

1 **Factors Influencing the Bioaccumulation of Methylmercury in**
2 **Largemouth Bass from California Lakes and Reservoirs**

3
4 Melwani, A.R., Negrey, J., Stephenson, M., Solomon, M.,
5 Bezalel, S.N., Allen, R., and Davis, J.A.
6

7 **Abstract**

8
9 The objective of this study was to develop a statistical model that describes the
10 driving factors of fish methylmercury (MeHg) bioaccumulation in California lakes and
11 reservoirs. MeHg concentrations in largemouth bass from 17 lakes were examined for
12 relationships to water and sediment chemistry, lake morphometry, and land use. A
13 combination of correlation and multivariate analysis was used to evaluate the controlling
14 factors. Average MeHg concentrations in length-standardized largemouth bass varied
15 from 0.06 – 1.3 µg/g at the 17 lakes included in the analysis. Several predictor variables
16 were significantly correlated with length-standardized largemouth bass MeHg
17 (Pearson $r > 0.5$, $p < 0.05$) and were a significant component of the multivariate
18 regression model. The driving factors included total Hg in sediment, total Hg in soils,
19 forested area, specific conductivity, and methylmercury in surface water. Results of the
20 statistical model were consistent with several studies from across the nation that have
21 suggested that broad land use characteristics can potentially influence MeHg
22 bioaccumulation in lakes and reservoirs. However, the limited sample size and spatial
23 distribution of lakes evaluated here preclude extending our interpretations beyond the
24 population of lakes included in this analysis.
25

26 **Introduction**

27
28 Mercury (Hg) is a widespread pollutant that has impacted aquatic ecosystems for
29 more than a century. Hg sources in California include historic mercury, gold, and silver
30 mining areas, wastewater, urban runoff, agricultural runoff, and atmospheric deposition.
31 The most significant Hg source by mass is attributed to mining activity during the 1800s,
32 when significant releases to the environment occurred (Domagalski 1998). On a national
33 scale, atmospheric deposition is considered the predominant source of Hg to aquatic
34 environments distant from mines (U.S. EPA 1997). Hammerschmidt and Fitzgerald
35 (2006) recently suggested that up to two-thirds of the MeHg accumulation in fish from
36 water bodies in the United States could be attributed to wet deposition of atmospheric
37 Hg. Hg deposition to the eastern United States has been indicated in many studies as the
38 primary vector for Hg accumulation in the food web (e.g., Fitzgerald and Clarkson 1991;
39 Hammerschmidt and Fitzgerald, 2006). Despite limited data, it has been estimated that
40 deposition to the west coast (California, Washington, Oregon) remains at background
41 levels. In California, runoff and weathering from historic gold and mercury mining areas
42 has mobilized legacy Hg from the landscape into many of the lakes and reservoirs. The
43 legacy contamination is considered the most likely source of high background
44 concentrations of Hg in water bodies throughout the state. Mining, atmospheric
45 deposition, and other sources predominantly release Hg to the aquatic environment in
46 inorganic forms, including elemental Hg (Hg⁰), cinnabar (HgS), and ionic Hg (Hg²⁺)

1 (USGS 2000). When transformed to its organic form, methylmercury (MeHg) by sulfate-
2 reducing bacteria (Gilmour *et al.* 1992), Hg becomes a significant toxicological concern
3 for biota. The primary pathways for increased methylation of inorganic Hg in lakes and
4 reservoirs are thought to relate to wetlands (St. Louis *et al.*, 1994), forests (St. Louis *et*
5 *al.*, 1996), and lake sediments (Ramlal *et al.*, 1993). Under certain conditions, Hg
6 methylation may also occur in the water column. But, a direct correlation between
7 sources of Hg and biota MeHg concentrations has yet to be shown for California lakes
8 and reservoirs.

9 Many studies have identified levels of MeHg that pose risks to wildlife and
10 humans that consume fish (e.g., Melwani *et al.* 2009; Eagles-Smith *et al.* 2009). MeHg
11 contamination has also been the principal driver for recent 303(d) water body listings,
12 Total Maximum Daily Loads (TMDLs), and many human health advisories in the state
13 (e.g., Fairey *et al.* 1997, Davis *et al.* 2008, 2009, 2010). Therefore, the bioaccumulation
14 of MeHg in fish tissue is considered a significant threat to the health of both wildlife and
15 humans, and is one of the primary indicators used to monitor water quality in the state.

16 The objective of this study was to develop a statistical model that describes the
17 driving factors of MeHg bioaccumulation in fish from California lakes and reservoirs.
18 MeHg concentrations in largemouth bass (*Micropterus salmoides*) were examined for
19 relationships to water and sediment chemistry, lake morphometry, and land use.

20 21 **Methods**

22
23 Total mercury in sport fish, from a statewide survey of 272 lakes and reservoirs in
24 California, sampled in 2007 and 2008, were considered for the statistical model (Davis *et*
25 *al.* 2009, 2010). Of these lakes, Moss Landing Marine Laboratories (MLML) prioritized
26 water and sediment sampling at 21 lakes based on guidance from the Regional Water
27 Quality Control Boards. The majority of these lakes (11 of 21) were located in the
28 Central Valley (Region 5), with the remainder in Region 1 (North Coast), Region 3
29 (Central Coast), and Region 8 (Santa Ana). Overall, fifteen of the 21 lakes selected by
30 MLML had data on Hg in largemouth bass (Table 1). In addition, MLML provided
31 largemouth bass MeHg data for an additional two lakes (Lake Hemet and Big Bear lake)
32 sampled in 2009 (Negrey and Stephenson, 2010), where water and sediment data had also
33 been collected. Therefore, 17 lakes were used to assess patterns related to MeHg
34 bioaccumulation in largemouth bass (Figure 1).

35 36 Prediction of Lake-specific Largemouth Bass MeHg Concentrations

37
38 In previous studies, largemouth bass have exhibited a strong size:MeHg
39 relationship when collected over a wide (spanning 150 mm or more) size range (Davis *et*
40 *al.* 2008; Melwani *et al.* 2009). In this study, MeHg in 350 mm largemouth bass was used
41 as a typical lake-specific estimate of MeHg. The 350 mm value was selected to represent
42 the middle (median) of the typical size distribution above the legal limit of 305 mm (12
43 in) for largemouth bass in California. The 350 mm concentration for 15 lakes was
44 estimated by employing a general linear model with maximum likelihood (PROC
45 MIXED in SAS v. 9.1; Littell *et al.* 1996). The approach was used to evaluate the “best”
46 regression model from which to estimate MeHg concentrations in largemouth bass

1 among lakes. In addition, two lakes (Lake Hemet and Big Bear Lake) sampled for
2 largemouth bass MeHg in 2009, were used to estimate 350 mm concentrations using a
3 simple linear regression approach. The resulting regression equations were used to
4 predict MeHg concentrations (mean and 95% confidence interval) for each lake in a 350
5 mm (total length) largemouth bass (Table 1). Further details of the general linear model
6 have been described in Davis *et al.* (2010).

7 8 Environmental Data

9
10 Data on lake morphometry (e.g., depth, volume, surface area) were obtained from
11 literature sources, websites, and lake management agencies, including the California
12 Department of Fish and Game, Regional Water Quality Control Boards, and the
13 Department of Water Resources (Appendix 1). A single attribute value representing
14 characteristics of lake morphometry was assigned to each lake used in the statistical
15 analysis (Table 2).

16 Land use data were calculated based on the lake catchment (sub-watershed) areas
17 (Figures 1 and 2). Catchment areas were determined from the Watershed Boundary
18 Dataset (WBD). Polygons from the “6th level” delineation (the smallest division of sub-
19 watersheds within WBD) were used in the GIS layer. Sampling locations for each study
20 basin were plotted, and the upstream sub-watershed area of each point was delineated.
21 The newly generated catchment areas were checked against other GIS data for accuracy.
22 These included the 4th & 5th level (more aggregated) of WBD, aerial photography, a 30
23 m hillshade, and the National Hydrography Dataset (NHD). In some cases, the WBD 6th
24 level delineations were coincident with lake-forming dams. In such cases, the polygons
25 were edited to exclude dams that would represent barriers to fish and environmental
26 variables. Once the watersheds were developed for each lake, land use data were overlaid
27 to allow for calculation of attributes related to mercury contamination of the catchment
28 area, such as number of mines, proportion of wetland area, and total watershed area
29 (Figure 2, Table 2, Appendix 1). In particular, three primary datasets were used to
30 represent habitat and mining information: CalVeg, National Wetlands Inventory (NWI),
31 and Mine Resources Data System (MRDS). The metadata behind each of these datasets
32 have been described (Melwani *et al.* 2007). In addition to the 17 lakes used for statistical
33 analysis, land use attributes were determined for an additional 21 lakes from the
34 statewide dataset where length-standardized largemouth bass MeHg concentrations had
35 been determined (Davis *et al.* 2010), to allow a qualitative assessment of sub-watershed
36 variables on a larger sample size of lakes. Lake morphometry and water chemistry data
37 were not readily available for this subset of lakes.

