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Historical and Contemporary Patterns of Mercury in a Hydroelectric Reservoir and Downstream Fishery: Concentration Decline in Water and Fishes

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Abstract Mercury (Hg) contamination can pose risks to human and animal health as well as commercial fisheries. Reservoir construction in riverine systems produces flooded conditions amenable to Hg(II)-methylating bacteria, which can transform this relatively benign environmental contaminant into the bioaccumulative, environmentally relevant, and neurotoxic methyl-Hg (MeHg). Hg concentrations ([Hg]) in fishes from reservoirs can take decades to decrease to pre-dam levels, but less is known about Hg exported downstream and its dynamics within downstream fish populations. We examined and compared the multidecadal rates of biotic [Hg] decrease and contemporary factors affecting [Hg] in fish collected from a hydroelectric reservoir (Tobin Lake) and a related downstream fishery (Cumberland Lake) along the Saskatchewan River, Canada. Rates of [Hg] decrease were considered in four species—northern pike (*Esox lucius*), sauger (*Sander canadensis*), goldeye (*Hiodon alosoides*), and walleye (*S. vitreus*)—all of which showed a significant decrease over time ($p < 0.001$) and are now lower than Health Canada consumption guidelines (0.5 µg/g). Rates of decrease ranged from 0.5 to 3.9 %/year and were similar between sites in the cases of northern pike and sauger. Contemporary factors affecting [Hg] in walleye collected downstream include fish length ($p < 0.001$), fish age ($p < 0.001$), and trophic magnification through the food web ($p < 0.001$), and relationships between [Hg] and trophic level in predatory and prey fish are now similar to those found in non-Hg-inundated systems at a similar latitude. Together, these results suggest connected contamination between the two sites and delineate the timeline during which [Hg] in a variety of fish species decreased to nontoxic levels in both locations.

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Inorganic trace mercury (Hg) is a common and fairly benign metallic environmental contaminant that can be methylated into the potent and more environmentally relevant neurotoxin, teratogen, and potential chronic cardiovascular toxin, methyl-Hg (MeHg) (Karagas et al. 2012). Natural processes are responsible for a background signature of Hg in the environment; however, human industrial activities and the burning of fossil fuels has substantially increased the global pool of Hg that is transported across the globe by way of wind and water cycles (Keating et al. 1997; Goodarzi et al. 2008; Pirrone et al. 2010). The result is aquatic Hg pollution both near and far from industrial sources (Mierle 1990; Lucotte et al. 1995; Engstrom and Swain 1997) as well as concentrations in fish that may affect the health of fish-eating consumers (Mergler et al. 2007). When compared with consumption guidelines for total Hg concentrations ([Hg] in fish (e.g., 0.5 μg/g wet weight [Health Canada 2007]), data from large-scale sampling has indicated that many higher trophic level fish species regularly approach or exceed this limit (Depew et al. 2013) suggesting toxicological risk to consumers. Exacerbating this exposure, fish that are the most heavily contaminated are among those commonly consumed (Hammerschmidt and Fitzgerald 2006; Depew et al. 2013).

Hg accumulation in aquatic food webs depends largely on local conditions, specifically those conducive to the activity of methylating bacteria. They include high dissolved organic carbon (DOC) under anoxic conditions (Graham et al. 2012), relatively warm temperatures (Wright and Hamilton 1982), reducible sulfate and/or iron(III) (Yu et al. 2011), and large amounts of decaying flooded vegetation and soil organic matter, which are the primary contributors to DOC (Louchouarn et al. 1993; St. Louis et al. 1994; Kelly et al. 1997). Reservoirs can be contaminated by Hg from atmospheric deposition, runoff from the surrounding land, and upstream point sources (Mason et al. 1994; Selin 2009), and reservoir construction produces the aforementioned conditions conducive to Hg methylation and consequently results in Hg contamination in fish (Louchouarn et al. 1993; Jackson 1998; Bodaly et al. 2007).

Influxes of inorganic Hg and DOC, followed by subsequent increases in methylation from flooding or point sources, can contaminate areas within a short time frame, although [Hg] in biota are known to decrease once the influx ceases (Hrabik and Watras 2002). Time-series analyses of [Hg] in fish from within newly flooded reservoirs show that concentrations may continue to increase for approximately one decade followed by slow decreases over approximately 20 to 30 years after impoundment (Bodaly et al. 2007) with larger, older

organisms feeding at the top of long food chains showing the greatest degree of contamination (Cabana et al. 1994; Kidd et al. 1995).

Far less is known about the downstream transport of MeHg produced in reservoirs and its long-term fate in downstream food webs (Rosenberg et al. 1997; Schetagne et al. 2000; Kasper et al. 2012), although point-source reservoirs of MeHg may lead to increased [Hg] as far as 320 km downstream, which can accumulate within and subsequently decline from the food web (Anderson 2011). Given the proliferation of dams throughout the developing world (Finer and Jenkins 2012; Turkenburg et al. 2012), information on the consequences of reservoir formation for downstream fisheries is needed for responsible hydroelectric implementation and management.

The Saskatchewan River Delta (SRD) is a large wetland complex located in the boreal plain of north-central Canada. The SRD and its largest lake, Cumberland Lake (CL), currently supports a commercial fishery that is largely focused on walleye (*Sander vitreus*) and northern pike (*Esox lucius*) (Royer 1966; Wallace 1999), which are also highly represented in the local diet (Waldram 1988). A now-defunct fishery for lake sturgeon (*Acipenser fulvescens*) was closed briefly in the early 1970s because of high [Hg] (Waldram 1988), reopened in 1973, and fished until 1997 when it was closed permanently due to population decreases. The Upper Delta of the SRD is approximately 50 km downstream from the E. B. Campbell hydroelectric facility, which was constructed in 1963 creating the large, shallow Tobin Lake reservoir (TL). This study site thus represents an opportunity to examine long-term trends in fish [Hg] in an area downstream of an impoundment. Although the source of the Hg to the reservoir itself is unknown, it is possible that some Hg may have been contributed by an upstream chlor-alkali plant, which operated between 1964 and 1978 and led to high [Hg] in fishes in the South Saskatchewan River (Wobeser et al. 1970). However, the large distance between the plant and TL (approximately 400 km), the additional dilution from the North Saskatchewan River, and the high deposition of sediment-bound Hg expected at the transition zone between the Saskatchewan River and TL suggest that MeHg within TL is comprised of atmospheric Hg deposition with minor contributions from upstream runoff, which was subsequently methylated within the reservoir (Turner and Lindberg 1978; Thornton et al. 1990; Schetagne et al. 2000; Kinghorn et al. 2007).

The objective of this study was to evaluate historical trends in and contemporary drivers of [Hg] in fish communities in three areas influenced by a hydroelectric dam: the reservoir, the river immediately below the dam, and the

downstream delta. We sought to estimate the annual percent change of [Hg] in commercial species since monitoring began in the 1970s. We predicted (1) that concentrations would show decreases consistent with those observed within reservoirs (Bodaly et al. 2007) and (2) that known factors dictating fish [Hg] (i.e., age, size and trophic level), as well as the relationships within and among contemporary fish populations, would suggest that Hg methylation within the TL reservoir was a common source of contamination for both TL and CL fish populations. Furthermore, we predicted that these data would suggest that downstream transport of Hg to CL has ceased and that Hg has largely returned to background concentrations (Simoneau et al. 2005; Jardine et al. 2012). We performed these analyses to evaluate the potential for adverse health effects of fish consumers in local communities and to better understand the likely consequences of reservoir creation on active commercial and subsistence fisheries elsewhere in the developing world.

