

PRELIMINARY ASSESSMENT

Black Butte Mine  
Township 23 South, Range 3 West, Section 8, W. M.  
Approximately 10 miles south of Cottage Grove, Oregon  
EPA ID # OR0000515759

April 1, 1996

Prepared for: David Bennett, Cleanup and Site Assessment Unit 2  
U.S. Environmental Protection Agency  
Region 10  
Seattle, Washington 98101

Prepared by: Keith Andersen, Remedial Action Specialist  
Oregon Department of Environmental Quality  
Site Assessment Section, Cleanup Programs  
Western Region  
Eugene, Oregon 97401

130161



## Introduction

Pursuant to a Cooperative Agreement (V-990519-01-0) between the U.S. Environmental Protection Agency (EPA) and the Oregon Department of Environmental Quality (DEQ), the DEQ conducted a Preliminary Assessment (PA) of the site known as the Black Butte Mine located near Cottage Grove, Lane County, Oregon.

PAs are intended generally to identify potential hazards at a site, identify sites that require immediate action, and to establish priorities for sites requiring in-depth investigations. The PA is based on readily available information about the site and is not a full investigation or characterization of the site.

The Black Butte Mine PA was conducted to identify potential public health and environmental threats related to the site. The PA is based on data derived from the sources listed in the back of this report. The scope of the investigation includes review of available file information, interviews, a target survey, and an on-site reconnaissance inspection.

### General Site Information

Site Name: Black Butte Mine

Site CERCLIS Number: OR0000515759

DEQ ECSI Number: 1657

Site Address: T23S, R3W, Sec. 8, and T23S, R3W, Sec. 16  
Approx. 10 Miles south of Cottage Grove, Oregon 97424

Site Coordinates: 43° 34' 42" N, 123° 03' 58" W

Date: April 1, 1996

### Site Description:

The site is located in southern Lane County, in the Coast Fork Willamette River basin, approximately 10 miles south of Cottage Grove, Oregon off London Road. Land use in the vicinity is primarily rural residential and recreational. The site is located on the northeast flank of Black Butte. The mine itself is in the NW 1/4 of Section 16, T.23 S., R.3 W. The abandoned kiln and tailings pile are located in the SE 1/4 of Section 8, T.23 S., R.3 W. (see Figure 1). Several buildings remain on site, including two dilapidated, unoccupied houses, and several sheds associated with mine adits. The tailings pile, located along the south bank of Dennis Creek, contains an estimated 300,000 cubic yards of material. Topographic maps and aerial photos also show several unimproved roads on the site.

The mine was first operated in the late 1890s. Ore was extracted and crushed in preparation for processing. The crushed ore was heated in the kiln to drive off mercury vapor. The mercury vapor was then condensed and liquid mercury bottled for shipment. The mine operated intermittently through the late 1960s, with peak production occurring during the period 1927-1943. The mine was the second largest mercury producer in Oregon. Much of the mine area was logged in the early 1990s, and some features of the mine were obscured by logging debris.

The site is currently owned by the Land and Timber Company. The site contact is Roger Villanueva, in Coos Bay, Oregon, phone number (541) 269-0222. According to the registered agent for the Land and Timber Company, Robert J. Custis, there is another group associated with the Land and Timber Company, represented by Sandy Simmons. There is no additional information available regarding Ms. Simmons.



### Site Geology:

The Black Butte Mine and surrounding vicinity is underlain by a sequence of hydrothermally altered mercury-bearing andesitic lavas, silicic ash tuff, and volcanic breccias belonging to the late Eocene-early Oligocene Fisher Formation (Tertiary). Locally these deposits have been injected by sills, plugs, and feeder dikes of Pliocene, Miocene, and possibly Oligocene age basalt and andesite. The bedrock is faulted and fractured. The Black Butte fault is exposed on the summit of Black Butte and in the underground mine workings and trends approximately N69W and dips 65 northeast. The mercury ore deposit (cinnabar) appears to have formed along this fault, thought to be the primary conduit for ascending hydrothermal solutions.

The site is characterized by primarily shallow soils on steep slopes on the hillsides; thicker accumulations of recent alluvium occur along the streams.

### Hydrogeology

The aquifers in the vicinity of the mine are the Fisher Formation (bedrock aquifer) and the alluvial aquifer along Dennis Creek, Garoutte Creek, and the Coast Fork Willamette River. Depth to groundwater and hydraulic conductivity of these aquifers at the mine site is unknown. Well logs in the vicinity of the mine indicate that the shallowest depth to water bearing strata in bedrock is at 29 feet below ground surface (bgs). Local groundwater gradients are unknown but are likely toward the streams with a component toward Cottage Grove Reservoir. The nearest spring is London Springs, located approximately 4 miles north of the site. Its source, use, and quality are unknown.

### Investigation History

The Black Butte Mine site was identified as a potential source of mercury contamination in Cottage Grove Reservoir by the Mercury Working Group of DEQ's Water Quality Division during an evaluation of Oregon's lakes. In 1994 the site was referred to DEQ's Site Assessment Section (SAS) staff for review. The SAS review resulted in a recommendation for further investigation at the site at a medium priority. DEQ is performing this federal Preliminary Assessment at the site under a cooperative agreement with EPA Region 10.

### Background

There has apparently been no other formal environmental investigation of the site to date. However, several organizations have undertaken sampling efforts in the vicinity of the site. These sampling efforts included limited sediment and tissue sampling done by the Oregon State University (OSU) Department of Fisheries and Wildlife in 1990, tissue analysis on an eagle's egg by the U.S. Fish and Wildlife Service in 1992, limited sediment and tissue analyses by U.S. Geological Survey (USGS) in 1993, and somewhat more extensive sediment sampling by the OSU Department of Fisheries and Wildlife in 1992 and 1994.

The initial OSU study compared samples from three Oregon reservoirs. Samples from Cottage Grove Reservoir included tissue samples from five largemouth bass and several sediment samples. The tissue from the oldest two fish (four and five years, respectively) showed mercury levels of 1.49 and 1.79 parts per million (ppm) in muscle tissue. The U.S. Food and Drug Administration limit for mercury in commercially caught fish is one ppm. Sediment concentrations averaged 0.84  $\mu\text{g/g}$ , dry weight.

In 1992 an addled egg was collected from a bald eagle's nest adjacent to the reservoir by the U.S. Fish and Wildlife Service. The egg was analyzed for trace elements and mercury was discovered at 2.9  $\mu\text{g/g}$  dry wt. and 0.76  $\mu\text{g/g}$  wet or fresh weight. According to the Fish and Wildlife Service fact sheet *Results of Cottage Grove Bald Eagle Egg Analysis*, these levels are significantly higher than nationally reported mercury levels for bald eagle eggs.

The USGS sediment data was generated as part of periodic state-wide sampling. Analysis of three samples in the vicinity of Back Butte Mine indicated 2.5 ppm mercury in sediment in Dennis Creek below the Mine, and .87 ppm in Sculpin (a small fish species) tissue at the same location. A sediment sample taken from the Coast Fork at London, Oregon, downstream from the mine and upstream from Cottage Grove Reservoir, had a mercury concentration of 1.4 ppm, and Sculpin tissue sampled there had a maximum concentration of 0.52 ppm. A sediment sample taken from Cottage Grove Reservoir had a concentration of 0.50 ppm.

OSU completed additional sampling in 1992. This work was documented in a report titled *Mercury Dynamics and Methylmercury Accumulation by Fish in Three Oregon Reservoirs* (Curtis and Allen-Gil, March 10, 1994) and detailed apparent elevated levels of methylmercury in fish tissue in Cottage Grove Reservoir. This report noted that the tissue concentrations approached or exceeded the United States Food and Drug Administration limit for human consumption for commercially caught fish.

In 1994, researchers with Oregon State University obtained soil samples from near the mine and sediment samples from Cottage Grove Reservoir and its tributaries. The sampling data from the recent OSU research appears to support the conclusion that elevated mercury levels in sediments can be traced to the Dennis Creek drainage, and may result from off-site transport of soils and mine tailings from the Black Butte Mine.

The soil levels detected at the mine and in the vicinity of the kiln ranged from approximately 100 ppm to 350 ppm. The residential soil maximum for mercury in the DEQ Soil Cleanup tables is 80 ppm, and the Industrial cleanup level is 600 ppm. The EPA Region III Risk-Based Concentration (RBC) Tables show an industrial soil ingestion RBC of 610 mg/kg and a residential soil ingestion RBC of 23 mg/kg. The soil screening level for transfers from soil to groundwater is 3 mg/kg. No information on leachate concentrations at the site is available.

The analysis results are found below in tables 1 & 2, and corresponding sampling locations are attached as figures 2 & 3.

Sample Site	Number of Samples	Mercury Concentration in Soil or Sediment, $\mu\text{g/g}$ dry wt.
1	1	190
2	2	$271 \pm 7$
3	2	$267 \pm 29$
4	2	$223 \pm 119$
5	2	$22 \pm 21$
6	2	$19 \pm 1$
7	3	$3 \pm 1$
8	3	$0.3 \pm 0.2$
9	3	$1.3 \pm 0.6$

**Table 1:** Mercury Content in Soil and Sediment Samples in the Vicinity of Black Butte Mine. Modified from Jeong-Gue, Park, OSU Department of Fisheries and Wildlife, unpublished data, 1994. Also see Figure 2.

Sample Site	Mercury Concentration in Sediment, $\mu\text{g/g}$ dry wt.
1	$0.83 \pm 0.12$
2	$< 0.01$
3	$0.08 \pm 0.01$
4	$1.75 \pm 0.10$
5	$< 0.01$
6	$0.35 \pm 0.05$
7	$0.18 \pm 0.03$
8	$0.68 \pm 0.07$
9	$1.03 \pm 0.02$
10	$1.11 \pm 0.01$

**Table 2:** Mercury Concentrations in Sediment Samples from Cottage Grove Reservoir. Modified from Jeong-Gue, Park, OSU Department of Fisheries and Wildlife, unpublished data, 1994. Also see Figure 3.

Figure 2

Soil and Sediment Sampling Site Locations  
Near Black Butte Mine. From Jeong-Gue,  
Park, OSU Department of Fisheries and  
Wildlife, unpublished data, 1994.

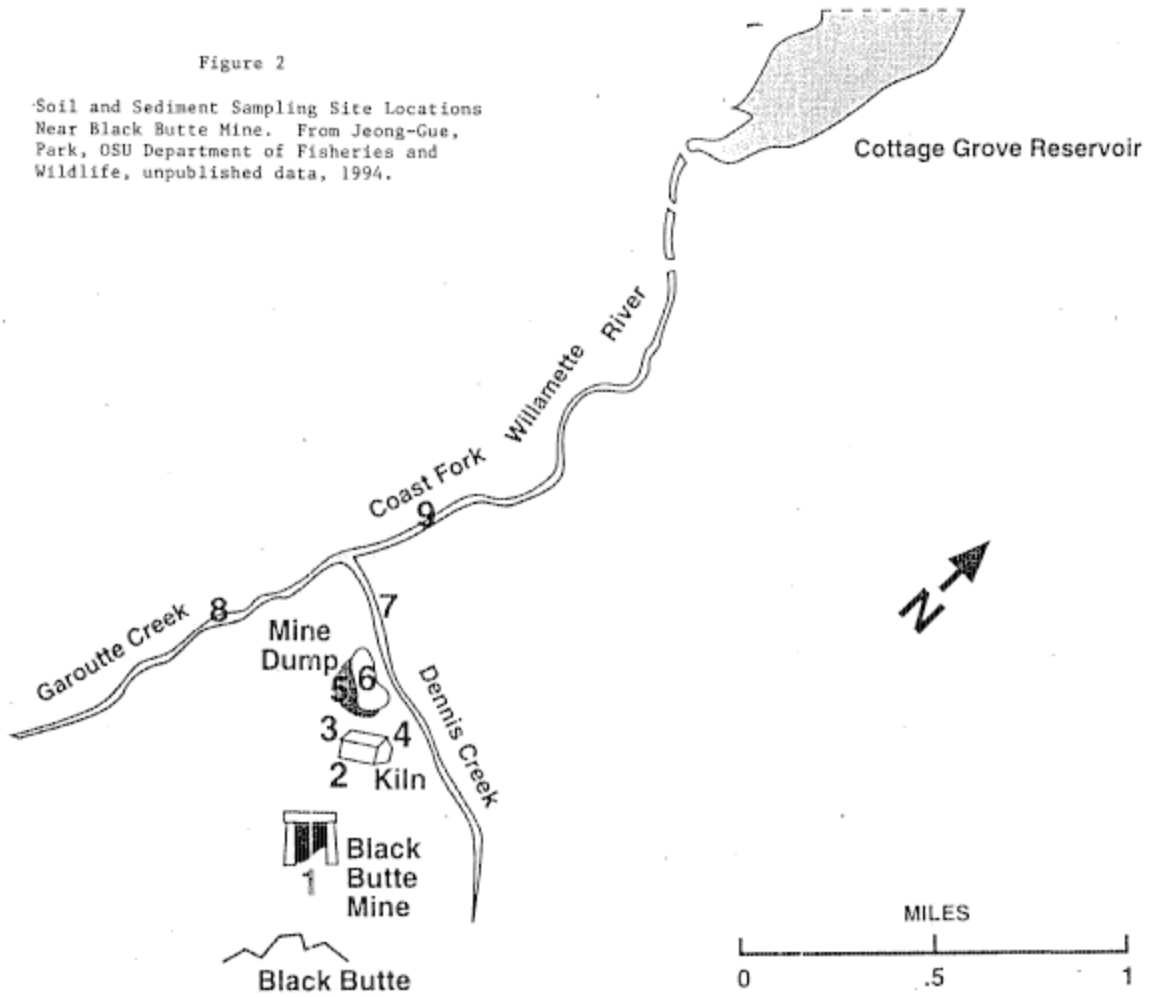
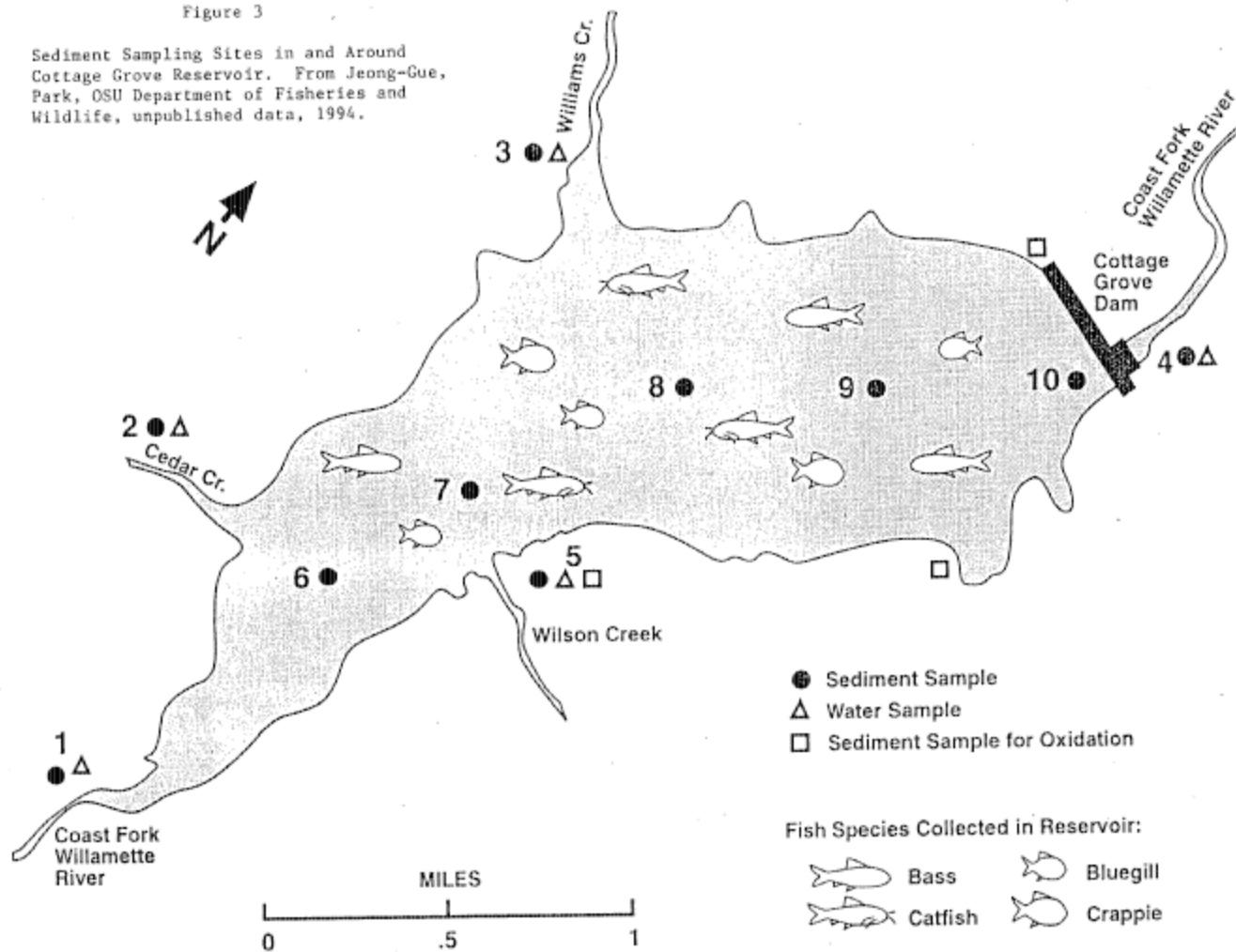




Figure 3

Sediment Sampling Sites in and Around Cottage Grove Reservoir. From Jeong-Gue, Park, OSU Department of Fisheries and Wildlife, unpublished data, 1994.



In March 19, 1996, DEQ staff visited the site in conjunction with this PA. Public access to the site is limited by a gate on the property entrance road. Permission to enter the property was obtained from the property owner prior to the DEQ visit. The purpose of the site visit was to evaluate current site conditions, confirm anecdotal information about the tailings pile and the road system at the site and to identify other potential environmental issues associated with the site.

Several of the roads that currently traverse the site appear to have been surfaced with the reddish mine tailings (see Photos 1&2, Appendix A). As noted previously, much of the site was logged recently, and road building for the logging efforts apparently made extensive use of the tailings. A failing portion of one of the main roads at the site (see Photo 3) also shows clearly the use of the distinct reddish mine tailings that appear to have been used over the years for roads at the site.

The tailings pile is quite extensive. The top of the pile is fairly level, and has been used by woodcutters at the site (see Photo 4). There is also evidence of excavation of the pile over time, apparently to provide the road surfacing rock as described above. Part of the toe of the tailings pile slopes directly into Dennis Creek at a very steep angle, and appears to be eroding directly into the creek (see Photo 5). The tailings pile stands approximately 50 feet above the creek and appears to front the creek for approximately 600 feet (see Photos 6&7). A gravel bed in the creek shows signs of the red-colored ore (see Photo 8) at a location downstream from the tailings pile.

The mine adits observed at the site are full of water, and at one location the water is flowing out of the adit toward a road (see Photo 9). As noted above, some parts of the roads at the site are subject to mass wasting, which may cause additional sediment loading to surface water features.

Based on the indications of elevated mercury levels in fish at the reservoir, the Lane County Health Department, in cooperation with the Oregon Health Division, has posted health advisories recommending that fish consumption be limited for fish caught in the reservoir (see Photo 10). The warning recommends no fish consumption for pregnant women and children under six years and 8 ounces per week for healthy adults. Based on weekday recreational use of the reservoir, this warning may not be heeded (see Photos 11 & 12).

### Pathway Information:

#### Soil Pathway

The soil pathway has been impacted, and elevated levels of mercury are found in soils around the mine and associated structures. While the site is fairly remote, and is accessible only via a steep gravel road, there are indications that the site is used for woodcutting, which could result in dermal exposure to mercury-bearing tailings. See Appendix B for Pathway summary sheets.

### Surface Water Pathway

The surface water pathway has also been impacted. It appears that surface transport of soil and sediment has resulted in widespread contaminant distribution downstream of the mine site in Dennis Creek, the Coast Fork Willamette River, and in Cottage Grove Reservoir. The contamination is reflected in downstream sediment mercury concentrations that appear to be higher than mercury concentrations in upstream sediment samples. Cottage Grove USGS and OSU researchers have also discovered elevated mercury levels in fish tissue in some of the water bodies noted above. It is believed that inorganic mercury available in sediments is biologically transformed to methylmercury, a form that is available to benthic organisms, plants and other aquatic organisms. As mercury tends to bio-accumulate, this results in increasing concentrations in fish tissues over time. US Army Corps of Engineers data indicates that in 1993, 350,000 visitors used the reservoir and that 24% of these visitors fished the lake. In addition, recreational use of downstream water bodies may result in dermal exposure to sediments. One surface water intake is present within a 5 mile radius of the site. The London Water Co-op utilizes a reach of Beaver Creek before its' confluence with the Coast Fork Willamette River. Therefore, there is very low risk of mercury contamination from the Black Butte Mine affecting this water supply. There are no other drinking water intakes within 15 miles downstream of the site. As noted previously, Cottage Grove Reservoir is used as a nesting area for the bald eagle, a federally designated threatened species. See Appendix B.

### Groundwater Pathway

It is not clear whether the groundwater pathway has been impacted. Two aquifers have been identified in the site vicinity: a shallow alluvial aquifer along the creeks and rivers and a deeper, bedrock aquifer. There are no public groundwater supply wells within a 5 mile radius of the site. There are 13 households present within a 1 mile radius of the site, which use groundwater for domestic water supply. Well logs for the vicinity of the site show that the first water bearing strata encountered is at depths ranging from 29 to 93 feet below ground surface (bgs), in the bedrock (see Appendix C). There is no information to indicate that mine activities have impacted groundwater at the site, and it seems unlikely that potential mine-related mercury contamination of groundwater could be differentiated from groundwater that comes into contact with naturally occurring mercury in area soils. See Appendix B.

### Air Pathway

The air pathway may have historically been impacted while the mercury kiln was operating, but there does not appear to be a major current impact to the air pathway. Wind-born dust transport from tailings piles or exposed soils may have a potential impact to nearby streams, and could potentially impact down-wind residences, however, the prevailing winds in this area are from the west to southwest, and would tend to blow away from the nearest residences. The air pathway will not be evaluated further.

