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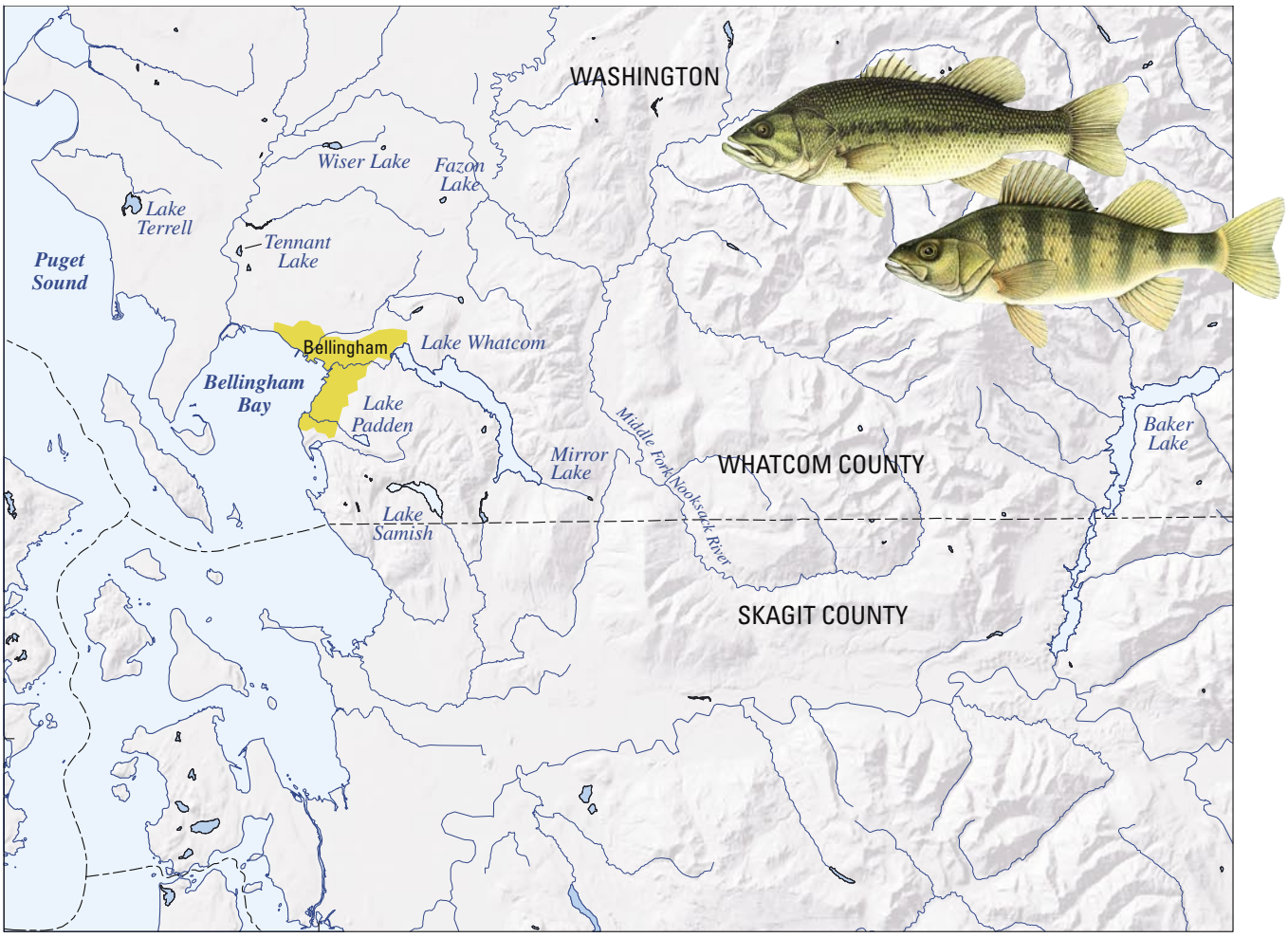
Prepared in cooperation with the Whatcom County Health Department, and the Washington State Department of Ecology

Sources of Mercury in Sediments, Water, and Fish of the Lakes of Whatcom County, Washington



Scientific Investigations Report 2004–5084

U.S. Department of the Interior
U.S. Geological Survey

Cover: Illustrations of largemouth bass, and yellow perch have been supplied courtesy of the Ohio State Environmental Protection Agency.

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By Anthony J. Paulson

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U.S. Department of the Interior
U.S. Geological Survey

U.S. Department of the Interior
Gale A. Norton, Secretary

U.S. Geological Survey
Charles G. Groat, Director

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Conversion Factors, Datums, and Abbreviations

CONVERSION FACTORS

Metric to Inch-Pound		
Multiply	**By**	**To obtain**
	Length	
centimeter (cm)	0.3937	inch
millimeter (mm)	0.03937	inch
meter (m)	3.281	foot
kilometer (km)	0.6214	mile
	Area	
square meter (m^2)	10.76	square foot
square centimeter (cm^2)	0.1550	square inch
	0.001076	square foot
square kilometer (km^2)	0.3861	square mile
	247.1	acre
	Volume	
cubic meter (m^3)	264.2	gallon
liter (L)	0.2642	gallon
	Flow rate	
liter per year (L/yr)	0.2642	gallon per year
	Mass	
gram (g)	0.03527	ounce, avoirdupois
gram per hour (g/hr)	0.03527	ounce per hour
kilogram (kg)	2.205	pound, avoirdupois
kilogram (kg/d)	2.205	pound per day
megagram (Mg)	1.102	ton, short
	Loads	
gram per hour (g/hr)	0.0529	pound per day
	19.295	pound per year
Inch-Pound to Metric		
ton per hour (ton/hr)	0.9072	metric ton per hour
mile (mi)	1.609	kilometer
acre	0.4047	hectare
cubic foot per second (ft^3/s)	0.02832	cubic meter per second
pound, avoirdupois (lb)	0.4536	kilogram
pound per year (lb /yr)	0.4536	kilogram per year
ton, short (2,000 lb)	0.9072	megagram (Mg)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$°F=1.8\ °C+32.$$

DATUMS

Vertical coordinate information is referenced to NGVD29; horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Altitude, as used in this report, refers to distance above the vertical datum.

ABBREVIATIONS

dscm	dry standard cubic meter
g/kg	gram per kilogram
g/ton	gram per ton
µg/L	microgram per liter
µg/g	microgram per gram
µg/kg	microgram per kilogram
$(\mu g/m^2)/yr$	microgram per square meter per year
mg/L	milligram per liter
(mg/kg)	milligram per kilogram
$(mg/cm^2)/yr$	milligram per square centimeter per year
ng/L	nanogram per liter
ng/mg	nanogram per milligram
ng/m^3	nanogram per cubic meter
ng/cm^2	nanogram per square centimeter
$(ng/cm^2)/yr$	nanogram per square centimeter per year
pCi/cm^2	picocuries per square centimeter

Sources of Mercury in Sediments, Water, and Fish of the Lakes of Whatcom County, Washington

By Anthony J. Paulson

Abstract

Concerns about mercury (Hg) contamination in Lake Whatcom, Washington, were raised in the late 1990s after a watershed protection survey reported elevated concentrations of Hg in smallmouth bass. The U.S. Geological Survey, the Whatcom County Health Department, and the Washington State Department of Ecology (Ecology) cooperated to develop a study to review existing data and collect new data that would lead to a better understanding of Hg deposition to Lake Whatcom and other lakes in Whatcom County, Washington.

A simple atmospheric deposition model was developed that allowed comparisons of the deposition of Hg to the surfaces of each lake. Estimates of Hg deposition derived from the model indicated that the most significant deposition of Hg would have occurred to the lakes north of the City of Bellingham. These lakes were in the primary wind pattern of two municipal waste incinerators. Of all the lakes examined, basin 1 of Lake Whatcom would have been most affected by the Hg emissions from the chlor-alkali plant and the municipal sewage-sludge incinerator in the City of Bellingham. The length-adjusted concentrations of Hg in largemouth and smallmouth bass were not related to estimated deposition rates of Hg to the lakes from local atmospheric sources.

Total Hg concentrations in the surface sediments of Lake Whatcom are affected by the sedimentation of fine-grained particles, whereas organic carbon regulates the concentration of methyl-Hg in the surface sediments of the lake. Hg concentrations in dated sediment core samples indicate that increases in Hg sedimentation were largest during the first half of the 20th century. Increases in Hg sedimentation were smaller after the chlor-alkali plant and the incinerators began operating between 1964 and 1984. Analysis of sediments recently deposited in basin 1 of Lake Whatcom, Lake Terrell, and Lake Samish indicates a decrease in Hg sedimentation.

Concentrations of Hg in Seattle precipitation and in tributary waters were used to calculate current (2002-03) loadings of Hg to Lake Whatcom. Hg in tributaries contributed 59 percent of the total Hg, whereas non-local atmospheric deposition was estimated to have contributed 41 percent of the 303 grams of Hg entering Lake Whatcom each year. However, these inputs cannot be verified without a better understanding of the sources of sediment to Lake Whatcom.

Introduction

Lake Whatcom is a large, natural lake in Whatcom County, Washington, near the western edge of the Cascade Range foothills (fig. 1). The lake is a source of drinking water for about 86,000 Whatcom County residents and is used for sport fishing, swimming, and other types of recreation. Concerns about mercury (Hg) contamination in Lake Whatcom were raised in the late 1990s after a watershed protection survey (Serdar and others, 1999) reported a concentration of 0.50 µg/g (microgram per gram, wet weight) in a composite sample of smallmouth bass fillets. For comparison, the national average concentration of Hg is 0.36 µg/g for sport fish fillets, and 0.34 µg/g for smallmouth bass (U.S. Environmental Protection Agency, 1992). Hg concentrations in sediment samples also were higher than the silt-clay fraction of reference sediments from the Puget Sound Basin (Serdar and others, 1999).

To address concerns about possible Hg contamination in Lake Whatcom, Whatcom County Health and Human Services Department and the Washington State Departments of Health, Ecology, and Fish and Wildlife conducted a second study to help determine if consumers of Lake Whatcom fish were at risk from Hg exposure (Serdar and others, 2001). The second study confirmed that Hg concentrations in smallmouth bass from Lake Whatcom were elevated relative to the national average concentration. The average Hg concentration was 0.49 µg/g, whereas the maximum concentration was 1.84 µg/g. These concentrations can be compared to the tissue-residue criterion of 0.3 µg/g methyl-Hg issued recently by the U.S. Environmental Protection Agency (2001).

Environmental managers require information on the current loadings of Hg to Lake Whatcom in order to focus any future action in an effective manner. Sources of Hg in Lake Whatcom are thought to include atmospheric deposition from global and local sources, discharges from tributaries (including the diversion from the Nooksack River), landfills, dumpsites, and local mining operations. Local interest has focused on a chlor-alkali plant that operated in the City of Bellingham and emitted mercury to the atmosphere from the early 1960s until 2000. There has been particular interest in assessing the rate and speciation of emissions from the chlor-alkali plant and the resulting atmospheric deposition of Hg to Lake Whatcom.

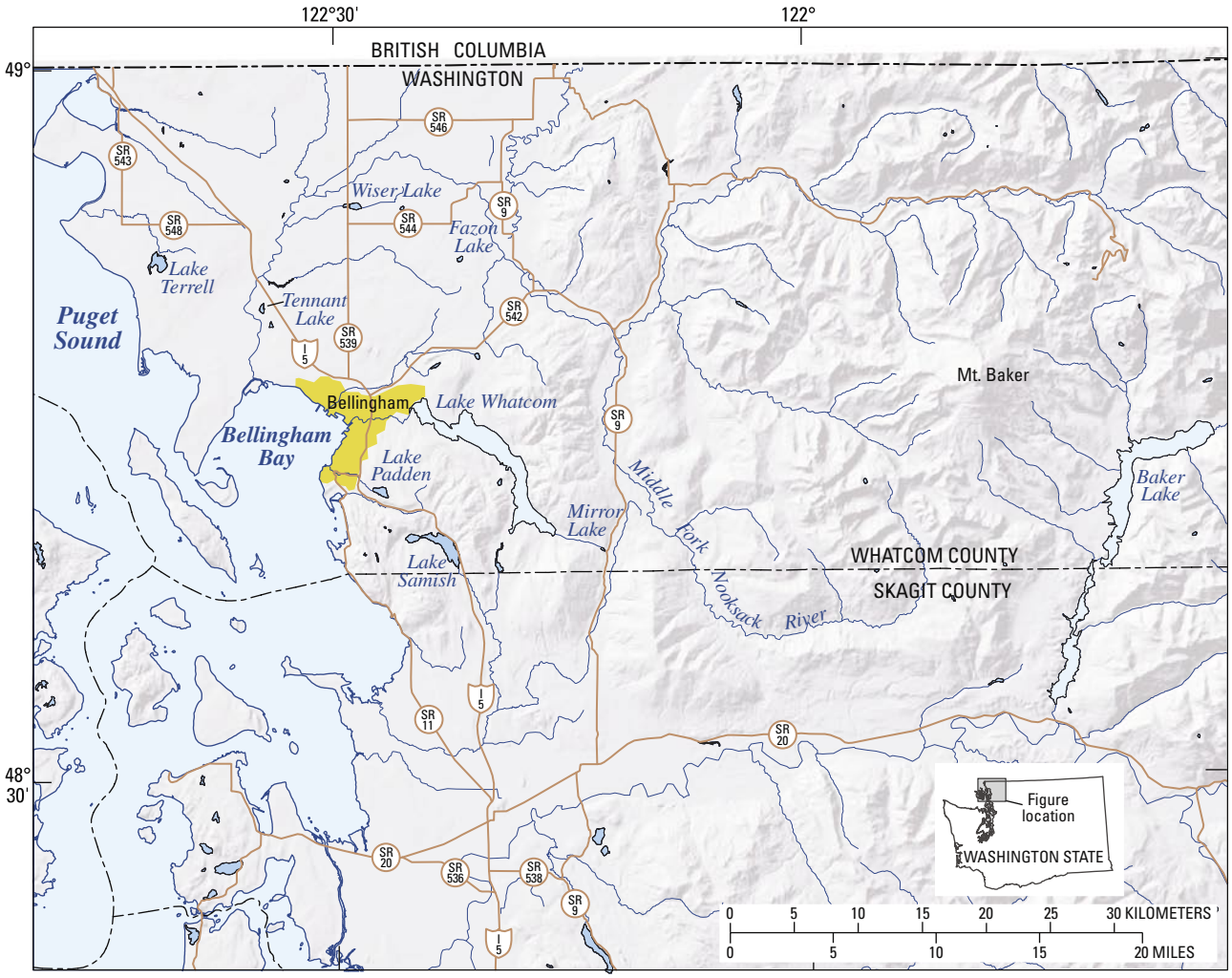

Figure 1. Location of study area and lakes in Whatcom County, Washington.

Purpose and Scope

In 2002, the Whatcom County Health Department (WCHD) contacted the U.S. Geological Survey (USGS) to evaluate existing data, with the broad objectives of determining the presence of Hg in Lake Whatcom and its possible sources. During discussions on the scope of the project, USGS became aware that the Washington State Department of Ecology (Ecology) was planning a survey of surface sediments of Lake Whatcom for Hg and also would be collecting three sediment cores in Lake Whatcom to determine the historical sedimentation of Hg in the lake. In addition, Ecology was planning to conduct year-long water sampling in 10 tributaries to Lake Whatcom for the analysis of total Hg and ancillary physical parameters (pH, specific conductance, and total suspended solids) and a statewide survey of Hg in sports fish that included several lakes in Whatcom County.

As both USGS and Ecology began to plan their efforts to understand the sources, transformation, and fate of Hg in Lake Whatcom, the agencies decided to coordinate sampling efforts. A joint Quality Assurance Project Plan (QAPP) was developed in which USGS personnel were to assist Ecology in the collection of surface-sediment samples and sediment cores in Lake Whatcom (Norton, 2002). With the efficiencies gained from the joint effort, the USGS was able to collect a core from each of five additional lakes in Whatcom County in order to assess the regional deposition of Hg in lake sediments. WCHD contracted with Ecology to analyze the USGS cores from the five other lakes in order to obtain analytical data that were consistent with the cores from Lake Whatcom. In addition, fish tissue samples were analyzed for Hg concentrations in a variety of fish species collected from five other lakes in Whatcom County.

The overall goal of the project reported here was to determine the potential sources of Hg to Lake Whatcom and other lakes in Whatcom County and the transformation and fate of Hg in Lake Whatcom. The more specific objectives for this project were as follows:

1. Use differences in Hg accumulation rates in other lakes in Whatcom County along with other data to infer the Hg deposition from known local and regional sources of airborne mercury and to determine if accumulation rates can be attributed to global sources.
2. Use available data on concentrations of Hg in the tissue of fish from Lake Whatcom and other lakes in Whatcom County to help identify sources of mercury.
3. Estimate the relative importance of potential sources of Hg in Lake Whatcom tributaries and in water diverted from the Middle Fork of the Nooksack River by examining the relation between concentrations of Hg in surface sediments of the lake and the proximity of potential sources.
4. Investigate the possibility that geochemical, biological, and physical processes occurring in Lake Whatcom or in connected wetlands affect the distribution of Hg in sediment and fish tissue.
5. Suggest additional sampling or other information needed to confirm Hg sources and refine the understanding of processes identified after completing the first four objectives.

Acknowledgments

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- Dean Momohara, Randy Knox, Sara Sekerak, Sally Cull, and Meredith Jones of the Ecology Manchester Laboratory conducted the analysis of metals and conventional constituents.
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- Karen Payne (USGS) and Dale Norton, Randy Coots, Brandee Era-Miller, Morgan Roose, and Dave Serdar (Ecology) assisted in collection of the sediment and core samples under a wide range of weather conditions.
- Joan Vandersypen (Western Washington University) provided flow data on several of the tributaries and sediment data on Lake Whatcom, and Peg Wendling (City of Bellingham) provided data on the operations of the Nooksack Diversion and the municipal sewage sludge incinerator.
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- Theresa Olsen (USGS) and Randy Coots (Ecology) provided GIS technical assistance.
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- Karen Payne (USGS) managed spreadsheets for initial compilation and interpretations of the data used in this report.
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Description of Study Area

The surface area of Lake Whatcom covers 20.3 km^2, and the total drainage area is about 142 km^2 (table 1), with more than 3,600 housing units, of which about 785 have on-site septic systems (Morris Arthur, Whatcom County Health Department, oral commun., 2003). The lake is divided into three distinct basins by glacial sills. Basins 1 and 2 (fig. 2) are relatively small and shallow (maximum depths of 29 and 21 m, respectively). Basin 1 is in the most urbanized part of the drainage basin, lying largely within the city limits of Bellingham, and therefore has few on-site septic systems along the shoreline (less than 50 m from the lake). In contrast, most of the 198 on-site septic units within 50 m of the lake are along the shore of basin 3 between Olsen and Smith Creeks, along South Bay to Brannian Creek, and near the outflow of Blue Canyon Creek (Morris Arthur, Whatcom County Health Department, oral commun., 2003). Basin 3, which has a maximum depth of about 103 m, contains 96 percent of the lake volume. Several streams drain to Lake Whatcom, and to maintain optimal lake levels, water is diverted to the lake from the Middle Fork Nooksack River.

After Lake Whatcom, Lake Samish and Baker Lake were the largest and deepest of the lakes sampled within Whatcom County (fig. 1). Like Lake Whatcom, Lake Samish has a highly developed shoreline. In addition, a major interstate highway passes adjacent to Lake Samish. Small perennial streams that drain into Lake Samish provide only a portion of the outflow of the lake. In contrast, the water balance for Baker Lake is dominated by flow from 13 creeks draining 769.2 km^2. Fazon Lake, Wiser Lake, and Lake Terrell (referred to as the three northern lakes) are small catchment lakes north of Bellingham, are surrounded primarily by agricultural and forest lands, and have little channelized surface-water inflow. Wiser Lake is more developed than either Lake Terrell or Fazon Lake, and a major arterial street that bisects Wiser Lake permits only limited water exchange between the two parts. In contrast, Lake Terrell is part of a wildlife refuge and its surface area comprises 24 percent of the area of the drainage basin. Tennant Lake also is surrounded by a wildlife refuge. The land around Lake Padden (south of Bellingham) has been developed as a park and golf course.

Table 1. Statistics on lakes in Whatcom County, Washington

[**Source of data:** WCP&DS, Whatcom County Planning and Development Services. km^2, square kilometer; m, meter; m^3, cubic meter. NA, not applicable; –, no data]

Lake	Lake area (km^2)	Drainage area (km^2)	Watershed use (percentage of total)				Shoreline percentage developed	Number of homes
			Lake	Agriculture	Forest	Urban/ suburban		
Whatcom								
Basin 1	2.1	–	–	–	–	–	–	–
Basin 2	1.6	–	–	–	–	–	–	–
Basin 3	16.6	–	–	–	–	–	–	–
Total	20.3	142.0	14	1.3	78	7	–	–
Terrell	1.8	7.4	–	–	–	–	0	NA
	–	[1]33.7	7	50	36	7	–	–
Wiser	.4	9.7	–	–	–	–	47	30
	–	[1]32.3	1.9	80.2	11.7	6	–	–
Fazon	.1	2.5	–	–	–	–	0	NA
		[1]8.7	2.2	42	48	7		
Samish–east arm	2.8	23.8	–	–	–	–	91	171
Total for both arms	88.2	[1]95	4	6	88	3	–	–
Baker	14.6	769.2	–	–	–	–	–	–
Tennant	.1	2.4	5	78	17	0	0	NA
Padden	.6	6.8	–	–	–	–	0	NA
	–	[1]15.6	4.2	1.1	49	45	–	–

Lake	Depth (m)		Volume (10^7 m^3)	Inflows	Outflows	Source of data
	Median	Maximum				
Whatcom						
Basin 1	9	29	1.94	3 gaged	one	Pelletier, 1998
Basin 2	11	21	1.80	none	–	Pelletier, 1998
Basin 3	54	103	88.4	6 gaged; 1 ungaged	–	Pelletier, 1998
Total	–	–	–	–	–	WCP&DS, written commun., 2004
Terrell	2	3	.37	intermittent	one ungaged	Bortleson and others, 1976
	–	–	–	–	–	WCP&DS, written commun., 2004
Wiser	2	3	.075	intermittent	one	Bortleson and others, 1976
	–	–	–	–	–	WCP&DS, written commun., 2004
Fazon	3	5	.037	none	none	Bortleson and others, 1976
Samish–east arm	9	23	2.94	perennial	one	Bortleson and others, 1976
Total for both arms	–	–	–	–	–	WCP&DS, written commun., 2004
Baker	–	86		13	–	Puget Sound Energy, 2002
Tennant	.9	1.8	.011	1 ungaged	–	Dion and others, 1976
Padden	8	18	.53	intermittent	one	Bortleson and others, 1976
	–	–	–	–	–	WCP&DS, written commun., 2004

[1]Drainage area calculations from WCP&DS include drainages downstream of lake.

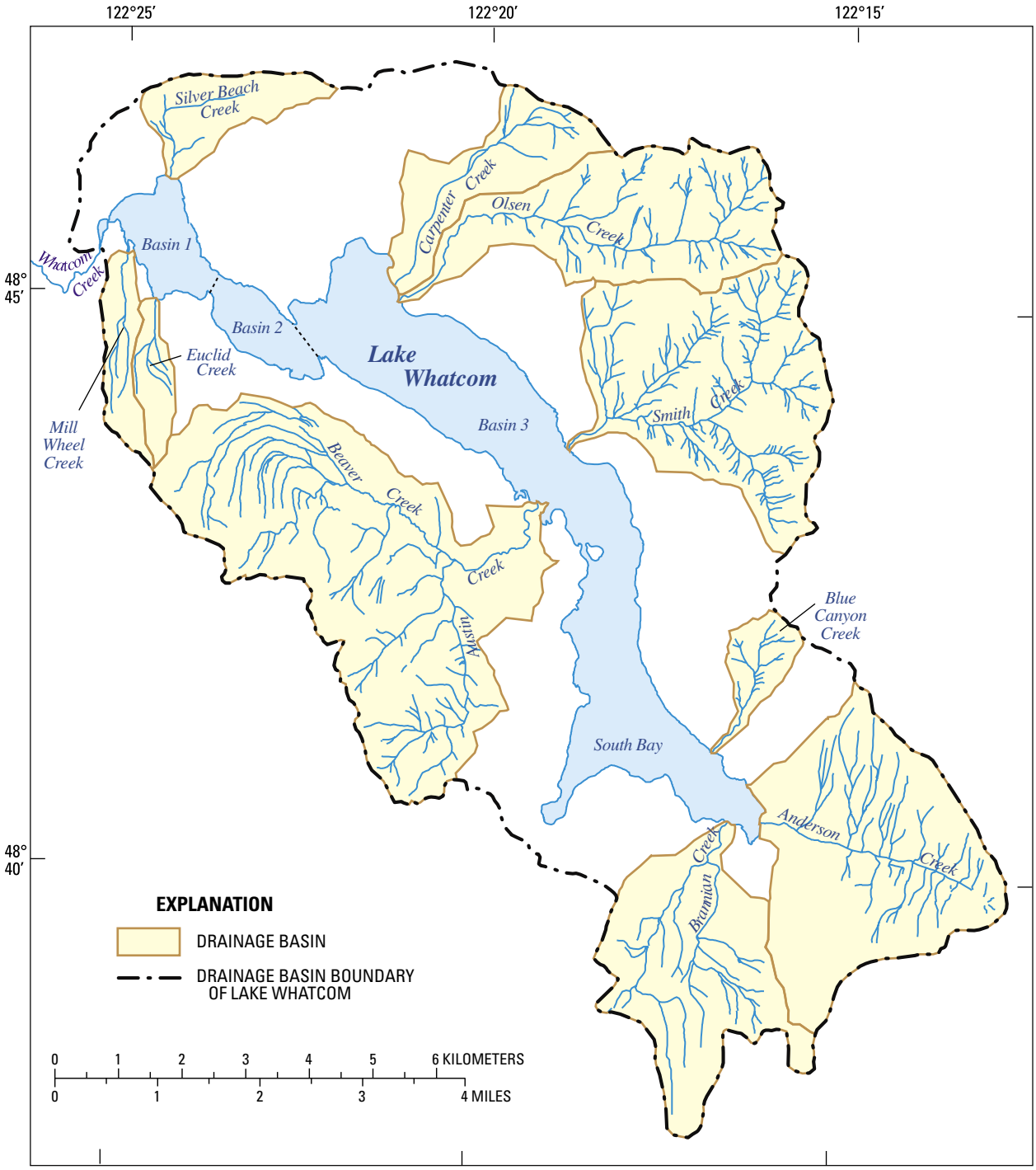


Figure 2. Location of the basins of Lake Whatcom and its tributaries, Whatcom County, Washington (from Norton, 2004).

Study Methods

Methods for analyses of Hg in fish reported in this document are described in appendix A (at back of report). Most data presented in this report were obtained from previous studies. All data related to Hg sources or existing conditions of fish, water, and sediment in the lakes of Whatcom County used in this report were assessed for the quality of sampling, processing, and analytical procedures. The results of those quality-assurance assessments also are included in appendix A.

Evaluation of Possible Sources of Mercury to Lake Whatcom and Other Lakes in Whatcom County

Background inputs of aqueous Hg to any body of water are determined by deposition from the global reservoir of atmospheric Hg. Deposition from local natural and anthropogenic sources of atmospheric Hg is added to this global deposition. Atmospheric deposition of Hg to land surfaces also serves as the initial source of Hg from surface- and ground-water inputs of Hg. As precipitation containing Hg percolates through the aquifer, the Hg can be sequestered by organic matter or inorganic minerals. In contrast, additional Hg can be released from natural aquifer materials. Anthropogenic sources of Hg in ground water could include historical application of pesticides on lawns and agricultural lands, release from dumps and landfills, or discharges from septic systems. Direct precipitation and ground water that contain both natural and anthropogenic Hg combine to form surface waters. Particulate matter that contains Hg is added to these surface waters by runoff from roads and buildings and by processes that erode natural geologic formations. In addition, precipitation washes various surfaces that may contain Hg, such as paint on residential siding, driveways, roads, lawns, and motor vehicles.

Atmospheric Inputs

Deposition of Mercury from Non-Local Atmospheric Sources

Accurate, direct measurement of Hg in precipitation has been achieved only in the last decade, with the advent of ultra-trace analytical techniques combined with the sensitive cold-vapor atomic fluoresce spectrometry (CVAFS) analysis. Such data are not available for Hg concentrations in precipitation or in dryfall in Whatcom County. In 1997, the first Pacific Northwest precipitation sampler in the national Mercury Deposition Network (MDN) was installed at a site located in an open area on the National Oceanic and Atmospheric Administration Sand Point facility in Seattle, Washington. Annual deposition of total Hg was fairly constant at 6.2 μg/m^2 between 1998 and 2002 (National Atmospheric Deposition Program, 1999, 2000, 2001, 2002, 2003). The annual precipitation-averaged concentration of Hg in precipitation ranged from about 6 to 10 nanograms per liter (ng/L) during 1998-2002 (table 2). In 2001, a precipitation sampler was installed on Reifle Island, British Columbia, and

Table 2. Measured deposition of atmospheric mercury (Hg) in the Pacific Northwest, 1997-2002

[To convert annual deposition to ng/cm^2 multiply by 0.1. ng/cm^2, nanogram per square centimeter; μg/m^2, microgram per square meter; ng/L, nanogram per liter]

Year	Annual deposition of Hg (μg/m^2)	Precipitation-averaged concentration of Hg in precipitation (ng/L)	Source of data
Seattle, Washington			
1997	17.3	18.3	National Atmospheric Deposition Program, 1998
1998	5.3	5.9	National Atmospheric Deposition Program, 1999
1999	8.0	8.5	National Atmospheric Deposition Program, 2000
2000	6.2	9.8	National Atmospheric Deposition Program, 2001
2001	5.9	6.3	National Atmospheric Deposition Program, 2002
2002	5.7	9.1	National Atmospheric Deposition Program, 2003
Reifle Island, British Columbia			
2001	5.5	6.0	National Atmospheric Deposition Program, 2002
2002	3.2	4.8	National Atmospheric Deposition Program, 2003

data collected indicated an annual Hg deposition and annual precipitation-averaged Hg concentration similar to that at the Seattle sampling site. The deposition of Hg at the Seattle and Reifle Island sites represents Pacific Northwest deposition plus deposition from sources local to the two sites. A Hg-deposition value of 6.2 ($\mu g/m^2$)/yr is used as an upper limit for regional deposition in the Pacific Northwest.

The annual Hg deposition at the Seattle sampling site (17.3 $\mu g/m^2$) during 1997 (National Atmospheric Deposition Program, 1998) was considerably higher than the annual Hg deposition from 1998 to 2002. The higher annual Hg deposition and precipitation-averaged concentration may have been an artifact of the initial start-up of the precipitation sampler in 1997, or may have been due to the general decrease in Hg deposition with time observed around the world. Hg deposition in North America is thought to be decreasing. Hg in ice cores from the Upper Fremont Glacier in Wyoming (fig. 3) peaked in 1984 (Schuster and others, 2002), whereas deposition in upper Midwest peat bogs peaked in the 1960s (Benoit and others, 1994). Hg concentrations in recently accumulated ice in Greenland also were only 60 percent of the Hg concentrations in ice that accumulated between 1940 and 1960 (Boutron and others, 1998).

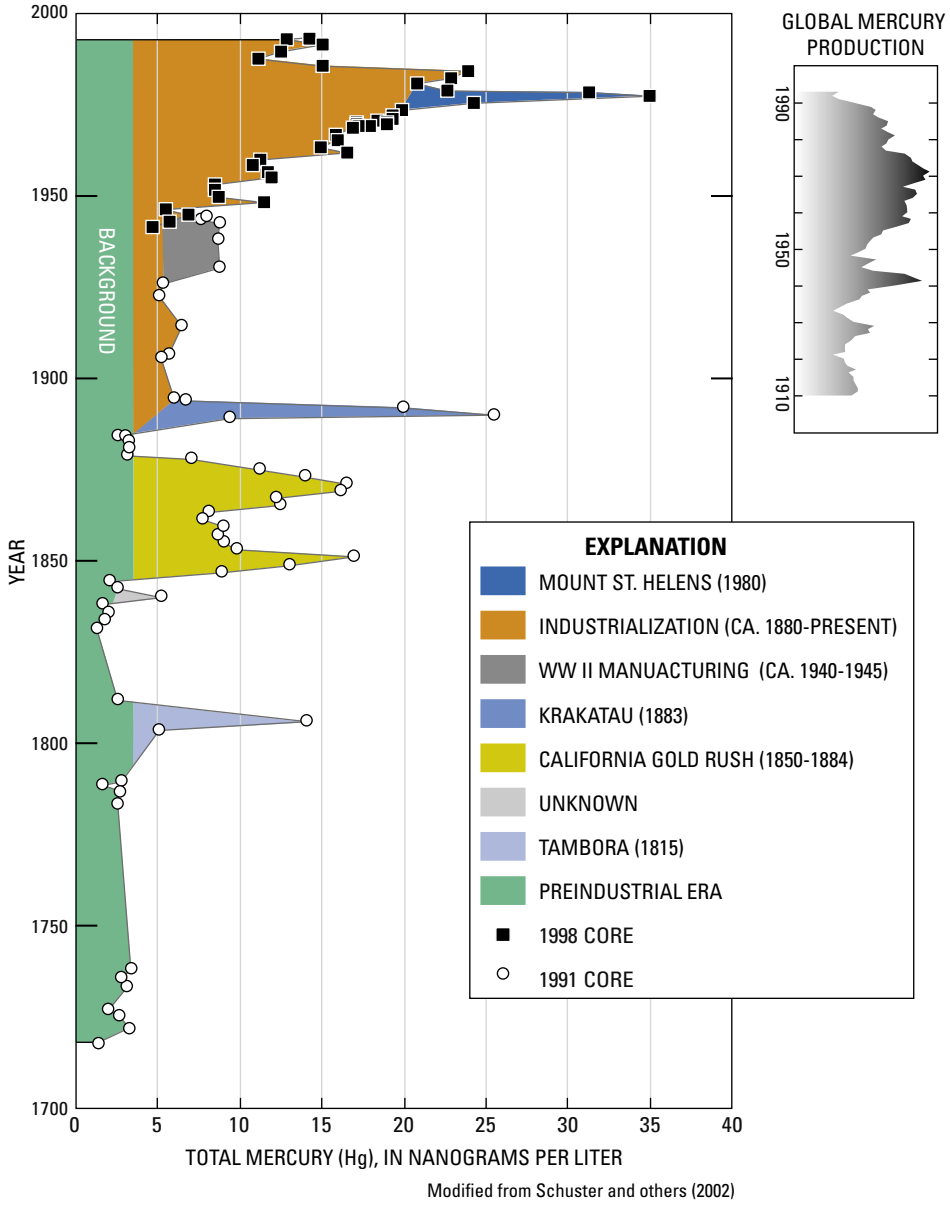


Figure 3. Historical concentrations of mercury in ice core from Upper Fremont Glacier, Wyoming.

Local Atmospheric Sources and Total Mercury Emission Rates

Municipal and Sludge Incinerators

Nationwide, utility and commercial boilers that burn coal and municipal and medical waste incinerators contribute about 80 percent of the total Hg loading to the atmosphere (U.S. Environmental Protection Agency, 1997a). Between 1974 and 1997, one municipal waste incinerator operated continuously within Whatcom County and a second municipal incinerator operated intermittently from 1984 to 1997. A third incinerator operated in Skagit County, to the south of Whatcom County, and its deposition of Hg is considered part of the regional deposition. Municipal solid waste contains mercury originating from common household items, with household batteries being the major contributor (87 percent in 1989). By 1991, Hg content in all batteries, except button or "coin" cells, had decreased from about 1 to 0.025 percentage by weight. The Mercury-Containing and Rechargeable Battery Management Act was intended to eliminate Hg disposal in municipal solid waste after 1996. Similarly, the Mercury Chemical Action Plan (Peele, 2003) describes elements of a statewide campaign "to virtually eliminate the use and release of human-caused mercury in Washington State and take steps to further minimize human exposure to mercury." Hg content in paint residues is thought to have peaked around 1975 and decreased to insignificant levels by 2000, as a result of the 1990s ban of Hg in interior and exterior paints. Other sources of Hg in municipal solid waste include electrical equipment, fluorescent bulbs, personal thermometers, thermostats, dental supplies, and special paper coatings.

The Thermal Reduction Company, Inc. of Ferndale, Washington (also known as Recomp, Inc.) started incinerating municipal and medical waste in 1974 (fig. 4) with eight modular incinerators that provided little pollution control (Northwest Air Pollution Authority, Mount Vernon, Washington, written commun., 2003). The results of a source emissions test in 1985 indicated that the facility was not in compliance for emissions of particulate matter. In 1986, the eight modular units were replaced with two CS 2000 incinerator units equipped with an electrostatic precipitator as the primary pollution-control device. On December 15, 1988, three 2-hour emissions tests were conducted at a burn rate of about 4 to 5 tons of waste per hour, with an exit temperature of 255 °C at the electrostatic precipitator. Hg emissions averaged 39.7 g/hr (table 3) and between 0.002 and 0.065 percent was captured by a filter (AM Test, Inc., Redmond, Washington, written commun., 1989). The results of this test indicated an average Hg specific-emission release of 9.1 g/metric ton of solids burned. In 1993, the air-pollution control device on the two-unit incinerator was modernized to include a dry sorbent scrubber equipped with a baghouse. Emission tests were not conducted between the time the new devices were installed and when the incinerator stopped reporting operational data in 1997.

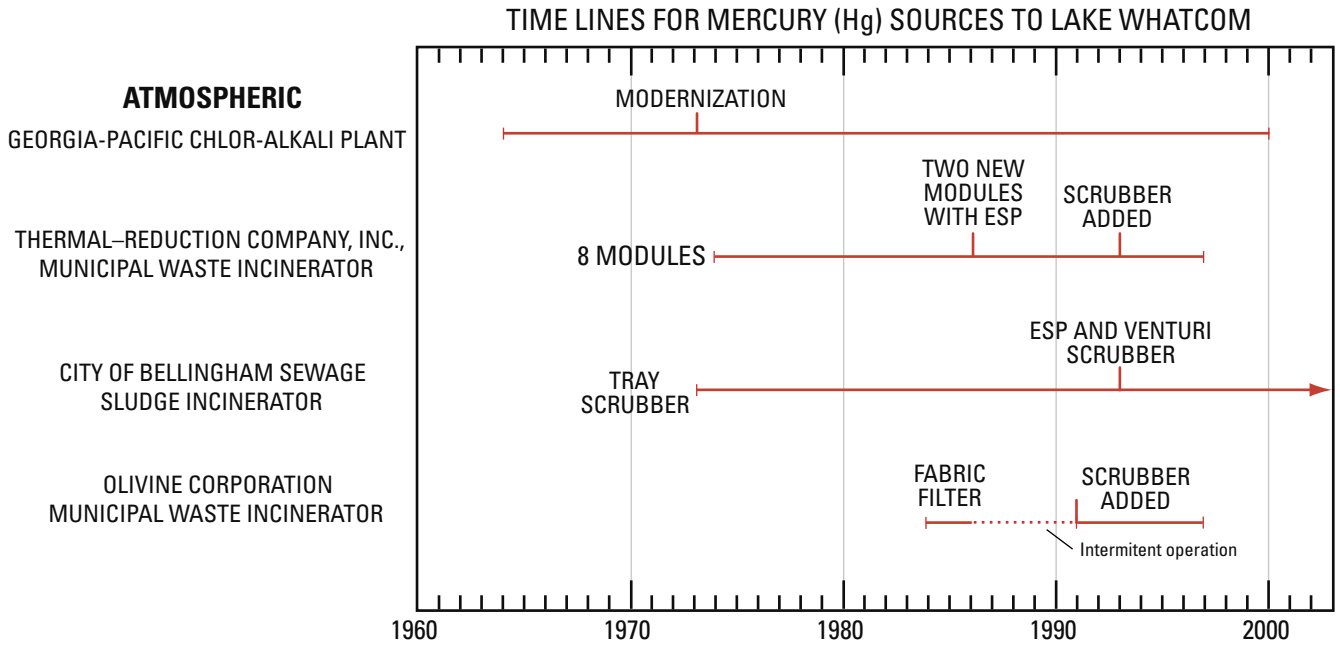


Figure 4. Timelines of local atmospheric sources of mercury.

Table 3. Rate of emission of mercury (Hg) and other characteristics related to local sources of mercury in Whatcom County, Washington

[**Annual tonnage:** Incinerators is in tons of waste burned; chlor-alkali plants is in tons of chlorine produced. **Mercury emission:** Values in parentheses () are estimates described in text section "Estimates of Mercury Deposition to Lakes in Whatcom County from Known Sources." **Form of emitted mercury**: RGM, reactive gaseous mercury. g/hr**,** gram per hour. NA, not analyzed; –, unknown]

Source	Year	Annual tonnage	Temperature	Mercury emission (g/hr)	Form of emitted mercury (particulate:RGM: vaporous)	References and notes
Incinerators						
Thermal Reduction Company, Inc. (municipal waste)	1974-96	–	–	(39.7)	[5:65:30]	Emission from 1989 test, 5 percent particulate Hg based on particulate violations
	1986-92	78,500	256	39.7	[0:70:30]	AM Test, Inc., Redmond, Washington, written comm., 1989
	1992-97	76,900	–	(19)	[0:62:38]	Estimated from reductions in RGM and vaporous Hg (van Velzen and others, 2002)
Olivine Corporation (municipal waste)	1984-90	31,500	–	(25)	[10:60:30]	Hg emissions estimated from Thermal Reduction Company's emission factor
	1991-97	43,600	–	(25)	[0:62:38]	Estimated from reductions in RGM and vaporous Hg (van Velzen and others, 2002)
City of Bellingham (municipal sewage sludge)	1974-92	–	–	(4.5)	–	Estimated from reductions in RGM and vaporous Hg (van Velzen and others, 2002)
	1993	–	–	.0037	Particulate emission = 0.2 percent of total	City of Bellingham, written commun., 1993
	1993	9,550	–	3.4	–	AmTest-Air Quality, Inc., Preston, Washington, written commun., 1994
	1993-2002	6,825	–	2.4	–	Above test and five days a week operation
	2003	3,622	–	1.16	–	Yearly average based on 33 percent usage reported in 2002
	2003	–	–	–	[0.2:10: 90]	Speciation based on estimated chlorine concentration
Chlor-Alkali plant						
Georgia-Pacific Hypo Stack	1975	–	–	5.7	[0:100:0]	Georgia-Pacific, Inc., written comm., 1975
	1981	79,200	9	.0139	[0:100:0]	Chris Lee, Georgia-Pacific, Inc., written comm., 1981
	1995	76,700	15	<.0003	[0:100:0]	AmTest-Air Quality, Inc., written comm., 1995
Georgia-Pacific hydrogen steam from chlor-alkali plant	1975	–	–	17.9	Unknown	Georgia-Pacific, Inc., written comm., 1975
	1981	79,200	9	14.8	–	Chris Lee, Georgia-Pacific, Inc., written comm., 1981
	1987	90,200	9	17.4	–	Ed Dahlgren, Georgia-Pacific, Inc., written comm., 1988
	1995	76,700	15	22.5	–	AmTest-Air Quality, written comm., 1995
Fugitive emissions						
Sweden		82,300	Ambient	10.5	[0.05:1:99]	Wangberg and others, 2003.
North Carolina		–	–	21	[NA:2:98]	Kinsey, 2003; Landis and others, 2004
Refinery						
British Petroleum, Cherry Point	2002	–	–	0.47	–	Sally Otterson, Ecology, Redmond, Washington, written commun., 2004

Olivine Corporation, located on Thomas Road to the east of the Bellingham Airport, received conditional approval to operate a 50-ton-per-day incinerator in 1984. Olivine Corporation received a number of extensions of their conditional approval to operate a municipal waste incinerator and operated it intermittently through 1990 (reported annual hours of operation ranged from 3,650 to 4,050). By March 4, 1991, a dry sorbent scrubber with a baghouse had been installed at the incinerator. Operational data were not available for 1988, 1989, and 1993, and Olivine Corporation stopped reporting operational data to the Northwest Air Pollution Authority in 1998. Hg emission tests were not conducted at this incinerator before or after the installation of the dry scrubbers.

The City of Bellingham began incinerating the sludge from its Post Point wastewater-treatment plant onsite in 1973. The primary air pollution control device for this incinerator was a tray scrubber, in which water is dripped into the stream of exiting gases from the incinerator. With the expansion of the wastewater-treatment plant in 1993, the air-pollution devices were upgraded to include a wet Venturi scrubber and an electrostatic precipitator (Larry Bateman, City of Bellingham, oral commun., 2003). On November 4 and 5, 1993, an atmospheric Hg emission rate was 3.4 g/hr at the Post Point wastewater-treatment plant. Simultaneous with the emissions test, the Hg content of the sludge being incinerated was measured (Am Test-Air Quality, Inc., Preston, Washington, written commun., 1994). Hg content of the sludge is measured bimonthly and the Hg emissions rates are reported annually, based on the 1993 emissions test adjusted for changes in the Hg content of the sludge relative to the 1993 sludge measurements. The average emission rate for 2002 reported to Ecology was 0.31 g/hr (Sally Otterson, Washington State Department of Ecology, oral commun., 2004).

Chlor-Alkali Plant

Chlor-alkali plants, such as the Georgia-Pacific plant in downtown Bellingham, are known to be significant sources of atmospheric Hg, and a number of studies have documented elevated concentrations of Hg in the air, soils, and vegetation near chlor-alkali plants (Maserti and Ferrara, 1991; Ebinghaus and Kruger, 1996; and Lodenius, 1998). Hg concentrations in soils were elevated by a factor of six near the boundary of an Italian chlor-alkali plant compared to background soils (Maserti and Ferrara, 1991). Concentrations of Hg in leaves of bushes near the chlor-alkali plant were correlated with soil Hg concentrations. Hg concentrations in air decreased with distance from the plant, and the authors state, "Typical background values (3 to 5 ng/m^3) are reached within a radius of 4 to 5 km from the emission point with an asymmetry in the spatial distribution due to the presence of prevailing north and north-east winds."

In a study of soils near three chlor-alkali plants in Europe, Biester and others (2002) suggested that direct uptake of atmospheric Hg by foliage and litterfall is one process by which Hg is incorporated into forest soils. At one chlor-alkali plant, "the highest concentrations were found downwind (main wind direction) from the plant in a distance of about 250 m from the plant." At the second plant, background Hg concentrations were found within 4 km downwind from the plant. Biester and others (2002) noted a correlation of Hg soil concentrations with landscape cover, soil type, and ground elevation of the soils in relation to the elevation of the stack of the chlor-alkali plant. The relation with elevation is demonstrated in their statement "Moreover, Hg concentrations in soils sampled in the valley at distance of more than 500 m from the plant rapidly decrease to values near the background, whereas Hg concentrations in forest soils on the mountain exceed 1,000 μg/kg even at a distance of 1 km." The intensity of their soil sampling allowed an estimate of the total inventory of Hg retained in the soils. Only 1 to 2 percent of the estimated total atmospheric emissions were found in the soils (upper 20 cm) within a distance of 1 km downwind (within a 90º arc) of three chlor-alkali plants that have been operating for 25 to 30 years.

Near a 10-year-old Cuban chlor-alkali plant, rapid decreases in Hg concentrations in soils and vegetation also were observed with increasing distance from the site. Gonzalez (1991) states "… that from 5 km on, it is not possible to find differences in concentrations with respect to more remote sites, even along the main wind direction (E-NE)." In this location, a lake is located 3 to 15 km upstream of the plant, but the lake is downwind along one of the prevailing wind directions. Gonzalez (1991) concludes "the dam waters are not polluted by Hg, suggesting that the atmospheric emissions of Hg by the plant are not significant."

In the chlor-alkali industrial process, electrolysis is used to generate chlorine gas (Cl_2) and caustic soda (NaOH) from NaCl in a two-step process. The Georgia-Pacific chlor-alkali plant in Bellingham went into production with 26 mercury electrolysis cells in 1964 with a capacity of 122 tons of chlorine per day (Perry, 1974). The capacity of the plant was increased by about 50 tons of chlorine per day in late 1977 with the construction of six additional mercury cells (Georgia-Pacific Inc., Bellingham, Washington, written commun., 1975). The chlorine production had increased to 220 tons by 1990 due to more efficient electrolysis.

In the first step of the chlor-alkali process, mercury electrolysis cells produced chlorine gas and an intermediate sodium-mercury amalgam (Na(Hg)) according to the equation:

$$NaCl + Hg \Rightarrow \tfrac{1}{2}Cl_2 + Na(Hg). \quad (1)$$

Mercury-laden vapors associated with the outlet end box and inlet end box on each cell were collected by a fume air system under a slight vacuum. The gas passed through a water heat exchanger and then through a glycol/water heat exchanger. A demister then was used to recover particulate mercury from the vapor stream and additional mercury was removed from the air in the brine de-chlorination stack. In February 1975, the Hg emission rate from the chlorine side of the process (fume air system) was 5.7 g/hr. A scrubber was installed during the modernization and expansion of the plant in the late 1970s. After the modernization, the fume air stream passed through this scrubber and was discharged through the hypo stack at a temperature of about 23 ºC (Ed Dahlgren, Georgia-Pacific, Inc., Bellingham, Washington, written commun., 1988). In 1981, a mercury source-emissions test (Chris Lee, Georgia-Pacific, Inc., written commun., 1981) indicated a release of 0.0139 g/hr from the hypo stack. In 1995, the Hg emission rate from the hypo stack based on test results was less than 0.0003 g/hr (AmTest-Air Quality, Inc., Preston, Washington, written commun., 1995).

In the second step of the process, caustic soda was produced and mercury was regenerated during the dissociation of the sodium-mercury amalgam that was produced during the first step:

$$Na(Hg) + H_2O \Rightarrow NaOH + \tfrac{1}{2}H_2 + Hg. \quad (2)$$

After being generated in the decomposers, hydrogen gas containing some Hg vapor passed through several heat exchangers to reduce the temperature to between 8 and 15 ºC, at which point Hg condenses and drops out of the gas stream. Hg vapor concentrations decrease with decreasing temperature of the air stream if the Hg vapor is in equilibrium with the glycol-cooled hydrogen heat exchanger. In the final stage of pollution control, the hydrogen gas passes through a demister to reduce particulate Hg in the air stream before exiting the hydrogen stack (Ed Dahlgren, Georgia-Pacific, Inc., written commun., 1988; Cazzaro, 2002). A Hg source test in 1975 indicated an emission rate of 17.9 g/hr from the hydrogen stream. A second demister was added to the hydrogen stack during the late 1970s expansion and renovation. In the late 1970s, the graphite anodes were replaced with titanium anodes, which are thought to reduce Hg emissions from chlor-alkali plants due to reduced maintenance. Three emission tests of the hydrogen stream were performed at the Bellingham Georgia-Pacific chlor-alkali plant between 1981 and 1995 (table 3) and indicated a release of between 14.8 and 22.5 g/hr, depending on production rate and outlet temperature.

