Draft Guidance for Implementing the January 2001 Methylmercury Water Quality Criterion

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Appendix A. Synopsized Mercury TMDLs Developed or Approved by EPA

I. **Ochlockonee Watershed, Georgia**
II. **Arivaca Lake, Arizona**
III. **McPhee and Narraguinep Reservoirs, Colorado**
VI. **Clear Lake, California**

I. Ochlockonee Watershed, Georgia

Description of the Applicable Water Quality Standards

TMDLs are established to attain and maintain the applicable narrative and numerical water quality standards. The state of Georgia's Rules and Regulations for Water Quality Control do not include a numeric criterion for the protection of human health from methylmercury, but they do provide a narrative "free from toxics" water quality standard. Because mercury can cause toxicity in humans, a numeric "interpretation" of the narrative water quality standard was used to assure that a TMDL will protect human health. The state of Georgia has made a numeric interpretation of their narrative water quality standard for toxic substances at a numeric concentration of no more than 0.3 mg/kg methylmercury in fish tissue. This numeric interpretation protects the "general population," which is the population that consumes 17.5 grams per day or less of freshwater fish.

This approach is consistent with EPA's recently adopted guidance value for the protection of human health from methylmercury described in the document titled, *Water Quality Criterion for the Protection of Human Health: Methylmercury* (USEPA 2001c). The methodology uses a "weighted consumption" approach. When only trophic level 3 and 4 fish have been collected, the methodology assumes that 8 grams per day (58.4 percent) of the total fish consumption is trophic level 3 fish (e.g., catfish and sunfish) and 5.7 grams per day (41.6 percent) are trophic level 4 fish (e.g., largemouth bass). EPA collected site-specific data from the Ochlockonee River on ambient mercury in fish tissue and in the water column in the summer of 2000 and in March and April 2001 at two locations. Using a weighted consumption approach, site-specific fish tissue concentration data collected in the Ochlockonee River yields a weighted fish tissue concentration of 0.6 mg/kg, which is greater than the state's current applicable water quality criterion of 0.3 mg/kg. This was calculated as

> *Weighted Fish Tissue Concentration = (Avg Trophic 4 Conc. x .416) + (Avg Trophic 3 Conc. x .584)*

where:

Avg. Trophic Level 3 Concentration = 0.2 mg/kg
Avg. Trophic Level 4 Concentration = 1.0 mg/kg
Weighted Fish Tissue Concentration = 0.6 mg/kg

To establish the TMDL, EPA determined the maximum allowable concentration of mercury in the ambient water that will prevent accumulation of methylmercury in fish tissue above the applicable water quality standard of 0.3 mg/kg level. To determine this EPA used the *Methodology for Deriving Ambient Water Quality Criteria for the Protection of Human Health* (USEPA 2000e). EPA also used the recommended national values from the Human Health Methodology, including the reference dose of 0.0001 mg/kg/day methylmercury; a standard average adult body weight of 70 kg; and the consumption rate for the general population of 17.5 grams per day. For the other factors in the calculation, bioaccumulation and fraction of methylmercury, EPA used site-specific data from the Ochlockonee River collected in summer of 2000 and March and April of 2001. From this site-specific data, EPA determined a representative weighted BAF. This BAF was calculated by taking the average calculated BAF from each of the two trophic levels to determine a “weighted” BAF on the basis of the different consumption rates for trophic levels and a median measured fraction methylmercury of 0.17. Using this approach, an allowable concentration of mercury in the ambient water of Ochlockonee River for the protection of human health is 1.6 ng/L. This was calculated as

$$WQS = \frac{((\text{Reference Dose} - \text{RSC}) \times \text{Body Weight} \times \text{Units Conversion})}{(\text{Consumption Rate} \times \text{Weighted BAF} \times \text{Fraction MeHg})}$$

Where:

WQS = water quality standard = 1.6 ng/L
Reference Dose = 0.0001 mg/kg/day MeHg
RSC = relative source contribution from other fish species = 0.000027 mg/kg/day MeHg
Body Weight = 70 kg
Units Conversion = 1,000,000 mg/kg
Consumption Rate = 0.0175 kg/day Fish
Weighted Bioaccumulation Factor = 1,063,270 l/kg
Fraction of the Mercury as Methylmercury = 0.17 as measured

Source Assessment

A TMDL evaluation must examine all known potential sources of the pollutant in the watershed including point sources, nonpoint sources, and background levels. The source assessment was used as the basis of development of a model and the analysis of TMDL allocation options. This TMDL analysis includes contributions from point sources, nonpoint sources, and background levels. There are 16 water point sources in the Ochlockonee River watershed that could potentially have mercury in their discharge.

According to a review of the *Mercury Study Report to Congress* (USEPA 1997a), significant potential air emission sources include coal-fired power plants, waste incinerators, cement and limekilns, smelters, pulp and paper mills, and chlor-alkali factories. In the report, a national airshed model (RELMAP) was applied to the continental United States. This model provides a distribution of both wet and dry deposition of mercury as a function of air emissions and global sources and was used to calculate dry and wet deposition rates for south Georgia as derived by RELMAP.

The MDN includes a national database of weekly concentrations of mercury in precipitation and the seasonal and annual flux of mercury in wet deposition. EPA reviewed the MDN data for a sampling station near south Georgia. This data was compared with the RELMAP deposition predictions and was found to be substantially higher. Using the MDN data, the average annual wet deposition rate was determined to be 12.75 μg/square meter. The dry deposition rate was determined to be 6.375 μg/square meter on the basis of the RELMAP results.

Loading Capacity—Linking Water Quality and Pollutant Sources

The link between the fish tissue endpoint and the identified sources of mercury was the basis for the development of the TMDL. This helped estimate total assimilative capacity of the river and any needed load reductions. In this TMDL, models of watershed loading of mercury were combined with a model of mercury cycling and bioaccumulation in the water. This enabled a translation between the endpoint for the TMDL (expressed as a fish tissue concentration of mercury) and the mercury loads to the water. The loading capacity was then determined by the linkage analysis as a mercury loading rate that was consistent with meeting the endpoint fish tissue concentration.

Watershed-scale loading of water and sediment was simulated using the WCS. The complexity of this loading function model falls between that of a detailed simulation model, which attempts a mechanistic, time-dependent representation of pollutant load generation and transport, and simple export coefficient models, which do not represent temporal variability. The WCS provides a mechanistic, simplified simulation of precipitation-driven runoff and sediment delivery, yet is intended to be applicable without calibration. Solids load, runoff, and ground water can then be used to estimate pollutant delivery to the receiving waterbody from the watershed. This estimate is based on pollutant concentrations in wet and dry deposition and processed by soils in the watershed and ultimately delivered to the receiving waterbody by runoff, erosion, and direct deposition. The WCS calculated loads for each subbasin are shown in Table A1.

Table A1. Annual average mercury load from each subbasin

Watershed name	Total Hg load (mg)	Areal load (mg/ha)	Impervious area (mg/yr)	Sediment (mg/yr)	Runoff (mg/yr)	Deposition on water (mg/yr)
Barnett Creek	786098.4	25.6	116614.69	422879.88	177553.9	68850
Middle/Lower Ochloclonee	307965.8	21.24	125771.73	89440.3	54786.29	37867.5
Tired Creek	827172.8	22.03	252386.89	317969.16	194751.7	61965
Lower Ochlockonee	359317.5	15.62	100125.11	130407.68	97802.16	30982.5
Little Ochlockonee	873773.4	19.89	140023.69	433136.75	219614.2	80898.75
Bridge Creek	454417.5	23.11	53496.45	261042.44	98468.66	41310
Upper/Middle Ochlockonee	627746.1	20.67	152881.42	254746.48	182250.7	37867.5
Upper Ochlockonee	766396.8	20.1	164465.44	320337	186825.6	94668.75

WASP5 (Ambrose, et al. 1988) was chosen to simulate mercury fate in the Ochlockonee River. WASP5 is a general, dynamic mass balance framework for modeling contaminant fate and transport in surface waters. Environmental properties and chemical concentrations are modeled as spatially constant within segments. Each variable is advected and dispersed among water segments and exchanged with surficial benthic segments by diffusive mixing. Sorbed or particulate fractions can settle through water column segments and deposit to or erode from surficial benthic segments. Within the bed, dissolved variables can migrate downward or upward through percolation and pore water diffusion. Sorbed variables can migrate downward or upward through net sedimentation or erosion.

The toxics WASP model, TOXI5, combines a kinetic structure adapted from EXAMS2 with the WASP5 transport structure and simple sediment balance algorithms to predict dissolved and sorbed chemical concentrations in the bed and overlying waters. TOXI5 simulates the transport and transformation of chemicals as a neutral compound and up to four ionic species, also for particulate material. Local equilibrium is assumed so that the distribution of the chemical between each of the species and phases is defined by distribution or partition coefficients. The predicted mercury concentrations are shown in Table A2.

Table A2. Predicted mercury for annual average load and flow

Calculated concentrations	River reach					
	1	2	3	4	5	6
Total Hg: Water column (ng/L)	6.33	5.84	5.55	5.76	5.65	5.17
Total Hg: Sediment (ng/g)	7.05	9.07	9.81	8.17	7.63	6.97
Methyl Hg: Water column (ng/L)	0.90	0.82	0.77	0.79	0.77	0.71

Allocations

To determine the total maximum load that can come into the Ochlockonee River, the current loading conditions are evaluated and the instream concentration is determined using the modeling approach described above. This allows the development of a relationship between load and instream mercury concentrations. Using this developed relationship, the total maximum load could be determined. Because the water column mercury concentration response is linear with respect to changes in load, a proportion could be developed to calculate the total maximum mercury load from the watershed that would achieve the derived water quality target of 1.6 ng/L. The TMDL was calculated as the ratio of the water quality target to the highest segment concentration (1.6 ng/L divided by 6.3 ng/L) applied to the current annual average load of 5.00 kg/yr. This gives a TMDL load of 1.22 kg/yr mercury. This represents a 76 percent reduction from the current annual average load.

