CALIFORNIA ENVIRONMENTAL PROTECTION AGENCY

REGIONAL WATER QUALITY CONTROL BOARD
CENTRAL VALLEY REGION

Clear Lake TMDL for Mercury

Staff Report

Final Report

February 2002

State of California

California Environmental Protection Agency

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3443 Routier Road, Suite A
Sacramento, California 95827-3003

Phone: (916) 255-3000
CalNet: 8-494-3000

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REPORT PREPARED BY:

JANIS COOKE, PH. D.
Environmental Scientist

And

Staff of the Sacramento River TMDL Unit

CLEAR LAKE TMDL FOR MERCURY
EXECUTIVE SUMMARY

Section 303(d) of the Federal Clean Water Act requires States to identify waterbodies that are not meeting water quality standards despite use of technology-based controls and to develop programs to correct the impairments. These requirements will be met through Total Maximum Daily Load (TMDL) programs. A TMDL represents the total loading rate of a pollutant that can be discharged to a waterbody and not result in impairments. The Clear Lake TMDL water quality management plan includes: establishment of a water quality numeric target; assessment of pollutant sources; linkage between the numeric target and loads; assignment of load reductions; and a margin of safety.

The Central Valley Regional Water Quality Control Board has determined that Clear Lake in Lake County, California is impaired because of high levels of mercury in fish. The California Department of Health Services issued a fish consumption advisory in 1987. Based upon levels of mercury in fish tissue and the fish consumption advisory, the Regional Board placed Clear Lake on the Clean Water Act 303(d) List of Impaired Waterbodies in 1988. The goal of this TMDL is to lower mercury levels in Clear Lake so that the beneficial uses of fishing and wildlife habitat are attained.

Clear Lake is a shallow, eutrophic waterbody that is comprised of three basins, the Upper, Lower and Oaks Arms. It is the largest natural lake located entirely within California's boundaries. Tourism and sport fishing are important sectors of the local economy. Five Native American Tribes utilize resources of the lake and its watershed.

The Clear Lake watershed lies within a region naturally enriched in mercury. The Sulphur Bank Mercury Mine (SBMM) site, on the shore of Oaks Arm, was a highly productive source of mercury between 1872 and 1957. Several smaller mines were located in the Clear Lake watershed, all of which are now inactive. Levels of mercury in Clear Lake sediments rose significantly after 1927, when open pit operations became the dominant methodology used at SBMM. The Bradley Mining Company currently owns SBMM. The U.S. Environmental Protection Agency (USEPA) declared the SBMM a federal Superfund site in 1991. Since then, several remediation projects have been completed, including regrading and vegetation of mine waste piles along the shoreline and construction of a diversion system for surface water runoff. At one time, the steep, unvegetated slopes of waste rock piles were a notable source of mercury entering Clear Lake. Remediation of the waste piles appears to have significantly reduced erosion of mine material into the lake. The USEPA is currently conducting a remedial investigation to fully characterize the SBMM site in order to propose final remedies. The USEPA Superfund Program expects to release the Remedial Investigation/Feasibility Study Report for the terrestrial area of the site in 2002.

Numeric Targets

Various media for development of numeric targets were considered, including sediment, water and biota. The primary route of exposure of humans and wildlife to methylmercury, which is the most toxic form of mercury, is through consumption of contaminated fish and other aquatic organisms. Therefore, staff selected a target of mercury in fish tissue because it provides protection to fish consumers and the most direct assessment of fishery conditions and improvement.

The key variables needed for the calculation of fish tissue targets are: acceptable daily intake level of methylmercury (reference dose); age and body weight (bwt) of the consumer; amount of fish consumed; trophic level or size of fish consumed; and portion size. The following basic equation was used to calculate the fish tissue targets:

$$\frac{\text{Reference Dose} * \text{Consumer's Body Weight}}{\text{Consumption Rate}} = \text{Acceptable level of mercury in fish tissue}$$

The reference dose is the quantity at or below which humans consuming methylmercury are expected to be protected from adverse effects. Regional Board staff used the USEPA reference dose of 0.1 micrograms mercury/kg bwt/day, an adult body weight of 65 kg, and the USEPA default consumption rate of 17.5 g/day of locally caught fish. Proportions of trophic level 2, 3 and 4 fish within the 17.5 g/day consumption rate were adjusted using Clear Lake-specific data. Consumers were assumed also to eat an average rate of commercial fish. For the Clear Lake Mercury TMDL, **Regional Board staff proposes numeric targets of 0.13 and 0.30 mg mercury/kg wet weight of fish tissue in trophic levels 3 and 4 fish, respectively**. These targets apply to the average of mercury concentrations in each trophic level. Currently, concentrations of mercury in Clear Lake fish average 0.2 mg/kg in trophic level 3 fish (includes bluegill and hitch) and 0.5 mg/kg in trophic level 4 fish (includes bass, catfish and crappie).

Wildlife species potentially at risk from toxic effects of mercury are those that eat fish or other aquatic organisms that contain mercury. Species at risk at Clear Lake include river otter, raccoon, mink, herons, grebes, bald eagles and osprey. The same method described above for humans can be used for wildlife to determine safe fish tissue concentrations. Due to uncertainties in reference doses, consumption patterns of wildlife, and whether wildlife at Clear Lake are being adversely impacted by mercury, Regional Board staff did not recommend separate fish tissue targets to protect wildlife. Regional Board staff will evaluate new information relative to wildlife risks prior to amending the Clear Lake TMDL into the Water Quality Control Plan for the Central Valley Region (Basin Plan).

Source Analysis

Inorganic mercury loads entering Clear Lake were estimated for the following sources: groundwater and surface water from the SBMM site; tributaries and other surface water that runs directly into the lake; and atmospheric deposition, including atmospheric flux from SBMM. Outputs of mercury were estimated for: flux to the atmosphere from the lake surface; Cache Creek downstream flow; and burial in sediment. The lakebed sediment consists of an active, surficial layer, in which mixing, resuspension, deposition, chemical cycling and methylation occur. Below the active layer, mercury becomes buried and removed from the cycle. **The linkage analysis and load allocations focus on removing mercury from the active layer of lakebed sediment.**

Sulphur Bank Mercury Mine

Mercury from SBMM continues to enter Clear Lake through groundwater, surface water, and atmospheric routes. The groundwater and surface water estimations include mercury in acid rock drainage from the site. A major route of transport is in groundwater through the waste rock dam (WRD), a large pile of waste rock between the main mine pit and Clear Lake.

The total of inputs from SBMM to the active sediment layer includes ongoing releases, as described above, and contributions of previously deposited mercury. Mercury in the active sediment layer may also derive from remobilization of mercury deposited in the past due to mine-related processes. While most mercury from SBMM that was deposited in the past was likely buried under incoming sediment, some historically-deposited mercury may still be contributing to the upper sediment layer. The amount and effects of this remobilized mercury need to be evaluated. SBMM-related mercury from these various sources will require different remediation activities and may have different degrees of bioavailability. These inputs were combined, however, for the purpose of determining total inputs from SBMM.

Inputs from SBMM to the surficial sediment layer (includes historical sediment remobilization and ongoing inputs) have a broad range of 2-695 kg/year. The USEPA Superfund estimate of 2 kg/year of mercury flux through the WRD is used as the lower bound of mercury inputs from SBMM. Based upon our analysis of data from the USEPA Superfund Program and the UC Davis Clear Lake Environmental Research Center, Regional Board staff believes that the 2 kg/year load underestimates the amount of mercury coming through the WRD. Regional Board staff estimate that 695 kg/year is the upper bound of all inputs from SBMM. This upper bound is the total amount of mercury deposited to surficial sediment annually, minus other inputs. This estimate of mercury in surficial sediment deposited yearly is based on mercury concentrations in lakebed sediment and annual sedimentation rates. The upper bound is likely a worst case scenario of inputs from SBMM. Regional Board staff will refine this estimate when additional information becomes available. The upper and lower bounds were calculated using different data and methodologies and should not be directly compared. The USEPA has not yet published an estimate of the total, ongoing inputs of mercury to Clear Lake or of the remobilized, historically-deposited sediment.

The USEPA is continuing to investigate routes and effects of mercury transported from the SBMM site. These studies include measuring levels of mercury fluxed into the air from mine waste piles and estimating local deposition of the air-borne mercury. The USEPA Superfund program is also continuing to examine mercury transported through wetlands north of the mine site. The wetlands are currently used for cattle grazing and as a source of fish, tules, and other resources utilized by Members of the Elem Pomo Tribe. One waste rock pile extends into the wetlands. The USEPA has also indicated its intention to investigate the contaminated lakebed sediments associated with SBMM.

Although decreasing total mercury loads is important, implementation of the TMDL should eliminate sources of mercury that contribute most significantly to methylmercury produced in the active sediment layer. Groundwater from SBMM appears to contribute mercury that is more readily methylated than mercury from other inputs. Groundwater moving through the WRD is acidic. Under conditions of acidity and high redox potential, groundwater accumulates high concentrations of mercury that are very soluble. The groundwater from SBMM is high in sulfate, which also facilitates methylation. This assertion is supported by data showing that methylation rates near the mine are significantly higher than in other parts of Clear Lake. Hydrodynamic modeling of currents and particle movement in the lake demonstrates that particles formed near the mine site can be carried relatively rapidly into other arms of Clear Lake. In contrast to mercury in SBMM groundwater, mercury in lakebed and tributary sediments originates primarily as cinnabar, which has low solubility in water.

Other Mercury Inputs

Mercury entering from tributaries originates in runoff from naturally mercury-enriched soils and mercury deposited in the watershed from the atmosphere. Geothermal springs may contribute to tributary loads,

particularly in the Schindler Creek tributary to Oaks Arm. Average loads from the tributaries were determined for a ten-year period, including low and high water years. Tributary and watershed runoff loads ranged from 1 to 60 kg/year, with an average of 18 kg/year.

Small amounts of mercury deposit directly on the surface of Clear Lake from the global atmospheric pool and potentially from local, mercury enriched sources. Atmospheric loads to the lake surface from the regional and global pool were estimated using data from monitoring stations in Mendocino County and San Jose. Estimates ranged from 0.6 to 2.0 kg/year.

Geothermal springs and lava tubes that discharge directly in the lake do not appear to be significant sources of mercury to Clear Lake. Mercury concentrations in surficial sediment samples collected near lakebed geothermal springs were not elevated, relative to levels in sediment away from geothermal springs.

Outputs of Mercury from Clear Lake

Mercury is removed from active cycling in flow downstream to Cache Creek, in water extracted for municipal and agricultural uses, in biota removed from the lake for human and wildlife consumption, through flux to the atmosphere, and by deep burial in lakebed sediment. Burial is the most significant route of mercury removal from the system. An average of 5 kg of mercury is estimated to be removed from Clear Lake annually through the outputs other than sediment burial.

Linkage Analysis

A linkage analysis describes the association of numeric targets with identified sources of mercury. This relationship provides a basis to estimate total assimilative capacity and identify load reductions. Key steps in the linkage are the relationship between methylmercury in fish tissue and methylmercury in the water column and the association between methylmercury and total mercury in the sediment. Many factors influence methylation and uptake, including concentration and activity of methylating and demethylating bacteria, water temperature, pH, sulfate, organic carbon, chemical form of mercury, and prey availability. Mathematical relationships describing effects of these factors are difficult to obtain. Research is ongoing at Clear Lake and in other waterbodies to better define these associations.

The linkage analysis for Clear Lake assumes a directly proportional relationship between methylmercury in fish tissue and inorganic mercury in the lake sediments and water column. This approach has been used in modeling efforts for other mercury TMDLs. Research conducted in the Everglades showed significant relationships between methylmercury and total mercury. Although other factors are involved in modulating methylation and bioaccumulation, inorganic mercury is a necessary component and can be controlled through load reductions.

Meeting the numeric targets proposed in this TMDL would require reduction of existing fish tissue concentrations of mercury by 40%. The linear relationship dictates that overall mercury loads to Clear Lake sediment must be reduced by 40% in order to reduce methylmercury concentrations in fish tissue by the equivalent amount. However, staff is establishing the assimilative capacity of inorganic mercury in Clear Lake sediments as 50% of existing levels to account for the considerable uncertainties in the linkage analysis.

Load Allocations

The linkage analysis showed that concentrations of mercury in surficial sediment must be reduced by one half. Therefore, ongoing contributions to the active layer of sediment must be reduced by 50%. Using available data, Regional Board staff estimates that approximately 715 kg of mercury is present in sediment deposited annually to the lakebed. This estimate is based on surficial sediment samples and the annual deposition rate of sediment to the lake bottom. The best estimate available of the acceptable load in sediment to meet the targets is approximately 358 kg/year. Loads of mercury to the active layer come from the following sources: tributaries; atmospheric deposition; the SBMM site; and mercury that was previously deposited in the sediment and becomes remobilized. The total load reduction will come from reductions in ongoing inputs and by controlling mercury in sediment that is remobilized. Load allocations are as follows:

a) Atmospheric Deposition. The allocation for atmospheric deposition is capped at the maximum load estimated to accumulate from the global atmospheric pool, 2 kg/year. Atmospheric mercury originating outside of the Clear Lake watershed is considered to be uncontrollable under this TMDL. Mercury from SBMM that fluxes into the air and deposits locally should be controlled by USEPA Superfund remediation activities.

b) Tributaries and Surface Water Runoff. The load allocation of mercury from tributaries and direct surface water runoff is 90% of existing input. Mercury inputs from tributaries and surface runoff will vary with precipitation and water flow. For an average water year, the load allocation is 16 kg/year.

   Sediment from tributaries contains less mercury per unit sediment than in lakebed sediment. It is proposed to allow cleaner sediment to cover the more contaminated sediment in the lake. Nevertheless, reducing total mercury loads to Clear Lake is required under the TMDL program. There may be "hot spots" of mercury loading within the tributaries that can be eliminated. In the first phase of TMDL implementation, Lake County, U.S. Bureau of Land Management, and U.S. Forest Service will be asked to partner with the Regional Board to develop tributary monitoring plans to identify potential hot spots of mercury loading. In the second phase, load reductions for hot spots will be developed and implemented. Proposed watershed restoration projects are likely to reduce overall sediment inputs to the lake, which will decrease input of mercury adhered to the sediment. Ecosystem restoration or preservation projects on tributaries to Clear Lake must not increase loads of methylmercury beyond existing levels.

c) Sulphur Bank Mercury Mine. The remainder of load reductions will come from reducing inputs to surficial lakebed sediments from existing discharges and historical deposits from SBMM. The TMDL requires that ongoing contributions to the active sediment layer from SBMM be reduced by 49%. The best available data suggests that past and present processes at SBMM contribute a maximum of 695 kg/year to the active sediment layer. Based on these data, the total load allocation for SBMM is 340 kg/year, which is the difference between acceptable load in sediment (358 kg/year) and the other allocations. Ongoing inputs include local deposition of mercury fluxed into the air and mercury in groundwater and surface water discharges. Because mercury in groundwater from the mine site is preferentially methylated, the load from SBMM groundwater is limited to 0.1 kg/year. The rest of the reduction should come from limitations in input from other ongoing discharges and resuspension of mercury deposited in prior years. Remobilization of mercury deposited in the past, particularly in the area directly offshore of SBMM, has not been sufficiently investigated. The USEPA Superfund Program should examine and address the effects on existing sediment

concentrations of past loading of mercury to the lake from the mine site. The implementation plans for reductions at SBMM will come from the Superfund Program Remedial Investigation and Feasibility Study Reports (RI/FS) and subsequent Records of Decision for Operable Unit 1, the mine site itself, and later operable units that include the sediments and biota of Clear Lake. In order to fully assess the effects of mercury from SBMM, Regional Board staff recommends that all operable units be completed.

Table ES-1. Existing Loads and Load Allocations for Mercury in Surficial Sediment of Clear Lake			
	Existing Loads (kg/year) *	**Load Allocation**	Estimated Acceptable Load under TMDL (kg/year)
Deposition from the global atmospheric pool	2	**(no change)**	2
Tributaries and direct surface water runoff	18	**90% of existing loads**	16
Sulphur Bank Mercury Mine (includes estimates of ongoing inputs and remobilization of mercury previously deposited in the lake)	Maximum estimated input: 695	**49% of existing loads**	340
Totals (Load allocation is 50% of existing loads)	715		358
* Existing and acceptable loads are based upon best available data for mercury in surficial sediment of Clear Lake. Meeting the numeric targets requires that surficial sediment concentrations and, therefore, total loads to surficial sediment be reduced by 50%. Acceptable loads may be revised as better data becomes available.			

Margin of Safety

A margin of safety was incorporated into the TMDL at several points. The most significant margin of safety is the 10-fold uncertainty factor in the reference dose of methylmercury for humans, which was used for the numeric targets. The USEPA human health reference dose is ten times lower than the lowest level of methylmercury intake observed to have adverse effects in children. The second margin of safety included with the numeric target is another 5% reduction in fish tissue mercury concentrations to account for some of the variability in consumption rates by humans. The third margin of safety is a 10% factor incorporated into the linkage analysis and assimilative capacity to account for considerable uncertainties in the linkage analysis.

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LIST OF ACRONYMS

303(d) List	Clean Water Act 303(d) List of Impaired Waterbodies
ATSDR	U.S. Agency for Toxic Substances and Disease Registry
BAF	bioaccumulation factor
Basin Plans	Water Quality Plans
BCF	bioconcentration factor
bwt	body weight
CDFG	California Department of Fish and Game
CDHS	California Department of Health Services
CTR	California Toxics Rule
CWA	federal Clean Water Act
GLWQI	Great Lakes Water Quality Initiative Final Rule
Hg	Mercury
LOAEL	lowest-observable adverse effect level
MOE	Margin of Exposure
MRC	Mercury Study Report to Congress
MRL	ATSDR Minimal Risk Level
N	population size
NAS	National Academy of Sciences
NOAEL	no-observable adverse effect level
NRC	National Research Council
OEHHA	Office of Environmental Health Hazard Assessment
PCBs	polychlorinated biphenyls
Regional Board	Central Valley Regional Water Quality Control Board
RfD	reference dose
SBMM	Sulphur Bank Mercury Mine
SDCDHS	San Diego County Department of Health Services
State Board	State Water Resources Control Board
Target Report	Clear Lake Mercury TMDL Numeric Target Report
TL3	trophic level 3
TL4	trophic level 4
TMDL	Total Maximum Daily Load
TMDL Report	Clear Lake Mercury TMDL Report
UC Davis	University of California-Davis

UC Davis CLERC	University of California, Davis Clear Lake Environmental Research Center
USDA	U.S. Department of Agriculture
USEPA	U.S. Environmental Protection Agency
USFDA	U.S. Food and Drug Administration
USFWS	U.S. Fish and Wildlife Service
WHO	World Health Organization
WRD	Waste Rock Dam (waste rock pile on the SBMM site)

UNITS OF MEASURE

µg	microgram
µg/g	microgram per gram
µg/L	microgram per liter
µm	micrometer
cm	centimeter
g	gram
g/day	gram per day
g/L	gram per liter
kg	kilogram
L	liter
m	meter
mg	milligram
mg/g	milligram per gram
mL	milliliter
mm	millimeter
ng	nanograms
ng/L	nanograms per liter
ppm	Parts per million; usually mg/kg or µg/g.

1 PROBLEM STATEMENT

1.1 Regulatory Background

1.1.1 Clean Water Act 303(d) Listing and Total Maximum Daily Load Development

Section 303(d) of the federal Clean Water Act requires States to:

1. Identify those waters not attaining water quality standards (referred to as the "303(d) list").
2. Set priorities for addressing the identified pollution problems.
3. Establish a "Total Maximum Daily Load" (TMDL) for each identified waterbody and pollutant to attain water quality standards.

The 303(d) List of Impaired Waterbodies for the Central Valley is prepared by the Regional Board and approved by the State Water Resources Control Board (State Board) and the USEPA. Waterbodies on the 303(d) List are not expected to meet water quality standards even if dischargers of point sources comply with their current discharge permit requirements. A TMDL represents the total loading rate of a pollutant that can be discharged to a waterbody and still meet the applicable water quality standards. A TMDL report describes the reductions needed to meet water quality objectives and allocates those reductions among the sources in the watershed. Elements of a TMDL report include:

1. problem statement;
2. numerical water quality target;
3. identification and quantification of sources and source loads;
4. analysis of the linkage between the water quality target and amount of contaminant;
5. maximum load of the contaminant that will not adversely impact beneficial uses;
6. allocation of portions of the necessary load reduction to the various sources;
7. margin of safety that takes into account uncertainties and seasonal variations;
8. plan and program of implementation to achieve the needed load reductions; and
9. monitoring plan to assess progress of the TMDL program.

1.1.2 Porter-Cologne Basin Plan Amendment Process

In general, the Regional Board will develop a water quality management strategy for each waterbody and pollutant in the Central Valley identified on California's 303(d) List. The management strategy will include several phases:

1. <u>TMDL Development</u>: involves the technical analysis of the sources of pollutant, the fate and transport of those pollutants, the numeric target(s), and the amount of pollutant reduction that is necessary to attain the target.
2. <u>Implementation Planning</u>: involves an evaluation of the practices and technology that can be applied to meet the necessary load reductions, the identification of potentially responsible parties, a description of the implementation framework (e.g. incentive-based, waste discharge requirements, and prohibitions), a time schedule for meeting the target(s), and a consideration of cost.
3. <u>Basin Planning</u>: focuses on the development of a Basin Plan Amendment and a Functionally Equivalent Document for Regional Board consideration. The Basin Plan Amendment will include those policies and regulations that the Regional Board believes are necessary to attain water quality

objectives. The Functionally Equivalent Document includes information and analyses required to comply with the California Environmental Quality Act.

4. Implementation: focuses on the establishment of a framework that ensures that appropriate practices or technologies are implemented (§13241 and §13242 of the Porter-Cologne Water Quality Act), including those elements necessary to meet federal TMDL requirements (CWA Section 303(d)).

1.1.3 Timeline and Process for the Clear Lake Mercury Management Strategy

Regional Board staff completed the TMDL Development phase of the Clear Lake mercury management strategy in February. A draft version of the *Clear Lake Mercury TMDL Final Report* was distributed to involved agencies and the interested public in early December 2001 for review. Comments received before 7 January 2002 were incorporated into the final version of this report. Comments received after 7 January will be incorporated during development of the Basin Plan Amendment and implementation plan. The Implementation Planning phase will rely heavily on the evaluation of remedial options being conducted by the USEPA's Superfund program for the Sulfur Bank Mine site. The results of USEPA's evaluation, and other public input on implementation options, could provide support for modification of the recommendations in the TMDL Report. The Implementation Planning phase should be complete by May 2002.

Comments received on the TMDL Report will be incorporated into the Basin Planning Phase. The Basin Plan Amendment Staff Report will contain alternatives for water quality objectives and implementation plans, evaluation and recommendations for objectives and implementation plan, proposed Basin Plan Amendment language, economic considerations and environmental analysis. The final TMDL Report will be an appended to the Basin Plan Amendment Staff Report. Should an evaluation of implementation options indicate that the beneficial uses could not be reasonably attained, Regional Board staff may prepare a Use Attainability Analysis as part of the Basin Plan Amendment. Regional Board staff anticipates that the Regional Board will adopt a Basin Plan Amendment for the mercury numeric targets and TMDL implementation plan by Fall 2002. The Basin Plan Amendment for the Clear Lake TMDL will be legally applicable once the amendment is adopted by the Regional Board and approved by the State Board, State Office of Administrative Law and the USEPA. Implementation will begin after the Basin Plan Amendment is legally applicable.

Regional Board staff intends to seek public input throughout the TMDL Development and Implementation Planning phases. In May 2002, Regional Board staff anticipates holding a public workshop in the Clear Lake area to receive comments on the proposed Basin Plan Amendment.

1.2 Watershed Characteristics and TMDL Scope

Clear Lake is located in the Coast Range in Lake County, California. It is a shallow, eutrophic[1] waterbody that has a length of approximately 18 miles, a surface area of approximately 43,000 acres, and a surface elevation of 1,326 feet above mean sea level (MSL) (USEPA, 1994; (USGS, 1990-1993). It is the largest natural lake located entirely within California's boundaries. Clear Lake is comprised of three distinct basins: the northern large, circular Upper Arm, the elongated southeast-trending Lower Arm, and the relatively small Oaks Arm located to the east (Figure 1). The mean depth of the basins ranges from

[1] A *eutrophic* waterbody is enriched in dissolved nutrients that stimulate the growth of aquatic plant life.

23 feet in the Upper Arm to 36 feet in Oaks Arm. The lake empties at the southern end of the Lower Arm into the South Fork of Cache Creek.

The Clear Lake watershed has an area of approximately 337,000 acres, approximately 75% of which drains into the Upper Arm. The mountains surrounding Clear Lake vary in elevation from 2,000 to 4,600 feet above MSL. The weather and vegetation of the Clear Lake region is typical of Mediterranean climates (USEPA, 1994). Grassland, scrub oak, stands of cypress, manzanita, and other chaparral-type plants are distributed between the lowlands and moderately high ridges, with stands of evergreen conifers and some deciduous trees in the higher elevations. Moderate to heavy precipitation can locally exceed 100 inches per year in the mountains, and can be as low as 20 inches per year in the Clear Lake basin. The mean annual precipitation at the Sulphur Bank Mercury Mine (SBMM) located on Oaks Arm is 24 inches, with 80% of the rain falling between November and March. During the winter, snow commonly falls in the mountains above the 3,000-foot elevation. The mean annual lake evaporation is 48 inches. The mean monthly precipitation usually exceeds mean monthly evaporation from November through February. Mean annual temperatures for the Clear Lake region are about 60 degrees Fahrenheit (^{o}F), with summer temperatures exceeding 100 ^{o}F and winter temperatures dropping below freezing.

The Clear Lake basin located in the northern Coast Range geomorphic province, approximately 60 miles east of the San Andreas Fault. The basin is a fault-bounded subsiding depression, believed to be a pull-apart basin related to a releasing bend in the San Andreas Fault. The regional bedrock of the Coast Range consists of a structurally complex group of rocks known as the Franciscan Formation, which formed during the Late Jurassic to Cretaceous period when sediments on the sea floor were scraped off and piled onto the continent as the Pacific plate was subducted beneath the North American Continental plate. Regional volcanic activity since that time may be related to the extensional faulting in the Clear Lake basin. The shallow magma chamber beneath the Geysers-Clear Lake area is the source of geothermal activity throughout the region.

Groundwater in the Clear Lake region is typically characterized by shallow aquifers that flow from the mountains into Clear Lake (USEPA, 1994). It is believed that there is little groundwater seepage lost from Clear Lake due to the low permeability of the underlying Franciscan Formation. The U.S. Geological Survey has mapped numerous hot springs discharging in the area. A large number of these springs vent directly into Clear Lake (USEPA, 1994).

Several small communities and resorts surround the perimeter of Clear Lake. The largest in the area is the City of Clearlake (population 15,200), located adjacent to the Lower Arm, north of the South Fork of Cache Creek. The communities of Nice, Lucerne, and Lakeport are located adjacent to the Upper Arm; Clearlake Oaks is located adjacent to the Oaks Arm; and Lower Lake is adjacent to the Lower Arm, south of Cache Creek. The Elem Colony of Southeastern Pomo Native Americans (Elem Tribal Colony or Sulphur Bank Rancheria) is located along the eastern perimeter of Oaks Arm, adjacent to the SBMM. The local economy is heavily dependent upon tourism, fishing and agriculture (USEPA, 1994).

The Clear Lake watershed lies within a region naturally enriched in mercury. The Sulphur Bank Mercury Mine (SBMM), on the shore of Oaks Arm, was a highly productive source of mercury between 1880 and 1957. Several smaller mines were located in the Clear Lake watershed, all of which are now inactive. Levels of mercury in Clear Lake sediments rose sharply after around 1927, when open pit operations

began at SBMM. The Bradley Mining Company currently owns SBMM. The mine is also a federal Superfund site.

1.3 Mercury Chemistry and Accumulation in Biota

Mercury (Hg) can exist in various forms in the environment. It has the properties of a metal in that it persists in the environment and doesn't break down. Mercury also has some properties of a hydrophobic organic chemical because it can be methylated via a bacterial process. Physically, mercury may be present in air as mercury vapor, dissolved in the water column, or associated with solid particles in air, water, or soil. Chemically, mercury can exist in three oxidation states: elemental (Hg^0), mercurous ion (monovalent, Hg_2^{+2}), or mercuric ion (divalent, Hg^{+2}). Ionic mercury can react with other chemicals to form both organic and inorganic compounds. Hg^{2+} generally predominates in aquatic systems. Organic forms include methylmercury (CH_3Hg^+), dimethylmercury and manufactured compounds such as organic mercury pesticides.

Sulfate-reducing bacteria convert inorganic mercury to methylmercury as a by-product of their normal respiration (Gilmour et al., 1992). Other types of bacteria in Clear Lake also participate in methylation (Mack, 1998). Important factors controlling the methylation rate include temperature, percent organic matter, redox potential, salinity, pH, ratio of sulfate to sulfide, and mercury concentration (Barkay et al., 1997; Xun et al., 1987). In lakes, methylation occurs mainly at the sediment water interface and at the oxic-anoxic boundary within the water column. Methylmercury can also be converted back into Hg^{+2}, primarily via bacterial degradation, in a process known as demethylation (Oremland et al., 1995). Dissolved methylmercury is quickly taken up into the food web or demethylated.

The ore mined at Clear Lake contained cinnabar (red HgS) and metacinnabar (black HgS). Cinnabar is the predominant natural ore in the Coast Range mercury belt. HgS has low solubility and settles out to bottom sediments. However, under aerobic conditions, bacterial-mediated oxidation can release Hg^{2+}, which would be available for methylation (Morel, 1998).

Mercury and methylmercury form strong complexes with organic substances (including humic acids) and strongly sorb onto soils and sediments. Once sorbed to organic matter, invertebrates can ingest mercury, thus entering it into the food chain. Some of the sorbed mercury will settle to the lake bottom; if buried deeply enough, mercury in bottom sediments will become unavailable to the mercury cycle. Burial in bottom sediments is an important route of removal of mercury from the aquatic environment. A conceptual model of mercury cycling in Clear Lake is shown in Figure 2.

Although it is important to identify sources of mercury to the lake, there may be fluxes of mercury within the lake that would continue nearly unabated for some time even if all sources of mercury to the lake were eliminated. In other words, compartments within the lake probably currently store a significant amount of mercury, and this mercury can continue to cycle through the system even without an ongoing outside source of mercury. The most important store of mercury within the lake is the bed sediment. Mercury in the bed sediment may cause exposure to biota by being resuspended into the water column.

Of the various forms of mercury, it is methylmercury that poses the real threat to biota due to its strong tendency to accumulate in biota and magnify up the food chain. Both Hg^{2+} and methylmercury are

ingested, but methylmercury is preferentially retained by and passed to the next trophic level (inorganic mercury is relatively easily egested). Invertebrates eat both algae and detritus, thereby accumulating any methylmercury that has sorbed to these. Fish then eat the invertebrates and accumulated mercury.

For low trophic level[2] species such as phytoplankton, most mercury is obtained directly from the water. *Bioconcentration* describes the net accumulation of mercury directly from water. The *bioconcentration factor* is the ratio of mercury concentration in an organism to mercury concentration in water. However, predatory species such as piscivorous (fish-eating) fish and birds obtain most mercury from mercury-containing prey rather than directly from the water (USEPA, 1997d). A *bioaccumulation factor* describes the degree to which mercury accumulates from water and prey, relative to mercury concentration in the water. Repeated consumption and accumulation of mercury from contaminated food sources result in tissue concentrations of mercury that are higher in each successive level of the food chain. This process is termed *biomagnification*. The processes of bioaccumulation and biomagnification produce high levels of mercury in organisms high on the food chain, despite nearly immeasurable quantities of mercury in the water column.

The proportion of total mercury that exists as the methylated form generally increases with level of the food chain, approaching greater than 90% in top trophic level fish (Nichols et al., 1999). Field studies indicate that diet is the primary route of mercury uptake by fish (Wiener and Spry, 1996). Methylmercury is the predominant form of organic mercury present in biological systems. Dimethylmercury, which is an unstable compound that dissociates to methylmercury at neutral or acid pH, is not considered to be a concern in freshwater systems (USEPA, 1997a).

