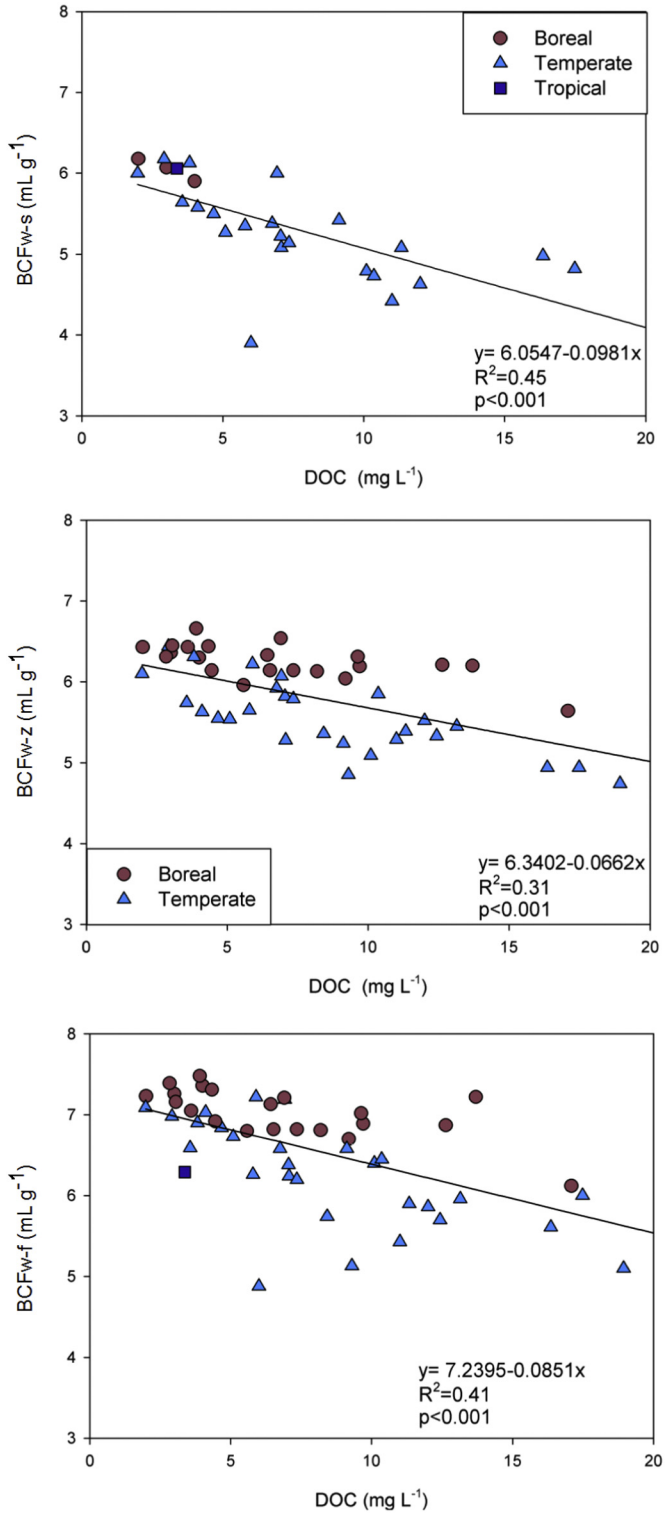

Fig. 4. Relationships between DOC and BCF ($logBCF_{w\text{-}s}$, $logBCF_{w\text{-}z}$, $logBCF_{w\text{-}f}$). Dots are sites within the boreal region, triangles are sites within the temperate region, and the only square is from the single tropical site (Site 9).

Nova Scotia, Canada. A few years later Pouilly et al. (2013), working with an Amazonian food web from Iténez River in Bolivia, revealed that periphyton contribution at the food web base is more critical to the elevated Hg bioaccumulation than food chain length. Clayden et al. (2013) also emphasized the role of lower-trophic-level organisms in driving changes of the trophic magnification slope intercepts under a gradient of DOC concentrations in 11 Canadian boreal lakes.

4.2. Classifying MeHg transfer steps

The importance of $BCF_{w\text{-}s}$ relative to the bioaccumulation from zooplankton into preyfish ($BMF_{z\text{-}f}$) leads us to conceptualize MeHg biomagnification in aquatic ecosystems (the total increase in MeHg from water to subsequent trophic levels) as having two distinct steps. The first step is transferring bioavailable MeHg from the water column into the base of the pelagic food web, represented by the $BCF_{w\text{-}s}$. The next step is MeHg biomagnifciation within the food web from one trophic level to the next ($BMF_{s\text{-}z}$, then $BMF_{z\text{-}f}$), and onwards. We characterize this trophic transfer as an inclined "stairway" of MeHg bioaccumulation (Fig. 5). The movement from water into the base of the pelagic food web ($BCF_{w\text{-}s}$), on the other hand, is more of a vertical "elevator", highlighting the significance that first step MeHg takes from the water column into the base of the food web (Fig. 5). This "elevator" ($BCF_{w\text{-}s}$) takes the aqueous [MeHg] and increases it by anywhere from three to six orders of magnitude, which is decisive for [MeHg] at subsequent trophic levels.