Although these Hg emission rates reflected the potential amount of airborne Hg leaving the chlor-alkali plant, they did not reflect the actual atmospheric loading from the Georgia-Pacific industrial complex. During the emission tests in 1975, 1981, 1987, and 1995, the hydrogen gas stream from the chlor-alkali plant was directed to the stack for accurate Hg emissions measurements. As early as 1972, the hydrogen gas stream from the chlor-alkali plant was piped to the nearby lignin plant of the Georgia-Pacific paper mill, where the hydrogen was burned as an energy source. When accounting for all equipment operation rates in the lignin plant from 1971 to 1999, an estimated 80 to 85 percent of the hydrogen produced by the chlor-alkali plant was burned in the lignin plant spray dryers [(Roger J. (Chip) Hilarides, Georgia-Pacific, Inc., oral commun., 2003].

The hydrogen line that ran from the chlor-alkali plant to the lignin plant was uninsulated and unheated. Seven traps were placed in the line to collect condensed elemental Hg. The line generally was at ambient temperature or lower. As a result, there were periodic operational problems associated with freezing of residual moisture in the line during the winter. The maintenance crews replaced the traps in the late autumn and put steam heat through the lines to keep the traps warm during cold spells. Chlor-alkali plant cell renewers and maintenance personnel routinely drained elemental Hg from these hydrogen line traps and returned it to the plant for reuse. The exact amount or frequency of the Hg recovered from the hydrogen line was not documented. According to facility maintenance personnel, about 10 lb of Hg was recovered from each of seven traps that were drained each autumn. Additionally, during the course of the year, six other traps (three on the chlor-alkali plant side and three on the lignin plant side) were drained twice a month of about 10 lb of Hg, or a total of 240 lb/yr from these traps. Therefore, by the estimate of Georgia-Pacific, Inc., personnel, 310 lb of liquid mercury were removed each year from the hydrogen line to the lignin plant (Roger J. (Chip) Hilarides, oral commun., 2003), which reduces the Hg loading to the lignin plant by 16.1 g/hr. This estimate of Hg drained from the line is larger than the flow of Hg entering the line in hydrogen stream as measured by the 1981 Hg emission test and represented at least a 70-percent reduction to the Hg loading to the lignin plant for the other three Hg emission tests. Although the amount of Hg estimated to have been collected in the line to the lignin plant may have been too high, most of the Hg going to the lignin plant during the winter months probably condensed in the line and was collected by Georgia-Pacific, Inc., personnel, thus, most of the Hg measured during the stack test did not enter the atmosphere.

The third source of Hg is general evasion of solid Hg from the Hg electrolysis cell room. If the operator follows the general housekeeping rules outlined in Review of National Emission Standards for Mercury (U.S. Environmental Protection Agency, 1984), an Hg emission rate of 1.3 kg/d can be assumed (40 CFR 61 Subpart E). The Hg emission rate from the mercury cell room at the Georgia-Pacific plant in Bellingham was never measured. Hg emission rates from cell rooms of chlor-alkali plants are difficult to measure and have been measured with modern analytical instrumentation only at a few plants. At a Swedish chlor-alkali plant built in 1924 with a chlorine production similar to that of the Bellingham plant, the Hg emissions from the cell house averaged 10.5 g/hr in 2001 (Wangberg and others, 2003). However, little information about the operation of the Swedish plant is given except that plant-reported Hg emissions per metric ton of chlorine were less than the European average. The emission rate of Hg from the cell room of a North Carolina chlor-alkali plant under controlled conditions was 21.6 g/hr (Kinsey, 2003). These measured Hg emission rates are considerably less than the U.S. default emission rate (51.6 g/hr) listed in 40 CFR 61 Subpart E. Given the differences in industrial processes and institutional controls, caution is advised when generalizing the Hg emissions on the basis of data from different chlor-alkali plants. Even within a single plant, Hg emission rates may vary over short periods due to variable operational schedules (routine versus invasive maintenance). The Hg emission rates between similarly equipped plants also are probably affected by the diligence of the operators and the maintenance of industrial and pollution devices.

Other atmospheric releases of Hg may be associated with the handling of clarifier solids from the treatment of wastewater generated from the Georgia-Pacific chlor-alkali plant and stockpiling of brine sludge in the early 1970s. Brine sludge was stockpiled in an impoundment basin until a recovery system was developed in 1973. The initial method of treatment was roasting the brine sludge and recovering the Hg in condensers. About 9.5 g/hr of Hg left the condensers, where 90 percent was removed (Perry, 1974). The 1 g/hr of Hg leaving the stack that originated from the roasting of brine sludge would not have been included in the total Hg emissions that were measured in 1975. Roasting was abandoned in late 1974 in favor of chemical stabilization.

Georgia-Pacific personnel (Roger J. (Chip) Hilarides, oral commun., 2003) verified that wood burned in the hog fuel burner was not contaminated with Hg. Burning of wood waste and old clarifier sludge was reported in the 5000 block of Everson-Goshen Road in 1979 (Glen Hallman, Northwest Air Pollution Authority, written commun., December 3, 1979; Bob Bishop, Washington State Department of Ecology, Olympia, Washington, written commun., December 7, 1979).

Other Mercury Air Sources in Whatcom County

The estimate of the Hg emissions from the Cherry Point refinery west of Ferndale was 0.47 g/hr for 2002, based on production rates and emission factors (Sally Otterson, written commun., 2004). Hg emissions from the release of dental fillings (about 1 mg/body) during the cremation of bodies (U.S. Environmental Protection Agency, 1997a) are not further considered. Although cement plants had operated in Whatcom County, they had closed by the 1970s before air emission measurements were required, and therefore the historical emissions from cement plants will not be assessed and considered. Mercury is not included in the Ecology's Toxic Release Inventory and Hazardous Waste databases for the Intalco aluminum smelter or a second refinery at Ferndale.

Model of Mercury Deposition to Lakes in Whatcom County from a Unit Mercury Emission

There is considerable debate in the scientific literature as to whether deposition of Hg to lakes is a result of global or local sources (Hanisch, 1998). "Assuming constant emission rates, the quantity of mercury deposited on a regional and local scale can vary depending on source characteristics (especially the species of mercury emitted), meteorological and topographical attributes, and other factors" (Expert Panel on Mercury Atmospheric Processes, 1994). Hg emitted to the atmosphere in the vapor phase (Hg^0) generally will remain in the atmosphere between 6 months and 2 years (Fitzgerald, 1989; Lindqvist, 1994). The residence time of Hg in the atmosphere reflects a combination of direct diffusion into water and the oxidation to Hg(II), which is rapidly removed by rainfall and dryfall. Therefore, local Hg sources emitted to the atmosphere in the Hg^0 form will likely not result in significant increases in near-field deposition of Hg, but will simply add to the global atmospheric pool.

Only Hg emitted in the particulate or oxidized (Hg(II)) reactive gaseous mercury (RGM) forms are likely to be deposited in significant amounts near the source. The atmospheric residence time of particulate-phase mercury is on the order of about a week (Pacyna and others, 1996). An estimated 5 to 10 percent of the initial reactive Hg emissions is deposited within 100 km of the sources (U.S. Environmental Protection Agency, 1997b). Contact with ground vegetation results in removal of atmospheric reactive Hg. However, methods to quantify this removal are difficult. Scavenging parameters based on vertical gradients of atmospheric Hg suggest removal mechanisms of Hg that are similar to those of nitric acid; however, Lindberg and Stratton (1997) state, "it is not possible [to] prove whether the concentrations gradients are statistically significant." The deposition of particulate or reactive forms of Hg is known to be influenced by rainfall.

Although single rainfall events have been known to remove one-half the RGM, the relatively uniform rainfall concentrations across southern Florida have been interpreted to mean that scavengable Hg (that is, RGM) is uniformly distributed on the time scales (residence times) of days to weeks during the wet season (Guentzel and others, 2001). In the absence of significant upwind local sources of particulate or reactive Hg, the direct deposition of Hg to the surfaces of lakes in the greater Bellingham area will be similar.

Data are not available on the forms of atmospheric Hg in the gases emitted by sources in Whatcom County or on the concentrations and forms of Hg in air masses above lakes of interest in Whatcom County after major Hg emitters began operating. Most of the worldwide effort in characterizing the speciation of Hg for atmospheric sources has concentrated on major energy-combustion sources, such as coal-fired electrical plants. A literature search indicated only two studies in which the forms of Hg emitted from the Hg electrolysis cell rooms of chlor-alkali plants were determined, and only one study in which the forms of Hg emitted by a municipal waste incinerator were investigated. Because the forms of Hg in a gas stream depend on the complex interaction between materials and industrial processes, the operations of a specific plant may affect the forms of Hg being emitted. Given the paucity of total Hg emission data for major Hg sources in Whatcom County, the lack of any local speciation data, and few worldwide measurements of Hg speciation for the types of plants operating in Whatcom County, a simple model that provides a rough estimate of Hg deposition for the emission sources described above is warranted, rather than applying a complex model with numerous input terms for which local data are not available.

A simple model based on first principles was developed for this project and is described in appendix B (at back of report). The model is used primarily to estimate, in a consistent manner, the deposition of Hg to lakes in Whatcom County from among past local sources of Hg. The simplifications of the plume dynamics allow use of an analytical solution to estimate the deposition rate without having to numerically estimate the concentration of each Hg species, i, in the air mass. Deposition to the aquatic and terrestrial surfaces depends primarily on the reactivity constant for species i, - k_i, which is the measure of how fast Hg is removed from the atmosphere and is inversely proportional to the atmospheric half-life of each species. In the model, the deposition of the three forms of Hg (vaporous elemental, reactive gaseous, and particulate) initially emitted from a source are considered independently, and the total deposition is the sum of the deposition of the three species (Dep_i).

$$Dep_i = \frac{k_i \bullet Q_{i,0} e^{-\frac{k_i}{u} \bullet x}}{(u \bullet \tan(\alpha) \bullet x)}, \quad (3)$$

where

$Q_{i,0}$ is the emission of species i from the source,

x is the distance between the source and lake of interest, and

u is the wind speed, and α is the angle of the plume dispersion.

The only simplification of the model is that the Hg from the source laterally disperses across the width of the plume as defined by $\tan(\alpha) \bullet x$. Equation 3 can be broken down into terms that easily relate the emissions rate to transport concepts (fig. 5). The $Q_{i,0}/u$ represents the dilution of the emission source of species i by the wind field, much as the initial concentration of a pollutant in a river resulting from a point source discharge is dependent on the velocity of a river. The term $e^{-k_i/u \bullet x}$ represents the decrease in the transport of the Hg species with distance from the source resulting from the deposition of species i between the source and distance x. The term $1/(\tan(\alpha) \bullet x)$ represents the decrease in the concentration of species i in the plume, and therefore deposition over an area, by the lateral dispersion of the plume with distance from the source. In most cases, deposition is independent of the plume angle. As the degree of lateral dispersion is increased (increasing α), the hourly deposition at a given site under a given plume will decrease. Because deposition will occur over a greater area, the number of hours that a given site is under a plume will increase. Only when a site is in a zone of wind shear will deposition be dependent on α. Note that the deposition also is independent of the thickness of the plume. Although expanding the thickness of the plume will decrease the concentration of species i in the plume, deposition will occur over a taller air column.

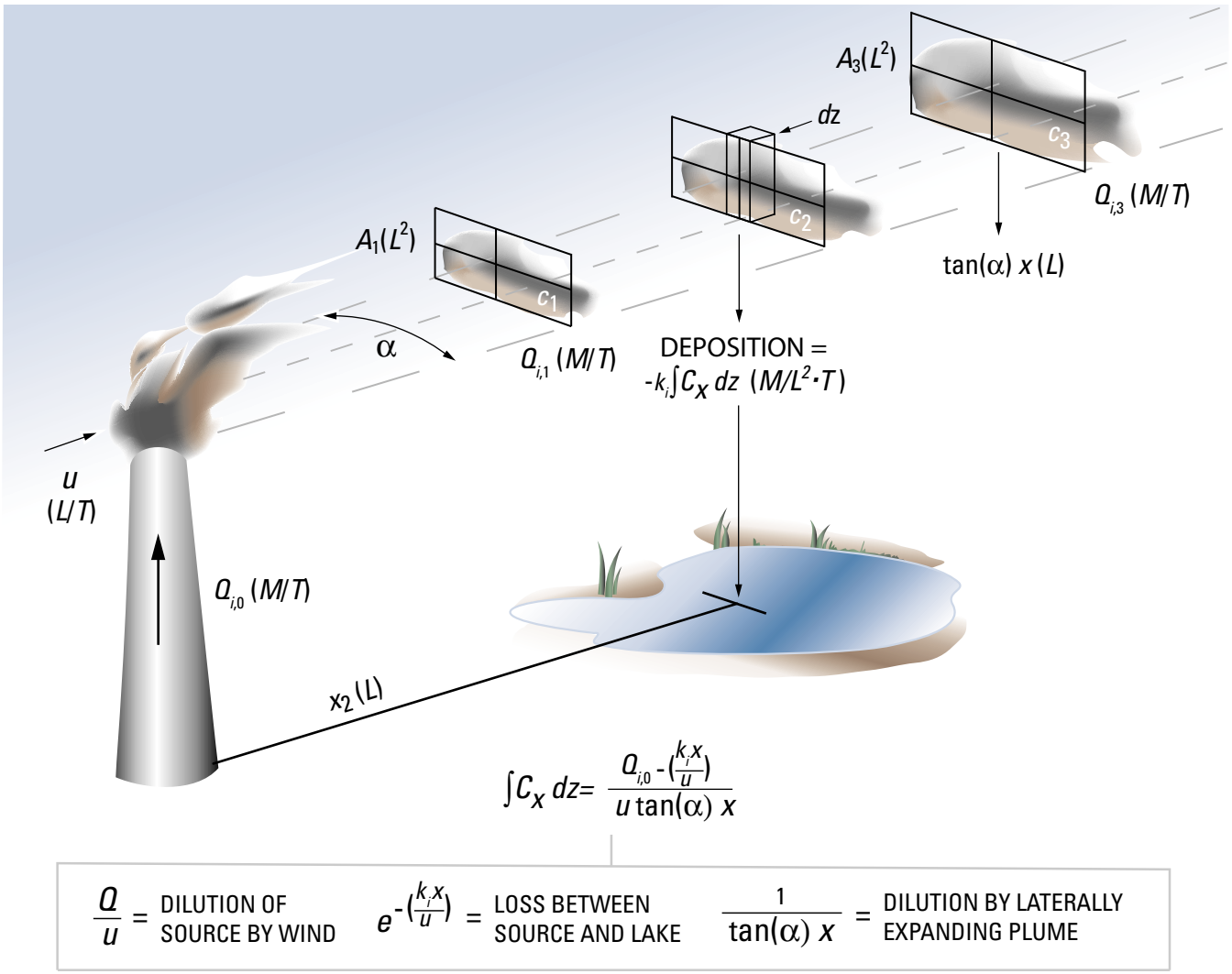


Figure 5. Concepts of a simple model of the deposition of mercury. (See the accompanying text for explanation of symbols.)

Given a specific wind field, deposition at a given point from the source was calculated knowing the emission rate of species *i* ($Q_{i,o}$) and the inverse of the half-life (k_i). The model was applied using hourly wind speed and wind direction data from the Georgia-Pacific (G-P) Bellingham meteorological site (Axel Franzmann, Northwest Air Pollution Authority, oral commun., 2003) during 1996 because the wind data for 1995, the year of the last emission test, were incomplete. If the plume was above the target receptor (target-receptor direction is within wind direction ± ½α), direct deposition of Hg to the site (fig. 6) was calculated for that hour. Deposition for a unit emission of Hg (Q_o = 1 g/hr) for an atmospheric half-life of 1 day, 1 week, 1 month, 1 quarter-year, and 1 year was calculated using equation 3 with the measured wind speeds at the Bellingham site, the distance from the source to the target receptor (from table B1 in appendix B), and a standard plume angle (α) of 20 degrees (table B1 and figs. B1-B2 in appendix B).

The deposition rates to target core sites in the eight lakes/basins are described in appendix B for unit emissions of Hg (1.0 g/hr) for Hg species with a range of half-lives. The total deposition of Hg (TotDep) to the water surface was calculated as the sum of the depositions from three species:

$$\text{TotDep} = \text{Dep}_P + \text{Dep}_R + \text{Dep}_V, \quad (4)$$

where

Dep_P is the deposition of particulate mercury,

Dep_R is the deposition of reactive Hg, and

Dep_V is the deposition of vaporous mercury.

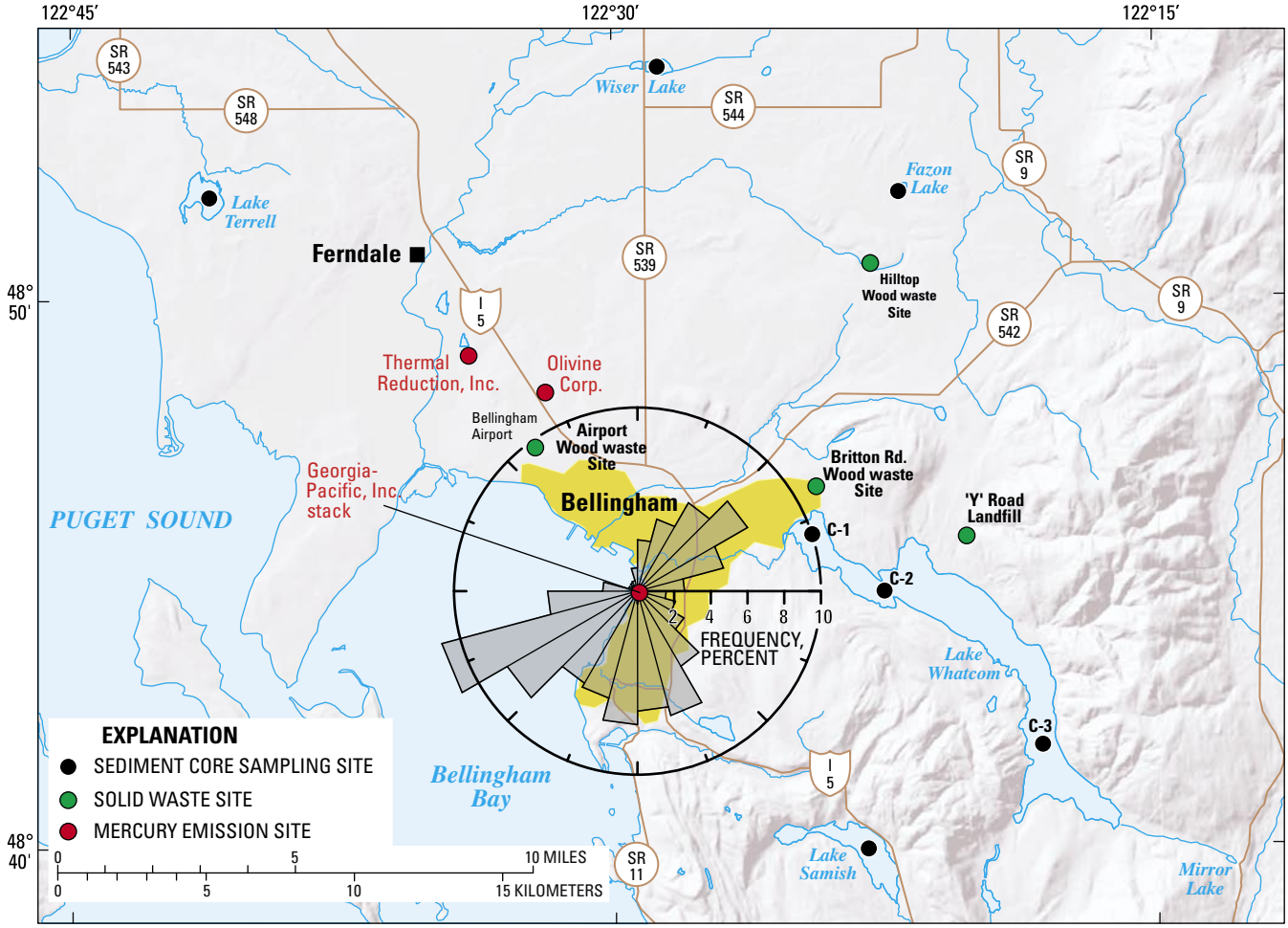


Figure 6. Location of sources of mercury emissions, sediment core sampling sites, and solid waste sites, with the 1996 wind field at Georgia-Pacific meteorological station superimposed, Bellingham, Washington.

Estimates of Mercury Deposition to Lakes in Whatcom County from Known Sources

The model deposition rates for a unit Hg source under a variety of residence times provided in appendix B allow for the calculation of estimated total deposition to any of the eight lakes/basins in Whatcom County from a source at any of three locations: downtown Bellingham, Ferndale, or Bellingham airport. This calculation requires knowledge of the Hg emission rate to the atmosphere, the form of Hg in the plume once it was cooled to ambient temperatures, and the residence time of each of the three species. Minimum and maximum estimates were made by varying the residence times within the range reported in the literature. Residence times used ranged from 1 day to 1 week for particulate Hg; 1 week to 1 month for reactive Hg, and 3 to 12 months for vaporous Hg. These times reflect the overall residence of a specific species once emitted to the atmosphere from a source, even though there may be some conversion among species (for example, the oxidation of vaporous Hg to Hg(II) proceeds slowly before the Hg(II) is more rapidly removed from the atmosphere by rainfall).

The same meteorological data sets and the same assumptions about the reactivity of the various forms of atmospheric Hg were applied consistently across Hg sources and receptor sites. The model required total Hg emissions data and the speciation of Hg from that specific source. Confidence in the model estimates was categorized on the basis of the quality of the best available data used in the estimates (table 4). Simulations were assigned to

- Category A if both Hg emission and speciation data were available for a source in Whatcom County.
- Category B when actual total Hg emissions data from the source and speciation data from the literature for the specific type of source were used.
- Category C when total Hg emission rates from another source of similar type and speciation data from the literature for the type of source were used.
- Category D when actual emission data were used and the Hg speciation was estimated from the actual air chemistry of the source gases.
- Category E when actual total Hg emissions were used and the speciation was estimated on the basis of concentrations of constituents other than Hg in the emissions gases obtained from the literature for facilities of the same type.

Table 4. Matrix of confidence category for estimates of mercury (Hg) deposition based on source of emission and speciation data

[**Emission Data**: Category C emission data for Thermal Reduction Company, Inc., were generated from the category B emission data and changes in Hg emission due to changes in control devices. Category F emission data for the City of Bellingham sewage sludge incinerator were generated from the Category E estimates and changes in Hg emission due to changes in control devices. –, no instances of speciation/emission combination]

		EMISSION DATA		
		Total emissions measured	**Estimated from changes in control devices**	**Similar type of facility measured**
SPECIATION DATA	Speciation measured	**Category A:** none	–	–
	Similar type of facility measured	**Category B:** 1) Thermal Reduction Company, Inc.; 1986-92	–	**Category C:** 1) Georgia-Pacific fugitive emissions from cell room 2) Olivine Corp.; 1984-90
	Estimated from change in control devices from measured speciation with similar type of facility	–	**Category C:** 2) Thermal Reduction Company Inc.; 1974-85 2) Thermal Reduction Company, Inc.; 1993-97	**Category D:** 1) Olivine Corp.; 1991-97
	Estimated from measured air chemistry	**Category D:** Georgia-Pacific fume air	–	–
	Estimated from estimated air chemistry	**Category E:** City of Bellingham sewage sludge incinerator; 1993-present	**Category F:** 1) City of Bellingham sewage sludge incinerator; 1974-1993	–
	No air chemistry information	–	**No estimates made:** 1) Georgia-Pacific hydrogen stream	**No estimates made:** 1) Cherry Point oil refinery, British Petroleum

A change in operation and(or) the extent of abatement of atmospheric emissions occurred during the operations of all major Hg sources in Whatcom County (fig. 4). Changes in Hg deposition to lakes in Whatcom County were assessed as a result of these improvements in operations. Species-specific adjustments in Hg emissions published in the literature were used to estimate Hg emissions either before or after the installation of a specific type of air-pollution control device. When species-specific efficiencies of air-pollution control devices from the literature were used to adjust the species emission rates as a result of changes in operations, confidence in the simulation was downgraded one category.

Thermal Reduction Company, Inc. Municipal Waste Incinerator

The 1988 Hg emission test for the Thermal Reduction Company, Inc., municipal waste incinerator (MWI) (39.7 g/hr or 9.1 g/metric ton of Hg of waste burned) was used to estimate Hg deposition for the period when the two modules were equipped with only an electrostatic precipitator (1986–92). The one measurement in the hot flue gas of a MWI indicates that 70 percent of the Hg is emitted in the reactive form (Prestbo and Bloom, 1995) because chlorine gas, originating from the burning of plastics (for instance, vinyl chlorides) stabilizes the Hg as Hg(II). Electrostatic precipitators only are effective for particulates and not for reactive or vaporous Hg (van Velzen and others, 2002), and therefore the remaining 30 percent was assumed to be vaporous Hg (Category B simulation). The maximum estimate of Hg deposition to Lake Whatcom from the Thermal Reduction Company, Inc., MWI from 1986 to 1992, based on the lower bounds of residence times for each Hg species, ranged from 0.77 ($\mu g/m^2$)/yr at the basin 1 coring site to 0.30 ($\mu g/m^2$)/yr for the basin 3 coring site (table 5, fig. 7*A*). In contrast, the maximum estimate of Hg deposition to the three northern lake sites from 1986 to 1992 ranged from 3.6 to 4.7 ($\mu g/m^2$)/yr, with 99 percent of the deposition originating from the RGM emissions (fig. 7*B*).

Dry sorbent injection and filtration through a baghouse (fiber filter) was estimated to reduce RGM emissions by 50 percent and vaporous Hg emission by 30 percent (van Velzen and others, 2002). For the Thermal Reduction Company, Inc., MWI, the Hg emissions from 1993 to its closure were estimated to be 5 g/metric ton or 19 g/hr with RGM decreasing to 62 percent of the total emissions. The deposition model indicated that the installation of the pollution devices in 1993 reduced Hg deposition from the Thermal Reduction Company, Inc. by more than 50 percent, with the contribution from RGM decreasing to 95 percent of the total deposition to lakes in Whatcom County (Category C).

Because particulate emissions were high during earlier operation of the eight smaller module units for Thermal Reduction Company, Inc. (1974–85), Hg could have condensed on the large mass of particles that was emitted. To account for higher particulate air emission during the earliest stage of operations, an estimate of 5 percent of the total Hg emissions (Category C) measured by the 1988 source test was modeled as particulate Hg, based on general knowledge of the efficiencies of air-pollution control devices. This change increased maximum estimated Hg deposition to lakes in Whatcom County by about 30 percent to 1.0 ($\mu g/m^2$)/yr for the basin 1 site because particulate Hg is deposited nearer the source than RGM.

Olivine Corporation Municipal Waste Incinerator

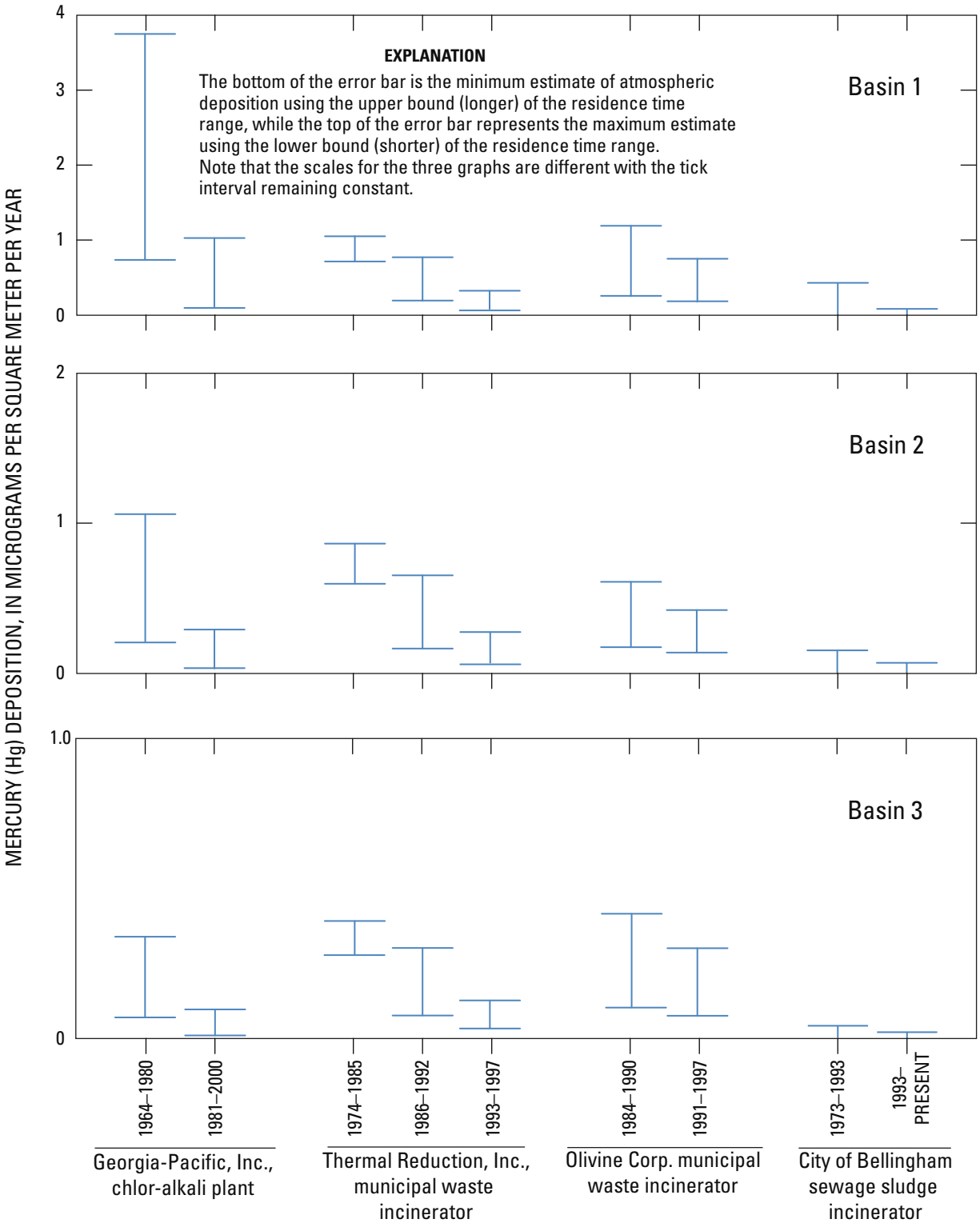
Hg emissions tests were not conducted at the Olivine Corp. facility either before or after the installation of the dry injection sorbent with baghouse filtration in 1991. Between 1984 and 1990, when the Olivine Corp. municipal waste incinerator was operating with only a fabric filter, data on the amount of waste burned and the number of hours of operation could be documented only for the years 1984 and 1990; those data indicated a burn rate of about 10 ton/hr while operating 40 percent of the time. The Hg content of the municipal waste from Olivine Corp. incinerator was assumed to be the same as that of the Thermal Reduction Company Inc., incinerator during the 1988 emission test. Because air-pollution devices at both facilities prior to the installation of dry injection sorbents were ineffective in removing RGM and vaporous Hg, the Hg emission rate per ton for the Olivine Corp. incinerator was estimated to be 9.1 g/metric ton, from the 1988 Thermal Reduction Company Inc., emission test, which converts to an annual average of 25 g/hr (Category C). Because the Olivine Corp. incinerator was operating under a conditional approval with a fabric filter and no emission tests of any kind were conducted, problems with particulate emissions were assumed to be the same as that of the earlier Thermal Reduction Company, Inc., operations. Particulate Hg emissions were estimated to contribute 10 percent of total Hg emissions, with 60 percent in the reactive form, and 30 percent in the vaporous form. The maximum deposition of Hg to Lake Whatcom basin 1 sediment coring site from the Olivine Corp. incinerator was estimated to have been 1.2 ($\mu g/m^2$)/yr, with 50 percent originating from deposition of particulate Hg with a residence time of 1 day. Maximum deposition for the northern lakes ranged from 2.3 ($\mu g/m^2$)/yr for Lake Terrell to 6.4 ($\mu g/m^2$)/yr for Fazon Lake.

Table 5. Estimates of deposition rates of mercury to lakes in Whatcom County from local emission sources, Washington

[All values are given in micrograms per square meter per year [($\mu g/m^2$)/yr]. Minimum estimates are based on the upper bound (longer) within the range of residence times for each species, while maximum estimates are based on the lower bound (shorter) within the range of residence times for each species (to convert annual deposition to (ng/cm^2)/yr multiply by 0.1). (ng/cm^2)/yr, nanogram per square centimeter per year; g/hr, gram per hour]

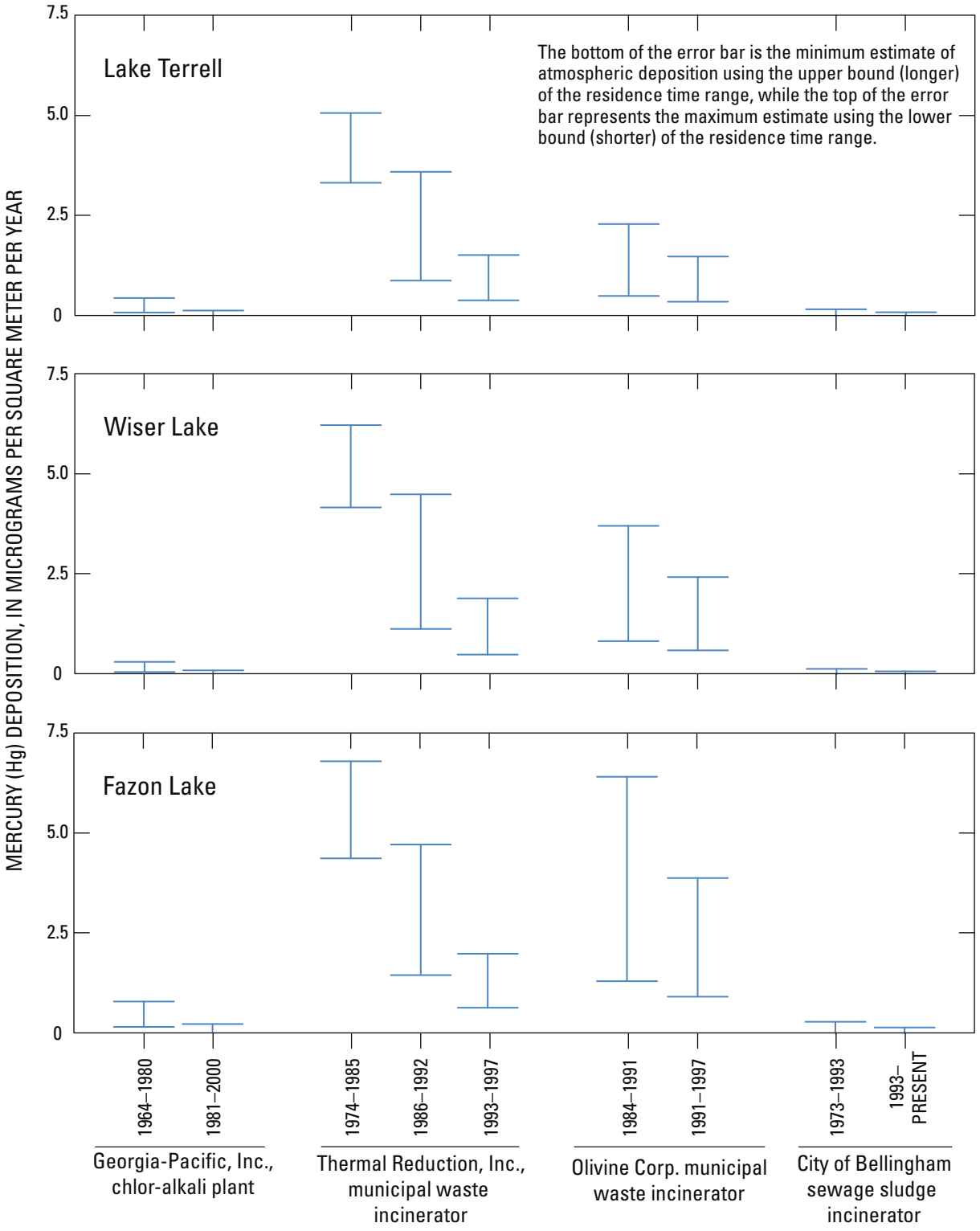
	Lake Whatcom			Lake Samish	Baker Lake	Fazon Lake	Lake Terrell	Wiser Lake	Confidence category (see table 4)
	Basin 1	Basin 2	Basin 3						
Georgia-Pacific Chlor-Alkali Plant[1]									
Fume Air System Before Modernization									
Minimum	0.63	0.18	0.06	0.07	0.02	0.14	0.08	0.05	D
Maximum	2.7	.77	.24	.28	.08	.56	.31	.21	D
Fugitive Emissions from Mercury Cell Room									
Minimum using 10 g/hr emission	0.11	0.03	0.01	0.01	0.00	0.02	0.01	0.01	C
Maximum using 20 g/hr emission	1.03	.29	.10	.11	.03	.22	.12	.08	C
Thermal Reduction Company, Inc., Municipal Waste Incinerator									
Minimum: 1993-97	0.08	0.07	0.03	0.08	0.04	0.63	0.38	0.48	C
Maximum: 1993-97	.32	.27	.13	.30	.12	2.0	1.5	1.9	C
Minimum: 1986-92	.19	.17	.08	.19	.09	1.5	.89	1.1	B
Maximum: 1986-92	.77	.65	.30	.72	.29	4.7	3.6	4.5	B
Minimum: 1974-85	.72	.61	.28	.67	.27	4.4	3.3	4.2	C
Maximums: 1974 -85	1.0	.86	.39	.92	.36	6.8	5.1	6.2	C
Olivine Corporation Municipal Waste Incinerator									
Minimum: 1991-97	0.15	0.12	0.06	0.14	0.05	0.79	0.31	0.51	D
Maximum: 1991-97	.68	.38	.27	.56	.19	3.5	1.3	2.2	D
Minimum:1984-90	.25	.17	.10	.22	.08	1.3	.50	.83	C
Maximum: 1984-90	1.2	.61	.42	.85	.26	6.4	2.3	3.7	C
City of Bellingham Sewage Sludge Incinerator									
Maximum in 2003	0.10	0.03	<0.01	0.01	<0.01	0.02	0.01	0.01	E
Maximum After 1993 Expansion	.21	.06	.01	.02	.01	.04	.02	.02	E
Maximum Before 1993 Expansion	.42	.13	.04	.05	.01	.10	.05	.03	F
Total Deposition Based on Maximum Estimates									
1964-74	3.75	1.06	0.34	0.39	0.12	0.78	0.43	0.29	
1974-84	2.50	1.29	.53	1.08	.41	7.12	5.23	6.33	
1986-91	3.41	1.68	.85	1.73	.61	11.44	6.05	8.29	
1993-97	2.24	1.00	.49	.99	.36	5.72	2.98	4.16	
1997-2000	1.24	.35	.10	.13	.04	.26	.14	.10	
2000-2002	.21	.06	<.01	.02	.01	.04	.02	.02	
2003	.10	.03	<.01	.01	.01	.02	.01	.01	

[1]Does not include hydrogen gas stream.



A. Lake Whatcom

Figure 7. Estimates of atmospheric deposition of mercury as a function of times of significantly different operations for the three basins of Lake Whatcom and Lake Terrell and Fazon and Wiser Lakes, Whatcom County, Washington. Emissions from Georgia-Pacific chlor-alkali plant does not include hydrogen stream. Times of significantly different operations are shown in figure 4.



B. Lake Terrell, Wiser Lake, and Fazon Lake

Figure 7.—*Continued.*

After the lime injection scrubber was installed at the Olivine Corp. incinerator in 1991, between 30,000 (1992 and 1997) and 39,800 tons (1996) of municipal waste were reported to have been burned while operating between 40 and 94 percent of the time, with no data reported for 1993. A burn rate of 9 metric ton/hr was assumed in 1996 for the hours of operation because tonnage of waste burned was not reported. Like the calculations for Thermal Reduction Company, Inc., the emission rates were estimated based on the efficiencies of lime injection sorbent in removing RGM and vaporous Hg according to (van Velzen and others, 2002) (Category D). Even though the Hg emission per ton was reduced to about 5 g/metric ton after installation of the lime injection system, the annual average of metric tons burned increased. Although the average Hg emission remained at 25 g/hr, the percentage of Hg in the RGM decreased to 63 percent, with the remainder emitted as vaporous Hg. The assumed elimination of particulate Hg emission shifts the dominant depositional species to RGM, which accounts for 95 percent of the Hg deposition. The maximum Hg deposition to all lakes using this change in speciation is estimated to reduce deposition between 1991 and 1997 by about 40 percent less than the 1986–91 levels (figs. 7*A* and 7*B*).

City of Bellingham Sewage Sludge Incinerator

The speciation of Hg in the City of Bellingham sewage sludge incinerator was not measured. In a German study, sewage sludge was amended with chlorine-rich solids and fluids to achieve an average chlorine content between 0.15 and 0.5 percentage by weight (Saenger and others, 1999). When the average Cl content in the incinerated sludge was less than 0.2 percentage by weight (dry weight basis), the percentage of Hg in the vaporous state in the flue gas increased substantially. With increasing chlorine content of the incinerated sludge, the loss of vaporous Hg presumably was balanced by an increase in RGM due to the stabilization of Hg by Cl in the flue gas, as has been observed with MWI flue gas. Thus, the chlorine chemistry of the sludge and incinerator flue gas is needed to accurately estimate Hg speciation in the absence of measured values. Data on chlorine chemistry were not available for the City of Bellingham sewage sludge incinerator. Hydrochloric-acid emission factors for flue gas in municipal sewage sludge incinerators (U.S. Environmental Protection Agency, 1995) have been estimated to be about 0.003 percentage of Cl by wet weight, based on little empirical data. This compares to 0.3 percentage of Cl by wet weight in municipal waste incinerators. Therefore, the percentage of Hg in the RGM form in sewage-sludge emission is likely much less than the 70-percent RGM found for municipal waste incinerators. Given the knowledge of the operation of the City of Bellingham sewage sludge incinerator and its air-pollution devices, the estimated maximum percentage of Hg emissions as RGM was 10 percent (0.34 g/hr) and the estimated maximum percentage of Hg emissions as particulate Hg was 0.2 percent (table 3). The estimated maximum present-day (2003) Hg deposition to Lake Whatcom basin 1 from the municipal sewage sludge incinerator operating at 33-percent capacity was 0.10 ($\mu g/m^2$)/yr (Category E), with 55 percent of the deposited Hg originating from emissions of RGM. Estimated maximum deposition to the three northern lakes ranged from less than 0.01 to 0.02 ($\mu g/m^2$)/yr.

Air emissions of Hg from the City of Bellingham sewage sludge incinerator before the expansion of the plant in 1993 probably were higher than present-day (2003) emissions because the tray scrubber was not as efficient as the Venturi scrubber at removing reactive gaseous and particulate Hg. To account for the less efficient pollution-control devices prior to 1993, when the tray scrubbers were the only air-pollution control devices, estimated maximum RGM emissions were three times higher than present-day (2003) RGM emissions. Likewise, estimated maximum particulate Hg emissions were twice as high as present-day particulate emissions. Because both tray scrubbers and Venturi scrubbers are inefficient at removing vaporous Hg, estimated vaporous Hg emissions before modernization were only 10 percent higher than present-day (2003) emissions (Category F). Estimated maximum Hg deposition to Lake Whatcom basin 1 was 0.42 ($\mu g/m^2$)/yr, and the estimated maximum deposition for the northern lakes was 0.10 ($\mu g/m^2$)/yr.

Georgia-Pacific Chlor-Alkali Plant

Atmospheric Hg was emitted from three major sources at the Georgia-Pacific facility: the hypochlorite side of the chlor-alkali process (also known as fume air source), Hg hydrolysis cell room emissions, and the alkali side of the chlor-alkali process that generates hydrogen gas. Although three measurements of the fume air emissions were made, only the 1975 emission of 5.7 g/hr was significant. The 1975 measurement was assumed to be 100 percent in the RGM form (Category D) because (1) this gas stream originated from the part of the plant where hypochlorite was produced and could have easily reacted with Hg to form RGM, and (2) the greater-than-99-percent reduction in the Hg emission from this stream after the installation of the scrubber in the path of this air stream strongly indicates a form that effectively was removed by the scrubbers. If Hg was in the vaporous form, the installation of the scrubbers would not have caused such an effective reduction in emissions. Estimated maximum deposition of Hg to basin 1 of Lake Whatcom was 2.7 ($\mu g/m^2$)/yr (table 5; fig. 7*A*), and estimated maximum deposition to the three northern lakes was 0.21 ($\mu g/m^2$)/yr for Wiser Lake, 0.31 ($\mu g/m^2$)/yr for Lake Terrell, and 0.56 ($\mu g/m^2$)/yr for Fazon Lake (table 5 and figs. 7*A* and 7*B*).

Although regulations allow an operator to assume a rate of 54 g/hr for cell-room emissions, modern measurements of Hg emission indicate rates between 10 and 20 g/hr. The speciation of Hg fugitive emissions from Hg hydrolysis cell rooms at two plants (Wangberg and others, 2003; Landis and others, 2004) indicate that about 2 percent of the Hg is emitted as RGM. Maximum deposition rates of 1.03 ($\mu g/m^2$)/yr for Lake Whatcom were calculated for basin 1 using the short residence time and the higher emission rate, with 80 percent of the deposition originating from vaporous Hg deposition. Because the northern lakes are not in the direction of the primary wind pattern for the City of Bellingham, estimated maximum deposition was 0.22 ($\mu g/m^2$)/yr for Fazon Lake.

An understanding of the air chemistry of atmospheric emissions from lignin spray dryers is required to predict the speciation of the Hg in the hydrogen stream that was combusted as a fuel source for the dryers, and to estimate the deposition rates to local lakes. The speciation of Hg in the combusted hydrogen stream emitted from the lignin spray dryers could not be assessed because data on the air chemistry of the emissions from the lignin spray dryers are not available. The lignin plant did not measure mercury emissions during their stack emission test or at any other time during the plant's operation. Only emissions of particulate matter were monitored in the gas stream leaving the lignin plant.

The spray dryers at the Bellingham pulp plant were the main combustion units, with three or four units operating during the nearly 28 years of operation when hydrogen was used as the primary fuel source. Natural gas (at least 10 percent) was used to supplement the hydrogen gas in the burner operations. The combustion temperature was maintained in the range of 820 to 1,100 ºC, at which time combustion exhaust gas was mixed with fresh air. This mixture entered the spray dryer, where the operating temperature was maintained between 230 to 240 ºC to assure product quality. Hg in the air stream entering the lignin spray dryers after combustion of the H_2 gas and mixing with ambient air to a temperature of 230 to 240 ºC likely was in the vaporous phase. The calcium- or sodium-based lignin products in liquid form were then sprayed into the hot gas stream. The unit used a cyclonic flow system in which the lignin was sprayed near the top of the cyclone in a manner that is described as spiral flow down, such that the dried lignin particles were recovered in the lower part of the cyclone. Exit temperatures for gases from the spray dryer ranged from 115 and 128 ºC. Reduced sulfur species from the residual sulfite pulping reagents or chlorine from bleaching process that can stabilize the Hg in the RGM phase may have been released into the moist air as the lignin was dried and the gas stream was cooled to 115-128 ºC as it left the lignin spray dryer.

The gases then passed through a multiple-stage cyclone where more lignin was recovered. The air exhaust from the cyclones was then sent to cold-water scrubbers, most of which also had demisters, before being discharged to the atmosphere. The particulate concentration of the exhaust typically ranged from 20 and 65 milligrams per dry standard cubic meter (dscm) and an average particulate-matter emission rate of 5,200 g/hr for all spray dryers. The burning of H_2 gas in lignin spray dryers of a Ca-based sulfite mill is a unique operation in the U.S. The only two other U.S. Ca-based sulfite mills are in Wisconsin. Neither mill complex is associated with a chlor-alkali plant. Although one Ca-based sulfite mill is associated with a lignin spray dryer, emissions data are not available for chloride or sulfur species for this lignin spray dryer.

The only information about the speciation of Hg in emissions from combusted hydrogen gas stream is derived from a North Carolina Kraft paper mill that burned the hydrogen gas stream from the adjacent chlor-alkali plant. Within 1 km of a chlor-alkali plant/Kraft pulp mill complex, RGM in air associated with the complex accounted for about 2 percent of the total atmospheric Hg (Hayward and others, 2003). However, these air mass samples likely contained the RGM emitted from the air venting of the mercury cell room and the hypochlorite air stream of the chlor-alkali plant. The deposition of Hg to local lakes from the hydrogen stream of the chlor-alkali plant was not estimated because data are not available for the air chemistry of hydrogen streams combusted in lignin spray dryers.

Summary of Deposition from Local Air Emissions

The simple model of Hg deposition that was developed for this project provides a tool to estimate the relative contributions of Hg from the different local emission sources to the basins of Lake Whatcom and other lakes in Whatcom County. The absolute values of these estimates of deposition rates are subject to considerable uncertainty as a result of our lack of knowledge of atmospheric residence times of Hg. The degree of uncertainty is evaluated by estimating both minimum and maximum deposition rates, based on the upper and lower bounds, respectively, of the range of residence times of the three Hg species reported in the literature. The deposition from the hydrogen gas stream could not be evaluated because of a complete lack of information on the air chemistry of the exhaust gas, and only the maximum emissions from the City of Bellingham sewage sludge incinerator were estimated because of a lack of speciation and air-chemistry data from the stack gas.