Diet is also the primary route of methylmercury exposure for organisms that consume fish and aquatic invertebrates. Although a few studies have indicated that methylmercury impairs reproduction of some fish (Huber, 1997; Wiener and Spry, 1996), the greatest concern for mercury toxicity is in higher trophic-level organisms that consume aquatic life. The aquatic food web provides more than 95% of humans' intake of methylmercury (USEPA, 1997c). Wildlife in the Clear Lake area potentially at risk for mercury toxicity include herons, egrets, grebes, mergansers and other fish-eating waterfowl, kingfishers, bald eagles, osprey, mink, raccoons, bats, and otter.

1.4 Toxicity of Mercury

1.4.1 Effects on Humans

Mercury is a potent neurotoxin in humans. Developing fetuses and young children are at greatest risk of toxicity from mercury (NRC, 2000). Although the inhalation of elemental mercury fumes can cause harm, exposure to levels of concern most frequently occurs through the consumption of methylmercury in fish tissue. Toxicity of mercury to humans has been documented in populations consuming contaminated

[2] Trophic levels are the hierarchical strata of a food web characterized by organisms that are the same number of steps removed from the primary producers. The USEPA Mercury Study Report to Congress used the following criteria to designate trophic levels based on an organism's feeding habits (USEPA 1997c):

Trophic level 1: Phytoplankton.
Trophic level 2: Zooplankton, benthic invertebrates, and fish that eat phytoplankton.
Trophic level 3: Organisms that consume zooplankton, benthic invertebrates and/or herbivorous fish.
Trophic level 4: Organisms that consume trophic level 3 organisms.

fish (Davidson et al., 1998; Grandjean et al., 1997; Kjellstrom et al., 1989; Tsubaki and Irukayama, 1977) and grains treated with methylmercury-containing fungicide (Bakir et al., 1973). Consumption of highly contaminated fish caused multiple effects, including tingling or loss of tactile sensation (paresthesia), loss of muscle control, blindness, paralysis, birth defects and death. Children whose mothers ate fish during pregnancy may be at risk for more subtle behavioral and neurodevelopmental impairments (Crump et al., 1998; Davidson et al., 1998; NRC, 2000). Children who eat fish themselves are also believed to be more sensitive than adults to mercury because their neural systems are still developing and they tend to consume more fish per body weight than adults (Grandjean et al., 1999; Mahaffey, 1999). Effects in children exposed early in development appear at dose levels five to ten times lower than dose levels associated with toxicity in adults (NRC, 2000).

Although the largest body of literature addresses effects of mercury on neurodevelopment, studies have found impairment of other human organ systems as well. Exposure to mercury has been found to cause reduced fertility, adverse cardiovascular effects, and immunotoxicity, and to alter cell division (NRC, 2000; Speirs and Speirs, 1998).

Effects of mercury are dependent upon the dose received. Levels of mercury in fish from Clear Lake are much lower (0.2 to 1.9 microgram per gram (µg/g), wet weight for top predator fish) (CVRWQCB, 1985) than levels in fish that poisoned consumers in Minamata Bay, Japan (mercury levels up to 50 µg/g) (Bakir et al., 1973; Marsh et al., 1987; Tsubaki and Irukayama, 1977). There is no current evidence of acute or chronic mercury toxicity to humans due to consumption of fish from Clear Lake or Cache Creek. However, researchers have not yet conducted extensive fish consumption and effect studies in the region. Existing fish consumption advisories for Clear Lake, presented in terms of pounds of fish that can be safely consumed, are based upon the risk for average adult consumers of developing a non-fatal, neurologic impairment of parasthesia (Stratton et al., 1987). Pregnant women, women who may soon become pregnant, nursing mothers, and children under age six are advised not to eat fish from Clear Lake (Stratton et al., 1987).

1.4.2 Effects on Wildlife

Wildlife species also exhibit detrimental effects from mercury exposure. Researchers have observed behavioral effects – such as impaired learning, reduced social behavior and impaired physical abilities – in mice, otter, mink and a primate species (crab-eating macaques) exposed to methylmercury (Wolfe et al., 1998). Researchers have also observed reproductive impairment following mercury exposure in multiple species, including common loons and western grebe (Wolfe et al., 1998), walleye (Huber, 1997), and mink (Dansereau et al., 1999). In wildlife, sensitive endpoints are reproductive success, motor control and damage to the neural system.

1.5 Beneficial Uses and Applicable Standards

1.5.1 Clear Lake Beneficial Uses Cited in the Basin Plan

Both the Federal Clean Water Act and the State Water Code (Porter-Cologne Water Quality Act) require identification and protection of beneficial uses. The beneficial uses designated in Table II-1 of the Water Quality Control Plan for the Sacramento and San Joaquin Basins (CVRWQCB, 1998) are intended to

meet all applicable State and Federal requirements. Table 1 lists the existing and potential beneficial uses of Clear Lake. Clear Lake provides water for domestic, municipal and agricultural uses within its watershed. It is also a source of agricultural, domestic and industrial waters downstream in the Cache Creek watershed. The beneficial uses that are impaired by mercury in Clear Lake are wildlife habitat and sport/recreational fishing. Elevated mercury levels in fish from Clear Lake pose a risk for humans and wildlife that consume fish taken from the lake.

Table 1. Existing and Potential Beneficial Uses of Clear Lake (CVRWQCB, 1998)

Beneficial Use	Status
Municipal and domestic supply (MUN)	Existing
Agriculture – irrigation and stock watering (AGR)	Existing
Recreation – contact (REC-1) and other non-contact (REC-2)	Existing (a)
Freshwater habitat (Warm)	Existing
Spawning (SPWN) – warm	Existing
Wildlife habitat (WILD)	Existing (a)
Freshwater habitat (Cold)	Potential

(a) Beneficial uses impaired by mercury in Clear Lake.

1.5.2 Water Quality Objectives and Criteria

The narrative water quality objective for toxicity in the Basin Plan states, in part, "All waters shall be maintained free of toxic substances in concentrations that produce detrimental physiological responses in human, plant, animal, or aquatic life." The narrative toxicity objective further states that "The Regional Water Board will also consider … numerical criteria and guidelines for toxic substances developed by the State Water Board, the California Office of Environmental Health Hazard Assessment, the California Department of Health Services, the U.S. Food and Drug Administration, the National Academy of Sciences, the U.S. Environmental Protection Agency, and other appropriate organizations to evaluate compliance with this objective." (CVRWQCB, 1998)

Researchers have developed numeric criteria for mercury in fish tissue and water for both human health and wildlife protection. The USEPA recently established a criterion of 0.3 µg/g methylmercury per wet weight in the edible portions of fish for protection of human health (USEPA, 2001b). The USEPA has also established wildlife criteria for the Great Lakes Water Quality Initiative (USEPA, 1995a, b) and the Mercury Study Report to Congress (USEPA, 1997a). These USEPA criteria suggest that a range of mercury in fish tissue of 0.08 µg/g (trophic level 3 fish; TL3) to 0.35 µg/g (trophic level 4 fish; TL4) should be protective of wildlife. Because wildlife generally consumes lower trophic level (and smaller) fish, the human health and wildlife criteria are not directly comparable.

In the past, the Regional Board has used other guidelines for identifying impaired waterbodies or as screening values. The National Academy of Sciences-National Academy of Engineering (NAS) numeric mercury guideline of 0.5 µg/g (parts per million; ppm) (NAS, 1973) applied to whole, freshwater fish and marine shellfish, for purposes of wildlife protection. The United States Food and Drug Administration (USFDA) action level of 1.0 ppm applies to the edible portion of commercially caught freshwater and marine fish; the action level applies to human health (USFDA, 1984). The USFDA levels were designed only to protect adults consuming a variety of commercial fish and shellfish. While still used, these two guidelines do not incorporate recent, improved information about mercury toxicity.

The USEPA promulgated the California Toxic Rule (CTR) in April 2000 (USEPA, 2000a). The CTR contains a water quality objective of 0.05 µg/L (50 ng/L) total recoverable mercury for freshwater sources of drinking water. The CTR criterion protects humans from exposure to mercury in drinking water and contaminated fish. The standard is enforceable for all waters with a municipal and domestic water supply and/or any aquatic beneficial use designation. Clear Lake has such a beneficial use designation. The federal rule did not specify duration or frequency terms; however, researchers have previously employed a 30-day averaging interval with an allowable exceedance frequency of once every three years for protection of human health, which is recommended for this effort (Personal communication from J. Marshack, CVRWQCB). The U.S. Fish and Wildlife Service and U.S. National Marine Fisheries Service were concerned that the USEPA's mercury objective in the CTR would not be sufficiently protective of threatened and endangered species. The USEPA has committed to revising its water quality objective to include protection of wildlife. Regional Board staff will monitor progress in the development of a USEPA water quality criterion for wildlife protection and will incorporate new information during the Basin Planning process.

1.6 Existing Conditions

Since 1970, several agencies, including the Regional Board, have monitored mercury in Clear Lake by collecting water, lakebed sediment, fish tissue, and other biota samples. In 1987, the California Department of Health Services (CDHS) issued an advisory for consumption of sport fish from Clear Lake (Stratton et al., 1987). In 1988 the Regional Board identified Clear Lake as impaired due to mercury and placed it on the 303(d) List of Impaired Waterbodies. The Regional Board based its decision to list Clear Lake on the elevated levels of mercury in fish tissue and the existence of a fish consumption advisory. The sections below summarize the available environmental data and describe the extent of mercury impairment.

Additional data on mercury levels in wildlife feeding in Clear Lake, particularly some fish-eating birds, suggest that wildlife habitat may also be impaired (CVRWQCB, 1985; Elbert, 1996; Elbert and Anderson, 1998; Suchanek et al., 1997). Limited studies conducted to date with Clear Lake wildlife have found no conclusive link between mercury levels in the environment and reproductive or other impairments of wildlife (Elbert, 1996; Suchanek et al., 1997; Wolfe and Norman, 1998).

1.6.1 Mercury Levels in Fish Tissue

Between 1970 and 1984, the CDHS, California Department of Fish (CDFG), and USFDA collected and analyzed more than 400 fish samples from Clear Lake for mercury. Species tested for mercury included largemouth bass, channel catfish, white catfish, brown bullhead, white and black crappie, bluegill, carp, hitch, Sacramento blackfish and inland silverside.

In 1970 CDHS collected and analyzed two composite fish samples from Clear Lake, one largemouth bass sample and one white catfish sample, each a composite of ten fish. This analysis provided the first indication that fish from Clear Lake might contain excessive levels of mercury (CVRWQCB, 1985). The USFDA analyzed additional fish-tissue samples in 1976 (CVRWQCB, 1985). The Toxic Substances Monitoring Program of the State Water Resources Control Board then collected and analyzed fish

samples from 1980 to 1983 (Rasmussen, 1993). Most data were reported for individual fish, although some data were reported for composite samples. All data reported were for mercury per wet weight in edible tissue. In 1985, Regional Board staff prepared a summary report that contained statistical evaluations and tables of data (CVRWQCB, 1985). Fish tissue data collected through 1985 were also summarized in the report that recommended guidelines for consumption of Clear Lake fish (Stratton et al., 1987).

Staff of the UC Davis Clear Lake Environmental Research Center (UC Davis CLERC) continued the sampling for fish tissue analyses in the 1990s and in 2000 (Suchanek et al., 2000a; Suchanek et al., 1997; Suchanek et al., 1993). Collections in 1994-96 were focused on young fish, mainly inland silversides and juvenile largemouth bass. Larger fish were included in the sampling in 1992 and in 2000.

Concentrations of mercury in fish from Clear Lake are shown in Table 2. More detailed data are shown in Appendix A. Fish-eating (pisciverous, trophic level 4) fish accumulated the highest levels of mercury and that concentrations generally increased with age and size of fish. Concentrations of mercury in fish are, in general, not significantly different between the arms of the lake (Suchanek et al., 1997). Analysis of juvenile largemouth bass and inland silversides caught in 1998 and 1999 showed no decline in mercury concentrations, as compared to 1970-1984 mercury concentrations (Suchanek et al., 2000a). Concentrations in adult largemouth bass also show no decrease with time (Personal Communication from T. Suchanek, 9/01).

Humans consume fish in trophic levels 3 and 4 fish from Clear Lake (Harnly et al., 1997; Macedo, 1991). The most frequently consumed TL4 species are largemouth bass, channel and white catfish, and black crappie. The most frequently consumed TL3 species are bluegill, black bullhead, brown bullhead, carp, hitch, and Sacramento blackfish.

Table 2. Concentrations of Mercury Tissue Clear Lake Fish

Fish species	Mercury concentration, μg/g wet weight (ppm)	
	mean	Standard deviation
Inland silverside	0.09	0.03
Largemouth bass, juvenile	0.18	0.04
Bluegill	0.19	0.20
Hitch	0.19	0.13
Carp	0.20	0.17
Black bullhead	0.22	0.09
Sacramento blackfish	0.28	0.10
Brown bullhead	0.28	0.11
Black crappie	0.36	0.19
White crappie	0.48	0.36
Channel catfish	0.48	0.37
White catfish	0.51	0.18
Largemouth bass, adults	0.54	0.32

Sources: CVRWQCB, 1985; Suchanek et al., 1993; Suchanek et al., 1997

1.6.2 Data for Other Wildlife

A complete ecological assessment of mercury effects has not been completed for Clear Lake. In particular, there is no information on potential sublethal, behavioral or reproductive effects of mercury on resident mammals or on other fish-eating birds. However, some samples from birds, raccoons, minks, and crayfish have been analyzed for mercury. The results of these analyses are described below.

CDFG collected twenty western grebe samples and twenty American coot samples from Clear Lake in March 1984 (CDFG, 1984d). The average concentrations of mercury in grebe breast muscle and liver were 2.0 ppm and 6.4 ppm, respectively. Mercury in breast muscle of coots ranged from below the detection limit of 0.2 ppm to 0.6 ppm. CDFG staff concluded that mercury levels in grebe livers bordered on toxic levels.

Mercury concentrations in tissue samples from grebes (CVRWQCB, 1985; Elbert and Anderson, 1998), herons (Elbert, 1996), and ospreys (Suchanek et al., 1997) from Clear Lake cohorts are elevated compared to mercury concentrations in tissue samples from cohorts in pristine areas. Nesting success of herons and cormorants (Wolfe and Norman, 1998) and ospreys (Suchanek et al., 1997) does not appear to be affected by mercury. However, the numbers of healthy offspring per nest of western grebes at Clear Lake were found to be significantly less than numbers at two other remote California lakes not contaminated by mercury (Elbert and Anderson, 1998). The authors concluded that nesting may be adversely impacted by mercury as well as other factors, such as human disturbance and boating.

Feathers were collected from nesting, fish-eating birds at Clear Lake in the early 1990s (Suchanek et al., 1997). Adult osprey showed the highest mercury values with an average of 20 ppm dry weight. Mercury in feathers of some adult western grebes and great blue herons sampled were at or above 20 ppm, although average levels in feathers of these birds were less than 20 ppm. Feathers from adult double crested cormorants were not tested, but levels in juvenile cormorant feathers suggested levels in adults would be around 25 ppm. A concentration of mercury in feathers of 20 ppm is considered a toxic risk level for birds (Scheuhammer, 1991).

Mercury has been measured in tissues of some mammals caught near the shores of Clear Lake (Wolfe and Norman, 1998). All raccoons and seven of eight mink examined had levels of mercury in brains and fur that were below no-observable effect levels reported in the literature. There are no field data available on reproductive effects of mercury in mammalian wildlife at Clear Lake.

Mercury levels in crayfish sampled from 1994 to 1996 ranged from 0.04 to 0.5 ppm wet weight. Mercury levels were higher in crayfish caught near the mine site than in similarly-sized crayfish caught in Upper or Lower Arms (Suchanek et al., 1997).

A preliminary assessment of hazards to wildlife from mercury and arsenic at Clear Lake was prepared for the 1994 Remedial Investigation Report for Sulphur Bank Mercury Mine (Elbert, 1993). Mercury concentrations in tissues of Clear Lake wildlife were compared with tissue concentrations and effects in published literature. Elbert concluded that mercury concentrations in prey fish from Clear Lake are unlikely to cause lethality of top-trophic level wildlife species. Mercury concentrations in prey fish could be high enough to cause reduced hatching success and/or behavioral abnormalities and reduced survival

of young. Mercury concentrations in adult wildlife could be enough to cause behavioral abnormalities, such as reduced nest attendance, which can result in reduced reproductive success (Elbert, 1993).

1.6.3 Water and Sediment Data

During the 1990s, researchers from the University of California, Davis obtained numerous measurements of mercury in Clear Lake water and sediment (Suchanek et al., 1997; Suchanek et al., 1993). Extensive amounts of sediment and water data were collected in 1994-1996 after USEPA stabilized shoreline waste rock piles at SBMM. Mercury levels in lakebed sediments showed a statistically significant, exponential decline as a function of distance from the SBMM. Surficial sediment just offshore of the mine site contained approximately 300 ppm of mercury; sediment from sites elsewhere in the Oaks Arm contained approximately 40 ppm of mercury. Mercury concentrations in surficial sediment ranged from 10 to 15 ppm in the Narrows, and from 0 to 5 ppm in the Upper and Lower Arms (See Figure D-4 in the Appendix).

Like the sediment samples, unfiltered water samples collected near the SBMM had the highest concentrations of mercury, with concentrations decreasing exponentially as a function of distance from the mine. The CTR criterion for total recoverable mercury has been exceeded in Clear Lake. Of water samples collected May 1994 through August 1996 (every 6-12 weeks), 25% (29/114) of deep water samples and 11% (13/114) of surface water samples contained mercury concentrations greater than 50 ng/L. Most samples with levels above 50 ng/L were collected from Oaks Arm, with only three samples coming from the Narrows, one from Lower Arm and none from Upper Arm. Mercury in water samples from Oaks Arm ranged up to 400 ng/L (Suchanek et al., 1997). A database of several hundred records for total mercury in water collected from 1992 to 1998 (including the above data) lists additional exceedances in Oaks Arm. Of the additional samples collected at the other locations, only one sample exceeded 50 ng/L; that sample was collected from Lower Arm (Suchanek, 2000c).

Levels of mercury in filtered water (i.e., the dissolved fraction) average around 1.0 to 2.0 ng/L. A peak concentration of 8.7 ng/L was measured near the mine site in April 1996, following a winter of heavy rains and overflow of water from Herman Impoundment (Suchanek et al., 1997). Average concentrations of methylmercury were 0.05 - 0.1 ng/L in filtered and 0.1 – 0.2 ng/L in unfiltered water samples taken throughout the lake. The peak of methylmercury production occurred in late summer or fall and was reflected by methylmercury concentrations up to 0.7 ng/L in unfiltered samples (Suchanek et al., 1997).

1.6.4 Humans

One study exists of human exposure to mercury at Clear Lake (Harnly et al., 1997). The sixty-three study participants included members of the Elem Indian Colony and neighbors of the SBMM site. The study showed that the participants consumed fish from the top and middle trophic levels. Asked to recall their consumption of local and commercial fish over the previous six months, some individuals reported consumption in excess of the fish advisory. Mercury levels in hair samples from study participants were less than levels linked with damage to unborn children (See Appendix D for details of the study).

1.6.5 Summary of Existing Conditions

Available data indicate that elevated levels of mercury exist in fish, lakebed sediment, water, birds and other wildlife from Clear Lake. In particular, fish-tissue data collected between 1970 and 1998 indicate that mercury levels in Clear Lake fish frequently exceed numeric criteria established for human health and wildlife protection. High levels of mercury in fish are of concern to humans and wildlife that eat fish from Clear Lake. The California Department of Health Services issued a fish-consumption advisory in 1987. The Regional Board identified Clear Lake as impaired due to mercury and placed it on the 303(d) List of Impaired Waterbodies in 1988. The Regional Board based its decision to list Clear Lake as impaired due to the elevated levels of mercury in fish tissue and the existence of a fish-consumption advisory. Mercury analyses of fish from Clear Lake began in 1970 and continue to the present. Concentrations of mercury in top-predatory fish species (largemouth bass, channel catfish, white catfish, and black crappie) ranged from 0.1 to 1.9 ppm in wet weight of tissue. Average mercury levels in these species were approximately 0.5 ppm. Elevated levels of mercury have also been found in the water, lakebed sediments and other biota at Clear Lake, including western grebes and osprey.

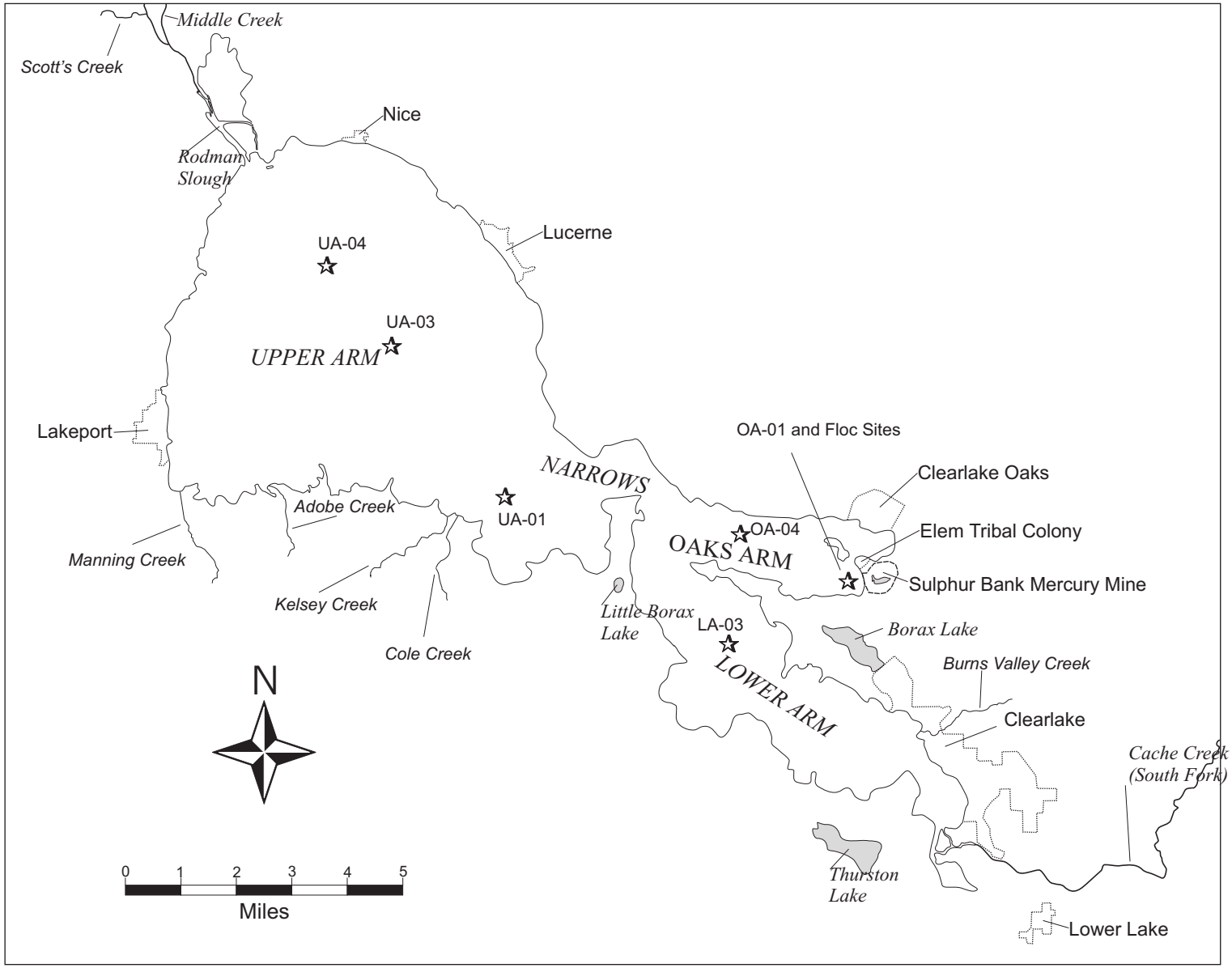

Figure 1. Map of Clear Lake and selected U.C. Davis Sampling Sites
(Sources: U.S. Geological Survey 7.5 Minute Quadrangles, 1990-1993, Suchanek et al., 1997.)

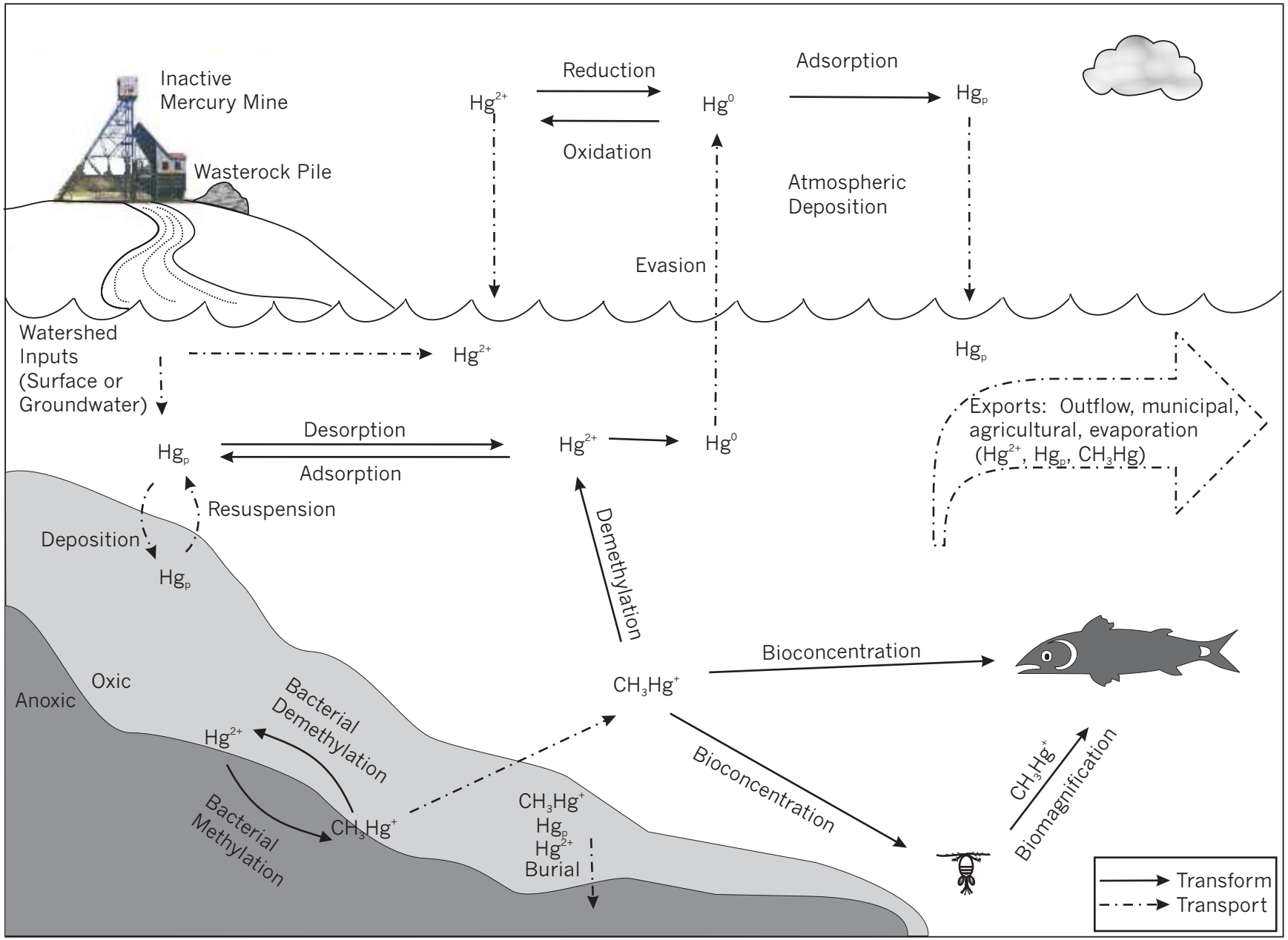

Figure 2. Conceptual Model of Mercury Cycling in Clear Lake

2 NUMERIC TARGET

Derivation of the numeric targets is discussed more fully in a separate document, the "Clear Lake TMDL for Mercury: Numeric Target Report, Final Draft Report" (Appendix H). The Numeric Target Report describes the methodology that the Regional Board staff used in determining water quality targets for mercury, presents options for targets that protect humans and wildlife and options for various factors that are incorporated into final target values, and recommends the final numeric targets. The Numeric Target Report Preliminary Draft was released in August 2000. The preliminary draft was revised based upon information available since August 2000 and comments from public and peer reviewers.

2.1 Types of Targets

Several media for development of numeric targets were considered, including sediment, water column and biota. Measurements of mercury in the target media should be able to assess fairly directly whether beneficial uses are being met. The major beneficial uses of Clear Lake that are currently unmet are as a safe fishery for humans and wildlife. A target of mercury in fish tissue was determined to be the most appropriate because it provides the most direct assessment of fishery conditions and improvement. Mercury data in fish from Clear Lake that have been collected since 1970 provide a good baseline from which to evaluate the success of future load reductions.

The California Toxics Rule (CTR) mercury criterion does apply to Clear Lake. This criterion of 50 ng/L total recoverable mercury in water is intended to protect human health from consuming contaminated organisms and drinking water. Regional Board staff does not consider the CTR value to be sufficiently protective of humans or wildlife consuming fish from Clear Lake because of the low practical bioconcentration factors used to determine the CTR value. However, because the criterion is a definite goal that needs to be met in Clear Lake, it could be used as a secondary target for the TMDL.

2.2 Fish Tissue Targets to Protect Human Health

A fish tissue target can be calculated using the following basic equation:

$$\frac{\text{Daily intake} * \text{Consumer's body weight}}{\text{Consumption rate}} = \text{Acceptable level of mercury in fish tissue}$$

Units in this equation are:

$$\frac{\mu\text{g mercury /kg bwt/day} * \text{kg bwt}}{\text{g fish/day}} = \mu\text{g mercury/ g fish (ppm)}$$

Where: g = gram
μg = microgram
bwt = consumer's body weight
kg = kilogram
ppm = parts per million

The acceptable daily intake is the quantity at or below which humans consuming methylmercury are expected to be protected from adverse effects. Regional Board staff used the USEPA reference dose of

0.1 μg mercury/kg bwt/day for calculation of the TMDL targets. The USEPA reference dose was revised in 2000 and is now based upon data from a population in the Faroe Islands that consumes fish regularly (USEPA, 2001b). The USEPA revision was guided by a comprehensive review of the literature and recommendations by a panel of the National Academy of Sciences (NRC, 2000).

An average bodyweight of 65 kg is recommended for use in developing the target. This is USEPA's standard bodyweight for pregnant females. To best ensure that a mercury target protects the unborn, it is logical that an average adult consumer be represented as a pregnant female. Conversion factors are available to adjust the target for other bodyweights (OEHHA, 1999). Children would only be at risk of mercury toxicity if they consumed more than the average portion for their body size.

Consumption rates of fish and other seafood determined in various national and regional studies vary widely. Mean consumption rates for consumers-only (people who eat no fish were not included) range from 9 to 111 g/day (Gassel et al., 1997). Consumers are exposed to mercury in sport as well as commercial fish. The USEPA recommends default consumption rates that are based on a 1993-94 nationwide dietary survey conducted by USFDA (USEPA 2000b). The USEPA default rate for the general population is 17.5 g/day of locally caught fish, which is the consumption rate for the 90th percentile of those surveyed. The USFDA national survey found that the average consumption rate of commercial fish was 12.5 g/day and that many consumers eat both commercial and locally caught fish. Creel surveys at Clear Lake suggest that more TL4 fish and fewer TL3 and TL2 fish are caught and consumed from Clear Lake than the national average (Macedo, 1991).

One small consumption study has been completed for members of the Elem Tribe and several neighbors of the Sulphur Bank Mercury Mine at Clear Lake (Harnly et al., 1997). Consumption rate by the 90th percentile of study participants was 30 g/day of Clear Lake fish. At least some participants ate commercial fish as well. Species consumed in the greatest amounts were catfish and perch. Consumption information for the general population at Clear Lake has not been collected. More detailed information on consumption rates and mercury measurements obtained from the study participants is provided in Appendix B.

For numeric targets for the Clear Lake Mercury TMDL, Regional Board staff is using 0.13 and 0.30 mg methylmercury/kg wet weight of fish tissue (ppm) in trophic levels 3 and 4 fish, respectively. To obtain these targets, fish tissue concentrations corresponding to safe consumption of 17.5 g/day of local fish were calculated, then lowered by a small, additional safety factor. These targets are derived from consumption patterns of trophic level 3 and 4 fish that are based upon creel surveys conducted in Clear Lake. Meeting these targets would require a 40% reduction from current fish tissue levels. These targets assume that consumers eat an additional 12.5 g/day of commercial fish, such as scallops and tuna.

