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Mercury in the Muscle Tissue of Fish from Three Northern Maine Lakes

John J. Akielaszek and Terry A. Haines

U.S. Fish and Wildlife Service, Columbia National Fisheries Research Laboratory, Field Research Station, Department of Zoology, University of Maine, Orono, ME 04469

There is evidence that fish in Canadian wilderness areas exhibit elevated mercury levels because of the oligotrophic nature of the lakes they inhabit (BROUZES et al. 1977). D'ITRI et al. (1971) reported higher levels of mercury in trout from oligotrophic waters than in trout from eutrophic waters in unpolluted areas. They attributed part of the difference to differences in the food of the fish in the two types of waters. Position on the trophic scale is an important factor determining the degree of bioaccumulation of mercury by aquatic organisms, especially fish.

Elevated mercury levels have been reported in fish from three wilderness lakes in northern Maine (NORMANDEAU ASSOCIATES 1978). Concentrations exceeding 0.50 µg/g were detected in the muscle tissue of many of the fish.

We report the levels of mercury in the muscle tissue of brook trout (*Salvelinus fontinalis*) and lake trout (*S. namaycush*) from three northern Maine lakes. Mercury levels in fish from two wilderness lakes in the same drainage basin were compared with each other, and in turn with those in fish from a lake in a separate drainage basin. The fish species composition in one of the wilderness lakes, Cliff Lake, is different from that in the other two lakes, enabling us to analyze the effects of trophic structure on mercury concentration in top carnivores. It is unlikely that mercury from agricultural, geological, or local industrial sources occurs in these lakes.

STUDY LAKES

Some previous information on mercury levels in lake trout was available for Eagle Lake (Allagash River drainage) and St. Froid Lake (Fish River drainage; NORMANDEAU ASSOCIATES 1978). Fish species compositions in these two lakes are similar. Mercury in fish from Cliff Lake had not been studied previously. This lake lacks an important forage fish, rainbow smelt (*Osmerus mordax*), on which lake trout commonly feed. Some physical and chemical characteristics of the lakes are given in Table 1.

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TABLE 1. Some physical and chemical characteristics of three northern Maine lakes (October 1979).

Lake	Area (ha)	Maximum depth (m)	pH	Alkalinity (mg/l as $CaCO_3$)	Specific Conductance (μmhos/cm)
Eagle	2153	38	6.75	14.3	37
Cliff	228	20	6.90	26.0	51
St. Froid	972	35	7.15	25.2	56

Eagle Lake, known locally as Big Eagle Lake, is a large, deep oligotrophic lake in the upper reaches of the Allagash River drainage. It is protected as a part of the Allagash Wilderness Waterway and is accessible only by canoe and trail in the summer and by plane, snowmobile, and trail in the winter. Except for two camps (cabins), there is no human development in the lake watershed.

Cliff Lake, which is 9.6 km east of Eagle Lake's northern basin, is also deep and oligotrophic but much smaller than Eagle Lake. Cliff Lake is accessible by road but the shoreline is undeveloped except for a few campsites. Both Cliff and Eagle lakes drain north into Churchill Lake.

St. Froid Lake, which is in the Fish River drainage, lies 86 km northeast of Cliff and Eagle lakes; it is large, deep, and oligotrophic. Most of the watershed is undeveloped, but some sportsmen's camps and residences are present.

MATERIALS AND METHODS

Most of the fish were collected with graded-mesh experimental gillnets during summer and fall 1978, and some by angling through the ice in March 1979. Lake trout and brook trout were preferentially sought for analysis because of their importance as game fish. Other species such as rainbow smelt, lake whitefish (*Coregonus clupeaformis*), and burbot (*Lota lota*) were collected when possible. The fish were weighed, measured, placed into sterilized plastic bags and frozen for transport, usually by immersion in liquid nitrogen. The fish were kept frozen until analyzed.

For analysis, a strip of muscle extending from the opercular region to the caudal penduncle was removed from each fish and subsequently homogenized. A 0.5-g subsample of the homogenate was taken for analysis. The total mercury concentration in this muscle subsample was determined by flameless atomic absorption spectrophotometry (HOOVER et al. 1970). Blanks and spiked samples were included with each batch of 15 samples. Recovery of mercury added to spiked samples ranged from 89 to 106%, and the mercury content of the blanks was always below the limit of detection (<0.01 μg/ml). Each fish was analyzed for total mercury in muscle at least twice.

