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Biomagnification of Mercury in Aquatic Food Webs: A Worldwide Meta-Analysis

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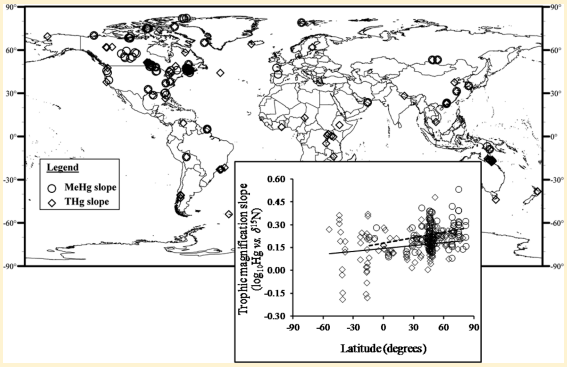
Supporting Information

ABSTRACT: The slope of the simple linear regression between $\log_{10}$ transformed mercury (Hg) concentration and stable nitrogen isotope values (δ^{15}N), hereafter called trophic magnification slope (TMS), from several trophic levels in a food web can represent the overall degree of Hg biomagnification. We compiled data from 69 studies that determined total Hg (THg) or methyl Hg (MeHg) TMS values in 205 aquatic food webs worldwide. Hg TMS values were compared against physicochemical and biological factors hypothesized to affect Hg biomagnification in aquatic systems. Food webs ranged across 1.7 ± 0.7 (mean ± SD) and 1.8 ± 0.8 trophic levels (calculated using δ^{15}N from baseline to top predator) for THg and MeHg, respectively. The average trophic level (based on δ^{15}N) of the upper-trophic-level organisms in the food web was 3.7 ± 0.8 and 3.8 ± 0.8 for THg and MeHg food webs, respectively. For MeHg, the mean TMS value was 0.24 ± 0.08 but varied from 0.08 to 0.53 and was, on average, 1.5 times higher than that for THg with a mean of 0.16 ± 0.11 (range: −0.19 to 0.48). Both THg and MeHg TMS values were significantly and positively correlated with latitude. TMS values in freshwater sites increased with dissolved organic carbon and decreased with total phosphorus and atmospheric Hg deposition. Results suggest that Hg biomagnification through food webs is highest in cold and low productivity systems; however, much of the among-system variability in TMS values remains unexplained. We identify critical data gaps and provide recommendations for future studies that would improve our understanding of global Hg biomagnification.



INTRODUCTION

Humans have altered the natural biogeochemical cycling of the toxic metal mercury (Hg), with coal combustion and gold mining accounting for the majority of atmospheric emissions over the past 150 years.[1] The bulk of contemporary Hg emissions are currently re-emitted legacy anthropogenic emissions.[2] Mercury contamination is a global issue due to its long-distance transport[3] and potential toxicity.[4] Of particular concern is methylmercury (MeHg), which can be converted from inorganic forms of mercury in aquatic ecosystems. Efficient trophic transfer of MeHg through aquatic food webs (i.e., biomagnification) results in Hg concentrations in predator species which can be millions of times higher than those observed in surface waters.[5] Biomagnification of Hg has been consistently observed in freshwater and marine food webs[6−9] and can lead to toxic concentrations in fish and fish-eating wildlife.[10]

Stable nitrogen isotopes (δ^{15}N) are used to characterize trophic relationships in aquatic food webs.[11,12] Increasing δ^{15}N values typically indicate increasing trophic position within a food web as most organisms consistently excrete lighter nitrogen isotopes (^{14}N) and retain heavier isotopes (^{15}N), which results in increasing tissue ratios of ^{15}N/^{14}N through the food web. Since THg and MeHg tend to biomagnify in ecosystems, the simple linear relationship between Hg and δ^{15}N values in organisms within a food web can be used to quantify the degree of biomagnification (i.e., the average change in tissue Hg concentration with trophic position). Biomagnification of Hg is typically calculated by using the following equation:

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Table 1. Summary of Factors Known to Affect Hg Bioaccumulation, Hypotheses about Their Effects on Trophic Magnification Slopes (TMS), and Predicted and Observed Relationships with THg and MeHg Trophic Magnification Slopes (TMS) in Freshwater Sites[a]

controller	hypothesis	measured variable	predicted outcome	outcome[b]	
				THg	MeHg
acidity	greater Hg bioavailability in acidic waters;[10,77] slower growth of biota in acidic waters.[10]	pH[c]	−	ns/ns	ns/+
dissolved organic carbon (DOC)	higher Hg concentrations in higher DOC waters under wetland influence[68] (+) but lower bioavailability,[58] reduced trophic transfer[58,70] and enhancement of demethylation[77] (−).	DOC[c]	+ and −	+/ns	ns/+
		% wetland[c]	+	ns/	ns/
Hg availability	greater dissolved uptake of Hg lower in the food web and reduced biomagnification when dietary concentrations are high.[17−19] intracellular competitive uptake kinetics and regulation mechanisms.[20−22]	Hg in water[c]	−	ns/ns	−/ns
		Hg in sediments[c]	−	ns/	ns/
		Hg deposition[d]	−	ns/−	−/−
		$Hg_{baseline}$[e]	−	ns/ns	ns/−
growth/ productivity	rapid growth causes dilution of Hg in body tissues.[71] high productivity causes biomass dilution of Hg.[25]	latitude[c]	+	+/	+/
		chlorophyll-*a*[c]	−	+/	−/
		total phosphorus[c]	−	ns/−	ns/−
		total nitrogen[c]	−	ns/	ns/
		productivity status[e]	−	ns/	ns/
		phosphorus loading[d]	−	+/+	−/−
		lake and watershed area[c]	+	ns/	ns/
energy efficiency	Hg is retained more readily than biomass; top predator endotherms magnify Hg more readily.[67]	species composition[c]	+	ns/	ns/
		% of endotherms[e]	+	ns/	ns/