In a TMDL assessment, the total allowable load is divided and allocated to the various pollutant sources. The calculated allowable load of mercury that can come into the Ochlockonee River without exceeding the applicable water quality target of 1.6 ng/L is 1.22 kilograms/year. Because EPA's assessment indicates that over 99 percent of the current loading of mercury is from atmospheric sources, all the load reduction is being assigned to the load allocation and no reduction is required of the wasteload allocation. Therefore, the load allocation and the wasteload allocation for the Ochlockonee River are:

Load allocation (atmospheric sources) = 1.16 kilograms/year
Wasteload allocation (NPDES sources) = 0.06 kilograms/year

EPA estimates that atmospheric deposition contributes over 99 percent of current mercury loadings to the river; therefore, significant reductions in atmospheric deposition will be necessary if the applicable water quality standard is to be attained. On the basis of the total allowable load of 1.22 kilograms per year, a 76 percent reduction of mercury loading is needed to achieve the applicable water quality standard. EPA believes that an estimated 31 percent to 41 percent reduction in mercury deposition to the Ochlockonee River watershed can be achieved by 2010 through full implementation of existing CAA requirements. In addition, there are a number of activities planned or underway to address remaining sources of mercury, and EPA expects that further reductions in mercury loadings will occur over time as a result of these activities. EPA is not able to estimate the reductions in mercury deposition to the Ochlockonee River watershed that will be achieved from future activities. However, as contemplated by CWA section 303(d)(1)(C), this TMDL quantifies the water quality problem facing the Ochlockonee River watershed and identifies the needed reductions in loadings from atmospheric deposition—by CAA initiatives or under other authorities—for the watershed to achieve applicable standards for mercury. In addition, as EPA collects additional data and information for the Ochlockonee River watershed and as new legal requirements are imposed under the CAA, EPA will continue to evaluate the effectiveness of regulatory and nonregulatory air programs in achieving the TMDL's water quality target.

The analysis of NPDES point sources in the watershed indicates that the cumulative loading of mercury from these facilities is less than 1 percent of the total estimated current loading. Even if this TMDL allocated none of the calculated allowable load to NPDES point sources (i.e., a wasteload allocation of zero), the waterbody would not attain the applicable water quality standards for mercury because of the very high mercury loadings from atmospheric deposition. At the same time, however, EPA recognizes that mercury is an environmentally persistent bioaccumulative toxic with detrimental effects to human fetuses even at minute quantities, and should be eliminated from discharges to the extent practicable. Taking these two considerations into account, this TMDL provides a wasteload allocation applicable to all Georgia NPDES facilities in the watershed in the amount of 0.06 kg/year. The TMDL was written so that all NPDES permitted facilities will achieve this wasteload allocation either through the discharge of mercury at concentrations below the applicable water quality standard of 1.6 ng/L or through the implementation of a pollutant minimization plan.

In the context of this TMDL, EPA believes it can reasonably offer the choice of the two approaches to the permitting authority for the following reasons. First, on the basis of EPA's analysis, the Agency expects either wasteload allocation option, in the aggregate, to result in point source mercury loadings less than the wasteload allocation. Second, EPA believes this flexibility is the best way of ensuring that the necessary load reductions are achieved without causing significant social and economic disruption. EPA recognizes that NPDES point sources contribute a small share of the mercury contributions to the Ochlockonee River. However, EPA also recognizes that mercury is a highly persistent toxic pollutant that can bioaccumulate in fish tissue at levels harmful to human health. Therefore, EPA has determined, as a matter of policy, that NPDES point sources known to discharge mercury at levels above the amount present in their source water should reduce their loadings of mercury using appropriate, cost-effective mercury minimization measures to ensure that the total point source discharges are at a level equal to or less than the wasteload allocation specified in this TMDL. The point sources' WLA will be applied to the increment of mercury in their discharge that is above the amount of mercury in their source water. EPA recommends that the permitting authority make this choice between the two options in consultation with the affected discharger because EPA is not able to make the case-by-case judgments in this TMDL that EPA believes are appropriate.

II. Arivaca Lake, Arizona

Description of the Applicable Water Quality Standards

Authorities develop TMDLs to meet applicable water quality standards. These may include numeric water quality standards, narrative standards describing designated uses, and other associated indicators supporting designated uses (beneficial uses apply only to California). A numeric target identifies the specific goals or endpoints for the TMDL that equate to attainment of the water quality standard. The numeric target may be equivalent to a numeric water quality standard (where one exists) or it may represent a quantitative interpretation of a narrative standard.

The applicable numeric targets for the Arivaca TMDL are the Arizona water quality standard of 0.2 µg/L mercury in the water column and the Arizona Fish Consumption Guideline criterion of 1 mg/kg mercury concentration in fish tissue. Arizona has adopted water quality standards for mercury that apply to a number of the designated uses specified for Arivaca Lake, including protection of aquatic life and wildlife and protection of human and agricultural uses. Of these numeric criteria, the most stringent is the chronic aquatic life criterion of 0.01 µg/L dissolved mercury (see Table 7 on page 15 in the TMDL). Arizona has also issued a fish consumption advisory for this lake because mercury concentrations in fish tissue exceed 1 mg/kg mercury.

Mercury bioaccumulates in the food chain. Within a lake fish community, top predators usually have higher mercury concentrations than forage fish, and tissue concentrations generally increase with age class. Top predators (such as largemouth bass) are often target species for sport fishermen. Arizona bases its Fish Consumption Guideline on average concentrations in a sample of sport fish. Therefore, the criterion should not apply to the extreme case of the most-contaminated age class of fish within a target species; instead, the criterion is most applicable to an average-age top predator. Within Arivaca Lake, the top predator sport fish is the largemouth bass. The selected target for the TMDL analysis is an average tissue concentration in 5-year-old largemouth bass of 1.0 mg/kg.

Source Assessment

A TMDL evaluation must examine all known potential sources of the pollutant in the watershed, including point sources, nonpoint sources, and background levels. The source assessment is used as the basis of development of a model and the analysis of TMDL allocation options. There are no permitted point source discharges and no known sources of mercury-containing effluent in the Arivaca watershed. External sources of mercury load to the lake include natural background load from the watershed, atmospheric deposition, and possible nonpoint loading from past mining activities.

Watershed background load: The watershed background load of mercury was derived from mercury in the parent rock and from the net effects of atmospheric deposition of mercury on the watershed. Some mercury also exists within the parent rock formations of the Arivaca watershed, although no concentrated ore deposits are known. The net contributions of both atmospheric deposition and weathering of native rock were assessed by measuring concentrations in sediment of tributaries to Arivaca Lake. EPA collected 25 sediment and rock samples from dry tributaries in the Arivaca watershed and analyzed them for mercury. From these data, most of the sediment samples from the Arivaca watershed were considered at or near background mercury levels.

Nonpoint loadings from mining: No known mining for mercury itself has occurred in the watershed. However, mining activities for minerals other than mercury, especially historical mining practices for gold, might contribute to mercury loading in the watershed. Gold and silver mining commonly occurred in the area surrounding Arivaca Lake but apparently not within the watershed itself. The U.S. Bureau of Mines identified only one exploratory prospect, for manganese and uranium, within the Arivaca watershed itself.

Ruby Dump: Ruby Dump is in the southern portion of Arivaca watershed at the very upstream end of Cedar Canyon Wash. The dump apparently served the town of Ruby and the Montana Mine. The waste is characterized by numerous mining artifacts (e.g., crucibles) but also includes many common household items such as bottles and plates. Samples were taken at three different locations of the Ruby Dump: top of the hill (just below the fire pit), the middle of the hill, and the base of the dump. The mercury results for these samples, from the top of the hill to the bottom, were 1,467 ppb, 1,244 ppb (blind duplicate was 495 ppb), and 486 ppb. The average of these four samples is 918 ppb, which is the number used in the watershed modeling to represent mercury concentration in sediment eroding from this site.

Near-field atmospheric deposition: Significant atmospheric point sources of mercury often cause locally elevated areas of near-field atmospheric deposition downwind. After a review of *Mercury Study Report to Congress* (USEPA 1997a) and a search of EPA's AIRS database of permitted point sources, there are no significant U.S. sources of airborne mercury within or near the Arivaca watershed. Also, the most nearby parts of Mexico immediately to the southwest (prevailing wind direction) of the watershed are sparsely populated. Because of the lack of major nearby sources, especially sources along the axis of the prevailing wind, EPA does not believe that near-field atmospheric deposition of mercury attributable to individual emitters is a major component of mercury loading to the Arivaca watershed. Because no significant near-field sources of mercury deposition were identified, mercury from atmospheric deposition onto the watershed is treated as part of a general watershed background load in this analysis.

Far-field atmospheric deposition: In May 1997, the MDN began collecting deposition data at a new station in Caballo, in the southwestern quadrant of New Mexico. This station is the closest MDN station to the Arivaca Lake and was used to estimate loads to Arivaca Lake. Because the climate at Arivaca is wetter than at Caballo, the distribution of wet and dry deposition is likely to be different. Monthly wet deposition rates at Arivaca were estimated as the product of the volume-weighted mean concentration for wet deposition at Caballo times the rainfall depth at Arivaca. This approach was used because volume-weighted mean concentrations are usually much more stable between sites than wet deposition rates, which are sensitive to rainfall amount. Dry deposition at Arivaca was then calculated as the difference between the total deposition rate at Caballo and the estimated Arivaca wet deposition rate. The estimates derived for Arivaca were 5.3 $\mu g/m^2/yr$ by wet deposition and 7.1 $\mu g/m^2/yr$ by dry deposition. In sum, mercury deposition at Arivaca is assumed to be equivalent to that estimated for Caballo, New Mexico, but Arivaca is estimated to receive greater wet deposition and less dry deposition than Caballo because more of the particulate mercury and reactive gaseous mercury that contribute to dry deposition will be scavenged at a site with higher rainfall.