Before the modernization of the Georgia-Pacific chlor-alkali plant in 1970s, the greatest deposition of Hg from local sources [3.75 (μg/m^2)/yr] was estimated to have occurred within basin 1 of Lake Whatcom because of the prevailing wind pattern. Most of the estimated deposition originated from RGM emitted from the fume air system of the plant. As the chlor-alkali plant was being modernized in 1970s and the fume air emissions were virtually eliminated, the maximum estimate of the deposition from emissions of the Hg hydrolysis cell room of the plant was 1.03 (μg/m^2)/yr. At the same time, the City of Bellingham sewage sludge incinerator with its simple control device [maximum deposition of 1.65 (μg/m^2)/yr] and the two-unit Thermal Reduction Company, Inc., incinerator that violated particulate emissions standards (maximum estimate of 1.0 (μg/m^2)/yr began operations. The upgrade of the Thermal Reduction Company, Inc., incinerator in 1985 [a decrease in the estimated maximum deposition of Hg to basin 1 of Lake Whatcom from 1.0 to 0.77 (μg/m^2)/yr] was counteracted by the beginning of the operation of the Olivine Corp. incinerator [estimated maximum of 1.2 (μg/m^2)/yr]. In the early 1990s, the upgrades in the pollution-control devices in the two municipal incinerators and the municipal sludge incinerator led to a chronic reduction of 1.18 (μg/m^2)/yr to a total maximum deposition of Hg to basin 1 of 2.24 (μg/m^2)/yr. With the closing of the two municipal incinerators and the chlor-alkali plant, the estimated maximum deposition of Hg to basin 1 of Lake Whatcom from the City of Bellingham incinerator was 0.21 (μg/m^2)/yr between 2000 and 2002. The lack of speciation data from the oil refinery emissions precludes estimating Hg deposition from the two refineries, the only other known large facilities emitting Hg. This present-day estimate of Hg deposition to basin 1 likely will change as better information about the air chemistry of sludge incinerators and refineries becomes available. The estimated maximum of Hg deposition to the basin 3 sediment coring site from Bellingham sources was a factor of 10 less than the estimates for the basin 1 site because basin 3 is out of the primary wind pattern of the City of Bellingham.

The largest deposition of Hg to lakes in Whatcom County from local emission sources was estimated to have fallen on lakes north of Bellingham (Lake Terrell, Wiser Lake, and Fazon Lake) that are in the primary wind pattern of the two municipal waste incinerators. At the time that both incinerators were operating with minimal air-pollution devices, estimated maximum deposition of Hg to Fazon Lake was 11.6 (μg/m^2)/yr, and the estimates for Wiser Lake and Lake Terrell were 8.38 and 6.15 (μg/m^2)/yr, respectively. These maximum deposition estimates decreased by 50 percent with the addition of dry sorbent air-control devices in the early 1990s. Deposition from Bellingham sources to these three lakes was an order of magnitude less than the deposition from the municipal waste incinerators because of the dominant wind patterns.

Wind blows toward Lake Samish from Bellingham only under light and variable wind; wind conditions under which this model does not simulate Hg deposition well. The maximum deposition from 1986 to 1991 was 1.73 (μg/m^2)/yr, originating primarily from the municipal incinerators. The maximum estimate of deposition to Baker Lake from local sources from 1984 to 1991 was 0.61 (μg/m^2)/yr, but was subject to greater uncertainty because of the distance between source and target receptor.

Tributary Riverine Inputs

Ecology (Norton, 2004) collected six water samples for the analyses of total Hg from each of three tributaries of basin 1 to Lake Whatcom and from seven tributaries to basin 3 (fig. 2) from July 2002 to May 2003 (Norton, 2004). Total Hg concentrations of individual creek samples ranged from the detection limit (<2 ng/L) to 17 ng/L (table 6). The concentration of 17 ng/L from Blue Canyon Creek in November 2002 was the only concentration that exceeded the water-quality standard of 12 ng/L (Norton, 2004). Blue Canyon Creek drains a basin containing historical coal mines, but the lack of ancillary environmental parameters precludes assessing the impact of these mines.

Median total Hg concentrations for the six measurements for each creek ranged from the detection limit for Blue Canyon Creek to 5.7 ng/L for Carpenter Creek. Median Hg concentrations were 3.8 ng/L in Austin Creek and 3.3 ng/L in Anderson Creek. Total Hg concentrations generally were higher at the end of the low-flow season, during September and November, when specific conductance was higher than during the wet season (January and March); this condition also was observed in Wisconsin forests (Stoor, 2002). The wet season arrived late in 2002 and only one minor precipitation event occurred before the November 17 creek sampling (figs. 8*A* and 8*B*). The ratio of average Hg concentrations of dry season:wet season ranged from 1.2 for Austin Creek to 2.3 for Smith Creek (a comparison was not made for Blue Canyon Creek because of the few detectable concentrations).

For each stream, detectable total Hg concentrations were regressed against specific conductance. A significant (p <0.05) correlation was observed only for Olsen Creek (fig. 9) with higher total Hg and specific conductance being observed during the end of the dry season and low detectable total Hg concentration observed in March and May (table 6). Increased specific conductance in surface waters has been attributed to greater ground-water inputs (Winter and others, 1999). Therefore, the high concentrations of Hg in Olsen Creek probably were a result of greater influence of ground-water inputs when ground water contributed most of the base flows at the end of the low-flow season. The highest specific-conductance values were measured at Silver Beach Creek, but total Hg concentrations were not correlated with specific conductance.

Table 6. Summary of analyses of water samples from the mouth of the tributaries to Lake Whatcom, Washington, July 2002 to May 2003

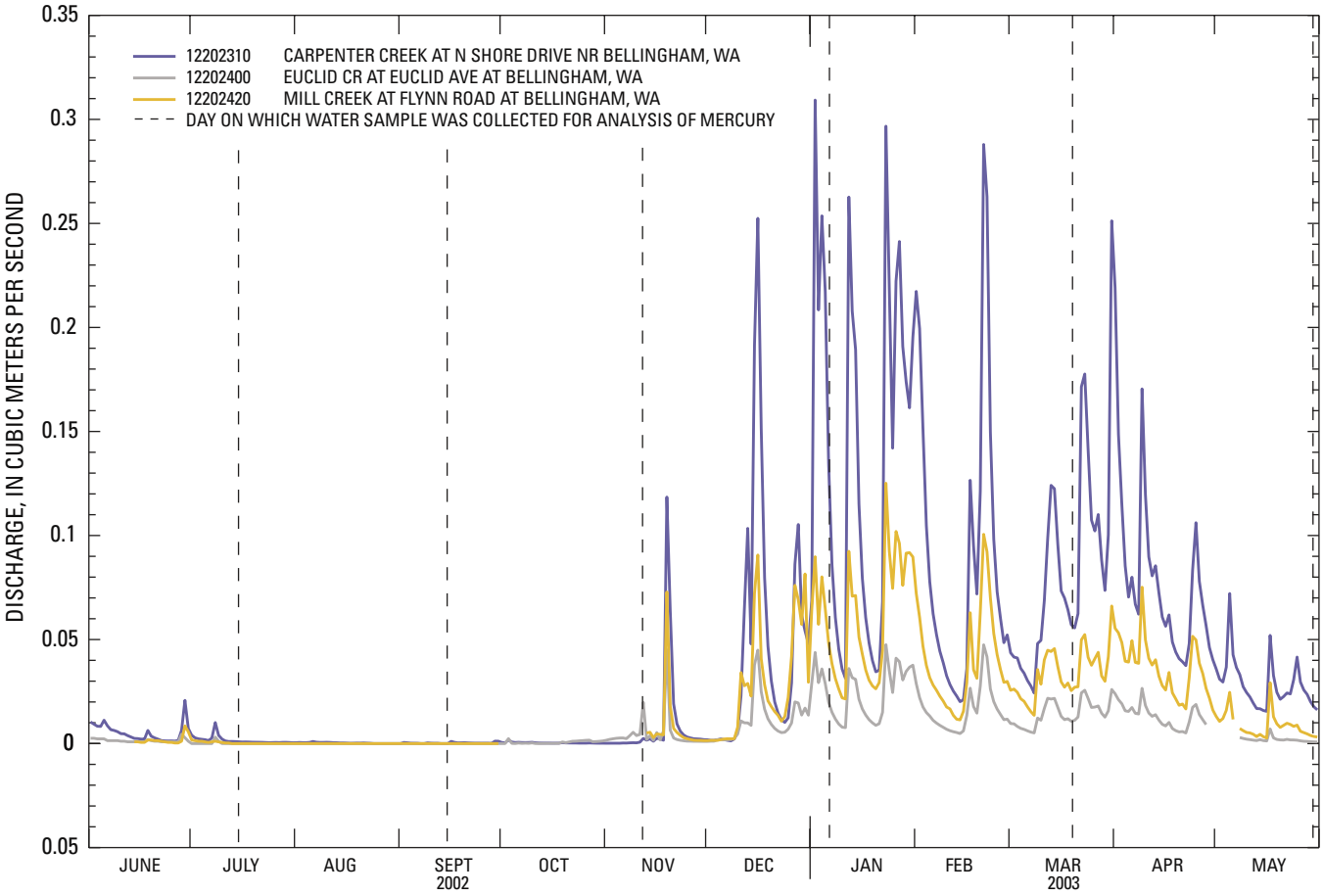
[Data from Norton (2004). Location of drainage basins are shown in figure 2.
ND, not determined; NS, not sampled; <, not detected at detection limit shown. ft^3/s, cubic feet per second; µS/cm, microsiemens per centimeter at 25°C; mg/L, milligram, per liter; ng/L: nanogram, per liter; –, no data]

Sampling round	Collection date	Time	Flow (ft^3/s)	Specific conductance (µS/cm)	Total suspended solids (mg/L)	Total mercury (ng/L)	Total mercury (ng/L) (duplicate)
Anderson Creek							
1	07/16/02	8:15	ND	35.2	19	5.5	7.2
2	09/17/02	9:50	6.78	52.7	2	<2	–
3	11/12/02	9:30	31.38	69	7	4.3	–
4	01/07/03	9:25	71.47	38.7	6	4.1	–
5	03/19/03	12:15	69.33	43.4	5	<2	–
6	05/28/03	13:30	62	30	6.5	2.6	2.4
Median						3.3	
Austin Creek							
1	07/16/02	9:30	1.19	77.2	1	4	4.5
2	09/17/02	10:40	.67	102.3	<1	3.8	–
3	11/12/02	10:25	2.88	125	2	7.3	–
4	01/07/03	10:15	15.96	46.9	1	3	–
5	03/19/03	11:25	3.24	52.6	3	3.8	–
6	05/28/03	12:40	–	66.5	1	<2	–
Median						3.8	
Brannian Creek							
1	07/16/02	8:50	1	37.00	<1	4.5	–
2	09/17/02	10:05	.59	54.6	<1	5.8	–
3	11/12/02	9:50	ND	63.0	<1	9.2	8.8
4	01/07/03	9:45	11	35.6	<1	<2	–
5	03/19/03	11:45	10	37.0	2	4.6	–
6	05/28/03	13:00	1.2	32.9	2	<2	–
Median						4.5	
Carpenter Creek							
1	07/16/02	11:30	0.03	100.9	2	7.9	–
4	01/07/03	12:15	3	45.1	<2	7	–
5	03/19/03	9:50	2	49.4	2	4	–
6	05/28/03	9:20	.68	53.1	3	4.4	–
Median						5.7	
Euclid Creek							
1	07/16/02	10:08	ND	94.5	2	6.0	–
3	11/12/02	11:00	0.7	90.0	<1	7.4	–
4	01/07/03	10:45	.53	57.4	<1	4.3	–
5	03/19/03	11:00	.34	65.2	3	3.2	–
6	05/28/03	12:00	ND	83.3	3	2.7	–
Median						4.3	

Table 6. Summary of analyses of water samples from the mouth of the tributaries to Lake Whatcom, Washington, July 2002 to May 2003—*Continued*

[Data from Norton (2004). Location of drainage basins are shown in figure 2.
ND, not determined; NS, not sampled; <, not detected at detection limit shown. ft^3/s, cubic feet per second; µS/cm, microsiemens per centimeter at 25ºC; mg/L, milligram, per liter; ng/L: nanogram, per liter; –, no data]

Sampling round	Collection date	Time	Flow (ft^3/s)	Specific conductance (µS/cm)	Total suspended solids (mg/L)	Total mercury (ng/L)	Total mercury (ng/L) (duplicate)
Mill Wheel Creek							
4	01/07/03	11:20	1.4	64.6	2	8.9	–
5	03/19/03	10:50	.9	72.8	5	5.1	–
6	05/28/03	11:40	.12	101.4	4	4.9	–
Median						5.1	
Olsen Creek							
1	07/16/02	11:40	0.95	73.3	2	6.7	–
2	09/17/02	11:50	.93	102.0	<1	4.9	–
3	11/12/02	12:45	3.1	118.0	7	10	–
4	01/07/03	12:40	9.4	44.4	1	3.8	–
5	03/19/03	9:40	8.3	46.5	2	2.4	–
6	05/28/03	8:55	3.1	51.6	3	2.9	–
Median						4.4	
Silver Beach Creek							
1	07/16/02	10:50	0.07	255.0	8	11	–
2	09/17/02	11:20	.09	310.0	5	4.1	3.1
3	11/12/02	12:20	1.1	150	10	11	–
4	01/07/03	11:45	.95	121.8	2	6.4	–
5	03/19/03	10:10	ND	128.4	5	4.4	–
6	05/28/03	9:45	ND	176.0	6	3.1	–
Median						5.4	
Smith Creek							
1	07/16/02	12:00	1.61	56.9	1	5.1	–
2	09/17/02	12:20	.79	83.6	<1	4.2	–
3	11/12/02	13:10	11.37	85	4	10	–
4	01/07/03	13:00	12.66	45.2	1	2.6	–
5	03/19/03	9:00	3.05	45.6	2	<2	2.3
6	05/28/03	8:20	ND	47.0	2	2.6	–
Median						3.4	
Blue Canyon Creek							
2	09/17/02	9:30	0.79	NS	NS	<2	–
3	11/12/02	9:10	NS	NS	NS	17	–
4	01/07/03	9:10	11.4	NS	NS	<2	<2
5	03/19/03	12:30	12.7	NS	NS	4.6	–
6	05/28/03	13:50	3.06	NS	NS	<2	–
Median						<2	

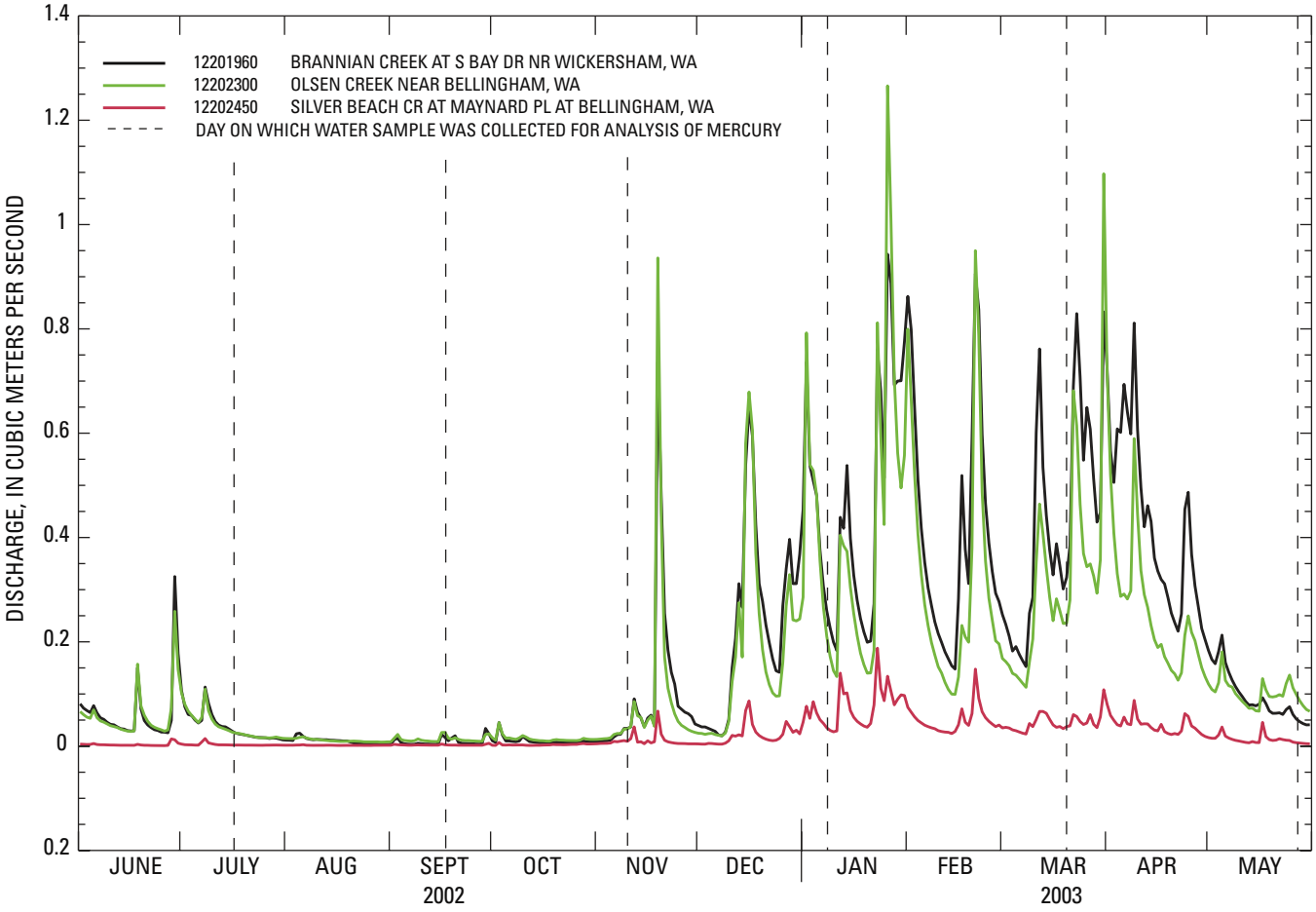


A. Carpenter, Euclid, and Mill Creeks

Figure 8. Daily mean flows of tributaries to Lake Whatcom, Washington, June 2002 – May 2003.

Total suspended-solids (TSS) concentrations for most creeks generally were low, and average TSS concentrations from each creek were less than 3 mg/L. However, the TSS concentration for Anderson Creek was 19 mg/L during the July 2002 sampling, with an average TSS concentration of 8 mg/L. The average TSS concentration for Silver Beach Creek was 6 mg/L, with a maximum of 10 mg/L during the November 2002 sampling. Samples for TSS concentrations were not collected for Blue Canyon Creek. Because total Hg concentrations were not correlated with TSS concentration in any creek, particulate Hg concentrations probably were not the major fraction of the total Hg during the sampling periods. Samples were not collected, however, as the flow of the creeks increased after a rainfall event, a time when high TSS concentrations are expected.

Exponent, Inc., an environmental consulting firm, collected water samples in October 2001 for analysis of total Hg concentrations in four of the creeks that were sampled by Ecology (Gary Bigham, Exponent, Inc., written commun., 2002). Total Hg concentrations ranged from 2.15 to 2.89 ng/L. Average concentrations of Hg in samples collected during the dry season by Ecology for these creeks ranged from 60 percent higher (Anderson Creek) to 130 percent higher (Silver Beach Creek) than the average concentrations reported by Exponent, Inc. The total methyl-Hg concentrations in two creeks draining into basin 1 (Silver Beach and Smith) were about 0.05 ng/L and were about 0.13 ng/L for two creeks draining into basin 3 (Anderson and Austin).



B. Brannian, Olsen, and Silver Beach Creeks

Figure 8.—*Continued.*

Fine-grained sediment (less than 0.004 mm) containing elevated concentrations of Hg (1-8 mg/kg) are known to be present in the Middle Fork Nooksack River (Babcock and Kolby, 1973; Falley, 1974). The City of Bellingham diverts water from the Middle Fork Nooksack River through Mirror Lake (fig. 1) and Anderson Creek to Lake Whatcom to ensure an adequate drinking-water supply to its residents. In October 2001, particulate Hg concentrations of 1.9 ng/L in the water entering the terminus of the water diversion system (table 7) were calculated using the total and dissolved Hg concentrations determined by Exponent, Inc. (Gary Bigham, oral commun., 2002) according to:

Recoverable Particulate Hg = Total Hg – Dissolved Hg. (5)

Particulate Hg in the Nooksack River water contributes to the loading of Hg to Lake Whatcom during periods when this water is being diverted. Suspended solids in the diverted water from the Middle Fork Nooksack River are removed to some degree both in a sedimentation pond at the diversion dam as well as within Mirror Lake before Anderson Creek flows into Lake Whatcom (Tracy, 2001). During the 2002-2003 Ecology sampling year, the volume of the Nooksack Diversion was 28 billion liters (7,404 million gallons), which amounted to 88 percent of the total flow of Anderson Creek. The total Hg concentrations at the terminus of the diversion structure at Mirror Lake averaged 3.15 ± 1.34 ng/L (table 7), whereas the concentrations of total Hg in Anderson Creek that discharges into Lake Whatcom averaged 3.22 ± 2.12 ng/L during periods when the diversion was operating (calculated from data in table 6). Although the Nooksack Diversion may have supplied most of the Hg discharged into Lake Whatcom from Anderson Creek from June 2002 to May 2003, the average total Hg concentration in Anderson Creek was lower than the average for the Lake Whatcom drainage basin.

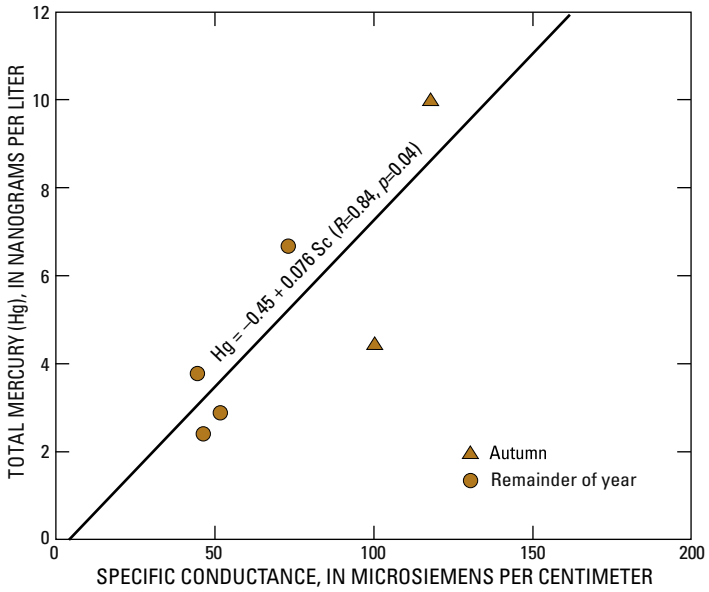


Figure 9. Correlation of total mercury and specific conductance in Olsen Creek, tributary to Lake Whatcom, Washington.

Table 7. Concentrations of mercury (Hg) at the terminus of the Nooksack River diversion, Whatcom County, Washington, October 2001–August 2003

[mg/L, milligrams per liter; µS/cm, microsiemens per centimeter at 25ºC; ng/L, nanograms per liter. –, no data]

Source/Date	Terminus of Nooksack Diversion (Mirror Lake)					
	Total suspended solids (mg/L)	Specific conductance (µS/cm)	Total Hg (ng/L)	Dissolved Hg (ng/L)	Total methyl-Hg (ng/L)	Dissolved methyl-Hg (ng/L)
Gary Bigham, Exponent, Inc., written commun., 2002						
October 2001	9.2	–	4.81	2.92	0.026	0.026
Peg Wendling, City of Bellingham, oral commun., 2003						
August 2002	8.1	66	2.65	–	–	–
October 2002	–	–	–	–	–	–
November 2002	3.7	64	2.37	–	–	–
December 2002	11.0	56	3.35	–	–	–
January 2003	3.8	50	5.1	–	–	–
February 2003	–	–	–	–	–	–
March 2003	2.8	64	1.99	–	–	–
April 2003	3.2	56	1.76	–	–	–
June 2003	76.2	46	7.1	–	–	–
July 2003	72.0	49	7.8	–	–	–
August 2003	85.0	75	–	–	–	–
Mean (June 2002 –May 2003)			3.15	–	–	–
Standard deviation			1.34	–	–	–

The sampling period for which the annual loadings from the terminus of the Nooksack Diversion were calculated may not have been a representative year. In June 2003, total Hg concentrations increased to 7.1 ng/L and remained high during the summer of 2003 (7.8 ng/L) (Peg Wendling, City of Bellingham, written commun., 2003). TSS concentrations of about 80 mg/L were observed during the summer of 2003, which were much higher than those for the period used to calculate annual loads. These high TSS concentrations in the Middle Fork Nooksack River probably were a result of glacial melting. If none of Hg was assumed to be in the dissolved phase, then the particulate Hg concentration on particles would be (7.5 ng/L) / (80 mg/L) or 0.094 ng/mg (or 0.094 mg/kg). However, about 3 ng/L of Hg was in the dissolved phase in October 2001 (Gary Bigham, written commun., 2002). If only 4.5 ng/L of Hg was in the particulate phase, then the Hg concentration on the suspended solids would be 0.05 mg/kg. Fine-grained particles contain higher concentrations of organic matter and more mineral surfaces that adsorb Hg than coarse particles. Therefore, the fine-grained sediments that settle in the deepest parts of Lake Whatcom due to differential settling likely have higher Hg concentration than the estimated average concentration of Hg in particles originating from the Nooksack Diversion.

The Park Place settling pond (basin 1) and a street drain in Cable Street (basin 2) were sampled to assess the input of Hg to Lake Whatcom from stormwater sources. The settling pond on Park Place in basin 1 was sampled three times. In 1998, water from the settling pond contained total Hg concentrations between 8 and 10 ng/L when the TSS concentrations were between 18 and 31 mg/L (Serdar and others, 1999). Using CVAFS methods, Exponent, Inc. (Gary Bigham, written commun., 2002) found total Hg concentrations of 1.2 ng/L, of which 0.25 ng/L was methyl-Hg in 2001. Stormwater entering a street drain on Cable Street was sampled during two 1998 storm events, and total Hg concentrations were about 6 ng/L, even though the TSS concentrations were as high as 220 mg/L. During a different sampling trip, suspended matter in water entering the Cable Street drain was collected using a sediment trap and the Hg concentration on the suspended matter was 0.062 mg/kg.

The loading of Hg (mass/time) can be calculated by multiplying the Hg concentration (mass/volume) times the flow rate (volume/time). The calculations of Hg loadings are based on the average Hg concentrations from the wet season (December 2002 to April 2003) and the dry season (June to November 2002 and May 2003) because of the limited set of Hg data. The flow data were obtained from Institute of Watershed Studies of the Western Washington University (Anderson, Austin, and Smith Creeks) and the USGS (Silver Beach, Euclid, Mill Wheel, Brannian, Carpenter, and Olsen Creeks). The methods for estimating missing flow data are explained in appendix C. The total annual volumes of water are shown in table 8. The flow of Anderson Creek is anomalous in that the dry-season flow is higher than the wet-season flow because of the addition of water diverted from the Nooksack Diversion during the dry season.

The loadings of Hg were roughly proportional to the total annual water volumes of each creek. Anderson Creek contributed 98.6 g of Hg to Lake Whatcom from June 2002 to May 2003. Using the average Hg concentration at the terminus of the Nooksack Diversion system, 88 ± 37 g of the 98 g of Hg was attributed to the flow of the Nooksack Diversion. In contrast, the low-flow Brannian Creek annually contributed only 0.8 g of Hg to Lake Whatcom.

Likewise, TSS concentrations were averaged over the year by season, and the seasonal TSS averages were multiplied by the total seasonal flows to obtain an estimate of the annual TSS loadings that are representative of the sampling period (June 2002-May 2003). By this calculation, Anderson Creek contributes 230 Mg of sediment of the estimated 272 Mg loading to Lake Whatcom. Using monthly averages of the TSS and the monthly diversion volume, the Nooksack diversion contributed 153 Mg to the TSS loading of Anderson Creek. However, the 272 Mg annual loading estimate may have been on the lower end of the range of TSS annual loading. In the summer of 2002, the average TSS concentration for the Nooksack Diversion was 8 mg/L, and the average for the summer of 2003 was about 80 mg/L. If the year on which the annual TSS loading was calculated was shifted forward by 3 months to include the summer of 2003 (September 2002–August 2003), the TSS loading of the Nooksack Diversion would nearly double from 153 to 293 Mg.

Table 8. Annual loadings of mercury (Hg) to Lake Whatcom, Washington, from tributary inflows, June 2002 – May 2003

[A megagram is 10^6 grams, which is commonly known as a metric ton]

Tributaries	Water (liters × 10^9)	Hg (grams)	Total suspended solids (megagrams)
Anderson	31.76	98.6	230
Austin	8.58	30.0	16.3
Olsen	4.78	17.4	8.9
Smith	5.64	16.1	8.7
Carpenter	1.33	7.4	2.4
Mill Wheel	.52	3.6	2.1
Silver Beach	.59	3.3	1.8
Euclid	.27	1.1	.5
Brannian	.27	.8	.4
Total		178.5	272

The Nooksack Diversion clearly has a significant impact on the loadings of Hg and TSS to Lake Whatcom. However, this impact is due primarily to the large volume of water flowing through the diversion relative to the natural flow of Anderson Creek rather than to the Hg concentration in the water or on the particles being transported through the Nooksack Diversion. In fact, the total Hg concentration of the Nooksack Diversion is less than the average for the tributaries of Lake Whatcom.

Ground-Water Sources

Assessments of Hg inputs from ground water to lakes in Whatcom County require knowledge of both the concentration of Hg in ground water and the flow rates of direct discharge of ground water to the lakes. Ground-water models that would provide estimates of freshwater inputs from direct ground-water discharges have not been developed for the area. In addition, sensitive high-quality measurements of Hg in ground water have not been made for the drainage basins of the lakes.

Hg present in ground water could originate from Hg in precipitation, from desorption of Hg, from natural aquifer materials, or from human inputs. Anthropogenic inputs include releases of leachate solutions from regulated and unregulated solid-waste facilities, and releases from septic systems. Two municipal solid-waste sites have been investigated using low-sensitivity screening methods with detection limits ranging from 100 to 500 ng/L for the analysis of Hg (BEK Engineering & Environmental, Inc., 2002a). Total Hg concentration of one of two samples of shallow ground water collected within the confines of the Y-road Landfill I (fig. 6) was 300 ng/L. The total suspended-solid concentration in this sample was 1,300 mg/L, indicating that much of the total Hg in this sample may have been associated with the particles. The dissolved Hg concentrations for these two ground-water samples were less than the detection limit of 100 ng/L. Hg concentrations in samples from five private wells within 608 m of the landfills were less than the detection limit of 100 ng/L. Dr. Susan Cook (Toledo, Washington, written commun., 2003) collected a water sample originating from a domestic well within 608 m of the landfill in 1990 and reported an Hg concentration of 220 ng/L using a more sensitive ICP-MS method. The concentrations of total iron (Fe) and aluminum (Al) in this sample were low, indicating that the sample contained few suspended-sediment particles. Although the ICP-MS provided a more sensitive analyses for Hg, the lack of quality-control data for the samples precluded an assessment of any contamination from the well structures and sampling procedures. Water pumped from wells and stored in the holding tank of the Y-Squalicum Water Association that was provided to Dr. Cook by the Association contained 90 ng/L of total Hg. However, this sample contained 11 mg/L of total Fe and 0.3 mg/L of total Al, indicating the presence of a significant amount of suspended sediments.

Wood-yard waste from the Georgia-Pacific pulp mill was deposited in the Lake Whatcom drainage basin on a lot located on the 3200 block of Britton Road (fig. 6). Hg concentrations in soils on the site were less than 0.05 mg/kg, whereas Hg concentrations in ground water collected from the site were less than the detection limit of 500 ng/L (Melinda Miller, Whatcom County Department of Health, oral commun., 2003).

Two other sites containing wood waste from Georgia-Pacific outside of the Lake Whatcom drainage basin have been studied more extensively, and Hg concentrations in ground water are presented here as examples of the water quality surrounding wood-waste sites. At the Hilltop Woodwaste Landfill, Hg concentrations in soil were greater than the method detection limit (MDL) in three of seven soil samples, with a maximum concentration of 0.7 mg/kg. Total Hg was not detected at 11 of the 21 ground-water sampling sites at concentrations significantly greater than the detection limits for analyses in which the detection limit ranged from 100 to 1,000 ng/L (BEK Engineering & Environmental, Inc., 2002b). Total Hg concentrations were greater than the MDL of 200 ng/L in 11 samples collected at the other 10 sites. However, total Fe concentrations were greater than 10 mg/L in 9 of these 11 samples, which indicates possible particulate contamination of the ground water. The average total Hg concentration for the two samples that had detectable Hg with total Fe concentrations less than 1.0 mg/L was 400 ng/L (MDL = 100 ng/L). Total Hg was detected in 1 of 38 samples that were analyzed with a resulting detection limit of 200 ng/L; however, this one sample also had a total Fe concentration of 3.3 mg/L.

The landfill containing Georgia-Pacific wood waste located at the Bellingham Airport (fig. 6) also was investigated (BEK Engineering & Environmental, Inc., 2002c). Hg concentrations in most of the ground-water samples were less than or near the detection limits (detection limit for analyses ranged from 200 to 1,000 ng/L). One of the four samples with Hg concentrations greater than the detection limit of 200 ng/L also had a high Fe concentration. The average Hg concentration of the three other samples was 300 ng/L.

The release of Hg from septic systems surrounding the lakes of Whatcom County could not be assessed because of a lack of local or regional data. Very little is known about the concentrations and sources of Hg discharged from domestic sewage sources. The mean concentration of Hg in wastewater collected by publicly owned treatment works servicing residential areas was 138 ng/L, with TSS concentrations ranging from 39 to 245 mg/L for one set of residential areas (Association of Metropolitan Sewerage Agencies, 2000). Although many household products contain low concentrations of Hg, the major source of Hg in domestic wastewater was thought to be excretion of Hg released into the body from dental fillings. Skare (1995) suggests that 25 mg of Hg per year is excreted by people with a moderate amount of dental fillings, of which 1.6 mg of Hg per year is excreted in urine. If septic tanks were totally efficient in trapping the fecal and other solid materials, 1 g of Hg would be released into ground water for each 600 people having a moderate number of dental fillings and living in houses served by septic systems surrounding the lake. If colloidal material containing Hg escaped the septic tank and was released into the drain field, the number of people with fillings served by septic systems required to release 1 g of Hg into the ground water would be less. In the reducing condition of a septic tank, geochemical principles suggest that methylation of Hg would likely occur and some of the Hg released into the ground water from a functioning septic system would be in the form of methyl-Hg. About 30 percent of the Hg in the surface discharge from a failing septic system was in the methyl-Hg form (Dr. Susan Cook, written commun., 2003.) Once discharged to the ground-water system, the Hg could be adsorbed by aquifer materials, enter the lake, or bypass the lake, depending on the ground-water flow path.

Mercury in Lake Whatcom and Other Lakes in Whatcom County

Sediments

Surface Sediments

As part of the companion study, Ecology (Norton, 2004) collected five random surface-sediment samples (0 to 2 cm) in Lake Whatcom basins 1 and 2, and 11 random samples from basin 3 in September 2002. Targeted samples within the deltas of tributaries were collected in basin 1 (n = 3) and basin 3 (n = 7). All surface-sediment samples were analyzed for grain size, total organic carbon (TOC), and total mercury by cold-vapor atomic absorption spectrometry (CVAAS). In addition, all random samples from basins 1 and 2 and one-half of the random samples from basin 3 were analyzed for methyl-Hg. The mean Hg concentration of the 31 samples was 0.14 mg/kg (ranging from 0.014 to 0.22 mg/kg). The Washington State Freshwater Sediment Quality Values (FSQV) are being updated, and Norton (2004) compared the Hg concentrations in Lake Whatcom with other FSQVs. Norton (2004) reported that no surface sediment samples exceeded Environment Canada's Probable Effects Level of 0.49 mg of Hg per kilogram. Adverse biological effects frequently occur at concentrations greater than the Environment Canada' threshold effects level (TEL) of 0.17 mg/kg and adverse biological effects rarely occur at concentrations less than the TEL. Hg concentrations in 36 percent of the surface-sediment samples exceeded the TEL.

The delta of the tributaries contained coarser grained sediments than the random samples from the respective basins (fig. 10). Concentrations of total organic carbon generally were higher in basin 1 than basin 2, with basin 3 having the least TOC. Relations between TOC and percentage of sediment in the finest clay fraction (less than 0.004 mm) were not significant. An attempt was made to identify biogeochemical processes that result in higher Hg concentrations in the sediments. A strong linear correlation (r^2 = 0. 78, p <0.01) between total Hg and TOC was found for the subset set of samples containing basin 3 tributary samples, and a weaker correlation was found for the entire data set (r^2 = 0.22, p <0.01). In contrast, a relation for the entire data set that seemed to be non-linear was observed between total Hg and the percentage of fine sediment, with total Hg in two samples significantly less than the pattern would indicate (fig. 10). Methyl-Hg was not linearly correlated with percentage of fines (r^2 = 0.13, p =0.17), but was highly linearly correlated with TOC (r^2 = 0. 80, p <0.01). Because TOC concentrations were highest in basin 1 sediments, concentrations of methyl-Hg also were highest in basin 1 sediments (fig. 11).

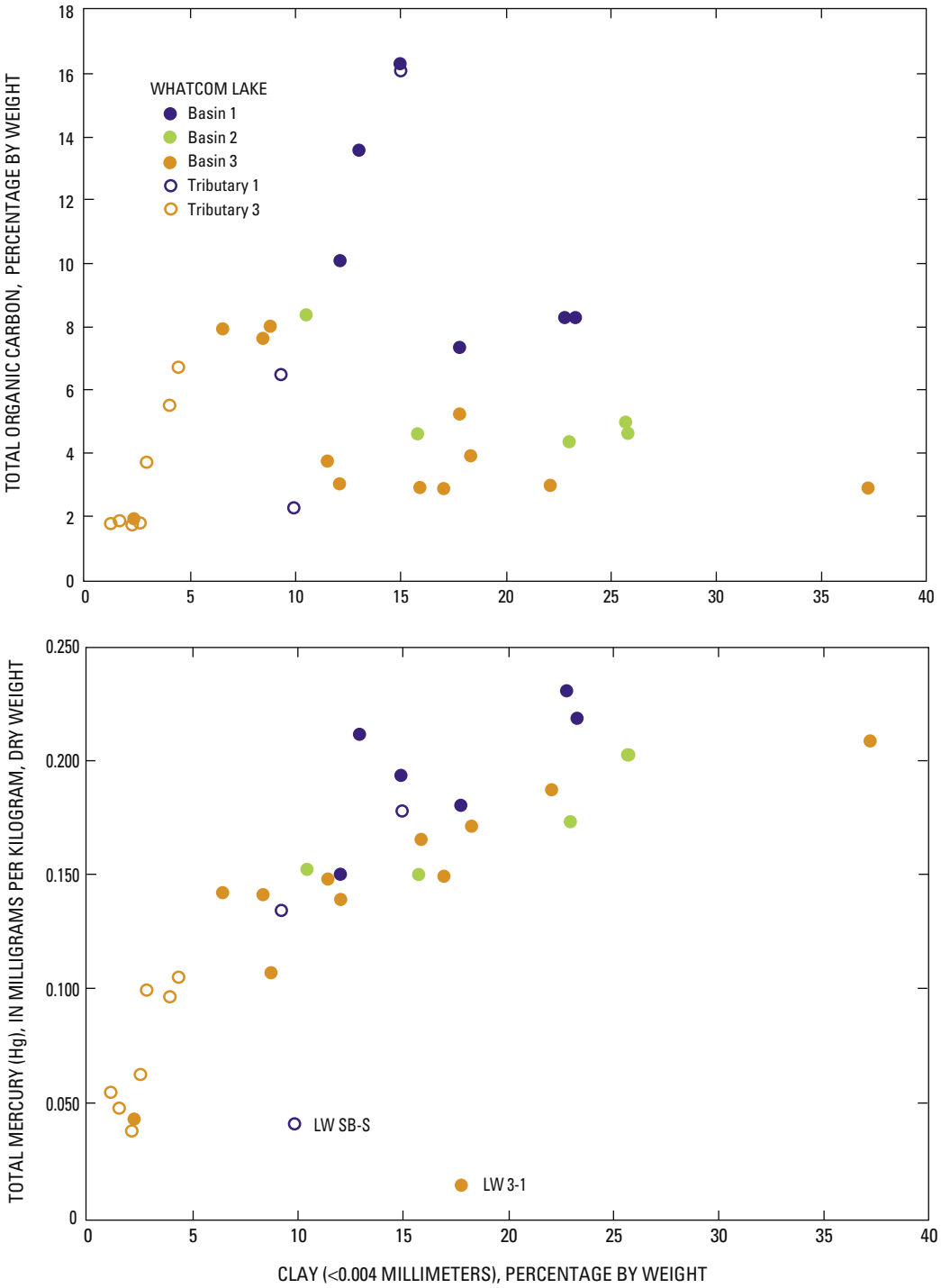


Figure 10. Relation of total organic carbon and total mercury to percentage of fines (<0.004 mm) in samples of surface sediments collected in 2002 near tributaries and in the three basins of Lake Whatcom, Washington. Data are from Norton (2004). The two outlying samples are labeled with the site No.

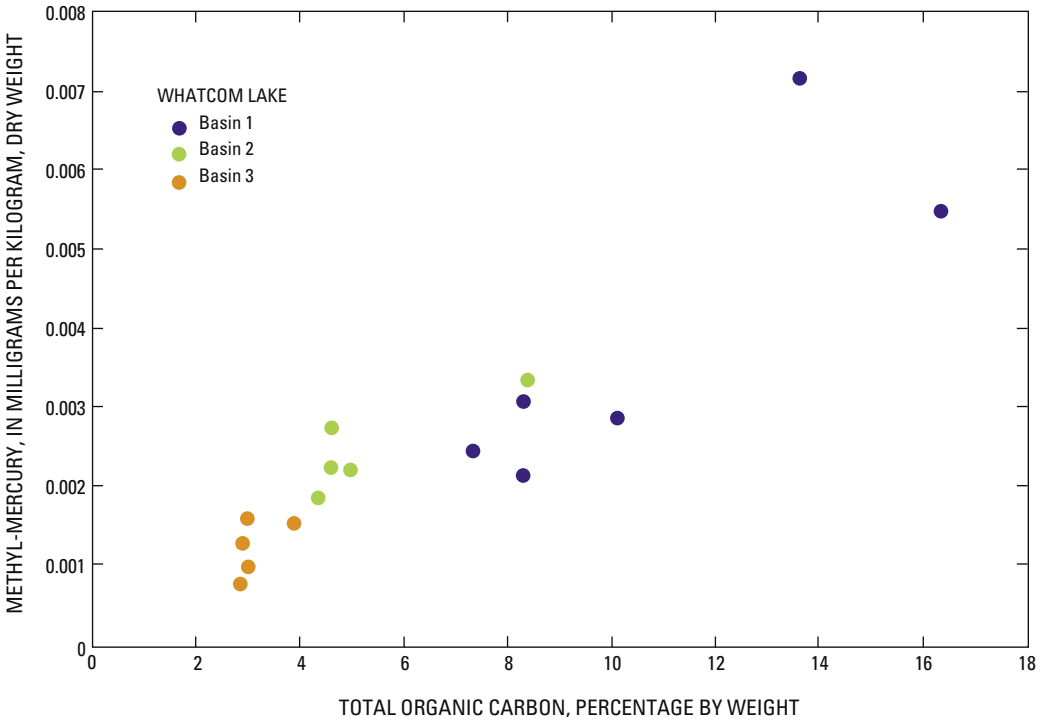


Figure 11. Relation of methyl-mercury to total organic carbon in samples of surface sediments from the three basins of Lake Whatcom, Washington, 2002.
Data are from Norton (2004).

Mercury Sedimentation in Lakes

Hg sedimentation rates were derived from sediment cores from the lakes in Whatcom County. Cores from Lake Whatcom are described in Norton (2004) and sediment cores from Lake Samish, Lake Terrell, Fazon Lake, Wiser Lake, and Baker Lake are described in appendix D (table D1). The method for deriving mass sedimentation rates from the stable lead (Pb), radioactive Pb (^{210}Pb), and radioactive cesium (^{137}Cs) data (table D2) are described in appendix D, along with the derived sedimentation rates for the eight cores (table D3).

Calculations based on a constant sedimentation rate for each core generally provided consistent sedimentation rates within the error of each measurement (about 50 percent). Four of the cores exhibited peaks in ^{137}Cs at a consistent peak concentration of about 3 pCi/g (table 9). The ^{137}Cs peak of 0.97 pCi/g for the core from basin 3 of Lake Whatcom was disregarded because the analytical signal was weak compared to the peak for other cores. The recent deposition of ^{210}Pb also was consistent among the lakes [0.3 to 0.7 (pCi/cm^2)/yr]. The higher deposition of ^{210}Pb in Baker Lake probably was a result of the much higher suspended-matter input from its much larger drainage basin. The total organic carbon data in the surface sediments also are given in table 9.

TOC concentrations were highest in the three northern lakes, ranging from 18.0 to 25.5 percent. These three smaller lakes have only intermittent freshwater flows and are considered catchment lakes. The three larger lakes (Lake Whatcom, Lake Samish, and Baker Lake) have much lower surface TOC concentrations, with Baker Lake having the lowest surface concentrations. Within Lake Whatcom, the concentration of TOC in the top section of these cores follows the same decreasing trend in TOC from basin 1 to basin 3 that was observed with the surface sediments (Norton, 2004). Peak and historical Pb concentrations are given to provide insight into the overall contamination of the site from atmospheric sources (table 9). The surface, peak, and historical concentrations of Hg and Hg sedimentation rates also are given.

Linear sedimentation rates (cm/yr) can be calculated given the constant mass-sedimentation rate [g/cm^2)/yr] and the porosity (measure of water content) of the surface sediments (see appendix D). The surface linear-sedimentation rate ranged from 0.20 cm/yr for basin 3 of Lake Whatcom to 0.99 cm/yr for Baker Lake. The inverse of the surface linear-sedimentation rate provides a measure of the number of years of sedimentation that the top 1-cm section represents. Thus, the top 1 cm of the core from basin 3 of Lake Whatcom represents 5 years, whereas the top 1 cm of the core from Baker Lake represents 1 year.

Table 9. Summary of concentrations and sedimentation rates of mass, total organic carbon, mercury (Hg), lead, lead-210, cesium-137, and total organic carbon in sediment cores from lakes in Whatcom County, Washington

[**Mass sedimentation rate: Bold** number indicates high confidence in the value (see appendix D); *Italicized* number indicates a moderate confidence, and an underlined number indicates a low confidence. pCi/g, picocurie per gram; mg/kg, milligrams per kilograms; g, grams; cm, centimeters; mg, milligrams; pCi, picocuries; cm^2/yr, square centimeters per year; (mg/cm^2)/yr, milligrams per square centimeter per year. –, not calculated because of insufficient data]

Lake	Surface concentration				Peak concentration		
	Porosity	Total organic carbon (percent)	Lead-210 (pCi/g)	Hg (mg/kg)	Lead (mg/kg)	Cesium-137 (pCi/g)	Hg (mg/kg)
Whatcom Lake							
Basin 1	0.944	9.2	8.9	0.229	95	3.78	0.259
Basin 2	.937	5.2	7.8	.202	48	3.13	.204
Basin 3	.927	4.3	8.5	.204	14.4	.97	.217
Lake Samish	.943	5.7	10	.130	63	–	.19
Fazon Lake	.972	25.5	[1]13.4	.180	88	3.33	.27
Lake Terrell	.965	19.4	13	.160	91	3.23	.22
Wiser Lake	.956	18.0	6.5	.230	68	–	.403
Baker Lake	.862	2.5	3.1	.056	43	5.03	.166

Lake	Historical concentration			Sedimentation						
				Recent				Hg [(ng/cm)^2/yr]		
	Porosity	Lead (mg/kg)	Hg (mg/kg)	Mass [(g/cm^2)/yr]	Linear (cm/yr)	Total organic carbon [(mg/cm^2)/yr]	Lead-210 [(pCi/cm^2)/yr]	Recent	Peak	Historical
Whatcom Lake										
Basin 1	0.928	4.6	0.086	**0.045**	0.30	4.1	0.40	10.3	11.7	3.9
Basin 2	.897	5.0	.082	**.040**	.24	2.1	.31	8.1	8.2	3.3
Basin 3	.693	10.0	.085	*.040*	.20	1.7	.34	8.2	8.7	3.4
Lake Samish	.916	6.1	.088	*.072*	.47	4.1	.72	9.4	13.7	6.3
Fazon Lake	.969	13.3	.14	**.039**	.52	9.9	[1].52	7.0	10.5	5.5
Lake Terrell	.927	6.6	.079	*.031*	.33	6.0	.40	5.0	6.8	2.4
Wiser Lake	.953	2.6	.23	*.049*	.41	8.8	.34	11.3	19.7	–
Baker Lake	.749	27.3	–	.370	.99	9.3	1.15	20.7	61.4	–

[1]Estimated from subsurface samples.

The average time before the core collection date of 2002 that a specific core section was deposited within the sediment column was estimated by dividing the cumulative mass sedimentation at the mid-point of the section by the assigned mass sedimentation rate (table 9). Likewise, the time interval over which the core section was deposited was estimated by dividing the cumulative mass sedimentation at the top and bottom of the core section by the assigned mass sedimentation rate. Having transformed the depths (table 10) to their dates of deposition, the trend in Hg sedimentation in each core section was examined back in time by dividing the Hg concentration in each section by the historical background concentrations observed within the deepest sections of that core (fig. 12). The resulting ratio is called an "enrichment ratio" and accounts for the different sediment types that accumulate Hg at a specific location as a result of differences in grain size or organic content. This treatment of the Hg data with depth in the core facilitates the comparisons among lakes. The sedimentation rates are subject to uncertainties described in appendix D.

The histories of Hg inputs to the lakes of Whatcom County are recorded in the sediments of each lake. Prior to 1910, Hg concentrations in sediments from the cores in basins 1 and 2 of Lake Whatcom were fairly constant back to the early 1800s (enrichment ratio about 1 in fig. 12*A*). The average background concentrations were 0.086 and 0.095 mg/kg, respectively (Norton, 2004). In the core from basin 3 of Lake Whatcom, Hg concentrations ranged from 0.055 to 0.113 mg/kg for the period between about 1590 (30 cm) and 1925 (7 cm), corresponding to Hg enrichment ratio between 0.65 and 1.17. After 1910, enrichment ratios increased rapidly in all three basins. In basin 1, the enrichment ratios increased to 2.34 in 1953 (±5 years) before increasing more slowly between 1953 and 1996, at which time a maximum enrichment ratio of 2.95 was observed. In basin 2, the enrichment ratio reached 2.04 in 1975 and only increased to 2.15 by 1995. The maximum enrichment ratio for basin 3 (2.55) was found in 1958 (±6.6 years) in the 4-5 cm section, before decreasing slightly to 2.4 in the surface 2 cm (1988–2002). Hg appears to be decreasing in sediments recently deposited in basin 1.