[a]TMS values represent individual slopes (b) of simple linear regressions between $\log_{10}$[Hg] and δ^{15}N for several sites worldwide. [b]Not significant (ns). Results of multiple statistical tests are separated by a slash (regression or correlation/multiple linear regression). [c]Extracted from reviewed or parallel studies (in situ physico-chemistry). [d]Extracted by GIS (in silico physico-chemistry). [e]Calculated from food web values provided in reviewed studies (see Methods for calculations).

$$\log_{10}[\mathrm{Hg}] = \delta^{15}\mathrm{N}(b) + a \quad (1)$$

where the slope (b) of this regression, hereafter called the Trophic Magnification Slope (TMS), has been routinely used as an indicator of biomagnifying potential of Hg in food webs since the early 1990s[13] (see also Supporting Information, SI, Table S1). A significant and positive slope (TMS > 0) indicates Hg biomagnification in a food web. This approach has been widely applied to assess Hg biomagnification worldwide (SI Table S1).

It is well established that the concentrations of Hg in fish and invertebrates are positively related to atmospheric Hg deposition,[14,15] but Hg concentrations in biota are also influenced by the intrinsic physical and chemical characteristics of a given ecosystem since they play an important role in Hg bioavailability at the water/primary production interface. For example, low pH, elevated dissolved organic carbon (DOC) and high aqueous Hg concentrations are typically associated with high concentrations of Hg in invertebrates and fish.[10,14,16] However, the influence of these variables on Hg biomagnification in food webs is unclear. It is especially unknown if biomagnification is only affected by food web processes (e.g., growth rate, species diversity and length of the food chain) or if physico-chemistry variables are also modulating Hg biomagnification. For example, elevated concentrations of MeHg in prey may in fact reduce the transfer of Hg to predators[17−19] (e.g., by intracellular competitive uptake kinetics and regulation mechanisms[20−22]). Moreover, ecosystem characteristics and food web processes may interact to influence Hg TMS: low productivity, acidic pH and cold temperatures could indirectly increase Hg TMS by reducing feeding and growth efficiency[23] and lowering excretion rates of MeHg in biota.[24] In addition, higher nutrient concentrations may reduce MeHg at the base of the food web[25] and can lead to dilution of Hg in upper-trophic-level consumers.[26−28] These effects lead to the prediction that ecosystems in equatorial regions, where temperature, primary productivity and growth rates are higher[29,30] and food webs are generally shorter,[31] may exhibit lower Hg TMS compared to those in temperate or polar regions.[32]

Several authors have noted that TMS values often fall within a similar range; however, among-system differences in Hg TMS values occur for reasons that are not yet well understood.[5] In this study, we compiled and queried a global database of the large and growing body of literature on Hg biomagnification to quantify average Hg TMS values (slope (b) in eq 1) from freshwater and marine ecosystems. Our objective was to test hypotheses related to physico-chemistry (e.g., productivity and acidification) as well as food web processes (e.g., growth rate and species composition) that are thought to explain variation in Hg biomagnification (rather than Hg concentration) among ecosystems on a global scale (see Table 1 for a list of hypotheses). We also identified critical data gaps, and methodology and conceptual issues in the literature and provided recommendations for future biomagnification studies that could contribute to our understanding of Hg biomagnification.

MATERIALS AND METHODS

Mercury Biomagnification and Trophic Magnification Factor. In our meta-analysis, the variable of interest was the TMS value, which is the slope (b) of the relationship between logarithm transformed (to the base 10) Hg concentration (THg or MeHg) and δ^{15}N values of biota from several trophic levels

Table 2. Trophic Magnification Slopes (TMS Values, Mean ± SD, *n*) Based on Wet Weight for All Sites Reviewed in This Study[a]

	THg[b]					MeHg[b]				
	mean	±	SD	*n*		mean	±	SD	*n*	
freshwater only										
latitudinal classes										
polar	0.19			1		0.28	±	0.09	24	*a*
temperate	0.16	±	0.10	65		0.24	±	0.07	78	*a*
tropical	0.12	±	0.12	35		0.16	±	0.07	8	*b*
species composition										
fish only	0.16	±	0.13	30		0.28			1	
fish and other species[c]	0.14	±	0.09	68		0.24	±	0.08	106	
other species, no fish[c]	0.19	±	0.10	3		0.31	±	0.10	3	
productivity status based on in situ phosphorus										
hypereutrophic	0.17	±	0.08	4		0.16	±	0.05	3	
eutrophic	0.22	±	0.09	9		0.26	±	0.06	8	
mesotrophic	0.14	±	0.09	11		0.24	±	0.06	19	
oligotrophic	0.13	±	0.12	33		0.24	±	0.07	35	
type of ecosystem										
lentic	0.16	±	0.10	60	*a*	0.23	±	0.08	77	*b*
lotic	0.12	±	0.11	41	*b*	0.27	±	0.08	33	*a*
total for freshwater sites	**0.15**	**±**	**0.11**	**101**	**B/b**	**0.24**	**±**	**0.08**	**110**	**A/a**
marine										
latitudinal classes										
polar	0.21	±	0.07	8		0.21	±	0.09	7	
temperate	0.22	±	0.11	13		0.26	±	0.08	6	
tropical	0.16	±	0.08	5		0.14			1	
species composition										
fish only	0.09	±	0.11	2		0.20	±	0.07	3	
fish and other species[c]	0.21	±	0.09	23		0.24	±	0.09	10	
other species, no fish[c]	0.21			1		0.14			1	
type of ecosystem[d]										
coastal	0.19	±	0.08	14		0.20		0.09	7	
oceanic	0.21	±	0.11	12		0.25		0.08	7	
total for marine sites	**0.20**	**±**	**0.10**	**26**	**A/a**	**0.22**	**±**	**0.09**	**14**	**A/a**
all sites										
latitudinal classes										
polar	0.21	±	0.07	9	*a*	0.26	±	0.10	31	*a*
temperate	0.17	±	0.10	78	*a*	0.24	±	0.07	84	*a*
tropical	0.13	±	0.12	40	*b*	0.15	±	0.07	9	*b*
species composition										
fish only	0.15	±	0.13	32		0.22	±	0.07	4	
fish and other species[c]	0.16	±	0.10	91		0.24	±	0.08	116	
other species, no fish[c]	0.20	±	0.09	4		0.26	±	0.12	4	
total for all sites	**0.16**	**±**	**0.11**	**127**	**B**	**0.24**	**±**	**0.08**	**124**	**A**

