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Relating Fish Hg to Variations in Sediment Hg, Climate and Atmospheric Deposition

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Abstract

This article addresses total fish Hg concentrations (THg) by variations in lake Sediment THg, atmospheric Hg deposition ($atmHg_{dep}$), and climate, *i.e.*, mean annual precipitation (ppt) and air temperature. The Fish THg data were taken from the 1967-to-2010 Fish Mercury Datalayer (FIMDAC). This compilation was standardized for 12-cm long Yellow Perch in accordance with the USGS National Descriptive Model for Mercury in Fish (NDMMF [1]), and documents Fish THg across 1936 non-contaminated lakes in Canada. About 40% of the standardized Fish THg variations related positively to increasing ppt and Sediment THg, but negatively to increasing mean annual July temperature (T_{July}). Only 20% of the Fish THg variations related positively to $atmHg_{dep}$ alone. Increasing T_{July} likely influences Fish Hg through increased lake and upslope Hg volatilization, in-fish growth dilution, and temperature-induced demethylization. FIMDAC Fish THg effectively did not change over time while $atmHg_{dep}$ decreased. Similarly, the above Fish Hg trends would likely not change much based on projecting the above observations into the future using current 2070 climate-change projections across Canada and the continental US. Regionally, the projected changes in Fish Hg would mostly increase with increasing ppt. Additional not-yet mapped increases are expected to occur in subarctic regions subject to increasing permafrost decline. Locally, Fish THg would continue to be affected by upwind and upslope pollution sources, and by lake-by-lake changes in water aeration and rates of lake-water inversions.

Keywords

Total Hg Concentrations, Fish, Sediments, Atmospheric Hg Deposition, Precipitation, Air Temperature, Climate-Change Projections, North America

1. Introduction

There is much uncertainty about how climate and atmospheric Hg deposition affect Fish THg [2] [3] [4] [5]. In part, this uncertainty is due to large and yet to be quantified land-to-lake and in-lake variations by which Hg uptake by fish increases or decreases. The primary concern is that the bioaccumulation of toxic methyl Hg (MeHg) in fish and other aquatic organisms may increase with increasing atmospheric Hg deposition and climate warming. In principle, the presence and accumulation of Hg and MeHg in fish and sediments starts with the sequestration of atmospheric Hg deposition on land and water, and with the release of surface-exposed geogenic Hg minerals. A part of land-retained Hg is gradually released into streams and lakes through 1) direct Hg-containing litter inputs (detritus), 2) upslope soil and stream bank erosion, and 3) transfer of Hg bound by water-carried particulate and dissolved Hg-containing matter [6] [7] [8] [9]. With regard to detrital Hg inputs, tree foliage, twigs, branches, bark, and wood generally have lower Hg concentrations than mosses, fungi, and lichens [10] [11]. In soils, THg decreases from the organic litter layers on the surface to the subsoil layers below [10]. In downstream, the total amount of Hg generally increases with increasing organic matter transfer [12], which in turn leads to increasing Hg accumulations in stream and lake sediments [13] [14] [15] [16]. Due to particulate Hg retention in wet areas and wetlands, first-order stream and lake sediments downslope from forested and wetland dominated watersheds have higher THg and organic matter contents than elsewhere [17]-[22].

Within lakes, Hg accumulation in sediments and trophic Hg bioaccumulation depend on many physical and chemical conditions and their combined effects on biological processes [17] [23] [24] [25] [26] [27]. For example, physical and chemical properties of water (e.g., temperature, aeration, pH, color, organic matter, and suspended mineralcontent) all influence the fate of Hg with regard to biological uptake, re-precipitation and settling, methylation, demethylation, and volatilization [28] [29] [30]. From a simplifying perspective, total Hg concentrations in water, sediments andaquatic organism are co-variants [13] [22] [31] [32] [33].

This article focuses on analyzing the extent to which standardized data for total Hg concentrations (THg) in fish—as compiled within the Fish Mercury Datalayer (FIMDAC [1])—co-vary with lake Sediment THg, atmospheric Hg deposition (atmHg$_{dep}$), mean annual precipitation (ppt), and mean annual air temperatures for January (winter, T$_{Jan}$) and July (summer, T$_{July}$). This analysis was enabled by cross-referencing the Fish THg data to the modelled and mapped Sediment THg, atmHg$_{dep}$, ppt, T$_{Jan}$ and T$_{July}$ variations across Canada, with special reference to potential climate-induced changes up to 2070. The hypothesis is that at least some of the Fish THg variations can be quantified in this way for tentative trend mapping across Canada, the continental USA, and over time. Data pertaining to lakes downwind and downslope from major anthropogenic

Hg release locations (e.g. [34]) are not part of this analysis.

