ADEQ
Arizona Department of Environmental Quality

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Lake Mary Regional TMDL For Mercury in Fish Tissue

Upper Lake Mary, Lower Lake Mary, Soldiers Lake, Soldiers Annex Lake, and Lower Long Lake

Little Colorado River Watershed
Coconino County

Arizona Department of Environmental Quality
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List of Acronyms

303(d)	Clean Water Act Section – Water Quality Impairment
305(b)	Clean Water Act Section - Water Quality Assessment
319	Clean Water Act Section – Nonpoint Source Program
A&W	Aquatic and Wildlife Designated Use
A.A.C.	Arizona Administrative Code
A.R.S.	Arizona Revised Statutes
ADEQ	Arizona Department of Environmental Quality
AGFD	Arizona Game and Fish Department
AgI	Agriculture Irrigation Designated Use
AgL	Agriculture Livestock Designated Use
B/C	Soil Horizons below Top Organic Layer
BAF	Biological Accumulation Factor
C_{avg}	Average Trophic Level Fish Mercury Concentration
CMAQ	Community Multiscale Air Quality Modeling System
CWA	Clean Water Act
DEM	Digital Elevation Model
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
DWS	Domestic Water Source Designated Use
FBC	Full Body Contact Designated Use
FC	Fish Consumption Designated Use
GWLF	Generalized Watershed Loading Function
Hg^{2+}	Ionic Mercury
Hg^0	Elemental Mercury
THg	Total Mercury
Me-Hg	Methyl-mercury
LA	Load Allocation
LMR	Lake Mary Region
M	meter
MDN	Mercury Deposition Network
Mg/kg	Milligrams per Kilogram
MOS	Margin of Safety
MP	Malcolm Pirnie, Inc.
NADP	National Atmospheric Deposition Program
NAU	Northern Arizona University
NB	Natural Background
Ng/g	Nanograms per Gram
NRCS	National Resource Conservation Service
ppb	Parts per Billion
R(USLE)2	Revised(Universal Soil Loss Equation) Second Version
REMSAD	Regional Modeling System for Aerosols and Deposition
SERAFM	Spreadsheet-based Ecological Risk Assessment for the Fate of Mercury
Tekran	Dry Deposition Mercury Monitor

TL-4 (3,2,1)	Trophic Levels (4 highest)
TMDL	Total Maximum Daily Load
TOC	Total Organic Carbon
TSS	Total Suspended Solids
*u*g/m^2	Microgram per Square Meter
USEPA	United States Environmental Protection Agency
USFS	United States Forest Service
USGS	United States Geological Survey
WLA	Wasteload Allocation
WRCC	Western Region Climate Center
WTP	Water Treatment Plant

I. EXECUTIVE SUMMARY

This Total Maximum Daily Load (TMDL) reflects a regional approach to fish tissue mercury contamination. Five lakes within the lower Little Colorado River watershed in northern Arizona were listed as impaired for mercury in fish tissue between 2002 and 2003. Not only are these five lakes within the same water and airsheds, they are also close to the same elevation 6,500-7,000 ft and located within similar surficial volcanic geology and soils. All five TMDL lakes were constructed between 1904 and 1954, display similar water chemistry, contain no known point sources, and share similar historical land uses. Because of these similarities, they have been treated collectively as to mercury contamination. Some differences do exist, however, most notably lake morphology, periodicity of water level (climate and water management), and fish stocking practices.

The first of the TMDL lakes to be constructed was Lower Lake Mary in 1904 for timber and stock water supply. The remaining four TMDL lakes were constructed in the 1940s and 1950s for similar reasons, although Upper Lake Mary has been used as a supplemental water supply for the City of Flagstaff.

The soils in the region contain mostly silt and clay and are extremely erodible. A history of timber harvest and grazing has resulted in some areas with heightened runoff due to loss of topsoil and vegetation. In large part today, the lakes are still surrounded by the Coconino National Forest with minimal timber harvest and moderate livestock grazing. Some private land exists, particularly in the immediate watershed of Upper and Lower Lake Mary.

The fish species which were sampled include walleye, northern pike, largemouth bass, yellow bass, crappie, channel catfish, bluegill and rainbow trout. Many lakes in the Lake Mary region (LMR) are stocked with trout in the summer, however, the lakes are really cool-water rather than cold-water lakes, so trout populations are not likely to survive from year to year. This TMDL addresses mercury levels in all species, with a focus on walleye as the top predator species.

There are two critical periods for mercury loading in this region, the monsoon season for intensity of runoff, and the spring snowmelt/runoff season for duration of runoff. Both wet and dry aerial deposition and geologic background mercury concentration were factored into this TMDL. The TMDL model used regional wet and dry air deposition data collected at the Sycamore Canyon Mercury Deposition Network (MDN) station (AZ02). Sediment cores showed pre-impoundment levels of mercury that were later confirmed with watershed soil sampling.

Four different types of models were developed and linked for this project:

- A watershed loading model;
- A lake hydrologic model;
- An in-lake mercury cycling model; and
- Mercury bioaccumulation calculations.

Site-specific biological accumulation factors (BAFs) were used to link model simulated water column concentrations to fish tissue concentrations. Model predictions of average mercury concentrations in adult walleye were made for various levels of anthropogenic input loads to the lakes.

In order to calculate load reductions on a lake system basis, ADEQ used the trophic level-weighted geometric mean approach described in the *Guidance for Implementing the January 2001 Methyl-mercury Water Quality Criterion*, (EPA, 2009). Based on trophic-level geometric mean concentrations, the following reductions in mercury loading are necessary to meet the 0.3 mg/kg mercury fish tissue standard.

Upper and Lower Lake Mary:

1) 25 percent reduction in methyl-mercury and
2) 32 percent reduction in total mercury.

Soldiers Complex:

1) 40 percent reduction in methyl-mercury and
2) 46 percent reduction in total mercury

The major source of mercury to the lakes in the LMR is atmospheric deposition with some mercury originating from natural geologic materials. As there are no known local atmospheric mercury sources in the LMR, it is not likely that aerial deposition can be significantly reduced in the near future through local efforts. Improvement can be made, however, by reducing soil erosion and transport of organic material from the watersheds. TMDL implementation will focus on decreasing sediment delivery to the lakes, lake level stability and fishery management.

II. INTRODUCTION AND PROBLEM STATEMENT

A. Description of TMDL Process

The goal of the federal Clean Water Act (CWA) is to "protect and preserve the physical, chemical, and biological integrity of the nations' waters." This is often termed the "fishable/swimmable" goal of the CWA and is understood to mean that a surface water is meeting the designated use standards for fishing and public recreation. In cases where waters do not meet this goal, Section 303(d) of CWA requires states to develop TMDLs for the pollutants causing impairment with oversight from the U.S. Environmental Protection Agency (EPA). A TMDL allocates pollution loads among pollution sources in a watershed, and is the basis for actions taken to restore the chemical, physical and biological integrity of a waterbody that has been classified as "impaired" for one or more designated uses.

A TMDL represents the total load of a pollutant that can be discharged to a water body on a daily basis and still meet the applicable water quality standard. The TMDL can be expressed as the total mass or quantity that can enter the water body within a unit of time. In most cases, the TMDL determines the allowable mass per day of a constituent and divides it among the various contributors in the watershed as waste load (i.e., point source discharge) and load (i.e., nonpoint source) allocations. The TMDL must also account for natural background sources, seasonal variation and provide a margin of safety.

B. Clean Water Act Section 305 (b), 303 (d) and significance

Surface water quality standards are reviewed and revised by states every three years as criteria are refined. These criteria, or threshold levels, are developed for various potential pollutants based on the particular designated uses of a water body and the degree of exposure or risk to humans, animals and plants. Standards may be numeric or narrative, meaning they can be numbers, ranges of numbers, or narrative descriptions. Arizona's Surface Water Quality Standards contain both numeric and narrative criteria (Arizona Administrative Code (A.A.C.) Title 18, Chapter 11).

Every two years, each state must submit an accounting of how well its water bodies are meeting the applicable standards. This report is known as the Water Quality Assessment Report or "305(b) Report" after the section of the CWA requiring a report to Congress. Waters are classified as follows:

Category 1- attaining their uses (full support),
Category 2- attaining some uses (partial support),
Category 3- inconclusive (insufficient data to assess),
Category 4- not attaining, and
Category 5- impaired.

Based on the 305(b) assessment report, the state generates a list of impaired waters (Arizona Revised Statutes (A.R.S.) Title § 49-232 through 234; A.A.C. Title 18, Chapter 11, Article 6). The list is referred to as the Water Quality Limited List or "303(d) List", after the relevant CWA section. Waters on this list require a TMDL to be completed.

Issuance of a fish advisory does not automatically result in a lake being listed as "impaired" for fish tissue. ADEQ and the Arizona Game and Fish Department (AGFD) issued fish advisories for Upper and Lower Lake Mary in 2002, and for Soldiers, Soldiers Annex and Lower Long Lakes, in 2003. In 2002, EPA added the five LMR lakes to Arizona's 303(d) List as impaired for mercury in fish tissue. This TMDL is the result of that listing, and will use the target of 0.3 mg/kg (wet weight) mercury, the fish tissue standard adopted by ADEQ in January 2009.

III. WATERSHED CHARACTERIZATION

A. Description of Study Area

LMR is located on the Coconino National Forest, within the Little Colorado River Watershed in north-central Arizona. Land in the LMR is primarily rugged and undeveloped, with 98% under the jurisdiction of the U.S. Forest Service (USFS) and the remaining 2% as private holdings. TMDL and background lakes are listed in Table 1 and depicted on Figure 1.

Table 1. LMR Lakes

TMDL Lakes	Waterbody ID	Background Lakes	Waterbody ID
Upper Lake Mary	15020015-0900	Ashurst Lake	15020015-0090
Lower Lake Mary	15020015-0890	Willow Springs Lake	15020010-1670
Soldiers Lake	15020008-1440	Mormon Lake	15020015-0970
Soldiers Annex Lake	15020008-1430		
Lower Long Lake	15020008-0820		

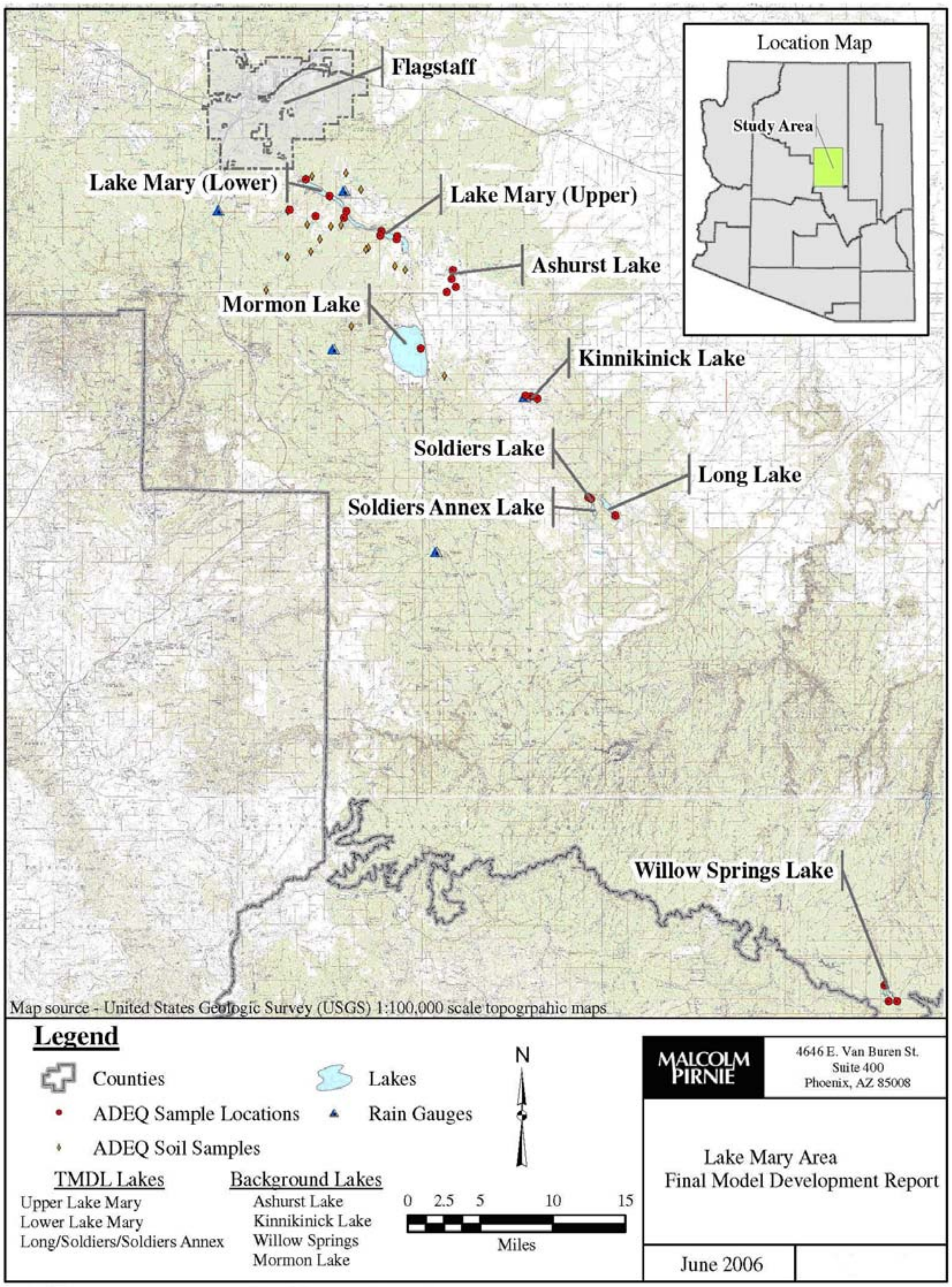

Figure 1. Vicinity Map (adapted from Malcolm Pirnie, 2006)

1. Upper and Lower Lake Mary

All of the lakes within the LMR, except Mormon Lake, are man-made and were originally created to provide additional water sources for either people or livestock in the Flagstaff area. Upper and Lower Lake Mary are located 6 miles southeast of Flagstaff. The majority of Upper and Lower Lake Mary watersheds are located to the south of the lakes, with elevations ranging from 6,800 to 8,500 feet.

Lower Lake Mary was created in 1904 after an eight-year drought, to support the Arizona Lumber and Timber Company, local community and stock industry. At full capacity Lower Lake Mary is approximately 765 acres in size, but it is really more of a wetland in many years with a pool by the dam.

Upper Lake Mary was constructed in 1940, and at full capacity is 860 acres in size, making it the larger of Flagstaff's twin lakes. It is 8 miles long and over one-half mile wide at its widest point. However, due to the shallow depth, the aerial extent of the lake varies widely with precipitation.

Upper and Lower Lake Mary are hydraulically connected and support heavy recreational use in the forms of fishing (Upper – largemouth and yellow bass, crappie, sunfish, channel catfish, walleye, tilapia and yellow perch; Lower – rainbow trout, sunfish, channel catfish, and northern pike), camping, wildlife viewing, boating (canoeing, sailing, rafting and power boats) and swimming.

Both Lakes have been assigned the following designated uses according to the A.A.C. Title 18, Chapter 11:

- Domestic Water Source (DWS),
- Aquatic and Wildlife Cold Water (A&W cold),
- Full Body Contact (FBC),
- Fish Consumption (FC), and
- Agricultural Livestock Watering (AgL).

Although Upper and Lower Lake Mary are designated as domestic water sources, the levels of total mercury observed do not approach drinking water maximum contaminant levels.

2. Soldiers and Lower Long Lake Complex

Soldiers Lake, Soldiers Annex Lake and Lower Long Lake were constructed in the 1940-1955 time period and are located 35 miles southeast of Flagstaff and 33 miles southwest of Winslow at an elevation of approximately 6,700 feet. Soldiers Lake is approximately 28 acres, Soldiers Annex is approximately 122 acres, and Lower Long Lake at maximum pool is 320 acres. All three lakes are located within the Coconino National Forest. The first lake in the series, Soldiers Lake, has the highest watershed-to-lake ratio of the lakes studied: 14,672 watershed acres: 28 lake acres, with a ratio of

525. Most likely due to this ratio, Soldiers Lake fish species were all found to be above the criterion of 0.3 mg/kg, with the exception of trout. Because they are in such close proximity to Soldiers Lake and interconnected, Soldiers Annex and Lower Long have the smallest watershed-to-lake ratios at 3 and 4 respectively.

These lakes have been assigned the following designated uses according to the A.A.C. Title 18, Chapter 11:

- Aquatic and Wildlife Cold Water (A&W cold),
- Full Body Contact (FBC),
- Fish Consumption (FC),
- Agricultural Livestock Watering (AgL), and
- Agricultural Irrigation (AgL).

3. Background Lakes

Ashurst Lake, Kinnikinick Lake, Mormon Lake, and Willow Springs Lake were originally selected as background lakes because fish tissue mercury results were lower than those measured in the TMDL lakes. Ashurst, Kinnickinick, and Mormon lakes are within the LMR, so they share the same airshed and major watershed (Little Colorado River) as the TMDL lakes. With the exception of Willow Springs Lake which resides in karst topography, the other lakes reside in similar volcanic geology as the TMDL lakes. All the lakes are surrounded by Pinyon, Juniper and Ponderosa Pine forests.

The original intent of the background lakes was to determine why some lakes in the LMR contained fish with high levels of mercury while others did not. Unfortunately the lakes do not all contain the same species, making this type of analysis inconclusive. In addition, the average fish tissue mercury in catfish from Kinnickinick Lake, was greater than the 0.3 mg/kg criterion (at 0.35 mg/kg), so Kinnickinick Lake has been removed from the background lake category.

Water quality sampling data indicate that tributary inputs of mercury are comparable among all of the lakes (see Section IV) studied, indicating that in-lake processes and the fish species contained within each lake, play an important role in the bioaccumulation of mercury. As fish tissue collection and bioaccumulation studies continue on a statewide basis ADEQ hopes to determine the specific factors leading to mercury methylation.

B. Climate

1. Upper and Lower Lake Mary

Data retrieved for the Western Region Climate Center (WRCC) indicate the Flagstaff Airport, near Upper and Lower Lake Mary watersheds, reports a high average temperature of 61.2˚F (16.2 C), an average of 21.26 inches of precipitation and 99.6 inches of snowfall a year (WRCC, 2005). Historic flow data indicates that runoff occurs during two periods of the year. The majority occurs in response to snowmelt during the period of mid-March to early April. The second period occurs from mid-July to early September during the summer monsoon storm season. Monsoon storms often produce brief but intense runoff to the lakes.

2. Soldiers and Lower Long Lake Complex

The closest weather station to the Soldiers and Long complex is the Happy Jack Ranger Station, which reports a high average temperature of 58.7˚F (14.8 C) and receives an average of 26.55 inches of precipitation and 93.8 inches of snow fall a year (WRCC, 2005). The majority of the runoff, primarily snowmelt, occurs during the winter and spring months. During the summer months very little runoff occurs, with the exception of summer monsoon storms which may produce some flow.

C. Hydrology

Bathymetric maps for all the lakes within the LMR study were provided by Dr. Paul Gremillion of the Northern Arizona University (NAU) Civil and Environmental Engineering Department. For Upper Lake Mary and Ashurst Lake, bathymetric contours were digitized from paper maps and elevation contours from USGS topographic maps. For Lower Long, Soldiers and Soldiers Annex Lakes, contours were digitized from USGS 7.5' topographic series maps to augment collected field data. Bathymetric maps for Lower Lake Mary were available from a study conducted by Dr. Wilbert Odem (2002), also from NAU.

The Upper Lake Mary watershed has a drainage area of approximately 34,650 acres (Figure 2). Within the Upper Lake Mary watershed five major ephemeral drainages exist (Figure 3), including: Hoxworth/Babbit Creek (3,900 acres), Newman Creek (14,200 acres), Sinkhole/Railroad Creek (3,800 acres), Pine Creek (4,300 acres) and Walnut Creek (4,000 acres).

The Lower Lake Mary watershed has a smaller drainage area of approximately 20,100 acres (Figure 2), not including the drainage area of Upper Lake Mary. Lower Lake Mary tends to be more susceptible to drought than Upper Lake Mary and can dry up during periodic droughts that affect the area. There are three major ephemeral drainages that contribute to Lower Lake Mary (Figure 3): two unnamed drainages (2,457 acres and 7,142 acres), and Priest and Howard Draw, which merge above the lake (5,875 acres).

The Soldiers Lake complex probably existed in some playalike form before western settlement. However, now the lakes are hydraulically connected by an extensive series of manmade dikes and canals. Historically, these waterways were constructed by the Tremaine Cattle Company to supply water to the Bar T Bar Ranch. To define the hydrologic connections among the lakes, Dr. Gremillion, conducted a topographic survey of the channels connecting the lakes and of the spillways draining the lakes (Figure 4).

