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MERCURY CONCENTRATIONS IN FISH FROM FOREST HARVESTING AND FIRE-IMPACTED CANADIAN BOREAL LAKES COMPARED USING STABLE ISOTOPES OF NITROGEN

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Abstract—Total mercury (Hg) concentration was determined in several piscivorous and nonpiscivorous species of fish from 38 drainage lakes with clear-cut, burnt, or undisturbed catchments located in the Canadian Boreal Shield. Mercury concentrations increased with increasing fish trophic position as estimated using stable isotopes of nitrogen (N; $r^2 = 0.52$, 0.49, and 0.30 for cut, reference, and burnt lakes, respectively; $p < 0.01$). Mercury biomagnification per ‰ $\delta^{15}N$ varied from 22 to 29% in the three groups of lakes. Mercury availability to organisms at the base of the food chain in lakes with cut catchments was higher than that in reference lakes. In cut lakes, Hg concentrations in fish were significantly related to ratio of the clear-cut area to lake area (or lake volume; $r = +0.82$ and $+0.74$, respectively, $p < 0.01$). Both impact ratios were, in turn, significantly correlated with dissolved organic carbon. These findings suggest that differential loading of organic matter–bound Hg to lakes can affect Hg cycling. In addition, Hg concentrations exceeded the advisory limit for human consumption (0.5 μg/g wet wt) from the World Health Organization in all top predatory species (northern pike, walleye, and burbot) found in cut and in two partially burnt lakes. Thus, high Hg concentrations in fish from forest-harvested and partially burnt lakes may reflect increased exposure to Hg relative to that in lakes not having these watershed disturbances.

Keywords—Mercury Clear-cut Wildfire Fish Nitrogen isotopes

INTRODUCTION

High mercury (Hg) concentrations have been reported in predatory fish from pristine boreal lakes in North America and Europe [1,2]. Deposition of atmospheric Hg originating from anthropogenic emissions is considered to be an important source of Hg even to remote lakes and their catchments [3]. In the catchments, Hg tends to associate with the organic matter in the upper layer of the soil [4]. Therefore, major watershed perturbations, such as wildfire and clear-cut logging, may alter Hg export rates to lakes and influence in-lake reactions that determine its fate. For example, clear-cut logging increases the export of dissolved organic carbon (DOC) and Hg [5,6] and can stimulate the bacterial production of methylmercury (MeHg). Wildfire may produce a nutrient pulse in lakes, influencing the formation and availability of MeHg to the biota. Forest fires also may reduce the pool of Hg in catchment soils through elemental Hg volatilization to the atmosphere [7].

In previous studies, we compared the effects of wildfire and clear-cut logging on MeHg in zooplankton and on total Hg in northern pike (*Esox lucius*), a top predatory fish [8,9]. Those results showed that high MeHg and total Hg were associated with clear-cut logging but not with wildfires. In the present study, we extended those findings to the most abundant species of fish in the same lakes. Our objective was to compare total Hg concentrations in piscivorous as well as in nonpiscivorous species from clear-cut logging and fire-impacted lakes. Assessing the effect of watershed disturbance in Hg levels in biota collected at different lakes is not a straightforward exercise, however. Because Hg is biomagnified along food chains, among-lake differences in Hg concentrations for a given species can be the result of variations in trophic position rather than the effect of disturbances. In the present study, stable isotopes of nitrogen (N) were used to determine the trophic position of fish. This approach is based on the enrichment in ^{15}N from prey to predator because of the preferential excretion of ^{14}N [10]. We then compared the relationships between Hg levels and fish trophic position in both disturbed and undisturbed lakes.

MATERIALS AND METHODS

Study site

The 38 study lakes were located on the Canadian Shield in a 30,000 km^2 area surrounding Réservoir Gouin in Haute-Mauricie, Québec (48°50′N, 75°00′W; see map in [11]). Cut lakes ($n = 9$) had 9 to 72% of their catchment area clear-cut in 1995. Seven burnt lakes had approximately 100% of their catchments cleared by high-intensity fires, also in 1995, whereas two others lakes, FP2 and FP30, had only 50 and 75%, respectively, of their catchments burnt. The catchments of reference lakes ($n = 20$) had remained undisturbed for at least 70 years. The 38 drainage lakes were selected on the basis of comparable size, depth, and catchment morphometry (Table 1). A detailed description of the selection criteria and the morphometric characteristics of the three groups of lakes has been given previously [11]. Wetlands were present in the catchments of 23 lakes and occupied in average 1.7% of the catchment area. In one cut lake (C23) and in two reference lakes (N88 and N107), wetland area represented approximately 6% of the catchment area.

Ancillary water samples

Water-quality sampling was conducted during the summer of 1996 and 1997. Duplicate integrated samples were taken

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Table 1. Average morphometric, chemical, and physical characteristics of the study lakes[a]