Methods

Study Area

The E. B. Campbell Dam (53°41′19″N 103°20′50″W) was commissioned upstream of the SRD in 1963 forming TL (Fig. 1). The SRD is located at the border of Saskatchewan and Manitoba, encompasses 10,000 km^2 (Smith et al. 2014), and is divided into the Upper Delta and Lower Delta with the lower portion in Manitoba and the upper located primarily in Saskatchewan. The Upper Delta contains the affected fishery site at CL (54°3′0″N 102°18′2″W). The majority of flow to the lake is from the Saskatchewan River flowing through the E. B. Campbell Dam, along with minor contributions from three tributaries: the Torch, Mossy-Grassberry River, and Sturgeon-Weir rivers (Smith et al. 2014). The areas surrounding CL, including the Cumberland Marshes, are characterized by numerous wetlands and both abandoned and active river channels. Although
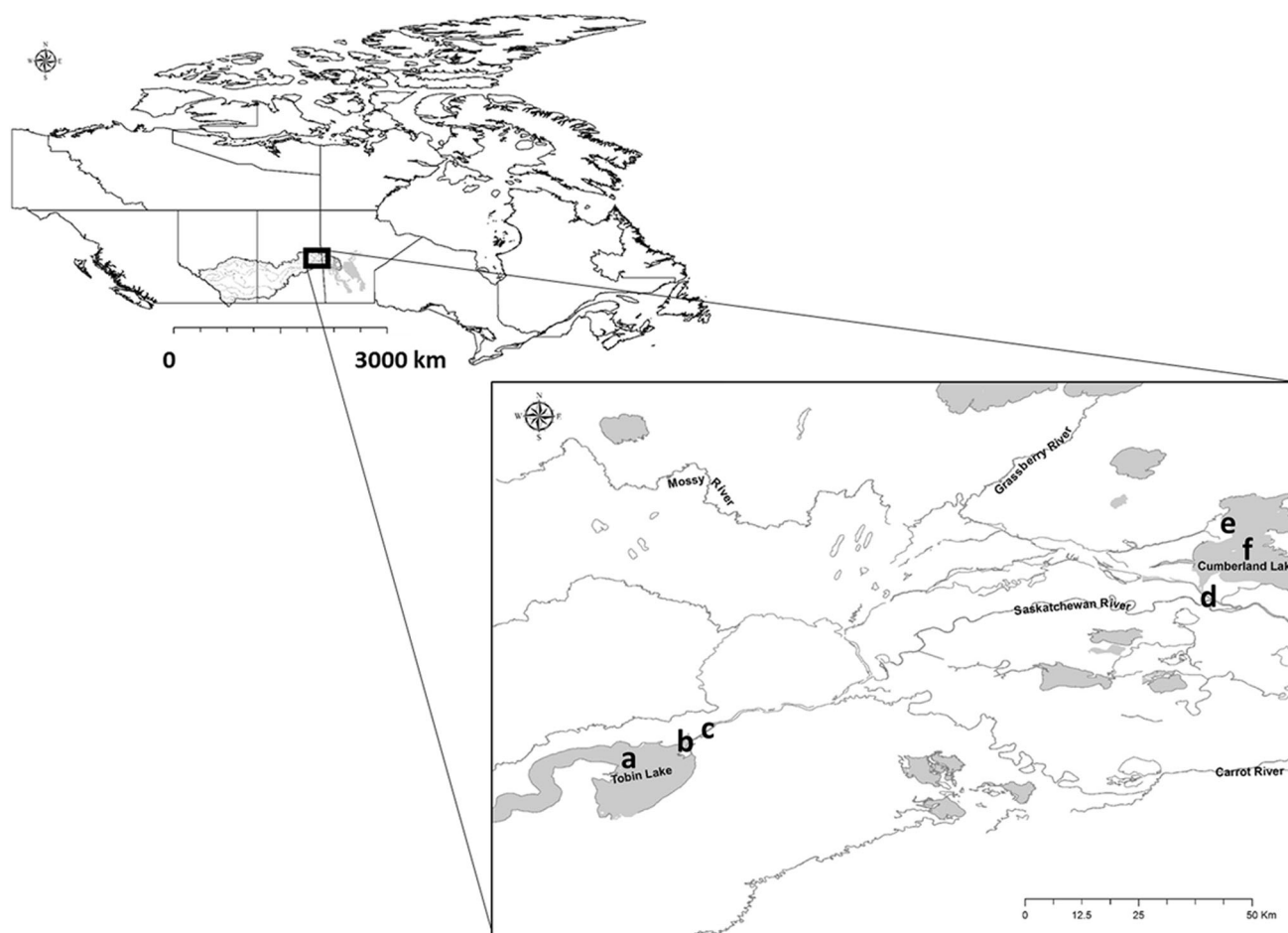

Fig. 1 Study sites: *a* Tobin Lake (TL), *b* E.B. Campbell Dam (formerly "Squaw Rapids Dam"), *c* Immediately below the E.B. Campbell Dam (EBC), *d* Saskatchewan River junction with Cumberland Lake (CL-SR), *e* Mossy River junction with Cumberland Lake (CL-MR), *f* Cumberland Lake (CL)

upstream water-resource development has decreased the magnitude of the annual summer flood, the delta still experiences significant flood–drought cycles (Sagin et al. 2015). A lack of industrial activity in the SRD means that contributions from local point sources of Hg are minimal, and an additional description of the quality and quantity of water in the Saskatchewan River can be found in Wheater and Gober (2013).

Water Sampling and Analysis

Historical water samples were collected by the provincial government of Saskatchewan between 1975 and 1990, and a total of 70 water samples were collected from immediately below the E. B. Campbell Dam (EBC; 53°42′59″N 103°17′11″W) and analyzed for Hg. We collected contemporary water samples (2013–2014) in 250-ml narrow-mouth FPE bottles and preserved the water at pH 2 with trace-metal analysis-grade nitric acid using clean-hands techniques (United States Environmental Protection Agency [USEPA] 1996). Collection vessels were prepared, and samples were analyzed by SRC Analytical Laboratories, Saskatoon, Saskatchewan. Additional water-chemistry parameters of contemporary samples were provided by SRC Analytical Laboratories and can be found in the supplementary information.

Throughout the collection period, [Hg] detection limits in water varied substantially, ranging from 0.1 µg/L in 1973 to 0.01 µg/L in 2014, and were therefore inconsistent between dates. Horizontally linear clusters of concentrations were presumed to represent censored values lower than detection within the relevant time frame, and we substituted half the detection limit for data-display purposes. Contemporary concentrations were initially analyzed using inductively coupled plasma mass spectrometry, which showed concentrations to be lower than the detection limit of this method (0.01 µg/L). A subset of water samples were then analyzed for low-level Hg by SRC Analytical Laboratories in accordance with USEPA 1631 methodology and using MERX Automated Total Analytical System, which allows for the detection of concentrations as low as 1 ng/L (USEPA 2002).