The key point of this conceptualization is that the variation in the vulnerability of aquatic ecosystems to MeHg biomagnification can be predicted from MeHg bioconcentration at the water-seston interface, since entry into the base of the food web varies much more than the subsequent trophic magnification ($BMF_{s\text{-}z}$ and $BMF_{z\text{-}f}$). As seen in our results, [MeHg] in planktivorous fish is already strongly predicted by [MeHg] in seston (SI Fig. S2a). This conceptualization can be related to the formula of MeHg biomagnification described in Borga et al. (2012),

$$\log[\mathrm{MeHg}]_{biota} = TMF \times TL + \mathbf{intercept} \qquad \text{(Borga et al. (2012))} \quad \text{(A)}$$

where TMF stands for trophic magnification factor and TL represents the trophic level of organisms in the food web. TMF and BMF are quite similar given that they are both determined empirically, using field measures of [MeHg] and relative trophic levels. We suggest an extension of this formula that makes explicit the role of the $BCF_{w\text{-}s}$ in the "intercept":

$$\log[\mathrm{MeHg}]_{biota} = TMF \times TL + \mathbf{log}(\mathbf{BCF_{w\text{-}}\,s} \times [\mathbf{MeHg}]_{\mathbf{water}}) \qquad \text{(this review)} \quad \text{(B)}$$

The predictive power of $BCF_{w\text{-}s}$ for [MeHg] in preyfish, found in this study (and in earlier regional studies) has been overshadowed by intensive investigation of MeHg bioaccumulation through well-defined trophic transfers along the food web. The interest in biomagnification, as summarized by Borga et al. (2012) (Formula A), focuses primarily on the complexity of the food web properties, particularly with regards to seasonal and between-year variation in food availability (Chen et al., 2012; Drevnick et al., 2012). The variation that arises from these processes is a major challenge for modeling biotic [MeHg] (Borga et al., 2012; Eklöf et al., 2016). Nevertheless, more work has focused on the TMF, relative to the intercept as stated in Formula A. The extended model (Formula B) specifically recognizes the role of bioconcentration into the base of the pelagic food web. The results from our meta-analysis show that this BCF is a predictor of $[\mathrm{MeHg}]_{fish}$, and the BCF is the key determinant of the "intercept" in Formula A. Compared to the $[\mathrm{MeHg}]_{water}$, which generally varies by one order of magnitude in natural ecosystems (hundredths to tenths of a ng L^{-1}, SI Table S2), the $BCF_{w\text{-}s}$ varies over 3–7 orders of magnitude.

Our conceptualization (Fig. 5) indicates that the magnitude of the $BCF_{w\text{-}s}$, rather than the $[\mathrm{MeHg}]_{water}$, is the key determinant of [MeHg] in planktivorous fish. Importantly, this model further suggests that [MeHg] in subsequent consumers at higher trophic levels depends to a large extent on the magnitude of the MeHg $BCF_{w\text{-}s}$ (or the height of the "MeHg elevator ride" into the base of the pelagic food web). This is evident from the strong correlation between $BCF_{w\text{-}s}$ and [MeHg] in planktivorous or small omnivorous fish. However, with increasing