	Reference (n = 20)				Burnt (n = 9)				Logged (n = 9)			
	Mean[a]	SE	Min.	Max.	Mean	SE	Min.	Max.	Mean	SE	Min.	Max.
Lake area (km^2)	0.4	0.1	0.2	0.8	0.4	0.1	0.2	0.6	0.6	0.1	0.2	2.3
Drainage area (km^2)	2.2	0.2	0.5	4.8	4.9	1.9	0.6	19.7	3.4	1.0	0.6	10.3
Maximum depth (m)	12.5	0.7	7.0	23.0	17.0	1.8	10.0	34.0	14.0	1.9	5.0	30.0
% Drainage area as wetlands	2.0	0.4	0	7.0	0.9	0.4	0	3.6	2.3	0.5	0.0	6.0
Disturbed area (km^2)	0	0	0	0	4.6	0.3	0.6	19.5	1.7	1.9	0.1	7.5
pH	6.5	0.1	5.8	7.0	6.6	0.1	5.7	7.2	6.4	0.1	5.8	7.0
Alkalinity (μEq/L)	50.3	4.9	18.7	110.0	64.2	9.8	17.2	146.1	54.2	9.4	15.3	47.6
Dissolved organic carbon (mg/L)	5.2	0.3	2.8	9.2	5.8	0.5	3.1	8.7	7.5	0.8	3.0	12.4
Dissolved organic carbon loading to lakes (mol C/m^2 lake/year)	2.7	0.4	1.0	9.3	4.7	2.1	0.4	9.7	5.1	0.9	0.8	9.5
Light attenuation coefficient (m^{-1})	1.0	0.1	0.5	1.7	1.2	0.1	0.5	1.8	1.5	0.2	0.6	2.6
Secchi depth (m)	4.0	0.2	2.2	6.0	3.4	0.4	2.1	5.6	3.0	0.3	1.4	5.0
Chlorophyll *a* (μg/L)	1.9	0.1	1.0	3.2	3.3	0.3	1.6	4.7	2.2	0.2	1.3	2.7
Total phosphorus (μg/L)	6.7	0.3	4.7	10.3	11.8	0.9	7.6	17.3	9.6	0.9	5.6	15.8
Total nitrogen (μg/L)	219.1	7.9	149.5	332.1	332.2	36.1	160.0	746.6	255.5	12.6	160.0	303.0
Ca^{2+} (mg/L)	1.5	0.1	1.0	2.5	2.1	0.1	1.4	2.7	1.5	0.1	1.1	2.2
SO_4^{2-} (mg/L)	0.9	0.1	0.5	1.2	1.6	0.1	1.0	2.1	0.8	0.1	0.5	1.1

[a] For each group of lakes, water-quality variables correspond to means obtained using, for each lake, summer values of the year when fish were caught (1996 or 1997). SE = standard error.

from the euphotic zone of the deepest point of the lakes. Measured parameters included DOC, dissolved oxygen, pH, alkalinity, total phosphorus (P) and N, sulfate, calcium, chlorophyll *a*, light attenuation, and Secchi depth. The analytical methods have been described previously [11]. Dissolved organic carbon loading to the lakes (mol C/m^2/year) was estimated by Lamontagne et al. [5].

Fish and invertebrates collection

Fish were sampled during the summer of 1996 or 1997 simultaneously with a study of fish communities in the 38 lakes. Fish collection, relative abundance, length, and species distribution in the three groups of lakes have been given previously [12]. Data regarding fish composition in each of the study lakes (Table 2) were provided by Pierre Magnan (Université de Québec à Trois Rivières, PQ, Canada, unpublished data). Fish for Hg analysis were selected among all individuals collected in each lake. Between 6 and 23 specimens of up to the three most abundant species in each lake were selected to maximize the range in size of the different species. A total of 642 individuals distributed in 63 fish populations were analyzed. Total length, weight, and sex were recorded for all fish. A boneless, skinless filet of dorsal muscle was removed from each fish, frozen, and freeze-dried. Fish assemblages varied from lake to lake. The most abundant species collected were northern pike (*Esox lucius*), walleye (*Stizostedion vitreum*), yellow perch (*Perca flavescens*), white sucker (*Catostomus commersoni*), whitefish (*Coregonus clupeaformis*), lake trout (*Salvelinus namaycush*), brook trout (*Salvelinus fontinalis*), and burbot (*Lota lota*). The mean total length for all individuals from a given species was used as a standardized length in the among-populations comparisons. These standardized lengths were as follows: Northern pike, 560 mm; walleye, 318 mm; yellow perch, 135 mm; white sucker, 325 mm; lake whitefish, 310 mm; lake trout, 365 mm; brook trout, 240 mm; and burbot, 590 mm.

Mayfly nymphs (*Cloeon* sp., Ephemeroptera) were used for correction of among-lake variations in baseline $\delta^{15}N$. These mayflies were considered to be primary consumers [13], and they were found associated with the vegetation in the littoral zone of the lakes (depth, <2 m). Between 2 and 45 individuals were collected in two stations in each lake using handheld dip nets (mesh size, 500 μm). The nymphs were sorted and counted. Nymphs for taxonomic identification were preserved on 4% formaldehyde, and subsamples for $\delta^{15}N$ determination were kept frozen until they could be freeze-dried.

Analytical methods

In the present study, we assumed that MeHg, the form of Hg that is biomagnified, accounted for the major part of total Hg in fish [14], particularly in those species occupying higher trophic levels. Mercury concentrations were measured on subsamples (0.1 g) of fish dorsal muscle by cold-vapor atomic absorption spectrometry [15] after hot acid digestion in Teflon® vessels using trace metal–grade nitric acid. The average coefficient of variation for Hg measured in 70 duplicate samples was 6%. Standard Hg solutions of 1.0 ng/ml were analyzed every 15 samples to ensure accuracy and precision throughout analyses in each batch of samples. Reference material (Hg = 4.64 ± 0.26 mg/kg [mean ± SD throughout]; DORM-2; National Research Council of Canada, Montreal, PQ) was run every 25 samples. The accuracy of analyses of the reference material was 98.7% ± 4.1% (n = 55). Recovery of spikes averaged 95% ± 6% (n = 27). Unless specified, all values are reported on a dry-mass basis. To allow comparisons with literature data, a conversion factor of 0.2 was used to express concentrations on a wet-mass basis; this factor corresponds to the average of values observed for 25 individuals from different fish species collected in the study lakes (0.197 ± 0.017).