Fish Sampling, Processing, and Analysis

Two sets of fish data were considered in this study. Historical records of fish muscle [Hg] (µg/g wet weight [ww]) were obtained from the provincial government of Saskatchewan and span from 1970 to 2013. Datasets derived from these data included muscle [Hg], fork length, and date of capture for several commercially and recreationally important fish species including northern pike, walleye, sauger (*S. canadensis*), and goldeye (*Hiodon alosoides*). Field-collected historical samples were filleted on site, and muscle

samples were frozen immediately and subsequently processed and analyzed in accordance with Environment Canada guidelines using cold-vapour atomic absorption techniques (Environment Canada 1977). Historical fish concentrations were used to derive rates of [Hg] decrease over time. Datasets were derived for TL (53°37′11″N 103°31′43″W) and CL as well as for several lakes nearby, but not connected to the Saskatchewan River to serve as reference sites. These unconnected reference lakes included Candle (53°48′59″N 105°18′00″W), Montreal (54°15′36″N 105°44′37″W), and Big Sandy (54°25′26″N 104°5′30″W) lakes, and they were chosen for their proximity to the SRD and their robust datasets, which included adequate samples spanning multiple years in the cases of northern pike and walleye.

Contemporary samples came from TL, EBC, and from two sites near CL, the mouth of the Mossy River where it empties into CL (CL-MR [54°04′48″N 102°21′3″W]) and along the Saskatchewan River south of CL (CL-SR [53°57′16″N 102°23′9″W]) (Fig. 1). Contemporary fish samples were provided by the Cumberland House Fisherman's Coop in spring and summer 2013 and 2014. Additional samples of small-bodied, low trophic level species (shiners, perch, minnows) were collected by way of bag seine. Because the Fisherman's Coop operates as a commercial fishery, often only heads were available for analysis. In these cases the total length of the walleye was extrapolated using the following formula:

$$\text{Total length } = 1.526 + (3.777 \times \text{head length})$$

where head length is the distance between the tip of the snout and the edge of the operculum. This isometric relationship was derived during preliminary analysis of fish of known length ($n = 113$, $r^2 = 0.87$). To standardize fish lengths in our contemporary data set, all contemporary walleye lengths were extrapolated using this formula. All contemporary pike and walleye [Hg] was derived from muscle samples that were excised from the filet remaining at the posterior of the heads supplied by the coop. Hg concentrations in muscle taken from this area correlate strongly with those from fillet samples (Bank et al. 2007). Whole-body Hg analysis, with head and tail removed, was used to determine [Hg] in shiner, minnow, and perch species.

Where available, data collected on contemporary fish samples included total length, fork length, and mass. To better understand contemporary trophic magnification of Hg at these sites, we measured [Hg] and trophic level of the northern pike and/or walleye provided by Cumberland House fishermen as well as four of their common prey species: spottail shiner (*Notropis hudsonius*), blackchin shiner (*N. heterodon*), brassy minnow (*Hybognathus hakinsoni*), and yellow perch (*Perca flavescens*). Muscle [Hg] in pike

and walleye of a given size within a given geographic location is relatively robust to seasonal variation and short-term annual cycles (approximately 1–2 % change/year Rasmussen et al. 2007; Neff et al. 2012); therefore, samples were pooled from summer collections in 2013 and 2014 for this analysis. Only prey species collected in summer 2013 were considered for analysis in an attempt to minimize seasonal influences over the whole body [Hg] in these fish (Korthals and Winfrey 1987; Greenfield et al. 2013).

All contemporary samples were desiccated for approximately 48 hours in a drying oven set to 50 °C to maximize Hg recovery (Cabañero Ortiz et al. 2002) and ground to a fine powder using an acid-washed mortar and pestle.

Each contemporary sample (50 mg) was subsequently analyzed in a direct mercury analyzer (DMA 80; Milestone, Connecticut, USA). Hg recovered from the collected samples was referenced against certified reference materials (CRMs—dogfish muscle DORM and lobster hepatopancreas TORT—from National Research Council, Ottawa, Canada, and human hair IAEA-85 from International Atomic Energy Agency, Vienna, Austria). Recovery of these CRMs was 100 ± 14 % ($n = 20$), 93 ± 7 % ($n = 8$), and 97 ± 1 % ($n = 4$), respectively. Blank sample boats contained less than half the detection limit of Hg (0.04 ng), and therefore the test samples were not blank corrected. To remain analytically consistent between historical and contemporary samples, wet-weight concentrations of muscle were subsequently derived from contemporary dry-weight concentrations by applying a 0.25 multiplier assuming 75 % moisture (May et al. 2009; Lavoie et al. 2010). For the purposes of this study, [Hg] is assumed as a proxy measure for [MeHg] because analyses of numerous fish species from different habitats confirm that virtually all Hg in fish tissues is in the methylated form and bound to thiol groups in proteins (Bloom 1992; Lemes and Wang 2009; Greenfield and Jahn 2010).

Heavy nitrogen isotopes (^{15}N) bioaccumulate at a rate of approximately 3.4 ‰ per trophic level compared with their lighter counterpart ^{14}N (Post 2002). To assess trophic magnification of Hg (Lavoie et al. 2013), nitrogen-stable isotope ratios (^{15}N/^{14}N [expressed as δ^{15}N]) were determined by combusting 1.0 (± 0.2) mg of dried, powdered samples of predators (walleye, pike) and prey fish (minnows) in a PDZ Europa ANCA-GSL elemental analyzer followed by delivery of N_2 gases to a PDZ Europa 20-20 isotope ratio mass spectrometer at the University of California Davis Stable Isotope Facility. Samples analyzed in duplicate differed by an average of 0.2 ‰.

To determine the effects of age on [Hg] in a subset of fish, otoliths were collected from contemporary walleye sampled from CL-MR. Otoliths were extracted, processed, and aged using the procedures developed by the Fisheries Management Branch of Alberta Environment and Sustainable Resource Development (Watkins and Spencer 2009) and analyzed by North Shore Environmental Services, Thunder Bay, Canada.

Statistics

Much of the historical water sampling was performed without the use of clean-hands techniques, and there was appreciable ambiguity in the detection limits over time. For these reasons, trends in water [Hg] over time were not analyzed statistically and instead were only included for display purposes.

To compare spatial and temporal changes in fish [Hg], we used historical data for four species (northern pike, sauger, walleye, and goldeye) from both TL and CL. Log-transformed [Hg] was compared against time (year) for each species across each location using analysis of covariance (ANCOVA) with location as the factor and year as the covariate after testing for parallel slopes. All ANCOVAs were considered significant when $p < 0.01$. Rates of [Hg] decrease from each species were determined from regression formulae derived from Log[Hg] in fish muscle over time (year) with the slope of the line taken as a measure of percent decrease per year. Because larger fish within species are known to have greater [Hg] in some, but not all, cases (Scott and Armstrong 1972; Simoneau et al. 2005; Fowlie et al. 2008), and preliminary analyses showed significant increases in fish length sampled over time in some populations, we accounted for this potential confound by standardizing to [Hg] of a uniform size. Log[Hg] per centimeter of fish length was calculated as Log[Hg]/Log Length(cm) for both TL and CL data on all species, and ANCOVAs were rerun to compare these adjusted rates of [Hg] decrease over time with location as the factor and time (year) as the covariate. Reference-lake data were analyzed using linear regression to assess whether their trends over time were comparable with our study sites. Because these reference analyses were run on pike and walleye, and the length of these species is known to correlate positively with muscle [Hg] in Saskatchewan (Saskatchewan Ministry of Environment 2014), these data were length adjusted as previously described before being analyzed against the year of capture. Reference-lake mean fish [Hg] is presented as either Log[Hg] or length adjusted Log[Hg] as indicated.

