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## **Enhancement of Methylmercury Uptake in Fish by Lake Temperature, pH, and Dissolved Oxygen Gradients: Hypothesis**

### **Abstract**

Mercury analyses of fish tissue from two Oregon, U.S.A., lakes showed higher mercury concentrations in fish from a "eutrophic" lake than in fish from an adjacent "oligotrophic" type. This finding was attributed to differences between lakes regarding certain physical-chemical vertical gradients. In summer, the eutrophic lake developed thermal, pH, and dissolved oxygen conditions which may have contributed to higher rates of methylmercury uptake in fish. Conversely, fish in the oligotrophic lake were not subjected to these conditions, and consequently accumulated less mercury.

### **Introduction**

Mercury levels in fish are reportedly higher for oligotrophic lakes than eutrophic ones, provided both lake types are similarly contaminated with mercury (Jernelöv *et al.* 1975). This fact is caused by (1) the large biomass unit<sup>-1</sup> volume eutrophic lakewater which makes less mercury available unit<sup>-1</sup> biomass, (2) the supposed higher efficiency of energy transfer between trophic levels in oligotrophic lakes which concentrates more mercury in the higher carnivores and (3) the generally lower pH of oligotrophic lakes which tends to form biologically-available monomethyl mercury rather than the volatile dimethyl type.

Lake studies in Oregon, U.S.A., indicate that other factors promote mercury uptake in fish. These factors may have caused the high mercury concentrations in fish from a "eutrophic" lake which exceeded levels found in fish from an adjacent "oligotrophic" type.

### **Lake Descriptions**

The study lakes, Big Squaw and Upper Squaw lakes, are located in southwestern Oregon, approximately 4 km north of the Oregon-California border, and 105 km east of the Pacific Coast. The lakes lie within a few hundred meters of one another in the vicinity of at least one former mercury mining site. Cinnabar deposits, associated with serpentine and diorite rocks, are found in the general area.

Upper Squaw Lake lies at elevation 777 m, has a surface area of 0.082 km<sup>2</sup>, reaches a maximum depth of 16 m and drains into Big Squaw Lake via Squaw Creek. Big Squaw Lake, elevation 762 m, has a surface area of 0.196 km<sup>2</sup>, reaches its maximum depth at 34 m and is impounded by a small earth-filled dam at its west end.

Big Squaw Lake is less productive biologically than the upper lake, caused, in part, by its deep, steep-sided morphometry. Upper Squaw Lake is truly eutrophic, featuring substantial blooms of bluegreen algae (*Anabaena sp.*) which did not appear in Big Squaw during the study.

Mercury concentrations in Big Squaw Lake sediment samples differed substantially from Upper Squaw sediments. Average total mercury concentration per three sediment samples from the upper lake (0.51, 0.61, 0.56  $\mu\text{g g}^{-1}$ ) was 0.56  $\mu\text{g g}^{-1}$ , or double the amount for Big Squaw (0.20, 0.30, 0.26  $\mu\text{g g}^{-1}$ ;  $\bar{x} = 0.25 \mu\text{g g}^{-1}$ ), (unpublished data courtesy Umpqua Research Company, Myrtle Creek, Oregon).

### Procedures

Several fish species were collected from Squaw Lakes on 23, 24 April and 21, 22 August, 1975. Collections were made using graduated mesh gill nets (stretch mesh 1.3-5.1 cm) set at right angles to the shoreline for overnight periods of about 14 hours. Nets were retrieved the following day. Gilled fish were identified, weighed, measured (fork length), tagged, placed individually in plastic bags and stored in ice. Mercury analyses of fish tissues were initiated within four hours of collection. Whole fish were placed in a blender and reduced to a fine slurry. This slurry was subsampled for total mercury analysis by sulfuric acid digestion and flameless atomic absorption technique (Uthe *et al.*, 1970; Thorpe, 1971).

Some fish from the April study were delivered to the U.S. Food and Drug Administration (FDA) lab in Seattle, Washington, U.S.A., for mercury analyses. These were frozen, and analyzed for total mercury about six weeks later. Fish were grouped according to species, and each species was composited for analysis. Analyses were performed on "edible" parts of composites of each species (*i.e.*, only muscle tissue). Determination of total mercury was by cold vapor atomic absorption method, using sulfuric/nitric acid digestion with vanadium pentoxide catalyst.