Nitrogen isotopic ratios were measured on 0.7 mg of freeze-dried, powdered samples of mayfly nymphs and fish muscle. One or two composite samples of mayflies (n = 2–5 individuals) were analyzed for each lake. For fish, at least three individuals (range, 3–11) per species collected in each lake were used for isotopic determinations (total, 234 individuals). The analyses were performed in the GEOTOP Laboratory (Université du Québec à Montréal, Montréal, PQ, Canada) on a continuous-flow Isoprime mass spectrometer coupled to a CHN analyzer (model NC 1500; Carlo Erba, Milan, Italy). Replicate analyses of five composite samples of mayflies and

Table 2. Mercury (Hg) concentrations (mg/kg dry biomass) in fish of standardized length and their $\delta^{15}N$ signature (‰, corrected for variations in baseline) for sampled fish (Sp) and other fish species (other spp.) in the study lakes[a]

Reference lakes (n = 19)					Burnt lakes (n = 9)					Logged lakes (n = 9)				
Lake	Sp	$\delta^{15}N$	Hg ± SD	Other spp.	Lake	Sp	$\delta^{15}N$	Hg ± SD	Other spp.	Lake	Sp	$\delta^{15}N$	Hg ± SD	Other spp.
N5	WS	7.80	1.13 ± 0.14	CY	FP2	NP	8.33	6.44 ± 1.13	GS, WS, WF, PE	C2	NP	9.34	3.06 ± 0.46	CY, WS, PE
N16	BT	7.66	0.53 ± 0.08	CY	FP15	WF	7.70	1.68 ± 0.05	CY, WS, PE	C9	NP	9.05	2.71 ± 0.31	WS
N35	PE	8.18	0.68 ± 0.07	WA		WA	9.22	2.00 ± 0.15			PE	7.43	1.51 ± 0.13	
N43	WF	6.46	0.68 ± 0.03	WS, PE, LO		NP	9.40	2.48 ± 0.57		C12	WS	6.36	1.18 ± 0.03	CY, WF, PE, NP
	NP	8.25	1.05 ± 0.14		FP24	NP	8.90	2.13 ± 0.11	PE		WA	9.39	1.87 ± 0.30	
N55	NP	10.73	2.07 ± 0.44	CY, GS, WS, PE	FP27	WS	4.02	0.78 ± 0.07	CY, LO	C23	WA	8.15	2.78 ± 0.24	WF, PE, NP
N56	NP	11.62	2.55 ± 0.43	CY, WS		LT	5.48	0.59 ± 0.09		C24	LO	9.33	2.56 ± 0.38	PE, CY
N63	NP	8.00	2.10 ± 0.33	CY, WS	FP30	WF	4.37	1.47 ± 0.23	WS, PE		NP	9.94	3.58 ± 0.40	
N70	WF	6.45	0.86 ± 0.08	CY, WS, PE, LO		NP	7.20	5.15 ± 0.33		C29	WS	6.37	1.08 ± 0.12	BT
	WA	7.97	1.00 ± 0.07			WA	6.65	3.73 ± 0.78		C40	NP	7.79	2.39 ± 0.28	GS, PE
N82	WS	6.81	1.11 ± 0.12	CY	FP31	BT	6.60	0.67 ± 0.12	PD	C44	WF	6.24	0.99 ± 0.05	CY, GS, WS, PE, NP
N84	WA	8.25	0.98 ± 0.38	CY, WS, PE, NP	FP32	WF	6.02	0.51 ± 0.05	PD, LO		PE	6.62	0.99 ± 0.09	
N88	WS	7.26	0.90 ± 0.08	PD		NP	8.40	2.24 ± 0.35		C48	WS	5.16	2.41 ± 0.05	CY, WF, LO
	NP	9.27	1.56 ± 0.27			WA	8.27	1.29 ± 0.07			BT	7.07	1.35 ± 0.08	
N89	WF	6.09	0.54 ± 0.09	CY, WS, PE		PE	6.94	0.81 ± 0.04			LT	7.34	1.63 ± 0.13	
	WA	7.52	1.00 ± 0.12			WS	5.67	0.35 ± 0.03						
	NP	8.37	1.00 ± 0.07		FBP9	WS	5.38	1.02 ± 0.08	PD					
N106	WS	4.66	0.55 ± 0.11	CY, PD, BT		NP	8.95	1.94 ± 0.43						
N107	NP	8.36	2.59 ± 0.10	CY, WS, PE	FBP10	WS	5.52	1.38 ± 0.17						
	PE	6.42	1.08 ± 0.19			PE	6.93	0.84 ± 0.08						
N120	WS	5.80	0.80 ± 0.07	CY, PE, LO, BT		NP	8.33	1.90 ± 0.35						
N122	NP	7.97	1.48 ± 0.11											
P25	WF	5.90	1.18 ± 0.15	WS, PE, NP										
P109	WS	6.01	1.05 ± 0.31	CY, PD										
	PE	6.42	1.08 ± 0.19											
P110	WS	6.97	1.23 ± 0.15	CY, BT, PE										

[a] All study locations are in Quebec (Canada). BT = brook trout (*Salvelinus fontinalis*); CY = minnows (*Cyprnus* sp.); GS = golden shiners (*Notemigonus crysoleucas*); LO = burbot (*Lota lota*); LT = lake trout (*Salvelinus namaycush*); NP = northern pike (*Esox lucius*); PD = pearl dace (*Semotilus margarita*); PE = yellow perch (*Perca flavescens*); WA = walleye (*Stizostedion vitreum*); WF = lake whitefish (*Coregonus clupeaformis*); WS = white sucker (*Catostomus commersoni*).

17 individual samples of fish from different species agreed within 0.3‰. Ratios of heavy to light isotopes were expressed as δ values (i.e., deviations in terms of ‰ from a standard reference material, in this case atmospheric N).

Data analysis

Mercury concentrations were regressed against total length for each species of fish in each lake. Significant correlations between these two variables were observed in 53 of the 63 populations (average $r = +0.82 \pm 0.11$) (Fig. 1). These regressions were used to estimate Hg levels in fish of a standardized length. For five white sucker and four whitefish populations, the relationship between Hg and fish length was not significant, and Hg mean values unadjusted for fish size were used. Northern pike length in one reference lake (N59) was not significantly correlated with Hg concentrations either, likely because of the small sample size ($n = 6$). All northern pike collected in Lake N59 were larger than the 560-mm standardized length for this species. Therefore, Hg in pike from this lake was excluded from the analyses.

As with Hg concentrations, simple regressions were used to estimate $\delta^{15}N$ for fish of a standardized length. For nonsignificant length–$\delta^{15}N$ regressions, the mean $\delta^{15}N$ for individuals with a total length equal or close to the standardized length was calculated.