2.3 Fish Tissue Targets to Protect Wildlife Health

Wildlife species potentially at risk from toxic effects of mercury are those that eat fish or other aquatic organisms that contain mercury. Species of concern at Clear Lake include river otter, raccoon, mink, herons, mergansers, grebes, bald eagles and osprey. The same method described above for humans can be used for wildlife to determine safe fish tissue concentrations. Reference doses for mammalian wildlife and birds and consumption rate and body weights for the species of concern were obtained from

published literature (USEPA, 1993b; USEPA, 1995a; USEPA, 1997c). The limited amount of data available on wildlife at Clear Lake was described in a preceding section.

Because of uncertainties in reference doses, consumption patterns of wildlife at Clear Lake, and whether wildlife at Clear Lake are being adversely impacted by mercury, Regional Board staff is not recommending separate fish tissue targets to protect wildlife. Instead of setting targets for wildlife, the effects on wildlife of meeting the proposed human health targets were examined. If mercury concentrations in fish eaten by wildlife were reduced by 40%, all species of concern except for river otter and kingfisher are expected to be protected. Using the recommended Clear Lake numeric targets and published literature values for consumption (not site-specific to Clear Lake; USEPA 1993), it is estimated that river otter and kingfisher would still exceed the safe daily intake levels of methylmercury for mammals and birds, respectively. To achieve safe intake levels for river otters, an additional 10% reduction from current fish tissue levels would be required. No information is available on health of river otters at Clear Lake. Methylmercury intake by kingfishers may be overestimated. Kingfishers likely eat the smallest fish available, which have less methylmercury than the average concentration in trophic level 2-3 fish used to calculate the intake for kingfishers. Because the Regional Board is committed to protecting all species at risk at Clear Lake, staff will evaluate any new information relative to wildlife risks prior to amending the Clear Lake TMDL targets into the Basin Plan.

3 SOURCE ANALYSIS

The Clear Lake watershed is located in the Coast Range of California, a region naturally enriched in mercury. Extensive mining of mercury in the Coast Range began in the early 1800s. Much of the mercury produced in the Coast Range was used to recover gold in the Sierra Nevada mountains during the Gold Rush period. Mining for mercury and gold has resulted in high levels of mercury in some streams and reservoirs in the Coast Range and Sierra Nevada mountains and the Sacramento River, and the Delta. Mercury mining exacerbated the amount of mercury entering some waterbodies due to erosion, weathering, and mass dumping of mercury-containing ores. Clear Lake contributes mercury downstream into Cache Creek, the Yolo Bypass and the Sacramento–San Joaquin River Delta (Foe and Croyle, 1998).

Sources of mercury entering Clear Lake include leaching and erosion from historic mining operations, geothermal vents and hot springs, urban and agricultural runoff, erosion of naturally mercury-enriched soils, and atmospheric deposition. One large mine, the Sulphur Bank Mercury Mine (SBMM), is located on the shore of Oaks Arm. Several smaller mercury mines or prospects were located in the Clear Lake watershed. All mercury mines in the area are now inactive. Tributaries to Clear Lake contribute mercury and methylmercury.

Sediment in the lake acts as a source for mercury to be methylated and as a sink as mercury is buried below the active sediment layer. Lakebed sediment consists of a surficial, active layer, in which mixing, resuspension and deposition occur and an inactive layer containing a reservoir of mercury. Loose sediment in the active layer of Clear Lake is easily resuspended and mixed by currents and sediment-dwelling invertebrates. Mercury is resuspended with these particles. The sediment active layer in Clear Lake is approximately 10-15 centimeters in depth (Suchanek et al., 1997). Maximal sulfate reduction and rates of methylation occur within the top four centimeters of sediment (Mack, 1998). Beneath the active layer, sediment containing mercury and methylmercury becomes buried. Unless the buried mercury is re-exposed by erosion of the overlying sediment, mercury in the inactive layer is considered to be removed from the mercury cycle. The active sediment layer is a site of bacterial methylation and demethylation and of cycling between different forms of mercury.

Mercury in the active layer, which is available for methylation, is the focus of the source assessment and load allocations for this TMDL. Methylmercury on sediment particles fluxes into the water column, where it may sorb to particulate matter, be ingested by biota, be demethylated or deposit to sediment (Figure 2). Flux of methylmercury into overlying water from the sediment is assumed to represent a net input of methylmercury to the water, taking into account methylation and demethylation in the sediment and in the water column.

A source analysis has been prepared for mercury and methylmercury in Clear Lake. The source analysis is presented as a mercury mass balance for the active layer of lakebed sediment. A map with features of the Clear Lake watershed is shown in Figure 1. Mercury loads entering Clear Lake were estimated for the following sources: groundwater and surface water from the Sulphur Bank Mercury Mine site, tributary and other surface water runoff into the lake, and atmospheric deposition, including atmospheric flux from SBMM. Outputs of mercury include flux to the atmosphere from the lake surface and flow downstream in Cache Creek. Sources and outputs are discussed below.

3.1 Mercury Inputs

3.1.1 Atmospheric Deposition on Lake Surface from the Regional and Global Mercury Pool

Inputs of mercury via atmospheric deposition are separated into two categories based on origin of mercury. Mercury that deposits from air may originate locally or beyond the boundaries of the Clear Lake basin (the regional and global atmospheric pool). Airborne mercury from local sources is treated in Section 3.1.3.4. Mercury from the global atmospheric pool is examined below.

In order to determine the total average annual deposition on the surface of Clear Lake of mercury from the global atmospheric pool, the following parameters were used: annual precipitation at Clear Lake, average concentration of mercury in precipitation (wet deposition concentration), area of Clear Lake, and amount of dry deposition. Atmospheric deposition is calculated only for the surface of Clear Lake. Mercury deposited atmospherically on the rest of the Clear Lake watershed and then transported into Clear Lake is accounted for in the estimates of tributary loads.

The following equation was then used to determine the total average annual deposition of mercury on Clear Lake:

$$Dt = (CwPyA)(1+Kd)$$

Where: A = area of Clear Lake in m^2
Cw = concentration of mercury in precipitation in ng/L
Dt = total annual mercury deposition to Clear Lake in ng/yr (then converted to kg/yr)
Kd = Coefficient used for dry deposition (ratio of dry deposition to wet deposition)
Py = Annual precipitation at Clear Lake in m

The area of Clear Lake is 1.77×10^8 m^2. The average annual precipitation at Clear Lake is 0.686 m (Richerson, 1994). Determining wet deposition concentration involved the use of a lower and upper limit estimate from two different sources due to the fact that no wet deposition is available for the Clear Lake area itself. The lower limit value of 3.9 ng/L is based on the average of data from 1998 and 1999 taken at the Covelo, California, Mercury Deposition Network station (Sweet, 2000). The upper value of 8.0 ng/L is based on an average concentration from three Bay Area collection sites where precipitation was collected from September 1, 1999 through August 31, 2000 (Tsai, 2001). Precipitation was collected in collection devices that open automatically during a precipitation event and close after moisture is no longer detected. After a precipitation event, the contents were taken and laboratory analyzed for mercury. Mercury concentration values at Covelo and the three Bay Area sites were based on volume-weighted concentration averages from the collection and analysis events. (Sweet, 2001; Tsai, 2001). As expected, mercury concentrations are higher in the San Francisco Bay area than at Covelo. Localized industrial sources in the Bay area add to the amount of mercury being deposited from the global atmospheric pool.

Directly measured dry deposition data is not available; therefore, as discussed by Tsai (2001) and Sweet (2001), dry deposition was estimated as a percentage of wet deposition. Due to uncertainty in these numbers, a large range of percentages was used to determine the overall lower and upper limit of total annual deposition. Dry deposition varies as a function of particulate matter in the air and mercury

concentrations on particulates. Values were calculated assuming dry deposition equivalent to 25% and 100 % of wet deposition (Table 3).

Table 3. Atmospheric Deposition of Mercury to Surface of Clear Lake

Wet Deposition Hg Concentration (ng/L) (a, b)		Average Precipitation (m/yr) (c)	Area of Clear Lake (m2)	Annual Wet Hg Deposition (kg/yr)
lower limit wet	3.9	0.686	1.77E+08	0.47
upper limit wet	8.0	0.686	1.77E+08	0.97

Annual wet Hg Deposition (kg/yr)		Dry Deposition Percent of Wet Deposition (d)	Total Annual Hg Deposition Wet + dry (kg/yr)
lower limit wet	0.47	25%	0.58
lower limit wet	0.47	100%	0.93
upper limit wet	0.97	25%	1.2
upper limit wet	0.97	100%	1.9

a) Lower limit of 3.85 ng/L is average wet deposition recorded by National Mercury Deposition Network at the Covelo, CA station (Sweet, Clyde. Personal Communication, 2001).
b) Upper limit of 8.0 ng/L is average wet deposition at three stations in San Francisco Bay area (Tsai, Pam. Personal Communication, 2001).
c) Richerson, P. J. et al., 1994. Page V-5
d) Lower bound (25%) and upper bound (100%) estimates based on values given by Tsai (2001) and Sweet (2001)

The estimates in Table 3 are similar to nation-wide estimates of mercury deposition prepared for the Mercury Study Report to Congress (MRC) (USEPA, 1997a). Authors of the MRC used the RELMAP (Regional Lagrangian Model of Air Pollution) model to predict the average annual atmospheric mercury concentration and the wet and dry deposition flux for each cell in a 40 km square grid over the continental United States. Emission, fate and transport of airborne mercury over the continental US was modeled using meteorological field data from 1989. Although the model incorporated over 10,000 mercury-emitting units such as municipal waste combustors and chlor-alkali plants, no data specific to mercury in air-borne dust or vapor from inactive mercury mine sites was available. Results of modeling with RELMAP were compared with measurements of mercury deposited and in air, available mainly for points in the upper Midwest, Florida and the Northeast (USEPA, 1997a). Mercury deposition estimates for western states developed using RELMAP were 0.86, 2.32, and 8.00 μg Hg/m^2/yr for the 10th, 50th and 90th percentiles of deposition, respectively. A deposition rate of 8.00 μg Hg/m^2/yr applied to the surface area of Clear Lake results in an estimated total annual deposition from the global and regional pool of 1.4 kg/year. This estimate does not include mercury from local sources, such as SBMM.

The mercury mass balance for Clear Lake (Table 11) shows a range of atmospheric deposition of 0.6 to 1.9 kg/year. The lower bound is the lowest estimate from Table 3 of atmospheric deposition directly on the lake surface from the global mercury pool. The upper bound is the highest estimate of deposition from the global pool.

3.1.2 Tributary Streams and Direct Surface Water Runoff

Mercury entering from tributaries likely originates in runoff from naturally mercury-enriched soils and mercury deposited in the watershed from the atmosphere. Geothermal vents and hot springs may contribute to tributary loads, particularly in Schindler Creek.

The California Department of Conservation of the Department of Mine Reclamation has identified several mercury mines or prospects, in addition to SBMM, that are in the Clear Lake watershed. Based upon limited mining activity at these sites, they are thought to contribute little mercury to Clear Lake. Field data should be collected to test this assumption. All mercury mines in the Clear Lake area are inactive. The Lucitta Mine is located on the south-east slope of Mount Konocti. The mine is visible from State Highway 29 as a white scar on the mountainside. Water flows from this side of Mt. Konocti into the Thurston Lake Basin, which is unconnected by surface water routes to Clear Lake (Personal communication, Steve Cannata, California Department of Fish and Game). The Thurston Basin may be connected to Clear Lake through groundwater flow. The Utopia Mine is located on a hillside above Clear Lake near Bartlett's Landing, just north of Lucerne. The Utopia Mine, which consisted of a single shaft, closed after two years of operation when the shaft flooded (Personal communications with Tom Smythe, Lake County Department of Public Works, and Ron Churchill, California Department of Conservation).

Mercury loads from tributary streams are calculated using estimated water flow rates and mercury concentrations in stream water. Data collection and calculations of water flow and mercury concentrations are described below.

3.1.2.1 Tributary Flow Rates and Water Budget

Tributary inflow, Cache creek outflow, and rainfall data were retrieved for water years 1990 to 1999 to calculate a water budget for Clear Lake. A water year begins 1 October of the previous year to 30 September of that year. The consecutive water years were chosen for data availability and because they include drought years as well as wet years. A water budget for water years 1990-1999 is shown in Appendix C.

Flow gauges are operated on three of the main tributaries to Clear Lake (Kelsey, Middle and Scott's Creeks) and on Cache Creek just downstream of the Cache Creek Dam. The dam is approximately three miles downstream of the outlet of Clear Lake. Flow gauges on Kelsey and Cache creeks are operated by USGS and data was accessed from the US Geological Survey (USGS) water homepage (http://water.wr.usgs.gov/). Department of Water Resources (DWR) operates flow gauges on Middle and Scott's Creeks as well as a rain gauge at Clear Lake Highlands. DWR's northern field office provided the flow data. Rainfall data was found on the California Data Exchange Center (CDEC) web site using monthly-accumulated rainfall.

The watersheds of Middle, Kelsey and Scott's creeks account for 43% of the entire Clear Lake Basin[3]. Runoff into the ungauged streams was assumed to be proportional to runoff in the gauged portion of the

[3] Watershed areas above the Kelsey, Middle and Scott's Creek gauges incorporate 36% of the surface-draining area of the Clear Lake basin. The surface-draining area of the Clear Watershed is 1,086 square km, excluding the surface area of the lake itself. An additional 98 square km of area near Mt. Konocti volcanics and Borax Lake drain to the sub-surface (Richerson et al., 1994). For calculations of TSS and water budgets, the area between the gauges and the lake was added for a total percentage

watershed. To determine total tributary inflow, yearly average flow from the three creeks was totaled and added to the estimated flow from the remaining 57% of the ungauged creeks for each of the water years.

Other components of the water budget include evaporation, groundwater, downstream flow, and municipal and agricultural extraction. Estimates of average annual evaporation from the lake surface (1.07 m/year), municipal and agricultural extraction (660,000 l/s) and average groundwater inflow to the lake (43,000 l/s) were obtained from the Clean Lakes Report (Richerson et al., 1994). Outputs were subtracted from inputs to obtain the change in lake storage for each of the ten water years.

3.1.2.2 Mercury Loads in Tributaries and Direct Surface Water Runoff

Very little mercury data exists for creeks within the Clear Lake watershed. Kelsey, Middle, Scott's, and Cache creeks were sampled five times for total mercury, methyl mercury, and total filtered mercury between 1998 and 2001 by staff of the UC Davis Clear Lake Environmental Research Center (Suchanek, 2001a). Samples were collected during low, medium and high flows and during one first flush event. Samples were analyzed for total suspended solids, total and filtered mercury and total and filtered methylmercury. Other creeks within the Clear Lake Basin were also sampled for mercury but flow data are not available for the corresponding water years. Regression analyses of the available data for Kelsey, Scott's and Middle Creeks were done to determine a correlation between flow and mercury concentration. Natural log regression produced the best R^2 values when compared to linear regression. Based on the regression equations for individual, gauged creeks, daily flow data from each of the creeks were then used to calculate mercury loading for the day. A simple mass balance equation (flow x concentration = mass) was used to determine mercury mass loading concentrations going into and out of Clear Lake. Daily loads were summed to estimate a mass loading (kg/yr) for each of the water years.

Mercury loads from the ungauged portion of the Clear Lake watershed were calculated in a manner similar to that used for the water budget. It was assumed that mercury load from a tributary is proportional to area of the drainage basin for that tributary. Mercury loads from the gauged streams were summed, then extrapolated to the remaining 57% of ungauged watershed area.

Mercury loads from Clear Lake tributaries are shown in Table 4. Loads are based on concentrations in unfiltered water. Depending upon volume of water flow, mercury loads to Clear Lake from the tributaries range from 1 to 60 kg/yr. The ten-year average load is 18 kg/year of mercury. Methylmercury loads from the tributaries ranged from 0.05 to 0.90 kg/yr. The ten-year average of methylmercury loading was 0.35 kg/yr.

These data should be refined with additional measurements of mercury concentrations at different seasons. Only fifteen data points were used to produce the regression lines of stream flow versus concentration (five sampling events x three gauged streams = 15 data points). The annual load value may overestimate the load coming from Scott's Creek, because mercury-laden sediment may deposit in the Tule Lake basin. Alternatively, these calculations could slightly underestimate actual average annual loads, because the calculation assumed that mercury concentrations in the gauged streams, which all flow

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for the portion of the Clear Lake watershed that is gauged of 43%. In making this estimation, it was assumed that the Tule Lake wetlands area, located downstream of the gauge, did not alter volume of water but did remove approximately 70% of suspended sediments from Scott's Creek.

into the Upper Arm, are the same for other streams. Schindler Creek, which drains into Oaks Arm, has relatively low flow but has higher concentrations of mercury.

Table 4. Mercury Loads from Tributaries into Clear Lake and Output Through Cache Creek

		Total Raw Mercury									
		Water Year									
		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Mass Loading	Kelsey Creek	0.228	0.726	0.423	1.970	0.178	5.307	1.477	2.336	3.593	1.597
Kg/Yr	Scott's Creek	0.134	0.331	0.335	1.450	0.192	2.830	1.408	1.629	3.035	1.107
	Middle Creek	0.225	0.747	1.205	5.750	0.212	9.824	5.021	2.835	17.749	4.955
	Total Gauged Streams 43.4%	0.586	1.803	1.963	9.169	0.582	17.960	7.906	6.799	24.377	7.659
	Total Ungauged Streams 56.6%	0.765	2.351	2.560	11.958	0.759	23.423	10.311	8.867	31.791	9.988
	Total	**1.351**	**4.154**	**4.522**	**21.128**	**1.342**	**41.383**	**18.218**	**15.667**	**56.168**	**17.647**
	Cache Creek Outflow	0.003	0.158	0.313	3.313	0.319	6.452	4.316	4.389	7.572	3.207
		Methyl Mercury									
		Water Year									
		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Mass Loading	Kelsey Creek	0.002	0.005	0.003	0.012	0.002	0.030	0.009	0.014	0.021	0.010
Kg/Yr	Scott's Creek	0.016	0.034	0.035	0.141	0.021	0.263	0.139	0.150	0.288	0.112
	Middle Creek	0.002	0.004	0.006	0.026	0.002	0.043	0.024	0.013	0.077	0.023
	Total Gauged Streams 43.4%	0.020	0.042	0.044	0.179	0.025	0.336	0.172	0.177	0.386	0.145
	Total Ungauged Streams 56.6%	0.026	0.055	0.058	0.234	0.032	0.438	0.224	0.231	0.503	0.189
	Total	**0.046**	**0.097**	**0.102**	**0.413**	**0.057**	**0.774**	**0.396**	**0.408**	**0.889**	**0.334**
	Cache Creek Outflow	0.000	0.007	0.015	0.163	0.015	0.322	0.213	0.218	0.379	0.158

3.1.3 Sulphur Bank Mercury Mine

Possible transport pathways of mercury entering Clear Lake from the mine site are:

- Mercury in surface water and groundwater flowing westward into the lake from the waste rock dam (WRD) area.
- Mercury in surface water and groundwater flowing through wetlands north of the site.
- Mercury fluxing to air and being deposited on the lake surface or locally to be carried in runoff.
- Mercury previously deposited in the lake that is remobilized into the active sediment layer.

Although mercury in Clear Lake derives from several sources, the Sulphur Bank Mercury Mine (SBMM) is clearly the largest historical source of mercury now residing in lakebed sediments (Chamberlin et al., 1990; Suchanek et al., 1997). Inactive since 1957, the SBMM was one of the largest mercury production sites in California and has been described as one of the most productive hot spring mineral deposits in the world (USEPA, 1994). Over 1.2 million tons of material were estimated to have been removed, processed, and disposed during nearly a century of mining activity. Surface sulfur deposits were mined between in 1865 and 1871 (USEPA, 1994). Shaft mining operations for cinnabar began between 1872 and 1883 when the Herman Shaft was sunk to approximately 450 feet below the ground surface. Six more shafts were sunk before underground operations were completely abandoned in 1944. Open pit mining techniques were utilized from 1927 to 1957. After 1957, the mine pit filled with water, forming the existing Herman Impoundment.

Sediment core data for Clear Lake show an elevation in mercury beginning near to the time that operations began at SBMM. Sediment mercury levels rose sharply (five to ten times above pre-mining period) after approximately 1927 (Suchanek et al., 1997). Open-pit mining methods utilized heavy earth-moving equipment, which greatly increased erosion of mining materials. During mining operations, excavated overburden and tailings from on-site ore processing were disposed in piles on the mine site, along the Clear Lake shoreline, and bulldozed directly into Clear Lake (USEPA, 1994). Due to releases related to mercury mining at Sulphur Bank between 1872 and 1957, an estimated 100 metric tons of mercury reside in the aquatic ecosystem of Clear Lake (Chamberlin et al., 1990).

Mercury from SBMM continues to enter Clear Lake through groundwater, surface erosion, and possibly atmospheric routes (Suchanek et al., 1997; Tetra Tech EMI, 2001). The USEPA declared the SBMM a federal Superfund site in 1991 (USEPA, 1994). Two remediation projects have been completed since then: regrading and vegetation of mining waste piles along the shoreline in 1992 that decreased mass erosion into the lake, and construction of a surface water runoff diversion system in 1999. The USEPA is currently conducting a remedial investigation to fully characterize the SBMM site and propose a final remedy. The USEPA Superfund Program expects to release the SBMM Remedial Investigation/Feasibility Study Report in 2002.

Ongoing inputs of mercury from SBMM through the WRD, north wetlands and air are discussed in sections below. In cases of the wetlands and atmospheric deposition, little information is available on mercury loads moving through these routes. The USEPA Superfund Program is continuing to investigate the mine site and wetlands. As more information becomes available, sections below will be updated.

Remobilization of previously deposited mercury from runoff or mining operations at SBMM is a possible source of mercury in the active layer of sediment. Waste material and tailings were bulldozed directly into the lake during open pit operations, such that the contours of the shoreline were changed (Chamberlin et al., 1990). Mercury-containing material continued to erode from steeply sloped shoreline waste piles until USEPA remediated the piles in 1992. In samples collected in 1994-96 at site OA-01 (0.3 km from SBMM), mercury concentrations in the sediment averaged 238 μg/g with a range of 42–425 μg/g (Suchanek et al., 1997). The extent of remobilization of this material is unknown. Surficial sediments collected offshore of the mine site contained more silt and sand and fewer clay particles than surficial sediments from other parts of the lake (Suchanek et al., 1993). These data suggest that fine particles may accumulate directly offshore of SBMM at a lesser rate than in other Arms. The contribution of the historically deposited mercury to surficial loads should be examined. While most mercury from SBMM that was deposited in the past was likely buried under incoming sediment, some historically-deposited mercury may still be contributing to the upper sediment layer.

The total of inputs from SBMM to the active sediment layer includes ongoing releases, as described above, and contributions of previously deposited mercury. SBMM-related mercury from these various sources will require different remediation activities and may have different degrees of bioavailability. These inputs were combined, however, for the purpose of determining total inputs from SBMM. As USEPA completes its investigations of mercury from atmospheric fluxes, surface water runoff, the north wetlands and contaminated lakebed sediments, these input estimates will be adjusted.

Summary of SBMM Inputs: Estimation of the amount of mercury contributed to the active sediment layer from past and ongoing processes at SBMM is complex. The USEPA and Tetra Tech EMI estimate of 2 kg/year (based on WRD well samples) is taken by Regional Board staff as the lower bound of mercury inputs from SBMM (Tetra Tech EMI, 2001; USEPA, 2001c). This estimate is based on analysis of samples from wells in the WRD and is the USEPA estimate of mercury discharged only from the WRD. The upper bound on inputs of 695 kg/year is based on the amount of mercury estimated to be in surficial sediment, minus loads from inputs other than SBMM.

3.1.3.1 Sulphur Bank Mercury Mine Site Description

The Sulphur Bank Mercury Mine site covers approximately one square mile. It abuts the Eastern Shore of the Oaks Arm of Clear Lake. The site contains approximately 120 acres of exposed mine overburden and tailings (hereafter referred to as waste rock). Two small, unprocessed ore piles are also on the site (USEPA, 1994). Mercury in samples of mine materials ranged from 50 to 4,000 ppm (USEPA, 2001c). Ores that contained 1,000 ppm or more of mercury could be economically processed during mine operations. As expected, samples from the ore stockpiles have the highest median concentrations of mercury. All piles of mine materials exhibit the potential to generate acid rock drainage (USEPA, 2001c).

The abandoned mine pit, called Herman Impoundment, is filled with acidic water (approximately pH 3) to a depth of 90 feet and has a surface area of about 20 acres. The average mercury concentrations in Herman Impoundment water and sediment are around 0.8 μg/L and 26 μg/g, respectively (Columbia Geoscience, 1988; USEPA, 1994).

A geothermal vent located at the bottom of Herman Impoundment continues to discharge gases, minerals, including mercury, and fluids into the pit (White and Roberson, 1962). Gas bubbles can be seen in Herman Impoundment and in other basins of water on the site. Mercury in the sediment of Herman Impoundment comes from the geothermal vents and in acid rock drainage that flows through waste rock on the north, east and southern sides of the pit into the pit (Tetra Tech EMI, 2001). Mercury in geothermal fluids is thought to precipitate upon coming in contact with the acidic pit water (Columbia Geoscience, 1988). Mining operations changed groundwater levels and the zone of transition between oxidizing and reducing environments. Acid generation and subsequent leaching of minerals in water entering and leaving the Herman Impoundment increased after mining began (Suchanek et al., 2001a; (White et al., 1973).

A large pile of waste rock, known as the waste rock dam (WRD), stretches about 2000 feet along the shore of Clear Lake. Although the WRD separates Herman Impoundment from Clear Lake, it does not control the flow of groundwater from the pit into the lake. The waste rock was deposited over the hydrothermal spring area that was originally mined for sulfur. Material in the WRD ranges in size from clay and silt to large boulders. Sizes, origin and extent of decomposition of material in the WRD are extremely variable, both vertically and horizontally (Tetra Tech EMI, 2000).

A wetland area lies directly to the north of SBMM. The wetland was created around 1915, when two levees were built in Clear Lake to create pasture. Historical records indicate that during mining between 1955 and 1957, water from Herman Impoundment was pumped into an abandoned, smaller pit. Overflow from the smaller pit apparently flowed into the wetlands (Columbia Geoscience, 1988). Surface runoff from the north waste rock piles is directed through culverts into the north wetland. Currently, the wetland is used for cattle grazing and as a source of fish, tules, and other resources utilized by Members of the Elem Pomo Tribe. One mine waste rock pile extends into the wetlands.

3.1.3.2 Waste Rock Dam

The WRD is a source and transportation pathway of mercury entering Clear Lake (Chamberlin et al., 1990; Suchanek et al., 1997; Tetra Tech EMI, 2001; USEPA, 1994). Mercury may be released from the WRD in surface water and groundwater. Surface and groundwater transportation routes are treated separately in the following paragraphs.

3.1.3.2.1 Mercury in Surface Water from the Waste Rock Dam

Surface water flowing over the WRD was, at one time, a significant source of mercury entering Clear Lake. Steep, unvegetated sides of the WRD were in direct contact with the lake and were highly susceptible to erosion. In a report to the RWQCB, Chamberlin and others (1990) estimated that sheetwash erosion and slope failures contributed at least 100 kg mercury per year to Oaks Arm. Their actual estimate of 132 kg/yr was made in a dry year. In heavy rainfall years, transport of mercury in eroded soil would likely have been much higher. In 1992, USEPA undertook emergency action to stabilize the WRD to reduce erosion of mine wastes into Clear Lake. The WRD was regraded, capped and vegetated. Rip rap was placed at the foot of the WRD. This remedial action appears to have significantly reduced erosion of WRD material into the lake (ICF Technology, 1995 in: (Harding Lawson Associates, 1999). Since the remediation, surface runoff into the lake from the western side of the mine site has not been measured.

3.1.3.2.2 Mercury in Groundwater in the Waste Rock Dam

Groundwater remains a source of mercury transported into Clear Lake from the WRD. Estimation of mercury fluxing out of the waste rock dam is complex. The USEPA and Tetra Tech EMI estimate of 2 kg/year (based on WRD well samples) is taken by Regional Board staff as the lower bound of mercury flux from SBMM (Tetra Tech EMI, 2001; USEPA, 2001c). Regional Board staff believes, because of surficial sediment concentrations and uncertainties in methods and timing of well sample collections, that inputs from the WRD may be much higher. WRD loads and uncertainties are explained in the following sections.

Beginning in 1999, USEPA and the US Army Corps of Engineers contracted with Tetra Tech EMI to conduct an extensive hydrogeologic investigation of SBMM. One purpose of the study was to determine groundwater flow rates and directions in various water-bearing geologic units. The conceptual site model developed by Tetra Tech EMI for water sampling at SBMM proposes three origins of mercury fluxing through the WRD (Tetra Tech EMI, 2001):

- Mercury in Herman Impoundment water flowing through the WRD
- Discharge from the hydrothermal vent beneath the WRD
- Mobilization of mercury in waste rock by the interaction of rock with shallow groundwater flowing through the WRD.

Mercury loads in groundwater are calculated as groundwater flow rate times the concentration of mercury in groundwater.

3.1.3.2.3 Rates of Groundwater Flow through the Waste Rock Dam

Investigations of groundwater flow at SBMM were conducted for Bradley Mining Company (Columbia Geoscience, 1988) and USEPA (ICF Technology, 1994 in: (Tetra Tech EMI, 2001). These investigations are in agreement regarding groundwater flow directions. Groundwater flows towards Herman Impoundment from the north, east and south. Groundwater flows westward from the west side of Herman Impoundment toward Clear Lake. The surface of Herman Impoundment is above the surface of Clear Lake, with the difference being about 10-14 feet. This difference in surface elevation contributes to a hydraulic gradient in the direction of Clear Lake from the pit. Groundwater in the WRD is approximately one third hydrothermal water (mainly from Herman Impoundment) and two-thirds meteoric water (rainfall and other precipitation) that infiltrates into the WRD (Tetra Tech EMI, 2001).

Through analysis of well borings, Tetra Tech EMI identified five distinct geologic layers or units below the WRD. In descending order from the surface, the geologic layers are: waste rock; upper deposit of lakebed sediments; andesite (lava flow); lower deposit of lakebed sediments; and the Franciscan geologic formation (Tetra Tech EMI, 2001). Because of similarities in conductivity and geology, Tetra Tech EMI analysts treated the waste rock and upper lake sediments as one geologic unit for purposes of estimating groundwater flow. As mentioned above, the 2000 Hydrogeologic Study by Tetra Tech EMI confirmed the groundwater flow pathway from Herman Impoundment toward Clear Lake. The upper portion of the WRD area (waste rock and upper historical deposit of lakebed sediments) is by far the most permeable unit to groundwater flow, followed by the andesite and lower lake sediments. Except near fractures, very little groundwater has been found in the Franciscan Formation.

In order to estimate groundwater flow, Tetra Tech EMI conducted several types of aquifer pumping tests in wells screened in the five geologic units and located throughout the site. Twelve wells in the WRD were tested on two dates (9 February and 25 April, 2000). The resulting groundwater flow estimates ranged from 7 to 81 gpm (Tetra Tech EMI, 2001; USEPA, 2001c). The variation was due to maximum and minimum values determined for hydraulic conductivity and hydraulic gradients[4].

Maximum and minimum groundwater flow rates determined by Tetra Tech EMI in 2001 can be compared with previous groundwater flow estimates. These estimates are shown in Table 5. ICF Kaiser has also conducted aquifer pumping tests. Other estimates have been made using the inflow to Herman Impoundment or change in surface water levels. These alternative methods of estimating flow can serve as "reality checks" for the aquifer pumping test estimates.

As shown in Table 5, estimates of groundwater flow rates have varied widely. Groundwater flow estimates based on pit dewatering, pit filling, surface level drop, and chemical composition of waters are all around 100 gpm. In comparison with Herman Impoundment inflow and water budget estimates, the flow rate estimates based upon data from well pumping tests are much lower. Regional Board staff considers flow estimates based on tracer studies to be highly uncertain. Tracers are generally used to obtain qualitative information about groundwater location and direction of flow, rather than quantitative estimates of flow rates.