### Recommendation/Action:

As noted previously, mercury tends to bio-accumulate. Although there is limited data available, and this data is of uncertain quality, it appears that mercury loading in streams and in Cottage Grove Reservoir is exacerbated by historic and current surface water transport of sediments from the Black Butte Mine area and the Black Butte Mine tailings pile. Fish tissue samples downstream of the mine have been found to contain elevated levels of methylmercury.

Based on the demonstrated impact to fish in Cottage Grove Reservoir, and the associated potential threat to human health, it is recommended that a Site Inspection be carried out to more fully evaluate the threat associated with the mine and tailings pile. Concern exists about potential impacts to soil, surface water and groundwater pathways, therefore sampling of domestic wells in the vicinity of the site and sediment and fish tissue samples from downstream surface water bodies appears warranted.

### References:

*An Ecoregion Approach to Mercury Dynamics in Three Oregon Reservoirs*, Susan M. Allen and Lawrence R. Curtis, Oak Creek Laboratory of Biology, Department of Fisheries and Wildlife, Oregon State University, June 10, 1991.

*Mercury Dynamics and Methylmercury Accumulation by Fish in Three Oregon Reservoirs*, Lawrence R. Curtis and Susan Allen-Gil, Department of Fisheries and Wildlife, Oregon State University, March 10, 1994.

Unpublished data collected by the Oregon State University Department of Fisheries and Wildlife, Jeong-Gue, Park.

*Quicksilver Deposits in Oregon*, H. Brooks, Oregon Department of Geology and Mineral Industries, Miscellaneous Paper # 15, 1971.

*Soil Survey of Lane County Area, Oregon*, USDA Soil Conservation Service.

*Results of Cottage Grove Bald Eagle Egg Analysis*, U.S. Fish & Wildlife Service, Portland Field Office.

USGS data on mercury, multiple station analysis, 1992 and 1993.

## Appendix A

Appendix A

**Photo Log: Black Butte Mine Site**

**Date:** 3/19/96 (Note: Due to the Leap Year, photo date feature printed 3/20/96)

**Location:** Black Butte Mine, Lane County

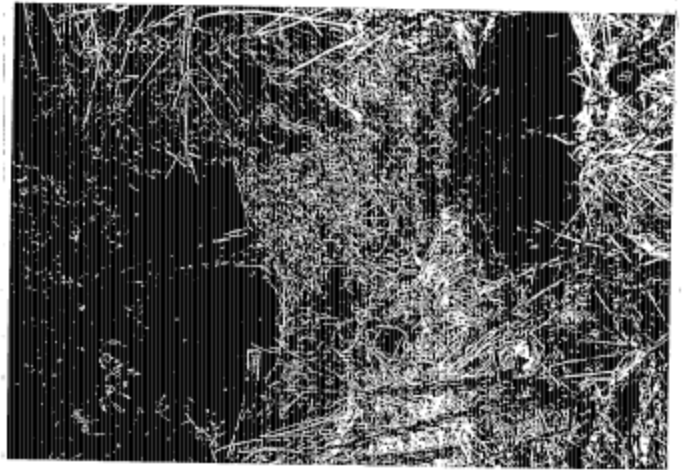
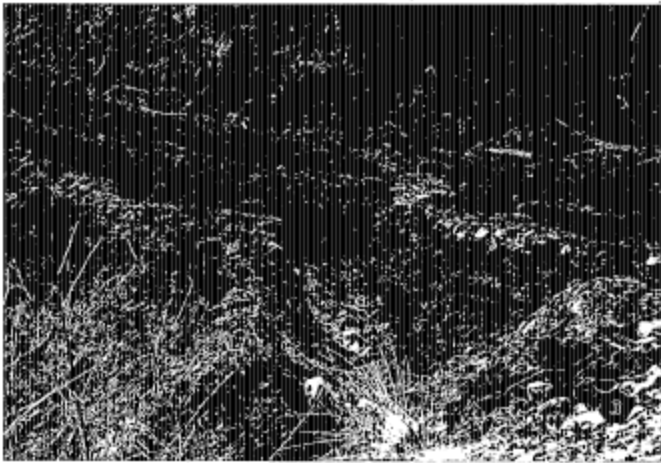
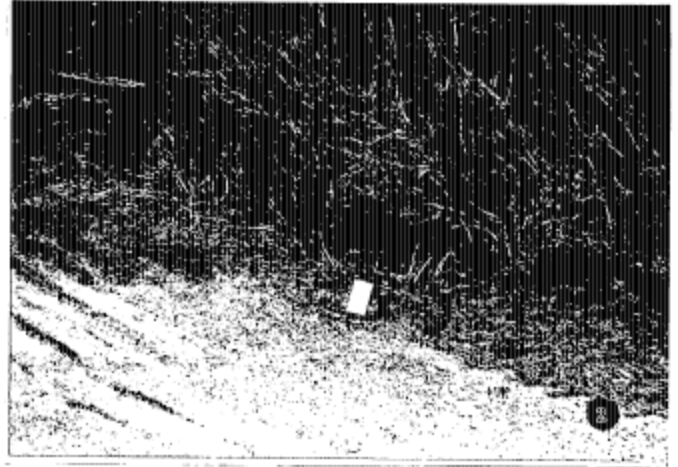
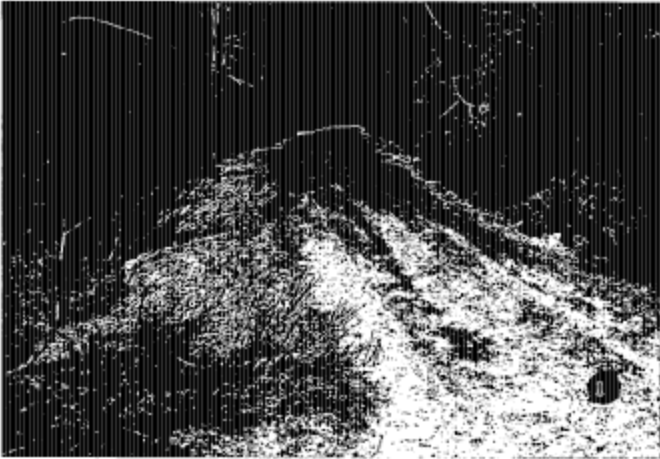
**Weather:** Partly cloudy, cool

**Photographer:** Keith Andersen

**Camera:** Pentax IQ Zoom WR90    **Lens:** Zoom 35-90

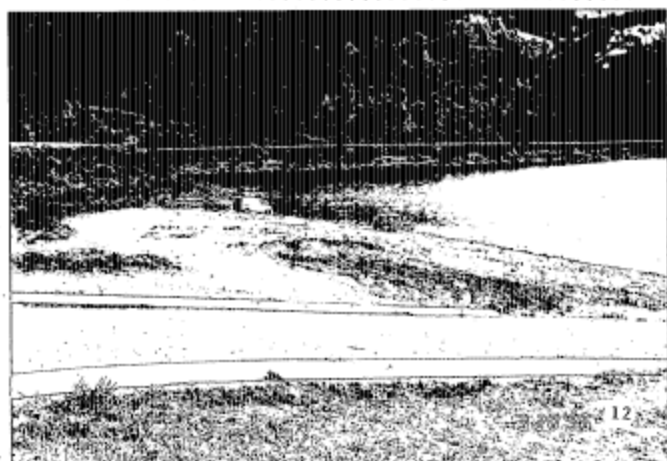
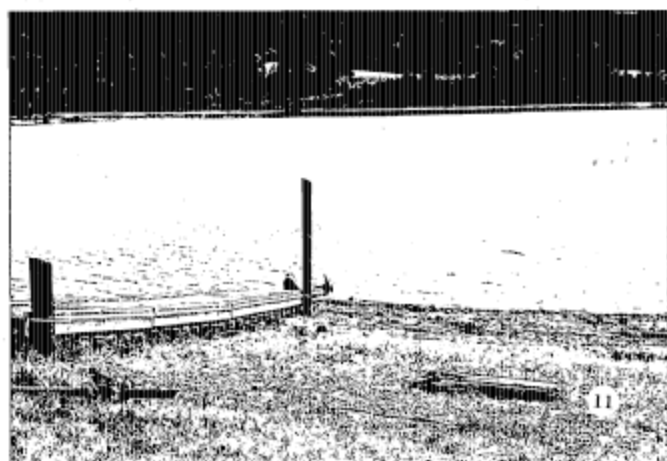
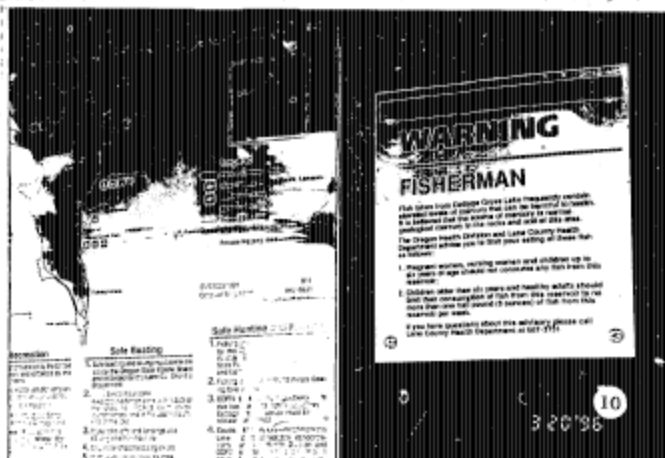
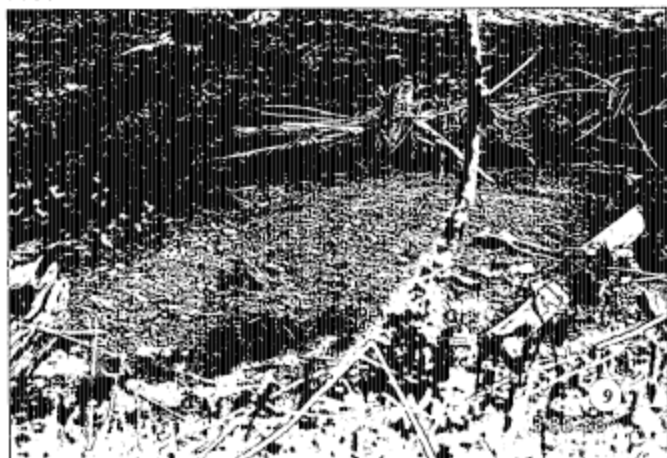
**Film:** Kodak ASA200

<u>No.</u>	<u>Direction Facing</u>	<u>Description</u>
1	N	Road surfaced with tailings
2	NE	Erosion of road surface
3	SE	Mass wasting exposing layers of tailings
4	SE	Adit with water draining towards road
5	W	Excavated area of tailings pile
6	N	Steep slope of tailings pile above creek
7	E	Side view of toe of tailings pile
8	S	Front view of tailings pile
9	E	Sediment bar in Dennis Creek
10	S	Health warning at Cottage Grove Lake
11	SE	Recreational anglers at C.G. Lake
12	E	Recreational use at C.G. Lake









## Appendix B

## Groundwater Pathway

No.	Aquifer Name	Type	Overlaying No.	Interconnected with
1	Alluvial	Unconfined	NA	NA
2	Fisher Formation	Confined	NA	NA

### Containment

No.	Source ID	Containment Value
1	Tailings Pile	10 (from Table 3.2)
1	Roads	10 (from Table 3.2)

### Net Precipitation

Net Precipitation: 60 inches/year

### Depth to Aquifer

A.	Depth to Hazardous Substances -	0 feet bgs
B.	Depth to Aquifer from Surface -	> 29 feet bgs
C.	Depth to Aquifer (A - B) -	> 29 feet

### Travel Time

All layers Karst?	No
Thickness of layer(s) with Lowest conductivity?	29 feet
Hydraulic Conductivity?	Unk*, use $10^{-2}$ to $10^{-6}$ cm/sec

\* - estimated ranges for fractured igneous or metamorphic bedrock

HRS Groundwater Form  
Page 2

Population by Well

No. of Wells	Well ID #	Sample Type	Distance (miles)	Contamination Level	Population
1	NA	NA	0-1/4	NA	1
3	NA	NA	1/4-1/2	NA	3
3	NA	NA	1/2-1	NA	12
21	NA	NA	1-2	NA	46
24	NA	NA	2-3	NA	46
36	NA	NA	3-4	NA	82
31	NA	NA	4-5	NA	71

Potential Contamination by Distance Category

Distance Category ( miles)	Population
0-1/4	1
1/4-1/2	4
1/2-1	16
1-2	62
2-3	108
3-4	190

Resources: NA

Wellhead Protection Area: NA

POTENTIAL TO RELEASE

## Containment

→

## Distance to Surface Water 0 feet

A. Drainage Area	< 50 acres
B. 2-year, 24-hour Rainfall	3.2 inches
C. Soil Group	B

#### Potential to Release by Flood

No.	Flood Containment Value	Flood Frequency Value
1	10	25
2	10	7

#### Potential Contamination

Intake ID	Average Annual Flow (cfs)	Population Served
NA	NA	NA

#### Nearest Intake

Location of Nearest Drinking Water Intake: Nearest surface water intake is 4.2 miles distant, on a tributary of the Coast Fork Willamette, and is not impacted by the site contamination. There are no other known surface water intakes within 15 miles down stream of the site.

#### Resources

Resources Use: Cottage Grove Reservoir and the Coast Fork Willamette River are used for recreational fishing

#### Potential Contamination

Fishery	Annual Production (pounds)	Type of Surface Water Body	Average Annual Flow (cfs)
NA	NA	NA	NA

#### Sensitive Environments

Type of Surface Water Body	Sensitive Environment	Sensitive Environment Value
Large Stream (17090002-013)	Endangered species habitat	75

Wetlands

Type of Surface Water Body	Wetlands Frontage
NA	NA

## SOIL EXPOSURE PATHWAY

Workers: NA

Resources:

Terrestrial Sensitive Environment	Value
NA	Table 5-5

### Likelihood of Exposure

No.	Source ID	Level of Contamination	Attractiveness/ Accessibility	Area of Contamination (sq. feet)
1	Tailings Pile	50 mg/kg mercury	10	162, 000
2	tailings on road surfaces	50 mg/kg mercury	10	113,000



## Appendix C

---

## REFERENCES

U.S. FISH & WILDLIFE SERVICE  
PORTLAND FIELD OFFICE

RESULTS OF COTTAGE GROVE BALD EAGLE EGG ANALYSIS

BACKGROUND

Cottage Grove Reservoir, located in Lane County, Oregon, exists in a geological region characterized by volcanic formations containing naturally occurring mercury. As a result, extensive mercury mining once occurred in this area. The second largest mercury mine in Oregon, Black Butte Mine, lies approximately 2 miles south of Cottage Grove Reservoir in its headwaters and was active intermittently from 1882 to 1966 (Brooks 1971, as cited in Allen and Curtis 1991).

Aquatic organisms can bioaccumulate mercury to dangerous levels in bodies of water in areas containing high levels. Bacteria living in sediment can convert mercury into an organic form known as methylmercury. Methylmercury is absorbed directly by aquatic organisms and is also taken in through the food they eat. Mercury burdens have been detected in water, sediments, and fish in Cottage Grove Reservoir and are most likely derived from natural mercury deposits exacerbated by past mining within the basin (Allen and Curtis 1991).

A substantial proportion of fish collected from Cottage Grove Reservoir show mercury concentrations which exceed the U.S. Food and Drug Administration maximum limit for mercury in commercially sold fish (1 ppm) [Allen and Curtis 1991]. Consequently, a public health advisory has been issued for a number of years for consumption of fish taken from the Cottage Grove Reservoir. Concerns have developed regarding mercury accumulation in piscivorous birds foraging in Cottage Grove Reservoir and the potential for adverse impacts.

A bald eagle (*Haliaeetus leucocephalus*) nest, located on the Cottage Grove Reservoir, has been occupied since 1986. Breeding attempts were unsuccessful in 1988, but successful at producing 2 young in both 1989 and 1990. In 1991, the original nest was blowdown and a new nest was built, successfully producing 1 young. The nest failed in 1992 and an addled egg was collected in May for chemical analysis of trace elements.

METHODS

Specialized tree-climbing equipment was utilized to enter the eagle nest and collect the addled bald eagle egg. The egg was cooled on ice during transport to the Portland Field Office where it was refrigerated at 4°C until processing. Egg length, width, whole weight, and volume were measured. The egg was scored at the equator with a scalpel and contents were released into a chemically-cleaned glass jar. Embryonic development was noted. Egg contents were frozen at -13°C until shipping to the Patuxent Analytical Control Facility (PACF) for trace element analysis.

Analytical methods included sample homogenization followed by digestion for Inductively Coupled Plasma Emission (ICP) and Graphite Furnace Atomic Absorption Measurements (GFAA). ICP measurements were quantified using a Leeman Labs Plasma Spec I sequential or ES2000 simultaneous spectrometer and GFAA measurements were quantified using a Perkin-Elmer Zeeman 3030 or 4100ZL atomic absorption spectrometer. CVAA was used to quantify levels of mercury using SNC14 as the reducing agent employing a Leeman PS200 Hg Analyzer.

Quality control and quality assurance of analytical data was reviewed by PACF. Acceptable performance (recovery variation averaged <20% for all chemicals detected) on spikes, blanks, and duplicates was documented in the laboratory quality control report.

## RESULTS

Results of the chemical analysis are presented in Table 1. Many of the trace elements including arsenic, barium, beryllium, cadmium, chromium, molybdenum, nickel, lead, and vanadium were below detection levels. Concentrations of selenium, and zinc detected in the egg sample did not surpass levels associated with adverse impacts (Heinz *et al.* 1989, Gasaway 1972, respectively). Other elements detected in the egg (aluminum, iron, magnesium, manganese, strontium) are not typically associated with impacts to fish-eating birds and information is lacking regarding residue levels related to adverse impacts.

The concentration of boron detected in the eagle egg does not indicate a level of concern. Although the residue detected in the Cottage Grove egg exceeds the median lethal dose ( $LD_{50}$ ) for borax injected into domestic chicken eggs, it does not exceed the  $LD_{50}$  for boric acid injected in chicken eggs (Eisler 1990). Further, the boron concentration in the Cottage Grove bald eagle egg did not exceed the level Smith and Anders (1989) associated with deleterious affects in mallard ducklings.

Mercury concentrations in the bald eagle egg were 2.9  $\mu\text{g/g}$ , dry weight (0.76  $\mu\text{g/g}$ , wet weight). The significance of mercury residues in bald eagles is not fully understood, and it is not currently possible to predict a mercury level associated with unsuccessful reproduction (Eisler 1987). For example, bald eagle eggs collected nationwide contained mean mercury concentrations of 0.15  $\mu\text{g/g}$  (fresh weight) from successful nests in contrast to 0.11  $\mu\text{g/g}$  in eggs from unsuccessful nests (Wiemeyer *et al.* 1984, as cited in Eisler 1987). Although the precise significance of the mercury residue in the Cottage Grove eagle egg cannot be determined, the egg concentration is up to 7 times greater than nationwide levels reported above (Wiemeyer *et al.* 1984). Residues were also higher than those found in eagle eggs collected from the Columbia River in 1991 (0.11 and 0.25  $\mu\text{g/g}$  wet weight) [U.S. Fish & Wildlife Service unpublished data] and 1986 (geometric mean = 0.19  $\mu\text{g/g}$  wet weight;  $n=13$ ) [Garrett *et al.* 1988]. A mercury threshold level for raptor species is unavailable, therefore, data for other avian species were used for comparison. The mercury residue in the Cottage Grove bald eagle egg approaches levels associated with reproductive impairment in a variety of other avian species: white-tailed sea eagles, common loon, and several seed eating species were affected at residues of 1.3 to 2.0  $\mu\text{g/g}$  fresh weight (Fimreite 1979 as cited in Eisler 1987); ring-necked pheasants between 0.9 to 3.1  $\mu\text{g/g}$  fresh weight (Spann *et al.* 1972 as cited in Eisler 1987); and mallard ducks 0.79 to 0.86  $\mu\text{g/g}$  fresh weight (Heinz 1979).

## CONCLUSIONS

The majority of trace element concentrations detected in the bald eagle egg collected from Cottage Grove do not appear at levels of concern. However, bald eagle egg mercury residues surpass averages for the nation and the Columbia River and approach levels associated with reproductive impairment in various other avian species. Additional investigation would be needed to determine if Cottage Grove eagles are being impacted by elevated mercury concentrations.

Table 1. Trace element concentrations in a bald eagle egg<sup>a</sup> collected near the Cottage Grove Reservoir, Oregon, 1992.

Trace Element	Contaminant Concentration	
	( $\mu\text{g/g}$ , dry wt.)	( $\mu\text{g/g}$ , wet wt.)
Al	22.6	5.96
As	<.457	<.120
B	2.01	.530
Ba	<.455	<.120
Be	<.0911	<.024
Cd	<.0911	<.024
Cr	<.455	<.120
Cu	4.19	1.10
Fe	39.2	10.3
Hg	2.9	.765
Mg	493	130
Mn	.634	.167
Mo	<.455	<.120
Ni	<.455	<.120
Pb	<.455	<.120
Se	2.04	.538
Sr	7.99	2.10
V	<.455	<.120
Zn	45.3	11.9

<sup>a</sup>Percent moisture = 73.6

# LITERATURE CITED

- Allen, S.M., and L.R. Curtis. 1991. An ecoregion approach to mercury dynamics in three Oregon reservoirs. Final Report to Oregon Department of Environmental Quality. June 10, 1991. 24 pp.
- Brooks, H. 1971. Quicksilver deposits in Oregon. Department of Geology and Mineral Industries, miscellaneous paper # 15.
- Eisler, R. 1987. Mercury hazards to fish, wildlife, and invertebrates: a synoptic review. U.S. Fish Wildl. Serv. Biol. Rep. 85(1.10). 90 pp.
- 1990. Boron hazards to fish, wildlife, and invertebrates: a synoptic review. U.S. Fish Wildl. Serv. Biol. Rep. 85(1.20). 32 pp.
- Fimreite, N. 1979. Accumulation and effects of mercury on birds. Pages 601-627 in J.O. Nriagu (ed.). The biogeochemistry of mercury in the environment. Elsevier/North-Holland Biomedical Press, New York.
- Garrett, M., R.G. Anthony, J.W. Watson, and K. McGarigal. 1988. Ecology of bald eagles on the Lower Columbia River. U.S. Army Corps of Engineers Contract No. DACW57-83-C-0100. 189 pp.
- Gasaway, W.C. and I.O. Buss. 1972. Zinc toxicity in the mallard duck. J. Wildl. Mgmt. 36(4): 1107-1117.
- Heinz, G.H. 1979. Methylmercury: reproductive and behavioral effects on three generations of mallard ducks. J. Wildl. Manage. 43(2):394-401.
- Heinz, G.H., D.J. Hoffman, and L.G. Gold. 1989. Impaired reproduction of mallards fed an organic form of selenium. J. Wildl. Manage. 53(2):418-428.
- Smith, G.J., and V.P. Anders. 1989. Toxic effects of boron on mallard reproduction. Environ. Toxic. Chem. 8:943-950.
- Spann, J.W., R.G. Heath, J.F. Kreitzer, and L.N. Locke. 1972. Ethyl mercury p-toluene sulfonanilide: lethal and reproductive effects on pheasants. Science 175:328-331.
- Wiemeyer, S.N., T.G. Lamont, C.M. Bunck, C.R. Sindelar, F.J. Gramlich, J.D. Fraser, and M.A. Byrd. 1984. Organochlorine pesticide, polychlorobiphenyl, and mercury residues in bald eagle eggs--1969 - 1979--and their relationships to shell thinning and reproduction. Arch. Environ. Contam. Toxicol. 13:529-549.