2. Methods

The 1967 to 2010 FIMDAC Fish THg data (reported in ng·g^{-1}, wet-weight) were compiled for 1936 pristine lakes across Canada from 1967 to 2010. These data were standardized for one fish species (Yellow Perch) and one fork length (12 cm), done in accordance with USGS National Descriptive Model for Mercury in Fish (NDMMF [1] [35] [36] [37]). Fish sampling per lake varied from one time to multiple times. Total sample size used for the analysis below was n = 3179. Fish THg data from water bodies other than lakes were not included; also excluded: Fish THg ≤ 1 ug/g.

The data for lake Sediment THg (in ng·g^{-1}) were obtained from the open geochemical survey files of the Geological Survey of Canada [38] [39], and were compiled for the provinces of Quebec and Nova Scotia as well [40]. This compilation produced 147,910 THg one-time sampling points per lake, all based on 30-cm deep sediment cores collected, dried, sieved, and analyzed from 1960 to 2008. The resulting Sediment THg data were averaged per National Topographic System (NTS) tile (1:50,000 scale [41]).

The data layers for atmHg$_{dep}$, ppt, T$_{Jan}$ and T$_{July}$ (shown and contoured in **Figure 1**) were obtained as follows:

1) total net wet and dry atmHg$_{dep}$, in μg·m^{-2}·a^{-1}, from the Global/Regional Atmospheric Heavy Metals Model (GRAHM), at 25×25 km^2 grid resolution, Canada only [42].

2) ppt (in m), and T$_{July}$ and T$_{Jan}$ (in °C) for Canada and the USA, from the Coupled Model Intercomparison Project (CMIP5 [43]), Scenario rcp8.5, downloaded for 2011 and 2070 (http://climate-scenarios.canada.ca/?page=download-intro).

With ArcMap, cross-referencing was done by data-layer extracting the lake Sediment THg, atmHg$_{dep}$, ppt, T$_{Jan}$ and T$_{July}$ values for each FIMDAC Fish THg location, row-by-row. This was followed by multivariate regression and factor analysis, with Fish THg, Sediment THg, atmHg$_{dep}$, ppt, T$_{July}$ and T$_{Jan}$ as variables.

3. Results

Table 1 provides a basic statistical summary of the variables used in this article. Most notably, these variables differ across Canada in range, such that the lake-by-lake entries for Fish THg have a wider maximum to minimum ratio (*i.e.* 38) than the corresponding model-derived entries for Sediment THg (*i.e.* 15) and for atmHg$_{dep}$ (*i.e.* 12) For mean annual precipitation, the maximum/minimum ratio is 20. Mean annual T$_{Jan}$ is about three times more variable than mean annual T$_{July}$. The longitudinal range is 4 times larger than the latitudinal range.