Table 5. Estimates of Groundwater Flow Rates into Herman Pit and Through the WRD

Reference	Flow Rate (gpm)	Type of Measurement & Conditions
(RWQCB, 1956 in: Tetra Tech EMI, 2001)	139	dewatering of Herman Impoundment during mining, 1956
(White and Roberson, 1962)	95	filling of Herman Impoundment after mining stopped, 1947-1954
(Suchanek et al., 1997)	109	Calculated from the net change in Herman Impoundment surface level (rainfall minus evaporation) plus hydrothermal spring flow (as estimated by White and Roberson).
(Goff and Bergfeld, 1997 in: Tetra Tech EMI, 2001)	100	preliminary estimate of seepage through WRD, based on chemical composition of water in the pit, WRD and Clear Lake adjacent to the WRD.
(ICF Kaiser, 1999 in: Tetra Tech EMI, 2001)	15	average flow rate through WRD based on aquifer pumping tests
(Tetra Tech EMI, 2001)		estimated flow rates based on aquifer pumping tests:
	7-9.2	uses minimum hydraulic conductivity and low hydraulic gradient estimate
	27-37	uses maximum hydraulic conductivity and low hydraulic gradient estimate
	18.6-20.7	uses minimum hydraulic conductivity and high hydraulic gradient estimate
	73-81.2	uses maximum hydraulic conductivity and high hydraulic gradient estimate
(Oton et al., 1998)	6,000-8,000	Based on disappearance of inert tracer from the pit and appearance in monitoring wells

[4] Groundwater flow rate was calculated as:

(average hydraulic conductivity) * (average hydraulic gradient) * (distance across the aquifer) = flow rate

There are several reasons why groundwater flow estimates from well pumping tests may be biased low. The first possibility that the Tetra Tech EMI and ICF Kaiser estimates are low is that these estimates assume uniform hydraulic conductivity throughout each geologic unit. Sizes of material in the WRD are not uniform, however. Small channels between boulders or gravel in the WRD could be preferential pathways for groundwater flow. Heterogeneous flow rates are expected as water flows through heterogeneous WRD material. Average flow rates may adequately characterize heterogeneous flow, provided that sufficient measurements are taken to obtain a true average conductivity for each unit.

The presence of large channels in the WRD is unlikely. Airborne and ground-based geophysical remote sensing surveys conducted at SBMM by the US Department of Energy suggest that large flow channels do not exist in the WRD (Hammack et al., 2000). The geophysical surveys conducted in 2000 found no large conductivity anomalies within the waste rock and upper lakebed sediments of the WRD. There may be small channels or fracture zones that extend through all or a portion of the WRD, which were not detected by geophysical sensing.

The second possible reason that Tetra Tech EMI estimates of groundwater flow rates are lower than estimates of other types is that their reported hydraulic conductivities may not represent long-term averages, for either mean or maximal conditions. Precipitation in Water Years 2000 and 2001 was near normal. Rainwater infiltration and subsequent groundwater flow through the WRD would likely be greater during a high water year.

Based on estimates in Table 5, average groundwater flow rates from the WRD range from 10 to more than 100 gpm. Regional Board staff believes that a conservative estimate is between 70 and 100 gpm, based on seasonal considerations and comparison of pumping tests with historical evaluations.

3.1.3.2.4 Mercury Concentrations in Waste Rock Dam Groundwater

Under typical Coast Range mercury mine conditions, mercury concentrations increase substantially as mine drainage flows through and reacts with tailings and waste rock (Rytuba, 2000). Water in contact with exposed, mineralized rock forms acid rock drainage. Mercury concentrations in groundwater increase as acidic drainage flows through and reacts with tailings and waste rock. The groundwater flowing into and out of Herman Impoundment is heavily contaminated with acid rock drainage (Suchanek et al., 2001a).

Concentrations of mercury in groundwater samples from the WRD have been measured several times (ICF Technology, 1994; Tetra Tech EMI, 2001; USEPA, 1994). The most extensive sampling was conducted by Tetra Tech EMI in May and June 2000. Groundwater concentrations of mercury for the different geologic units are shown in Table 6. Most samples were filtered in order to analyze for dissolved mercury. In some cases, the concentrations of mercury in unfiltered samples are much higher than dissolved concentrations from the same well. The validity of the unfiltered mercury measurements is uncertain, due to the possibility that the well sampling process disturbed soil that would not normally move through the aquifer. Regional Board staff hydrogeologists estimate that there is probably little mercury moving through the waste rock dam on particulates (except in possible small channels between rocks) and that the dissolved mercury concentrations serve as an adequate measures of total mercury concentrations in groundwater at this point.

Table 6. Mercury in Filtered and Unfiltered Water Samples in the SBMM Waste Rock Dam

			USEPA, 1994	ICF Kaiser, 1999 in: Tetra Tech EMI, 2001	Tetra Tech EMI, 2001 (b)	USEPA, 1994	ICF Kaiser, 1999 in: Tetra Tech EMI, 2001
ground-water contour (a)	Monitoring Well	Geologic Unit	Mercury in filtered samples (ug/l)	Mercury in filtered samples (ug/l)	Mercury in filtered samples (ug/l)	Mercury in unfiltered samples (ug/l)	Mercury in unfiltered samples (ug/l)
1	MW-11	Waste Rock		0.56	0.38		22.4
1	MW-3S	Upper Lake Seds.			22.4		
1	MW-2	Upper Lake Seds.	21	12.8	45.6	462	
1	MW-13	Waste Rock		49.2	62.4		619
1	MW-5	Waste Rock	123	96.7	164	1040	
1	HP14	Upper Lake Seds.		251	350		4920
1	MW-4	Upper Lake Seds.	0.74	0.34	0.23	4650	
1	MW-3*	Upper Lake Seds.	0.31	0.02J		297	NA
2	MW-6	Waste Rock	1.4	2.7/2.4	5.2	3.4	
2	MW-14	Waste Rock		35.5/38	59.2		6440/6360
3	MW-7	Waste Rock	14.6	2	6.1	8410	
4	SB-8S	Upper Lake Seds.			80		
4	MW-9	Waste Rock	32.1	<0.01J	0.3 U	41.8	
5	MW-16S	Waste Rock			9.8		
5	MW-10	Upper Lake Seds.		9.4	63.1		38.3
5	SB-8D			<0.01 J	0.2 U		NA
5	HP10 (.75)	Waste Rock		6.1	5.6		16,200
5	HP15	Upper Lake Seds.		<0.21 J	0.12 J		48,300
5	MW-1	Upper Lake Seds.	0.13	<0.01 J	0.051 J	6.2	
	MW-11I	Andesite			0.068 J		
	MW-16I	Andesite			0.2 U		
	SB-8I	Andesite			0.096 J		
	MW-15NWS	Lower Lake Seds.			0.2 U		
	MW-11D	Lower Lake Seds.			3.8 J		
	MW-16D	Lower Lake Seds.			0.2 U		
	MW-3D	Lower Lake Seds.			0.2 U		
	MW-15D	Franciscan Complex			0.2 U		
	MW-31D	Franciscan Complex			0.2 U		

a) Groundwater contours of waste rock dam/upper lake sediments from Tetra Tech EMI, 2001. Numbers added by Regional Board staff. Contour 1 is closest to the Clear Lake shoreline.

b) Laboratory analysis codes. U = undetected at the stated value. J = stated value is estimated.

Groundwater data collected in December 2000 through May 2001 is consistent with the hypothesis that overall, particulate transport of mercury is not a significant component of mercury flux through the WRD. The USEPA Superfund Program and Tetra Tech EMI collected groundwater samples monthly from six wells, three of which were in the WRD, to evaluate the impacts of particulate transport on mercury flux off site from SBMM (USEPA, 2001c). Samples were collected of unfiltered groundwater and groundwater that was passed through a series of filters (25 microns down to 0.45 microns). Generally, concentrations of mercury in filtered samples were equivalent to or slightly less than concentrations in unfiltered samples, which indicated that most mercury was in a dissolved form.

Groundwater samples with the highest concentrations of dissolved mercury have been collected from the waste rock and upper lakebed sediments (Tetra Tech EMI, 2001; USEPA, 1994). The waste rock/upper lakebed sediment unit was also the most permeable to groundwater. Therefore, it is assumed that the largest amount of mercury is transported in this unit. Concentrations of dissolved mercury measured in wells in the waste rock/upper lake sediment unit vary over three orders of magnitude (Tetra Tech EMI, 2001). Wells that were sampled more than once tended to show similar mercury concentrations when sampled in other studies or over multiple sample dates. Table 6 shows wells in the waste rock/upper lake sediments, ordered by the groundwater contour in which each well is located. Groundwater contours were identified by Tetra Tech EMI (2001) and labeled by Regional Board staff.

As shown in Table 6, wells having the highest dissolved mercury concentrations are located in groundwater contours that are closest to the lake. Mercury is thought to accumulate in groundwater as water moves through the waste rock dam. As shown by low concentrations of dissolved mercury in some wells close to the lake, this process of accumulation does not occur uniformly across the WRD. Tetra Tech EMI found that waters with relatively high concentrations of dissolved mercury (> 10.0 micrograms/L) consistently had low pH and high redox potential (Eh, measured as millivolts) (Tetra Tech EMI, 2001). Under low pH and high Eh conditions, mercury can form highly soluble complexes with chloride, which are readily taken up by methylating bacteria (Morel, 1998). Factors controlling redox potential in the WRD will likely be the focus of further investigations by the USEPA Superfund Program.

USEPA and Tetra Tech EMI have estimated that the average mercury flux in groundwater from the WRD is 1-3 kg/year (Tetra Tech EMI, 2001; USEPA 2001c). This estimate is based on groundwater pumping test data and mercury concentrations in well samples collected by Tetra Tech EMI. USEPA used minimal average groundwater flow rates and the average mercury concentration in all wells in the WRD. An estimate of mercury flux made by Regional Board staff using the Tetra Tech EMI and Superfund Program data is shown in Appendix G. No analyses within Clear Lake, such as of sediment porewater, have been made that could be compared with estimates of flux from the WRD well samples.

3.1.3.2.5 Uncertainties in Mercury Inputs Based on WRD Well Samples

Regional Board staff believes that the concentrations of dissolved mercury in groundwater flowing through the waste rock dam are underestimated by the USEPA Superfund program. Two lines of reasoning support this assertion. First, mercury concentrations may not have been measured at the peak season of the year. Mercury concentrations in well samples increase with groundwater level in the WRD. In WRD wells sampled in 2000, mercury concentrations were higher in samples collected in May/June than in December, which corresponds to an increase in groundwater levels from winter rains (USEPA, 2001c). Data from two wells in the WRD that were sampled monthly from December through April showed a consistent increase in mercury concentrations as the winter progressed. Mercury samples have not been collected for all wells when groundwater levels are expected to be at their maximum. When wells were sampled in May and June 2000, groundwater levels had already declined from the peak in March and April (USEPA, 2001c).

The second reason that mercury concentrations in the WRD may be underestimated is that data is not yet available for a year of extensive rainfall. As mentioned above, the annual precipitation levels for Water

Years 2000 and 2001 was average. To date, well sampling for mercury has not been conducted in a period of significant rainfall and infiltration into the WRD. In addition to the increased volume of water available for transporting mercury, oxygenated rainwater infiltrating into the WRD likely contributes to increased redox potential of the groundwater. WRD wells with the highest concentrations of dissolved mercury also exhibited high redox potential (Tetra Tech EMI, 2001). Because redox potential varies widely across the WRD, the amount of rain water infiltrating the WRD is not the only influence on redox potential. USEPA has indicated a need to better understand factors controlling redox potential.

A third possible reason that mercury loads from the WRD are underestimated is that groundwater may continue to pick up mercury as it moves through lakebed sediments into the lake. By examining porewater in short sediment cores collected near the mine site, UC Davis researchers have located "hotspots" of contaminated water upwelling into the lake. Porewater at the hotspots has low pH and contains high sulfate and other chemical constituents characteristic of acidic drainage from the mine site (Shipp, 2001). Sediments near the mine contain waste rock that was directly pushed into the lake during mining operations. The acidic groundwater may accumulate additional mercury as it moves through the lakebed mine wastes. Concentrations of mercury in groundwater at the end of its pathway, prior to mixing with lake water, have not been measured.

3.1.3.3 North Wetlands

Data needed to quantify mercury releases from the north wetlands are currently lacking. USEPA is continuing to investigate the levels and impacts of mercury in the wetlands. The methylation efficiency index (ratio of methylmercury to total mercury in the sediment) for summer months is high in the north wetlands, relative to open water areas of the lake (See methylation efficiency section in the Linkage Analysis). The amount of methylmercury transported from the wetlands to the lake is uncertain. Methylmercury produced in the wetlands may impact biota that feed on aquatic organisms in the wetland.

3.1.3.3.1 Mercury in groundwater from the wetlands

Groundwater contours developed by Tetra Tech EMI showed that from the peak of the northern waste rock piles, groundwater flows to the north toward the wetlands (Tetra Tech EMI, 2001). Qualitatively, it appears that the mercury load from SBMM to the wetland area via groundwater is of low significance. This conclusion is based on the following evidence:

e) From geologic data cross sections provided in the Tetra Tech EMI 2001 report, groundwater from SBMM to the wetlands area would flow mostly through the andesite unit, which has low hydraulic conductivity (1.63×10^{-5} ft/s to 7.51×10^{-5} ft/s; Tetra Tech EMI, 2001). Therefore, there would be low flow to the wetland area from groundwater.
f) The groundwater to the wetland area does not primarily come from Herman Pit or from the shear zone in which Herman Pit was excavated. This would mean that water quality is likely of higher quality than groundwater related to the Herman Pit and shear zone. The groundwater that flows towards the wetlands area comes from an east/west trending topographic mound north of Herman Pit that divides water discharges northward to the wetlands and southward to the Herman Pit.
g) Concentrations measured in wells up-gradient of the wetlands area show concentrations of dissolved mercury below the practical quantitative limit of 0.2 μg/L. Water samples from these wells also had low pH and high redox potential (Tetra Tech EMI, 2001). Although the well samples had little

mercury, the acidity and redox potential suggest that groundwater moving into the wetlands could release mercury from surrounding rocks.

USEPA is continuing to investigate the levels and impacts of mercury in the wetlands.

3.1.3.3.2 Mercury in surface water from the wetlands

Estimates are lacking of the quantity of mercury transported in surface water to the wetland area from SBMM. Surface water does run off of unvegetated waste rock piles into the wetland. In 1990, rock and geofabric barriers were installed at the mouths of culverts carrying surface water. The barriers were intended to trap suspended solids in the water. Effectiveness of the barriers is unknown.

3.1.3.4 Local Deposition of Mercury Fluxed to the Atmosphere

There are no major industrial sources in the Clear Lake watershed that emit mercury to the atmosphere, but mercury may be emitted from mine waste or disturbed rock that is naturally enriched in mercury. Based on measurements of mercury fluxing from soil at 22 locations on the SBMM site, Gustin and colleagues estimated an annual flux of 6.5 kg mercury from the mine site (Gustin et al., 2000). The flux estimates were of mercury emitted from the soil. Comparable estimates of the amount of emitted mercury that redeposits locally have not been made. Mercury fluxing from the soil may be in the form of elemental mercury, which is relatively stable and can travel long distances in air, or reactive gaseous mercury, which is more likely to be deposited soon after emission. Under direction of the Superfund Program, researchers from the University of Nevada at Reno will collect additional flux measurements that will differentiate between total and reactive gaseous mercury. Estimates of local deposition of mercury will be incorporated into the TMDL and Basin Planning elements as they become available.

3.1.3.5 Mercury from SBMM Based on Lakebed Sediment Concentrations

Mercury loading to the lake can be estimated using sediment concentrations and sediment accumulation rates, which is an entirely different method than using estimations of groundwater and surface water flow rates. The UC Davis Clear Lake Environmental Research Center collected deep cores of lakebed sediment in 1996 (Suchanek et al., 1997) and 2000 (Suchanek et al., unpublished data collected for the Regional Board). The cores were analyzed for mercury, methylmercury and other analytes and dated using lead isotope levels. Plots of core data from 1996 and 2000 are presented in Appendix D.

Annual deposition of mercury in Clear Lake sediment can be estimated as the product of annual volume of sediment deposited and the concentration of mercury in the top layer of sediment. This calculation is shown in Section 3.3.1. Because top sediments are mixed by waves and sediment-dwelling invertebrates, mercury loads are estimated as the average deposited in the past six to seven years. UC Davis researchers estimate that a significant decline in mercury loading would be clearly visible in the sediment record eight to ten years in the future (Suchanek et al., 1997; Suchanek et al., 2001a). Concentrations of mercury in the sediment measured in 2000 suggest that substantial loading from the mine is still occurring, despite the remediation of lakeshore waste rock piles in 1992. Calculation of ongoing, annual deposition of mercury based on surface sediment concentrations is explained in detail in the section on mercury outputs. The lake sediment calculations indicate that over 700 kg mercury is being deposited to lake sediments annually, with approximately 375 kg/year deposited in Oaks Arm. In contrast, tributary stream loading to Oaks arm is estimated to be less than 2 kg/year (See section on tributary loading below).

These loads could come from ongoing inputs from the mine site or remobilization of mercury previously deposited due to mine-related processes.

There are uncertainties in this estimate of mercury inputs to the lake, namely, that average sediment concentrations and sedimentation rates were applied across the surface areas of each Arm. Although the sedimentation rates and mercury concentrations were consistent between cores collected in different years, all cores were collected from sites near the middle of each arm. Sedimentation rates and mercury concentrations undoubtedly vary across each arm. Despite uncertainties, however, the sediment calculation produces an estimate of mercury loading that is two orders of magnitude higher than the calculations using groundwater flow estimates from the waste rock dam.

No other sources of mercury, such as lakebed springs, geothermal vents, or lava tubes that discharge directly into the lake, are able to account for the difference in loading to Oaks Arm. In 1992, UC Davis collected surficial sediment samples at 35 sites in the lake, including 8 sites near geothermal springs (Suchanek et al., 1993). The study was designed to test whether the geothermal springs contributed significant amounts of mercury to Clear Lake. Surficial sediment samples collected near lakebed geothermal spring did not show any elevation in total mercury concentrations, relative to sites apart from geothermal springs.

Based on the lakebed sediment calculations, the mercury mass balance in Table 11 shows an upper bound of mercury loads coming from the SBMM of 695 kg/year. This is the difference between the total amount of mercury estimated to be deposited to surficial sediment annually, and the annual loads from other inputs. Deep cores indicate that sediment mercury concentrations have gradually declined over the past 15 to 40 years, depending on Arm and core sample. Average rates of decline from the 1996 and 2000 cores were less than 0.1, 0.1 and 1 ppm mercury per year for Upper, Lower and Oaks Arms, respectively (See graphs of core data). Even taking into account the rates of decline, concentrations of mercury in the sediment are still higher than expected from estimates of loading from SBMM that are based on groundwater well tests. Regional Board staff expects that the USEPA Superfund Program will work to clarify the differences between methods of calculating loads of mercury from the mine site prior to completion their investigation at the mine site.

3.2 Mercury Outputs

3.2.1 Cache Creek Outflow

A flow gage is located just downstream of the Cache Creek Dam, approximately four miles downstream of the outflow point of Clear Lake. This is the closest gauge to the outflow of Clear Lake. Flows and mercury concentrations obtained just below Cache Creek Dam provide the best available estimates of mercury loads leaving Clear Lake through Cache Creek.

Loads of mercury exported down Cache Creek were estimated using the same method that was used for tributary inputs. Mercury concentrations in Cache Creek were measured by UC Davis CLERC during five different flow regimes. Natural log regression analyses were used to determine the relationship between flow and mercury concentration. Based on the regression equations for Cache Creek flow, daily flow data were then used to calculate daily mercury loads. Daily loads were summed to estimate a mass

loading (kg/yr) for each of the water years. Cache Creek gauge data were obtained from USGS for water years 1990 to 1999. Dam operation interrupts the flow pattern, sometimes causing wide ranges of flow rates during one month or season. For this reason, it was important to calculate daily loads rather than average monthly loads.

Mercury loads carried through the Cache Creek Dam are shown in Tables 4 and 11. The range of mercury outputs calculated using the UC Davis CLERC data was less than 0.1 to 7 kg/year. The ten-year average from this data set was 3 kg/year. These estimates were compared with loads calculated from a separate set of concentration measurements, collected by Regional Board staff between February, 1996 and July, 1997 (Foe and Croyle, 1998). Using data from Foe and Croyle, the annual load of mercury in Cache Creek at the dam was estimated to be about 11 kg/year. This TMDL report, therefore, uses a range of mercury loads transported through the Clear Lake output as in Cache Creek of 0.1 – 11 kg/year. Loss of methylmercury through Cache Creek ranges from less than 0.001 to 0.4 kg/year, with a ten-year average of 0.15 kg/yr.

3.2.2 Flux to the Atmosphere

Loss of mercury by volatilization from the water column to the atmosphere was estimated by Bale in a model of the fate of aquatic mercury in Clear Lake (Bale, 2000). Mercury in its elemental form (Hg^0) is able to volatilize to the atmosphere. Rate of loss depends upon temperature, concentration of elemental mercury in the water column and the background atmospheric concentration of mercury. The calculation assumed an average temperature of 20 degrees Celsius, background atmospheric mercury concentration of 0.002 ng/L, and concentrations of mercury in surface water sampled between May 1994 and March, 1998 (Suchanek et al., 1997; personal communication from T. Suchanek to J. Cooke regarding 1998 data). Estimated total loss of mercury to the atmosphere from all arms of Clear Lake was 1.6 kg/year (Bale, 2000).

3.2.3 Water Diversions

Water for municipal and agricultural uses is extracted from Clear Lake at an average rate of 20.8×10^6 m^3/year (Richerson et al., 1994). The average concentration of mercury in the water column is 20 ng/L (Suchanek et al., 1997). Multiplying these values together gives an average mass of mercury removed from the lake in agricultural and municipal diversions of 0.4 kg/year.

3.2.4 Biota

A small amount of mercury is eliminated from Clear Lake in fish and other organisms. A conservative estimate was made of mercury taken out of Clear Lake in biota, mainly in fish removed by human anglers and wildlife. According to creel survey and fish harvest data collected by the California Department of Fish and Game, sport anglers remove an estimated 17,300 kg of fish from Clear Lake per year (Cannata, 2000; Macedo, 1991). To obtain this estimate, anglers were surveyed on weekends once per month. Data was then extrapolated to the rest of the year. According to Fish and Game staff, the survey likely underestimated the catch by sport anglers on shore, especially of catfish, sunfishes and hitch. The commercial fishery in 1993 and 1994 removed an average of 12,300 kg/year of carp and Sacramento Blackfish (Bairrington, 2000). Average concentrations of mercury in the various species fished are

known (CVRWQCB, 1985; (Stratton et al., 1987; Suchanek et al., 1997). Multiplying the catch of each species by concentration of mercury results in an estimated 7 grams of mercury removed from Clear Lake by human anglers annually. This estimate was doubled to 14 g/year, to account for the underestimate of human catch and for intake by wildlife removing their prey from Clear Lake. Presumably, nearly all mercury in fish removed from Clear Lake is in the form of methylmercury.

3.3 Mercury Reservoirs in the Lake

3.3.1 Mercury in the Sediment Active Layer

Anthropogenic mercury contamination of Clear Lake exists over a background level of mercury. The UC Davis core data shows that average concentrations of mercury in sediment in the early 1800s (prior to the start of mining at SBMM) were 0.3, 1 and 5 ppm dry weight in Upper, Lower and Oaks Arms, respectively. USGS researchers found the average concentrations of mercury in the Oaks Arm core prior to the middle 1800s to be around 7 ppm (Sims and White, 1981). Higher levels of mercury in deep Oaks Arm sediments are not unexpected relative to the other Arms, given the presence of the hydrothermal system that formed the SBMM mercury deposit. A deep core collected about 3.5 km from SBMM showed a mercury peak of 65 ppm at a depth of 599 cm, which correlates to approximately 7400 years ago (Sims and White, 1981).

Sediment cores collected by UC Davis CLERC (Suchanek et al., 1997 and unpublished data collected for the Regional Board in 2000) and USGS (Sims and White, 1981) show that concentrations of mercury in Oaks Arm increased to 50 ppm or greater in the top 40-80 centimeters of sediment deposit. Concentrations of mercury in sediment of Upper Arm have increased about 10-fold, relative to pre-mining levels. The start of the mercury increase in the cores was dated around 1930, which coincides with the use of heavy machinery on the mine site and the beginning of open pit mining. Plots of the UC Davis CLERC 1996 and 2000 cores are shown in Figures D-1 and D-2 of Appendix D. Since the cessation of mine operations, mercury concentrations appear to have declined slightly in all arms of the lake. However, surface concentrations of mercury are virtually the same in the 1996 and 2000 cores. Samples of surface sediment collected by UC Davis CLERC in 1994-96 showed a clear, inverse relationship between concentration of mercury in sediment and distance from the SBMM site (Suchanek et al., 1997; Figure D-4). Although intensive sampling has not been conducted since 1996, samples from selected sites in Oaks Arm indicate that the relationship between distance and concentration still exists.

Lakebed sediments are a sink as well as a source of mercury in the water column. Sediment deposition rates are relatively high in Clear Lake. Recent, average, sediment deposition rates are 0.9 cm/year with a range of 0.7 – 1.2 cm/year (Chamberlin et al., 1990; Richerson et al., 1994). Mercury attached to particulates becomes deposited with the sediment. Much of the bottom surface of Clear Lake is comprised of very fine-grained sediments. As currents resuspend these very loose sediments, mercury is returned to the water column. The top 5-10 cm of sediment comprises an active sediment layer in which resuspension and mixing occur due to physical processes and bioturbation. Burial of mercury in sediment below the active layer constitutes an output of mercury from the system. For the mercury budget, the amount of mercury that collects in the top of the active layer in one year was calculated. This allowed comparison of current inputs with the amount of mercury in surficial sediment.

An estimate of the amount of total inorganic mercury in one centimeter of sediment was made by the following procedure. Mercury concentrations were calculated for the top five centimeters and divided by the deposition rate to obtain an estimate of the amount of mercury deposited in sediment annually. Because the lakebed slopes from shore to center of the lake, sedimentation rates are likely less in the near-shore zones than in the middle of each Arm where they were measured. To adjust for this sediment focusing effect, the sedimentation rate of 0.9 cm/year was applied to 75% of the total surface area for each Arm. Sediment was not assumed to accumulate permanently in the remaining 25% of the surface areas. Masses of mercury in surficial sediment were calculated for each of the Arms, then summed to obtain the total amount of mercury in the surficial sediments.

Surficial sediment samples were collected by UC Davis CLERC seasonally in 1996 through 1998 (Suchanek et al., 1997; unpublished data from T. Suchanek). Concentrations of mercury in these samples and in the top centimeters of the sediment cores collected in 1996 and 2000 were averaged to obtain sediment concentrations of mercury in each arm. Sediment data collected in 1992-1995 results were not used in this calculation because sediment concentrations were likely affected by erosion from the WRD. Remediation of the WRD slopes occurred in 1992. For estimating current load of mercury in the surficial sediment, it was preferable to use data collected at least several years after the remediation.

For the calculation of mercury in the Upper and Lower Arms, the range of concentrations was small enough such that the average sediment in Upper Arm could be used. For Lower and Upper Arms, total mercury in sediment was determined as:

Mass of mercury = average mercury concentration * surface area * depth * f

(Where f = volume-weighted sediment density conversion factor)

Mercury levels in sediment decline with distance from SBMM. The relationship is most pronounced in Oaks Arm. To more accurately estimate the mass of mercury in Oaks Arm, average mercury concentrations were determined for the area from the shore of SBMM to sampling site OA-01, and for the area from OA-01 to the mouth of Oaks Arm at the Narrows (See Figure 1).

Because sediment mercury concentrations are expressed in dry weight and the values for lakebed area and depth are for "whole" sediment (particles plus porewater), all of the load equations included a conversion of dry weight sediment to wet weight sediment. It was also necessary to convert from sediment weight (kg wet sediment) to sediment volume (cubic meters wet sediment). A single, volume-weighted sediment density factor was calculated for each arm that used percent total solids in sediment and standard densities for water (1000 kg/m^3) and dry aluminosilicate sediment (2650 kg/m^3). On average, surficial sediment in Clear Lake is comprised of 10% solids and 90% water. Depth for these equations was a constant 5 centimeters.

The above equations resulted in mass of mercury per five centimeters of surface sediment. These values were then divided by the annual deposition rate to estimate the recent, annual deposition of mercury to lakebed sediments. Estimates are shown in Table 7.

Table 7. Annual Deposition of Mercury in Clear Lake Sediment.

	Average mercury concentration in surface sediment (mg/kg dry weight)	Total Surface Area (km2)	Annual sediment deposition rate (cm/year)	Annual deposition of mercury in sediment (kg/yr)
Lower Arm	3.5	34.0	0.9	90
Upper Arm	2.8	111.6	0.9	250
Oaks Arm (a): OA-01 to mouth SBMM shore to OA-01	 25 200	 15.0 0.6	 0.9 0.9	 295 80
Total				**715**

a. Concentrations of mercury decline with distance from SBMM, ranging from 3 ppm at the Narrows to more than 300 ppm in mining debris offshore of the mine site.

The same exercise can be done using average concentrations of mercury in sediment prior to the beginning of mining operations in the watershed. Assuming the same sediment density and deposition rate as above, annual loads to sediment were calculated for the pre-mining period. Pre-mining loads were estimated to be 30, 30 and 60 kg/year for Lower, Upper and Oaks Arms, respectively, for a total of 120 kg/year across the lake (See Appendix D for mercury concentrations in sediment cores prior to mining activities).

Regional Board staff assumes that the estimation of mercury deposited in one year to surficial sediments is be the upper bound of annual loading of mercury to Clear Lake sediments. Mercury in the active layer comes from new inputs (the global atmospheric pool, tributaries and ongoing inputs from SBMM). Mercury in the active layer may also come from remobilization of mercury in previous loads from SBMM. The amount of historically deposited mercury that does remobilize should be investigated. The total deposition of 715 kg/yr, minus inputs from the global atmospheric pool and tributaries, is assumed to be the upper bound of loads from SBMM, including past and ongoing inputs.

3.3.2 Mercury in the Water Column

An estimate can be made of the mass of mercury that is contained in the water of Clear Lake (Table 8). This estimation uses average concentrations of mercury in unfiltered water collected over a three-year period and during all seasons (Suchanek et al., 1997). During periods of significant runoff into the lake or high winds, the water column would contain more mercury. Mercury mass in the water of each Arm is calculated by multiplying average mercury concentration by water volume.

Table 8. Average Mass of Mercury and Methylmercury in Clear Lake Water

	Average mercury concentration, ng/L (a)	Average methylmercury concentration, ng/L (a)	Water volume $m^3 \times 10^6$ (b)	Average mercury mass kg	Average methylmercury mass, g
Lower Arm	12	0.08	384	4	30
Oaks Arm	66	0.11	138	9	20
Upper Arm	10	0.09	904	9	90
Totals				22	140

a) Averages of mercury and methylmercury concentrations in unfiltered water samples from surface and deep water levels, collected 1994-1996 (Suchanek et al., 1997)
b) Richerson et al., 1994

3.4 Significance of Mercury in Groundwater from SBMM

Regional Board staff assumes that, in general, mercury entering the lake can be methylated. Data from the ongoing CALFED-funded study in the Delta support this assumption, by showing that in an environment conducive to methylation, mercury from various sites was readily methylated (Bloom and Katon, 2001). Therefore, all mercury entering and in surficial sediment of Clear Lake is of concern. One source, however, that must be particularly addressed in this TMDL is the ongoing release of mercury in groundwater from the Sulphur Bank Mercury Mine.

3.4.1 Porewater analyses have verified that groundwater from the waste rock dam flows upward through Clear Lake sediments near the mine site. A flocculent precipitate (floc) forms when acidic groundwater mixes with lake water is easily moved by currents. Most floc samples contained high concentrations of methylmercury.

Groundwater flow from the mine site has been determined to be entering Clear Lake by subsurface flow through lake sediments (Shipp, 2001). This investigation involved chemical analyses of porewater in sediment cores collected near the mine site and further into Oaks Arm. The determinations are based upon the fact that various sources of porewater, including lake water, acid mine drainage, Herman Impoundment water and hydrothermal fluids from springs on the mine site, have unique "signatures" in their ratios of chemical elements and stable isotopes. Relative proportions of source water in a porewater sample are reflected in the geochemistry of the sample.