Variations in the $\delta^{15}N$ of primary producers can result in a wide range of $\delta^{15}N$ values for consumers [16] and invalidate the use of $\delta^{15}N$ in among-system comparisons. Such variations were accounted for by subtracting the $\delta^{15}N$ measured in mayflies from the $\delta^{15}N$ measured in fish. We used primary consumers rather than primary producers for baseline corrections, because the latter are more prone to variability in $\delta^{15}N$ as a result of their shorter life span [17]. We assumed that the difference between a given species of fish and mayflies within a lake should be proportional to the trophic position of fish relative to primary consumers. Under the above assumptions, the trophic position of the fish was given as

$$\Lambda_{\text{fish}} = \left[\frac{(\delta^{15}N_{\text{fish}} - \delta^{15}N_{\text{mayfly}})}{3.4}\right] + 2 \qquad (1)$$

where Λ_{fish} is the trophic position and $\delta^{15}N$ is the $\delta^{15}N$ measured for fish or mayfly. The constant 3.4 represents the $\delta^{15}N$ increment per unit of trophic level [16], and the constant 2 is the trophic level of mayflies.

Mercury concentrations and $\delta^{15}N$ values for fish of a standardized length from each population were used to compare the trends in Hg accumulation in cut, burnt, and reference lakes. This relationship was represented by equations in the form $\log_e Hg = a + b \cdot \delta^{15}N$, as proposed by Broman et al. [18]. In this model, $\log_e$ is the natural logarithm of Hg, the a term reflects Hg bioavailability, which is influenced, in part, by Hg load to the lakes, whereas the slope b provides an estimation of the biomagnification power of Hg by ‰ $\delta^{15}N$. Analysis of covariance (ANCOVA) with fish $\delta^{15}N$ as a covariate was used to compare Hg concentrations in fish populations from the three groups of lakes. Pearson correlation coefficients (r) between Hg contents in fish and environmental and biological variables were calculated separately for cut, burnt, and reference lakes, as well as for pooled lakes. Among-treatment differences in water quality and biological variables were tested using analysis of variance or the nonparametric Kruskal–Wallis test. When necessary, statistical analyses were conducted using log-transformed data to achieve normality and homoscedasticity.

RESULTS

Water quality

Chemical and physical characteristics of the study lakes are shown in Table 1. Several significant differences in water-quality variables have been observed among the three groups of lakes and discussed in detail previously [11]. In summary, concentrations of DOC and light extinction tended to be the highest in logged lakes, whereas concentrations of chlorophyll *a*, Ca^{2+}, SO_4^{2-}, total P, and total N generally were highest in the burnt group. Among-group differences in pH were not significant.

Hg concentration in fish

Mercury concentrations ranged from 0.2 to 20.2 μg/g dry mass among all fish sizes and species. For all species excluding whitefish and white sucker, Hg concentrations tended to increase as a function of fish length within a given population. Examples of these correlations are shown in Figure 1. Average Hg concentrations for fish of a standardized length were the highest in top predatory northern pike in the three groups of lakes (Fig. 2). Other piscivorous species, walleye and burbot, also showed high mean Hg levels in lakes with logged and fire-impacted catchments. In undisturbed lakes, the mean Hg concentration in walleye was not significantly different from that in species from lower trophic levels. Although Hg concentrations in general tended to be higher in fish from impacted lakes, one should note that the among-group variations in Figure 2 are not directly comparable because of variations in fish trophic position, as discussed below.

$\delta^{15}N$ signal and trophic level characterization

The 38 lakes were quite different in terms of fish composition, with the number of species in each lake varying from one to six (Table 2). Consequently, food-chain length as well as the $\delta^{15}N$ signature of a given species varied from lake to lake. The trophic position occupied by the different species (Table 3) indicated a progressive $\delta^{15}N$ enrichment up the food chain and followed the pattern of white sucker ≤ whitefish ≤ brook trout ≤ lake trout ≤ yellow perch ≤ burbot ≤ walleye ≤ pike.

The $\delta^{15}N$ in mayfly nymphs ranged from −2.7 to +3.3‰ (Table 3), which represented a 6‰ variation. Assuming that $\delta^{15}N$ increases, on average, by 3.4‰ from prey to predator [16], this variation corresponded to 1.8 trophic levels. The average $\delta^{15}N$ of mayfly from burnt lakes (1.8‰) was significantly higher (analysis of variance, $p < 0.05$) than in cut (−0.2‰) and reference (−0.5‰) lakes. Similar to mayfly nymphs, all species of fish found in burnt lakes showed consistently higher $\delta^{15}N$ than the same species collected in reference and cut lakes (Kruskal–Wallis, $p < 0.0001$). Differences in $\delta^{15}N$ between reference and cut groups were not significant except regarding whitefish, for which $\delta^{15}N$ was significantly higher in cut lakes. For white sucker, whitefish, yellow perch, walleye, and northern pike populations, differences between minimum and maximum values of $\delta^{15}N$ for fish of standardized length varied from 3.3 to 4.5‰ (Table 3), which is equal or superior to one trophic level. For other species only found in a small number of lakes (brook trout, lake trout, and burbot), differences between minimum and maximum values of $\delta^{15}N$

