

FINAL REPORT

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Mercury in the Tongue River Reservoir

By

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INTRODUCTION

Mercury in Aquatic Environments

The fates of mercury in aquatic systems have received considerable attention in recent years because mercury is readily accumulated by aquatic organisms and is highly toxic to humans. Mercury is the only metal, that I am aware of, for which human poisonings have occurred as a direct result of consuming metal-contaminated fish. Over 1200 victims of mercury poisoning have been identified in Japan, including more than 200 that have died (National Academy of Sciences 1978). Humans suffering from mercury poisoning have also been identified in Quebec and Ontario, Canada.

Victims of mercury poisoning suffer from paresthesia, incoordination, losses of vision and hearing, and intellectual deterioration (Study Group on Mercury Hazards 1971). Methylmercury has also been implicated as being teratogenic, mutagenic and carcinogenic to mammals. Consumption of mercury by pregnant women presents an anomalous hazard because mercury tends to concentrate in the red blood cells of the fetus. In Japan, several babies were born suffering from mercury poisoning although the mother suffered no ill effects.

The U.S. Food and Drug Administration (FDA) has established 0.5 $\mu\text{g Hg/g}$ wet tissue as the maximum safe concentration for mercury in food (FDA 1974). This guideline is based on clinical evidence from Minamata Bay victims, methylmercury dose-retention experiments with human subjects, and the fish eating habits of U.S. citizens. Enforcement of the guideline has resulted in closures of several commercial fisheries since 1969 and in marketing restrictions on tuna and swordfish. The U.S. Environmental Protection Agency (EPA) has recommended that mercury in natural waters not exceed 0.05 $\mu\text{g Hg/l}$ (EPA 1977),

based on laboratory experiments demonstrating that fish exposed to this concentration of methylmercury in water eventually exceed the FDA guideline.

Although many forms of mercury exist in natural waters, methylmercury ($\text{CH}_3\text{-Hg}^+$), an alkylmercury derivative, is the most toxic and bioaccumulative species. The bioaccumulative properties of methylmercury are due to its relatively small size, its lipophilicity, and its affinity for sulfhydryl groups in proteins (Clarkson 1973). Because of its cumulative tendencies, methylmercury is the predominant form of mercury present in fish muscle tissue (Westoo 1973; Uthe et al. 1973) and is the form causing clinical symptoms in humans. In water, inorganic mercury can be bacterially converted to methylmercury (Wood et al. 1968; Jensen and Jernelov 1969). Thus, less toxic forms of mercury that enter natural waters can potentially be converted to the most toxic and bioaccumulative form.

Energy and Water

Energy shortages in this country have resulted in expanded exploitation of the vast coal reserves of the western United States. Southeastern Montana, southwestern Wyoming, and western North and South Dakotas contain portions of the Fort Union coal formation; nearly 19 billion tons of economically recoverable coal underlie this region.

Major problems resulting from coal exploitation in the western United States are associated with water. Not only is groundwater disturbed and mineralized during the mining operation (Van Voast and Hedges 1975), but also, large quantities of water are consumed during energy conversion processes. The problem is compounded by the scarcity of water in the arid west.

Study Area

One of the major water courses in the mining area is the Tongue River which originates on the east slopes of the Bighorn Mountains in northcentral Wyoming. The Tongue River moves northeastward into southeastern Montana, finally reaching its confluence with the Yellowstone River near Miles City, Montana. The character of the river changes from a high gradient, cold water trout stream in its upper reaches to a meandering warm water stream as it flows through the western prairie.

A major agricultural storage impoundment, the Tongue River Reservoir, is situated about 10 km north of the Montana-Wyoming border near the town of Decker, Montana. The Decker Coal Company (jointly owned by Peter Kiewit and Sons, Sheridan, Wyoming and Pacific Power and Light, Portland, Oregon) began mining near the reservoir in 1972; sites are currently being mined on both the east and west sides of the reservoir and lease applications for a northward extension of the western site are currently under consideration.

An earlier study (Phillips 1978) has shown that some gamefishes from the Tongue River Reservoir contain mercury concentrations in edible portions that exceed the U.S. Food and Drug Administration (FDA) guideline for human consumption. However, major sources of mercury to the reservoir have not been identified.

This study was initiated to try and determine potential source(s) of mercury to the Tongue River Reservoir and to further examine some of the fates of mercury in this reservoir system.

Project Tasks

Tasks completed during this project included the following:

(1) Total and dissolved mercury concentrations in water entering (inflow, mine discharges) and leaving (reservoir outflow) the Tongue River Reservoir were monitored at regular intervals for 11 months and the percentage of mercury of mining origin was calculated.

(2) Total and dissolved mercury concentrations in water entering (inflow, mine discharges) and leaving (reservoir outflow) the Tongue River Reservoir were monitored (6 h intervals) over a 24 h period to determine if short-term fluctuations occurred.

(3) Mercury concentrations in water samples collected from different depths along a vertical transect of the Tongue River were measured to delineate the uniformity of mercury distribution in the river relative to depth.

(4) Sediment cores (analyzed for total mercury) were collected from various locations in the reservoir to determine the relationship between sediment depth and total mercury concentration.

(5) Surficial sediment samples were collected from throughout the impoundment (and analyzed for total mercury) to discover the distribution of mercury in the reservoir's sediments.

(6) Sediment samples (analyzed for total mercury) were collected from various reaches of the Tongue River and its tributaries to determine the distribution of mercury in the watershed upstream from the reservoir.

(7) Northern pike, smallmouth bass, and walleye were collected from the reservoir and the relationship between fish length (mm) and total mercury concentration in axial muscle tissue was noted for each species.

(8) Crappie (both white and black) were collected from two locations in the Tongue River downstream from the reservoir and the relationship between fish length (mm) and total mercury concentration in axial muscle tissue was determined; the mercury concentrations in the river crappie were compared to the mercury concentrations in crappie of similar sizes collected from the reservoir.

(9) Stomachs were removed from several northern pike and the contents were identified and analyzed for total mercury to determine the mercury concentrations in food organisms of pike.

(10) Age and growth studies of Tongue River Reservoir fishes (conducted by previous investigators) were used to derive the relationship between fish age and total mercury concentration in muscle tissues for the various species.

(11) The relationship between growth and mercury accrual for individual northern pike that were captured, tagged, and analyzed for total mercury in muscle tissue in both 1978 and 1979 was described.

MATERIAL AND METHODS

Water (Collection and Analytical)

Water samples were collected at regular intervals throughout the year from the East Decker mine discharge and from the Tongue River Reservoir inflow and outflow. Samples were also taken from the West Decker mine outflow during months when discharging occurred.

On May 17-18, water was collected from each location at four 6 h intervals. At each sampling interval and at each site, two samples were collected, one for dissolved mercury and one for total mercury. Water was collected in one liter polyethylene bottles and stored in 500 ml glass bottles that were previously spiked with nitric acid preservative (1 ml/l). Samples to be analyzed for total mercury were added directly to the glass bottles but dissolved mercury samples were filtered immediately after collection and then added to the bottles containing preservative. Filtration was first through a glass fiber filter and then through a 0.45 μ cellulose acetate millipore filter; all samples were refrigerated after collection and analyzed for mercury within 38 days. Prior to analysis, samples were oxidized with potassium permanganate and potassium persulfate; mercury was then vaporized, collected on a gold plated tube, and quantified by atomic absorption spectrophotometry (Varian model AA-6) after atomization with a carbon rod (Siemer and Woodriff 1974).

Sediment (Collection and Analytical)

Surficial sediment samples (top 5 cm) were collected from 21 locations (fall, 1978) in the Tongue River drainage upstream from the reservoir and from 170 locations (summer, 1979) in the reservoir. Sediment cores were also taken

(through the ice) from 11 reservoir sites during winter, 1979. Surficial sediments were collected with an Ekman dredge and cores were taken with a Phleger sampler equipped with removable polycarbonate liners. Surficial samples were placed in labeled plastic bags and cores were stored in the polycarbonate liners; all samples were frozen soon after collection. Prior to mercury analyses, cores were partially thawed, removed from the liners, and divided into 5 cm sections. Core sections and surficial sediments from the Tongue River were dried at 45-60 C, pulverized with a mortar and pestle, and sifted through a 100 mesh sieve (Tyler equivalents) prior to mercury analyses. Surficial sediments from the reservoir were handled as above but were not sieved. All samples were analyzed for total mercury using the method described for fish tissue samples (Siemer and Woodriff 1974). Mercury concentrations in sediments are reported on a dry weight basis.

Fish (Collection and Analytical)

Fish were collected from the reservoir using trapnets or gillnets and from the river by electrofishing or gillnetting. In a few instances, fish tissue samples were taken (from fish caught by anglers) at a creel census station operated by the Montana Department of Fish, Wildlife and Parks. At the time of collection, most fish were sacrificed, weighed, measured, and a portion of axial muscle tissue was removed and frozen for subsequent mercury analysis; however, most northern pike were anesthetized with MS 222 at the time of capture, a muscle sample was surgically removed, and the fish were returned to the reservoir, enabling us to resample individual pike.

Stomach contents of sacrificed northern pike were frozen and homogenized in a high speed blender with a few grams of dry ice. Samples were then warmed

to room temperature resulting in a homogeneous paste. Total mercury concentrations in muscle tissue samples, stomach walls, and in aliquots from stomach content homogenates were determined with a Varian model AA-6 atomic absorption spectrophotometer equipped with a carbon rod atomizer (Siemer and Woodriff 1974). Precision was estimated at $\pm 0.01 \mu\text{g Hg/g}$ based on the standard deviation of repeated analyses of the same samples. All mercury concentrations for fish tissue are reported on a wet weight basis.

Statistics

Paired sample means were compared using Student's t-test and multiple means by the Bonferroni multiple comparison procedure (Neter and Asserman 1974). Regressions of fish length vs. mercury concentration in axial muscle tissue were derived both linearly and after a log transformation of mercury content (Steel and Torrie 1960); the form of regression resulting in the best correlation coefficient was chosen for portrayal. Degree of difference between regression lines was determined by F-test using the method outlined by Neter and Asserman (1974).

RESULTS AND DISCUSSION

Mercury in Water

Mercury concentrations were consistently low in water from the Tongue River Reservoir inflow and outflow and from the East and West Decker mine discharges (Table 1); mercury in virtually all samples occurred near and below the lower detectable limit (0.05 $\mu\text{g Hg/l}$). At these low concentrations it is impossible to make valid comparisons between mercury occurring in dissolved and suspended forms. Mean mercury concentrations in water from the four stations sampled were not statistically different (Students t-test). The two mines accounted for only 0.4% of the total surface runoff (thus, 0.4% of the mercury) entering the reservoir from November, 1978 to September, 1979. No trends were noted between mercury concentration in the water and time of sample collection on a given day (Table 2) or between depth and distance from shore (inflow) of sample collection (Fig. 1).

The origin of mercury in the reservoir is not clear; several conceivable sources exist. Metal smelting is common in Western Montana and large amounts of mercury are known to be discharged to the atmosphere from this industry. Mercury in the reservoir may originate from atmospheric fallout. Matlick and Buseck (1978) have determined that there is a strong positive correlation between mercury concentrations in soil and intensity of geothermal activity. Conceivably, the underground fires that are common in the coal field of the Northern Great Plains have volatilized mercury present in buried strata, causing mercury to be emitted to the atmosphere or to concentrate in surface soil horizons where it is subject to natural leaching and runoff. The high concentrations of mercury in fishes from the reservoir may result because the reservoir possesses a unique set of limnological conditions that facilitate

Table 1. Total and dissolved mercury concentrations in water samples from the East and West Decker mine discharges and the Tongue River Reservoir inflow and outflow.

Date	Mercury concentration ($\mu\text{g Hg/l}$) ^a							
	Reservoir inflow		Reservoir outflow		East Decker		West Decker	
	Total	Dissolved	Total	Dissolved	Total	Dissolved	Total	Dissolved
11-28-78	0.1	0.1	0.1	---	0.1	0.1	---	---
12-20-78	0.1	0.1	0.1	0.1	0.1	0.1	---	---
1-12-79	0.1	0.1	0.1	0.1	0.1	0.1	---	---
1-26-79	0.0	0.0	0.1	0.1	0.0	0.0	---	---
2-15-79	0.0	0.0	0.1	0.1	0.0	0.0	---	---
3-9-79	0.1	0.0	0.1	0.0	0.0	0.0	---	---
3-23-79	0.0	0.0	---	---	0.0	0.0	---	---
4-5-79	0.0	0.0	0.0	0.1	0.1	0.1	---	---
4-16-79	0.0	0.0	0.0	0.0	0.0	0.0	---	---
4-27-79	0.0	0.0	0.1	0.1	0.0	0.0	---	---
5-7-79	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.1
5-17-79	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
6-1-79	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0
6-11-79	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
6-26-79	0.0	0.1	0.1	0.0	0.1	0.1	---	---
7-16-79	0.0	0.1	0.1	0.1	0.1	0.1	---	---
7-30-79	0.0	0.0	0.0	0.0	0.0	0.0	---	---
8-14-79	0.1	0.1	0.1	0.1	0.1	0.1	---	---
8-27-79	0.1	0.0	0.0	0.0	0.0	0.0	---	---
9-11-79	0.0	0.0	0.0	0.0	0.0	0.0	---	---
Mean \pm SD	0.04 \pm 0.05	0.03 \pm 0.05	0.06 \pm 0.05	0.06 \pm 0.05	0.04 \pm 0.05	0.04 \pm 0.05	0.00 \pm 0.00	0.04 \pm 0.05

^a Values are rounded off to the nearest 0.01 $\mu\text{g Hg/l}$; thus 0.1 denotes 0.05 - 0.14 $\mu\text{g Hg/l}$.

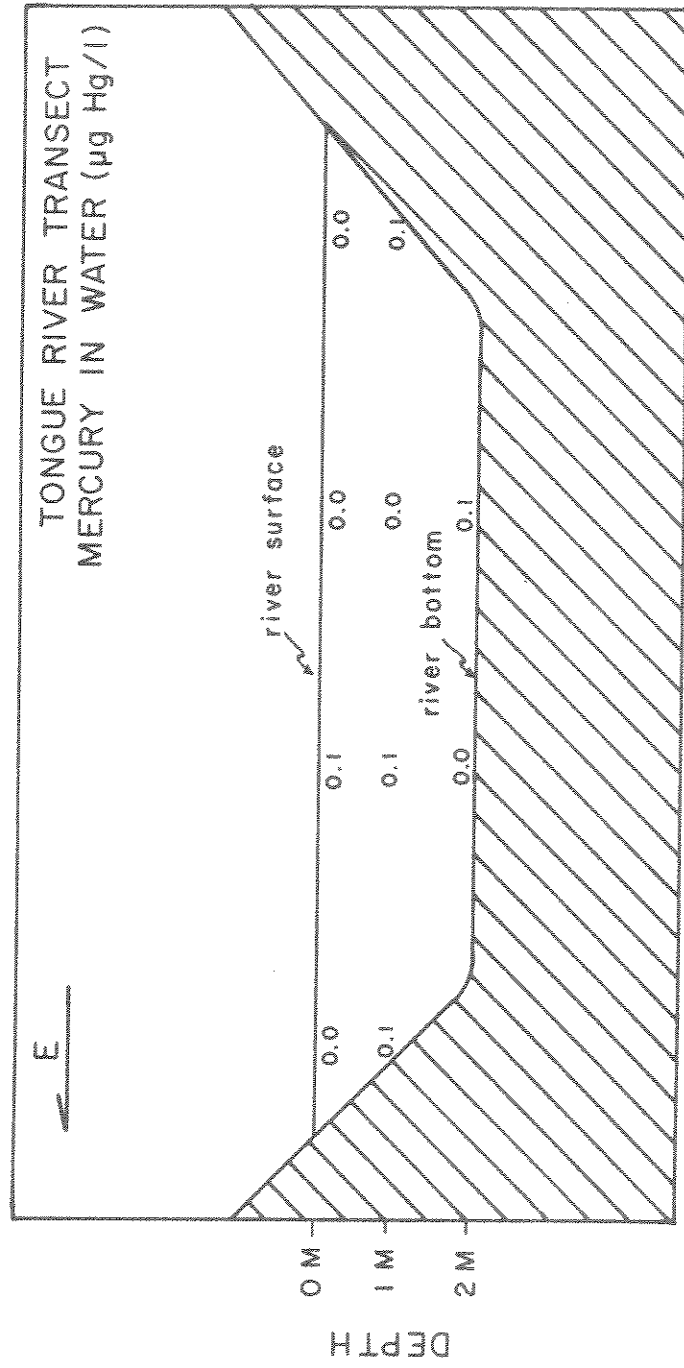


Figure 1. Total mercury concentrations in water samples taken from various locations in a vertical transect of the Tongue River.