Using Sediment THg as dependent variable, and atmHg$_{dep}$, ppt, T$_{Jan}$ and T$_{July}$ as independent variables produced:
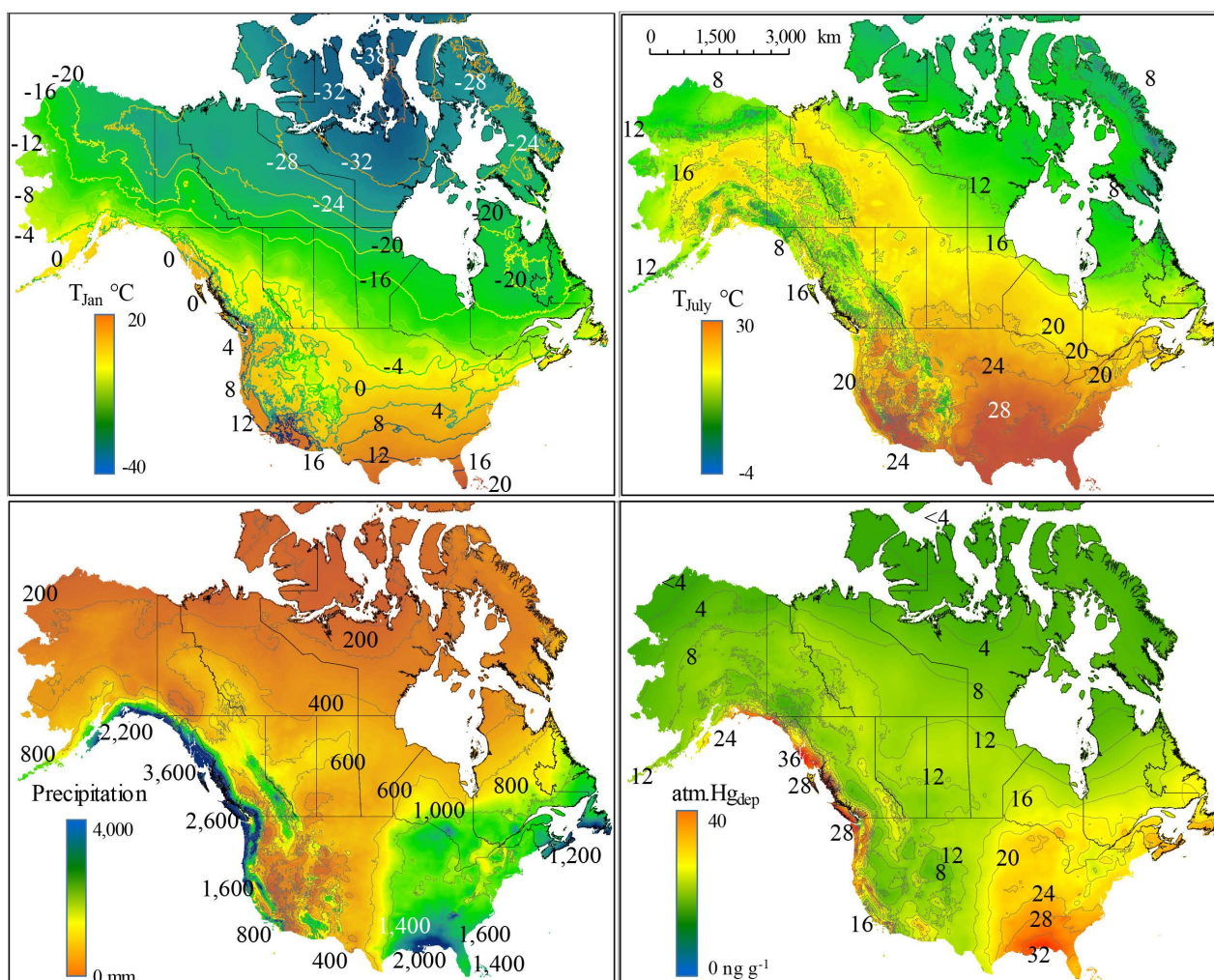
Figure 1. Mean annual January and July air temperature (T_{Jan}, T_{July}) and mean annual precipitation (ppt) according to the Coupled Model Intercomparison Project (CMIP5-rcp8.5) at http://climate-scenarios.canada.ca/?page=download-intro with contours. Also shown: mean annual net atmospheric Hg deposition (atmHg$_{dep}$), based on the Global/Regional Atmospheric Heavy Metals Model (GRAHM2005). Extent: across Canada (provinces and northern territories outlined except most northern parts) and the continental USA.

Table 1. Basic statistics concerning the variables in Equations (1) to (8); Fish THg > 10 ppb; n = 3179.

Variable	Units	Mean	Std. Dev.	Minimum	Maximum
Fish $\log_{10}$THg	$\log_{10}$ (ng·g^{-1}), wet weight	1.81	0.30	1.30	2.88
Lake $\log_{10}$THg (modelled)	$\log_{10}$ (ng·g^{-1}), dry weight	1.76	0.15	1.54	2.72
Precipitation	m·a^{-1}	0.65	0.30	0.14	2.81
atmHg$_{dep}$	µg·m^{-2}·a^{-1}	11.3	4.5	2.5	31.1
T_{July}	˚C	16.6	1.8	6.2	20.9
T_{Jan}	˚C	−19.6	5.8	−33.0	4.6
Latitude	Degrees	53.4	4.6	43.9	69.4
Longitude	Degrees	−93.9	17.0	−138.7	−53.8

$$\text{Sediment } \log_{10} \text{THg}$$
$$= (1.380 \pm 0.048) + (0.406 \pm 0.059) \text{ppt} \tag{1}$$
$$+ (0.012 \pm 0.004) \text{atmHg}_{\text{dep}}, \quad R^2 = 0.432$$

with ppt and atmTHg$_{\text{dep}}$ as significant predictor variables.