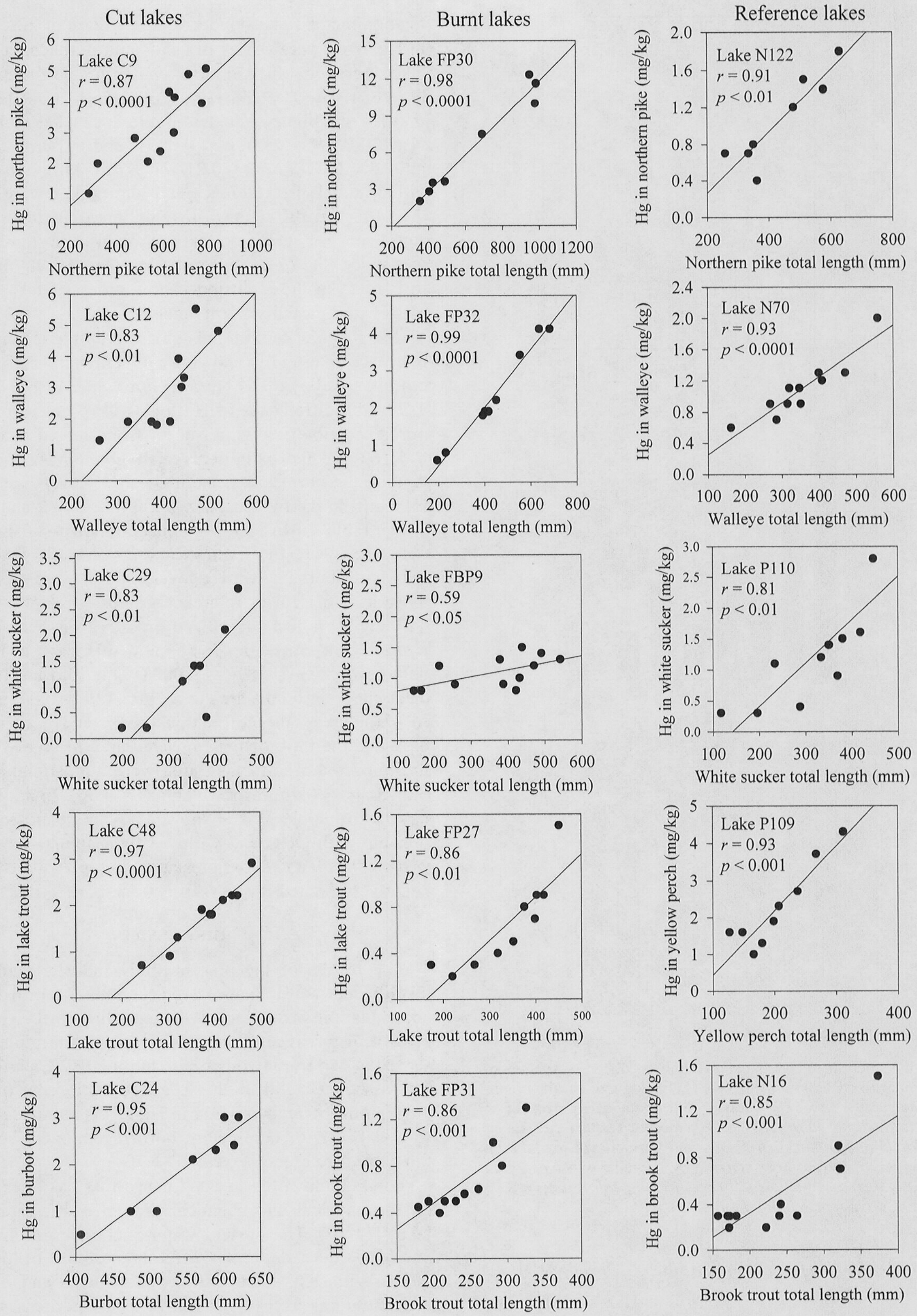

Fig. 1. Relationships between mercury (Hg) concentration, on a dry-mass basis, and fish length for 15 of the study populations from lakes with logged, burnt, or undisturbed catchments (PQ, Canada).

corresponded to significantly less than one trophic level (Table 3).

In 11 populations of pike, walleye, and perch, $\delta^{15}N$ was determined in more than six individuals spanning a broad range in length. For these populations, significant correlations (average $r = +0.87$) were observed between $\delta^{15}N$ and fish length (Fig. 3). For whitefish and white sucker populations, $\delta^{15}N$ was not significantly correlated to fish length.

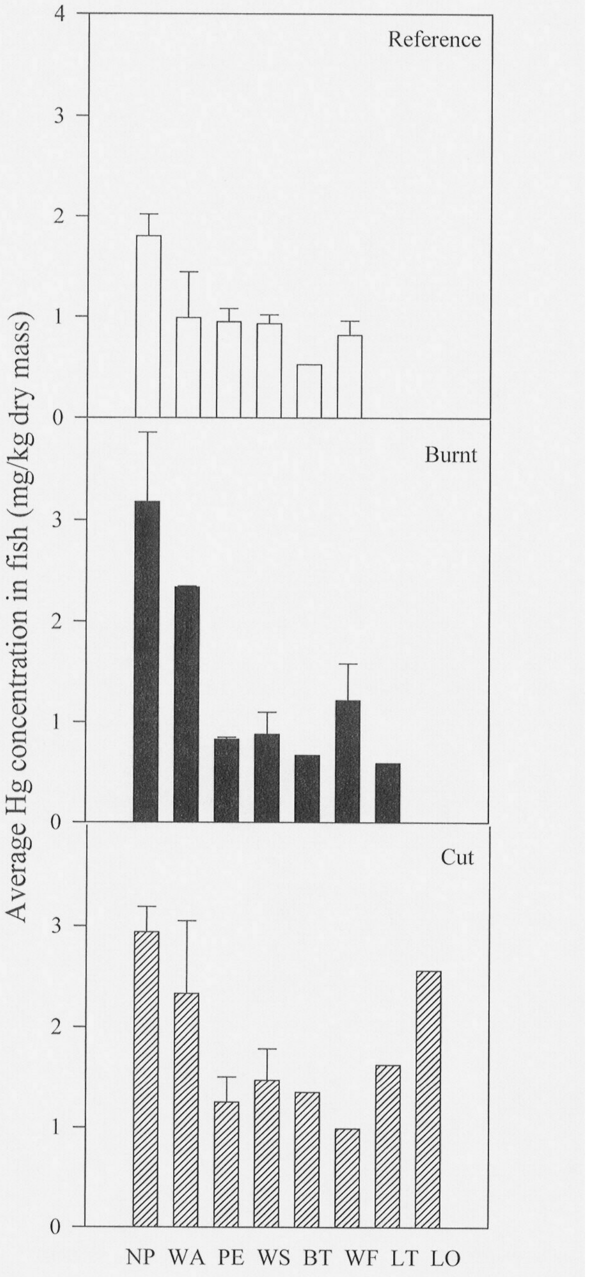

Fig. 2. Average mercury (Hg) concentrations in different species of fish of a standardized length from lakes with logged, burnt, or undisturbed catchments. Error bars correspond to standard error. BT = brook trout (*Salvelinus fontinalis*); LO = burbot (*Lota lota*); LT = lake trout (*Salvelinus namaycush*); NP = northern pike (*Esox lucius*); PE = yellow perch (*Perca flavescens*); WA = walleye (*Stizostedion vitreum*); WF = lake whitefish (*Coregonus clupeaformis*); WS = white sucker (*Catostomus commersoni*).