Porewater samples indicated that fluids from Herman Impoundment and other acid rock drainage are present in porewater collected near the mine face (Shipp, 2001). Water flowing through sediments near the mine site contains ion and stable isotope concentrations indicative of a mixture of Herman Impoundment water and meteoric water that is acidified and mineralized as it flows through the WRD. Fluids from the mine site were detected in lakebed sediments as far as 1.2 km from the mine shoreline. In contrast, porewater at site OA04 (about three km from the mine shore) was characteristic of aged lake water and did not appear influenced by acid rock drainage. Surveys of shallow water conducted along the length of the mine face have revealed "hotspots" with low pH and very high sulfate concentrations (approximately ten or more times higher than Herman Pit) that are suggestive of preferential pathways of acidic flow into the lake (Suchanek et al., 2001a).

Upon contact with lake water, acidic water from the mine appears to form a flocculent precipitate (floc) on the lakebed adjacent to the mine site. Floc was discovered in Spring of 1995, after heavy rains caused Herman Impoundment to overflow into the lake (Suchanek et al., 1997). The floc covered an area of approximately $1x10^6$ square meters offshore of the mine site. Chemical analysis showed the floc to be a clay-based, aluminosilicate mineral. In the laboratory, floc was formed by mixing water from the pit and the lake. Floc has been observed yearly since 1995 despite the fact that the pit has not overflowed. Distribution of the floc seems to vary by season and with rainfall (Suchanek et al., 2000b). The discovery of floc provided the first evidence that groundwater, as well as surface runoff, is a route of mercury transport into Clear Lake.

The porewater and precipitate data demonstrate a link between mercury in groundwater on the SBMM site and transport of that mercury into the lake. More data is needed to quantify the amounts of mercury

fluxing up through lakebed sediments and to establish a baseline for evaluating the success of future groundwater controls at the mine site.

UC Davis researchers have hypothesized that floc containing large quantities of methylmercury is a significant source of bioavailable mercury to the entire Clear Lake ecosystem (Suchanek et al., 1997). Concentrations of mercury in newly formed floc were equivalent to or lower than concentrations in underlying sediment. As floc consolidated in late spring and summer, concentrations of methylmercury in floc were higher than in underlying sediment, by an order of magnitude or more. Floc is light and fluffy, with buoyancy near neutral. It is easily suspended in currents. As will be described below, particles in the eastern end of Oaks Arm are readily transported to other Arms. The degree of influence of floc on production of methylmercury at sites distant from SBMM is unknown. Mercury may be methylated in floc and transported across the lake. It also may be that mercury, sulfate, bacteria, and/or dissolved organic carbon in floc modulate methylmercury formation in sediment of other Arms of the lake.

3.4.2 Most mercury leaving the mine site in groundwater is dissolved, probably as a chloride or oxychloride or oxide complex. These forms of mercury are more readily methylated than cinnabar.

Acid rock drainage creates an environment with high sulfate and low pH that facilitates methylation. Cinnabar has extremely low solubility in water. Its solubility is significantly increased in oxidized, low pH, high sulfate waters, which are typical of acid rock drainage. Recent groundwater sampling by Tetra Tech showed that concentrations of dissolved mercury in the waste rock dam are positively correlated with redox potential. (Tetra Tech EMI, 2001).

Dissolved mercury entering the lake may be a significant source of mercury that is preferentially methylated, relative to large amounts of cinnabar in the lake sediments. Mercury is likely moving through the waste rock dam in chloride or other highly-soluble complexes. Mercuric chloride ($HgCl_2$) and bisulfide complexes are hypothesized to be key chemical species that cross cell membranes and are taken up into methylating bacteria (Morel, 1998). Acidic drainage from the mine site also contains high sulfate concentrations (Shipp, 2001), which enhance the rates of methylation by sulfate-reducing bacteria (Rytuba, 2000). High concentrations of methylmercury in floc near the mine site suggest that the dissolved mercury is readily methylated. Additional research in Clear Lake is needed to determine relative bioavailabilities of mercury compounds and factors that control methylation.

3.4.3 Microcosm experiments demonstrate that flux of methylmercury from floc-laden sediments near the mine are significantly higher than in other parts of Clear Lake.

Flux of methylmercury out of Clear Lake sediments has been measured in laboratory experiments in 1996, 2000 and 2001 (Suchanek, 2001b; Suchanek et al., 1997). Microcosm trials were designed to evaluate current methylmercury flux during predicted seasons of low and high methylmercury production. These data were used to set lower and upper estimates on the contribution that sediments from various locations in Clear Lake make to methylmercury production during different seasons. For each experiment, methylmercury released from sediments into overlying water in experimental core tubes was

measured over a five-day period in the laboratory. Trials were conducted under anoxic (bubbled with nitrogen) and oxic (bubbled with air) conditions.

Results from the 2000 and 2001 microcosm trials are reported below (Table 9). Two separate collections of short cores were made from five sites in Clear Lake for two five-day-long lab experiments, one set in fall 2000 and one set in Spring 2001. Sediment was collected from sites in the middle of Oaks, Upper and Lower Arms and from floc-containing sediment approximately 0.3 km offshore from the mine site. The experiments were conducted during a predicted high methyl-Hg production period (fall) and a predicted low methyl-Hg production period (winter/spring). Surficial sediment and water samples were also collected from the same sites.

Under anoxic conditions, October sediment samples from sampling sites OA-01, UA-03, LA-03 and the black floc site in Oaks Arm (OA-FB) had similar net rates of methylmercury production (0.004–0.008 ng/cm3, over a five-day period) (Suchanek, 2001b). Net methylmercury production at the white floc (OA-FW) site was five times higher (0.034 ng/cm3) and at OA-04 two times higher (0.016 ng/cm3) than rates at the other sites. As expected, methylmercury production was much lower in April (maximum 0.005 ng/cm3 at OA-FB). In April, the floc sites continued to produce methylmercury, whereas methylmercury flux rates from Oaks Arm and Upper Arm sediments were less than in the water-only controls. Possibly demethylation was the controlling reaction in the Oaks and Upper Arm sediments in April. Under oxic conditions, methylmercury production was low (less than 0.004 ng/cm3 for all samples and time points).

A similar pattern was seen in microcosm experiments conducted in October 1996. Under anoxic conditions, methylmercury production in the Oaks Arm-floc sediment was about five times higher than rate from the Upper Arm site and eleven times higher than production at Oaks Arm sites lacking floc (Mack et al., 1997).

Methylation activity is hypothesized to be high in the floc for several reasons (Suchanek et al., 1997):

- it has a large surface area available for support of methylating bacteria,
- floc contains elevated levels of divalent mercury,
- floc is rich in sulfate, and
- floc forms at the eastern end of Oaks Arm, which is also a collection point for drifting organic matter. Winds drive organic material to the end of Oaks Arm, providing a plentiful nutrient source for bacterial growth

Despite high rates of methylmercury production occurring at the floc sites, the surface area covered by floc is small relative to the rest of the lake. Methylmercury production is likely dominated by smaller rates of methylation occurring over the large surface area of Upper Arm. If the results of the microcosm experiments are extrapolated to the entire lake, the influence of Upper Arm sediments on methylmercury inputs can be seen (Table 10). This comparison assumes that the microcosm results represent average net methylation rates across each Arm.

Table 9. Estimated Methylmercury Production in Clear Lake Sediments for a Five-Day Period, Extrapolated from Microcosm Studies.

	Average net methylmercury production rate, ng/cm^2		Surface Area (b)	Methylmercury produced in 5 day period (c)	
	October 2000	April 2001		October 2000	April 2001
	ng/cm2	ng/cm2	km2	g	g
Oaks Arm -Floc	0.034	0.005	0.05	0.02	0.003
Oaks Arm	0.01	0 (a)	15.5	1.55	0
Upper Arm	0.007	0 (a)	111.6	7.81	0
Lower Arm	0.006	0.002	33.9	2.04	0.679

(a) In microcosm trials, methylmercury flux was less from these sediments than in the control tubes (no sediment). Demethylation rate may have been greater than the methylation rate.
(b) The surface area of the floc site was estimated as the average surface area recorded in surveys of floc conducted in 1998 (Suchanek et al., 2000b).
(c) Assumes that methylation rates can be generally applied across the corresponding surface area.

Table 10. Estimated Yearly Input of Methylmercury from Sediment, Based on Extrapolation from Microcosm Studies.

	Methylmercury produced in 5 day period across surface area of each region (From Table 9)		Lower Bound Estimate: summer rate occurs for 2 months of the year.		Upper Bound Estimate: summer rate occurs for 6 months of the year.		Sums: Lower Bound of Estimated Sediment Input	Sums: Upper Bound of Estimated Sediment Input
	Summer rate (represented by October microcosm data)	Winter rate (represented by April microcosm data)	Summer (2 months)	Winter (10 months)	Summer (6 months)	Winter (6 months)		
	g	g	g	g	g	g	g/yr	g/yr
Oaks Arm - Floc	0.02	0.0027	0.2	0.2	0. 7	0.1	0.4	0.8
Oaks Arm	1.55	0	19	1	57	0.6	20	58
Upper Arm	7.81	0	95	7	285	4	100	290
Lower Arm	2.04	0.679	25	41	74	25	66	99

For purposes of the methylmercury budget in Table 12, the microcosm data was used to estimate upper and lower bounds of methylmercury flux from sediment. Ranges of methylmercury inputs from the sediment were obtained by assuming that the October flux rates represented the average net methylmercury input for two months of the year (lower bound estimate) and for six months of the year (upper bound estimate). This estimation is highly uncertain, but gives a possible range of methylmercury inputs to the water column from sediment.

3.4.4 A hydrologic model of Clear Lake indicates that particles originating in Oaks Arm and near SBMM are readily transported into Lower and Upper Arms.

Clear Lake is polymictic, meaning that the water column mixes vertically at multiple times during the year. As radiant energy warms the lake surface, the water column stratifies with respect to temperature.

In summer and fall, wind from the northwest blows nearly daily across the lake. Clear Lake is stratified for only short periods of time because of the wind and shallow depths, which facilitate remixing (Horne, 2000).

Movements of currents and particles in Clear Lake have been modeled by Rueda and colleagues at the Department of Environmental Engineering at UC Davis (Rueda, 2001a). Rueda's 3-dimensional, hydrodynamic model addressed water flow and particle transport within and out of the Oaks Arm. The model was validated using detailed field measurements of water temperature gradients collected in spring and summer of 1999 and 2000. The movement of neutrally buoyant particles was tracked from a release point on the northern shore of the Oaks Arm. This model was not designed to examine particle transport during storm events.

Results of the modeling showed that the maximum amount of particle transport occurs at the end of a stratification period (Rueda, 2001a; Rueda, 2001b). Water is pushed on the surface to the end of the Oaks Arm by northwest winds and out of the Oaks Arm along the bottom by return flow. Temperature stratification allows masses of water on the surface and bottom to move in different directions with the least amount of friction. Buoyant particles, therefore, move most rapidly when the lake is stratified. Clear Lake stratifies at least partially on a daily basis, linked to the diurnal wind cycle. The lake likely stratifies more completely for brief periods at least four times per year.

Rueda and colleagues released neutrally-buoyant particles near the northern shore of the Oaks Arm, northwest of Rattlesnake Island at a depth of 1.5 m. The particles were detected in surface water at the eastern end of the Oaks Arm within two days after release and well into the Lower Arm within three days. Wind-driven mixing also drove the tracer particles downward. The particles moved with bottom currents (7 m depth) to the mine face within two days and into Upper Arm in three days. Because particles entering Upper Arm likely moved past the mine face first, particle transport from the mine face to the Upper Arm could occur in as little as one day.

Current measurements collected during a four-day period in August 1995 under a range of wind conditions showed a similar result in terms of flow patterns (Lynch and Schladow, 1996). A continuous exchange was observed between the Upper Arm, oxygenated surface water and Oaks Arm bottom water low in dissolved oxygen. A clockwise circulation pattern observed in the Upper Arm suggests that particles entering the Upper Arm at the Narrows would disperse in the Upper Arm. Current velocities were used to estimate average volumes of water exchanged each day. Based on current velocities, the authors estimated that a volume equivalent to 10% of the volume of water in the Oaks Arm was exchanged on a daily basis at the Narrows and at the mouth of the Lower Arm.

Data and modeling described above demonstrate that particles formed near the mine site can be relatively rapidly carried into other Arms. This information is particularly important because dissolved mercury and/or floc near the mine site are available for transport.

3.5 Mass Balance for Mercury in Clear Lake

The mass balance for mercury in the active layer of sediment includes annual estimated of inputs, outputs and storage within the lake (Table 11). Mercury in lakebed sediment is expressed as a surficial sediment reservoir of mercury that is deposited in one year. The surficial sediment load can be compared to the sum of annual inputs. Mercury loads in surficial sediment were estimated from samples collected in 1996-2000, in order to most closely match inputs estimated from data collected in the same period. Sediment on the surface today is expected to be removed from the system (an output term) when it becomes buried five or more years in the future. Inputs and outputs to the sediment would be equivalent if loading remained constant over the time period needed for burial below the active layer to occur.

Table 11. Annual Mass Balance for Mercury in Clear Lake

	Annual Mercury Load (kg/yr)
INPUTS	
Deposition from Global Atmospheric Pool	2
Tributaries and Direct Surface Water Runoff (a)	18
Total input from SBMM. Includes ongoing inputs in surface and groundwater flow and remobilization of previously deposited mercury from the mine site (b)	2 - 695
OUTPUTS	
Cache Creek (a)	3
Flux from lake surface to atmosphere	1.6
Removal in biota	0.015
Surface water diversions for agricultural and municipal use	0.4
RESERVOIRS	
Water column	22
Surficial sediment reservoir (annual deposition, based on averages of surface sediment concentrations measured in 1996– 2000) (c)	
Lower Arm	90
Upper Arm	250
Oaks Arm	375
Total of the sediment reservoir	715

(a) The ten-year averages for tributary loading and Cache Creek outflow are given. Loads into Clear Lake vary from 1 to 60 kg/year and are correlated with low versus high water years. Cache Creek outflow varied from less than 0.01 to 11 kg/yr.
(b) The lower bound from SBMM is the USEPA Superfund Program's estimate of mercury transported in groundwater through waste rock only. The upper bound is the total of mercury in surficial sediment, minus other inputs.
(c) The sediment reservoir becomes buried approximately 10 cm below the active, surficial sediment layer

3.5.1 Uncertainties in the Mass Balance for Mercury in the Active Layer

One source of uncertainty is that current inputs are compared with sediment concentrations that, due to mixing of sediments by wave action and bioturbation, likely reflect an average of the inputs of five or more years ago. In particular, the reduction in mercury inputs that occurred as a result of the 1992 remediation at SBMM may not yet be reflected in the surficial sediment concentrations. UC Davis

researchers have suggested that it could take eight years or more beyond the remediation for surficial sediment levels to reflect the change in load (Suchanek et al., 1997). If mercury that eroded from waste rock piles did not become buried as quickly as expected, sediment concentrations may not yet have responded to the reduced load. One check of this hypothesis would be to include the estimated annual load from surface water runoff and erosion from the SBMM waste rock dam prior to remediation. Refined estimates of this load contribution, however, do not exist. Chamberlin and coworkers estimated the annual load was 132 kg/year. Using their annual estimate, they suggested that during the 35 years between closure of the mine and emergency remediation action by USEPA to regrade and vegetate the waste piles, between 3720 and 5270 kg mercury was transported into Clear Lake (Chamberlin et al., 1990). This estimate is likely low. The erosion measurements made by Chamberlin and colleagues were made during a drought year. Erosion and mercury loading during normal and high precipitation years could have been much greater. If shoreline remediation significantly reduced the loading of mercury from SBMM, we anticipate that the load reduction should be apparent in sediment concentrations within the next ten years. More sediment data could help to refine this uncertainty.

Previously deposited sediment may also be resuspended by currents. During periods of high turbidity, portions of the sediment load of mercury are resuspended such that part of the "load" is carried in the water column. This load may be redeposited again over time. Particularly in the Oaks Arm, the mine wastes dumped into the lake may be eroding and depositing in surficial sediment elsewhere in the lake.

Uncertainties in the load estimates have been discussed in previous sections of the Source Analysis. Additional data being gathered by the USEPA Superfund Program on wetland transport and flux from mercury in soils to the atmosphere may help to refine estimates of ongoing inputs from SBMM. With the exception of atmospheric flux, other potential sources (hydrothermal vents and lava tubes in the lake) have been investigated and found to emit little mercury (Suchanek et al., 1997).

3.6 Mass Balance for Methylmercury in Clear Lake

A very rough budget for methylmercury in Clear Lake is shown in Table 12. This estimate of methylmercury inputs and outputs needs considerable refinement but can be used for semi-quantitative assessment. The magnitudes of inputs of methylmercury from the tributaries and the lakebed sediment appear to be similar. However, inputs from the sediment are very difficult to measure. The extrapolation of the existing laboratory studies across the entire lakebed introduces considerable uncertainty into the sediment estimate.

Methylmercury is produced in sediment, where it cycles between methylation and demethylation and between flux to the water column and deposition. The estimates for net methylmercury production from lake sediments are based on flux of methylmercury from sediment into overlying water, as measured in the laboratory (Suchanek, 2001b). Microcosm experiments were conducted in October 2000, a peak methylation period, and in April 2001, a time of reduced methylmercury production in lakebed sediments. Sediments were collected from one site each in Upper and Lower Arms and for several sites in Oaks Arm. In the laboratory, methylmercury released into overlying water was measured over a five-day period. For this methylmercury budget, the flux rates were extrapolated to the entire surface area of each corresponding Arm of the lake. This calculation was described more fully in Tables 9 and 10 of Section 3.4.3. Ranges of methylmercury inputs from the sediment were obtained by assuming that the October

flux rates represented the average net methylmercury input for two months of the year (lower bound estimate) and for six months of the year (upper bound estimate). Flux of methylmercury into overlying water from the sediment is assumed to represent a net input of methylmercury to the water, taking into account methylation and demethylation in the sediment.

The estimates of methylmercury input and outputs from tributary streams are ten-year averages of yearly data shown in Table 4. Small amounts of methylmercury likely enter the lake from air (Lindberg and al., 2001) and in groundwater from mine waste (Rytuba, 2000). Methylmercury from these sources has not been quantified.

The estimate of methylmercury in fish removed from the lake by humans and wildlife is the same as that used in the total mercury budget (Table 11). This follows the assumption used throughout this TMDL, that essentially all mercury in trophic level 3 and 4 fish is methylmercury.

Mass of methylmercury in the water column is based on multi-year averages of methylmercury concentrations, as shown in Table 8. During typical, high methylation periods in August, September and October, methylmercury in the water column would likely be higher (Estimated 50, 30 and 170 grams in Lower, Oaks and Upper Arms, respectively, based on water column concentrations in Aug. – Oct.). Water concentrations of methylmercury decrease in winter and early spring.

An estimate of masses of methylmercury in surficial sediments of each Arm was prepared by multiplying the average concentration of methylmercury in the top three centimeters of sediment collected in 1994-1998 (Suchanek et al., 1997; unpublished 1998 data provided by T. Suchanek) by the surface area of each Arm. The majority of activity by methylating bacteria occurs in the top three to four centimeters of sediment (Mack and Nelson, 1997). For this calculation, then, a sediment depth of three cm was used. Significant uncertainties in this estimation exist in: 1) extrapolating methylmercury concentrations from collection points to the surface area of an Arm and 2) quantitative relationship between methylmercury production in the sediment, flux into overlying water and burial. Microcosm experiments conducted in 1996 indicated that less methylmercury fluxed into overlying water than was present in sediment porewater (Mack, 1998). Therefore, mass of methylmercury in surficial sediments is not expected to be equivalent to the result of inputs minus outputs and storage.

An unknown amount of methylmercury is stored in the sediment. In the mercury budget, mercury mass in surficial sediment burial will become an output term, as mercury is buried below the active or resuspended layer of sediment. In the methylmercury budget, the sediment reservoir of methylmercury refers to methylmercury in the surficial sediment layer, where methylmercury is both deposited and produced (net production is methylation minus demethylation). Analysis of deep sediment cores collected in September 2000 showed methylmercury throughout the core and generally declining with depth (T. Suchanek, unpublished data collected for the Regional Board). The methylmercury core data is still being interpreted with respect to stability of methylmercury in sediment and rates of burial.

Table 12. Annual Methylmercury Mass Balance for Clear Lake

	Average, g/year
INPUTS	
Total tributary input	350
Flux from sediment (extrapolated from net methylation rates from microcosm experiments) (a)	
Upper Arm	100 - 300
Lower Arm	70 - 100
Oaks Arm	20 - 60
OUTPUTS	
Cache Creek	150
Biota (estimation of methylmercury removed through wildlife consumption or sport fishing)	15
Water diversions - municipal and agricultural	2
RESERVOIRS	
Water Column (based on average water concentrations of methylmercury and volume of each Arm)	
Upper Arm	90
Lower Arm	30
Oaks Arm	20
Sediments - top 3 cm (based on average surface sediment concentrations of methylmercury and surface area of each Arm)	
Upper Arm	1,100
Lower Arm	420
Oaks Arm	740

(a). Lower bound assumes that the peak methylation period is 2 months long. Upper bound assumes that it is 6 months long. See Tables 9 and 10.

4 LINKAGE ANALYSIS

A linkage analysis describes the association of numeric targets with identified sources of mercury. Definition of this relationship provides a basis to estimate total assimilative capacity and identify any needed load reductions. The linkage analysis determines assimilative capacity as a loading rate that is consistent with meeting the target fish tissue concentration.

The linkage analysis for mercury describes the relationship between inorganic mercury in water and sediment and methylmercury in fish tissue. The purpose of the linkage analysis is to answer the question, "How much should mercury loads be reduced in order to meet the target concentrations of mercury in fish tissue?" A conceptual model of mercury loading and transformation is presented in Figure 2. Mercury cycling between inorganic and organic forms and from water to sediment is highly complex. Quantitative links between methylmercury in fish and water, and between water and acceptable sediment concentrations are difficult to achieve with existing data. The focus of the TMDL will be to control loads of inorganic mercury in the active sediment layer. Although other factors influence rates of methylation, information is lacking to design an implementation plan to control other factors. This focus on total mercury is represented by the following equation:

[methylmercury] = k * [total mercury]

Where:
[methylmercury] = concentration of methylmercury in fish tissue
[total mercury] = concentration of total mercury in the active sediment layer
k = constant representing all factors modifying methylation or uptake rates

The above equation essentially asserts that the relationship between concentrations of methylmercury and total mercury is a simple proportionality. It is very likely that this relationship is not, in fact, linear. Many factors affect methylation, including sulfide and sulfate concentrations, temperature, levels of anoxia, organic carbon, concentrations of methylating and demethylating bacteria, rate of demethylation, chemical form of mercury, sunlight, pH, sediment grain size, and other nutrients (Barkay et al., 1997; Morel, 1998; Regnell et al., 1998; Xun et al., 1987). Factors that affect accumulation of methylmercury in fish tissue include species, growth rate, prey availability and preference, and methylmercury intake by prey (Harris and Bodaly, 1998; Wiener and Spry, 1996). At a given point in time, the cumulative effect of all factors that affect methylation rate could be a constant. In that case, the concentration of methylmercury would vary solely as a function of mercury concentration. Over multiple seasons or water quality conditions, however, the relationship is more complex. We lack enough information on the factors modifying methylation and uptake to adjust the above equation so that "k" is more than a constant.

One piece of information available for Clear Lake is regarding the sources of mercury that are being methylated. Several types of data indicate that dissolved mercury entering Clear Lake from the SBMM is preferentially methylated, relative to cinnabar, in lakebed sediment. The significance of ongoing inputs of mercury in groundwater from SBMM was described in Section 3.4. Patterns of current movement and high levels of methylmercury in biota distant from SBMM indicate that the effects of loading from SBMM are felt throughout the lake. Part of the load allocation focuses on groundwater from SBMM as an important source of mercury that must be controlled.

The linkage analysis for Clear Lake was actually separated into several steps. Two steps are elaborated in sections below. Concentrations of methylmercury in water and methylmercury in biota are related by bioaccumulation factors. Concentrations of methylmercury and total mercury in sediment are related through calculation of a methylation efficiency index. The limited amount of data available on relationships between methylmercury in the water column and in sediment (flux rate of methylmercury from sediment) were described previously in Section 3.4.3. In each of these steps, one variable is related to another by a simple ratio or linear equation. Understanding of the methylation and uptake processes is lacking to refine the equations to incorporate effects of other factors. Hence, the end result becomes the equation shown above, that methylmercury in biota is related in direct proporation to mercury in sediment.

Although this simplified linkage assumes a linear relationship between methylmercury in fish tissue and inorganic mercury in surficial sediment, the relationship is not 1:1. The linear relationship implies proportionality between mercury in various environmental compartments. For example, the use of BAFs assumes that methylmercury in fish tissue is directly proportional to methylmercury in water.

Assumptions of a linear relationship between methylmercury concentrations in fish tissue and concentrations of mercury in sediment, or between steps in this linkage have been used previously. This assumption has been made in the mercury TMDL for the Savannah River in Georgia, the draft TMDL for San Francisco Bay, and in preliminary modeling for the Florida Everglades TMDL (Abu-Saba and Tang, 2000, Tetra Tech, 2001 #150). Researchers working in the Experimental Lakes Area in Ontario found that within a given ecological system (such as a lake), the concentration of mercury in the water column was a good predictor of methylmercury levels. The relationship broke down during comparisons across different types of lakes (Kelly et al., 1995; Waldron et al., 2000). Researchers in the Florida Everglades also found that within ecosystem types (such as eutrophic wetland or oligotrophic wetland), significant relationships existed between methylmercury in the water and total mercury in the water or sediment (Stober et al., 2001). Clear Lake is a single, shallow, eutrophic waterbody. With the exception of the area of the Oaks Arm influenced by acid rock drainage from SBMM, conditions for methylation and bioavailability are thought to be rather uniform throughout the lake.

There have been two, more sophisticated models used for Clear Lake. One was a models of aquatic fate and transformation of mercury in Clear Lake (Bale, 2000). Application of this model showed that total mercury and methylmercury concentrations in the water could be reasonably modeled as functions of total mercury in surficial sediment. The model was unable to accurately predict concentrations of methylmercury in sediment and water near SBMM, perhaps because of the low acidity and high sulfate conditions resulting from groundwater flow from the mine. Bale modeled exchanges of mercury between the atmosphere, water, active sediment layer and burial in deep sediments, but did not model inputs of mercury from SBMM or the watershed.

A second model has been developed of particle transport in the lake. Movements of currents and particles were modeled by Rueda and colleagues at the Department of Environmental Engineering at UC Davis (Rueda, 2001a; Rueda, 2001b). Rueda's 3-dimensional, hydrodynamic model addressed water flow and particle transport within and out of Oaks Arm. The hydrodynamic model is relevant to mercury transport, because much of the mercury in the lake is sorbed to particles. Results showed that particles formed near the mine site can be relatively rapidly carried into other Arms.

Summary: Meeting the numeric targets of 0.3 and 0.13 mg/kg wet weight for trophic level four and three fish, respectively, would require reducing existing fish tissue concentrations by 40%. A linear linkage relationship dictates that overall mercury loads to Clear Lake sediment be reduced by 40%, in order to reduce methylmercury concentrations in fish tissue by the equivalent amount. Reducing inorganic mercury levels in sediment by 50% provides a margin of safety for the considerable uncertainties in the linkage analysis.

4.1 Bioaccumulation Factors

Mercury in fish can be related to mercury in water through the use of bioaccumulation factors (BAFs). Following is a discussion of Clear Lake-specific BAFs and derivation of a range of water column concentrations that correspond to the fish tissue targets. Because of uncertainties in Clear Lake food webs and seasonal fluctuations in methylmercury levels in water, setting a single water column concentration for Clear Lake that is "safe" is questionable and is not attempted here. Future investigations may reduce the uncertainties. By using BAFs, an acceptable range of water column concentrations is produced that can be used to assess the progress of load reductions. BAFs imply a linear relationship between methylmercury in the water column and in fish. Thus, to reduce fish tissue concentrations by forty percent, the goal must be to reduce water column concentrations by forty percent.

Bioaccumulation refers to the uptake and retention of a substance by an organism from its surrounding medium and from food. BAFs are indices that relate the concentration of mercury in a target organism to the concentration in a single source of contamination (water or prey). They are calculated by simply dividing the concentration of mercury in the organism by the concentration in the exposure source. For example, the BAF for methylmercury from water to channel catfish is:

$$\frac{\text{Methylmercury concentration in catfish}}{\text{Methylmercury concentration in water}} = \text{BAF}$$

Clear Lake-specific BAF's have been reported by UC Davis and the Clear Lake Environmental Research Center (UCD-CLERC; Suchanek et al., 1993). Samples of water, sediment and biota were collected simultaneously in Fall 1992. The purpose was to examine trophic transfer of mercury through benthic (i.e., sediment-chironomids-fish) and water column (i.e., water-phytoplankton-zooplankton-fish) pathways. A simple food web diagram is shown in Figure 3. This figure is adapted from diagrams showing food webs and accompanying BAFs for various Arms of the lake prepared by Suchanek and colleagues (1993).

BAFs were originally calculated by UC Davis using concentrations of methylmercury in filtered water. Raw data in the report was used by Regional Board staff to calculate corresponding BAFs from methylmercury in unfiltered water. For the linkage analysis, the focus is on BAFs relative to unfiltered water concentrations. Levels of methylmercury in filtered and unfiltered water samples from Clear Lake are correlated to approximately the same degree with concentrations in biota from sites in different arms (Suchanek et al., 1993). Also, data for the rest of the linkage analysis is available for methylmercury in

unfiltered water. For these reasons, BAFs for this linkage analysis were calculated from the concentrations of methylmercury in unfiltered water (Table 13).

Table 13 shows BAFs for trophic level three and four fish, using average, multi-year, lake-wide concentrations of methylmercury in fish and unfiltered water. The mercury concentration in a fish is the integration of mercury absorbed over the lifetime of the fish. Most fish samples used for calculation of average fish tissue concentrations in Clear Lake were collected in 1976-84, whereas water column samples were collected in 1994-96. Deriving BAFs based on these averages assumes that fish tissue concentrations have not changed over this time period.

The linkage analysis focuses on accumulation of methylmercury from the water column, not sediment. Fish feeding extensively on chironomids (sediment-dwelling midge larvae) clearly accumulate mercury through a benthic route. Data are lacking to calculated biota-sediment accumulation factors (BSAF) for Clear Lake. The sediment concentrations of methylmercury are measured in 1992 are questionable, due to use of a standard analytical procedure which may have produced methylmercury during the analysis. For calculating criteria to protect humans and wildlife, the Mercury Study Report to Congress (USEPA, 1997c) and the Great Lakes Water Quality Initiative (USEPA, 1995b) used only BAFs representing a water column transport pathway.

BAFs for average-sized trophic level four fish range from about 3×10^6 to 4×10^7. This range for trophic level four fish is based on average, multi-year concentrations of methylmercury in unfiltered water. BAFs for the largest channel catfish and largemouth bass (mercury concentrations 1.1–3.3 ppm) range up to 3×10^8. BAFs for trophic level three fish, ranged from about 1×10^6 to 2×10^7. This range for TL3 fish includes the BAFs for inland silversides, which are representative of fish likely eaten by wildlife.

Table 13. Concentrations of Methylmercury in Water Corresponding to Fish Tissue Targets

	TL4		TL3	
Target fish tissue concentrations, ppb	300	300	130	130
Bioaccumulation factors (a)	3.9 E+07	3.0 E+06	1.7 E+07	1.3 E+06
Corresponding water column concentration of methylmercury, ng/L (pptr)	0.008	0.099	0.008	0.098
Percent of 1994-96 water samples less than corresponding water column concentration (b)	0	66	0	65

(a) There is a range of BAFs for each of the trophic levels, because of variations in concentrations of methylmercury in water and in individual fish within each trophic level.

(b) Water samples were collected seasonally from multiple sites in each Arm in 1994-1996 (Suchanek et al., 1997). Methylmercury concentrations in surface and deep water samples, not including marsh sites, ranged from 0.013 to 1.49 ng/L. In general, highest concentrations were observed in August through October.