38 Lake chemistry data collected by Moss Landing Marine Laboratories (MLML)
39 were obtained for the 17 lakes included in the statistical analysis. Aqueous MeHg was
40 measured in surface (above thermocline) and deep (below thermocline) samples in both
41 summer (May – Sept) and winter (Oct – April) in 2008 and summer 2009. During each
42 monthly sample event, surface water samples were also measured for chlorophyll a,
43 sulfate (SO₄), and dissolved organic carbon (DOC). Total mercury in water was
44 measured on a single sampling event, once in the summer and once in the winter. Profile
45 data of temperature, conductivity, pH, and oxygen data in 3 m increments from the
46 surface to near-bottom at all lakes (50 ft. max depth) were also collected. Finally, total

1 mercury in sediment was collected once from the deepest portion of each lake in 2008.
2 Detailed methods for sampling and analysis have been described in Negrey and
3 Stephenson (2010).

4 5 Statistical Approach

6
7 Log-10 transformed lake-specific 350 mm largemouth bass MeHg concentration
8 (LMB₃₅₀ MeHg) was used as the response variable in all statistical analyses. Analysis
9 was conducted with predictor variables that were log-10 transformed and standardized
10 (i.e., mean centered and scaled to the standard deviation, often known as the z-score).
11 This allowed for all predictors to be evaluated with the same weight in the statistical
12 models. Therefore, each variable had the same influence, despite being measured on
13 different scales. Log transformations were used to improve linear relationships and
14 ensure normally distributed error values and equal variances in the analysis. Pearson
15 correlation analysis was used to evaluate the relationships among the predictor variables
16 and to LMB₃₅₀ MeHg.

17 Partial Least Square regression models (also called Projection of Latent Structures
18 or PLS) were used to assess the influence of 23 explanatory variables on standard-size
19 largemouth bass MeHg concentrations from 17 California lakes. PLS is a multivariate
20 regression method similar to Principal Component Regression (PCR). The main
21 difference is that PLS uses the information in the response variable to extract the useful
22 variance among the explanatory variables to form its components (Carrascal *et al.* 2009).
23 In other words, PLS maximizes the covariance between the explanatory and response
24 variable, whereas in PCR, the components are extracted independent of the relationship
25 in the predictor variables to the response.

26 PLS model development was initiated with the full complement of 23 predictor
27 variables included. Model evaluation consisted of identifying the following statistics:

- 28 1) the minimum number of components required to minimize the root mean
29 square error of predicted values;
- 30 2) the minimum number of predictors required to maximize the total variance
31 explained by the model;
- 32 3) loading results (correlation structure between the explanatory and response
33 variables);
- 34 4) regression coefficients (direction and strength of the predictors in the model);
35 and,
- 36 5) variance influence on projection (VIP; strength of influence among all PLS
37 components).

38 In subsequent PLS model runs, variables were removed based on their regression
39 coefficients, VIP, and relative contribution to the model. Although no defined limit exists
40 for statistical significance in PLS models, VIP limits of 0.8 or 1.0 have often been used
41 (Eriksson *et al.* 1995; Sonesten, 2004). In this study, significant predictor variables were
42 defined as having a VIP > 1.0. Predictors with small regressions coefficients or VIP < 1.0
43 were removed in the final model. Predictive ability of the different PLS models was
44 estimated by cross-validation (Eriksson *et al.*, 1995). The PLS models were developed
45 using the R Statistical Software Package (<http://cran.r-project.org>).
46

1
2 **Results and Discussion**
3

4 MeHg concentrations in 350 mm largemouth bass (here on referred as LMB₃₅₀
5 MeHg) at 17 lakes used in the statistical analysis, varied from 0.06 – 1.3 µg/g (Table 1).
6 There was a regional pattern to the concentrations; lakes in Region 1 (North Coast, 3 of
7 17 lakes) exhibited the highest concentrations, while lakes in Region 8 (Santa Ana, 5 of
8 17 lakes) exhibited the lowest (Figure 1). This pattern was consistent with the overall
9 distribution of largemouth bass MeHg concentrations found in the full statewide dataset
10 (Davis *et al.* 2010).
11

12 Correlation Structure
13

14 Pearson correlation analysis was used to evaluate relationships among the
15 variables included in the statistical models. Nine of the predictor variables were
16 significantly related to LMB₃₅₀ MeHg (Table 3). Conductivity had the strongest
17 correlation, exhibiting a Pearson $r = -0.79$. Total mercury (THg) in sediment was the
18 single most important environmental variable ($r = 0.69$) related to LMB₃₅₀ MeHg.
19 Latitude and longitude were also highly correlated with LMB₃₅₀ MeHg reflecting the
20 regional pattern in the concentrations. Generally, lakes in the north-east and north-west
21 portions of the state had higher LMB₃₅₀ MeHg than central coast and southern California
22 lakes. In addition, percent forested area ($r = 0.63$), THg in soils ($r = 0.65$), chlorophyll a
23 ($r = -0.51$), sulfate ($r = -0.50$), and annual water level flux ($r = 0.51$) were all significantly
24 correlated to LMB₃₅₀ MeHg. Pearson correlation analysis also indicated that average
25 concentrations of surface water MeHg and ancillary parameters (pH, DOC, etc.) collected
26 during the summer season had the highest correlation coefficients with LMB₃₅₀ MeHg
27 (seasonal comparison not presented). Among the predictor variables, many significant
28 correlations were also detected. The variables that were most commonly associated with
29 other predictors were conductivity, sulfate, latitude, watershed area, and forested area.
30 This is indicative of the multi-collinearity of the dataset and supports the use of a latent
31 structure approach in the modeling described below.
32

33 Summary of Partial Least Square Regression (PLS) Models
34

35 The influence of 23 environmental variables on LMB₃₅₀ MeHg was evaluated
36 using several PLS models. Each model sought to narrow the factors that ‘best’ describe
37 the variation in LMB₃₅₀ MeHg at the 17 lakes used in the analysis. All of the PLS model
38 runs were generally consistent regarding the significant environmental factors explaining
39 LMB₃₅₀ MeHg. Therefore, only two models are summarized in detail here, the most
40 complex model (Model 1) and the simplest model (Model 4). PLS model outputs for
41 Model 2 and Model 3 can be found in Appendix 2. All four models explained
42 approximately 80% of the variance in LMB₃₅₀ MeHg (Table 4). The models also
43 indicated that 18-49% of the variation in the predictor variables was ‘noise’, and thus did
44 not contribute significantly to the model results. In multiple regression analysis, this
45 variation in the predictor variables would have been included in the model parameters,
46 resulting in over-fitting and an inflated coefficient-of-determination (R^2). In all models,

1 the first two components were significant ($p < 0.05$) and explained the vast majority ($>$
2 70%) of the variance in LMB_{350} MeHg. However, in the simpler models (with fewer
3 variables), only component 1 was necessary to predict LMB_{350} MeHg. The regression
4 coefficients and VIP for Model 1 indicated that many of the predictor variables had little
5 effect on LMB_{350} MeHg. Twelve variables had relatively small regression coefficients
6 and low VIP (Table 5). In contrast, the variables included in the final model (Model 4)
7 were all relatively significant ($VIP > 1.0$) and contributed equally to the model (similar
8 regression coefficients; Table 6). Finally, the predictive ability of the models was highest
9 in Model 4. Cross-validation was used to evaluate the predictive ability and stability of
10 the PLS models. Model 4 indicated a model R^2 of 0.81 and a cross-validated R^2 of 0.75,
11 which was the highest prediction R^2 of the models evaluated.

12
13 The final model equation (Model 4) was:

$$\begin{aligned} \text{Log}_{10}(LMB_{350} \text{ MeHg}) = & 0.15 + 0.010(\text{Latitude}) + 0.012(\text{Longitude}) + 0.021(\text{THg-} \\ & \text{Sediment}) + 0.025(\text{MeHg-Water}) + 0.013(\text{THg-Soil}) + 0.010(\text{Forested-area}) \\ & - 0.019(\text{Conductivity}) \end{aligned}$$

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19 The majority of mean predicted values of LMB_{350} MeHg were within $0.25 \mu\text{g/g}$ of the
20 observed mean concentration (Table 7). The majority of predicted values were higher
21 than the observed concentration, suggesting that the model estimates are more
22 conservative than the observed values. The least deviation from observed was evident at
23 lakes in the low to moderate concentrations range ($0.1 - 0.6 \mu\text{g/g}$). This is likely because
24 the majority of lakes used in the model corresponded to this range in concentration. The
25 three lakes with the highest degree of bias (Thermalito Afterbay, Lake Hemet, O'Neill
26 Forebay) were outliers because their land use characteristics were anomalous. For
27 example, Thermalito Afterbay was biased by 1.1 times the observed concentrations
28 because it had relatively high proportion of forested area (78%) in its catchment, but the
29 observed concentration was not proportionally high. Similarly, Lake Hemet was biased
30 by 0.83 times the observed concentrations because it had a relatively low observed
31 LMB_{350} MeHg, but did not indicate the lowest values of THg in sediment or MeHg in
32 water. Therefore, this lake was predicted to have a higher LMB_{350} MeHg. Overall, the
33 majority of predicted values were reasonable given the limited sample size of lakes and
34 variables employed in the final model.