Loading Capacity—Linking Water Quality and Pollutant Sources

The linkage analysis defines the connection between numeric targets and identified sources. The linkage is defined as the cause-and-effect relationship between the selected indicators, associated numeric targets, and the identified sources. This provided the basis for estimating total assimilative capacity and any needed load reductions. Specifically, models of watershed loading of mercury were combined with a model of mercury cycling and bioaccumulation in the lake. This enabled a translation between the numeric target (expressed as a fish tissue concentration of mercury) and mercury loading rates. The loading capacity was then determined via the linkage analysis as the mercury loading rate that is consistent with meeting the target fish tissue concentration.

Watershed model: Watershed-scale loading of water and sediment was simulated using the Generalized Watershed Loading Function (GWLF) model. The complexity of this loading function model falls between that of detailed simulation models, which attempt a mechanistic, time-dependent representation of pollutant load generation and transport, and simple export coefficient models, which do not represent temporal variability. GWLF provides a mechanistic, simplified simulation of precipitation-driven runoff and sediment delivery yet is intended to be applicable without calibration. Solids load, runoff, and ground water seepage can then be used to estimate particulate and dissolved-phase pollutant delivery to a stream, on the basis of pollutant concentrations in soil, runoff, and ground water. Applying the GWLF model to the period from October 1985 through September 1998 yielded an average of 11.0 cm/year runoff and 2,520,000 kg sediment yield by sheet and rill erosion. The sediment yield estimate is likely to be less than the actual yield rate from the watershed because mass wasting loads were not accounted for; however, mass wasting loads are thought to be of minor significance for loading of bioavailable mercury to the lake.

Estimates of watershed mercury loading were based on the sediment loading estimates generated by GWLF by applying a sediment potency factor. These estimate are shown in Table A3. A background loading estimate was first calculated, then combined with estimates of loads from individual hot spots. The majority of the EPA sediment samples showed no clear spatial patterns, with the exception of the hot spot area identified at Ruby Dump. Therefore, background loading was calculated using the central tendency of sediment concentrations from all samples excluding Ruby Dump. The background sediment mercury concentrations were assumed to be distributed lognormally, as is typical for environmental concentration samples, and an estimate of the arithmetic mean of 70.9 ppb was calculated from the observed geometric mean and coefficient of variation. Applying this assumption to the GWLF estimates of sediment transport yields an estimated rate of mercury loading from watershed background of 178.9 g/yr.

Loading from the Ruby Dump was calculated separately, but was also based on the GWLF estimate of sediment load generated per hectare of "rangeland" (the land use surrounding the hot spots), as reduced by the sediment delivery ratio for the watershed. The extent of the hot spot was observed to be 200 feet by 50 feet. The mercury concentration assigned to surface sediments at the dump was the arithmetic average of the four EPA samples taken in October 1997, or 918 ppb. From these assumptions, less than 1 percent of the watershed mercury load to Arivaca Lake appears to originate from Ruby Dump, which is the only identified hot spot in the watershed.

Table A3. Annual total mercury load to Arivaca Lake

Watershed year	Mercury loading to lake (grams per year)			
	From watershed	From Ruby Dump	From direct atmospheric deposition to lake	Total
1986	170.16	0.65	4.208	175.018
1987	184.34	0.7	4.208	189.248
1988	205.61	0.79	4.208	210.608
1989	70.9	0.27	4.208	75.378
1990	198.52	0.76	4.208	203.488
1991	99.26	0.38	4.208	103.848
1992	163.07	0.62	4.208	167.898
1993	233.97	0.89	4.208	239.068
1994	141.8	0.54	4.208	146.548
1995	219.79	0.84	4.208	224.838
1996	170.16	0.65	4.208	175.018
1997	191.43	0.73	4.208	196.368
1998	276.51	1.06	4.208	281.778
Grand Total	2,325.52	8.88	54.704	2,389.10
Annual Average	**178.89**	**0.68**	**4.21**	**183.78**

The direct deposition of mercury from the atmosphere onto the Arivaca Lake surface was calculated by multiplying the estimated atmospheric deposition rates times the lake surface area, resulting in a load of 4.2 g/yr.

Lake hydrology model: The water level in Arivaca Lake is not actively managed, and releases occur only when storage capacity is exceeded. Therefore, lake hydrology was represented by a simple monthly water balance. Applying the water balance model requires pan evaporation data as an input in addition to the watershed meteorological data. Because no evaporation data were available at the local Cooperative Summary of the Day meteorological station, pan evaporation data for Tucson were used. Pan evaporation for 1980 through 1995 was obtained from the BASINS 2.0 Region 9 data files. Later pan evaporation data were not available for Tucson, so monthly averages were used for the 1996 through 1998 water balance. The water balance model was run for the period 1985 through 1998. This water balance approach provides a rough approximation of the seasonal cycle of changes in volume and surface area of Arivaca Lake and of the amount of water released downstream over the spillway. It cannot capture daily or event scale movement of water in and out of the lake.

Mercury cycling and bioaccumulation model: Cycling and bioaccumulation of mercury within the lake were simulated using the D-MCM (EPRI 1999). D-MCM predicts the cycling and fate of the major forms of mercury in lakes, including methylmercury, Hg(II), and elemental mercury. D-MCM is a time-dependent mechanistic model, designed to consider the most important physical, chemical, and biological factors affecting fish mercury concentrations in lakes. It can be used to develop and test hypotheses, scope field studies, improve understanding of cause/effect relationships, predict responses to changes in loading, and help design and evaluate mitigation options.

Because strong anoxia in the hypolimnion is a prominent feature during summer stratification for the Arizona lakes simulated in this study, D-MCM was modified to explicitly allow significant methylation to occur in the hypolimnion. In previous applications of D-MCM, the occurrence of methylation was restricted to primarily within surficial sediments. That the locus of methylation likely includes or is even largely within the hypolimnion is supported by (1) the detection of significant very high methylmercury concentrations in the hypolimnia of Arivaca Lake and (2) almost complete losses of sulfate in Arivaca Lake in the hypolimnion resulting from sulfate reduction. An input was added to the model to specify the rate constant for hypolimnetic methylation, distinct from sediment methylation.

Results of the model calibration are shown in Table A4. The model calculations are the predicted annual ranges after the model has reached steady state. The observed concentrations are from July 1997.

Table A4. Predicted and observed mercury for annual average load and flow

	Predicted	Observed
Methyl Hg: Water column (ng/L)	0.00–12.07	14.3
Hg II: Water column (ng/L)	0.00–6.28	1.46–8.3
Methyl Hg: 5-year-old largemouth bass (mg/kg)	1.18	1.18

Allocations

A TMDL represents the sum of all individual allocations of portions of the waterbody's loading capacity. Allocations may be made to point sources (wasteload allocations) or nonpoint sources (load allocations). The TMDL (sum of allocations) must be less than or equal to the loading capacity; it is equal to the loading capacity only if the entire loading capacity is allocated. In many cases, it is appropriate to hold in reserve a portion of the loading capacity to provide a margin of safety (MOS), as provided for in the TMDL regulation. The allocations and MOS are shown in Table A5. These allocations, from the best currently available information, predict attainment of acceptable fish tissue concentrations within a time horizon of approximately 10 years. A delay in achieving standards is unavoidable because time will be required for mercury to cycle through the lake and food chain after load reductions occur.

Table A5. Summary of TMDL allocations and needed load reductions (in g-Hg/yr)

Source	Allocation	Existing load	Needed reduction
Wasteload allocations	0.0	0.0	0.0
Load allocations			
Atmospheric deposition	4.2	4.2	0
Ruby dump	0.7	0.7	0
Watershed background	111.2	178.9	67.7
Total	116.1	183.8	67.7
Unallocated reserve	38.7		
Loading capacity	154.8		

The model was used to evaluate the load reductions necessary to meet the numeric target. The response of concentrations of mercury in 5-year-old largemouth bass to changes in external mercury loads is nearly linear. This is because the sediment burial rates are high and sediment recycling is low, with the majority of the methylmercury that enters the food chain being created in the anoxic portion of the water column. The model calculates that the numeric target of 1 mg/kg in 5-year-old largemouth bass is predicted to be met with a 16 percent reduction in total watershed loads to Arivaca Lake, which results in a loading capacity of 154.8 grams mercury per year.

There are uncertainties associated with mercury sources and the linkage between mercury sources and fish tissue concentrations in Arivaca Lake. As a result, the TMDL reserves 38.7 g-Hg/yr (25 percent of the loading capacity) for the MOS and allots the remaining load of 116.1 g-Hg/yr for sources. Because no permitted point source discharges occur within the Arivaca watershed, the wasteload allocation is zero and the load allocation is 116.1 g-Hg/yr.

The load allocation provides loads for three general sources: direct atmospheric deposition onto the lake surface, hot spot loading from Ruby Dump, and generalized background watershed loading, including mercury derived from parent rock and soil material, small amounts of residual mercury from past mining operations, and the net contribution of atmospheric deposition onto the watershed. Direct deposition to the lake surface is a small part of the total load and is believed to derive from long-range transport of global sources which are not readily controllable. The load from Ruby Dump is also small. As a result, the TMDL does not require reductions from these sources, and their load allocations are their existing loads.