To assess spatial and biological factors driving [Hg] in our contemporary data set (2013 and 2014), we first ran an ANCOVA to compare the influence of location (factor) and length (covariate) on Log[Hg] in a single species—walleye—across three locations (EBC, CL-MR, and CL-SR). Next, we compared the relative influence of length, age, and δ^{15}N on [Hg] within CL-MR walleye where we had a range of body sizes and age data. We used forward stepwise linear regression to compare the fit and significance of the three variables.

Finally, we determined if trophic level, as estimated from δ^{15}N, provided additional explanatory power for [Hg] across fish species. We examined Hg transfer through the fish food web by calculating trophic magnification factors (TMFs) for three locations (TL, EBC and CL with the latter including both CL-MR and CL-SR) by regressing log-transformed [Hg] against δ^{15}N values for commercial fishes and prey fishes (Kidd et al. 2003; Jardine et al. 2006, 2012; Lavoie et al. 2013). These trophic magnification slopes (TMSs) can be converted to TMFs using TMF = 10^m where m is the TMS multiplied by the average increase in δ^{15}N across trophic levels (3.4‰ [Post 2002]). Differences in TMSs were tested among the three locations using ANCOVA with δ^{15}N as the covariate and location as a factor.

Results

Water [Hg] and Chemistry

Concentrations of Hg in water were occasionally above detection in the historical period, reaching as high as 2 to 4 µg/L in the 1970s, although data collected before the advent of clean-hands techniques are to be interpreted cautiously (Fig. 2). Although the frequency of detection appears to have decreased over the study period, ambiguities in the detection limits over time precludes any statistical analyses. All contemporary water samples contained [Hg] <0.01 µg/L, and subsequent low-level analyses showed that current concentrations range from 1 to 5 ng/L (data not shown). Because water-concentration data were only presented for illustrative purposes and were not analyzed statistically, our historical and contemporary concentrations are shown on a single plot in Fig. 2.

Because the Saskatchewan River is the common water source for both the reservoir and the downstream delta, there is limited variability in water chemistry among locations sampled in this study. Contemporary water samples are neutral to basic with moderate solute and DOC concentrations (Appendix Table 1 in supplementary material). Nutrient levels from each location can be classified as eutrophic with the exception of mesotrophic levels of chlorophyll *a* observed across sites (Nürnberg 1996).

Historical Trends in Fish [Hg]

Initial sampling in 1970 showed mean [Hg] above or nearing modern consumption guidelines (0.5 µg/g ww Health Canada 2007) in fillets of all species at both TL and
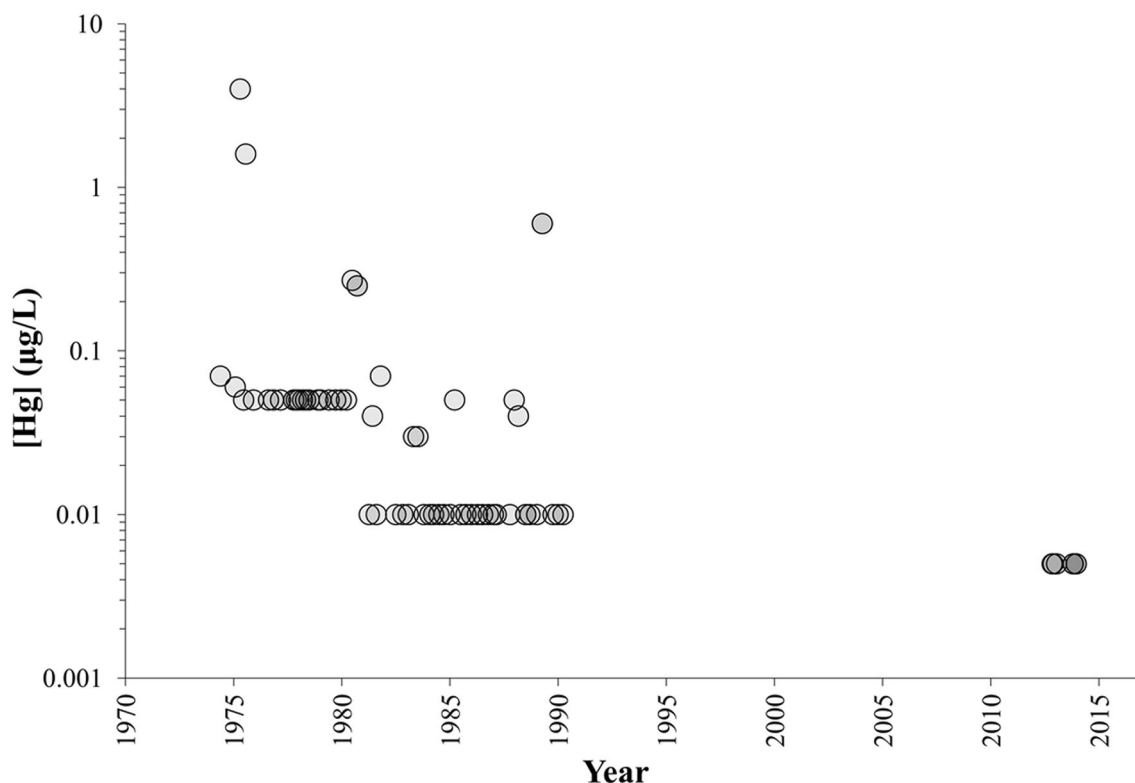