The history of Hg deposition to the basins of Lake Whatcom are examined in relation to the human development of the drainage basin and the City of Bellingham. The pre-industrial sedimentation of Hg in cores from the deepest parts of each basin ranged from 3.4 to 3.9 (ng/cm^2)/yr (fig. 13). Most of the increases in Hg sedimentation in Lake Whatcom and the five other lakes occurred before major facilities emitting Hg to the atmosphere began operating in Whatcom County. This observation suggests that the general global air pollution resulting from the Industrial Revolution during the first one-half of the 20th century was responsible for a significant amount of Hg loading to Lake Whatcom. For basin 3 of Lake Whatcom, the sedimentation between 1952 and 1965 represents the peak rate of sedimentation of Hg. For basins 1 and 2, the sedimentation of Hg immediately prior to the onset of major local emissions sources represents 80 percent of the peak sedimentation for sediments deposited in the 1990s (fig. 13). The small increases in Hg in basins 1 and 2 after 1964 could have been a result of either further increases in deposition from global sources, the Hg emissions of the Bellingham chlor-alkali plant, and the municipal waste incinerators near Bellingham, or increased tributary loadings to the lake from the Nooksack Diversion. The fact that the enrichments in Hg after 1964 among the basins of Lake Whatcom do not correlate with the estimated atmospheric deposition from local atmospheric Hg sources indicates that local atmospheric Hg emissions were not the dominant sources of Hg to the lake. The small decrease in the Hg concentrations in the most recently deposited sediment to basin 1 (fig. 13) might possibly have been the result of the closing of the two incinerators in 1997. The concentration of Hg in the top 1-cm section of the core basin 1, which represents 3-4 years of sedimentation, would unlikely have had time to respond to the closing of the chlor-alkali plant just two years prior to the collection of the core.

From 1890, when Hg concentrations were 0.088 mg/kg, Hg enrichment ratio (fig. 12*B*) of the Lake Samish core increased erratically to a value of 1.6 in 1954 (±5 years). Between 1975 and 1998, the enrichment ratio averaged about 2.0. Hg enrichment was observed as early as 1920 in the Fazon Lake core, and an enrichment ratio of 1.86 was found for the section dated 1950 (±4 years). Between 1950 and 1998, enrichment ratios ranged from 1.64 to 1.93. Significant decreases in enrichment ratios were observed for recently deposited sediments in both Samish Lake and Fazon Lake.

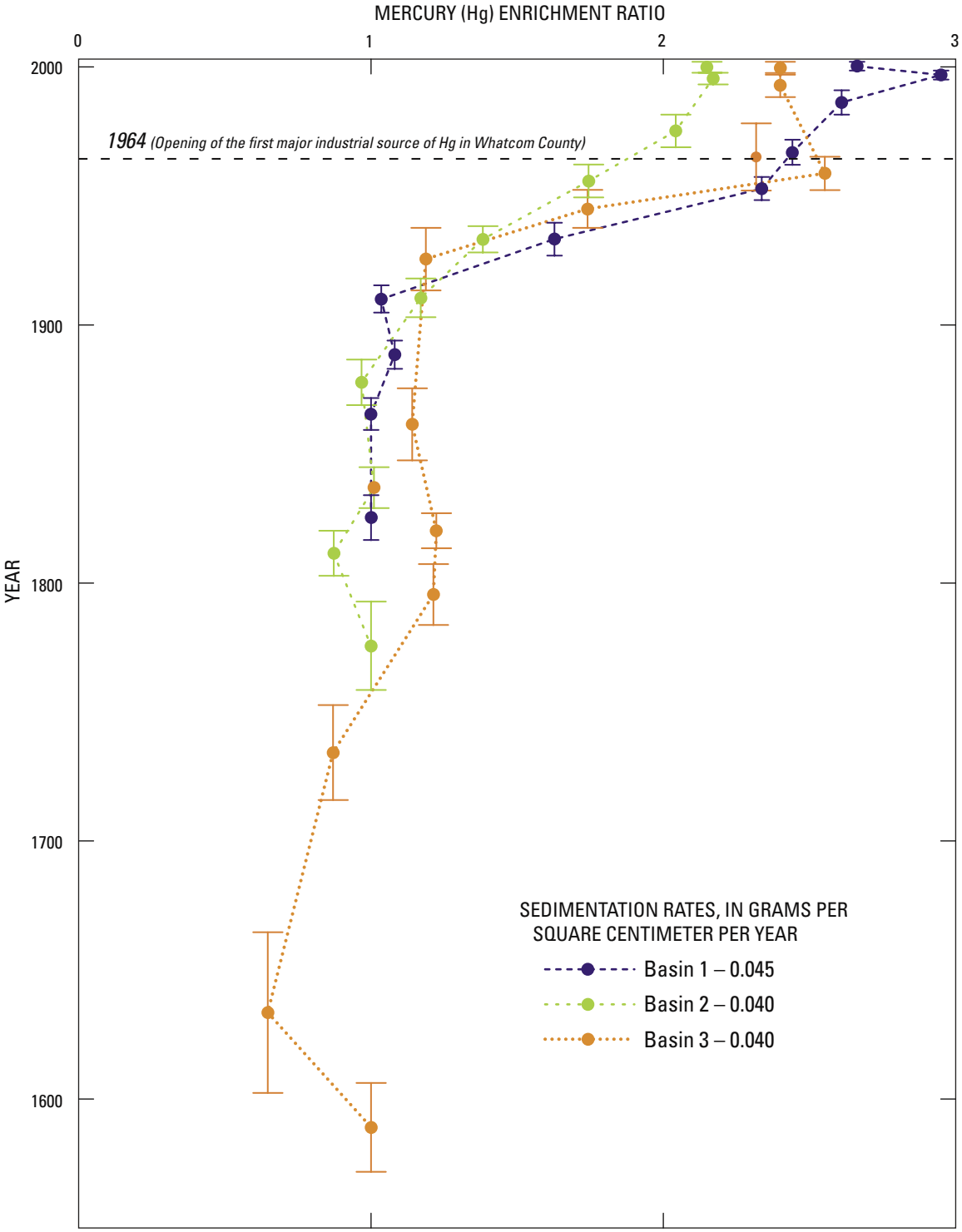
The enrichment ratio for Terrell Lake sediments increased to 1.2 by 1920 and increased to 2.8 in 1958 (fig. 12*B*). Between 1968 and 1989, the enrichment ratio was fairly constant, at a lower value of about 2.6. The enrichment ratio for sediments deposited within the last 8 years was near 2.0.

The Wiser Lake core appeared to show numerous disturbances (table D1), including numerous dramatic shifts in the subsurface distribution of Hg (table 10), making interpretation difficult. Enrichments for Baker Lake could not be calculated because the bottom of the core did not reach a depth that represented dates prior to global industrialization. Hg concentrations decreased from 0.121 mg/kg in 1967 to 0.056 mg/kg after 2000, except for a peak of 0.166 mg/kg in 1977. The peak possibly was a result of adsorption of Hg released from soils covered with water during the filling of Baker Lake as it was expanded to a reservoir in the early 1960s. Hg releases from unsaturated soils have been observed after filling of other reservoirs (Therriault and Schneider, 1998). The fairly gradual decrease in Hg in the core from Baker Lake might reflect either the trend in global deposition or a time lag of 1960s atmospheric Hg deposition reaching the core location as a result of focusing within the reservoir after its filling.

Table 10. Summary of analyses of mercury (Hg) in sediment cores from lakes in Whatcom County, Washington, September 2002

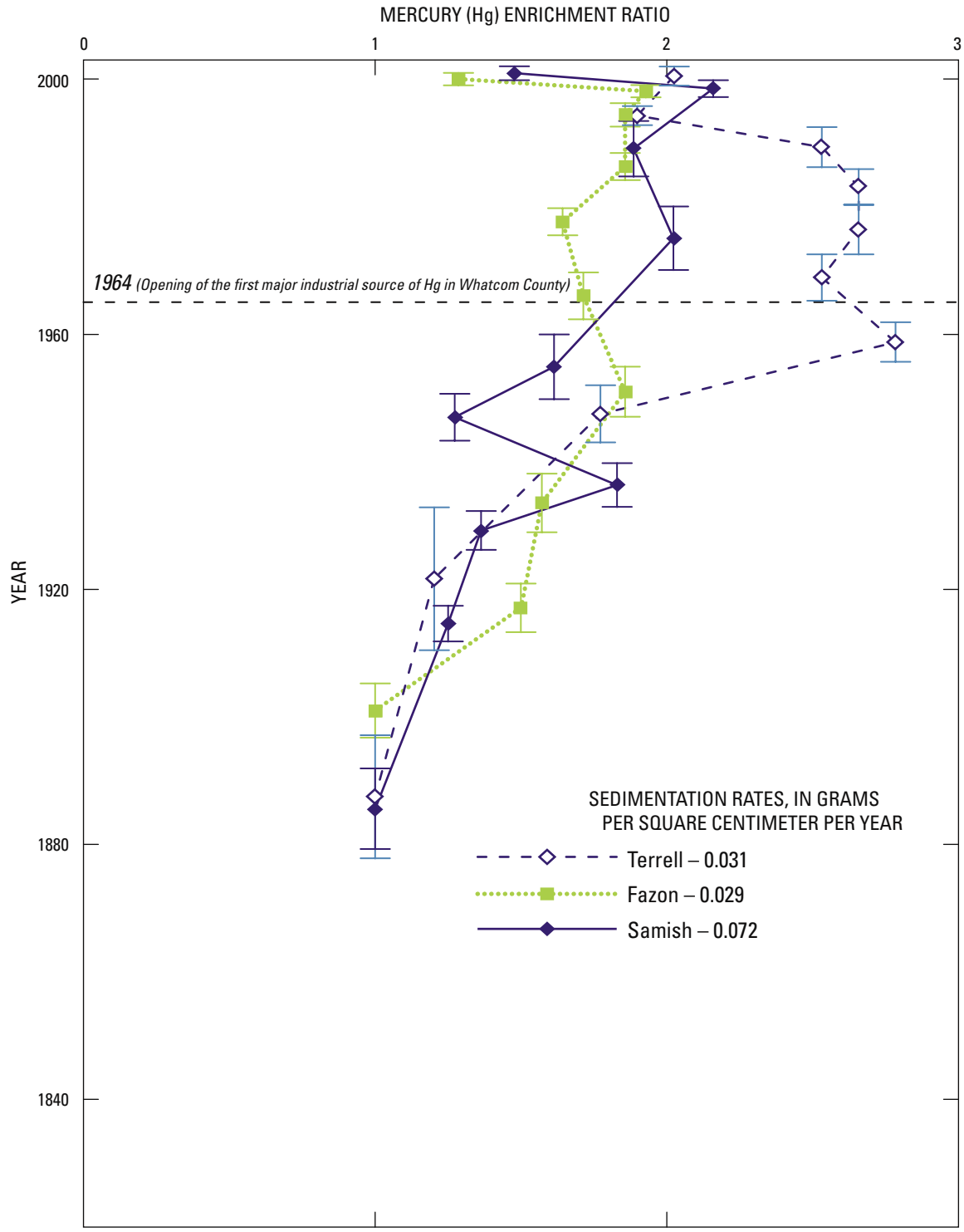
[**Total mercury:** J, estimated value because holding time elapsed. cm, centimeter; mg/kg, dw, milligrams per kilograms in dry weight]

Location	Collection date	Depth interval (cm)	Total mercury (mg/kg, dw)
Lake Samish	09/23/02	0-1	0.13
		1-2	.19
		3-5	.166
		6-8	.178
		9-11	.142
		12-14	.112
		15-17	.161
		18-20	.12
		22-24	.11
		27-31	.088
Fazon Lake	09/24/02	0-1	.18J
		1-2	.27J
		3-5	.26J
		7-9	.26J
		11-13	.23J
		15-18	.24J
		21-24	.26J
		27-30	.22J
		33-36	.21J
		40-44	.14J
Lake Terrell	09/23/02	0-1	.16J
		1-2	.15J
		3-5	.2J
		5-7	.21J
		7-9	.21J
		9-11	.2J
		12-14	.22J
		15-17	.14J
		18-21	.095
		23-25	.079J
Wiser Lake	09/24/02	0-1	0.23J
		1-2	.16J
		3-5	.403J
		7-9	.13J
		11-13	.16J
		14-16	.372J
		20-23	.14J
		26-29	.33J
		32-35	.367J
		40-44	.23J
Baker Lake	09/26/02	0-1	.056
		1-2	.057
		3-5	.065
		6-8	.0728
		9-11	.0762
		12-14	.0636
		15-17	.166
		18-20	.105
		21-23	.121J
		25-27	.1



A. Lake Whatcom

Figure 12. Relation of enrichment ratio of mercury with time in sediment cores from the three basins of Lake Whatcom, and Lake Terrell and Fazon and Wiser Lakes, Whatcom County, Washington, 2002.



B. Lake Terrell and Fazon and Samish Lakes

Figure 12.—*Continued.*

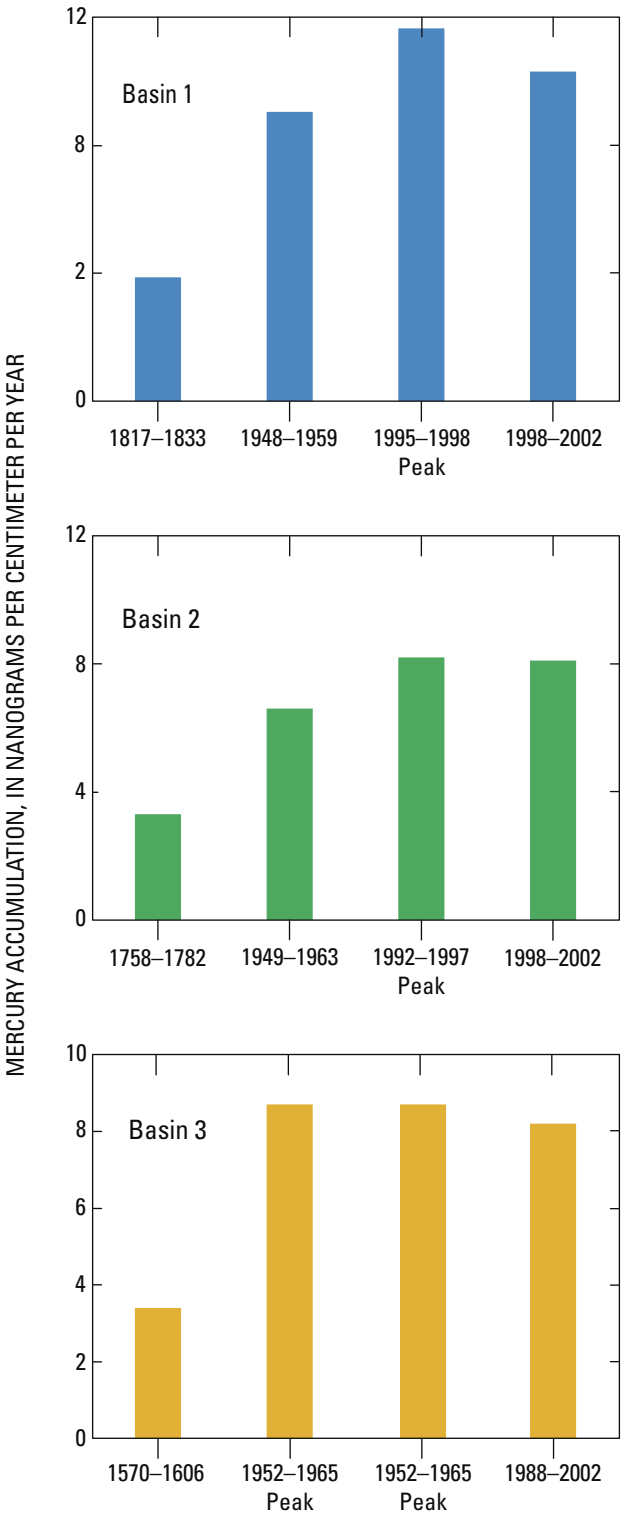


Figure 13. Historical, pre-1964, peak and recent rates of mercury accumulation in core sections from Lake Whatcom, Washington. The dates when the core section was deposited (on the x-axis) were estimated from the mass accumulation rate and the thickness of the core section. Pre-1964 rates of mercury accumulation predate the operation of first major Hg emission source.

The enrichment ratios for the cores from basin 2 of Lake Whatcom, Lake Samish, and Fazon Lake reached a maximum of about 2 in the 1980s. These values are low compared to enrichment ratios as high as 6 in Midwest lakes (Benoit and others, 1994). The core from Lake Samish exhibited decreases in Hg concentrations in the 1-cm section, which represents 2 years (linear sedimentation rate of about 0.5 cm/yr). In contrast, no decrease was observed in the top 1-cm section of the core from basin 2 of Lake Whatcom, which represents 4 years. Basin 2 of Lake Whatcom, Lake Samish, and Fazon Lake have little or no channelized surface water inflow. Basin 2 of Lake Whatcom receives very little surface water inflow from creeks, and Fazon Lake is a catchment basin. Although Lake Samish receives some water from a small creek, the flows from the outflow creek were always greater than surface inflows.

If local atmospheric inputs were a major source of Hg to the lakes, a general relation between the estimated atmospheric deposition from local emission sources and increases in enrichment would be expected. The sum of the estimated peak Hg deposition from atmospheric local sources in the mid-1980s, primarily from incinerators, amounted to 7 percent of the peak sedimentation of Hg of Fazon Lake. In contrast, the sum of the estimated peak Hg deposition from local atmospheric sources to Lake Samish was equal to 1.6 percent of peak Hg sedimentation. Even if the value of the estimated Hg atmospheric deposition were low because of uncertainty in atmospheric residence of Hg species, the enrichment ratio for Fazon Lake would be expected to be much higher than those of Samish Lake if deposition from local Hg sources was the primary source of Hg. Yet, the enrichment ratios for Fazon Lake and Lake Samish were similar. The decrease in Hg emissions over time because of improvements in local air-pollution control devices also are not reflected in decreases in enrichment ratios in the 1980 and 1990s.

Mercury enrichment ratios were from 2.5 to 3.0 for basins 1 and 3 of Lake Whatcom, which receive significant amounts of surface waters. The maximum enrichment ratios for Lake Terrell also were near 3. The timing of the enrichment plateau in Lake Terrell between 1960 and 1990, however, was unique among the lakes that were studied and did not seem to coincide with changes in local emission rates, which indicates a source within the drainage basin. The higher enrichment ratios of basins 1 and 3 of Lake Whatcom, and Lake Terrell, probably are related to the inflow of suspended sediments from their respective drainage basins.

Water Column

On October 11, 2001, seven water samples (table 11) were collected by Exponent, Inc. (Gary Bigham, written commun., 2002) from Lake Whatcom using ultra-clean techniques, and the samples were analyzed for Hg and methyl-Hg using the most sensitive CVAFS technique available. Samples were collected from the surface layer (epilimnion) of each basin and near the bottom layer of basins 1 and 2.

In the surface layer of all three basins, total Hg concentrations ranged from 0.37 to 0.61 ng/L, with the highest surface concentration in the sample from basin 3 (table 11). Dissolved Hg concentrations ranged from 0.29 to 0.66 ng/L, with the highest concentration in the sample from basin 1 (fig. 14). Dissolved methyl-Hg concentrations in the epilimnion were less than or near the detection limit of 0.020 ng/L in all three basins. Total methyl-Hg concentrations were detectable, indicating that methyl-Hg on particles represents between 39 percent and greater than 50 percent of the total methyl-Hg in the epilimnion of basins 1 and 2, respectively. The concentrations and biogeochemistry of Hg in the two samples from basin 3 within (23 m) and just below (30 m) the mixed layer resembled those of epilimnion samples, probably because these samples were collected at least 67 m above the bottom.

The only quality-assurance discrepancy for this data set was the fact that the concentration of dissolved Hg for basin 1 (0.66 ng/L) was higher than total Hg (0.51 ng/L). Particulate Hg concentrations estimated from equation 5 have large uncertainties because they usually are based on small differences between two large numbers, each with relative standard deviations of 10 percent or more. Given these unknown uncertainties, the particulate Hg concentrations in the epilimnion were estimated to be 0.08 ng/L for basin 2 and 0.23 ng/L for basin 3. Recoverable particulate Hg in the epilimnion of basin 2 and 3 represented 22 and 38 percent, respectively, of the total Hg. Although particulate Hg concentrations on the suspended particles are more accurately determined by direct analysis of particles, these data can be used to estimate particulate concentrations by the equation:

$$\text{Particulate Hg (mg/kg = ng/mg)} = \text{Particulate Hg (ng/L)/ TSS (mg/L)}. \quad (6)$$

These calculations result in particulate concentrations ranging from 0.10 to greater than 0.46 mg/kg.

The biogeochemistry of Hg of the near-bottom water samples from 5 meters above bottom (MAB) in basin 1 and 2 MAB in basin 2 was significantly different than those in the epilimnion. Total Hg concentrations were higher than those in the epilimnion in near-bottom samples, partly because of higher TSS concentrations (2.8 to 6.4 mg/L) in the epilimnion. Recoverable particulate Hg concentrations were 0.52 and 1.16 ng/L, respectively, and constituted about 45 percent of the total Hg. The estimated Hg concentrations on the particles ranged from 0.08 to greater than 0.46 mg/kg, which were higher than Hg concentrations of the randomly collected surface sediments, which ranged from 0.014 to 0.23 mg/kg. Although the dissolved Hg concentration in the 5-MAB sample from basin 1 (0.54 ng/L) was similar to those in the epilimnion, the dissolved Hg concentration of the 2-MAB sample from basin 2 (1.4 ng/L) was much higher than that of any other sample.

The most significant result was the much higher portion of the dissolved Hg found as methyl-Hg in near-bottom waters (fig. 14*B*). Dissolved methyl-mercury concentrations in near-bottom samples were from 5 to 50 times higher than those in the epilimnion and constituted 64 and 95 percent of the dissolved Hg in basins 1 and 2, respectively. In October 2001, dissolved oxygen was depleted at depths below 12 and 15 m in basins 1 and 2, respectively (Mathews and others, 2003). In 2001, hydrogen sulfide (H_2S) was present in the bottom waters of basins 1 and 2, indicating that sulfate reduction was occurring. Maximum rates of methyl-Hg production have been shown to occur under reducing conditions when the sulfate reduction rates are low (1-8 nanomoles cm^2/d) (Marvin-DiPasquale and Agee, 2003).

Calculated particulate methyl-Hg concentrations in near-bottom samples in basins 1 and 2 (0.03 and 0.11 mg/kg) were higher than those in the epilimnion (0.016 mg/kg) and an order of magnitude higher than methyl-Hg concentrations in the sediments of basins 1 and 2.

The City of Bellingham (William McCourt, written commun., 2003) collected monthly water samples for Hg analysis by CVAFS at the intake gatehouse at the shore end of the wood-stave intake pipe extending about 366 m into Lake Whatcom for drinking-water withdrawals beginning in November 2002. The concentration of total Hg in six of the eight samples ranged from 0.69 to 0.89 ng/L, and the concentration in two samples was 1.7 ng/L (table 12). These values are within the general range of total Hg concentrations in basin 2 of Lake Whatcom observed by Exponent, Inc. In comparison, the Maximum Contaminant Levels for total mercury in drinking water are 2,000 ng/L (U.S. Environmental Protection Agency, 2002; 2004).

Fish

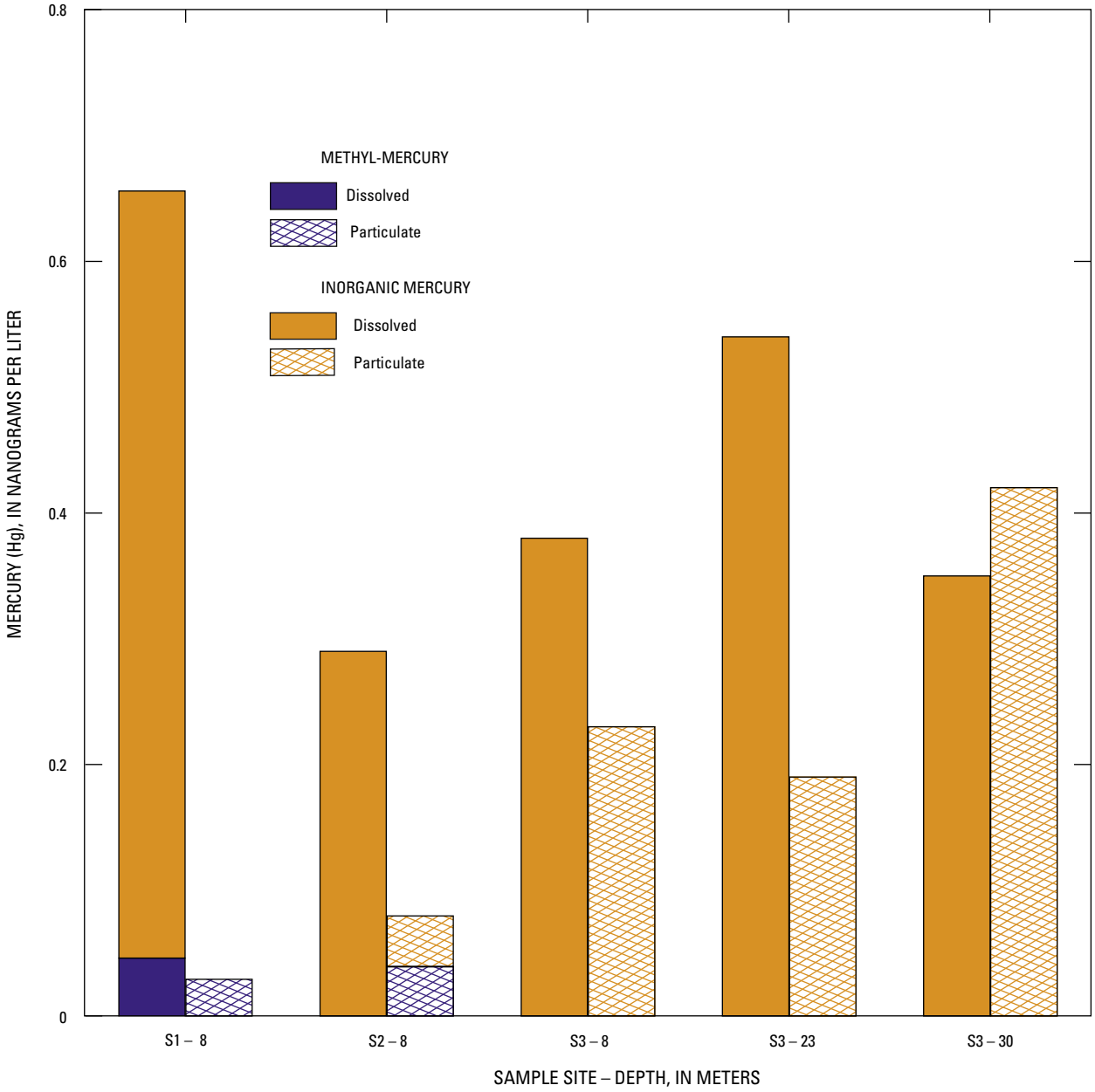
Four studies have investigated Hg concentrations in largemouth bass, smallmouth bass, cutthroat trout, yellow perch, and brown bullhead in lakes in Whatcom County (Serdar and others, 1999, 2001; Seiders, 2003; and this study). The slow elimination of Hg from freshwater game fish results in the accumulation of Hg with age, weight, and length in these fish (Munn and Short, 1997; Gilmour and Riedel, 2000; Serdar and others, 2001). Hg concentrations for specific fish species are examined to determine differences among lakes and among basins within Lake Whatcom that take into consideration the differences in the length of the fish specimens analyzed. The bioaccumulation of Hg in the trophic levels of the food web of Lake Whatcom have been summarized in Mueller and Serdar (2002).

Table 11. Concentrations of total and dissolved mercury (Hg) and methyl-mercury in Lake Whatcom, Washington, October 2001

[**Reported concentrations:** Data from Gary Bigham, Exponent, Inc., written commun., 2002. m, meters; ng/L, nanograms per liter; mg/kg, milligrams per kilogram. >, actual value is greater than value shown; <, not detected at the detection limit shown; NA, not applicable]

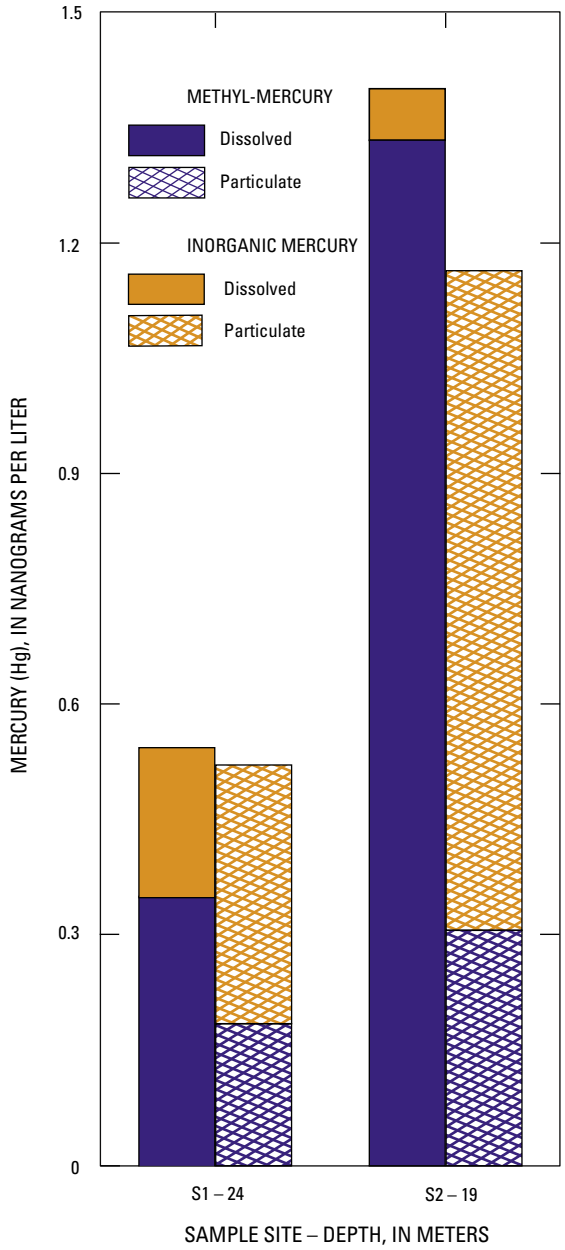
Water body	Latitude	Longitude	Sample ID.	Depth (m)	Meters above bottom (m)	Reported concentrations				
						Total suspended solids (mg/L)	Hg (ng/L)		Methyl-Hg (ng/L)	
							Total	Dissolved	Total	Dissolved
Lake Whatcom										
Basin 1	48º 45.67’	122º 24.69’	SW-S1-111401-8	8	NA	1.8	0.51	0.66	0.075	0.046
			SW-S1-111401-24	24	5	6.4	1.06	.54	.527	.347
Basin 2	48º 44.56’	122º 22.81’	SW-S2-111401-8	8	NA	.8	.37	.29	.039	>.020
			SW-S2-111401-19	19	2	2.8	2.56	1.4	1.63	1.33
Basin 3	48º 41.60’	122º 28.19’’	SW-S3-111401-8	8	NA	<.5	.61	.38	<.020	<.020
			SW-S3-111401-23	23	NA	.8	.73	.54	<.020	<.020
			SW-S3-111401-30	30	67	.8	.77	.35	<.020	<.020

Water body	Depth (m)	Calculated methyl-Hg		Calculated particulate			Calculated particulate methyl-Hg		
		Percentage of total Hg	Percentage of dissolved Hg	ng/L	Percentage of total	mg/kg	ng/L	Percentage of total methyl-Hg	mg/kg
Lake Whatcom									
Basin 1	8	15	7	NA	NA	NA	0.03	39	0.02
	24	50	64	0.52	49	0.08	.18	34	.03
Basin 2	8	11	<7	.08	22	.10	>.019	>49	NA
	19	64	95	1.16	45	.40	.30	18	.11
Basin 3	8	<3	<5	.23	38	>.46	NA	NA	NA
	23	<3	<4	.19	26	.2	NA	NA	NA
	30	<3	<6	.42	56	.5	NA	NA	NA



A. Upper water columns

Figure 14. Estimated partitioning of mercury between the dissolved and particulate phases in five samples in the upper water column and two samples in the near-bottom water column of the three basins of Lake Whatcom, Washington.
Data are taken from calculated concentrations in table 11 from the original data of Exponent, Inc. (Gary Bigham, written commun., 2002).



B. Near-bottom water column

Figure 14.—*Continued.*

Table 12. Concentrations of total mercury (Hg) at the intake gatehouse on Lake Whatcom, Washington

Month	Year	Total Hg (nanograms per liter)
November	2002	0.77
December	2002	.65
January	2003	.79
February	2003	.73
March	2003	.83
April	2003	1.65
May	2003	.89
June	2003	1.67
	Average	1.00
	Standard deviation	.41

Smallmouth Bass

Concentrations of Hg greater than 0.100 µg/g (wet weight) in the tissue of Lake Whatcom smallmouth bass were first reported for two composites of eight fish fillets collected from basins 2 and 3 in the autumn of 1998 (Serdar and others, 1999). A more detailed study of smallmouth bass in May and June 2000 included analyses of fillets from 30 individual specimens collected from the near-shore region of each of the three Lake Whatcom basins (Serdar and others, 2001).

More than 75 percent of the smallmouth bass collected from Lake Whatcom had total Hg concentrations greater than the tissue residue criterion for methyl-Hg of 0.30 µg/g. Chemists analyzed for total Hg, the criterion is based on methyl-Hg concentrations. Literature data suggests that most of the Hg in fish is methyl-Hg (Bloom, 1991). Significant correlations of Hg concentration with fish length (fig. 15) and with age were noted for each of the three basins. The study design included selection of 10 individual specimens within three size classes from each of the three basins. Mean and median concentrations of Hg in fish collected from basin 3 were higher than those from basins 1 and 2 for all three classes (Serdar and others, 2001). Differences in average and median lengths and in average and median ages did not appear to account for the high Hg concentrations in smallmouth bass from basin 3.

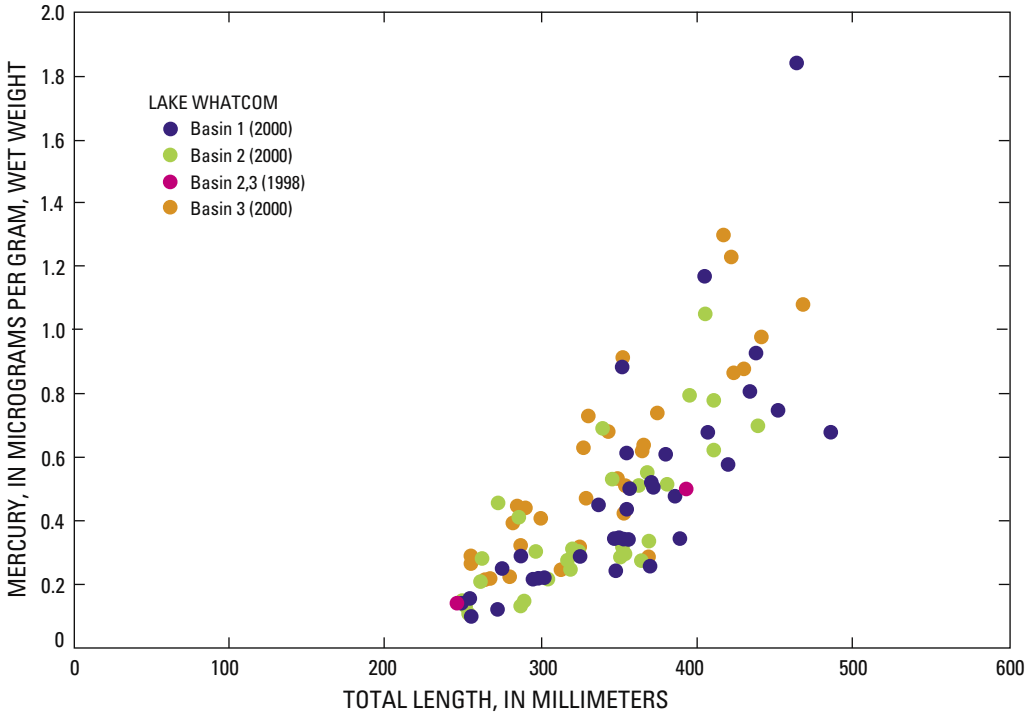


Figure 15. Relation of concentration of mercury in fillets to total length of smallmouth bass in Lake Whatcom, Washington.

In order to investigate whether smallmouth bass collected from basin 3 of Lake Whatcom had statistically higher Hg concentrations than fish from basins 1 and 2, an analysis of covariance was applied to the entire data set of Hg concentrations from smallmouth bass from the lake, taking into account the confounding variable of length. No significant difference (p <0.05) in the slope of the Hg increase with length was found for the three basins. Hg concentrations in fish from basin 3 were higher than those in fish from basins 1 and 2 (p <0.05) when length was considered as a confounding factor, regardless of whether an outlier value from basin 1 was included. A cursory examination did not reveal any obvious relations between the Hg concentration in specific specimens and the proximity of their collection location relative to the density of septic systems within 50 m of the lake. Therefore, the higher concentration of Hg in smallmouth bass from basin 3 than smallmouth bass from basins 1 and 2 must be related to the biogeochemistry of the environmental systems or to differences in the physiology of the fish collected.

Largemouth Bass

Largemouth bass from lakes around the State of Washington (Fischnaller and others, 2003; Seiders, 2003) and from Tennant Lake (table 13) were analyzed for total Hg. For 17 Washington lakes that had sufficient variation in the size and age of fish specimens, the Hg concentrations in largemouth bass from 16 lakes showed a significant correlation with length and age, although a significant correlation with weight was observed for 15 lakes. Concentrations of Hg in fillets of largemouth bass from six lakes in Whatcom County are plotted in figure 16. This data set was subjected to an analysis of covariance, with length being the confounding factor. Because all specimens from Padden Lake except one were within a narrow length range, the data for Lake Padden was not included in the analysis of covariance. Hg concentrations in the fish from Lake Terrell, Lake Samish, Fazon Lake, Tennant Lake, and Wiser Lake were subjected to an analysis of covariance after disregarding the one outlier from Lake Samish. The slope of the Hg concentration versus length relation for Lake Terrell was lower than the slope of the relation for the other four lakes and was not included in the second step of the analysis of covariance. The ad hoc comparison of the results of the analysis of covariance indicates that Hg concentrations in largemouth bass from lakes in Whatcom County were highest in Tennant and Fazon Lakes, with concentrations in Lake Samish being significantly lower (p <0.05). In addition, the Hg concentrations in largemouth bass from Wiser Lake were the lowest and were significantly less than in the fish from Samish Lake.

Table 13. Concentrations of total mercury (Hg) and other characteristics of largemouth bass and yellow perch collected from Tennant Lake, Washington, May 10, 2002

[**Gender:** f, female; m, male. **Age**: Determined by Washington State Fish and Wildlife (WA F&W). **Total Hg**: Analyzed by Frontier Geosciences, Inc. mm, millimeter; g, gram; µg/g, micrograms per gram]

WA F&W scale No.	Sample No.	Gender	Length (mm)	Weight (g)	Age (year)	Total Hg (µg/g)
Largemouth Bass						
1	1	m	327	587	8	0.309
2	2	f	327	680	6	.242
3	3	f	329	652	5	.195
4	4	f	337	677	7	.328
5	6	m	223	154	3	.138
6	7	f	225	154	3	.124
7	8	m	203	117	3	.103
Yellow Perch						
13	14	m	259	243	5	0.126
14	15	m	203	108	3	.628
15	16	m	191	100	3	.0609
16	17	m	200	107	3	.0541
17	18	m	217	128	3	.0664
18	19	m	208	117	3	.0500
19	20	m	205	107	3	.0591
20	21	m	198	103	3	.0615

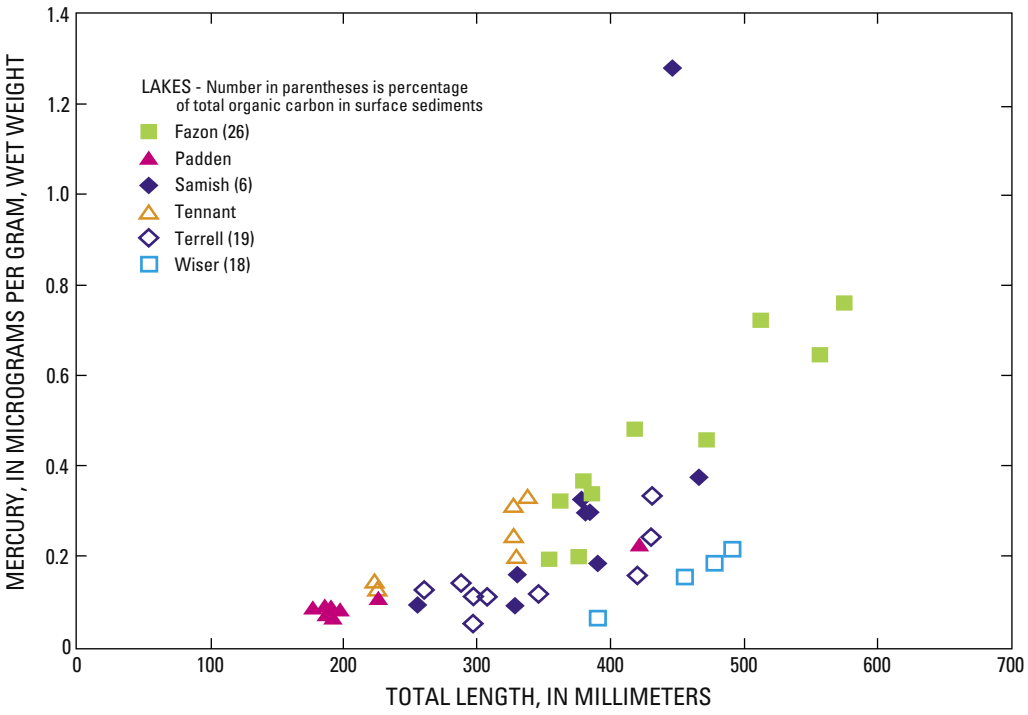


Figure 16. Relation of concentrations of mercury in fillets to total length of largemouth bass in selected lakes of Whatcom County, Washington.

Cutthroat Trout

Cutthroat trout collected from the three basins of Lake Whatcom (Serdar and others, 2001; Seiders, 2003) and Lake Samish (this study) listed in table 14 were analyzed for Hg concentrations (fig. 17). No significant differences in the slope of the relation between Hg concentrations and fish length or the average Hg concentrations adjusted for length among the three Lake Whatcom basins were detected by the analysis of covariance tests. The Hg result for the single Lake Samish specimen that was analyzed fits among those found for Lake Whatcom. Cutthroat trout was the only species examined that exhibited increasingly higher Hg concentrations with decreasing conditioning factor for larger fish (fig. 18). The conditioning factor (mass divided by the total length to the third power) reflects the relative leanness of the fish specimen (Munn and Short, 1997). Fish with a low conditioning factor are lean (low weight for a given length) and may have lost mass without excreting their Hg.

Table 14. Average concentrations of mercury (Hg) and other characteristics of cutthroat trout collected from Lake Whatcom and Lake Samish, Washington, 2000-01

[Values in parenthesis are range. Average concentrations are ± one standard deviation. µg/g, micrograms per gram; mm, millimeters; g, gram]

Lake	Number of fish samples collected	Hg (µg/g)	Length (mm)	Weight (g)	Age range (years)	Reference
Lake Whatcom						
Basin 1	10	0.056 ± 0.014 (0.043 – 0.070)	190 ± 30 (173 – 274)	65 ± 47 (38 – 195)	1 – 2	Serdar and others, 2001
Basin 2	10	0.065 ± 0.031 (0.031 – 0.116)	248 ± 51 (191 – 339)	162 ± 90 (58 – 320)	2 – 3	Serdar and others, 2001
Basin 3	10	0.067 ± 0.025 (0.032 – 0.116)	248 ± 51 (184 – 312)	130 ± 85 (47 – 260)	2	Serdar and others, 2001
	12	0.083 ±0.026 (0.047 – 0.124)	317 ± 40 (241 – 373)	299 ± 80 (183 –453)	3 – 5	Seiders, 2003
Lake Samish	1	0.033	241	136	1	This study

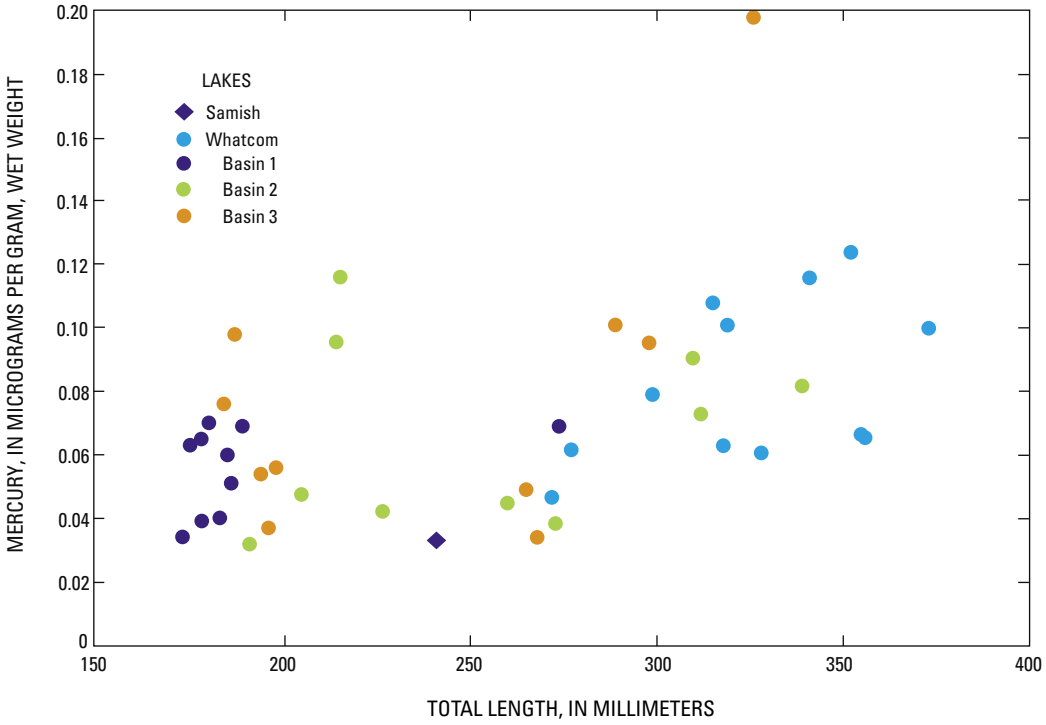


Figure 17. Relation of concentration of mercury in fillets to total length of cutthroat trout in Lake Samish and the three basins of Lake Whatcom, Washington.

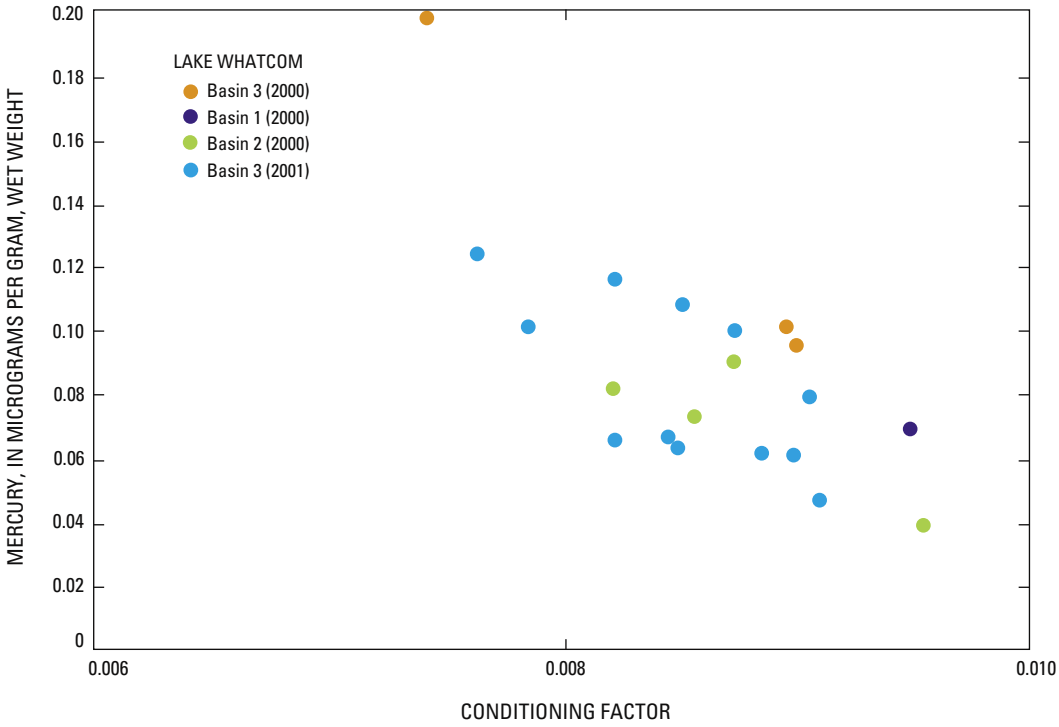


Figure 18. Relation of concentration of mercury in fillets to a conditioning factor for large cutthroat trout in Lake Whatcom, Washington.

Yellow Perch

Hg concentrations and other characteristics of yellow perch collected from Lakes Whatcom, Padden, Samish, and Tennant Lake are summarized in table 15. Relation of Hg concentrations to total length of yellow perch in Tennant Lake and Lake Whatcom are shown in figure 19. Correlations of Hg concentrations with fish length were significant for all three Lake Whatcom basins, and with age in basins 2 and 3 (Serdar and others, 2001). The Hg concentrations in fish from Lake Padden (0.059 ± 0.015 µg/g) and Tennant Lake (0.068 ± 0.024 µg/g) were similar, even though yellow perch from Tennant Lake (126 g) were twice as heavy, on average, as those analyzed from Lake Padden (62 g). Although the average length and weight of yellow perch that were analyzed from Lake Samish and Tennant Lake were similar, the average Hg concentration in fish from Lake Samish was 0.123 µg/g (±0.033 µg/g).

The data from individual specimens from the three basins of Lake Whatcom and Tennant Lake were subjected to an analysis of covariance. The slope of the Hg versus length relation for basin 3 of Lake Whatcom was significantly higher (p <0.05) than those of basins 1 and 2 of Lake Whatcom and Lake Tennant.