Background watershed loading appears to be the major source of mercury to Arivaca Lake. The intensive watershed survey conducted for this TMDL did not identify any significant terrestrial sources of mercury. Regarding air deposition to the watershed land surface, insufficient data were available to calculate reliable estimates of the proportion of mercury deposited from the air that actually reaches Arivaca Lake. Therefore, a load allocation of 111.2 g-Hg/yr was established for overall background watershed loading.

This requires a 38 percent reduction from existing estimated loads from this source. This reduction is believed feasible for several reasons.

Potential for erosion control: Reduction of mercury loading from the watershed to Arivaca Lake depends on reduction in sediment erosion rates. Improved livestock management practices could obtain significant reductions in erosion rates. As a side benefit, implementation of livestock BMPs could result in significant reductions in loadings of DOC and nutrients to the lake. The availability of high levels of DOC and nutrients in the lake appears to affect the methylation process. Reduction of DOC and nutrient levels should reduce the efficiency of the methylation process at Arivaca Lake, effectively increasing the lake's mercury loading capacity.

Reductions in atmospheric deposition of mercury: Although reliable estimates are unavailable, new mercury air emissions to the environment appear to be declining. U.S. mercury emissions have declined significantly since 1990 and are expected to decline further upon implementation of new emission limits on incinerators as required by recent EPA regulations. Reductions in air deposition in Arivaca Lake watershed would eventually result in decreases in mercury loading to the lake itself.

Potential location and remediation of undiscovered mercury sources: Although investigation of the watershed did not reveal any significant localized sources of mercury in the watershed (with the possible exception of Ruby Dump), additional site investigation is warranted to ensure that no significant sources were missed. From past experience with mine site remediation in similar circumstances in Arizona, newly discovered sites could be effectively eliminated as ongoing mercury sources.

Alternative management strategies: Any alterations in rates of methylation or in rates of mercury loss to deep sediments will change the relationship between external mercury load and fish tissue concentration and would thus result in a change in the loading capacity for external mercury loads. The loading capacity could be increased by management intervention methods that decrease rates of bacterial methylmercury production within the lake or increase rates of burial and sequestration of mercury in lake sediment. Selection of such an approach would require further research and feasibility studies. Some alternative strategies that may be suitable for further investigation include the following:

- Hypolimnion aeration or mixing
- Sulfur chemistry modification
- Alum treatment
- Reduce DOC and nutrient levels
- Dredge lake sediments

III. McPhee and Narraguinnep Reservoirs, Colorado

Description of the Applicable Water Quality Standards

The TMDL for McPhee and Narraguinnep Reservoirs in southwestern Colorado was based on the Fish Consumption Advisory action level of 0.5 mg/kg mercury concentration in fish tissue. Colorado Department of Public Health and the Environment listings are based on the risk analysis presented in the May 6, 1991 Disease Control and Epidemiology Division Position Paper for *Draft Colorado Health Advisory for Consumption of Fish Contaminated with Methylmercury*. This paper, using a toxicity value RfD of 0.3 µg/kg/day, establishes a fish tissue concentration of 0.5 mg/kg as the approximate center of the range at which the safe consumption level is 4 meals per month for nonpregnant adults and 1 meal per month for women who are pregnant, nursing, or planning to become pregnant and children 9 years of age or younger. The criterion is applied to an average-age top predator. Within McPhee Reservoir, the top predator among sport fish regularly taken is the smallmouth bass (19 percent of the total catch in 1993). The top predator sport fish in Narraguinnep Reservoir is the walleye. The lake water quality model D-MCM (EPRI 1999) is capable of predicting mercury concentrations in fish tissue for each age class at each trophic level. Average mercury concentrations in fish tissue of target species are assumed to be approximated by the average concentration in 15-inch smallmouth bass in McPhee and the 18-inch walleye in Narraguinnep. Therefore, the selected target for the TMDL analysis in McPhee Reservoir is an average tissue concentration in 15-inch smallmouth bass of 0.5 mg/kg or less. The selected target in Narraguinnep Reservoir is the 18-inch walleye of 0.5 mg/kg or less.

Source Assessment

McPhee and Narraguinnep Reservoirs have several sources of mercury. The sources external to the reservoirs separate into direct atmospheric deposition onto the lakes (from both near- and far-field sources) and transport into the lakes from the watershed. The watershed loading occurs in both dissolved and sediment-sorbed forms. Ultimate sources in the watershed include mercury in parent rock, mercury residue from mine tailings and mine seeps, point source discharges, and atmospheric deposition on to the watershed, including deposition and storage in snowpack.

Table A6. Summary of mercury load estimates for McPhee Reservoir

Reservoir	Watershed runoff (g/yr)	Watershed sediment (g/yr)	Interbasin transfer (g/yr)	Atmos. deposition (g/yr)	Total (g/yr)	Load per volume (mg/ac-ft)	Load per surface area (mg/m^2)
McPhee	2,576	222		251	3,049	4.66	0.098
Narraguinnep	2.7	22.7	15.9	36.8	78.1	4.59	0.035

Past mining activities likely provide an important source of mercury load to the McPhee and Narraguinnep watershed. Three large mining districts exist in the Dolores River watershed, the LaPlata, the Rico, and the area around Dunton on the West Dolores River. The quantity of mercury loading from mining operations has been estimated through a combination of observed data in the water column and sediment coupled with the watershed linkage analysis.

Significant atmospheric point sources of mercury often cause locally elevated areas of near-field atmospheric deposition downwind. Two large coal-fired power plants are in the Four Corners area within about 50 miles of the McPhee and Narraguinnep Reservoirs. The plants in the Four Corners area (2,040 megawatt (MW) capacity) and the Navajo plant (1,500 MW capacity) are upwind of McPhee and Narraguinnep Reservoirs. It is likely that the mercury emitted from these plants contributes to the mercury loading of McPhee and Narraguinnep Reservoirs. No direct measurements of atmospheric deposition of mercury are available, therefore EPA cannot assess the significance of this loading and must await further investigation, including the establishment of a mercury deposition monitoring site in the area.

Loading Capacity—Linking Water Quality and Pollutant Sources

Models of watershed loading of mercury are combined with a model of mercury cycling and bioaccumulation in the lake to translate the numeric target, expressed as a fish tissue concentration of mercury, to mercury loading rates. The coupled models estimate mercury loading to the reservoirs and predict mercury cycling and speciation within the reservoir. An estimated load reduction of 52 percent is needed for long-term average mercury concentrations in a standardized 15-inch smallmouth bass to drop to 0.g mg/kg wet muscle.

Allocations

The loading capacity for McPhee Reservoir was estimated to be 2.59 kilograms mercury per year. Narraguinnep Reservoir loading capacity was estimated at 39.1 grams of mercury per year. This is the maximum rate of loading consistent with meeting the numeric target of 0.5 mg/kg in fish tissue. Due to the uncertainties regarding the linkage between mercury sources and fish tissue concentrations in McPhee and Narraguinnep Reservoirs, an allocation of 70 percent of the loading capacity was used for this TMDL. The TMDL calculated for McPhee Reservoir is equivalent to a total annual mercury loading rate of 1,814 g/yr (70 percent of the loading capacity of 2,592 kg/yr), while Narraguinnep Reservoir is equivalent to a total annual mercury loading rate of 27.3 g-Hg/yr (70 percent of 39.1 g-Hg/yr).

Table A7. Summary of TMDL allocations and needed load reductions for McPhee Reservoir

Source	Allocation	Existing load	Needed reduction
Atmospheric deposition	63	251	188
Rico/Silver Creek mining area	507	1030	523
Dunton mining area	348	708	360
La Plata mining area	69	141	72
Watershed background	827	919	92
Total	**1,814**	**3,049**	**1,235**
Unallocated reserve	778		
Loading capacity	2,590		

Measurements in g-Hg/yr

Table A8. Summary of TMDL allocations and needed load reductions for Narraguinnep Reservoir

Source	Allocation	Existing load	Needed reduction
Atmospheric Deposition	9.2	36.8	27.6
Interbasin Transfer from McPhee Reservoir	9.5	15.9	6.4
Watershed Background	8.6	25.4	16.8
Total	27.3	78.1	50.8
Unallocated Reserve	11.8		
Loading Capacity	39.1		

Measurements in g-Hg/yr

IV. Clear Lake, California

Description of the Applicable Water Quality Standards

The EPA promulgated the CTR in May 2000 (65 FR 31682). The CTR contains a water quality criterion of 50 ng/L total recoverable mercury for water and organism consumption and is intended to protect humans from exposure to mercury in drinking water and fish and shellfish consumption. This criterion is enforceable in California for all waters with a municipal or domestic water supply designated use and is applicable to Clear Lake. However, the state of California does not consider this criterion to be sufficiently protective of the consumers of fish from Clear Lake.

The water quality management plan or Basin Plan for the Central Valley Regional Water Quality Control Board adopted new water quality standards for mercury for Clear Lake at the same time it adopted mercury TMDLs for Clear Lake. The state's water quality criteria are for fish tissue and are intended to protect designated uses for fishing and wildlife habitat. The applicable criteria are: 0.09 mg/kg and 0.19 mg/kg of mercury in fish tissue for trophic levels 3 and 4 fish, respectively. These levels were recommended by the U.S. Fish and Wildlife Service to protect wildlife, including osprey and bald eagles, at Clear Lake; these levels allow adults to safely consume about 3.5 fish meals per month (26 grams/day) if eating mainly trophic level 4 fish such as catfish and bass. The 26 grams/day assumes a diet comprised of 70 percent trophic level 4 fish and 30 percent trophic level 3 fish. The 90th percentile consumption rate of a small group of residents of Clear Lake, primarily members of the Elem Pomo Indian Tribe, is 30 grams/day of Clear Lake fish, as reported in 1997.