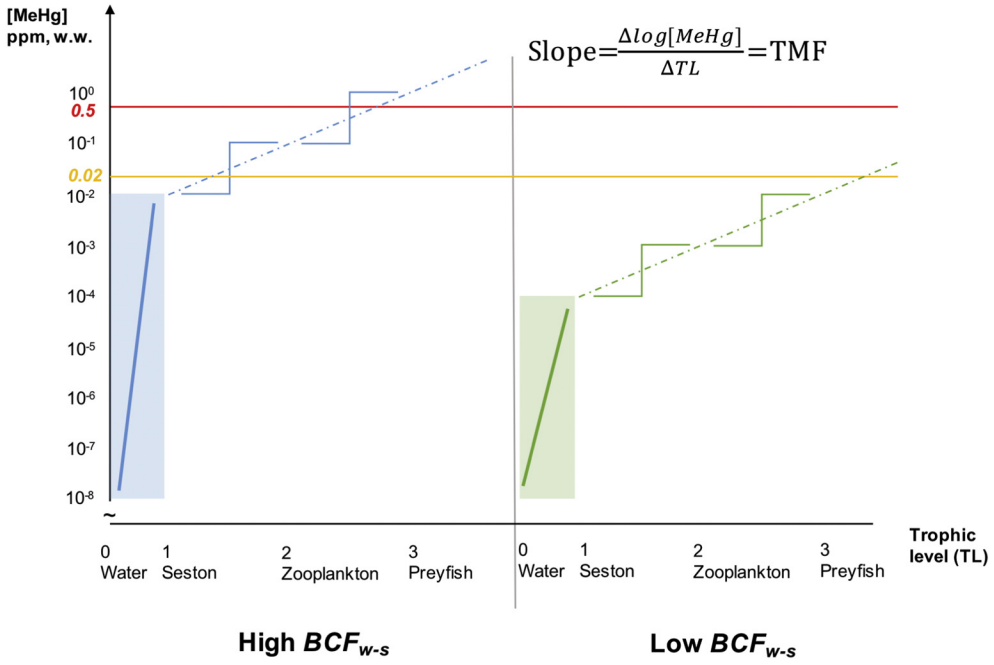

Fig. 5. Conceptualization of the "MeHg elevator" from water to the base of the pelagic food web (the $BCF_{w\text{-}s}$), and the subsequent "MeHg staircase" of increase along the trophic levels in a pelagic food web. The X-axis defines the conceptualized trophic level. The Y-axis is [MeHg] (ppm, equivalent to μg MeHg mL^{-1} in aqueous phase, μg MeHg g^{-1} in solid phase). Blue solid lines indicate changes of [MeHg] in aquatic ecosystems with high $BCF_{w\text{-}s}$, while green solid lines indicate the changes in low $BCF_{w\text{-}s}$ systems. Shaded blue and green areas highlight the contrasts for aquatic food webs with different $BCF_{w\text{-}s}$. The slopes of the dashed lines represent the TMF, similar to BMF. The horizontal orange solid line is EU's EQS of 0.02 μg g^{-1} w.w. for freshwater biota (EC, 2013). The horizontal red solid line is the more often adopted safe consumption WHO EQS for [Hg] in fish (assuming 100% MeHg) of 0.5 μg MeHg g^{-1} w.w. (IPCS, 1990; USEPA, 2012). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

food web lengths, the number of stairs in the trophic transfer between the start and end of the bioaccumulation "staircase" become more important in determining the concentrations at the top of the aquatic food web.

4.3. Correlating MeHg transfer with environmental factors

The investigated environmental factors, especially DOC and trophic status, influenced the MeHg transfer from seston to zooplankton ($BMF_{s\text{-}z}$) more than the transfer from zooplankton to fish ($BMF_{z\text{-}f}$). Our PLS BCF model (Table 1) and linear regressions (Fig. 4) implicated the importance of in-lake water chemistry parameters for BCF. Specifically, DOC—which may influence the accumulation of MeHg from water to zooplankton by strengthening food web links due to bacteria in the microbial loop (Jonsson et al., 2017); climate-related factors of temperature and latitude that can affect individual growth efficiency, MeHg production, and fluxes at the base of the food web, indirectly influencing on MeHg bioaccumulation (Dijkstra et al., 2013); nutrient loadings under seasonal precipitation patterns which are also crucial to productivity of the food web and thus MeHg production and bioaccumulation (Driscoll et al., 2012). This is a reminder that also within $BCF_{w\text{-}s}$ and $BCF_{w\text{-}z}$ there are additional factors that have not been explicitly introduced into the PLS model (e.g., temperature, nutrient loadings, and precipitation).

Regional differences in MeHg transfer within the base of the food web as defined by seston can be influenced by various characteristics of aquatic ecosystems. Nutrient availability greatly affects both plankton's growth efficiency and somatic growth of fish, which is the amount of new somatic biomass produced per unit dietary energy consumed. This influences biomass dilution that can play a significant role in decreasing both BCF and BMF (Arnot et al., 2008), especially in temperate and tropical regions (Karimi et al., 2007; Liu et al., 2012). Low growth efficiency in plankton can also be influenced by catchment characteristics: low pH in weakly buffered aquatic ecosystems can be associated with low nutrient availability which will affect growth efficiency in both fish and plankton due to limited food consumption, which is the opposite of growth dilution. This can be linked with more efficient MeHg biomagnification and higher fish [MeHg] (Iivonen et al., 1992; Le Faucheur et al., 2011), all related to higher BCF values. As reported in this meta-analysis, pH in freshwater sites is negatively correlated with $BCF_{w\text{-}s}$ and low pH sites exhibited higher zooplankton and preyfish [MeHg].

Our results showed some relationship between $BCF_{w\text{-}s}$, eutrophy, and lower MeHg bioaccumulation in fish. This is in line with previous findings that MeHg bioaccumulation is typically higher in meso- and oligotrophic aquatic ecosystems compared to eutrophic systems (Kainz et al., 2003; Kehrig et al., 2009; Dieguez et al., 2013). The lower MeHg bioaccumulation often reported in eutrophic waters may also be related to higher pH in eutrophic waters (Cheng et al., 2011; Liu et al., 2012). This inter-relationship was also found in our PLS analysis (SI Fig. S2a).

The effect of DOC on MeHg $BCF_{w\text{-}s}$ is complex. Despite a well-known chemical coupling between DOC and Hg in freshwaters (Meili et al., 2003; Ravichandran, 2004; Eklöf et al., 2012), DOC can also interact with biota to decrease $BCF_{w\text{-}s}$ through passive uptake in plankton because of Hg binding to aquatic humic substances (Sjöblom et al., 2000; Gorski et al., 2006). Terrigenous organic matter (OM) plays an important role in MeHg production and transfer in boreal aquatic food webs when it contains low molecular mass fulvic acids that associate strongly with Hg (French et al., 2014). Recently, Jonsson et al. (2017) argued that increases in terrigenous OM induced a shift in the trophic structure by stimulating bacterial growth and consequently favoring phagotrophic protozoa, essentially a microbial loop, that eventually increased the length of the aquatic food chain. This implies an indirect influence on zooplankton growth that enhances MeHg bioaccumulation in consumers. Such influences may contribute to why our statistical analyses found that DOC was more important than pH and biome regions for BCF, especially $BCF_{w\text{-}s}$. Similarly, the negative influence of DOC on the MeHg bioaccumulation at the food web base was observed in eastern