Table 15. Average concentrations of mercury (Hg) and other characteristics of yellow perch collected from Lakes Whatcom, Padden, Samish, and Tennant Lake, Washington, 2000-02

[Values in parenthesis are a range. Average concentrations are ± one standard deviation. ND, not determined. µg/g, micrograms per gram; mm, millimeters; g, gram]

Lake	Number of fish samples collected	Hg (µg/g)	Length (mm)	Weight (g)	Age range (years)	Reference
Lake Whatcom						
Basin 1	10	0.124 ± 0.070 (0.047 – 0.307)	190 ± 32 (154 – 257)	79 ± 52 (35 – 210)	2 – 6	Serdar and others, 2001
Basin 2	10	0.174 ± 0.109 (0.070 – 0.374)	227 ± 60 (165 – 333)	200 ± 223 (44 – 680)	2 – 8	Serdar and others, 2001
Basin 3	10	0.290 ± 0.260 (0.085 – 0.869)	219 ± 56 (157 – 310)	142 ± 124 (40 – 440)	2 – 7	Serdar and others, 2001
Lake Padden	8	0.059 ± 0.015 (0.046 – 0.092)	170 ± 26 (150 – 229)	62 ± 34 (39 –141)	1 – 3	This study
Lake Samish	10	0.123 ± 0.033 (0.063 – 0.197)	212 ± 10 (203 – 231)	111 ± 18 (95 – 144)	3 – 4	This study
Tennant Lake	8	0.068 ± 0.024 (0.050 – 0.126)	210 ± 21 (191 – 259)	126 ± 48 (100 – 243)	3 – 5	This study

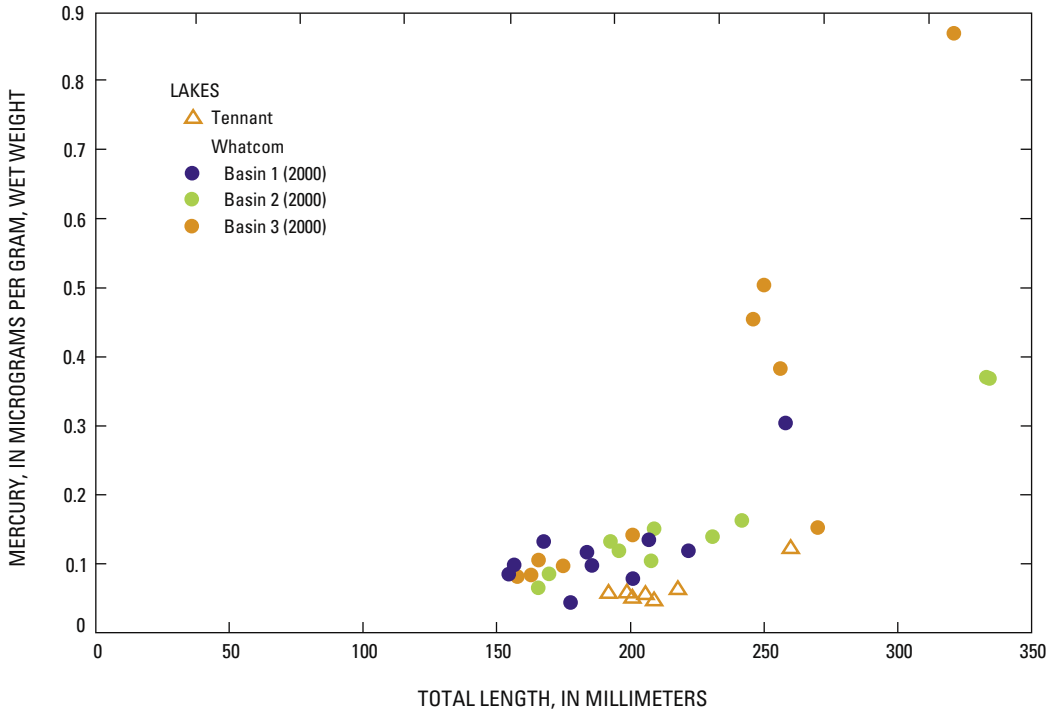


Figure 19. Relation of concentration of mercury in fillets to total length of yellow perch in Tennant Lake and Lake Whatcom, Washington.

Brown Bullhead

Ten brown bullhead (lengths of 186-288 mm) collected from basin 1 of Lake Whatcom were analyzed for Hg (Serdar and others, 2001) (fig. 20). The Hg concentrations of the fillets ranged from 0.031 to 0.121 µg/g and averaged 0.071 µg/g (table 16). One brown bullhead collected from basin 3 (275 mm long) had a similar Hg concentration (0.138 µg/g). Two other brown bullhead from basin 3 were larger (335 and 356 mm) and had much higher mercury concentrations (0.408 and 0.784 µg/g).

Average Hg concentrations were 5-fold less in eight brown bullhead collected from Lake Terrell (lengths of 264-310 mm) than those from basin 1 of Lake Whatcom (table 16). Average Hg fillet concentrations were only 0.014 µg/kg (ranging from 0.0063 to 0.023 µg/g) in 10 brown bullhead from Wiser Lake (lengths of 315-404 mm). Different analytical techniques used by the two studies could be partly responsible for the differences in Hg concentrations of brown bullhead. However, lack of analyses of standard reference material from both studies precluded the assessment of procedures.

Other Species

Signal crayfish, pumpkinseed, and kokanee from Lake Whatcom also were analyzed for Hg (Serdar and others, 2001). Correlations of Hg concentration with length were not significant for the three species, nor with age for pumpkinseed and kokanee. Hg concentrations in the tail muscle of signal crayfish generally were from 0.020 and 0.050 µg/g wet weight. Hg concentrations of pumpkinseed specimens were highly variable for basin 1 (ranging from 0.040 to 0.230 µg/g) and basin 3 (ranging from 0.030 to 0.28 µg/g), compared to the range from 0.040 to 0.090 µg/g for basin 2. Hg concentrations in kokanee collected in 2000 were similar to those in specimens collected in 1998 (Serdar and others, 1999); basin-wide average Hg concentrations in kokanee from the three basins of Lake Whatcom were within 30 percent of each other.

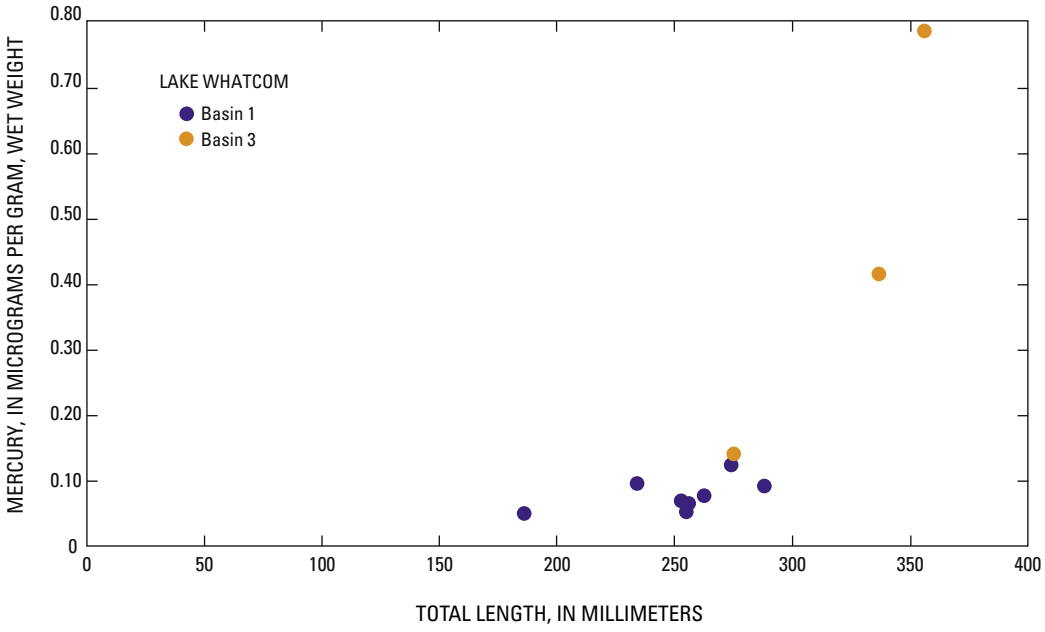


Figure 20. Relation of concentration of mercury to total length of brown bullhead in Lake Whatcom, Washington.

Table 16. Average concentrations of mercury (Hg) and other characteristics of brown bullhead collected from Lakes Whatcom and Terrell and Wiser Lake, Washington, 2000-01

[Values in parenthesis is a range. Average concentrations are ± one standard deviation. µg/g, micrograms per gram; mm, millimeters; g, gram]

Lake	Number of fish samples collected	Hg (µg/g)	Length (mm)	Weight (g)	Reference
Lake Whatcom					
Basin 1	10	0.071 ± 0.026 (0.031 – 0.121)	251 ± 27 (186 – 288)	0.232 ± 0.068 (0.087 – 0.032)	Serdar and others, 2001
Basin 3	3	0.443 ± 0.325 (0.138 –0.785)	322 ± 42 (275 – 356)	0.572 ± 0.271 (0.269 – 0.790)	Serdar and others, 2001
Lake Terrell	8	0.0186 ± 0.0072 (0.0134 – 0.0335)	282 ± 14 (264 – 310)	294 ± 54 (232 – 404)	This study
Wiser Lake	10	0.0141 ± 0.0065 (0.0063 – 0.023)	368 ± 24 (315 – 404)	795 ± 133 (483 – 987)	This study

Biogeochemical Behavior of Mercury in Lake Whatcom

Present-day (2003) concentrations of Hg in various media and the observed relations provide a qualitative understanding of the behavior of Hg in Lake Whatcom. If no Hg is lost from the water column of Lake Whatcom, the volume-weighted total Hg concentration of the water inputs (regional and local atmospheric deposition, ground-water discharge, and tributary discharge) would equal the volume-weighted total Hg concentrations of the water outputs (Whatcom Creek and drinking-water withdrawals). The annual volume-averaged Hg concentrations of precipitation from the two nearest samplers of the national Mercury Deposition Network were greater than 6 ng/L. The average Hg concentrations of tributaries discharging to Lake Whatcom ranged from 3.2 to 5.8 ng/L. No data with low detection limits for Hg exist that could be used to assess the general concentration of Hg in ground water discharging directly into Lake Whatcom. In contrast, the average concentrations of total Hg in both the surface waters of Lake Whatcom (table 11) and of water pumped from Lake Whatcom for public supply (table 12) were significantly lower (1 ng/L or less) than any known inputs of water (tributary or regional precipitation). Because the Hg concentrations of the identifiable inputs of aqueous Hg to Lake Whatcom were significantly higher than the concentrations in the water column of the lake (the source of the removed water), total Hg must have been lost from the water column of Lake Whatcom.

The likely losses of total Hg from the water column of Lake Whatcom would include degassing of dissolved vaporous Hg (evasion), physical settling of Hg attached to particles of suspended sediment in tributary inflows, and chemical adsorption of dissolved Hg onto terrestrial or detrital particles that then settle to the lake bottom. Evasion of dissolved vaporous Hg (Hg^o) was determined to be 7 percent of the total atmospheric deposition of Hg in another lake study (Watras and others, 1996), and thus is not considered an important loss term in the Hg mass balance of Lake Whatcom. In contrast, the strong relation between Hg and the percentage of fines in the surface sediments throughout all three basins (fig. 10) indicates that Hg is transported to the sediments primarily on fine-grained particles. This observation could be attributed either to the physical winnowing effect on the original tributary suspended matter as the coarser material settle out near the mouths of Lake Whatcom tributaries (as seen in fig. 10) or to the geochemical adsorption of dissolved Hg onto small organic and inorganic particles in the water column preceding settling. Assessing the relative importance of the physical settling of the original tributary particles versus the settling of particles on which additional Hg from that water column has adsorbed requires a well-constrained mass balance of particles.

Bioaccumulation of methyl-Hg through the food web is the primary pathway of Hg to the tissue of fish. Although methyl-Hg is present in both the bottom waters (fig. 14*B*) and sediments (fig. 11) of Lake Whatcom, there is insufficient information to develop an accurate conceptual model that could account for bioaccumulation of Hg in fish. In many lakes, total Hg in the sediments is correlated with TOC (Watras and others, 1996; Ullrich and others, 2001). Fine-grained sediments seem to be more important than TOC in transporting Hg to the surface sediments of Lake Whatcom (fig. 10). For core samples in the deeper parts of the three basins, surface Hg concentrations varied by only 10 percent, whereas TOC varied by a factor of 2. Production of methyl-Hg occurs primarily at the oxic/anoxic interface, whether in the water column or in the sediments. The production rate of methyl-Hg depends on temperature, pH, the availability of organic carbon, and the rate of sulfate reduction. The distribution of methyl-Hg in the sediments of the three basins of Lake Whatcom follows the same trend as TOC:

basin 1 > basin 2 > basin 3.

The increasing concentration of methyl-Hg in the sediments with increasing TOC concentration may be a result of an increasing rate of sedimentary diagenesis. Increasing TOC concentrations in the sediments result in increasing rates of sulfate reduction. In turn, the rate of sulfate reduction has been shown to be a key factor in controlling the rates of methylation of Hg in the sediment column (Marvin-DiPasquale and Agee, 2003). An alternative explanation suggests that the relationship between methyl-Hg and TOC is not related to the methylation rate but to the characteristics of the sediment. Methyl-Hg is known to have a higher binding affinity for organic-rich sediments. Therefore, this alternative explanation suggests that organic-rich sediments will more efficiently sequester the methyl-Hg in the interstitial porewaters of the sediments regardless of the Hg methylation rate.

Data on the concentration and speciation of Hg in near-bottom and shoreline waters of basin 3 would lead to a better understanding of Hg cycling in the largest basin of Lake Whatcom and might help to explain the high concentrations of Hg in smallmouth bass collected in basin 3 relative to Hg concentrations in smallmouth bass collected in basins 1 and 2. Three observations provide some insight into the pathways of methyl-Hg to fish. The first set of observations relates to the input of methyl-Hg from sources of water to Lake Whatcom. The total methyl-Hg concentrations in Anderson Creek (0.15 ng/L) and Austin Creek (0.13 ng/L), which drain into basin 3, were higher than those in Silver Beach Creek, which drains into basin 1 (about 0.05 ng/L). However, methyl-Hg concentration was highest in the wetlands of the settling pond on Park Place in basin 1 (0.25 ng/L), but flow data were

not available for this source. In contrast, the majority of the nearshore (within 50 m of the lake) onsite septic systems are located within basin 3. Human waste and residential wastewater are known to contain small concentrations of Hg. It is expected that a higher proportion of the Hg discharged from these systems will be in the methyl-Hg form relative to other human-related sources of Hg. The lack of ground-water flow and Hg chemistry data precludes the evaluation of the loading of methyl-Hg from ground water, and whether it is from natural or anthropogenic sources. Secondly, the higher calculated particulate Hg in the surface waters of basin 3 of Lake Whatcom (0.2 to 0.4 ng/L) than in the other basins (0.1 ng/L or less) could have led to a higher uptake of Hg by detrital feeding organisms that form the base of the food web (fig. 14*A*). Concentrations of Hg on the particles in the upper water column also were calculated to be higher in basin 3 (0.2 to 0.5 mg/kg) than in basin 2 (0.1 mg/kg) (table 11). Finally, the higher ratio of Hg to TOC in surface sediments of basin 3 (4.3 ± 2.0 mg of Hg per kilogram of TOC) relative to basin 1 (1.9 ± 0.6 mg of Hg per kilogram of TOC) also could explain the higher concentrations of Hg in the tissue of smallmouth bass. In order for smallmouth bass or its prey to grow, they must eat organic carbon for both tissue growth and respiration. If that organic carbon enters the food web through benthic processes, the Hg:TOC signal in the sediments would be magnified up the food web as the major fraction of the organic carbon (that is, energy) is used for respiration. Thus, smallmouth bass and its prey would consume more Hg from the given amount of organic carbon of basin 3 than from the same amount of consumed organic carbon originating from the sediments of basin 1.

Hg concentrations in smallmouth bass and yellow perch collected from the near-shore regions of basin 3 had higher Hg concentrations than other basins, but did not show any correlation with conditioning factor. In contrast, the native cutthroat trout did not exhibit higher Hg concentrations in basin 3 relative to basins 1 and 2, but the Hg concentrations were correlated with conditioning factor. Perhaps this observation is related to the home range, foraging behavior, and physiology of the various species.

The concentration of total Hg in the sediments throughout Lake Whatcom is controlled by the Hg input to the drainage basin of Lake Whatcom and the grain-size distribution of the particles, which in turn is controlled by the physical processes of transport and settling acting on the particles. However, the presence of Hg in the water and sediment columns is not the limiting factor controlling Hg uptake by biota. Organic carbon plays a major role in controlling the transfer of Hg to the biota. The oxidation of organic matter in the sediments drives the reduction-oxidation processes that produce methyl-Hg in the sediment. Some of this methyl-Hg diffuses out of the sediment, as seen in the bottom waters in basins 1 and 2 (fig. 14*B*), and can be directly absorbed by the biota through respiration. The Hg associated with particulate organic carbon in the sediments or in the water column that enters the food web will be magnified at higher trophic levels.

Mass Balances of Mercury in Lake Whatcom

The annual Hg inputs to Lake Whatcom were calculated using 2002 and 2003 data (table 17). The estimate for annual discharge of Hg from the tributaries was 179 g/yr (table 8); this is an underestimate, however, because storm events were not represented. For non-local atmospheric sources, the annual deposition rate of 6.2 μg/m^2 of Hg (based on the average 1998–2002 for the Seattle Station in table 2) was applied over the 20 km^2 surface of Lake Whatcom (table 1) to yield a non-local atmospheric deposition of 124 g/yr. Known local sources of atmospheric Hg in Whatcom County in 2003 include the Bellingham sewage-sludge incinerator and the two oil refineries. Atmospheric deposition calculations that were developed for the sewage-sludge incinerator are categorized as having the lowest confidence because Hg speciation assumptions were based on indirect knowledge of the chlorine content of air chemistry (table 5). When the maximum estimated deposition for the three basins from the City of Bellingham sewage-sludge incinerators, which operated at 33 percent of total capacity in 2003 (table 5), was applied to the surface areas of the three basins (fig. 1), a total input of 0.27 g/yr was estimated for local atmospheric sources. Deposition calculations were not attempted for the oil refineries because of the lack of data on the Hg species emitted by refineries. Likewise, the lack of precise Hg concentrations in ground water precluded an assessment of Hg inputs from direct ground water discharge to Lake Whatcom. Thus, the recent total input of Hg to Lake Whatcom was estimated to be 303 g/yr.

Table 17. Summary of recent annual inputs of sediment and mercury (Hg) from known sources to Lake Whatcom, Washington, 1998-2003

[Mg/yr, megagrams per year; g/yr, grams per year]

Inputs	Data (years)	Sediment sources (Mg/yr)	Hg sources (g/yr)	Percentage of known total Hg
Tributaries	2002-2003	272	179	59
Non-local atmospheric	1998-2002	negligible	124	41
Local atmospheric	2003	negligible	.27	.1
Known total		272	303	

In order to verify the sources of Hg to Lake Whatcom and understand the importance of any sources that have not been assessed or identified, the total sources of Hg must be balanced by the sinks of Hg. The foundations of a mass balance for any constituent in any lake are the mass balances of water and particles. The City of Bellingham has an extensive understanding of the water balance in Lake Whatcom, as a result of providing drinking water to its customers and replenishing water to Lake Whatcom through the Nooksack Diversion. However, little is known about the sources of particles to the lake sediments and their transport among the basins. A secondary product of a recent study of tributary Hg loading to Lake Whatcom (Norton, 2004) was the first lake-wide investigation of suspended-sediment concentrations in the tributaries to the lake. Using the TSS data from Norton (2004), the annual input of suspended sediment to Lake Whatcom was estimated to be 272 Mg in 2002-03. The sinks of suspended matter from Lake Whatcom include sedimentation within the basins, and particles suspended in the water withdrawn from the lake. As a first approximation of the sinks of particles, the sedimentation rates obtained from the cores in each of the three basins of the lake were used to calculate the sink of particles from the water column. If the basin sedimentation rates from the cores are assumed to represent the average sedimentation rate for the entire area of the basin, then a total sedimentation of 8,225 Mg is calculated for Lake Whatcom.

In most lakes, topographic relief of the lake bottom results in an uneven distribution of sediment accumulation. The accumulation of fine-grained sediments in the deep areas of lakes at rates greater than the average sedimentation for a lake is called sediment focusing. For a specific core location, a focusing factor is defined as the sediment accumulation rate at the site divided by the sediment accumulation rate averaged over the whole lake bottom. Because there was not enough material in each 1-cm section of the cores to determine grain size, the geomorphology of the core sites could not be related to other parts of the lake where sediment grain size has been measured. However, sediment focusing in the basins of Lake Whatcom was estimated by comparing the ^{210}Pb inventory in the upper section of each core with the atmospheric deposition of ^{210}Pb measured in western Washington (appendix D). The average sedimentation rate for each basin was then adjusted by dividing the sedimentation rate for the core taken from the deep areas of each basin by its focusing factor. The total sedimentation rate in Lake Whatcom then was estimated by summing the total sedimentation in each basin, which was calculated by multiplying the focus-adjusted average sedimentation rate for each basin by its surface area. After adjusting the sedimentation rates in the three cores for sediment focusing, the total annual sedimentation within Lake Whatcom was estimated to be 1,243 Mg, which was five times the estimated fluvial input. The total sinks of Hg from the Lake Whatcom water column cannot be calculated without a better understanding of the mass balance and dynamics of particles within Lake Whatcom. Likewise, the estimated total mass loading of sources of Hg to Lake Whatcom (table 17) cannot be verified until the sources of Hg are balanced by the total sinks of Hg.

Suggestions for Future Studies

The understanding of the sources and behavior of Hg in Lake Whatcom and the other freshwater lakes of Whatcom County is incomplete because of a lack of critical data and other information. Although the data collected since the inception of this project provide an initial understanding of the sources of Hg to Lake Whatcom, additional data are needed to reduce the uncertainty in and to identify processes that control the concentration of Hg in fish, water, and sediments of Lake Whatcom and other lakes in Whatcom County. This additional data could be obtained in study efforts with the following objectives:

A. Developing a better understanding of the sources, sinks, and dynamics of sediment and particulate Hg in Lake Whatcom. This would require:

   1. Determining the TSS and concentrations of particulate and dissolved Hg in the tributaries of Lake Whatcom during the rising phase of the hydrograph of storm events;

   2. Collecting a series of sediment samples from the Nooksack Diversion and analyzing Hg in the numerous size fractions to assess whether differential settling and/or focusing of fine-grained sediments in the deep basin(s) of Lake Whatcom contribute to enriched Hg sediment concentrations; and

   3. Resampling the sediments at the location of the cores and performing grain-size analyses to determine how the core sites relate to the overall sediment texture map of Lake Whatcom. If a relation between grain size and sedimentation rates can be established, then the total sedimentation of the lake can be quantified more easily.

B. Better quantifying the sources of Hg to Lake Whatcom. This would require:

   1. Obtaining better Hg emissions and speciation data for current Hg sources in Whatcom County, including the City of Bellingham sewage sludge incinerator, the two refineries and the aluminum smelter when operating. Better emissions estimates of Hg from the industrial sources would be achieved by Hg emissions testing rather than estimates based on emission factors. In lieu of sophisticated Hg speciation measurements in the stack gases, the speciation of Hg could be estimated from better air chemistry data (e.g., chloride and sulfide concentrations) in the stack emissions. For the City of Bellingham sewage sludge incinerator, chloride air chemistry could be estimated based on the chloride concentration of sewage sludge that is incinerated.

   2. Using sensitive analytical techniques, determine Hg concentrations in ground water entering Lake Whatcom in a manner that would also assess the impact of Hg from septic systems.

C. Developing a better understanding of the biogeochemistry of Hg in Lake Whatcom and its effect on the bioaccumulation of fish. This would require:

   1. Measuring the concentrations of dissolved, particulate, and methyl-Hg, and other important biogeochemical parameters (dissolved organic carbon, hydrogen sulfide, ammonium, pH, and TSS) in the water column of the deeper parts of basin 3 of Lake Whatcom; and

   2. Evaluating the methylation of Hg in habitats critical to fish growth and survival, especially if near-shore or benthic habitats are critical to the fish species of interest.

Summary and Conclusions

Mercury (Hg) in the lakes of Whatcom County originated from atmospheric deposition of global sources of Hg, from atmospheric emissions of Hg from local incinerators and a chlor-alkali plant, and from erosion of drainage basin soils. Deposition of Hg from global sources has been decreasing since the 1980s.

The operators of four major air-emission sources of Hg in Whatcom County that were examined in detail improved the control of their air emissions during the lifetime of the operations through changes in operating procedures or improvements in air-pollution control devices. Hg speciation data were not available from the four local air emission sources and source-specific Hg emissions data were available only for limited periods of the operational stages of the facilities. A simple air-deposition model with an analytical solution was developed that allowed relative comparisons of the Hg deposition to lakes or basins within a lake, based on measured or estimated Hg emissions, speciation of Hg in the emission source, prevailing annual wind pattern for the region, location of lakes or basins with respect to the sources, and residence times of the particulate, reactive, and vaporous Hg species in the atmosphere.

The results of the estimates of atmospheric deposition derived from the model indicated that the three lakes north of Bellingham (Lake Terrell, Wiser Lake, and Fazon Lake) received the largest deposition of Hg from local air emission sources, primarily from the municipal waste incinerators during early stages of operation, when air pollution control was minimal (1974–91). The City of Bellingham sewage sludge incinerator, which was modernized in 1993, and the two oil refineries are the only known major local air-emission sources operating in the region in 2003. Of the lakes modeled, the estimates of the air-deposition model indicated that basin 1 of Lake Whatcom is most affected by emission from the municipal sewage sludge incinerator. However, the estimated deposition of Hg to basin 1 from the sewage sludge incinerators was much less than the deposition to basin 1 from the municipal waste incinerators when they were operating.

The chlor-alkali plant in the City of Bellingham began operation in 1964 and emitted Hg from three major sources until its closure in 2000: two vented air streams from the two sides of the electrolysis process (fume air and hydrogen) and fugitive emissions from the cell room. By 1979, the chlor-alkali plant had undergone a major expansion and modernization of its air-pollution control devices. Because basin 1 of Lake Whatcom is in the path of the prevailing winds passing through Bellingham, the deposition of Hg from any of the three emission sources from the chlor-alkali plant would be greatest for basin 1, with basin 2 of Lake Whatcom and Fazon Lake receiving 4-fold and 5-fold less deposition, respectively. Emissions of Hg from the fume air vent prior to the modernization in the late 1970s were likely in a chemical form that was deposited regionally, and this emission was virtually eliminated after modernization. Recent measurements of fugitive emissions of Hg from Hg hydrolysis cell rooms of chlor-alkali plants were 2- to 5-fold lower than estimates codified in Federal regulations. These modern measurements indicate that Hg escaping from cell rooms primarily was in the vaporous form, which adds to the global atmospheric inventory and does not settle near the source to any great extent. Estimates of maximum deposition of Hg to basin 1 from fugitive emissions from the cell room of the chlor-alkali plant were higher than the maximum estimates from the City of Bellingham sewage sludge incinerator before modernization in 1993. The emissions of Hg from the hydrogen gas stream

of the chlor-alkali plant were comparable to the fugitive emissions from the cell room. However, the hydrogen gas stream was not normally sent directly up the stack into the atmosphere as in the emission test, but the hydrogen gas was used as a fuel source for heating the lignin spray dryers. This plant configuration is unique among chlor-alkali plants in the United States and the deposition of Hg to nearby lakes could not be estimated because the speciation of the emitted Hg could not be evaluated.

Hg concentrations in largemouth bass and the enrichment of Hg in the sediments of six lakes in Whatcom County over time did not seem to be related to the estimated deposition from local emission sources. For instance, the estimated Hg depositions to Wiser and Fazon Lakes from local Hg sources were similar, but Hg concentrations in largemouth bass from Fazon Lake were much higher than bass from Wiser Lake for a given length. Likewise, the estimated deposition of Hg to Fazon Lake from local air emissions was slightly higher than those to Lake Terrell, yet maximum enrichments in Hg in the sediments of Lake Terrell from 1950 to 1990 were significantly higher than those of Fazon Lake. The enrichment in Hg in the sediments of a lake was dependent on the importance of surface-water inflows to the lake, rather than on estimated deposition of Hg from local air emissions. Lake Samish and Fazon Lake have little or no surface-water input and they exhibit similar enrichments of Hg in their sediments.

The recent high-sensitivity Hg data from the City of Bellingham, Exponent, Inc., and Washington State Department of Ecology provide some insight into the behavior and sources of Hg in Lake Whatcom. Because the total Hg concentration of 1 nanogram per liter (ng/L) in the surface water of Lake Whatcom is considerably smaller than that in either the volume-averaged precipitation (from a Seattle site) or in tributary inflow water, Hg is being removed from the water column of Lake Whatcom and deposited within the sediments. Sedimentation of Hg in Lake Whatcom is controlled by the settling of fine-grained particles, either through the physical process of size fractionation or subsequent to chemical adsorption. In the low-oxygen, near-bottom waters of basins 1 and 2 during autumn 2001, methyl-Hg was the dominant form of Hg. The lack of Hg data from the bottom waters of basin 3 precludes addressing any processes that resulted in the higher Hg concentrations in smallmouth bass and yellow perch in basin 3 relative to those in basins 1 and 2. However, the cycling of organic matter within the lake is ultimately controlling Hg concentrations in fish tissue.

A lake-wide mass balance of Hg was attempted using data collected between June 2002 and May 2003. The Hg loadings in known sources of water to Lake Whatcom (precipitation and 10 tributaries) amounted to 303 grams per year (g/yr), with tributaries contributing 59 percent and direct precipitation contributing 41 percent. Although the concentration of total Hg in the water diverted from the Middle Fork Nooksack River through Anderson Creek was less than the average Hg concentration in the other creeks in the drainage basin, the total loading of Hg from the Nooksack Diversion was equal to the sum of other surface-water sources because of the large volume of water diverted. The estimated inputs of Hg to Lake Whatcom cannot be verified until they are balanced by the sinks of Hg to the sediments and by water withdrawals. The ability to develop accurate estimates of the sinks of Hg is hampered by an incomplete understanding of the sources, sinks, and dynamics of particles of Lake Whatcom.

References Cited

Association of Metropolitan Sewerage Agencies, 2000, Evaluation of domestic sources of mercury, August 2000: Washington, D.C., accessed September 2, 2003, at URL http://www.amsa-cleanwater.org/pubs/mercury/mercury.cfm

Babcock, R.S., and Kolby, N.I., 1973, Distribution of mercury in sediments of the Nooksack River Drainage: Northwest Science, v. 47, no. 3, p. 180-184.

BEK Engineering & Environmental Inc., 2002a, Phase II Hydrogeologic investigation: Y-road landfills, Whatcom County, Washington: Ferndale, WA, 21 p. plus appendixes.

BEK Engineering & Environmental Inc., 2002b, Annual report 2002 landfill monitoring test results, Hilltop Woodwaste Landfill: Georgia-Pacific West, Inc., Ferndale, WA, 10 p. plus appendixes.

BEK Engineering & Environmental Inc., 2002c, Annual report 2002 landfill monitoring test results, Airport Woodwaste Landfill, Georgia-Pacific West, Inc., Ferndale, WA, 10 p. plus appendixes.

Benoit, J.M., Fitzgerald, W.F., and Damman, A.W.H., 1994, Historical atmospheric mercury deposition in the Mid-Continental U.S. as recorded in an Ombrotraphic Peat Bog, in Watras, C.J. and Huckabee, J.W., eds., Mercury Pollution: Integration and Synthesis: Ann Arbor, MI, Lewis Publishers, p. 187-202.

Biester, H., Müller, G., and Schöler, H.F., 2002, Estimating distribution and retention of mercury in three different soils contaminated by emissions from chlor-alklai plants, Part I: Science of the Total Environment, v. 284, p. 177-189.

Bloom, N.S., 1991, On the chemical form of mercury in edible fish and marine invertebrate tissue: Canadian Journal of Fisheries and Aquatic Sciences, v. 49, p. 1010-1017.

Bortleson, G.C., Dion, N.P., McConnell, J.B., and Nelson, L.M., 1976, Reconnaissance data on lakes in Washington, v. 1: Olympia, WA, Washington State Department of Ecology, no. 43 (1), 248 p.

Boutron, C.F., Vandal, G.M., Fitzgerald, W.F., and Ferrari, C.P., 1998, A forty year record of mercury in central Greenland snow: Geophysical Research Letter, v. 25, no. 17, p. 3315-3318.

Cazzaro, F., 2002, Industrial contamination by mercury: Present State and Perspectives, *in* Integrated Approach for the Remediation of Polluted Sites, Trieste, Italy, April 22-23, 2002.

Dion, N.P., Bortleson, G.C., McConnell, J.B., and Innes, J.K., 1976, Data on selected lakes in Washington: Part 5: Olympia, WA, Washington State Department of Ecology, no. 42, 108-111 p.

Ebinghaus, R., and Kruger, O., 1996, Emissions and local deposition estimates of atmospheric mercury in North-Western and Central Europe, *in* Baeyens, W., Ebinghaus, R., and Vasiliev, O., eds., Global and Regional Mercury Cycles: Sources, Fluxes, and Mass Balances: Boston, Massachusetts, Kluwer Academic Publishers, Series 2, Environment, v. 21, p. 135-159.

Expert Panel on Mercury Atmospheric Processes, 1994, Mercury atmospheric processes: A Synthesis Report: Palo Alto, California, 94304, Electric Power Research Institute TR-104214, 23 p.

Falley, R.T., 1974, Distribution and transport of mercury within the Nooksack River Drainage, Whatcom County, Washington: Bellingham, WA, M.S. Thesis, Western Washington University.

Fischnaller, S., Anderson, P., and Norton, D., 2003, Mercury in edible fish tissue and sediments from selected lakes and rivers of Washington State: Olympia, WA, Washington State Department of Ecology, no. 03-03-026, 104 p.

Fitzgerald, W.F., 1989, Atmospheric and oceanic cycling of mercury, *in* Riley, J.P., and Chester, R., eds., Chemical Oceanography: London, Academic Press, v. 10, p. 151-186.

Fitzgerald, W.F., Vandal, G.M., and Mason, R.P., 1991, Atmospheric cycling and air-water exchange of Hg over mid-continental lacustrian regions: Water, Air, and Soil Pollution, v. 56, p. 745-767.

Gilmour, C.C., and Riedel, G.S., 2000, A survey of size-specific mercury concentrations in game fish from Maryland fresh and estuarine waters: Archives of Environmental Contamination and Toxicology v. 39, no. 1, p. 53-59.

Gonzalez, H., 1991, Mercury pollution caused by a chlor-alkali plant: Water, Air, and Soil Pollution, v. 56, p. 83-93.

Guentzel, J.L., Landing, W.M., Gill, G.A., and Pollman, C.D., 2001, Processes influencing rainfall deposition of mercury in Florida: Environmental Science and Technology, v. 35, p. 863-873.

Hanisch, C., 1998, Where is mercury deposition coming from?: Environmental Science and Technology, v. 32, no. 7, p. 176A-179A.

Hayward, J., Clevenger, K., and Crawford, T., 2003, Long-term atmospheric mercury trends in Eastern North Carolina: Relationship between local source activities and ambient air mercury concentrations: Raleigh, NC, North Carolina Department of Environment and Natural Resources, accessed September 30, 2003, at URL http://daq.state.nc.us/toxics/studies/mercury/aq2final.pdf.

Kinsey, J.S., 2003, Characterization of mercury emissions at a chlor-alkali plant: Cincinnati, OH, National Risk Management Research Laboratory, U.S. Environmental Protection Agency EPA/600/SR-02/007, 4 p.

Landis, M.S., Keeler, G.J., Al-Wali, K.I., and Stevens, R.K., 2004, Divalent inorganic reactive gaseous mercury emissions from a mercury cell chlor-alkali plant and its impact on near field atmospheric dry deposition: Atmospheric Environment, v. 38, no. 4, p. 613-622.

Lawson, N.M., and Mason, R.P., 2001, Concentrations of mercury, methyl-mercury, cadmium, lead, arsenic, and selenium in the rain and stream water of two contrasting watersheds in western Maryland: Water Research, v. 35, no. 17, p. 4039-4052.

Lindberg, S.E., Turner, R.R., Meyers, T.P., Taylor, G.E., and Schroeder, W.H., 1991, Atmospheric concentration and deposition of Hg to the deciduous forest at Walker Beach Watershed, Tennessee, USA: Water, Air, and Soil Pollution, v. 56, p. 577-594.

Lindberg, S.E., and Stratton, W.J., 1997, Atmospheric mercury speciation: Concentrations and behavior of reactive gaseous mercury in ambient air: Environmental Science and Technology, v. 32, no. 1, p. 49-57.

Lindqvist, O., 1994, Atmospheric cycling of mercury: An Overview, in Watras, C.J., and Huckabee, J.W., eds., Mercury Pollution: Integration and Synthesis: Ann Arbor, MI, Lewis Publishers, p. 181-185.

Lodenius, M., 1998, Dry and wet deposition of mercury near a chlor-alkali plant: Science of the Total Environment, v. 213, p. 53-56.

Marvin-DiPasquale, M., and Agee, J.L., 2003, Microbial mercury cycling in sediments of the San Francisco Bay-Delta: Estuaries, v. 26, no. 6, p. 1517-1528.

Maserti, B.E., and Ferrara, R., 1991, Mercury in plants, soil, and atmosphere near a chlor-alkali complex: Water, Air, and Soil Pollution, v. 56, p. 15-20.

Mathews, R.A., Hilles, M., Vandersypen, J., Mitchell, R.J., and Mathews, G.B., 2002, Lake Whatcom Monitoring Project, 2000/2001 Final Report: Bellingham, Washington, Western Washington University, 423 p.

Mathews, R.A., Hilles, M., Vandersypen, J., Mitchell, R.J., and Mathews, G.B., 2003, Lake Whatcom Monitoring Project, 2001/2002 Final Report: Bellingham, Washington, Western Washington University, 195 p.

Mueller, K., and Serdar, D., 2002, Total mercury concentrations among fish and crayfish inhabiting different trophic levels in Lake Whatcom, Washington: Journal of Freshwater Ecology, v. 17, no. 4, p. 621-633.

Munn, M.D., and Short, T.M., 1997, Spatial heterogeneity of mercury bioaccumulation by walleye in Franklin D. Roosevelt Lake and the Upper Columbia River, Washington: Transactions of the American Fisheries Society, v. 126, p. 677-487.

National Atmospheric Deposition Program, 1998, National Atmospheric Deposition Program 1997 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed June 3, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/97as.pdf.

National Atmospheric Deposition Program, 1999, National Atmospheric Deposition Program 1998 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed June 3, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/98as.pdf.

National Atmospheric Deposition Program, 2000, National Atmospheric Deposition Program 1999 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed May 27, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/99as.pdf.

National Atmospheric Deposition Program, 2001, National Atmospheric Deposition Program 2000 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed May 27, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/2000as.pdf.

National Atmospheric Deposition Program, 2002, National Atmospheric Deposition Program 2001 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed May 27, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/2001as.pdf.

National Atmospheric Deposition Program, 2003, National Atmospheric Deposition Program 2002 Wet Deposition: Champaign, IL, NADP Program Office, 16 p., accessed February 4, 2003, at URL http://nadp.sws.uiuc.edu/lib/data/2002as.pdf.

Norton, D., 2002, Quality assurance project plan: Mercury in sediments from Lake Whatcom and surrounding lakes: Determination of spatial and temporal patterns and characterization of tributary inputs: Olympia, Washington, Washington State Department of Ecology, No. 02-03-079.

Norton, D., 2004, Mercury in sediments from Lake Whatcom: Determination of spatial and temporal patterns and characterization of tributary inputs: Olympia, Washington, Washington State Department of Ecology, No. 04-03-018, 38 p. plus appendixes.

Pacyna, J. M., Sofiev, M.A., Ebinghaus, R., Henden, E., Petersen, G., Shroeder, W., Slemr, F., Sokolov, V., Yagolnitser, M., 1996, Atmospheric mercury working group report in Baeyens, W., Ebinghaus, R., and Vasielieve O., eds., Global and Regional Mercury Cycles: Sources, Fluxes and Mass Balances: Boston, Massachusetts, Kluwer Academic Publishers, p. 523-530.

Peele, Cheri, 2003, Washington State mercury chemical action plan: Olympia, Washington, Washington State Department of Ecology, Publication No. 03-03-001, 190 p.

Pelletier, G., 1998, Dissolved oxygen in Lake Whatcom: Trend in the depletion of hypolimnetic oxygen in Basin I, 1983-1997: Olympia, Washington, Washington State Department of Ecology, Report No. 98-313.

Perry, R., 1974, Recovery from contaminated waste water and sludges: Washington, D.C., U.S. Environmental Protection Agency, EPA/660/2-74-086, 130 p.

Prestbo, E.M., and Bloom, N.S., 1995, Mercury speciation adsorption (MESA) method for combustion flue gas: Methodology, artifacts, intercomparison, and atmospheric implications:Water, Air, and Soil Pollution, v. 80, p. 145-158.

Puget Sound Energy, 2002, Regional setting, *in* Baker River project relicense: Initial consultation, accessed March 30, 2004, at URL http://www.pse.com/hydro/baker/project/projectinfo.html.

Saenger, M., Werther, J., and Hanben, H., 1999, Concentration and mass balances of mercury in a fluidized bed sewage sludge incinerator plant: 15th International Conference on Fluidized Bed Combustion, Savannah, Georgia, American Society of Mechanical Engineers, May 16-19, 1999.

Schroeder, W.H., 1996, Estimates of atmospheric input and evasion fluxes of mercury to and from the Great Lakes, *in* Baeyens, W., Ebinghaus, R., and Vasiliev, O., Global and Regional Mercury Cycles: Sources, Fluxes, and Mass Balances: Kluwer Academic Publishers, p. 109-121.

Schuster, P.F., Krabbenhoft, D.P., Naftz, D.L., Cecil, L.D., Olson, M.L., Dewild, J.F., Susong, D.D., Green, J.R., and Abbot, M.L., 2002, Atmospheric mercury deposition during the last 270 years: A glacial ice core record of natural and anthropogenic sources: Environmental Science and Technology, v. 36, no. 11, p. 2303-2310.

Seiders, K., 2003, Washington State Toxics Monitoring Program: Toxic fish tissue and surface water in freshwater environments, 2001: Washington State Department of Ecology, No. 03-03-012, 39 p. plus appendixes.

Serdar, D., Davis, D., and Hirsch, J., 1999, Lake Whatcom watershed cooperative drinking water protection project: Results of 1998 water, sediment, and fish tissue sampling: Olympia, Washington, Washington State Department of Ecology, Report No. 99-337, 66 p. plus appendixes.

Serdar, D., Johnston, J., Mueller, K., and Patrick, G., 2001, Mercury concentrations in edible muscle of Lake Whatcom Fish: Olympia, Washington, Washington State Department of Ecology, No. 01-03-012, 28 p. plus appendixes.

Skare, I., 1995, Mass balance and systemic uptake of mercury released from dental amalgam fillings: Water, Air, and Soil Pollution, v. 80, p. 59-67.

Stoor, R.W., 2002, Ground-water contributions of methyl-mercury to a Lake Superior Watershed: Environmental Chemistry and Technology, Madison, Wisconsin, M.S. Thesis, University of Wisconsin, 73 p.

Therriault, T.W., and Schneider, D.C., 1998, Predicting change in fish mercury concentrations following reservoir impoundment: Environmental Pollution, v. 101, p. 33-42.

Tracy, K., 2001, Changes in Mirror Lake, northwestern Washington, as a result of the diversion of water from the Nooksack River: Bellingham, Washington, Western Washington University, 90 p.

Ullrich, S.M., Tanton, T.W., and Abdrashitova, S.V., 2001, Mercury in the aquatic environment: A review of factors affecting methylation: Critical Reviews *in* Environmental Science and Technology, CRC Press, v. 31, p. 241-293.

U.S. Environmental Protection Agency, 1984, Review of National Emissions Standards for Mercury: Research Triangle Park, North Carolina, Emissions Standards and Engineering Division, U.S. Environmental Protection Agency, EPA 450/3-84-014.

U.S. Environmental Protection Agency, 1990, Specifications and guidance for obtaining contaminant-free sample container institution: Washington D.C., Office of Solid Waste and Emergency Response, U.S. Environmental Protection Agency, Directive #93240.0-05.

U.S. Environmental Protection Agency, 1992, National study of chemical residues in fish: Washington, D.C., Office of Science and Technology, U.S. Environmental Protection Agency, EPA 823 823-R-92-008a, 166 p. plus appendixes.

U.S. Environmental Protection Agency, 1995, Solid waste disposal, sewage sludge incineration: AP-42, 5th edition, v. I, chap. 2.2, accessed June 23, 2004, at URL http://www.epa.gov/ttn/chief/ap42/ch02/final/c02s02.pdf.

U.S. Environmental Protection Agency, 1997a, Anthropogenic point sources of mercury emissions, Mercury study report to Congress: An Inventory of Anthropogenic Emissions in the United States: Washington, D.C., Office Air Quality Planning and Standards and Office of Research and Development, U.S. Environmental Protection Agency, v. 4, p. 1-40.

U.S. Environmental Protection Agency, 1997b, Overview of mercury fate and transport, Mercury Study Report to Congress: Fate and Transport of Mercury in the Environment: Washington, D.C., Office Air Quality Planning and Standards and Office of Research and Development, U.S. Environmental Protection Agency, v. 3, chap. 2., p. 2-14, accessed July 30, 2003, at URL http://www.epa.gov/ttn/oarpg/t3/reports/volume3.pdf.

U.S. Environmental Protection Agency, 2001, Water-quality criterion for the protection of human health: Methyl-mercury: Washington, D.C., Office of Science and Technology, EPA-823-R-01-001, accessed June 10, 2004 at URL http://www.epa.gov/waterscience/criteria/methylmercury/document.html.

U.S. Environmental Protection Agency,U.S. Environmental Protection Agency, 2002, Maximum contaminant levels for inorganic contaminants: Washington D.C., Code of Federal Regulations 40CFR141.62 accessed June 10, 2004 at URL http://a257.g.akamaitech.net/7/257/2422/14mar20010800/edocket.access.gpo.gov/cfr_2002/julqtr/40cfr141.62.htm.

U.S. Environmental Protection Agency, 2004, Consumer fact sheet on: Mercury, accessed June 10, 2004, at URL http://www.epa.gov/OGWDW/contaminants/dw_contamfs/mercury.html.

van Velzen, D., Langenkamp, H., and Herb, Georg, 2002, Review: Mercury in waste incineration: Waste Management and Research, v. 20, p. 556-568.

Wangberg, I., Edner, H., and others, 2003, Atmospheric mercury near a chlor-alkali plant in Sweden: Science of the Total Environment, v. 304, p. 29-41.

Winter, T.C., Harvey, J.W., Franke, O.L., and Alley, W.M., 1999, Ground water and surface water — A single resource: U.S. Geological Survey Circular 1139, 79 p.

Washington State Department of Ecology, 2003, Water quality standards for surface waters of the State of Washington, Chapter 173-201A WAC: 101.

Watras, C.J., Morrision, K.A., and others, 1996, Mass balances studies of mercury and methyl-mercury in small temperate/Boreal Lakes in the Northern Hemisphere, in Baeyens, W., Ebinghaus, R., and Vasielieve O., eds., Global and Regional Mercury Cycles: Sources, Fluxes and Mass Balances: Boston, Massachusetts, Kluwer Academic Publishers, p. 319-358.

Appendix A. Analysis of Mercury in Fish and Assessment of Quality of Data Used in the Report

Methods for analyses of new data on Hg concentrations in fish from lakes in Whatcom County presented in this report are described below.

Analysis of Mercury in Fish

Yellow perch from Lake Padden, Lake Samish, and Tennant Lake; brown bullhead from Lake Terrell and Wiser Lake; largemouth bass from Tennant Lake; and one cutthroat trout from Lake Samish were collected by the Washington State Department of Fish and Wildlife. After the fish samples were filleted and homogenized by En Chem (Green Bay, Wis.), Frontier Geosciences, Inc. (Seattle, Wash.) digested about 0.5 g of tissue from each sample with 10 mL of hot-refluxing 70-percent HNO3: 30-percent H_2SO_4 for about 2 hours. The digests were then diluted to final volume of 40 mL with a solution of 10-percent (v/v) 0.2 N BrCl. Aliquots of each sample were analyzed by stannous chloride ($SnCl_2$) reduction, dual gold amalgamation, and analyzed by cold-vapor atomic fluorescence spectrometry (CVAFS) according to standard operation procedures (Frontier Geosciences Inc., Seattle, Washington, written commun., 2003). The Hg concentrations in yellow perch and brown bullhead from Lake Padden, Lake Samish, and Wiser Lake could not be assigned to specific fish, and only averages of the individual specimens are reported.

The Hg concentrations in fillets of smallmouth bass and cutthroat trout from Lake Whatcom and in fillets of largemouth bass and yellow perch in lakes in Whatcom County were subjected to the statistical analysis of covariance relative to fish length using the General Linear Models (GLM) procedure without transformation (SAS Institute, 1989). Significant differences in the slopes of the relation of Hg concentrations to fish length among basins and(or) lakes were evaluated. For those lakes in which differences in slopes were not significant, the differences in Hg concentrations among basins and(or) lakes were evaluated taking into consideration the lengths of the populations of fish analyzed as the confounding factor.

Quality of Data

Mercury in Riverine Sources

Storm water entering Lake Whatcom was collected at three sites in the spring and autumn of 1998 by Washington State Department of Ecology (Ecology) and analyzed for total recoverable Hg with cold-vapor atomic absorption spectrometry (CVAAS) using ultra-clean techniques and the CETAC instrument (Serdar and others, 1999). The detection limit was 3 ng/L and the instrument calibration verification standard was 118 percent. Recovery of matrix spikes was 96 percent (±20 percent) in the spring and 91 percent (±1 percent) in the autumn. The relative percent difference (RPD) of one field duplicate at 6 ng/L was 14 percent. Analyses of laboratory control samples were reported to have been within the prescribed acceptable range, but data were not provided.