Source Assessment

Clear Lake is in Lake County in northern California. It is a shallow, eutrophic waterbody that is comprised of three basins, the Upper, Lower, and Oaks Arms. It is the largest natural lake entirely within California's boundaries. Tourism and sport fishing are important sectors of the local economy. Five Native American Indian Tribes use 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 shores of Oak Arm, was a highly productive source of mercury between 1872 and 1957. Similar smaller mines were 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 mining method at SBMM. EPA declared the SBMM a federal Superfund site in 1991, and 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. EPA is conducting a remedial investigation to fully characterize the SBMM site to propose final remedies.

Inorganic mercury loads entering Clear Lake come from: ground water and surface water from the SBMM site; tributaries and other surface water that flows directly into the lake; and atmospheric deposition, including atmospheric flux from SBMM. Some mercury deposited historically in the lake due to mining operations or erosion at SBMM might also contribute to mercury concentrations in fish today.

Ground water and surface water from the SBMM site: SBMM covers approximately 1 square mile on the east shore of the Oaks Arm of Clear Lake. The site contains approximately 120 acres of exposed mine overburden and tailings (referred to as waste rock). Two small unprocessed ore piles are also on the site. Mercury in samples of mine materials ranged from 50 to 4,000 mg/kg. All piles of mine materials exhibit the potential to generate acid rock drainage. The abandoned mine pit, the Herman Impoundment, is filled with 90 feet of acidic water (pH = 3), and has a surface area of about 20 acres. The average concentrations in the Herman Impoundment of water and sediment are around 800 ng/L and 26 mg/kg, respectively. A geothermal vent located at the bottom of the impoundment continues to discharge gases, minerals (including mercury), and fluids into the pit.

A large pile of waste rock, known as the waste rock dam (WRD) stretches about 2,000 feet along the shore of the western side of the SBMM site. The WRD lies between Herman Impoundment and Clear Lake. The surface water in the impoundment is 10–14 feet above the surface of Clear Lake, which creates a gradient of ground water flow toward the lake. Surface runoff from the northern side of the site is bounded by a wetland that drains to Clear Lake. Surface runoff from the northern waste rock piles is directed through culverts into the northern wetland. In 1990, rock and geofabric barriers were installed at the culverts to reduce transport of suspended solids. The northern wetland is used for cattle grazing and as a source of fish, tules, and other resources used by the members of the Elem Pomo Tribe. Waste rock piles extend into the wetlands.

Inputs of mercury from SBMM are estimated to be between 1 and 568 kg/year. EPA Superfund Program’s estimate of mercury transported in ground water from the WRD is used as the lower bound input. Regional Board staff estimate that 568 kg/year is the maximum upper bound estimate of all inputs from SBMM, including past and continuing contributions to the active sediment layer. This is approximately 96.5 percent of total sources.

Ground water from SBMM appears to contribute mercury that is readily methylated, relative to mercury from other inputs. Ground water flow from the mine site has been detected entering Clear Lake by subsurface flow through lake sediments. Mercury in ground water from the WRD is solubilized and likely in chemical forms that are easily taken up by methylating bacteria. Acidic drainage from the mine site also contains high sulfate concentrations that enhance the rates of methylation by sulfate-reducing bacteria. This assertion is supported by data showing that methylation rates near the mine site are significantly higher than in other parts of the Clear Lake. In contrast to mercury in SBMM ground water, mercury in lakebed and tributary sediments originates primarily as cinnabar, which has low solubility in water.

Tributaries and other surface water flowing directly into the lake: Mercury entering Clear Lake from its tributaries originates in runoff from naturally mercury-enriched soils, sites of historical mining activities, and mercury deposited in the watershed from the atmosphere. Geothermal springs might contribute to tributary loads, especially in the Schindler Creek tributary to Oaks Arm. Tributary and watershed runoff loads of mercury range from 1 to 60 kg/year, depending upon flow rates. Loads in average water years are 18 kg/year. This is approximately 3 percent of total sources.

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

Atmospheric deposition including flux from the SBMM site: 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 global pool were estimated using data from MDN monitoring stations in Mendocino County and San Jose. Estimates ranged from 0.6 to 2.0 kg/year. This is approximately 0.3 percent of total sources.

Loading Capacity–Linking Water Quality and Pollutant Sources

The Regional Board staff assumes that there is a directly proportional relationship between methylmercury in fish and mercury in the surficial sediment. This is a simplification of a highly complex process. Many factors affect methylation or concentrations of methylmercury, including sulfide and sulfate concentrations, temperature, organic carbon, and so on. Factors that affect accumulation of methylmercury in fish include species, growth rate, prey availability, and the like. To reduce levels of methylmercury in fish, loads of mercury to the lake must be reduced. Section 5.3.1 of the Staff Report provides examples of remediation projects that demonstrate removal of inorganic mercury from a range of aquatic environments has been effective in reducing concentrations of mercury in fish.

A set of first order relationships, each controlled by a single variable of concentration of mercury or methylmercury provide basis for the assumption of a directly proportional relationship between mercury in fish and in surficial sediment in Clear Lake. Concentrations of methylmercury in water and methylmercury in biota are related by BAFs. Relationships between methylmercury in the water column and in sediment can be described as a flux rate of methylmercury from sediment. Concentrations of methylmercury and mercury in sediment are related through calculation of a methylation efficiency index (ratio of methylmercury to mercury in surficial sediment).

In each of these steps in the linkage analysis, one variable is related to another by a simple ratio or linear equation. For example, BAFs are calculated by dividing the concentration of methylmercury in fish by the concentration of methylmercury in the water. Data are available to determine BAF and methylation indices that are specific for Clear Lake. With the current understanding of the transport, methylation, and uptake processes in Clear Lake, staff is unable to refine these relationships to incorporate effects of other factors. The end result was that methylmercury in biota was related linearly to mercury in surficial sediment.

Meeting the recommended water quality standards would require reduction of existing fish tissue concentrations by 60 percent. Using the linear relationship, the linkage analysis indicates that overall mercury loads to Clear Lake sediment must be reduced by 60 percent in order to reduce methylmercury concentrations in fish tissue by the proportional amount. The Regional Board is establishing the assimilative capacity of inorganic mercury in Clear Lake sediments as 70 percent of existing levels to include a margin of safety of 10 percent to account for the uncertainties in the linkage analysis.

Allocations

The strategy for meeting the fish tissue criteria is to reduce the inputs of mercury to the lake from tributaries and the SBMM site, combined with active and passive remediation of contaminated lake sediments. The load allocations for Clear Lake will result in a reduction in the overall mercury sediment concentration by 70 percent of existing concentrations. The load allocations are assigned to the active sediment layer of the lakebed, the SBMM terrestrial site, the tributary creeks and surface water runoff to Clear Lake, and atmospheric deposition. Table A9 summarizes the load allocations. The load allocation to the active sediment layer is expressed as reducing concentrations of mercury in the active sediment layer to 30 percent of current concentrations. The load allocation to the SBMM terrestrial site is 5 percent of the ongoing loads from the terrestrial mine site. The load allocation for the mine also includes reducing mercury concentrations in surficial sediment to achieve the sediment compliance goals for Oaks Arm shown in Table A10. The load allocation to tributary and surface water runoff is 80 percent of existing loads. These load allocations account for seasonal variation in mercury loads, which vary with water flow and rainfall. The analysis includes an implicit margin of safety in the reference doses for methylmercury that were used to develop the fish tissue objectives. It also includes an explicit margin of safety of 10 percent to account for uncertainty in the relationship between fish tissue concentrations and loads of mercury. The reductions in loads of mercury from all sources are expected to result in attainment of water quality objectives.

Table A9. Summary of mercury load allocations

Source	Existing load (kg/year)	Needed reduction
Clear Lake sediment	695	70% of existing concentration
Sulphur Bank Mine		95% of existing load
Tributaries	18	20% of existing load
Atmosphere	2	no change

Table A10. Sediment goals for mercury in Clear Lake

Site designation	Location	Sediment mercury goal (mg/kg dry weight)[a]
Upper Arm UA-03	Center of Upper Arm on transect from Lakeport to Lucerne	0.8
Lower Arm LA-03	Center of Lower Arm, north and west of Monitor Point	1.0
Oaks Arm		
OA-01[c]	0.3 km from SBMM 0.3 km from SBMM	16[b]
OA-02[c]	0.8 km from SBMM	16[b]
OA-03[c]	1.8 km from SBMM	16
OA-04[c]	3.0 km from SBMM	10
Narrows O1	7.7 km from SBMM	3

a. Sediment goals are 30 percent of existing concentrations. Existing concentrations are taken as the average mercury concentrations in samples collected in 1996–2000 (Clear Lake Basin Plan Amendment Staff Report).

b. Due to the exceptionally high concentrations existing at the eastern end of Oaks Arm, sediment goals at OA-01 and OA-02 are not 70 percent of existing concentrations. These goals are equal to the sediment goal established for OA-03.

c. Sediment goal is part of the load allocation for SBMM.

Clear Lake sediment: Reducing mercury concentrations in surficial sediment by 70 percent is an overall goal for the entire lake. To achieve water quality objectives, extremely high levels of mercury in the eastern end of Oaks Arm near SBMM must be reduced by more than 70 percent. To evaluate progress in lowering sediment concentrations, the following sediment compliance goals are established at sites that have been sampled previously.

Sulphur Bank Mercury Mine: Current and past releases from the SBMM are a significant source of mercury loading to Clear Lake. Ongoing annual loads from the terrestrial mine site to the lakebed sediments occur through ground water, surface water, and atmospheric routes. Loads from ongoing releases from the terrestrial mine site should be reduced to 5 percent of existing inputs. Because of its high potential for methylation relative to mercury in lakebed sediments, mercury entering the lake through ground water from the mine site should be reduced to 0.5 kg/year.