Mercury Dynamics and Methylmercury  
Accumulation by Fish in Three Oregon Reservoirs

March 10, 1994

Lawrence R. Curtis  
and  
Susan Allen-Gil

Department of Fisheries and Wildlife  
Oregon State University

A Final Report for  
Oregon Department of Environmental Quality  
Portland, Oregon

## TABLE OF CONTENTS

	<u>Page</u>
Abstract .....	1
Introduction .....	3
Site Description .....	5
Geology .....	5
Historical Mining Practices .....	8
Methods .....	10
Field Sampling .....	10
Chemical Analysis .....	12
Results .....	16
Mercury in Water .....	16
Mercury in Sediment .....	18
Mercury in Fish .....	20
Discussion .....	22
Mercury in Water .....	22
Mercury in Sediment .....	23
Mercury in Fish .....	26
Factors Affecting Bioaccumulation .....	27
Conclusions .....	31



## ABSTRACT

MERCURY DYNAMICS AND METHYLMERCURY ACCUMULATION BY FISH IN THREE OREGON RESERVOIRS. L.R. Curtis. Department of Fisheries and Wildlife, Oregon State University, Corvallis, OR 97331

Bioaccumulation of methylmercury by fish, sediment and water organic and inorganic mercury concentrations and potential inorganic mercury sources were assessed in three Oregon reservoirs (Ochoco, Owyhee, and Cottage Grove) which occur in distinct ecoregions. Ecoregions were distinguished by topography, geology, soil type and composition, and land use patterns. We examined pH, dissolved oxygen, hardness, and conductivity of the water; complexing agent levels, volatile solids and cinnabar content of sediment to quantitate differences in environmental conditions between study sites. Mercury concentrations in water were below detection (0.2 ppb) in most water samples from the 3 reservoirs. In samples above detection, mercury was probably predominantly in the particulate fraction, and thus not in true solution. Mercury in sediment occurred in the inorganic state; methylmercury was below detection (0.2 ppb) in 83% of all samples. Significant differences ( $p=0.05$ ) in sediment mercury concentrations were found between the reservoirs and between sampling dates within each reservoir. Despite the low mercury levels in water and sediment, mercury in fish over 4 years old tested above the FDA limit of 1.0 ppm. More than 90% of the mercury was organic mercury in all fish. Mercury in lateral muscle did not differ significantly between smallmouth bass in Owyhee Reservoir and largemouth bass in Cottage Grove

Reservoir. We conclude that although ecoregion parameters alone do not explain mercury dynamics, they may influence the methylation rate in areas of similar loading rates. Future work should consider multiple fish species and examine within ecoregion variability in lakes with different histories of mercury occurrence in their watersheds. (Supported by USEPA, grant S-000397/01).

## INTRODUCTION

Mercury is a ubiquitous metal, occurring in different concentrations in the soil, rocks, air and water throughout the world. Elevated concentrations of mercury in surface water can be derived from many sources, including natural processes and anthropogenic losses. Natural processes include volcanic and atmospheric deposition, degassing, and surface runoff and erosion of mercuric soils; anthropogenic sources include mercury mining and processing, processing of gold and silver ores, energy related activities, pesticide application, chloro-alkali operations, and smaller emissions from other industrial processes (Andren and Nriagu, 1979).

Methylmercury (MM) concentrations in some Oregon reservoir fish exceed the 1.0 ppm U.S. Food and Drug Administration limit for human consumption (DEQ, pers comm.: Worcester, 1979; Lowe et al., 1985). The goal of this project was to identify abiotic and biotic factors contributing to accumulation of MM in fish tissue in three Oregon reservoirs in different ecoregions. The specific objectives were to identify probable sources of mercury in the reservoir systems and parameters affecting MM dynamics and bioavailability, and to examine the extent to which ecoregion-level parameters influence MM dynamics in these reservoir systems. An ecoregion is defined as an area in which within-region variation is less than between-region variation (Gallant et al., 1989). While ecoregions can be distinguished based on almost any physiogeographical feature, a common suite of parameters is land surface form, potential natural vegetation, land use and soil characteristics

(Omernik, 1986). Limnological and biological features of aquatic ecosystems are largely determined by these broad-scale parameters.

Ecoregion phenomena directly and indirectly influence the dynamics of compounds in the environment. Mineral composition of soils and land use practices within a watershed have major impacts on mercury loading rates. Indirectly, the physical, chemical, and biological features of an aquatic system influence mercury dynamics (Hakanson, 1980; Akielaszek and Haines, 1981). Given these observations, an ecoregion approach to mercury dynamics and bioaccumulation seems to be an appropriate model. We have developed this model with the assumptions that mercury burdens in the reservoirs are largely derived from their watersheds and that mercury bioaccumulation by fish is generally related to the concentration of total bioavailable MM.

To investigate the validity of an ecoregion approach, we examined both qualitative and quantitative ecoregion parameters as they relate to mercury concentrations in water, sediment and fish. The qualitative parameters are those which define ecoregions (land surface form, potential natural vegetation, land use and soils). The quantitative parameters examined were pH, conductivity, hardness and alkalinity of the water column, and the clay and organic matter content of the sediment. In addition, characteristics of the specific drainage basins were compared; these include basin area, relief, annual precipitation and land uses. Limnological features of each reservoir were also considered. The interactions between mercury concentrations in sediment and MM in fish were also investigated.

## SITE DESCRIPTION

Cottage Grove, Owyhee and Ochoco reservoirs are located in three distinct ecoregions (Fig. 1). Drainage basin and limnological characteristics of the study reservoirs were compared to other reservoirs within the ecoregions (Johnson et al., 1985) and were considered representative of reservoirs of similar size within the ecoregions. The specific characteristics of each drainage basin are summarized in Table 1.

### *Geology*

The geology of all three areas is characterized by a combination of sedimentary and volcanic formations. The area surrounding Cottage Grove reservoir is older Cenozoic marine and estuarine sedimentary deposits with minor amounts of volcanic elements; the Ochoco area predominantly contains metasedimentary and metavolcanic formations; and the Owyhee region is a mixture of deposits of sedimentary and volcanic origin (Baldwin, 1976).

Geothermal activity is high in the Owyhee area, moderate in the Ochoco area, and virtually undetectable in the Cottage Grove basin. Therefore, the potential contribution of geothermal venting to mercury loading is likely to vary accordingly among the three reservoirs based on the differences in geothermal activity.

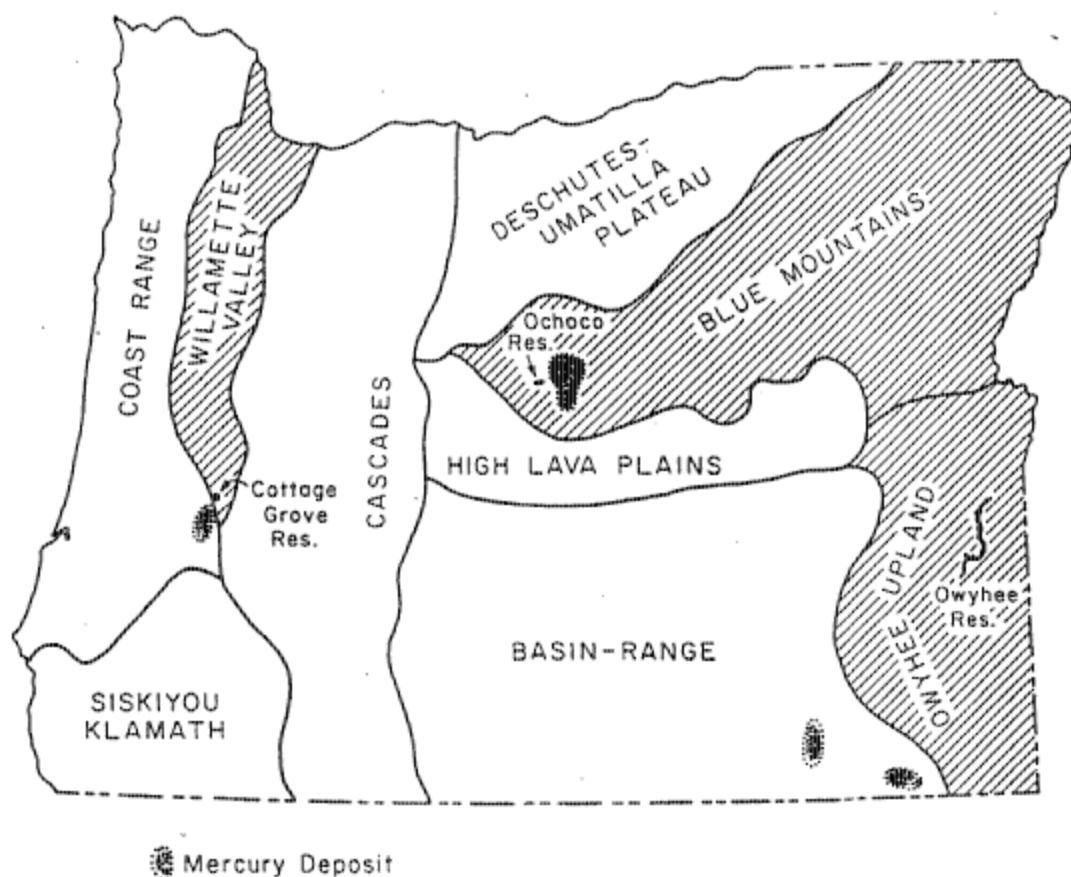


Figure 1 - Location of study areas and mercury deposits within the different ecoregions. Compiled from : Baldwin, 1976.

---

Table 1 - Drainage basin characteristics

---

	Cottage Grove	Ochoco	Owyhee
Drainage basin area (km <sup>2</sup> )	257	288	11,160
Annual Precipitation (cm)	122-157	43-64	25-64
Land use (% total area)			
Forest	96.5	73.8	0.5
Range	1.0	21.3	93.2
Surface area (ha)	461	388	5625
Average depth (m)	9	8.4	24.6
Shoal area (%)	17	29	4
Retention time	2 mo	5 mo	1.7 yr
pH	7.7	8.4	8.4
Conductivity (μmhos/cm)	63	197	160
Sulphide (SO <sub>4</sub> mg/l)	1.2	3.9	11.7
Dissolved oxygen (mg/l)	7.2	-	8.3
Trophic status	mesotrophic	eutrophic	eutrophic

---

Compiled from: Johnson et al., 1985.

While mercury most frequently occurs as deposits in rock fractures and veins, it may also be found in low concentrations in other geologic formations. In the Owyhee River area, mercury is commonly found as an anomaly, present in 12 of 23 random outcrop rock-chip samples where concentrations averaged 0.3 ppm (Gray et al., 1983).

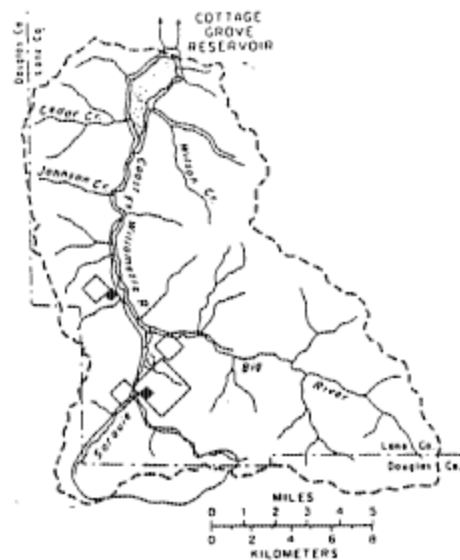
#### *Historical Mining Practices*

Extensive mercury mining was performed in all three areas (Fig. 2). As is true for all mercury mines in the state, production occurred primarily from the late 1800s until 1950, with peak production coinciding with war years. Since 1950 the increased Korean mercury production and the surplus from war years have saturated the mercury market and decreased prices such that Oregon mines have ceased production (Brooks, 1971).

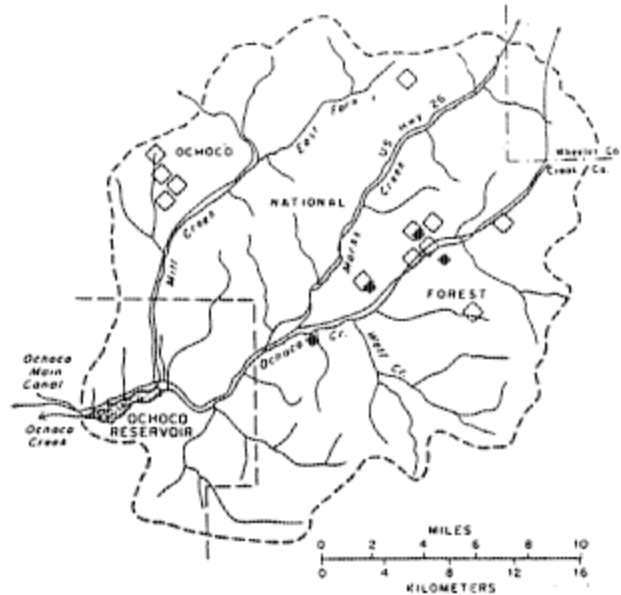
The second largest mercury mine in Oregon, Black Butte Mine, is located 2 miles south of Cottage Grove Reservoir, within the drainage basin (Brooks, 1971). Active intermittently from 1882 to 1966, this mine produced 18,156 flasks (Brooks, 1971). The ore in this area has been low-grade, approximately 0.175% by weight (Brooks, 1971). Although there are no mercury mines in Oregon in the Owyhee basin, Brooks (1971) reported that one of the leading national mercury producing mines in the country is in the Nevada section of the drainage basin. This could not be confirmed by maps of mercury occurrences in Nevada (see Lawrence and Wilson, 1962).



COTTAGE GROVE RESERVOIR DRAINAGE BASIN



OCHOCO RESERVOIR DRAINAGE BASIN



OWYHEE RESERVOIR DRAINAGE BASIN

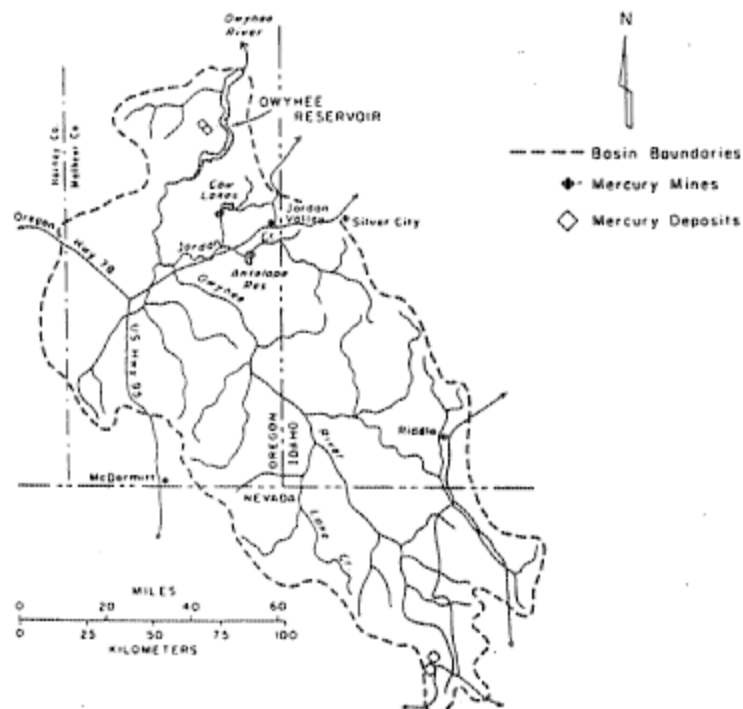


Figure 2 - Mercury mines and deposits in the three study areas.  
Compiled from: Johnson et al., 1985; Lawrence and Wilson, 1962; Ferns and Huber, 1954; and State Water Resources Board, 1960a, 1960b.

Mercury in the Owyhee basin may be derived from its use in gold and silver extraction. Extensive gold and silver mining took place in the Jordan Creek region of Owyhee basin between 1860 and 1920 (Hill, 1973). Mercury used in the amalgamation process can be lost to the environment through inefficient recovery after distillation. It has been estimated that 76 pounds of mercury were lost daily during mining years in Idaho (Hill, 1973).

Mercury production in Ochoco basin has been from several small mines, due to the discontinuous faulting in this region (Brooks, 1971). Prior to 1943, the four mines in the Ochoco basin (Byram-Oscar, Staley, Champion, and Taylor Ranch) collectively produced 857 flasks (Brooks, 1971).

## METHODS

### *Field Sampling*

Two or three sites were sampled for water and sediment at each reservoir, depending on the reservoir size and water level (Fig. 3). Water and sediment samples were collected in September 1989, and June and September, 1990, and sediment only in September 1992. Water samples were collected 0.5 m from the surface in 1 l polyethylene bottles, pre-washed with  $\text{HNO}_3$ . Core sediment samples were obtained using a 1" d PVC pipe. The uppermost 5 cm of the core was transferred into a pyrex plate, mixed and placed in 250 ml polyethylene bottles, pre-washed with  $\text{HNO}_3$ . All samples were frozen immediately, and stored frozen until subsequent analysis. The following parameters were also measured at each site: water pH (Orion Model 250 pH meter), conductivity, and dissolved oxygen (Winkler method).

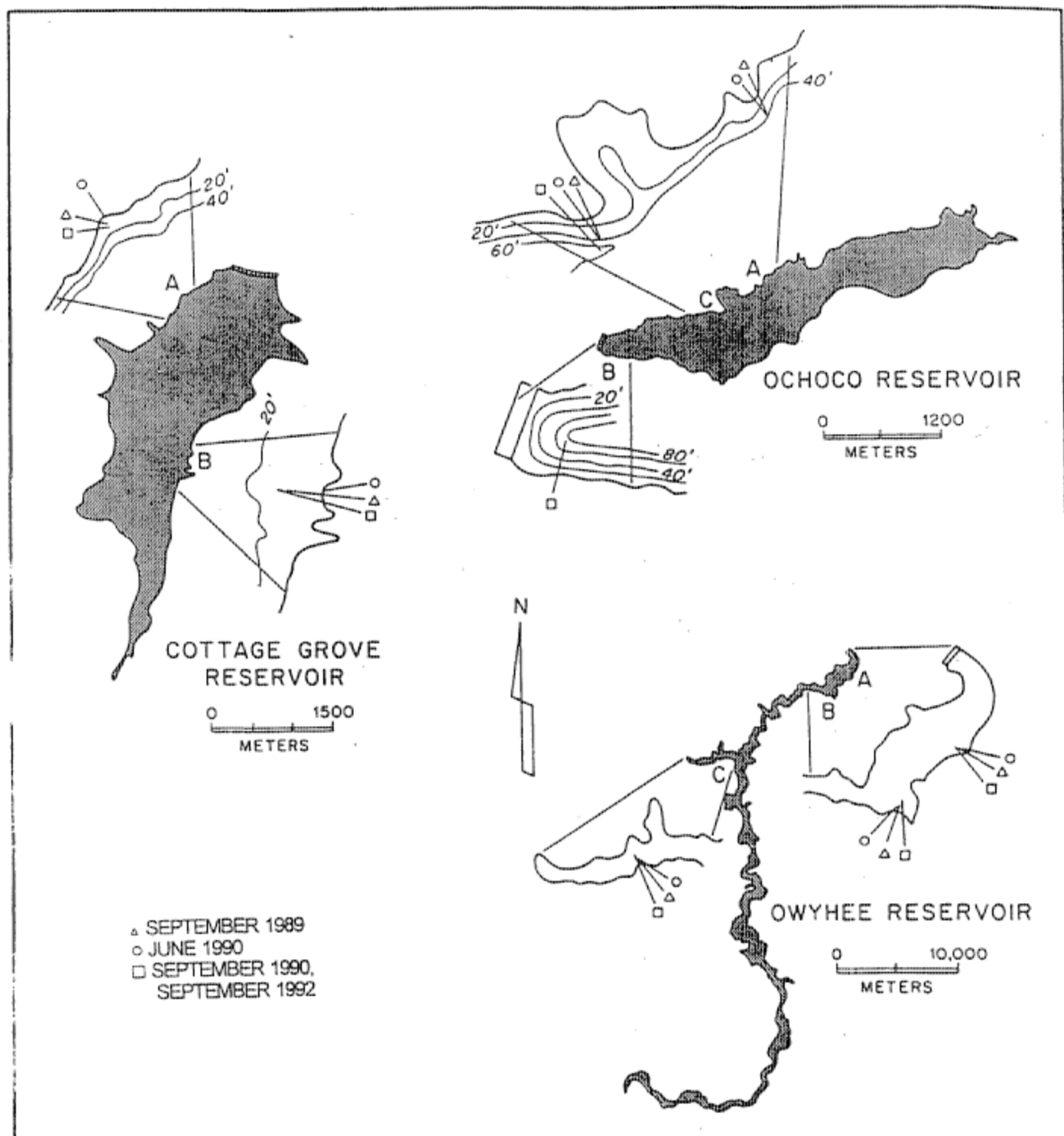


Figure 3. Locations of sites from which sediment samples were collected for mercury and other analyses.

Fish were collected by electroshocking in cooperation with the Oregon Department of Environmental Quality in September and October, 1990, or by angling in September and October of 1992. Fish were filleted in the field, and fillets (with skin) were immediately frozen and stored frozen.