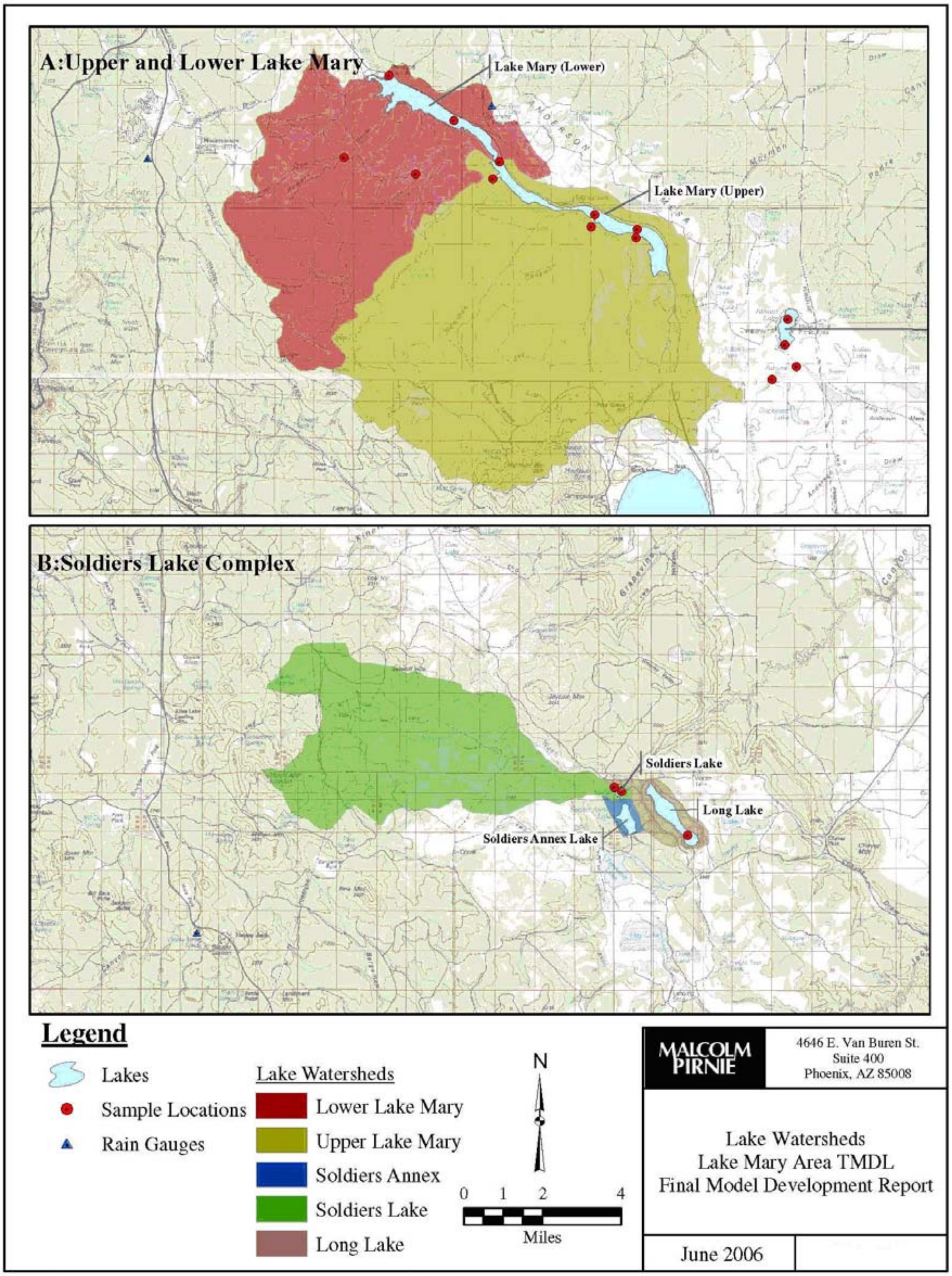

Figure 2. LMR Lake Watersheds (adapted from Malcolm Pirnie, 2006)

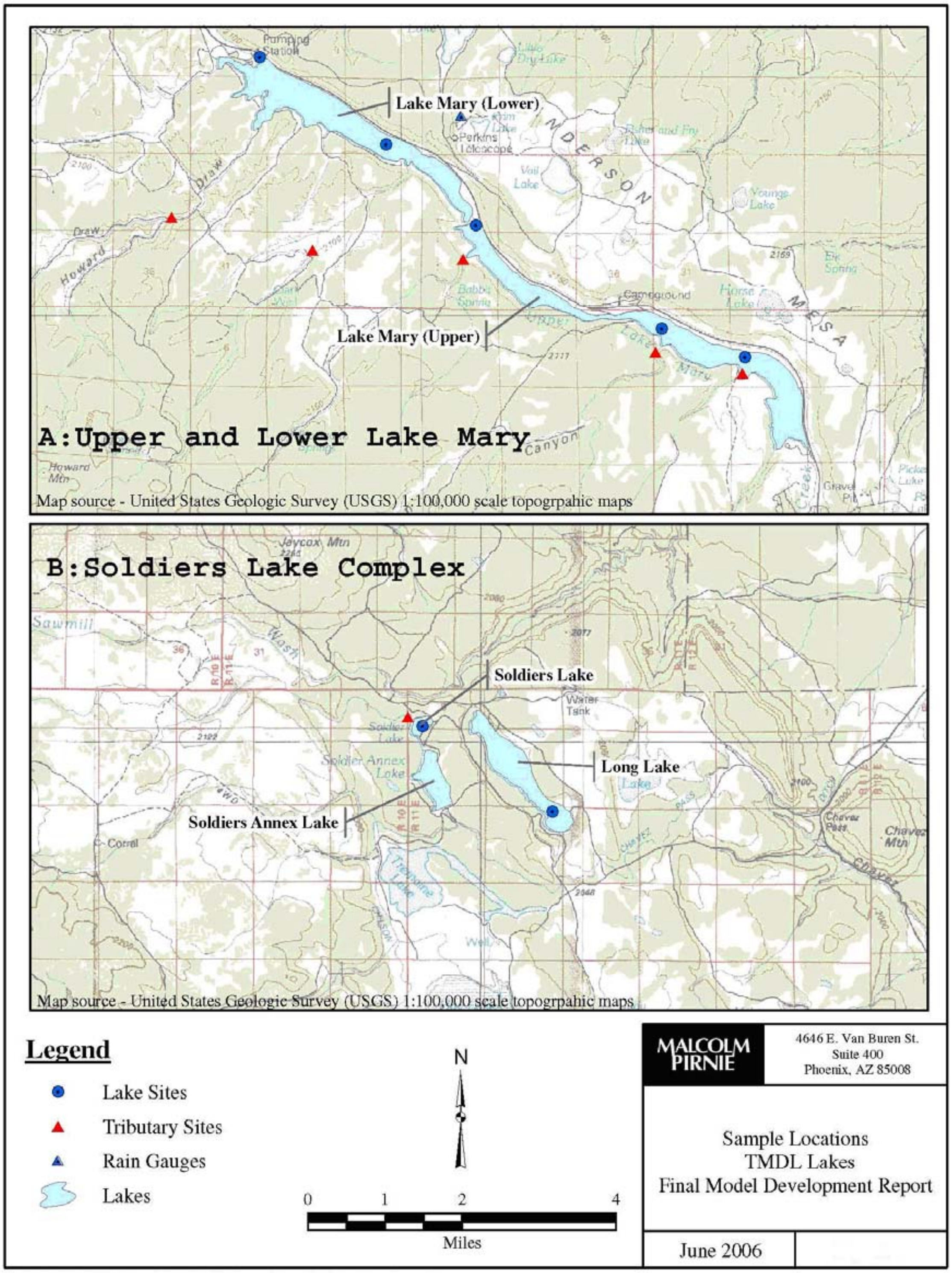

Figure 3. Sampling Locations (adapted from Malcolm Pirnie, 2006)

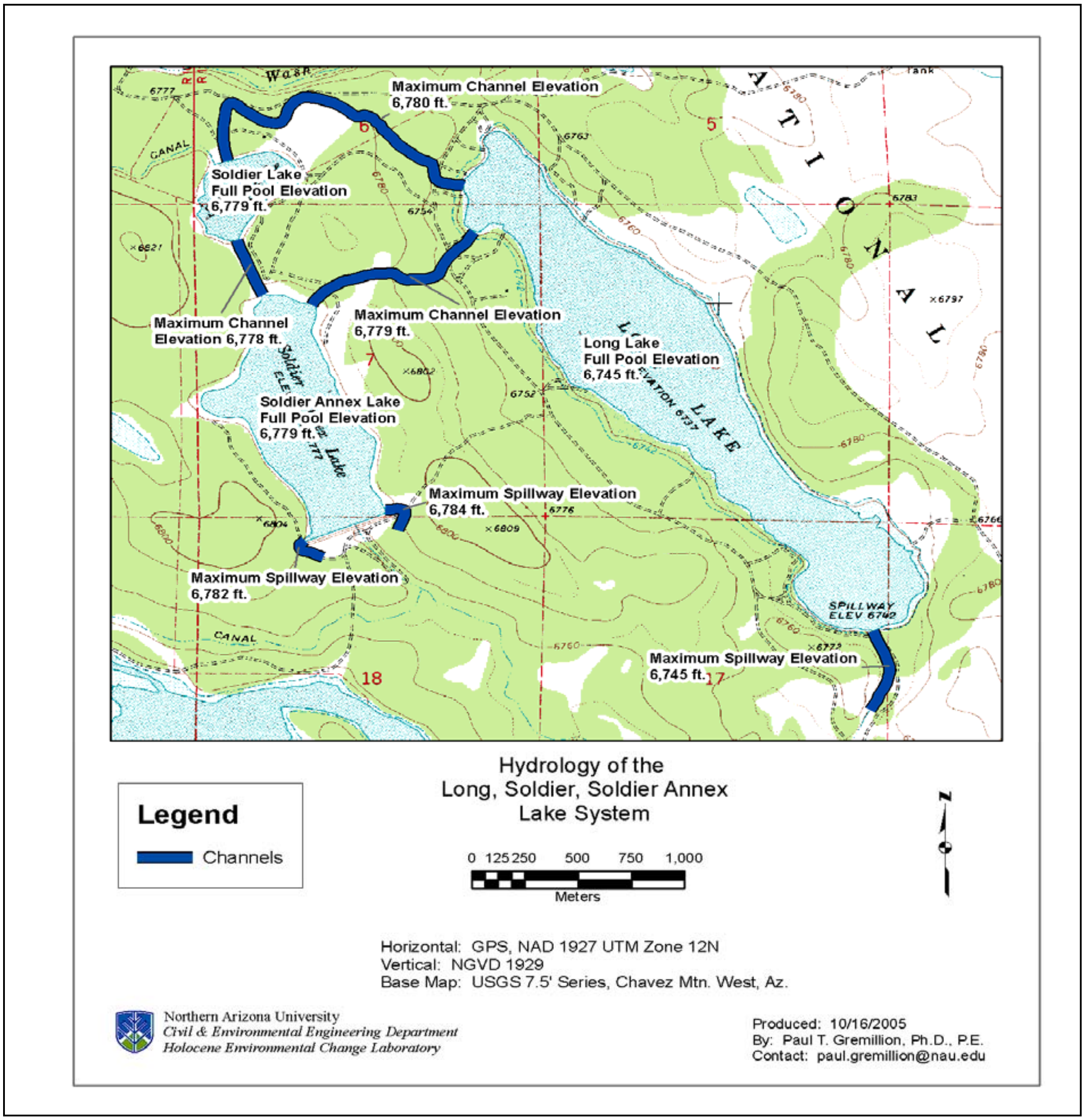

Figure 4. Hydrology of Soldiers-Long Complex (adapted from Gremillion and Toney, (2005)

In the upper Soldiers Lake watershed, a diversion dam was built partway across Sawmill Wash to divert water to a dike system directing the flow southeasterly to Soldiers Lake. Above an elevation of 6,778 feet Soldiers Lake is connected to Soldiers Annex Lake by a channel at the south end of Soldiers Lake. Below that elevation the lakes are separated. That elevation corresponds roughly to the full pool elevation of both lakes. When Soldiers Annex Lake fills to an elevation of 6,779 feet or above, water spills into Lower Long Lake via a channel built between Soldiers Annex and Lower Long Lake. With the exception of winter 2005, water levels in the past six years have not been high enough in Soldiers Annex Lake to result in flow to Lower Long Lake. Above an elevation of 6,780 feet, water flows through a channel connecting Soldiers Lake with Lower Long Lake. At an elevation of 6,782 feet, water then flows out of Soldiers Annex Lake over the west spillway. Additionally, the elevation of Soldiers Annex Lake may be drawn down through a valve at the dam. The hydrology of Lower Long Lake is influenced by direct runoff from its immediate watershed and from flows from Soldiers and Soldiers Annex Lakes.

D. Geology

On a regional scale, the entire LMR is surrounded by recent volcanism. With the exception of Willow Springs Lake, which is in a sedimentary rock setting, all the other lakes included in this study are underlain by a series of basalt and sedimentary rocks that have been structurally altered by folds, faults and joint fractures (Koval, 1976). Volcanic geology has the highest potential of any of the rock types to contain mercury compounds (USGS, 2003).

E. Land Cover/Vegetation

Vegetation types within the watersheds vary with elevation: the Upper and Lower Lake Mary watersheds are characterized by Ponderosa Pine, Gambel Oak, Pinyon Pine and Juniper. Vegetation types in Soldiers, Soldiers Annex and Lower Long Lake Complex watersheds consist primarily of Ponderosa Pine, Gambel Oak, Pinyon Pine and Juniper. However, a small portion of the watershed is characterized by arid grasslands. Historic grazing and timber harvest practices have resulted in areas of increased runoff and sedimentation.

IV. DATA COLLECTION & DATA SUMMARY

This section summarizes the geographic, hydrologic, meteorological, atmospheric mercury, water quality, soil, and biota data that were compiled for the LMR TMDL. Data were collected by ADEQ, NAU, AGFD, and the City of Flagstaff Water Treatment Plant (WTP). Data used in the models are summarized in the Lake Mary Regional TMDL Data Summary Report (Malcolm Pirnie, 2005). Lakes were sampled from 3 to 5 times at 1 to 3 locations and two depths, including sediment. Tributary runoff and sediment samples were collected 1 to 5 times. Spatial and temporal data gaps exist, particularly due to storm frequency and duration, and site accessibility.

Spatial data available for this TMDL included: DEM-delineated sub-watersheds, land use, land ownership, USGS topography, vegetation, soils, geology, and precipitation. In addition, modelers referenced the Coconino National Forest Terrestrial Ecosystem Survey (2001) and a rainfall-runoff study from the adjacent Wet Beaver Creek watershed (Baker, 1982).

Collectively, these data are used to define the sources and transport characteristics of mercury in LMR lakes in support of mercury TMDL modeling. The following sub-sections describe origin, scale and other characteristics of each data type. Additional tabular data may be found in the Lake Mary Regional Model Development Report (Malcolm Pirnie, 2006).

A. Fish Tissue Sampling

Between 2001 and 2008, fish tissue data were collected by ADEQ and AGFD from Upper Lake Mary, Lower Long Lake, Soldiers and Soldiers Annex lakes, as well as Ashurst Lake and Kinnikinick Lake. These data were collected from a variety of fish species including rainbow trout, northern pike, largemouth bass, yellow bass, walleye, catfish, bluegill, and crappie. All samples collected between 2001 and 2005 were submitted to the Arizona Department of Health Services (ADHS) laboratory for total mercury analysis of filets. ADHS has a detection limit of 0.25 mg/kg. The 2008 samples, six trout and one pike, were sent to the Region 9 EPA laboratory in San Francisco, which has a detection limit of 0.025 mg/kg. Only two trophic level-4 fish (northern pike) were collected from the background lakes; results averaged less than 0.30 mg/kg mercury. Five trout and eight catfish were also analyzed from the background lakes and the average was 0.30 and 0.35 mg/kg respectively. The following observations can be made:

- The highest fish tissue concentrations were observed in walleye collected in the TMDL lakes.
- All species in Soldiers Lake exceeded the human health threshold, with the exception of rainbow trout.

A summary of the existing mercury concentrations in fish tissue is presented in Table 2.

Table 2. Summary of Fish Tissue Data: Number and Average Mercury per Species (mg/kg)

Species	Upper Lake Mary	Lower Lake Mary	Soldiers	Soldiers Annex	Lower Long Lake
Walleye	(9) 1.01	NA	(10) 1.65	NA	(7) 0.71
Northern Pike	(7) 0.60	NA	(2) 1.21	NA	(5) 0.25
Largemouth Bass	NA	NA	(1) 0.36	NA	NA
Yellow Bass	(10) <0.25	NA	NA	NA	NA
Crappie	(2) <0.25	NA	NA	NA	NA
Channel Catfish	(3) 0.18	NA	(2) 0.42	NA	NA
Bluegill	NA	NA	(2) 0.45	NA	
Rainbow Trout	NA	NA	(1) 0.14	NA	(5) 0.07

(#) - number of samples per species

Scatterplots of fish length versus fish mercury concentration in walleye samples for Upper Lake Mary, Lower Long Lake and Soldiers Lake show a significant positive linear relationship. Soldiers Lake results show the highest mercury values and steepest slope (Figure 5). This may be due to the fact that Soldiers has the highest watershed-to-lake ratio of the lakes studied.

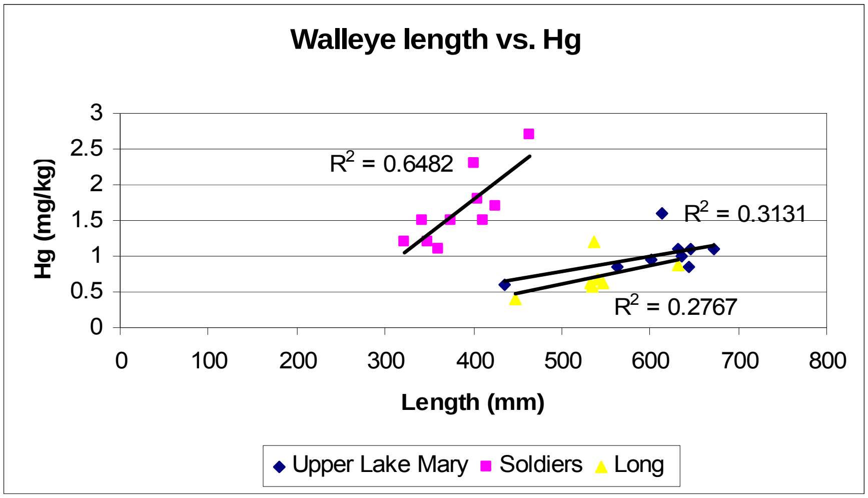

Figure 5. Comparison of Walleye Mercury Levels Between Lakes

B. Lake and Tributary Sampling

Analytical data for the lakes and their tributaries were available from ADEQ and the AGFD. The AGFD data included lake chemistry samples collected in 1983, 1987-88, 1993, 1998, and 2001-03. ADEQ data, which included lake water, lake sediment, tributary runoff, and soils, were collected in 1993-94 and 2000-05.

ADEQ lake data were collected from one to three stations per lake depending on lake size as well as several depths within each lake. Samples were collected over several seasons. A profile of basic field parameters was collected at depth intervals of one meter at each lake location prior to sample collection. Basic parameters for lab analysis of lake water included suites for general chemistry, nutrients, and total and dissolved metals. Chlorophyll-a and phytoplankton species identification were also run. Sediment was analyzed for total metals and physical characteristics.

Clean Sampling field methods (EPA Method 1669) were used to collect total, dissolved, and methyl-mercury in water, and total and methyl-mercury in sediment. Sulfate and sulfur-reducing bacteria as well as total and dissolved organic carbon data were also collected. These “clean” mercury samples were shipped overnight to Brooks Rand Laboratory in Seattle Wash., for low-level analysis.

ADEQ obtained lake water quality data during periods of thermal stratification, usually mid-summer, and also during periods when the lake was unstratified (well-mixed), typically the winter season. The results of the lake water quality sampling are summarized below.

1. Thermal Stratification

All the lakes were stratified with respect to temperature beginning in April through August, and in some cases into early September. The degree of stratification and depth at which the temperature change occurred were dependent on the water level in the lake. In Upper Lake Mary at the dam, temperature change began at a depth of 4 meters, and by September, the water was well-mixed with respect to temperature. At Soldiers Lake, the temperature profile in August 2005 indicated that the water temperature decreased from about 12 C at the surface to about 8.5 Celsius below 2 meters.

2. Dissolved Oxygen

The dissolved oxygen (DO) concentrations in the TMDL lakes were mostly above 4 mg/L throughout the stratified period. Notably, the ADEQ dissolved oxygen data indicate that the water column in the TMDL lakes remained oxygenated during the stratified period despite the thermal stratification. However, unpublished data obtained from the Flagstaff WTP suggest that the bottom waters of Upper Lake Mary can go hypoxic or anoxic where the lake has a depth of 10 meters or greater, depending on climatic conditions. The maximum depth observed at Upper Lake Mary during this TMDL study was 7.7 m.

3. Mercury

Total, dissolved, and methyl-mercury concentrations ranged from 0.9 to 29.7 ng/L, 0.07 to 18.5 ng/L and <0.02 to 16.2 ng/L, respectively. The highest mercury concentrations were observed in July 2004 at Lower Long Lake, corresponding with high organic carbon and total suspended solids (TSS). This measured value at Lower Long Lake should be interpreted with caution because the field observations indicate that the lake was almost dry and the sample may have included surface sediment. Concentrations of methyl-mercury in upper and bottom waters were only available for Upper Lake Mary at the dam in June and August 2003, and the observed values did not indicate significant methyl-mercury accumulation in bottom waters during the stratified period. For Upper and Lower Lake Mary, which have multiple sample locations within each lake, there were no significant differences in average total, average dissolved, and average methyl-mercury concentrations among the different sites within each lake. On average the background lakes contained less mercury (total, dissolved and methyl-mercury) than the TMDL lakes.

4. Other Water Quality Constituents

Total (TOC) and dissolved (DOC) organic carbon ranged from 10.8 mg/L to 159.9 mg/L, and from 12.5 mg/L to 152.7 mg/L, respectively. However, values of organic carbon in the hundreds are not common in surface water; only Lower Long Lake results were this high and it is likely the results reflect capture of sediment in the water sample. The highest organic carbon concentrations were observed in Lower Long Lake in 2004 when the lake was nearly dry.

TSS ranged from non-detect to 600 mg/L, with the highest value observed at Lower Long Lake in July 2004, again as a result of sediment capture. Average TSS concentrations were above 40 mg/L in Lower Long and Soldiers lakes and below 25 mg/L in the other lakes. Sulfate concentration in lake water ranged from non-detect to 65 mg/L. Average sulfate concentrations were comparable among the different lakes. Sulfide concentrations in lake water were mostly less than the detection limit of 0.1 mg/L.

5. Correlations between Constituents

Total mercury in lake water had a weak positive correlation with sulfate concentration, whereas, methyl-mercury had weak but positive correlations with DOC and sulfate, and a moderate correlation with chlorophyll-a. In general, higher productivity, carbon and sulfate appear to promote formation of methyl-mercury. The average water column methyl-mercury in TMDL lakes was 0.802 ng/L; TMDL lake summaries are in Table 3.