Canadian lakes, where low DOC lakes have higher sensitivity to Hg and higher invertebrate [MeHg] (Chételat et al., 2018). As mentioned in the beginning of this paper, MeHg PD in freshwater lakes is also mediated by DOC, resulting in differentiated MeHg flux across waters along a gradient of DOC concentrations. Although UV radiation more efficiently contributed to MeHg PD activity in colored water compared to clear water, this process occurs mainly in the surface water layers of humic-rich lakes due to light attenuation (Lehnherr and St. Louis, 2009). A more recent work by Klapstein et al. (2018) argued further that MeHg PD activity is largely inhibited in high DOC environments due to the quick turnover of photoreactive DOM competing for photons with MeHg-DOM complexes in the water column. Thus high DOC environments may preserve much of the MeHg flux from being photodemethylated, making biota in colored water more sensitive to MeHg flux changes over time.

The differences in the mechanisms controlling the "MeHg elevator" (water to the base of the food web) and "MeHg staircase" (seston-zooplankton-fish) mean that MeHg bioavailability from the water column at the food web base and further biomagnification can be influenced in different ways by different environmental factors. The base of the food web also needs to be clearly defined to capture functional differences in MeHg trophic transfer between different types of plankton. It is quite likely that key features of these functional differences are related to the likelihood that there is already trophic transfer occurring within the base of the food web (Kainz and Mazumder, 2005), due to the fact that most studies of plankton MeHg involve sampling by size fractions that do not accurately separate primary producers from primary consumers. This results in clouding the distinction between bioconcentration and biomagnification within the planktonic "base" of the food web.

The understanding of the interaction between environmental factors and functional organism groups at the base of the food web is crucial to improving the understanding and predictability of overall Hg biomagnification. Nutrient-poor aquatic ecosystems, many of which also have low pH, such as those in remote areas of Northern America and northern Europe and Asia, exhibited both the highest $BCF_{w\text{-}s}$, and the highest $[MeHg]_{fish}$. Increases in nutrients will result in higher primary production, thus the aqueous MeHg present is taken up into a larger amount of higher phytoplankton biomass (i.e., MeHg biodilution, Pickhardt et al. (2002)). This lowers the $BCF_{w\text{-}s}$, leading to lower MeHg in consumers at higher trophic levels. The trophic status of aquatic ecosystems influences growth of primary producers and consumers, and thereby trophic MeHg transfer within the planktonic food web. Such processes (i.e., the interactions of autotrophs, heterotrophs and mixotrophs) at the base of the aquatic food web require further elucidation to distinguish true bioconcentration from trophic transfers at the base of the food web when trying to understand the variation in BCF.

This study highlights the importance of bioconcentration into the base of the food web as a key step in determining fish [MeHg]. Understanding how to control this "elevator increase" from water into the seston could be especially useful for evaluating measures to reduce fish Hg. Regional variation in Hg bioaccumulation is an indication of the potential range of food web vulnerability to Hg biomagnification. Further investigations into the complex ecological interactions influencing MeHg transfer at the base of the food web are needed to transform the observed variation of MeHg biomagnification into knowledge that can improve strategies to reduce the risks of MeHg exposure to humans and wildlife from freshwater fish.

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Appendix A. Supplementary data

Publication selection process with a detailed list of screened literature (SI 1); map of the world with selected sites (SI 2); summary of MeHg concentrations from selected aquatic ecosystems, including water chemistry and site characteristics (SI 3); PLS BCF model details with loading scores and scatter plots (SI 4). Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.07.328.