Table 2. Total and dissolved mercury concentrations in water samples collected at 6 h intervals from the East and West Decker mine discharges and the Tongue River Reservoir inflow and outflow (May 17-18, 1979).

Date	Reservoir inflow		Reservoir outflow		Mercury concentration ($\mu\text{g Hg/l}$) ^a			
	Total		Dissolved		Total		Dissolved	
	Total	Dissolved	Total	Dissolved	East Decker Total	East Decker Dissolved	West Decker Total	West Decker Dissolved
5-17 (10:00 A.M.)	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.1
5-17 (4:00 P.M.)	0.1	0.1	0.1	0.0	0.0	0.0	0.0	0.0
5-17 (10:00 P.M.)	0.0	0.0	0.1	0.1	0.1	0.1	0.0	0.1
5-18 (4:00 A.M.)	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0

^aValues are rounded off to the nearest 0.1 $\mu\text{g Hg/l}$; thus 0.1 denotes 0.05 - 0.15 $\mu\text{g Hg/l}$.

the methylation of mercury although the total pool of mercury present in the sediments and water column is relatively small. Future studies at the Tongue River Reservoir should quantify atmospheric fallout of mercury.

Water Flow and Suspended Sediments

During the period November 1978 to September 1979 the combined discharges from the mines at East and West Decker averaged only 0.40% of the flow volume of the Tongue River (Fig. 2) and only 0.15% of the suspended solids entering the reservoir (Table 3). Only 20% of the suspended solids that entered the reservoir were discharged at the dam; thus the reservoir is acting as a sediment trap. Based on the mean flow rates and suspended solids concentrations for the dates sampled, suspended solids are entering the reservoir at a rate of 4.4×10^3 kg/yr from mining (East Decker only) and 3.0×10^7 kg/yr from the Tongue River; 6.0×10^6 kg/yr are being discharged from the dam.

Literature reports are conflicting as to whether most of the mercury transported in river systems is associated with particulate material (Nelson et al. 1977; Ottawa River Project Group 1979) or dissolved in water (Cranston and Buckley 1972 and Thomas et al. 1975). Since we were not able to distinguish between particulate and dissolved mercury, we have no way of confirming which was the case for the Tongue River Reservoir. This question has obvious implications regarding whether the reservoir is acting as a mercury sink.

Mercury in Sediments

River (surficial) - Previous investigators have shown that high concentrations of mercury usually occur in river sediments near major sources of mercury to rivers (Cranston and Buckley 1972; Langley 1973). In response to the

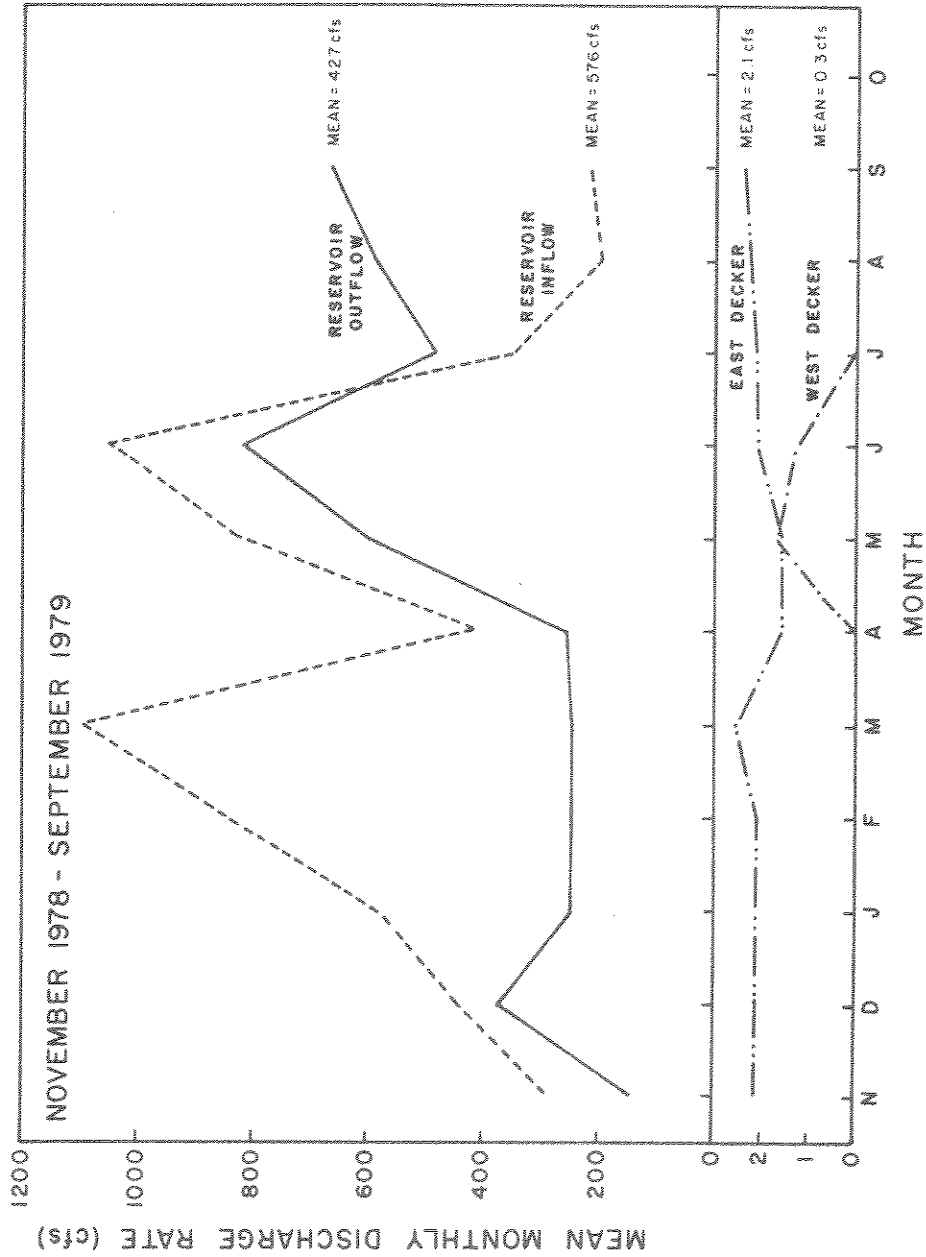


Figure 2. Monthly flow rates (cfs) for the Tongue River Reservoir inflow and outflow and mine discharges at East and West Decker (November 1978 - September 1979).

Table 3. Suspended solids concentrations in and flow rates of the East and West Decker mine discharges and the Tongue River Reservoir inflow and outflow.

Date	Flow Rate (cfs)				Suspended Solids (mg/l)			
	East	West	Inflow ^a	Outflow ^a	East	West	Inflow ^a	Outflow ^a
11-28-79	2.6	---	324	224	13	---	6	1
12-20-78	1.8	---	512	316	80	---	83	61
1-12-79	---	---	473	259	38	---	---	12
1-26-79	1.9	---	747	250	7	---	2	4
2-15-79	2.1	---	866	253	33	---	24	1
3-9-79	4.4	---	1450	241	34	---	73	12
3-23-79	1.0	---	1120	---	12	---	150	---
4-5-79	---	---	323	259	2	---	74	4
4-16-79	---	---	374	262	59	---	65	22
4-27-79	2.5	---	437	251	2	---	22	19
5-7-79	2.3		596	454	---	---	---	---
5-17-79	1.2	1.6	785	462	20	21	50	10
6-1-79	1.2	1.2	1420	1600	19	0	93	8
6-11-79	1.1	1.2	957	999	16	2	52	4
6-26-79	2.4	---	702	496	20	---	10	2
7-16-79	2.2	---	269	430	11	---	24	---
7-30-79	2.2	---	193	516	20	---	108	6
8-14-79	2.6	---	229	520	3	---	44	12
8-27-79	2.9	---	269	677	27	---	46	33
9-11-79	2.4	---	---	660	20	---	25	19
Mean±2SE	2.1±0.5	1.3±0.2	634±177	480±155	12±9	8±14	53±18	14±7

^aProvisional data of United States Geological Survey.

possibility that most of the mercury in the Tongue River system originates in the upstream portion of the watershed, we collected surficial sediment samples from 21 locations in the upper Tongue River drainage. Mercury concentrations in most of the samples were relatively low (Fig. 3). However, a few observations are noteworthy: (1) Higher mercury concentrations were detected in the sediments of Big Goose Creek directly below the Sheridan sewage outfall than in the sediments upstream from the sewage treatment plant. (2) Mercury occurred at a low concentration in Big Goose Creek sediment near the confluence of Big Goose Creek and the Tongue River. (3) All of the sediment samples collected in the Tongue River upstream from the Bighorn Mine contained less than $0.06 \mu\text{g Hg/g}$ while all of the samples collected downstream from the mine contained at least $0.15 \mu\text{g Hg/g}$. These data suggest that most of the mercury in the Tongue River (upstream from the reservoir) originates relatively near the reservoir. More extensive sampling is required to determine if higher mercury concentrations are present in sediments throughout this portion of the river.

Reservoir (cores) - Sediment cores were collected from eleven locations in the Tongue River Reservoir (Fig. 4). Mercury concentrations (dry basis) at all depths of all cores were relatively low (Table 4); mean mercury concentrations for entire cores ranged from $0.04 - 0.09 \mu\text{g Hg/g}$. The mean mercury concentration at station 1 was significantly lower ($P < 0.10$) than that reported for all other stations except 2, 6 and 10. This difference may result because station 1 is shallow and exposed to the air for part of the year. The station 10 core also contained a significantly lower mercury concentration ($P < 0.10$) than the core from station 11. In all other comparisons of mean mercury concentrations between cores, differences were not statistically significant.

In general, little significant difference existed between sample depth and mercury content; although the mean mercury concentration in sections

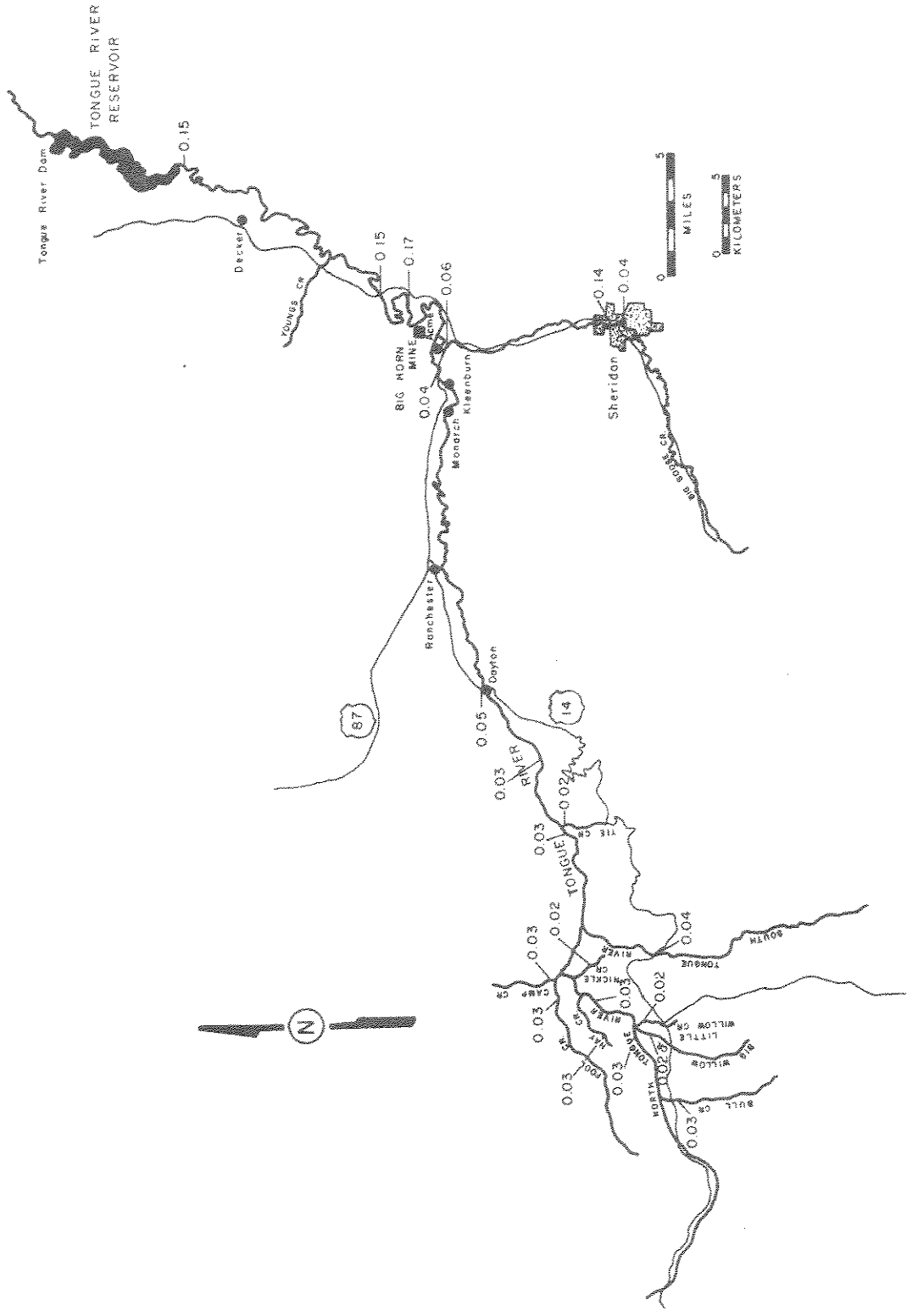


Figure 3. Total mercury concentrations ($\mu\text{g Hg/g}$ dry sediment) in surficial sediment samples collected from the Tongue River watershed upstream from the Tongue River Reservoir.