Temperature and dissolved oxygen vertical profiles were obtained for each lake on 25 July and 22 August. Temperature was measured with a Yellow Springs Model 43TD tele-thermometer (scale calibrated in 1.0°C units). D.O. determinations were by Winkler method (azide modification). pH was measured with Corning Model 610A pH meter (expanded scale) on 25 July.

### Results and Discussion

Average concentrations of total mercury in Upper Squaw Lake fish were double the levels detected in the same species from the lower, more "oligotrophic" lake (Table 1). Composited fish analyses by FDA produced similar results (Table 2), although FDA values were relatively higher than those obtained by the private lab. This difference was caused, perhaps, by differences between laboratory digestion techniques or analytical methods (*e.g.*, composite versus individual fish, "edible" portion versus whole fish, delayed versus immediate analyses).

Limnological studies indicated that the lakes were gradually depleted of dissolved oxygen in deep waters during summer (Table 3). By August, Upper Squaw Lake was anaerobic, or nearly so, between nine meters and the bottom. The concentration of dissolved oxygen in the 6-9 m stratum, particularly during early morning, appeared to be intolerably low for fish (Table 3). Big Squaw Lake was well-oxygenated ( $>5.0 \text{ mg/l}$ ) in the top 20 m (Table 3), but was anaerobic along the bottom.

Both lakes developed relatively high temperature and pH conditions in the 0-4 m stratum during summer. Temperatures in the upper layers exceeded 16.0°C, and pH reached 9.1 in Upper Squaw Lake during a July algal bloom (Table 3).

TABLE 1. Concentrations of total mercury (wet weight basis) in Squaw Lakes Fish.

23-24 April				
Lake <sup>a</sup>	Species <sup>b</sup>	Weight (g)	Fork length (cm)	Mercury conc (p.p.m.)
U.S.	CT	74.1	19.6	0.216
U.S.	CT	45.6	17.3	0.047
U.S.	CT	122.5	24.1	0.220
U.S.	CT	109.0	22.9	0.268
U.S.	CT	102.9	22.1	0.240
U.S.	CT	120.3	23.6	0.222
U.S.	CT	82.8	20.8	0.190
U.S.	CT	88.3	21.6	0.213
U.S.	CT	81.4	20.6	0.264
U.S.	CT	88.0	21.6	0.218
U.S.	CT	72.8	19.3	0.159
U.S.	CT	91.6	20.8	0.174
U.S.	CT	69.3	19.1	0.312
U.S.	CT	69.0	18.3	0.167
U.S.	CT	47.8	17.3	0.222
				0.205 ( $\bar{x}$ )
B.S.	CT-RB	175.0	25.9	0.078
B.S.	CT-RB	203.7	26.2	0.099
B.S.	CT-RB	182.8	25.9	0.100
B.S.	CT-RB	150.0	24.9	0.082
				0.090 ( $\bar{x}$ )
B.S.	BH	63.4	17.3	0.046
B.S.	BH	84.5	18.3	0.051
B.S.	BH	90.9	19.1	0.049
				0.049 ( $\bar{x}$ )
B.S.	BC	35.0	13.0	0.067
B.S.	BC	39.7	13.7	0.075
B.S.	BC	53.0	15.2	0.044
B.S.	BC	49.1	14.7	0.053
B.S.	BC	41.1	14.2	0.044
B.S.	BC	46.9	14.2	0.064
B.S.	BC	50.8	15.0	0.054
B.S.	BC	45.4	14.7	0.078
B.S.	BC	45.4	14.2	0.083
B.S.	BC	46.2	15.5	0.068
B.S.	BC	46.4	14.5	0.064
B.S.	BC	43.8	14.2	0.050
B.S.	BC	38.0	14.5	0.048
B.S.	BC	40.5	14.2	0.063
B.S.	BC	40.8	14.0	0.070
B.S.	BC	41.5	14.0	0.056
B.S.	BC	41.1	14.2	0.066
B.S.	BC	41.3	14.0	0.120
B.S.	BC	37.2	13.2	0.061
B.S.	BC	41.1	14.0	0.048
B.S.	BC	50.0	15.0	0.050
B.S.	BC	41.2	14.0	0.074
B.S.	BC	44.9	14.0	0.048
B.S.	BC	44.8	14.0	0.071
B.S.	BC	36.1	13.7	0.048