References

Åkerblom, S., Bignert, A., Meili, M., Sonesten, L., Sundbom, M., 2014. Half a century of changing mercury levels in Swedish freshwater fish. Ambio 43, 91–103.
Arnot, J.A., MacKay, D., Bonnell, M., 2008. Estimating metabolic biotransformation rates in fish from laboratory data. Environ. Toxicol. Chem. 27, 341–351.
Barkay, T., Kroer, N., Poulain, A.J., 2011. Some like it cold: microbial transformations of mercury in polar regions. Polar Res. 30.
Baumann, Z., Mason, R.P., Conover, D.O., Balcom, P., Chen, C.Y., Buckman, K.L., Fisher, N.S., Baumann, H., 2017. Mercury bioaccumulation increases with latitude in a coastal marine fish (Atlantic silverside, *Menidia menidia*). Can. J. Fish. Aquat. Sci. 74, 1009–1015.
Bellanger, M., Pichery, C., Aerts, D., Berglund, M., Castaño, A., Čejchanová, M., Crettaz, P., Davidson, F., Esteban, M., Fischer, M.E., Gurzau, A.E., Halzlova, K., Katsonouri, A., Knudsen, L.E., Kolossa-Gehring, M., Koppen, G., Ligocka, D., Miklavčič, A., Reis, M.c., Rudnai, P., Tratnik, J.S., Weihe, P., Budtz-Jørgensen, E., Grandjean, P., 2013. Economic benefits of methylmercury exposure control in Europe: monetary value of neurotoxicity prevention. Environ. Health 12, 3.
Borga, K., Kidd, K.A., Muir, D.C., Berglund, O., Conder, J.M., Gobas, F.A., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: considerations of ecology, ecosystems, and study design. Integr. Environ. Assess. Manag. 8, 64–84.
Bravo, A.G., Loizeau, J.L., Bouchet, S., Richard, A., Rubin, J.F., Ungureanu, V.G., Amouroux, D., Dominik, J., 2010. Mercury human exposure through fish consumption in a reservoir contaminated by a chlor-alkali plant: Babeni reservoir (Romania). Environ. Sci. Pollut. Res. 17, 1422–1432.
Bravo, A.G., Cosio, C., Amouroux, D., Zopfi, J., Cheualley, P.A., Spangenberg, J.E., Ungureanu, V.G., Dominik, J., 2014. Extremely elevated methyl mercury levels in water, sediment and organisms in a Romanian reservoir affected by release of mercury from a chlor-alkali plant. Water Res. 49, 391–405.
Brett, M.T., Bunn, S.E., Chandra, S., Galloway, A.W.E., Guo, F., Kainz, M., Kankaala, P., Lau, D.C.P., Moulton, T.P., Power, M., Rasmussen, J.B., Taipale, S., Thorpe, J.H., Wehr, J.D., 2017. How important are terrestrial organic carbon inputs for secondary production in freshwater ecosystems? Freshw. Biol. 62, 833–853.
Brooks, J.L., Dodson, S.I., 1965. Predation, body size, and composition of plankton. Science 150, 28–35.
Cabana, G., Tremblay, A., Kalff, J., Rasmussen, J.B., 1994. Pelagic food chain structure in Ontario lakes: a determinant of mercury levels in lake trout (*Salvelinus namaycush*). Can. J. Fish. Aquat. Sci. 51, 381–389.
Chen, C., Kamman, N., Williams, J., Bugge, D., Taylor, V., Jackson, B., Miller, E., 2012. Spatial and temporal variation in mercury bioaccumulation by zooplankton in Lake Champlain (North America). Environ. Pollut. 161, 343–349.
Chen, C.Y., Borsuk, M.E., Bugge, D.M., Hollweg, T., Balcom, P.H., Ward, D.M., Williams, J., Mason, R.P., 2014. Benthic and pelagic pathways of methylmercury bioaccumulation in estuarine food webs of the Northeast United States. PLoS One 9.
Cheng, Z., Liang, P., Shao, D.D., Wu, S.C., Nie, X.P., Chen, K.C., Li, K.B., Wong, M.H., 2011. Mercury biomagnification in the aquaculture pond ecosystem in the Pearl River Delta. Arch. Environ. Contam. Toxicol. 61, 491–499.
Chételat, J., Richardson, M.C., MacMillan, G.A., Amyot, M., Poulain, A.J., 2018. Ratio of methylmercury to dissolved organic carbon in water explains methylmercury bioaccumulation across a latitudinal gradient from north-temperate to Arctic lakes. Environ. Sci. Technol. 52, 79–88.
Clarkson, T.W., Magos, L., Myers, G.J., 2003. The toxicology of mercury – current exposures and clinical manifestations. N. Engl. J. Med. 349, 1731–1737.
Clayden, M.G., Kidd, K.A., Wyn, B., Kirk, J.L., Muir, D.C.G., O'Driscoll, N.J., 2013. Mercury biomagnification through food webs is affected by physical and chemical characteristics of lakes. Environ. Sci. Technol. 47, 12047–12053.
Da Silva, D.S., Lucotte, M., Roulet, M., Poirier, H., Mergler, D., Santos, E.O., Crossa, M., 2005. Trophic structure and bioaccumulation of mercury in fish of three natural lakes of the Brazilian Amazon. Water Air Soil Pollut. 165, 77–94.
Danielsson, S., Hedman, J., Miller, A., Bignert, A., 2011. Mercury in Perch From Norway, Sweden and Finland – Geographical Patterns and Temporal Trends. Swedish Natural History Musuem, Stockholm, Sweden.
Di Prinzio, Z.Y., Omad, G., Miserendino, M.L., Casaux, R., 2015. Selective foraging by non-native rainbow trout on invertebrates in Patagonian streams in Argentina. Zool. Stud. 54.
Dieguez, M.C., Queimalinos, C.P., Guevara, S.R., Marvin-DiPasquale, M., Cardenas, C.S., Arribere, M.A., 2013. Influence of dissolved organic matter character on mercury incorporation by planktonic organisms: an experimental study using oligotrophic water from Patagonian lakes. J. Environ. Sci. (China) 25, 1980–1991.
Dijkstra, J.A., Buckman, K.L., Ward, D., Evans, D.W., Dionne, M., Chen, C.Y., 2013. Experimental and natural warming elevates mercury concentrations in estuarine fish. PLoS One 8, e58401.

