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Factors influencing methylmercury contamination of black bass from California reservoirs[☆]



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ABSTRACT

Understanding how mercury (Hg) accumulates in the aquatic food web requires information on the factors driving methylmercury (MeHg) contamination. This paper employs data on MeHg in muscle tissue of three black bass species (Largemouth Bass, Spotted Bass, and Smallmouth Bass) sampled from 21 reservoirs in California. During a two-year period, reservoirs were sampled for total Hg in sediment, total Hg and MeHg in water, chlorophyll *a*, organic carbon, sulfate, dissolved oxygen, pH, conductivity, and temperature. These data, combined with land-use statistics and reservoir morphometry, were used to investigate relationships to size-normalized black bass MeHg concentrations. Significant correlations to black bass MeHg were observed for total Hg in sediment, total Hg and MeHg in surface water, and forested area. A multivariate statistical model predicted Largemouth Bass MeHg as a function of total Hg in sediment, MeHg in surface water, specific conductivity, total Hg in soils, and forested area. Comparison to historical reservoir sediment data suggested there has been no significant decline in sediment total Hg at five northern California reservoirs during the past 20 years. Overall, total Hg in sediment was indicated as the most influential factor associated with black bass MeHg contamination. The results of this study improve understanding of how MeHg varies in California reservoirs and the factors that correlate with fish MeHg contamination.

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1. Introduction

Historical gold, silver, and mercury (Hg) mining in the Coast and Sierra Nevada ranges of California have resulted in widespread Hg contamination. The most significant Hg source in California, by mass, is attributed to mining activity during the 1800s (Domagalski, 1998). Between 1850 and 1980, California was the leading producer of Hg (~100 million kg) in the continental United States (Churchill, 2000). Inefficient Hg mine processing and placer and lode gold mining resulted in an estimated loss of 41 million kg between the Coast and Sierra ranges (Churchill, 2000). As a result, inorganic Hg

concentrations in sediment and water in the Central Valley and Sacramento-San Joaquin Delta are elevated above background levels (Davis et al., 2008; Heim et al., 2007; Wiener et al., 2003).

On a national scale, atmospheric deposition is considered the predominant source of Hg to aquatic environments distant from mines (USEPA, 1997a). Hammerschmidt and Fitzgerald (2006) suggested that up to two-thirds of the Hg contamination in fish from water bodies in the United States could be attributed to wet deposition of atmospheric Hg. Mercury deposition to the eastern United States has been indicated in many studies as the primary vector for accumulation in the food web (e.g., Fitzgerald and Clarkson, 1991; Hammerschmidt and Fitzgerald, 2006). Despite limited data, atmospheric Hg deposition to the western coast of the U.S. is relatively low compared to other U.S. regions (Prestbo and Gay, 2009).

Mining, atmospheric deposition, and other sources predominantly release Hg to the aquatic environment in inorganic forms (USGS, 2000). When transformed to organic methylmercury

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(MeHg) by anaerobic bacteria (e.g., sulfate-reducing bacteria; Gilmour et al., 1992), Hg becomes a significant toxicological concern for biota. The primary pathways for increased methylation of inorganic Hg in lakes and reservoir systems are thought to relate to wetlands (St Louis et al., 1994), changes in water level (Sorensen et al., 2005), redox state (Whalin et al., 2007), forested landscape (St Louis et al., 1996), and sediments (Ramlal et al., 1993). Under certain conditions, Hg methylation may also occur in the water column (Lee and Iverfeldt, 1991). However, a direct correlation between water-column or sedimentary sources of Hg and biota MeHg concentrations has yet to be shown for California lakes and reservoirs.

Mercury contamination in California lakes and reservoirs has resulted in the posting of health advisories and interim health advisories for consumption of sports fish. These advisories recommend no consumption of large Striped Bass and limited consumption of other sport fish (Gassel et al., 2008). Results in a recent report to the California State Water Resources Control Board showed 68% of 272 lakes and reservoirs sampled had a fish species with an average MeHg concentration above $0.07 \text{ mg} \cdot \text{kg}^{-1}$ (threshold to consider recommending consumption of less than three servings per week; Davis et al., 2010). Twenty-one percent of the 272 locations were above $0.44 \text{ mg} \cdot \text{kg}^{-1}$, the threshold to consider recommending no consumption of contaminated species for women between 18 and 45 and children between 1 and 17 years of age.