There is substantial evidence indicating a positive correlation between mercury concentration and length in many species (SCOTT & ARMSTRONG 1972). To determine the effect of trophic structure differences on mercury concentration in fish, we compared brook trout and lake trout from each lake. Analysis of covariance and the Newman-Keuls multiple range test (ZAR 1974) were used to test for differences among the slopes for the three lakes.

RESULTS

Total mercury in the muscle tissue of the fish examined ranged from 0.08 to 2.17 µg/g, wet weight. It exceeded 0.50 µg/g in 12 lake trout, 8 lake whitefish, 6 burbot, 2 rainbow smelt and 1 brook trout (Table 2).

TABLE 2. Mercury concentrations in the species examined from three northern Maine lakes.

Lake and Species	Total Length (cm)	Mercury Concentration (µg/g)
Eagle		
Lake Trout	18 - 69	0.13 - 1.11
Brook Trout	28 - 34	0.13 - 0.23
Lake Whitefish	25 - 51	0.30 - 2.17
Burbot	43 - 64	0.40 - 1.29
St. Froid		
Lake Trout	40 - 59	0.34 - 0.84
Brook Trout	24 - 43	0.08 - 0.58
Burbot	32 - 52	0.35 - 0.89
Rainbow Smelt	18 - 22	0.28 - 0.59
Cliff		
Lake Trout	35 - 47	0.10 - 0.23
Brook Trout	33 - 38	0.12 - 0.21

Slopes and correlation coefficients for mercury concentration in muscle vs. total length of brook trout and lake trout were closely similar for Eagle and St. Froid lakes (Fig. 1, Table 3) and significantly different from the same relationship for these species in Cliff Lake ($P < 0.001$), which also had a relatively low correlation coefficient.

DISCUSSION

High mercury concentrations appear to be characteristic of certain species of fish in certain locations in the Allagash Wilderness Waterway and also from other areas in northern Maine. Mercury concentrations in lake trout, burbot, and lake whitefish are higher than might be expected for fish inhabiting remote lakes.

Data on mercury levels in northern Maine fish did not exist before the study by NORMANDEAU ASSOCIATES (1978), and thus it is

TABLE 3. Equations and correlation coefficients of lines fitted for graph of mercury concentration vs. total length for lake trout and brook trout from three northern Maine lakes.

Lake	Equation[a]	Correlation coefficient
Eagle	Hg = 0.019 TL - 0.423	0.90
St. Froid	Hg = 0.021 TL - 0.411	0.88
Cliff	Hg = 0.005 TL - 0.022	0.44

[a]Hg = µg/g; TL = total length in centimeters

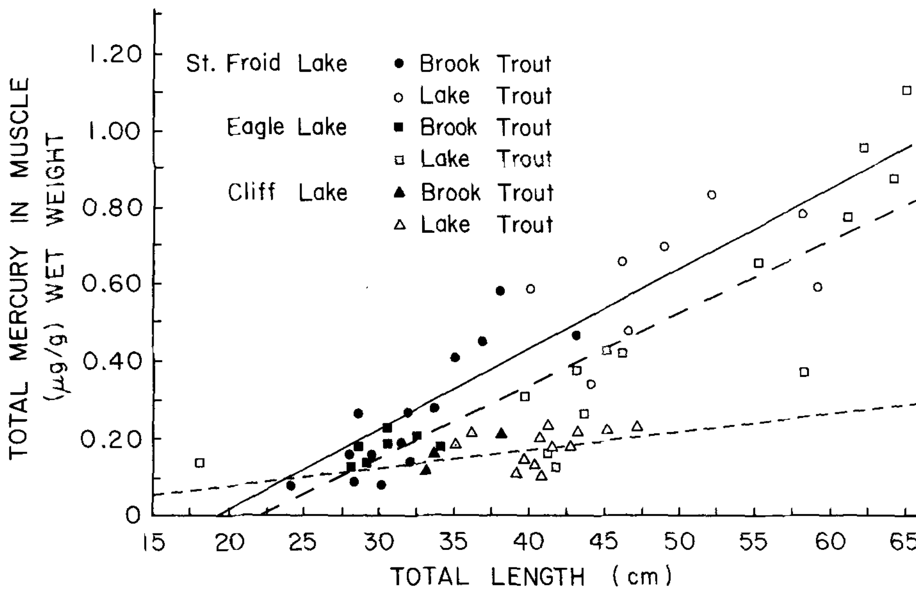

Figure 1. Total mercury concentration in muscle (µg/g wet weight) vs. length of brook trout and lake trout for each study lake (regression lines: St. Froid Lake —, Eagle Lake — —, Cliff Lake - - -).