Collection of monthly samples from terminus for the diversion of the Middle Fork Nooksack River were initiated by the City of Bellingham in August 2002; and the samples were analyzed for total Hg by Frontier Geosciences, Inc. using CVAFS (William McCort, City of Bellingham, written commun., 2003). Trip blanks were collected during six sampling periods and all trip blank concentrations were less than the reporting level of 0.15 ng/L. For each sampling period, Hg in the standard reference material (SRM) 1641d [National Institute of Standards and Technology (NIST), Gaithersburg, MD] was measured and resulted in an average recovery of 96 percent (±7 percent, n=11). The average recovery of two duplicate Nooksack River samples spiked at the 7 ng/L concentrations was 100 percent (±2 percent, n = 4). The recovery of nine duplicate matrix samples spiked at concentrations ranging from 5 to 80 ng/L was 96 percent (±10 percent, n = 18). The RPD of one laboratory duplicate was 9.9 percent at a concentration of 1.85 ng/L.

Bimonthly samples were collected by Ecology from 10 tributaries between July 2002 and May 2003 (Norton, 2004). The detection limit of the method was 2 ng/L, and concentrations in the field blanks were less than the detection limit. The RPD of four field duplicates ranged from 4 to 27 percent at Hg concentrations from 2.5 to 6.4 ng/L. The two analyses of SRM NIST #1641d, diluted to 30.8 ng/L, were within 3 percent of the certified value.

Mercury in Ground Water

Ground-water samples were collected from the Y-Road landfills by direct push sampling between October 19 and November 2, 2001, and from domestic wells by BEK Engineering and Environmental, Inc. (2002a). A method blank of 100 ng/L was reported. The recoveries of two matrix spikes were 110 percent at 2,200 ng/L.

BEK Engineering & Environmental, Inc. (formerly BEK Purnell Engineering, Inc.) performed soil and ground-water assessments at the Hilltop (BEK Engineering & Environmental, Inc., 2002b) and Airport Woodwaste (BEK Engineering & Environmental, Inc., 2002c) landfill sites for Georgia-Pacific West, Inc. The detection limit for total Hg was 1,000 ng/L through 1995, when it decreased to 200 ng/L. At the Hilltop Woodwaste Landfill, soil samples also were analyzed with a method with a detection limit of 0.1 mg/kg. No other quality-assurance (QA) data were provided for this report.

Dr. Susan Cook collected four whole domestic water samples in the Whatcom Lake drainage basin in 1990 and sent them to Elemental Research of North Vancouver, British Columbia, Canada, for analysis of inductively-coupled plasma-mass spectrometry (ICP-KMS). This method has a detection limit of about 50 ng/L of Hg, depending on the concentration of dissolved solids. Samples were collected from the faucet of the well into acid-cleaned bottles provided by the analytical laboratory, after running water for 5 minutes. No other quality-assurance data were provided. Concentrations of aluminum (Al) and iron (Fe) concentrations in the whole-water samples were used to assess the presence of particles, and by inference, particulate Hg, in the samples.

Mercury in Lakes

Sediments

For the study in which lake sediments were collected (Serdar and others, 1999), the RPD for one field replicate was 1 percent and the RPD of one laboratory duplicate was 5 percent. Analyses of laboratory control samples were reported to be within the acceptable range for Hg, but no data were presented. The recovery of one matrix spike was 102 (±1 percent, n = 2).

The quality-assurance program for the collection, processing, and analyses of sediment core samples from five lakes in Whatcom County reported in this report and three core sediments in Lake Whatcom reported by Ecology (Norton, 2004) are grouped together because the samples were collected by the same USGS and Ecology personnel, using the same collection and processing techniques, and analyzed by, or contracted through, the same Ecology Manchester Environmental Laboratory and are described by Norton (2002). In addition, the procedures used for the analyses of 30 grab samples of surface sediments are identical to those used to analyze the cores samples. Therefore, the quality-control data reported here will reflect the overall quality of the analytical data for samples from both the eight cores and the 30 grab samples. Whereas, each of the field replicates for the surface grab samples were collected from a completely different sample, the duplicates from the core samples were taken from the same core section.

The RPD in the determination of percentage of solids from two field replicates was less than 1 percent at percentage of solids contents of 17 and 27 percent. The RPD of the percentage of material in the dominant size fractions (fractions greater than 20 percent by weight) ranged from 0.4 to 11.3 percent for duplicates of two surface sediment samples collected from Lake Whatcom. The relative standard deviation for triplicate laboratory analyses of total organic carbon (TOC) ranged from 0.3 to 6.9 percent for six samples. The RPD of field duplicates of two surface samples was less than 4 percent, and the RPDs for TOC results of field splits from core samples from basins 1, 2, and 3 of Lake Whatcom were 0.9, 1.1, and 33.4 percent, respectively. The stable Pb data was excellent, with the RPD for three field splits being less than 4 percent, and 7 percent for one laboratory duplicate. The Pb concentrations of all laboratory blanks were below the reported value of 0.003 mg/kg. The recoveries for matrix spikes was 102 ± 6.7 percent (n = 9) and 105 ± 7.6 (n = 9) for the recovery of a laboratory control standard.

Quality-assurance data for total Hg consisted of laboratory blanks, laboratory replicates, laboratory control standards, field replicates (surface grab samples), field splits (core samples), blind standard reference materials, and interlaboratory comparisons. The quality-control data for analyses of total Hg generally was good, although the core samples from Fazon, Terrell and Wiser Lakes, and the bottom two sections from the Baker Lake core were analyzed in a second batch of analyses that was qualified because they exceeded the holding time. The Hg concentration of all laboratory blanks were less than the reporting value of 0.003 mg/kg. The recovery of the laboratory control standards was 99.9 ±3.4 percent. Although the matrix spike recoveries satisfied the data-quality objectives, there seemed to be a small difference in the matrix spike recoveries among the batches of core samples analyzed. For the first three sets that were analyzed within the holding times, the matrix recoveries were 99.8 ± 4.2 percent. The matrix recoveries for the last two sets that were qualified because of expired holding time were 91 ± 1.4 percent.

The RPD of total Hg in laboratory duplicates for two surface grab samples was 4 percent. At moderate Hg concentrations of 0.224 and 0.142 mg/kg, the RPDs of Hg in field duplicates of two surface grab samples were 5.4 and 0.7 percent, respectively. The RPDs of total Hg in duplicate field splits from core samples from Lake Whatcom were less than 2 percent for basins 1 and 2, and 19 percent for basin 3.

Because the three aliquots of the two SRM were submitted to the laboratory as dried samples, these SRMs samples (IAEA-405 and MESS-3) can only be considered blind samples because the analysts probably knew they were SRMs but did not know the identity of the SRM. Three SRM aliquots were analyzed with the earlier batch of samples and three SRM aliquots analyzed in the latter batch that was qualified because of the expired holding time. The results of the Hg analyses of three aliquots of SRM MESS-3 averaged 0.108 ± 0.038 mg/kg, compared to the certified value of 0.091 mg/kg (confidence interval between 0.082 and 0.100 mg/kg). The results of the SRM IAEA-405 averaged 0.85 ± 0.06 mg/kg, and compared favorably with the certified value of 0.81 mg/kg (confidence interval 0.77 to 0.85). The three analyses of SRM aliquots analyzed with the earlier batch of samples were within the certified values, and the RPD of Hg concentration for the one SRM samples analyzed in duplicate was 40 percent. The analyses of the three SRMs aliquots analyzed with the later batch of samples generally seemed to produce higher concentrations than the earlier batch. Interlaboratory comparison of three sediment samples were analyzed by both the USGS Mercury Laboratory in Wisconsin and the Ecology Laboratory. Total Hg concentrations for the Ecology Lab generally were slightly higher than those from the USGS Mercury Laboratory at total Hg concentrations from 0.1 to 0.2 mg/kg. The bottom 30-34 cm section of the basin 3 core of Lake Whatcom appeared to be contaminated, possibly from the bottom plunger of the core extractor. The Pb and Hg analyses were repeated for the 29-31 cm section. The percentage of solids and TOC analyses from the 30-34 cm section were used for the 29-31 cm section.

Quality-control data for methyl-Hg in the surface-sediment grab samples (Norton, 2004) consisted of the analysis of field replicates, blind standard reference materials, and interlaboratory comparisons. The RPD of methyl-Hg concentrations of one field duplicate sample, with an average concentration of 0.0025 mg/kg, was 36 percent. For two blind aliquots of the SRMs samples [International Atomic Energy Agency (IAEA-405)], the concentration of methyl-Hg reported by the Ecology Laboratory was 0.0047 ± 0.00051 (1 standard deviation) mg/kg compared to the recommended value of 0.00549 mg/kg (95-percent confidence range from 0.00496 to 0.00602 mg/kg) released by IAEA. Although the laboratory results were slightly below the confidence interval, the set of analyses from which the IAEA recommended value was calculated contained three analyses for which methyl-Hg concentrations were less than 0.0050 mg/kg.

Two interlaboratory comparison surface grab samples were analyzed by the Ecology Laboratory and the USGS Mercury Laboratory. The methyl-Hg concentrations in aliquots from basin 3 were comparable between the two laboratories; however, the methyl-Hg concentration in the samples from basin 1 (LW 1-4) determined by the USGS Mercury Laboratory was twice as high as that of the Ecology Laboratory. This difference is likely due to the inhomogeneity of the surface samples. Samples of about 5 to 10 mL for methyl-Hg analyses were collected directly from the top 2 cm of the grab, and transported separately to the respective laboratories. Only at the laboratory were the materials from the three grab samples mixed for analyses. Because there was no mixing in the field, variations in methyl-Hg within or between grab samples at a single station could result in the differences observed by the two laboratories.

Water

The Institute of Watershed Studies has been monitoring the water quality of Lake Whatcom since 1981. Prior to 2001, water-sample analysis for Hg by CVAAS (Mathews and others, 2002). Concentration greater than the detection limit of 200 ng/L was not detected in any samples. In 2001, special analytical tests were conducted using ICP-MS in which the method detection limit (MDL) was 50 ng/L. Hg concentrations were reported to be less than the MDL in 24 of the 25 samples. Hg concentration in the one remaining sample was reported to be 80 ng/L. Because the detection of Hg in just one sample represents 4 percent of the sample population and is only 60 percent greater than the MDL, it is statistically possible that the detectable value of 80 ng/L was a false positive.

Exponent, Inc., collected 7 water samples from Lake Whatcom on October 11, 2001, using ultra-clean techniques and analyzed the samples for Hg and methyl-Hg by cold-vapor atomic fluorescence spectrometry (CVAFS) (Gary Bigham, written commun., 2002). The detection limit for total Hg was 0.20 ng/L, and the RPD for one duplicate was 11.6 percent at a mean Hg concentration of 2.42 ng/L. The recovery of 10.1 ng/L of Hg spiked into a lake sample was 94.1 percent, and the recovery of a SRM with a very high Hg concentration (NIST-1641d) averaged 94 percent. The detection limit for methyl-Hg was 0.020 ng/L and the RPD for a duplicate of one near-bottom lake water sample with an average methyl-Hg concentration of 0.504 ng/L was 9.3 percent. The recoveries of methyl-Hg in replicate near-bottom water samples spiked at 2 ng/L of methyl-Hg were 78 and 85 percent.

The quality-assurance data for monthly samples of Lake Whatcom water from the drinking-water intake gatehouse, which were collected by the City of Bellingham beginning in November 2002, are given in the section, “Mercury in Riverine Sources.”

Mercury in Fish

Hg in fish in Lake Whatcom and other lakes in Whatcom County was examined in four studies. Quality-assurance data were summarized in a limited study of eight fish (Serdar and others, 1999). For Hg, the concentration of Hg in the procedural blank was less than the detection limit, recoveries were within 25 percent of the spiked levels, the RPD of duplicate tissue samples was reported to be within 20 percent, and the analysis of SRM DORM-2 was within the accepted range. In the most extensive study, the method detection limits for the analyses of Hg in smallmouth bass, yellow perch, cutthroat trout, brown bullhead, kokanee, pumpkinseed, and signal crayfish from Lake Whatcom (Serdar and others, 2001) ranged from 0.005 to 0.010 μg/g wet weight. The average relative standard deviation for 15 laboratory replicates was 9 percent and ranged from 2 to 19 percent. The recovery of 19 matrix spikes was 84 percent and ranged from 72 to 98 percent. Analyses of standard reference materials were not reported.

Smallmouth bass, largemouth bass, and cutthroat trout caught by the Washington State Department of Fish and Wildlife were analyzed for Hg by the Washington State Toxics Monitoring Program of Ecology (Seiders, 2003). The median RPD of Hg in laboratory duplicates (n = 10) was 2 percent and the average was 8 percent (ranging from 0 to 31 percent for a largemouth bass from Fazon Lake). The median RPD for field duplicates (n = 11) was 4 percent with an average of 8 percent (ranging from 0 to 42 percent for largemouth bass from Lake Samish). The average recovery of Hg from fish matrix spiked at an unknown Hg concentration was 90 percent (±14 percent recovery) and the RPD of matrix spikes averaged 2 percent (ranging from 0 to 13 percent). The average recovery of Hg in the dogfish liver SRM (DOLT from National Research Council of Canada) was 100 percent (±8 percent) and was 90 percent (±15 percent) for the dogfish muscle SRM (DORM)).

In this study, the method detection limit for analyses of Hg in cutthroat trout, yellow perch, brown bullhead, and largemouth bass collected from lakes in Whatcom County by the Washington State Department of Fish and Wildlife and analyzed by Frontier Geosciences, Inc. was 0.0005 μg/g (wet weight). The RPD for two laboratory duplicates was 7 and 10 percent. The average recovery of two yellow perch spiked with 0.3 μg/g Hg and two brown bullhead samples spiked with 0.07 μg/g Hg was 99 percent (±4 percent). The average recovery of duplicate aliquots of the SRM DORM-2 was 86 percent (±5 percent).

References Cited

BEK Engineering & Environmental Inc., 2002a, Phase II Hydrogeologic investigation: Y-road landfills, Whatcom County, Washington: Ferndale, WA, 21 p. plus appendixes.

BEK Engineering & Environmental Inc., 2002b, Annual report 2002 landfill monitoring test results, Hilltop Woodwaste Landfill: Georgia-Pacific West, Inc., Ferndale, WA, 10 p. plus appendixes.

BEK Engineering & Environmental Inc., 2002c, Annual report 2002 landfill monitoring test results, Airport Woodwaste Landfill, Georgia-Pacific West, Inc., Ferndale, WA, 10 p. plus appendixes.

Mathews, R.A., Hilles, M., Vandersypen, J., Mitchell, R.J., and Mathews, G.B., 2002, Lake Whatcom Monitoring Project, 2000/2001 Final Report: Bellingham, Washington, Western Washington University, 423 p.

Norton, D., 2002, Quality assurance project plan: Mercury in sediments from Lake Whatcom and surrounding lakes: Determination of spatial and temporal patterns and characterization of tributary inputs: Olympia, Washington, Washington State Department of Ecology, No. 02-03-079.

Norton, D., 2004, Mercury in sediments from Lake Whatcom: Determination of spatial and temporal patterns and characterization of tributary inputs: Olympia, Washington, Washington State Department of Ecology, NPI.

SAS Institute, 1989, SAS/STAT User's guide, 4th Edition: Cary, North Carolina, v. 2.

Seiders, K., 2003, Washington State Toxics Monitoring Program: Toxic fish tissue and surface water in freshwater environments, 2001: Washington State Department of Ecology, No. 03-03-012, 39 p. plus appendixes.

Serdar, D., Davis, D., and Hirsch, J., 1999, Lake Whatcom watershed cooperative drinking water protection project: Results of 1998 water, sediment, and fish tissue sampling: Olympia, Washington, Washington State Department of Ecology, Report No. 99-337, 66 p. plus appendixes.

Serdar, D., Johnston, J., Mueller, K., and Patrick, G., 2001, Mercury concentrations in edible muscle of Lake Whatcom Fish: Olympia, Washington, Washington State Department of Ecology, No. 01-03-012, 28 p. plus appendixes.

Appendix B. Estimates of Atmospheric Deposition to Lakes in Whatcom County for Sources Emitting a Hypothetical Unit of Mercury Emission in Bellingham, Ferndale, and at the Bellingham Airport

The simple model described here was developed to be used primarily to evaluate in a consistent manner the possible atmospheric deposition of mercury (Hg) to lakes in Whatcom County from past and present local sources of Hg. In the model, the three forms (species) of Hg (vaporous elemental, reactive gaseous, and particulate) initially emitted from a source are considered independently and the total deposition is the sum of the deposition of the three species. The reactivity of a species is characterized by its overall residence time in the atmosphere (τ_i) once it has been discharged from the source, cooled to ambient temperatures and is traveling within the local wind field. The depositional half-life of a specific species is the time it takes to remove one-half of that species of Hg from the atmosphere. The general kinetic rate constant for removal of each specific species from the atmosphere is defined as

$$k_i = \ln(2) / \tau_i \,, \tag{B1}$$

where k_i is in units of the inverse of time (T^{-1}).
The change in concentration of Hg for species i in any parcel of air within an expanding plume is defined as

$$\partial C_{i,x,y,z} / \partial t = - k_i \bullet C_{i,x,y,z} \,, \tag{B2}$$

where the concentration of Hg of species *i* in the parcel of air ($C_{i,x,y,z}$) is in units of mass/length^3 ($M\ L^{-3}$), and $\partial C_{i,x,y,z} / \partial t$ is in units of (mass/length^3)/time or ($M\ L^{-3}\ T^{-1}$).

Because the change in the concentration of species *i* ($-\partial C_{i,x,y,z}/\partial t$) is due to the removal of species *i*, the contribution of this parcel of air to the gain of Hg to the earth's surface (deposition) is simply $-k_i \bullet C_{i,x,y,z}$. The deposition rate of Hg species *i* (Dep_i) to an area of land or water below ($M\ L^{-2}\ T^{-1}$) is the integration of the Hg losses from all parcels of air over the height of plume (*dz*).

$$Dep_i = -\int \partial C_{i,x,y,z} / \partial t \, dz = k_i \int C_{i,x,y,z} \, dz \,. \tag{B3}$$

In order to calculate the Hg concentration in the parcel of air once the plume has buoyantly risen and is traveling horizontally, the transport of Hg (Q in units of $M\ T^{-1}$) through any plane perpendicular to the wind direction is

$$Q_i = u \iint C_{i,x,y,z} \, dy \, dz \,, \tag{B4}$$

where u is the wind speed in units of $L\ T^{-1}$ and $dy\ dz$ is the planar extent of the plume.

Because the change in the mass transport of Hg with time is simply the summation of losses of Hg over all the parcels of air in the plume that are moving past the plane perpendicular to the wind direction:

$$dQ_i / dt = -k_i \, Q_i \,. \tag{B5}$$

Equation B5 can be transformed from time coordinates to space (x) coordinates by recognizing that $dt = dx/u$,

$$d\, Q_i / dx = -(k_i / u)\, Q_i \,, \tag{B6}$$

whose analytical solution is

$$Q_{i,x} = Q_{i,0} e^{-k_i/u \bullet x} \,, \tag{B7}$$

where $Q_{i,x}$ is the mass transport of species *i* at distance *x* in the direction of the wind field and $Q_{i,o}$ is the transport at source, which will be defined as the mass emission from the source.

The derivations up to this point are generalized for any plume behavior in a steady-state wind field. Plume dynamics in this model were simplified by assuming that (1) species *i* is completely mixed across the plane of the plume at a given *x*, and (2) the lateral expansion of the plume is constant, forming a cone in the *xy* plane with total angle of α. The width of the plume (y) at any given x is $2*\tan(\alpha/2) \bullet x$, which is approximated at small angles as $\tan(\alpha) \bullet x$. Equation B4 is then simplified to

$$Q_{i,x} = u \bullet \tan(\alpha) \bullet x \int C_{i,x} \, dz \,. \tag{B8}$$

Substituting the mass flux at a given x from equation B7 on the left side of equation B8, and substituting Dep_i / k_i (from equation B3) for $\int C_{i,x}, dz$ yields

$$Q_{i,0} e^{-k_i/u \bullet x} = u \bullet \tan(\alpha) \bullet Dep_i / k_i \,. \tag{B9}$$

Solving equation B9 for deposition of species *i* yields

$$\mathrm{Dep}_i = k_i \bullet Q_{i,0} e^{-k_i/u \bullet x} / (u \bullet \tan(\alpha) \bullet x). \quad \text{(B10)}$$

Note that the deposition is independent of the thickness of the plume. Although expanding the thickness of the plume will decrease the concentration of species i in the plume, deposition will occur over a longer air column (*dz*).

The simplifications of the plume dynamics allows one to use an analytical solution to estimate the deposition rate without having to numerically calculate the concentration of Hg species *i* in the air mass. Given a specific wind field, deposition at a given point from the source was calculated knowing only the emission rate of species *i* ($Q_{i,0}$) and its residence time (τ_i). Equation B10 can be broken down into terms that are easily understandable. The $Q_{i,0}/u$ represents the dilution of the emission source by the wind field, much as the initial concentration of a pollutant in a river resulting from a point source discharge is dependent on the velocity of a river.

The term $e^{-k_i/u \bullet x}$ represents the decrease in the transport of the Hg species with distance from the source resulting from the deposition of species *i* between the source and distance *x*. The term $1/(\tan(\alpha) \bullet x)$ represents the decrease in the concentration of species *i* in the plume by the lateral dispersion of the plume with distance from the source. The decrease in concentration in the plume results in a decrease in the estimated deposition at a point under the plume.

Distances and directions from each major air emission source to eight target receptors (i.e., the location of the eight lake sediment cores) were determined (table B1). The model was applied using hourly wind speed and wind direction data from the Georgia-Pacific, Inc. (G-P) meteorological station (Axel Franzmann, Northwest Air Pollution Authority, Mount Vernon, Washington, written. commun., 2003) during 1996 (fig. 6) because the wind data for 1995, the year of the last emission test, were incomplete. If the plume was above the target receptor (target receptor direction is within wind direction ± ½α), direct deposition of Hg to the site was calculated for that hour using equation B10 with the measured wind speeds at the G-P station in miles per hour, the distance from the source to the target receptor from table B1 in miles, and a standard plume angle (α) of 20 degrees. The hourly deposition amounts were summed over a year and the annual deposition was converted to SI units. The percentage of hours recording deposition at the specific receptor site (location of lake cores) in the year was calculated and the wind speeds for those hours recording deposition were averaged. Deposition for a unit emission of Hg (Q_0 = 1 g/hr) for residence times of 1 day, 1 week, 1 month, 1 quarter-year, and 1 year were calculated. The sensitivity of the model results was tested by varying the plume angle (α) from 5 to 20 degrees at a residence time of 1 week. This simplified model assumes steady-state conditions and assumes that a parcel of air will continue traveling in recorded wind direction at the recorded wind speed even if it takes several hours to reach the receptor. This model probably is not valid during light and variable wind conditions (about 20 percent of the time), when a radially expanding plume model would be more appropriate. Caution is advised when interpreting deposition calculations in which infrequent deposition occurs with low wind speeds.

Table B1. Direction and distances from source of atmospheric mercury to receptor for lakes in Whatcom County, Washington

Receptor	Direction (degrees)	Distance (miles)
Downtown Bellingham Source		
Lake Whatcom		
Basin 1	71	3.75
Basin 2	91	4.97
Basin 3	111	9.08
Baker Lake	91	39.80
Lake Terrell	312	12.21
Lake Samish	139	7.26
Wiser Lake	1	10.98
Fazon Lake	33	10.00
Ferndale Source		
Lake Whatcom		
Basin 1	118	8.13
Basin 2	120	9.94
Basin 3	124	14.62
Baker Lake	98	43.78
Lake Terrell	300	6.31
Lake Samish	142	13.39
Wiser Lake	33	7.21
Fazon Lake	69	9.73
Bellingham Airport Source		
Lake Whatcom		
Basin 1	119	6.31
Basin 2	121	8.12
Basin 3	126	12.82
Baker Lake	97	42.07
Lake Terrell	299	8.13
Lake Samish	145	11.76
Wiser Lake	18	7.22
Fazon Lake	60	8.59

As equation B10 indicates, the inverse of the residence time of the species being modeled ($\ln(2)/\tau_i$ or k_i) is the primary factor controlling deposition (fig. B1). Because basin 1 of Lake Whatcom is in the direction of a primary wind pattern from Bellingham, deposition occurs 13 percent of the year at an a of 20 degrees, leading to the highest level of estimated deposition (table B2). Estimated deposition for a unit Hg emission decreases rapidly in the southerly direction down Lake Whatcom because the frequency of depositional events decreases to about 4 percent in basin 2 and less than 1 percent in basin 3. As with most source-receptor pairs, the model estimates of Hg deposition are insensitive to the value of the plume angle. As α decreases, the deposition per a depositional event increases because the plume over the receptor is more concentrated, but the frequency of deposition events likewise decreases because of the narrowness of the plume. Deposition rates of Hg in Lake Whatcom from Ferndale and Bellingham Airport sources are more than an order of magnitude less per unit Hg emission than a Bellingham source because the frequency of deposition is only 0.5 percent at average wind speeds of 1.8 mph (table B2). Because the location of the Georgia-Pacific source and the City of Bellingham sludge incinerator are relatively close to each other, deposition from the City of Bellingham sludge incinerator is approximated by the unit deposition calculated from the downtown Bellingham source (Georgia-Pacific, Inc., site).

Lake Terrell and Wiser and Fazon Lakes, all north of Bellingham and referred to as three northern lakes throughout, are in a secondary wind pattern for a Bellingham source. These lakes register deposition events between 4 and 8 percent of the year with average wind speeds from 7 to 10 mph (fig. B2*A*). The estimated deposition for a unit Hg emission is about an order of magnitude less than that of basin 1 of Lake Whatcom with annual deposition decreasing in order from Fazon Lake to Lake Terrell to Wiser Lake (table B2). The annual depositions to the three northern lakes from a unit Hg source in Ferndale (table B2) are slightly greater than that from a Bellingham source. The estimated deposition of Hg to Fazon Lake and Lake Wiser from a Ferndale source (fig. B2*B*) are about equal, whereas that to Lake Terrell is considerably less because of its location west of the source. When the wind data collected from a refinery in Ferndale (Axel Franzmann, Mount Vernon, Wash., written commun., 2003) was used to calculate deposition for a residence time of 1 week to the three northern lakes, the frequency of deposition decreased from about 6 percent to about 2.4 percent. This led to a decrease in deposition by factors between 2.5 and 3.0, and the trend among the lakes remained the same as the trend with a Bellingham wind field.

The deposition to Fazon Lake for a unit Hg emission near the Bellingham Airport (table B2) is slightly greater than that from the Ferndale source because of the shorter distance to Fazon Lake, and therefore less lateral dispersion is modeled. In contrast, deposition to Wiser Lake from a unit emission of Hg from Ferndale is slightly greater than that from a Bellingham Airport unit source (fig. B2*C*).

Caution should be used when interpreting the estimates of Lake Samish deposition (table B2). For all three source locations, deposition registered only under light and variable wind conditions (less than 2 mph and less than 1 percent of the time) because little wind comes from the north, the direction of these sources in relation to Lake Samish. Deposition to Lake Samish from known Hg sources is best estimated with a radially expanding plume model.

Although Baker Lake is in the direction of a primary wind pattern (2.6 to 3.9 percent frequency at about 5 mph), the long distance to Baker Lake (about 55 mi) and the effects of Mount Baker on the meteorology of the area make these estimates susceptible to large uncertainty.

The deposition rates to target lakes described above is for a unit emission of Hg with a specific half-life. Only after (1) the emission rate from a specific source is known or estimated, (2) the speciation among the three species, *i*, has been assessed, and (3) half-lives of each of the three species have been assigned to each species can the total deposition of Hg (TotDep) to the water surface be calculated as the sum of the depositions from three species:

$$\mathrm{TotDep} = \mathrm{Dep}_P + \mathrm{Dep}_R + \mathrm{Dep}_V, \qquad \text{(B11)}$$

where Dep_P is the deposition of particulate mercury, Dep_R is the deposition of reactive Hg, and Dep_V is the deposition of vaporous mercury.

Table B2. Deposition of mercury from an emission of 1.0 gram per hour from downtown Bellingham, Ferndale, and Bellingham Airport, Washington

[(μg/m^2/)yr, micrograms per square meter per year; mph, miles per hour]

Lake	Plume angle (degrees)	Frequency of deposition (percentage of year)	Windspeed (mph)	Deposition of mercury [(μg/m^2)/yr]				
				Residence time (days)				
				1	7	30	92	365
Downtown Bellingham								
Whatcom								
Basin 1	20	13.1	5.2	3.3	0.51	0.12	0.039	0.0099
	10	7.35	5.0		.54			
	5	3.01	5.0		.41			
Basin 2	20	3.93	5.3	.88	.14	.034	.011	.0028
	10	2.00	5.7		.15			
	5	.94	6.1		.14			
Basin 3	20	.96	3.0	.20	.045	.011	.0038	.0010
	10	.39	2.0		.039			
	5	.22	1.8		.048			
Samish	20	.59	1.5	.23	.052	.013	.0043	.0011
	10	.36	1.6		.053			
	5	.14	1.5		.043			
Baker	20	3.93	5.3	.055	.015	.0040	.0014	.0003
	10	2.00	5.7		.015			
	5	.94	6.1		.014			
Fazon	20	7.67	9.3	.49	.10	.026	.0088	.0022
	10	3.89	9.3		.094			
	5	1.62	9.7		.067			
Terrell	20	4.37	6.8	.26	.057	.014	.0049	.0012
	10	2.27	6.4		.057			
	5	.80	6.3		.039			
Wiser	20	5.42	10	.22	.040	.0094	.0032	.0008
	10	3.02	9.5		.044			
	5	1.78	9.6		.047			
Ferndale								
Whatcom								
Basin 1	20	0.54	1.8	0.13	0.028	0.0067	0.0023	0.0006
	10	.25	1.7		.026			
	5	.09	2.1		.016			
Basin 2	20	.51	1.8	.098	.023	.0058	.0019	.0005
	10	.22	2.1		.015			
	5	.09	2.6		.0089			
Basin 3	20	.39	1.8	.041	.011	.0027	.0009	.0002
	10	.21	1.9		.0098			
	5	.01	2.1		.0099			
Samish	20	.63	1.5	.092	.026	.0067	.0025	.0006
	10	.38	1.6		.027			
	5	.06	.58		.0152			

Table B2. Deposition of mercury from an emission of 1.0 gram per hour from downtown Bellingham, Ferndale, and Bellingham Airport, Washington—*Continued*

[(μg/m²/)yr, micrograms per square meter per year; mph, miles per hour]

Lake	Plume angle (degrees)	Frequency of deposition (percentage of year)	Windspeed (mph)	Deposition of mercury [(μg/m²)/yr]				
				Residence time (days)				
				1	7	30	92	365
Ferndale—*Continued*								
Baker	20	2.56	5.5	0.028	0.011	0.0031	0.0011	0.0003
	10	1.2	5.7		.0088			
	5	.05	5.7		.0084			
Fazon	20	12.1	5.0	.97	.17	.051	.017	.0043
	10	4.81	4.8		.18			
	5	2.88	5.0		.15			
Terrell	20	3.66	6.3	.69	.13	.031	.010	.0026
	10	1.92	6.5		.11			
	5	.91	6.7		.13			
Wiser	20	7.99	8.1	.82	.16	.039	.013	.0033
	10	4.23	8.8		.18			
	5	2.03	9.3		.14			
Bellingham Airport								
Whatcom								
Basin 1	20	0.53	1.8	0.22	0.042	0.0099	0.0033	0.0008
	10	.24	2.1		.026			
	5	.10	2.3		.025			
Basin 2	20	.51	1.8	.098	.023	.0077	.0019	.0005
	10	.22	2.1		.015			
	5	.09	2.6		.0089			
Basin 3	20	.42	1.8	.063	.017	.0042	.0014	.0004
	10	.21	1.9		.015			
	5	.06	1.2		.015			
Samish	20	.72	1.7	.13	.035	.0088	.0023	.0006
	10	.38	1.6		.027			
	5	.06	.58		.015			
Baker	20	2.68	5.5	.033	.012	.0034	.0012	.0003
	10	1.36	5.9		.011			
	5	.58	6.4		.0088			
Fazon	20	14.0	6.3	1.2	.21	.051	.017	.0043
	10	7.79	6.5		.22			
	5	3.65	6.6		.21			
Terrell	20	3.66	6.2	.41	.081	.020	.0067	.0017
	10	1.79	6.3		.067			
	5	.86	6.0		.064			
Wiser	20	8.80	1.4	.65	.13	.033	.011	.0028
	10	4.54	1.6		.12			
	5	2.22	1.3		.11			

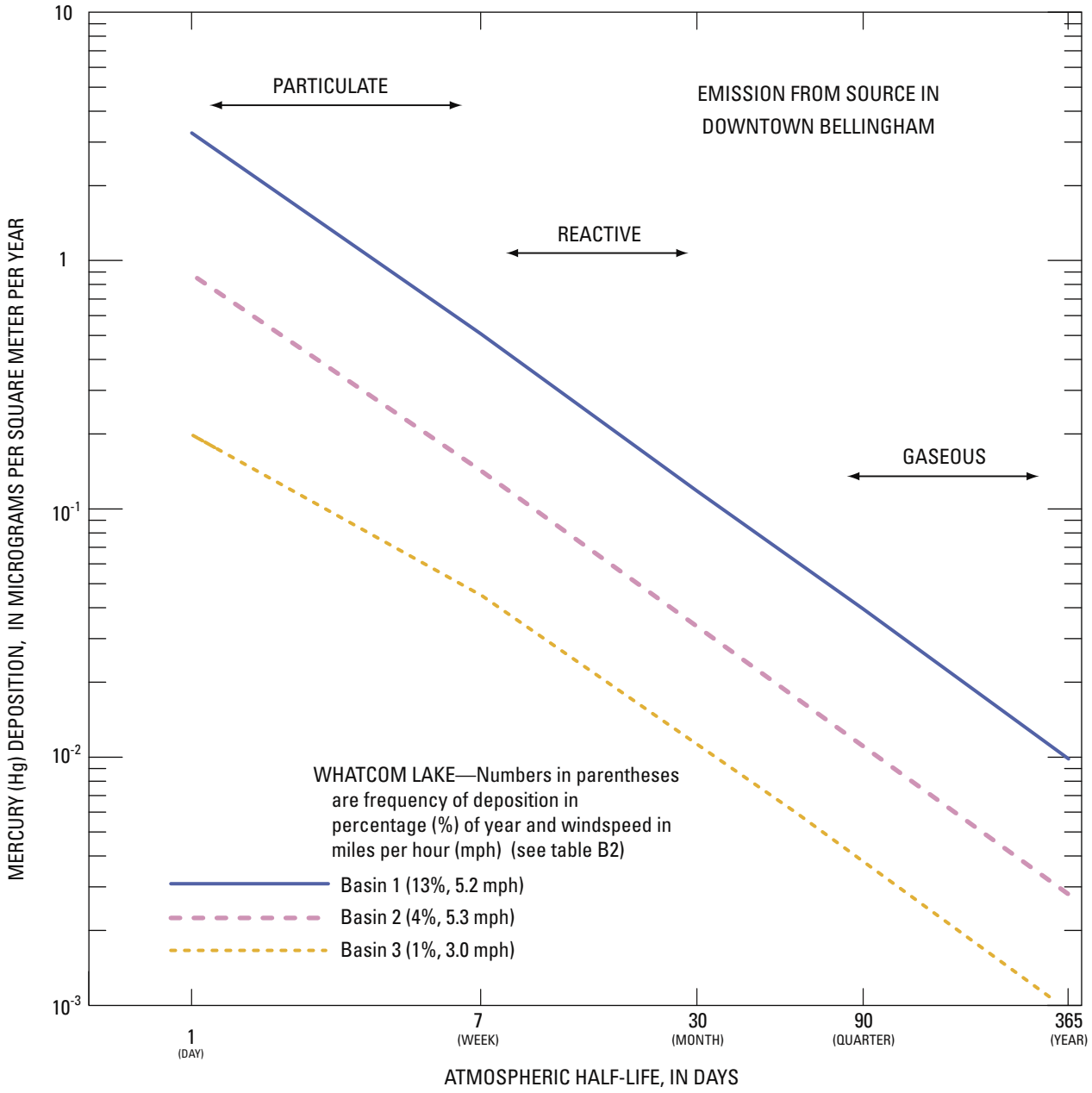
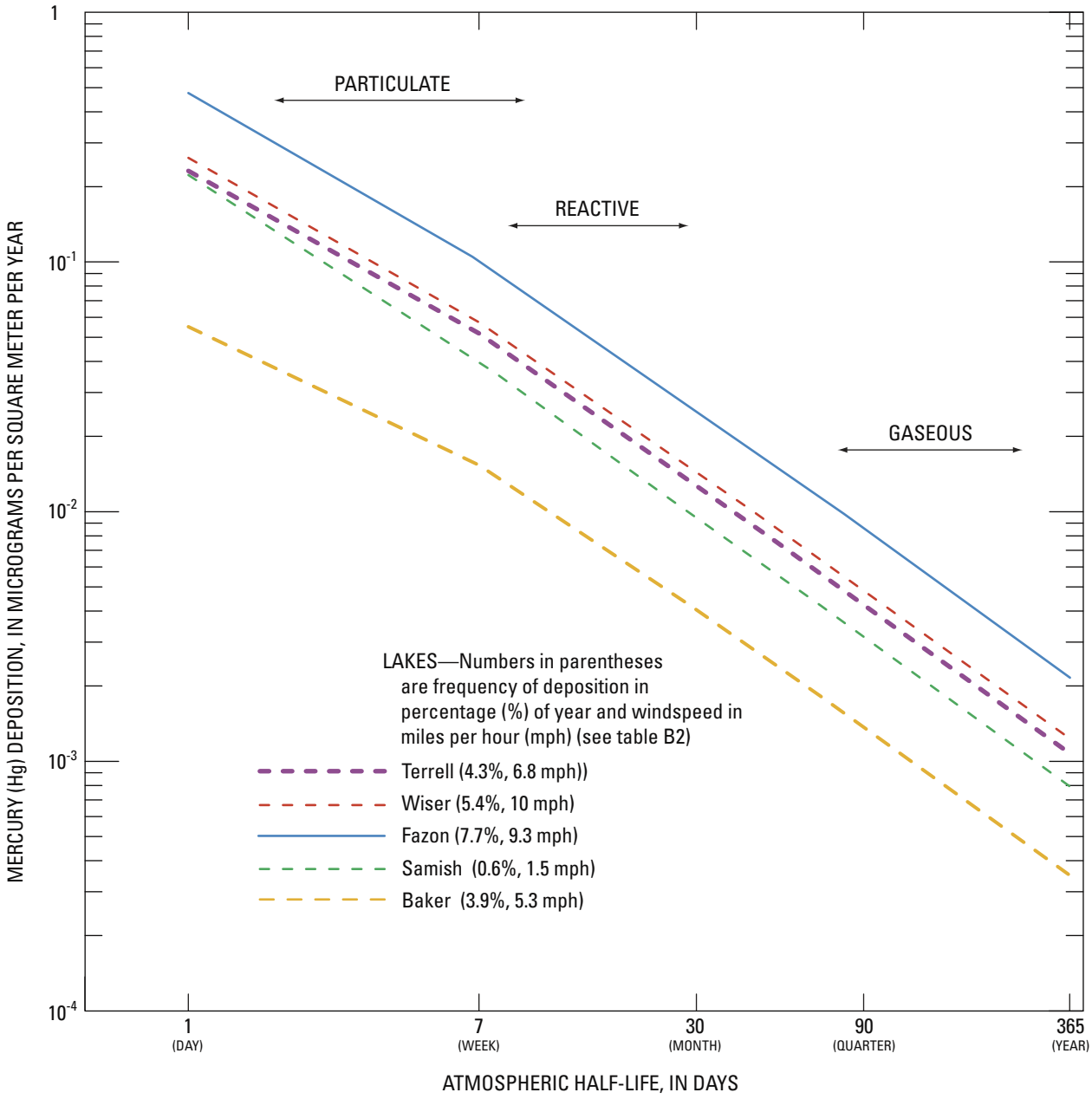
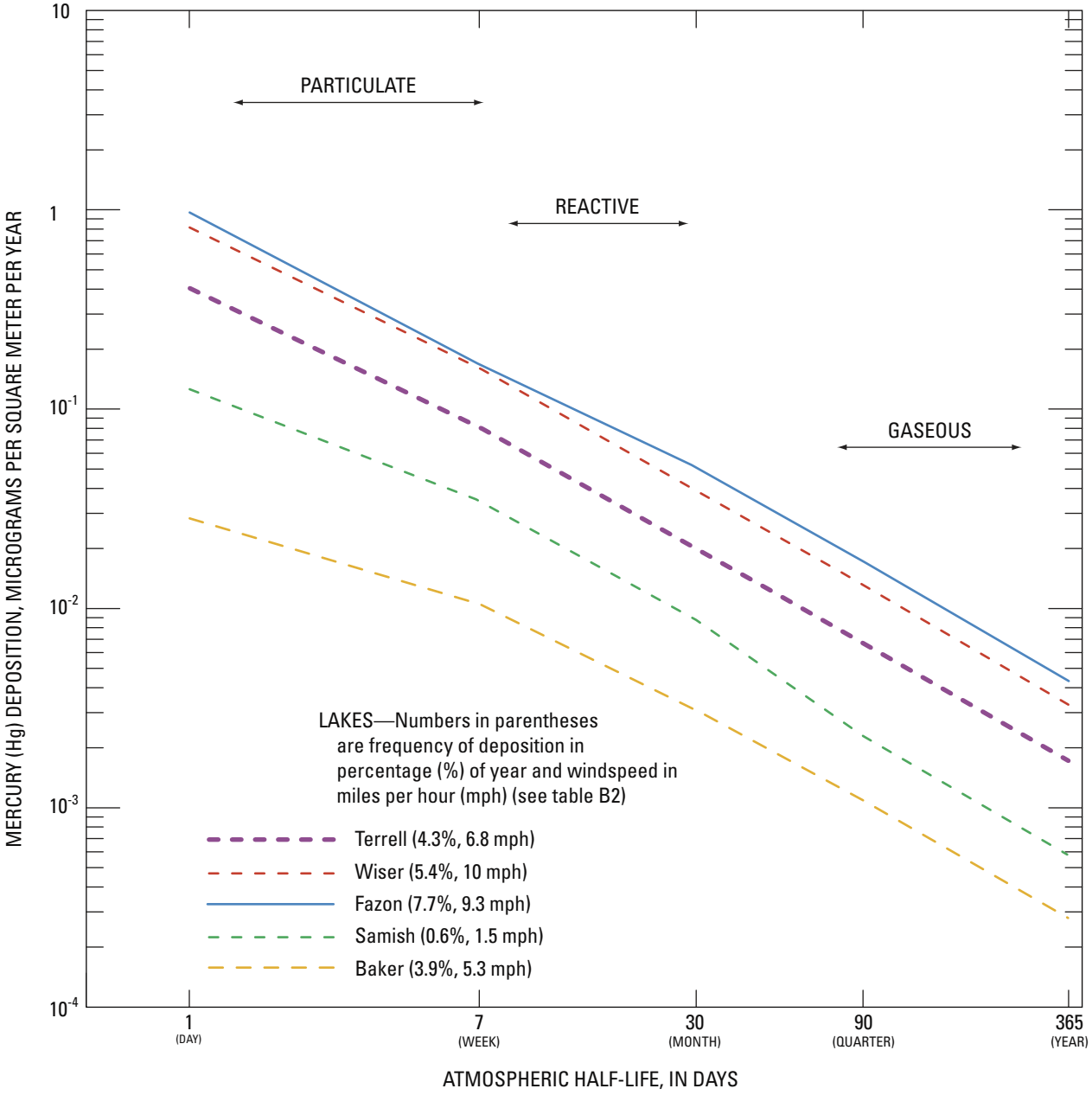


Figure B1. Deposition of mercury in Lake Whatcom from a source in downtown Bellingham, Washington.



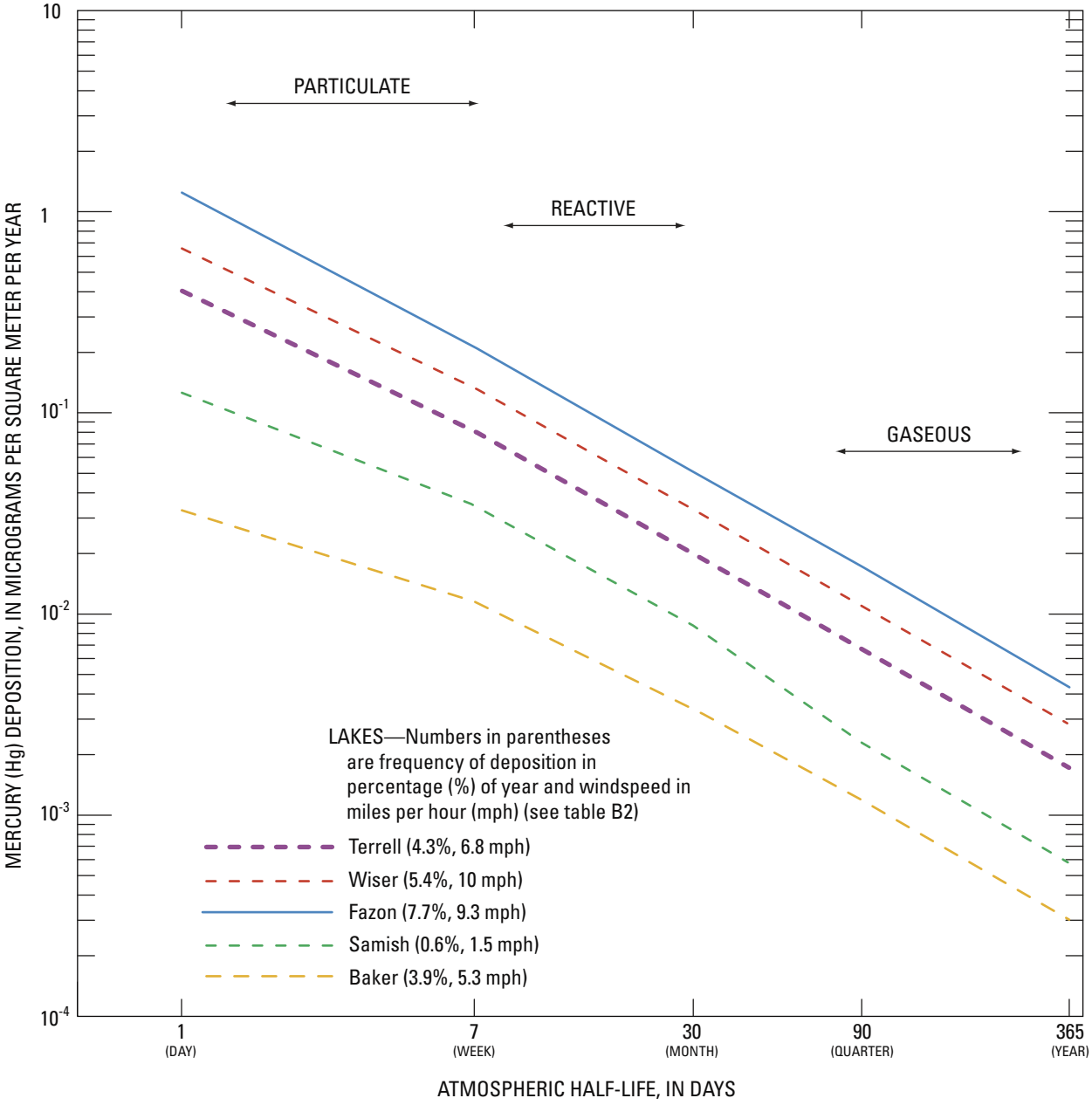
A. EMISSIONS FROM SOURCE IN DOWNTOWN BELLINGHAM

Figure B2. Deposition of mercury in lakes in Whatcom County from emission sources in downtown Bellingham, Ferndale, and Bellingham airport, Washington.



B. EMISSIONS FROM SOURCE IN FERNDALE

Figure B2.—*Continued.*



C. EMISSIONS FROM SOURCE NEAR BELLINGHAM AIRPORT

Figure B2.—*Continued.*

Appendix C. Method for Calculating Missing Flow Data for the Tributaries of Lake Whatcom

The calculations of mercury (Hg) fluxes from each tributary of Lake Whatcom require knowledge of the annual discharge when used with total Hg concentrations determined six times between June 2002 and May 2003. Flow data from Smith, Austin and Anderson Creeks were not available during some periods within this one-year interval. Flow data from both Smith and Austin Creeks were not available until after February 5, 2003. Monthly averages of the ratios of the flows of Smith Creek to the flow of Austin Creeks that were calculated from water year 2002 were used to estimate missing data during the water year 2003. Missing flow data from Smith Creek in October 2002, November 2002, and December 1-10, 2002, were estimated by multiplying the daily flows from Austin Creek by the ratio of Smith:Austin flows in October 2001 (0.538), November 2001 (0.627), and December 2001 (0.54), respectively. To account for the five days in December 2002 for which both Smith and Austin data were missing, the total flows for the 26 days were increased by 19 percent.

To estimate the flows in Austin Creek for December 11- 31, 2002, January 2003, and February 1-4, 2003, the daily flow from Smith Creek was divided by the monthly Smith: Austin flow ratio in December 2001 (0.54), January 2002 (0.758) and February 2002 (0.61), respectively. During July 2002, when the Nooksack Diversion was operating most of the time, the Anderson Creek gaging station did not record until July 21. The total monthly flow was based on the estimate of the diversion volume (2,260 million gallons; Peg Wendling, City of Bellingham, written commun., 2003) plus the base flows of Anderson Creek. A base flow of 5.5 ft^3/s was obtained based on gaging station data during the morning of July 29, after flows had decreased as a result of the closing of the diversion at 1:45 pm on July 28. Data for Anderson Creek were not available for December 14, 2002 and the monthly volume calculation based on 30 days was increased by 3 percent.

Flows from Anderson Creek between February 4 and 14, 2003, at a time when the diversion had been off except between 0:00 am and 10:00 am February 4, were estimated by multiplying the Austin Creek flows by the ratio of Austin: Anderson Creek flows (1.32 ± 0.48). The daily volume flow from Anderson Creek on May 26, 2003 was estimated from the average daily volume flows for May 25 and May 27, during which time the diversion was operating.

On the basis of specific conductance, total Hg concentrations, and flow data, the sampling year was divided into a dry season (May through November) and a wet season (December through April). Monthly averages were used to calculate total monthly flows for the gaging stations monitored by the USGS (Silver Beach, Euclid, Mill Wheel, Brannian, Carpenter, and Olsen). Total dry season flows were calculated from the sums of the daily flows from June 1, 2002 through November 30, 2002 and from May 2003. Total wet season flows were calculated from the daily flows between December 1, 2002 and April 30, 2003. When monthly flows could not be calculated, flows for the appropriate months from water year 2002 and 2003 were substituted. In the worst case, the only available data from Mill Wheel Creek for the dry season was collected during July 2002. The July 2002 total monthly flow was multiplied by 7 to calculate the total dry season flow.