35 36 Factors Controlling Fish MeHg Concentrations

37
38 The seven predictor variables included in the final model can be separated into
39 three distinct groups for interpretation: spatial location (latitude and longitude); land
40 use/geology (THg in soils, THg in sediment, percent forested); and methylation (MeHg in
41 water and specific conductivity). These groups of variables explained 81% of the
42 variance in LMB_{350} MeHg. Exclusion of latitude and longitude from the final model
43 reduced the model variance by $< 5\%$ (results not presented). Therefore, despite latitude
44 and longitude having relatively strong influence in the final model (indicated by 3rd and
45 4th highest VIP), the variables did not contribute much variance to the prediction of
46 LMB_{350} MeHg. This suggests that the vast majority of the variation due to spatial or
47 regional differences was captured by other predictors, such as land use and geology.

1 Component 1 of the final model exhibited a LMB₃₅₀ MeHg explanatory variance
2 of 75%. Therefore, the variables driving the separation of lakes along this axis were
3 assumed to be the most dominant factors in the model. The regression coefficients
4 suggest the main controlling factors along Component 1 were spatial location, land use,
5 and conductivity (Figure 3). As described above, spatial location was an important factor,
6 but did not contribute much variance to the model. Therefore, forested area, THg in
7 sediment, and THg in soils, indicated the strongest, positive influences on LMB₃₅₀ MeHg,
8 while conductivity indicated a strong negative effect. The placement of land use
9 variables, as well as latitude and longitude, in a similar latent component space suggests
10 similar effect on LMB₃₅₀ MeHg. The equal importance of the variables is also illustrated
11 by the similar PLS regression coefficients for the first significant component (Table 6).
12 THg in sediment diverged from the other land use variables in Figure 3 mainly due to the
13 higher average concentration of THg in sediment at Lake Pillsbury. The comparable
14 regression coefficient and VIP of THg in sediment suggests that the influence of the
15 variable in the model is similar to the other land use variables. Overall, 11 of 17 lakes
16 exhibited LMB₃₅₀ MeHg > 0.3 µg/g, and seven of these were positively related to land use
17 variables.

18 Component 2 of the model contributed an additional 6% to the final model
19 variance. Only two lakes (Lake Irvine and Lake Pillsbury) separated strongly along this
20 axis suggesting these observations may be the result of outliers. MeHg in water and THg
21 in sediment had the two largest regression coefficients in Component 2, and thus
22 contributed most to the separation of lakes. Lake Pillsbury likely separated along
23 Component 2 due to much higher MeHg in water and LMB₃₅₀ MeHg than any of the
24 other lakes. The separation of Lake Irvine was more likely an artifact of data availability
25 for the model, as it lacked information on THg in soil. If Lake Irvine were to have
26 followed the pattern in THg in soils observed at other lakes, it is likely this lake would
27 have corresponded to the positive side of Component 1. Overall, the results along
28 Component 2 did not drive much of the model variance, and thus have not been
29 emphasized in the following interpretations.

30 *Land Use and Geology*

31 Lakes and reservoirs located in Region 1 (Lake Sonoma and Lake Mendocino)
32 and Region 5 (Folsom Lake, Lake Natomas, Don Pedro Reservoir, Lake McSwain, and
33 Lake McClure) were associated with catchments with higher LMB₃₅₀ MeHg, and
34 relatively high forested area (> 70%) and THg in soils. These parameters suggest that
35 land use and habitat may be critical to understanding the higher concentrations at these
36 lakes. Several previous studies of habitat influences on fish mercury concentrations in
37 lake catchments have found significant correlations with land cover and habitat features
38 of the watershed (e.g., Hurley *et al.* 1995; Wiener *et al.* 2006; Chumchal *et al.* 2008).
39 Furthermore, differences in mercury methylation efficiency have been reported on both
40 land use scales (e.g., agricultural, industrial, urban; Krabbenhoft *et al.* 1999) as well as
41 finer, habitat scales (e.g., wetland, forested, un-vegetated; Hurley *et al.* 1995; Wiener
42 *et al.* 2006). The limited sample size of lakes used in this analysis precluded separation on
43 such scales in the model, but could be attempted on a larger sample size in future work.

44 Many types of wetland and forested habitat can exhibit conditions that are
45 favorable for Hg methylation (Zillioux *et al.* 1993). Understanding the many interacting
46

1 factors that control MeHg production in these habitat types is still an area of ongoing
2 research (Wiener *et al.* 2003). One focus area has been to understand the activity of
3 sulfur-reducing bacteria, which thrive in anoxic soils and sediment (Gilmour and Henry
4 1991, Gilmour *et al.* 1992). Forested areas have been observed to transport the more
5 reactive forms of Hg relative to other land use types (Chumchal *et al.* 2008), and sulfur-
6 reducing bacteria are primarily responsible for this transformation. Accordingly, positive
7 correlations between forested area and MeHg concentration in water and largemouth bass
8 have been observed previously (St Louis *et al.* 1994; Hurley *et al.* 1995; Krabbenhoft *et*
9 *al.* 1999; Chumchal *et al.* 2008). The indication from these results is that the association
10 between LMB₃₅₀ MeHg and land use may occur on a regional scale, but lakes in certain
11 areas of the state deviate from the observed pattern.

12 Mining was not indicated to have a direct correlation with LMB₃₅₀ MeHg in this
13 analysis. In addition to the lack of influence in the PLS models, a qualitative assessment
14 of an additional 21 lakes indicated that many lakes that had relatively high LMB₃₅₀ MeHg
15 coincided with watersheds with very few mines. Twelve lakes had LMB₃₅₀ MeHg > 0.8
16 µg/g, but only three of these had more than three mines in their watershed (Table 8).
17 Furthermore, two lakes with the lowest LMB₃₅₀ MeHg (< 0.03 µg/g) were indicated to
18 have more than 30 mines within their watershed. The lack of consistent correlation to
19 number of gold and mercury mines suggests that other variables are responsible for
20 driving elevated LMB₃₅₀ MeHg at the majority of these lakes. It has been suggested that
21 THg in sediments and soil from mine sites are not as bioavailable as those from diffuse
22 sources, such as atmospheric deposition or urban runoff (Krabbenhoft *et al.* 1999), which
23 may somewhat explain the lack of correlation to LMB₃₅₀ MeHg. It is also possible that
24 the mining data layer is not precise enough to support this type of analysis (see Davis *et*
25 *al.* [2010] for further discussion).

26 The qualitative assessment of 21 lakes further pointed towards forested area being
27 associated with many of the lakes of higher LMB₃₅₀ MeHg in northern California. New
28 Melones, Crystal Lake, and Eastman Lake had LMB₃₅₀ MeHg between 0.95 – 1.12 µg/g
29 and greater than 75% forested area in their watersheds. This was similar to the pattern
30 evident at the initial set of lakes used for modeling. However, in southern California,
31 where lake catchments have relatively little forested area, no obvious pattern could be
32 deduced, with some lakes having relatively high (> 0.6 µg/g) or low (< 0.3 µg/g) LMB₃₅₀
33 MeHg. In addition, patterns of wetland area associated with elevated LMB₃₅₀ MeHg were
34 inconclusive. Overall, the results suggests a hypothesis for at least some lakes, where Hg
35 may be leached more readily from forested areas than in catchments dominated by other
36 habitat types. However, the potential sources of Hg to these forested catchments still
37 remains an open question, due to the lack of correlation to mine data, and the inability to
38 examine atmospheric or other watershed-scale sources (e.g., POTWs) in this analysis.

39 40 *Mercury Methylation*

41 Hg methylation rates are controlled by both the physical and biogeochemical
42 conditions of a water body. Lakes and reservoirs are often environments of elevated Hg
43 methylation due to daily and seasonally changing environmental conditions. Fluctuations
44 in the water level of reservoirs can cause spikes in Hg methylation. For example, when
45 the perimeter of a reservoir periodically dries out and then submerges, it can become an
46 area of intense MeHg production (Johnson *et al.* 1991). Additionally, during the summer,

1 lake conditions can often enhance the methylation process by the presence of warmer,
2 slow moving water, which promotes algal growth and can reduce oxygen levels. These
3 features of lakes were examined using several characteristics of lake morphometry and
4 water chemistry variables, but were generally not significant (Table 3). The only
5 significant morphometric parameter was annual water level change ($r = 0.49$). However,
6 in the final model other variables were more closely related to LMB₃₅₀ MeHg.