Regressing Fish $\log_{10}$THg versus Sediment THg, atmHg$_{\text{dep}}$, ppt, T$_{\text{July}}$, and T$_{\text{Jan}}$ per lakegenerated the following regression equation:

$$\text{Fish } \log_{10} \text{THg}$$
$$= (1.32 \pm 0.02) \text{Sediment } \log_{10} \text{THg} - (0.031 \pm 0.002) \text{T}_{\text{July}}, \quad R^2 = 0.382 \tag{2}$$

By climate variables alone, the following was obtained:

$$\text{Fish } \log_{10} \text{THg}$$
$$= (1.54 \pm 0.07) + (0.77 \pm 0.02) \text{ppt} - (0.022 \pm 0.003) \text{T}_{\text{July}} \tag{3}$$
$$- (0.007 \pm 0.001) \text{T}_{\text{Jan}}, \quad R^2 = 0.385$$

Using atmHg$_{\text{dep}}$ as only predictor accounted for only one-half of the Equation (2) and (3) captured fish $\log_{10}$THg variations:

$$\text{Fish } \log_{10} \text{THg} = (1.47 \pm 0.01) + (0.030 \pm 0.001) \text{atmHg}_{\text{dep}}, \quad R^2 = 0.206 \tag{4}$$

Using Sediment THg and ppt each as only fish $\log_{10}$THg predictors yielded:

$$\text{Fish } \log_{10} \text{THg} = (1.02 \pm 0.03) \text{Sediment } \log_{10} \text{THg}, \quad R^2 = 0.342 \tag{5}$$

and

$$\text{Fish } \log_{10} \text{THg} = (1.42 \pm 0.01) + (0.60 \pm 0.01) \text{ppt}, \quad R^2 = 0.354 \tag{6}$$

thereby accounting for about 90% of the variations captured by Equations (2) and (3). Using atmHg$_{\text{dep}}$, T$_{\text{July}}$ and T$_{\text{Jan}}$ as predictor variables produced

$$\text{Fish } \log_{10} \text{THg}$$
$$= (2.67 \pm 0.06) + (0.025 \pm 0.001) \text{atmHg}_{\text{dep}} \tag{7}$$
$$- (0.050 \pm 0.003) \text{T}_{\text{July}} + (0.016 \pm 0.01) \text{T}_{\text{Jan}}, \quad R^2 = 0.303$$

Hence, using atmHg$_{\text{dep}}$ together with T$_{\text{July}}$ and T$_{\text{Jan}}$ improved the results, but using Sediment $\log_{10}$THg or ppt with and without T$_{\text{July}}$ as additional predictor variables was more effective in capturing more of the Fish $\log_{10}$THg variations (Equations (2), (3), and (6), (7)). In noting that 80% of the GRAHM2005-modelled atmHg$_{\text{dep}}$ variations can also be attributed to climatic and geographic variations via Equation (8), *i.e.*,

$$\text{atmHg}_{\text{dep}} = -(24.7 \pm 0.8) + (26.8 \pm 1.0) \text{ppt}^{0.5}$$
$$+ (0.80 \pm 0.06) \text{T}_{\text{July}} (0.25 \pm 0.03) \text{T}_{\text{Jan}} - (7.7 \pm 0.7) \text{Pacific Rim} \tag{8}$$
$$+ (7.5 \pm 1.0) \text{High Arctic Coastal Areas}, \quad R^2 = 0.803$$

it follows that climate and geography affect Fish THg, Sediment THg and atmospheric Hg in quantitatively and quantitatively different ways. For example, the positive effect of increasing ppt on atmHg$_{\text{dep}}$ levels off with increasing ppt. The T$_{\text{July}}$ effect on atmHg$_{\text{dep}}$ is most likely a surrogate for the geographic Hg

emission and subsequent Hg deposition pattern, which is highest along the southeastern USA-Canada border, and least for the snow- and ice-covered alpine and arctic areas. T_{Jan} likely accounts for increased Hg volatilities from snow-and ice-covered surfaces.

The location-specific adjustments for the Pacific Rim and High-Arctic coastal areas in Equation (8) can be used to compensate 1) for the ppt-induced $atmHg_{dep}$ dilution, and 2) for the T_{Jan}-induced underestimation for $atmHg_{dep}$ along the arctic coastlines, where oceanic Hg upwelling contributes to land-based and aquatic Hg sequestration and bioaccumulation [44] [45]. With respect to Fish THg, increases in ppt and Sediment THg affect Fish $log_{10}THg$ in direct proportions. Checking the correlations among the Equation (1) to (8) variables reveals that T_{July} only has a weak direct effect on Fish $log_{10}THg$ (**Table 2**). Nevertheless, factor analyzing this matrix revealed that T_{July} is an important negative co-variant of Fish $log_{10}THg$ as per factor 2 in **Table 3**.

Projecting the Equation (2) results across Canada and overlaying the Fish THg data produced the map in **Figure 2** and the corresponding map-to-data residual conformance plot in **Figure 3**. According to **Figure 3**, about 80% of the standardized Fish THg variations conform to the Equation (2) projections within a factor of two, 8 times out of 10. In comparison, about 90% of the recorded lake Sediment THg variations conform to the Equation (1) projections within a factor of two, 8 times out of 10. This indicates that Fish THg is somewhat less predictable than Sediment THg, likely due to lake-specific factors that affect Fish Hg uptake and retention, such as, e.g., lake-to-lake variations in biogeochemical composition and trophic Hg uptake dynamics [46] [47].