Table 3. Comparison of Average Water Quality Results between Lakes

Constituent	Units	Lower Lake Mary	Lower Long Lake	Soldiers Lake	Upper Lake Mary	Avg. TMDL Lakes
DOC	mg/L	18.1	152.7	12.5	10.1	34.9
TOC	mg/L	21.2	159.9	14.5	10.8	40.9
Chl-a	mg/L	13.9	6.1	8.6	4.6	7.1
Hg, unfiltered	ng/L	6.61	14.01	8.72	13.63	11.72
Hg, filtered	ng/L	2.99	1.47	3.86	5.34	4.24
Hg, particulate	ng/g	51	427	758	1037	725
MeHg	ng/L	0.368	5.839	0.119	0.165	0.802
TDS	mg/L	34.7	132.8	53.5	13.1	54.4
Sulfate	mg/L	4.6	18.7	12.7	26.3	20.1

C. Lake Sediment Results

ADEQ collected lake sediment samples at each lake site between 2003 and 2005, and analyzed them for total and methyl-mercury, and other analytes such as organic carbon, sulfates, sulfur-reducing bacteria count, and other physical parameters.

1. Mercury

Total mercury concentrations in sediment from TMDL lakes ranged from a minimum of 9.0 ng/g, detected at the Lower Long Lake dam, to a maximum of 80.9 ng/g, detected at the Upper Lake Mary dam. Methyl-mercury concentrations in sediments ranged from a minimum detection of 0.3 ng/g to a maximum detection of 0.54 ng/g (Table 4). Lake sediment total mercury concentrations between the TMDL and background lakes were, on average, equal. The exception was Willow Springs Lake, which had the highest total and methyl-mercury values in sediment, but low values in the water column and no sulfur-reducing bacteria. Set in limestone geology instead of volcanic geology, Willow Springs Lake has very low hardness, conductivity, and sulfate in the water column. The one largemouth bass analyzed from Willow Springs Lake showed mercury at only 0.17 mg/kg. TMDL lake methyl-mercury concentrations were, on average, higher than the background lakes. Other mercury TMDLs in Arizona have found that the magnitude of sediment mercury does not necessarily correspond to fish tissue impairment.

Table 4. Average Lake Surface Sediment Mercury Concentrations

Lake	Methyl-mercury (ng/g)	Total mercury (ng/g)
Upper Lake Mary	0.3	50
Lower Lake Mary	0.54	60
Soldiers Lake	0.35	34
Lower Long Lake	0.4	29

2. Other Sediment Constituents

TOC concentrations in sediments ranged from a minimum of 0.83 mg/kg detected at the midlake location of Soldiers Lake to a maximum of 3.11 mg/kg detected at the Lower Lake Mary dam. Average TOC concentrations in sediments ranged from 1.3 mg/kg at Soldiers Lake to 2.71 mg/kg at Upper Lake Mary. The sediment TOC concentration was of the same order of magnitude in all lakes.

Sulfate concentrations in sediments ranged from a minimum of 24 mg/kg detected at the upper lake sample site of Upper Lake Mary to a maximum of 593 mg/kg detected at the Upper Lake Mary dam. The average sulfate concentration equaled 223 ng/g in LMR lake sediments.

3. Correlations between Constituents

No significant correlation was observed between total mercury and TOC in lake sediments. Similarly, no significant correlation was observed between methyl-mercury and TOC in lake sediments. Total mercury had a weak inverse correlation with percent solids, but methyl-mercury did not. Neither mercury species had a significant correlation with oxidation-reduction potential or sulfate.

D. Watershed Sub-surface and Surficial Soil Results

The first set of watershed sediment samples were collected by ADEQ and analyzed for total mercury using EPA Method 7471A with a detection limit of 100 ng/g (ppb). Only three out of 14 samples had detectable total mercury concentrations (Table 5), with an average detection of 140 ng/g.

Table 5. Surface Soil/Sediment Collected During Runoff

Site	Hg (ng/g)
Marshall Lake	< 100
Upper Ashurst watershed	110
Coulter Ridge	< 100
Vail Lake	190
Anderson Mesa	< 100
Lower Elk Meadows	< 100
Upper Elk Meadows	< 100
Newman Sub Tributary	120
Newman Canyon	< 100
Near Railroad Tank	< 100
Clarks Well Area	< 100
Near Thomas	< 100
Priest	< 100
Howard	< 100

* Note high detection limit

Crustal mercury averages for basaltic or other volcanic rocks have been reported nationally to be in the range of 80 - 90 ppb. Additional sediment data, including sedimentation rates and metals deposition in each of the lakes, was provided in the study conducted for ADEQ by Gremillion and Toney (2005).

To establish background levels, soil samples were collected from the “B”-“C” soil horizons (10-12 in below the surface) at 20 upland sites in 2007. Sites were chosen randomly within the various watershed lithologies and spanned both the Lake Mary and Soldiers Lake complex watersheds. Analysis was conducted using low-level (EPA Method 1631) detection for total mercury (0.06-0.23 ng/g). Data are shown in Table 6 and Figure 6; average results are shown in Figure 7 for four geology types. No sample was greater than 40 ng/g and the overall average was 23 ng/g. In general, the range of values corresponded well to the average pre-impoundment value of 30 ng/g found at the bottom of lake sediment cores (Gremillion & Toney, 2005).

Table 6. Sub-surface Background Soil Data

Rock Type	Sub-type	Hg (ng/g)	Site ID
Basalt/Alluvium	Tby	10.07	LMR-1
Sedimentary	P	19.88	LMR-2
Basalt	Tby	31.63	LMR-3
Sedimentary	P	10.48	LMR-4
Basalt	Qtb	40.62	LMR-5
Sedimentary	P	18.77	LMR-6
Basalt	Tby	19.09	LMR-7
Basalt	Tby	36.48	LMR-8
Basalt	Qtb	39.31	LMR-9
Basalt	Tby	18.77	LMR-10
Volcanic	Qtv	39.02	LMR-11
Alluvium	Q	10.91	LMR-12
Basalt	Tby	11.20	LMR-13
Basalt	Tby	42.45	LMR-14
Basalt	Tby	39.05	LMR-15
Basalt	Tby	10.93	LMR-16
Basalt	Tby	14.03	LMR-17
Basalt	Tby	14.68	LMR-18
Basalt	Tby	15.78	LMR-19
Basalt	Tby	15.67	LMR-20
Arithmetic mean			
Geometric mean			

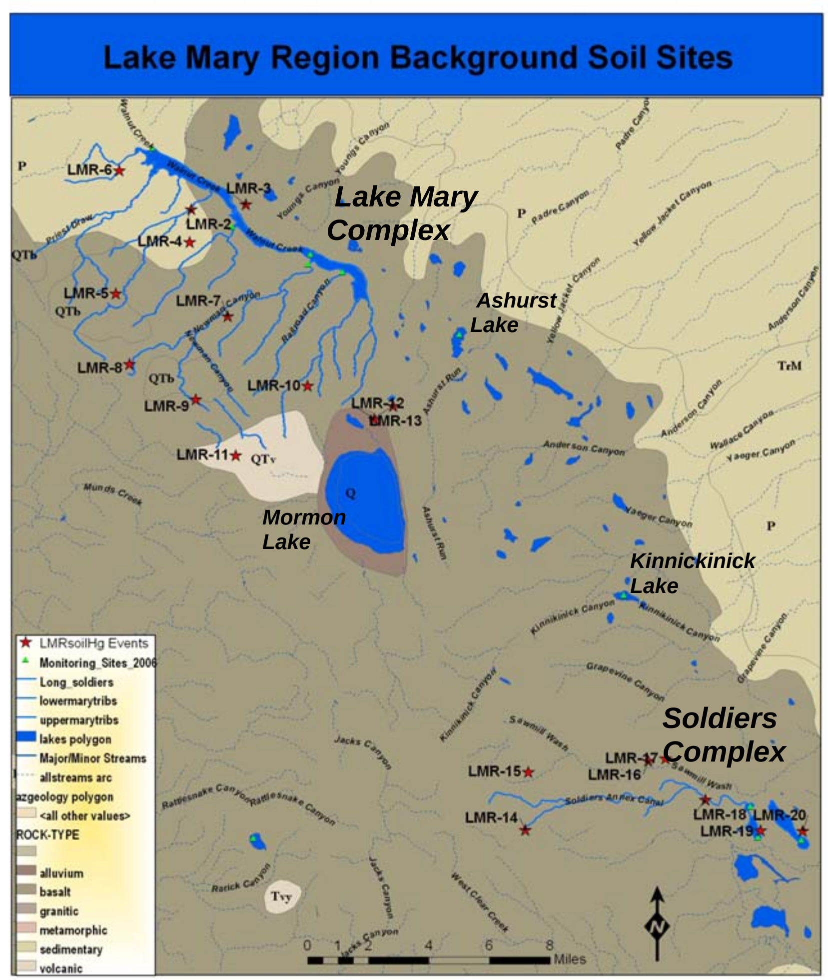

Figure 6. Locations of Background Soil Data and Other Data

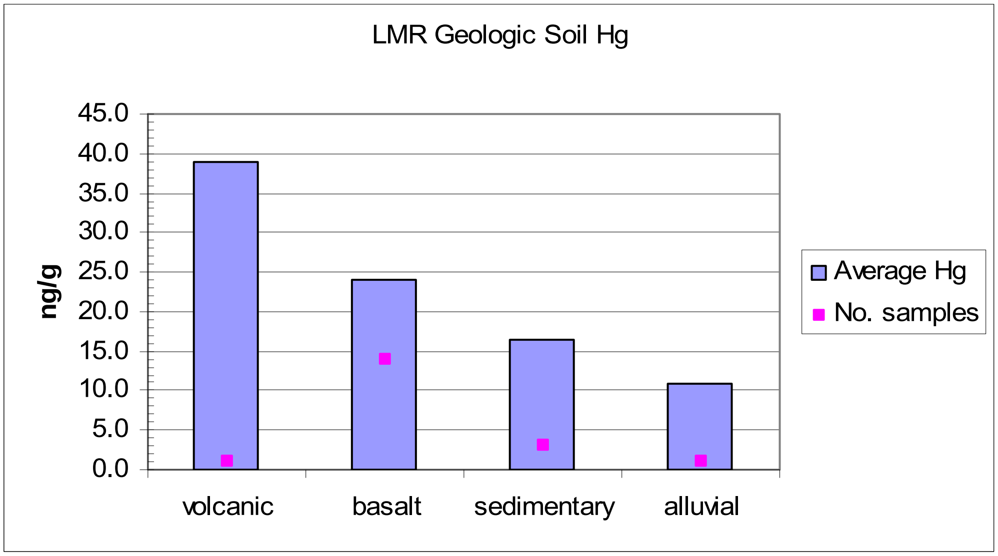

Figure 7. Results of Total Mercury Found in B/C Horizon Soils

E. Runoff and Stream Flow Results

ADEQ conducted water quality sampling during the spring runoff (snowmelt) period and storm event (summer monsoon) season. Monitoring sites were selected based on sub-watershed size and sampling was conducted when sufficient runoff was observed. ADEQ sample results are available from 2003 to 2005. Most tributary flows in the study area are ephemeral to intermittent, including the manmade canal flowing into Soldiers Lake.

Total mercury concentrations ranged from 1.2 ng/L in Babbit Spring Wash (tributary to Upper Lake Mary), to 22.7 ng/L in Kinnikinick Canyon (tributary to Kinnikinick Lake). Dissolved mercury concentrations ranged from 1.02 ng/L to 13 ng/L, with both values observed in Babbit Spring Wash.

Dissolved mercury results from monsoon flows in September 2003, at both Babbit Spring Wash and Newman Canyon Wash, exceeded the A&W chronic standard of 10 ng/L (range 13 – 15 ng/L), as did Soldiers Canal in September of 2004 (10 – 12 ng/L).

Methyl-mercury concentrations ranged from 0.02 ng/L to 0.23 ng/L with both values observed in Babbit Spring Wash. The average concentrations of total, dissolved mercury, and methyl-mercury were comparable in runoff to the TMDL lakes and runoff to the background lakes with the exception of the unnamed tributary #1 to Lower Lake Mary (Figure 8 and Figure 9).

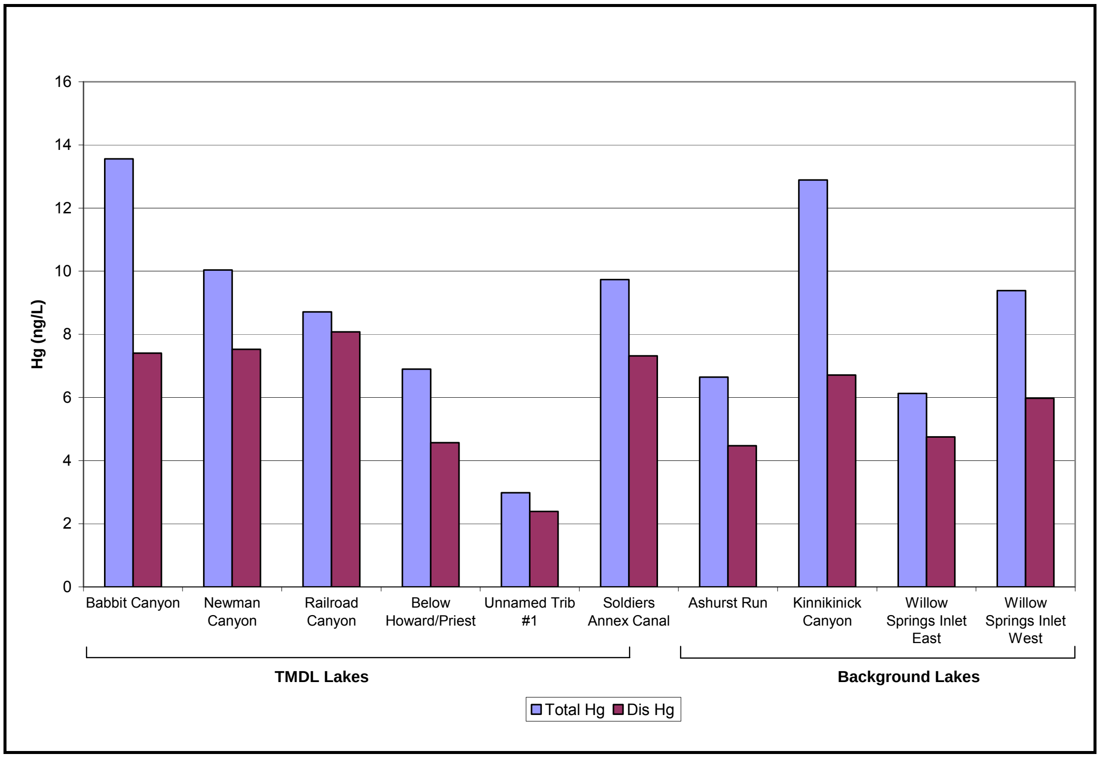

Figure 8. Average Total and Dissolved Mercury in Tributaries

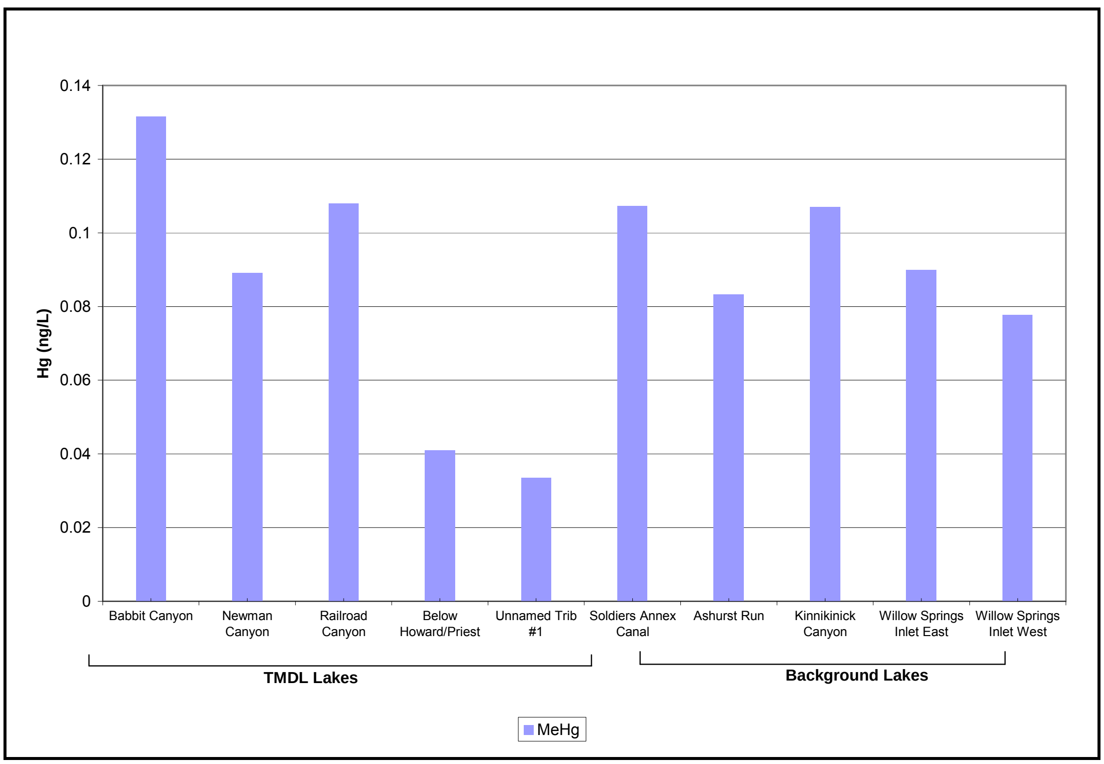

Figure 9. Average Methyl-mercury in Tributaries

The total mercury content of suspended solids was calculated using the total and methyl-mercury concentrations and the TSS concentrations from tributary samples. The calculations were performed for samples with data reported for all three analytes, and the results were expressed on a mass ratio basis (ng mercury per g solids). Total mercury concentrations associated with suspended solids ranged from 96 ng/g to 624 ng/g, with a mean of 200 ng/g (Table 7).

Table 7. Total Mercury in Runoff as Suspended Solids

TMDL- related Sites	**ng Hg per g TSS**
Babbit Spring Wash - Near Upper Lake Mary	571
Babbit Spring Wash - Near Upper Lake Mary	624
Babbit Spring Wash - Near Upper Lake Mary	576
Newman Canyon - Near Upper Lake Mary Inlet	420
Newman Canyon - Near Upper Lake Mary Inlet	276
Newman Canyon - Near Upper Lake Mary Inlet	394
Railroad - Near Upper Lake Mary Inlet	210
Below Howard-Priest Draw - Near Lower Lake Mary	179
Unnamed Trib #1	NA
Soldiers Canal - Near Soldiers Lake	418
Soldiers Canal - Near Soldiers Lake	235
Soldiers Canal - Near Soldiers Lake	209
Ashurst Run - Above Coconino Reservoir	498
Ashurst Run - Below Coconino Reservoir	252
Willow Springs E Inlet	161
Willow Springs W Inlet	96
Kinnikinick Canyon - Near Terminus	93
Kinnikinick Canyon - Near Terminus	86

As was the case for total mercury, the low methyl-mercury concentrations for the Lower Lake Mary tributaries can be attributed to the fact that samples were only collected during the January to April period, when relatively lower values were observed for all runoff sampled. Measurements of runoff pH were mostly alkaline, with the exception of a few observations associated with Willow Springs, where the influence of karst buffers pH. DO observed at the time of sampling was high in all sampling locations.

Mercury species in runoff had weak to moderate correlations with DOC, TOC, and TSS. The strongest correlation (Kendall's tau (t) = 0.71) was observed between dissolved mercury and DOC. There were few significant correlations between mercury species and flow, dissolved oxygen, pH, or water temperature. The DOC-dissolved mercury correlations support the concept that suspended sediments and organic carbon play a role in regulating the transport of mercury in the watershed (USGS, 2009).

V. SOURCE ASSESSMENT

A key component of a TMDL evaluation is the determination of known and potential sources of contamination to the watershed. Contaminant sources can include point sources, non-point sources, and background levels. There are no permitted mercury point sources to the lakes in the LMR. Potential external loads of mercury to the lake include direct atmospheric deposition to the lake, indirect atmospheric deposition via watershed inputs, natural background (geologic), and groundwater. The assessment of sources serves as the basis for development of a model, and as a basis for the allocation of the TMDL.

A. Atmospheric Deposition

Mercury in the atmosphere is present primarily in four forms:

- Gaseous elemental mercury vapor (Hg^0 or zero valence mercury);
- Gaseous divalent mercury (Hg^{2+}), also called reactive gaseous mercury;
- Particulate or particle-bound mercury (both Hg^0 and Hg^{2+}, relative proportion not known, and likely varying with type of particle); and
- Organic mercury (mostly mono-methyl mercury) which can be measured in rainfall, but in amounts so small, that the inputs are negligible in watershed studies.

There are two mechanisms of atmospheric deposition of mercury as specified in Volume III of the Mercury Study Report to Congress (EPA, 1997); these are:

Wet deposition - In this mechanism, reactive gaseous mercury dissolved in precipitation is deposited onto land and/or the surface of water bodies. Particle-bound mercury is also deposited by this mechanism, but is a relatively minor constituent in rain in most areas.

Dry deposition - In this mechanism, both gaseous and particulate forms of mercury are deposited on land, vegetation and/or the surface of water bodies by atmospheric mixing and adsorption, plus settling by gravity. Land uses and type of vegetation cover can affect the net dry deposition.

Important factors controlling deposition of mercury emitted to the atmosphere include weather patterns, the mercury species, and the distance of the emission source. It has been reported that gaseous divalent mercury released to the atmosphere has a relatively short residence time in the lower atmosphere, with the majority of this form of mercury deposited within 100 kilometers of the source (Dvonch et. al., 1999). Particulate bound mercury has somewhat longer residence time in the atmosphere and is generally deposited over a few thousand kilometers, while gaseous elemental mercury has the longest residence time and can be deposited over international or global scale distances.

One source of atmospheric deposition information is the Mercury Deposition Network (MDN) of the National Atmospheric Deposition Program (NADP). The MDN contains a national database of weekly concentrations of total mercury in precipitation and the seasonal and annual flux of total mercury in wet deposition. Arizona established its first MDN site in 2006 at the Sycamore Canyon (AZ02) air monitoring station near Williams. Aside from the Sycamore Canyon site, the closest stations, which are several hundred miles away from LMR, include: Chapin Mesa Station (CO99) in Montezuma County, Colorado, and Caballo Station (NM10) in Sierra County, New Mexico. The temporal patterns in wet deposition at these two remote MDN stations indicate the majority of the wet deposition typically occurs from June through October. Average weekly wet deposition rate of total mercury at these two stations, from 2000 to 2005, was 80 ng/m^2; on an annual basis and the average wet deposition rate was 4.2 *u*g/m^2.