Appendix D. Analysis of Core Sections and Estimates of Sedimentation Rates in Lakes in Whatcom County

Collection of Sediment Cores and Preparation for Analysis

Sediment cores were collected from Lake Terrell, Lake Samish, Baker Lake, Wiser Lake, and Fazon Lakes from the Washington State Department of Ecology's (Ecology) 8-m research vessel *R.V. Skookum* using a Wildco stainless-steel box corer fitted with a new, pre-cleaned 13×13×50-cm acrylic liner for each station. Stations were located and positions recorded using a differentially corrected global positioning system (GPS). The length of sediment cores recovered ranged from 50 cm in Wiser and Fazon Lakes to 27.5 cm in Lake Terrell.

After retrieving an undisturbed core, overlying water was siphoned off and the core liner was removed from the stainless-steel box corer. The sediment core was extruded using a gear-driven piston that pushed the sediment column up at 1-cm increments and out the top of the liner. Each extruded 1-cm section then was sliced off at the top of the core line using aluminum plates. The sides of the 13×13×1-cm layer were trimmed to prevent contamination from the liner or shallower sediment adhering to the liner. Each section was placed into an 8-oz glass jar with Teflon® lid liners that had been cleaned to USEPA specifications (U.S. Environmental Protection Agency, 1990), placed in plastic bags, and stored in coolers on ice pending processing in the laboratory. This process resulted in a maximum of 42 subsections per core. The bottom portion of each core (typically 4-6 cm) was sectioned into 2-cm intervals and the sediment section in contact with the bottom plate of the piston plunger was discarded.

Sections for analysis were selected to represent current conditions (top layer), background conditions to calibrate radio-dating method, and representative intervals along the core length to reflect sediments deposited over several decades. Horizons not selected for initial analysis were frozen and archived to allow for future analysis. Sections selected for analysis were homogenized in the laboratory, and subsamples were split into various containers for analysis of total organic carbon (TOC), Hg, stable Pb, and radioactivity.

Before the collection of each core, the core liners and all utensils (aluminum plates and sampling spoons) that contacted samples were pre-cleaned by washing with Liquinox detergent, followed by sequential rinses with tap water, diluted (10 percent) nitric acid, deionized distilled water (DDI), and methanol. Between stations, the corer was thoroughly cleaned by brushing with onsite water.

Methods of Analyses

Total Organic Carbon (Analyzed by Ecology)

Chilled samples were thawed to room temperature and homogenized. About 5-10 g of sample was added to weighed crucibles that had been dried at 70 °C and stored in a desiccator, and the weight of wet sample + crucible was recorded. Samples were then dried in the oven at 70 °C for at least 18 hours. The dried samples were placed into a desiccator to cool, and the weights of each of the dried samples + crucibles were recorded after coming to constant weight (about 3 hours). Samples were ground in a mortar and pestle and sieved. Several drops of 10-percent HCl were added to the blank and each of the samples to remove carbonates. Additional acid was added until no further effervescence occurred. The acid-treated samples were dried again at 70 °C for at least 18 hours before being re-ground, sieved, and stored in a desiccator until analysis.

Sample boats were scraped cleaned, rinsed with DDI water, heated for 30 minutes at 900 °C in a muffle furnace, and cooled. Five grams of the Standard Ottawa Sand was used as the method blank. Sediment samples were analyzed for total organic carbon (TOC) using a Shimadzu Model TOC-5050A Total Organic Carbon Analyzer equipped with a Shimadzu Model SSM-5000 Solid Sample Module. In this method, subsamples were combusted at 900 °C using high-purity oxygen as the carrier gas. The CO_2 formed by the combustion is measured by the non-dispersive infrared detector (NDIR) and compared to a calibration curve. A 4-point calibration curve was developed using the peak area response of 0 to 15 mg of glucose as the standard and was required to have a regression coefficient of at least 0.997. One-gram samples of standard reference materials (SRM) that were representative of the type of samples being analyzed were used for verification. Two aliquots of each sample weighing enough to result in a peak that is close to the upper limit of the calibration curve (250 to 500 mg) were added to clean sample boats. Quality-control procedures included analyses of laboratory blanks, laboratory replicates, and SRMs. The resulting total carbon (TC) value is considered to be total organic carbon (TOC) because inorganic carbon was removed in the acidification step.

Stable Lead (Analyzed by Ecology)

Samples for stable lead (Pb) analyses were oven dried at 105 ± 4 °C (usually overnight) and cooled. One gram of dried sample was transferred to a tared 250-mL beaker, to which 10 mL of 1:1 mixture of trace-metal grade HNO_3 and DDI water was added. The sediment slurry was mixed, covered with a watch glass, heated to 95 °C, and refluxed for 10 minutes without boiling. The slurry was allowed to cool, and an additional 5 mL of concentrated HNO_3 was added. The beaker was covered and slurry was refluxed for 30 minutes. This last step was repeated to ensure complete oxidation. The solution was allowed to evaporate to 5 mL without boiling, while maintaining a covering of solution over the bottom of the beaker that was covered with a ribbed watch glass. After the solution was cooled, 2 mL of DDI water and 3 mL of 30 percent H_2O_2 (low-metal content) was added. The beaker was covered with a watch glass and warmed on a hotplate to start the peroxide reaction, in such a manner as to ensure that losses did not occur due to excessively vigorous effervescence. Additional l-mL aliquots of 30-percent H_2O_2, up to a total volume of 10 mL, were added while warming until the effervescence was minimal or until the general sample appearance did not change. After effervescence subsided and the beaker was cooled, 5 mL 1:1 mixture of HCl and DDI water and 10 mL DDI water was added. The covered beaker was returned to the hotplate and heated for an additional 10 minutes. The slurry was filtered through a 0.8-micron pre-acid washed membrane filter into a tared 125-mL polyethylene sample bottle and diluted to 100 mL, 101 g, with DDI water.

Sample material in solution was introduced by pneumatic nebulization into the Inductively Coupled Plasma Mass Spectrometer (ICP-MS) VG PQ ExCell, where a radio-frequency plasma transferred energy that caused desolvation, atomization, and ionization. The ions were extracted from the plasma through a vacuum interface and separated on the basis of their mass-to-charge ratio by a quadrupole mass spectrometer. The ions passing through the mass spectrometer are counted by a dual-stage discrete dynode electron multiplier detector. Daily startup procedures for the ICP-MS analysis included tuning for mass calibration, sensitivity, background, oxide formation, and double ionization. The instrument was calibrated daily using 1- to 20-µg/L standards prepared from 100-µg/mL multi-element stock solutions (Spex QC-19 and QC-7, High Purity Standards and Inorganic Venture). The sediment digests were diluted 1:10 with DDI, and internal standards having similar analytical behavior to the elements being determined were added before the solution was introduced into the ICP-MS. During the analytical run, instrumental drift and sample-matrix effects were corrected with changes in the sensitivity of the internal standards and the calibration verified by independent multi-element stock standard for ICP-MS.

Radioactive Tracers (Analyzed by Severn Trent Services, Richland, Washington)

^{210}Pb was determined by low-energy photon gamma spectrometry following drying and sieving using EPA method 901.1. ^{137}Cs in sediments were determined in a similar manner using intrinsic hyperpure germanium gamma spectrometry. Detection limits for some samples, especially those at the top of the organic-rich cores near the sediment-water interface, were high because the samples sent to the laboratory did not contain enough solid material owing to the high percentage of water.

Total Mercury (Analyzed by Ecology)

Samples for analysis of total mercury (Hg) were pulverized and mixed well before weighing 1.00 ± 0.01 g of sediment into a stoppered, tared 250-mL biological oxygen demand bottle. DDI water (5.0 ± 0.1 mL) and 5.0 mL of aqua regia (three volumes of concentrated trace-metal-grade HCl to one volume of concentrated trace-metal-grade HNO_3) was added and the bottle was placed in the covered water bath, where it was held for 2 minutes and 40 seconds at 91.5 °C. After removing from the water bath, the bottle was cooled to room temperature. DDI water (50.0 ± 0.5 mL) and

15.0 ± 0.1 mL 5-percent potassium permanganate reagent was added and allowed to stand for 15 minutes. If the permanganate color did not persist, an additional 5 mL of permanganate was added. After adding 8.0 ± 0.1 mL of 5-percent potassium persulfate reagent, the bottle was heated in a covered water bath at 91.5 °C for 40 minutes. After removing from bath and cooling, the excess solid brown permanganate at the bottom of the bottle was reduced with the addition of 6.0 ± 0.1 mL of a 1.73-M hydroxylamine hydrochloride solution and swirling vigorously under a hood. Additional hydroxylamine hydrochloride was added in 1.0 ± 0.1 mL increments and the bottle was swirled vigorously after each addition until the brown solid was no longer visible. After adding 55 ± 0.5 mL of DDI, samples were allowed to stand for at least 1 hour. If the supernatant was cloudy after standing, the sample was filtered through a pre-rinsed, 0.8-µm syringe filter before transferring to an analysis tube.

With the CETAC M-6000A cold vapor Hg absorbance analyzer, a $SnCl_2$ solution and the sample solution were pumped simultaneously into a mixing tee. The reaction of $SnCl_2$ with Hg(II) resulted in the reduction of Hg(II) to Hg0. The mixture was pumped through a gas-liquid separator and drying tube into the sample absorbance cell with high-purity argon. Prior to analysis, the drying tube and flows on the CETAC M-6000A were checked. The amount of light absorbed at 253.7 nanometers was proportional to the Hg level in the sample.

Calibration was performed using 4.0, 2.0, 1.0, 0.5, 0.25, and 0.0 µg Hg/L standards prepared from a secondary 1,000 µg/L Hg standard diluted from the primary 100 µg/mL SPEX (Metuchen, NJ) Hg standard. The instrument was recalibrated if the correlation coefficient was less than 0.995. During the analytical run, calibration and procedural blanks, calibration verification standards, independent calibration verification standards, laboratory replicates, and laboratory control standards were performed. Concentrations calculations were based on peak height modeled from four highest readings. Quality-control samples for the core samples included procedural blanks, field splits, blind standard reference material, and interlaboratory duplicates.

The 30-34 cm section of the basin 3 core of Lake Whatcom appeared to be contaminated, possibly from the bottom plunger of the core extractor. The Pb and Hg analyses were repeated for the 29-31 cm section. The percentage of solids and TOC analyses from the 30-34 cm section were used for the 29-31 cm section.

Age Dating

The rates of sediment accumulation in the lakes must be known in order to correlate Hg concentrations in section cores at different depths with the dates in which the sediments initially were deposited, and to calculate Hg sedimentation rates. Sedimentation rates based on linear distances down the sediment core are sometimes skewed because interstitial water is squeezed out of the sediments by the weight of the overlying sediments, causing compression. In this report, sediment rates are based on sediment mass accumulation. In order to convert from a weight-to-weight basis of the percentage of solids measurement to a weight-to-volume basis that is needed to calculate mass accumulation, the porosity is defined as the percentage of volume occupied by water. The percentage of solids by weight is a function of the porosity of the section sample (Φ) and the densities of water (ρw= 1 g/cm^3) and sediment grain (ρs) according to the equation:

Equation D1 is solved for porosity according to

$$\text{percentage of solids} = \rho s \times \frac{(1-\Phi)}{((\rho w \times \Phi) + \rho s(1-\Phi))} \quad \text{(D1)}$$

$$\Phi = \frac{(1 - \text{percentage of solids})}{(\text{percentage of solids}/(\rho s - \text{percentage of solids-1}))} \quad \text{(D2)}$$

Given a sediment density of 2.7 g/cm^3, the mass accumulation of a 1-cm section is calculated according to

$$\text{Mass accumulation} = \rho s(1-\Phi) \quad \text{(D3)}$$

Where percentage of solids were not measured, extrapolation between known values was used. The mass accumulation with depth in the core (g/cm^2) is the cumulative masses of the overlying 1-cm thick sections. Six estimates of sediment accumulation rates [(g/cm^2)/yr] were attempted using cumulative mass accumulation: (1) The decay of the ^{210}Pb (Schell, 1986), (2) the first appearance of stable Pb between 1920 and 1940, (3) the shape of the rise in elevated concentrations of stable Pb from first appearance to peak concentrations, (4) the peak concentration in 1975 of stable Pb due primarily to tetraethyl Pb in gasoline (Yake, 2001), (5) the first appearance of elevated concentration in 1953 of ^{137}Cs as a result of atmospheric testing of thermonuclear weapons, and (6) peak concentrations of ^{137}Cs in 1964 (Charles and Hites, 1987). The peak concentrations and first appearances of stable Pb and ^{137}Cs are considered markers because these four features are associated with specific time marks. Constant sedimentation rates were assumed because consistent values were obtained on the basis of the above events and processes that represented time intervals ranging from 25 to 100 years.

^{210}Pb in the atmosphere is produced from the decay of atmospheric ^{222}Rn and thus has a different pathway to lakes than natural stable Pb. ^{210}Pb in the atmosphere is quickly scavenged onto atmospheric particles, which settle to the water and land surfaces. Once in the water column of lakes, the geochemical behavior of ^{210}Pb will be similar to stable Pb and will quickly settle to the sediment-water interface by attaching onto particles. Once delivered to the sediment column, the ^{210}Pb on the particles will be buried by deposition of newly settling particles. ^{210}Pb decays with a half-life of 22.6 years. Assuming that the sedimentation rate remained constant, the depth in the sediment at which the ^{210}Pb radioactivity decreases to one-half of the surface value represents 22.6 years of sedimentation. The sedimentation rate based on ^{210}Pb activity is calculated from the slope of the linear regression of the natural log of the ^{210}Pb activity, after correcting for the low level of the blank and activity supported by radium decay in the sediments, versus the mass accumulation rate with depth. For most cores for which ^{210}Pb sedimentation rates were calculated, the sedimentation rates were based on only a few detectable measurements of ^{210}Pb activity. In addition, there was significant scatter in the ^{210}Pb data in some cores. Therefore, no assessment of the variation in sedimentation rate based on ^{210}Pb activity was made.

The sedimentation rate based on the peak in stable Pb was determined by dividing the accumulated mass sedimentation at the mid-point of the interval in which the peak Pb was found by 27 years (2002-1975). The confidence interval for the sedimentation rate based on stable Pb peak was determined by the broadness of the peak. The peak was defined as the mid-point of those sections in which Pb values are within 10 percent of the highest value. Because not all depth intervals were analyzed, the upper boundary of the peak is defined as the 1-cm section farthest away from the peak before measured Pb less than 90 percent of the peak value was

encountered as the depth interval was decreased. Likewise, the lower boundary of the peak was defined by the 1-cm section prior to encountering a Pb value less than 90 percent of the peak value with increasing depth intervals. The same approach was used for the ^{137}Cs peak as a marker for 1964 (38 years before present).

The first appearances of elevated concentrations of ^{137}Cs and stable Pb as a result of anthropogenic atmospheric inputs were used to assess the upper and lower limits of the average sedimentation rates during the last 50 and 75 years, respectively. The first appearance of elevated Pb values in sediments in western Washington lakes occurred between 1920 and 1940 (Yake, 2001). In some cores, the natural variability in pre-industrial Pb concentrations makes it difficult to identify anthropogenic increases. The confidence intervals for long-term sedimentation rates based on first appearance of ^{137}Cs and stable Pb incorporated both uncertainty in identifying first appearance of elevated concentrations in cores and uncertainty in the dates of anthropogenic influences within the region. The upper limit of the sedimentation rate for each core was calculated by dividing the accumulated mass sedimentation for the deepest section in which stable Pb first appeared to increase by 60 years (about 1940). The lower limit was calculated by dividing the accumulated mass sedimentation for the shallowest section in which stable Pb definitely increased above background by 80 years (about 1920). Likewise, confidence intervals for sedimentation rates based on the first appearance of ^{137}Cs were based on identifying the possible 1-cm section in which ^{137}Cs first increased as defined by the years between 1952 and 1958.

Given the range of sedimentation rates estimated from the five methods above, stable Pb concentrations were plotted with preliminary dates. Because grain size affects the magnitude of the enrichment in Pb, the concentrations were normalized to the maximum concentration. The sedimentation rates for the three Lake Whatcom cores then were adjusted manually until the shape of the rise of stable Pb matched among the three cores. Likewise, the shapes of the rise of Pb in the cores from the three lakes north of Bellingham (Terrell, Wiser, and Fazon) were matched.

Each sedimentation rate for a specific core was assigned a weighting factor (low, moderate, and high) based on the quantity and quality of the available data on which the estimate was based. A single estimate of the sedimentation rate for each core was derived by giving more consideration to estimates with highly weighted factors, while still satisfying the range for estimates with low weighting factors. A confidence level of high, moderate, or low was assigned the overall sedimentation rate, based on the consistency among the sedimentation rates derived from the different methods and the weighting factor of each result.

General Characteristics of the Sediment Column and Estimates of Sedimentation Rates in Lakes in Whatcom County

Eight sediment cores were collected from lakes in Whatcom County by USGS and Ecology personnel and the overall quality of the quality-assurance data was good. The data for the three cores from Lake Whatcom are reported in Norton (2004). Descriptions of the cores from five other lakes in Whatcom County are given in table D1 and the analytical results for selected core sections are summarized in table D2.

Lake Whatcom Basin 1

Percentage of solids increased from 13.9 percent at the surface to about 19 percent in the sections between 3 and 8 cm (fig. D1). Below 10 cm, percentage of solids ranged from 14 to 18 percent with a minimum observed at about 25 cm. The total organic carbon remained fairly constant throughout the core at values between 8 and 10 percent, except for one anomalous value of 18.1 percent at 11-13 cm. The top 16 cm appeared to be a layer of uniform brown silt. Wood debris was found between 16 and 19 cm. Between 32 and 44 cm, the sediment appeared to be a layer of compacted light brown clay.

Three sections within the top 10 cm had detectable ^{210}Pb values, and all six sections below 10 cm had undetectable values with an average 1/2 method detection limit (MDL) of 0.13 pCi/g. The ^{210}Pb data indicate a sedimentation rate of 0.046 (g/cm^2)/yr, which was assigned a low weighting factor because of the few detectable values. Stable Pb peaked in the 3-5 cm section and decreased to consistent low concentrations below 21 cm. Using Pb at 4 cm as the 1975 marker, a sedimentation rate of 0.034 (g/cm^2)/yr was calculated and given a high weighting factor. The confidence interval for this calculation was quite large [0.018 to 0.074 (g/cm^2)/yr] because measured values on both sides of the Pb peak were within 10 percent of the peak value. ^{137}Cs exhibited a 3-point increase, reaching a peak concentration in 7-9 cm section and undetectable concentration in two sections below 15 cm. Assigning 8 cm as the 1964 ^{137}Cs marker yielded a sedimentation rate of 0.050 (g/cm^2)/yr [confidence level of 0.032 to 0.072 (g/cm^2)/yr], which was assigned a moderate weighting factor.

The first appearance of elevated stable Pb between 14 and 21 cm yielded a confidence interval ranging from 0.035 to 0.068 (g/cm^2)/yr when the marker was assigned dates between 1920 and 1940, which was given a high weighting factor. The first appearance of ^{137}Cs between 9 and 15 cm for the dates between 1953 and 1958 yielded a confidence interval between 0.045 and 0.062 (g/cm^2)/yr, which was assigned a moderate weighting factor. The increase of stable Pb using an initial estimate of the sedimentation rate of 0.045 (g/cm^2)/yr matched the increase in stable Pb of the core from basin 2 of Lake Whatcom.

The maximum sedimentation rates for the four marker events were similar to the average rate calculated from the ^{210}Pb calculation (fig. D2 and table D3). Weighting the stable Pb profile higher than the ^{137}Cs profile, a sedimentation rate of 0.045 (g/cm^2)/yr was assigned to this profile.

Lake Whatcom Basin 2

Percentage of solids increased from 15.3 percent at the surface to about 22 percent from 3 to 9 cm. Between 11 and 17 cm, the percentage of solids were stable at 17.8 percent before increasing in the bottom 20 cm of the 44-cm core (fig. D3). TOC values were relatively stable between 5 and 6 percent in the top 5 cm, with slightly higher values of 6 percent found between 11 and 18 cm. Within the top 4 cm, small wood fragments and other black particles were observed, but otherwise the core appeared to be uniform gray silt throughout its length.

Four values of ^{210}Pb were detectable within the top 9 cm, and six undetectable values below 11 cm were determined to have an average 1/2 MDL of 0.4 pCi/g. Average sedimentation within the top 9 cm was calculated to be 0.105 (g/cm^2)/yr. Because the regression coefficient of the slope of ln(excess ^{210}Pb) versus accumulated mass was only 0.916, the ^{210}Pb sedimentation rate was assigned a low weighting factor. The stable Pb profile was uniform within the top 5 cm. Although an average or minimum sedimentation rate was not assigned, assigning the 1975 Pb peak marker to the 6-7 cm section that was not analyzed yielded a maximum sedimentation rate of 0.059 (g/cm^2)/yr. Assigning the 1964 ^{137}Cs marker to the 4-cm horizon yielded a sedimentation rate of 0.023 (g/cm^2)/yr, with confidence intervals between 0.016 and 0.051 (g/cm^2)/yr. The sedimentation rate based on the ^{137}Cs peak marker was given a high weighting factor.

Stable Pb increased above background concentrations between the 14 and 17 cm horizons, which yielded minimum and maximum sedimentation rates of 0.039 and 0.063 (g/cm^2)/yr, respectively. The 1953 to 1958 first appearance of ^{137}Cs occurred somewhere within the 10-16 cm section, yielding sedimentation rates between 0.047 and 0.079 (g/cm^2)/yr. The range of sedimentation rates calculated from the first appearance of the ^{137}Cs and stable Pb were consistent and were given a moderate weighting factor. The increase of stable Pb using an initial estimate of 0.040 (g/cm^2)/yr matched the increase in stable Pb in the core from basin 1 of Lake Whatcom.

Maximum sedimentation rates for the four marker events ranged from 0.051 to 0.079 (g/cm^2)/yr, which was three times lower than the average rate calculated from the ^{210}Pb calculation (fig. D2 and table D3). Therefore, the ^{210}Pb data were disregarded. Satisfying all the minimum and maximum values for the highly weighted factors limited sedimentation rates between 0.016 and 0.051 (g/cm^2)/yr, whereas a value greater than 0.039 (g/cm^2)/yr is required to satisfy the moderately weighted first-appearance estimates (table D3). A sedimentation rate of 0.040 (g/cm^2)/yr is assigned.

Lake Whatcom Basin 3

The percentage of solids increased from 17.6 percent at the surface to 50 percent in the 6-8 cm section (fig. D4). Below 10 cm, percentage of solids ranged from 37 to 41 percent. TOC decreased from 4.8 percent at the surface to about 2.5 percent between 3 and 8 cm. Between 10 and 22 cm, TOC ranged from 4 to 5 percent, but decreased to 2 percent in the 25-28 cm section. The percentage of solids, stable Pb, and ^{210}Pb data all indicated a mixed layer in the top 2 cm. The stable Pb data indicated that the most contaminated mixed layer may extend down to 5 cm. Between 20 and 30 cm, the core is uniform silt containing visible plant material. Below 31 cm, black layering was observed in the core.

The ^{210}Pb sedimentation rate determined from data in the top 6 cm was 0.034 (g/cm^2)/yr. Because the correlation coefficient was 0.97 for four points, this value was given a high weighting factor. Below 6 cm, the ^{210}Pb concentration was undetectable at a MDL of 0.88 pCi/g in the 6-8 cm section, but the 10-12 cm section contained a detectable ^{210}Pb concentration of 1.53 pCi/g. If the 10-12 cm data were used for the ^{210}Pb dating calculation, the correlation coefficient decreased to 0.72 and the sedimentation rate increased to 0.080 (g/cm^2)/yr. This sedimentation rate was assumed to be the upper limit of the ^{210}Pb method, and the upper limit is given a low weighting factor. The 1975 Pb peak marker lay within the top 5 cm, which yielded a maximum sedimentation rate of 0.074 (g/cm^2)/yr. The one detectable ^{137}Cs concentration and a small Pb peak in the 15-17 cm section are disregarded in the calculation of the sedimentation rate. When the increase in the normalized Pb concentration from basin 3 is matched to the increase in Pb for basins 1 and 2, the resulting sedimentation rate is 0.052 (g/cm^2)/yr, which is given a moderate weighting factor. Given slightly more weighting to the ^{210}Pb sedimentation value than the Pb matching value (fig. D2), a sedimentation rate of 0.040 (g/cm^2)/yr is assigned (table D3).

Lake Samish

The percentage of solids were 14.1 percent at the surface and were 29.2 percent between 6 and 11 cm (fig. D5). The data from two sections (1-2 cm and 3-5 cm, table D2) were disregarded. TOC decreased from 5.7 percent at the surface to 4.7 percent within the 6-8 cm section. The percentage of solids between 2 and 6 cm were linearly extrapolated using the TOC content. The uncertainty in the mass accumulation with depth adds considerable uncertainty to the sedimentation rate calculations, especially for those markers within or just below the zone of disregarded percent solid values (1-5 cm). The core appeared to be brown silt down to 25 cm, and the bottom 6 cm appeared to be finer light brown silt.

Four samples in the upper 8 cm contained detectable ^{210}Pb concentrations, but the decrease with depth appeared to be stepwise rather than a smooth decay. No attempt was made to calculate a sedimentation rate based on ^{210}Pb. Stable Pb exhibited a distinct peak of 63 mg/kg within the 6-8 cm section, which yields a sedimentation rate of 0.072 (g/cm^2)/yr (fig. D6 and table D3). ^{137}Cs showed peaks both at the surface and within the 6-8 cm section, and no calculation based on the 1964 ^{137}Cs peak was attempted. The first appearance of ^{137}Cs could have occurred somewhere between the 9-cm and 12-cm horizons, translating into maximum and minimum sedimentation rates of 0.046 and 0.067 (g/cm^2)/yr, respectively. Low concentrations of stable Pb were found below 20 cm and the first appearance of anthropogenic stable Pb could be located somewhere between 21 and 25 cm, yielding minimum and maximum sedimentation rates of 0.056 and 0.082 (g/cm^2)/yr, respectively (fig. D6).

The Lake Samish core was assigned a sedimentation rate of 0.072 (g/cm^2)/yr, primarily on the basis of the position of the stable Pb peak.

Lake Terrell

Percentage of solids ranged from 8 to 11 percent within the top 14 cm, before increasing to about 20 percent at the bottom of the 25-cm core (fig. D7). TOC increased smoothly from a concentration of 19.4 percent at the surface to 33 percent in the 12-14 cm section, and then decreased to about to 16 percent at depth. The top of the core contained a silty layer. Wood fibers and plant material were observed between 16 and 21 cm, with a limb lodged vertically within the core between 9 and 17 cm. Below 22 cm, the sediment appeared to be compacted silt and clay.

Seven sections had detectable ^{210}Pb concentrations and two concentrations at depth had an average 1/2 MDL of 0.62 pCi/g. The decrease in ^{210}Pb with depth was not smooth, resulting in a regression coefficient of 0.86 for the slope of the ln(excess ^{210}Pb) versus depth. The average sedimentation rate was calculated as 0.095 (g/cm^2)/yr.

There was a broad maximum of about 90 mg/kg in stable Pb in the upper 11 cm, with a decrease in the 1-2 cm section. Below 12 cm, stable Pb decreases to 6.6 mg/kg within the bottom section (23-25 cm). If the 1975 horizon is assigned to the middle of the broad peak, a sedimentation rate of 0.029 (g/cm^2)/yr is calculated, although this value is given a low weighting factor because of the broadness of the peak and the second peak at the surface. A maximum sedimentation rate based on 1975 stable Pb peak being no deeper than 12 cm was 0.046 (g/cm^2)/yr, which was assigned a moderate weighting factor. Three sections between 3 and 14 cm contained detectable amounts of ^{137}Cs with the peak located within the 5-7 cm section, yielding a sedimentation rate of 0.029 (g/cm^2)/yr that was assigned a high weighting factor. Given that no ^{137}Cs data was collected between 7 and 12 cm, the ^{137}Cs peak could have been as deep as 12 cm, yielding a maximum sedimentation rate of 0.047 (g/cm^2)/yr.

The first appearance of ^{137}Cs occurred somewhere between 15 and 18 cm, yielding minimum and maximum sedimentation rates of 0.031 and 0.048 (g/cm^2)/yr, respectively, which were assigned a moderate weighting factor. Stable Pb concentrations in the bottom section (6.6 mg/kg) were near background concentrations, yielding minimum and maximum sedimentation rates of 0.038 and 0.054 (g/cm^2)/yr, respectively. Because lower values could have been present below 25 cm, sedimentation rates based on first appearance of elevated stable Pb were assigned a low weighting factor. Matching the rise of stable Pb with the profiles from Fazon and Wiser yielded a sedimentation rate of 0.025 (g/cm^2)/yr.

Maximum sedimentation rates from three markers ranged from 0.046 to 0.054 (g/cm^2)/yr (fig. D6), whereas ^{210}Pb decay indicates a sedimentation rate of 0.095 (g/cm^2)/yr. The core was assigned a sedimentation rate of 0.031 (g/cm^2)/yr, based primarily on the 1964 ^{137}Cs peak marker (0.047 (g/cm^2)/yr) with a slight adjustment downward to match the stable Pb increase (table D3).

Wiser Lake

The percentage of solids varied within a narrow range between 9 and 12 percent for this 44-cm core (fig. D8). TOC increased from 18 percent at the surface to 23.6 percent in the 32-35 cm section. The top appeared to be disturbed, with the underlying sediment having a uniform appearance of black to brown silt with some organic materials. Woody debris was present between 14 and 16 cm.

^{210}Pb concentrations in the top 2 cm were lower than those in the 7-9 cm section, which is consistent with the disturbed appearance of the top of the core. Below 20 cm, four core sections had undetectable ^{210}Pb concentration with a 1/2 MDL of 0.44 pCi/g. Given the ^{210}Pb profile, a sedimentation rate based on ^{210}Pb decay was not attempted. Stable Pb exhibited a narrow peak of about 66 mg/kg between 11 and 16 cm. Assigning the 1975 stable Pb peak to the 12-cm horizon yields a sedimentation rate of 0.049 (g/cm^2)/yr, with a confidence interval between 0.041 and 0.079 (g/cm^2)/yr and a high weighting factor. Stable Pb decreased to 2.55 mg/kg in the 40-44 cm section. If this Pb concentration was the background level, then minimum and maximum sedimentation rates based on the first appearance of anthropogenic Pb were estimated to have been 0.049 and 0.072 (g/cm^2)/yr, respectively. Because it is possible, but not likely, that lower Pb concentrations could be present below 44 cm, sedimentation rates based on first appearance of elevated stable Pb were assigned a moderate weighting factor. ^{137}Cs was not detected in four sections of the core at a maximum MDL of 0.99 pCi/g.

Based solely on the stable Pb profile, the sedimentation rate for Wiser Lake was estimated to be 0.049 (g/cm^2)/yr (fig. D8 and table D3).

Fazon Lake

Percentage of solids generally increased from about 7 percent at the surface to 11 percent within the 27-30 cm section before decreasing to 8 percent at the 40-44 cm section (fig. D9). TOC concentrations were high, ranging from 22 to 28 percent. The core appeared uniform throughout its length.

^{210}Pb concentrations in the top 5 cm were either not detectable at MDL of 5 pCi/g or were lower than those in the 7-9 cm section. ^{210}Pb generally decreased down core, but was constant at detectable concentrations between 27 and 44 cm. A calculation based on the ^{210}Pb decay between 7 and 30 cm yielded a sedimentation rate of 0.044 (g/cm^2)/yr, which was assigned a low weighting factor.

Stable Pb exhibited a sharp peak of 88 mg/kg within the 11-13 cm section, which corresponds to a sedimentation rate of 0.035 (g/cm^2)/yr (range 0.029 to 0.045 (g/cm^2)/yr), which was assigned a high weighting factor. ^{137}Cs was not found in the upper 5 cm at a maximum MDL of 2.6 pCi/g. Consistent with the stable Pb, a broad three-point peak in ^{137}Cs was centered within the 15-18 cm section, yielding a sedimentation rate of 0.039 (g/cm^2)/yr, which was assigned a moderate weighting factor. The minimum sedimentation rate calculated from the ^{137}Cs peak was estimated to be 0.022 (g/cm^2)/yr, but no maximum rate for either the peak or first appearance of ^{137}Cs could be calculated because no core sections below the ^{137}Cs peak were analyzed.

Stable Pb concentration at the bottom of the core was 13.3 mg/kg, which was higher than concentrations in the bottom of all the cores thought to represent a date before 1940. Because the bottom of the core may not have represented a pre-anthropogenic date, only a minimum sedimentation rate of 0.051 (g/cm^2)/yr could be estimated and was given a low weighting factor. The sedimentation rate of 0.039 (g/cm^2)/yr obtained from matching the shape of the stable Pb peak was consistent with the range obtained for the stable Pb peak and the ^{137}Cs peak, and 0.039 (g/cm^2)/yr was assigned a high weighting factor (fig. D6). A sedimentation rate for Fazon Lake was estimated to have been 0.049 (g/cm^2)/yr, and was assigned a moderate confidence factor (table D3).

Baker Lake

Percentage of solids increased from 30 percent at the surface to values between 40 and 48 percent between 8 cm and the bottom of the core at 27 cm (fig. D10). TOC concentrations were low (1.5 to 2.8 percent) throughout the core. The sediment texture was uniformly silt and clay with black layers first appearing at 3 cm. Wood debris was present below 16 cm.

^{210}Pb and stable Pb showed no distinctive patterns. In contrast, a concentration of ^{137}Cs that was below the MDL (0.73 pCi/g MDL) was present at the surface, with a smooth increase of five core sections that were above the MDL reaching the maximum within the 21-23 cm section. Only a minimum mass-accumulation rate was calculated, based on the ^{137}Cs peak at the bottom of the core representing 1964 (table D3).

These calculated time intervals are subject to same uncertainty of both individual estimates of sedimentation rates (figs. D2 and D6), as well as differences in the ranges among the six methods. Because each method is sensitive to sedimentation rates during different periods, difference in sedimentation rate among methods also may reflect changes in sedimentation rates with time or mixing of sediments near the sediment-water interface. Reconciling the differences in the ranges of sedimentation rates among the methods should decrease the uncertainty of the assigned sedimentation rate.

Sediment Focusing in Lake Whatcom

In most lakes, the topographic relief of the lake bottom results in an uneven distribution of sediments. The accumulation of sediments in the deep areas of lakes at rates above the average sedimentation for a lake is called sediment focusing. Because concentrations of particulate reactive metals, such as Pb and Hg, on suspended matter generally increase with decreasing particle size, differential settling of the different size fractions also focuses metal accumulation in deep depositional zones. Larger particles with lower-than-average metal concentrations settle out in shallow waters near mouths of the tributaries to Lake Whatcom, whereas fine-grained particles with higher concentrations of particulate reactive metals travel farther and settle in deeper waters (Norton, 2004). Sediment focusing is addressed in this report solely for the purpose of assessing one possible reason for the imbalance of particle mass in Lake Whatcom.

The effect of sediment focusing of sediment and particulate reactive metals in Lake Whatcom was attempted by comparing the atmospheric flux of ^{210}Pb to the accumulation in the sediments (Appleby and Oldfield, 1992; Crusius and Anderson, 1995). The inventory of excess ^{210}Pb down to a depth at which the concentration was undetectable (10-13 cm) was calculated for each core (table D4). Missing values within the core were estimated by using logarithmic extrapolations between the three to four detectable concentrations within each core. The steady state flux of excess ^{210}Pb (table D4) was calculated by dividing ^{210}Pb inventory by the mean life of ^{210}Pb (32.2 years). A focusing factor then was derived by dividing the steady-state flux of excess ^{210}Pb by atmospheric and fluvial inputs of ^{210}Pb. The fluvial input of ^{210}Pb was found to be insignificant (less than 0.005 (pCi/cm^2)/yr) in Lake Sammamish, a lake of similar size in western Washington, and will be disregarded in the calculations for Lake Whatcom. The atmospheric deposition of ^{210}Pb in western Washington has been measured at 0.13 ± 0.08 (pCi/cm^2)/yr (Balistrieri and others, 1995), 0.14 (pCi/cm^2)/yr (Barnes and others, 1979), and 0.20 ± 0.11 (pCi/cm^2)/yr (Nevissi, 1985). Using an average value of 0.155 (pCi/cm^2)/yr, focusing factors of 11, 12, and 6 were estimated for basins 1, 2, and 3 respectively. The focusing factors estimated above should not be applied to the mass balance of a constituent because the factors were calculated with core inventories of ^{210}Pb derived from sediment cores with incomplete concentration data and high ^{210}Pb method detection limits.

References Cited

Appleby, P.G., and Oldfield, F., 1992, Application of lead-210 to sedimentation studies, *in* Ivanovich, M., and Harmon, R.S., eds., Uranium-series disequilibrium: Applications to Earth, Marine and Environmental Sciences (2nd ed.): Oxford, Clarendon Press, p. 731-778.

Balistrieri, L.S., Murray, J.W., and Paul, B., 1995, The geochemical cycling of stable Pb, ^{210}Pb, and ^{210}Po in seasonally anoxic Lake Sammamish, Washington, USA: Geochimica Cosmochimica Acta, v. 59, no. 23, p. 4845-4861.

Barnes, R.S., Birch, P.B., Spyridakis, D.E., and Schell, W.R., 1979, Changes in the sedimentation histories of lakes using lead-210 as a tracer for sinking particles, *in* International Symposium on Isotope Hydrology, p. 875-898.

Charles, M.J., and Hites, R.A., 1987, Sediments as archives of environmental pollution trends, *in* Hites, R.A., and Eisenreich, S.J., eds., Sources and Fate of Aquatic Pollutants: Washington, D.C., American Chemical Society, p. 365-389.

Crusius, J., and Anderson, R.F., 1995, Sediment focusing in six small lakes inferred from radionuclide profiles: Journal of Paleolimnology, v. 13, p. 143-155.

Nevissi, A.E., 1985, Measurements of ^{210}Pb atmospheric flux in the Pacific Northwest: Heatlh Physics, v. 48, p. 169-174.

Norton, D., 2004, Mercury in sediments from Lake Whatcom: Determination of spatial and temporal patterns and characterization of tributary inputs: Olympia, Washington, Washington State Department of Ecology, NPI.

Schell, W.R., 1986, Deposited atmospheric chemicals: Environmental Science & Technology, v. 20, no. 9, p. 847-853.

U.S. Environmental Protection Agency, 1990, Specifications and guidance for obtaining contaminant-free sample container: Office Solid Waste and Emergency Response, U.S. Environmental Protection Agency Directive #93240.0-05.

Yake, B., 2001, The use of sediment cores to track persistent pollutants in Washington State: Olympia, WA, Washington State Department of Ecology, No. 01-03-001, 44 p.

Table D1. Sediment core logs for lakes in Whatcom County, Washington

[**Analysis**: Pb-210, lead-210; Cs-137, cesium-137; T. Hg, total mercury; T. Pb, total lead; TOC, total organic carbon. **Analytical sample No.**: Sample in core sent out for analysis. Up to two sections may have been combined to form an analytical sample. cm, centimeter. –, no data]

Section	Analysis	Analytical sample No.	Ecology sample No.	Length (cm)	Description	Notes
Lake Samish (East Arm) **ID: Sam-1** **Date collected: 9/23/02 12:30 PM** **Latitude/Longitude: 48°39.898 N 122°23.099 W** **Depth of water: 6.6 meters**						
1	Pb-210, Cs-137, T. Hg, T. Pb, TOC	1	40-8577	1	Brown silt with some organics	No archive sample
2	Pb-210, T. Hg, T. Pb, TOC	2	40-8578	1	Brown silt with some organics	No archive sample
3	Archive	–	–	1	Brown silt with some organics	–
4	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	40-8579	1	Brown silt with some organics	No archive sample
5	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	–	1	Brown silt with some organics	No archive sample
6	Archive	–	–	1	Brown silt with some organics	–
7	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	40-8580	1	Brown silt with some organics	–
8	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	–	1	Brown silt with some organics	–
9	Archive	–	–	1	Brown silt with some organics	–
10	Pb-210, T. Hg, T. Pb, TOC	5	40-8581	1	Brown silt with some organics	–
11	Pb-210, T. Hg, T. Pb, TOC	5	–	1	Brown silt with some organics	–
12	Archive	–	–	1	Brown silt with some organics	–
13	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	40-8582	1	Brown silt with some organics	–
14	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	–	1	Brown silt with some organics	–
15	Archive	–	–	1	Brown silt with some organics	–
16	Pb-210, T. Hg, T. Pb, TOC	7	40-8583	1	Brown silt with some organics	–
17	Pb-210, T. Hg, T. Pb, TOC	7	–	1	Brown silt with some organics	–
18	Archive	–	–	1	Brown silt with some organics	–
19	Pb-210, Cs-137, T. Hg, T. Pb, TOC	8	40-8584	1	Brown silt with some organics	–
20	Pb-210, Cs-137, T. Hg, T. Pb, TOC	8	–	1	Brown silt with some organics	–
21	Archive	–	–	1	Brown silt with some organics	–
22	Archive	–	–	1	Brown silt with some organics	–
23	Pb-210, T. Hg, T. Pb, TOC	9	40-8585	1	Brown silt with some organics	No archive sample
24	Pb-210, T. Hg, T. Pb, TOC	9	–	1	Brown silt with some organics	No archive sample
25	Archive	–	–	1	Brown silt with some organics	–
26	Archive	–	–	2	Fine light brown silt	–
27	Pb-210, T. Hg, T. Pb, TOC	10	40-8586	2	Fine light brown silt	–
28	Pb-210, T. Hg, T. Pb, TOC	10	–	2	Fine light brown silt	–
Lake Terrell **ID: Ter-1** **Date collected: 9/24/02 9:30 AM** **Latitude/Longitude: 48°51.819 N 122°41.182 W** **Water depth: 0.6 meter**						
1	Pb-210, Cs-137, T. Hg, T. Pb, TOC	1	41-8566	1	Brown silt with organic material	No archive samples
2	Pb-210, T. Hg, T. Pb, TOC	2	41-8567	1	Brown silt with organic material	No archive samples
3	Archive	–	–	1	Brown silt with organic material	–
4	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	41-8568	1	Brown silt with organic material	No archive samples
5	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3		1	Brown silt with organic material	No archive samples
6	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	41-8569	1	Brown silt with organic material	No archive samples
7	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4		1	Brown silt with organic material	No archive samples

Table D1. Sediment core logs for lakes in Whatcom County, Washington—*Continued*

[**Analysis**: Pb-210, lead-210; Cs-137, cesium-137; T. Hg, total mercury; T. Pb, total lead; TOC, total organic carbon. **Analytical sample No.**: Sample in core sent out for analysis. Up to two sections may have been combined to form an analytical sample. cm, centimeter. –, no data]

Section	Analysis	Analytical sample No.	Ecology sample No.	Length (cm)	Description	Notes
8	Pb-210, T. Hg, T. Pb, TOC	5	41-8570	1	Brown silt with organic material	No archive samples
9	Pb-210, T. Hg, T. Pb, TOC	5	–	1	Brown silt with organic material – limb vertical (9-17)	No archive samples
10	Pb-210, T. Hg, T. Pb, TOC	6	41-8571	1	Brown silt with organic material – limb vertical (9-17)	No archive samples
11	Pb-210, T. Hg, T. Pb, TOC	6	–	1	Brown silt with organic material – limb vertical (9-17)	No archive samples
12	Archive	–	–	1	Brown silt with organic material- limb vertical (9-17)	–
13	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	41-8572	1	Brown silt with organic material – limb vertical (9-17)	No archive samples
14	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Brown silt with organic material – limb vertical (9-17)	No archive samples
15	Archive	–	–	1	Brown silt with organic material – limb vertical (9-17)	–
16	Pb-210, T. Hg, T. Pb, TOC	8	41-8573	1	Wood fibers and plant material in silt –limb vertical	No archive samples
17	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Wood fibers and plant material in silt –limb vertical	–
18	Archive	–	–	1	Wood fibers and plant material in silt	–
19	Pb-210, Cs-137, T. Hg, T. Pb, TOC	9	41-8574	1	Wood fibers and plant material in silt	No archive samples
20	Pb-210, Cs-137, T. Hg, T. Pb, TOC	9	–	2	Wood fibers and plant material in silt	–
21	Archive	–	–	2	Wood fibers and plant material in silt	–
22	Pb-210, T. Hg, T. Pb, TOC	10	41-8575	2	Compacted silt and clay	No archive samples
Wiser Lake **ID: Wis-1** **Date collected: 9/24/02 5:50 PM** **Latitude/Longitude: 48º 54.207 N 122º 28.79 W** **Water depth: 0.7 meter**						
1	Pb-210, Cs-137, T. Hg, T. Pb, TOC	1	41-8544	1	Black to brown silt with some organic material – slightly disturbed layer	No archive sample
2	Pb-210, T. Hg, T. Pb, TOC	2	41-8545	1	Black to brown silt with some organic material	No archive sample
3	Archive	–	–	1	Black to brown silt with some organic material	–
4	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	41-8546	1	Black to brown silt with some organic material	No archive sample
5	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	–	1	Black to brown silt with some organic material	–
6	Archive	–	–	1	Black to brown silt with some organic material	–
7	Archive	–	–	1	Black to brown silt with some organic material	–
8	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	41-8547	1	Black to brown silt with some organic material	No archive sample
9	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	–	1	Black to brown silt with some organic material	–
10	Archive	–	–	1	Black to brown silt with some organic material	–
11	Archive	–	–	1	Black to brown silt with some organic material	–
12	Pb-210, T. Hg, T. Pb, TOC	5	41-8548	1	Black to brown silt with some organic material	No archive sample
13	Pb-210, T. Hg, T. Pb, TOC	5	–	1	Black to brown silt with some organic material	–
14	Archive	–	–	1	Woody debris	–
15	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	41-8549	1	Woody debris	No archive sample

Table D1. Sediment core logs for lakes in Whatcom County, Washington—*Continued*

[**Analysis**: Pb-210, lead-210; Cs-137, cesium-137; T. Hg, total mercury; T. Pb, total lead; TOC, total organic carbon. **Analytical sample No.**: Sample in core sent out for analysis. Up to two sections may have been combined to form an analytical sample. cm, centimeter. –, no data]

Section	Analysis	Analytical sample No.	Ecology sample No.	Length (cm)	Description	Notes
16	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	–	1	Woody debris	–
17	Archive	–	–	1	Black to brown silt with some organic material	–
18	Archive	–	–	1	Black to brown silt with some organic material	–
19	Archive	–	–	1	Black to brown silt with some organic material	–
20	Archive	–	–	1	Black to brown silt with some organic material	–
21	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	41-8550	1	Black to brown silt with some organic material	–
22	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Black to brown silt with some organic material	–
23	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Black to brown silt with some organic material	–
24	Archive	–	–	1	Black to brown silt with some organic material	–
25	Archive	–	–	1	Black to brown silt with some organic material	–
26	Archive	–	–	1	Black to brown silt with some organic material	–
27	Pb-210, T. Hg, T. Pb, TOC	8	41-8551	1	Black to brown silt with some organic material	–
28	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Black to brown silt with some organic material	–
29	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Black to brown silt with some organic material	–
30	Archive	–	–	1	Contact change brown silt layer	–
31	Archive	–	–	1	Light brown silt layer	–
32	Archive	–	–	1	Light brown silt layer	–
33	Pb-210, T. Hg, T. Pb, TOC	9	41-8552	1	Light brown silt layer	–
34	Pb-210, T. Hg, T. Pb, TOC	9	–	1	Light brown silt layer	–
35	Pb-210, T. Hg, T. Pb, TOC	9	–	1	Light brown silt layer	–
36	Archive	–	–	1	Light brown silt layer	–
37	Archive	–	–	1	Light brown silt layer	–
38	Archive	–	–	1	Light brown silt layer	–
39	Archive	–	–	1	Light brown silt layer	–
40	Archive	–	–	1	Light brown silt layer	–
41	Pb-210, T. Hg, T. Pb, TOC	10	41-8553	2	Light brown silt layer	–
42	Pb-210, T. Hg, T. Pb, TOC	10	–	2	Light brown silt layer	–
Fazon Lake **ID: Faz-1** **Date collected: 09/24/02 1:40 PM** **Latitude/Longitude: 48º 51.908 N 122º 22.039 W** **Depth of water: 1.5 meters**						
1	Pb-210, Cs-137, T. Hg, T. Pb, TOC	1	41-8555	1	Uniform fine brown silt	No archive sample
2	Pb-210, T. Hg, T. Pb, TOC	2	41-8556	1	Uniform fine brown silt	No archive sample
3	Archive	–		1	Uniform fine brown silt	–
4	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	41-8557	1	Uniform fine brown silt	No archive sample
5	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	–	1	Uniform fine brown silt	No archive sample
6	Archive	–	–	1	Uniform fine brown silt	–
7	Archive	–	–	1	Uniform fine brown silt	–
8	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	41-8558	1	Uniform fine brown silt	No archive sample
9	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	–	1	Uniform fine brown silt	No archive sample
10	Archive	–	–	1	Uniform fine brown silt	–
11	Archive	–	–	1	Uniform fine brown silt	–
12	Pb-210, T. Hg, T. Pb, TOC	5	41-8559	1	Uniform fine brown silt	No archive sample
13	Pb-210, T. Hg, T. Pb, TOC	5	–	1	Uniform fine brown silt	No archive sample
14	Archive	–	–	1	Uniform fine brown silt	–

Table D1. Sediment core logs for lakes in Whatcom County, Washington—*Continued*

[**Analysis:** Pb-210, lead-210; Cs-137, cesium-137; T. Hg, total mercury; T. Pb, total lead; TOC, total organic carbon. **Analytical sample No.:** Sample in core sent out for analysis. Up to two sections may have been combined to form an analytical sample. cm, centimeter. –, no data]