Dominik, J., Tagliapietra, D., Bravo, A.G., Sigovini, M., Spangenberg, J.E., Amouroux, D., Zonta, R., 2014. Mercury in the food chain of the Lagoon of Venice, Italy. Mar. Pollut. Bull. 88, 194–206.
Drevnick, P.E., Engstrom, D.R., Driscoll, C.T., Swain, E.B., Balogh, S.J., Kamman, N.C., Long, D.T., Muir, D.G.C., Parsons, M.J., Rolfhus, K.R., Rossmann, R., 2012. Spatial and temporal patterns of mercury accumulation in lacustrine sediments across the Laurentian Great Lakes region. Environ. Pollut. 161, 252–260.
Driscoll, C.T., Han, Y.J., Chen, C.Y., Evers, D.C., Lambert, K.F., Holsen, T.M., Kamman, N.C., Munson, R.K., 2007. Mercury contamination in forest and freshwater ecosystems in the northeastern United States. Bioscience 57, 17–28.
Driscoll, C.T., Chen, C.Y., Hammerschmidt, C.R., Mason, R.P., Gilmour, C.C., Sunderland, E.M., Greenfield, B.K., Buckman, K.L., Lamborg, C.H., 2012. Nutrient supply and mercury dynamics in marine ecosystems: a conceptual model. Environ. Res. 119, 118–131.
EC, 2013. In: E. Commission (Ed.), Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 Amending Directives 2000/60/EC and 2008/105/EC as Regards Priority Substances in the Field of Water Policy.
Eklöf, K., Fölster, J., Sonesten, L., Bishop, K., 2012. Spatial and temporal variation of THg concentrations in run-off water from 19 boreal catchments, 2000–2010. Environ. Pollut. 164, 102–109.
Eklöf, K., Lidskog, R., Bishop, K., 2016. Managing Swedish forestry's impact on mercury in fish: defining the impact and mitigation measures. Ambio 45, S163–S174.
FAO/WHO, 2015. Codex general standard for contaminants and toxins in food and feed. Codex Standard 193–1995. Codex Alimentarius, International Food Standards, pp. 47–48.
Farkas, A., Salánki, J., Varanka, I., 2000. Heavy metal concentrations in fish of Lake Balaton. Lakes Reserv. Res. Manag. 5, 271–279.
French, T.D., Houben, A.J., Desforges, J.P.W., Kimpe, L.E., Kokelj, S.V., Poulain, A.J., Smol, J.P., Wang, X.W., Blais, J.M., 2014. Dissolved organic carbon thresholds affect mercury bioaccumulation in Arctic Lakes. Environ. Sci. Technol. 48, 3162–3168.
Fujiki, M., Hirota, R., Yamaguchi, S., 1976. The mechanism of methylmercury accumulation in fish. 2nd U.S.-Japan Experts Meeting, pp. 89–95 (Tokyo, Japan).
Gandhi, N., Bhavsar, S.P., Tang, R.W.K., Drouillard, K.G., Arhonditsis, G.B., 2014. Significance of toxaphene in Great Lakes fish consumption advisories. J. Great Lakes Res. 40, 71–79.
Gobas, F.A.P.C., Morrison, H.A., 2000. Bioconcentration and biomagnification in the aquatic environment. Handbook of Property Estimation Methods for Chemicals. CRC Press.
Gorski, P.R., Armstrong, D.E., Hurley, J.P., Shafer, M.M., 2006. Speciation of aqueous methylmercury influences uptake by a freshwater alga (*Selenastrum capricornutum*). Environ. Toxicol. Chem. 25, 534–540.
Hammerschmidt, C.R., Finiguerra, M.B., Weller, R.L., Fitzgerald, W.F., 2013. Methylmercury accumulation in plankton on the continental margin of the Northwest Atlantic Ocean. Environ. Sci. Technol. 47, 3671–3677.
Hirota, R., Fujiki, M., Tajima, S., 1979. Mercury contents of zooplankton collected in the Tropical Pacific Ocean. Nippon Suisan Gakkaishi 45, 1449–1451.
Iivonen, P., Piepponen, S., Verta, M., 1992. Factors affecting trace-metal bioaccumulation in Finnish headwater lakes. Environ. Pollut. 78, 87–95.
IPCS, 1990. Methylmercury. In: I.P.O.C.S. World Health Organization (Ed.), Environmental Health Criteria 101. International Programme on Chemical Safety, Geneva, Switzerland.
Jonsson, S., Andersson, A., Nilsson, M.B., Skyllberg, U., Lundberg, E., Schaefer, J.K., Åkerblom, S., Björn, E., 2017. Terrestrial discharges mediate trophic shifts and enhance methylmercury accumulation in estuarine biota. Sci. Adv. 3.
Kainz, M., Mazumder, A., 2005. Effect of algal and bacterial diet on methyl mercury concentrations in zooplankton. Environ. Sci. Technol. 39, 1666–1672.
Kainz, M., Lucotte, M., Parrish, C.C., 2003. Relationships between organic matter composition and methyl mercury content of offshore and carbon-rich littoral sediments in an oligotrophic lake. Can. J. Fish. Aquat. Sci. 60, 888–896.