7 The final model indicated MeHg in water to be an important factor related to
8 LMB₃₅₀ MeHg, which was the only direct proxy for methylation evaluated. The
9 correlation to LMB₃₅₀ MeHg may point towards enhanced exposure of largemouth bass to
10 MeHg in the water column under certain environmental conditions. A recent mercury
11 food web model for river systems indicated that largemouth bass tend to forage more
12 during the summer, and exhibit higher activity and metabolism rates (Greenfield and
13 Lent, 2008). As a result, largemouth bass may have higher MeHg exposure during the
14 summer due to feeding more at the surface, where their piscivorous prey reside (Moyle
15 2002). Therefore, movement and behavior of largemouth bass during the summer may
16 partly explain the correlations observed with MeHg in water at some lakes (particularly,
17 Lake Pillsbury and Lake Irvine). MeHg water data collected in the Sacramento-San
18 Joaquin Delta have also indicated strong relationships ($R^2 = 0.91$) with 350 mm
19 largemouth bass (Wood *et al.* 2006, Davis *et al.* 2008). Further discussion of the water
20 column MeHg data and the relationship to MeHg bioaccumulation is reported in a
21 companion paper by Negrey and Stephenson (2010).

22 Finally, conductivity has been found to be associated with lower methylation rates
23 in some aquatic environments and was indicated as an important factor in the statistical
24 analysis. Many of the lakes with LMB₃₅₀ MeHg $< 0.3 \mu\text{g/g}$ exhibited conductivity values
25 $> 300 \text{ mS}$. Moreover, the lake with the highest LMB₃₅₀ MeHg (Lake Pillsbury, $1.31 \mu\text{g/g}$),
26 had the lowest conductivity ($< 1 \text{ mS}$). MeHg in fish tissue has often been found to have a
27 negative correlation with conductivity, hardness, and alkalinity (e.g., Hanten Jr. *et al.*
28 1998, Wren *et al.* 1991, Sonesten 2004). Enhanced microbial production of MeHg has
29 also been shown under low pH and hardness conditions (Xun *et al.* 1987). Therefore, it is
30 plausible that the negative correlation of LMB₃₅₀ MeHg with conductivity observed here
31 relates to some underlying relationship to Hg methylation. A previous study of boreal
32 lakes in Sweden also suggested that higher concentrations of ions such as Ca^{2+} and Mg^{2+}
33 were related to lower MeHg in the food web (Sonesten, 2004). High Ca^{2+} levels have also
34 been suggested as a potential inhibitor of MeHg production and direct water-borne uptake
35 by fish (Hanten Jr. 1998).

36 37 Limitations and Further Work

38
39 It should be acknowledged that the lack of clear relationships of some predictor
40 variables in these analyses may be due to the limitations of some of the underlying
41 datasets rather than a true absence of influence. Specifically, datasets on both mining and
42 wetlands were inconsistent and limited. The National Wetlands Inventory (NWI) was
43 used to display wetland area, but had many areas of missing or incomplete coverage. The
44 wetland polygons often differed depending on the individual cartographers who
45 performed the digitizing, and some regions completely lacked NWI coverage. The
46 regional variability in NWI likely inhibited the ability to detect correlations to LMB₃₅₀

1 MeHg. Similarly, the MRDS data used to enumerate gold and mercury mines, lacked
2 consistent information across the state. Due to limitations in the mining dataset, it was not
3 possible to account for mining quantities or Hg loadings to the environment. The mine
4 layer appeared useful for distinguishing broad areas of intense mining from water bodies
5 without mining influence, but did not correlate well with LMB₃₅₀ MeHg at a regional or
6 watershed-specific scale.

7 Another data gap was the lack of atmospheric deposition information for the
8 watersheds of interest. Atmospheric deposition is considered to be a principal source of
9 inorganic mercury to most aquatic systems in the United States. However, few
10 atmospheric deposition datasets have been collected in California, and only a handful of
11 monitoring stations have gathered long-term data (e.g., San Jose, CA; SFEI 2001). It is
12 thought that wet and dry deposition of Hg is relatively low and homogenous across
13 California (e.g., NADP 2004), but attempts to determine relative contributions to the food
14 web have been limited by a lack of data. Future attempts to relate lake and watershed
15 attributes with MeHg in the food web should consider prioritizing work around the
16 factors found to be significant in this study (particularly, forested area, THg in soil and
17 sediments, and MeHg in water). However, the influence of some watershed attributes that
18 could not be examined fully here, such as wetlands and atmospheric deposition, also
19 warrant further investigation.

20 Finally, one major caveat to the analyses presented here that needs acknowledgment,
21 is the limited sample size and distribution of lakes used in the statistical model. The lakes
22 used for the statistical analysis presented in this report were based on Regional Board
23 priorities and were not initially selected with the goal of developing a statistical model. A
24 more robust approach to the study design would have been to adequately represent the
25 range of land use, lake morphometry, and Hg methylation conditions across the state.
26 Therefore, the statistical model described here may only be appropriate for the population
27 of lakes included in the analysis, and should not be extrapolated outside this sample
28 space.

29 Conclusion

30
31
32 Methylmercury concentrations in largemouth bass from 17 lakes across California
33 varied from 0.06 – 1.3 µg/g, with highest concentrations in the northern portion of the
34 state. Lake variables that were related to MeHg concentrations in 350 mm largemouth
35 bass were THg in sediment and soils, forested area, specific conductivity, and MeHg in
36 surface water. The strong influence of land use, geology, and conductivity on fish MeHg
37 concentrations warrants further investigation. The results of this analysis are consistent
38 with several studies that have suggested that broad land use characteristics can potentially
39 influence MeHg bioaccumulation in lakes and reservoirs. Furthermore, the transport of
40 Hg from source to watershed may be a critical step leading to high MeHg levels in fish.
41 However, the limited sample size and spatial distribution of lakes evaluated here preclude
42 extending our model beyond the population of lakes included in this analysis.

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1 **References**

- 2
- 3 Alpers, C. N., and M. P. Hunerlach. 2000. Mercury contamination from historic gold
4 mining in California. Fact Sheet FS-061-00, USGS.
- 5
- 6 Carrascal, Partial least squares regression as an alternative to current regression
7 methods used in ecology, *Oikos* 118: 681-690
- 8
- 9 Chumchal *et al.* 2008 Habitat-specific differences in mercury concentration in a
10 top predator from a shallow lake, *Transactions of the American Fisheries Society*
11 137:195–208
- 12
- 13 Davis, J.A., Greenfield, B.K., Ichikawa, G., Stephenson, M., 2008a. Mercury in sport
14 fish from the Sacramento–San Joaquin delta region, California. *Science of the*
15 *Total Environment* 391, 66–75.
- 16
- 17 Davis, J.A., Melwani, A.R., Bezalel, S.N., Ichikawa, G., Bonnema, A., Lamerdin, C.,
18 Heim, W.A., Crane, D., Stephenson, M., 2009. The first year of a two-year screening
19 survey of contaminants in sport fish from California lakes and reservoirs. San Francisco
20 Estuary Institute, Oakland, CA.
- 21
- 22 Davis, J.A., Melwani, A.R., Bezalel, S.N., Ichikawa, G., Bonnema, A., Lamerdin, C.,
23 Heim, W.A., Crane, D., Stephenson, M., 2010. Two-year screening survey of
24 contaminants in sport fish from California lakes and reservoirs. San Francisco Estuary
25 Institute, Oakland, CA.
- 26
- 27 Domagalski, J. 1998. Occurrence and transport of total mercury and methyl mercury in
28 the Sacramento River Basin, California. *Journal of Geochemical Exploration* 64:277-291.
- 29
- 30 Eagles-Smith, C.A, Ackerman, J.T., De La Cruz, S.E.W., and Takekawa, J.Y., 2009.
31 Mercury bioaccumulation and risk to three waterbird foraging guilds is influenced by
32 foraging ecology and breeding stage, *Environmental Pollution*, vol. 157(7): 1993-2002
- 33
- 34 Eriksson, L., Hermens, J. L. M., Johansson, E., Verhaar, H. J. M. and Wold, S. 1995.
35 Multivariate analysis of aquatic toxicity data with PLS', *Aquat. Sci.* 57, 217–241
- 36
- 37 Fairey, R., K. Taberski, S. Lamerdin, E. Johnson, R. P. Clark, J. W. Downing, J.
38 Newman, and M. Petreas. 1997. Organochlorines and other environmental contaminants
39 in muscle tissues of sport fish collected from San Francisco Bay. *Marine Pollution*
40 *Bulletin* 34:1058-1071.
- 41
- 42 Fitzgerald, W.F. and Clarkson, T.W. 1991. Mercury and monomethylmercury: present
43 and future concerns, *Environ Health Perspect*, vol. 96: 159–166
- 44
- 45 Gilmour, C. C., and E. A. Henry. 1991. Mercury methylation in aquatic systems affected
46 by acid deposition. *Environmental Pollution* 71:131-170.