Relationship between Hg concentration in fish and $\delta^{15}N$ signature

Mercury concentrations in fish from the three groups of lakes increased with δ^{15}N following the model $\log_e$Hg = a + $b \cdot \delta^{15}$N (r^2 = 0.52, 0.49, and 0.30 for cut, reference, and burnt lakes, respectively; $p < 0.01$) (Fig. 4). The slopes of the models (i.e., the b term) did not vary significantly (ANCOVA, F = 0.06, p = 0.81) and were equal to 0.20 ± 0.042 (reference lakes), 0.22 ± 0.062 (cut lakes), and 0.26 ± 0.089 (burnt lakes). These slopes, when exponentially transformed, gave an estimation of the biomagnification factor. The biomagnification factor corresponded to an increase of 22, 25, and 29% in Hg levels per ‰ δ^{15}N in fish from reference, cut, and burnt lakes, respectively. Mercury availability to organisms at the base of the food chain (i.e., the a term) in cut lakes (−1.07 ± 0.16) was higher than that in reference lakes (−1.43 ± 0.32, ANCOVA, F = 21.9, $p < 0.0001$) and in burnt lakes (−1.46 ± 0.65, ANCOVA, F = 0.21, p = 0.65). Although the a values in the models for reference and burnt lakes were very close, the observed differences in relation to cut lakes were only significant for reference lakes. Variability in the burnt group was twice that observed in the reference group and resulted, in part, from the inclusion in this group of lakes with completely and partially burnt catchments.

Mercury concentrations tended to be higher in cut than in reference lakes in fish showing a δ^{15}N higher than 7.5‰ (northern pike, walleye, and burbot). For values of δ^{15}N lower than 7.5‰, the differences in Hg levels were not significant. The only exception was white sucker from the cut lake C48 ($\delta^{15}N$ = 5.16‰), which exhibited a relatively high Hg concentration. Among the burnt lakes, the highest Hg levels were observed in fish from the two partially burnt lakes, FP2 and FP30 (Fig. 4 and Table 2). In fact, Hg concentrations in fish from these two lakes were the highest among all study lakes. For the remaining burnt lakes, Hg levels in fish were comparable to those in either cut or reference groups.

Mercury in fish normalized for δ^{15}N from cut lakes showed a significant correlation with ratio of clear-cut area to lake volume (r = +0.82, p = 0.0003) (Fig. 5a) and the ratio of clear-cut area to lake area (r = +0.74, p = 0.0023) (Fig. 5b). In burnt lakes, the correlations between Hg normalized for δ^{15}N and the ratio of fire-impacted area to lake volume or of fire-impacted area to lake area were almost significant (r = +0.45 and +0.46, respectively; p = 0.06). In addition, Hg in fish in the pooled lakes showed a significant positive relationship with DOC and with the light attenuation coefficient (r = +0.41 and +0.42, respectively; $p < 0.001$) and was negatively related to pH (r = −0.38, $p < 0.01$).

DISCUSSION

The results of the present study indicate that watershed disturbances can influence Hg accumulation in fish. Mercury accumulation varied, however, according to the type and degree of disturbance. The highest Hg concentrations were observed in fish from two partially burnt lakes. Clear-cut logging was associated with high Hg concentrations in fish as well. No significant augmentation in Hg levels was observed in fish from lakes with completely burnt catchments compared to those from reference lakes.

Forest removal by fire and harvesting has different effects on the physical and chemical properties of watersheds and lakes [11,19]. The present water-quality results indicate that one of the most important differential effects of logging is an increase in DOC loading from catchments and in the DOC concentrations in lakes [5,11]. In clear-cut areas, the organic soil is preserved, whereas intense fires tend to mineralize this layer. Therefore, higher DOC concentrations in cut lakes could explain the higher Hg levels observed in fish sampled in these lakes. Dissolved organic carbon may play an important role here, because it acts as a vector of Hg from terrestrial to aquatic systems [20]. Furthermore, DOC influences the chemical speciation of Hg by stimulating bacterial methylation [21], by decreasing MeHg photodegradation [22], and possibly, by decreasing the photoreduction of Hg(II) to Hg(0) in lakes [23].

Table 3. Average measured $\delta^{15}N$ (‰) of Ephemeroptera and fish of a standardized length in the study lakes

	Individuals (*n*)	Lakes (*n*)	Mean	Standard deviation	Min.	Max.
Mayfly nymphs	172	38	0.2	1.6	−2.4	3.3
White sucker	69	13	7.1	1.3	4.8	9.1
Whitefish	48	9	7.8	1.1	5.7	9.6
Brook trout	11	3	7.8	0.4	7.1	8.4
Lake trout	6	2	8.2	0.4	7.6	8.7
Yellow perch	50	7	8.1	1.0	6.1	10.6
Burbot	3	1	8.2	0.4	8.0	8.5
Walleye	35	8	9.9	1.6	7.9	12.0
Northern pike	93	19	8.9	0.7	7.1	10.4

These mechanisms support the positive relationships between Hg concentrations in fish and DOC (or correlated variables: DOC loading, light attenuation) observed in our lakes and in other studies [24–26]. Similarly, the positive correlations between Hg in fish and the extent of catchment area logged when normalized to lake area or volume seem to reflect the influence of organic matter in Hg loading to lakes. Indeed, these two impact ratios showed significant correlations with DOC in the cut group [11]. These findings are consistent with those of Porvari et al. [6], who observed significant increases in total Hg and MeHg concentrations and a positive correlation between total Hg and organic carbon in runoff water of boreal forest catchments following clear-cutting.