The focus of this study was an exploratory analysis of factors related to the contamination of MeHg in black bass sampled from 21 reservoirs distributed throughout California. All of the study sites were located in reservoirs; no natural lakes were included. Black bass include the Largemouth Bass (*Micropterus salmoides*), Smallmouth Bass (*M. dolomieu*), and Spotted Bass (*M. punctulatus*), which are commonly present in many California inland waterbodies. Furthermore, Largemouth Bass are widely used as an bio-indicator of MeHg contamination throughout freshwaters of the United States (e.g., Barber, 2008; Chasar et al., 2009; Goodchild and Gerstenberger, 2011; Greenfield et al., 2009). Information from this study leads to a greater understanding of how MeHg varies in California reservoirs and the significant factors related to MeHg contamination in black bass species.

2. Materials & methods

2.1. Lake selection

This study investigated MeHg contamination in black bass from 21 reservoirs sampled during 2007–2008, and during follow-up sampling in 2009. Eleven (11) of the sites were located in the Sierra Nevada Mountain Range, five locations were in the California Coast Range, and five were in Southern California (Fig. 1).

2.2. Sample collection

Black bass (Largemouth Bass, Spotted Bass, Smallmouth Bass) were sampled from 19 reservoirs during a statewide survey of fish contamination conducted in 2007 and 2008 (Davis et al., 2010, 2009). Fifteen reservoirs were sampled for Largemouth Bass and two reservoirs were each sampled for Spotted Bass and Smallmouth Bass. To supplement this dataset for the current study, two reservoirs (Lake Hemet and Big Bear Lake) were sampled for Largemouth Bass in 2009. At each reservoir, fish were collected across a wide size range and analyzed as individuals to facilitate estimation of length-normalized wet-weight MeHg concentrations (Table 1). Dissection and compositing of muscle tissue filet samples were performed following USEPA guidance (USEPA, 2000a).

At each reservoir, surface water quality measurements and

surface water samples were collected from the limnetic zone monthly or twice monthly, depending on the season, from August 2008 to October 2009. Summer season sampling was performed during May–October, and winter season sampling during November–March. When reservoirs stratified during the summer, hypolimnion water samples were also collected from June to October 2009. A multi-parameter YSI 600 XL sonde was used to measure depth profiles (0–6 m) of temperature, conductivity, pH, and dissolved oxygen at each sampling event prior to water sample collection.

Near surface water was sampled, by hand, less than a meter below the surface of each reservoir. Hypolimnion water (>2 m depths) was only collected when reservoirs were stratified, using a pre-cleaned (5% HCl) 1.2-L Wildco Teflon Kemmerer sampler. Near-surface water samples were analyzed for total Hg, MeHg, chlorophyll *a*, sulfate (SO_4), and dissolved organic carbon (DOC). Hypolimnion water was analyzed for total Hg and MeHg only. Water samples collected for Hg analysis (total Hg or MeHg) were stored dark and acidified within 24 h of collection in I-Chem clear glass bottles (#229–0250). Total Hg samples bottles were acid washed prior to use and samples preserved with BrCl. Samples collected for MeHg were stored cold and preserved using HCl. All water samples were collected following EPA method 1669 (USEPA, 1996) for the collection of ambient water for trace metal analysis (clean hands/dirty hands technique).

Fine grain sediments were collected from all 21 reservoirs between the summer of 2009 and 2011. Samples were collected in the deepest depositional area of each reservoir using a Van Veen grab (0.5 m^2). The top 2 cm of sediment was collected from the grab using an acid cleaned Teflon scoop. Samples were stored in iChem 60 mL glass jars and frozen ($-20 \text{ }^\circ\text{C}$) until analysis. Sediment samples were not sieved.

Historical sediment Hg data from five reservoirs were used to assess changes over time in locations that overlapped with the current investigation (Camp Far West, Folsom Lake, Lake Engelbright, Lake Oroville, and Lake McClure). The historical sediment Hg data were collected and analyzed in 1987 by the Central Valley Regional Water Quality Control Board (CVRWQCB, 1987).