Section	Analysis	Analytical sample No.	Ecology sample No.	Length (cm)	Description	Notes
15	Archive	–	–	1	Uniform fine brown silt	–
16	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	41-8560	1	Uniform fine brown silt	–
17	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	–	1	Uniform fine brown silt	–
18	Pb-210, Cs-137, T. Hg, T. Pb, TOC	6	–	1	Uniform fine brown silt	–
19	Archive	–	–	1	Uniform fine brown silt	–
20	Archive	–	–	1	Uniform fine brown silt	–
21	Archive	–	–	1	Uniform fine brown silt	–
22	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	41-8561	1	Uniform fine brown silt	–
23	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Uniform fine brown silt	–
24	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Uniform fine brown silt	–
25	Archive	–	–	1	Uniform fine brown silt	–
26	Archive	–	–	1	Uniform fine brown silt	–
27	Archive	–	–	1	Uniform fine brown silt	–
28	Pb-210, T. Hg, T. Pb, TOC	8	41-8562	1	Uniform fine brown silt	–
29	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Uniform fine brown silt	–
30	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Uniform fine brown silt	–
31	Archive	–	–	1	Uniform fine brown silt	–
32	Archive	–	–	1	Uniform fine brown silt	–
33	Archive	–	–	1	Uniform fine brown silt	–
34	Pb-210, T. Hg, T. Pb, TOC	9	41-8563	1	Uniform fine brown silt	–
35	Pb-210, T. Hg, T. Pb, TOC	9	–	1	Uniform fine brown silt	–
36	Pb-210, T. Hg, T. Pb, TOC	9	–	1	Uniform fine brown silt	–
37	Archive	–	–	1	Uniform fine brown silt	–
38	Archive	–	–	1	Uniform fine brown silt	–
39	Archive	–	–	1	Uniform fine brown silt	–
40	Archive	–	–	1	Uniform fine brown silt	–
41	Pb-210, T. Hg, T. Pb, TOC	10	41-8564	2	Uniform fine brown silt	–
42	Pb-210, T. Hg, T. Pb, TOC	10	–	2	Uniform fine brown silt	–
Baker Lake **ID: Bak-1** **Date collected: 9/26/2002 11:39: AM** **Latitude/Longitude: 48 º 43.700 N 122º 37.082 W** **Water depth: 15.3 meters**						
1	Pb-210, Cs-137, T. Hg, T. Pb, TOC	1	41-8534	1	Light brown fine silt and clay	No archive sample
2	Pb-210, T. Hg, T. Pb, TOC	2	41-8535	1	Light brown fine silt and clay	No archive sample
3	Archive	–	–	1	Beginning of reducing layer (black streaks)	–
4	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	41-8536	1	Light brown fine silt and clay	No archive sample
5	Pb-210, Cs-137, T. Hg, T. Pb, TOC	3	–	1	Light brown fine silt and clay	–
6	Archive	–	–	1	Light brown fine silt and clay	–
7	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	41-8537	1	Light brown fine silt and clay	–
8	Pb-210, Cs-137, T. Hg, T. Pb, TOC	4	–	1	Light brown fine silt and clay	–
9	Archive	–	–	1	Light brown fine silt and clay	–
10	Pb-210, Cs-137, T. Hg, T. Pb, TOC	5	41-8538	1	Light brown fine silt and clay	–
11	Pb-210, Cs-137, T. Hg, T. Pb, TOC	5	–	1	Light brown fine silt and clay	–

Table D1. Sediment core logs for lakes in Whatcom County, Washington—*Continued*

[**Analysis:** Pb-210, lead-210; Cs-137, cesium-137; T. Hg, total mercury; T. Pb, total lead; TOC, total organic carbon. **Analytical sample No.**: Sample in core sent out for analysis. Up to two sections may have been combined to form an analytical sample. cm, centimeter. –, no data]

Section	Analysis	Analytical sample No.	Ecology sample No.	Length (cm)	Description	Notes
12	Archive	–	–	1	Light brown fine silt and clay	–
13	Pb-210, T. Hg, T. Pb, TOC	6	41-8539	1	Light brown fine silt and clay	–
14	Pb-210, T. Hg, T. Pb, TOC	6	–	1	Light brown fine silt and clay	–
15	Archive	–	–	1	Light brown fine silt and clay	–
16	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	41-8540	1	Wood debris begins	–
17	Pb-210, Cs-137, T. Hg, T. Pb, TOC	7	–	1	Light brown fine silt and clay	–
18	Archive	–	–	1	Light brown fine silt and clay	–
19	Pb-210, T. Hg, T. Pb, TOC	8	41-8541	1	Light brown fine silt and clay	–
20	Pb-210, T. Hg, T. Pb, TOC	8	–	1	Light brown fine silt and clay	–
21	Archive	–	–	1	Light brown fine silt and clay	–
22	Pb-210, Cs-137, T. Hg, T. Pb, TOC	9	41-8542	1	Light brown fine silt and clay	–
23	Pb-210, Cs-137, T. Hg, T. Pb, TOC	9	–	1	Light brown fine silt and clay	–
24	Archive	–	–	2	Light brown fine silt and clay	–
25	Pb-210, T. Hg, T. Pb, TOC	10	41-8543	2	Light brown fine silt and clay	–

Table D2. Summary of analysis of sediment cores from lakes in Whatcom County, Washington, September 2002

[<, not detected at detection limit shown (detection limit varied because of the mass of sediment used in the analyses); J, estimated value because holding time elapsed; J2, disregarded—used 19 percent for 2-3 cm section and 25 percent for 5-6 cm section. mg/kg, dw, milligrams per kilograms, dry weight; piC/g, picocurie per gram. – no data]

Lake	Collection date	Ecology sample No.	Interval (cm)	Percentage of solids	Total organic carbon (percentage by weight)		Total mercury (mg/kg, dw)	Total lead (mg/kg, dw)	Lead-210 (piC/g)	Cesium-137 (piC/g)
Lake Samish	09/23/02	02408577	0-1	14.1	5.7		0.13	36.8	10.4	2.44
		02408578	1-2	51.3J2	5.5		.19	42.4	1.6	–
		02408579	3-5	7.9J2	4.9		.166	45.6	5.42	1.45
		02408580	6-8	29.2	4.7		.178	62.8	5.25	1.91
		02408581	9-11	29.7	5.1		.142	36.5	<1.32	–
		02408582	12-14	22.7	6.3		.112	35.4	<.95	<.18
		02408583	15-17	21.3	6.6		.161	32.5	<2.56	–
		02408584	18-20	19.3	7.1		.12	17.8	<.93	<-.29
		02408585	22-24	17.9	7.1		.11	8.06	<3.65	–
		02408586	27-31	19.9	6.6		.088	6.1	<.89	–
Lake Terrell	09/24/02	02418566	0-1	8.8	19.4		.16J	89.8	<2.67	<3.37
		02418567	1-2	8.6	2.6		.15J	57.9	7.74	–
		02418568	3-5	1.2	2.4		.2J	9.5	8.25	2.97
		02418569	5-7	8.2	21.0		.21J	89.6	5.33	3.23
		02418570	7-9	11.2	25.1		.21J	91.4	5.92	–
		02418571	9-11	1.5	25.0		.2J	86	4.96	–
		02418572	12-14	9.1	33.0		.22J	74	4.42	1.45
		02418573	15-17	12.8	24.8		.14J	3.3	<1.49	–
		02418574	18-21	2.2	15.9		.095J	17.4	<.96	<.15
		02418575	23-25	17.7	17.6		.079J	6.63	2.73	–
Wiser Lake	09/24/02	02418544	0-1	11.1	18.0		.23J	54.5	5.12	<.66
		02418545	1-2	1.0	18.2	J	.16J	48.1	5.53	–
		02418546	3-5	1.0	18.5		.403J	49.8	<3.15	<.17
		02418547	7-9	1.6	18.0		.13J	5.3	6.48	<.99
		02418548	11-13	9.3	2.0		.16J	67.6	<4.57	–
		02418549	14-16	8.9	18.9		.372J	66	2.83	<.73
		02418550	20-23	11.0	2.9		.14J	26	<.62	<.78
		02418551	26-29	1.1	21.6		.33J	16.4	<1.39	–
		02418552	32-35	1.8	23.6		.367	8.71	<1.26	–
		02418553	40-44	11.8	21.6		.23J	2.55	<.43	–
Fazon Lake	09/24/02	02418555	0-1	7.3	25.5		.18J	48.6	5.02	<2.56
		02418556	1-2	7.2	27.0		.27J	38.3	5.8	–
		02418557	3-5	6.9	27.3		.26J	43.4	3.99	<1.99
		02418558	7-9	8.1	24.3		.26J	54.8	8.48	2.39
		02418559	11-13	7.9	26.8		.23J	88.2	5.97	–
		02418560	15-18	9.0	25.0		.24J	56.7	4.99	3.33
		02418561	21-24	9.6	24.1		.26J	58.7	5.34	3.11
		02418562	27-30	11.1	22.2		.22J	47.1	3.37	–
		02418563	33-36	9.3	26.1		.21J	29.2	2.22	–
		02418564	40-44	7.9	27.7		.14J	13.3	3.44	–
Baker Lake	09/26/02	02418534	0-1	3.2	2.5		.056	37.4	3.11	<.73
		02418535	1-2	39.2	1.8		.057	31.1	4.33	–
		02418536	3-5	38.6	2.3		.065	37	6.29	.97
		02418537	6-8	43.3	2.1		.0728	37.5	4.2	1.47
		02418538	9-11	45.1	2.0		.0762	43.2	6.05	2.29
		02418539	12-14	48.5	1.5		.0636	31.7	4.75	–
		02418540	15-17	43.7	2.4	J	.166	32	2.56	2.81
		02418541	18-20	39.8	2.8		.105	39.7	<5.17	–
		02418542	21-23	46.9	2.0		.121J	3.5		5.03
		02418543	25-27	47.5	.2		.1 J	27.3	<2.08	–

Table D3. Estimates of mass sedimentation rates based on anayses of sediment cores from lakes in Whatcom County, Washington

[All values are in grams per square centimeter per year. Values in **bold** are assigned a high weighting factor; Values in *italic* are assigned a moderage weighting factor; Values underlined are assigned a low weighting factor]

Lake		Lead-210 slope	Lead peak			Cesium-137		Weight of evidence
			Peak	Appearance	Matching	Peak	Appearance	
Lake Whatcom								
Basin 1	avg.	0.046	**0.034**	–	0.045	*0.050*	–	**0.045**
	min	–	**.018**	**.035**	–	*.032*	0.045	–
	max	–	**.074**	**.068**	–	*.072*	.062	–
Basin 2	avg.	.105	–	–	.040	**.023**	–	**.040**
	min	–	–	*.039*	–	**.016**	*.047*	–
	max	–	*.059*	*.063*	–	**.051**	*.079*	–
Basin 3	avg.	.034	–	–	.052	–	–	.040
	min	–	–	–	–	–	–	–
	max	.080	.074	–	–	–	–	–
Lake Samish	avg.	–	*.072*	–	–	–	–	.072
	min	–	*.046*	.056	–	–	.046	–
	max	–	*.085*	.082	–	–	.067	–
LakeTerrell	avg.	.095	.029	–	.025	**.029**	–	*.031*
	min	–	–	.038	–	–	*.031*	–
	max	–	*.046*	.054	–	**.047**	*.048*	–
Wiser Lake	avg.	–	**.049**	–	.040	–	–	.049
	min	–	**.041**	*.049*	–	–	–	–
	max	–	**.079**	*.074*	–	–	–	–
Fazon Lake	avg.	.044	**.035**	–	.039	*.039*	–	.039
	min	–	**.029**	.051	–	*.022*	–	–
	max	–	**.045**	–	–	–	–	–
Baker Lake	avg.	–	–	–	–	–	–	–
	min	–	–	–	–	.370	–	.370
	max	–	–	–	–	–	–	–

Table D4. Estimated rates of sedimentation in Lake Whatcom, Washington, after adjusting for focusing factor obtained from lead-210 inventories of sediment cores

[$(g/cm^2)/yr$, grams per cubic centimeter per year; cm, centimeters; $(pCi/cm^2)/yr$, picocuries per cubic centimeter per year; Mg/yr, megagrams per year]

Lake Whatcom	Core sedimentation rate [$(g/cm^2)/yr$]	Lead-210 inventory (pCi/cm^2)	Depth of inventory (cm)	Lead-210 steady state flux [$(pCi/cm^2)/yr$]	Estimated focusing factor	Estimated average basin sedimentation rate [$(g/cm^2)yr$]	Estimated basin sedimentation (Mg/yr)
Basin 1	0.045	52.6	10	1.63	10.6	0.0042	89
Basin 2	.040	58.3	13	1.81	11.7	.0034	55
Basin 3	.040	30.0	11	.93	6.04	.0066	1,099
Total							1,243

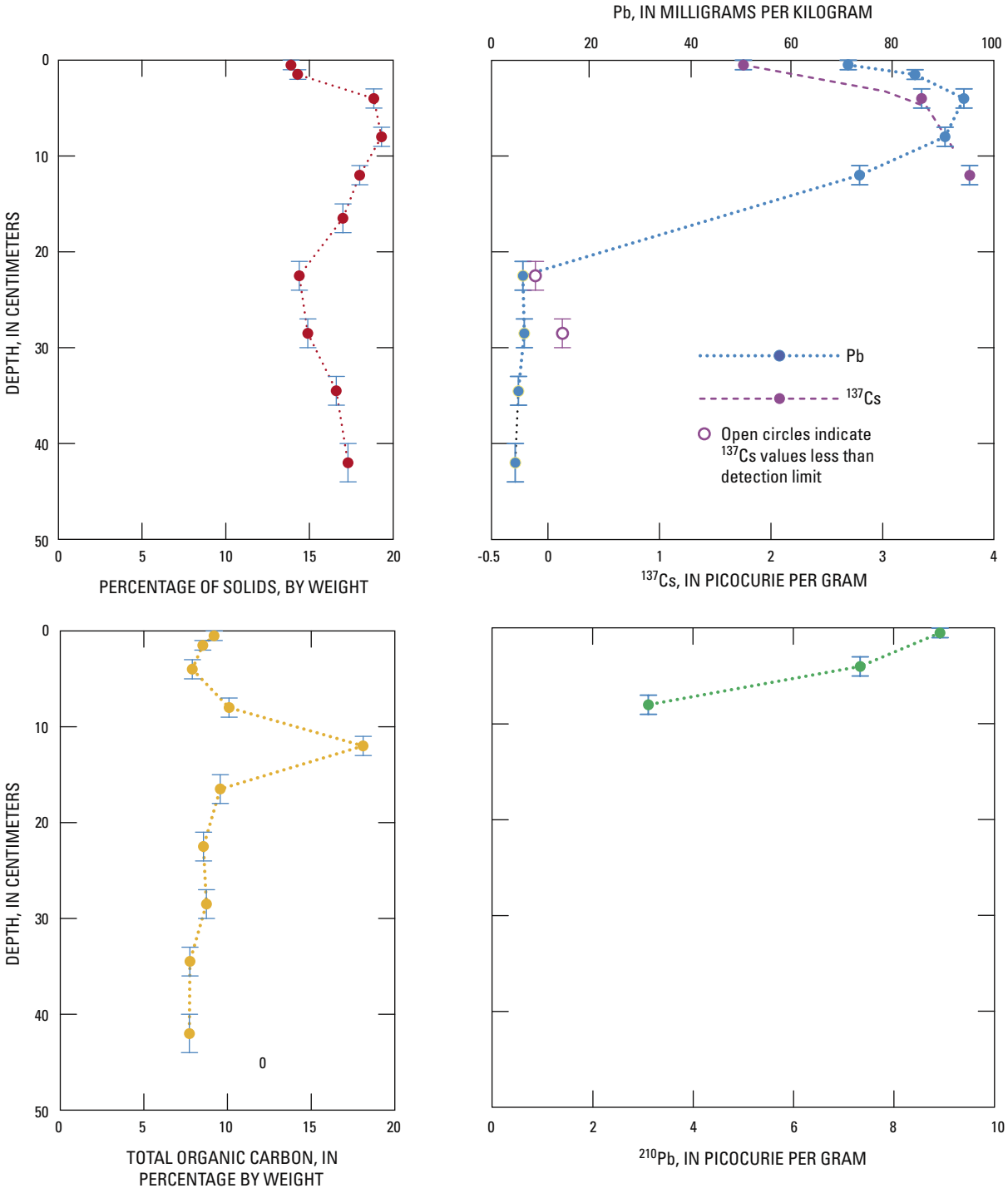


Figure D1. Characteristics of sediment core from Basin 1 of Lake Whatcom, Washington.

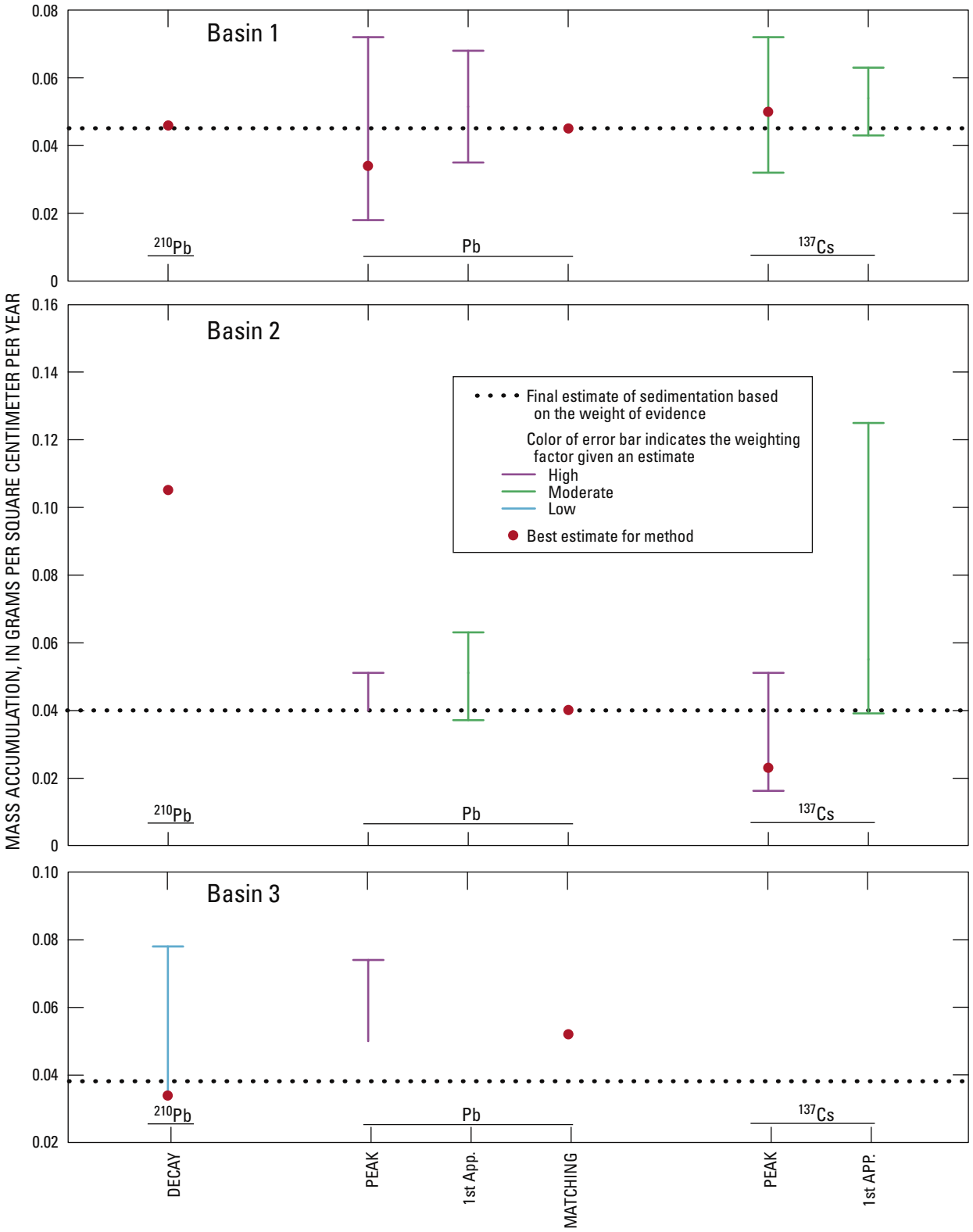


Figure D2. Summary of sedimentation rates estimated from analyses of sediment cores from Lake Whatcom, Washington. The upper error estimates for sedimentation rates calculated from ^{210}Pb core from basin 3 was calculated by including the detectable ^{210}Pb concentration from the 10-12 cm section. The minimum thickness of the core section in which the peak or first appearance of elevated stable Pb or ^{137}Cs could have occurred determined the range of sedimentation rates (error bars).

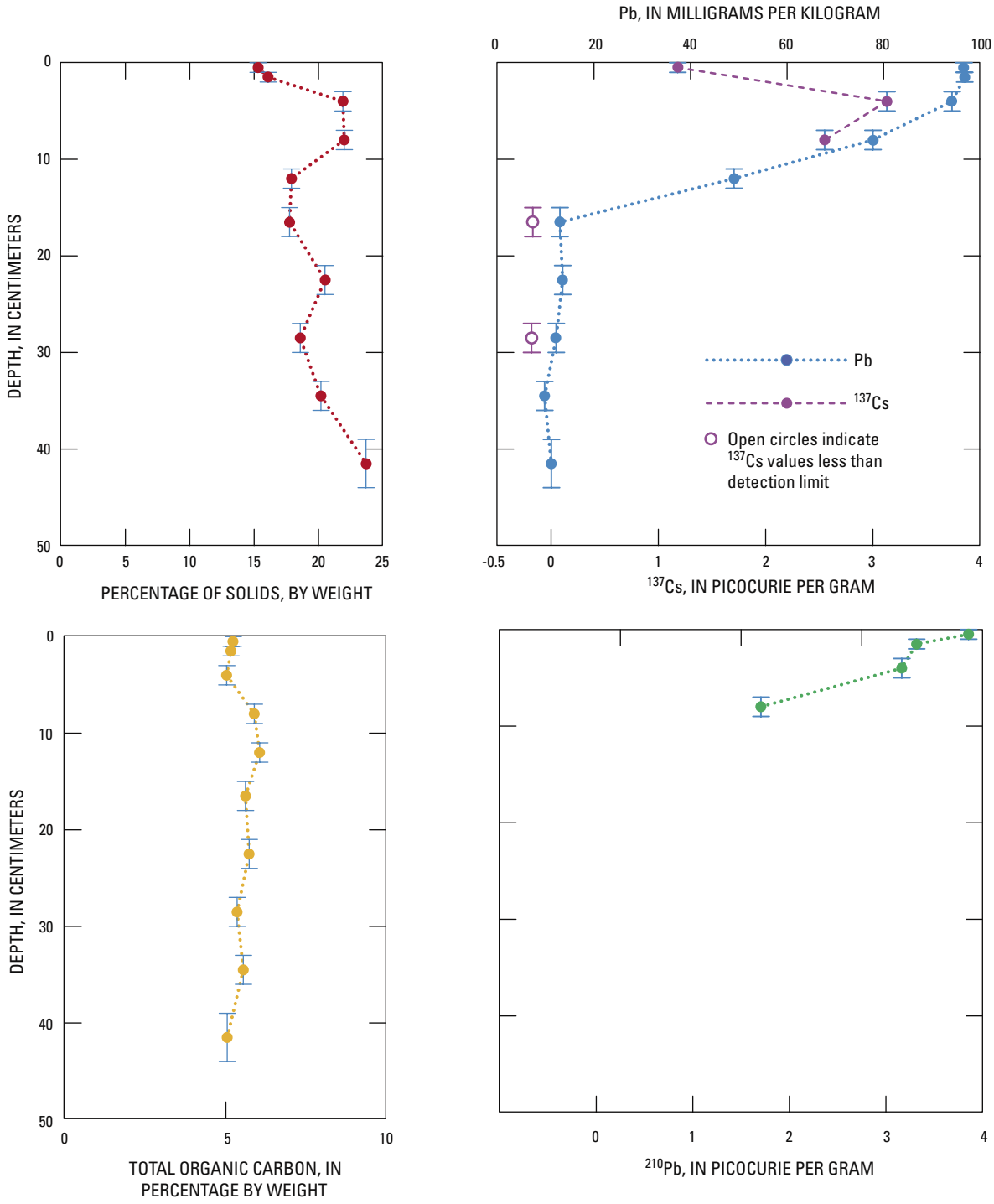


Figure D3. Characteristics of sediment core from Basin 2 of Lake Whatcom, Washington.

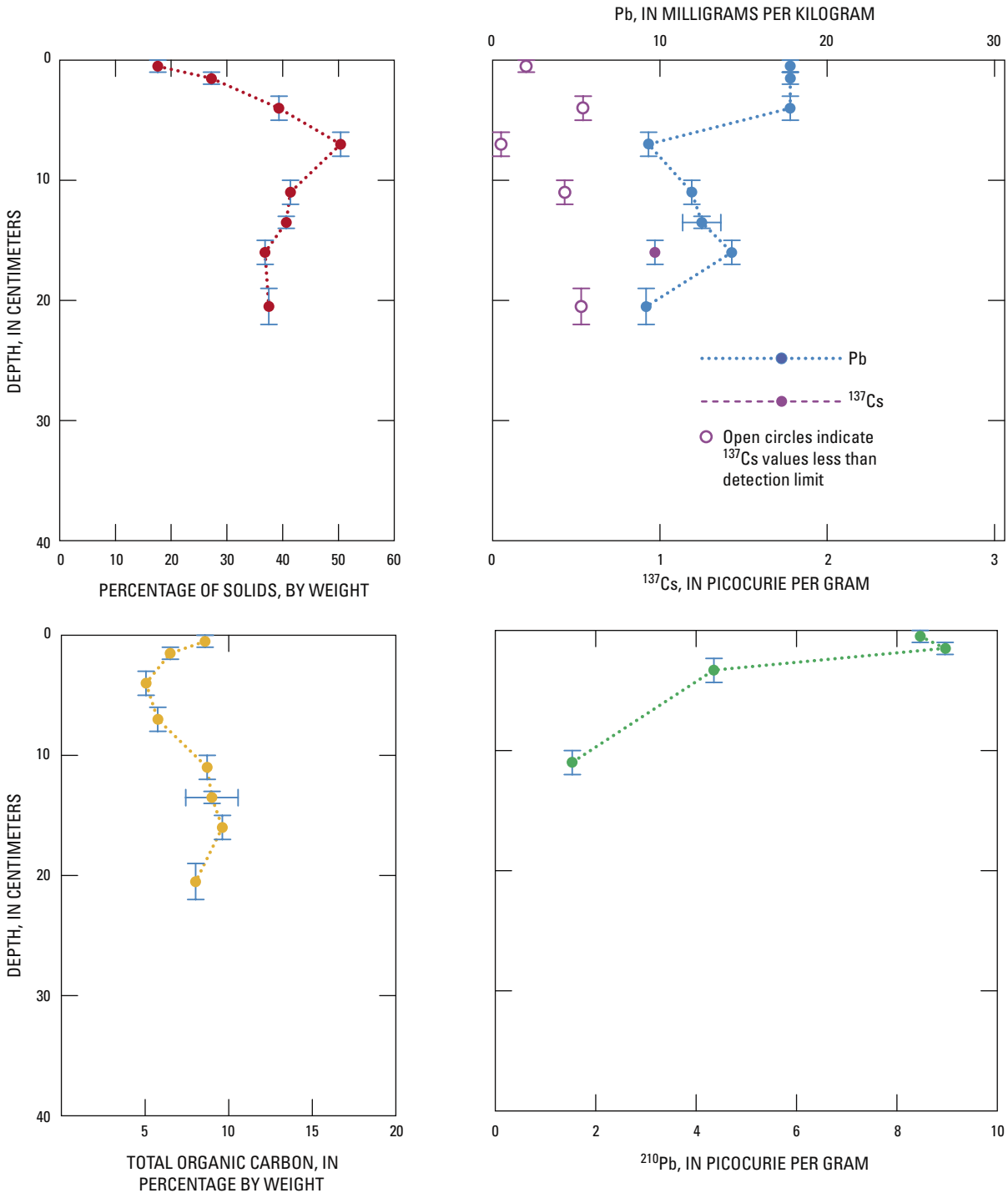


Figure D4. Characteristics of sediment core from Basin 3 of Lake Whatcom, Washington.

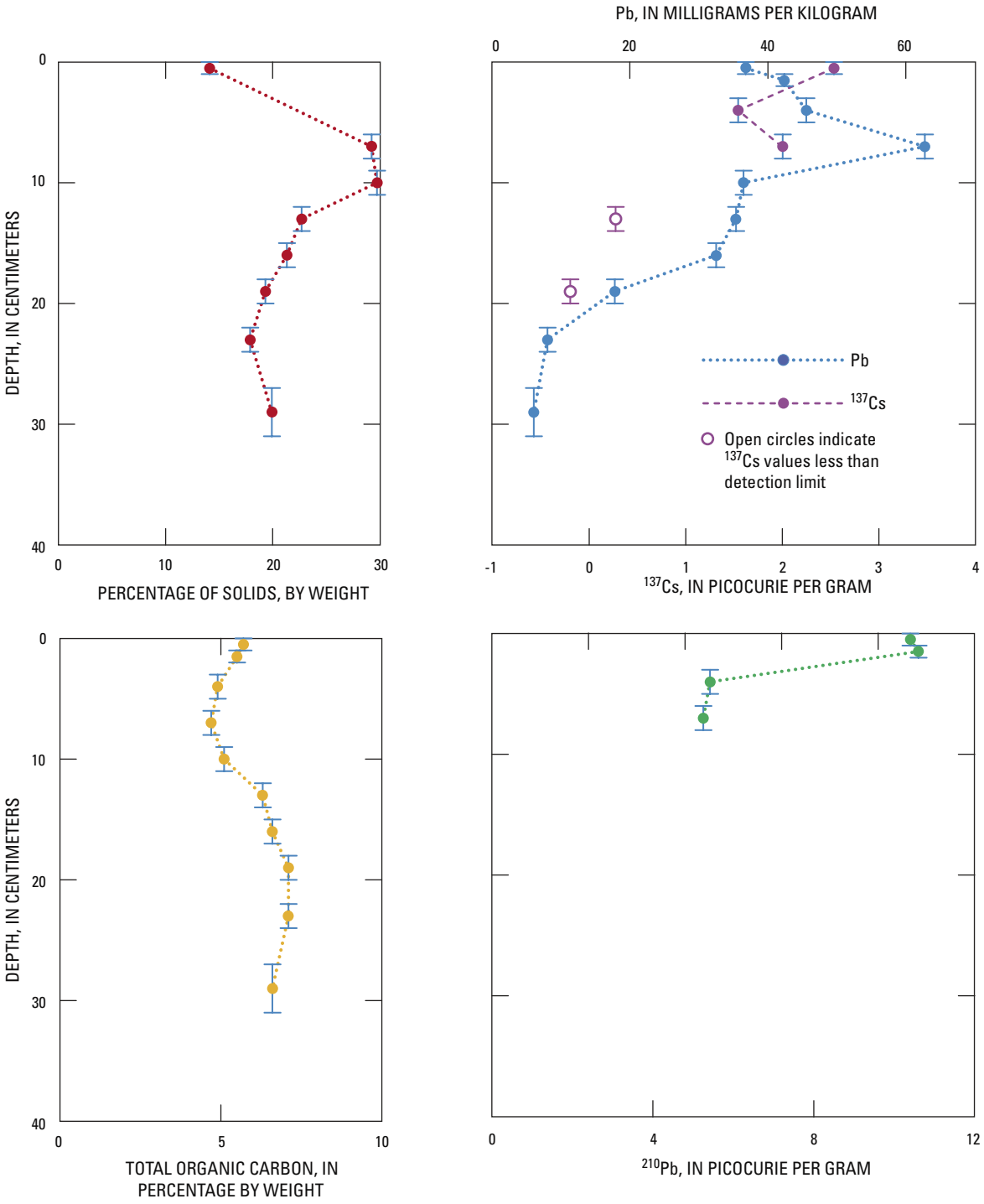


Figure D5. Characteristics of sediment core from Samish Lake, Washington.

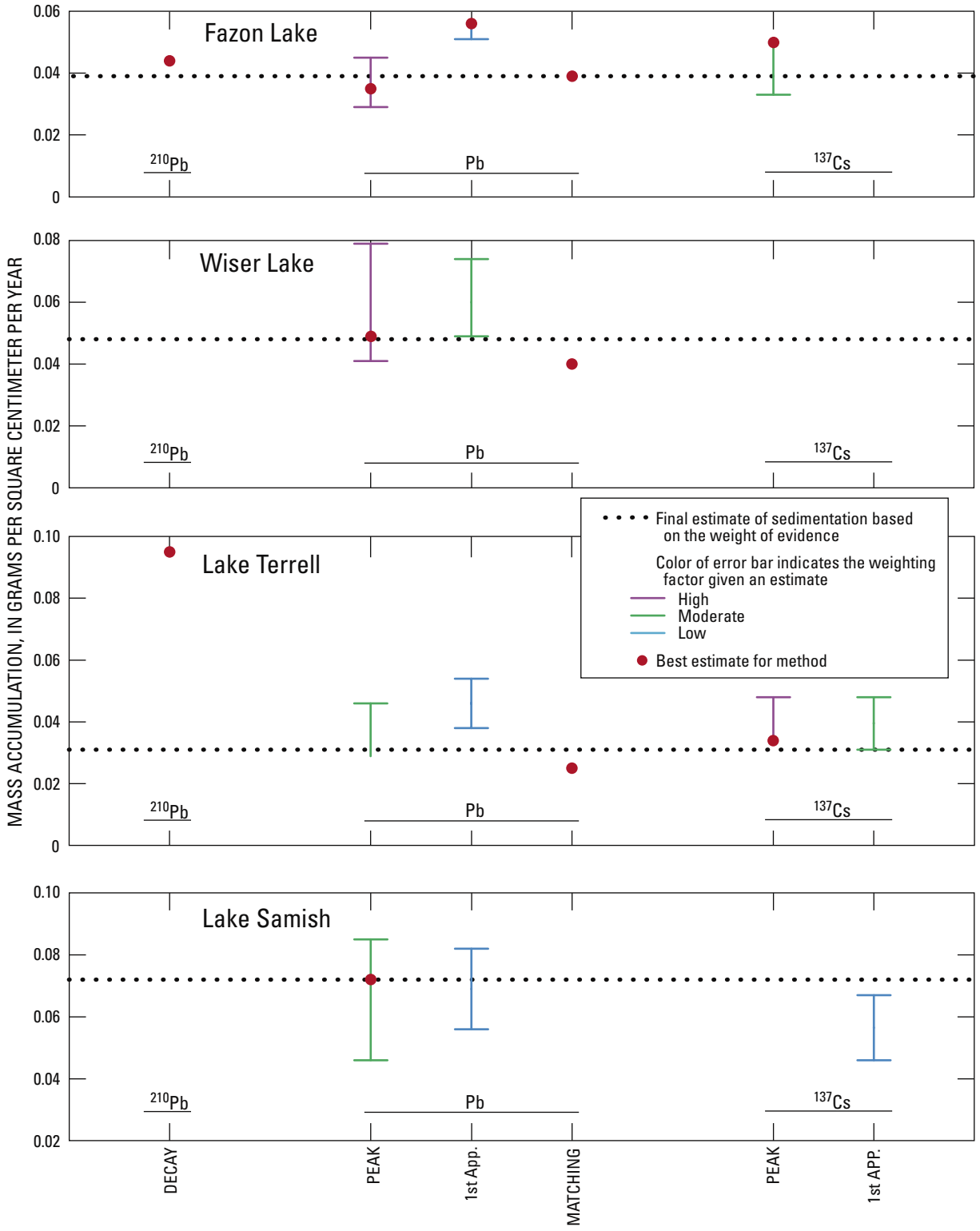


Figure D6. Summary of sedimentation rates estimated from analysis of sediment cores from Lake Samish, Lake Terrell, Wiser Lake and Fazon Lake. The minimum thickness of the core section in which the peak or first appearance of ^{137}Cs and elevated stable Pb could have occurred determined the range of sedimentation rates (error bars).

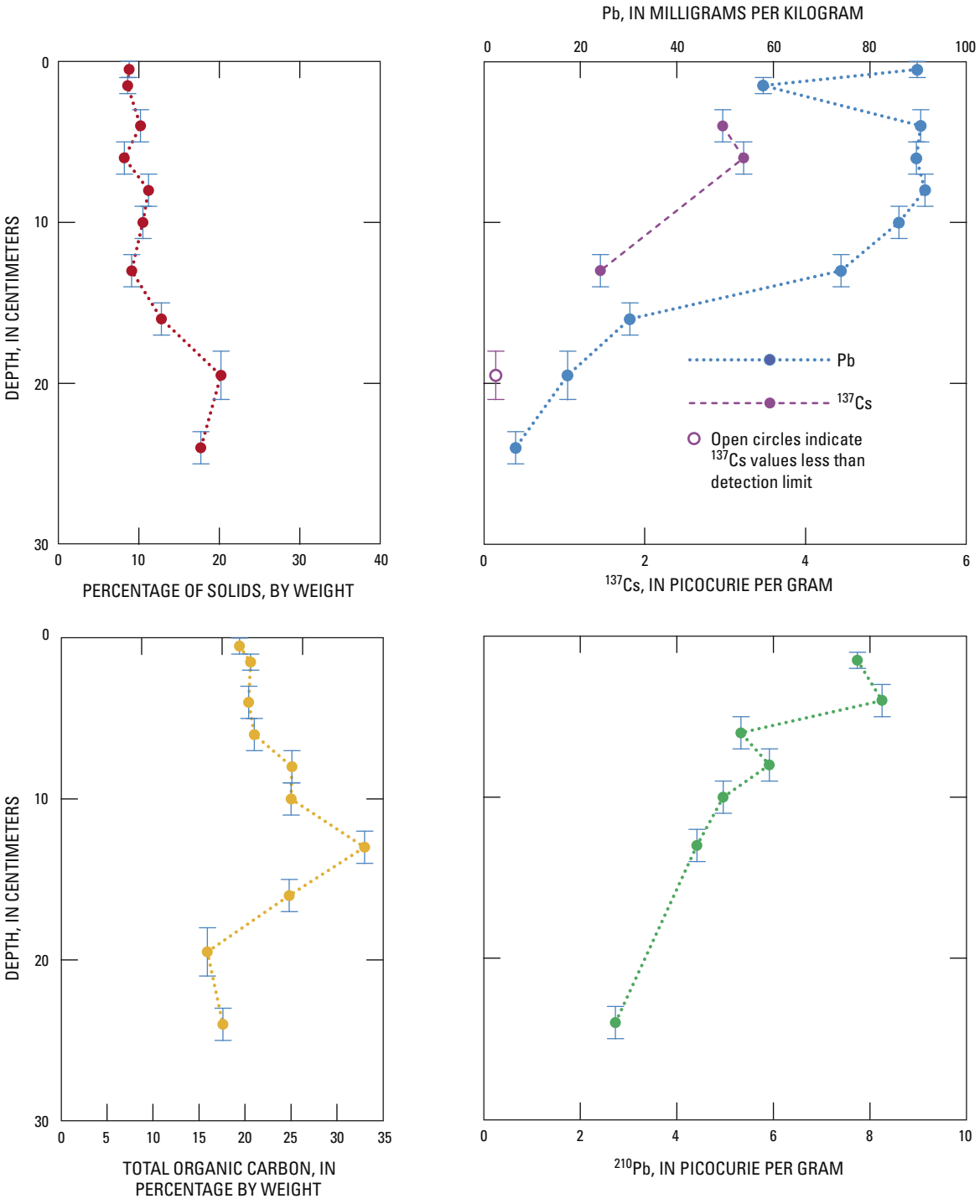


Figure D7. Characteristics of sediment core from Terrell Lake, Washington.

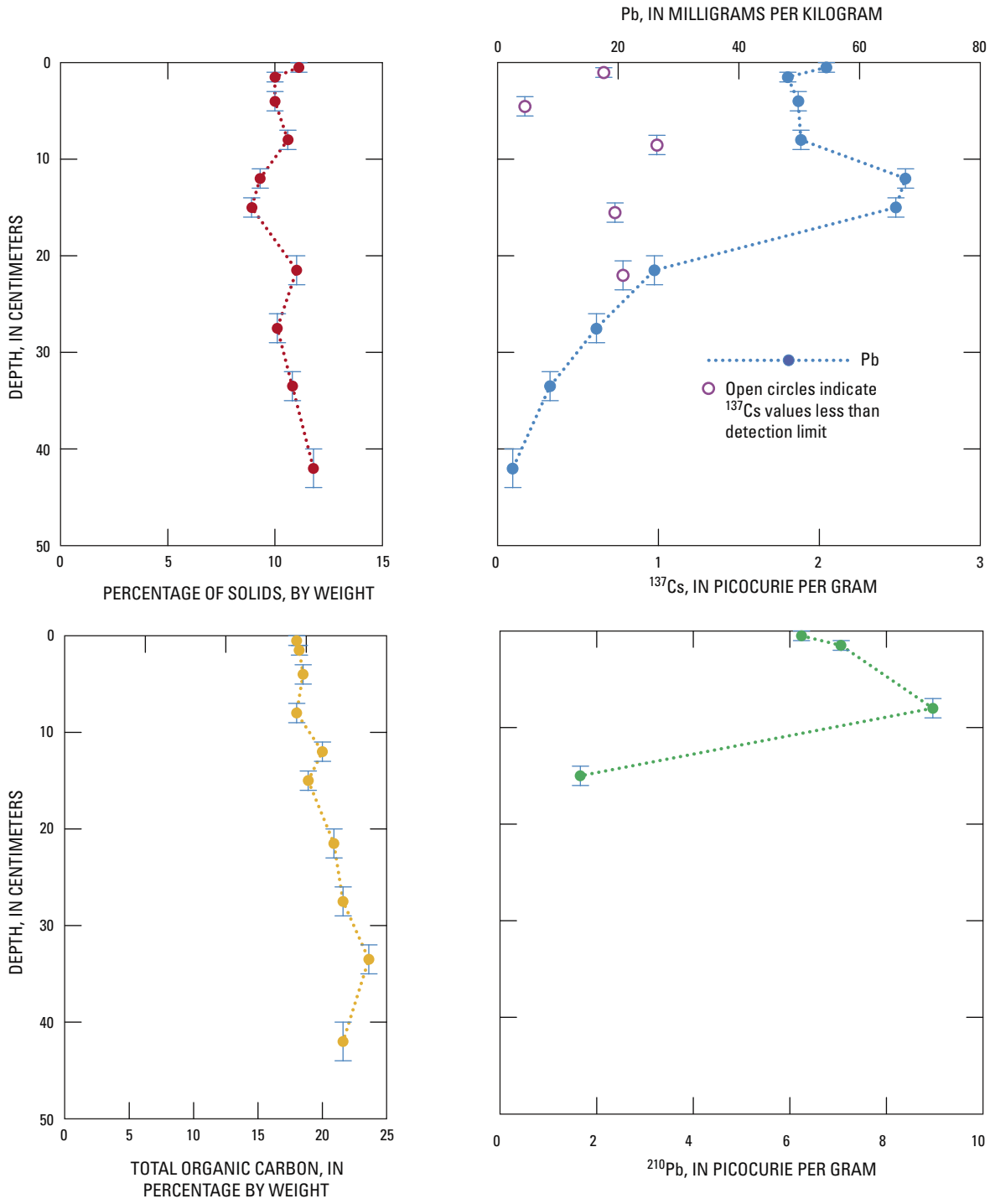


Figure D8. Characteristics of sediment core from Wiser Lake, Washington.

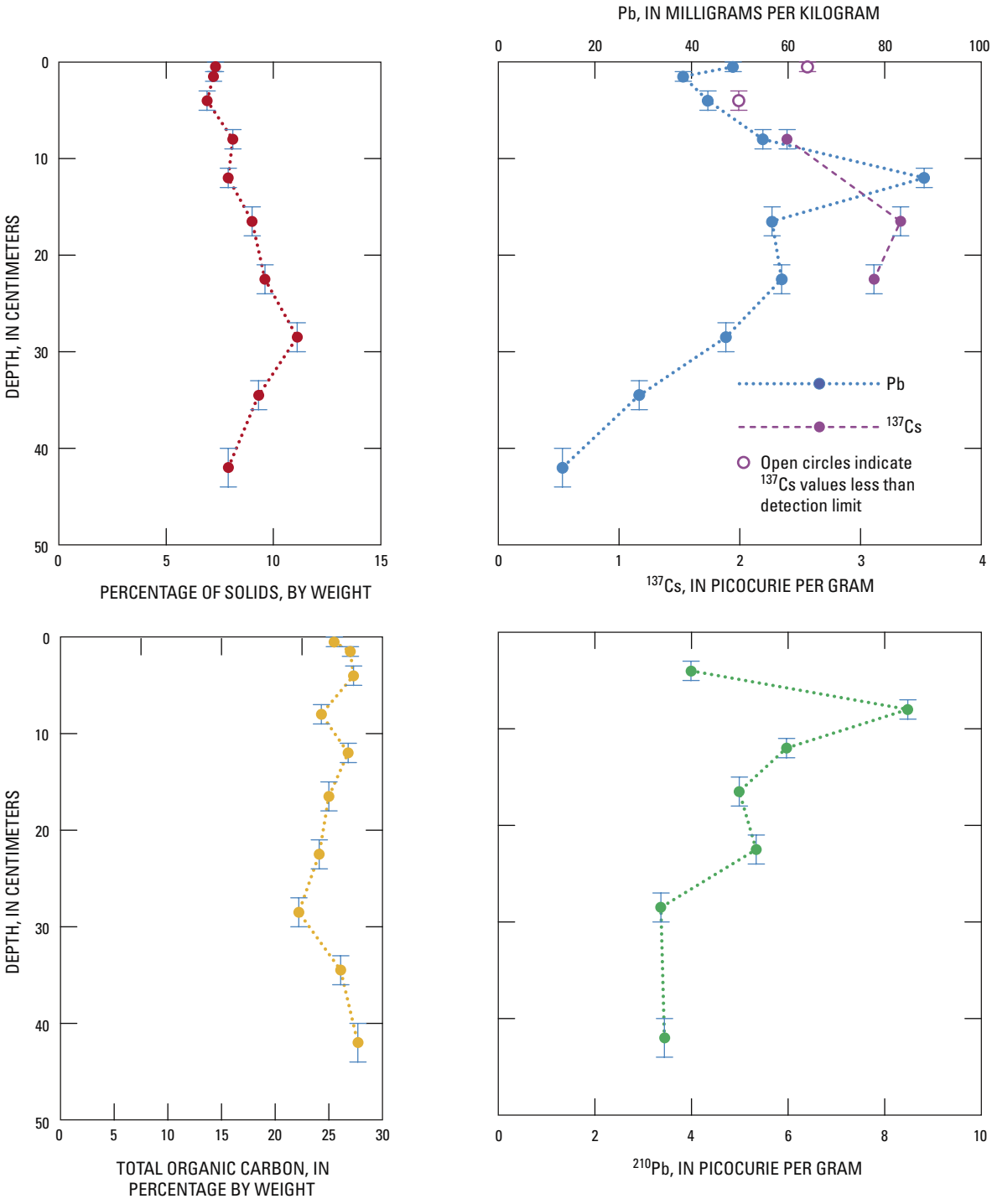


Figure D9. Characteristics of sediment core from Fazon Lake, Washington.

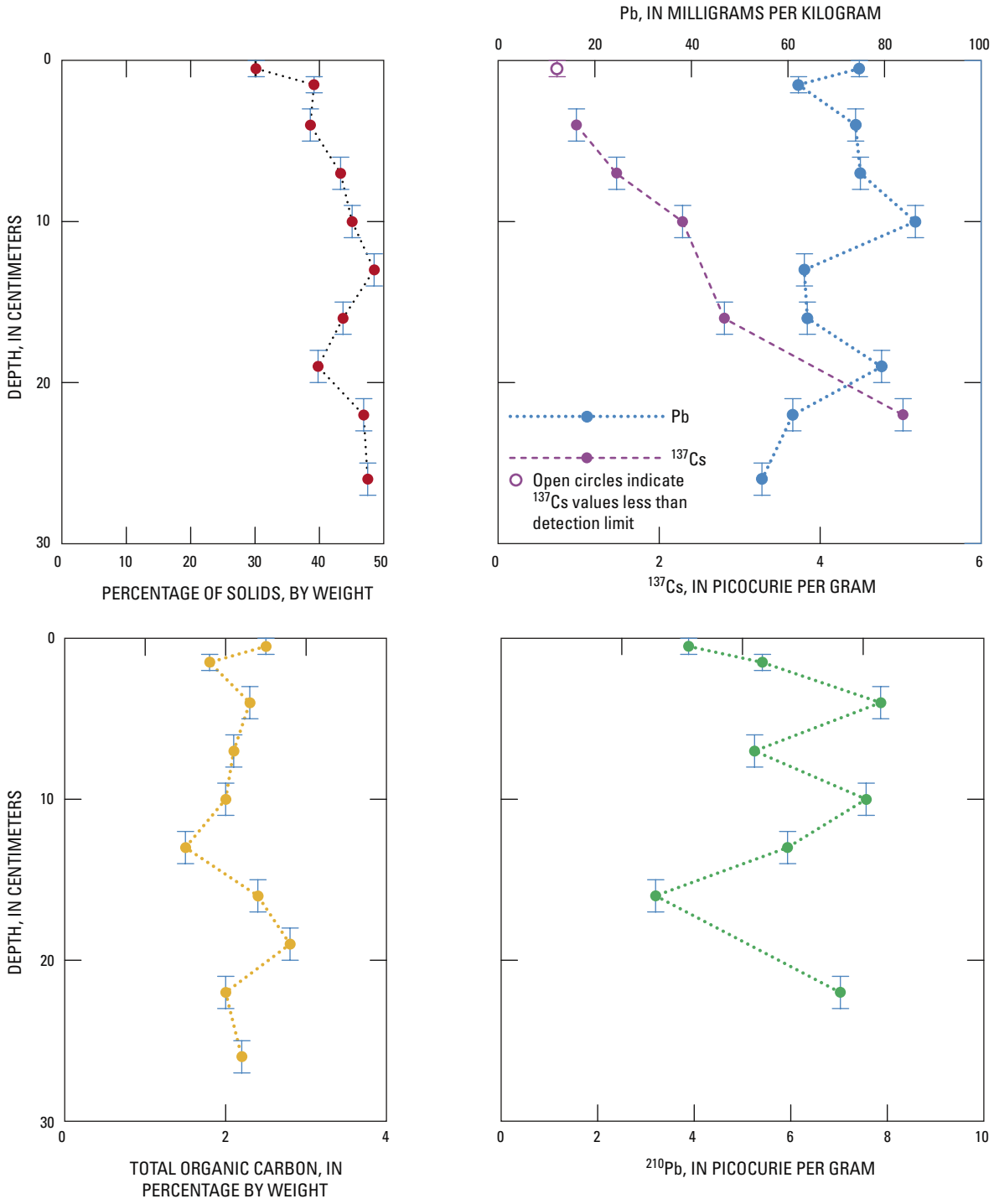


Figure D10. Characteristics of sediment core from Baker Lake, Washington.

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