difficult to determine whether mercury levels in these fish have always been elevated. There is evidence that these levels are not typical for fish in remote areas. Analyses in Scandinavia established that fish in unpolluted lakes may accumulate mercury up to 0.20 µg/g (JOHNELS et al. 1969), and ABERNATHY & CUMBIE (1977) believed that levels less than 0.50 µg/g should be characteristic of fish from unpolluted areas. In the present study 0.20 µg/g was near the lower end of the range of mercury levels measured, and mercury in 28% of the fish examined exceeded 0.50 µg/g. Mercury concentrations in lake trout from Eagle and St. Froid lakes were similar to those in lake trout from unpolluted lakes near sources of mercury contamination (Table 4). Mercury concentrations in lake whitefish from Eagle Lake were high and similar to those in lake whitefish from Ontario's heavily mercury-contaminated Wabigoon-English River system.

TABLE 4. Comparison of data on mercury in lake trout and lake whitefish collected by other researchers with data on these species from the present study.

Species and Location	Size Range	Mercury range (μg/g)
Lake Trout		
St. Froid Lake, ME	25-60 cm	0.45-0.94[a]
		0.34-0.84[b]
Eagle Lake, ME	50-70 cm	0.71-2.00[a]
		0.37-1.11[b]
Allagash Lake, ME	50-75 cm	0.72-1.01[a]
Umsaskis Lake, ME	25-40 cm	0.50-0.78[a]
Ontario lakes (near Wabigoon R.)	1500-2400 g	0.31-1.33[c]
St. Froid and Eagle Lakes	1500-2400 g	0.34-1.11[b]
Lake Whitefish		
Ball Lake, Ontario	---	0.20-1.70[d]
Eagle Lake, ME	---	0.30-2.17[b]
Clay Lake, Ontario	---	0.75-2.60[d]

[a]NORMANDEAU ASSOCIATES (1978); data for four lake trout from each of four northern Maine lakes.
[b]Present study.
[c]FIMREITE & REYNOLDS (1973); data for Ontario lakes near the heavily polluted Wabigoon-English River system.
[d]SHERBIN (1979); data for Ball and Clay lakes of the Wabigoon River.

The elevated mercury levels characteristic of certain species of fish examined in this study are probably the result of a combination of factors, including trophic status of the fish, increased uptake of mercury by fish under oligotrophic conditions, and some source of mercury input into these aquatic systems.

Rainbow Smelt and Mercury Uptake

Mercury content was high in lake trout and burbot from St. Froid and Eagle lakes but low in lake trout from Cliff Lake, which had levels similar to those in brook trout. The presence or absence of rainbow smelt may be one of the factors determining differences or similarities in mercury content of the piscivorous species in the three lakes. Fish species in the lakes are similar but Cliff Lake lacks rainbow smelt. Mean mercury levels in St. Froid Lake rainbow smelt were much higher than those reported in the literature for smelt from unpolluted areas in Canada, which ranged from 0.01 to 0.20 μg/g Hg (SHERBIN 1979). Smelt can apparently accumulate substantial amounts of mercury. They are eaten by burbot and lake trout in St. Froid Lake and, since their introduction in about 1958, they have been an important food of lake trout, burbot, and the larger lake whitefish in Eagle Lake, as determined by stomach analyses and direct observations (P. BOURQUE, Maine Department of Inland Fisheries and Wildlife, personal communication). The consumption of smelt could explain the high mercury concentrations in lake whitefish from Eagle Lake (0.30-2.17 µg/g; mean length 45 cm) compared with concentrations of 0.04-0.28 µg/g Hg reported by BISHOP & NEARY (1976) for lake whitefish (mean length 50 cm) in Ontario lakes. The food of lake trout in Cliff Lake is probably similar to that of brook trout; in the absence of smelt the mercury content of lake trout remains significantly lower than that in lake trout from Eagle and St. Froid lakes where smelt are available.