Past releases from the mine site are a current source of exposure through remobilization of mercury that exists in the lakebed sediments as a result of past releases to the lake from the terrestrial mine site. Past active mining operations, erosion, and other mercury transport processes at SBMM have contaminated sediment in Oaks Arm. The load allocation assigned to SBMM includes reducing surficial sediment concentrations in Oaks Arm by 70 percent (more at sites nearest the mine site) to meet the sediment compliance goals in Table A10.

EPA anticipates implementing additional actions to address the ongoing surface and ground water releases from the SBMM over the next several years. These actions are expected to lead to significant reductions in the ongoing releases from the mine pit, the mine waste piles, and other ongoing sources of mercury releases from the terrestrial mine site. EPA also plans to investigate what steps are appropriate under Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) to address the existing contamination in the lakebed sediments from past releases from the SBMM. The Regional Board will continue to work closely with EPA on these important activities. In addition, the Regional Board will coordinate monitoring activities to investigate other sources of mercury loads to Clear Lake. These investigations by EPA and the Regional Board should reduce the uncertainty that currently exists regarding the annual load of mercury to the lake, the contribution of each source to that load, and the degree to which those sources lead to methylmercury exposure to and mercury uptake by fish in the lake. This information should lead to more refined decisions about what additional steps are appropriate and feasible to achieve the applicable water quality criteria.

Tributaries and surface water runoff: Past and current loads of mercury from the tributaries and direct surface water runoff are also a source of mercury loading to the lake and to the active sediment layer in the lakebed. This section excludes loads from surface water runoff associated with the SBMM, which are addressed separately above. The loads of mercury from the tributaries and surface water runoff to Clear Lake should be reduced by 20 percent of existing levels. In an average water year, existing loads are estimated to be 18 kg/year. Loads range from 1 to 60 kg/year depending upon water flow rates and other factors. The load allocation applies to tributary inputs as a whole, instead of to individual tributaries. Efforts should be focused on identifying and controlling inputs from hot spots. The U.S. Bureau of Land Management, U.S. Forest Service, other land management agencies in the Clear Lake Basin, and Lake County will submit plans for monitoring and implementation to achieve the necessary load reductions. The Regional Board will coordinate with those agencies and other interested parties to develop the monitoring and implementation plans. The purpose of the monitoring is to refine load estimates and identify potential hot spots of mercury loading from tributaries or direct surface runoff into Clear Lake. Hot spots can include erosion of soils with concentrations of mercury above the average for the rest of the tributary. If significant sources are identified, the Regional Board will coordinate with the agencies to develop and implement load reductions. The implementation plans will include a summation of existing erosion control efforts and a discussion of feasibility and proposed actions to control loads from identified hot spots. The agencies will provide monitoring and implementation plans within 5 years after the effective date of this amendment and implement load reduction plans within 5 years thereafter. The goal is to complete the load reductions within 10 years of implementation plan approval.

The Regional Board will work with the Native American Tribes in the Clear Lake watershed on mercury reduction programs for the tributaries and surface water runoff. They will solicit the tribes’ participation in the development of monitoring and implementation plans.

Wetlands: The Regional Board is concerned about the potential for wetland areas to be significant sources of methylmercury. Loads and fate of methylmercury from wetlands that drain to Clear Lake are not fully understood. The potential for production of methylmercury should be assessed during the planning of any wetlands or floodplain restoration projects within the Clear Lake watershed. The Regional Board established a goal of no significant increases of methylmercury to Clear Lake resulting from such activities. As factors contributing to mercury methylation are better understood, the Regional Board should examine the possible control of existing methylmercury production within tributary watersheds.

Atmospheric deposition: Atmospheric loads of mercury originating outside of the Clear Lake watershed and depositing locally are minimal. Global and regional atmospheric inputs of mercury are not under the jurisdiction of the Regional Water Board. Loads of mercury from outside of the Clear Lake watershed and depositing from air onto the lake surface are established at the existing input rate, which is estimated to be 1 to 2 kg/year.

Appendix B. Tables from Methylmercury Criteria Document

This appendix contains several tables taken directly from the 2001 methylmercury criteria document. These are repeated here to help the reader understand the development of the 2001 criterion.

Table 5-1. Exposure parameters used in derivation of the water quality criterion. (References cited in this table can be found in the 2001 methylmercury criteria document.)

Parameter	Population			Source
	Children (0-14 years)	**Women of Childbearing Age (15-44 years)**	**Adults in the General Population**	
Body Weight, kg	30	67	70	U.S. EPA (2000a)
Drinking Water Intake, L/day	1.0	2.0	2.0	U.S. EPA (2000a)
Freshwater/Estuarine Fish Intake, g/day	156.3[b]	165.5[b]	17.5[c][24]	U.S. EPA (2000a)
Inhalation, m^3/day	10.4	11	20	U.S. EPA (1994, 1997h)[d]
Soil Ingestion, g/day	0.0001, 0.01[a]	0.00005	0.00005	U.S. EPA (1997h)
Mean Marine Fish Intake, g/day	74.9[b]	91.04[b]	12.46[c]	U.S. EPA (2000b)
Median Marine Fish intake, g/day	59.71[b]	75.48[b]	0[c]	U.S. EPA (2000b)
90th Percentile Marine Fish Intake, g/day	152.29[b]	188.35[b]	49.16[c]	U.S. EPA (2000b)

[a] Pica child soil ingestion

[b] For children and women of childbearing age, intake rates are estimates of "consumers only" data (as described in U.S. EPA, 2000b)

[c] For adults in the general population, intake rates are estimates of all survey respondents to derive an estimate of long-term consumption (U.S. EPA).

[d] Inhalation rates for children and women of childbearing age from U.S. EPA, 1997h. Inhalation rates for adults in the general population from U.S. EPA (1994).

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[24] This is the 90th percentile freshwater and estuarine fish consumption value.

Table 5-14. Average Mercury Concentrations in Marine Fish and Shellfish[25] (References cited in this table can be found in the 2001 methylmercury criteria document)

Species	Concentration[a] (µg Hg/g Wet Wt.)	Species	Concentration (µg Hg/g Wet Wt.)
Finfish			
Anchovy	0.047	Pompano*	0.104
Barracuda, Pacific	0.177	Porgy*	0.522[b]
Cod*	0.121	Ray	0.176
Croaker, Atlantic	0.125	Salmon*	0.035
Eel, American	0.213	Sardines*	0.1
Flounder*,[e]	0.092	Sea Bass*	0.135
Haddock*	0.089	Shark*	1.327
Hake	0.145	Skate	0.176
Halibut*	0.25	Smelt, Rainbot*	0.1
Herring	0.013	Snapper*	0.25
Kingfish	0.10	Sturgeon	0.235
Mackerel*	0.081	Swordfish*	0.95[c]
Mullet	0.009	Tuna*	0.206
Ocean Perch*	0.116	Whiting (silver hake)*	0.041
Pollock*	0.15	Whitefish*	0.054[d]
Shellfish			
Abalone	0.016	Oysters	0.023
Clam*	0.023	Scallop*	0.042
Crab*	0.117	Shrimp	0.047
Lobster	0232	Other shellfish*	0.012[b]
Molluscan Cephalopods			
Octopus*	0.029	Squid*	0.026

*Denotes species used in calculation of methylmercury intake from marine fish for one or more populations of concern, based on existence of data for consumption in the CSFII (U.S. EPA, 2000b).

[a] Mercury concentrations are from NMFS (1978) as reported in U.S. EPA (1997d) unless otherwise noted, measured as µg of mercury per gram wet weight of fish tissue.

[b] Mercury concentration data are from Stern et al. (1996) as cited in U.S. EPA (1997c).

[c] Mercury concentration data are from U.S. FDA Compliance Testing as cited in U.S. EPA (1997c).

[d] Mercury concentration data are from U.S. FDA (1978) as cited in U.S. EPA (1997c).

[e] Mercury data for flounder were used to estimate mercury concentration in marine flatfish for intake calculations.

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[25] More current information on commercial fish and shellfish is provided by the Food and Drug Administration at http://www.cfsan.fda.gov/%7Efrf/sea-mehg.html

Table 5-30. Exposure estimates for methylmercury and percent of total exposure based on adults in the general population

Exposure Source	Exposure Estimate (mg/kg-day)	Percent of Total Exposure	Percent of RfD
Ambient water intake	4.3×10^{-9}	0.0047	0.004
Drinking water intake[a]	5.6×10^{-8}	0.0605	0.006
Nonfish dietary intake	0	0	0
Marine fish intake	2.7×10^{-5}	29.33	27
Air intake	4.6×10^{-9}	0.005	0.005
Soil intake	1.3×10^{-9}	0.0014	0.001

[a] This represents the high-end of the range of estimates. Because the contribution of ambient water or drinking water intake to total exposure is so negligible in comparison to the sum of intake from other sources, there is not difference in the total exposure estimated using either of these two alternatives.