Variations in other environmental variables related to watershed disturbances also could affect Hg concentrations in fish from cut and burnt lakes. For example, the increase in nutrient loading (total P and total N) following fire resulted in significantly higher algal biomass in burnt lakes compared to that in reference lakes [27]. High phytoplankton biomass may cause a biological dilution of the available Hg pool, thereby reducing Hg concentrations in organisms, including those occupying higher trophic levels [28]. Alternatively, lower light penetration may account for the lower algal biomass in cut lakes as compared with burnt lakes [27]. In addition, during

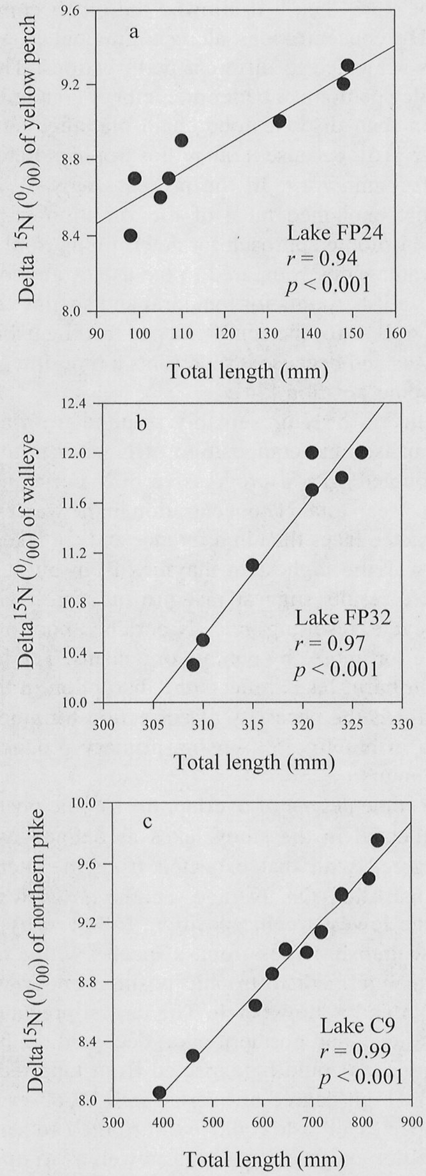

Fig. 3. Relationships between $\delta^{15}N$ and fish length for yellow perch, walleye, and northern pike from two fire-impacted lakes (FP24 and FP32) and from one lake with logged catchment (C9).

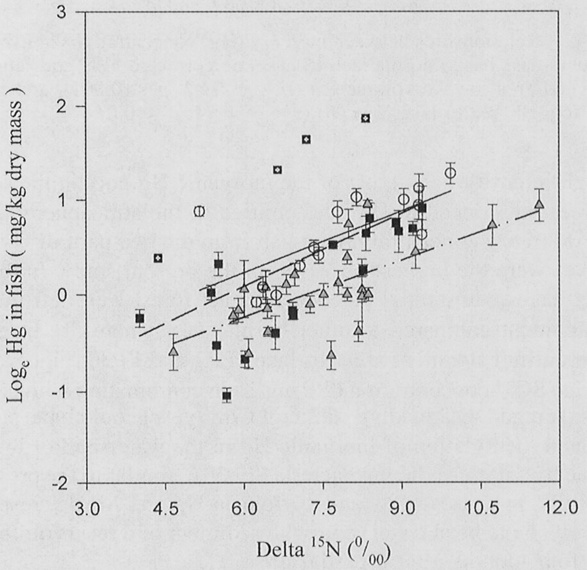

Fig. 4. Relationships between mercury (Hg) concentrations and fish $\delta^{15}N$ for reference lakes (triangles and dash-dot-dot line), and for lakes with cut (open circles and solid line) and burnt catchments (black and dotted squares for completely and partially burnt catchments, respectively, and dashed lines). Points represent fish of a standardized length from 62 populations. The $\delta^{15}N$ values are corrected for baseline variations. Error bars correspond to standard deviation.

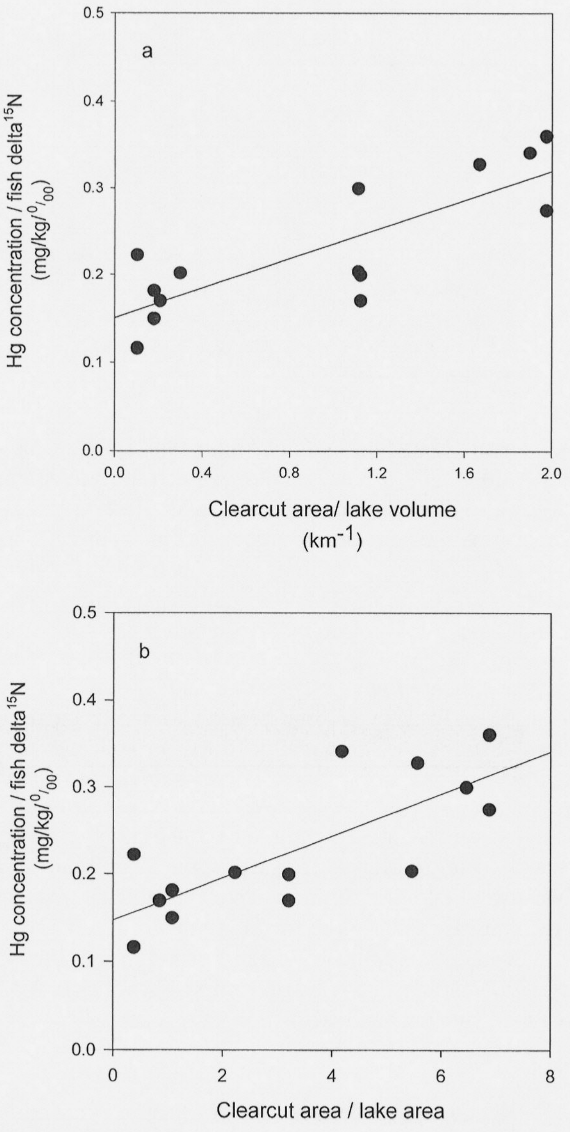

Fig. 5. Relationships between mercury (Hg) concentrations in fish (on a dry-mass basis) normalized to baseline corrected $\delta^{15}N$ and ratio of logged area to lake volume (**a**) ($r = +0.82$, $p < 0.001$), and ratio of logged area to lake area (**b**) ($r = +0.74$, $p < 0.01$).