[a]TMS values represent individual slopes (*b*) of simple linear regressions between $Log_{10}[Hg]$ and $\delta^{15}N$ for several sites worldwide. [b]Categories that share common letters do not differ significantly: **BOLD CAPITAL** is for comparisons between Hg species (THg vs MeHg), **lower case bold** is for comparison between freshwater and marine sites and *lower case italics* is for comparisons within levels of a factor for a given Hg species. [c]Other species correspond to phytoplankton, invertebrates, reptiles, birds or mammals. [d]Coastal food webs were within 20km from the coast and depths less than 50m.

within a food web (eq 1, SI Figure S1). Higher TMS values indicate greater biomagnification potential within a food web.

Biomagnification can also be quantified using trophic level (TL) in place of $\delta^{15}N$ values in eq 1. In this approach, an organism's raw $\delta^{15}N$ value is converted into TL using the following equation:

$$TL_{consumer} = (\delta^{15}N_{consumer} - \delta^{15}N_{baseline})/\Delta^{15}N + \lambda \quad (2)$$

where λ is the trophic level of the baseline organism (TL = 1 for primary producers and TL = 2 for primary consumers), $TL_{consumer}$ is the trophic level of a given consumer, and $\delta^{15}N_{consumer}$ and $\delta^{15}N_{baseline}$ are $\delta^{15}N$ values of a given consumer and the baseline organism, respectively. A trophic discrimination factor for $\delta^{15}N$ ($\Delta^{15}N$) of 3.4‰ was the most frequently used for aquatic organisms in the reviewed studies (see also refs 33,34) and was therefore chosen for TL calculation.

The trophic magnification factor (TMF), calculated as the antilog of the slope of the relationship between logarithm transformed (to the base 10) Hg concentration and TL, represents the average biomagnification per TL through the entire food web and can be calculated from the slope (*b*) of eq 1 by the following:

$$TMF = 10^{(b*3.4‰)} \quad (3)$$

where 3.4‰ is the trophic discrimination factor for $\delta^{15}N$ ($\Delta^{15}N$). TMF represents the increase of Hg concentration per trophic level (reported in Table S2).

Mercury at the Base of the Food Web. From eq 1, the intercept (a) has been previously considered as an estimate of the concentration of Hg that is incorporated at the base of the food web. However, the intercept value is intrinsically linked to the slope[33] (see SI 1 and Table S3 for the complete procedure to test the validity of using the intercept for this purpose). Instead of the intercept to estimate Hg concentrations at the base of the food chain, we developed three approaches. First, we examined average Hg concentrations of baseline species ($Hg_{baseline}$) designated as primary consumers (e.g., mussels or snails) that are longer-lived and integrate temporal $\delta^{15}N$ variations in the environment[12,35] although other primary consumer taxa such as zooplankton[6] have been used. Baseline organisms were usually specifically characterized in studies and their average Hg and $\delta^{15}N$ values were incorporated in the database. In cases where baseline organisms were not specified, values from a primary consumer in the food web from the observations in the individual study were selected and included in the database. When more than one food web was identified at a given site (e.g., pelagic and benthic), corresponding baselines of analogous taxa were used. Second, we estimated Hg concentration of the baseline organism using the $\delta^{15}N$ value of the baseline organism ($Hg_{\delta^{15}N\text{-}baseline}$) using eq 1 (see SI 1 and Table S3). Third, we estimated Hg at trophic level 2 (Hg_{TL2}) using the slope of the relationship between Hg and TL (when available in studies).

Data Acquisition and Database Preparation. We identified published freshwater and marine studies for this meta-analysis using a Web of Knowledge (Thompson Reuters) search for the following keywords: "mercury", "biomagnification", and "nitrogen isotope" as well as a review of references therein. Biomagnification regression equation parameters as well as average Hg concentrations and $\delta^{15}N$ values for identified baseline organisms were extracted. Authors were contacted to obtain information that was unavailable in the publications and these unpublished data were also incorporated in the global database. In cases where we could not obtain raw data directly from the authors, data were extracted from published figures[36−39] using the program Datagrabber[40] to generate eq 1. For studies that reported equations in another format (e.g., reduced major axis[36]), data were extracted using Datagrabber and were transformed into a consistent format (i.e., simple linear regression). Datagrabber was also used to estimate average $\delta^{15}N$ and Hg values of baseline organisms.[41−43] Data accuracy and precision for the Datagrabber software are presented in SI 2. When no other option was available, regression equations based on average data were included in the meta-analysis.[38,44] Many of the authors of this meta-analysis, as well as colleagues, contributed unpublished data sets (see acknowledgments, SI Table S1 and Figure S2). Results of graduate theses were also included.[45−50] Studies were incorporated in the database, regardless of the significance of the biomagnification regression. Methods for unpublished studies are presented in SI 3.