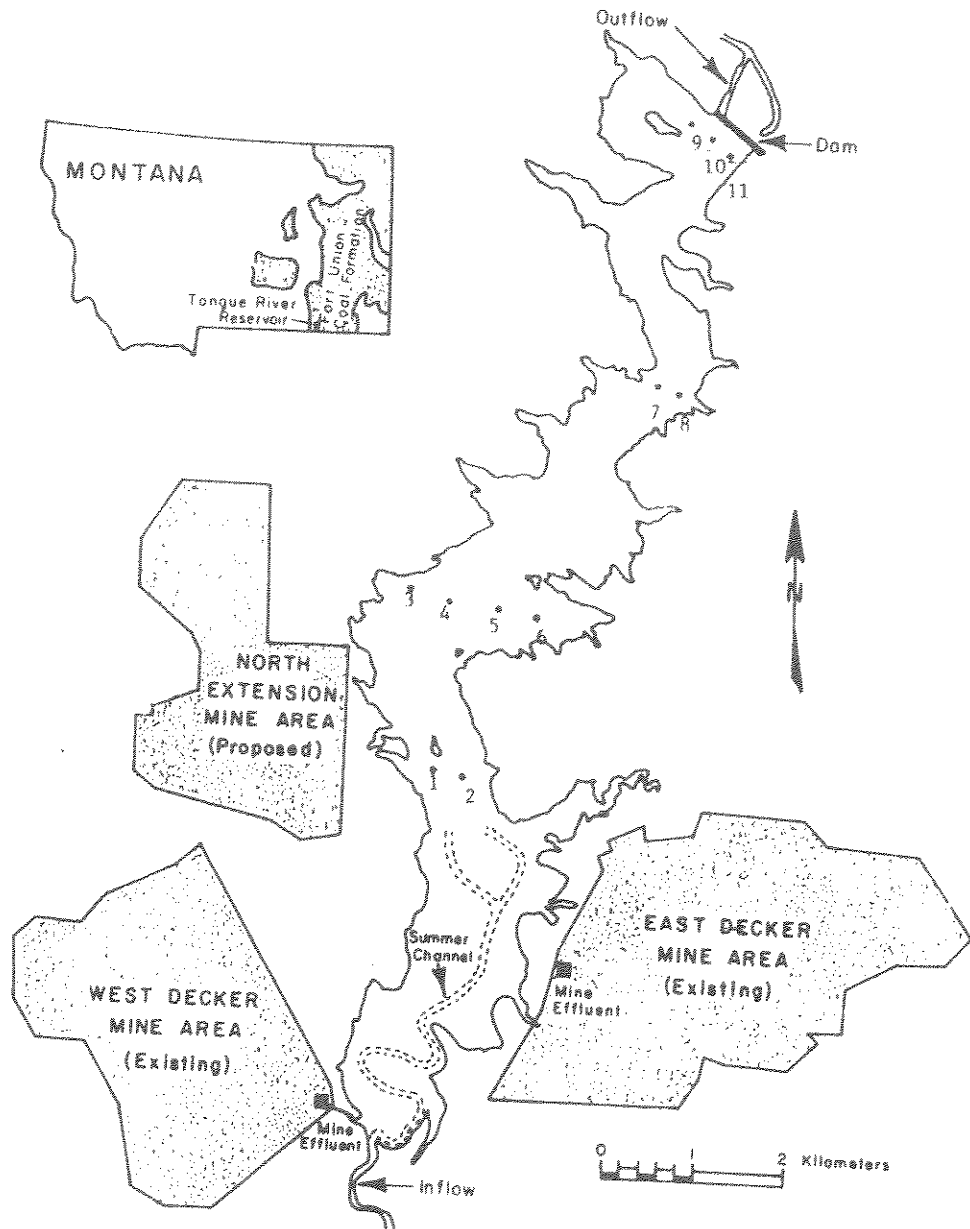


Figure 4. Map of the Tongue River Reservoir showing nearby surface coal mining activity and reservoir sampling sites for sediment core samples.

Table 4. Mercury concentrations ($\mu\text{g Hg/g}$ dry sediment) relative to depth in sections from sediment cores collected from the Tongue River Reservoir.

Sample depth (cm)	Sample number and location in reservoir ^a											$\bar{x} \pm 2SE^b$ (each depth)
	Upper			Lower-mid			Lower			10	11	
	1	2	3	4	5	6	7	8	9			
0-5	0.02	0.07	0.06	0.11	0.15	0.10	0.12	0.05	0.12	0.06	0.08	0.09 \pm 0.02
5-10	0.04	0.06	0.06	0.12	0.08	0.08	0.06	0.07	0.09	0.05	0.08	0.08 \pm 0.01
10-15	0.04	0.06	0.08	0.09	0.11	0.05	0.08	0.07	0.07	0.07	0.10	0.08 \pm 0.01
15-20	0.04	0.04	0.04	0.08	0.06	0.04	0.05	0.10	0.08	0.07	0.10	0.07 \pm 0.01
20-25	0.03	0.06	0.06	0.04	0.09	0.06	0.05	0.07	0.06	0.08	0.06	0.06 \pm 0.01
25-30	0.03	0.09	0.06	0.04	0.07	0.06	0.05	0.08	0.08	0.08	0.10	0.08 \pm 0.01
30-35	0.07	0.07	0.07	0.08	0.08	0.07	0.06	0.08	0.07	0.07	0.08	0.07 \pm 0.01
35-40	0.06		0.07	0.08	0.08		0.06	0.07	0.03	0.03	0.09	0.07 \pm 0.02
40-45			0.10				0.08	0.07	0.05	0.05	0.07	0.07 \pm 0.02
45-50			0.07				0.08	0.06	0.09	0.09	0.07	0.07 \pm 0.01
50-55							0.08	0.04	0.05	0.05	0.09	0.07 \pm 0.02
55-60							0.08	0.09	0.05	0.05	0.09	0.07 \pm 0.04
$\bar{x} \pm 2SE$ (each station)	0.04 ± 0.01	0.06 ± 0.01	0.07 ± 0.01	0.09 ± 0.03	0.09 ± 0.02	0.07 ± 0.02	0.07 ± 0.02	0.07 ± 0.01	0.08 ± 0.01	0.06 ± 0.02	0.08 ± 0.01	0.07 ± 0.01

^aSee Fig. 4 for exact sample location.

^bStation 1 was not used to derive means because it is a shallow water station that is not submerged for the entire year.

taken at a depth of 20-25 cm was significantly lower than means for samples taken at 0-5, 5-10, and 10-15 cm. Armstrong et al. (1972) reported that surficial sediments from Clay Lake, Ontario contained higher mercury concentrations than underlying layers; this does not appear to be true for the Tongue River Reservoir.

Reservoir (surficial) - Surficial sediment samples were collected from 170 locations from throughout the reservoir and analyzed for mercury. Sampling locations and mercury concentrations found are listed in Appendices I and II. The average mercury concentration in surficial sediment was 0.04 $\mu\text{g Hg/g}$, an extremely low accumulation considering the high concentrations of mercury in fish. A number of investigators have collected data on mercury concentrations in both fishes and sediments from aquatic environments (Table 5). In no case was there such a large discrepancy between mercury in fish and in sediments as in the Tongue River Reservoir, suggesting that conditions must be highly conducive for the methylation of what little mercury is available.

Figure 5 describes the distribution of mercury (dry basis) in surficial sediments from the reservoir. In general, highest mercury concentrations ($> 0.05 \mu\text{g Hg/g}$) were present in sediments near the downstream end of the reservoir, intermediate concentrations of mercury ($0.03 - 0.05 \mu\text{g Hg/g}$) occurred from mid reservoir to the upstream end, and lowest mercury concentrations ($< 0.03 \mu\text{g Hg/g}$) were present in sediments along the shores. Mercury is known to have a higher affinity for fine clay and silt-like particles ($< 1/16 \text{ mm}$ diameter) than for coarser sands and gravels (Thomas and Jaquet 1976). The distribution of mercury in the Tongue River Reservoir sediments is related to the distribution of sediment types, i.e., the coarsest sediments are found near shore, the intermediate sized particles settle out of suspension near the upstream end and the fine suspended materials settle out near the downstream end.

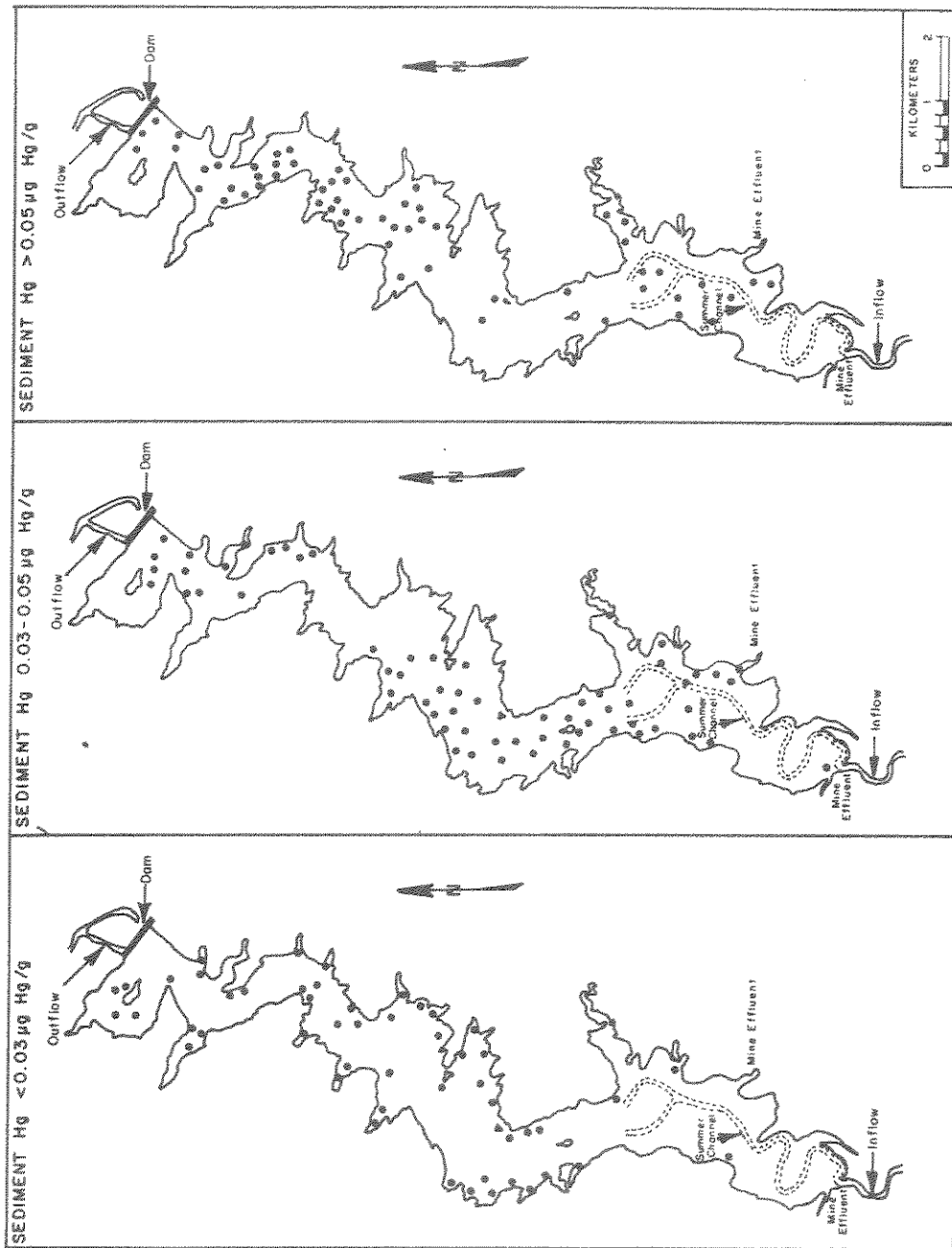


Figure 5. Map showing the distribution of mercury in surficial sediments of the Tongue River Reservoir.

Table 5. Reports from the literature of maximum mercury concentrations in fish muscle tissue relative to mercury concentrations found in sediments from the same environment.

Location	Mercury in Sediment µg Hg/g	Max. Hg in Fish ^d µg Hg/g	Fish Species	Reference(s)
Clay Lake (Ontario)	0.14-7.83 ^a	16.0	northern pike (<u>Esox lucius</u>)	Armstrong <u>et al</u> (1972); Bligh (1970)
Lahontan Reservoir (Nevada)	0.12-1.35 ^a	2.72	white bass (<u>Marone chrysops</u>)	Richins and Risser (1975)
Lake Powell (Arizona)	0.30 ^b	0.76	walleye (<u>Stizostedion vitreum</u>)	Potter <u>et al.</u> (1975)
Hemlock Lake (Michigan)	0.02-1.25 ^a	0.42	rainbow trout (<u>Salmo gairdneri</u>)	D'Itrie <u>et al.</u> (1971)
Section Four Lake (Michigan)	0.03-0.12 ^a	0.45	rainbow trout (<u>Salmo gairdneri</u>)	D'Itrie <u>et al.</u> (1971)
Unspecified river (Eastern Canada)	0.01-109.0 ^a	7.0	not specified	Langley (1973)
Antelope Reservoir (Oregon)	17.1 ^c	1.79	rainbow trout (<u>Salmo gairdneri</u>)	Phillips and Buhler (1979); Hill <u>et al.</u> (1975)
Tongue River Res. (Montana)	0.003-0.075 ^a	2.5	northern pike (<u>E. lucius</u>)	This study

^aRange.

^bMean.

^cOnly one sample taken.

^dReported for axial muscle on a wet weight basis.

Mercury in Fishes

Relative to size and species - The mercury concentrations in axial muscle tissue from all fish species collected from the Tongue River Reservoir during 1979 increased as fish size increased (Figs. 6-9 and Table 6). A similar trend was reported for fish sampled from the reservoir the previous year (Phillips 1978). For the species sampled, (with the exception of smallmouth bass) regressions of the form $\log_{10} Y = a + bX$ (Y = mercury in tissue, X = fish length) resulted in better correlations than did linear regressions. A regression of fish length vs. mercury concentration was not derived for largemouth bass because only six individuals were sampled.

The trend toward increasing mercury content with fish size has been noted by others (e.g., Bache et al. 1971; Fagerstrom et al. 1974; Scott 1974; Potter et al. 1975) and is primarily a result of the long biological half-time of methylmercury in fishes (Jarvenpaa et al. 1970; Giblein and Massaro 1972); fish retain previously assimilated mercury while continuing to accumulate more. However, the exponential shape of our curves is highly unusual; most workers have observed mercury uptake curves (for a variety of fishes) that initially proceeded linearly or curvilinearly and finally became asymptotic (Bache et al. 1971; Fagerstrom et al. 1974). The pattern of mercury uptake observed for Tongue River Reservoir fishes may indicate that their mercury exposure regime is becoming progressively more severe. Future studies should seek to answer this question.