2.3. Mercury analysis

Davis et al. (2010) followed the USEPA assumption (USEPA, 2000a) that all Hg present in fish tissue occurs as MeHg, in order to be most protective of human health. Total Hg in fish tissue was determined by Moss Landing Marine Laboratories (MLML) using EPA method 7473 (USEPA, 2000b). Fish samples were dissected using standard trace metal clean procedures and analyzed on a Milestone Direct Mercury Analyzer (DMA-80). Detailed description of quality assurance protocols and results can be found in Davis et al. (2010, 2009), but generally followed the procedures described below for water and sediment analyses by the same analytical laboratory.

Water samples were analyzed for MeHg and total Hg using modified EPA method 1630 (USEPA, 1998) and method 1631 (USEPA, 2002), respectively. Total Hg in sediment was measured using an aqua regia digestion and aqueous-phase reduction with stannous chloride solution, followed by atomic absorbance detection using an automated PerkinElmer flow injection mercury system (Heim et al., 2003).

Analysis of Hg and MeHg was run in batches of 20 samples. Each batch had associated quality assurance/quality control samples: 3 method blanks, 1 reference material, 1 analytical native and duplicate pair, and 1 analytical spike and spike duplicate pair. No CRM for MeHg or total Hg in water is available. Therefore, a second known standard was used as a “second source” of Hg, and analyzed