TABLE 1. (continued)

B.S.	BC	49.5	14.5	0.048
B.S.	BC	51.2	15.2	0.068
B.S.	BC	37.0	13.7	0.048
B.S.	BC	43.9	13.7	0.061
B.S.	BC	33.0	13.5	0.052
				0.059 ( $\bar{x}$ )
21-22 August				
U.S.	CT-RB	61.8	18.8	0.018
U.S.	CT-RB	66.4	19.1	0.053
U.S.	CT-RB	61.7	19.4	0.123
U.S.	CT-RB	55.8	18.8	0.158
U.S.	CT-RB	96.6	22.9	0.193
U.S.	CT-RB	57.1	18.1	0.066
U.S.	CT-RB	62.0	18.7	0.098
U.S.	CT-RB	133.6	24.1	0.218
U.S.	CT-RB	119.1	24.8	0.143
U.S.	CT-RB	126.4	24.4	0.175
U.S.	CT-RB	146.0	26.7	0.169
U.S.	CT-RB	137.6	25.2	0.146
U.S.	CT-RB	129.6	25.7	0.149
U.S.	CT-RB	133.6	24.9	0.285
				0.149 ( $\bar{x}$ )
U.S.	BH	87.4	20.0	0.125
U.S.	BH	57.0	17.5	0.093
U.S.	BH	74.2	19.6	0.100
				0.103 ( $\bar{x}$ )
B.S.	CT-RB	77.0	20.8	0.030
B.S.	CT-RB	225.1	31.5	0.067
				0.049 ( $\bar{x}$ )
B.S.	BH	59.4	17.3	0.038
B.S.	BH	134.8	22.6	0.068
B.S.	BH	126.8	22.6	0.078
B.S.	BH	88.4	19.8	0.055
B.S.	BH	112.6	21.6	0.059
B.S.	BH	76.7	19.8	0.120
B.S.	BH	94.4	20.8	0.095
B.S.	BH	174.6	24.9	0.093
				0.076 ( $\bar{x}$ )
B.S.	BC	47.4	15.5	0.026
B.S.	BC	91.8	19.1	0.048
B.S.	BC	38.6	15.0	0.048
B.S.	BC	69.7	17.3	0.024
				0.037 ( $\bar{x}$ )
B.S.	BG	68.2	16.5	0.061
B.S.	BG	69.2	16.3	0.038
B.S.	BG	70.5	16.3	0.061
				0.053 ( $\bar{x}$ )

<sup>a</sup> U.S. = Upper Squaw Lake; B.S. = Big Squaw Lake

<sup>b</sup> CT = Cutthroat trout *Salmo clarki* Richardson  
 CT-RB = Cutthroat-Rainbow trout cross  
 RB = Rainbow trout *Salmo gairdneri* Richardson  
 BH = Brown bullhead *Ictalurus nebulosus*  
 BC = Black crappie *Pomoxis nigromaculatus*  
 BG = Bluegill *Lepomis macrochirus*

The rate at which fish assimilate toxicants (e.g., methylmercury) may be accelerated when the fish are stressed by increased temperatures, reduced dissolved oxygen concentration, high pH, or a combination of all three factors (Lloyd, 1961; Warren, 1971). Accordingly, as temperatures rise, the fish may respond with a sudden increase in dissolved oxygen consumption. Increased respiration rates will result in proportionately higher exposure to methylmercury and a consequent higher assimilation of the substance as respiratory surfaces receive larger quantities of mercury-laden water. Likewise, under conditions of low dissolved oxygen, fish may be forced to respire more rapidly to acquire sufficient oxygen. As the quantity of water increases across respiratory surfaces, the fish may be exposed to more methylmercury.