As part of the Mercury Report to Congress, a national airshed model, the Regional Modeling System for Aerosols and Deposition (REMSAD) was applied to the continental United States. Deposition analysis in the REMSAD system is conducted using three global-scale models and two continental-scale models to both derive boundary conditions and likely background conditions from other countries.. This model provides a distribution of both wet and dry deposition of mercury as functions of air emissions and global sources. This model was based on the existing emissions inventory in 1995 and 1996 and it did not include any foreign airshed data (e.g. Mexico) that may impact Arizona. Because REMSAD is believed to underestimate mercury deposition in the arid Southwest, the EPA has recently developed a new three-dimensional grid-based Eulerian air quality model called the Community Multiscale Air Quality Model (CMAQ) (EPA, 2005). Emissions (deposition) output are provided with comparisons of global background contributions from both REMSAD and CMAQ models The CMAQ model incorporates spatial and temporal variations and complex transport and reactions of mercury in the atmosphere, and it is the best available model for evaluating total mercury deposition.

Mercury emission data are extremely limited and no known mercury point source currently exists in the local airshed. Based on simulated mercury tagged modeling results from the REMSAD model (Tetra Tech, 2008), Figure 10, at least 95 percent of atmospheric mercury originates from long-range transport from global mercury sources. This conclusion is similar to that reached in the 2007 study of mercury in fish from western U.S. streams and rivers (EPA, 2007). The remaining 5 percent is interpreted as the regional contribution.

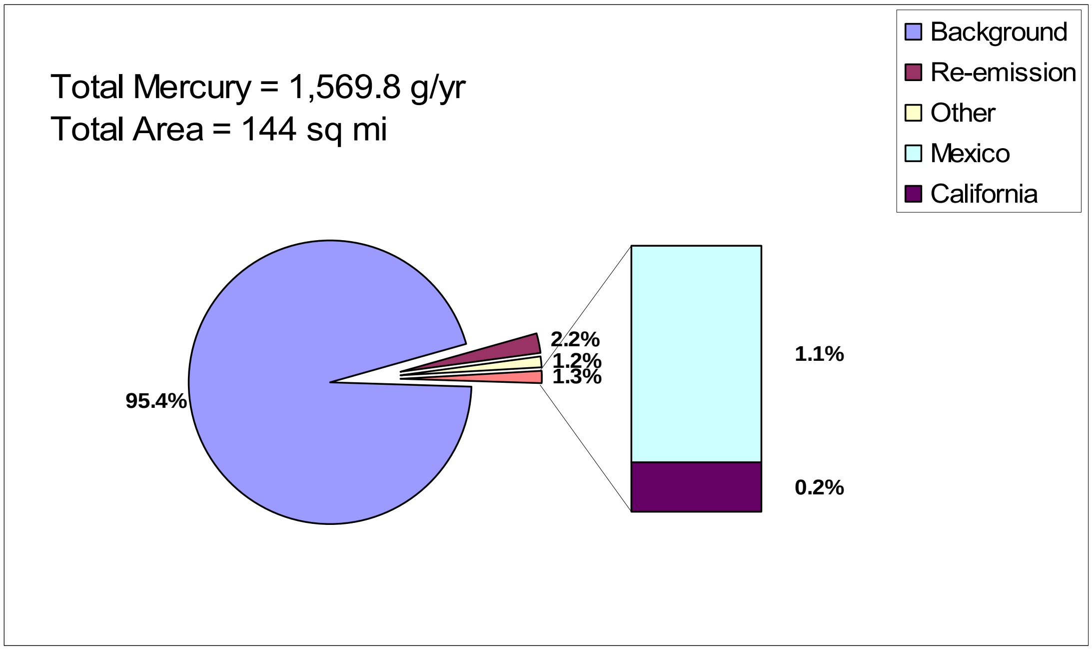

Figure 10. REMSAD Atmospheric Mercury Source Predictions

Although the REMSAD model underpredicts the total mercury deposition input to the Sycamore Canyon area, it is expected that the relative contribution of the atmospheric deposition from sources determined by REMSAD currently represents the majority of the anthropogenic mercury input to the LMR.

In 2008, Tetra Tech re-evaluated the national mercury air deposition models: CMAQ 2001 (meteorological data from 2001), CMAQ 2002 (meteorological data from 2002), and REMSAD. They also reviewed two years of data from the Sycamore MDN wet deposition station and Tekran dry deposition data collected at the Sycamore site, as well as data collected at Lyman Lake, Lake Pleasant, and Parker Canyon Lake in Arizona. Initial estimates used in the LMR model were 5 *u*g/m^2 wet deposition and 13 *u*g/m^2 dry deposition, for a total of 18 *u*g/m^2. Following the 2008 Tetra Tech review, the LMR model was updated using regional data from the Sycamore site: 11 *u*g/m^2/yr wet deposition and 24 *u*g/m^2/yr dry deposition (total of 35 *u*g/m^2). Both wet and dry deposition at the Sycamore station were higher than previously modeled.

Also in 2008, Tetra Tech summarized the mercury emissions reported in the 2006 Toxic Release Inventory (TRI). Table 8 shows mercury emissions in pounds for all reporting facilities in Arizona. Note that the Cholla Power Plant is the closest facility to the LMR, never-the-less, it is fifty to eighty miles east and downwind for most of the year (Figure 11 in TT, 2008).

Table 8. 2006 TRI Mercury Emissions (Tetra Tech, Inc., 2008)

Facility Name	**Total Mercury Air Emissions 2006 (lbs)**
Coronado Generating Station	551
Cholla Power Plant	321
Salt River Project Navajo Generating Station	283
Arizona Electric Power Cooperative	129
Tucson Electric Power Co Springerville	122
Phelps Dodge Miami Inc.	47.0
Phoenix Cement	41.6
Abitibi Consolidated Snowflake Division	33.5
Asarco LLC Ray Complex & Hayden Smelter/Concentrator	13.0
Irvington Generating Station	9.69
Veolia Es Technical Solutions LLC	0.69
Arizona Portland Cement Company	0.25
Honeywell Air Transport	0.12
Phelps Dodge (Freeport-McMoRan) Bagdad Mine	0.05
World Resources Company	0.01
Asarco LLC Ray Mine Operations	0
Chemical Lime Nelson	0
Earth Protection Services Inc.	0
Phelps Dodge (Freeport McMoRan) Sierrita Mine	0
TOTAL	1,551.9 lbs as of 2006

In theory, the mercury deposited to the LMR watersheds originates from sources both within the local airshed, and from regional/global sources located beyond the local airshed. Potential local and regional sources include coal-fired power plants, cement plants, kilns, smelters, historic sawmills and wood treatment facilities, and volcanism (Figure 11).

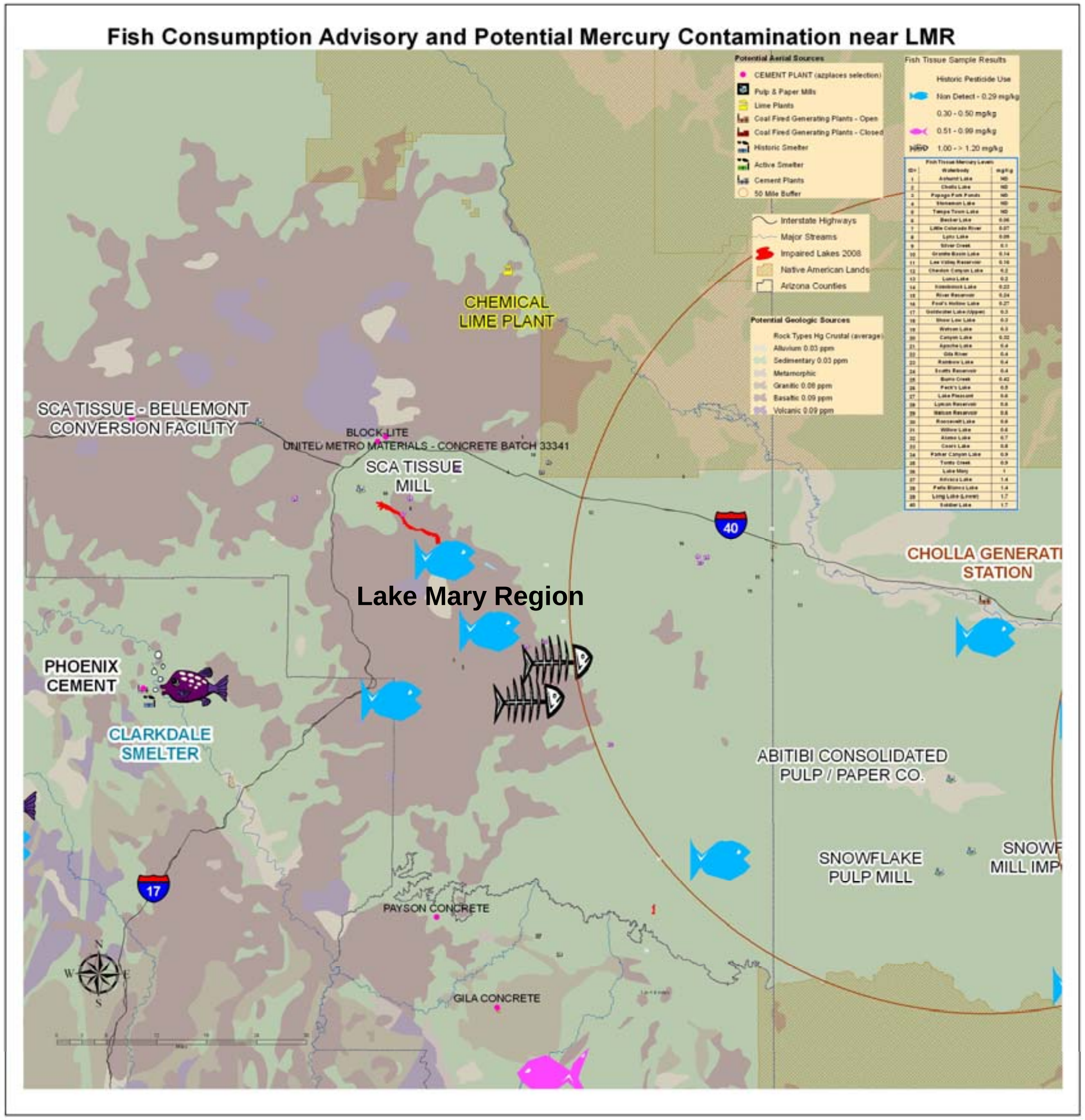

Figure 11. Potential Aerial Sources of Mercury near LMR

B. Timber Industry

In particular, the timber industry around Flagstaff thrived for many decades; one or more sawmills were present from the 1880s to the 1970s. Prior to creation of any of the LMR lakes, from 1880 until 1900, a sawmill was located where Upper Lake Mary is now in what was then called Clark's Valley (Figure 12).

Mills that produce pulp or paper would potentially release more mercury than a sawmill, but the early mill in the Upper Clarks Valley primarily produced railroad ties for local timber rail lines as well as supplying the construction of the Atlantic-Pacific Railroad (later the Sante Fe Railroad) that passes through Flagstaff. The process for preservation of railroad ties may have included treatment with mercuric chloride, which was popular before the turn of the century. In addition, mercury has commonly been used in switches and in mercury halide vapor lamps.

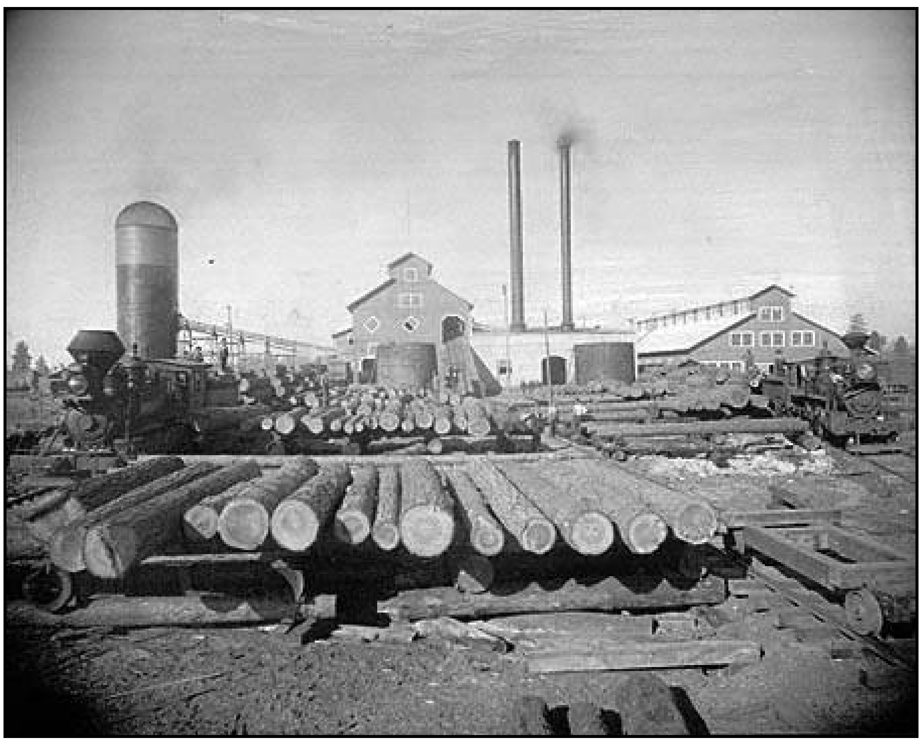

Figure 12. Arizona Lumber and Timber Company Sawmill: logging docks,1899.
(Courtesy of the Cline Library Digital Archives, NAU, 2010)

C. Fires

Aggressive harvesting of old growth Ponderosa Pine forests led to replacement with dense stands of smaller trees. A 2003 USGS study shows increases in stand replacement fires in Arizona and New Mexico between 1915 and 2000, in part due to diminished natural grassland breaks (Figure 13). Fires may contribute mercury to the air and to the watershed by release of mercury bound up in plant tissue, known as foliar mercury.

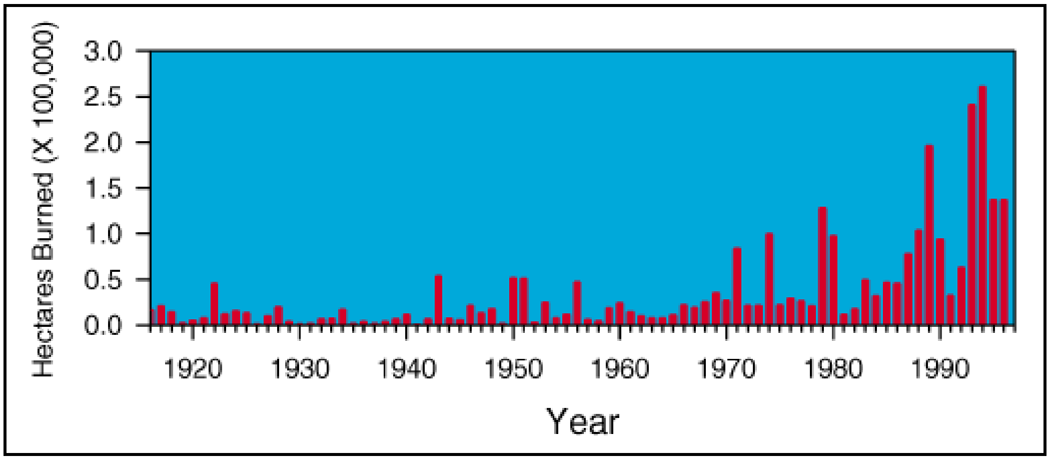

Figure13. Increase in Forest Fires in Southwest Region 3: 1915-2000, USGS, 2003

A Coconino County listing of historical hazards shows a 25-yr fire history within the County, many of them within or close to the LMR airshed. (Table 9).

Table 9. A 25-yr History of Forest Fires in Coconino National Forest

Date	**Name**	**Category**	**Damage**	**Address City**
1977		Natural		
June 1977	Radio Fire	Campfire	4,594 acres	Mt. Elden, Flagstaff
May 1986		Natural	198 acres	
July 1986		Natural	150 acres	
1987		Natural	100 acres	
1987		Natural	225 acres	
1987		Natural	300 acres	
1989		Natural	125 acres	
1989		Natural	765 acres	
1989		Natural	110 acres	
1990		Natural	145 acres	
1990		Natural	150 acres	
1990		Natural	320 acres	
May 1991		Natural	456 acres	
October 1991		Natural	300 acres	Flagstaff
June 1993		Natural	1762 acres	
August 1993		Natural	150 acres	
September 1993		Natural	250 acres	
November 1993		Natural	1000 acres	
1995		Natural	200 acres	
April 1996	Slate Fire	Human/Wind driven	275 acres	Flagstaff
1996	Hochderfer Fire	Natural	15,000+	Flagstaff outskirts

			acres	
May 1996	Switzer Fire	Natural	100+ acres	Flagstaff N. central
May 1996	Walnut Fire	Natural	100+ acres	Flagstaff S. side
1996	Side Fire	Human/wind driven	320 acres	Flagstaff E. side
1996	Horseshoe Fire	Slashpile/wind	8,650 acres	
May 1996		Natural	8,200 acres	
June 1996		Natural	700 acres	
June-July 1996		Lightning-caused:	82,000+ acres	
	Bridger Knolls/Jump		53,500 acres	N. Rim Grand Canyon
	Hochderfer Fire		16,400 acres	12 mi NW of Flagstaff
	Pot Fire		7,000 acres	10 mi NE of Sedona
	Cottonwood Fire		1,586 acres	1 mi S of Pinedale
June 1996		Natural	285 acres	
June 1996		Natural	48,000 acres	
May 2000	Outlet Fire	Prescribed burn/wind	13,350 acres	S of Jacob Lake N. Rim Grand Canyon
2000	Clover Fire	Natural	150 acres	Happy Jack
June 2000	Pumpkin Fire	Lightening-caused	14,760 acres	N/NE of Williams
2000	Pipe Fire	Lightening-caused	600 acres	NW of Flagstaff
2000	Power Fire	Lightening-caused	1,500 acres	E of Flagstaff
2000		Lightening-caused	84 acres	N of Sedona
2001	Leroux Fire	Abandoned camp/wind	1,250 acres	San Francisco Peaks
2002	Springer Fire	Natural	840 acres	Happy Jack
2002	Hart Fire	Natural	50 acres	NW of Flagstaff
2002	Tram Fire	Human-caused	191 acres	S of Happy Jack
2002	Big Fire	Natural	100 acres	N. Coconino County
2002	Pack Rat Fire	Lightening-caused	3,470 acres	N of Payson/Clint's Well
2002	Trick Fire	Natural	100 acres	S Coconino County
2002	Antelope Fire	Natural	100 acres	S. Central Coconino Co

D. Mining

An article from the Northern Arizona University website, entitled, *Land Use History of North America, Mining on the Colorado Plateau,* states that sedimentary rocks of the Plateau include pockets of coal, oil, natural gas, and uranium. However, the LMR is located along the southern border of the Plateau within a zone of surficial volcanism. Mining in the LMR has consisted almost exclusively of cinders, pumice, gravel, and underlying limestone, not likely sources of mercury (Figure 14).

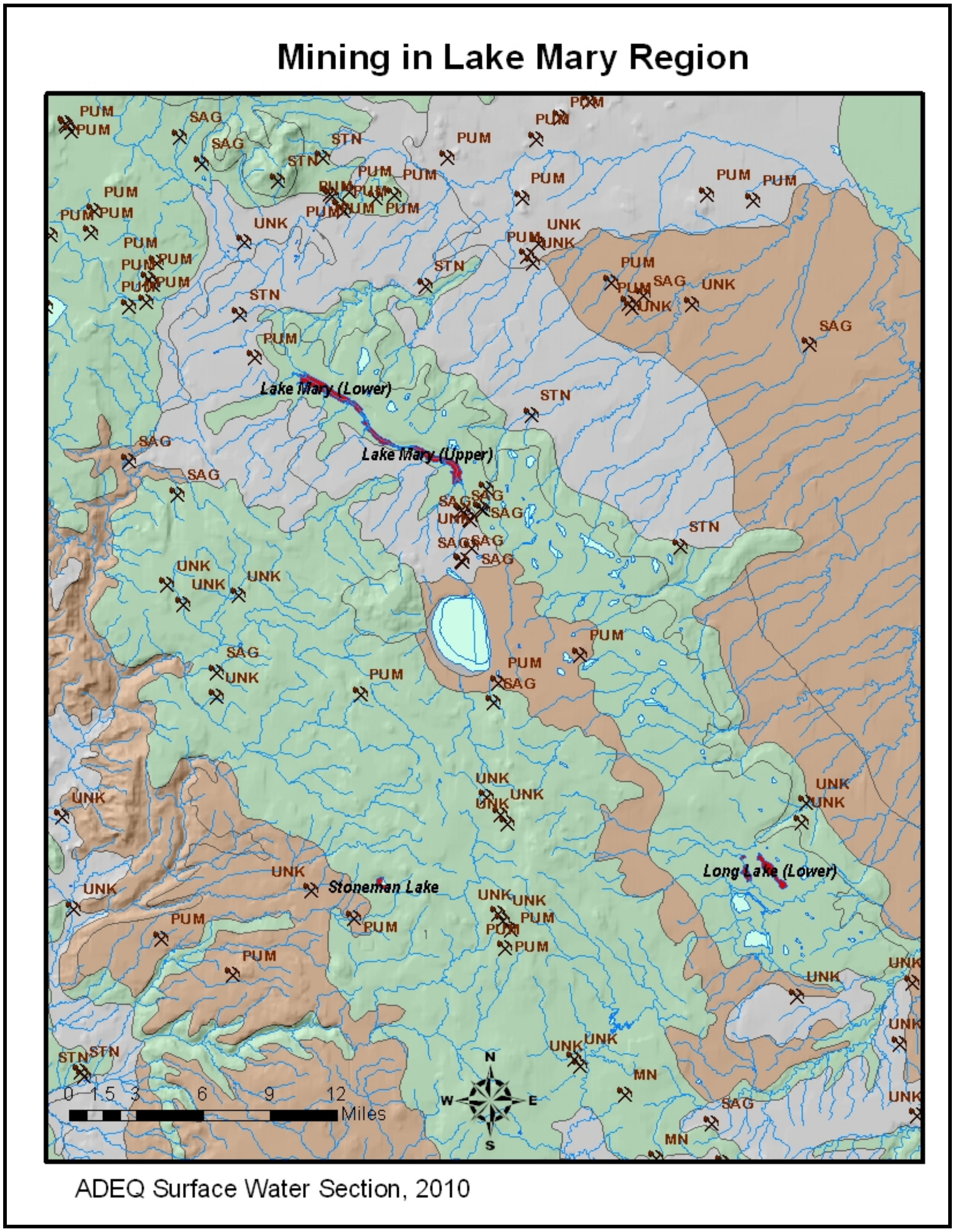

Figure 14. Extent of Mining Activities within the Lake Mary Region

E. Summary of Lake Coring Study

To understand the sources of mercury to lakes in Arizona, ADEQ contracted Gremillion & Toney (2005) to collect sediment cores from Upper Lake Mary and 11 other reservoirs in northern and central Arizona. All of the LMR TMDL lakes and background lakes were included in this analysis, with the exception of Soldiers Annex Lake, due to the fact that no mercury data had been collected in that lake at the time. Physical and geochemical analyses were performed on these cores to determine the chronology of sediment deposition, the concentrations of total mercury, lead, and zinc, and the stable isotope ratios of lead and plutonium. The two age markers used were radioisotopic analysis and date of impoundment. Radiometric dating provides a marker for the peak of above-ground atomic testing in 1963 (Beck and Bennett, 2002). The characteristic change in radioactive fallout over the period of 1954 to about the 1970s also provides an indication of sediment disturbance and remixing. A plot of plutonium$^{239+240}$, versus depth in the core, indicates that sediments have been deposited sequentially over time with minimal disturbance.