**Table 2.** Correlation coefficients for the variables in Equations (1) to (6).

Dependent variable	Independent variable	Correlation coefficient	95% Lower	95% Upper	P-Value
	Lake $log_{10}THg$	0.588	0.565	0.611	<0.0001
	T_{July}	−0.035	−0.070	0.000	0.049
Fish $log_{10}THg$	$T_{January}$	0.382	0.352	0.412	<0.0001
	$atmHg_{dep}$	0.454	0.426	0.481	<0.0001
	Precipitation	0.595	0.572	0.617	<0.0001
	T_{July}	0.251	0.218	0.283	<0.0001
Lake $log_{10}THg$	$T_{January}$	0.764	0.750	0.779	<0.0001
	$atmHg_{dep}$	0.847	0.837	0.857	<.0001
	Precipitation	0.984	0.983	0.985	<0.0001
	$T_{January}$	0.462	0.434	0.489	<0.0001
T_{July}	$atmHg_{dep}$	0.318	0.287	0.349	<0.0001
	Precipitation	0.211	0.178	0.244	<0.0001
	$atmHg_{dep}$	0.545	0.521	0.569	<0.0001
T_{jan}	Precipitation	0.788	0.775	0.801	<0.0001
$atmHg_{dep}$	Precipitation	0.744	0.728	0.759	<0.0001

Table 3. Factor analysis of the correlations among the Equations (1) to (6) variables, with additional reference to latitude and longitude.

Variable	Factor 1	Factor 2	Communality
Fish $\log_{10}$THg	**0.64**	**0.47**	0.63
Lake $\log_{10}$THg	**0.97**	0.09	0.95
Precipitation	**0.95**	0.11	0.92
atmHg$_{dep}$	**0.84**	0.00	0.70
T_{jan}	**0.81**	−0.32	0.76
T_{July}	0.38	**−0.86**	0.88
Latitude	**−0.92**	0.25	0.91
Longitude	**0.80**	0.40	0.80
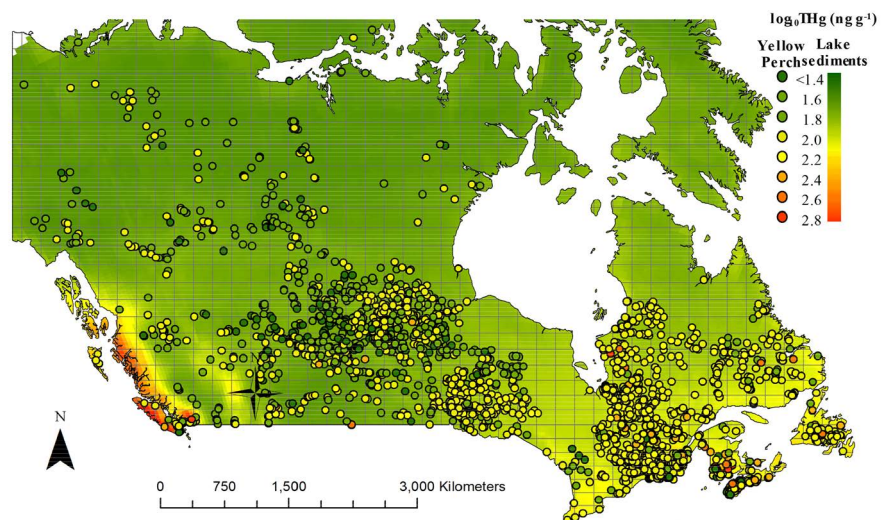
Figure 2. Overlaying FIMDAC locations and standardized Fish THg concentrations (Yellow Perch, wet weight basis, 12-cm long) on the corresponding Equation (2) generated map, using map-projected lake Sediment THg (ng·g^{-1}) and mean annual July temperature (°C) as only projection variables. Also shown: NTS tile grid.