#### *Chemical Analyses*

Total mercury in water: Total mercury concentration in water was determined using cold vapor atomic absorption according to the procedure in Standard Methods for the Examination of Water and Wastewater. A 100-ml volume of each sample was transferred to a 250 ml Erlenmeyer flask, to which 5 ml concentrated  $\text{H}_2\text{SO}_4$  and 2.5 ml concentrated  $\text{HNO}_3$ , and 15 ml of 5%  $\text{KMnO}_4$  were added. After fifteen minutes 8 ml of 5%  $\text{K}_2\text{S}_2\text{O}_8$  was added to each sample, and the flasks were heated in a 95°C water bath for 2 hours. The samples were cooled to room temperature, transferred to 250 ml gas-scrubbing reaction flasks, treated with 8 ml of 24% NaCl-hydroxylamine sulfate to reduce excess  $\text{KMnO}_4$ . Immediately following the addition of 5 ml 10%  $\text{SnCl}_2$  in dilute HCl, the flasks were supplied with flow-through nitrogen gas (2 l/min). Mercury vapor was passed through a Coleman Model 50 mercury analyzer (Perkin-Elmer Co., Maywood, IL), connected to a Microscribe 4500 recorder set at 5 mV. (The Recorder Company, San Marcos, TX). Peak area was calculated as height X width at half-height. Unknowns were determined using a standard curve, based on  $\text{HgCl}_2$  in  $\text{HNO}_3$  (0.25 - 1.0  $\mu\text{g/l}$ ). More than 75% of water samples were analyzed in duplicate.

Total mercury in sediment: Sediment samples were analyzed following the methods outlined in Buhler et al. (1984). After initial preparation and hot-acid digestion, this procedure is very similar to the analysis of water. Thoroughly mixed sediment samples (3-5 g) were dried to a constant weight in a 60°C oven (approx. 96 h). Samples were crushed using a ceramic mortar and pestle, and passed through a 1 mm mesh screen. Particles that did not pass through the screen were pulverized a second time, and screened. Any remaining material was discarded. Subsamples (0.5 - 1.5 g) were weighed and transferred to glass 250 ml BOD bottles, to which 5 ml deionized H<sub>2</sub>O and 5 ml aqua regia (3 vol. conc. HCl: 1 vol. conc HNO<sub>3</sub>) were added. Samples were placed in a 95°C water bath. After two minutes, 50 ml deionized H<sub>2</sub>O and 50 ml 5% KMnO<sub>4</sub> were added to each sample. Samples were digested in the water bath for 30 min, and cooled to room temperature. Because of the strong HCl fumes released during the heating, the water bath was placed in a hood. Fifteen min before analysis, samples were treated with 50 ml deionized H<sub>2</sub>O and 8 ml 24% NaCl-hydroxylamine and placed in a hood to allow the evolved oxygen gas to escape. Samples were transferred to the 250 ml reaction flasks used for water analysis, and analyzed in the same manner. Gas flow was set at 1.5 l/min. Recorder sensitivity was set at 20 mV.

Sediment mercury concentrations were determined based on a standard curve of HgCl<sub>2</sub> in HNO<sub>3</sub> (0.05 - 0.90 µg). The accuracy of the standard curve and recovery efficiency was tested using reference material from the National Institute of Standards and Technology. Most samples (>75%) were analyzed in duplicate.

Methylmercury in sediment: Sediment samples were analyzed for MM by gas chromatography using a modification of the multi-stage extraction and concentration procedure developed by Uthe et al. (1972). The modified technique for sediment is outlined by Furutani and Rudd (1980).

Two aliquots of wet sediment (approximately 1 g) were weighed to the nearest 0.001 g. One set of samples was dried to a constant weight in a 60°C oven to determine the water content so that measurements could be standardized on a dry weight basis.

The second set of samples were transferred to 50 ml centrifuge vials. Two ml of 0.5M  $\text{CuSO}_4$  and 10 ml of 3M NaBr in 22% conc.  $\text{H}_2\text{SO}_4$  were added to the samples. After shaking vigorously for two minutes, samples were centrifuged for 5 min and transferred to 60 ml glass separatory funnels. Twenty ml of toluene were added and samples were shaken for three min. Following removal of the aqueous phase, the toluene phase was treated with 1 g anhydrous  $\text{NaSO}_4$  and decanted into a 50 ml erlenmeyer flask and further dried with 0.5 g anhydrous  $\text{NaSO}_4$ . A 10 ml sample of the extract was transferred to a clean separatory funnel, and 5 ml of 0.0025 M  $\text{Na}_2\text{S}_2\text{O}_3$  in 20% ethanol was added. After shaking and standing, 3 ml of the lower aqueous phase was collected into a calibrated, glass-stoppered centrifuge tube, to which 1 ml of 3M KI and 1 ml hexane were added.

Subsamples (6  $\mu\text{l}$ ) were injected into a Hewlett-Packard 5700 gas chromatograph equipped with a  $^{63}\text{Ni}$  electron capture detector. The column was packed with 7% Carbowax 20M on Chromosorb W, acid-washed DMCS-treated.

Argon/methane gas was supplied at 50 ml/min, and GC attenuation was set at 2 mV. The operating temperatures for the column and detector were 175°C and 200°C respectively. The Microscribe recorder was set at 1 mV, and 1 cm/min. Peak area was calculated by height times width at half-height. Unknowns were compared to standards prepared from methylmercuric chloride in hexane (6-20 ng).

Total and organic mercury in fish: Mercury concentration in fish muscle was determined using a hot-base digestion followed by cold vapor atomic absorption. Fillet samples (1-2 g) were placed in screw-top test tubes, to which 2 ml 10N NaOH was added. Samples were then heated for 30 min in a heat block (95°C) and cooled to room temperature. One percent NaCl (8 ml) was added to each sample.

Total mercury was determined by placing 1 ml subsample in a reaction flask along with 3 ml 1% NaCl, 1 ml 1% cysteine, 4 drops octanol, and 1 ml 50% SnCl<sub>2</sub> and 10% CdCl<sub>2</sub> in 4 N HCl. Inorganic mercury was determined by adding 1 ml of 50% SnCl<sub>2</sub> in place of the SnCl<sub>2</sub>- CdCl<sub>2</sub> solution. The flask opening was then covered with a septum, through which 4 ml 10N NaOH was injected by syringe. After thirty seconds, N gas was supplied at 1.5 l/min. The recorder was set at 5mV. Standards were prepared as Hg in HNO<sub>3</sub> (10-100 µg/ml), prepared from a commercially available standard (1000 ppm, Johnson & Mathey, Seabrook, NH). Organic mercury was calculated as the difference of total and inorganic mercury.

#### Additional analyses

1. All water samples were analyzed in the lab for hardness, following the EDTA titrimetric method in Standard Methods for the Examination of Water and Wastewater (1985).
2. All sediment samples were analyzed for percent volatile solids by dried sediments in a 600°C muffle furnace for 4 hours, after Buhler et al. (1984).
3. Sediment physical characteristics were performed by hydrometer method by the OSU Soil Physics Laboratory.
4. Sediment samples from 9/90 were sent to the U.S. Bureau of Mines (Albany, OR) for determination of cinnabar content.

#### RESULTS

##### *Mercury in water*

Twelve water samples and 24-32 sediment samples were analyzed from each reservoir. Results of the limnological and sediment analyses are summarized in Table 2. Mercury was detected in 25%, 6%, and 15% of Cottage Grove, Ochoco, and Owyhee unfiltered water samples respectively. Differences in mean total mercury concentration in water among reservoirs were not statistically significant ( $p=0.05$ ).



Table 2. Water chemistry and sediment parameters

Parameter	Cottage Grove	Ochoco	Owyhee
<u>Water</u>	(n=12)	(n=12)	(n=12)
pH	7.70 <sup>a</sup>	8.53 <sup>b</sup>	8.44 <sup>b</sup>
Dissolved oxygen(mg/l)	8.20 <sup>a</sup>	7.8 <sup>a</sup>	7.24 <sup>b</sup>
Conductivity ( $\mu$ mhos)	56 <sup>a</sup>	260 <sup>b</sup>	254 <sup>b</sup>
Hardness (mg CaCO <sub>3</sub> /l)	39 <sup>a</sup>	101 <sup>b</sup>	72 <sup>c</sup>
Hg ( $\mu$ g/l)	0.78 $\pm$ 0.40	0.17 $\pm$ 0.10	0.37 $\pm$ 0.30
<u>Sediment</u>	(n=24)	(n=24)	(n=32)
Soil texture	*sand-sandy loam	*clay-clay loam	*sandy loam - clay loam
Clay content (%)	10 $\pm$ 7	34 $\pm$ 5	12 $\pm$ 9
Carbon content (%)	7.11 $\pm$ 0.63 <sup>a</sup>	5.71 $\pm$ 0.4 <sup>ab</sup>	5.16 $\pm$ 0.42 <sup>b</sup>
Total Hg ( $\mu$ g/g)	0.84 $\pm$ 0.2	1.0 $\pm$ 0.2	1.4 $\pm$ 0.3
Fine grain Hg <sup>**</sup> ( $\mu$ g/g)	0.28 $\pm$ 0.07 <sup>a</sup>	0.14 $\pm$ 0.02 <sup>b</sup>	0.11 $\pm$ 0.01 <sup>b</sup>

\* Soil texture was determined by hydrometer analysis on the >2 mm fraction of 600 g samples. Cottage Grove and Ochoco reservoirs(n=2), Owyhee Reservoir(n=3).

\*\* Fine grain sediment was defined as the fraction <1 mm in diameter.

Values are presented are the mean or mean  $\pm$  standard error.

All sediment values are presented on a dry weight basis.

Superscripts designate significant differences at p=0.05.

#### *Mercury in sediment*

The highest fine-grain sediment mercury level was associated with Cottage Grove Reservoir ( $0.28 \pm 0.07$ ), which was significantly different from Ochoco ( $0.14 \pm 0.02$ ) and Owyhee ( $0.11 \pm 0.01$ ) reservoirs values (Fig. 4). This finding, however, is largely attributable to the high mercury levels found at both sites in the September 1989 samples. To confirm the observation that 1989 levels were higher than the other two dates, particularly for Cottage Grove Reservoir, 1989 samples were reanalyzed in a mixed batch with samples from later dates; the results were consistent with previous analyses. Methylmercury was detected in sediment from all three reservoirs, but not in every sample (4 of 15 samples analyzed). The average percent of total Hg was 1.5%; the maximum (4.20%) was from Cottage Grove reservoir.

Sediment mercury concentrations did not significantly correlate with organic content (% volatile solids, PVS) for any of the reservoirs. Organic content (PVS) exhibited a seasonal pattern, modified by water level fluctuations. Higher organic content was observed in the fall samples than in the spring samples for Cottage Grove Reservoir. In Ochoco, this pattern was superseded by water level fluctuations. The water level was unchanged between fall 1989 and spring 1990, and the sediment organic content was similar for both dates. However, a 34-ft decline in water level between the June 1990 and September 1990 sampling dates resulted in a higher percent organic content associated with the low water level (7.4% vs. 5.1%). Organic matter in Owyhee Reservoir sediment was consistently between 4.6% and 5.8%.

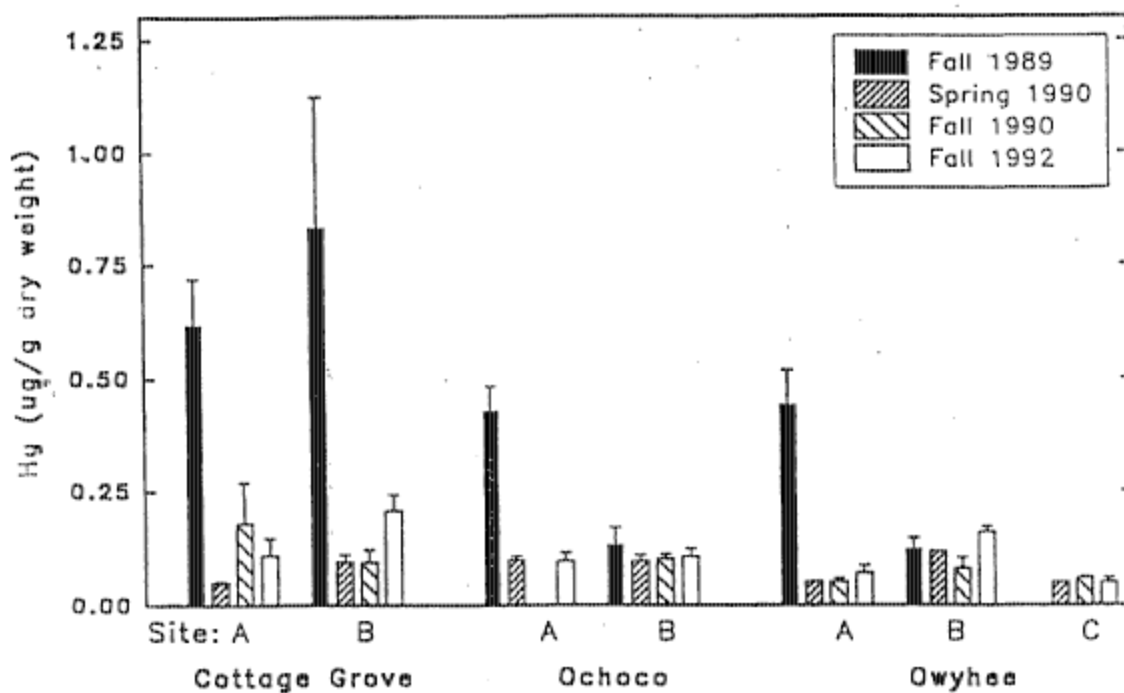


Figure 4. Concentration of mercury in fine-grain sediment by sampling date for Cottage Grove, Ochoco and Owyhee reservoirs.

Mercury values were more influenced by sampling date than organic content, suggesting the importance of parameters other than organic content. Sediment mercury levels in Owyhee and Ochoco reservoirs were consistent for all four sampling dates. In Cottage Grove Reservoir, the fall 1989 sediment samples had statistically higher mercury levels than fall 1990 samples, despite no change in organic content. This information, combined with the observation that levels of Hg in bulk, unfiltered sediment was an order of magnitude higher than in fine grain sediment, suggests that most Hg is not associated with the organic matter in the sediment, but is probably bound to sulphur as cinnabar.

#### *Mercury in fish*

Smallmouth bass (*Micropterus dolomieu*) from Owyhee, largemouth bass (*Micropterus salmoides*) from Cottage Grove, and rainbow trout (*Salmo gairdneri*) from Ochoco were analyzed for total and organic mercury content in lateral fillets. Five year old Owyhee Reservoir Cottage Grove Reservoir bass exceeded the United States FDA limit for human consumption (1.0 µg/g wet weight) for commercially-caught fish (Fig. 5). Mercury concentrations in fish from Owyhee and Cottage Grove reservoirs were not significantly different from each other, and both were higher than rainbow trout from Ochoco Reservoir. Organic mercury comprised >90% of the total mercury in all cases in which both analyses were run on the same fish (n=11).

MM concentration in fish muscle increased with age for bass from Owyhee and Cottage Grove reservoirs (Fig. 5). There was little or no bioaccumulation of mercury between 2 and 3 years, followed by a linear increase for

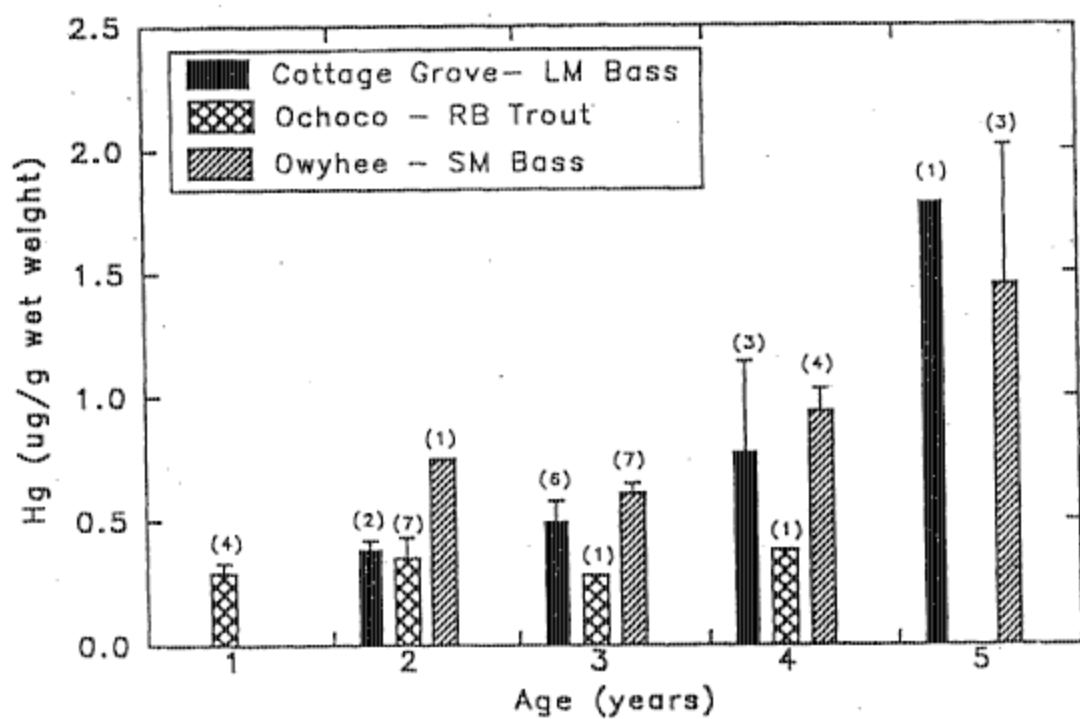


Figure 5. Mercury concentration in fish tissue by fish age for the three species from the three reservoirs. Numbers in parentheses represent number of fish sampled.

largemouth from Cottage Grove and smallmouth bass from Owyhee reservoir. All rainbow trout from Ochoco reservoir contained less than 0.5 ppm MM and there was no evidence of MM accumulation with age (Fig. 5).

## DISCUSSION

### *Mercury in water*

Surface water concentrations of mercury in areas with mercury deposits, such as the Pacific Northwest, may be considerably higher than areas without natural deposits. In lakes where the only significant source of Hg is atmospheric transport, mean surface water Hg concentrations are 0.07  $\mu\text{g/l}$  (Sorensen et al. 1990). Sampling of the Columbia and Willamette rivers in 1970-71 yielded mercury concentrations between 1-35  $\mu\text{g/l}$  for filtered waters (Jenne 1973), indicating the presence of mercury sources. Our results indicate an elevated frequency (6-26% of unfiltered samples) and magnitude (0.12-1.0  $\mu\text{g/l}$  for samples above the detection limit) of surface water contamination above background levels derived from atmospheric deposition. Thus, atmospheric inputs alone probably do not account for the mercury concentrations in Cottage Grove, Ochoco and Owyhee surface waters. The large range of concentration values (0.12 - 1.0  $\mu\text{g/l}$ ) suggests that the highest concentrations of mercury are probably found in the particulate fraction. This is consistent with results in which mercury in unfiltered water was <0.04  $\mu\text{g/l}$ , while suspended matter in the stream after a storm event had a maximum value of 0.61 mg/kg Hg (Lacerda et al. 1991). This does not, however, indicate that bioavailability to reservoir fish is enhanced,

since mercury in the particulate fraction may not be as readily absorbed in biota as dissolved inorganic mercury or MM (Stokes and Wren 1987).

#### *Mercury in sediment*

Mercury was routinely detected in sediment from all three reservoirs. Mean mercury concentrations for all three lakes were higher than those reported for numerous other lakes in the Northwest and elsewhere (as reviewed by Phillips et al. 1987). Differences in sediment mercury concentrations are most likely due to variations in loading rates and patterns from the drainage basins as atmospheric transport alone do not account for the observed mercury levels. Factors that affect loading rates include the quantity of available mercury as natural deposits or mining waste, and sediment transport rates. Sediment transport rates are, in turn, affected by geological, climatological and physiographical forces. In this study the quantity of available mercury in the drainage basin seems to be more influential than sediment transport rates. Ongoing research on mercury concentrations in stream sediments flowing into Cottage Grove Reservoir is expected to provide additional information on sediment transport of mercury.

Drainage basin area was not a significant determinant in sediment mercury concentrations. This is evidenced by the fact that despite the comparative enormity of the Owyhee drainage basin, it did not have elevated mercury loads relative to the other two systems. Based on overall relief of the basins, we would expect sediment transport rates to be highest in the Owyhee area because of the steep relief, relatively barren vegetation, and erosional force of extensive snowmelt. The amount and cycle of annual

precipitation and flooding events also affect sediment transport (Lacerda et al. 1990). Precipitation in the Owyhee area is primarily snow (Johnson et al. 1985). Sparse vegetation and the timing of the snow melt produce conditions favoring high surface runoff and sediment transport. This, however, may be offset by differences in the total amount of precipitation. The high rainfall in the Willamette Valley ecoregion may exert a greater influence on relative loading rates. The difference between sediment mercury concentrations in Cottage Grove Reservoir between Fall samplings in 1989, 1990 and 1992 may relate to timing of storm events. Local flooding, resulting in a surge of mercury and other materials into reservoirs, may enhance bioaccumulation. Phillips et al. (1987) found that MM concentrations in northern pike were significantly higher in a year following a severe flood compared to previous or succeeding years (Phillips et al. 1987). One explanation is higher methylation rates in flooded shorelines and nearshore sediments (Ramlal et al. 1986, Johnston et al. 1991).

Land use patterns may also play a role in determining the behavior of mercury in reservoir systems. Plant biomass has been reported to be positively correlated with mercury concentration in areas where mercury is predominantly of atmospheric origin (Sorensen et al. 1990). Land use patterns also influence the rate of transport to surface waters and movement within reservoirs. Unfortunately, the relative enhancement of erosional forces from forest clearing, agricultural irrigation, and range practices are not known for these areas. However, based on slope and vegetational cover we would expect erosion to be highest in Owyhee Reservoir.



Thus, our observation that cinnabar in sediment was highest in Owyhee Reservoir is consistent with expectations based on sediment transport rates. We also suspect that the available reserves within the three reservoir systems are very different, and that this also influences sediment mercury concentrations.

Mercury burdens in all three reservoirs are most likely derived from natural mercury deposits exacerbated by past mercury, gold and silver mining in the drainage basins. The relative size and mining effort of the Black Butte Mine in the headwaters of Cottage Grove Reservoir may explain the elevated sediment mercury concentrations. This mine produced 25 times more mercury than all the mines in the Ochoco District combined (Brooks 1971).