Results indicate that Upper Lake Mary and most of the other TMDL lakes experienced non-point increases in mercury and lead over their history. No evidence could be detected of point sources of either metal. Although changes in zinc over time were observed, significant zinc loading was not detected. Stable isotope data point to coal and gasoline combustion as likely sources of lead in lake sediments. Total mercury at the base of each lake core, at pre-impoundment depths, averaged 30-36 ng/g. Total mercury at the top of the cores averaged 80 ng/g (maximum of 140 ng/g).

Trends were similar for Upper Lake Mary (TMDL lake) and Ashurst (background lake). Lead ratio plots clearly showed variation in both lakes between the two end members used, coal (Pb 204/206) and leaded gasoline (Pb 208/206). This does not exclude other sources for the lead in these sediments but is consistent with a combination of these two. Additionally, a coal combustion source for lead may also explain some of the sources of mercury loading, as mercury is released with coal combustion (e.g., Yudovich and Ketris, 2005).

The non-point sources of mercury include geologic mercury from within the watershed and atmospheric deposition. Any patterns of change detected in one lake were also detected in other lakes in the same geographic area. An exception was the high concentrations of lead and zinc at the 39-centimeter position in the Upper Lake Mary core. This excursion, however, was clearly related to organic trapping of metals in peat during the pre-impoundment period in the lake. Plots of zinc, mercury, and lead versus loss-on-ignition, a measure of organic carbon, indicated generally positive correlations between metals and organic carbon on a lake-specific basis. These correlations do not reveal the origins of the metals, but do provide a mechanism to explain how it may be possible for crustal or anthropogenic metals to accumulate in concentrations higher than their original mineral concentration. With regard to mercury, Gremillion & Toney (2005) concluded that,

> "The growing consensus among the scientific community is that practically all watershed mercury has atmospheric deposition as the ultimate source. Because of the high affinity mercury has for organic matter, understanding the patterns of storage and release of organically-bound mercury from watersheds may be of far greater importance in managing lakes than detecting the ultimate source. Further study should be directed toward better understanding the delivery of mercury from watersheds and the biological availability of these

mercury forms. Observations are consistent with a non-point source of mercury. Upper Lake Mary and Ashurst have unconnected watersheds, but mercury concentrations varied similarly. Mercury flux calculations separate the lakes somewhat in terms of their delivery of mercury to the sediments. Lakes with low sedimentation or high bulk density tended to have higher mercury budgets".

Figures 15 and 16 demonstrate that Soldiers Lake with a higher sedimentation rate has lower bulk density and a much lower mercury budget than Upper Lake Mary (Gremillion & Toney, 2005). Because sedimentation is much higher at Soldiers Lake, the process of mercury loading and lake response at Soldiers Lake is condensed; consequently, bioaccumulation is accelerated.

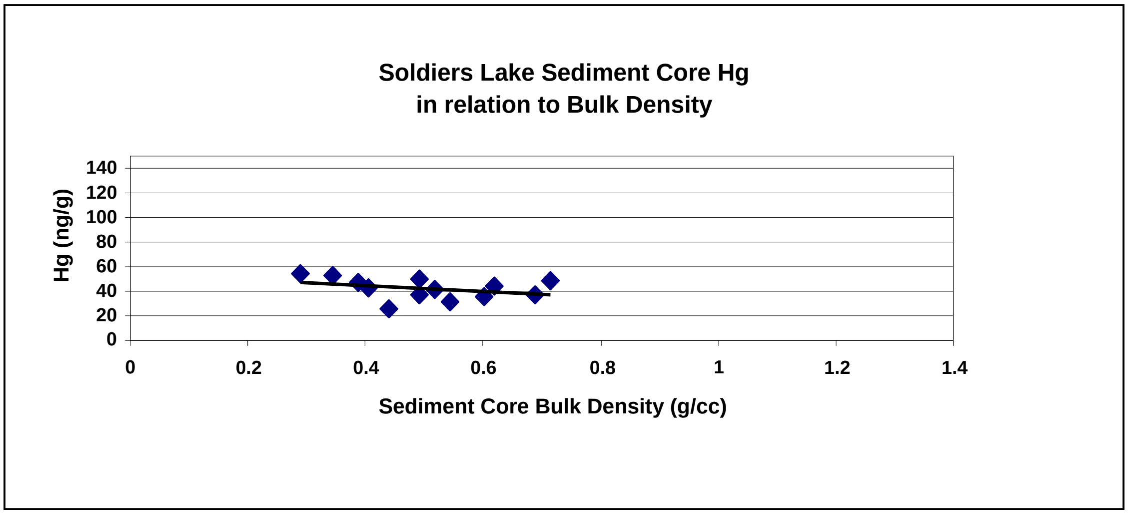

Figure 15. Soldiers Lake Sediment Core

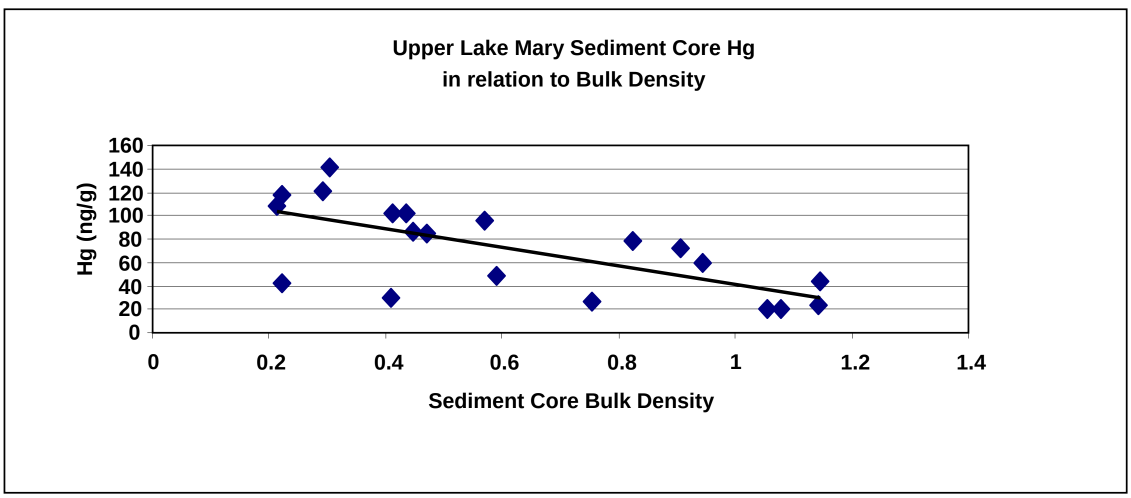

Figure 16. Upper Lake Mary Sediment Core

F. Determination of Watershed Background Mercury

The watershed input of mercury to the lakes consists of mercury derived from the parent geologic material and the net release of mercury deposited from the atmosphere to the watershed. Given that geologic weathering contributes to soils, a portion of the mercury in soils would come from mercury sources in the underlying geology. The USGS national crustal averages for mercury by rock type cite a concentration of 80-90 ppb for basaltic volcanic rocks versus 30-40 ppb for sedimentary rocks. Although the LMR lakes are located in surface volcanics, those volcanic rocks are underlain by limestone and sandstone. In addition, ongoing and historical atmospheric mercury deposition would also contribute to the mercury in the soils. Although some of the mercury deposited to the watershed is likely re-emitted to the atmosphere, some of it becomes particulate bound and is transported to the lakes through erosion and runoff.

It is generally difficult to differentiate between naturally occurring and anthropogenic-derived mercury concentrations in watershed ecosystems. To estimate background geologic mercury contribution, additional soil samples were collected from 20 locations within the "B/C" soil horizon (Figure 6). Collected from a depth of 10-12 inches, these soils were considered to be below the surface organic soil layer. Low level mercury analysis revealed a range of 10 ng/g to 43 ng/g (average of 23 ng/g, Table 6) of total mercury, which corresponded well with pre-impoundment lake sediment core values (30 ng/g to 36 ng/g) found by Gremillion and Toney in 2005.

Results from a USGS study (Gustavsson et al., 2001), provides an interpolated map of mercury soil concentrations in the continental United States, showing a range of 75 ppb to 150 ppb for the LMR study area. However, this report does not attempt to distinguish the geologic input from the net release of atmospheric deposited mercury. The mercury loading model for LMR required input of the mercury concentrations for soils eroded and delivered to the lake. The mercury concentration modeled for this purpose was 200 ng/g, derived as the average of surface soil sample results and suspended sediment mercury results (whole water and dissolved phase) found in runoff samples. A value of 30 ng/g was selected to represent background soil mercury. Subtracting 30 ng/g from 200 ng/g left 170 ng/g to account for in the indirect atmospheric mercury watershed load.

VI. MERCURY CYCLING AND SITE CONCEPTUAL MODEL

Mercury is a complex element that exists in elemental, inorganic and organic forms in aquatic systems. Each of these species has different ecological and toxicological impacts. Depending on the environmental conditions, mercury compounds can be inter-converted, released from sediments into aqueous phase, taken up by biota, released into the atmosphere, or transported to other locations. Methyl-mercury is the species that is easily absorbed by organisms and is effectively biomagnified up the food web. The mercury dynamics in the lakes of the LMR can be summarized in the conceptual diagram in Figure 17 (Malcolm Pirnie, 2006). The ellipses represent pools of mercury, while the arrows and rectangular figures represent fluxes and transformations.

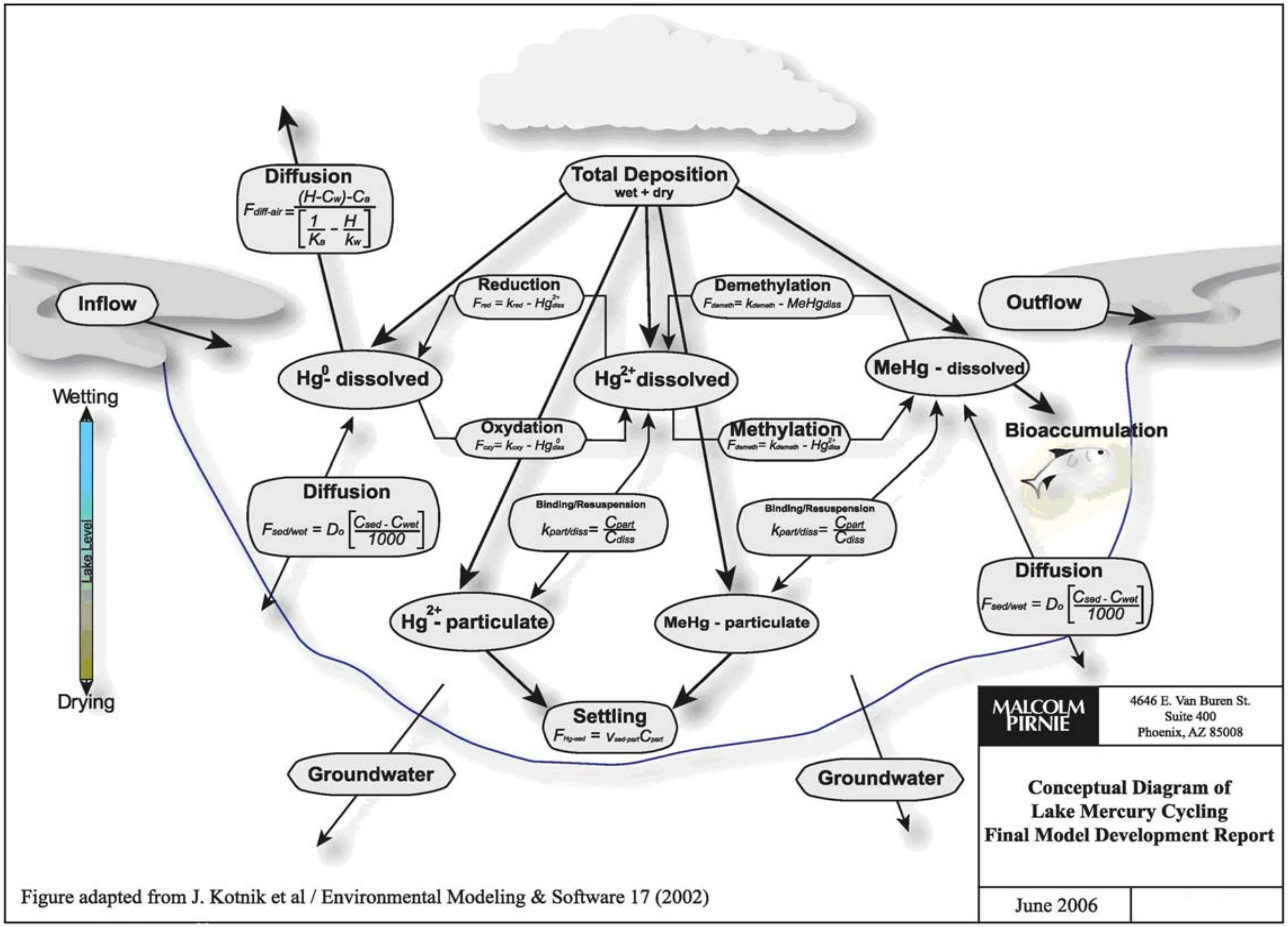

Figure 17. Conceptual diagram of mercury dynamics (Malcolm Pirnie, 2006)

The major sources of mercury to the lakes in the LMR system are atmospheric deposition and natural geologic materials. Anthropogenic disturbances such as grazing and timber harvesting have resulted in increased soil erosion and sediment delivery during winter storms and episodic rainfall events. A moderate correlation was observed between total and methyl-mercury species and organic carbon in the watershed, in particular DOC. This suggests that one of the important factors controlling the delivery of mercury through these watersheds is organic carbon.

Morphologically, Upper Lake Mary, Lower Lake Mary, and Lower Long Lake are elongated, whereas, Soldiers and Soldiers Annex are circular. However, there were no significant spatial differences in the water column concentrations of mercury species within the elongated lakes, therefore, the conceptual site model assumed for all the lakes represents each lake as a well mixed system (Gremillion and Toney, 2005).

Temperature profiles based on data collected by ADEQ indicate that the lakes are thermally stratified in summer. However, the related dissolved oxygen data showed no indication of strong anoxia. In wetter years with higher lake levels, Upper Lake Mary by the dam has been known to go anoxic. Mercury samples in top and bottom waters during the stratified period were only collected twice at Upper Lake Mary at the dam site and they did not show significant difference in methyl-mercury conditions between the epilimnion and hypolimnion. The lack of prominent anoxia suggests that the water body can be depicted as a single well mixed system and seasonal effects on water column mercury methylation can be captured through seasonal methylation rates.

Studies conducted by the USGS (Blee, 1988) suggested that groundwater seepage from Upper Lake Mary can account for 45 percent of the water lost from the lake. In general, the lakes in the Lake Mary Region are not hydraulically connected with the regional water table, and seepage rates are controlled by the porous media underlying the lakes. The USGS study developed a strong linear relation between the seepage losses and the elevation of water in the lake. Lower Lake Mary goes dry in most years because it contains a sinkhole that enhances infiltration to groundwater. It is also documented that Lower Long Lake went completely dry during the summer of 2004 (Gremillion and Piastrini, 2005).

The process of wetting and drying may play a role in methyl-mercury dynamics in the TMDL lakes. This process has been shown to enhance the production of methyl-mercury in the Florida Everglades soils and sediments (Gilmour et al, 2004). There it was observed that high levels of methyl-mercury production coincided with high levels of sulfate in surface water and sediment pore water following rewetting of dry areas. Based on this observation, it was hypothesized that the release of sulfate from the soils, as well as release of sediment-bound mercury, stimulated methyl-mercury production following the drought period. Modeling such processes requires dynamic modeling of sulfate and sulfur reduction. Therefore, this process, as hypothesized in the Florida Everglades study, could not be evaluated in the LMR study.

Within each lake the processes that control the transformation of mercury are included in the conceptual diagram (Figure 17). The predominant form of mercury is inorganic Hg^{2+}, which is partly reduced into Hg^0, which can be evaporated back to the atmosphere, and partly methylated into methyl-mercury, which can be de-methylated to Hg^{2+} and further reduced to Hg^0. Methylation and de-methylation in water and sediment is primarily a biological process carried out by different aerobic or anaerobic micro-organisms. Also possible, but less frequent, is abiotic methylation. This conceptual model assumes that the fish tissue concentrations are related to the water column methyl-mercury concentrations through the Biological Accumulaton Factors (BAFs). It is assumed that these site-specific BAFs capture the effect of all the important factors.

VII. MODEL DEVELOPMENT

In order to formulate meaningful TMDLs for lakes in the LMR, it was necessary to quantitatively link the fish tissue mercury concentrations to the sources of mercury to the lakes. This linkage was achieved through a mechanistic model or sets of models that can be used to simulate cause and effect relations between fish concentrations and changes in mercury loading or other lake parameters (Malcolm Pirnie, 2006 with updates in 2007, 2008, and 2009). Four different types of models were developed and linked for this project:

- A watershed loading model;
- A lake hydrologic model;
- An in-lake mercury cycling model; and
- Mercury bio-uptake calculations.

A. Watershed Hydrologic and Sediment Loading Model

In order to model the mercury concentration in the lakes within the study area, it was necessary to estimate the total input of mercury from external sources, including sediment and water transported via runoff, atmospheric deposition to the watershed, mercury from geologic sources, groundwater inputs, and any inputs related to historical land disturbance activities. Determination of the watershed loads requires estimation of flows and mercury concentrations in the input waters to the lakes. Because there were only a few measurements of flow and mercury concentration for tributaries to the lakes in the LMR, and a significant relation between flow and concentrations was not observed, a watershed loading model was needed.

Watershed loading models range from simple empirical estimations of precipitation-runoff relations to complex deterministic models. A simple method was used for the LMR watershed because:

- There are no continuous stream flow or runoff records by which to calibrate a complex hydrologic model.

- With the exception of Upper Lake Mary, which has some historical lake levels measured by the Flagstaff WTP (Jack Ricken, personal and electronic communication, 2005), there are no available historical records of lake level for the other lakes.
- A deterministic hydrologic model would require information on complex hydrologic processes (such as snow pack dynamics and groundwater flow) for which little to no watershed-specific information exists.

Therefore, the benefits of hydrologic complexity would be nullified by difficulties of parameter estimation and calibration. Several methods exist for estimating hydrologic and pollutant contributions from a watershed. These include the Simple Method, the Generalized Watershed Loading Functions (GWLF), and EPA Screening Procedures. All of these methods are similar in concept and any would serve the purposes of this project. Similar to the models mentioned above, a spreadsheet-based hydrologic and mass loading model was created using empirical hydrologic and pollutant transport functions to support TMDL calculations.

B. Runoff and Groundwater Inputs

Hydrologic inputs to the lakes were empirically estimated using hydrologic data collected by the USFS in the Beaver Creek Watershed, located about 50 miles south of Flagstaff in Coconino and Yavapai counties. The USFS performed various hydrologic and water quality studies on this watershed between 1956 and 1982; primarily watershed management research within the Pinyon-Juniper and Ponderosa Pine vegetation of the Coconino National Forest. Hydrologic data from the Beaver Creek Watershed are useful for the LMR due to the proximity and similar geologic and vegetative characteristics (Baker, 1982).

For this study, the watershed runoff/groundwater inputs to the lakes were empirically estimated using monthly watershed yield coefficients derived from the Beaver Creek Watershed. In this report, a watershed yield coefficient is defined as the proportion of precipitation on a watershed that becomes stream flow at the mouth of the watershed. It represents all water falling on the watershed that does not evapotranspirate or infiltrate to deep groundwater. However, it includes groundwater that re-emerges as spring flow or base flow to a stream within the watershed. The average monthly watershed yields were determined by examining the relation between precipitation and stream flow in the Beaver Creek Watershed 20, where the USFS maintained a stream gauge between 1962 and 1983. Watershed 20 was selected because it was undisturbed and it is the largest watershed of all the Beaver Creek watersheds.