Best-fitted absolute residuals for fish and lake sediment $\log_{10}$THg (ng g^{-1})
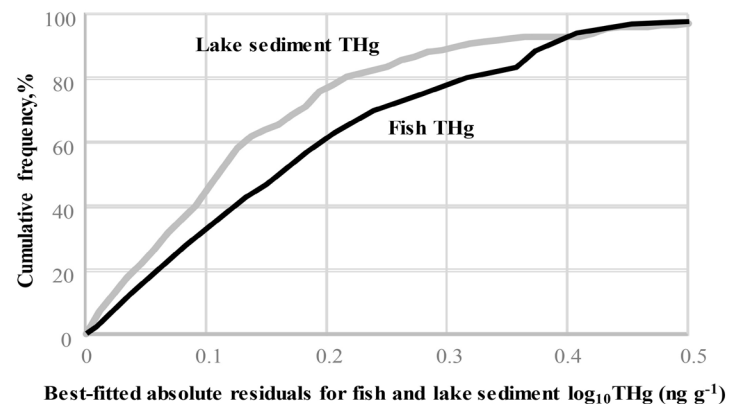
Figure 3. Standardized fish and lake sediment $\log_{10}$THg map-to-data conformance plot based on Equations (1) and (2); **x-axis:** best-fitted absolute residuals for fish and lake sediment $\log_{10}$THg (ng·g^{-1}); **y-axis:** cumulative frequency of the $\log_{10}$THg residuals.

Using Equations (1), (2) and (8) produced the Sediment THg, Fish THg, and atmHg$_{dep}$ projections in **Figure 4** across all of Canada and the continental USA for current conditions. To what extent these projections require further data calibrations needs to be determined. This includes analyzing forthcoming total dry atmospheric Hg deposition and Hg in forest litter data and maps from the Mercury Deposition Network (MDN; http://nadp.sws.uiuc.edu/mdn/). At this stage, the Equations (8)—projected pattern in **Figure 4** resembles USA maps for wet Hg deposition in general, with highest Hg deposition occurring along the south-eastern States, low deposition rates for the mid-eastern States, and increased deposition along the southern Pacific coast of Alaska. Currently available data on dry deposition [48] [49] increase the wet deposition pattern by a factor of 2, as is the case for the GRAHM-based net atmTH$_{dep}$ projections via Equations (8). This was also the case for the earlier North American Hg deposition study by [50] and [51].

4. Discussion

The above results account for about 40% of the standardized Hg concentrations in fish, with Sediment THg, atmTHg$_{dep}$, ppt, T$_{July}$, and T$_{Jan}$ as significant co-variants in varying combinations. The co-varying influences on Fish THg are particularly strong for Sediment THg and ppt, likely due to direct and indirect precipitation-induced Hg inputs into lake water from the atmosphere and surrounding land. While these inputs would have the most direct and causative
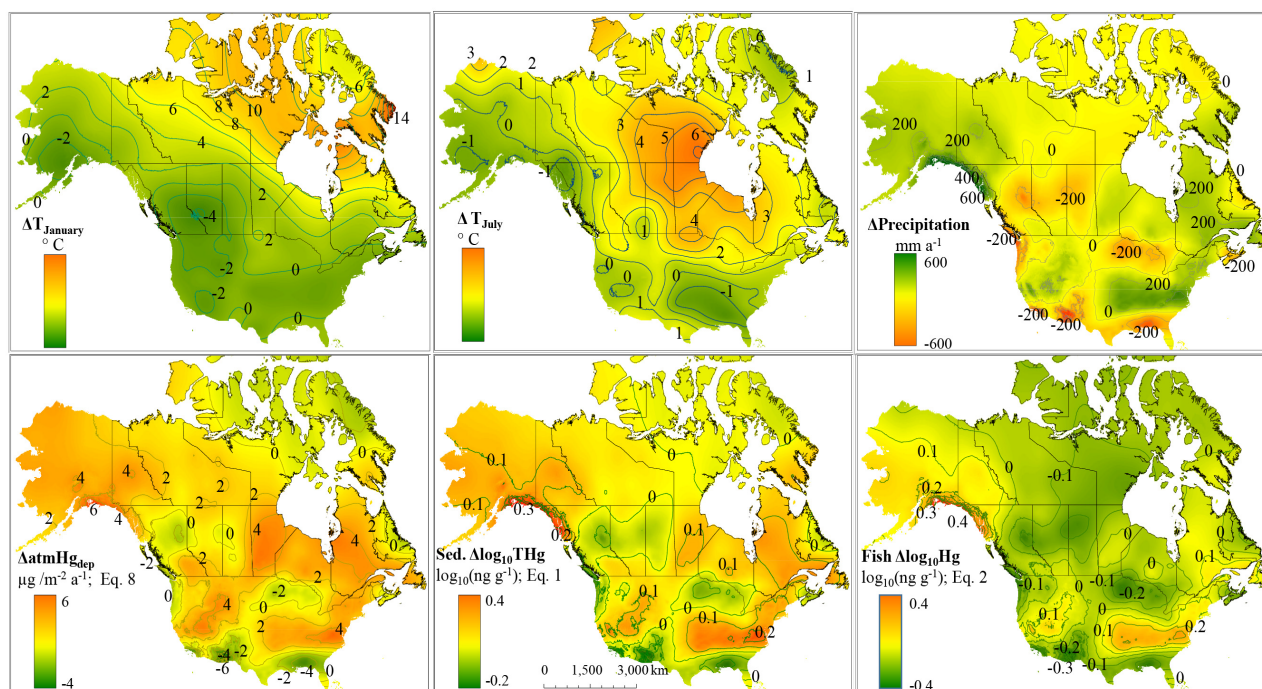
Figure 4. Projected 2011 to 2070 changes in mean annual January and July temperature (ΔT$_{Jan}$, ΔT$_{July}$), mean annual precipitation (Δppt), net atmospheric Hg deposition (ΔatmHg$_{dep}$), Sediment THg (Δlog$_{10}$Hg) and Fish THg (Δlog$_{10}$Hg) including contoursas outlined across the Canada (except most northern parts) and the continental USA.