The diffuse distribution of mercury in Owyhee Reservoir is consistent with a distant mercury source. Mercury in the Owyhee basin may be derived from its use in gold and silver extraction. Mercury levels in gold mining tailing piles can be as high as 5 mg/kg (Lacerda et al. 1990). Extensive gold and silver mining took place in the Jordan Creek region of Owyhee basin between 1860 and 1920; it has been estimated that 76 pounds of mercury were lost daily during mining years in Idaho (Hill 1973). Geothermal releases of mercury may also be a significant and persistent source of mercury (Eisler 1987, Nriagu 1979). This may be important in Owyhee Reservoir given the high level of geothermal activity in southeastern Oregon.

### *Mercury in fish*

Concentrations of MM in fish muscle were well above background levels, as calculated by Håkanson et al. (1990). The range of MM in Cottage Grove Reservoir largemouth bass (0.22-1.79  $\mu\text{g/g}$ ) overlaps the range reported by Worcester (1979), suggesting that mercury contamination in Cottage Grove Reservoir has neither increased nor dissipated. Of five species examined for mercury concentrations in past efforts, the highest values were observed in largemouth bass (Worcester 1979). Other species analyzed were chinook salmon, cutthroat, rainbow trout and brown bullhead.

Smallmouth bass from Owyhee Reservoir from this sampling effort also have MM burdens within the range of past investigations. According to the Oregon Department of Environmental Quality, MM concentrations in smallmouth bass sampled in 1987-89 ranged between 0.66 and 1.68  $\mu\text{g/g}$  (based on edible flesh); these levels were higher than those of carp and black crappie. Largemouth bass and channel catfish had similar MM concentrations as smallmouth bass.

MM concentrations in rainbow trout from Ochoco Reservoir were significantly lower than fish from Cottage Grove or Owyhee reservoirs. Since the sediment mercury levels were not significantly different from the other two reservoirs, this may reflect species differences in uptake or bioaccumulation. This is consistent with the findings of Worcester (1979) within Cottage Grove Reservoir.

Several studies have examined the importance of drainage basin size. McMurty et al. (1989) observed a positive correlation between MM in smallmouth bass tissue and watershed and lake area. Suns and Hitchin (1990) also found a positive correlation between drainage basin area/lake volume and MM in yellow perch, suggesting that watershed inputs are important. However, Weiner et al. (1990b) report that drainage basin area/lake volume was not a significant variable to explain MM in walleyes. Thus, it appears that this relationship is valid only where mercury inputs are approximately equal, as in the case of atmospherically-derived inputs.

#### *Factors affecting bioaccumulation*

Sedimentation rates and mercury inputs alone may not explain observed levels in fish. Microbial mercury methylation in sediment and water is considered to be an important determinant in the rate of bioaccumulation by fish (for example, Håkanson 1980, Lindberg et al. 1987, Stokes and Wren 1987). Conditions that influence methylation include pH, redox potential, organic substrate and temperature (Beijer and Jernelev 1979). In addition, we suspect that mercury in fine-grain sediment is more easily methylated by microorganisms than mercury bound to sulphur as cinnabar. If this is true, then Cottage Grove Reservoir has the highest levels of potentially-bioavailable mercury.

Sediment organic content was not significantly different in the three reservoirs, suggesting that the importance of this variable was overshadowed by other factors such as mercury inputs, fish species or age. Klein (1973) suggests that organic material may control mercury distribution.

Corresponding to greater microbial density and available energy, high organic content stimulates MM production (Jackson and Woychuk 1980).

Several water chemistry parameters (conductivity, turbidity, total dissolved solids, pH, temperature and trophic state (reviewed by Lindberg et al. 1987) also influence the availability of MM in surface waters and uptake by fish. Favorable conditions for MM accumulation are reviewed in Table 3. Furthermore, the relationship between water chemistry parameters and bioaccumulation by fish may not be the same for all fish species (McMurty et al. 1989, Wren et al. 1991).

The conductivity in Cottage Grove Reservoir was significantly lower than that of Ochoco or Owyhee, which may exacerbate the mercury problem in this reservoir. For 13 Canadian lakes, conductivity explained 54% of the total variation in MM concentrations in the crayfish (Allard and Stokes 1989). A negative correlation between conductivity and MM in fish muscle has also been reported by Wren et al. (1991).

The effect of pH on accumulation of MM in fish has been extensively studied (Jernelov and Asell 1975, Schlinder et al 1980, Beijer and Jernelov 1979, Phillips et al. 1987, Stokes and Wren 1987, Weiner et al. 1990b, Wren et al. 1991), although the driving mechanisms are still debated (Richman et al. 1988). Most studies in acidic lakes report an inverse correlation between pH and MM in fish (Sorensen et al. 1990, Weiner et al. 1990a, Grieb et al. 1990, Suns and Hitchin 1990, Håkanson et al. 1990). However, this relationship is less consistent in alkaline lakes. Rates of MM uptake in

Table-3. Conditions favorable to mercury bioaccumulation

---

Hydrologic

slow flow

frequent flooding

recent impoundment

Water chemistry

low conductivity

high dissolved organics

pH <6.0 or >8.5

high temperature

Sediment characteristics

mildly oxidizing environment

low clay content

high organic content

low level of complexing agents

Life history factors

large size

long life span

high trophic position

---

fish were positively correlated with pH in reservoirs of pH between 8 and 9 in the Upper Missouri River basin (Phillips et al. 1987). The pH of all three Oregon reservoirs is between 7.7 and 8.6, and thus fall into a range in which the relationship to mercury/MM dynamics is unclear. The significantly lower pH at Cottage Grove may enhance MM availability and bioaccumulation.

All three reservoirs are classified as mesotrophic or eutrophic. The effect of limnological trophic state has also not yet been clearly established (Håkanson 1980, Lindberg et al. 1987, Richman et al. 1988). Oligotrophic lakes are thought to generally have lower methylation and uptake rates because of the lower density of methylating bacteria, lower energy availability for metabolic activity and oxygen concentrations above the optimum for methylation (Phillips et al. 1987). Conversely, methylation rates should be higher under eutrophic conditions; however, Akielaszek and Haines (1981) argue that methylation rates are higher in oligotrophic conditions based on greater mercury availability because there is less organic matter for mercury complexation.

Food web structure and the position of a given species in the foodweb influences the bioaccumulation of MM because diet can be a significant exposure route. The percent of accumulated mercury from food varies with species. MM is the only metal for which bioaccumulation is widely documented (Lindberg et al. 1987). Phillips et al. (1980) showed that piscivorous fish accumulate MM faster than those eating plankton. All three species examined here are at equivalent levels in the trophic structure; their diets consist

of varying quantities of insects, crustaceans and fish (Scott and Crossman 1973). The lower levels of MM in rainbow trout in Ochoco Reservoir by age may reflect a relatively greater contribution of insects and crustaceans to rainbow trout diet than bass, as well as differences in mercury/MM dynamics between reservoirs.

### CONCLUSIONS

If mercury inputs were similar to all three reservoirs and water chemistry parameters were the most important determinant, then we would expect MM bioaccumulation in fish to be highest in Cottage Grove Reservoir. The pH, conductivity and hardness of this reservoir are significantly lower than the other two reservoirs. All three of these factors have been reported to favor MM bioaccumulation in fish (Lindberg et al. 1987, McMurty et al. 1989, Allard and Stokes 1989). As the MM concentrations in fish from Cottage Grove Reservoir were not significantly higher than Owyhee reservoir based on fish age, we believe that mercury deposits in the drainage basins and sediment transport rates are very different among the reservoirs, and that these are the dominant influences on mercury/MM dynamics.

It is evident that MM bioaccumulation in fish represents a management problem in Cottage Grove and Owyhee reservoirs. Despite the general pattern of low mercury concentrations in water and sediment, older fish have MM burdens in muscle tissue exceeding the U.S. Food and Drug Administration limit for human consumption. This supports the importance of methylation and bioaccumulation as critical determinants of MM levels in fish. The importance of ecoregion parameters in determining mercury/MM dynamics in

reservoir systems with natural and mined deposits and MM accumulation in fish is overshadowed by differences in mercury deposits and mining activities. We intend to investigate the sources and transport of mercury in Cottage Grove Reservoir in a continuation of this study.

*Acknowledgement-* We would like to acknowledge the assistance of the Oregon Department of Environmental Quality (ODEQ), Oregon Department of Fish and Game (ODFW), and the U.S. Bureau of Mines. The ODEQ staff, particularly Doug Drake and Eugene Foster, assisted in fish sampling. Kim Daily and Lisa Borgeson of ODFW performed scale analyses to determine fish ages. The U.S. Department of the Interior, Bureau of Mines analyzed sediment samples for cinnabar content. (Supported by USEPA, grant S-000397/01).



## REFERENCES

- Akielaszek JJ, Haines TA (1981) Mercury in the muscle tissue of fish from three northern Maine lakes. *Bull. Environ. Contam. Toxicol.* 27:201-208
- Allard M, Stokes PM (1989) Mercury in crayfish species from thirteen Ontario lakes in relation to water chemistry and smallmouth bass (*Micropterus dolomieu*) mercury. *Can. J. Fish. Aquat. Sci.* 46:1040-1046
- American Public Health Association, American Water Works Association, and Water Pollution Control Federation (1985) Standard methods for the examination of water and wastewater, 16th edition. (Eds: Greenberg, A.E., R.R. Trussell, L.S. Clesceri), American Public Health, Washington, DC
- Baldwin EM (1976) Geology of Oregon, Edwards Brothers Inc., Ann Arbor, Michigan
- Beijer K, Jernelov A (1979) Methylation of mercury in aquatic environments. Elsevier Press, Holland, p 203
- Bodaly, RA, Hecky RE, Fudge RJP (1984) Increases in fish mercury levels in lakes flooded by the Churchill River Diversion, Northern Manitoba. *Can. J. Fish. Aquat. Sci.* 41:682-691
- Brooks H (1971) Quicksilver deposits in Oregon. Department of Geology and Mineral Industries, miscellaneous paper #15
- Buhler D, Reed R, Caldwell R (1984) As assessment of projected mercury levels in the proposed Galloway Dam project on the Weisser River, Idaho. Report submitted to the U.S. Army Corps of Engineers
- Eisler R (1987) Mercury hazards to fish, wildlife, and invertebrates: a synoptic review. U.S. Fish Wildl. Serv. Biol. Rep. 85(1.10). 90 p
- Furutani A, Rudd JWM (1980) Measurement of mercury methylation in lake water and sediment samples. *App. Environ. Microbiol.* 40:770-776
- Gallant A, Whittier T, Larsen D, Omernik J, Hughes R (1989) Regionalization as a tool for managing environmental resources. EPA/600/3-89-060
- Grieb TM, Driscoll CT, Gloss SP, Schofield CL, Bowie GL, Porcella DB (1990) Factors affecting mercury accumulation in fish in the Upper Michigan Peninsula. *Environ. Toxicol. Chem.* 9:919-930
- Håkanson L, Andersson T, Nilsson A (1990) Mercury in fish in Swedish lakes - linkages to domestic and European sources of emission. *Water Air Soil Pollut.* 50:171-191
- Håkanson L (1980) The quantitative impact of pH, bioproduction and Hg-contamination on the Hg-content of fish (pike). *Environ. Poll.* 18:285-304

- Hill S (1973) Study of mercury and heavy metals pollutants in the Jordan Creek Drainage. Unpublished report, College of Mines, University of Idaho, Moscow, Idaho.
- Huckabee JW, Elwood JW, Hildebrand SG (1979) Accumulation of mercury in freshwater biota. In: Nriagu JO (ed) The biogeochemistry of mercury in the environment, Elsevier Press, Holland, p 277
- Jackson TA (1991) Biological and environmental control of mercury accumulation by fish in lakes and reservoirs of Northern Manitoba, Canada. Can. J. Fish. Aquat. Sci. 48:2449-2470
- Jackson TA, Woychuk RN (1980) The geochemistry and distribution of mercury in the Wabigoon river system. In: Jackson TA (ed) Mercury pollution in the Wabigoon-English river system of northeastern Ontario, and possible remedial measures: a progress report. 1-28
- Jenne EA (1973) Mercury in waters of the western U.S. In: Buhler DR (ed) Mercury in the western environment. Oregon State University, Corvallis, OR, 16-28
- Jernelov A, Asell B (1973) The feasibility of restoring mercury-contaminated waters. In: Krenkel PA (ed) Heavy metals in the aquatic environment: an international conference. Pergamon Press, NY, 299-309.
- Johnson DM, Petersen RR, Lycan DR, Sweet JW, Neuhaus ME (1985) Atlas of Oregon lakes. Oregon State University Press, Corvallis, OR.
- Johnston TA, Bodaly RA, Mathias JA (1991) Predicting fish mercury levels from physical characteristics of boreal reservoirs. Can. J. Fish. Aquat. Sci. 48:1468-1475.
- Klein DH (1973) Sources and present status of the mercury problem. In: Buhler DR (ed) Mercury in the western environment. Oregon State University Press, Corvallis, OR, 1-13.
- Lacerda LD, Pfeiffer WC, Marins RV, Rodrigues S, Souza CMM, Bastos WR (1991) Mercury dispersal in water, sediments, and aquatic biota of a gold mining tailing deposit drainage in Pocone, Brazil. Water Air Soil Pollut. 55:283-294
- Lindberg S, Stokes PM, Goldberg E, Wren C (1987) Group report: mercury. In: Hutchinson TC, Meema KM (eds) Lead, cadmium, mercury and arsenic in the environment. John Wiley & Sons Ltd., New York, 17-33
- Lowe, TP, May TW, Brumbaugh WG, Kane DA (1985) National contaminant biomonitoring program: concentrations of seven elements in freshwater fish, 1978-1981. Arch. Environ. Contam. Toxicol. 14:363-388
- McMurty MJ, Wales DL, Scheider WA, Beggs GL, Dimond PE (1989) Relationship of mercury concentrations in lake trout (*Salvelinus namaycush*) and smallmouth

- (*Micropterus dolomieu*) to the physical and chemical characteristics of Ontario lakes. Can. J. Fish. Aquat. Sci. 46:426-434
- Nriagu JO (1979) Production and uses of mercury. In: Nriagu JO (ed) The biogeochemistry of mercury in the environment, Elsevier Press, Holland, 23-40
- Omernik J, Gallant A (1986) Ecoregions of the Pacific Northwest. EPA/600/3-86-033
- Oregon Department of Geology and Mineral Industries (1982) Geothermal resources of Oregon (map)
- Phillips GR, Medvick PA, Skaar DR, Knight D (1987) Factors affecting the mobilization, transport, and bioavailability of mercury in reservoirs of the Upper Missouri River Basin. U.S. Department of the Interior, Fisheries and Wildlife Service, tech. rep. #10
- Phillips GR (1975) Some quantitative aspects of mercury accumulation by rainbow trout. PhD dissertation, Oregon State University
- Ramlal PS, Rudd JWM, Hecky RE (1986) Methods for measuring specific rates of mercury methylation and degradation and their use in determining factors controlling net rates of mercury methylation. Appl. Environ. Microbiol. 51:110-114
- Richman LA, Wren CD, Stokes PM (1988) Facts and fallacies concerning mercury uptake by fish in acid stressed lakes. Water Air Soil Pollut. 37:465-473
- Schlinder DW, Hesslein RH, Wagemann R, Broecker WS (1980) Effects of acidification on mobilization of heavy metals and radionuclides from the sediments of a freshwater lake. Can. J. Fish. Aquat. Sci. 37:1723-1729
- Scott WB, Crossman EJ (1973) Freshwater fishes of Canada. Fish. Res. Bd. Can. Bull. 184, 966 pp
- Sorensen, JA, Glass GE, Schmidt KW, Huber JK, Rapp GR Jr (1990) Airborne mercury deposition and watershed characteristics in relation to mercury concentrations in water, sediments, plankton, and fish of eighty Northern Minnesota lakes. Environ. Sci. Technol. 24:1716-1727
- Stokes PM, Wren CD (1987) Bioaccumulation of mercury by aquatic biota in hydroelectric reservoirs: a review and consideration of mechanisms. In: Hutchinson TC, Meema KM (eds) Lead, cadmium, mercury and arsenic in the environment. John Wiley & Sons Ltd., New York, 255-277
- Suns K, Hitchin G (1990) Interrelationships between mercury levels in yearling yellow perch, fish condition, and water quality. Water Air Soil Pollut. 50:255-265

- Utne JF, Solomon J, Grift B (1972) Rapid semi-micro method for the determination of methyl mercury in fish tissue. J. Assoc. Offic. Analyt. Chem. 55(3): 583-589
- Weiner JG, Fitzgerald WF, Watras CJ, Rada RG (1990a) Partitioning and bioavailability of mercury in an experimentally acidified Wisconsin lake. Environ. Contam. Toxicol. 9:909-918
- Wiener JG, Martini RE, Sheffy TB, Glass GE (1990b) Factors influencing mercury concentrations in walleyes in Northern Wisconsin Lakes. Trans. Am. Fish. Soc. 119:862-870
- Winfrey MR, Rudd JWM (1990) Environmental factors affecting the formation of methylmercury in low pH lakes. Environ. Toxicol. Chem. 9:853-869
- Worcester TC (1979) Mercury accumulation in fish from Cottage Grove Reservoir and its tributaries. M.S. thesis, Oregon State University
- Wren CD, Scheider WA, Wales DL, Muncaster BW, Gray IM (1991) Relation between mercury concentrations in walleye (*Stizostedion vitreum vitreum*) and northern pike (*Esox lucius*) in Ontario lakes and influence of environmental factors. Can. J. Fish. Aquat. Sci. 48:132-139

Appendix 1. Total mercury concentrations for individual sediment samples collected from three Oregon reservoirs. Sites 1, 2 and 3 are Cottage Grove, Ochoco and Owyhee Reservoirs, respectively. Sampling sites for each reservoir are as identified in the body of the report(Figure 3).

SITE	LOC	DUP	MONTH	YEAR	UG/G
1	1	1	9	92	0.04
1	1	2	9	92	0.17
1	1	3	9	92	0.13
1	2	1	9	92	0.2
1	2	2	9	92	0.27
1	2	3	9	92	0.15
2	1	1	9	92	0.12
2	1	2	9	92	0.06
2	1	3	9	92	0.11
2	2	1	9	92	0.1
2	2	2	9	92	0.14
2	2	3	9	92	0.08
3	1	1	9	92	0.07
3	1	2	9	92	0.04
3	1	3	9	92	0.1
3	2	1	9	92	0.13
3	2	2	9	92	0.19
3	2	3	9	92	0.16
3	3	1	9	92	0.07
3	3	2	9	92	0.04
3	3	3	9	92	0.04
3	1	1	9	90	0.099
3	1	2	9	90	0.056
3	1	3	9	90	0.086
3	2	1	9	90	0.065
3	2	2	9	90	0.043
3	2	2	9	90	0.056
3	2	3	9	90	0.037
3	3	1	9	90	0.060
3	3	2	9	90	0.055
3	3	3	9	90	0.066
1	1	1	9	90	0.099
1	1	2	9	90	0.358
1	1	2	9	90	0.353
1	1	3	9	90	0.080
1	2	1	9	90	0.040
1	2	2	9	90	0.134
1	2	3	9	90	0.109
2	2	1	9	90	0.084
2	2	2	9	90	0.119
2	2	3	9	90	0.102
2	3	1	9	90	0.068
2	3	2	9	90	0.080
2	3	3	9	90	0.077
3	1	1	6	90	0.053
3	1	2	6	90	0.053
3	1	3	6	90	0.054

SITE	LOC	DUP	MONTH	YEAR	UG/G
3	2	1	6	90	0.082
3	2	2	6	90	0.025
3	2	2	6	90	0.152
3	2	3	6	90	0.072
3	2	3	6	90	0.063
3	3	1	6	90	0.050
3	3	2	6	90	0.050
3	3	3	6	90	0.053
1	1	1	6	90	0.050
1	1	2	6	90	0.050
1	1	3	6	90	0.047
1	2	1	6	90	0.046
1	2	1	6	90	0.069
1	2	2	6	90	0.091
1	2	3	6	90	0.113
2	1	1	6	90	0.085
2	1	2	6	90	0.095
2	1	3	6	90	0.117
2	2	1	6	90	0.091
2	2	1	6	90	0.153
2	2	2	6	90	0.084
2	2	3	6	90	0.085
2	2	3	6	90	0.101
3	2	1	9	89	0.111
3	2	1	9	89	0.096
3	2	2	9	89	0.103
3	2	3	9	89	0.168
1	2	1	9	89	0.227
1	2	2	9	89	0.682
1	2	3	9	89	0.682
2	2	1	9	89	0.119
2	2	2	9	89	0.079
2	2	3	9	89	0.119

Appendix 2. Fish Tissue Mercury Concentration.

Site	Species	Year	Sample id	Weight (g)	Length (in)	Age (yr)	Hg (ug/g ww)	Sites:
1	1	1990	4				0.36	1=C. Grove
1	1	1990	1			3	0.44	2=Ochoco
1	1	1990	5			3	0.22	3=Owyhee
1	1	1990	2			4	1.49	
1	1	1990	3			5	1.79	
1	1	1992	1	158.5	9	2	0.42	Species:
1	1	1992	2	243.8	10.25	3	0.64	1=Largemouth bass
1	1	1992	3	312.3	11	3	0.59	2=Rainbow trout
1	1	1992	4	308.5	11.5	3	0.74	3=Smallmouth bass
1	1	1992	5	378.5	12	4	0.42	
1	1	1992	6	657.7	13.5	4	0.43	
1	1	1992	7	596.2	13.5	3	0.37	
2	2	1990	1			2	0.79	
2	2	1992	1	72.3	6.5	1	0.23	
2	2	1992	2	72	6.5	1	0.23	
2	2	1992	3	55.4	6	1	0.42	
2	2	1992	4	168.3	9	3	0.28	
2	2	1992	5	208.2	9.54	2	0.26	
2	2	1992	6	228	10	1	0.28	
2	2	1992	7	270	10.5	2	0.29	
2	2	1992	8	307.8	11.5	2	0.26	
2	2	1992	9	278.9	11.5	2	0.34	
2	2	1992	10	348.5	11.5	2	0.25	
2	2	1992	11	370.2	12.5	2	0.23	
2	2	1992	12	567.7	14.5	4	0.38	
3	3	1990	2			2	0.75	
3	3	1990	1			3	0.65	
3	3	1990	3			3	0.79	
3	3	1990	5			4	1.16	
3	3	1990	4			5	0.63	
3	3	1992	1			4	0.9	
3	3	1992	2			3	0.48	
3	3	1992	3			3	0.48	
3	3	1992	4			3	0.63	
3	3	1992	5			4	0.97	
3	3	1992	6			5	1.18	
3	3	1992	7			3	0.61	
3	3	1992	8			3	0.6	
3	3	1992	9			4	0.73	
3	3	1992	10			5	2.54	

Length and weight data are not available for individual fish where columns are blank.