high-intensity fires, most of the inorganic Hg pool in the soils could be reduced to Hg^0 and emitted to the atmosphere [7].

Mercury concentrations in fish from the two partially burnt lakes were the highest observed in the present study. In these two lakes, substantial patches of intact forest were left in the catchment compared to other burnt lakes, where fire cleared the entire catchment area. In lakes FP2 and FP30, higher average SO_4^{2-} concentration (2.0 mg/L) in combination with higher average DOC loading (6.2 mol C/m²/year) could have stimulated methylation of inorganic Hg in the watershed or in the lake by sulfate-reducing bacteria [29,30]. Similar to the present results, increases of 8- and 10-fold in Hg and MeHg, respectively, have been observed in the sediment of a reservoir three to four months after a partial fire [31].

The significant positive relationship between total Hg and fish $\delta^{15}N$ indicated an overall biomagnification of Hg in disturbed and undisturbed lakes. Biomagnification factors were comparable among cut, burnt, and reference groups and consistent with the range of values observed in temperate freshwater fish [32] as well as in an arctic marine food web [33]. Therefore, among-group variations in Hg concentrations of fish cannot be attributed to differences in the biomagnification power of Hg by ‰ $\delta^{15}N$. Rather, our findings suggest that the higher Hg in fish from lakes with logged catchments results from higher bioavailability of Hg in these lakes. In a previous study, we observed higher MeHg levels in zooplankton from cut lakes comparatively to reference and burnt lakes [8]. Alterations of Hg cycling linked to watershed disturbances therefore seem to affect primary consumers and, subsequently, organisms occupying higher trophic levels.

In comparisons of Hg levels in fish from different systems, it is particularly important to take trophic position into account in combination with watershed and water characteristics. In the present study, the use of $\delta^{15}N$ to estimate trophic position eliminated some of the problems encountered when comparing Hg concentrations in fish from different lakes. First, $\delta^{15}N$ measurements allowed the simultaneous treatment of different species. Second, it circumvented the statistical problems related to variations in fish community composition from one lake to another, with a given species seldom being found in all systems at the same time. Third, it allowed the direct comparison of trends in Hg concentrations along a gradient in $\delta^{15}N$ in fish from lakes subjected to different perturbations. The $\delta^{15}N$-defined trophic position is a better predictor of contaminant biomagnification than discrete food-chain classification based on dietary data [16], because it quantifies trophic interactions and accounts for omnivory. In the present study, $\delta^{15}N$ was the variable that explained most of the variation of Hg in fish (42%). The isotopic approach for determining trophic position also is advantageous compared to the use of gut content data, both because it accounts for temporal and spatial variations in feeding as well as for the complexity of feeding habits at lower trophic levels and because it represents a time-integrated measure of trophic position [34].

Variability in $\delta^{15}N$ presumably resulted from among-lake variations in isotopic composition of N sources to algae and bacteria coupled with a progressive $\delta^{15}N$ enrichment up the food chain. Mean total N concentration in the water was higher in fire-impacted lakes than in reference and cut lakes [11]. The $\delta^{15}N$ also was the highest in mayflies from burnt lakes. Together, these results suggest that fire interferes with the terrestrial N cycle, resulting in a ^{15}N-enriched pool of inorganic N available for uptake by benthic organisms. The higher $\delta^{15}N$ of fish from burnt lakes reflects the disruption in the N cycle and strengthens the necessity of correction for among-system differences in baseline $\delta^{15}N$ using primary producers or primary consumers.

Despite some degree of overlap, the trophic position of the species collected in the study lakes as defined using stable isotopes agreed with that expected from the literature data based on fish diet. On average, benthivorous white sucker occupied the lowest trophic position, followed by planktivorous lake whitefish. Brook trout, a species with a mixed diet, occupied an intermediate trophic position and was followed by lake trout and yellow perch. The largest predatory species (burbot, walleye, and northern pike) occupied the highest trophic positions, as could be expected from long-term diet averaging. The high degree of overlap in $\delta^{15}N$ observed for the different species of fish could be attributed to variations in trophic position of prey organisms as well as to opportunistic feeding habits. Analyses of the gut content of 3,000 specimens of pike, walleye, perch, and white sucker from the study lakes (P. Magnan et al., Université de Québec à Trois Rivières, PQ, Canada, unpublished data) showed a large variety of prey items. The diet of each of the above species included terrestrial invertebrates, littoral and profundal benthic invertebrates, aquatic pupae, leeches, and fish. Zooplankton was found in the stomach of all fish species except pike. These findings support the high level of omnivory suggested by the range of $\delta^{15}N$ for those species of fish.

As expected, piscivorous species occupying higher trophic levels showed the highest concentrations of Hg in the three groups of lakes. In all northern pike, walleye, and burbot of standardized length found in cut lakes and in the two partially burnt lakes, Hg concentrations exceeded the recommended limit for human consumption (0.5 μg/g wet biomass or 2.5 μg/g dry biomass) from the World Health Organization. This limit was surpassed only in 18% of piscivorous species populations found in reference lakes and in no species from completely burnt lakes. High Hg concentrations in fish from forest-harvested and partially burnt lakes thus may reflect increased exposure to MeHg relative to lakes not having these watershed disturbances.

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