In Upper Squaw Lake, fish may be forced to occupy depths around 5-6 m to avoid less favorable temperatures of the overlaying water. In occupying the 5-6 m stratum, however, the fish are exposed to exceedingly low concentrations of dissolved oxygen, and find themselves compressed between high temperatures and pH above and dangerously low D.O. below. This condition may force an abnormally high number of fish into a narrow band of water resulting in excessive competition for food and space, and predation. Consequently, stress develops which forces abnormally high respiration rates and possible increase in mercury assimilation. Conversely, Big Squaw Lake appears to offer suitable fish habitat from thermocline (i.e., 6-7 m) to 20 m (Table 3). Thus, Big Squaw fish are less likely to encounter temperature, pH, and oxygen-related stress which may account for the lower mercury concentrations found in their tissues.

The higher concentrations of mercury in Upper Squaw Lake sediments may have little relationship to the levels of mercury found in Upper Squaw Lake fish. Langley (1973) showed an inverse relationship between methylation rates and the concentration of mercury in sediments. Methylation rate, according to Langley, was dependent far more on the organic enrichment and microbial density of the sediments rather than on mercury content.

TABLE 2. Mercury content of Squaw Lakes fish (total mercury, wet weight basis) collected 23-24 April 1975. Analyses by U.S. Food and Drug Administration, Seattle, Washington, U.S.A.

23-24 April					
Lake <sup>a</sup>	Species <sup>b</sup>	Weight (g) <sup>c</sup>	Fork length (cm) <sup>d</sup>	Number fish composited	Mercury conc. (p.p.m.)
U.S.	CT	51-150	17.8-25.9	8	0.68
U.S.	BH	33	14.0	1	0.52
B.S.	CT-RB	217-282	27.4-30.2	5	0.31
B.S.	RB	54-292	18.8-29.7	14	0.32
B.S.	BH	108-241	20.3-26.7	2	0.29
B.S.	BC	40-243	13.5-24.9	26	0.26
B.S.	BG	39- 89	11.9-16.3	8	0.21

<sup>a</sup> U.S. = Upper Squaw Lake; B.S. = Big Squaw Lake

<sup>b</sup> CT = Cutthroat trout *Salmo clarki* Richardson  
 CT-RB = Cutthroat-Rainbow trout cross  
 RB = Rainbow trout *Salmo gairdneri* Richardson  
 BH = Brown bullhead *Ictalurus nebulosus*  
 BC = Black crappie *Promoxis nigromaculatus*  
 BG = Bluegill *Lepomis macrochirus*

<sup>c</sup> Range of individuals in composite

<sup>d</sup> Range of individuals in composite

TABLE 3. Temperature, pH and dissolved oxygen profiles in Squaw Lakes, Oregon, Summer, 1975. Values for temperature and dissolved oxygen (D.O.) reported in degrees centigrade and milligrams per liter, respectively.

Depth (m)	Big Squaw			Upper Squaw		
	Temp.	25 July pH	D.O.	Temp.	25 July pH	D.O.
Surface	24.2	7.9	7.7	23.6	8.9	13.1
1	23.6			22.1		
2	23.4			19.9		
3	23.0	8.2	7.8	18.3	9.1	14.4
4	20.8			16.4		
5	14.3			14.4		
6	11.3	7.8	9.1	13.1	8.4	10.7
7	8.5			11.9		
8	7.7			10.5		
9	7.1	7.6	6.7	8.8	8.2	4.1
10	6.8			7.8		
11	6.6			7.4		
12	6.4	7.6	6.8	7.0	8.3	1.2
13	6.2			7.0		
14	6.1			7.0		
15	5.9	7.5	6.8	7.0	8.3	0.08
16	5.9			7.7		
17	5.7			6.7		
18	5.6			6.1		
19	5.6			6.1		
20	5.5	7.5	6.2	6.1		
25	5.5			6.1		
27	5.4			6.1		
						2.2
						10.5
						10.2
						9.9
						7.0
						3.1
						0.22
						0.0
						5.2
						0.23

a 1500 hrs.

b 0500 hrs.

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