Hg in fish 1990 C.G., Ous, OC

AN ECOREGION APPROACH TO MERCURY DYNAMICS  
IN THREE OREGON RESERVOIRS

Final Report

June 10, 1991

Submitted to:

Oregon Department of Environmental Quality  
811 SW Sixth Avenue  
Portland, OR 97204-1390

Susan M. Allen and Lawrence R. Curtis  
Oak Creek Laboratory of Biology  
Department of Fisheries and Wildlife  
Oregon State University  
Corvallis, OR 97331

## ABSTRACT

AN ECOREGION APPROACH TO MERCURY DYNAMICS IN THREE OREGON RESERVOIRS. S.M. Allen and L.R. Curtis. Department of Fisheries and Wildlife, Oregon State University, Corvallis, OR 97331

Three Oregon reservoirs (Ochoco, Owyhee, and Cottage Grove) were evaluated for environmental parameters affecting mercury dynamics and bioaccumulation in fish using an ecoregion approach. Ecoregions are distinguished by topography, geology, soil type and composition and land use patterns. We examined pH, dissolved oxygen, hardness, conductivity and mercury concentration of the water, and complexing agent levels, volatile solid content, inorganic and organic mercury in sediments to mercury in several fish species. Mercury concentrations in water were below detection (0.2 ppb) in most water samples from the 3 reservoirs. In samples above detection, it is expected that mercury was predominantly in the particulate fraction, and thus not in true solution. Mercury in sediment occurred only in the inorganic state; methyl-mercury was below detection (0.2 ppb) in 83% of all samples. Significant differences ( $p=0.05$ ) in sediment mercury concentrations were found among the reservoirs and among sampling dates within each reservoir. Despite the low mercury levels in water and sediment, mercury in fish over 4 years old tested above the FDA limit of 1.0 ppm. More than 99.9% of the mercury was found as organic mercury in all fish. Mercury in lateral muscle did not differ significantly between smallmouth bass in Owyhee Reservoir and largemouth bass in Cottage Grove Reservoir. We conclude that although ecoregion parameters alone do not explain mercury dynamics, they may influence the methylation rate in areas of similar loading rates. (Supported by USEPA, grant S-000397/01).

**RECEIVED**  
JUN 17 1991

WATER QUALITY DIVISION  
DEPT. OF ENVIRONMENTAL QUALITY



#### ACKNOWLEDGEMENTS

We would like to acknowledge the assistance of the Oregon Department of Environmental Quality (ODEQ), Oregon Department of Fish and Wildlife (OSFW), and the U.S. Bureau of Mines. The ODEQ staff, particularly Doug Drake and Gene Foster, assisted in fish sampling. Kin Daily of ODFW performed scale analyses to determine fish ages. The U.S. Department of the Interior, Bureau of Mines analyzed sediment samples for cinnibar content.

## TABLE OF CONTENTS

Abstract

Acknowledgements

I.	INTRODUCTION .....	1
II.	SITE DESCRIPTION .....	2
	A. Geology .....	4
	B. Historical mining practices .....	6
III.	METHODS .....	8
	A. Field sampling .....	8
	B. Chemical analyses .....	8
	1. Total mercury in water .....	8
	2. Total mercury in sediment .....	8
	3. Methylmercury in sediment .....	10
	4. Total and organic mercury in fish .....	11
	5. Additional analyses .....	11
IV.	RESULTS .....	12
	A. Mercury in water and sediment .....	12
	B. Mercury in fish .....	14
V.	DISCUSSION .....	15
	A. Mercury in water .....	15
	B. Mercury in sediment .....	16
	C. Mercury in fish .....	18
VI.	LITERATURE CITED .....	22

## TABLES AND FIGURES

Table 1	- Ecoregion attributes .....	3
Table 2	- Drainage basin characteristics .....	4
Table 3	- Water chemistry and sediment parameters .....	13
Table 4	- Mercury concentrations in fish muscle .....	14
Table 5	- Conditions favorable for mercury bioaccumulation .	19

\*\*\*\*\*

Figure 1	- Ecoregions of Oregon .....	2
Figure 2	- Geothermal activity in the area of the reservoirs	5
Figure 3	- Location of mercury depsoits and mines .....	7
Figure 4	- Location of sample sites .....	9
Figure 5	- Sediment mercury concentrations by reservoir and sampling date .....	13
Figure 6	- Mercury concentration in fish tissue as a function of fish age .....	15

## I. INTRODUCTION

Mercury is a ubiquitous metal, occurring in different concentrations in the soil, rocks, air and water throughout the world. Elevated concentrations of mercury in surface water can be derived from many sources, including natural processes and anthropogenic losses. Natural processes include volcanic and atmospheric deposition, degassing, and surface runoff and erosion of mercuric soils; anthropogenic sources include mercury mining and processing, energy related activities, pesticide application, chloro-alkali operations, and smaller emissions from other industrial processes (Andren and Nriagu, 1979).

Mercury levels in Oregon reservoir fish have been found to exceed the 1.0 ppm U.S. Food and Drug Administration limit for human consumption (DEQ, pers comm.: Worcester, 1979; Lowe et al., 1985). The goal of this project was to identify abiotic and biotic factors contributing to accumulation of mercury in fish tissue in three Oregon reservoirs using an ecoregion approach. The specific objectives were to identify probable sources of mercury in the reservoir systems and parameters affecting mercury dynamics and bioavailability, and to examine the extent to which ecoregion-level parameters influence mercury dynamics in these reservoir systems.

An ecoregion is defined as an area in which within-region variation is less than between-region variation (Gallant et al., 1989). While ecoregions can be distinguished based on almost any physiogeographical feature, a common suite of parameters is land surface form, potential natural vegetation, land use and soil characteristics (Omernik, 1986). Limnological and biological features of aquatic ecosystems are largely determined by these broad-scale parameters.

Ecoregion phenomena directly and indirectly influence the dynamics of compounds in the environment. Mineral composition of soils and land use practices within a watershed have major impacts on mercury loading rates. Indirectly, the physical, chemical, and biological features of an aquatic system influence mercury dynamics (Hakanson, 1980; Akielaszek and Haines, 1981).

Given these observations, an ecoregion approach to mercury dynamics and bioaccumulation seems to be an appropriate model. We have developed this model with the assumptions that mercury burdens in the reservoirs are largely derived from their watersheds and that mercury bioaccumulation by fish is generally related to the concentration of total mercury (particularly methylmercury) that is bioavailable.

To investigate the validity of an ecoregion approach, we examined both qualitative and quantitative ecoregion parameters as they relate to mercury concentrations in water, sediment and fish. The qualitative parameters are those which define ecoregions (land surface form, potential natural vegetation, land use and soils). The quantitative parameters examined were pH, conductivity, hardness and alkalinity of the water column, and the clay and organic matter content of the sediment. In addition, characteristics of the specific drainage basins were compared; these include basin area, relief, annual precipitation and land uses. Limnological features of each

reservoir were also considered. The interactions between mercury reserves in water, sediment and fish were also investigated.

## II. SITE DESCRIPTION

Cottage Grove, Owyhee and Ochoco reservoirs are located in three distinct ecoregions (Fig. 1). The attributes of each ecoregion are presented in Table 1. Drainage basin and limnological characteristics of the study reservoirs were compared to other reservoirs within the ecoregions as reported by Johnson et al., 1985, and were considered to be representative of reservoirs of similar size within the ecoregions. The specific characteristics of each drainage basin are summarized in Table 2.

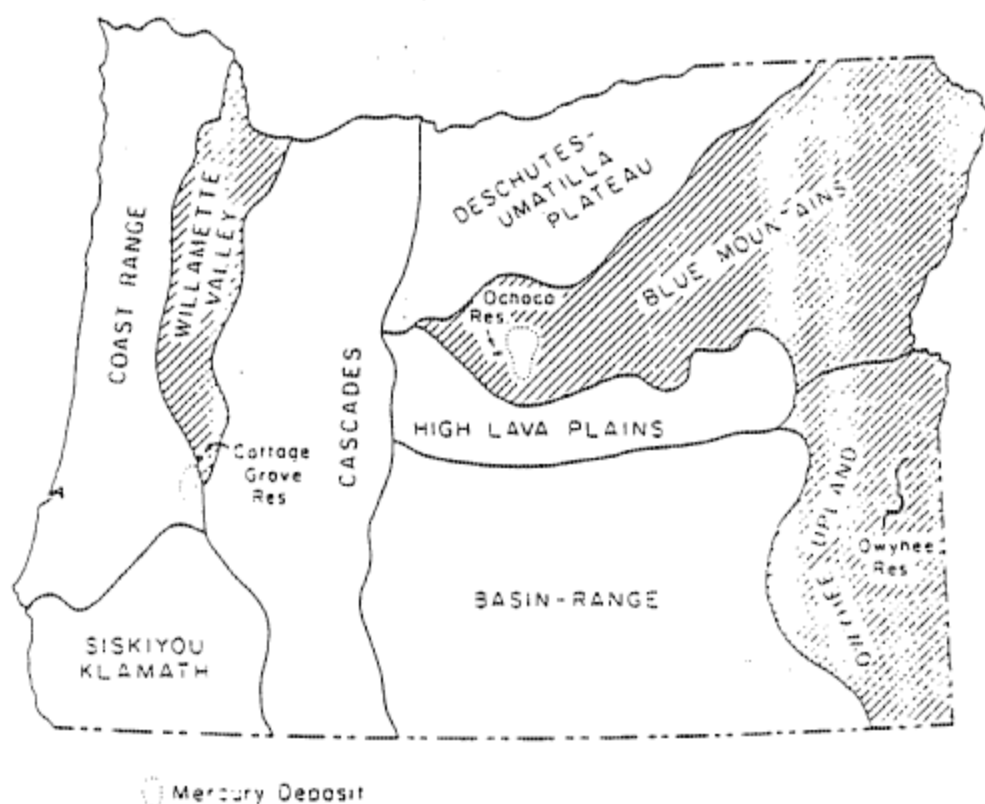


Figure 1 - Location of study areas and mercury deposits within the different ecoregions. Compiled from : Baldwin, 1976.

Table 1 - Qualitative characteristics of the three ecoregions

Ecoregion	Land Surface Form	Potential Natural Vegetation	Land Use	Soils
Willamette Valley (C. Grove Res.)	Plains with hills, or open hills  Elevation: 100 - 2000 ft.	Cedar/hemlock/Douglas fir, mosaic of Oregon oakwoods and cedar/hemlock/Douglas fir  Annual precipitation: 40 in.	Emphasis on cropland with some interspersions of pasture, woodland and forest	Xeric Mollisols, Vertisols, and Alfisols of interior valleys
Blue Mountains (Ochoco Res.)	Low to high open mountains, change in local relief can be 1000-3000 ft.  Elevation: 2700 - 10000 ft.	Grand fir/Douglas fir, western ponderosa pine, western spruce/fir, Douglas fir  Annual precipitation: 10 - 20 in.	Forest and woodland, grazed	Soils of eastern interior mountains, Mollisols, Inceptisols
Snake River Basin/ High Desert (Owyhee Res.)	Tablelands with moderate to high relief, plains with hills or low mountains  Elevation: 2500-9000 ft.	Sagebrush steppe (sagebrush, wheat-grass), saltbush/greasewood  Annual precipitation: 8 - 12 in.	Desert shrubland, grazed	Aridisols, aridic mollisols

Adapted from: Omernik, J.M. and A.L. Gallant. 1986. Ecoregions of the Pacific Northwest. EPA/600/3-86/033

Table 2 - Drainage basin characteristics

	Cottage Grove	Ochoco	Owyhee
Drainage basin area (km <sup>2</sup> )	257	288	11,160
Annual Precipitation (cm)	122-157	43-64	25-64
Land use (% total area)			
Forest	96.5	73.8	0.5
Range	1.0	21.3	93.2
Surface area (ha)	461	388	5625
Average depth (m)	9	8.4	24.6
Shoal area (%)	17	29	4
Retention time (months)	2	5	20
pH	7.7	8.4	8.4
Conductivity (umhos/cm)	63	197	160
Sulphide (SO <sub>4</sub> mg/l)	1.2	3.9	11.7
Dissolved oxygen (mg/l)	7.2	-	8.3
Trophic status	mesotrophic	eutrophic	eutrophic

Compiled from: Johnson et al., 1985.

#### A. Geology

The geology of all three areas is characterized by a combination of sedimentary and volcanic formations. The area surrounding Cottage Grove reservoir is older Cenozoic marine and estuarine sedimentary deposits with minor amounts of volcanic elements; the Ochoco area predominantly contains metasedimentary and metavolcanic formations; and the Owyhee region is a mixture of deposits of sedimentary and volcanic origin (Baldwin, 1976).

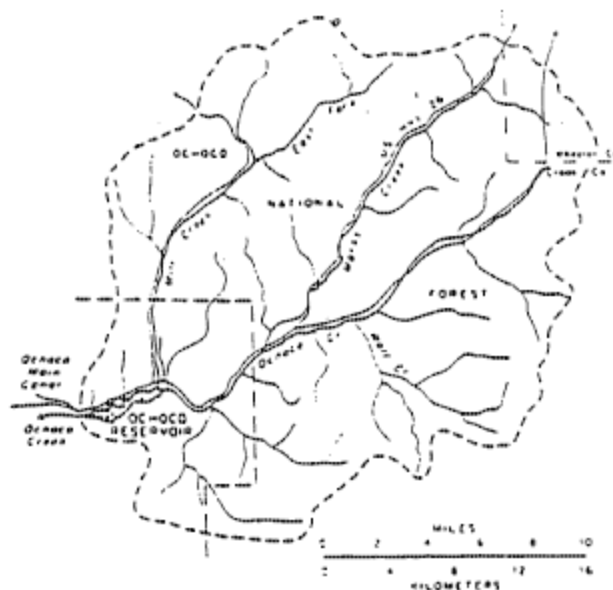
The geothermal conditions of the three areas are shown in Fig. 2. Geothermal activity is high in the Owyhee area, moderate in the Ochoco area, and virtually undetectable in the Cottage Grove basin. Therefore, the potential contribution of geothermal venting to mercury loading is likely to vary accordingly among the three reservoirs based on the differences in geothermal activity.

While mercury most frequently occurs as deposits in rock fractures and veins, it may also be found in low concentrations in other geologic formations. In the Owyhee River area, mercury is commonly found as an anomaly, present in 12 of 23 random outcrop rock-chip samples (Gray et al., 1983). Mercury concentrations averaged 0.3 ppm (Gray et al., 1983).

COTTAGE GROVE RESERVOIR DRAINAGE BASIN



OC-OCO RESERVOIR DRAINAGE BASIN



OWYHEE RESERVOIR DRAINAGE BASIN

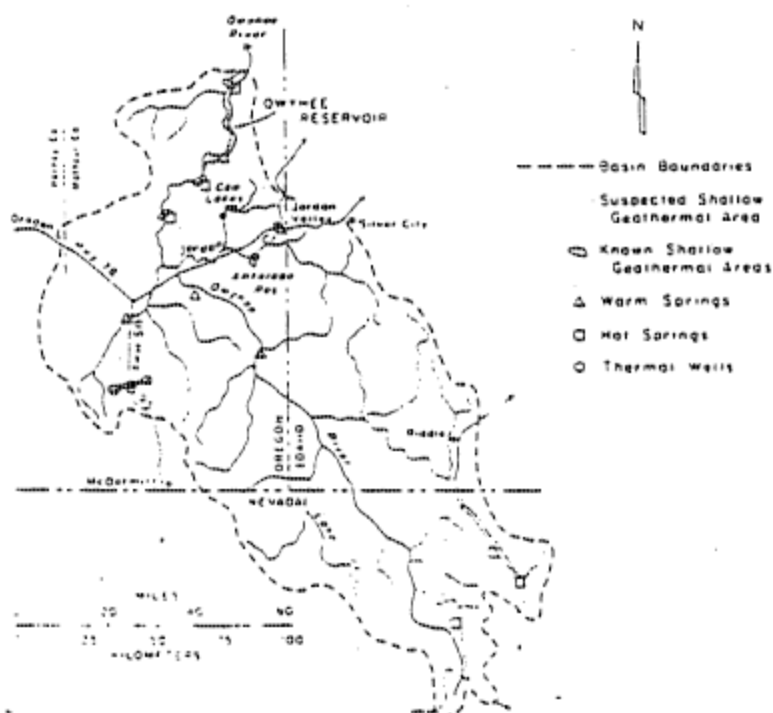


Figure 2 - Geothermal activity in the three study areas.  
Compiled from: Johnson et al., 1985; Horton, 1964;  
Oregon Department of Geology and Mineral Resources, 1982.



## 8. Historical Mining Practices

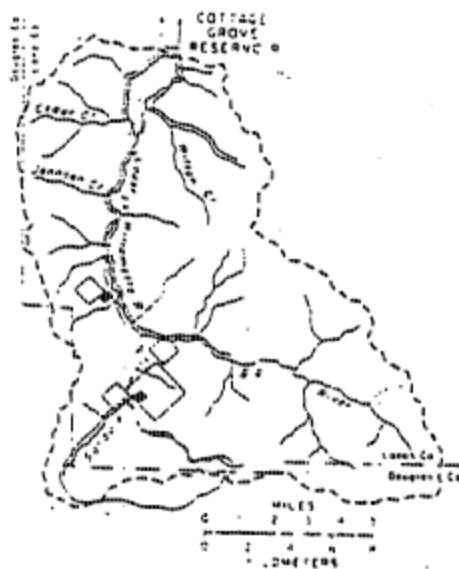
Extensive mercury mining occurred in all three areas, as shown in Fig. 3. As is true for all mercury mines in the state, production occurred primarily from the late 1800s until 1950, with peak production coinciding with war years. Since 1950 the increased Korean mercury production and the surplus from war years have saturated the mercury market and decreased prices such that Oregon mines have ceased production (Brooks, 1971).

The second largest mercury mine in Oregon, Black Butte Mine, is located 2 miles south of Cottage Grove Reservoir, within the drainage basin (Brooks, 1971). Active intermittently from 1882 to 1966, this mine produced 18,156 flasks (Brooks, 1971). The ore in this area has been low-grade, approximately 0.175% by weight (Brooks, 1971).

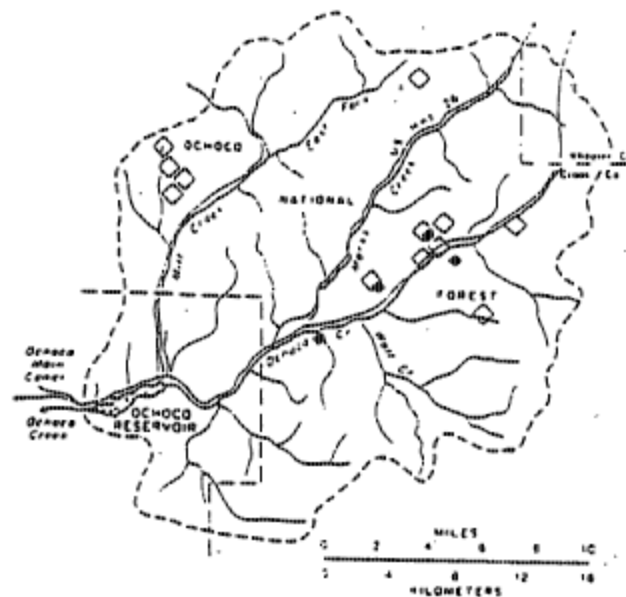
Mercury production in Ochoco basin has been from several small mines, due to the discontinuous faulting in this region (Brooks, 1971). Prior to 1943, the four mines in the Ochoco basin (Byram-Oscar, Staley, Champion, and Taylor Ranch) collectively produced 857 flasks (Brooks, 1971).

Although there are no mercury mines in Oregon in the Owyhee basin, Brooks (1971) reported that one of the leading national mercury producing mines in the country, is in the Nevada section of the drainage basin. This could not be confirmed by maps of mercury occurrences in Nevada (see Lawrence and Wilson, 1962).

COTTAGE GROVE RESERVOIR DRAINAGE BASIN



OCHOCO RESERVOIR DRAINAGE BASIN



OWYHEE RESERVOIR DRAINAGE BASIN

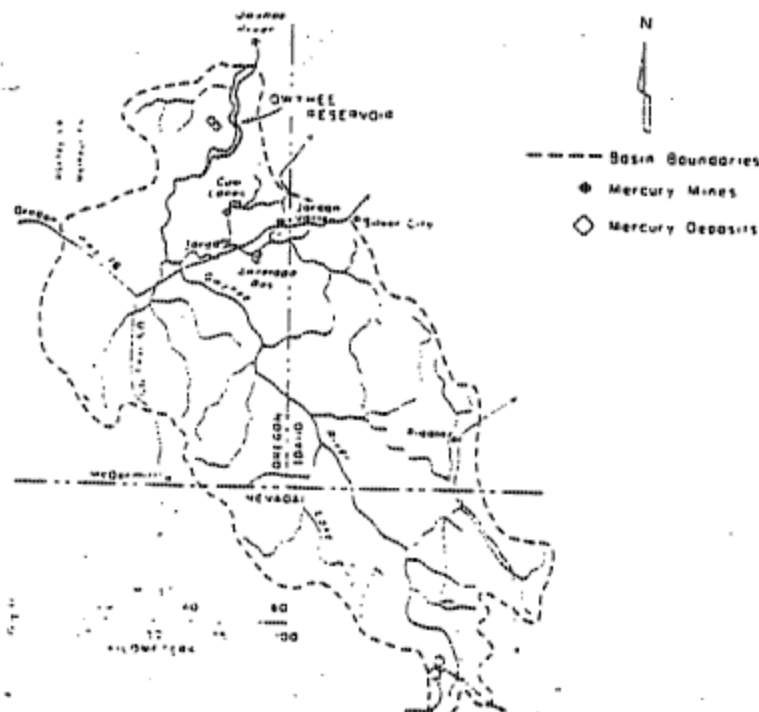


Figure 3 - Mercury mines and deposits in the three study areas. Compiled from: Johnson et al., 1985; Lawrence and Wilson, 1962; Ferns and Huber, 1954; and State Water Resources Board, 1960a, 1960b.