Enhanced Methylation and Uptake Under Oligotrophic Conditions

The biotic accumulation of trace elements is high in waters where dissolved organic compounds are not present in high concentration because the organic complexation that normally decreases the availability of trace elements is lacking. Hg^{2+} and methylmercury, which show a strong affinity for organic substances, are similar to trace elements in this regard. Studies have shown that accumulation of mercury in fish is correlated with decreasing water hardness and alkalinity. SCHEIDER et al. (1979) reported that the mercury content of walleyes was higher in fish from lakes with alkalinities less than 15 mg/l as $CaCO_3$ than in fish from lakes with higher alkalinities. Studies in Wisconsin showed that mercury in fish was highest when the total alkalinity was less than 50 mg/l as $CaCO_3$ and pH was less than 7.0 (KLEINERT & DEGURSE 1972). These studies indicate an increase in accumulation of mercury resulting from a lack of complex formations and precipitation reactions, which would tend to lower its availability. Oligotrophic lakes may be efficient media for the accumulation of mercury by aquatic organisms. Enhanced methylation of mercury in oligotrophic lakes, resulting in higher levels of mercury in fish of these lakes than in eutrophic lakes has been reported for unpolluted areas (D'ITRI et al. 1971). Fish in such lakes may accumulate two or more times the normal background level. These are also the findings of BROUZES et al. (1977), which strongly suggested a general lack of correlation between mercury in lake sediments and that in fish from oligotrophic lakes in northwestern Quebec. They believed that methylation by bacteria in the water column is the primary source of methylmercury found in fish.

Levels of mercury in the lake sediments from St. Froid, Eagle, Allagash, and Umsaskis lakes, which were determined by NORMANDEAU ASSOCIATES (1978), were all within the range of values for unpolluted lakes (<1.1 µg/g dry weight; SHERBIN 1979). Surface sediments contained more mercury than deep sediments, suggesting a recent increase in mercury input into these lakes. Regardless of the source of the mercury, conditions in the oligotrophic lakes in northern Maine are suitable for the enhanced methylation of mercury and uptake by aquatic organisms. This may partly explain the elevated concentrations found in northern Maine fish relative to the "natural" levels in the lake sediments.

Possible Sources of Mercury

Eagle and Cliff lakes are remote, and direct industrial sources are not responsible for the mercury in the fish. St. Froid Lake is more accessible but there is no industry on the lake. No intensive geological surveys have been conducted in the study lakes area, and therefore no data exist for mercury in the watersheds of the lakes. Levels in stream sediments north and west of the study area, determined by the U. S. Geological Survey (BOUDETTE et al. 1976), ranged from 0.01-0.15 μg/g Hg--lower than levels reported for unpolluted areas in Canada (0.1-1.0 μg/g Hg; SHERBIN 1979). An examination of the geological map of Maine indicated that the study area probably does not contain formations bearing anomalous concentrations of mercury (S. NORTON, Department of Geological Sciences, University of Maine, personal communication). Finally, the use of mercurial fungicides by the Maine timber industry has not been documented for this region (NORMANDEAU ASSOCIATES 1978), and it is unlikely that the source of mercury is agricultural runoff because the watersheds of the lakes are forested.

NORMANDEAU ASSOCIATES (1978) believed that a likely source of mercury in northern Maine is contaminated precipitation. There are no data on mercury in northern Maine precipitation, but atmospheric deposition is a conceivable source. BROUZES et al. (1977) detected levels of mercury in rain and snow that were an order of magnitude greater than those in rivers and lakes in northwest Quebec, and interpreted this relation to indicate that precipitation is the principal contributor of mercury to these aquatic systems. Similar studies in Maine may establish atmospheric deposition as the primary source of mercury in northern Maine lakes.

Acid Precipitation, pH, and Mercury

Storm tracks converging on the northeast U.S. have been implicated in acid precipitation problems in New York State and Maine. Acid precipitation may cause an "oligotrophication" process (GRAHN ET AL. 1974) by reducing the buffering capacity of lakes, thus lowering their alkalinities and pH. The degree to which this process occurs depends on the bedrock geology of the lake watershed. FAGERSTROM & JERNELOV (1972) determined that the methylation of mercury proceeds at an optimum rate at pH 6.0. Because mercury may accompany and be deposited with acidic precipitation due to a concomitant release from industrial sources, fish in poorly buffered waters may accumulate elevated levels of mercury.

In many large oligotrophic lakes in northern Maine the pH ranges from 6.2 to 7.5 and alkalinities are less than 40 mg/l as $CaCO_3$ (J. AKIELASZEK, unpublished data). The oligotrophic nature of most of these lakes would tend to enhance the accumulation of mercury by fish, regardless of the source. Some of these lakes are poorly buffered (alkalinity <15 mg/l) and may be susceptible to acid precipitation, which would further enhance methylation of mercury and its subsequent uptake by fish. The fact that the recovery process of a mercury contaminated oligotrophic lake is a slow one makes this a matter of some concern.

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