Hg concentrations were standardized for moisture content (into wet weight) and data transformation (Hg transformed into $\log_{10}$). If Hg concentrations were reported as dry weight, without specific moisture content, then we assumed a value of 75 ± 8% (n = 272),[51] as this was the most frequently reported value in the reviewed studies. Several studies estimated MeHg (rather than measuring it) in specific taxa such as higher-trophic-level fish species by measuring THg and assuming 95 or 100% as MeHg, and then measuring MeHg in lower-trophic-level species (SI Table S1). In such cases, TMS values were considered to be representative of MeHg TMS values.

The 69 studies in the database yielded 205 independent sites with 127 and 124 TMS values and over 7200 and 5400 tissue samples for THg and MeHg, respectively (Table 2). Study sites were distributed worldwide in several types of ecosystems: streams, rivers, wetlands, lakes, estuaries, and oceans (see Table 2 and Table SI for more details on site types and sample sizes), and ranged from low to high input of natural or anthropogenic Hg (SI Figure S3). Site was the unit of replication of TMS values derived from the data sets.

For freshwater sites, TMS values were paired with in situ physicochemical variables (when available) known to influence mercury trophodynamics. Variables of interest were pH, dissolved organic carbon (DOC), Hg in water and Hg in sediments, productivity (chlorophyll-a [Chl-a], total nitrogen and total phosphorus concentrations), lake surface area, watershed area, and percentage of wetland (Table 1). If physicochemical data were not directly available in the food web study, then values from parallel studies and government databases from the same site were used when available. In addition to measured in situ physico-chemistry variables mentioned immediately above, a complementary approach using GIS (ArcGIS Desktop 9.3.1) was applied to obtain data such as Hg deposition as a measure of input of Hg in the system and phosphorus loading as a measure of productivity, as described in SI 4. Atmospheric Hg deposition data were obtained from Environment Canada (A. Dastoor, unpubl. data; SI Figure S3). Phosphorus loading data were obtained from Vörösmarty et al.[52] In this present study, physico-chemistry variables will be referred to as "in situ physico-chemistry variables" when they were measured in the field directly and "in silico physico-chemistry variables" when they were estimated at each site using GIS. A comparison between in situ and in silico physico-chemistry data was done to ensure the validity of the latter for freshwater sites (SI Table S4). Unfortunately, in situ physico-chemistry data were generally not reported for marine sites or could not be obtained through other sources and consequently relationships related to physico-chemistry in marine sites could not be tested.

Data Analyses. Both discrete and continuous statistical tests were used to examine effects and trends in biomagnification for the entire data set and for freshwater sites. Discrete tests included t-tests of differences in THg and MeHg TMS values. Sites were grouped based on latitudinal classes (polar (66.6° to 90° N or S), temperate (23° to 66.6° N or S), tropical (0° to 23° N or S)), type of ecosystem (streams and rivers, lakes, and marine (coastal and open ocean)), species composition (*i*, fish only; *ii*, fish and other taxa; and *iii*, no fish) and productivity status (oligotrophic, mesotrophic, eutrophic, and hypereutrophic, based on total phosphorus values[53]). Those factors were tested using analysis of variances (ANOVA) with TMS values as the dependent variable, followed by a Tukey's test for differences among levels of factors.

For the continuous tests, simple linear regressions and correlations were used to test the relationships between TMS values and physicochemical variables (in situ and in silico). Multiple linear regressions examining the combined influence of different physicochemical variables on TMS were done with model selection by Akaike information criterion (AIC) to determine the best and most parsimonious model.[54] The model with the lowest AIC was selected. Multiple linear regressions were done on groups of variables with standardized (mean = 0,

SD = 1, to remove the effect of scale differences between variables[54]) TMS values as dependent variables and standardized physico-chemistry variables as independent variables. Multicollinearity was tested by variance inflation factor with values below 5 being considered acceptable.[54]

Statistical analyses were performed using version 2.13.2 of the R statistical program.[55] Lilliefors tests were done on residuals after each statistical test to ensure normality of variables. Nonparametric tests were used when assumptions were not met. When possible, statistical analyses were done separately on freshwater sites, marine sites, and all sites combined. However, emphasis was put on freshwater sites in this study because more sites and more physico-chemistry data were available for this type of ecosystem.

RESULTS

Food Web Characteristics. The average (±SD) range of trophic levels included in food webs (trophic level of top predator − trophic level of baseline) based on δ^{15}N (eq 2) was 1.7 ± 0.7 for THg and 1.8 ± 0.8 for MeHg. Trophic level of the upper consumer organism in the food web (eq 2) averaged 3.7 ± 0.8 for THg and 3.8 ± 0.8 for MeHg.

Average Biomagnification of THg and MeHg. Globally, THg TMS values (0.16 ± 0.11) were significantly lower than those based on MeHg (0.24 ± 0.08, Student's *t*-test: $t = 6.9$, $p < 0.001$; Table 2, Figure 1). When freshwater and marine sites were considered separately, freshwater food web THg slopes were still significantly lower than MeHg slopes ($t = 7.3$, $p < 0.001$), but marine sites did not have any significant differences between MeHg and THg TMS values ($t = 0.7$, $p = 0.50$, Figure 1).