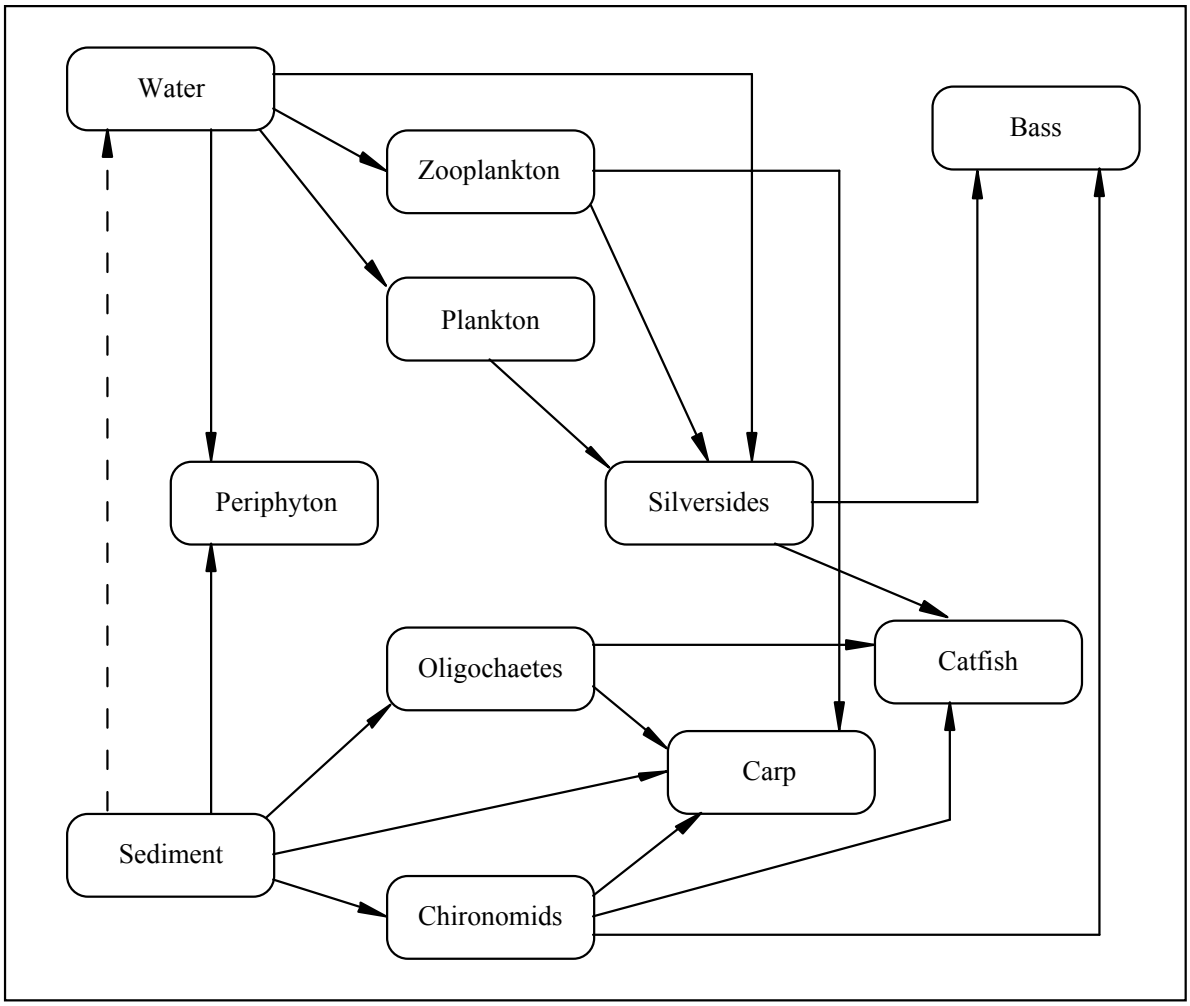
Target fish tissue concentrations were divided by bioaccumulation factors to calculate corresponding water concentrations of methylmercury (Table 13). The range of water column concentrations that correspond to fish tissue targets spans an order of magnitude. The water column ranges associated with targets (0.008 – 0.099 ng/L) overlap but are slightly less than the range of methylmercury concentrations measured under existing conditions. Methylmercury concentrations recorded during regular sampling of Clear Lake in 1994-96 ranged from 0.013 to 1.49 ng/L (Suchanek et al., 1997).

Site-specific BAFs can be used to identify a range of water column concentrations of methylmercury that correspond to acceptable levels of methylmercury in fish tissue. The range of methylmercury concentrations corresponding to the targets will be used to assess the progress of TMDL implementation. Uncertainties in the BAF model and the range of methylmercury concentrations in water and biota of Clear Lake make identification of a single water quality "target" for Clear Lake to be little more than speculation. Calculation of a BAF for fish from water imposes a simple, linear relationship on what is likely a complex, non-linear process. Key steps in defining the relationship between concentrations in water and fish remain unknown.

One major unknown is the methylmercury water concentration that is most closely correlated with mercury levels in fish. Should BAFs be calculated using long-term averages of water concentrations? Alternatively, does methylmercury production during a particular season drive accumulation? In Clear Lake, methylmercury concentrations in the water column vary by depth and season. Concentrations of methylmercury in water and sediment peak in late summer to fall, as do concentrations in plankton (mainly blue-green algae) and zooplankton (Suchanek et al., 1997). Presumably methylmercury levels in fish are a function of the varying levels of methylmercury in water, but a more precise relationship is as yet undefined for Clear Lake.

Clear Lake food webs are more complex than the simplified pathway shown in Figure 3. For example, largemouth bass may feed on a variety of smaller fish species, including Sacramento blackfish, sunfishes or juveniles of larger species. Mercury concentrations in largemouth bass prey items may range from less than 0.1 ppm to 0.3 ppm mercury in tissue. Prey of top-trophic level feeders will vary by season, abundance of prey, and size and feeding habits of the predator fish. The structure of the food web determines the efficiency of methylmercury biomagnification from algae to top predators. Increases in the complexity and/or number of trophic levels in the aquatic food web, as in a eutrophic waterbody like Clear Lake, lead to higher mercury concentrations in top predators (Morel, 1998).

Figure 3. Food Web Diagram for Biota in Clear Lake



Adapted from Suchanek et al., 1993.

4.2 Methylation Efficiency

Clear Lake-specific bioaccumulation factors allow us to translate numeric fish tissue targets to a range of corresponding water column concentrations of methylmercury. The next step is to assess the linkage between methylmercury and total mercury. Again, the quantitative link to an acceptable sediment concentration is difficult to achieve with existing data. Instead, our focus is on reducing inorganic mercury loads proportionally to the reduction needed in fish tissue.

Only a fraction of the mercury in sediment becomes methylated. Net methylmercury production can be described as a function of inorganic mercury concentration and methylation efficiency (proportion of total mercury that becomes methylated). The assumption is made that the relationship between total mercury and methylmercury in sediment is linear; that is that it depends only on the concentration of total mercury. Factors that control the methylation rate are the focus of ongoing investigations in Clear Lake and other waterbodies.

In different areas of Clear Lake, mercury appears to be more readily methylated than in other areas. This is seen in the ratios of methylmercury to total mercury in the sediment (methylation index). Methylation indices were prepared using sediment data collected by UC Davis CLERC in 1994-1998 (Suchanek et al., 1997; Suchanek et al., unpublished data collected in 1996-1998). The data was collected throughout the year. In order to simplify the data presentation, averages for generally high (summer) and low (winter) methylmercury production periods were calculated. Ratios of methylmercury to total mercury are presented in Table 14.

Table 14. Sediment Methylation Index (Ratio of Sediment Concentrations of Methylmercury to Mercury, 1994-1998)

	Summer Index, as percent (a)	Winter Index, as percent (b)
Oaks Arm – floc site near SBMM	0.049	0.027
Oaks Arm site OA-01	0.0055	0.0034
Wetland north of SBMM	0.97	NA
Oaks Arm OA-04	0.013	0.017
Lower Arm LA-04	0.109	0.077
Upper Arm UA-01	0.066	0.042
Upper Arm UA-04	0.104	0.073

(a) For all sites except wetlands, "Summer" refers to July through October. For wetlands, peak methyl mercury concentrations were recorded in June.
(b) "Winter" includes samples collected January through April.

Methylation indices should be examined together with methylmercury flux measurements presented in Section 3.4.3. Methylation indices in Upper and Lower Arms are relatively high in comparison with Oaks Arm sites, in large part because inorganic mercury concentrations in Upper and Lower Arms are relatively low. Oaks Arm site OA-01 tends to be rocky and may provide poor substrate for methylating

bacteria (Personal communication, T. Suchanek). As expected from a conceptual model of mercury cycling, the methylation index is highest in the wetland north of SBMM. The surface area of the wetland is small, however, relative to the large surface areas of Lower or Upper Arms. Presumably the methylation index is high in other wetlands around the lake as well. Methylation indices are high in the floc area. In laboratory microcosms, floc samples also exhibited the highest flux rates of methylmercury from sediment into overlying water (Suchanek et al., 2001b). At the wetland north of SBMM the period of peak methylation occurred earlier than at sites within the lake.

5 MARGIN OF SAFETY AND SEASONAL VARIABILITY

5.1 Margin of Safety

A margin of safety was incorporated into the TMDL at several points. The most significant margin of safety is the 10-fold uncertainty factor in the acceptable daily intake level (reference dose) of methylmercury for humans. The reference dose is directly used to develop the numeric targets. The USEPA human health reference dose is 10-fold lower than the lowest level of methylmercury intake observed to have adverse effects in children.

The numeric targets also contain a second, smaller margin of safety of 5%. Safe daily intake levels of methylmercury for human consumers would have been met by reducing fish tissue concentrations by 35% of existing levels. The numeric targets require reducing current levels by 40%.

A third margin of safety is incorporated into the linkage analysis and assimilative capacity. A linear linkage relationship dictates that overall mercury loads to Clear Lake sediment be reduced by 40%, in order to reduce methylmercury concentrations in fish tissue by the equivalent amount. Reducing inorganic mercury levels in sediment by 50% provides a margin of safety for the considerable uncertainties in the linkage analysis.

Although the numeric targets were originally calculated using a reference dose for humans, there is a margin of safety for wildlife that eat fish from Clear Lake. The avian and mammalian reference doses each contain an uncertainty factor of three. These uncertainty factors lower the reference doses below levels of mercury known to cause adverse effects to mallards and mink, respectively. Although the uncertainty factors were not applied to account for species differences, they do provide some measure of protection to wildlife that may be more sensitive than others to affects of mercury.

5.2 Seasonal Variability

Seasonal fluctuations in loading, concentrations and transport of mercury and methylmercury production were discussed throughout the report. Winter precipitation influences the loads of mercury entering Clear Lake. Most of the mercury coming from tributaries and direct surface water runoff enters during high flow events (unpublished data from T. Suchanek; Appendix C). Precipitation and groundwater levels also affect loads of mercury coming from SBMM. Rates of groundwater flow through the waste rock dam tend to peak when the difference between surface levels of Herman Impoundment and Clear Lake is at its maximum. The maximum difference occurs generally twice per year: 1) early in winter as rain fills Herman Impoundment more quickly than the lake; and 2) in late summer, when lake levels are lowest due to releases from Cache Creek Dam (USEPA, 2001c). Concentrations of dissolved mercury in the groundwater may peak in early spring, when soil of the waste rock dam reaches its highest saturation point with oxygenated rain water. Well samples have not been analyzed seasonally for mercury to test this supposition.

Methylmercury concentrations in sediment and water of Clear Lake are highest in August through October (Suchanek et al., 1997). Correspondingly, methylmercury production in the sediment appears greatest in these months and lowest in January through April (Suchanek, 2001b; Suchanek et al., 1997).

Production of methylmercury is, in part, correlated with water temperature. Methylmercury concentrations seem to peak earlier in the summer in wetland areas than in open water of the lake. Transport of particulate-bound mercury and methylmercury from the eastern end of Oaks Arm to other Arm likely occurs most readily in the spring, summer and early fall seasons. Periods of temperature stratification of the lake, followed by winds from the northwest facilitate flow of water and particles along the bottom out of Oaks Arm (Rueda, 2001a).

Although season influences loading rates and transport of mercury and methylmercury in Clear Lake, the association of season with uptake by biota is not known. Uptake by higher trophic level organisms is a function of prey availability and mercury concentration in prey. Both of these factors fluctuate with season. In top trophic level organisms that have bioaccumulated mercury for several years, however, seasonal fluctuations in mercury concentration are less apparent.

The load allocations focus on controlling all loads to the surficial sediment of Clear Lake, instead of loads entering in a particular season. This focus was determined for two reasons. One reason is the lack of information regarding effects of season on bioaccumulation. The second reason is that Regional Board staff assumes that all mercury entering Clear Lake has the potential to be methylated. Controlling the effects of season on loading rates, such as the effects of precipitation on maximum daily loading rates from the tributaries or SBMM waste rock dam, will be addressed during implementation of the TMDL.

6 LOAD ALLOCATIONS AND REASONABLE ASSURANCE OF MEETING LOADS

Methylmercury production was described as a function of inorganic mercury concentration in sediment and methylation efficiency. Given the current lack of understanding of methylation processes, we cannot expect to control methylation efficiencies in Clear Lake. The remaining option is to control inorganic mercury loads available for methylation, which are in the active layer of sediment.

As shown by the linkage analysis, meeting the fish tissue targets requires that concentrations of mercury in the active layer of Clear Lake sediment be reduced by 40% in order to reduce methylmercury concentrations in fish tissue by the equivalent amount. Reducing inorganic mercury concentrations in the active layer sediment by 50% provides a margin of safety for the considerable uncertainties in the linkage analysis. In order to meet the goal of reducing surficial sediment concentrations by 50%, ongoing contributions to the active layer of sediment must also be reduced by 50%. Using available data, Regional Board staff estimates that approximately 715 kg of mercury is present in sediment deposited annually to the lakebed. This estimate is based on surficial sediment samples and the annual deposition rate of sediment to the lake bottom. The best estimate available of the acceptable load in sediment that would meet the targets is approximately 358 kg/year. The total load reduction will come from reductions in ongoing inputs and by controlling mercury in sediment that is remobilized. An Implementation Plan for the following load allocations, including environmental and economic consequences, will be presented in the Staff Report for the Basin Plan Amendment. Load allocations are shown in Table 15.

6.1 Sulphur Bank Mercury Mine

The majority of load reductions will come from reducing inputs to surficial lakebed sediments from existing discharges and historical deposits from SBMM. The load allocation for SBMM is 49% of ongoing contributions to the active sediment layer from past and ongoing processes. The available data suggests that past and present processes at SBMM contribute a maximum of 695 kg/year to the active sediment layer. Based on this data, TMDL requires that the total input from SBMM to the active sediment layer be limited to 340 kg/year, which is the difference between acceptable load in sediment (358 kg/year) and the other allocations (18 kg/year). Existing discharges to the lake include local deposition of mercury fluxed into the air and mercury in groundwater and surface water inflows. While the load allocation for SBMM (49% of existing inputs to the active sediment layer from past and ongoing processes) will not change, estimates of the total load of mercury in the surficial sediment layer and loads from SBMM may be refined as new data is gathered.

Because mercury in groundwater from the mine site is preferentially methylated, the load from SBMM groundwater is limited to 0.1 kg/year. Mercury in acidic drainage from the mine site appears to have particularly high methylation rates in the drainage-affected area near the shore. Modeling of water currents indicates that mercury and methylmercury are carried from the eastern end of Oaks Arm to the rest of the lake. The USEPA Superfund Program has committed publicly to performing remediation to reduce mercury movement from the SBMM site, including routes through groundwater, surface water and erosion and atmospheric flux.

The rest of the reduction should come from limitations in input from other ongoing discharges and resuspension of mercury deposited in prior years. Remobilization of mercury deposited in the past,

particularly in the area directly offshore of SBMM, has not been sufficiently investigated. The USEPA Superfund Program should examine and address the effects on existing sediment concentrations of past loading of mercury to the lake from the mine site. The implementation plans for reductions at SBMM will come from the Superfund Program Remedial Investigation and Feasibility Study Reports (RI/FS) and subsequent Records of Decision for Operable Unit 1, the mine site itself, and future Operable Units, which will encompass the North Wetlands, lakebed sediments, and biota of Clear Lake.

6.2 Atmospheric Inputs

The allocation for atmospheric deposition is capped at the maximum load estimated to accumulate from the global atmospheric pool, 2 kg/year. Atmospheric mercury originating outside of the Clear Lake watershed is considered to be uncontrollable under this TMDL. Mercury from SBMM that fluxes into the air and deposits locally should be controlled by USEPA Superfund remediation activities.

6.3 Tributaries

The load allocation of mercury from tributaries and direct surface water runoff is 90% of existing input. Mercury inputs from tributaries and surface runoff will vary with precipitation and water flow. Therefore, the load allocation is expressed as a percentage of the load anticipated for each water year. For an average water year, the load allocation is 16 kg/year. Sediment from tributaries contains less mercury per unit sediment than in lakebed sediment. It is proposed to allow cleaner sediment to cover the more contaminated sediment in the lake. Nevertheless, reducing total mercury loads to Clear Lake is required under the TMDL program.

Tributaries to Clear Lake are a source of mercury and methylmercury. As is shown in the next section, they are also a source of sediment to Clear Lake. Reducing loads of mercury and methylmercury from the tributaries should focus on identifying upstream sources of mercury and, if possible, controlling releases from them. At this point, no upstream sources have been identified. There may be “hot spots” of mercury loading within the tributaries that can be eliminated. In the first phase of TMDL implementation, Lake County, U.S. Bureau of Land Management, and U.S. Forest Service will be asked to partner with the Regional Board to develop tributary monitoring plans to identify potential hot spots of mercury loading. In the second phase, load reductions for hot spots will be developed and implemented. Watershed restoration projects are expected to reduce overall sediment inputs to the lake, which will decrease input of mercury adhered to the sediment. The TMDL requires that ecosystem restoration or preservation projects on tributaries to Clear Lake must not increase loads of methylmercury beyond existing levels.

Total mercury loads from the tributaries are expected to decrease, due to an increase in erosion control and other activities in the Clear Lake watershed. These activities are designed to reduce sediment and nutrient loading to the lake, not to address mercury in particular. Regional Board staff encourages these activities because they will improve overall water quality in Clear Lake (Clear Lake is listed as impaired due to excess nutrient loading). One effect of balancing various water quality needs in Clear Lake may be that the length of time required for burial of contaminated sediments will lengthen, because sediment will accumulate less rapidly on the lakebed.

The U.S. Army Corps of Engineers, in partnership with Lake County, is in the feasibility study phase of an extensive ecosystem restoration project at the mouth of Middle Creek. The project would restore

wetland and open water habitat and eliminate one of the existing wild rice growing areas. The project is expected to reduce total mercury loads to Clear Lake by retaining about 40% of the sediment from the Scott's and Middle Creek watersheds (Jones & Stokes Associates, 1997). Restoration of wetlands also has the potential to increase methylmercury production and input to the lake, which is of significant concern to the Regional Board. Limited amounts of data on methylmercury from the existing rice fields and in Rodman Slough were gathered by UC Davis Clear Lake Environmental Research Center in 2000 (Suchanek, 2001c) and by Regional Board staff in 2001. The Regional Board staff is continuing to work the U.S. Army Corps of Engineers to ensure that requirements of the TMDL be met, specifically that methylmercury inputs not be increased.

Other activities to reduce sediment loads and improve overall water quality in Clear Lake are being conducted by Lake County and by watershed stakeholder groups. The Lake County Board of Supervisors recently enacted a grading ordinance for all construction projects and a shoreline ordinance (requires mitigation for development of any wetland or natural shorelines along the lake edge). Lake County is also working with farmers to apply best management practices during vineyard development. Streambank stabilization and erosion control projects under the direction of stakeholder groups are completed or in progress in Scotts and Middle Creek Watersheds.

Table 15. Existing Loads and Load Allocations for Mercury in Surficial Sediment of Clear Lake

	Existing Loads (kg/year) (a)	Load Allocation	Acceptable Load under TMDL (kg/year)
Deposition from the global atmospheric pool	2	(no change)	2
Tributaries and direct surface water runoff	18	90% of existing loads	16
Sulphur Bank Mercury Mine (includes estimates of ongoing inputs and remobilization of mercury previously deposited in the lake)	Maximum estimated input: 695	48% of existing loads	340
Totals (Load allocation is 50% of existing loads)	715		358

(a) Existing and acceptable loads are based upon best available data for mercury in surficial sediment of Clear Lake. Meeting the numeric targets requires that surficial sediment concentrations and, therefore, total loads to surficial sediment be reduced by 50%. Acceptable loads may be revised as better data becomes available.

6.4 Active Layer of Lakebed Sediment

Surficial sediment is a source of mercury that becomes methylated. Under existing conditions, annual load to surficial sediment is estimated to be about 715 kg for the entire lake (See Section 3.3.1). The sources of the surficial sediment load are new inputs (SBMM, atmosphere, and tributaries and surface water runoff) and resuspension and deposition of mercury loaded into the lake in previous years. An objective of the TMDL will be to reduce sediment concentrations to half of existing concentrations. This reduction in would be achieved by reducing loads to half of the existing load estimate, or to loads 358 kg/year. In comparison, the sediment load prior to mining and use of heavy equipment in the region is estimated to be 120 kg/yr (Section 3.3.1). Decreases in surficial sediment load will be achieved by reducing the loads from various inputs and allowing cleaner, incoming sediment to bury the contaminated material.

How much, if any, historically contaminated sediment and mine waste rock can be feasibly and effectively dredged is unknown. The USEPA Superfund Program will evaluate effects of mercury in the lake as part of the Feasibility Study for SBMM Superfund Site Operable Unit 2, the environment beyond the terrestrial mine site. It is possible to assume that for much of the lakebed, however, dredging will not be feasible or cost effective. In that case, cleanup of the sediment available for methylation (surficial sediments) will rely upon burial of the existing sediments under less contaminated material. As shown in Table 16, mercury concentrations in incoming sediment from the tributaries are considerably less contaminated than surface sediments in the lake.

Levels of mercury in incoming sediment can be estimated by examining the sediment mercury concentrations in depositional zones at the mouths of tributaries. Fine-grained sediments at the mouths would presumably have been deposited during the previous winter's flows. Mercury on suspended and fine, deposited sediments can be compared with levels of mercury in lake surficial sediments. In April 2001, Regional Board staff collected fine-grained sediments from depositional zones at the mouths of three tributaries to the Upper Arm. Mercury concentrations in deposited sediment from the tributaries are shown in Table 16. The streambed sediment concentrations from 2000 are comparable with previously published values (Varekamp and Waibel, 1987).

Table 16. Mercury Concentrations in Sediment Samples from the Mouths of Clear Lake Tributaries Compared with Lake Sediment Concentrations

Site	total mercury, mg/kg dry wt	
	2001 (a)	1987 (b)
Upper Arm Tributaries:		
Cole Creek	0.141	na
Kelsey Creek	0.058	0.046
Rodman Slough @ Nice-Lucerne Cutoff	0.072	0.044 - 0.184
Morrison Creek	na	0.119
Unnamed tributary to north side of Narrows	na	0.283
Shindler Creek (tributary to Oaks Arm)	na	0.73
Lakebed sediment concentrations from cores collected September 2000 (c)		
Surface concentrations (represents existing loads)		
Upper Arm UA-03	2.5	
Lower Arm LA-03	4	
Oaks Arm OA-04 (c)	45	
Average concentration prior to mining activity in the watershed		
Upper Arm UA-03	0.3	
Lower Arm LA-03	1	
Oaks Arm OA-04 (d)	5	

a. Regional Board staff focused on tributaries with sufficient fine-grained sediment for sampling. Tributary samples collected April 2001.
b. Varekamp, 1987.
c. Suchanek et at., 2001. Unpublished data collected for the Regional Board. See Appendix D of the Source Analysis
d. Oaks Arm sediment concentrations range from more than 100 ppm near the mine site to approximately 20 ppm near the Narrows.

6.5 Monitoring Plan

An essential element of the TMDL is a monitoring plan. The goal of monitoring is to measure whether loads have been reduced and to track progress in meeting the targets. A monitoring plan for Clear Lake should include:

6.5.1 Fish Tissue.

Fish tissue sampling should be conducted on two levels. One effort should focus on young fish that remain in a relatively defined home territory. Young fish are desired because their methylmercury uptake is largely the result of recent exposure. Juvenile fish will more quickly reflect changes in mercury bioavailability than will larger or older fish, which integrate mercury uptake across years and large spatial areas. Young-of-the-year largemouth bass and inland silversides are recommended for this effort. The largest silversides, greater than 65 mm in length, may be older than one year and should not be used. Some baseline data for these species have been collected by UC Davis CLERC (Suchanek et al., 2000a; Suchanek et al., 1997). The baseline data set should be expanded in order to understand individual and inter-annual variabilities in mercury concentrations.

Mercury levels should also be measured in fish of the species and sizes frequently consumed by humans. Largemouth bass and channel catfish are recommended because they are at the top of the aquatic food web, are regularly consumed and have the most extensive historical data set of mercury concentrations. Larger fish can be effectively sampled every 10 years. Because adult fish integrate methylmercury levels over a lifetime and changes in total sediment mercury concentrations are not expected to be discernable for more than five years, more frequent sampling of sport fish is not necessary. In order to remove the fish tissue advisory, presumably mercury levels would need to be evaluated in other species popular for sport fishing.

6.5.2 Sediment.

Total mercury and methylmercury sediment concentrations throughout the lake should be evaluated regularly, preferably on the same time schedule as small fish. Levels of total mercury in sediment can be used to indicate whether loads have diminished. Methylmercury levels in sediment will be compared with fish tissue levels to assess changes in methylation efficiency. Existing sediment data should be evaluated to determine if there is an adequate baseline of information. A profile of sediment concentrations in Oaks Arm with respect to distance from the mine site should be obtained for current conditions. The most recent profile of surficial sediment concentrations in Oaks Arm was completed in 1994-96. A better understanding is needed of sedimentation patterns, especially in Oaks Arm. Short cores of sediment should be collected in Oaks Arm to determine to what extent the waste rock and tailings pushed into the lake are being eroded or covered with sediment.

For all data collection effort described above, some baseline data is available. The existing data must be evaluated by a statistician for completeness, understanding variability in the study population and to design future collections. Statistical analysis is critical to being able to assess whether load reductions have decreased fish tissue levels. For example, more years of data are needed if the variability between yearly averages is thirty percent versus fifteen percent.

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APPENDIX A. MERCURY IN CLEAR LAKE FISH

Levels of mercury in various fish species are reported below as the average and standard deviation of total mercury per wet weight of tissue. Raw data collected by Department of Fish and Game, Department of Health Services and Central Valley Regional Water Quality Control Board (CVRWQCB, 1995) and the UC Davis-Clear Lake Environmental Research Center (Suchanek et al., 1993; Suchanek et al., 1997) were tabulated and combined for statistical analyses. Although the data were collected over a time period of 25 years, there are no obvious trends in mercury concentrations between the older and more recently collected data.

Abbreviations for fish species used in the following graphics are:

ILSS	inland silverside
juv LMB	juvenile largemouth bass
BG	blue gill
BLB	black bullhead
SBF	Sacramento blackfish
BB	brown bullhead
BC	black crappie
WCF	white catfish
WCR	white crappie
LMB	largemouth bass
CCF	channel catfish

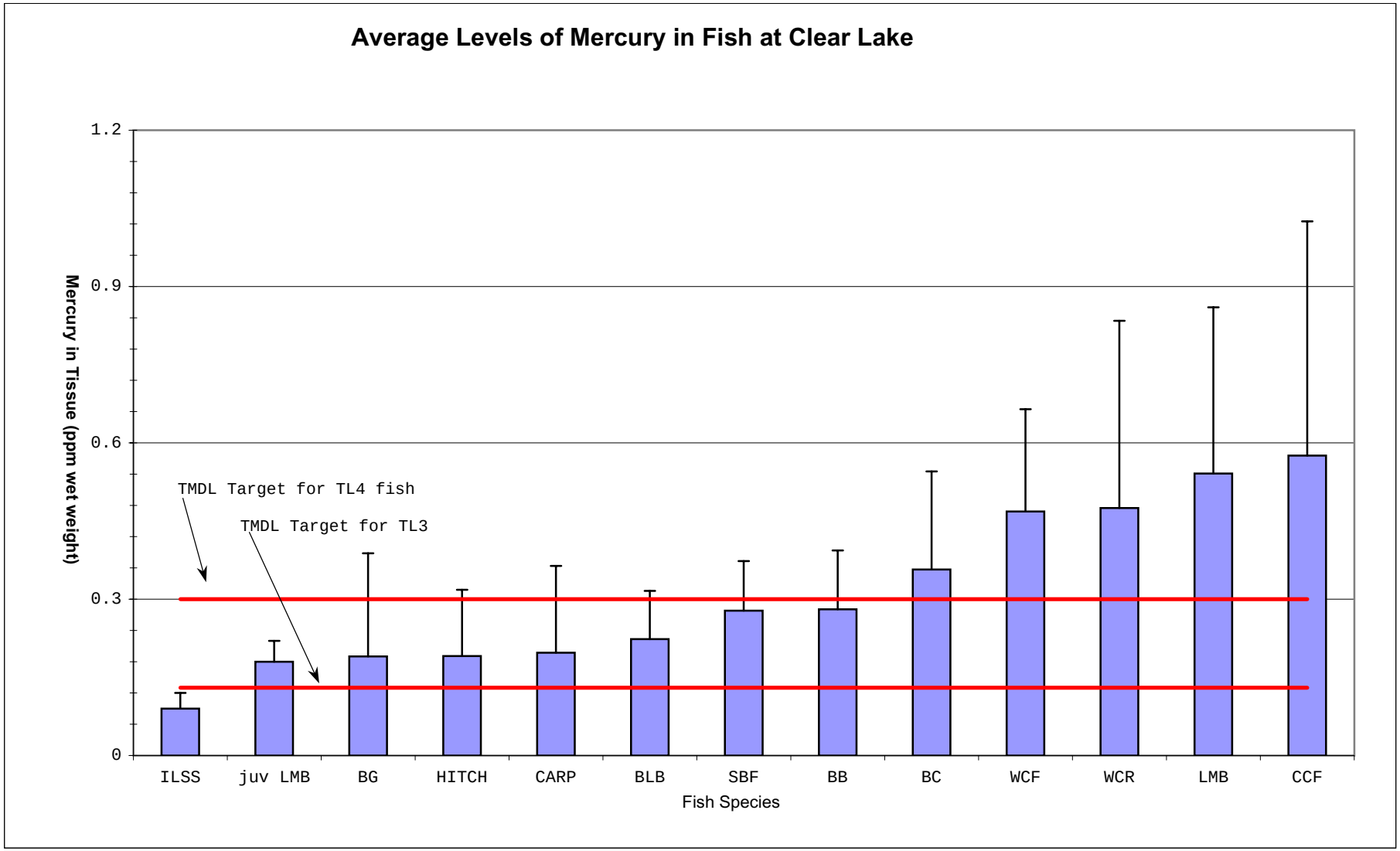
For inland silversides and juvenile largemouth bass, data reported is for size classes of 65 - 80 mm and 100-200 mm, respectively. These are size classes likely eaten by small, pisciverous birds.

Table A.1. Forklengths of Fish Caught by Anglers in March through June 1988 at Clear Lake

Species	Length Range (mm)
Black crappie	115 – 310
Bluegill	115 – 200
Brown bullhead	300 – 430
Channel catfish	390 – 780
Green sunfish	118 – 163
Largemouth bass	190 – 590
White catfish	245 – 455
White crappie	180 – 330

Source: (Macedo, 1991)

Figure A-1. Average Levels of Mercury in Fish at Clear Lake



Concentrations of Mercury in Clear Lake Fish

Sources: CVRWQCB, 1985; Suchanek et al., 1993; Suchanek et al., 1997

Table A-2. Mercury Tissue Concentrations in Clear Lake Fish, by Species (ppm wet weight):

Fish Species	ILSS	juv LMB	bluegill	hitch	carp	BLB	SBF	BB	black crappie
Mean	0.09	0.18	0.19	0.19	0.20	0.22	0.28	0.28	0.36
Standard Deviation	0.03	0.04	0.20	0.13	0.17	0.09	0.10	0.11	0.19
Fish Species	white crappie	channel catfish	white catfish	LMB					
Mean	0.48	0.48	0.51	0.54					
Standard Deviation	0.36	0.37	0.18	0.32					

Table A-3. Mercury Concentrations in Fish Categories Most Commonly Consumed by Humans (ppm wet weight):

Fish Species	Average Concentration of Trophic Level 3 (includes bluegill, hitch, carp, Sacramento blackfish and black bullhead; catfish less than 250 mm forklength and largemouth bass 150-175 mm) (a)	Average concentration of TL4 fish. (Includes black crappie and white crappie longer than 140 mm forklength; brown bullhead, white catfish and channel catfish longer than 250 mm; and largemouth bass longer than 175 mm)
Mean	0.22	0.50

(a). (Small bass and catfish categorized as TL3 fish because of probable prey consumed by these sizes of fish)

Table A-4. White and Channel Catfish Mercury Concentrations by Length Range (ppm wet weight)

	catfish length (mm)								
	126-250	251-300	301-350	351-400	401-450	451-500	501-550	600-655	701-750
Mean	0.29	0.40	0.39	0.41	0.30	0.39	0.51	1.26	0.97
Standard Deviation	0.18	0.19	0.20	0.15	0.12	0.21	0.15	0.22	0.52

Table A-5. Largemouth Bass Mercury Concentrations by Length Range (ppm wet weight)

	largemouth bass length (mm)								
	144-200	201-250	251-300	301-350	351-400	401-450	451-515		
Mean	0.32	0.41	0.46	0.47	0.56	0.73	1.14		
St Dev	0.15	0.18	0.20	0.34	0.26	0.40	0.41		
Std. Dev.	0.15	0.18	0.20	0.34	0.26	0.40	0.41		

APPENDIX B. ASSESSMENT OF HUMAN EXPOSURE TO MERCURY AT CLEAR LAKE

Following is summary data from the California Department of Health Services study of mercury exposure in Members of the Elem Tribe Elem and some non-tribal neighbors of the Sulphur Bank Mercury Mine (Harnly et al., 1997). The study was conducted in November 1992 by Harnly and colleagues. Sixty-three Tribal Members (46% of the resident tribal population) and four residents of neighboring, non-native homes participated in biological monitoring. The first centimeter of hair closest to the scalp was analyzed. For sport and commercial fish consumption, respondents were asked what type they had consumed over the past six months, the estimated average number of times per week they ate the fish, and the average amount (in pounds) they ate at each meal. Children under ten years of age were interviewed with their parents. Levels of mercury in blood and hair of study participants are shown in Table B-1.