Fig. 2 Water [Hg] from historical (1975–1990) and contemporary (2013–2014) samples collected immediately below the E.B. Campbell Dam ($n = 70$) versus sampling year. Data are presented using a log-scaled y-axis
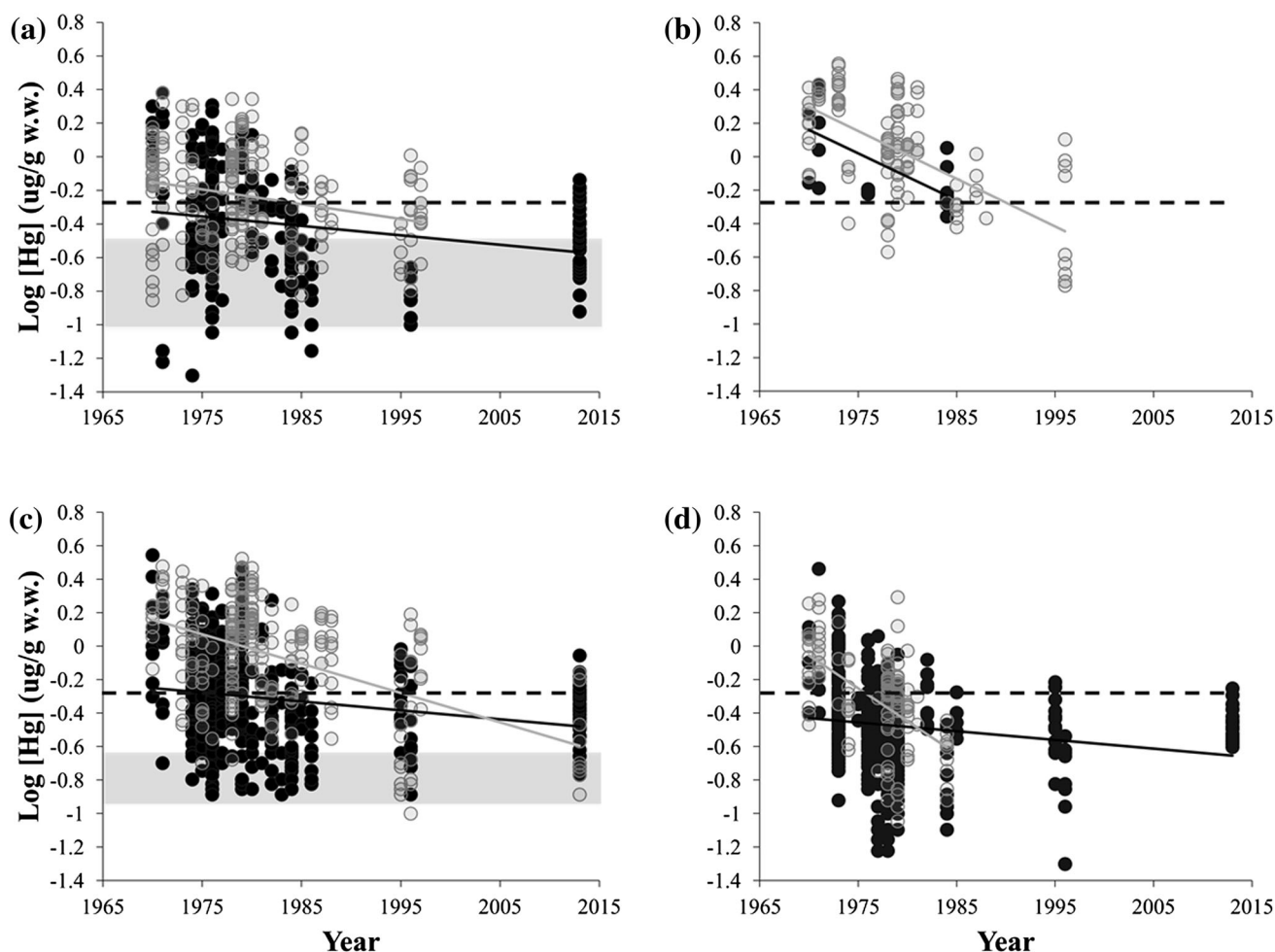
Fig. 3 Historical Log[Hg] in fish muscle versus sampling year (1970–2013) for species sampled in Tobin Lake (TL *grey*) and Cumberland Lake (CL *black*): **a** northern pike (TL, *n* = 215; CL *n* = 275), **b** sauger (TL *n* = 114; CL *n* = 21), **c** walleye (TL *n* = 232; CL *n* = 483), and **d** goldeye (TL *n* = 118; CL *n* = 396). The *horizontal dashed black line* represents the current consumption guideline, 0.5 µg/g (Health Canada 2007), *shaded areas* represent reference lake mean Log[Hg] (±SD) of fillets from northern pike (Fig. 3a; collection dates 1971–2002; *n* = 89) and walleye (Fig. 3c; collection dates 1970–2002; *n* = 96). See Table 1 for regression analyses and Table 2 for ANCOVA results

Table 1 Annual rates and regression results of Log[Hg] decrease over year from muscle of northern pike, sauger, walleye, and goldeye populations sampled from Tobin and Cumberland lakes between 1970 and 2013

Species	N	Annual [Hg] decline (%)	r^2	F	P
Northern pike$_{TL}$	215	0.9	0.06	14.70	<0.001
Northern pike$_{CL}$	275	0.6	0.05	14.56	<0.001
Sauger$_{TL}$	114	2.9	0.41	76.97	<0.001
Sauger$_{CL}$	21	2.8	0.47	16.62	<0.001
Walleye$_{TL}$	232	1.8	0.25	78.08	<0.001
Walleye$_{CL}$	483	0.5	0.04	21.63	<0.001
Goldeye$_{TL}$	118	3.9	0.33	55.80	<0.001
Goldeye$_{CL}$	396	0.5	0.03	11.63	<0.001

CL with values ranging from 0.3 to 1.8 µg/g in goldeye, <0.1 to 2.0 µg/g in northern pike, 0.5 to 3.5 µg/g in walleye, and 0.7 to 2.6 µg/g in sauger. Comparing rates of Log[Hg] decrease within species across TL and CL fishes showed significant rates of [Hg] decrease in both sites for all species (Fig. 3; Table 1), although the strength of the regression varied (r^2 0.03 to 0.47; p < 0.001).

Location explained significant variation in [Hg] in walleye and goldeye (Table 2), but the interaction term was significant for these two species, suggesting a more

Table 2 ANCOVA table comparing effects of location and year on Log[Hg] in fillets of northern pike ($n = 490$), sauger ($n = 135$), walleye ($n = 715$), and goldeye ($n = 514$) collected from Tobin and Cumberland lakes between 1970 and 2013

Species	df	MS	F	p
Northern pike				
Location	1	0.262	3.2	0.076
Year	1	2.123	25.7	<0.001
Location*year	1	0.110	1.3	0.248
Sauger				
Location	1	0.003	0.1	0.804
Year	1	2.242	41.4	<0.001
Location*year	1	0.000	0.0	0.956
Walleye				
Location	1	3.391	45.7	<0.001
Year	1	7.592	102.2	<0.001
Location*year	1	2.161	29.1	<0.001
Goldeye				
Location	1	2.440	34.7	<0.001
Year	1	3.760	53.5	<0.001
Location*year	1	2.204	31.3	<0.001

df degrees of freedom

rapid decrease in TL, and thus limited our ability to conclude that either site had greater concentrations for a given year. For the other two species (pike and sauger), [Hg] decrease was similar between locations (Tables 1 and 2, Fig. 3). Interaction terms were removed from pike and sauger analyses due to nonsignificance. Subsequent equal slopes analyses of the simplified model indicated effects of location and year for both species (pike location: $F = 29.77$, $p < 0.001$; year: $F = 27.46$, $p < 0.001$; sauger location: $F = 5.66$, $p = 0.019$; year: $F = 93.23$, $p < 0.001$). Holm–Sidak post hoc tests showed significant differences in mean [Hg] between locations in both pike ($t = 5.456$, $p < 0.001$) and sauger ($t = 2.379$, $p = 0.019$) with greater concentrations in TL fishes for both species.

Length-adjusted [Hg] in all fish showed similar trends to those observed in nonadjusted data sets, thus showing significant decreases in all species in both locations (Appendix Tables 2 and 3 and Fig. 1 in supplementary material). Specifically, significant rates of decrease were observed across sites and species ($p \leq 0.001$). Interaction terms were removed from the pike and sauger analyses due to nonsignificance. Subsequent equal slopes analyses indicated effects of location and year for both species (pike location: $F = 24.48$, $p < 0.001$; year: $F = 21.68$, $p < 0.001$; sauger location: $F = 4.75$, $p = 0.031$; year: $F = 89.97$, $p < 0.001$). Holm–Sidak post hoc analysis showed that significant differences remained between locations when considering length-adjusted Log[Hg]

against year in pike ($t = 4.948$, $p < 0.001$) and sauger ($t = 2.179$, $p = 0.03$) with greater concentrations in TL fishes for both species.