Some individuals of all species analyzed exceeded the FDA guideline. The highest concentration reported ($\mu\text{g Hg/g}$ wet tissue) for each species was: northern pike male-2.47, northern pike female-2.15, walleye-1.60, largemouth bass-0.77, and smallmouth bass-0.65. According to the regressions, the length (mm) at which the average fish of each species began to exceed the FDA guideline

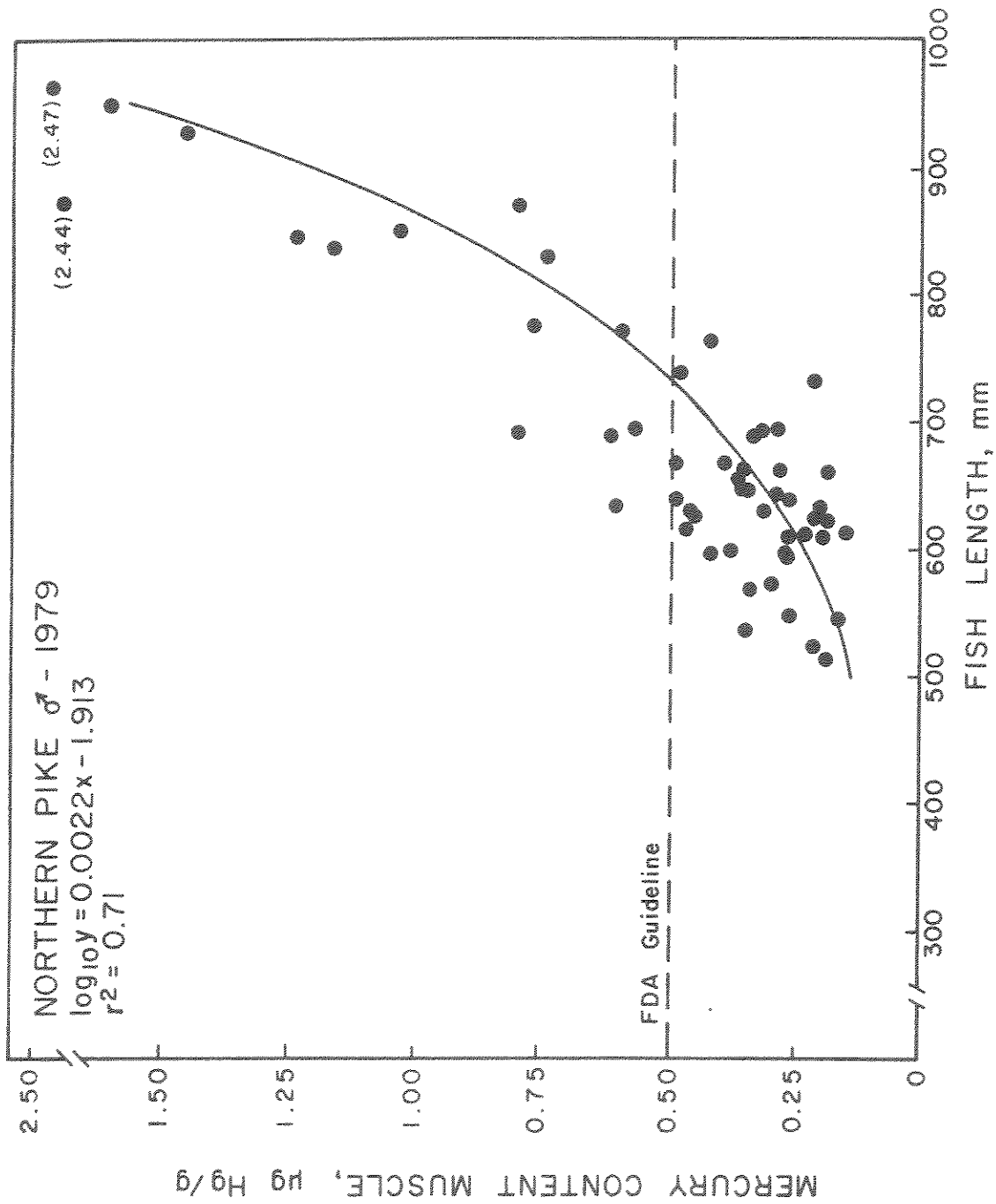


Figure 6. Relationship between total fish length and mercury concentration in axial muscle tissue for male northern pike collected from the Tongue River Reservoir during spring and summer 1979.

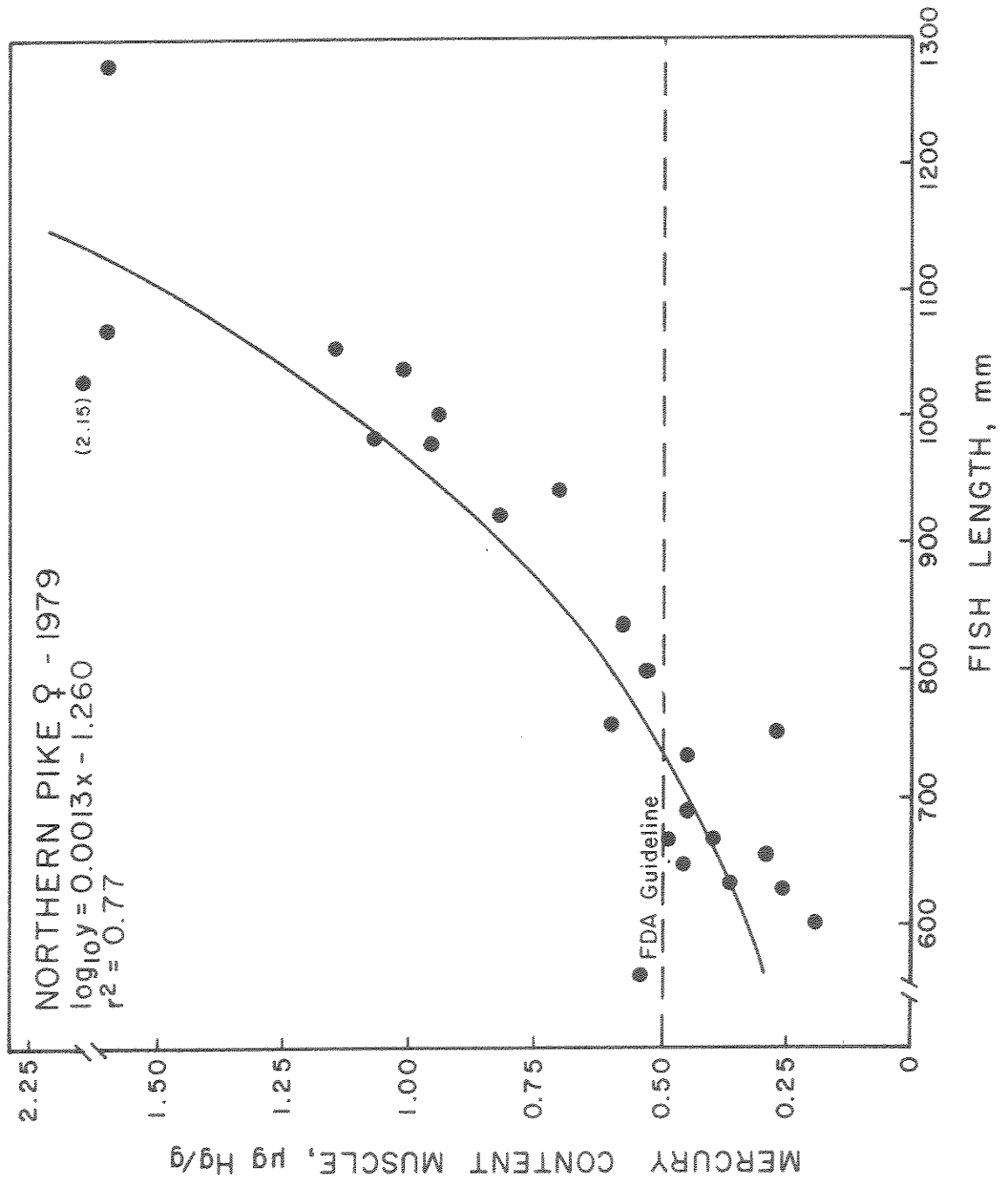


Figure 7. Relationship between total fish length and mercury concentration in axial muscle tissue for female northern pike collected from the Tongue River Reservoir during spring and summer 1979.

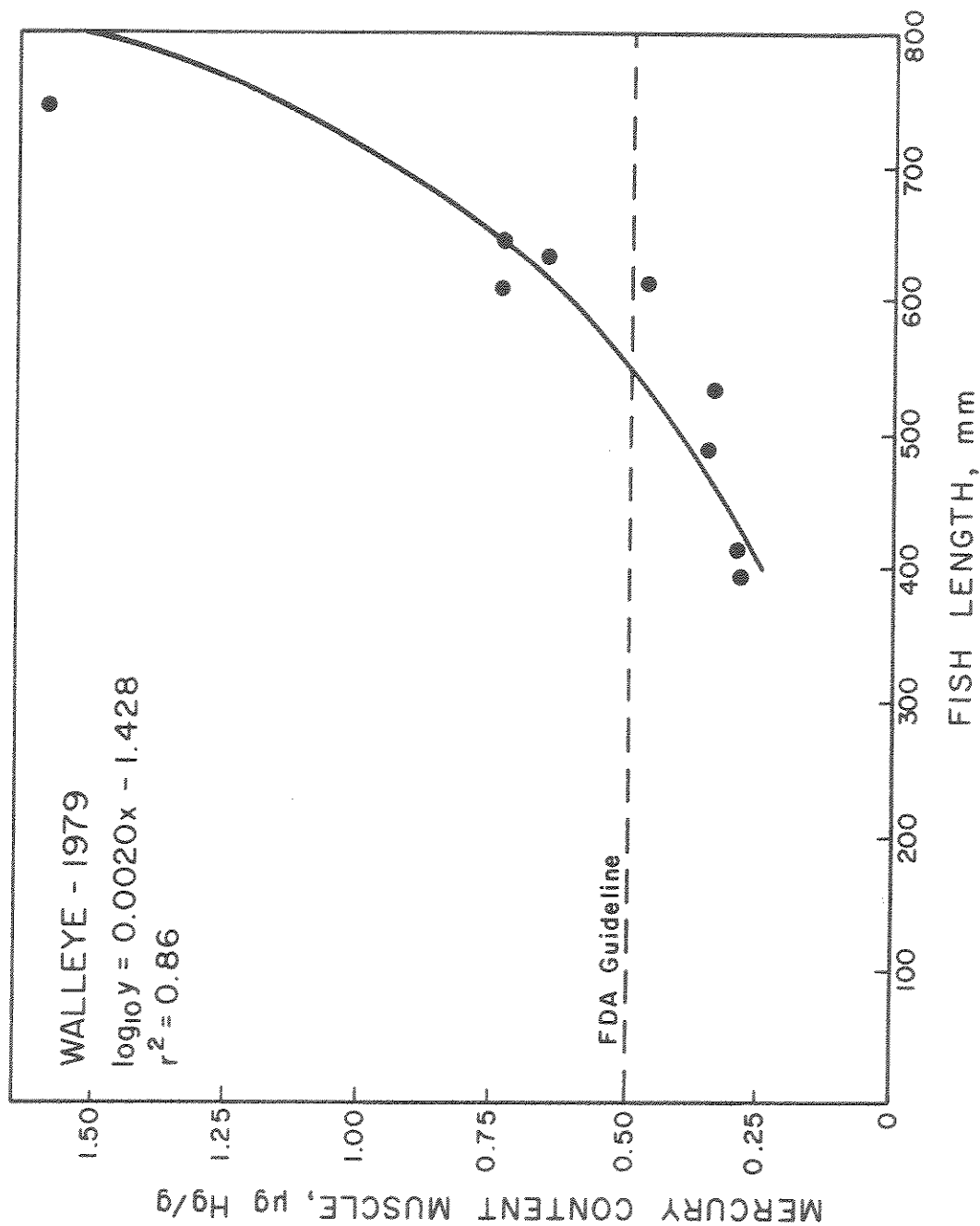


Figure 8. Relationship between total fish length and mercury concentration in axial muscle tissue for walleye collected from the Tongue River Reservoir during spring and summer 1979.

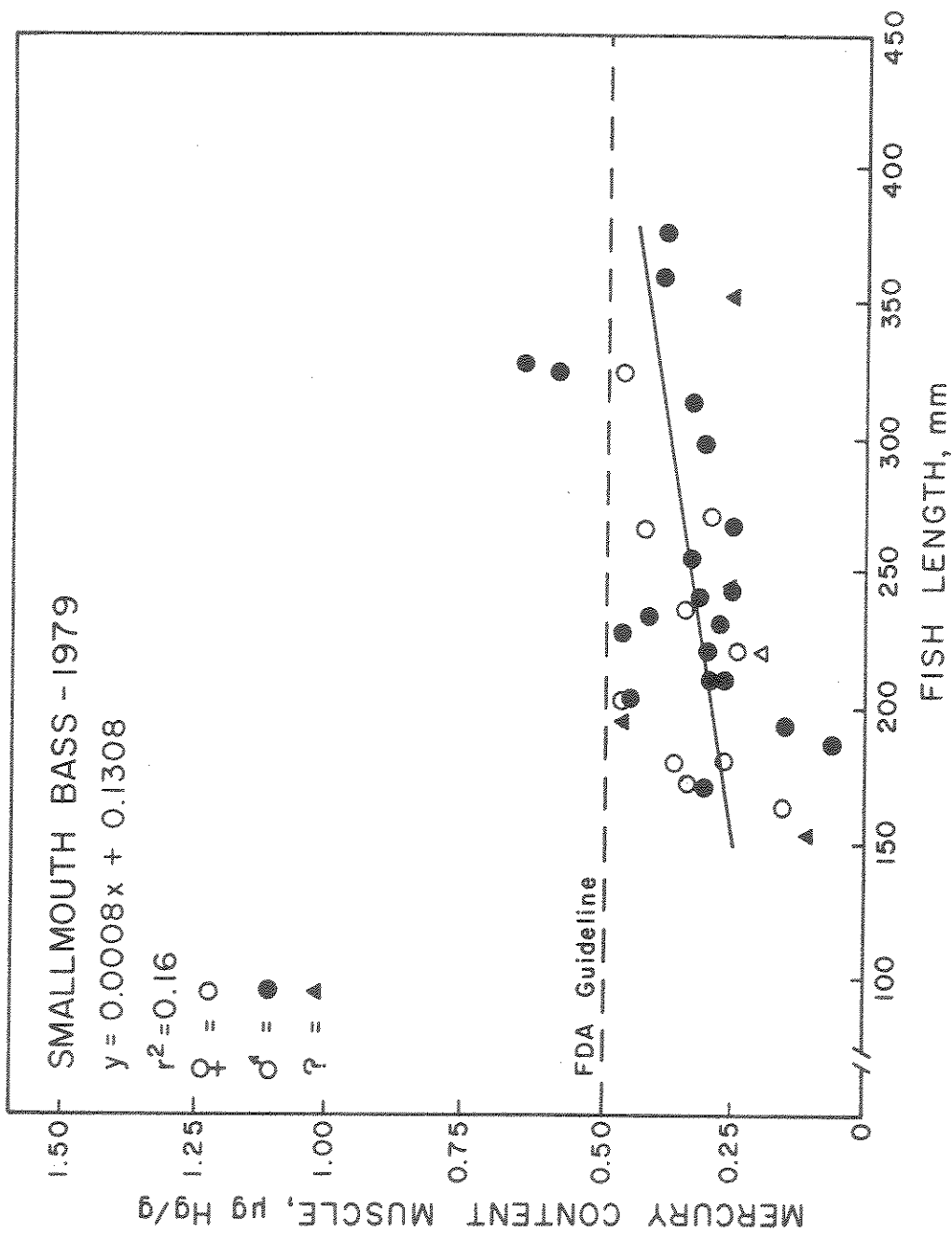


Figure 9. Relationship between total fish length and mercury concentration in axial muscle tissue for smallmouth bass collected from the Tongue River Reservoir during spring and summer 1979.