### III. METHODS

#### A. Field Sampling

Two or three sites were sampled for water and sediment at each reservoir, depending on the reservoir size and water level (Fig. 4). Water and sediment samples were collected in September 1989, and June and September, 1990.

Water samples were collected 0.5 m from the surface in 1 L polyethylene bottles, pre-rinsed with 10%  $\text{HNO}_3$ . Core sediment samples were obtained using a 1" d PVC pipe. The uppermost 5 cm of the core was transferred into a pyrex plate, mixed and collected in 250 ml polyethylene bottles, pre-rinsed with 10%  $\text{HNO}_3$ . All samples were frozen immediately, and stored frozen until subsequent analysis.

The following parameters were also measured at each site: water pH (Orion Model 250 pH meter), conductivity, and dissolved oxygen (Winkler method).

Fish were collected by electroshocking in cooperation with the Oregon Department of Environmental Quality in September and October, 1990. Fish were filleted in the field, and fillets (with skin) and liver samples were immediately frozen and stored frozen.

#### B. Chemical Analyses

1. Total mercury in water: Total mercury concentration in water was determined using cold vapor atomic absorption according to the procedure in Standard Methods for the Examination of Water and Wastewater. A 100-ml volume of each sample was transferred to a 250 ml Erlenmeyer flask, to which 5 ml concentrated  $\text{H}_2\text{SO}_4$  and 2.5 ml concentrated  $\text{HNO}_3$ , and 15 ml of 5%  $\text{KMnO}_4$  were added. After fifteen minutes 8 ml of 5%  $\text{K}_2\text{S}_2\text{O}_8$  was added to each sample, and the flasks were heated in a 95°C water bath for 2 hours. The samples were cooled to room temperature, transferred to 250 ml gas-scrubbing reaction flasks, treated with 8 ml of 24%  $\text{NaCl}$ -hydroxylamine sulfate to reduce excess  $\text{KMnO}_4$ . Immediately following the addition of 5 ml 10%  $\text{SnCl}_2$  in dilute  $\text{HCl}$ , the flasks were supplied with flow-through nitrogen gas (2 l/min). Mercury vapor was passed through a Coleman Model 50 mercury analyzer (Perkin-Elmer Co., Maywood, IL), connected to a Microscribe 4500 recorder set at 5 mV. (The Recorder Company, San Marcos, TX). Peak area was calculated as height X width at half-height. Unknowns were determined using a standard curve, based on  $\text{HgCl}_2$  in  $\text{HNO}_3$  (0.25 - 1.0  $\mu\text{g/l}$ ). More than 75% of water samples were analyzed in duplicate.

2. Total mercury in sediment: Sediment samples were analyzed following the methods outlined in Buhler et al. (1984). After initial preparation and hot-acid digestion, this procedure is very similar to the analysis of water. Thoroughly mixed sediment samples (3-5 g) were dried to a constant weight in a 60°C oven (approx. 96 h). Samples were crushed using a ceramic mortar and pestle, and passed through a 1 mm mesh screen. Particles that did not pass through the screen were pulverized a second time, and screened. Any

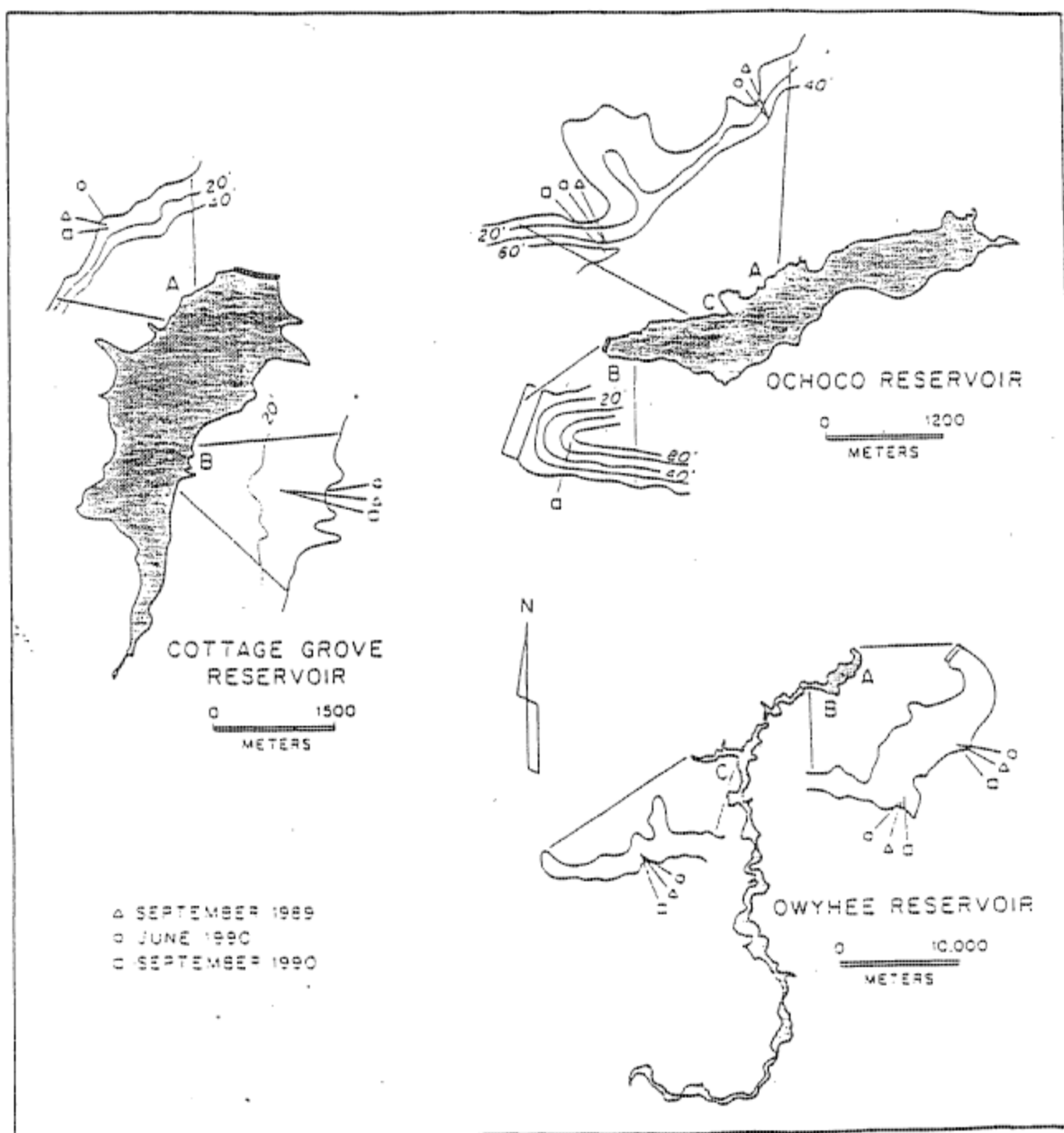


Figure 4 - Location of sample collection sites on each reservoir by date of collection.

remaining material was discarded. Subsamples (0.5 - 1.5 g) were weighed and transferred to glass 250 ml BOD bottles, to which 5 ml deionized  $H_2O$  and 5 ml aqua regia (3 vol. conc.  $HCl$ : 1 vol. conc.  $HNO_3$ ) were added. Samples were placed in a  $95^\circ C$  water bath. After two minutes, 50 ml deionized  $H_2O$  and 50 ml 5%  $KMnO_4$  were added to each sample. Samples were digested in the water bath for 30 min., and cooled to room temperature. Fifteen min. before analysis, samples were treated with 50 ml deionized  $H_2O$  and 8 ml 24%  $NaCl$ -hydroxylamine and placed in a hood to allow the evolved oxygen gas to escape. Samples were transferred to the 250 ml reaction flasks used for water analysis, and analyzed in the same manner. Gas flow was set at 1.5 l/min. Recorder sensitivity was set at 20 mV.

Sediment mercury concentrations were determined based on a standard curve of  $HgCl_2$  in  $HNO_3$  (0.05 - 0.90  $\mu g$ ). The accuracy of the standard curve and recovery efficiency was tested using reference material from the National Institute of Standards and Technology. Most samples (>75%) were analyzed in duplicate.

3. Methylmercury in sediment: Sediment samples were analyzed for methylmercury by gas chromatography using a modification of the multi-stage extraction and concentration procedure developed by Uthe et al. (1972). The modified technique for sediment is outlined by Furutani and Rudd (1980).

Two aliquots of wet sediment (approximately 1 g) were weighed to the nearest 0.001 g. One set of samples was dried to a constant weight in a  $60^\circ C$  oven to determine the water content so that measurements could be standardized on a dry weight basis.

The second set of samples were transferred to 50 ml centrifuge vials. Two ml of 0.5M  $CuSO_4$  and 10 ml of 3M  $NaBr$  in 22% conc.  $H_2SO_4$  were added to the samples. After shaking vigorously for two minutes, samples were centrifuged for 5 min. and transferred to 60 ml glass separatory funnels. Twenty ml of toluene were added and samples were shaken for three min. Following removal of the aqueous phase, the toluene phase was treated with 1 g anhydrous  $NaSO_4$  and decanted into a 50 ml erlenmeyer flask and further dried with 0.5 g anhydrous  $NaSO_4$ . A 10 ml sample of the extract was transferred to a clean separatory funnel, and 5 ml of 0.0025 M  $Na_2S_2O_3$  in 20% ethanol was added. After shaking and standing, 3 ml of the lower aqueous phase was collected into a calibrated, glass-stoppered centrifuge tube, to which 1 ml of 3M  $KI$  and 1 ml hexane were added.

Subsamples (6  $\mu l$ ) were injected into a Hewlett-Packard 5700 gas chromatograph equipped with a  $^{63}Ni$  electron capture detector. The column was packed with 7% Carbowax 20M on Chromosorb W, acid-washed DMCS-treated. Argon/methane gas was supplied at 50 ml/min, and GC attenuation was set at 2 mV. The operating temperatures for the column and detector were  $175^\circ C$  and  $200^\circ C$  respectively. The Microscribe recorder was set at 1 mV, and 1 cm/min. Peak area was calculated by height times width at half-height. Unknowns were compared to standards prepared from methylmercuric chloride in hexane (6-20 ng).

4. Total and organic mercury in fish: Mercury concentration in fish muscle was determined using a hot-base digestion followed by cold vapor atomic absorption.

Fillet samples (1-2 g) were placed in screw-top test tubes, to which 2 ml 10N NaOH was added. Samples were then heated for 30 minutes in a heat block (95°C) and cooled to room temperature. One percent NaCl (8 ml) was added to each sample.

Total mercury was determined by placing 1 ml subsample in a reaction flask along with 3 ml 1% NaCl, 1 ml 1% cysteine, 4 drops octanol, and 1 ml 50% SnCl<sub>2</sub> and 10% CdCl<sub>2</sub> in 4 N HCl. Inorganic mercury was determined by adding 1 ml of 50% SnCl<sub>2</sub> in place of the SnCl<sub>2</sub>- CdCl<sub>2</sub> solution. The flask opening was then covered with a septum, through which 4 ml 10N NaOH was injected by syringe. After thirty seconds, N gas was supplied at 1.5 l/min. The recorder was set at 5mV. Standards were prepared as Hg in HNO<sub>3</sub> (10-100 µg/ml), prepared from a commercially available standard (1000 ppm, Johnson & Mathey, Seabrook, NH).

Organic mercury was calculated as the difference of total and inorganic mercury.

#### 5. Additional analyses

- a. All water samples were analyzed in the lab for hardness, following the EDTA titrimetric method in Standard Methods for the Examination of Water and Wastewater (1985).
- b. All sediment samples were analyzed for percent volatile solids by dried sediments in a 600°C muffle furnace for 4 hours, after Buhler et al. (1984).
- c. Sediment physical characteristics were performed by hydrometer method by the OSU Soil Physics Laboratory.
- d. Sediment samples from 9/90 were sent to the U.S. Bureau of Mines (Albany, OR) for determination of cinnibar content.

#### IV. RESULTS

##### A. Mercury in water and sediment

Twelve water samples and 18-23 sediment samples were analyzed from each reservoir. Results of the limnological and sediment analyses are summarized in Table 3. Mercury was detected in 25%, 6%, and 15% of Cottage Grove, Ochoco, and Owyhee water samples respectively (detection limit of 0.1 ppb). The large range of concentration values (0.12 - 1.0 ppb) suggests that the highest concentrations of mercury are probably found in the particulate fraction. Differences in mean mercury concentration in water among reservoirs were not statistically significant ( $p=0.05$ ).

Sediment mercury concentration and site, date and carbon content differences were examined using Anova and Newman-Keuls multiple comparison analysis. The highest overall mercury level was associated with Cottage Grove reservoir, and this was significantly different from the values for Ochoco and Owyhee reservoir. This finding, however, is largely attributable to the high mercury levels found at both sites in the September 1989 samples (Fig. 5). To confirm the observation that 1989 levels were higher than the other two dates, particularly for Cottage Grove Reservoir, 1989 samples were reanalyzed in a mixed batch with samples from later dates; the results were consistent with previous analyses.

Mercury concentrations did not vary solely as a function of carbon content on a seasonal, or localized manner. For Ochoco, mercury concentration at the up-reservoir site (A) was higher than at site B or C despite no difference in carbon content. The opposite condition existed for Owyhee and Cottage Grove reservoirs; mercury concentrations at the sites within each reservoir were not significantly different, despite differences in carbon content.

Carbon content exhibited a pattern of seasonality, modified by water level fluctuations. Higher carbon levels were observed in the September samples than in the June samples for Cottage Grove Reservoir. In Ochoco, this pattern was overridden by water table fluctuations. The water level was unchanged between September 1989 and June 1990, and the carbon content in sediment was similar for both dates. However, a 34-ft decline in water level between the June 1990 and September 1990 sampling dates, resulting in a much higher carbon content associated with the low water level (7.4% vs. 5.1%). Carbon content in Owyhee Reservoir was consistently between 4.6% and 5.8%, reflecting the low level of carbon loading to the reservoir caused by low vegetational biomass in the watershed.

Mercury values were less influenced by sampling date, suggesting the importance of parameters other than carbon content. Sediment mercury levels in Owyhee and Ochoco reservoirs were consistent for all three sampling dates. In Cottage Grove reservoir, the September 1989 sediment samples had statistically higher mercury levels than September 1990 samples, despite no change in carbon content.

Table 3 - Water chemistry and sediment parameters

Parameter	Cottage Grove ( $\bar{x} \pm \text{SE}$ )	Ochoco ( $\bar{x} \pm \text{SE}$ )	Owyhee ( $\bar{x} \pm \text{SE}$ )
<u>Water</u>			
pH	7.70 <sup>a</sup>	8.53 <sup>b</sup>	8.44
DO	8.20 <sup>a</sup>	7.8 <sup>a</sup>	7.24 <sup>b</sup>
Conductivity ( $\mu\text{mhos}$ )	56 <sup>a</sup>	260 <sup>b</sup>	254 <sup>b</sup>
Hardness (mg $\text{CaCO}_3/\text{l}$ )	39 <sup>a</sup>	101 <sup>b</sup>	72 <sup>c</sup>
Hg (ng/l)	78 $\pm$ 40 <sup>a</sup>	17 $\pm$ 10 <sup>a</sup>	37 $\pm$ 30 <sup>a</sup>
<u>Sediment</u>			
Soil texture	sand- sandy loam	clay- clay loam	sandy loam- clay loam
Clay content (%)	9.5 $\pm$ 6.5	34 $\pm$ 5	12 $\pm$ 9.3
Carbon content (%)	7.11 <sup>a</sup>	5.71 <sup>ab</sup>	5.33 <sup>b</sup>
Total Hg ( $\mu\text{g/g}$ )	0.839 $\pm$ .17	1.010 $\pm$ .16	1.373 $\pm$ .33
Fine grain Hg ( $\mu\text{g/g}$ )	0.313 $\pm$ .03	0.156 $\pm$ .02	0.109 $\pm$ .01

All sediment values are presented on a dry weight basis.  
 Values with different superscripts are significantly different at  $p=0.05$

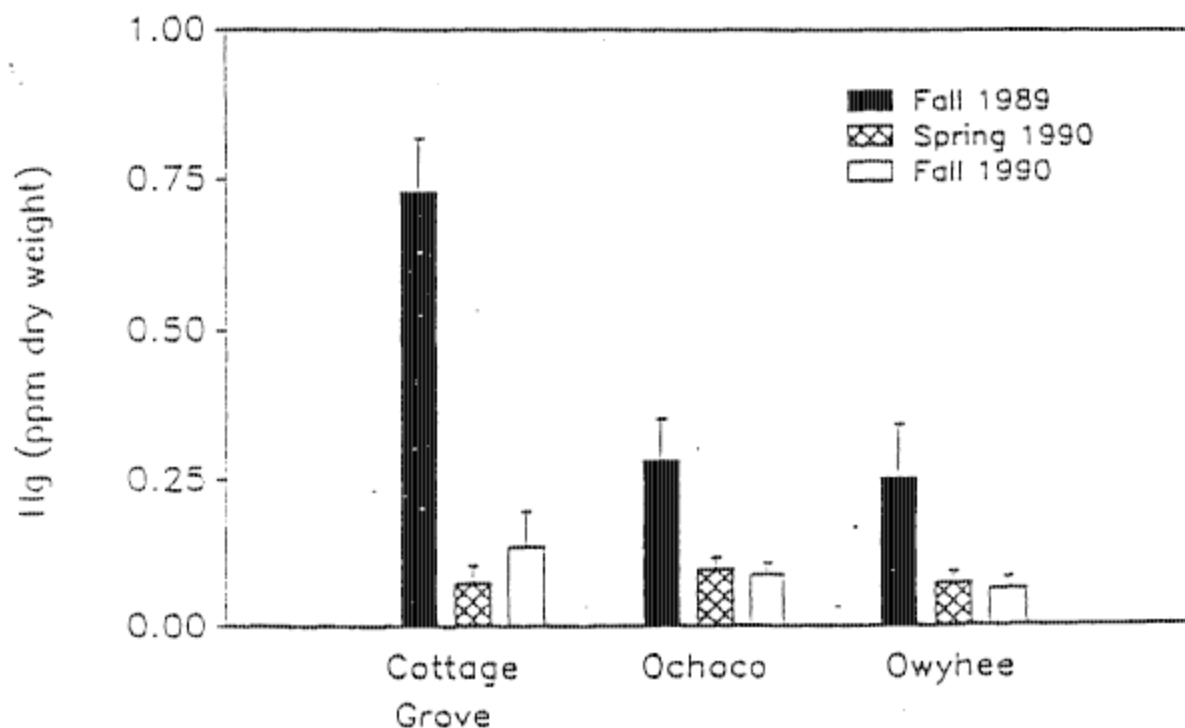


Figure 5 - Mercury concentration in sediment by sampling date.



## B. Mercury in Fish

Five smallmouth bass from Owyhee, five largemouth bass from Cottage Grove, and one rainbow trout from Ochoco were analyzed for total and organic mercury content in lateral fillets. The results are presented in Table 4. One of five fish from Owyhee reservoir and two of five fish from Cottage Grove reservoir exceeded the United States FDA limit for human consumption of 1.0 ppm. Mercury concentrations in fish from Owyhee and Cottage Grove reservoirs were not significantly different (t-test,  $p=0.05$ ).

Organic mercury comprised >99% of the total mercury in all cases.

Mercury concentration in fish muscle increased as a function of age for both bass species (Fig. 6). However, the pattern was not identical for the two reservoirs. This difference may be attributable either to species or reservoir differences, or the small sample size.

Table 4 - Mercury concentrations in fish muscle

<u>Reservoir</u>	<u>Species</u>	<u>Age</u> (years)	<u>Total Hg</u> (ppm)	<u>% MeHg</u>
Cottage Grove	LM Bass	2	0.36	100
		3	0.44	100
		3	0.22	100
		4	1.49	100
		5	1.79	99.9
Ochoco	RB Trout	2	0.79	99.9
Owyhee	SM Bass	2	0.75	100
		3	0.65	99.9
		3	0.79	100
		4	1.16	99.9
		5	1.16	100

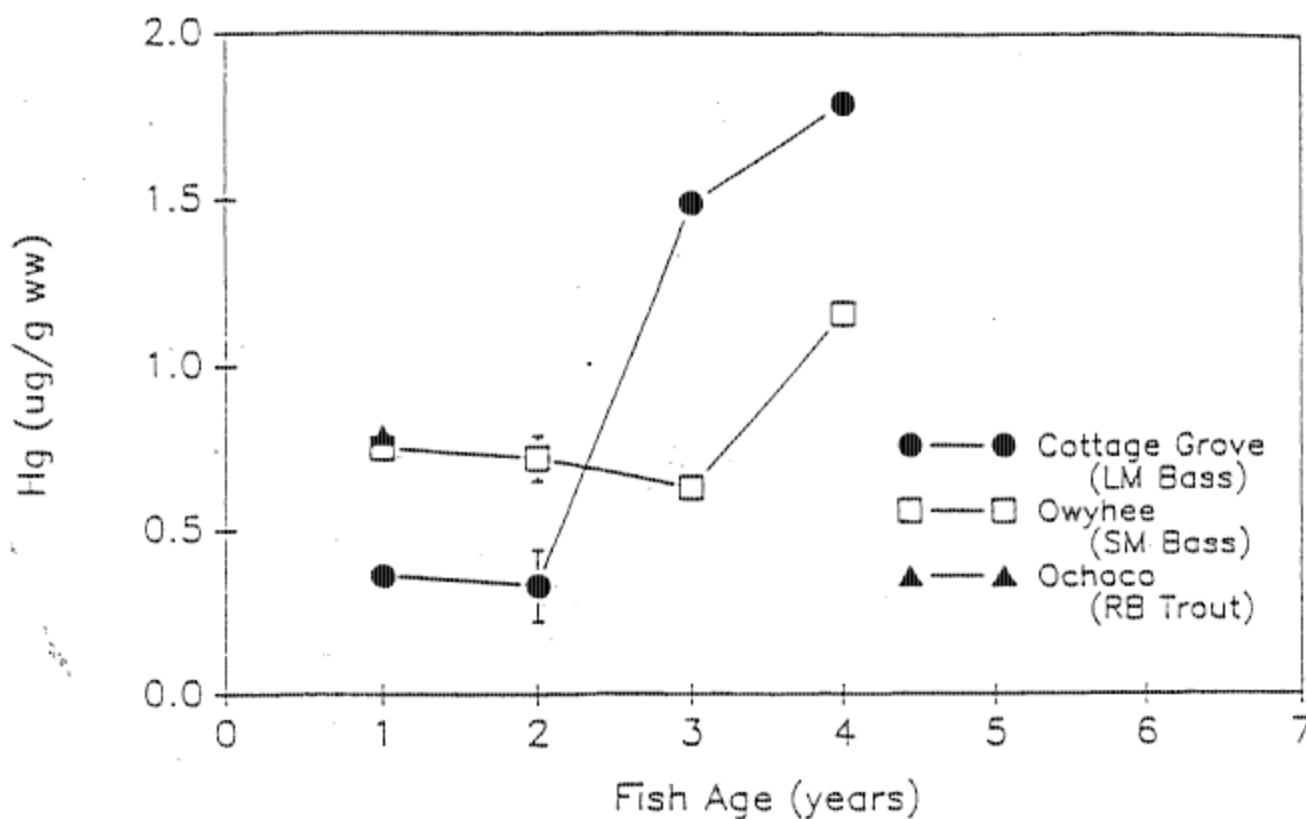


Figure 6 - Mercury concentration in fish tissue by fish age for the three species from the three reservoirs

## V. DISCUSSION

### A. Mercury in water

Surface water mercury concentrations in areas without mercury deposits range from 0.01-0.05 ppb (Wiklander, 1969). Background mercury may be derived from atmospheric deposition (estimated at 0.17 ppb). More than 50% of atmospherically derived mercury is evaporated from the soils and air (Klein, 1973), resulting in background surface water concentration of 0.09 ppb or lower. Jenne (1973) reported that mercury was detected in 9% of unfiltered

water and 3% in filtered water analyses reported in the literature. Thus, in more than half the waters sampled, most of the mercury was suspended in the particulate fraction.