- 1
2 Gilmour, C. C., Henry, E. A., and Mitchell, R. 1992. Sulfate stimulation of mercury
3 methylation in freshwater sediments. *Environ. Sci. Technol.*, 26, 2281-2287
4
5 Greenfield, B.K. and Lent, M.A. 2008. Modeling mercury bioaccumulation in
6 largemouth bass in the Sacramento-San Joaquin River Delta and tributaries, SFEI
7 Technical Report, San Francisco Estuary Institute, Oakland, CA
8
9 Hammerschmidt, C.R., and Fitzgerald, W.F. 2006. Methylmercury in freshwater fish
10 linked to atmospheric mercury deposition, *Environ. Sci. Technol.*, vol. 40(24): 7764–
11 7770
12
13 Hanten, Jr, R.P., Neumann, R.M., Ward, S.M., Carley, R.J., Perkins, C.R., and Pirrier, R.
14 1998. Relationships between concentrations of mercury in largemouth bass and physical
15 and chemical characteristics of Connecticut lakes, *Transactions of the American Fisheries*
16 *Society* 127:807–818
17
18 Hurley, J. P., J. M. Benoit, C. L. Babiarz, M. M. Shafer, A. W. Andren, J. R. Sullivan, R.
19 Hammond, and D. A. Webb. 1995. Influences of watershed characteristics on mercury
20 levels in Wisconsin Rivers. *Environmental Science and Technology* 29:1867-1875.
21
22 Johnson, T.A., R.A Bodaly, R.A., and Mathias, J.A. 1991; Predicting fish mercury levels
23 from physical characteristics of boreal reservoirs; *Canadian Journal of Fisheries and*
24 *Aquatic Science* 48:1468-1475
25
26 Krabbenhoft, D. P., J. G. Wiener, W. G. Brumbaugh, M. L. Olson, J. F. DeWild, and T. J.
27 Sabin. 1999. A national pilot study of mercury contamination of aquatic ecosystems
28 along multiple gradients. U.S. Geological Survey Toxic Substances Hydrology Program
29 Proceedings of Technical Meeting 2:147-160.
30
31 Littell, R.C., Milliken, G.A., Stroup, W.W., Wolfinger, R.D., 1996. SAS System for
32 Mixed Models. SAS Institute, Inc., Cary, NC.
33
34 Melwani, A.R., Bezalel, S.N., Grenier, J.L., Hunt, J.A., Robinson, A.H., and Davis, J.A.,
35 2007. The relationship between landscape features and sport fish mercury in the
36 Sacramento-San Joaquin Delta watershed, SFEI Contribution No. 534, San Francisco
37 Estuary Institute, Oakland, CA.
38
39 Melwani, A.R., Bezalel, S.N., Hunt, J.A., Grenier, J.L., and Davis, J.A. 2009. Spatial
40 trends and impairment assessment of mercury in fish from the Sacramento-San Joaquin
41 Delta watershed, *Environmental Pollution*, vol. 157(11): 3137-49
42
43 Moyle, P. B. 2002. *Inland Fishes of California*. University of California Press, Berkeley,
44 CA.
45

- 1 Negrey, J. and Stephenson, M. 2010. Mercury in water, sediment, and fish in California
2 Lakes, Draft Report
3
- 4 Ramlal, P. S., Kelly, C. A., Rudd, J. W. M. and Furutani, A. 1993. Sites of methyl
5 mercury production in remote Canadian Shield lakes', *Can. J. Fish. Aquat. Sci.* 50, 972–
6 979.
7
- 8 San Francisco Estuary Institute [SFEI]. 2001. San Francisco Bay Atmospheric Deposition
9 Pilot Study Part 1: Mercury. San Francisco Estuary Institute, Oakland, CA.
10
- 11 Sonesten, L. 2004. Catchment area composition and water chemistry heavily affects
12 mercury levels in perch (*Perca fluviatilis* L.) in circumneutral lakes, *Water, Air, and*
13 *Soil Pollution* 144: 117–139, 2003
14
- 15 St. Louis, V. L., Rudd, J. W. M., Kelly, C. A., Beaty, K. G., Bloom, N. S. and Flett, R. J.
16 1994. Importance of wetlands as sources of methyl mercury to boreal forest ecosystems,
17 *Can. J. Fish. Aquat. Sci.* 51, 1065–1076
18
- 19 St. Louis, V. L., Rudd, J. W. M., Kelly, C. A., Beaty, K. G., Flett, R. J. and Roulet, N. T.
20 1996. Production and loss of methylmercury and loss of total mercury from boreal forest
21 catchments containing different types of wetlands, *Environ. Sci. Technol.* 30, 2719–2729
22
- 23 United States Environmental Protection Agency [USEPA], 1997, Mercury Study Report
24 to Congress, United States Environmental Protection Agency, EPA-452-97-003 –010,
25 Office of Air and Radiation, 1980 pp.
26
- 27 United States Geological Survey [USGS]. 2000. Mercury contamination from historic
28 gold mining in California. Fact Sheet FS-061-00. U.S. Geological Survey, Sacramento,
29 CA.
30
- 31 Wiener, J.G., Krabbenhoft, D.P., Heinz, G.H., and Scheuhammer, A.M., 2003,
32 *Ecotoxicology of mercury*, chap. 16 of Hoffman, D.J., Rattner, B.A., Burton, G.A., Jr.,
33 and Cairns, J., Jr., eds., *Handbook of ecotoxicology* (2d ed.): Boca Raton, Fla., Lewis
34 Publishers, p. 409–463.
35
- 36 Wiener, J. G., Knights, B.C., Sandheinrich, M.B., Jeremiason, J.D., Brigham, M.E.,
37 Engstrom, D.R., Woodruff, L.G., Cannon, W.F., and Balogh, S.J. 2006. Mercury in soils,
38 lakes, and fish in Voyageurs National Park (Minnesota): Importance of atmospheric
39 deposition and ecosystem factors. *Environmental Science & Technology* 40: 6261-6268.
40
- 41 Wren, C. D., W. A. Scheider, D. L. Wales, B. W. Muncaster, and I. M. Gray. 1991.
42 Relation between mercury concentrations in walleye *Stizostedion vitreum vitreum* and
43 northern pike *Esox lucius* in Ontario lakes and influence of environmental factors.
44 *Canadian Journal of Fisheries and Aquatic Sciences* 48:132–139.
45

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- 1 Xun, L., N. E. R. Campbell, and J. W. M. Rudd. 1987. Measurements of specific rates of
2 net methyl mercury production in the water column and surface sediments of acidified
3 and circumneutral lakes. *Canadian Journal of Fisheries and Aquatic Sciences* 44:750–
4 757.
5
6 Wood ML, Foe C, and Cooke J. 2006. Sacramento–San Joaquin Delta Estuary TMDL for
7 Methylmercury: Draft Report for Scientific Peer Review. Regional Water Quality
8 Control Board, Central Valley Region. Rancho Cordova, CA, USA
9
10 Zillioux, E. J., D. B. Porcella, and J. M. Benoit. 1993. Mercury cycling and effects in
11 fresh-water wetland ecosystems. *Environmental Toxicology and Chemistry* 12:2245–
12 2264.
13



Figure 1. Map of 17 sub-watersheds used in this study. Blue boundaries delineate the upstream area for each lake. Dot colors correspond to 350 mm lake-specific MeHg concentration ranges for largemouth bass.