Table 6. Weights, lengths, sexes of and mercury concentrations in (axial muscle tissue) largemouth bass collected from the Tongue River Reservoir during 1979.

Weight (g)	Length (mm)	Sex	Mercury in muscle ($\mu\text{g Hg/g}$)
80	173	-	0.30
280	268	male	0.42
305	270	female	0.14
310	266	male	0.35
340	288	female	0.39
2050	475	-	0.77

was female northern pike-789, male northern pike-735, walleye-563, and small-mouth bass-462.

The FOA/WHO Joint Expert Committee on Food Additives (1972) recommends that humans should not consume more than 200 μg Hg/wk of methylmercury. This rate of mercury consumption would be achieved if a person were to consume 400 g (0.88 lb.) of fish per week that contained 0.5 μg Hg/g or 200 g (0.44 lb.) of fish per week that contained 1.0 μg Hg/g.

Fish from the Tongue River Reservoir of the sizes that exceed the FDA guideline (0.5 μg Hg/g) are relatively rare in the sportfishery. It is therefore highly unlikely that a fisherman at the Tongue River Reservoir could catch enough fish exceeding the guideline to allow extended consumption of mercury at a rate exceeding recommendations. I therefore do not advocate restrictive sportfishing regulations at this time. This recommendation is supported by studies in the literature of human populations with known mercury consumption rates in the United Kingdom (Haxton et al. 1979) and Sweden (Birke et al. 1972).

Relative to age and trophic position - To compare rates of mercury uptake among the various species, mercury concentration in tissue must be considered relative to fish age. Previous age and growth studies conducted on Tongue River Reservoir fishes provided estimates of age for black and white crappies (Elser et al. 1977) and walleye and sauger (Riggs 1978). No age and growth studies of northern pike in the Tongue River Reservoir have been completed; however, ages of pike were estimated from the work of Van Engel (1940), who recorded ages of pike from Wisconsin lakes of a latitude similar to that of the Tongue River Reservoir. Substituting the average length of each year class (for each species) into the regression equation for fish length vs. mercury concentration yields the average mercury concentration present in fish

of a given year class. Regressions determined for fish sampled during 1978 (Table 7) were used for the derivation because more species were sampled in 1978 than in 1979.

Figure 10 shows the relationship between fish age and mercury content for Tongue River Reservoir fishes. Northern pike and sauger accumulated mercury nearly twice as fast as did the two species of crappie; the rate for walleye was intermediate. Ages at which fish began to exceed the FDA guideline were VI for northern pike and sauger, VII for walleye, and VIII for the two crappie species.

Although the diets of these species vary with fish age (size), and seasonal food availability, in general, these fishes can be grouped according to food habits. Diets consist primarily of medium to large sized fish (northern pike), of small to medium sized fish and medium to large sized invertebrates (walleye and sauger), or of small to medium sized invertebrates (black and white crappies).

Numerous studies have shown that large prey organisms (both fishes and invertebrates) contain higher mercury concentrations than small prey organisms and that prey fish contain higher mercury concentrations than prey invertebrates e.g. Jernelov and Lann (1971); Phillips and Buhler (1979). Furthermore, much of the mercury in invertebrates (primarily aquatic insects) is inorganic mercury (Jernelov and Lann 1971; Cox et al. 1975), a form not readily accumulated by fish. Conversely, most of the mercury in fish tissue, prey or predator species, is methylmercury (Bache et al. 1971; Westoo 1973). Thus piscivores consume more methylmercury than insectivores or planktivores suggesting that differences in the mercury uptake rates among the various species from the Tongue River Reservoir may be related to differences in rates of dietary methylmercury intake.

Table 7. Regressions of mercury concentrations in axial muscle tissue vs. total fish length as reported for Tongue River Reservoir fishes collected during spring and summer 1978 (Phillips 1978).

Fish Species	Sample Size	Regression equation $\log_{10}Y = A+BX^a$	r^2
Northern pike	58	$\log_{10}Y = 0.0014X - 1.429^b$	0.76
Walleye	31	$\log_{10}Y = 0.0018X - 1.447$	0.70
Sauger	31	$\log_{10}Y = 0.0020X - 1.392$	0.32
Black crappie	7	$\log_{10}Y = 0.0027X - 1.390$	0.90
White crappie	34	$\log_{10}Y = 0.0034X - 1.468$	0.67

^aY = mercury concentration in fish muscle ($\mu\text{g Hg/g}$ wet tissue) and X = fish length (mm).

^bCombined regression for male and female northern pike.

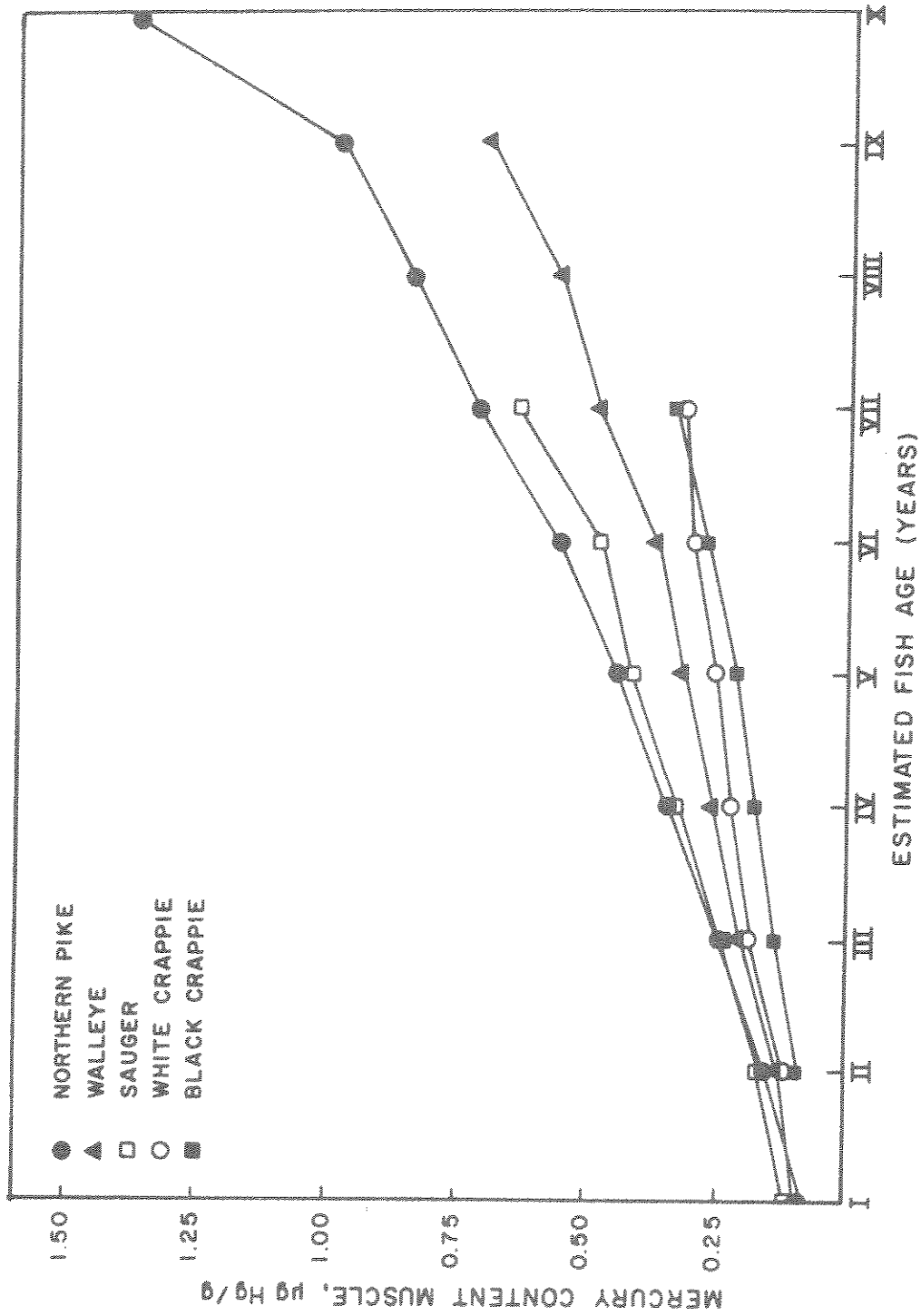


Figure 10. Relationship between total mercury concentration in axial muscle tissue and fish age for fishes sampled from the Tongue River Reservoir during 1978.

For pike that were captured in both 1978 and 1979 (Table 8) there was a positive correlation ($r^2 = 0.84$) between percentage increase in weight and percentage increase in total quantity of mercury in the fish (Fig. 11); the correlation was weaker for individuals that grew less. Errors in weight measurements resulting from stomachs being full or evacuated probably accounts for the greater variability among individuals that realized small percentage increases in weight. Increases in mercury concentration ranged from 27-152% while changes in quantity of mercury present ranged from 58-203%. Considering that weight increase is related to amount of food eaten (and thus amount of mercury eaten), a positive correlation further suggests that food is an important source of mercury to these fish.

Portions of the stomach walls from northern pike averaged 0.20 $\mu\text{g Hg/g}$ (Table 9); little variation existed between individuals (range: 0.12- 0.25 $\mu\text{g Hg/g}$) suggesting that the tissue was saturated with mercury. Mercury concentrations in axial muscle vs. mercury concentrations in stomach wall were poorly correlated ($r^2 = 0.07$). Possibly, stomach wall has less available binding sites for mercury than axial muscle or acid secretion in the stomach results in lower mercury retention.

Contents from the stomachs of northern pike that were sacrificed at the time of capture (or were procured during creel census) contained, on the average 0.28 $\mu\text{g Hg/g}$ (Table 9); values ranged from 0.10-0.59 $\mu\text{g Hg/g}$. Items in the stomachs included golden shiner, white crappie, shorthead redhorse and yellow perch. The stomach contents from one smallmouth bass (wt. = 460 g; length = 324 mm) and one walleye (wt. = 2230 g; length = 610 mm) were also analyzed for mercury. Mercury concentrations ($\mu\text{g Hg/g}$) were 0.20 for the smallmouth and 0.18 for the walleye. The walleye stomach contained a yellow perch while the smallmouth contained terrestrial insects.

Table 8. Changes in length, total mercury concentration in axial muscle, and total quantity of mercury present in male northern pike captured from the Tongue River Reservoir in spring 1978 and recaptured again in spring 1979.

Parameter	Fish number							
	1	2	3	4	5	6	7	8
Weight (g)								
1978	920	1,700	1,640	3,620	5,720	6,040	1,340	3,870
1979	1,580	3,375	2,160	3,680	5,811	6,401	2,600	4,800
difference ^a	660	1,675	520	60	91	361	1,260	930
% increase	72	99	32	2	2	6	94	24
Length (mm)								
1978	531	680	657	833	946	941	587	861
1979	631	800	740	842	961	949	661	870
difference ^a	100	120	83	9	15	8	74	9
% increase	19	18	13	1	2	1	13	1
Mercury conc. ($\mu\text{g Hg/g}$) ^b								
1978	0.24	0.21	0.35	0.67	1.25	0.82	0.19	0.63
1979	0.31	0.53	0.48	1.24	2.47	1.60	0.28	0.80
difference ^a	0.07	0.32	0.13	0.57	1.22	0.78	0.09	0.17
% increase	29	152	37	85	98	95	47	27
Quantity mercury ($\mu\text{g Hg}$) ^c								
1978	221	357	574	2,425	7,150	4,953	255	2,438
1979	490	1,080	1,037	4,563	14,353	10,241	728	3,840
difference ^a	269	723	463	2,138	7,203	5,288	473	1,402
% increase	122	203	81	88	101	107	185	58

^aDifference between 1978 and 1979.

^bMercury concentration in muscle tissue.

^cDerived by multiplying total weight(g) by the mercury concentration in muscle tissue ($\mu\text{g Hg/g}$); implicit assumption is that muscle is an average tissue with respect to mercury.

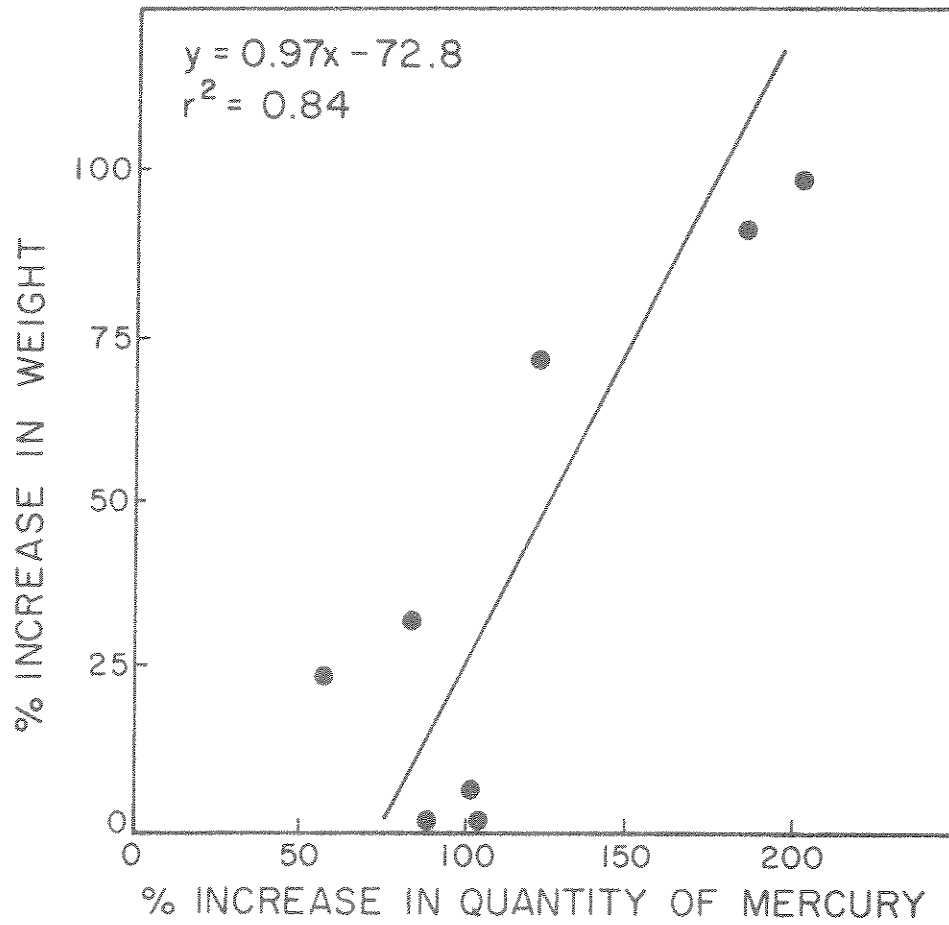


Figure 11. Relationship between percentage increase in body weight and percentage increase in quantity of mercury present in individual northern pike sampled in two consecutive years.

Table 9. Mercury in axial muscle, stomach muscle, and stomach contents of northern pike collected from the Tongue River Reservoir during spring and summer 1979.