Appendix C. Analytical Methods

Table C1. Analytical methods for determining mercury and methylmercury in tissue

Method	Form/species and applicable matrices	Sensitivity	Technique	Known studies or literature references using the techniques in this method
Draft Method 1630, with modifications for tissue	Methylmercury in tissue	Methylmercury in tissue	Tissue modification: digest tissue with acid solution, neutralize with acetate buffer, and analyze as per Method 1630 (i.e., distillation with heat and N_2 flow to separate methylHg from sample, ethylation with sodium tetraethyl borate, N_2 purging of methylethylHg onto graphite carbon (Carbotrap) column, thermal desorption of methylethylHg and reduction to Hg^0, followed by CVAFS detection.	• EPA Cook Inlet Contaminant Study • Lake Michigan fish and invertebrates, Mason and Sullivan 1997 • NE Minnesota lake plankton, Monson and Brezonik 1998[26] • Method performance testing in freshwater and marine fish, Bloom 1989
Method 1631, Draft Appendix A	Total mercury in tissue, sludge, and sediment	Total mercury in tissue, sludge, and sediment	Digest tissue with HNO_3/H_2SO_4. Dilute digestate with BrCl solution to destroy remaining organic material. Analyze digestate per Method 1631 (i.e., add BrCl to oxidize all Hg compounds to Hg(II). Sequentially pre-reduced with hydroxylamine hydrochloride to destroy the free halogens and reduced with $SnCl_2$ to convert Hg(II) to Hg(0). Hg(0) is purged from solution onto gold-coated sand trap and thermally desorbed from trap for detection by CVAFS.	• EPA National Fish Tissue Study (>1000 samples over 4-year period) • EPA Cook Inlet Contaminant Study • Lake Michigan fish and invertebrates, Mason and Sullivan 1997 • NE Minnesota lake plankton, Monson and Brezonik 1998[27] • Method performance testing in freshwater and marine fish, Bloom 1989
Method 245.6[28]	Total mercury in tissue	Total mercury in tissue	Sulfuric and nitric acid digestion, oxidation with potassium permanganate and potassium persulfate, $SnCl_2$ reduction, CVAAS detection	unknown
Draft Method 7474 (SW-846)[29]	Total mercury in sediment and tissue	Total mercury in sediment and tissue	Microwave digestion of sample in nitric and hydrochloric acids, followed by cold digestion with bromate/bromide in HCl. Hg purged from sample and determined by CVAFS.	Reference materials cited in method. Niessen et al. 1999.

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[26] Used similar techniques but used a methylene chloride extraction instead of the distillation.

[27] Used similar techniques but used a methylene chloride extraction instead of the distillation.

[28] Provided for reference purposes only. EPA recommends use of Method 1631 for mercury for analyzing water and fish tissue.

[29] Provided for reference purposes only. EPA recommends using Method 1631 for analyzing mercury for water and fish tissue.

Table C2. Analytical methods for determining mercury and methylmercury in water, sediment, and other nontissue matrices

Method	Forms/species and applicable matrices	Sensitivity	Sample preparation	Known studies or literature references using the techniques in this method
EPA 1630	Methylmercury in water	0.02 ng/L	Distillation with heat and N_2 flow, addition of acetate buffer and ethylation with sodium tetraethyl borate. Purge with N_2 onto Carbotrap. Thermal desorption and GC separation of ethylated mercury species, reduction to Hg° followed by CVAFS detection.	• USEPA Cook Inlet Study • USEPA Savannah River TMDL study • Northern Wisconsin Lakes, Watras et al. 1995 • Lake Michigan waters, Mason and Sullivan 1997 • Anacostia River Study, Mason and Sullivan 1998 • NE Minnesota lakes, Monson and Brezonik 1998[30] • Poplar Creek, TN CERCLA Remedial Investigation of surface water, sediment, and pore water, Cambell et al. 1998[31] • Scheldt estuary study of water, polychaetes, and sediments, Baeyens et al. 1998
UW-Madison SOP for MeHg Analysis	Methylmercury in water	0.01 ng/L	Distillation with heat and N_2 flow, with potassium chloride, sulfuric acid, and copper sulfate. Ethylation with sodium tetraethyl borate. Purge with N_2 onto Carbotrap. Thermal desorption and GC separation of ethylated mercury species, reduction to Hg^0 followed by CVAFS detection.	• Lake Michigan tributaries to support GLNPO's LMMB Study • Fox River, WI waters and sediments, Hurley et al. 1998
USGS Wisconsin - Mercury Lab SOPs 004	Methylmercury in water	0.05 ng/L	Distillation (heat), APDC solution, N_2 flow, potassium chloride, sulfuric acid, and copper sulfate. Ethylation with sodium tetraethyl borate. Purge with N_2 onto Carbotrap. Thermal desorption and GC separation of ethylated species, reduction to Hg^0, and CVAFS detection.	Aquatic Cycling of Mercury in the Everglades, (ACME) cofunded by USGS, EPA, and others
USGS Open-File Report 01-445:	Methylmercury in water	Detection limit cited as 0.04 ng/L	Distillation (heat) and N_2 flow, HCl and copper sulfate. Addition of acetate buffer and ethylation with sodium tetraethyl borate. Purge with N_2 onto Carbotrap. Thermal desorption and GC separation of ethylated mercury species, reduction to Hg(0) followed by CVAFS detection.	Formalized USGS method version of USGS Wisconsin Lab SOP 004. Report title is: *Determination of Methyl Mercury by Aqueous Phase Ethylation, Followed by GC Separation with CVAFS Detection*.

Note: The four methylmercury methods above are all based on the work of Bloom 1989 as modified by Horvat et al. 1993, and are virtually identical as a result.

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[30] Used similar techniques but used a methylene chloride extraction instead of the distillation.

[31] Used similar techniques but omitted the distillation procedure

Table C2. (continued)

Method	Forms/species and applicable matrices	Sensitivity	Sample preparation	Known studies or literature references using the techniques in this method
EPA 1631 (CVAFS)	Total or dissolved mercury in water	MDL=0.2 ng/L ML=0.5 ng/L	Oxidize all Hg compounds to Hg(II) with BrCl. Sequentially pre-reduce with hydroxylamine hydrochloride to destroy the free halogens and reduce with $SnCl_2$ to convert Hg(II) to Hg(0). Hg(0) is purged from solution with N_2 onto gold coated sand trap and thermally desorbed from trap for detection by CVAFS.	• USEPA Cook Inlet Study • State of Maine studies • USEPA Savannah River TMDL study • USEPA/U.S. Navy study for development of Uniform National Discharge Standards • Watras et al. 1995 • Anacostia River Study, Mason and Sullivan 1998 • Northeastern Minnesota lakes, Monson and Brezonik 1998 • Poplar Creek, TN CERCLA Remedial Investigation Study, Cambell et al. 1998 • Scheldt Estuary Study, Baeyens et al. 1998
EPA 245.1 (CVAAS)	Total or dissolved mercury in wastewater	200 ng/L	H_2SO_4 and HNO_3 digestion, $KMnO_4$, $K_2S_2O_8$ oxidation + heat, cool +NaCl-$(NH_2OH)_2 \cdot H_2SO_4$, $SnSO_4$, aeration. Detection by CVAAS.	Effluent guideline development studies for the Meat Products Industry, Metal Products and Machinery Industry, and Waste Incinerators
EPA 245.2 (CVAAS)	Total or dissolved mercury in wastewater and sewage	200 ng/L	H_2SO_4 and HNO_3 added, $SnSO_4$, NaCl-$(NH_2OH)_2 \cdot H_2SO_4$, $KMnO_4$, $K_2S_2O_8$, heat. Detection by CVAAS.	MPM Industry effluent guideline development study
EPA 245.5 (CVAAS)	Total or dissolved mercury in soils, sludge and sediment	200 ng/L	Dry sample, aqua regia, heat, $KMnO_4$ added, cool +NaCl-$(NH_2OH)_2 \cdot H_2SO_4$, $SnSO_4$, aeration. Detection by CVAAS.	Pharmaceutical industry effluent guideline development study
EPA 245.7 (CVAFS)	Total or dissolved mercury in water	ML = 5 ng/L; MDL = 1.8 ng/L	HCl, $KBrO_3$ /KBr, $NH_2OH \cdot HCl$, $SnCl_2$, liquid-vapor separation. CVAFS detection	Interlaboratory validation completed.
EPA 7470A (CVAAS)	Total or dissolved mercury in liquid wastes and Ground water	200 ng/L (IDL)	H_2SO_4 and HNO_3 added, $KMnO_4$ added, $K_2S_2O_8$ added + heat, cool +NaCl-$(NH_2OH)_2 \cdot H_2SO_4$, $SnSO_4$, aeration of sample. CVAAS detection.	Method is similar to and cites performance data given in EPA 245.5
EPA 7471B (CVAAS)	Total or dissolved mercury in solid wastes semisolid wastes	200 ng/L (IDL)	H_2SO_4 and HNO_3 added, $KMnO_4$ added, $K_2S_2O_8$ added + heat, cool +NaCl-$(NH_2OH)_2 \cdot H_2SO_4$, $SnSO_4$, aeration of sample. CVAAS detection.	Method is similar to and cites performance data given in EPA 245.5
EPA 7472 (Anodic Stripping Voltametry)	Total or dissolved mercury in water	100-300 ng/L	Acidify and chlorinate sample, GCE electrode	Unknown
EPA 7473 (Thermal decomposition, amalgamation, and CVAA)	Mercury in water, soil, and sediment	estimated to be as low as 20 ng/ L or 20 ng/kg	Sample aliquot decomposed at 750EC in oxygen atmosphere. Decomposition products carried into catalytical furnace for completed oxidations, then to algamated trap. Mercury is thermally desorbed and determined by AA.	Unknown

Table C2. (continued)