Note that this study was conducted after the fish consumption advisory was issued. Traditional consumption rates were likely higher and fishery use patterns may have been different in years prior to the consumption advisory.

Table B-2 shows average consumption rates as reported in the published paper. The text of the paper, however, states that 90% of those interviewed consumed at a rate equal to or less than 30 g/day. It is clear from this statement that the average consumption rates were heavily influenced by the amounts of fish eaten by a few, high-consuming individuals. Because the numbers of study participants that reported eating commercial and Clear Lake fish differed, the average consumption rates of fish from both sources cannot be added to determine consumption of all fish.

Table B.1 Biological Mercury Levels Among CDHS Study Participants (Harnly et al., 1997)

Participants	N	Mean	Minimum	Maximum	Standard Deviation
Blood inorganic mercury (μg/L)					
Tribal Members	44	2.9	0.7	4.7	1.0
Others	4	2.7	1.7	3.4	0.8
Blood organic mercury (μg/L)					
Tribal Members	44	15.6	3.3	38.8	6.6
Others	4	8.8	2.5	12.2	6.9
Hair mercury (μg/g) (a)					
Tribal Members	63	0.64	0.3	1.8	0.43
Others	4	1.6	0.3	2.3	0.88

(a). For samples less than the detection limit (0.3 – 0.6 μg/g), the value was taken as 0.3 μg/g.

Table B-2. Fish Consumption Rates Among Consumers in CDHS Study at Clear Lake.		
	Number of individuals reporting consumption	Average consumption among consumers (g/day)
Clear Lake fish		
Total of Clear Lake fish	23	60
Catfish	19	53
Hitch	4	12
Perch	4	74
Bass	2	5
Carp	1	1
Commercial fish		
Total of commercial fish	32	24
Tuna	15	9
Salmon	12	9
Crab	8	9
Snapper	5	10
Shrimp	5	6

APPENDIX C. CLEAR LAKE INFLOW AND OUTFLOW

Following is a water budget for Clear Lake, including tributary inflow and Cache Creek outflow, for water years 1990 to 1999. A water year begins 1 October of the previous year to 30 September of that year. The consecutive water years were chosen for data availability and because they include drought years as well as wet years.

Flow gauges are operated on three of the main tributaries to Clear Lake (Kelsey, Middle and Scott's Creeks) and on Cache Creek just downstream of the Cache Creek Dam. The dam is approximately thee miles downstream of the outlet of Clear Lake. Flow gauges on Kelsey and Cache creeks are operated by USGS. Data was accessed from the US Geological Survey (USGS) water homepage (http://water.wr.usgs.gov/). Department of Water Resources (DWR) operates flow gauges on Middle and Scott creeks as well as a rain gauge at Clear Lake Highlands. DWR's northern field office provided the flow data, which is preliminary and has not yet been reviewed for accuracy.

Watershed sizes for Middle, Kelsey and Scott's creeks account for 43% of the entire Clear Lake Basin. Runoff into the gauged streams was assumed to be proportional to runoff in the ungauged portion of the watershed. To determine total tributary inflow, yearly average flow from the three creeks was totaled and added to the estimated flow from the remaining 57% of the ungauged creeks for each of the water years.

Raw data on mercury concentrations and water flow used to calculate regression equations for estimating tributary loads are shown in Table C-2. The mercury and methylmercury concentration data were provided by Dr. Tom Suchanek and staff of the UC Davis Clear Lake Environmental Research Center.

Table C-1. Annual Flow Rates for Clear Lake Tributaries and Watershed

	Average Annual Flow (l/s)									
	Water Year									
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Kelsey Creek	674	968	802	2487	632	4633	2135	2585	4089	2285
Scott's Creek	377	754	794	3072	493	5528	3044	3146	6140	2486
Middle Creek	518	517	815	2587	484	3653	2457	1311	6313	2363
Total Gauged Streams 43.4%	1569	2240	2411	8147	1609	13814	7636	7042	16543	7134
Total Ungauged Streams 56.6%	2046	2921	3144	10625	2098	18016	9958	9183	21574	9303
Total	3615	5161	5555	18771	3707	31830	17594	16225	38117	16437
Cache Creek Outflow	377	754	794	3072	493	5528	3044	3146	6140	2486
Change in Inflow to Outflow	3238	4407	4761	15699	3214	26302	14550	13079	31977	13951

Table C-2. Mercury Concentrations and Flow rates Used to Determine Tributary Stream Loads

Kelsey Creek	Q (l/s)	Hg (ng/L)	MMHg (ng/L)	Filt Hg (ng/L)	Filt MMHg (ng/L)
2/19/98	38515.2	29.5	0.184	2.00	NA
5/21/98	1104.5	0.829	0.0318	0.377	0.0318
11/30/98	35400.0	57.4	0.300	7.53	0.0469
5/23/00	906.2	1.24	0.0507	1.24	0.0250
1/26/01	3625.0	8.20	0.0554	4.98	0.0504
Scott's Creek	Q (l/s)	Hg (ng/L)	MMHg (ng/L)	Filt Hg (ng/L)	Filt MMHg (ng/L)
2/19/98	68534.4	17.9	0.103	1.52	NA
5/21/98	906.24	0.962	0.0318	0.449	0.0318
11/30/98	13140.48	16.4	0.145	4.90	0.0428
5/23/00	192.576	2.83	0.649	1.20	0.386
1/26/01	4276.32	13.5	0.0859	4.33	0.0377
Middle Creek	Q (l/s)	Hg (ng/L)	MMHg (ng/L)	Filt Hg (ng/L)	Filt MMHg (ng/L)
2/19/98	42480	108	0.384	3.56	NA
5/21/98	1161.12	0.650	0.0706	0.456	0.0318
11/30/98	7929.6	82.4	0.502	6.94	0.0473
5/23/00	3001.92	1.41	0.0927	0.933	0.0304
1/26/01	1614.24	3.51	0.0323	2.57	0.025
Cache Creek	Q (l/s)	Hg (ng/L)	MMHg (ng/L)	Filt Hg (ng/L)	Filt MMHg (ng/L)
2/19/98	205320	7.50	0.418	1.13	NA
5/21/98	14641.4	7.50	0.307	0.737	0.0104
11/30/98	161.42	5.84	0.272	1.98	0.0338
5/23/00					
1/26/01		30.3	0.154	5.17	0.0773

Source: T. Suchanek, previously unpublished data
Hg: Mercury; MMHG: Monomethylmercury; Filt: Samples were passed through a 0.45 micron filter before analysis.

APPENDIX D. CLEAR LAKE SEDIMENT CORES AND SURFICIAL SEDIMENT CONCENTRATIONS OF MERCURY

Deep sediment cores were collected by the UC Davis Clear Lake Environmental Research Center in 1996 and 2000 (Suchanek et al., 1997; unpublished data collected for the Regional Board, 2000). Core sections were analyzed for mercury and other chemical constituents. The 1996 cores were dated using concentrations of lead-210. Dates for the 2000 cores are still being verified. Each core shows slightly different sedimentation rates.

Mercury concentrations in 1996 cores are shown in Figure D-1. Sharp increases in mercury levels in each core correspond to an estimated date of 1927, which was the beginning of open pit operations at SBMM. This period also corresponded to increases in sediment, total organic carbon and other parameters. Mercury concentrations peaked around an estimated date of 1961.

Mercury concentrations in sediment cores from 2000 are shown in Figure D-2 and in Table D-2. Cores were collected at approximately the same locations as the 1996 cores. Mercury levels do not appear to have declined during this period. Except for the Lower Arm cores, patterns of mercury concentration are generally the same between 1996 and 2000 cores when compared for each site. The Lower Arm core collected in 2000 exhibits a dramatic increase in mercury in the top ten centimeters of the core but does not show a peak in deeper portions of the core. A possible explanation for this discrepancy is that the top sediment layers were eroded away or somehow removed prior to collection of the LA-03 in 2000. Collection and analysis of the 2000 cores were conducted under a detailed Quality Assurance Project Plan approved by the Regional Board. Note also that the shape of the mercury profile in duplicate cores OA-03a and OA-03b is the same but the cores have different apparent sedimentation rates. Precise dating of these cores will determine whether one duplicate was compressed during processing or whether the difference is real.

The deep sediment cores indicate that mercury concentrations have declined somewhat since the cessation of mining at SBMM. Regional Board staff determined rough estimates of the rates of decline in each Arm using the following procedure. Figure D-3 shows near-surface portions of the deep sediment cores in greater detail in order to obtain slopes of the line (concentration/depth). The slope of the line was obtained for the portion of each core through which declines in mercury concentrations appear approximately linear. Assuming a uniform sedimentation rate of 0.9 cm/year, the linear slopes were converted into units of concentration/year. Rates of decline were averaged for two or more cores for each arm and are shown in Table D-1.

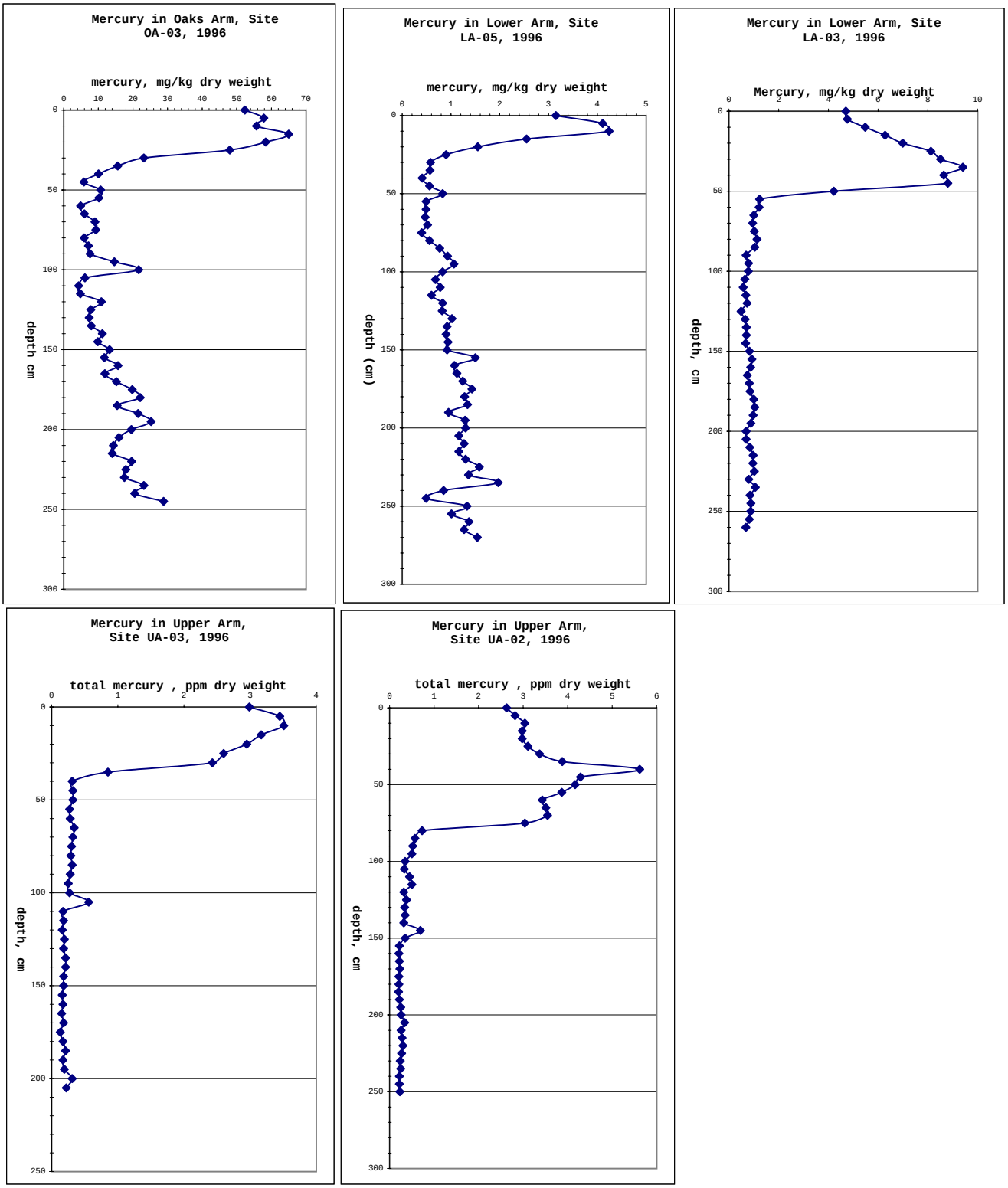
These rates of decline should be treated as semi-quantitative estimates, because of the small range of declines seen in some cores. Because of bioturbation and mixing of surface sediments by currents, the concentration at any depth is an integration of mercury deposited for a time period before and after that date. There is essentially no difference between concentrations of mercury in surface sediment of cores collected in 1996 and 2000.

Table D-1. Estimated Rates of Decline in Mercury Concentrations in Clear Lake Sediments

Site and year of core	Depth of sediment core showing generally consistent decline in mercury concentration (cm)	Estimated rate of decline (slope of the line on a plot of sediment depth versus mercury concentration, ppm/cm)	Estimated rate of decline (slope of the line converted to ppm/yr, assuming 0.9 cm/yr sedimentation rate)
LA03, 1996	0 - 35	0.14	0.13
LA05, 1996	0 - 10	0.11	0.099
UA02, 1996	0 - 35	0.030	0.027
UA03, 2000	0 - 40	0.047	0.042
OA03, 1996	0 - 15	0.72	0.97
OA03, 2000A	0 – 17.5	2.19	1.3

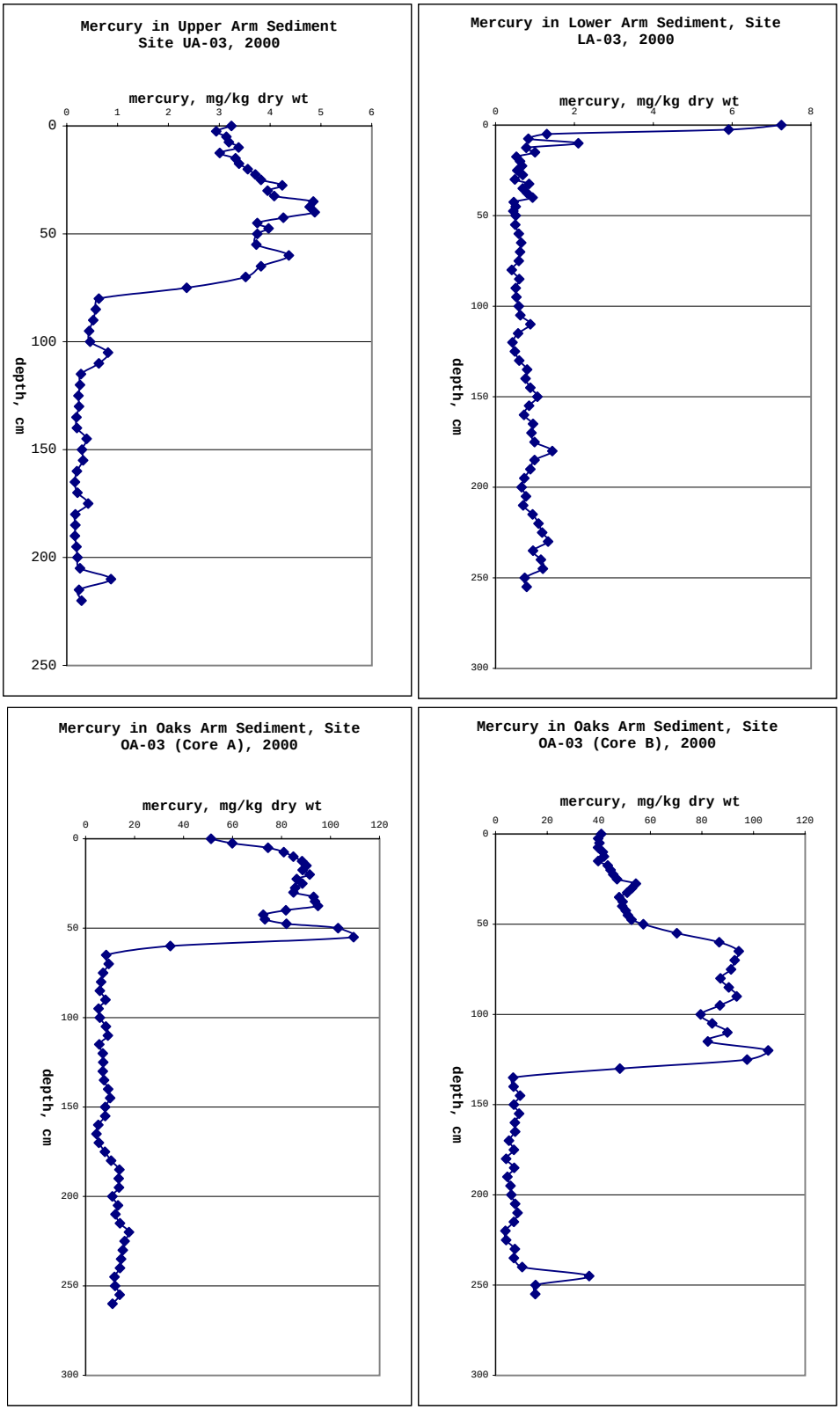
Mercury has also been measured in surficial sediments by the UC Davis Clear Lake Environmental Research Center (Suchanek et al., 1993; 1997). A large number of samples were collected in 1994-96, after the remediation of waste rock piles on the Clear Lake shoreline. Mercury levels in surficial sediments showed a statistically significant decline as a function of distance from the SBMM. Surficial sediment just offshore of the mine site contained approximately 300 ppm of mercury; sediment from sites in the Oaks Arm contained approximately 40 ppm of mercury. Mercury concentrations in surficial sediment ranged from 10 to 15 ppm in the Narrows, and from 0 to 5 ppm in the Upper and Lower Arms (See Figure D-4).

Figure D-1. Mercury in Sediment Cores Collected in Clear Lake, 1996



Source: Suchanek et al., 1997.

Figure D-2. Mercury in Sediment Cores Collected in Clear Lake, 2000



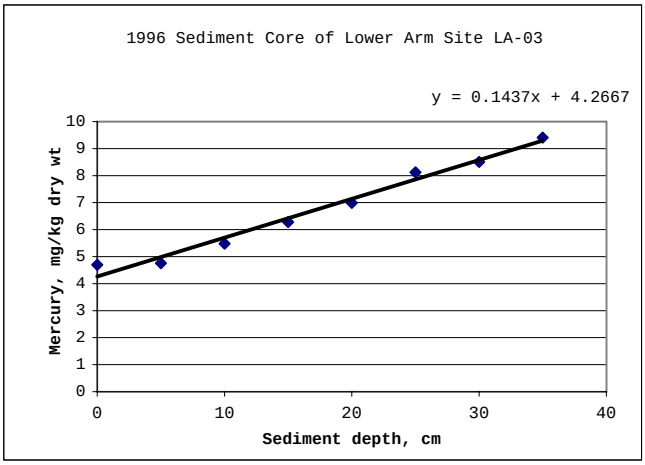
Source: T, Suchanek, Unpublished data collected for the Regional Board. Data is shown in Table D-2. Core LA-03 may not include the top sediment layer, due to malfunction of the core sampler. Lead 210 dating of the cores is in process. Dates will indicate whether one of the Oaks Arm cores may have been compressed or stretched relative to the other Oaks Arm core during sample collection.

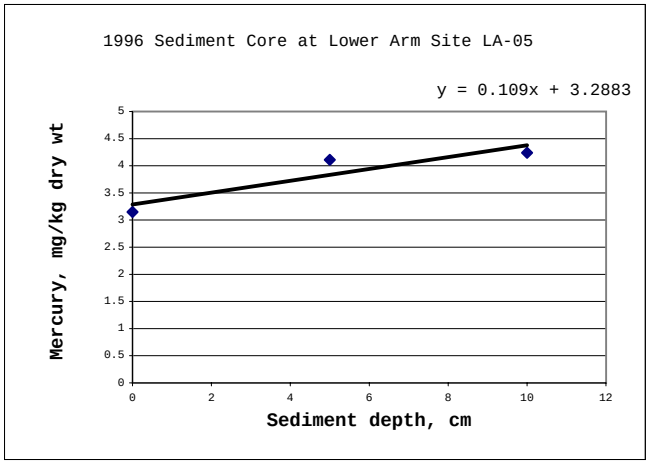
Table D-2. Concentration of Mercury in Deep Sediment Cores from Clear Lake. Cores were collected in September 2000 by the UC Davis Clear Lake Environmental Research Center for the Regional Board.

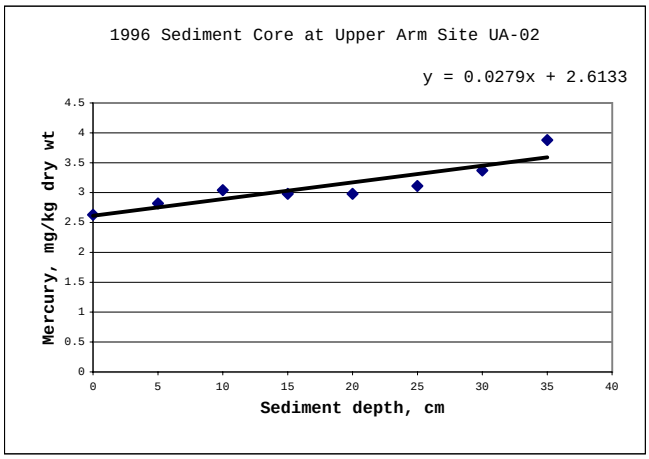
	Mercury Concentration, mg/kg dry weight			
depth (cm)	UA-03	LA-03 (*)	OA-03 (Core A)	OA-03 (Core B)
0	3.24	7.25	51.2	41
2.5	2.94	5.91	59.9	39.8
5	3.14	1.3	74.5	40.3
7.5	3.19	0.83	80.9	39.7
10	3.38	2.1	84.8	41.6
12.5	3.01	0.78	88.4	42
15	3.32	1	90.2	39.8
17.5	3.39	0.53	88.6	43.5
20	3.56	0.62	91.5	44.6
22.5	3.71	0.67	86.2	45.6
25	3.82	0.55	88.5	47.1
27.5	4.24	0.69	85.6	54.4
30	3.95	0.49	84.9	52.9
32.5	4.08	0.85	93.1	51
35	4.85	0.69	93.7	47.9
37.5	4.78	0.8	94.9	49.3
40	4.88	0.94	81.8	49.1
42.5	4.26	0.46	72.5	50.5
45	3.75	0.51	73.2	51.3
47.5	3.97	0.45	82	52.7
50	3.75	0.51	103.1	57.3
55	3.73	0.5	109.4	70.3
60	4.37	0.59	34.6	86.7
65	3.82	0.65	8.4	94.3
70	3.52	0.62	9.4	92.7
75	2.36	0.59	7.1	91.3
80	0.63	0.41	6.3	87.2
85	0.57	0.6	5.8	90.4
90	0.52	0.51	8.1	93.5
95	0.44	0.53	5.3	87
100	0.46	0.59	5.8	79.5
105	0.81	0.63	8.3	84
110	0.63	0.88	9.1	89.9
115	0.28	0.57	5.6	82.3
120	0.26	0.43	7	105.7
125	0.23	0.49	7.2	97.5
130	0.24	0.6	7	48.2
135	0.19	0.8	7.5	6.8
140	0.2	0.76	9.2	7
145	0.39	0.88	10	9.5
150	0.3	1.06	8	7.1
155	0.32	0.85	8	9.1
160	0.2	0.72	5.2	7.5
165	0.16	0.95	4.4	7.6
170	0.21	0.91	5.4	5.2
175	0.42	0.99	7.9	7.1
180	0.17	1.44	10.4	4.1
185	0.17	0.99	13.8	7.2
190	0.16	0.88	13.5	4.6
195	0.19	0.73	13.6	5.8
200	0.21	0.66	10.9	6.1
205	0.26	0.77	13.2	7.6
210	0.87	0.7	12.2	8.5
215	0.24	0.94	14	7.1
220	0.29	1.09	17.7	3.8
225		1.18	15.9	4.1
230		1.33	15.2	7.5
235		0.95	14.5	7.1
240		1.15	14	10.3
245		1.2	11.8	36.3
250		0.74	12	15.5
255		0.79	13.9	15.4
260			11	

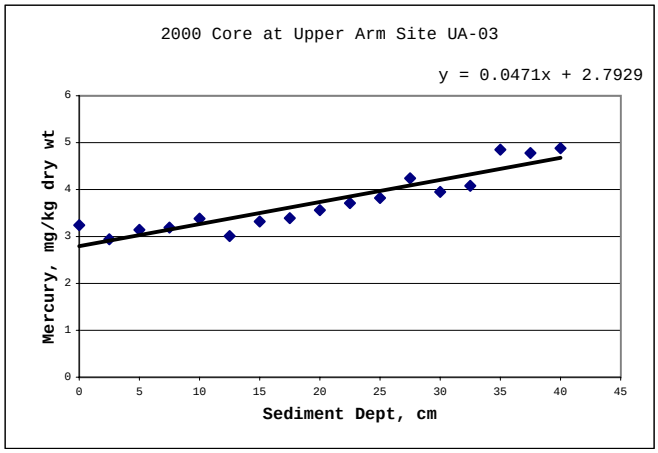
* See notes on Figure D-2

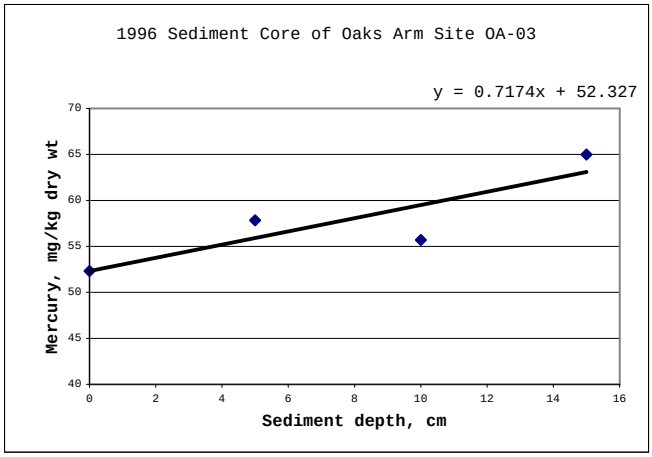
Figure D-3. Near-surface Sections of Sediment Cores Collected 1996 and 2000, Showing Approximate Rates of Decline in Mercury Concentrations

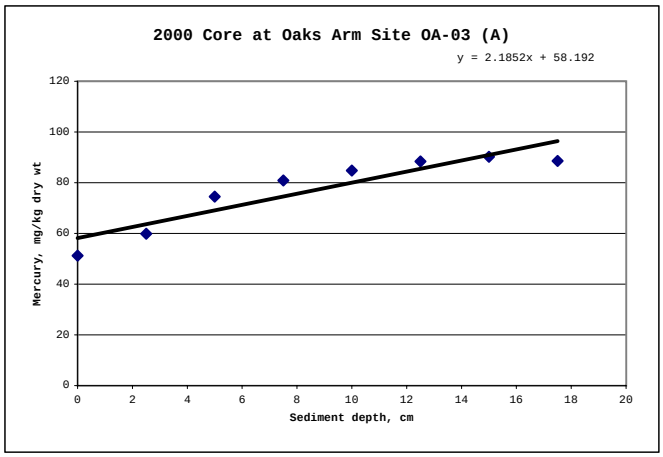












Suchanek et al., 1997 and unpublished data collected for the Regional Board

Figure D-4. Mercury in Surficial Sediment as a Function of Distance from Sulphur Bank Mercury Mine

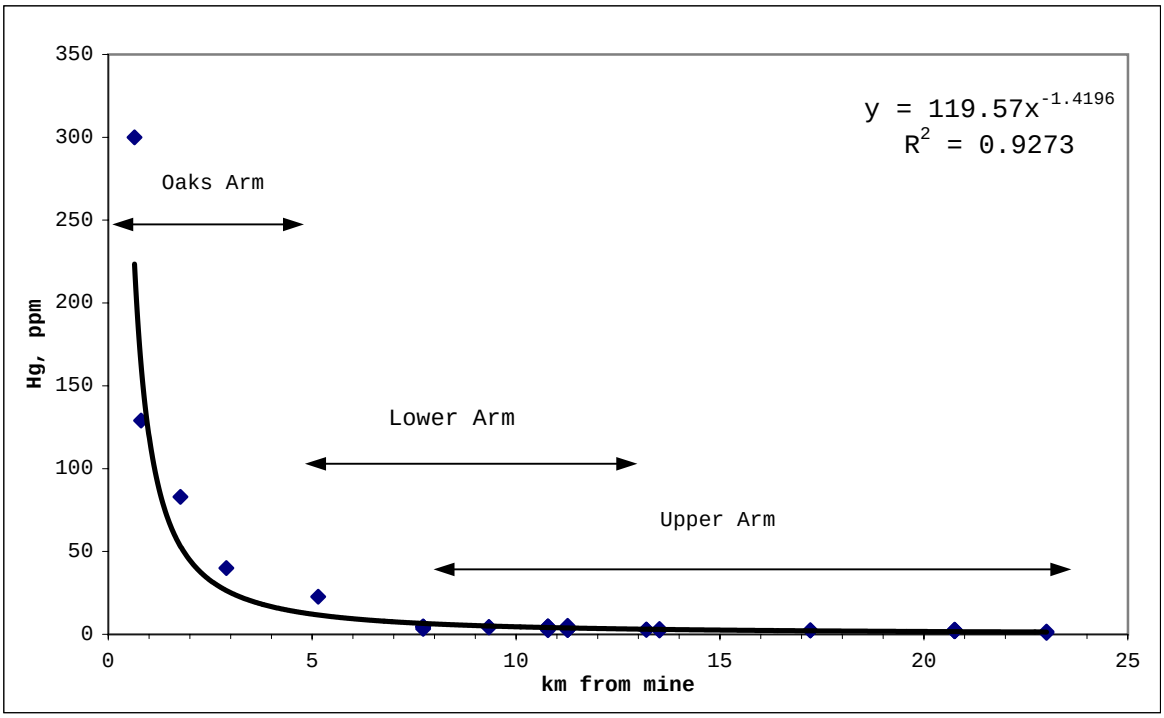


Figure shows approximate distances for each Arm from the SBMM Superfund site at the east end of Oaks Arm. Particle transport distance may be longer. This is particularly true for particles entering the Lower Arm, which may first circulate in the Upper Arm (Lynch and Schladow, 1996).

APPENDIX E. MERCURY CONCENTRATIONS AND LOADS IN THE MIDDLE CREEK/RODMAN SLOUGH WETLAND AREA

The wetland area at the northern end of Clear Lake receives water from the Middle, Scott's, Cooper and Clover Creek watersheds and agricultural drainage. It empties into Clear Lake through Rodman Slough. This area is of particular concern for the TMDL because of the potential for mercury methylation in the wetlands and in flooded fields irrigated for wild rice production. There are several areas that are used periodically or annually to produce wild rice. Water pumped out of the wild rice fields eventually enters Clear Lake. The wetlands and flooded agricultural fields may be a source of the methylmercury measured in water and biota of the Upper Arm. In comparison with concentrations in other Arms, methylmercury concentrations in the Upper Arm are unexpectedly high, relative to the amount of mercury in the sediment (Suchanek et al., 1997).