Weight (g)	Length (mm)	Mercury concentration ($\mu\text{g Hg/g}$)			Items in stomach
		Axial muscle	Stomach muscle	Stomach ^a contents	
3140	774	0.59	0.15	--	empty
1580	631	0.31	--	0.22	golden shiner
2710	--	--	--	0.16	golden shiner
2440	--	0.28	--	0.26	white crappie
7128	980	0.95	--	0.10	shorthead redhorse
3780	858	1.04	--	0.37	white crappie
2480	735	0.38	--	0.34	white crappie
1660	700	0.56	0.24	0.18	shorthead redhorse
3800	823	0.70	0.25	0.59	yellow perch
1920	637	0.25	0.21	--	empty
1340	591	--	0.14	--	empty
1680	648	0.34	0.21	--	empty
2040	675	0.49	0.18	--	empty
1100	568	0.53	0.12	--	empty
1830	642	0.38	0.23	--	empty
1900	620	0.39	0.22	--	empty
1620	640	0.35	0.15	--	empty
1000	541	0.34	0.25	--	empty
Mean \pm 2SE			0.20 \pm 0.00	0.28 \pm 0.12	

^aItems in the stomach were homogenized in a blender and a subsample was analyzed for mercury.

Realizing that the mercury concentrations in partially digested prey fishes removed from stomachs may have different mercury concentrations than would the same prey fishes freshly caught, it is, nevertheless, instructive to calculate the expected rate of mercury accumulation for a predator (such as northern pike in the Tongue River Reservoir) consuming a diet that averages 0.28 $\mu\text{g Hg/g}$. For this calculation, the following assumptions were made: (1) 90% of the mercury in the prey fish was methylmercury; (2) 19% of the consumed methylmercury was assimilated; (3) 30% of the assimilated methylmercury was cleared each year; (4) food consumption occurred at an average yearly rate of 4% body wt./day (40 g/kg/day); (5) weight increase in a given year resulted in a 40% dilution of mercury concentration.

The calculation is then as follows: The pike diet contained 0.28 $\mu\text{g Hg/g}$ of which 90% (0.25 $\mu\text{g Hg/g}$) was methylmercury. The diet was being consumed at a rate of 40 g/kg fish per day, thus, methylmercury was being consumed at a rate of 10 $\mu\text{g Hg/kg fish per day}$ or 3650 $\mu\text{g Hg/kg fish per year}$. However, only 19% of that which was consumed was assimilated, only 70% of that which was assimilated was retained (13.3%), and only 60% of that retained was realized (8%) because of growth dilution. Therefore, the fish realized a net increase of 292 $\mu\text{g Hg/kg per year}$ or 0.29 $\mu\text{g Hg/g per year}$. From Figure 10, the average rate of mercury accumulation by northern pike in the Tongue River Reservoir was only 0.13 $\mu\text{g Hg/g per year}$. These calculations further support the hypothesis that food is an extremely important (if not the predominant) source of methylmercury to northern pike in the Tongue River Reservoir. However, many of the coefficients used to make these calculations have not been verified and changes in some parameters in the calculations could change the results considerably. Confirmation of the diet's contribution to the body burden of methylmercury in fishes in nature will require a thorough assessment of the

growth rates, food consumption rates and methylmercury consumption rates of fish in a natural environment.

Relative to sex and year sampled - Male northern pike sampled from the Tongue River Reservoir in a given year contained significantly higher mercury concentrations in axial muscle tissue (relative to fish length) than did females (Fig. 12). This was true for pike samples in both 1978 ($P < 0.02$) and in 1979 ($P < 0.00$). This trend has been noticed by other workers (Johnels et al. 1967; Olsson and Jensen 1975) and occurs primarily because females grow faster than males; thus male northern pike of a given size are exposed to the mercury contaminated environment for a longer period than are similar sized females.

Male northern pike ($P < 0.01$), female northern pike ($P < 0.05$), and walleye ($P < 0.10$)^{*} sampled in 1979 contained significantly higher mercury concentrations (relative to fish length) than individuals of the same sex and species sampled the previous year (Fig. 12). Regressions are the same as those depicted in Figs. 6-8 and Table 7 but the individual data points have been omitted for clarity. This trend strongly suggests that conditions were more favorable for methylation of mercury during 1979 and/or that more mercury was available for methylation in 1979. Conditions may be becoming progressively more favorable for mercury uptake by fishes in the reservoir or yearly variations in conditions that influence the availability of mercury simply may result in fishes experiencing fluctuating mercury exposure regimes.

Relative to environment sampled - One of the objectives of this study was to determine if the Tongue River Reservoir possesses a higher capacity for methylation of mercury than does the Tongue River; the mercury content of fish from the two environments should reflect the respective methylating capacities.

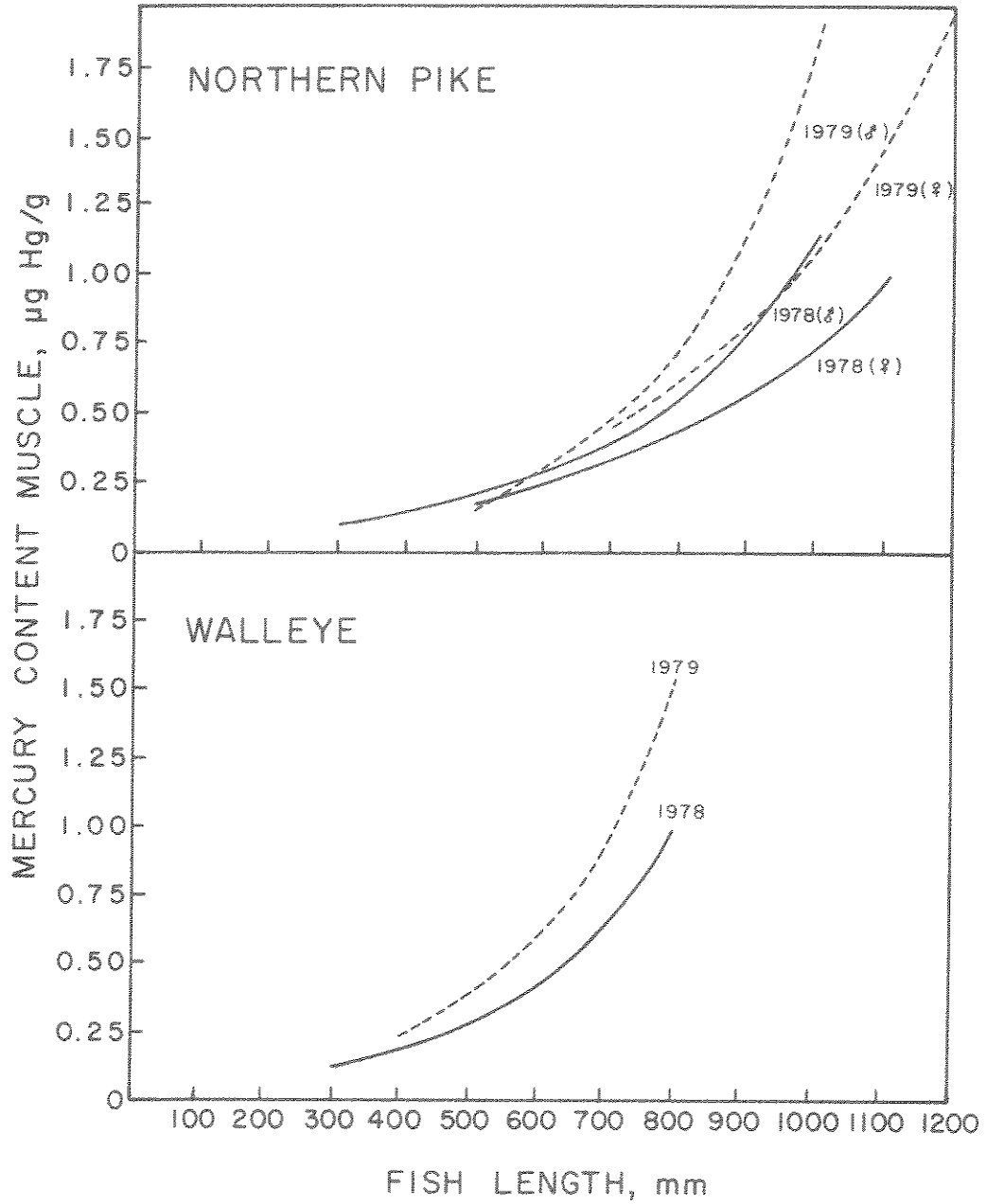


Figure 12. Relationships between sex, year sampled, and rate of mercury accumulation for northern pike and walleye from the Tongue River Reservoir.

We therefore collected crappie from two locations in the Tongue River (immediately below the dam and 32.4 river miles downstream, Fig. 13) and compared the mercury uptake patterns of these river populations (Figs. 14 and 15) to the mercury uptake pattern of crappie in the reservoir (Fig. 16). Regression lines for fish from the three locations are compared in Fig. 17. Crappie from the reservoir accumulated mercury significantly faster (relative to fish length) than crappie from the Tongue River below the dam ($P = 0.002$) or near Birney ($P = 0.001$).

A few northern pike, sauger and walleye were also collected at the station below the dam. For these species the mercury content of similar sized reservoir fish were interpolated from the regressions of mercury content ($\mu\text{g Hg/g wet tissue}$) vs. fish length (mm) reported in Table 7. Northern pike collected below the dam contained lower mercury concentrations than northerns of similar sizes collected from the reservoir (Table 10); no trend was apparent for the few walleye and sauger that were collected.

Although the ages of fish from the three sample locations are not known, it is probably safe to assume that the river fish were at least as old (and therefore exposed to mercury for at least as long) as fish of similar sizes from the reservoir. These findings support the contention that conditions are more conducive for the methylation of mercury in reservoirs than in rivers.

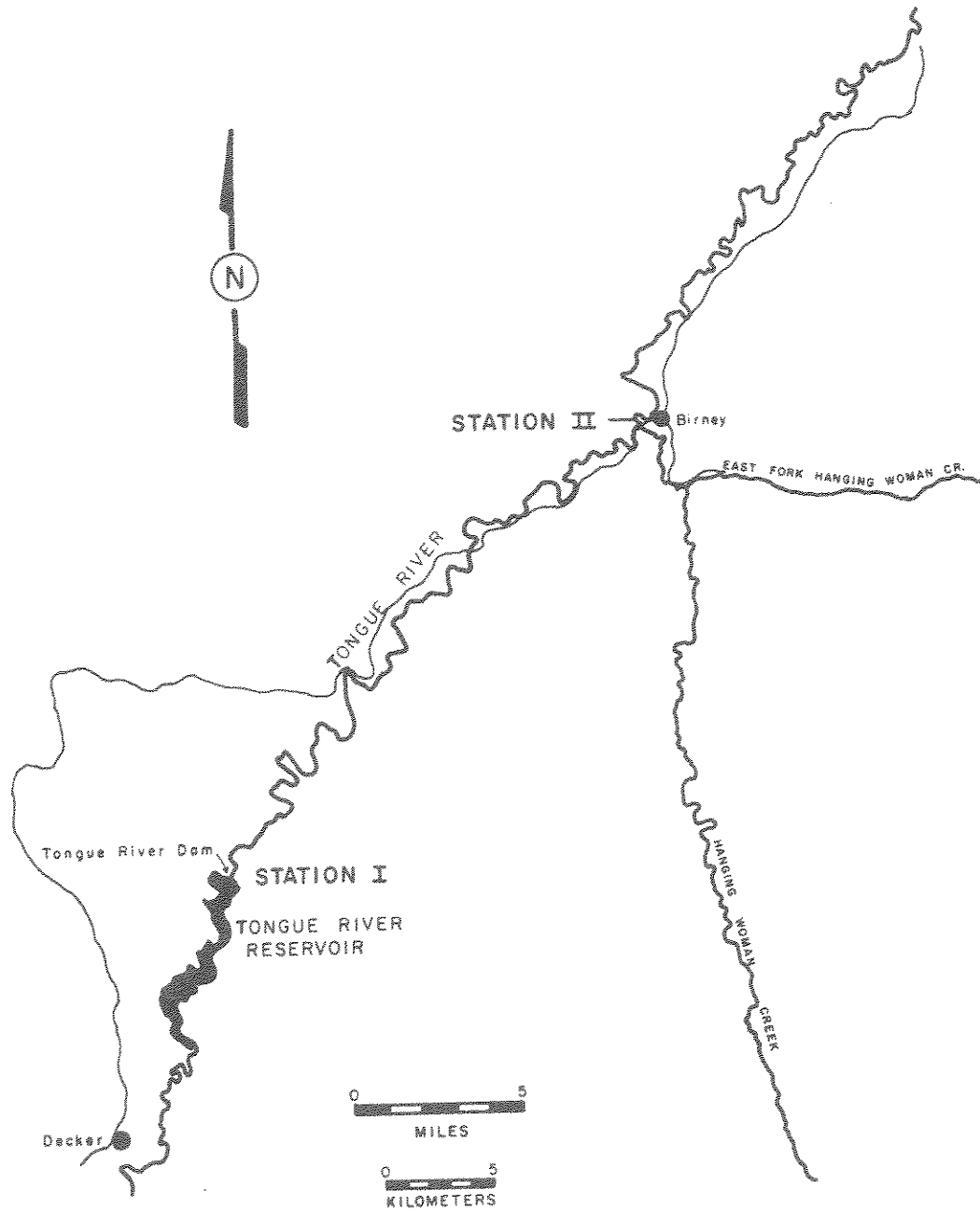


Figure 13. Map of the Tongue River showing fish sampling stations located downstream from the Tongue River Reservoir.

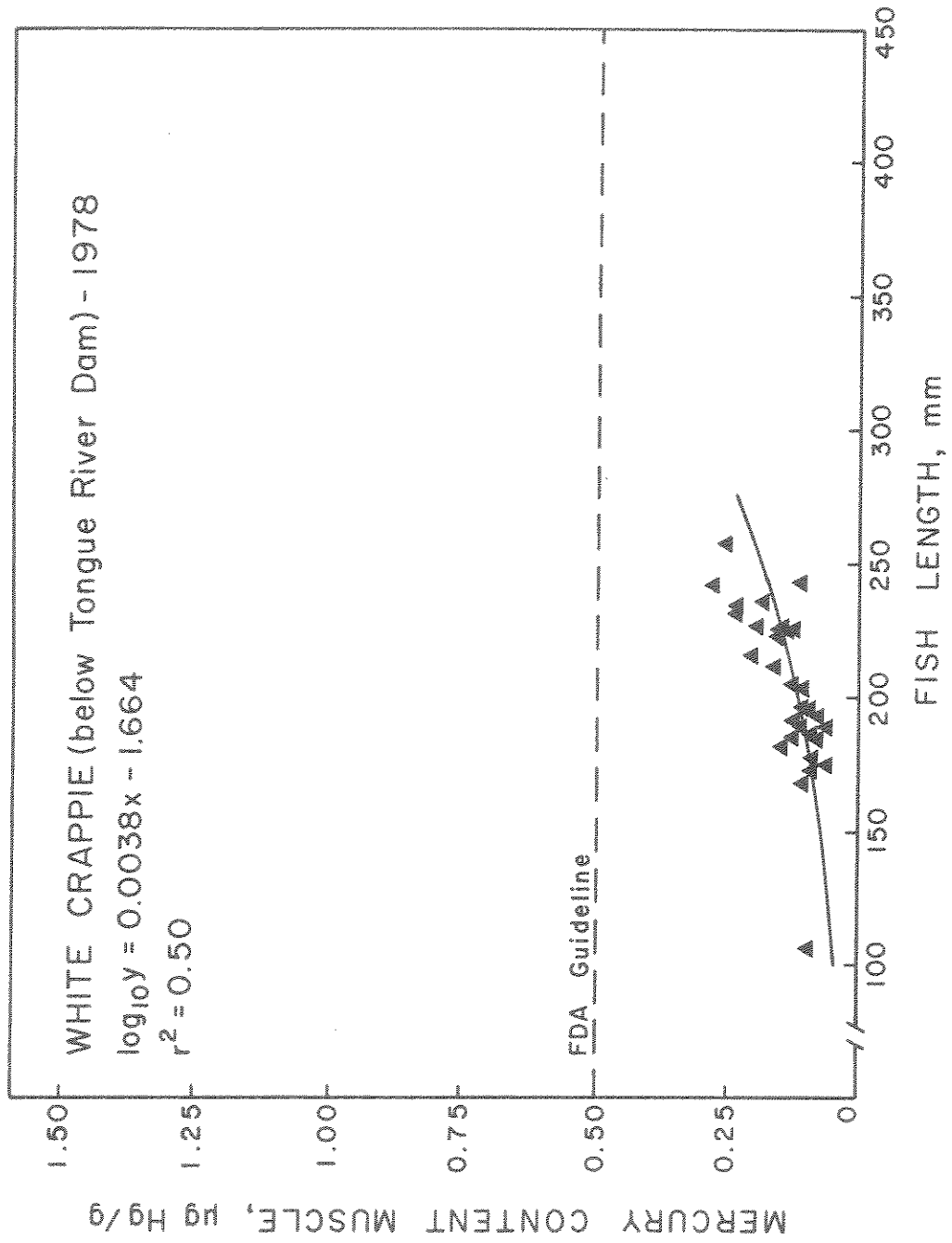


Figure 14. Relationship between total fish length and mercury concentration in axial muscle tissue for white crappie collected from the Tongue River immediately downstream from the Tongue River Dam during fall 1978.