influence on Hg methylization and fish uptake in the water column, trophic Hg uptake may also occur along biologically active and generally muddy top portions of lake sediments. However, the solid 30-cm deep cores of the GSC sediment survey represent about 100 years to sediment accumulation in northern lakes [52]. Hence, Sediment THg concentrations—as determined from the dried and ground-up sediment cores—would influence Fish Hg uptake only indirectly as a co-variate, with both Sediment THg and Fish THg influenced by external Hg lake inputs. In addition, while Equations (2) and (5) imply that Fish THg to Sediment THg co-vary to some extent across Canada, this is not necessarily the case within region-specific studies with limited range in climatic variations as noted for northern Ontario [53] and for the Great lakes region [54].

While atmospheric Hg emission and deposition rates have decreased over the last 25 years or so [55] [56], this is not reflected by the 1967-to-2010 FIMDAC data for which

$$\text{Fish} \log_{10} \text{THg} = (1.78 \pm 0.01) + (0.0020 \pm 0.0005)\text{Sampling year since 1970}, \ R^2 = 0.0047 \quad (9)$$

In part, this is due to increasing and decreasing trends by regions and lakes. In some areas, gradual declines in Fish THg have occurred [46]. Increases in Sediment THg with increasing atmTHg$_{dep}$—as implied by Equations (4) and (5)—have been reported for the upper layer of non-bulked mid- to high-latitude lake sediments by [52] and [57]. Fish THg followed a straight-line downward pattern for Lake Ontario from 1998 to 2009. For Lake Superior and Erie, 1998-to-2009 Fish THg decreased and then increased again. For Lake Huron and Michigan, the opposite occurred [58].

Using Equations (1), (2), and (8) to estimate changes in atmHg$_{dep}$, Sediment THg and Fish THg into the future based on current 2011-2070 T$_{Jan}$, T$_{July}$ and mean annual precipitation projections produced the changes mapped in **Figure 4** across Canada (except the northern most islands) and the continental USA. These maps suggest that—without further changes in anthropogenic Hg emission rates—Sediment THg and Fish THg would change up and down by at most a factor of 3 or 5, respectively. At the same time, atmHg$_{dep}$ would change regionally from 4 to 6 μg·m^{-2}·a^{-1}, which corresponds to about 10% to 15% of the current total Hg deposition range across Canada and the USA. For the most part, the projected atmHg$_{dep}$, Sediment THg and Fish THg changes follow the projected precipitation pattern, being generally positive along the northern Pacific and mid-latitude Atlantic regions. Decreasing precipitation would lower atmHg$_{dep}$, Sediment THg and Fish THg along, e.g., the Bay of Mexico, the Prairie provinces of Canada (Alberta, Saskatchewan, Manitoba), and the States along the Bay of Mexico and along the Great Lakes.

Together, the Equation (2) projected changes for Fish THg in **Figure 4** are relatively small except where precipitation levels would increase by 200 mm or more. The corresponding lack of major Fish THg changes—as represented by

Equation (9) and in Figure 4—is likely due to a cancelling of positive and negative lake-by-lake trends. For example, [59] and [46] reported declining to non-declining levels of Fish Hg with respect to climate warming. In contrast, [60] 1) reported increasing Fish THg levels for Ontario, varying over the last 15 years from 0.01 to 0.16 $\mu g \cdot g^{-1}$ per decade by fish species (4321 screened data), and 2) extrapolated these into the future, up to 2050.

Changing to non-changing levels in Fish THg are likely due to complex interactions involving changes in trophic Hg transferences, fish predation, lake-water conditions (temperature, aeration, chemical composition) and reductions in atmospheric and terrestrial Hg inputs into lakes. The latter, as implied by Equations (3) and (4), would especially be the case in areas subject to drought (*i.e.*, low ppt, high rates of Hg volatilization, low rates of Hg immobilization). On land, Hg retention and land-to-lake transfers are affected by upslope land use, extent and type of vegetation type including wetland coverage, and Hg-containing mineral exposures [22] [61]. Increased surface temperatures on land stimulate 1) Hg re-volatilization from open areas [62], 2) Hg losses due to forest fires [63], and 3) Hg immobilization in vegetation biomass and in soil organic matter due to increased warmer and longer summer growth, especially where soils remain moist to wet [8] [64].