Following are data on methylmercury concentrations in the Rodman Slough and preliminary estimates of loads entering Clear Lake. Regional Board staff is continuing to work with the U.S. Army Corps of Engineers and Lake County Department of Public Works staff to refine the load estimates. Load estimates and an understanding of methylmercury production in the wetland area are necessary for feasibility studies of a restoration project in the area. The Middle Creek Ecosystem Restoration Project would restore wetlands and wildlife habitat by breaching existing levees and reconnecting Scott's and Middle Creeks to the historic floodplain. The first phase of the project, the Reconnaissance Study, is complete (Jones & Stokes Associates, 1997). The feasibility study is expected to be released by Spring 2002.

Preliminary estimates of loads of mercury and methylmercury were obtained using the same method as described in Section 1.1.2 for estimating total tributary loads (Table E-1). Mercury data were collected during five flow events on three gauged tributaries. Although Middle and Scott's creeks are gauged, the gauge on Scott's Creek is considerably further upstream than the wetland area. Contributions from the ungauged portions were estimated using a simple ratio of watershed area and assuming equal rates of runoff. Tributary load estimates should be refined using land use data, elevations and runoff coefficients.

Table E-1. Methylmercury Loads from Middle Creek/Scott's Creek/Rodman Slough Watershed

	Methylmercury load, kg/year			
	Average Water Year	10 Year Average, 1990-1999	10 Year Minimum	10 Year Maximum
Middle Creek	0.026	0.022	0.002	0.077
Scott's Creek	0.161	0.120	0.016	0.288
Ungauged portion of the Rodman Slough watershed, including Clover and Alley Creeks (a)	0.027766	0.020996	0.00272	0.053078
Total	0.215	0.163	0.021	0.418

a. Assumes that ungauged portions of the Rodman Slough watershed are one eighth of the ungauged portion of the entire Clear Lake watershed.

Methylmercury concentration data have been collected in the Middle Creek Ecosystem Restoration Project area for two years. Water sampling occurred in late summer during times that water from rice

fields was being pumped into Rodman Slough. In 2000, samples were collected by staff of the UC Davis Clear Lake Environmental Research Center and funded by the Regional Board (Suchanek, 2001c). The UC Davis report is amended in Appendix F and includes a site map. Samples were collected in 2001 by Regional Board staff (Table E-2). Water samples were collected of water draining from rice fields and at sites above and below the drains, if possible. Analyses were performed for mercury and methylmercury in unfiltered, "raw" water and water passed through a 0.45 micron filter.

Table E-2 Mercury and Methylmercury in Water in the Rodman Slough Area, August 2001

Date	Site (a)	Methylmercury, ng/L		Mercury, ng/L		Total Suspended Solids, mg/L
		filtered	raw	filtered	raw	
8/2/01	Highline Slough Pipe	0.0769	0.597	0.443	3.36	29
8/2/01	Central Rice Field	0.254	0.615	0.854	2.83	24
8/2/01	Middle/Scotts Confluence	0.156	1.36	0.604	4.54	49
8/2/01	Rodman Slough 04	0.0487	0.430	(na)	2.53	18
8/2/01	Rodman Slough 03	0.0492	0.320	(na)	3.1	22
8/2/01	Rodman Slough 02	0.0516	0.207	0.393	2.15	11
8/23/01	Tule Lake pump-out 1	0.260	0.524	0.911	5.06	58
8/23/01	Tule Lake pump-out duplicate	0.231	0.542	0.831	6.57	(na)
8/23/01	Middle/Scotts Confluence	0.113	1.24	0.529	4.02	24
8/23/01	Rodman Slough 04	0.135	0.830	0.605	4.47	32
8/23/01	Rodman Slough 03	0.0851	0.311	0.350	3.19	20
8/23/01	Rodman Slough 02	0.0634	0.309	0.421	4.65	40

(a). Highline Slough pipe is the pump station for the Central Wild Rice Field. The pump shut off just after collecting the first sample on 8/2/01, so an additional sample was collected from an irrigation ditch within the rice field, near the pump inlet. RS04 is approximately 200 feet upstream of Highline Slough. RS03 is between RS04 and RS02. RS02 is the most downstream site, just upstream of the Nice-Lucerne Cutoff bridge.

APPENDIX F. CLEAR LAKE ENVIRONMENTAL RESEARCH CENTER MERCURY STUDIES, DATA REPORT 5/10/01

Prepared for SWRCB CONTRACT 0-059-150-0
TASK 5. METHYLMERCURY PRODUCTION IN WETLANDS DRAINING TO RODMAN SLOUGH AND IN SHORELINE AREAS OF THE UPPER ARM OF CLEAR LAKE, Part 1: Rice Fields Adjacent to Clear Lake.

OBJECTIVE:

The objective of this task is to determine the contribution of methyl Hg from rice fields and wetland sites that input to Clear Lake through Rodman Slough. This includes the wetland areas of Tule Lake and Robinson Lake. Previous data has shown that water and organisms in the Upper Arm contain levels of methyl Hg that are unexpectedly high, relative to Hg in Upper Arm lakebed sediments. Possible sources of methyl Hg are wetlands and shallow water areas that drain into or communicate with the Upper Arm of Clear Lake. These sources previously have not been investigated rigorously.

METHODS:

STUDY AREAS: The rice field systems take water from, or release water to, Clear Lake depending on several factors and the flow of water varies annually. A significant proportion of the water in rice fields evaporates, thus reducing the need to pump the water out. All of the water that is pumped out of these fields enters Clear Lake eventually. Lake water circulates into and within the rice fields throughout a system of ditches incorporating check gates to regulate the flow of water. Farmers do not drain water off of the fields for the purpose of harvesting, which is accomplished with water in the fields, and water is not pumped out according to any schedule. Inflows and outflows are regulated to maintain a certain water level throughout the season, which is related to weather conditions. Harvesting of the crops is done during the late summer/early fall. Crop planting is typically done in the spring around the month of April.

SAMPLING EVENTS: Collections were made directly from (1) water flowing out of the rice fields when they were actively being pumped out, in instances when pumping was not occurring collections were made at the pump-out pipe inlet, (2) sites above the rice fields when possible and (3) sites below the rice fields. Property owners were contacted prior to site visits to collect samples. Approval was given for sampling in both areas. Samples were taken over several days during August and September 2000. No GPS equipment was available, thus no GPS coordinates were obtained.

Water samples were obtained from the following sites during the seasonal pump-out period in August and September 2000 as identified in Figure F-1 below:

Tule Lake Rice Field	TLR-01
Middle Creek/Scott's Creek Confluence	MSC-01
Highline Slough Pipe	HSP-01
Rodman Slough/Rice Field South	RSRFS
Rodman Slough	RS-02

RAW (unfiltered) water was analyzed for total Hg, methyl Hg, Total Suspended Solids (TSS) and Sulfate. FILTERED water (to 0.45 µm) was analyzed for total Hg and methyl Hg.

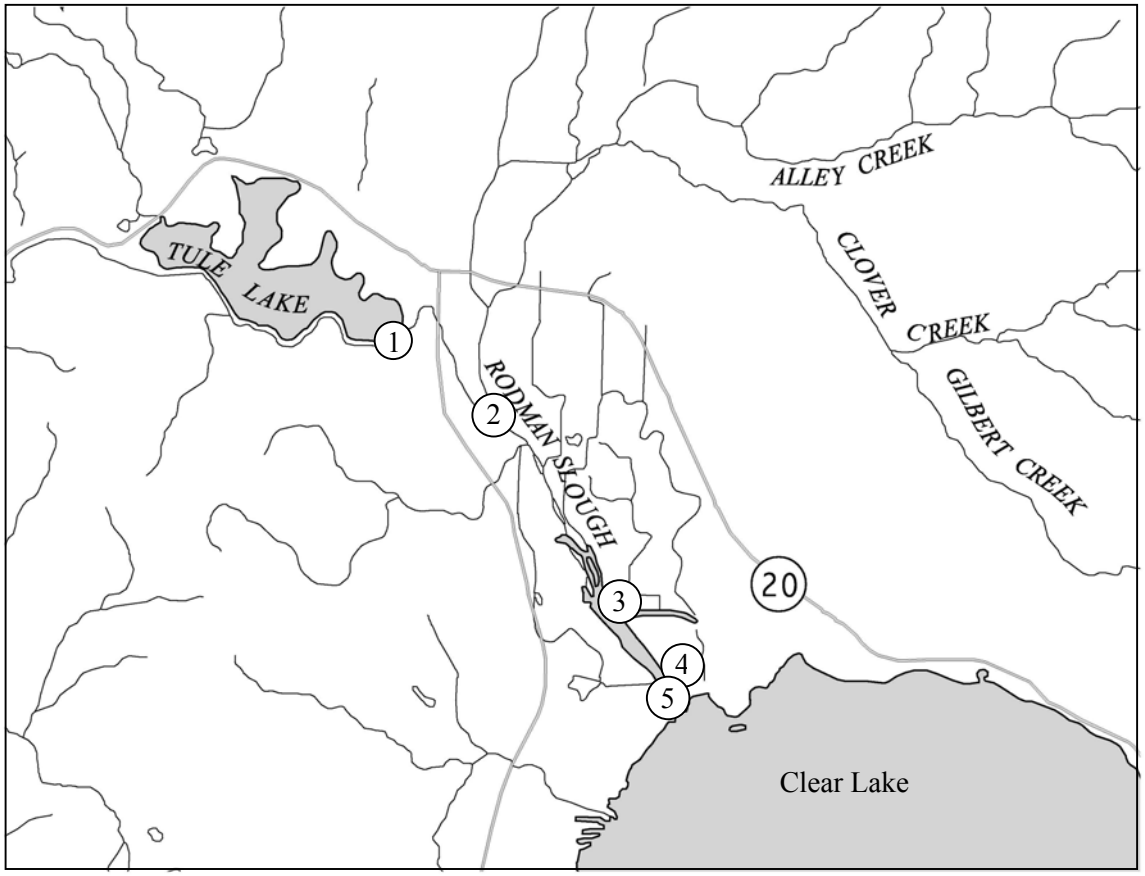


Figure F-1. Map of Clear Lake indicating sampling sites for rice field sites. #1 (TLR-01) = Tule Lake Rice Field; #2 (MSC-01) = Middle Creek / Scotts Creek Confluence; #3 (HSP-01) = Highline Slough Pipe; #4 (RSRFS) = Rodman Slough Rice Field South, #3 (RS-02) = UCD Rodman Slough Site.

Tule Lake

The Tule Lake rice field area is approximately 380 acres. Water circulates via a ditch surrounding the rice fields releasing water to Scotts Creek when necessary. The pump out area has two pipes, one large pipe for winter drainage and a small pipe for summer months if the water level gets too high. Generally lake water circulates through the field. There is a floater mechanism in the field and when the water gets too high water is released to Scotts Creek, which eventually flows into Middle Creek through Rodman Slough into Clear Lake.

Sampling at this site was conducted on September 12, 13, 14, 2000. The field was in the process of harvest. Samples were taken directly from the outflow pipe (TLR-01) on the Scotts Creek side of the

pump station and at the UCD Rodman Slough site (RS-02). The pump station was not running at the time of sampling on the 12th so the sample was taken in front of the outlet pipe near the pump house on the rice field side. The samples taken on the 14th were delayed by the overnight shipping company, arriving four days after shipment. No analysis was performed because this period exceeded the holding time for methyl Hg analysis in water. The RS-02 samples were taken slightly above the south area rice fields, although there was no water draining out of this field at the time of sampling and there was no evidence that harvesting had begun at this rice field. Water was flowing out of the outflow pipe during sampling on the 13th and samples were taken from the pipe as the water was exiting the field.

Robinson Lake: Central Area (managed by Jones)

This rice field is located in the middle of the Robinson Lake area rice fields and thus is named the Central Area. There are approximately 410 acres in this field that drains into Highline Slough. This ancillary slough flows into Rodman Slough, which directly enters Clear Lake. The upper end of Rodman Slough borders this field, water inlets to the rice fields are located along the Western edge. There are other areas located above (to the North) of the central area, however the field that might influence sample results via coincident drainage had been harvested and did not appear to be draining into the slough. There are two pipes that carry water pumped from the rice field into the slough. The larger pipe is utilized during periods of flooding during the winter.

Sampling at this site was conducted on August 8, 9, 10, 2000. Samples were taken above the rice fields just below the confluence of Middle Creek and Scott's Creek (MSC-01), from the rice field outflow pipe located in Highline Slough (HSP-01), and in Rodman Slough below the rice field at an established UCD site (RS-02). On 8/9/01 the outflow pipe was not running so the sample was taken from the pump intake location. The pump was in operation at the other sample times.

Robinson Lake: South Area (managed by Nielson)

This area is bordered by the Nice Lucerne Cutoff road and is physically the closest in proximity to Clear Lake and the mouth of Rodman Slough. 300 acres of this rice field drains into approximately 100 acres of wetland area which has a State of California maintained levee road running through it. The pipeline for the pump-out station located in the wetland area at this site is near the mouth of Rodman Slough. The water is pumped out periodically to keep the water lower than the road, particularly during the winter months. The field water equalizes with the surrounding slough and Clear Lake water in late summer. The pump-out is very close to one of the sampling sites RS-02. Irrigation water, when necessary, is drawn from Clear Lake.

Sampling at this site was conducted on September 25, 2000. Samples were taken from the pump out pipe that was turned on specifically for UCD to obtain a sample.

RESULTS:

RAW WATER:

Figure F.2 provides total Hg and methyl Hg data for RAW (unfiltered) water. Total Hg in RAW water ranged from 2.0-10.7 ng/L (= pptr) in August and from 2.2-7.6 ng/L in September, which is significantly lower than the average of Clear Lake concentrations (ca. 12 ng/L). However, methyl Hg in RAW water ranged from 0.2-2.2 ng/L in August and from 0.2-0.9 ng/L in September. These values, especially in August, were considerably elevated from the long term average methyl Hg concentrations in Clear Lake water as described in Suchanek *et al.* (1997). While there was significantly elevated methyl Hg (relative to average Clear Lake concentrations) in the August collections in water from the Highline Slough pipe draining the Central Area of Robinson Lake, this trend was not observed in waters downstream in Rodman Slough (RS-02) that eventually flow into Clear Lake in either August or September.

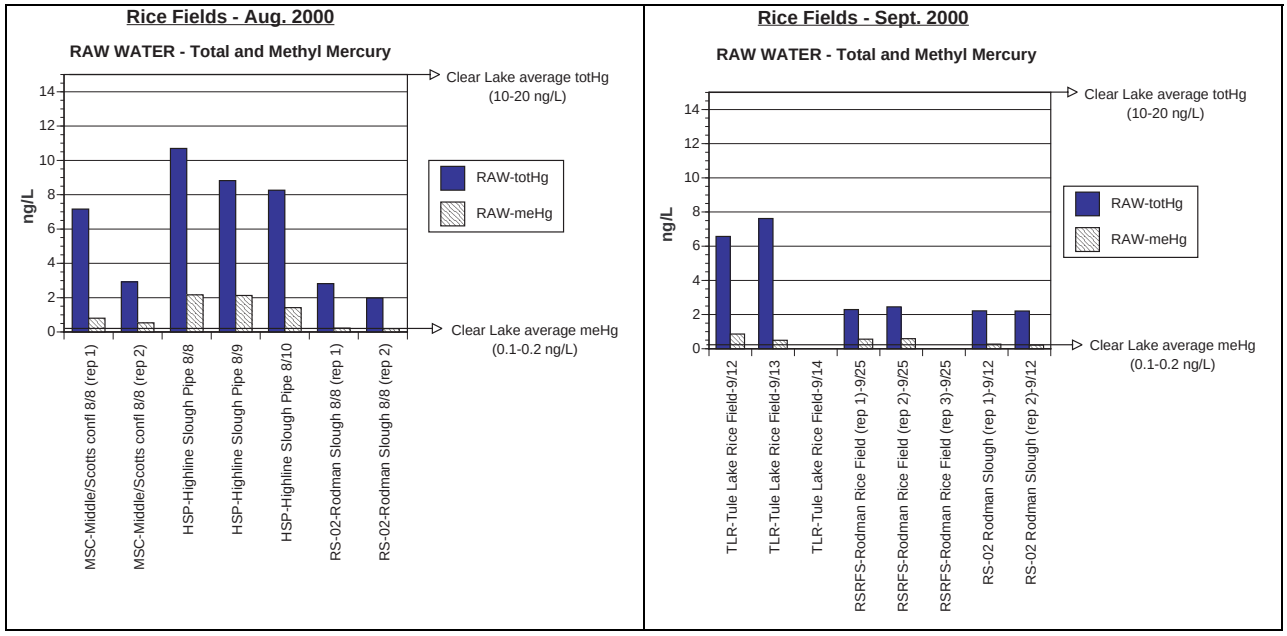


Figure F-2. Data plots for total Hg and methyl Hg in RAW water from rice field sites sampled in August and September 2000.

Figure F.3 provides data for Total Suspended Solids (TSS) and Sulfate (SO_4) concentrations in RAW water from the rice field sites. TSS concentrations ranged from 13-59 mg/L (ppm) in August and from 6-102 mg/L in Setpember. SO_4 concentrations ranged from 1.6-5.1 mg/L in August and from 5.3-23.0 mg/L in September. As might be expected, TSS concentrations from rice field waters are considerably elevated (especially in August) from average values in Clear Lake waters (5-10 mg/L), but the TSS concentrations in Rodman Slough waters are lower and nearly comparable to those found in Clear Lake. SO_4 concentrations (in all but the September Tule Lake Rice Field samples) are below those average values documented for Clear Lake in Suchanek *et al*. (1997).

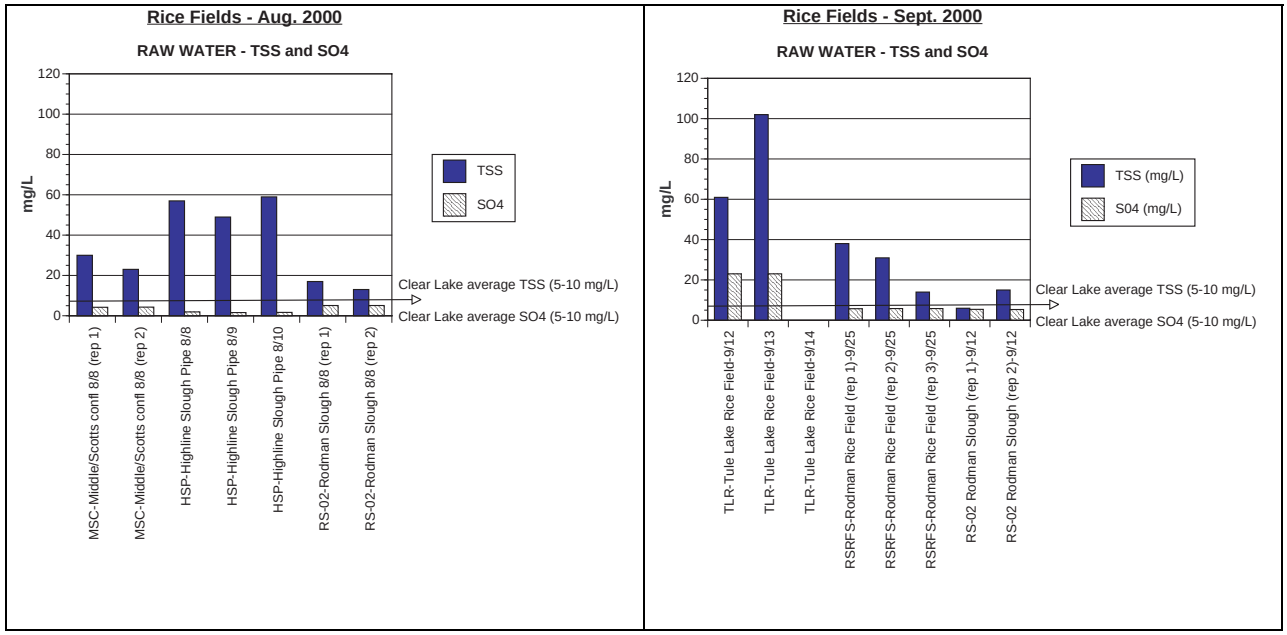


Figure F-3. Data plots for TSS and SO_4 in RAW water from rice field sites in August and September 2000 in comparison with average values for Clear Lake.

FILTERED WATER:

Figure F.4 provides total Hg and methyl Hg data for FILTERED water. Total Hg in FILTERED water ranged from 0.7-2.7 ng/L (pptr) in August and from 0.4-1.2 ng/L in September. The only samples which showed significantly higher concentrations than average Clear Lake water (ca. 1.0-2.0 ng/L) were the Highline Slough Pipe water samples. Methyl Hg concentrations in FILTERED water ranged from 0.1-0.6 ng/L in August and from 0.1-0.5 ng/L in September. All of these values were elevated from the typical average for FILTERED water (ca. 0.05-0.10 ng/L) from most Clear Lake sites, although the waters that flow out of Rodman Slough, as with other parameters, were significantly lower in methyl Hg than those from the rice fields and were nearly identical to those concentrations found in Clear Lake waters.

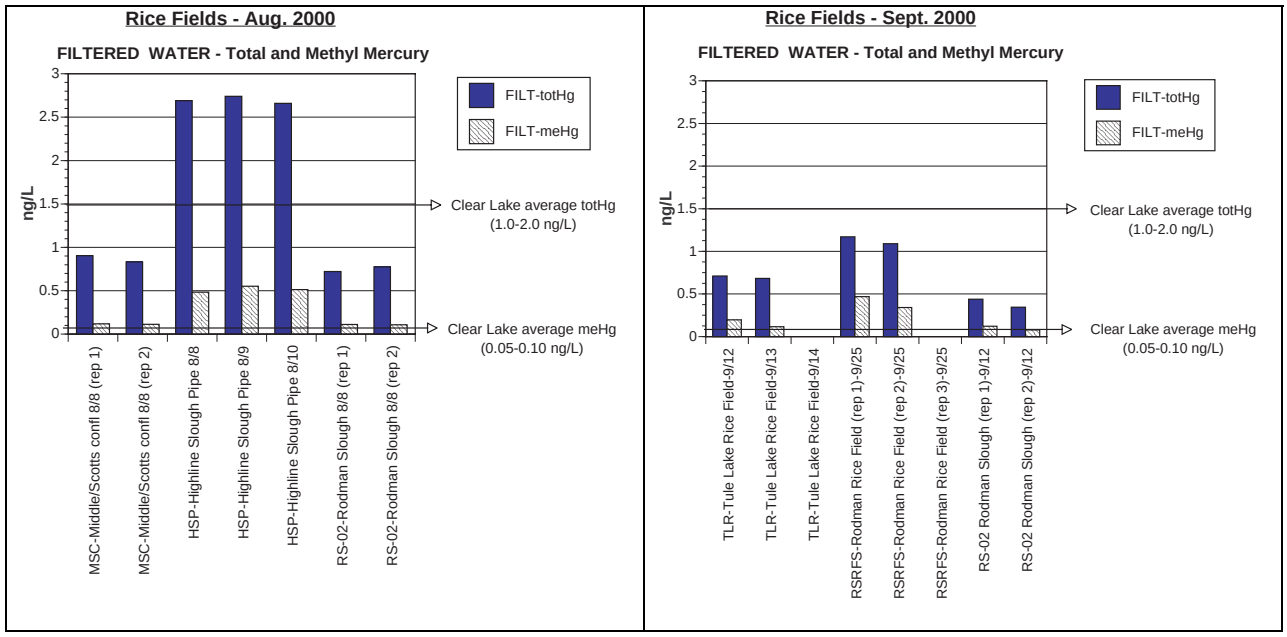


Figure F-4. Data plots for total Hg and methyl Hg in FILTERED water from rice field sites in August and September 2000.

CONCLUSIONS:

Data on total Hg, methyl Hg, TSS and SO_4 in water from rice field sites were compared with longer term time-series data for Clear Lake sites (from 1994-1996) by Suchanek *et al*. 1997.

Total Hg: These data indicate that for RAW (unfiltered) water there are no rice field sites sampled in this study which contribute significantly to total Hg loading into Clear Lake (Fig. 5.2). For FILTERED water, the Highline Slough pipe (draining the Central Area of the Robinson Lake rice field) had significantly elevated total Hg concentrations compared with the long term average for total Hg in FILTERED water for most Clear Lake sites. However, when this water mixed with water from Middle Creek and Scott's Creek, the final total Hg concentration of FILTERED water entering Clear Lake through Rodman Slough was significantly below the Clear Lake average (Fig. 5.4).

Methyl Hg: These data indicate that for RAW (unfiltered) water, all rice field sites exhibited elevated methyl Hg concentrations (up to 10-20X higher) compared with typical average concentrations in Clear Lake waters (Fig. F.2). However, concentrations of methyl Hg in water flowing through Rodman Slough (at RS-02) were close (ca. 0.2 ng/L) to those average values in Clear Lake (ca. 0.1-0.2 ng/L). As above, for total Hg this is likely caused by mixing with additional flow from Middle Creek and Scott's Creek. For FILTERED water, all rice field sites also exhibited elevated methyl Hg concentrations (up to 6-11X higher) compared with typical average values for Clear Lake sites. However, again, once mixed with additional flow from Middle Creek and Scott's Creek, these concentrations declined to values approximating those found in Clear Lake (ca. 0.1 ng/L).

TSS and SO_4: Data from this study indicate that rice fields exhibit significantly elevated (10-20X higher) TSS compared with typical Clear Lake concentrations. However, when mixed with additional water from Middle Creek and Scott's Creek, water flowing out of Rodman Slough is similar to that found in Clear Lake. SO_4 was typically lower than Clear Lake average values except for the September concentrations in the Tule Lake Rice Field. As with total Hg, methyl Hg and TSS, once these waters mix with additional Scott's Creek and Middle Creek waters, the final SO_4 concentrations in water that drains out of Rodman Slough are lower than the typical average concentrations in Clear Lake.

In conclusion, water from some rice fields appear to exhibit some elevated total Hg, TSS and SO_4 concentrations, and some rice fields generate a significant amount of methyl Hg. However, these elevated concentrations appear to be diluted by additional flows from Middle Creek and Scotts Creek before flowing into Clear Lake through Rodman Slough where the concentrations are very nearly equivalent to those observed over many sites and many seasons within Clear Lake.

Literature Cited:

Suchanek, T.H., P.J. Richerson, L.J. Mullen, L.L. Brister, J.C. Becker, A. Maxson, and D.G. Slotton 1997. The role of the Sulphur Bank Mercury Mine site (and associated hydrogeological processes) in the dynamics of mercury transport and bioaccumulation within the Clear Lake aquatic ecosystem. A report prepared for the USEPA, Region IX Superfund Program. 245pp, plus 9 Appendices and 2 Attachments.

Raw Data for Rice Fields Water Analyses:

Rice Fields Water Data

LOCATION - August 2000	RAW-totHg ng/L	RAW-meHg ng/L	FILT-totHg ng/L	FILT-meHg ng/L	TSS mg/L	SO4 mg/L
MSC-Middle/Scotts confl 8/8 (rep 1)	7.16	0.80	0.91	0.12	30	4.2
MSC-Middle/Scotts confl 8/8 (rep 2)	2.92	0.53	0.83	0.11	23	4.3
HSP-Highline Slough Pipe 8/8	10.70	2.17	2.69	0.48	57	1.9
HSP-Highline Slough Pipe 8/9	8.82	2.13	2.74	0.55	49	1.6
HSP-Highline Slough Pipe 8/10	8.26	1.42	2.66	0.51	59	1.7
RS-02-Rodman Slough 8/8 (rep 1)	2.82	0.22	0.72	0.11	17	5.1
RS-02-Rodman Slough 8/8 (rep 2)	1.98	0.20	0.78	0.11	13	5.1

LOCATION - Setember 2000	RAW-totHg ng/L	RAW-meHg ng/L	FILT-totHg ng/L	FILT-meHg ng/L	TSS mg/L	SO4 mg/L
TLR-Tule Lake Rice Field-9/12	6.57	0.86	0.71	0.20	61	23.0
TLR-Tule Lake Rice Field-9/13	7.62	0.50	0.68	0.12	102	23.0
RSRFS-Rodman Rice Field (rep 1)-9/25	2.29	0.56	1.17	0.47	38	5.7
RSRFS-Rodman Rice Field (rep 2)-9/25	2.45	0.59	1.09	0.34	31	5.8
RSRFS-Rodman Rice Field (rep 3)-9/25					14	5.8
RS-02 Rodman Slough (rep 1)-9/12	2.22	0.27	0.44	0.12	6	5.4
RS-02 Rodman Slough (rep 2)-9/12	2.21	0.21	0.35	0.08	15	5.3

APPENDIX G. REGIONAL BOARD STAFF ESTIMATE OF MERCURY FLUXING THROUGH THE WRD, USING WELL DATA COLLECTED FOR THE USEPA SUPERFUND PROGRAM

Quantification of mercury fluxing through the waste rock dam (WRD) of the Sulphur Bank Mercury Mine site is difficult. Uncertainties in this type of calculation are addressed in Sections 3.1.3.2.3 through 3.1.3.2.5. To generate one estimate, Regional Board staff examined and analyzed the data collected by Tetra Tech EMI in 2000 from wells in the WRD. Analysis of the well data by Regional Board staff resulted in higher flux rates than those reported by the Superfund Program (Tetra Tech EMI, 2001). It is important to note that the Tetra Tech EMI and Regional Board estimates were developed using the same set of data gathered by Tetra Tech EMI from wells within the WRD. No measurements have been made of mercury in groundwater at the points of entry into Clear Lake, which could be used to validate these well sampling data for estimations of mercury influx.

To calculate a mercury load released from the waste rock/upper lakebed sediment unit, Regional Board staff used a dissolved mercury concentration of 92 μg/l. This value is the average concentration of dissolved mercury in wells in the groundwater contour closest to the lake, as measured in June 2000 (Tetra Tech EMI, 2001). Assuming the average concentration in the first contour acknowledges both the variability in mercury concentrations between wells and the trend of mercury accumulation as water passes through the WRD.

Concentrations of dissolved mercury in other geologic units were much less variable than in the waste rock unit. Average mercury concentrations throughout the WRD, as calculated by Tetra Tech EMI, were used to estimate mercury loads for the other units. The mercury loads from the andesite and lower lake sediment units were insignificant.

For the TMDL, annual mercury load estimates were calculated using the basic equation of: mercury concentration times flow rate equals estimated annual load. Load calculations are shown in Table 5. Loads of mercury from the andesite and lower lakebed sediment units were very small. The estimated mercury load entering Clear Lake in groundwater flowing through the waste rock dam is about 9 kg/year. This calculation uses maximum groundwater flow measurements taken from wells in 2000 by Tetra Tech.

Table G-1. Regional Board Estimate of Annual Mercury Load from the Waste Rock Dam to Clear Lake, Estimated from Groundwater Flow Rate and Mercury Concentration Data Collected by Tetra Tech EMI for the USEPA Superfund Program

Geologic Unit	Average groundwater flow rate (gpm) (a)	Average dissolved mercury concentration (μg/L)	Annual Load of mercury to Clear Lake (kg/yr)
Waste rock/upper lake sediments	52	92 (b)	9.5
Andesite	20	0.07	0.003
Lower lake sediments	3	1	0.006
Totals	**75**		**9.5**

(a). Average groundwater flow rates are averages of the maximum rates presented by Tetra Tech EMI (2001).
(b). Average mercury concentration for the waste rock/upper lake sediments is the average of dissolved mercury in wells closest to the lake as measured in May/June 2000 (Tetra Tech EMI, 2001).

The USEPA and Tetra Tech EMI have estimated that the average mercury flux in groundwater from the WRD is 1-3 kg/year (Tetra Tech EMI, 2001; USEPA 2001c). Their estimate is based on the same groundwater pump data and mercury concentrations in well samples collected by Tetra Tech EMI and that were used for the estimate in Table 5. The difference is that USEPA used minimal average groundwater flow rates and the average mercury concentration in all wells in the WRD.

APPENDIX H. CLEAR LAKE MERCURY TMDL NUMERIC TARGET REPORT

This separate report is available at:

http://www.waterboards.ca.gov/centralvalley/programs/tmdl/num_tartget_report.pdf