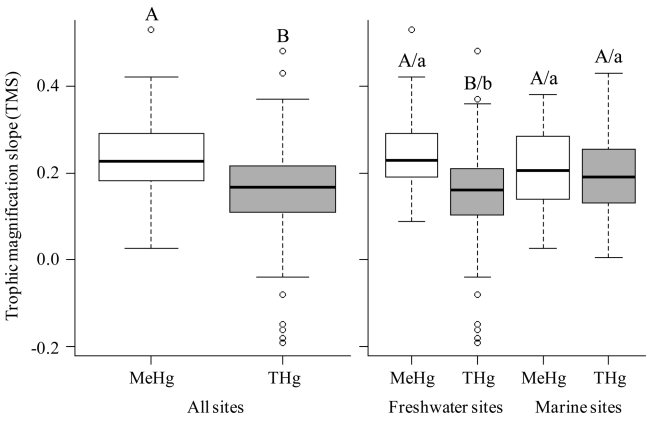

Figure 1. Boxplot of MeHg (white) and THg (gray) trophic magnification slopes (TMS values) based on wet weight for all sites (left panel) and freshwater sites and marine sites (right panel) reviewed in this study. Categories that share common letters do not differ significantly: capital letters are for comparisons between Hg species (THg vs MeHg) and lower case letters are for comparison between freshwater and marine site.

TMS values from marine sites were higher than those from freshwater sites for THg, but not for MeHg (Mann−Whitney−Wilcoxon, THg: $W = 881$, $p = 0.01$, MeHg: $W = 853$, $p = 0.06$, Figure 1). Within freshwater systems, TMS values for lentic ecosystems were higher than those of lotic ecosystems for THg ($W = 1552$, $p = 0.026$), but lower for MeHg ($W = 807$, $p = 0.003$). Within marine systems, there was no difference between coastal and open water sites (THg: $t = -0.5$, $p = 0.618$, MeHg: $t = -1.1$, $p = 0.291$; Table 2).

Effect of Latitude on Hg Biomagnification. THg and MeHg TMS values were both significantly higher in polar and temperate sites when compared to tropical sites (Kruskal−Wallis, THg: $\chi^2 = 7.0$, $p = 0.03$, MeHg: $\chi^2 = 12.1$, $p = 0.002$). Within freshwater sites, higher MeHg TMS values were also found in polar and temperate sites than in tropical sites ($\chi^2 = 12.4$, $p = 0.002$). Regression analyses also found a positive relationship between TMS values and latitude for all sites combined and for freshwater food webs only (Figure 2). In summary, both discrete statistics using latitudinal classes and continuous statistics demonstrate that polar and temperate sites consistently have higher Hg TMS values while tropical sites have among the lowest TMS values.

Effect of Food Web Composition on Hg Biomagnification. THg or MeHg biomagnification was not different for studies that examined food webs composed of fish only, fish and other taxa (phytoplankton, invertebrates, reptiles, birds, or mammals), or nonfish taxa in freshwater sites (ANOVA, THg: $F = 0.5$, $p = 0.59$, MeHg: $F = 1.2$, $p = 0.31$), marine sites (THg: $F = 1.7$, $p = 0.20$, MeHg: $F = 0.7$, $p = 0.52$) or all sites combined (THg: $F = 0.3$, $p = 0.73$, MeHg: $F = 0.3$, $p = 0.75$; Table 2). There was also no relationship between percentage of endotherms in a food web and TMS for freshwater sites (linear regression, THg: $F = 1.5$, $p = 0.22$, MeHg, $F = 1.0$, $p = 0.31$) or marine sites (THg: $F = 0.6$, $p = 0.46$, MeHg, $F = 1.0$, $p = 0.34$).

Effect of in Situ Physico-chemistry on Hg Biomagnification—Freshwater Sites. Using in situ water chemistry data (measured directly in the field at the reviewed study sites), THg and MeHg TMS values in freshwater sites were not affected by productivity status (from oligotrophic to hypereutrophic), categorized using total phosphorus (ANOVA, THg: $F = 1.7$, $p = 0.18$, MeHg: $F = 1.7$, $p = 0.17$, Table 2). When examined as continuous variables, there was a moderate positive correlation between Chl-*a* and THg TMS ($r^2 = 0.18$, $p = 0.010$), but a moderate negative correlation for MeHg TMS ($r^2 = -0.22$, $p = 0.003$; Table 1). In addition, a moderate positive relationship between DOC and slopes was found for THg ($r^2 = 0.14$, $p = 0.014$), but not for MeHg ($r^2 = 0.04$, $p = 0.087$). However, there were no significant relationships between total phosphorus and TMS values (THg: $r^2 = 0.01$, $p = 0.38$, MeHg: $r^2 = -0.02$, $p = 0.26$) or between total nitrogen and TMS values (THg: $r^2 < 0.01$, $p = 0.80$, MeHg: $r^2 < 0.01$, $p = 0.90$, Table 1).

There was a negative relationship between aqueous Hg concentration and MeHg TMS ($r^2 = 0.16$, $p = 0.006$), but not for THg TMS ($r^2 = -0.01$, $p = 0.58$). No other in situ water physico-chemistry variables (Hg in sediments, pH, percentage of wetland, lake area and watershed area) showed any relationship with TMS values (Table 1).

The multiple linear regression model for THg TMS values including pH, total phosphorus and DOC was not significant for the overall model ($F = 3.1$, $R^2 = 0.20$, $p = 0.06$), but total phosphorus showed a negative partial standardized regression coefficient in the model ($b' = -0.24$, $p = 0.035$, SI Table S5). However, 25% of the variation in MeHg slopes was accounted for (in decreasing order of hierarchical partitioning) by pH ($b' = 0.35$), total phosphorus ($b' = -0.32$) and DOC ($b' = 0.27$; overall model: $F = 4.4$, $R^2 = 0.25$, $p < 0.001$, SI Table S5). A multiple linear regression model including $Hg_{baseline}$ and aqueous Hg showed that $Hg_{baseline}$ ($b' = -0.52$) accounted for 21% of the variation of MeHg slopes ($F = 5.6$, $p = 0.007$), whereas aqueous Hg concentration was not significant ($b' = -0.27$, $p = 0.18$). $Hg_{baseline}$ and aqueous Hg concentration had no effect on THg slopes ($F = 0.3$, $R^2 = 0.04$, $p = 0.76$).