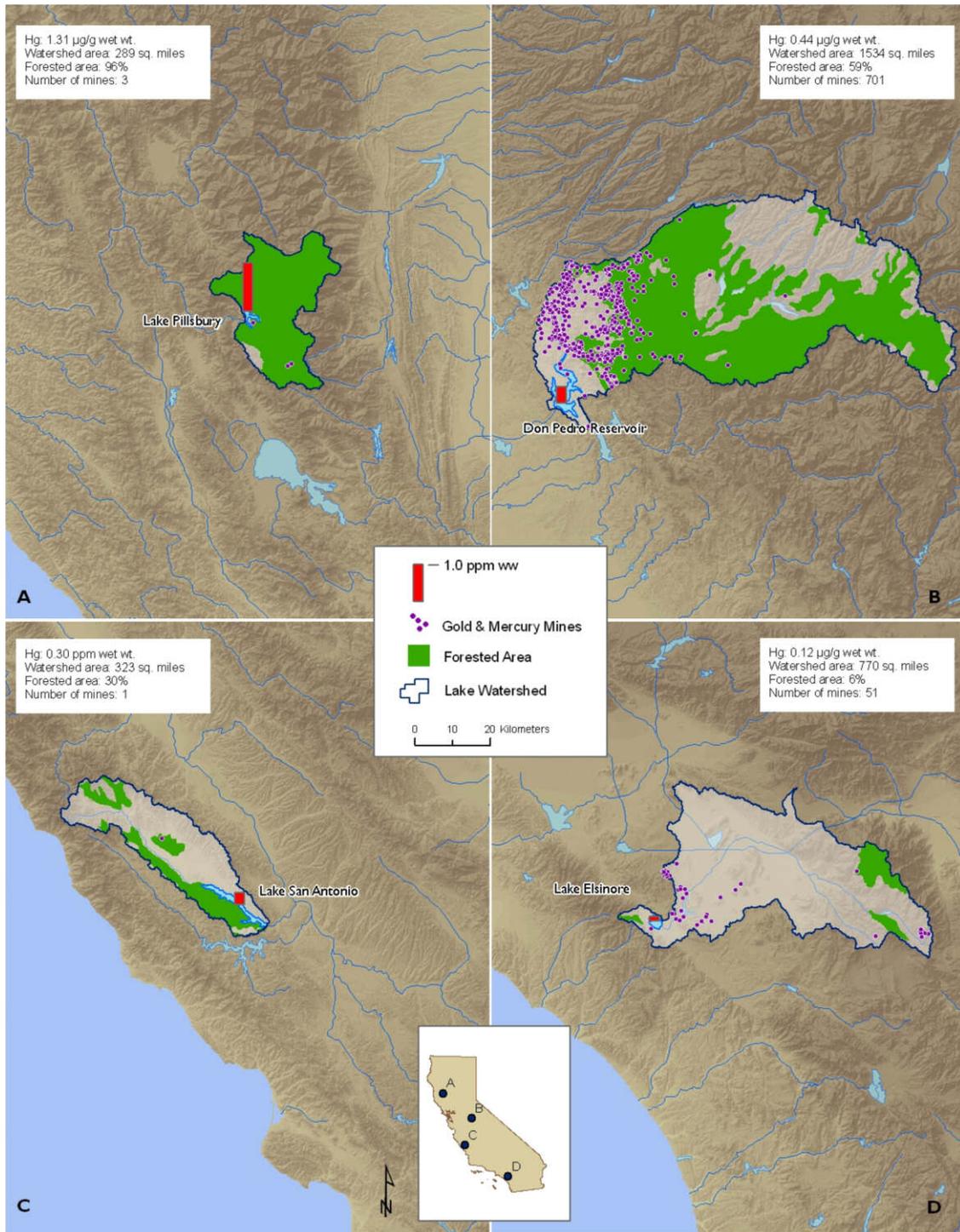


Figure 2. Example of land use attributes for four lakes used in the statistical analysis.

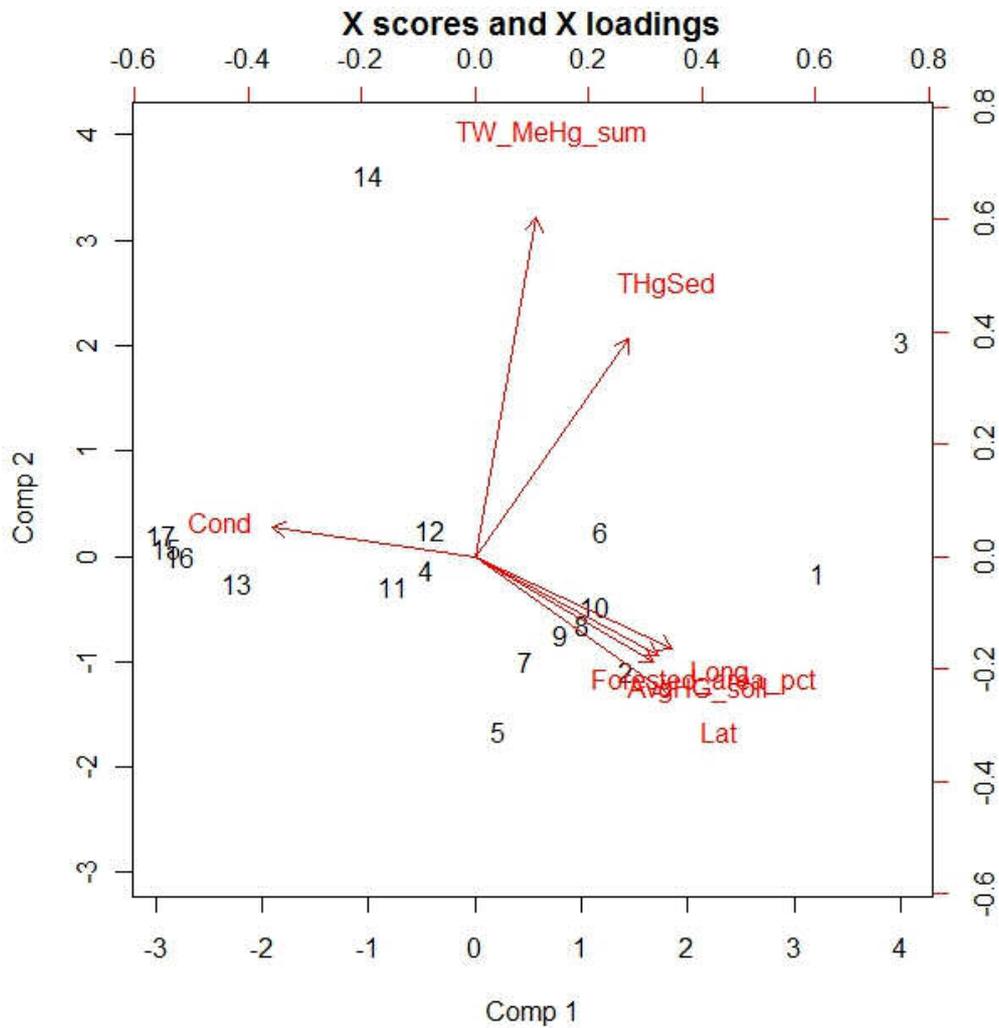


Figure 3. Biplot of latent x-scores and loadings from the final model (Model 4). Numbers correspond to lakes and reservoirs listed in Table 1. Length of each vector indicates the relative strength in the model. For example, a strong, negative relationship between conductivity and largemouth bass MeHg concentrations is indicated at lakes 15, 16, and 17.

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Table 1. MeHg concentrations in 350 mm largemouth bass from 17 lakes in California

Lake Number	Regional Board	Lake Name	350 mm Largemouth bass MeHg (µg/g)
1	1	Lake Sonoma	0.68
2	1	Lake Mendocino	0.54
3	1	Lake Pillsbury	1.31
4	3	Lake San Antonio	0.30
5	5	Thermalito Afterbay	0.21
6	5	Folsom Lake	0.47
7	5	Lake Natomas	0.54
8	5	Don Pedro Reservoir	0.44
9	5	Lake McClure	0.77
10	5	Lake McSwain	0.54
11	5	San Luis Reservoir	0.56
12	5	O'Neill Forebay	0.23
13	8	Big Bear Lake	0.18
14	8	Irvine Lake	0.48
15	8	Perris Reservoir	0.10
16	8	Lake Hemet	0.06
17	8	Lake Elsinore	0.12

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Table 2. Description of the dependent and independent variables evaluated in the statistical model.

Variable	Description	Unit	Mean	Min	Max
Largemouth bass MeHg	Largemouth bass MeHg concentration at 350mm	µg/g	0.44	0.06	1.31
Chlorophyll a	Chlorophyll a concentration of lake surface water (< 2m)	µg/L	6.95	0.92	50.49
Conductivity	Conductivity of lake surface water	mS	439	0.1	2933
DO	Dissolved oxygen content of lake surface water (< 6m)	mg/L	8.45	5.49	10.57
DOC	Dissolved organic carbon content of lake surface water (< 2m)	mg/L	25.19	2.05	154.0
Elevation	Lake elevation	Ft	1298	129	6760
Forested area	Forested coverage in the lake catchment area	%	0.5	0	1.0
Lake age	Age of lake relative to 2009	Years	57	27	121
Latitude	Latitude Coordinates	DMS	36.9	33.7	39.5
Longitude	Longitude Coordinates	DMS	120.2	116.7	123.2
Max annual depth change	Maximum annual water level fluctuation of lake	Ft	39	3	157
Max depth	Maximum depth of lake	Ft	155	17	530
Mines	Number of gold and mercury mines in catchment area	Count	510	0	2539
pH	pH of lake surface water (< 6m)	pH units	7.8	5.3	8.9
Storage capacity	Maximum volume of water storage of lake	acre-feet	434000	8000	2039000
Sulfate	Sulfate concentration in lake surface water (< 2m)	mg/L	37.42	3.45	181.33
Surface area	Surface area of lake	square-miles	6.8	0.5	20
Temperature	Temperature of lake surface water (< 6m)	centigrade	21.8	14.9	25.1
TOC	Total organic carbon content of lake sediment	%	8.11	2.17	15.33
Total sediment Hg	Total mercury concentration of lake sediment	µg/g	0.09	0.01	0.21
Total soil Hg	Total mercury content of soil in catchment area	µg/g	0.05	0.00	0.10
Total water Hg	Total mercury concentration of lake surface water (< 2m)	ng/L	0.74	0.25	1.54
Water methyl Hg	Methylmercury concentration of lake surface water (< 2m)	ng/L	0.05	0.01	0.18
Watershed area	Total area of lake catchment	square-miles	767	10	3639
Wetland area	Wetland coverage in the lake catchment area	%	0.70	0.00	3.44

Table 3. Correlation matrix of 350 mm largemouth bass MeHg concentrations with each predictor variable and of predictors to each other. Statistic calculated is the Pearson (r) correlation coefficient. Bold values were statistically significant ($p < 0.05$) and boxed values have Pearson $r \geq 0.8$ or ≤ -0.8 (at 1 significant digit).