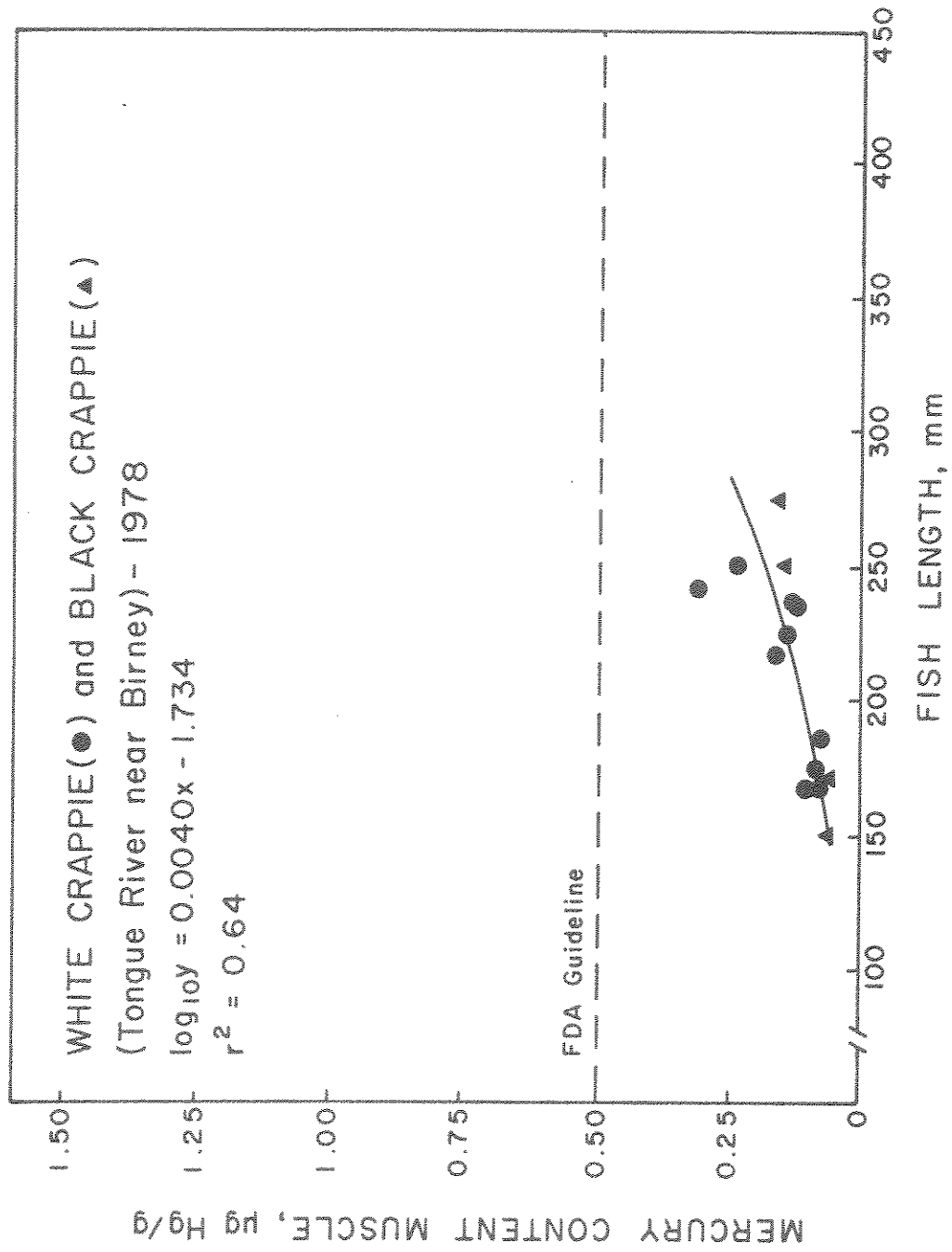


Figure 15. Relationship between total fish length and mercury concentration in axial muscle tissue for white and black crappie collected from the Tongue River near the town of Birney during fall 1978.

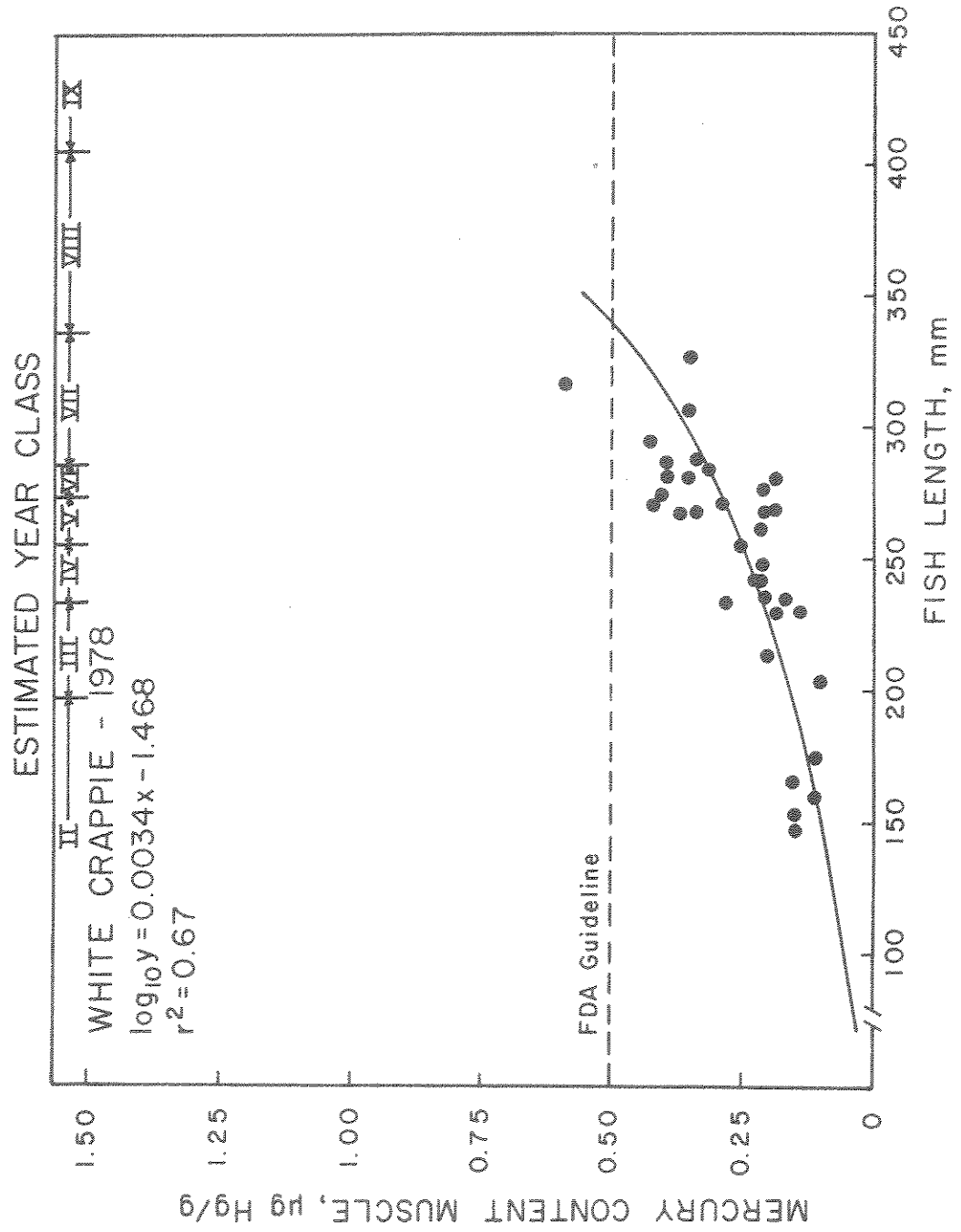


Figure 16. Relationship between total fish length and mercury concentration in axial muscle tissue for white crappie collected from the Tongue River Reservoir during summer 1978.

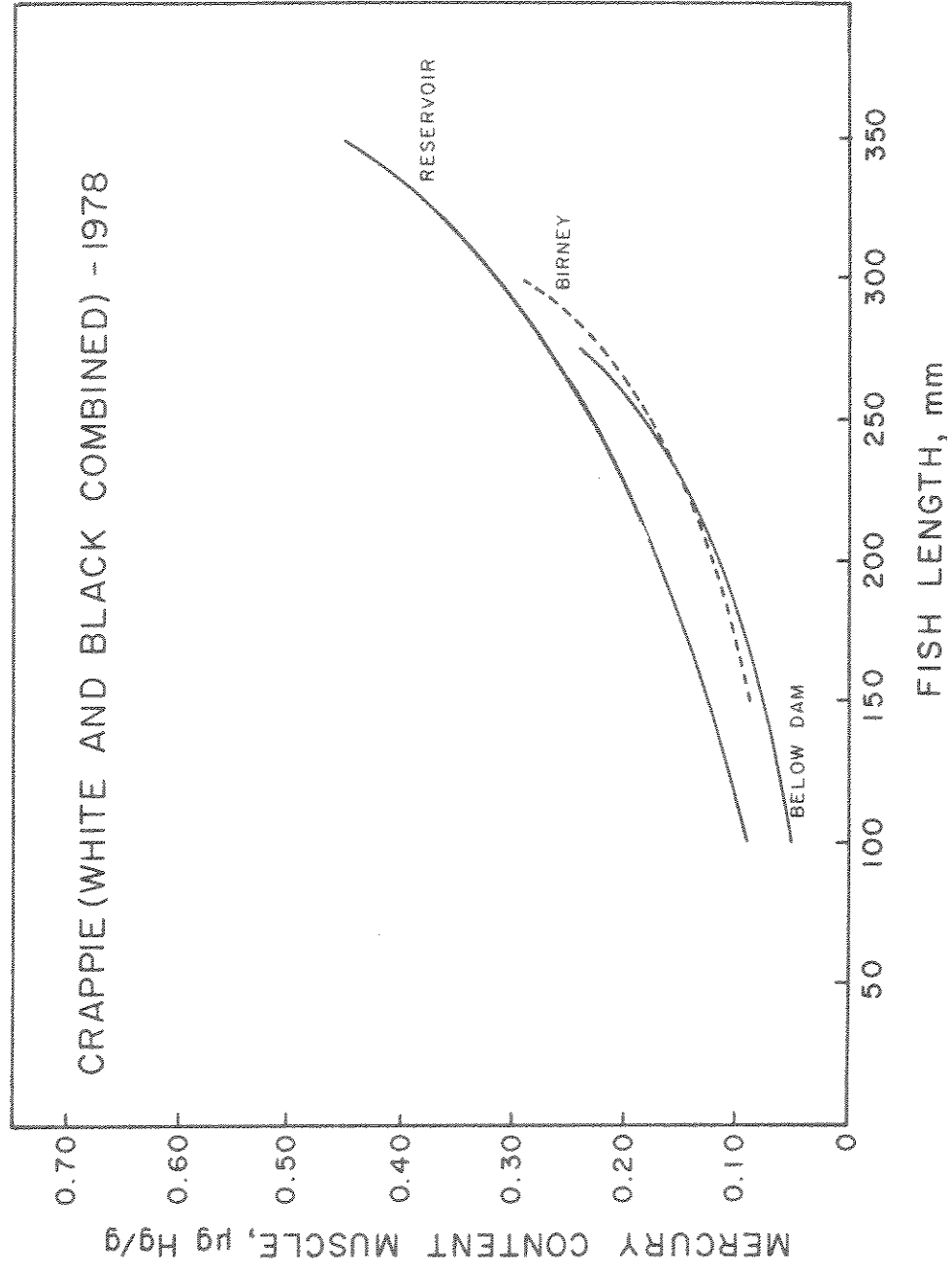


Figure 17. Comparison of the relationship between total fish length and mercury concentration in axial muscle tissue for crappie collected from the Tongue River Reservoir, from the Tongue River below the dam, or from the Tongue River near Birney during 1978.

Table 10. Mercury concentrations in axial muscle tissue from walleye, sauger and northern pike collected from the Tongue River immediately downstream from the Tongue River Dam compared to the mercury content of similar sizes of fish of the same species from the Tongue River Reservoir.

Species	Weight	Length	Mercury content ($\mu\text{g Hg/g}$) ^a	
			River fish	Reservoir fish
Walleye	454	372	0.25	0.17
	610	382	0.15	0.17
	1,700	553	0.41	0.35
Sauger	1,100	474	0.36	0.36
	1,140	540	0.48	0.49
Northern pike	1,200	590	0.11	0.25
	1,350	602	0.11	0.26
	1,780	652	0.18	0.30
	1,880	712	0.18	0.37

^aInterpolated from regressions of mercury content vs. fish length for reservoir fishes (Phillips 1978).

CONCLUSIONS AND RECOMMENDATIONS

1. The aqueous discharges from the mines at East and West Decker are not significant sources of mercury to the Tongue River Reservoir; mercury in the reservoir may originate from the atmosphere, groundwater or be of natural origin. Future studies should address these possible sources.
2. The reservoir is acting as a sediment sink; only 20% of the suspended solids entering the reservoir during water year 1978-1979 were discharged at the dam. Further data is needed to determine if the reservoir is acting as a mercury sink.
3. Higher concentrations of mercury occur in the Big Goose Creek sediments downstream from the Sheridan sewage treatment outfall and below the Bighorn Coal Mine in the Tongue River, than occur upstream from these points. If surface run-off is, indeed, a significant source of mercury to the Tongue River Reservoir, most of the mercury probably originates relatively near the reservoir. More intensive sampling of sediments immediately upriver and downstream from the reservoir will permit accurate mapping of mercury in sediments of the entire upper watershed.
4. The distribution of mercury in the surficial sediments in the Tongue River Reservoir is inversely correlated with sediment particle size; in general, lowest concentrations of mercury are found in coarse sediment particles found near shore, highest concentrations are present in fine sediments deposited near the downstream end, and intermediate mercury concentrations exist in medium size sediment particles deposited near the upstream end. Mercury concentration does not vary significantly with sediment depth.