Method	Forms/species and applicable matrices	Sensitivity	Sample preparation	Known studies or literature references using the techniques in this method
EPA 1620 (CVAAS)	Mercury in water, sludge, and soil	200 ng/L	H_2SO_4 and HNO_3 added, $KMnO_4$, $K_2S_2O_8$ + heat, cool +NaCl-$(NH_2OH)_2\cdot H_2SO_4$, $SnSO_4$, aeration. CVAAS detection.	Industry effluent guideline development studies
SM 3112B (CVAAS)	Total or dissolved mercury in water	500 ng/L	H_2SO_4 and HNO_3 added, $KMnO_4$ added, $K_2S_2O_8$ added + heat, cool +NaCl $(NH_2OH)_2\cdot H_2SO_4$, $SnCl_2$ or $SnSO_4$, aeration. CVAAS determination.	Unknown
*ASTM D3223-91(CVAAS)	Total or dissolved mercury in water	500 ng/L	H_2SO_4 and HNO_3 added, $KMnO_4$ added,$K_2S_2O_8$ added + heat, cool +NaCl $(NH_2OH)_2\cdot H_2SO_4$, $SnSO_4$, aeration. CVAAS determination.	Unknown
*AOAC 977.22 (Atomic absorption spectrometry)	Total or dissolved mercury in water	200 ng/L	H_2SO_4 and HNO_3 added, $KMnO_4$ added, $K_2S_2O_8$ added + heat, cool +NaCl $(NH_2OH)_2\cdot H_2SO_4$, $SnSO_4$, aeration. Determine mercury by CVAA.	Unknown

Notes: (1) CVAAS = cold vapor atomic absorption spectrometry
(2) CVAFS = cold vapor atomic fluorescence spectrometry
(3) ASTM and AOAC analytical methods are available from the respective organization

Appendix D. Examples of National Deposition Monitoring Networks

There are a number of national deposition monitoring networks that may be useful for developing TMDLs. Networks include the National Atmospheric Deposition Program - National Trends Network (NADP/NTN) and the MDN (a subset of the NADP network). The NADP/NTN is a nationwide network of precipitation monitoring stations. Operating since 1978, it collects data on the chemistry of precipitation for monitoring of geographical patterns and temporal long-term trends. NADP/NTN measures weekly average concentrations of sulfate, nitrate, ammonium, base cations, and acidity at approximately 230 monitoring stations across the United States. The MDN measures concentrations of total mercury in precipitation at approximately 45 monitoring stations across the United States NADP/NTN results for 2003 are shown in Figure D-1. For more information about NADP see http://nadp.sws.uiuc.edu.

Used in conjunction with NADP/NTN, the Clean Air Status and Trends Network (CASTNET) is the nation's primary source of atmospheric data on the dry deposition component of total acid deposition, ground-level ozone, and other forms of atmospheric pollution that enters the environment as particles and gases. CASTNET measures weekly average atmospheric concentrations of sulfate, nitrate, ammonium, sulfur dioxide, and nitric acid, and hourly concentrations of ambient ozone levels in rural areas. Dry deposition rates are calculated using the measured atmospheric concentrations, meteorological data, and information on land use, surface conditions, and vegetation. Seventy-nine monitoring stations operate across the United States. For more information about CASTNET, see http://www.epa.gov/castnet and http://nadp.sws.uiuc.edu.

Note that these national monitoring networks generally provide only estimates of wet deposition; estimates of dry deposition can be obtained from the literature. For more information on deposition monitoring networks, see *Deposition of Air Pollutants to the Great Waters: Third Report to Congress* (USEPA 2000f) (http://www.epa.gov/oar/oaqps/gr8water/3rdrpt) and the Air-Water Interface Plan (http://www.epa.gov/owow/oceans/airdep/airwater_plan16.pdf).

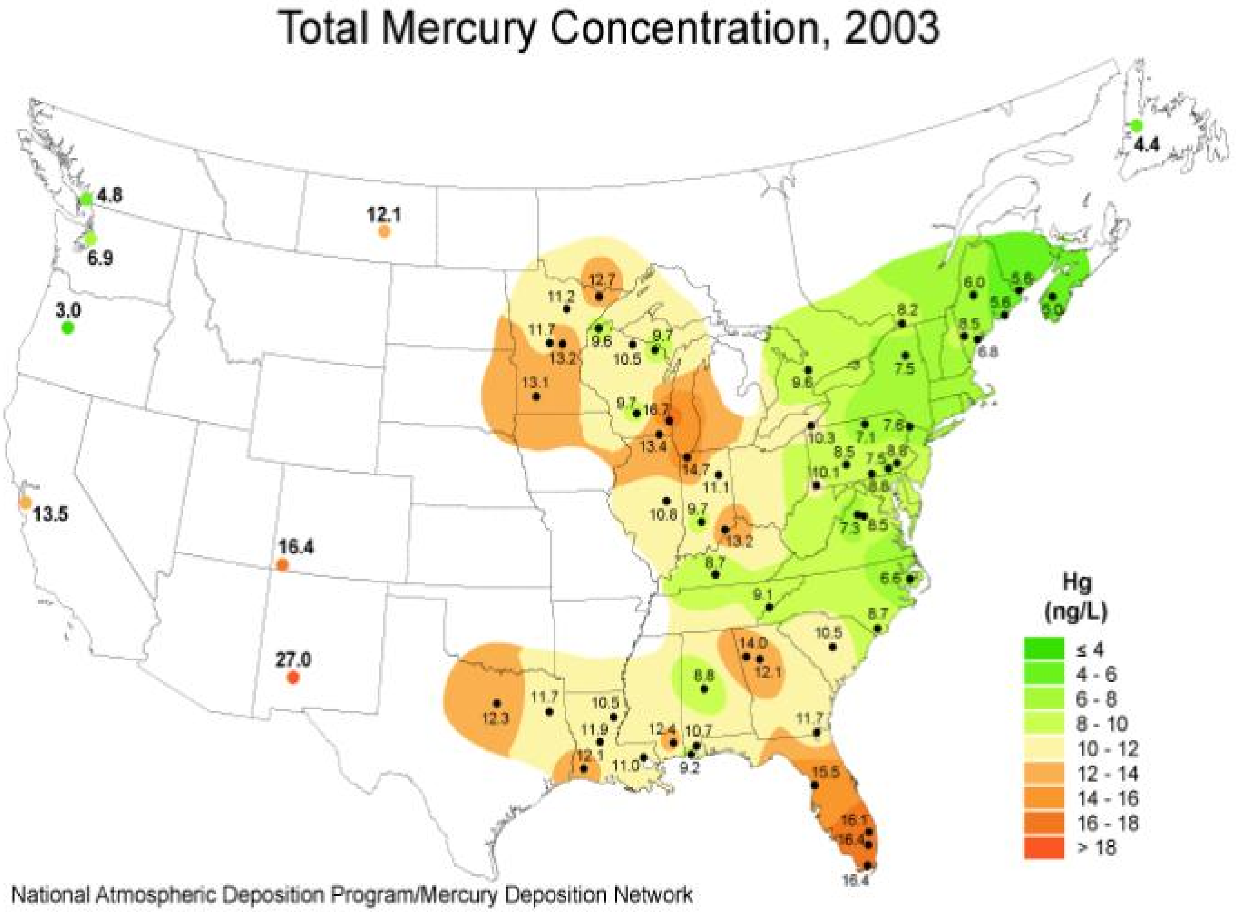

Figure D-1 MDN data for 2003

Appendix E. Methylmercury/Mercury Ratio Exhibited in Muscle Tissue of Various Freshwater Fish Species

Source	**Ecosystem type**	**Fish species**	**MethylHg/ total Hg ratio**
C.R. Hammerschmidt, J.G. Wiener, B.E. Frazier and R.G. Rada (1999)	Freshwater lakes in Wisconsin, USA	Yellow perch (Perca flavescens)	mean 0.95; range from 0.84 to 0.97
D.S. Becker and G.N. Bigham (1995)	Onondaga Lake, a chemically-contaminated lake in New York, USA	Gizzard shad (*Dorosoma cepedianum*) White perch (*Morone americana*) Carp (*Cyprinus carpio*) Channel catfish (*Ictalurus punctatus*) Bluegill (*Lepomis macrochirus)* Smallmouth bass (*Micropterus dolomieui*) Walleye (*Stizostedion vitreum*)	> 0.90 Note: authors did not provide specific percentages for individual species
T.M. Grieb, C.T. Driscoll, S.P. Gloss, C.L. Schofield, G.L. Bowie, and D.B. Porcella (1990)	Lakes in the Upper Michigan Peninsula, USA	Yellow perch (*Perca flavescens*) Northern pike (*Esox lucius*) Largemouth bass (*Micropterus salmoides*) White sucker (*Catostomus commersoni)*	0.99 Note: authors did not provide data for each species separately—only mean value observed over all species
N.S. Bloom (1992)	Freshwater fish species collected from remote midwestern lakes and one mercury contaminated site USA	Yellow perch (*Perca flavescens*) Northern pike (*Esox lucius*) White sucker (*Catostomus commersoni*) Largemouth bass (*Micropterus salmoides*)	0.99 1.03 0.96 0.99
B. Lasorsa and S. Allen-Gil (1995)	3 lakes in the Alaskan Arctic, USA	Arctic grayling Lake trout Arctic char Whitefish	1.00 all for species Note: authors did not provide species specific information on MeHg/Total Hg ratio
T. A. Jackson (1991)	Lakes and reservoirs in northern Manitoba, Canada	Walleye (*Stizostedion vitreum*) Northern pike (*Esox lucius*) Lake whitefish (*Coregonus clupeaformis*)	range: 0.806 to 0.877% range: 0.824 to 0.899% range: 0.781 to 0.923% Note: author sampled the 3 fish species at 4 lake locations
R. Wagemann, E. Trebacz, R. Hunt, and G. Boila (1997)	Sampling location not provided; presumed to be from Canadian waters	Walleye (*Stizostedion vitreum*)	mean 1.00 Note: authors did not provide more specific information

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Note: Bold numbers indicate where the term is defined (if applicable). If the term has been broken into subcategories, this is noted with a "defined" entry.

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