Although periods of data availability for the reference lakes were typically shorter than those for TL and CL, these former lakes exhibited limited change in fish [Hg] over time. There were no significant changes over time in Candle Lake pike (1992 to 2002, $n = 30$, $p = 0.131$), Montreal Lake pike (1971 to 1988, $n = 27$, $p = 0.308$), Montreal Lake walleye (1983 to 1988, $n = 15$, $p = 0.350$), Big Sandy Lake pike (1978 to 1999, $n = 32$, $p = 0.083$), or Big Sandy Lake walleye (1978 to 1999, $n = 39$, $p = 0.883$). Candle Lake walleye (1970 to 2002, $n = 42$, $p < 0.001$) had a slight positive increase in [Hg] over time.

Contemporary Predictors of Fish [Hg]

In the contemporary walleye samples collected near (EBC) and far (CL-MR and CL-SR) from the dam, the interaction term was not significant, suggesting similar [Hg]-versus-length relationships among sites ($F = 2.016$, $p = 0.136$; Fig. 4). With the interaction term removed, length and location were both significant predictors of [Hg] ($F = 91.046$, $p < 0.001$; $F = 13.648$, $p < 0.001$). Holm–Sidak post hoc analysis showed that fish from immediately below the dam have greater [Hg] than both CL-MR and CL-SR ($t = 3.665$, $p < 0.001$; $t = 5.224$, $p < 0.001$), whereas [Hg] from the two downstream locations were not significantly different from each other ($t = 1.876$, $p = 0.062$) (Fig. 4).

Within walleye from CL-MR, forward stepwise regression analysis showed both age and length positively predicted [Hg] ($F = 33.777$, $p < 0.001$; $F = 33.011$, $p < 0.001$), and derivation of our strongest simplified model included only age, which proved to be the single best predictor of [Hg] ($p < 0.001$, $r^2 = 0.35$). Within this species, δ^{15}N proved to be an ineffective predictor of [Hg] ($F = 1.17$, $p = 0.284$) (Fig. 5).

The strength of the Log[Hg]-versus-δ^{15}N regressions for food webs with multiple fish species varied among sites. There was a strong and significant relationship at TL ($r^2 = 0.85$, $F = 71.969$, $p < 0.001$), a weak but significant relationship at CL ($r^2 = 0.34$, $F = 50.212$, $p < 0.001$), and a nonsignificant relationship at EBC ($r^2 = 0.11$, $F = 3.093$, $p = 0.09$) (Fig. 6). Across sites, both δ^{15}N and location were significant predictors of [Hg] ($F = 55.273$, $p < 0.001$; $F = 8.135$, $p < 0.001$). However, the differences in slopes resulted in a significant interaction term ($F = 5.998$, $p = 0.003$), thus indicating different Hg biomagnification among sites, possible differences in the structure of the food webs, or insufficient characterization of the breadth of δ^{15}N at the EBC site. Estimated slopes at
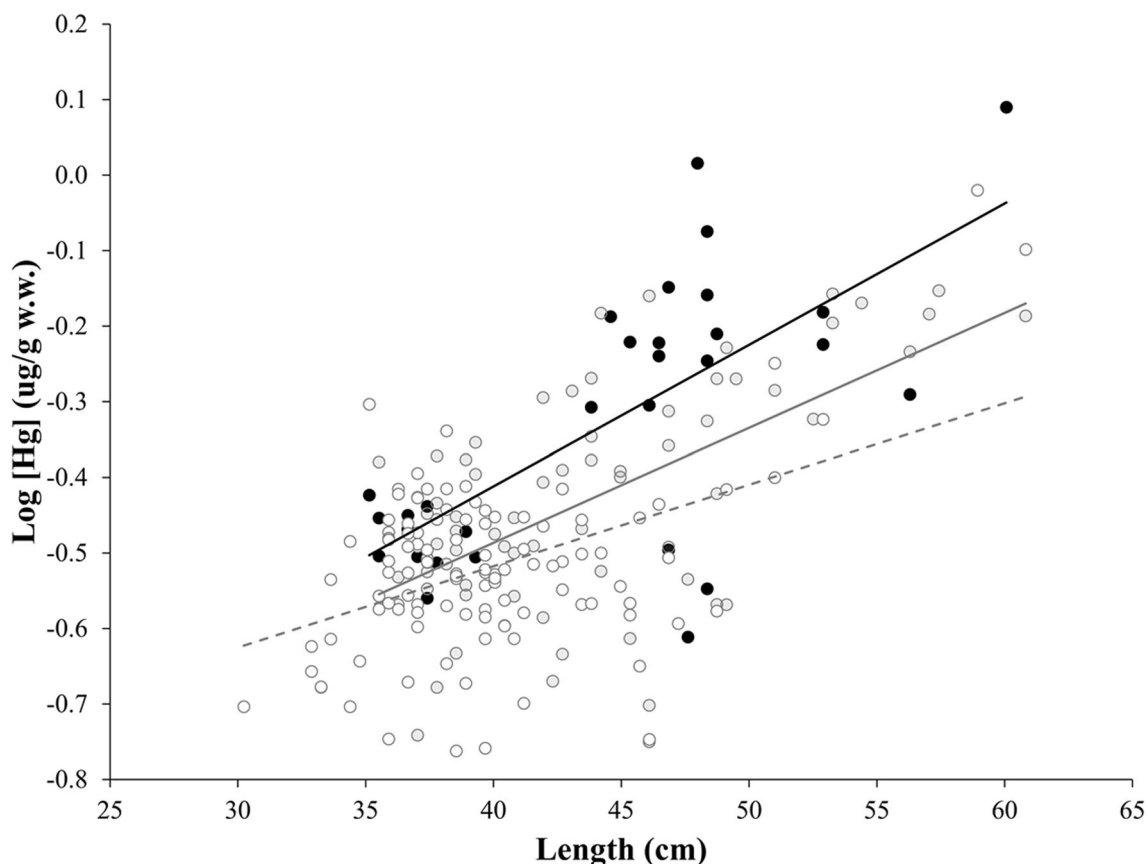
Fig. 4 Log[Hg] in contemporary muscle samples versus length (cm) for walleye collected from immediately below the E.B. Campbell Dam (*Solid circles*, *black best-fit line*; *n* = 30), Cumberland Lake juncture with Mossy River (*Shaded circles*, *grey best-fit line*; *n* = 66), and Cumberland Lake juncture with the Saskatchewan River (*Open circles*, *dashed best-fit line*; *n* = 113)

the three sites correspond to [Hg] TMFs of 9.0 (TL), 2.9 (CL), and 2.6 (EBC) (Table 3).

Discussion

Analyses of a 40-year historical record showed significant [Hg] decrease within the muscle of multiple fish species in the TL reservoir and a downstream fishery at CL, thus providing new information on rates of [Hg] decrease after impoundment in an array of commercially relevant species (Bodaly et al. 2007; Anderson 2011). Furthermore, analyses of contemporary samples found length and age to be strong significant predictors of [Hg] in walleye, which will allow consumers to use either parameter to avoid consuming walleye that may retain high [Hg] or to guide more efficient walleye sample selection in the ongoing monitoring of [Hg] recovery in this system. δ^{15}N analyses proved useful in predicting [Hg] across species within sites, but not within species or across sites, and TMS values suggested that current [Hg] are at or approaching a new

equilibrium in affected fish (Lavoie et al. 2013). Taken together, these data can help to delineate the effective timeline of [Hg] decrease from impounded and downstream fish contaminated as a result of hydro-electric reservoir construction as well as the factors controlling rates of Hg uptake and clearance. These data can inform expected consequences of reservoir-initiated Hg inundation as the world develops an increasing reliance on hydro-electric-power generation (Finer and Jenkins 2012; Turkenburg et al. 2012).