Increasing upstream lake-to-lake wetland coverage tends to increase THg in water and fish through brown-water transfer of organically complexed Hg [7] [65] [66]. In lakes, fish uptake of Hg varies by, e.g., extent and type of lake input from land and atmospheric sources, and by lake size and morphology. Daily, seasonal and annual water-intake and related aeration and de-aeration dynamics affect in-lake trophic bioaccumulation of methyl Hg directly [67]. For example, increased Hg uptake by fish would occur through increased net Hg methylization as hypolimnetic water temperatures and biological oxygen demands rise from cool to warm [68]. In this regard, brown-water discharge renders small lakes warmer than clear-water lakes, thereby reduxing epi- and hypolimnetic aeration which would stimulate Hg methylization. Hence, Fish THg (small brown-water lakes) > Fish THg (clear-water lakes). In addition, Fish Hg increases with decreasing lake-water pH, likely due to higher levels of Hg solubility and increased Hg methylization rates at lower pH [68] [69]. However, Fish THg can also increase with increasing pH, particular where in-lake de-aeration trumps the pH effect on Hg methylization [70] [71].

Detailed studies have shown that, while increasing water temperatures increase the production and uptake of methyl Hg from sediments and epilimnetic and anoxic hypolimnetic waters [72], there is also a concurrent temperature increase in Hg demethylation [53] [73]. [71] reported that deepening the thermocline and oxycline of one lake relative to a reference lake (done by way of pumping at about 15,000 $m^3 \cdot day^{-1}$) increased epilimnetic temperature and oxygen saturation leading to continuing reductions in epilimnetic MeHg and Fish THg, with post-experiment effects lasting for at least one year. Hence, future changes in Fish Hg are also related to climate-induced thermocline inversions, lake-by-lake.

Across northern Canada, the influx of Hg and MeHg into lakes and rivers peaks during the snowmelt season, and is lowest during the winter season [74].

Post-2000 Sediment THg fluxes decreased towards northern latitudes after showing a steady increase since about 1900. This was likely in response to the steadily falling of net atmospheric Hg deposition rates over the last 25 years [75] [76] [77]. Mercury levels in fish, birds, and mammals, however, varied, with inter-annual trends remaining uncertain across arctic inland and marine waters [5] [67] [78] [79]. In contrast, the subarctic region is currently subject to increasing permafrost decline. This decline produces water-filled collapse scars across peat plateaus [80], with each scar supporting algal growth, which in turn leads to peat-and mineral-released Hg accumulations in lakes and sediments [81] [82] [83].

Given that total Hg concentrations in retail fish should not exceed 500 ng·g^{-1} [84] (which corresponds to $\log_{10}$THg = 2.70), most of the standardized Fish THg values fell below this limit. The percentage extent to this value to be exceeded in 2070 would mainly be limited to southern Quebec where the maximum projected change in $\log_{10}$THg is ≈0.1, and more strongly so along the Pacific coast, where the maximum projected change in $\log_{10}$THg is ≈0.4.

Altogether, 60% of the Fish THg variations remain unexplained by way of the above trend analysis. This situation may improve by adding lake-by-lake variables such as lake size, morphology, upland watershed, topography and upslope wetland coverage to the analysis.

5. Concluding Remarks

As shown, climate affects in-lake Hg accumulations due to variations in weather, season, atmospheric Hg deposition, surrounding vegetation, and geological Hg exposure patterns. This would also include lake-by-lake variations in thermocline inversion and related recovery times. As quantified above, cross-regional changes among the co-varying patterns of atmospheric Hg deposition, Sediment THg and Fish THg will likely remain small, with potential changes more likely related to changes in precipitation amounts, frequencies and intensities than to co-varying changes in atmospheric Hg deposition and temperature. A major exception to this would be occurring across the subarctic, where sustained permafrost losses lead to a widening incidence of peat-plateau scars followed by mineral- and peat-accumulated Hg release into lakes and rivers. In total, lake-by-lake variations in Fish THg will remain large as documented, and will require continued monitoring due to changing Hg inputs and changing in-lake dynamics.

Whether or not future levels in sediment and Fish THg will trend as projected in Figure 4 remains unknown. The assumption made is that the co-dependencies of the variables in Table 1 are functionally linked to changing climate variations not only across space, but also across time, in the same way.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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