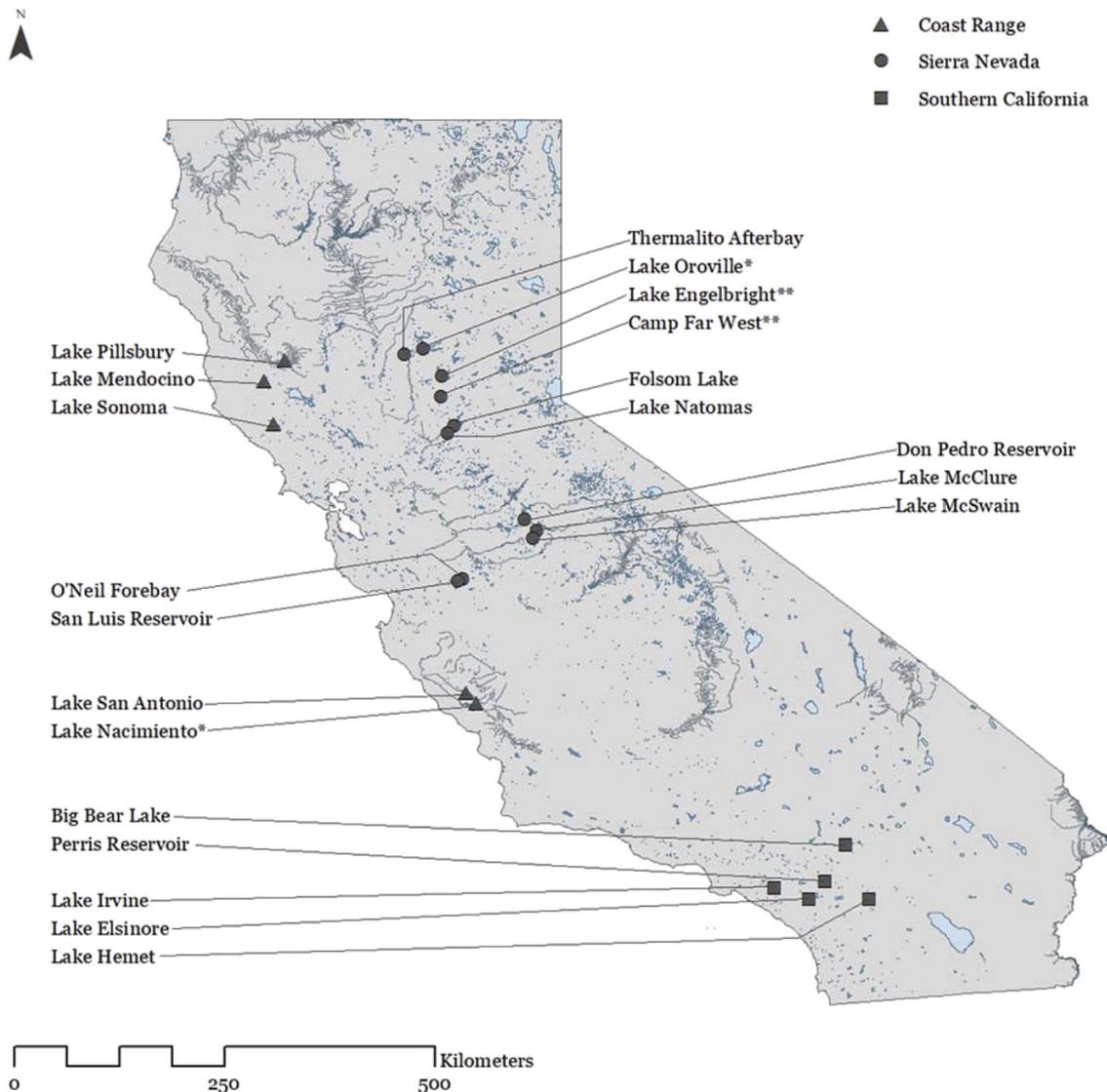


Fig. 1. Locations of the 21 California reservoirs sampled for Black Bass in this study. *Smallmouth Bass or **Spotted Bass only.

with the water samples to assess accuracy. All batches for Hg analysis passed quality control checks.

Field blanks and duplicates were taken for MeHg and total Hg a minimum of once per sampling event. All duplicate water samples analyzed for MeHg were within the data quality objective of 80–120% of native sample. All field blanks were below the method detection limit of $0.020 \text{ ng} \cdot \text{L}^{-1}$. All duplicate water samples analyzed for total Hg were within the data quality objective of 80–120% of native samples. None of the total Hg field blanks were above the method detection limit of $0.2 \text{ nng} \cdot \text{L}^{-1}$. All field duplicates ($n = 3$) for total Hg sediment were within the data quality objective of 80–120% native samples.

2.4. Chlorophyll *a*, sulfates, & organic carbon analysis

Chlorophyll *a* samples were analyzed at MLML using modified EPA method 445 (USEPA, 1997b). Sulfate (SO_4) and dissolved organic carbon (DOC) analysis were performed by Soil Control Laboratory located in Watsonville, Ca. using EPA method 300.0 (Pfaff, 1993) and method 415.1 (USEPA, 1979), respectively. Total organic carbon (TOC) in sediments was measured as percent loss on

ignition (LOI). Sediment samples were pre-dried at 37°C then placed in a glass 40 mL vial and weighed. Samples were heated in excess of 537°C for 24 h to volatilize and remove organic carbon. Samples were weighed again and a percent difference was calculated between the pre and post heat treatments for each sample.

All batches for chlorophyll *a*, SO_4 , DOC, and TOC analysis passed quality control checks for analysis, including CRM's, laboratory duplicates, and matrix spikes, where applicable. All laboratory blanks were non-detect, and the matrix spikes and SRM's met targets of 80–120%. Field blanks comprised ~10% and duplicate samples a minimum of 5% of all samples collected and analyzed. All of the duplicate chlorophyll *a* filters analyzed were within 80–120% of native samples and one field blank was above the reporting limit ($\text{RL} = 0.045 \text{ } \mu\text{g} \cdot \text{L}^{-1}$). All of the duplicate SO_4 water samples analyzed were within 80–120% of native samples and one field blank was above the reporting limit ($\text{RL} = 1.0 \text{ mg} \cdot \text{L}^{-1}$). Out of 17 duplicate samples analyzed for DOC, three fell outside 80–120% of native samples and one field blank was above the reporting limit ($\text{RL} = 1.0 \text{ mg} \cdot \text{L}^{-1}$). The two duplicate sediment samples analyzed for LOI and were within 80–120% of the native sample. Field duplicate samples that fell outside of the data quality objective

Table 1

Summary statistics of fish collection, average Hg, and size-normalized Hg concentrations in black bass from 21 California reservoirs.