Karimi, R., Chen, C.Y., Pickhardt, P.C., Fisher, N.S., Folt, C.L., 2007. Stoichiometric controls of mercury dilution by growth. Proc. Natl. Acad. Sci. 104, 7477–7482.
Kasper, D., Forsberg, B.R., Amaral, J.H.F., Leitão, R.P., Py-Daniel, S.S., Bastos, W.R., Malm, O., 2014. Reservoir stratification affects methylmercury levels in river water, plankton, and fish downstream from Balbina Hydroelectric Dam, Amazonas, Brazil. Environ. Sci. Technol. 48, 1032–1040.
Kehrig, H.A., Palermo, E.F.A., Seixas, T.G., Santos, H.S.B., Malm, O., Akagi, H., 2009. Methyl and total mercury found in two man-made Amazonian reservoirs. J. Braz. Chem. Soc. 20, 1142–1152.
Kidd, K.A., Clayden, M.G., Jardine, T.D., 2011. Bioaccumulation and biomagnification of mercury through food webs. In: Liu, G., Cai, C., O'driscoll, N.J. (Eds.), Environmental Chemistry and Toxicology of Mercury.
Kindaichi, M., Matsuyama, A., 2005. Change of the total mercury concentration in fish of Minamata Bay over the past 26 years. J. Jpn. Soc. Water Environ. 28, 529–533.
Klapstein, S.J., Ziegler, S.E., O'Driscoll, N.J., 2018. Methylmercury photodemethylation is inhibited in lakes with high dissolved organic matter. Environ. Pollut. 232, 392–401.
KumamotoPrefecture, 1998. An Outline of the Environmental Restoration of Minamata Bay (In Japanese: "水俣湾環境復元事業の概要"). p. 118 (Kumamoto Prefecture, Kumamoto, Japan).
Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A., Campbell, L.M., 2013. Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. Environ. Sci. Technol. 47, 13385–13394.
Le Faucheur, S., Tremblay, Y., Fortin, C., Campbell, P.G.C., 2011. Acidification increases mercury uptake by a freshwater alga, *Chlamydomonas reinhardtii*. Environ. Chem. 8, 612–622.
Le Faucheur, S., Campbell, P.G.C., Fortin, C., Slaveykova, V.I., 2014. Interactions between mercury and phytoplankton: speciation, bioavailability, and internal handling. Environ. Toxicol. Chem. 33, 14.
Lehnherr, I., 2014. Methylmercury biogeochemistry: a review with special reference to Arctic aquatic ecosystems. Environ. Rev. 1–15.
Lehnherr, I., St. Louis, V.L., 2009. Importance of ultraviolet radiation in the photodemethylation of methylmercury in freshwater ecosystems. Environ. Sci. Technol. 43, 5692–5698.
Lindqvist, O., Johansson, K., Aastrup, M., Andersson, A., Bringmark, L., Hovsenius, G., Hakanson, L., Iverfeldt, A., Meili, M., Timm, B., 1991. Mercury in the Swedish environment - recent research on causes, consequences and corrective methods. Water Air Soil Pollut. 55.
Liu, B., Yan, H., Wang, C., Li, Q., Guédron, S., Spangenberg, J.E., Feng, X., Dominik, J., 2012. Insights into low fish mercury bioaccumulation in a mercury-contaminated reservoir, Guizhou, China. Environ. Pollut. 160, 109–117.
Mackay, D., Arnot, J.A., Gobas, F.A.P.C., Powell, D.E., 2013. Mathematical relationships between metrics of chemical bioaccumulation in fish. Environ. Toxicol. Chem. 32, 1459–1466.
Mason, R.P., Reinfelder, J.R., Morel, F.M.M., 1995. Bioaccumulation of mercury and methylmercury. Water Air Soil Pollut. 80, 915–921.
Mason, R.P., Reinfelder, J.R., Morel, F.M.M., 1996. Uptake, toxicity, and trophic transfer of mercury in a coastal diatom. Environ. Sci. Technol. 30, 1835–1845.
Mason, R.P., Choi, A.L., Fitzgerald, W.F., Hammerschmidt, C.R., Lamborg, C.H., Soerensen, A.L., Sunderland, E.M., 2012. Mercury biogeochemical cycling in the ocean and policy implications. Environ. Res. 119, 101–117.
McMeans, B.C., Arts, M.T., Fisk, A.T., 2015. Impacts of food web structure and feeding behavior on mercury exposure in Greenland Sharks (*Somniosus microcephalus*). Sci. Total Environ. 509, 216–225.
Meili, M., 1991. Mercury in forest lake ecosystems - bioavailability, bioaccumulation and biomagnification. Water Air Soil Pollut. 55, 131–157.
Meili, M., Bishop, K., Bringmark, L., Johansson, K., Munthe, J., Sverdrup, H., de Vries, W., 2003. Critical levels of atmospheric pollution: criteria and concepts for operational modelling of mercury in forest and lake ecosystems. Sci. Total Environ. 304, 83–106.
Munthe, J., Bodaly, R.A., Branfireun, B.A., Driscoll, C.T., Gilmour, C.C., Harris, R., Horvat, M., Lucotte, M., Malm, O., 2007. Recovery of mercury-contaminated fisheries. Ambio 36, 33–44.