Although we lack pre-dam data for these systems, the patterns of [Hg] decrease beginning in 1970 agrees with the approximate 7- to 10-year post-impoundment peak [Hg] observed for pike and walleye in similar systems (Bodaly et al. 2007; Schetagne and Therrien 2013). The greater [Hg] in walleye immediately below the reservoir agrees with the persistent but attenuated export of Hg from reservoirs (Schetagne et al. 2000) despite the very low concentrations in contemporary water samples. The connection between TL and CL is strengthened by the consistent patterns of [Hg] decrease observed in the muscle of
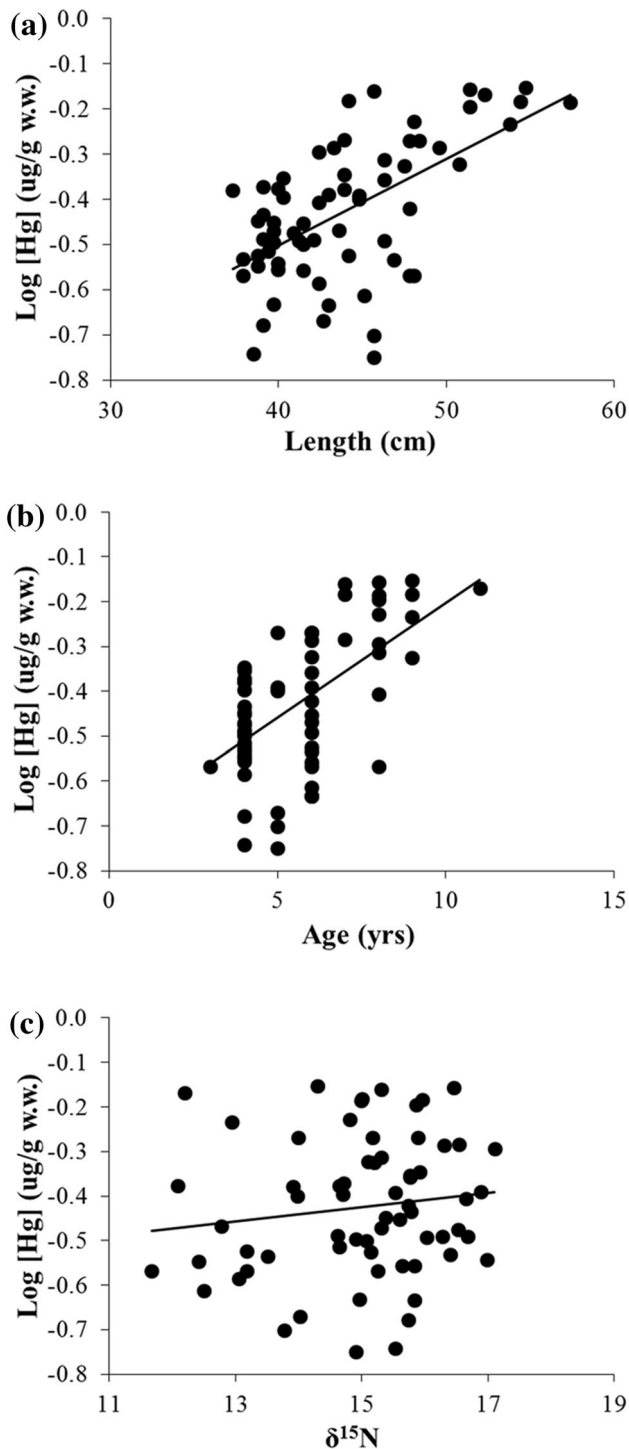
Fig. 5 Log[Hg] of muscle versus **a** Length (cm) **b** Age (years); and **c** δ^{15}N (‰) in walleye sampled from the juncture of Cumberland Lake with Mossy River (2014; *n* = 65)

Fig. 6 Log[Hg] of muscle (pike, walleye) and whole body homogenate (all others) versus δ^{15}N in **a** Tobin Lake (walleye, *n* = 7; yellow perch, *n* = 3; spottail shiner, *n* = 5), **b** Immediately below the E.B. Campbell Dam (walleye, *n* = 17; yellow perch, *n* = 8; spottail shiner, *n* = 3), **c** Cumberland Lake (including samples from the Cumberland Lake and Mossy River junctures with the Saskatchewan River; northern pike, *n* = 18; walleye, *n* = 65; spottail shiner, *n* = 6; blackchin shiner, *n* = 2; brassy minnow, *n* = 3; yellow perch, *n* = 6)

the four fish species studied, an occurrence unlikely in unrelated sites, and by the lack of similar chronological trends in fish [Hg] in reference lakes where concentrations were always lower than those in TL and CL (Lathrop et al. 1991; Harris and Bodaly 1998; Simoneau et al. 2005). As a
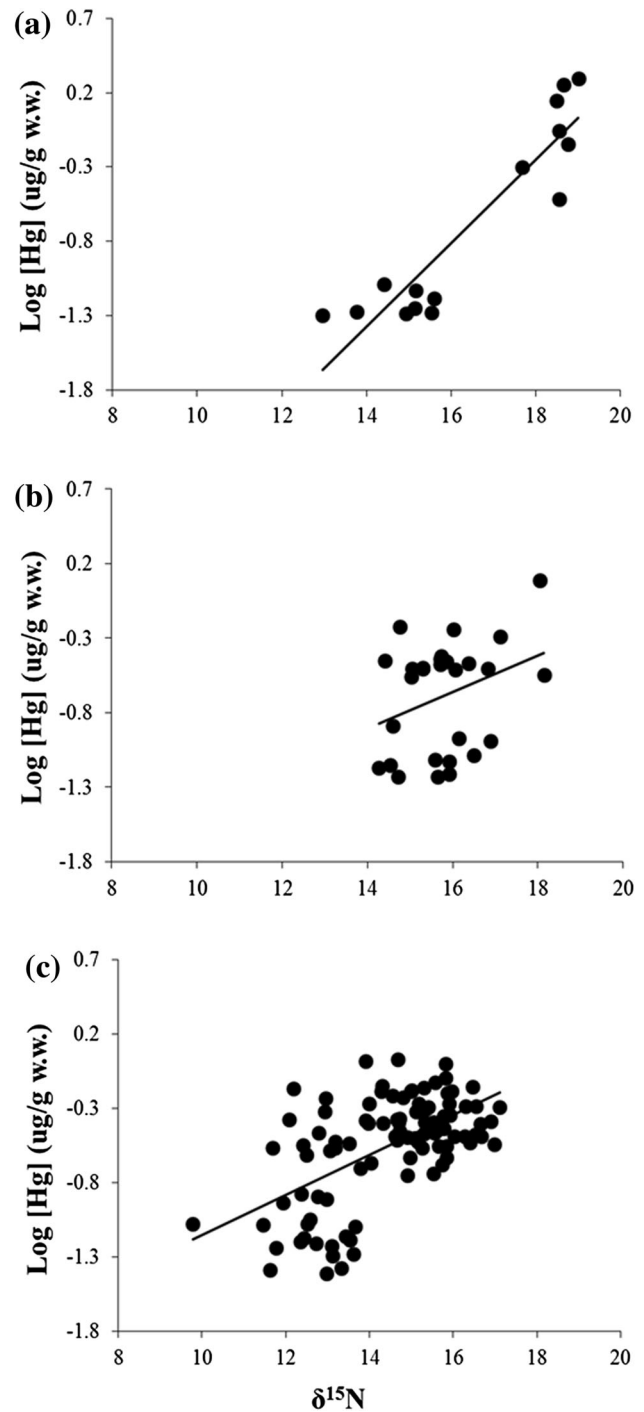
result of decreases in [Hg] from TL and CL fish during the last 40 years, Hg levels in the species studied are now, on average, lower than consumption guidelines (Fig. 3). The