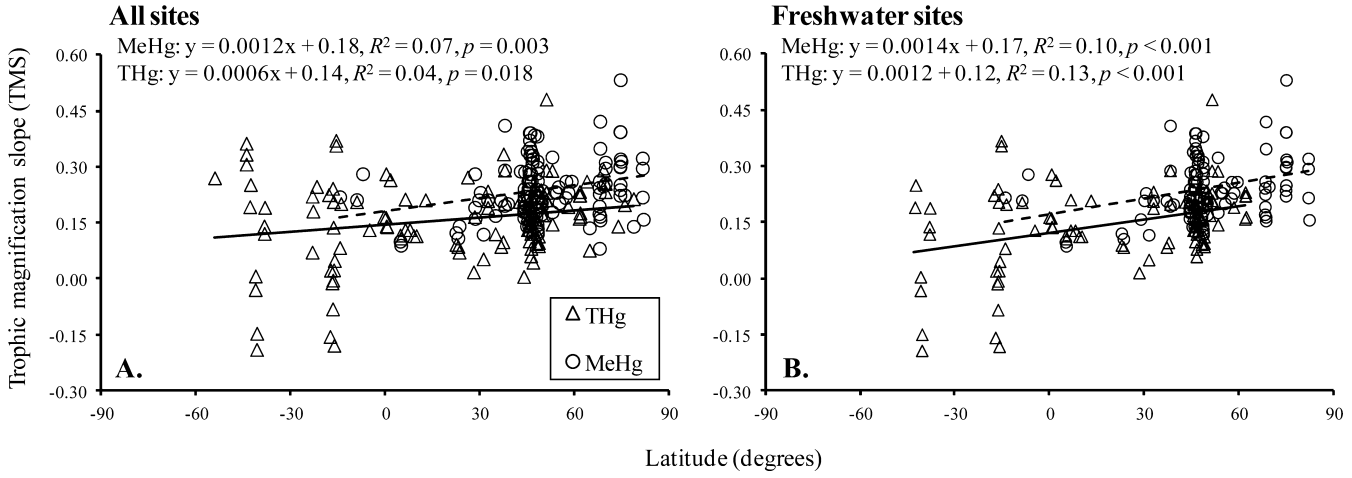

Figure 2. Relationships between THg (triangles) and MeHg (circles) trophic magnification slopes (TMS) against latitude for all sites (A) and freshwater sites (B). TMS values represent individual slopes (b) of simple linear regressions between $\log_{10}$[Hg] and δ^{15}N for several sites worldwide.

Effect of in Silico Physico-chemistry on Hg Biomagnification—Freshwater Sites. MeHg TMS values in freshwater sites showed a significant negative relationship with GIS-derived (in silico) atmospheric Hg deposition (A. Dastoor, Environment Canada, unpubl. data, linear regression, F = 12.9, R^2 = 0.11, p < 0.001), but it was not the case for THg TMS values (F = 2.4, R^2 = 0.01, p = 0.12, Table 1).

Freshwater THg TMS values demonstrated a positive significant relationship with in silico (GIS-derived) phosphorus loading (F = 6.9, R^2 = 0.07, p = 0.01), whereas MeHg TMS values were negatively related to phosphorus loading (F = 11.7, R^2 = 0.10, p < 0.001, Table 1).

In a multiple linear regression model, THg TMS values were explained by Hg deposition (b' = −0.26) and phosphorus loading (b' = 0.33) with an overall model accounting for 13% of the variation (F = 7.4, p = 0.001; SI Table S5). MeHg slopes were also explained by Hg deposition (b' = −0.19) and phosphorus loading (b' = −0.20) with an overall model accounting for 12% of the variation (F = 7.5, p < 0.001).

DISCUSSION

Biomagnification of THg and MeHg. Over the past three decades, many studies have used δ^{15}N to estimate Hg biomagnification, providing the opportunity for a comprehensive literature review of factors affecting Hg biomagnification in aquatic systems. We found that biomagnification was highly variable among all reviewed sites. When all sites were combined, the average (±SD) TMS for THg and MeHg were 0.16 ± 0.11 and 0.24 ± 0.08, respectively. These TMS values can be converted to TMF values using eq 3 (see SI Table S2 for a complete list of TMF values) and indicate that THg and MeHg increase by a factor of 4.7 ± 4.7 and 8.1 ± 7.2 per trophic level, respectively. The average TMS value for freshwater sites was 0.15 ± 0.11 (TMF = 4.3 ± 4.8) and 0.24 ± 0.08 (TMF = 8.3 ± 7.5) for THg and MeHg, respectively. For marine sites, the average TMS was 0.20 ± 0.10 (TMF = 6.2 ± 4.1) and 0.22 ± 0.09 (TMF = 7.0 ± 4.9) for THg and MeHg, respectively.

Despite intensive comparative studies on Hg biomagnification conducted at a regional scale,[7,18,49,56−63] no general consensus has emerged regarding the main variables affecting Hg biomagnification in aquatic ecosystems. Here, we present a synthesis of Hg biomagnification in aquatic food webs on a global scale and explore physical, chemical and biological factors that could explain the observed variability in TMS values.