Surface water concentrations in areas with mercury deposits, such as the Pacific Northwest, may be considerably higher than areas without natural deposits (Jenne, 1973). Sampling of the Columbia and Willamette rivers in 1970-71 yielded mercury concentrations between 1-35 ppb for filtered waters (Jenne, 1973), indicating input from sources other than background-level deposition.

Our results indicate an elevated frequency (6-26% of unfiltered samples) and magnitude (0.12-1.0 ppb) surface water contamination above background levels. Therefore, atmospheric inputs alone probably do not account for the elevated mercury concentrations in Cottage Grove, Ochoco and Owyhee reservoirs. This does not, however, indicate that bioavailability to reservoir fish is enhanced, since mercury in the particulate fraction may not be as readily absorbed in biota as dissolved inorganic mercury or methylmercury.

#### 8. Mercury in sediment

Mercury was routinely detected in sediment from all three reservoirs. Despite statistical differences in mercury concentrations in sediment, corresponding concentrations in fish were similar for all reservoirs, based on fish age.

Differences in sediment mercury concentrations are most likely due to differences in loading rates and patterns. Factors that affect loading rates include the quantity of available mercury as natural deposits or mining waste, and sediment transport rates in the drainage basin. Sediment transport rates are, in turn, affected by geological and climatological forces. In this study the quantity of available mercury in the drainage basin seems to be more influential than sediment transport rates.

Drainage basin area was not a significant determinant in sediment mercury concentrations. This is evidenced by the fact that despite the enormity of the Owyhee drainage basin, it did not have elevated mercury loads relative to the other two systems. Although McMurty et al. (1989) observed a positive correlation between mercury concentration in the smallmouth bass tissue and watershed and lake area, it appears that this relationship is valid only where available mercury reserves are approximately equal.

Based on overall relief of the basins, we would expect sediment transport rates to be highest in the Owyhee area. Although the overall relief in all three drainage basins is moderate (Johnson et al, 1985), significant localized differences are present. Buttes dominate in the Owyhee basin which are more susceptible to erosion and sediment transport than gently sloping hills, characteristic of the Cottage Grove basin.

It is difficult to assess the importance of land use patterns in determining the behavior of mercury in these systems. While land use patterns may

influence both the rate of mercury transport to surface waters and movement within the reservoirs, the relative enhancement of erosional forces from forest clearing, agricultural irrigation, and range practices are not known for these areas.

The amount and cycle of annual precipitation and flooding events affect sediment transport. Precipitation in the Owyhee area is primarily snow; 70% of the inflow the Owyhee reservoir occurs between March and May as snowmelt (Johnson et al., 1985). Sparse vegetation and the timing of the snow melt produce conditions favoring high surface runoff and sediment transport (Malheur Soil and Water Conservation District, pers. comm.). This, however, may be offset by differences in the total amount of precipitation. The high rainfall in the Willamette Valley ecoregion may exert a greater influence on relative loading rates. The difference between sediment mercury concentrations in Cottage Grove reservoir between September sampling in 1989 and 1990 may relate to timing of storm events. Local flooding results in a surge of mercury and other materials into reservoirs. This may enhance bioaccumulation; mercury concentrations in northern pike were significantly higher in a year following a severe flood compared to previous or succeeding years (Phillips et al., 1987). One possible explanation is high methylation rates in flooded shoreline sediments (Ramlal et al., 1986).

Given the inability of sediment transport rates to explain differences in sediment mercury concentrations, we suspect that the available stores of mercury within the three reservoir systems are very different, and that determine sediment concentrations.

Mercury burdens in all three reservoirs are most likely are derived from natural mercury deposits exacerbated by past mercury, gold and silver mining in the drainage basins. The relative size and mining effort of the Black Butte Mine in the headwaters of Cottage Grove Reservoir may explain the elevated sediment mercury concentrations. This mine produced 25 times as many flasks of mercury than all the mines in the Ochoco District combined. The lack of site differences in sediment mercury concentrations suggest a uniform, or diffuse mercury distribution within Cottage Grove Reservoir. By contrast, the fact that the upstream site in Ochoco Reservoir exhibited higher sediment mercury concentrations than other sites may reflect a distribution pattern from feeder streams.

The diffuse distribution of mercury in Owyhee Reservoir is consistent with a distant mercury source. Mercury in the Owyhee basin may be derived from its use in gold and silver extraction. Extensive gold and silver mining took place in the Jordan Creek region of Owyhee basin between 1860 and 1920 (Hill, 1973). Mercury used in the amalgamation process can be lost to the environment through inefficient recovery after distillation. It has been estimated that 76 pounds of mercury were lost daily during mining years in Idaho (Hill, 1973).

Geothermal releases of mercury may also be a significant and persistent source of mercury in Owyhee Reservoir given the high level of geothermal activity in Southeast Oregon. Mercury enters the environment during episodes of geothermal and volcanic activity (Gray, pers. comm., 1989).

Hydrothermal solutions transport mercury along rock fractures veins as sulfide and chloride complexes, as evidenced by the common occurrence of cinnibar (HgS) in hot spring deposits (Nriagu, 1979). The volatility of mercury causes it to vaporize from the heated solution and be transported as a gas (Gray, pers. comm.). Mercury halos are formed at varying distances from the geothermal center depending on temperature, pressure, and pH (Gray, pers. comm.). Close to the epicenter, mercury will bind with the gaseous sulfur and precipitate as cinnibar; at greater distances from the geothermal vent, mercury is transported in aqueous solution in a free form or complexed with Cl; a process that is influenced by pH, sulfur availability, temperature, pressure, and chemistry of the mineral solution (Gray, pers. comm.). Thus, further from the epicenter mercury is more available for transport in solution to surface waters and biota.

### C. Mercury in fish

The mercury concentrations in two fish from Cottage Grove Reservoir exceed maximum values reported in the past. Concentrations of mercury in largemouth bass muscle in 1974-5 ranged between 0.55-1.11 ppm (Worcester, 1979). Although this difference may be explained by fish age or seasonal variation, it may also reveal that mercury is accumulating with time in the reservoir. For this reason, it would be desirable to monitor mercury in fish in this reservoir in the future. It has been suggested that stream impoundment elevates mercury bioaccumulation. Following dam construction, mercury concentrations tend to be high initially before reaching an equilibrium; the proposed mechanism is that the surge of trapped soil favors methylmercury production and methylation (Phillips et al., 1987). Investigating the source of mercury in fish in new impoundments, Cox et al. (1979) concluded that insoluble mercury in soils is released through aquatic biological methylation once the soils are submerged. This phenomenon may also explain the increased bioaccumulation associated with flooding.

Of five species examined for mercury concentrations in past efforts (Worcester, 1979), the highest values were observed in largemouth bass. Other species analyzed were chinook salmon, cutthroat, rainbow trout and brown bullhead.

As only one fish could be analyzed from Ochoco Reservoir, it is impossible to perform any statistical analyses. Furthermore, no records were available of past analyses. However, it should be noted that the mercury burden in the 2-year old fish that was sampled was similar to those of 2-year old fish from Cottage Grove and Owyhee reservoirs. This suggests that older fish (4-5 years) in Ochoco Reservoir may also have muscle mercury burdens exceeding the FDA limit.

Smallmouth bass from Owyhee Reservoir in this sampling effort have mercury burdens within the range of past investigations. Mercury concentrations in smallmouth bass sampled in 1987-9 ranged between 0.66 and 1.68 ppm based on the whole fish (DEQ, pers. comm.). Mercury concentrations in smallmouth bass exceeded those of carp and black crappie. Largemouth bass and channel catfish had similar mercury concentrations to smallmouth bass.

As the mercury concentration in fish tissue did not differ among the reservoirs, we could not adequately evaluate several influential factors that may be operative in Oregon reservoirs. Conditions reported to influence mercury availability and bioaccumulation are presented in Table 5.

Table 5 - Conditions favorable for mercury bioaccumulation

<u>Hydrologic</u>	<u>Water chemistry</u>
slow flow	high conductivity
frequent flooding	high dissolved organics
recent impoundment	pH 6.0 or >8.5
	high temperature
<u>Sediment characteristics</u>	<u>Life history factors</u>
mildly oxidizing sediments	large size
low clay content	long life span
high organic content	(high trophic position)
low complexing agents	

By limiting the composition of methylating bacteria, the sediment redox potential affects the rate of methylation. Although methylation has been observed in anaerobes, facultative anaerobes, and aerobes (Beijer and Jernelov, 1979), the optimal condition for methylmercury bioavailability is a mildly oxidizing environment (-100 mV to +150 mV) (Phillips et al., 1987). Therefore, reservoirs with oxygenated sediments are expected to have less methylmercury bioavailable than those with anoxic sediments, and reservoirs with oxygenated bottom waters for more of the year are expected to have higher levels of methylmercury.

Sediment organic content was not significantly different in the three reservoirs ( $p=0.212$ ). Therefore the influence of organic content on bioaccumulation in fish cannot be evaluated. However, Klein (1973) suggests that organic material may actually control mercury distribution. Corresponding to greater microbial density and available energy, high organic content stimulates methylmercury production (Jackson and Woychuk, 1980).

In addition, the supply of nutrients and complexing agents affect the rate of methylation. Nutrient availability influences both the density of methylating microorganisms and their metabolic rates (Furutani and Rudd, 1980). By binding mercury inertly, complexing agents (especially sulphides) affect the rate of methylation (Beijer and Jernelov, 1979). Thus, methylation rates are reduced in the presence of high concentrations of complexing agents. Berman and Bartha (1986a) reported a causal relationship between high sulphide levels and low methylation rates.

As methylation rates are largely determined by levels of oxygen, nutrients, and organic material in sediments, it is not surprising that methylmercury

bioavailability has been shown to exhibit seasonal variation (Korthals and Winfrey, 1987).

Several water chemistry parameters also influence the availability of mercury in surface waters and uptake by fish. These factors include conductivity, turbidity, total dissolved solids, pH, temperature and trophic state.

The conductivity in Cottage Grove Reservoir was significantly higher than that of Ochoco or Owyhee. This may serve to exacerbate the mercury problem in this reservoir. For 13 Ontario lakes, conductivity explained 54% of the total variation in mercury concentrations in the crayfish (Allard and Stokes, 1989).

The pH of all three reservoirs is between 7.7 and 8.6. This corresponds to a pH range that is not optimal for mercury bioaccumulation. The effect of pH on accumulation of mercury in fish varies with the range of pH. Acidic conditions favor the formation of monomethylmercury (Jernelov and Asell, 1975) but also allow more binding of the mercury to particulate matter which lowers evaporation and sedimentation (Schlinder et al, 1980). The consequences of the two processes is elevated methylmercury production and availability to aquatic organisms. At higher pH values (>8), the formation of dimethyl mercury ( $\text{CH}_3\text{HgCH}_3$ ) is favored, but conditions for the uptake of available monomethyl mercury is enhanced (Beijer and Jernelov, 1979). The combined effect of these forces is a two-phase response in uptake, peaking once between pH 5.5 to 6.5, and again at pH >8.5 (Phillips et al., 1987).

In addition to pH, water temperature may affect mercury availability. Higher water temperatures at the water-sediment interface enhance methylation rates (Phillips et al., 1987). The rate of uptake of mercury by fish also increases with higher water temperatures (Huckabee et al., 1979).

All three reservoirs are classified as mesotrophic or eutrophic. The effect of limnological trophic state has not yet been clearly established. Characterized by low nutrient concentrations, low primary productivity and high levels of dissolved oxygen, oligotrophic lakes are thought to generally have lower methylation and uptake rates because of the lower density of methylating bacteria, lower energy availability for metabolic activity and oxygen concentrations above the optimum for methylation (Phillips et al., 1987). Conversely, methylation rates should be higher under eutrophic conditions; however, Akielaszek and Haines (1981) argue that methylation rates are higher in oligotrophic conditions based on greater mercury availability because there is less organic matter for mercury complexation.

Food web structure and the position of a given species in the complex influences the bioaccumulation of methylmercury because diet can be a significant exposure route. The percent of accumulated mercury from food varies with species. Food contributed less than 15% of the accumulated mercury in rainbow trout (Phillips, 1975); while it contributed 41-63% in walleyes and 51-73% in white crappies (Phillips et al., 1987). Furthermore, as a fish changes its diet as it matures, patterns of accumulation as a function of age may reflect these dietary changes. This may partially

explain the differences in this relationship between largemouth bass in Cottage Grove Reservoir and smallmouth bass in Owyhee Reservoir.

It is evident that mercury bioaccumulation in fish represents a management problem in Cottage Grove and Owyhee reservoirs. Additional monitoring of Ochoco Reservoir at higher water levels may reveal a similar situation. Despite the general pattern of low mercury concentrations in water and sediment, older fish consistently have mercury burdens in muscle tissue that exceed the FDA limit for human consumption. This supports the importance of methylation and bioaccumulation as critical determinants of mercury levels in fish. The importance of ecoregion parameters in determining the behavior of mercury in reservoir systems and accumulation in fish appears to be overshadowed by differences in mercury deposits and mining activities.



## VI. LITERATURE CITED

- Akielaszek, J.J. and T.A. Haines. 1981. Mercury in the muscle tissue of fish from three northern Maine lakes. *Bull. Environ. Contam. Toxicol.* 27:201-208.
- Allard, M. and P.M. Stokes. 1989. Mercury in crayfish species from thirteen Ontario lakes in relation to water chemistry and smallmouth bass (Micropterus dolomieu) mercury. *Can. J. Fish. Aquat. Sci.* 46:1040-1046.
- Andren, A.W. and J. O. Nriagu. 1979. The global cycle of mercury. In: *The biogeochemistry of mercury in the environment*, edited by J.O. Nriagu. Elsevier Press, Holland, pp.1-22.
- Baldwin, E.M. 1976. *Geology of Oregon*, Edwards Brothers Inc., Ann Arbor, Michigan.
- Brooks, H. 1971. Quicksilver deposits in Oregon. Department of Geology and Mineral Industries, miscellaneous paper # 15.
- Beijer, K. and A. Jernelov. 1979. Methylation of mercury in aquatic environments. In: *The biogeochemistry of mercury in the environment*, edited by J.O. Nriagu. Elsevier Press, Holland, pp.203-210.
- Berman, M. and R. Bartha. 1986. Levels of chemical versus biological methylation of mercury in sediments. *Bull. Environ. Contam. Toxicol.* 36:401-404.
- Buhler, D., R. Reed, and R. Caldwell. 1984. As assessment of projected mercury levels in the proposed Galloway Dam project on the Weisser River, Idaho. Report submitted to the U.S. Army Corps of Engineers.
- Ferns, M.L. and R.V. Wilson. 1984. Mineral resources map of Oregon, GMS-36.
- Furutani A. and J.W.M. Rudd. 1980. Measurement of mercury methylation in lake water and sediment samples. *App. Environ. Microbiol.* 40:770-776.
- Gallant, A., T. Whittier, D. Larsen, J. Omernik and R. Hughes. 1989. Regionalization as a tool for managing environmental resources. EPA/600/3-89-060.
- Gray, J.J., M.N. Peterson, J. Clayton and G. Baxter. 1983. Geology and mineral resources of 18 BLM wilderness study areas, Harney and Malheur counties, Oregon. Oregon Department of Geology and Mineral Industries, open-file report, O-83-2.
- Hakanson, L. 1980. The quantitative impact of pH, bioproduction and Hg-contamination on the Hg-content of fish (pike). *Environ. Poll.* 18:285-304.
- Hill, S. 1973. Study of mercury and heavy metals pollutants in the Jordan Creek Drainage. Unpublished report, College of Mines, University of Idaho, Moscow, Idaho.
- Horton, R.C. 1964. Hotspring, sinter deposits, and volcanic cinder cones in Nevada. Nevada Bureau of Mines, map #25.

Huckabee, J.W., J.W. Elwood, and S.G. Hildebrand. 1979. Accumulation of mercury in freshwater biota. In: The biogeochemistry of mercury in the environment, edited by J.O. Nriagu. Elsevier Press, Holland, pp.277-325.

Jackson, T.A. and R.N. Woychuk. 1980. The geochemistry and distribution of mercury in the Wabigoon river system. In: Mercury pollution in the Wabigoon-English river system of northeastern Ontario, and possible remedial measures: a progress report, edited by T.A. Jackson, pp.1-28.

Jenne, E.A. 1973. Mercury in waters of the western U.S. In: Mercury in the western environment, edited by D.R. Buhler. Oregon State University, Corvallis, OR, pp.16-28.

Jernelov, A. and B. Asell. 1973. The feasibility of restoring mercury-contaminated waters. In: Heavy metals in the aquatic environment: an international conference, edited by P.A. Krenkel. Pergamon Press, New York, pp.299-309.

Johnson, D.M., R.R. Petersen, D.R. Lycan, J.W. Sweet and M.E. Neuhaus. 1985. Atlas of Oregon lakes. Oregon State University, Corvallis, Oregon.

Klein, D.H. 1973. Sources and present status of the mercury problem. In: Mercury in the western environment, edited by D.R. Buhler. Oregon State University, Corvallis, Oregon, pp.1-13.

Korthals, E.T. and M.R. Winfrey. 1987. Seasonal and spatial variations in mercury methylation and demethylation in an oligotrophic lake. Appl. Environ. Microbiol. 53(10):2397-2404.

Lawrence, E.F. and R.V. Wilson. 1962. Mercury occurrences in Nevada. Nevada Bureau of Mines, map #7.

Longbottom, J.E., R.C. Dressman, and J.J. Lichtenberg. 1973. Gas chromatographic determination of methyl mercury in fish, sediment, and water. J. Assoc. Offic. Analyt. Chem. 56(6):1297-1303.

Lowe, T.P., T.W. May, W.G. Brumbaugh and D.A. Kane. 1985. National contaminant biomonitoring program: concentrations of seven elements in freshwater fish, 1978-1981. Arch. Environ. Contam Toxicol. 14:363-388.

McMurty, M.J., D.L. Wales, W.A. Scheider, G.L. Beggs and P.E. Dimond. 1989. Relationship of mercury concentrations in lake trout (Salvelinus namaycush) and smallmouth bass (Micropterus dolomieu) to the physical and chemical characteristics of Ontario lakes. Can. J. Fish. Aquat. Sci. 46:426-434.

Nriagu, J.O. 1979. Production and uses of mercury. In: The biogeochemistry of mercury in the environment, edited by J.O. Nriagu. Elsevier Press, Holland, pp.23-40.

Omernik, J. and A. Gallant. 1986. Ecoregions of the Pacific Northwest. EPA/600/3-86-033.

Oregon Department of Geology and Mineral Industries. 1982. Geothermal resources of Oregon (map).

Phillips, G.R., P.A. Medvick, D.R. Skaar and D. Knight. 1987. Factors affecting the mobilization, transport, and bioavailability of mercury in reservoirs of the Upper Missouri River Basin. U.S. Department of the Interior, Fisheries and Wildlife Service, technical report #10.

Phillips, G.R. 1975. Some quantitative aspects of mercury accumulation by rainbow trout. PhD dissertation, Oregon State University.

Ramlal, P.S., J.W.M. Rudd and R.E. Hecky. 1986. Methods for measuring specific rates of mercury methylation and degradation and their use in determining factors controlling net rates of mercury methylation. Appl. Environ. Microbiol. 51:110-114.

Schindler, D.W., R.H. Hesslein, R. Wagemann and W.S. Broecker. 1980. Effects of acidification on mobilization of heavy metals and radionuclides from the sediments of a freshwater lake. Can. J. Fish. Aquat. Sci. 37:1723-1729.

Standard methods for the examination of water and wastewater. 1989. Published jointly by: American Public Health Association, American Water Works Association, and Water Pollution Control Federation, Washington, D.C.

State Water Resources Board, Oregon. 1974. Oregon drainage basins, map #0.2

State Water Resources Board, Oregon. 1960a. Deschutes drainage basin- mineral deposits, map #5.65.

State Water Resources Board, Oregon. 1960b. Upper Willamette drainage basin- mineral deposits, map #2A.65.

Uthe, J.F., J. Solomon, and B. Grift. 1972. Rapid semi-micro method for the determination of methyl mercury in fish tissue. J. Assoc. Offic. Analyt. Chem. 55(3): 583-589.

Wiklander, L. 1969. The content of mercury in Swedish groundwater and river water. Geoderma 3:75-79.

Worcester, T.C. 1979. Mercury accumulation in fish from Cottage Grove Reservoir and its tributaries. M.S. thesis, Oregon State University.