Table 3 TMSs calculated from regressions of Log[Hg] versus δ^{15}N in fish muscle (northern pike, walleye) and whole-body homogenates (spottail and blackchin shiner, yellow perch, brassy minnow) with corresponding TMFs for Tobin Lake (TL) immediately below the E. B. Campbell Dam (EBC) and Cumberland Lake (CL)

Site	n	TMS ± SE	r^2	p	TMF ± SE
TL	15	0.28 ± 0.03	0.85	<0.001	9.0 ± 1.3
EBC	28	0.12 ± 0.07	0.11	<0.090	2.6 ± 1.7
CL	100	0.14 ± 0.02	0.34	<0.001	2.9 ± 1.2

exception is CL sauger, although this disparity is likely an artifact of the truncated data set for this species because the existing trend suggests that current [Hg] may be lower than the guidelines. Notably, rates of [Hg] decrease from pike in our data set (0.6 % year^{-1} or approximately 0.0016 d^{-1}) were on the same order of magnitude as those observed in pike in laboratory depurations (K = 0.00072–0.004 d^{-1}) as summarized by Trudel and Rasmussen (1997). Disparities between the sizes of walleye between Trudel and Rasmussen's (1997) data and the present study preclude similar comparisons for that species.

Within our three piscivorous species, sauger exhibited a relatively rapid decreases in [Hg] compared with northern pike and walleye, which is consistent with its relatively short life span (Scott and Crossman 1973) rapidly turning over cohorts exposed to cleaner waters after peak methylation. Because dietary uptake is the primary route of Hg exposure (Harris and Bodaly 1998; Pickhardt et al. 2006), the similarly rapid decrease of [Hg] in TL goldeye support conclusions from previous researchers that small-bodied, nonpiscivorous species should show the most rapid [Hg] decreases due the relatively short length of their food chain (Scott and Crossman 1973; Cabana et al. 1994; Moon et al. 1998; Bodaly et al. 2007). Significantly slower [Hg] decreases were observed in CL goldeye and walleye, and converging trends of pike [Hg] suggest slower decreases in the downstream population, although the interaction between rates was not significant for this species. These patterns may represent an attenuated and more persistent MeHg exposure as MeHg was transported between the sites (Schetagne et al. 2000). Although sauger data do not adhere to these trends, we suspect that a more robust sauger data set would have unveiled similar patterns. It is also possible that benthic-*versus*-pelagic feeding habits of these species may account for differences in downstream exposure post-inundation (Eagles-Smith et al. 2008). Unfortunately, there is considerable overlap in the dietary habits of our predatory species, and differences in feeding habits were not possible to confirm because we did not conduct detailed stomach content analyses. Where rates of [Hg] decrease were similar enough to be compared across sites within species (pike and sauger), TL [Hg] was consistently greater than CL.

As is the case with much of Northern Canada, the SRD is a critical foundation for the subsistence economies of Cree, Métis, and nonstatus First Nations peoples. Increased [Hg] in fish became a concern for Cumberland House fisheries in the 1970s, although no cause could be ascribed (Waldram 1988). The parallel decreases in [Hg] in TL and CL fish and greater [Hg] in walleye nearest the dam observed in this study, coupled with similar observations in other reservoir influenced systems (Bodaly et al. 1984, 2007; Anderson 2011), suggests that implementation of the TL reservoir upstream may be responsible for the biotic contamination. The fishery downstream was then susceptible to MeHg inundations from this source through aqueous and, to a lesser extent, biotic transport (Schetagne et al. 2000; Baker et al. 2009). At present, muscle [Hg] concentrations have fallen lower than consumption guidelines in three of four species, with trends in sauger data suggesting that contemporary sauger [Hg] is lower than guideline [Hg] as well (Health Canada 2007). The human population of Cumberland House faced increased [Hg] for decades after implementation of the dam, and although maximum concentrations in the 1970s were lower than those in the well-documented Grassy Narrows incident in Northwestern Ontario (e.g. 15 µg/g in walleye muscle from Clay Lake in 1973 Kinghorn et al. 2007), further studies may be required to determine if any subclinical symptoms are evident in the human population (Takoaka et al. 2014).

In agreement with previous research, age and length were strong predictors of muscle [Hg] (Simoneau et al. 2005; Rasmussen et al. 2007) in walleye from the downstream sites. In contrast to the strong effects of length and age, δ^{15}N failed to predict Log[Hg] concentrations within species (Jardine et al. 2012), likely reflecting the limited trophic variation in walleye >15 cm in length inhabiting similar habitats (Galarowicz et al. 2006). Taken together, these results support the standard use of walleye body length by various jurisdictions as a suitable proxy for [Hg] in fish consumption advisories (Ontario Ministry of the Environment 2013; Saskatchewan Ministry of Environment 2014).

With the exception of the EBC site, the observed significant TMSs were consistent with those predicted for northern latitudes (Lavoie et al. 2013), rather than being substantially lower, suggesting that current [Hg] in fish tissues has either reached or is nearing equilibrium. Average TMSs in freshwater are 0.16 ± 0.1 SD, i.e., values obtained from CL are well within expected ranges, whereas the TMS from TL slightly exceeds the upper range of the scale (Lavoie et al. 2013). The high value observed in TL may reflect that predators from TL available for this analysis were relatively large compared with those from EBC and CL because, by convention, [Hg] levels were not standardized against length for trophic analyses (Lavoie

et al. 2013). The divergence in these trends may also stem from sampling success because prey species sampled from TL were only successfully captured in June of 2013, whereas those from EBC and CL were captured in August, which may have artificially inflated the [Hg] of prey species at the latter two sites relative to the former (Korthals and Winfrey 1987; Greenfield et al. 2013).

Although previous studies have suggested that <30 years are required for [Hg] to return to baseline levels in impounded fish (Bodaly et al. 2007) and in those downstream of impoundments (Anderson 2011), present data suggest a less optimistic timeline upwards of 40 years. Further study will be required to fully describe the ultimate timeline of Hg inundation and decrease, particularly because we lack pre-impoundment data. Future studies may corroborate the influence of reservoir liberated Hg on downstream fisheries by constructing ^{137}Cs and/or ^{210}Pb [Hg] timelines from affected and reference sediment profiles (Van Metre et al. 2004; Muir et al. 2009). These data are necessary to ensure that the increasing reliance on hydroelectric power generation does not come at the cost of human and environmental health. A more robust understanding of the dynamics of Hg within and downstream of an affected reservoir and its biota hold promise to ensure more ecologically sound implementation of future reservoirs.

Compliance with Ethical Standards

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