	Hg_Lake_Log1	Latitude	Longitude	TOC	THgSed	TW_Chla	TW_SO4	TW_DOC	TW_MeHg	TW_THg	Watershed_area	Mines	Wetland_area	Wetland_pct	AvgHG_soil	MaxHG_soil	Forested_area_pct	Storage	Max.Depth.ft.	SArea	WA.SA	Elevation	Age	Max_DepthChange_abs	Temp	O2	pH	Cond	
Hg_Lake_Log1	1.00																												
Latitude	0.66	1.00																											
Longitude	0.69	0.93	1.00																										
TOC	-0.30	-0.62	-0.61	1.00																									
THgSed	0.69	0.29	0.39	0.09	1.00																								
TW_Chla	-0.51	-0.77	-0.62	0.41	-0.37	1.00																							
TW_SO4	-0.50	-0.80	-0.59	0.32	-0.18	0.85	1.00																						
TW_DOC	-0.41	-0.78	-0.58	0.36	-0.13	0.86	0.97	1.00																					
TW_MeHg	0.47	-0.04	0.08	-0.09	0.62	0.01	0.30	0.36	1.00																				
TW_THg	0.32	0.05	0.26	-0.15	0.45	0.21	0.31	0.36	0.50	1.00																			
Watershed_area	0.24	0.56	0.38	-0.46	-0.09	-0.29	-0.56	-0.58	-0.15	0.02	1.00																		
Mines	0.06	0.31	0.02	-0.20	-0.18	-0.31	-0.60	-0.61	-0.29	-0.27	0.85	1.00																	
Wetland_area	-0.03	0.42	0.19	-0.46	-0.30	-0.32	-0.55	-0.63	-0.36	-0.31	0.87	0.86	1.00																
Wetland_pct	-0.24	0.19	-0.01	-0.27	-0.43	-0.20	-0.40	-0.53	-0.45	-0.39	0.67	0.69	0.89	1.00															
AvgHG_soil	0.65	0.72	0.79	-0.21	0.49	-0.44	-0.55	-0.54	-0.11	0.27	0.34	0.05	0.14	0.00	1.00														
MaxHG_soil	0.42	0.67	0.56	-0.45	0.09	-0.45	-0.71	-0.74	-0.24	-0.03	0.80	0.69	0.73	0.52	0.61	1.00													
Forested_area_pct	0.63	0.76	0.64	-0.44	0.36	-0.74	-0.81	-0.79	-0.01	0.17	0.58	0.51	0.45	0.32	0.56	0.64	1.00												
Storage	0.29	0.30	0.35	0.01	0.23	-0.03	-0.19	-0.22	-0.13	-0.27	0.07	-0.04	0.08	0.00	0.45	0.26	0.09	1.00											
Max.Depth.ft.	0.32	0.22	0.23	0.18	0.28	-0.23	-0.39	-0.42	-0.21	-0.40	-0.01	0.04	0.03	0.06	0.44	0.33	0.22	0.76	1.00										
SArea	0.10	0.25	0.25	-0.03	-0.01	0.11	-0.10	-0.14	-0.19	-0.31	0.25	0.10	0.25	0.18	0.29	0.24	-0.01	0.92	0.54	1.00									
WA.SA	0.15	0.34	0.17	-0.37	-0.09	-0.33	-0.47	-0.46	-0.03	0.18	0.77	0.75	0.66	0.52	0.13	0.61	0.53	-0.52	-0.33	-0.42	1.00								
Elevation	-0.20	-0.62	-0.61	0.69	-0.02	0.37	0.34	0.38	0.05	-0.12	-0.53	-0.30	-0.49	-0.20	-0.40	-0.60	-0.24	-0.02	0.16	-0.05	-0.45	1.00							
Age	-0.40	-0.63	-0.67	0.40	-0.33	0.49	0.49	0.51	0.18	0.08	-0.10	0.04	-0.21	-0.04	-0.69	-0.50	-0.32	-0.52	-0.40	-0.33	0.12	0.56	1.00						
Max_DepthChange_abs	0.51	0.36	0.43	0.02	0.42	-0.18	-0.31	-0.27	0.14	-0.13	0.09	-0.03	0.00	-0.15	0.53	0.39	0.18	0.70	0.76	0.51	-0.22	-0.15	-0.47	1.00					
Temp	0.06	-0.06	0.08	-0.11	0.16	0.18	0.26	0.27	0.29	0.05	-0.18	-0.35	-0.21	-0.27	0.02	-0.21	-0.10	0.52	0.28	0.48	-0.51	0.18	-0.06	0.44	1.00				
O2	0.21	0.24	0.20	0.08	0.18	-0.41	-0.42	-0.40	-0.16	-0.40	-0.21	-0.04	-0.02	0.00	0.16	0.18	0.06	0.14	0.37	-0.04	-0.12	-0.11	-0.51	0.26	-0.48	1.00			
pH	-0.29	-0.34	-0.15	0.29	0.06	0.44	0.57	0.56	0.20	0.18	-0.48	-0.65	-0.46	-0.27	-0.17	-0.65	-0.40	0.24	-0.06	0.30	-0.67	0.45	0.15	0.00	0.61	-0.32	1.00		
Cond	-0.79	-0.78	-0.70	0.36	-0.54	0.79	0.81	0.74	-0.15	-0.12	-0.38	-0.27	-0.23	-0.10	-0.63	-0.50	-0.86	-0.15	-0.26	-0.01	-0.34	0.16	0.45	-0.28	0.07	-0.29	0.29	1.00	

Table 4. Summary of Partial Least Squares (PLS) regression models

Model #	Minimum Number of Components	Number of Predictors Evaluated	Variance in Predictors Explained By Model (%)*	Variance in <i>LMB Hg</i> Explained By Model (%)*
1	2	23	51%	83%
2	1	11	76%	82%
3	1	10	81%	81%
4	1	7	82%	81%

* Determined for two components for comparison among models

Table 5. Regression coefficients and variance in projection (VIP) for Model 1 (the full model). Horizontal lines separate predictors based on relative significance in VIP (i.e. < 0.08 , $0.8 - 1.0$, > 1.0)

Predictor	Regression Coefficients for Component 2	VIP
Conductivity	-0.014	1.28
THg Sediment	0.015	1.19
MeHg Water	0.016	1.13
Longitude	0.009	1.11
Latitude	0.008	1.08
THg Soil	0.009	1.04
Forested area	0.009	1.01
Sulfate	-0.004	0.88
Chlorophyll a	-0.004	0.88
DOC	-0.001	0.83
Annual Depth Change	0.009	0.82
Wetland area	-0.011	0.76
Age	-0.002	0.75
THg Water	0.010	0.71
Elevation	0.002	0.55
Max Depth	0.004	0.52
TOC	-0.003	0.52
Storage	0.004	0.47
pH	-0.004	0.46
DO	0.002	0.37
Mines	-0.003	0.33
Watershed area: surface area	0.0005	0.30
Temp	0.002	0.14

Table 6. Regression coefficients and variance in projection (VIP) for Model 4 (the simplest PLS Model). Component 1 explained 75% of the variation in LMB MeHg and Component 2 explained the remaining 6%. All predictors were relatively significant indicated by $VIP > 1.0$.

Predictor	Regression Coefficient for Component 1	Regression Coefficient for Component 2	VIP
Conductivity	-0.017	-0.019	1.69
THg Sediment	0.015	0.021	1.50
Longitude	0.015	0.012	1.49
Latitude	0.014	0.010	1.45
THg Soil	0.014	0.013	1.39
Forested area	0.013	0.010	1.37
MeHg water	0.010	0.025	1.31

Table 7. Observed and predicted mean MeHg concentrations in 350 mm largemouth bass. Predicted means were calculated using regression parameters estimated from the final model. Lakes are sorted by Bias (Predicted – Observed / Observed). Direction of Bias identifies if the predicted mean value was is higher (+), lower (-), or equal (=) to the observed mean concentrations.