Latitude and Hg Biomagnification. To the best of our knowledge, this is the first study to test and show that global biomagnification of THg or MeHg was positively related to latitude. Several mechanisms related to temperature may explain these latitudinal trends. First, warmer temperatures stimulate growth rates in aquatic organisms which in turn decreases the amount of Hg per unit of body mass (growth biodilution) as opposed to colder temperatures where growth rate is suppressed.[26] In warmer regions, trophic transfer efficiency of Hg would be reduced at every trophic step therefore reducing biomagnification (TMS). Second, colder temperatures lead to slower excretion rates of MeHg resulting in a higher accumulation in organisms.[24] It is expected that higher accumulation would affect all organisms along the food web, albeit not equally, and would have repercussions on the overall TMS value. Third, we hypothesize that simpler food webs at higher latitudes,[64] as characterized by a low species diversity,[65] could lead to higher Hg biomagnification than in more complex and more diverse food webs at lower latitudes. With Hg increasing exponentially with trophic levels, a slight change in dietary trophic level along the food chain would have implications for Hg concentrations and therefore on the overall TMS. Hence, high diversity (large choice of prey for a given consumer) could potentially reduce efficiency of Hg trophic transfer.

The latitudinal relationship with Hg biomagnification found in our review is likely conservative because lower δ^{15}N trophic discrimination factors (Δ^{15}N) may occur in the tropics[66] and should, in principle, lead to higher TMS values in that region compared to temperate or polar latitudes (see SI 5, Table S6 and Figure S4). On the contrary, our results show lower TMS in the tropics despite lower Δ^{15}N. These data therefore indicate that latitude plays a role in Hg biomagnification, although the mechanism remains unclear. Additional studies specifically testing the effect of food web composition or structure and temperature on Hg biomagnification are needed to address the mechanism responsible for the observed latitudinal effect.

Although latitude was linked to TMS in our study, there are likely other drivers than only temperature. Indeed, latitude was correlated with several physico-chemistry variables among THg sites (such as DOC, Chl-*a*, and total phosphorus) and among

MeHg sites (pH, DOC, Chl-*a*, total phosphorus, phosphorus loading, aqueous Hg, sediment Hg and Hg deposition, SI Table S4). It is therefore likely that several variables are acting in synergy to cause the observed latitudinal trends. It is especially hard to dissociate ecosystem productivity variables (e.g., total phosphorus and Chl-*a*) from each other and from latitude as they are not independent and are driven by temperature. Variables related to ecosystem productivity following a latitudinal gradient could also explain the observed trend in TMS values and are discussed below.

Physiology and Hg Biomagnification. Hg biomagnification was expected to be influenced by the energy requirement of organisms included in a given food web. Endotherms have higher energy requirements and lower food conversion efficiencies than ectotherms; thus, they have a higher potential intake of Hg (higher weight-specific metabolic rate) compared to ectotherms. Inclusion of endotherms in food webs was hypothesized to increase Hg biomagnification slopes. However, neither the species composition nor the percentage of endotherms in food webs affected TMS values. This is in contrast to a study by Hallanger et al.[67] who showed that marine food webs including endotherms have higher biomagnification of persistent organic pollutants than those excluding endotherms.

Physico-chemistry and Hg Biomagnification. It is well established that Hg concentrations in aquatic organisms are affected by the physicochemical characteristics of the systems they inhabit.[10,14,16,58] The focus of our study, however, was on determining if Hg biomagnification (rather than Hg concentrations) was affected by those variables. Herein several physico-chemistry variables were related to TMS values and relationships were most consistent (same direction of outcome for both THg and MeHg TMS in Table 1) for DOC (+), Hg deposition (−) and total phosphorus (−). Most of these trends agree with known relationships of these variables with Hg bioaccumulation in biota,[10,17,25] whereas contradictory results were found for DOC.

While DOC binds MeHg and thereby increases the transport of MeHg from wetlands to lakes,[68] DOC can also reduce trophic transfer of Hg from one organism to another,[10,69,70] with lower bioaccumulation in lakes with high DOC.[58,71] Although Rolfhus et al.[58] found that water MeHg concentrations increased with aqueous DOC concentrations across multiple lakes, bioaccumulation factors (BAF = Hg in biota/Hg in water) decreased while TMS values remained constant, suggesting that Hg in upper trophic levels was primarily defined at the base of the food web in those lakes.[58] Dittman and Driscoll[71] also found a negative correlation between DOC and BAF suggesting that DOC could limit uptake of Hg by organisms. Our study showed positive relationships between TMS values and DOC. Contradictory relationships between previous studies and our current meta-analysis reflect the complexity of DOC as a predictor of Hg in aquatic organisms and could suggest a role for DOC effects on higher-trophic-level organisms.

Although higher Hg concentrations at the base of a given system (Hg deposition, aqueous Hg, $Hg_{baseline}$) cause higher concentrations in aquatic organisms,[14,72−74] TMS values were expected to decrease when basal inputs increase.[17−19] This Hg accumulation paradox may be due to competitive uptake kinetics (with other elements) and regulation mechanisms at the cellular level of organisms.[20−22] Our study showed that Hg biomagnification was indeed higher in systems with lower Hg deposition (as modeled by GRAHM, A. Dastoor, Environment Canada, unpubl. data) and therefore with less Hg available in the system. Moreover, because Hg deposition was positively correlated to $MeHg_{baseline}$ (r^2 = 0.05, p = 0.025), this suggests that prey concentrations control Hg biomagnification as has been demonstrated in laboratory studies[17] and for predator−prey relationships in rivers.[18] Consequently, systems with increased Hg loading may not exhibit a proportional response in concentrations in top predators because of lower TMS values in systems with higher $Hg_{baseline}$. An alternative explanation for MeHg is that latitude (being negatively correlated with Hg deposition at MeHg sites, SI Table S4) has the highest impact on TMS and that low Hg deposition coincides with high latitudes where high MeHg TMS are found. That explanation would not be valid for THg sites however since there was no correlation between latitude and Hg deposition.