C. Solids in Runoff

The next component of the watershed input consists of the solids load from runoff. Soil erosion was estimated using the Revised Universal Soil Loss Equation (RUSLE), which was applied without calibration. This equation does not account for gully erosion and stream bank erosion because RUSLE calculates sheet and rill erosion only. The RUSLE equation for soil loss (NRCS, 1995) is given as:

$$A = R * K * (LS) * C * P$$

Where: A = estimated soil erosion
R = rainfall-runoff erosivity factor
K = soil erodibility factor
LS = length slope factor
C = cover and management factor
P = support practice factor

Because substantial trapping of sediment may occur during overland flow, the soil loss estimated above is not equivalent to sediment yield. The sediment yield was determined through a sediment delivery ratio (SD) and runoff transport capacity. The SD ratio is used to estimate the portion of the eroded soil that is delivered to the mouth of the watershed and is calculated based on watershed area (USDA-NRCS, 1998), as follows:

$SD = 0.42^{\text{area(sq mi)}} – 0.125$ if area < 200 acres
$SD = 0.417662 \text{ area (sq mi)}^{-0.134958} – 0.127097$ if area >/= 200 acres

The transport of sediment from the watershed to the lakes was based on the transport capacity of runoff during each month. This transport capacity was estimated using the formulation in the GWLF model.

Estimation of watershed sediment yield and watershed solids mercury loads were calculated within Microsoft Excel as follows:

- Monthly variability was captured in the rainfall-runoff erosivity factor (R). Each month, this factor was estimated as the product of the monthly runoff erosivity density and monthly precipitation. The monthly erosivity densities were obtained from monthly erosivity density surface maps given in the science documentation of RULSE Version 2 (Foster, 2005). The erosivity density is defined as the ratio of monthly erosivity to monthly precipitation amount.
- The product of the factors K, LS, C, and P factors (KLSCP) for each lake's watershed was estimated using data obtained from the USFS. In 2001, the USFS completed a Terrestrial Ecosystem Survey (TES) for the Coconino National Forest to map and evaluate the terrestrial ecosystems in the survey area, and determined current and potential soil erosion for the area. The USFS divided Coconino National Forest into 134 map units based on topography, geology, and vegetation and conducted field investigations to determine the soil type, erodibility, slope and slope length for each map unit. The USFS used the Universal Soil Loss Equation (USLE) to estimate the current soil loss for average rainfall conditions. Based on the USFS data, the product KLSCP was determined for each map unit. Using GIS, the map units that span each lake's watershed were identified. Using the KLSCP values for the map units identified within each lake's watershed, and their respective areas, an area-weighted average KLSCP value was estimated for each lake's

watershed. No seasonal or monthly variability was assumed for the KLSCP values.

- For each lake, the monthly soil erosion was estimated as the product of the monthly R factor and KLSCP composite value. The appropriate SD and a transport capacity based on average daily runoff were applied to the calculated erosion to obtain the sediment yield.
- Monthly sediment yields were multiplied by watershed soil concentration of total and methyl-mercury to obtain solids load for mercury. Soil concentrations of total mercury for the TMDL watersheds were based on the average of total mercury in runoff as suspended solids (200 ppb) cited in Table 7, rather than the estimated value of 120 ppb from the Gustavsson study (2001). Background mercury was derived by taking the average of deep core sample results yielding a pre-impoundment core value of 30 ng/g for the LMR lakes. The remaining 170 ng/g needed to be accounted for in the watershed load. Methyl-mercury soil concentrations were estimated using the ratio of total to methyl-mercury observed in sediments.

D. Lake Hydrologic Model

With the exception of Upper Lake Mary, which has some historical lake levels measured by the city of Flagstaff, lakes levels are not actively monitored. Therefore, a simple monthly water balance was used to represent the hydrology of the lakes (Malcolm Pirnie, 2005, with updates in 2006, 2007, and 2009). The details of the hydrologic balance are as follows:

- Inflow from the watershed was estimated by the watershed model described above in Sections A and B. Upper and Lower Lake Mary are connected such that water that overflows from Upper Lake Mary dam was included as inflow to Lower Lake Mary. For the Soldiers Lake Complex, the majority of the watershed runoff flows into Soldiers Lake; outflow from Soldiers Lake flows into both Soldiers Annex and Lower Long Lakes; outflow from Soldiers Annex Lake flows into Lower Long Lake or out of a spillway at the dam. The hydrology of the Soldiers Lake Complex has been described by Gremillion and Piastrini in Section III above (2005). The three lakes in the Soldiers Lake Complex are separated when the water level falls below an elevation of 6,778 feet.
- Direct precipitation was estimated from the following rain gauges: Happy Jack, Bear Seep, Coyote Park, Anderson Mesa, and Kinnikinick. The Anderson Mesa and Kinnikinick rain gauges only contain 2005 precipitation data. For Upper and Lower Lake Mary, monthly precipitation was obtained by averaging measured monthly values reported for Bear Seep, Coyote Park and Anderson Mesa. For the Solders Lake complex, monthly precipitation was obtained by averaging measured monthly values reported for Happy Jack and Kinnikinick.

- Evaporation from the lake surface was estimated from local pan evaporation for Flagstaff and a pan coefficient of 0.7 (WRCC, 2005).

Assumptions were made regarding the net loss of groundwater through the lake beds as follows:

- Groundwater seepage from Upper Lake Mary was estimated as a function of the lake water elevation, based on data reported by the USGS (Blee, 1988).
- For Lower Lake Mary, groundwater loss was also assumed to depend on elevation, but the rate of loss was obtained by calibrating the model to the few elevation data available. It was assumed that no overflows occurred during the simulation period.
- There is no information available for net groundwater loss through the lake bed of the Soldiers Lake Complex. In addition to the unknown groundwater losses, there are no accurate lake elevation data nor interconnecting flow data between the lakes. Given these unknowns the groundwater losses could not be estimated. Therefore, groundwater losses were assumed to be zero for the three lakes.

The water balance model calculated the water volume at the end of every month, using the initial volume, adding monthly inputs, minus evaporation and groundwater losses, and spilling any excess above spillway elevation in cases where spillage is allowed. The lake elevation, surface area, and volume were updated at the end of each month, using expressions developed from the results of the lake bathymetric surveys conducted for all the lakes in 2005 (Gremillion and Piastrini, 2005).

E. Mercury Cycling Model

A dynamic mercury cycling model was developed in Microsoft Excel in accordance with the conceptual diagram described in the previous section (Malcolm Pirnie, 2005 with updates in 2006, 2007, and 2009). Because of the significant fluctuations and lack of detailed data regarding lake volumes, the mass balance equations for each mercury species were written in terms of mass rather than concentrations. Therefore, the solution of the mercury mass balance was closely coupled with the solution of the water balance at each time step. The mercury transformations included: volatilization, methylation, reduction, oxidation, settling and others. These transformations were formulated as first order linear reactions as depicted in the conceptual model above, and the rate constants were obtained from Kotnik et al. (2002). The partitioning coefficients used were based on calculated values for the lakes and literature guidance from the EPA (Allison and Allison, 2005).

The mercury mass cycling model developed for this study used literature and published values of mercury transformation rates, and measured values for most parameters. The objective of the model application was to provide guidance in estimating whether management scenarios would provide low, moderate, or high degrees of benefit. Based on the objective of the model application, it was decided not to calibrate most parameters. Rather, only limited calibration was performed by adjusting the settling velocity.

The mercury model also required site specific data including:

- TSS and DOC concentrations in the water column, obtained from ADEQ lake water quality data.
- The settling velocity is the most sensitive parameter, and it was obtained through calibration. The model conducts a simple solids balance to determine re-suspension rates, using burial rates obtained from plutonium dating analysis of high resolution cores (Gremillion and Toney, 2005). Sediment bed porosity and sediment density were derived from (Gremillion and Toney, 2005), and literature values.
- Lake sediment concentrations were available from ADEQ data. Sediment concentrations were not simulated, but were held constant during the period of the model simulation. Pore water concentrations were not measured, but were derived by using literature values of sediment-water partitioning coefficient and measured sediment mercury concentrations.
- Concentration of gaseous elemental mercury in air was required to estimate volatilization fluxes. Literature guidance for the continental U.S. was used for all the lakes (Mason et al., 1994).

F. Mercury Bioaccumulation Estimation

The bioaccumulation of mercury in fish tissue depends on the food web dynamics, and ambient concentration and speciation of mercury. Bio-uptake models are used to simulate the linkage between mercury in water or sediment and mercury in fish or lower trophic level organisms. Site-specific BAFs were used to link model simulated water column concentrations to fish tissue concentrations (Section IX of this report).

VIII. TMDL CONSIDERATIONS

A. Numeric and Narrative Standards

The five LMR TMDL lakes have been monitored and assessed for all designated use parameters. Only mercury in fish tissue has exceeded its respective standard. Therefore, the numeric criteria of concern in this TMDL mercury in fish tissue and mercury in water. Table 10 shows numeric standards relevant to the LMR TMDL.

Table 10. Numeric Mercury Standards and Narrative Standards for the LMR TMDL Arizona Administrative Code Title 18, Chapter 11

NUMERIC Appendix A			
Designated Use	General Application	Acute Standard	Chronic Standard
Domestic Water Use	2.0 *u*g/L T-Hg		
Fish Consumption	3.0 mg/kg Methyl Hg		
Full Body Contact		280 *u*g/L T-Hg	
Agriculture Livestock Watering	10.0 *u*g/L T-Hg		
Aquatic & Wildlife (cold water)		2.4 *u*g/L D-Hg	0.01 *u*g/L D-Hg
Aquatic & Wildlife (ephemeral)	Tributaries only	5.0 *u*g/L D-Hg	
Partial Body Contact	Tributaries only	280 *u*g/L T-Hg	

T-Hg = Total Mercury; D-Hg = Dissolved Mercury; Me-Hg = Methyl-mercury

The part of the Narrative Standard relevant to this TMDL is R18-11-108(A)(5), which states:

> A surface water shall not contain pollutants in amounts or combinations that: are toxic to humans, animals, plants, or other organisms.

Only Upper and Lower Lake Mary and Mormon Lake carry the Domestic Water Use. The highest result for total mercury in watershed runoff was 0.014 *u*g/L from Babbit Spring Wash, tributary to Upper Lake Mary. The highest in-lake total mercury was 0.010 *u*g/L from Mormon Lake, 0.023 *u*g/L from Upper Lake Mary, and 0.030 *u*g/L from Lower Lake Mary. These values are two orders of magnitude lower than the DWS standard and are not considered a threat to drinking water use.

With regard to the dissolved mercury A&W (cold water) standards, no sample results exceeded the acute criterion, but one sample, collected from the upper portion of Upper Lake Mary on Sept. 8, 2004, measured 0.0185 *u*g/L and exceeded the chronic criterion. This sample result corresponds to 0.023 *u*g/L total mercury result cited above.

Since there is a linear relationship between fish tissue and water column mercury levels the targeted loads reductions will be calculated to meet the fish tissue standard. The Fish Consumption Advisories were issued between May 2002 and July 2003, prior to adoption of the current mercury fish tissue standard. The advisories and supporting data led EPA Region 9 to list these lakes as impaired for mercury in fish tissue.

B. Critical Conditions

The season of most intense but episodic sediment runoff is during monsoon rains from July through September, but these events are brief. By far, the greatest sediment load is delivered to the lakes during winter storms and spring snowmelt. The critical relationship appears to be the watershed to lake area ratio. The amount of lake surface area exposed to the wind may play a secondary role in keeping organic matter and sediments suspended. The highest direct aerial deposition to the lakes takes place during dry months. In modeling watershed and aerial loading, a monthly time step enabled the best fit annual estimation across all seasons.

C. TMDL Targets

Based on modeling results, the TMDLs for LMR lakes will reflect relative watershed and aerial load reductions needed to achieve the desired 0.3 mg/kg fish tissue target for methyl-mercury in fish tissue on a trophic-weighted basis.

IX. LINKAGE ANALYSIS

The linked models (as described in Section VII) were used to simulate current lake conditions as well as the implementation of scenario conditions. The major output results of interest under the existing conditions include predictions of watershed runoff and watershed mercury loading, lake water volume, and water column total and methyl-mercury. The model was applied with limited calibration and generalized parameter values obtained from the literature were used to characterize mercury transformation rates (Malcolm Pirnie, 2005 with updates in 2006, 2007, and 2009).

Although the majority of mercury entering lakes from the atmosphere and background sources are in the inorganic form, essentially all of the mercury accumulating in fish is methyl-mercury (Bloom, 1992). Therefore, understanding all the sources and conditions favoring the formation of methyl-mercury is important for management of mercury pollution. In general, sources of methyl-mercury to remote lakes include atmospheric deposition, watershed background load and in-lake methylation of inorganic mercury. The relative importance of these sources reportedly varies with rates of mercury deposition from the atmosphere, lake types and watershed hydrology (Rudd, 1995). In-lake inorganic mercury methylation occurs in the sediments (Ulrich et al., 2001), and for lakes that have anoxic hypolimnion, methylation occurs in the oxic/anoxic boundary in the water column (Eckley et al., 2005). The TMDL lakes modeled in this study did not show prominent anoxia or significant accumulation of methyl-mercury in bottom waters during the stratified period.

The fish tissue bioaccumulation analysis in this study focused exclusively on walleye because of the high observed mercury concentrations. As a trophic level four (TL-4) fish, the walleye represents the upper end of the food chain where the biomagnification of mercury would be the greatest. Walleye tissue was collected and analyzed for in only three lakes: Upper Lake Mary, Lower Long Lake and Soldiers Lake. A simple method for determining the BAF is the ratio of the tissue concentration to the water column concentration of mercury (usually dissolved methyl-mercury) in units of L kg^{-1}:

$BAF = C_T / C_W * 10^6$

Where:

C_T = MeHg concentration in the fish tissue, mg/kg
C_W= MeHg concentration in the water, ng/L

Table 11 presents a summary of the walleye tissue mercury concentrations, water column methyl-mercury concentrations, and the log-transformed values of the bioaccumulation factors (BAFs) derived in this manner.

Table 11. Average Tissue Concentrations, Water Column Methyl-mercury, and BAFs

Lake	Average Walleye Tissue Total Hg (mg/kg)	Average MeHg in Lake Water (ng/L)	Log (BAF, L/kg)
Lower Long Lake	0.71	0.70[1]	6.01
Upper Lake Mary	1.01	0.16	6.79
Lower Lake Mary	*1.01*	0.27	*6.57*
Soldiers Lake	1.65	0.12	7.14
Soldiers Annex Lake	*1.18*	*2.98*	*5.60*

1- excludes outlier of 16.2 ng/L from July 13, 2004; Italics indicate substituted/ interpolated data

ADEQ requested that Malcolm Pirnie (2009) revisit the simple BAF method in light of two other approaches used in previous mercury TMDLs: 1) Brumbaugh et al., 2001 regressions used in the Alamo Lake TMDL, and 2) the Spreadsheet-based Ecological Risk Assessment for the Fate of Mercury (SERAFM) model used in the Parker Canyon TMDL. Brumbaugh et al (2001) summarized data from across the United States and developed the following equation for length-normalized concentration of mercury for TL-4 fish as a function of methyl-mercury concentration in water:

$$\text{Ln} \; [\text{Hg-fish (mg/kg)/length(m)}] = 0.4923 * \text{Ln} \; [\text{MeHg}_{water} \text{ (ng/L)}] + 1.2189$$

Using this equation and the average of water column methyl-mercury concentration, the concentration of mercury in adult walleye was predicted for the various lakes. The predicted fish tissue mercury concentrations from the application of site-specific BAFs and the Brumbaugh et al (2001) equation, were compared to observed concentrations of adult walleye in the lakes (Table 12).

Table 12. Comparison of Observed and Model Simulation of Mercury Concentration under Current Loading Conditions for Average Adult Walleye

Lake	Mean Observed Fish Hg (Range) (mg/kg)	Model using Lake-specific BAF: Fish Hg (mg/kg)	Model using Brumbaugh et al. (2001) BAF: Fish Hg (mg/kg)	Model using ULM BAF: Fish Hg (mg/kg)
Lower Long Lake	0.71 (0.39 - 1.2)	0.19	0.79	1.12
Soldiers Lake	1.65 (1.1 - 2.7)	2.90	0.60	1.29
Upper Lake Mary	1.01 (0.6 - 1.6)	1.14	0.89	N/A

Excluding one outlier from Lower Long Lake, the lake-specific BAFs for Upper Lake Mary, Soldiers and Long, range from 1.0E+06 to 1.3E+07, consistent with the default value used in the SERAFM model (6.80E+0.6). The predicted BAF for Soldiers Lake is significantly higher that the other two lakes; given this anomaly, MP applied the site-specific BAF for Upper Lake Mary (6.3E+06 L/kg) to the Soldiers Lake Complex to predict walleye tissue concentrations.

X. MODELING RESULTS AND UNCERTAINTY

The linked watershed, hydrology, mercury cycling and bioaccumulation models described in the previous sections were used to simulate current lake conditions as well as the implementation of scenario conditions (Malcolm Pirnie, 2005, and updates from 2006, 2007, 2009). The major output results of interest under the existing conditions include predictions of the following:

1. Watershed runoff and watershed mercury loading;
2. Lake water volume; and
3. Water column total and methyl-mercury.

The major categories of uncertainty for this TMDL are:

- Limited tributary data and gauged flows
- Lack of lake elevation data except for Upper Lake Mary
- Limited number of fish collected
- Variability in application of RUSLE for deriving monthly erosivity density values
- Use of literature values where empirical data were missing
- Lack of data for dynamic in-lake mercury cycling model

Despite these inherent limitations, the TMDL is based defensibly on the level of information available: 1) the Forest Service rainfall/runoff research and TES data provided relevant empirical detail, 2) the modeling approach was kept simple to minimize amplification of uncertainty, and 3) the TMDL was supported by and incorporated additional studies (e.g., Gremillon et al – bathymetry and deep sediment cores; Tetra Tech – reevaluation of global and regional aerial deposition models and local MDN and Tekran data). Model results provide insight into several interesting characteristics of the TMDL Lakes and their watersheds:

A. Watershed Runoff and Sediment Yield

Application of the watershed model predicted an annual average runoff of approximately 10 cm for the Lake Mary Complex and approximately 7 cm for the Soldiers Lake Complex for the period 1996 to 2005. Model-simulated average annual sediment yields by sheet and rill erosion between 1996 and 2005 were: 1,101,000 kilograms, 921,000 kilograms, 1,065,000 kilograms, 144,000 kilograms and 303,000 kilograms for Upper Lake Mary, Lower Lake Mary, Soldiers Lake, Soldiers Annex, and Lower Long Lake, respectively.

Sediment yield may be episodically high during monsoon runoff. It was observed that average mercury concentrations in runoff in August and November were more than twice average values from January through April. However, overall, most of the annual water and sediment loading occurs during winter storms and spring runoff, setting up the potential for mercury methylation in the summer months when anoxic conditions may exist.

The annual sediments loads compare well to estimates made by the USFS under current vegetation and soil conditions. The temporal variability in sediment yield estimated in this analysis agrees with observations made in an undisturbed watershed from Beaver Creek, where sediment measurements indicated than at least 50 percent of the sediment is generated during the winter season. It should be noted that the RUSLE model (NRCS, 1995) was designed to predict long-term average annual soils erosion, and the monthly erosivity density values which attempt to express the erosion on a monthly basis are highly variable in Western states. In addition, the sediment erosion density formulation as implemented in RUSLE2 (Foster, 2005), the most recent version of the USLE family of models, is limited when snow cover is present for most of the winter months and doesn’t account for the impact of snowmelt for Western areas.

B. Lake Hydrology

For Upper Lake Mary, modeled lake volumes were compared to observations from the Flagstaff WTP. The model accurately simulated the temporal patterns observed for Upper Lake Mary, with increasing volumes during the snowmelt period and declining volumes afterwards (Figure 18). Overflows from Upper Lake Mary to Lower Lake Mary occurred only once in April 2005. Linear regression of the model-simulated volume versus observed volume had an R^2 of 0.74, indicating that the model captured 74 percent of the variability in Upper Lake Mary volume estimates. The other 26 percent can be attributed to errors such as year-to-year variations in evaporation and water yield coefficients, and to the fact that the precipitation measurements from the Bear Seep and

Coyote Park gauges may not be exactly representative of the average precipitation in Upper Lake Mary watershed.

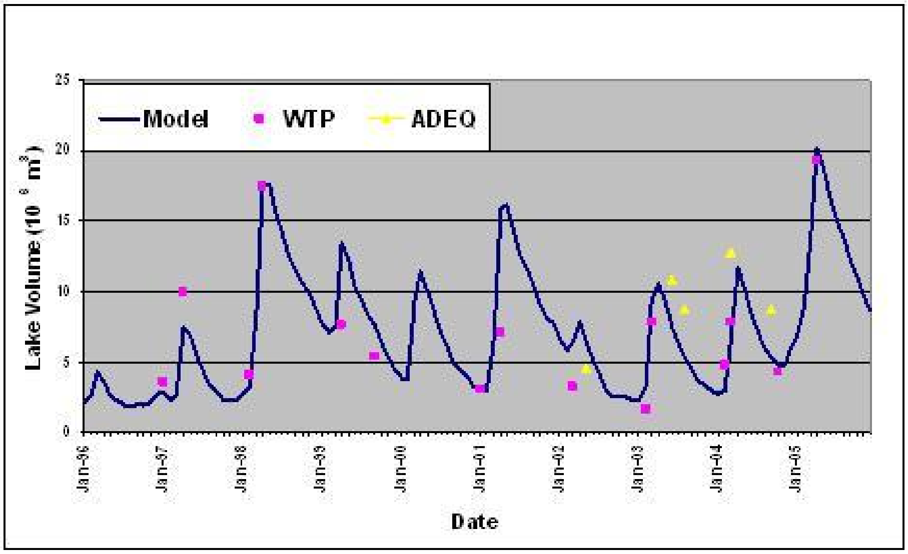

Figure 18. Model Simulated and Observed Hydrology for Upper Lake Mary

For Lower Lake Mary, lake volumes were calibrated against the few water depths obtained by the ADEQ during water quality monitoring. The model simulations indicate that Lower Lake Mary contains significant water volumes during the snowmelt period followed by almost dry conditions for the rest of the year (Figure 19). This result is consistent with observations made during the bathymetric surveys by Gremillion and Piastrini (2005), that typically the lake elevation does not exceed about 6,784 feet, corresponding to a volume of approximately 37,500 m^3, except during monsoon, rain on snow, or other exceptional wet events.