Nguetseng, R., Fliedner, A., Knopf, B., Lebreton, B., Quack, M., Rüdel, H., 2015. Retrospective monitoring of mercury in fish from selected European freshwater and estuary sites. Chemosphere 134, 427–434.
Nguyen, H.L., Leermakers, M., Kurunczi, S., Bozo, L., Baeyens, W., 2005. Mercury distribution and speciation in Lake Balaton, Hungary. Sci. Total Environ. 340, 231–246.
Paterson, M.J., Rudd, J.W.M., St Louis, V., 1998. Increases in total and methylmercury in zooplankton following flooding of a peatland reservoir. Environ. Sci. Technol. 32, 3868–3874.
Pickhardt, P.C., Folt, C.L., Chen, C.Y., Klaue, B., Blum, J.D., 2002. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. Proc. Natl. Acad. Sci. U. S. A. 99, 4419–4423.
Pouilly, M., Rejas, D., Pérez, T., Duprey, J.-L., Molina, C.I., Hubas, C., Guimarães, J.-R.D., 2013. Trophic structure and mercury biomagnification in tropical fish assemblages, Iténez River, Bolivia. PLoS One 8, e65054.
Ravichandran, M., 2004. Interactions between mercury and dissolved organic matter—a review. Chemosphere 55, 319–331.
Rohatgi, A., 2016. WebPlotDigitizer: HTML5 Based Online tool to Extract Numerical Data From Plot Images. Version 3.10 (In:).
Rolfhus, K., Hall, B., Monson, B., Paterson, M., Jeremiason, J., 2011. Assessment of mercury bioaccumulation within the pelagic food web of lakes in the western Great Lakes region. Ecotoxicology 20, 1520–1529.
Roulet, M., Lucotte, M., Guimaraes, J.R.D., Rheault, I., 2000. Methylmercury in water, seston, and epiphyton of an Amazonian river and its floodplain, Tapajos River, Brazil. Sci. Total Environ. 261, 43–59.
Schindler, D.W., 1990. Natural and anthropogenically imposed limitations to biotic richness in freshwaters. In: Woodwell, G.M. (Ed.), The Earth in Transition: Patterns and Processes of Biotic Impoverishment. Cambridge University Press, Cambridge, UK, pp. 425–462.
Selin, N.E., 2009. Global biogeochemical cycling of mercury: a review. Annu. Rev. Environ. Resour. 34, 43–63.
Selin, N.E., Jacob, D.J., Park, R.J., Yantosca, R.M., Strode, S., Jaegle, L., Jaffe, D., 2007. Chemical cycling and deposition of atmospheric mercury: global constraints from observations. J. Geophys. Res.-Atmos. 112.
Shapiro, S.S., Wilk, M.B., 1965. An analysis of variance test for normality (complete samples). Biometrika 52, 591–611.
Sheehan, M.C., Burke, T.A., Navas-Acien, A., Breysse, P.N., McGready, J., Fox, M.A., 2014. Global methylmercury exposure from seafood consumption and risk of developmental neurotoxicity: a systematic review. Bull. World Health Organ. 92, 254–269.
Simoneau, M., Lucotte, M., Garceau, S., Laliberté, D., 2005. Fish growth rates modulate mercury concentrations in walleye (*Sander vitreus*) from eastern Canadian lakes. Environ. Res. 98, 73–82.
Sjöblom, Å., Meili, M., Sundbom, M., 2000. The influence of humic substances on the speciation and bioavailability of dissolved mercury and methylmercury, measured as uptake by Chaoborus larvae and loss by volatilization. Sci. Total Environ. 261, 115–124.
Suchanek, T.H., Eagles-Smith, C.A., Harner, E.J., 2008. Is Clear Lake methylmercury distribution decoupled from bulk mercury loading? Ecol. Appl. 18, A107–A127.

USEPA, 2012. Integrated Risk Information System (IRIS) on methyl mercury. National Center for Environmental Assessment. Office of Research and Development. United States Environmental Protection Agency, Washington D.C., U.S.
Vignati, D.A.L., Polesello, S., Bettinetti, R., Bank, M.S., 2013. Mercury environmental quality standard for biota in Europe: opportunities and challenges. Integr. Environ. Assess. Manag. 9, 167–168.
Watras, C.J., Bloom, N.S., 1992. Mercury and methylmercury in individual zooplankton - implications for bioaccumulation. Limnol. Oceanogr. 37, 1313–1318.
Watras, C.J., Back, R.C., Halvorsen, S., Hudson, R.J.M., Morrison, K.A., Wente, S.P., 1998. Bioaccumulation of mercury in pelagic freshwater food webs. Sci. Total Environ. 219, 183–208.
Wyn, B., Kidd, K.A., Burgess, N.M., Curry, R.A., 2009. Mercury biomagnification in the food webs of acidic lakes in Kejimkujik National Park and National Historic Site, Nova Scotia. Can. J. Fish. Aquat. Sci. 66, 1532–1545.