Increased system productivity, in the form of algal blooms, can reduce the uptake of MeHg by higher-trophic-level organisms such as zooplankton[25] because the pool of Hg is diluted by a larger amount of biomass, therefore reducing concentrations in predators. In our study, we found negative relationships with phosphorus consistent with previous studies.[25] In contrast, Kidd et al.[7] found that biomagnification slopes were positively related to total phosphorus in 14 Canadian lakes. With the results of our study, we believe that biomass dilution (i.e., due to high productivity[25]) could reduce Hg uptake in lower trophic levels, but more importantly, high primary productivity could increase growth rate of higher-trophic-level species[26,28,75] (i.e., due to better food quality[27]) therefore reducing the overall TMS.

There were contradictory relationships between THg and MeHg TMS with other physicochemical variables such as Chl-*a*, pH, phosphorus loading, and Hg concentration in water. Unfortunately, since studies did not systematically report all in situ physico-chemistry variables, we could only examine pairwise relationships or a limited number of combined variables (i.e., multiple linear regressions, SI Table S5). Other considerations of the studies in our review are the different methods used to measure a specific in situ physicochemical variable, and that most existing data were from single measurements that may not be representative of a given system. It is therefore possible that some of those variables have an impact on Hg biomagnification, but our incomplete data set could not detect it.

Although studies that tested the effect of specific physico-chemistry variables on Hg concentrations in biota across multiple sites have found significant relationships,[10,14,16,58] those linking physico-chemistry to Hg biomagnification across multiple sites have not been as conclusive.[18,59,61,63] This may stem from insufficient statistical power in those studies to detect differences in biomagnification slopes. For example, a power analysis for THg TMS in freshwater sites vs latitude (Figure 2b) showed that 46 sites would be required to detect a significant effect (power = 0.8, effect size (R^2) = 0.13, well above the typical number of sites examined in a given study. This further highlights the important role of meta-analyses in evaluating Hg biomagnification trends and in generating new testable hypotheses.

In this present study, we found that MeHg biomagnified more efficiently than THg, and this was expected since MeHg is the Hg species known to bioaccumulate and biomagnify.[10] We also found that biomagnification increased with latitude and this phenomenon was likely due to a combination of interdependent variables related to temperature. The most plausible explanation is the combined effect of dilution due to growth rate and biomass dilution in highly productive, warm systems that could decrease the trophic transfer of Hg in aquatic food webs. In addition, concentrations of Hg in a system (as measured by Hg

deposition) could either act as a suppressor to biomagnification (Hg accumulation paradox[20−22]) or is simply the result of latitudinal trends with TMS being stronger than Hg deposition. Since physico-chemistry data were not systematically reported among the reviewed studies, we were unable to reach strong conclusions regarding specific processes and mechanisms regarding some of those variables on Hg biomagnification. We therefore urge for more studies that test specific sets of hypotheses on a large number of sites.

Recommendations for Future Studies. On the basis of our observations during this meta-analysis, we recommend following the guidelines for measuring biomagnification of contaminants in food webs provided by Borgå et al.[33] (see also SI 6) plus these additional recommendations for future studies of Hg biomagnification (see also SI 7):

- Examine a large number of sites that vary in specific physical and chemical characteristics (e.g., pH, DOC, Hg in water and productivity) to better understand the effects of ecosystem context on Hg biomagnification and to determine factors affecting TMS. As stated above, a power analysis should be done to determine the appropriate number of sites.
- Adequately characterize baselines of food webs using a representative primary consumer (trophic level = 2) species. We suggest using this baseline to interpret Hg at the base of the food web ($Hg_{baseline}$) and avoid erroneous interpretation of the Hg vs $\delta^{15}N$ intercept. We also suggest calculating and reporting the metrics described in our study, namely $Hg_{\delta^{15}N\text{-baseline}}$ and Hg_{TL2} (see Materials and Methods and SI 1).
- Report slope and intercept error estimates (e.g., confidence interval) and effect size (R^2) of the Hg vs $\delta^{15}N$ relationship as well as significance levels. In addition, trophic level should be calculated and the Hg vs TL relationship should be reported.
- Calculate regression models for each independent food web after confirming that species are operationally defined by dietary dependence among trophic levels[63] (e.g., pelagic food web vs benthic food web). Organisms from disconnected systems (e.g., different lakes) or those that are migratory or not linked to similar carbon sources should not be pooled together.
- Measure the entry of both THg and MeHg into the food chain at the water/primary production interface (e.g., bioaccumulation factor: BAF = Hg in biota/Hg in water[10] and biota-sediment accumulation factor: BSAF = Hg in biota/Hg in sediment[76]).

By following these recommendations, it should be possible to describe some of the currently unexplained variability in TMS and improve our understanding of the global cycling of Hg through food webs.

■ ASSOCIATED CONTENT

Supporting Information

Testing the validity of the equation intercept as an indicator of mercury concentration at the base of the food web (SI 1); quality assurance and quality control on the use of Datagrabber (SI 2); methods for unpublished results (SI 3); physico-chemistry data using GIS (SI 4); effect of $\delta^{15}N$ trophic dicrimination factor difference with latitude on TMS (SI 5); summary of guidelines for mercury studies (SI 6); and other recommendations for future studies (SI 7), including additional method descriptions, results, discussions, data tables, and figures. This information is available free of charge via the Internet at http://pubs.acs.org.

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Notes

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