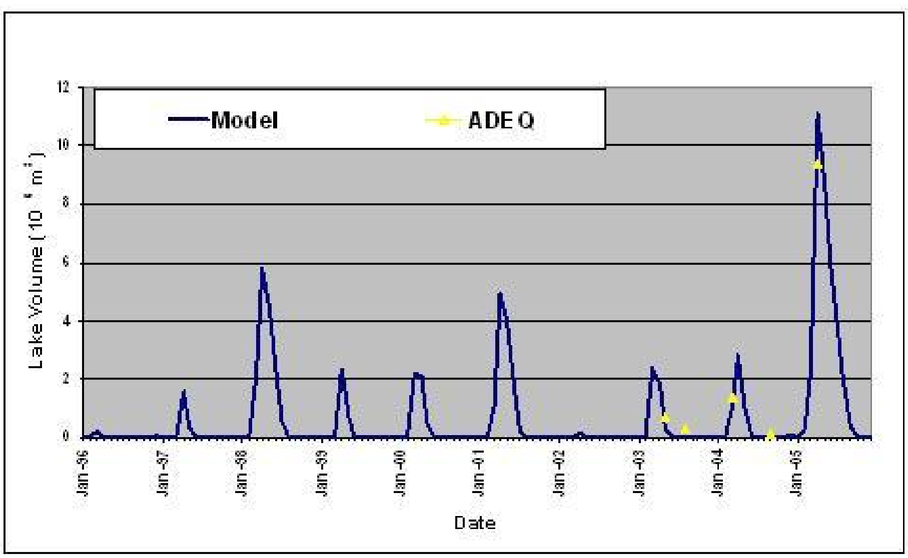

Figure 19. Model Simulated and Observed Lake Hydrology for Lower Lake Mary

For the Soldiers Lake Complex, an attempt to determine lake elevations or volumes from ADEQ's observed water depths was unsuccessful, as estimates were well above the lake spillways elevations. Therefore, model simulation of lake volumes for the Soldiers Complex are not well constrained given that groundwater loses and overflows from one lake to the next were also unknown. Given the lack of constraints, Soldiers Lake simulated lake volumes still showed similar temporal variation observed for Upper Lake Mary. Temporal variability in the model simulated lake water volumes was less pronounced in Soldiers Annex and Lower Long Lakes. Groundwater losses in the Soldiers Lake Complex were assumed to be zero.

XI. REVIEW OF MODEL REFINEMENTS

Between 2006 and 2009, refinements were made to the LMR model to reflect regionally acquired mercury air deposition and background soil data and the relationship of these data to watershed and aerial loading of mercury (Malcolm Pirnie 2006, 2007, 2009; Tetra Tech, Inc., 2008).

A. Mercury Loads

The external loads of mercury to the lakes were re-estimated using the revised atmospheric deposition flux and soil concentrations using new data as outlined above. The revised average annual loads for each lake are depicted below in Figures 20-24.

Based on refinements to aerial loading, model simulated watershed sediment deliveries to the various lakes were re-estimated. Some modifications on the timing of the delivery of the sediments to the lake were made. Results confirmed that increased sediment yield is closely associated with the large spring runoff events.

Mercury loading to Soldiers Annex and Lower Long Lake includes loading from Soldiers Lake, due to the hydrologic connectedness of the lake complex. Mercury from Soldiers Lake accounts for 65 percent of the total mercury loading to Soldiers Annex. Similarly 17 percent of the total mercury load to Lower Long Lake originates from Soldiers Lake and an additional 11 percent from Soldiers Annex.

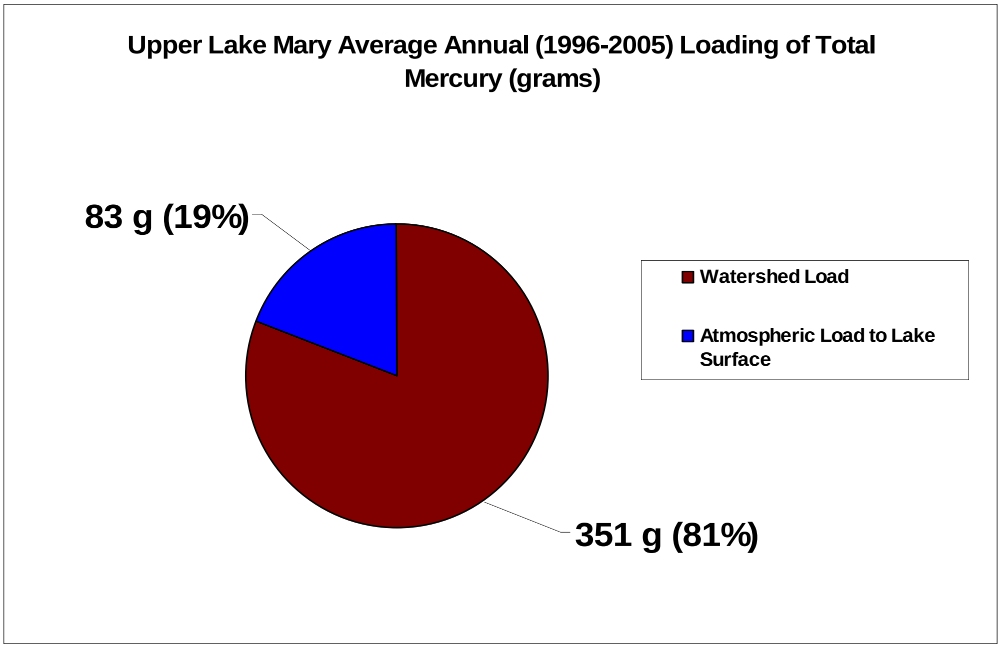

Figure 20. Upper Lake Mary Average Annual Total Mercury Load

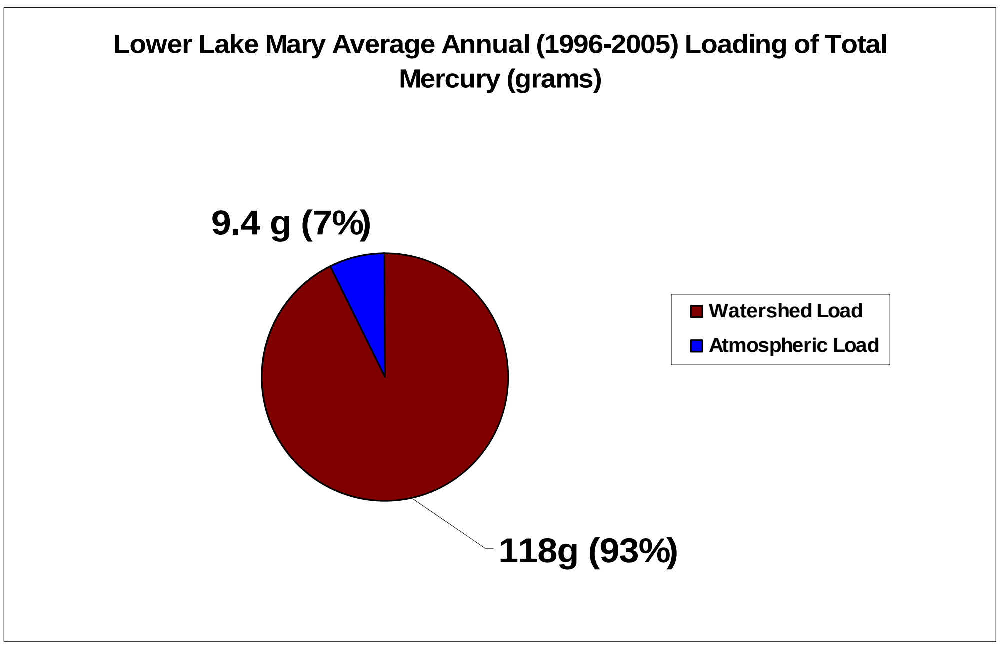

Figure 21. Lower Lake Mary Average Annual Total Mercury Load

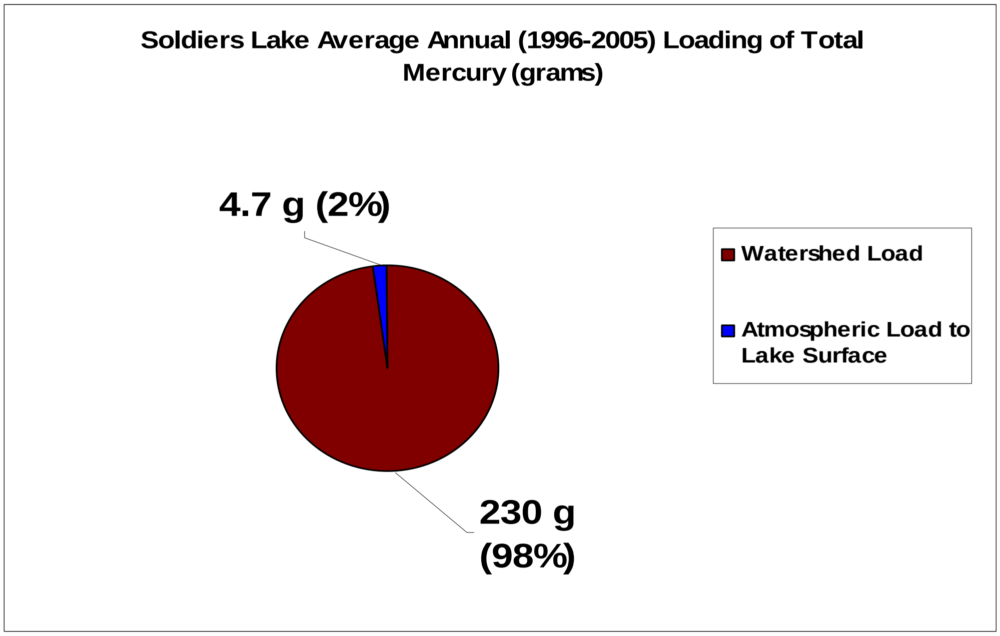

Figure 22. Soldiers Lake Average Annual Total Mercury Load

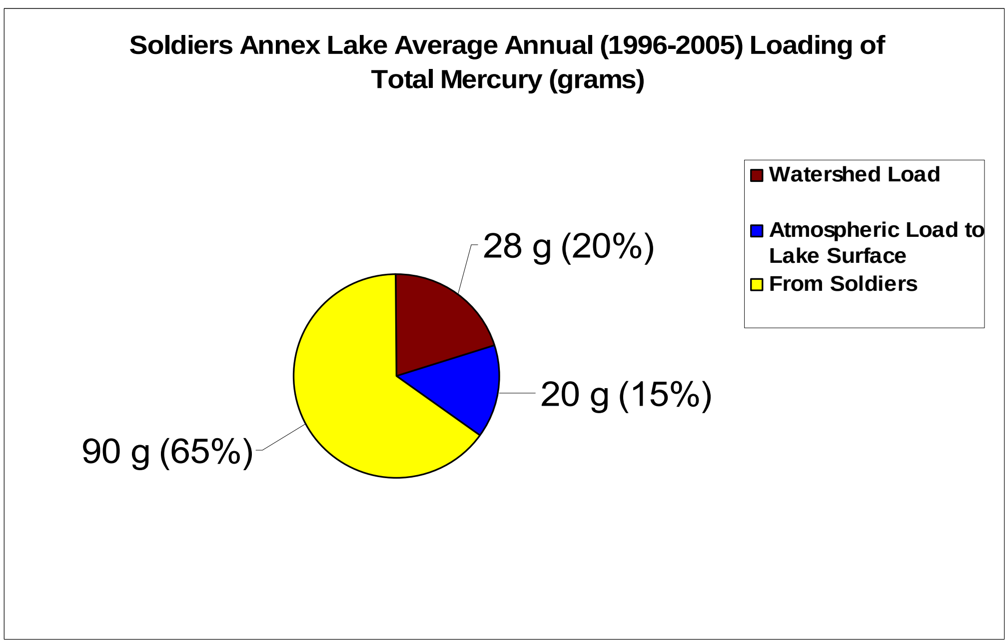

Figure 23. Soldiers Annex Lake Average Annual Total Mercury Load

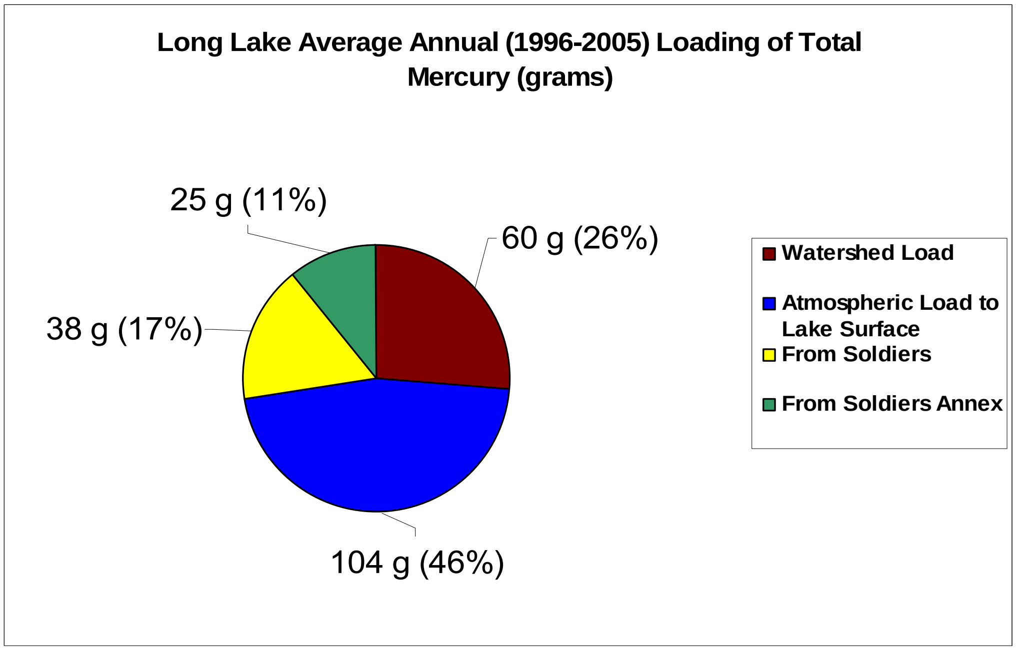

Figure 24. Lower Long Lake Average Annual Total Mercury Load

B. Model Simulated Lake Water Column Total and Methyl-mercury

The model was recalibrated by adjusting the settling velocity of particulate matter. Similar to previous observations; the concentrations of the mercury species reflect the temporal loading patterns from the watershed. For Upper Lake Mary, model predictions followed the observed increase in water column concentration resulting from the large runoff and sediment delivery events in spring. In the case of Soldiers Lake, the few observed data were well represented by the model, but there were no data points available to evaluate the peak concentrations predicted following the large runoff events. The lack of observed data during the peak mercury concentration period effects estimates of exposure concentrations used to estimate BAFs.

Overall, model simulation suggests that the major runoff events result in significant amounts of mercury delivered to these lakes from the watershed. In addition, these events also significantly increase the lake water levels, inundating previously exposed shoreline areas. This wetting of previously dry areas is likely to increase the sulfate and organic carbon concentrations in the lake, which is of importance in the production of methyl-mercury (Ullrich et al, 2001).

The lake-specific BAF performed better for Upper Lake Mary and Soldiers Lake compared to predictions based on the Brumbaugh et al (2001) empirical relationship. However, the reverse is true for Lower Long Lake. The use of Upper Lake Mary BAF for Soldiers and Lower Long Lakes improved model estimate for these lakes. This improvement in model predictions for Soldiers and Lower Long Lake suggests that the current measured data set are not representative of exposure concentrations of methyl-mercury in the water columns of the Soldiers Lake complex. Because a simple BAF was used to link water column methyl-mercury concentration to fish tissue concentration, model evaluations are based on the response of water column methyl-mercury concentrations between the existing and scenario conditions.

C. Model Forecasts of Fish Tissue Concentrations in Response to Decreases in Anthropogenic Mercury Loads

Model predictions of average mercury concentrations in adult walleye were made for various levels of anthropogenic input loads to the lakes. In these simulations, note that a zero percent anthropogenic mercury load reduction represent current conditions (deposition of 11 g/km^2/yr wet and 24 g/km^2/yr dry deposition, and mercury concentration of 200 ng/g in suspended sediment runoff) while 100 percent reduction represents a condition where soil mercury returns to background (30 ng/g) and long-range anthropogenic source contributions from the atmosphere is eliminated (wet and dry mercury deposition reduced by 95 percent of current values). The results are interpreted to indicate the following:

- Model forecast results show a linear response of simulated average adult walleye mercury concentrations to reductions in anthropogenic loads. The impact of the load reduction on fish tissue concentrations is dependent on the watershed area to lake area ratio as discussed in the modeling report. Soldiers Lake, which has a high watershed to lake area ratio showed the most rapid response to watershed load changes.
- There are significant differences in forecasted fish tissue concentrations based on the method used to relate fish tissue concentration to water column exposure concentration. In general, the Brumbaugh et al (2001) empirical relationship showed the slowest response to anthropogenic load reduction for each lake.
- Reductions in watershed mercury loads will result in significant reductions in fish tissue mercury concentrations.

D. Modeling Summary

In the LMR, the majority of runoff to the lakes occurs during the snowmelt period in March/April, although significant sediment can be delivered during brief monsoon storms. External sources to the lakes constitute direct atmospheric and watershed sources. Watershed sources include geologic, historic and ongoing atmospheric deposition.

Watershed loading is tied to soil erosion and transport of sediments. The majority of the load was estimated to occur in the winter/spring snowmelt, though in August/September short-term precipitation rates may generate large sediment pulses. The relative contribution of the direct atmospheric deposition and the watershed loads is dependent on the watershed area to lake area ratio.

The calibrated lake hydrology model is in good agreement with the observed lake volumes for Upper and Lower Lake Mary. Lower Lake Mary was simulated as usually dry following the snowmelt period making hydrologic simulations impossible without detailed lake volume data. The calibrated mercury model is generally in good agreement with the observed water column concentration in the lakes. The water column dynamics of mercury are controlled by the external inputs to the lakes.

Modeling was conducted on a monthly time-step, but loads will be presented as both annual average and daily average. These loads have been calculated to reflect the percent reduction in total mercury delivered to the lake needed to achieve the fish tissue target of 0.3 mg/kg methyl-mercury based on the relationship in Figure 25 and Table 13.

As stated, the impact of the load reduction is dependent on the watershed area to lake area ratio. Complete elimination of anthropogenic watershed mercury loads would result in approximately 67 percent, 75 percent, 37 percent, and 11 percent reduction in water column methyl-mercury concentrations in Upper Lake Mary, Soldiers Lake, Soldiers Annex Lake, and Lower Long Lake, respectively. Due to its intermittent nature Lower Lake Mary could not be included in the predictions directly. The Lower Lake Mary load reductions will be interpolated using Upper Lake Mary relationships.

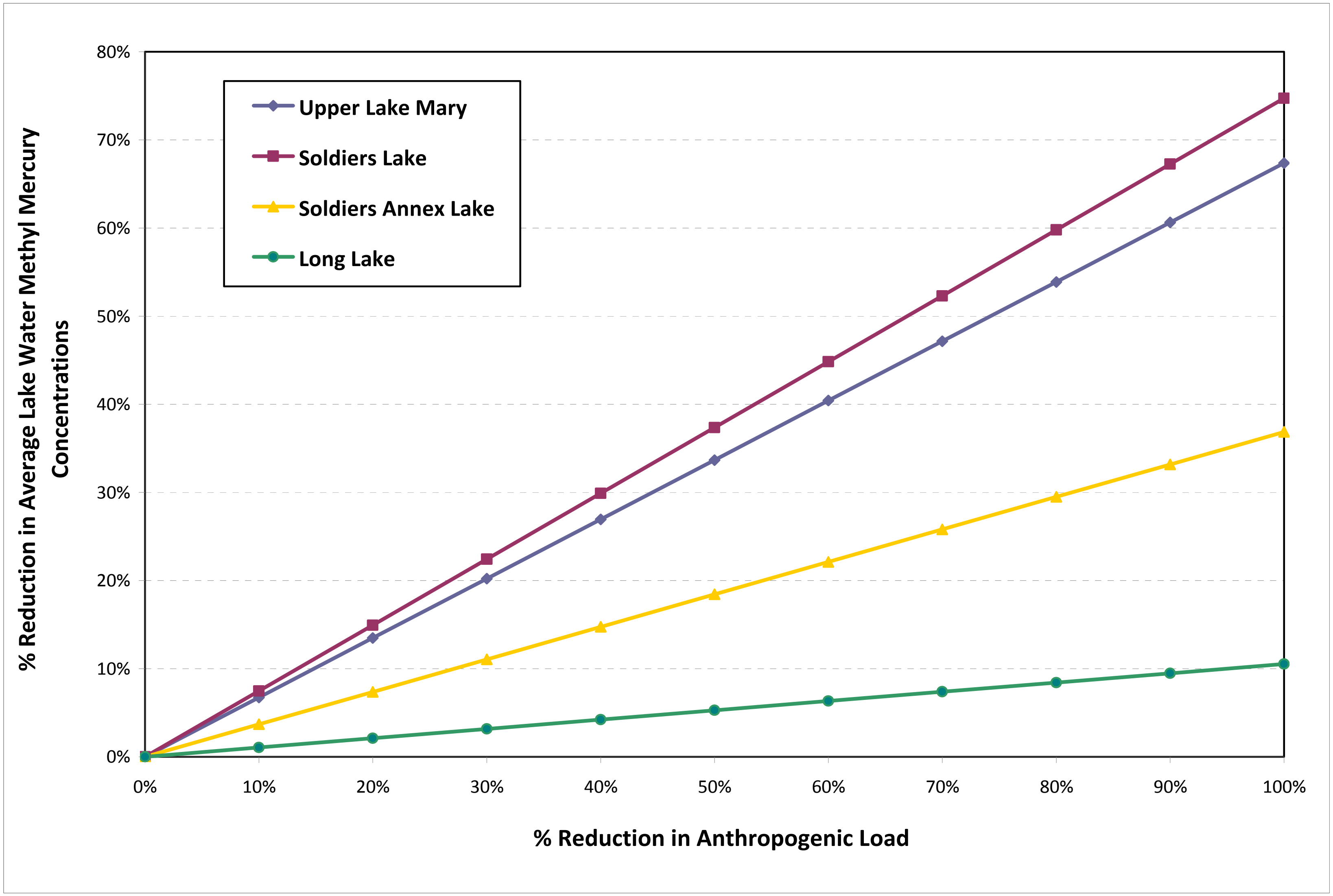

Figure 25. Response of Long-Term Average Water Column Methyl-mercury Concentration to Reductions in Anthropogenic Loads

Table 13. Long-Term Average Water Column Methyl-Mercury Concentration to Watershed Load Reduction

% Anthropogenic Reduction	Upper Lake Mary			Lower Lake Mary			Soldiers Lake			Soldiers Annex			Lower Long Lake		
	Total Hg load grams/yr	% Red Total	% Red MeHg	Total Hg load grams/yr	% Red Total	% Red MeHg	Total Hg load grams/yr	% Red Total	% Red MeHg	Total Hg load grams/yr	% Red Total	% Red MeHg	Total Hg load grams/yr	% Red Total	% Red MeHg
0[1]	434	0%	0%	127	0%	0%	235	0%	0%	138	0%	0%	227	0%	0%
10	395	9%	7%	116	9%	7%	214	9%	7%	127	8%	4%	207	9%	1%
20	360	17%	13%	105	17%	13%	195	17%	15%	115	17%	7%	186	18%	2%
30	321	26%	20%	94	26%	20%	174	26%	22%	104	25%	11%	168	26%	3%
40	282	35%	27%	83	35%	27%	155	34%	30%	91	34%	15%	148	35%	4%
50	234	44%	34%	69	44%	34%	134	43%	37%	80	42%	18%	127	44%	5%
60	208	52%	40%	61	52%	40%	115	51%	45%	68	51%	22%	107	53%	6%
70	169	61%	47%	50	61%	47%	94	60%	52%	57	59%	26%	86	62%	7%
80	130	70%	54%	38	70%	54%	75	68%	60%	44	68%	29%	68	70%	8%
90	96	78%	61%	28	78%	61%	54	77%	67%	33	76%	33%	48	79%	9%
100[2]	56	87%	67%	17	87%	67%	35	85%	75%	21	85%	37%	27	88%	11%

1- Current Conditions.
2- Natural Baseline Condition = Complete reduction of mercury from anthropogenic atmospheric deposition (i.e. reduce atmospheric deposition to 5% of its current value), and watershed soil reduced to 30 ng/g.