Lake Number	Lake Name	Observed μg/g	Predicted μg/g	Bias	Direction of Bias
15	Perris Reservoir	0.10	0.10	0.00	=
13	Big Bear Lake	0.18	0.17	0.06	-
7	Lake Natomas	0.54	0.47	0.06	-
10	Lake McSwain	0.53	0.56	0.11	+
2	Lake Mendocino	0.54	0.60	0.13	+
17	Lake Elsinore	0.12	0.10	0.17	-
3	Lake Pillsbury	1.31	1.00	0.20	-
4	Lake San Antonio	0.30	0.36	0.21	+
6	Folsom Lake	0.47	0.57	0.24	+
9	Lake McClure	0.77	0.52	0.25	-
8	Don Pedro Reservoir	0.44	0.55	0.28	+
1	Lake Sonoma	0.68	0.87	0.32	+
14	Irvine Lake	0.48	0.30	0.38	-
11	San Luis Reservoir	0.56	0.33	0.41	-
12	O'Neill Forebay	0.23	0.37	0.61	+
16	Lake Hemet	0.06	0.11	0.83	+
5	Thermalito Afterbay	0.21	0.44	1.10	+

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Table 8. Land use variables for 21 lake catchments. These data were used for a qualitative assessment of the relationship between land use and 350 mm largemouth bass MeHg concentrations.

Station Name	Regional Board	350 mm Largemouth bass MeHg ($\mu\text{g/g}$)	Mines	THg-soil ($\mu\text{g/g}$)	Watershed Area (sq. miles)	Forested (%)	Wetland (%)
Almaden Lake	2	2.15	47	0.2	53.0	0	0.4
Calero Reservoir	2	1.05	0		6.9	0	0.7
Soulejoule Lake	2	0.94	1	0.0	18.8	0	0.8
Upper San Leandro Reservoir	2	1.01	0		30.6	0	1.0
Lower Crystal Springs Res.	2	0.85	0		15.0	0	1.5
Uvas Reservoir	3	0.91	0	0.2	30.5	0	0.4
Chesbro Reservoir	3	1.04	3		19.3	0	0.5
Echo Lake - Reg 4	4	0.08	0		1.2	0	0.1
Westlake Lake	4	0.09	0	0.0	28.1	0	0.4
Toluca Lake	4	0.00	30	0.4	422.7	5	0.0
Crystal Lake	4	0.95	0		1.0	95	0.5
New Melones Lake	5	1.12	823	0.0	904.2	78	1.2
Eastman Lake_BOG	5	1.04	59	0.0	235.0	80	0.2
Bass Lake	5	0.09	2	0.0	49.6	97	0.9
Little Rock Reservoir	6	0.92	2		63.8	21	0.1
Ferguson Lake_BOG	7	0.09	1	0.1	19.6	0	3.5
Prado Lake	8	0.07	0		27.1	0	0.1
Lake Evans	8	0.03	58	0.0	760.9	24	0.1
Lake Poway	9	0.05	0		2.4	0	0.2
Lake Wohlford	9	0.05	0		8.2	0	1.4
Dixon Lake	9	0.06	1		4.0	0	1.6

Appendix Table 1a. Sediment and water quality data for 17 California lakes and reservoirs.

Lake Number	Station Name	THg-sed (µg/g)	TOC (%)	Chlorophyll a (µg/L)	Sulphate (mg/L)	DOC (mg/L)	MeHg-water (ng/L)	THg-water (ng/L)	Temp (deg. C)	O2 (mg/L)	pH	Conductivity (mS)
1	Lake Sonoma	0.21	7.1	1.5	7.6	5.7	0.03	1.54	23.1	8.6	8.4	0.1
2	Lake Mendocino	0.07	5.4	1.1	8.8	6.8	0.03	0.80	22.6	8.7	8.3	14.7
3	Lake Pillsbury	0.20	5.4	0.9	6.3	8.4	0.15	1.25	14.9	10.1	6.2	34.0
4	Lake San Antonio	0.07	4.7	13.7	65.1	25.9	0.05	1.16	24.0	8.2	8.0	37.5
5	Thermalito Afterbay	0.01	2.2	1.5	7.0	3.2	0.02	0.51	16.8	9.0	5.3	51.0
6	Folsom Lake	0.13	7.5	1.6	3.5	2.9	0.07	0.38	23.5	7.8	6.4	52.0
7	Lake Natomas	0.06	2.6	1.0	3.5	3.1	0.03	0.72	24.3	8.8	7.5	58.5
8	Don Pedro Reservoir	0.13	9.8	1.6	3.6	2.1	0.01	0.28	25.1	7.6	8.2	63.1
9	Lake McClure	0.08	6.2	2.5	4.5	2.7	0.03	0.32	20.9	7.8	8.1	84.5
10	Lake McSwain	0.12	12.2	2.2	3.6	3.2	0.03	0.93	23.8	8.1	8.6	385.1
11	San Luis Reservoir	0.07	9.8	12.2	35.0	28.7	0.03	0.44	17.8	8.4	8.1	407.0
12	O'Neill Forebay	0.11	7.9	2.3	36.5	16.6	0.05	0.61	20.2	8.8	8.2	414.6
13	Big Bear Lake	0.07	15.3	6.5	33.5	17.5	0.01	0.53	20.3	8.5	7.9	579.0
14	Irvine Lake	0.17	7.9	7.8	181.3	98.7	0.18	1.25	20.5	10.6	8.2	624.0
15	Perris Reservoir	0.05	8.8	6.7	39.5	34.7	0.02	0.25	24.5	9.6	8.4	625.4
16	Lake Hemet	0.04	14.2	4.6	21.9	14.2	0.02	0.25	23.9	7.5	8.1	1104.4
17	Lake Elsinore	0.03	10.9	50.5	175.0	154.0	0.04	1.45	24.3	5.5	8.9	2933.4

Appendix Table 1b. Land use attributes for 17 California lakes and reservoirs

Lake Number	Station Name	Watershed Area (sq. miles)	Mines	Wetland (%)	THg-soil ($\mu\text{g/g}$)	Forested (%)
1	Lake Sonoma	130	2	0.0	0.1	100
2	Lake Mendocino	105	0	0.0	0.06	79
3	Lake Pillsbury	289	3	0.0	0.06	96
4	Lake San Antonio	323	1	1.2	0.05	30
5	Thermalito Afterbay	3639	1009	3.4	0.03	78
6	Folsom Lake	1863	2510	0.7	0.03	76
7	Lake Natomas	1904	2539	0.7	0.03	75
8	Don Pedro Reservoir	1535	701	1.6	0.07	59
9	Lake McClure	1038	892	1.1	0.07	76
10	Lake McSwain	1063	904	1.1	0.07	74
11	San Luis Reservoir	82	0	0.0	0.05	0
12	O'Neill Forebay	102	0	0.1	0.05	0
13	Big Bear Lake	73	46	0.7		41
14	Irvine Lake	63	2	0.0		15
15	Perris Reservoir	10	0	0.0		0
16	Lake Hemet	66	10	1.0	0	18
17	Lake Elsinore	771	51	0.2	0.02	6

Appendix Table 1c. Lake morphometry data for 17 California lakes and reservoirs

Lake Number	Station Name	Storage Capacity (acre-feet)	Max Depth (ft)	Surface Area (acres)	Watershed Area: Surface Area	Elevation (ft)	Age (yrs since 2009)	Annual Water Level Flux (ft)
1	Lake Sonoma	381	223	4.2	31	452	27	50
2	Lake Mendocino	122.4	133	3	35	720	51	30
3	Lake Pillsbury	80.5	60	3.5	82	1807	53	18
4	Lake San Antonio	335	180	8.9	36	780	44	20
5	Thermalito Afterbay	57.04	20	6.7	542	139	41	3
6	Folsom Lake	977	261	17.9	104	468	54	157
7	Lake Natomas	8.76	54	0.8	2257	129	54	3
8	Don Pedro Reservoir	2030	530	20.3	76	830	38	40
9	Lake McClure	1024.6	465	11.1	94	867	42	115
10	Lake McSwain	9.73	56	0.5	2208	399	42	15
11	San Luis Reservoir	2039	270	19.8	4	543	40	120
12	O'Neill Forebay	56.4	40	4.2	24	225	42	7
13	Big Bear Lake	73.37	72	4.6	16	6760	97	
14	Irvine Lake	28	47	1.1	58	794	78	16
15	Perris Reservoir	125	80	3.4	3	1567	36	10
16	Lake Hemet	8.1	135	0.7	101	4339	114	12
17	Lake Elsinore	30	17	5.2	149	1240	121	4

Appendix 2a. Regression coefficients and variance in projection (VIP) for Model 2

Predictor	Regression Coefficients for Component 2	VIP
Conductivity	-0.016	1.40
THg Sediment	0.020	1.33
MeHg Water	0.023	1.34
Longitude	0.012	1.22
Latitude	0.008	1.20
THg Soil	0.012	1.15
Forested area	0.009	1.13
Sulphate	-0.002	1.02
Chlorophyll a	-0.002	1.02
DOC	0.003	1.00
Annual Depth Change	0.012	0.93

Appendix 2b. Regression coefficients and variance in projection (VIP) for Model 3

Predictor	Regression Coefficients for Component 2	VIP
Conductivity	-0.016	1.44
THg Sediment	0.020	1.40
MeHg Water	0.025	1.40
Longitude	0.013	1.25
Latitude	0.009	1.22
THg Soil	0.014	1.18
Forested area	0.009	1.16
Chlorophyll a	-0.002	1.06
Sulphate	-0.002	1.04
DOC	0.003	1.02