5. Mercury concentrations in Tongue River Reservoir sediments are extremely low considering the high mercury concentrations attained by fishes in the reservoir. Conditions in the reservoir must therefore be highly conducive for the methylation of available mercury.
6. The relationship between total fish length and mercury concentration in axial muscle tissue for fish sampled in 1979 was positive (this was also true of fish sampled the previous year); the estimated lengths (mm) at which fishes begin to exceed the FDA guideline are: female northern pike-789, male northern pike-735, walleye-563, and smallmouth bass-462. Since fishes of these sizes are rare in the sportfishery, restrictive regulations for the protection of human health are not currently recommended.
7. Male northern pike contain significantly higher mercury concentrations in relation to size than females; this is probably due to sex related growth differences.
8. Stomach contents from northern pike contained, on the average, $0.28 \mu\text{g}$ Hg/g; a hypothetical calculation of the mercury consumption rates of these pike suggests that much of their mercury body burden is of dietary origin. Data that verifies or refutes the assumptions used to make this calculation will permit a more accurate assessment of the importance of dietary mercury in overall mercury accrual.
9. Percentage increases in mercury concentrations for individual northern pike analyzed for mercury in both 1978 and 1979 are correlated with weight increases. Further, mercury uptake relative to fish age is correlated with dietary habit of the species; piscivores accumulated mercury the fastest and planktivores the slowest. These observations

further suggest that the diet is an important source of mercury to piscivores.

10. The relationship between fish length and mercury concentration in tissue for male northern pike demonstrates that pike collected during 1979 contained higher mercury concentrations than pike of similar sizes collected the previous year. Thus, more mercury was probably methylated in 1979. Future studies should determine if the mercury situation in the reservoir is becoming progressively more hazardous or if normal fluctuations in conditions that influence methylation of mercury are occurring.
11. Fishes residing in the Tongue River Reservoir accumulate mercury at a faster rate than the same species residing in the Tongue River downstream from the reservoir. This probably occurs because the reservoir possesses a greater capacity for methylation of inorganic mercury than the river. Future studies should attempt to correlate in situ methylation of mercury with limnological conditions known to influence methylation. Conceivably, methylation could be inhibited in the reservoir by manipulating the discharge regime.

REFERENCES

- Armstrong, F. A. J., D. Metner, and M. J. Capel. 1972. Mercury in sediments and water of Clay Lake northwestern Ontario. pp. 46-67 In J. F. Uthe (ed.) Mercury in the aquatic environment: A summary of research carried out by the freshwater institute 1970-1971. Fish. Res. Board Can. Manuscript Rep. Ser. No. 1167. 163 p.
- Bache, C. A., W. H. Gutenmann, and D. V. Lisk. 1971. Residues of total mercury and methylmercury salts in lake trout as a function of age. Science 172:951-952.
- Birke, G., L. O. Plantin, and S. Skerfving. 1972. Studies on humans exposed to methyl mercury through fish consumption. Arch. Environ. Health 25:77-91.
- Bligh, E. G. 1970. Mercury and the contamination of freshwater fish. Fish. Res. Board Can. Manuscript Rep. Ser. No. 1088. 27 p.
- Clarkson, T. W. 1973. The pharmacodynamics of mercury and its compounds with emphasis on the short-chain alkylmercurials. pp. 332-353 In D. R. Buhler (ed.) Mercury in the western environment. Continuing Education Publications, Oregon State Univ., Corvallis.
- Cox, M. F., H. W. Holm, H. J. Kania, and R. L. Knight. 1975. Methylmercury and total mercury concentrations in selected stream biota. pp. 151-155 In D. D. Hemphill (ed.) Trace Substances in Environmental Health. Vol. IX, University of Missouri, Columbia.
- Cranston, R. E. and D. E. Buckley. 1972. Mercury pathways in a river and estuary. Environ. Sci. Technol. 6(3):274-277.
- D'Itri, F. M., C. S. Annett, and A. W. Fast. 1971. Comparison of mercury levels in an oligotrophic and a eutrophic lake. Mar. Technol. Soc. J. 5:10-14.
- Elser, A. R., R. C. McFarland and D. Schwehr. 1977. The effects of altered streamflow on fish of the Yellowstone and Tongue Rivers, Montana. Tech. Rep. No. 8, Water Res. Div., Montana Dept. Nat. Res. 180 p.
- Fagerstrom, T., B. Asell, and A. Jernelov. 1974. Model for accumulation of methylmercury in northern pike (Esox lucius). Oikos 25:14-20.
- Giblin, F. J., and E. J. Massaro. 1972. Pharmaco-dynamics of methylmercury in the rainbow trout (Salmo gairdneri): tissue uptake, distribution and excretion. Toxicol. Appl. Pharmacol. 24:81-91.
- Haxton, J., D. G. Lindsay, J. S. Hislop, L. Salmon, E. J. Dixon, W. H. Evans, J. R. Reid, C. J. Hewitt, and D. F. Jeffries. 1979. Duplicate diet study on fishing communities in the United Kingdom: mercury exposure in a "critical group." Environ. Res. 18:351-368.

- Hildebrand, S. G., A. W. Andren, and J. S. Huckabee. 1976. Distribution and bioaccumulation of mercury in biotic and abiotic compartments of a contaminated river-reservoir system. pp. 211-232 In R. W. Andrew, P. V. Hodson and D. E. Konasewich (eds.) Toxicity to biota of metal forms in natural waters. Proc. Great Lakes Research Advisory Board Workshop, Duluth, Minnesota.
- Hill, S., A. Cochrane, D. Williams, M. Lucky, K. Greenfield, R. Farlee, B. Hudson, T. Tkachyk, D. Ugstad, P. Ugstad, and L. Wickman. 1975. Study of mercury and heavy metals pollutants in the Jordan Creek drainage. The Silver City Project, Student Originated Studies, Natl. Sci. Foundation. 113 pp.
- Jarvenpaa, T., M. Tillander, and J. K. Miettinen. 1970. Methylmercury: half-time of elimination in flounder, pike and eel. *Suom. Kemistil. B.* 43:439-442.
- Jensen, S. and A. Jernelov. 1969. Biological methylation of mercury in aquatic organisms. *Nature (Lond.)* 223:753-754.
- Jernelov, A. and H. Lann. 1971. Mercury accumulation in food chains. *Oikos* 22:403-406.
- Johnels, A. G., T. Westermark, W. Berg, P. I. Persson, and B. Sjostrand. 1967. Pike (Esox lucius L.) and some other aquatic organisms in Sweden as indicators of mercury contamination in the environment. *Oikos* 18:323-333.
- Joint FAO/WHO Expert Committee on Food Additives. 1972. Evaluation of certain food additives and the contaminants mercury, lead and cadmium. WHO Food Additives Series No. 4. FAO Nutrition Meetings Report Series No. 51A.
- Langley, D. G. 1973. Mercury methylation in an aquatic environment. *J. Water Pollut. Control Fed.* 45(1):44-51.
- Matlick, J. S. III, and P. R. Buseck. 1978. Exploration for geothermal areas using mercury: a new geochemical technique. *Geothermal Energy Mag.* 6(9):15-23.
- National Academy of Sciences. 1978. An assessment of mercury in the environment. *Natl. Acad. Sci., Wash., D.C.* 185 pp.
- Nelson, H., B. R. Larsen, E. A. Jenne, and D. H. Sorg. 1977. Mercury dispersal from lode sources in the Kuskokwim River drainage, Alaska. *Science* 198:820-824.
- Neter, J. and W. W. Asserman. 1974. Applied linear statistical models. Richard D. Irwin, Inc., Homewood, Ill. 842 pp.
- Olsson, M., and S. Jensen. 1975. Pike as the test organism for mercury, DDT and PCB pollution. A study of the contamination in the Stockholm Archipelago. *Fish. Board Swed. Inst. Freshwater Res. Drottningholm Rep.* 54:83-106.

- Ottawa River Project Group. 1979. Mercury in the Ottawa River. Environ. Res. 19:231-243.
- Phillips, G. R. 1978. The potential for long-term mercury contamination of the Tongue River Reservoir resulting from surface coal mining. Final Report to the U.S. Fish and Wildlife Service. 53 p.
- Phillips, G. R. and D. R. Buhler. 1979. Mercury accumulation and growth by rainbow trout (Salmo gairdneri) stocked in an eastern Oregon reservoir. Arch. Environ. Contam. Toxicol. (in press).
- Potter, L., D. Kidd, and D. Standiford. 1975. Mercury levels in Lake Powell. Environ. Sci. Technol. 9(1):41-46.
- Richins, R. T. and A. C. Risser Jr. 1975. Total mercury in water, sediment, and selected aquatic organisms, Carson River, Nevada - 1972. Pestic. Monit. J. 9(1):44-54.
- Riggs, V. L. 1978. Age and growth of walleye and sauger of the Tongue River Reservoir. M. S. Thesis, Montana State Univ. 53 pp.
- Scott, D. P. 1974. Mercury concentration of white muscle in relation to age, growth, and condition in four species of fishes from Clay Lake, Ontario. J. Fish. Res. Board Can. 31(11):1723-1729.
- Siemer, D. D., and R. Woodriff. 1974. Application of the carbon rod atomizer to the determination of mercury in the gaseous products of oxygen combustion of solid samples. Anal. Chem. 46(4):597-598.
- Steel, R. G. D., and J. H. Torrie. 1960. Principles and procedures of statistics. McGraw-Hill, New York. 481 pp.
- Study group on mercury hazards. 1971. Hazards of mercury. Environ. Res. 4(1):1-69.
- Thomas, R. L. and J. -M. Jaquet. 1976. Mercury in the surficial sediments of Lake Erie. J. Fish. Res. Board Can. 33(3):404-412.
- Thomas, R. L., J. -M. Jaquet, and A. Mudroch. 1975. Sedimentation processes and associated changes in surface sediment trace metal concentrations in Lake St. Clair, 1970-1974. pp. 691-708 In Internat. Conf. on Heavy Metals in the Environ. Ontario, Canada.
- U.S. Environmental Protection Agency. 1977. Quality criteria for water. Office of Water and Hazardous Materials, U.S. Environ. Protec. Agency, Washington D.C. 256 p.
- U.S. Food and Drug Administration. 1974. Action level for mercury in fish and shellfish. Fed. Register 39(236):42738-42740.
- Uthe, J. F., F. M. Atton, and L. M. Royer. 1973. Uptake of mercury by caged rainbow trout (Salmo gairdneri) in the South Saskatchewan River. J. Fish. Res. Board Can. 30:643-650.

- Van Engel, W. A. 1940. The rate of growth of the northern pike, Esox lucius Linnaeus, in Wisconsin waters. *Copeia* 1940(3):177-188.
- Van Voast, W. A., and R. B. Hedges. 1975. Hydrogeological aspects of existing and proposed strip coal mines near Decker, southeastern Montana. Bull. 97, State of Montana, Bureau of Mines and Geology, Butte, MT. 31 pp.
- Westoo, G. 1973. Methylmercury as a percentage of total mercury in flesh and viscera of salmon and sea trout of various ages. *Science* 181:567-568.
- Wood, J. M., F. S. Kennedy, and C. J. Rosen. 1968. Synthesis of methyl-mercury compounds by extracts of methanogenic bacterium. *Nature (Lond.)* 220:173-174.

APPENDICES

Appendix I. Total mercury concentrations (dry basis) in surficial sediment samples collected from various locations in the Tongue River Reservoir (see Appendix II for sample location relative to sample number).

Sample No. - $\mu\text{g Hg/g}$	Sample No. - $\mu\text{g Hg/g}$	Sample No. - $\mu\text{g Hg/g}$	Sample No. - $\mu\text{g Hg/g}$	Sample No. - $\mu\text{g Hg/g}$
1 - 0.053	36 - 0.050	71 - 0.048	106 - 0.029	141 - 0.048
2 - 0.057	37 - 0.055	72 - 0.054	107 - 0.011	142 - 0.050
3 - 0.054	38 - 0.055	73 - 0.057	108 - 0.022	143 - 0.039
4 - 0.047	39 - 0.061	74 - 0.074	109 - 0.053	144 - 0.056
5 - 0.042	40 - 0.061	75 - 0.064	110 - 0.051	145 - 0.057
6 - 0.045	41 - 0.049	76 - 0.013	111 - 0.044	146 - 0.057
7 - 0.042	42 - 0.046	77 - 0.078	112 - 0.027	147 - 0.053
8 - 0.017	43 - 0.059	78 - 0.075	113 - 0.013	148 - 0.040
9 - 0.003	44 - 0.051	79 - 0.016	114 - 0.024	149 - 0.055
10 - 0.021	45 - 0.013	80 - 0.039	115 - 0.036	150 - 0.046
11 - 0.053	46 - 0.050	81 - 0.039	116 - 0.010	151 - 0.056
12 - 0.053	47 - 0.027	82 - 0.025	117 - 0.010	152 - 0.075
13 - 0.034	48 - 0.043	83 - 0.007	118 - 0.044	153 - 0.054
14 - 0.035	49 - 0.023	84 - 0.071	119 - 0.030	154 - 0.046
15 - 0.035	50 - 0.007	85 - 0.038	120 - 0.019	155 - 0.045
16 - 0.073	51 - 0.015	86 - 0.043	121 - 0.019	156 - 0.047
17 - 0.050	52 - 0.060	87 - 0.051	122 - 0.017	157 - 0.017
18 - 0.051	53 - 0.058	88 - 0.022	123 - 0.046	158 - 0.038
19 - 0.060	54 - 0.057	89 - 0.003	124 - 0.026	159 - 0.038
20 - 0.020	55 - 0.021	90 - 0.055	125 - 0.044	160 - 0.059
21 - 0.009	56 - 0.054	91 - 0.046	126 - 0.031	161 - 0.051
22 - 0.015	57 - 0.054	92 - 0.058	127 - 0.013	162 - 0.031
23 - 0.016	58 - 0.053	93 - 0.042	128 - 0.036	163 - 0.028
24 - 0.020	59 - 0.056	94 - 0.040	129 - 0.008	164 - 0.056
25 - 0.014	60 - 0.015	95 - 0.045	130 - 0.043	165 - 0.051
26 - 0.014	61 - 0.020	96 - 0.007	131 - 0.044	166 - 0.048
27 - 0.042	62 - 0.053	97 - 0.016	132 - 0.063	167 - 0.031
28 - 0.031	63 - 0.053	98 - 0.011	133 - 0.031	168 - 0.031
29 - 0.011	64 - 0.010	99 - 0.033	134 - 0.035	169 - 0.052
30 - 0.059	65 - 0.029	100 - 0.037	135 - 0.055	170 - 0.037
31 - 0.055	66 - 0.058	101 - 0.044	136 - 0.050	
32 - 0.051	67 - 0.020	102 - 0.035	137 - 0.049	
33 - 0.030	68 - 0.024	103 - 0.044	138 - 0.044	
34 - 0.013	69 - 0.012	104 - 0.033	139 - 0.025	
35 - 0.056	70 - 0.063	105 - 0.048	140 - 0.031	

Appendix II. Map showing surficial sediment sampling locations in the Tongue River Reservoir.