XII. TMDL CALCULATIONS

This section provides the TMDL calculations based on the relationships established by modeling results summarized in Table 13 and the application of trophic-weighted fish tissue concentrations. A TMDL is the maximum allowable daily load that a waterbody can assimilate and still meet water quality, or in this case, fish tissue standards. TMDLs include load allocations (LA) for nonpoint sources, waste load allocations (WLA) for point sources, a margin of safety (MOS) and natural background (NB). TMDL calculations will follow the equation:

$$TMDL = LA + WLA + NB + MOS$$

TMDL calculations have been made using the modeling results for Upper Lake Mary and Soldiers Lake only. The rationale for this is as follows:

- Both of these lakes provide water and fish to the lower lakes within their respective systems so reductions to mercury loading would be realized throughout each lake complex;
- The majority of fish tissue data have been collected from these two lakes; and
- They have the most stable hydrology of all the LMR lakes.

A. Fish Tissue Criterion and Trophic Considerations

ADEQ formally adopted the 0.3 mg/kg fish tissue criterion in the Arizona Surface Water Quality Standards in January 2009. Modeling performed by Malcolm Pirnie took a conservative approach in using the average walleye (highest TL-4 species) tissue concentration to derive target reductions necessary for achieving the 0.3 mg/kg standard. However, as discussed previously, there are many factors affecting mercury bioaccumulation, including sulfate, sulfur-reducing bacteria, DOC, redox potential, and the specific structure and dynamics of a particular trophic system. Fish may fall into one TL category part of the year, or for part of its lifespan, and another category as they age. A juvenile TL-3 or TL-4 can slide down a level; similarly, a very large predator in TL-3 can slide up a level.

EPA cites the need to consider the trophic structure in setting TMDL reduction goals (EPA, 2009). Through application of TL-weighted geometric mean analysis, TMDL reductions reflect more realistic goals that will ensure that the fishery as a whole will meet the tissue criterion. Using the same default TL consumption rates as the fish tissue criterion (EPA, 2001), ADEQ derived reduction goals for LMR by calculating the geometric mean mercury concentration for all species (C_{avg}) within each lake complex and comparing it to the fish tissue standard. The average lake fish tissue concentration was calculated using:

$$C_{avg} = \frac{3.8 * C_2 + 8.0 * C_3 + 5.7 * C_4}{(3.8+8.0+5.7)}$$

Where:

C_2 = average weighted geometric mean mercury concentration for TL 2
C_3 = average weighted geometric mean mercury concentration for TL 3
C_4 = average weighted geometric mean mercury concentration for TL 4

Fish tissue data were aggregated between the Upper and Lower Lake Mary (Lake Mary complex), and Soldiers, Soldiers Annex and Lower Long Lakes (Soldiers complex). The calculation apportions the 17.5 g fish/day national default consumption rate into: 5.7 g/day of TL-4 fish, 8.0 g/day of TL-3 fish, and 3.8 g/day of TL-2 fish. Table 14 summarizes fish tissue data and shows TL-weighted geometric means for both lake complexes.

Table 14. TL-weighted Geometric Mean Mercury Concentrations

Lake Mary Complex						
	TL-4	TL-4	TL-3	TL-3	TL-3	TL-2
Fish Species	Walleye	Northern Pike	Yellow Bass	Channel Catfish	Crappie	none
# of samples	9	7	10	2	3	0
Species geometric mean Hg	1.01	0.6	0.13	0.18	0.13	NA
Weighted TL Hg geometric mean	0.80		0.13			NA

Soldiers Lake Complex						
	TL-4	TL-4	TL-4	TL-3	TL-3	TL-2
Fish Species	Walleye	Northern Pike	Largemouth Bass	Channel Catfish	Bluegill	Rainbow Trout
# of samples	17	7	1	2	2	6
Species geometric mean Hg	1.26	0.52	0.36	0.42	0.45	0.08
Weighted TL Hg geometric mean	0.87			0.43		0.09

B. Load Reductions

Applying these weighted TL geometric mean values to the formula cited above, yields an average fish tissue concentration of 0.40 mg/kg for the Lake Mary Complex and 0.50 mg/kg for the Soldiers Complex. Approaching TMDL reductions from this weighted approach normalizes the consumption risk, but also normalizes the degree of impairment. The reductions necessary will be the difference between the TL-weighted geometric means and the fish tissue standard of 0.3 mg/kg.

For Lake Mary: 0.3/0.4 = 0.75; need 25 percent reduction in methyl-mercury

For Soldiers: 0.3/0.5 = 0.6; need 40 percent reduction in methyl-mercury

Interpolating from Table 13, the methyl-mercury reductions equate to total mercury reductions of 32 percent for the Lake Mary complex and 46 percent for the Soldiers Lake complex. Based on a geologic natural background value of 30 ng/g total mercury, Table 15 shows that the natural background load currently equals 13 percent for the Lake Mary complex (NB load of 56 g/yr divided by current total load od 434 g/yr = 0.129, or 13%) and 15 percent for the Soldiers Lake complex (NB load of 35 g./yr divided by current total load of 235 g/yr = 0.149, or 15%). Based on the respective TMDLs, the corresponding natural background load will be 19% for the Lake Mary Complex, and 28% for the Soldiers Complex.

C. Margin of Safety

The BAF applied to all lakes was derived from the tissue results of the top predatory fish in the system, the walleye. Use of this BAF represents an implicit margin of safety because meeting 0.3 mg/kg in walleye should guarantee attainment of that target in lower trophic level fish. An explicit MOS equal to 10 percent of the TMDL value is also included in the TMDL calculations (Table 15).

Table 15. Reductions Needed by Lake Complex

Lake Mary Complex								
% MeHg reduction	% total Hg reduction	Current total Hg load (g/yr)	TMDL (g/yr)	WLA (g/yr)	LA (g/yr)	NB (g/yr)	MOS (g/yr)	TMDL (g/day)
25%	32%	434	295	0	209	56	30	0.80
Soldiers Complex								
% MeHg reduction	% total Hg reduction	Current total Hg load (g/yr)	TMDL (g/yr)	WLA (g/yr)	LA (g/yr)	NB (g/yr)	MOS (g/yr)	TMDL (g/day)
40%	46%	235	127	0	79	35	13	0.36

XIII. CONCLUSION

There are no known local watershed point sources or aerial point sources of mercury currently operating within the LMR. Past emissions from sawmills in Flagstaff and Clark Valley, in which Lake Mary is located, may have contributed mercury through aerial deposition between the 1880s and 1970s but these loads were not quantified. Additional contributions may have been made by smelters and cement plants within Arizona. Modeling estimates show approximately 95 percent of mercury in the region is from global sources. The remaining 5 percent is attributed to a combination of regional aerial sources from California, Mexico, and Arizona, including natural geological background based on REMSAD modeling results.

Most of the mercury that enters the lake comes from surface water runoff, particularly bound to clay sediments. While some mercury is lost to settling, a significant portion appears to remain suspended in the water column where sulfur-reducing bacteria mediate the transformation to methyl-mercury. Fish are exposed to methyl-mercury both directly and indirectly from eating prey containing methyl-mercury.

Analysis of the trophic distribution of fish tissue concentration demonstrates that, overall, both lake complexes are not meeting the 0.3 mg/kg fish tissue target for mercury. The level of reduction necessary to reach this target appears dependent upon which species are prominent and their trophic status.

Load reductions necessary to meet the fish tissue criterion have been established through modeling and empirical evidence. There are no known point sources of mercury in the LMR watershed so the WLA in all calculations is equal to zero. A MOS is implicitly contained within the conservative BAF used in modeling and explicitly by allocating 10% of the TMDL to MOS. The final TMDLs for each lake complex are shown below:

Lake Mary Complex:

TMDL = WLA(0) + LA(0.57 g/day) + NB(0.15 g/day) + MOS(0.08 g/day) = 0.80 g/day

Soldiers Lake Complex:

TMDL = WLA(0) + LA (0.22 g/day) + NB(0.10 g/day) + MOS(0.04 g/day) = 0.36 g/day

XIV. TMDL IMPLEMENTATION

Regardless of the initial source of mercury, watershed loading can potentially be reduced through management of sedimentation and vegetative stability. Implementation of this TMDL should include a review of upland and drainage stability, so that areas needing soil stabilization and channel improvements may be identified.

TMDL implementation plans are required by A.R.S 49-234, paragraphs G, H, & J requiring TMDL implementation plans to be written for those navigable waters listed as impaired and for which a TMDL has been completed pursuant to Section 303(d) of the Clean Water Act. Implementation plans provide a strategy that explains "how the allocations in the TMDL and any reductions in existing pollutant loadings will be achieved and the time frame in which compliance with applicable surface quality standards is expected to be achieved." Due to the nonpoint source of pollutants within the LMR, the voluntary implementation of this plan lies on the responsibilities of stakeholders to achieve necessary load reductions to maintain water quality standards within the described reach.

Congress amended the Clean Water Act in 1987 to establish the Section 319 Nonpoint Source Management Program. As a result of this federal guidance, states have an improved partnership in their efforts to reduce nonpoint source pollution. The ADEQ Water Quality Improvement Grant Program allocates 319 grant funds from the EPA to interested parties for implementation of nonpoint source management and watershed protection. Under Section 319, state, private/public entities, and Indian tribes receive grant money which support restoration projects to implement on-the-ground water quality improvement projects to control nonpoint source pollution.

When a grantee applies for 319 funding, a watershed based plan or implementation plan submitted with the proposal demonstrates that the project has been carefully planned, reveals technical-economic feasibility, and illustrates the milestones that need to be implemented within a clear timeline. Watershed-based plans, such as TMDL implementation plans, help 319 proposals gain the highest priority for funding.

Watershed-based or implementation plans define nine essential elements to help provide reasonable assurance to EPA, stakeholders, and the state of Arizona that load allocations identified in the TMDL will be achieved, waterbodies that have a completed TMDL and watershed-based plan or implementation plan receive high priority for 319 grant funds. These nine essential elements clearly define: causes and sources of pollutant(s), an estimate of load reductions, management measures that will need to be implemented, an estimate of technical and financial assistance needed, an information and education component, reasonable schedule of implementation, measurable milestones and events to determine if whether the management measures are being implemented, a set of criteria to evaluate pollutant reduction, as well as, a set of methods to monitor project effectiveness.

Stakeholder input is requested to promote collaboration and acceptance of the strategies proposed in this TMDL implementation plan. After the plan is adopted through a public participation process, ADEQ is required to revisit and review the TMDL every five years to determine if the TMDL implementation plan was successful. ADEQ will develop a separate LMR implementation plan document in collaboration with local stakeholders within six months of TMDL approval. Likely projects or recommendations will include reducing sediment entering the lakes and changes to fisheries and lake management. The plan will also establish effectiveness monitoring of methyl-mercury and total mercury in water, sediment and fish tissue within the lakes and total mercury in water and sediment on tributaries identified for erosion control.

XV. PUBLIC PARTICIPATION

Stakeholder and public participation for the Lake Mary Regional TMDL has been encouraged and received throughout the development of the TMDL. ADEQ has extended a request for input from the watershed groups, local residents, governmental agencies, and other interested parties related to their opinions and suggestions regarding the TMDL study and findings, current and future implementation plans, model selection and use, data collection, and the level of involvement that they might contribute to the decision making process.

In addition to informal meetings in the field with stakeholders, three formal public meetings were conducted during the LMR project. The public meetings were arranged with the assistance of the local stakeholders and watershed partners. The first was held on Sept. 29, 2005 at the Coconino County Board of Supervisors meeting room in Flagstaff followed by another meeting on Dec. 15, 2005. Discussions at these meetings included the introduction of the TMDL process to the attendees, a reporting on the ADEQ preliminary investigation and the modeling status at that time. Notice regarding guidance available to parties interested in pursuing development of other remediation projects, as well as the availability of federal (319) grants for that purpose, was provided. A question and answer period followed. The third meeting at the ADEQ Northern Regional Office occurred on September 9, 2008. The draft TMDL modeling report and the associated results were the main topics of discussion.

The draft TMDL report was made available for a 30-day public comment period from June 23, 2010 to July 23, 2010. Public notice of the availability of the draft document was made via a posting in a newspaper of general circulation Arizona Daily Sun; via email notifications, phone calls; and webpage postings.

Responses to questions and comments received during the 30-day public comment period were addressed in a Notice of Public Information submitted to the Arizona Administrative Register on September 17, 2010.

REFERENCES

Allison, J.D. and Allison, T.L. (2005). Partition Coefficients for Metals in Surface Water, Soil, and Waste. EPA/600/R-05/074.

Baker, M.B. (1982). Hydrologic Regimes of Forested Areas in the Beaver Creek Watershed, U.S. Dept. of Agriculture (http://ag.arizona.edu/OALS/watershed/beaver/index.html)

Beck, H.L. and Bennett, B.G., (2002). Historical Overview of Atmospheric Nuclear Weapons Testing and Estimates of Fallout in the Continental United States, *Health Phys*. 82(5):591-608.

Blee, J.W.H., (1988). Determination of Evaporation and Seepage Losses, Upper Lake Mary near Flagstaff, Arizona. U.S. Geological Survey Water-Resources Investigations Report 87-4250.

Bloom, N.S. (1992). On the Chemical Form of Mercury in Edible Fish and Marine Invertebrate Tissue. Canadian Journal of Fisheries and Aquatic Science 49: 1010–1017.

Brumbaugh, W.G., Krabbenhoft, D.P., Helsel, D.R. Wiener, J.G., Echols, K.R., (2001). A national pilot study of mercury contamination of aquatic ecosystems along multiple gradients: bioaccumulation in fish, USGS/BRD/BSR-2001-0009, iii+25pp.

Cline Library Special Collections and Archives Department (Viewed 2010). Northern Arizona University. The Cline Library Digital Archives

Dvonch, J. T., Graney , J. R., Keeler , G. J., and Stevens, R. K. (1999). Use of Elemental Tracers to Source Apportion Mercury in South Florida Precipitation. Environmental Science & Technology 33(24):4522-4527

Eckley, C.S., Watras, C.J., Hintelmann, H., Morrison, K., Kent, A.D., and Regnell, O. (2005). Mercury Methylation in the Hypolimnetic Waters of Lakes with and without connection to Wetlands in Northern Wisconsin. Canadian Journal of Fisheries and Aquatic Science, 62: 400-411

EPA (2009). Guidance for Implementing the January 2001 Methyl-mercury Water Quality Criterion. EPA 823-R-09-002

EPA (2005). Technical Support Document for the Final Clear Air Mercury Rule. Air Quality Modeling. Research Triangle Park, NC.

EPA (2001). Water Quality Criterion for the Protection of Human Health: Methylmercury. Office of Water, Washington D.C. EPA-823-R-01-001.

EPA (1997). Mercury Study Report to Congress. Volume 111: Fate and Transport of Mercury in the Environment. EPA-452/R-97-005

Foster, G.R. (2005). Revised Universal Soil Loss Equation Version 2. Draft Science Documentation. Prepared for USDA-Agricultural Research Service, Washington, D.C.

Gilmour, C.C., Krabbenhoft, D.P., Orem W, Aiken G. (2004). The influence of drying and rewetting on Hg and S cycling in Everglades soils. Presented at the Seventh International Conference on Mercury as a Global Pollutant, Ljubljana, Slovenia, June 27-July 2, 2004.

Gremillion, P.T and Toney, J. L. (2005). Metals Deposition in Northern Arizona Reservoirs. Draft Final Report prepared for ADEQ.

Gremillion, P.T and Piastrini, C. (2005). Bathymetric Study of Northern Arizona Lakes. Draft Final Report prepared for ADEQ.

Gustavsson, Bolviken, N., B., Smith, D.B., and Severson, R.C. (2001). Geochemical landscapes of the conterminous United States – New map presentations for 22 elements. USGS Professional Paper 1648. USGS. Denver, Co.: US Geological Survey.

Koval, D.B., (1976). Geophysical-structural analysis of the Lake Mary well field area with hydrologic interpretations. Northern Arizona University, M.S. thesis, 123 p.

Kotnik, J., Horvat, M., and Jereb, V. (2002). Modeling of mercury geochemical cycle in Lake Velenje, Slovenia. Environmental Modeling & Software 17: 593–611.

Malcolm Pirnie. (2005). Lake Mary Region Total Maximum Daily Load (TMDL) Data Summary Report.

Malcolm Pirnie. (2006). Model Development Report, Lake Mary Region, Mercury Total Maximum Daily Load (TMDL).

Malcolm Pirnie. (2007). Model Scenario for 30 ppb Background Concentration. Technical Memorandum dated May 14, 2007.

Malcolm Pirnie. (2009). Model Simulation of the Impact of Reduction in Anthropogenic Mercury Inputs on Average Adult Walleye Mercury Concentration in the Lake Mary Region, Technical Memorandum dated February 25, 2009.

Mason, R.P., Fitzgerald , W.F., and Morel, F.M.M. (1994). The biogeochemical cycling of elemental mercury: Anthropogenic Influences. Geochimica et Cosmochimica Act. 58(15): 3191-1918.

National Resource Conservation Service (NRCS) (1995). Revised Universal Soil Loss Equation (RUSLE) Version 1.04 User Guide.

Northern Arizona University article. 2010. *Land Use History of North America, Mining on the Colorado Plateau*. http://cpluhna.nau.edu/Change/mining.htm

Odem, W.I. (2002). Unpublished Bathymetric Map Data for Lower Lake Mary. NAU.

Ricken, Jack (2005). City of Flagstaff Water treatment Plant. Personal Comminication.

Rudd, J.W.M. (1995). Sources of Methylmercury to Aquatic Ecosystems: A Review. Water Air Soil Pollution, 80: 697–713.

Sustaining our Forest: A Photo Essay of the Cumulative Effects of 50 Years of Stand Replacement Wildfire. 2003. Viewed 2010 on the Greater Flagstaff Forests Partnership (GFFP) web site: http://www.gffp.org/pine/p_essay/p_essay.htm, Program in Community Culture and Environment, Northern Arizona University.

National Interagency Fire Center. 2002. Wildland Fire Statistics. 4/10/03. Viewed 2010. Statistics and graphs on: Total fires and Acres by Year (1960-2002); Average Number of Fires and Acres Burned By Decade; Number of Wildland Fires and Acres By Cause; Suppression Costs for Federal Agencies: http://www.nifc.gov/stats/wildlandfirestats.html

Swetnam, Thomas W. and Julio L.Betancourt. 1997. Mesoscale Ecological Responses to Climatic Variability in the American Southwest. In 2003 on-line article with statistics on forest structure changes and chages in fire frequency and severity: http://geochange.er.usgs.gov/sw/impacts/biology/fires_SOI/

Tetra Tech, Inc. (2008). Memorandum to USEPA Region 9: Arizona Mercury Air Deposition Data Analysis.

Ullrich, S.M., Tanton, T.W., and Abdrashitova, S.A. (2001). Mercury in the Aquatic Environment: A Review of Factors Affecting Methylation. Critical Reviews in Environmental. Science and Technology, 31: 241–193.

USDA-NFS. (2001). Terrestrial Ecosystem Survey of the Coconino National Forest. (http://alic.arid.arizona.edu/tes/tes.html)

USDA-NRCS. (1998). Field Office Technical Guide Notice No. IA-198. “Erosion and Sediment Delivery Procedure”, Section I, Erosion Protection.

USGS. (2003). Geologic Studies of Mercury by the U.S. Geological Survey. U.S. Geological Survey Circular 1248.

USGS. (2009). Mercury Cycling in Stream Ecosystems: 1) Water Column Chemistry and Transport; 2) Benthic Methylmercury Production and Bed Sediment – Pore Water Partitioning; 3) Trophic Dynamics and Methylmercury Bioaccumulation

Western Region Climate Center web site (WRRC) . (2005). http://www.wrcc.dri.edu/htmlfiles/westevap.final.html

Yudovich, Y.E., and Ketris, M.P. (2005). Mercury in coal: a review Part 2. Coal use and environmental problems. International J. of Coal Geology, Article in press, preprint.