Region	Station Name	n	Average Bass Hg (mg·kg ⁻¹)	Average Total Length (mm)	Size Range (mm)	Average Bass Hg Normalized to 350 mm (mg·kg ⁻¹)	Model R ²
Coast Range	Lake Mendocino	22	0.50 ± 0.05	335	191–485	0.54 ± 0.10	0.78
Coast Range	Lake Nacimiento**	33	1.00 ± 0.07	313	184–459	1.24 ± 0.25	0.65
Coast Range	Lake Pillsbury	22	1.47 ± 0.16	420	295–559	1.31 ± 0.42	0.69
Coast Range	Lake San Antonio	33	0.27 ± 0.02	336	224–492	0.30 ± 0.05	0.85
Coast Range	Lake Sonoma	22	0.61 ± 0.05	320	205–411	0.68 ± 0.15	0.50
S. California	Big Bear Lake	32	0.14 ± 0.02	328	235–519	0.18 ± 0.07	0.73
S. California	Lake Irvine	11	0.50 ± 0.09	365	300–440	0.48 ± 0.21	0.51
S. California	Lake Elsinore	22	0.05 ± 0.01	311	195–395	0.12 ± 0.02	0.49
S. California	Lake Hemet	11	0.06 ± 0.00	281	205–374	0.06 ± 0.02	0.01
S. California	Perris Reservoir	22	0.09 ± 0.01	332	202–466	0.10 ± 0.03	0.70
Sierra Nevada	Camp Far West*	21	0.65 ± 0.08	306	202–454	0.84 ± 0.16	0.81
Sierra Nevada	Don Pedro Reservoir	33	0.42 ± 0.03	344	201–539	0.44 ± 0.11	0.62
Sierra Nevada	Folsom Lake	33	0.64 ± 0.04	395	251–546	0.47 ± 0.15	0.63
Sierra Nevada	Lake Engelbright*	15	0.48 ± 0.21	335	224–469	0.52 ± 0.12	0.69
Sierra Nevada	Lake McClure	33	0.72 ± 0.04	326	221–533	0.77 ± 0.18	0.37
Sierra Nevada	Lake McSwain	9	0.67 ± 0.06	435	312–557	0.54 ± 0.12	0.65
Sierra Nevada	Lake Natomas	11	0.51 ± 0.08	345	200–450	0.54 ± 0.15	0.71
Sierra Nevada	Lake Oroville**	43	0.45 ± 0.03	320	151–529	0.51 ± 0.12	0.74
Sierra Nevada	O'Neill Forebay	22	0.20 ± 0.02	338	214–498	0.23 ± 0.07	0.56
Sierra Nevada	San Luis Reservoir	33	0.53 ± 0.04	325	207–442	0.56 ± 0.13	0.63
Sierra Nevada	Thermalito Afterbay	22	0.18 ± 0.03	342	229–514	0.21 ± 0.08	0.75

*Spotted Bass, **Smallmouth Bass.

(DQO) of 80–120% were below 10x the analytical MDL and were not thought to affect data quality. Samples were not blank corrected.

2.5. Reservoir morphometry & land use data

Data on reservoir morphometry (e.g., maximum depth, volume, surface area) were obtained from literature sources, websites, and water management agencies, including the California Department of Fish and Wildlife, Regional Water Quality Control Boards, and the California Department of Water Resources. Attributes representing characteristics of reservoir morphometry was obtained for all reservoirs where Largemouth Bass was collected (N = 17), and used in the multivariate statistical model (Table 2, Supplementary Table S1). A few attributes (e.g. maximum annual water depth) were not available for all reservoirs.

Land use statistics were calculated based on the reservoir catchment (sub-watershed) areas. Catchment areas were determined from the Watershed Boundary Dataset (WBD). Polygons from the “6th level” delineation (the smallest division of sub-watersheds within WBD) were used in the GIS layer. Sampling locations for each study basin were plotted, and the upstream sub-watershed area of each point was delineated. The newly generated catchment areas were checked against other GIS data for accuracy. These included the 4th & 5th level (more aggregated) of WBD, aerial photography, a 30 m hillshade, and the National Hydrography Dataset (NHD). In some cases, the WBD 6th level delineations were coincident with dams. In such cases, the polygons were edited to exclude dams that would represent barriers to fish and water quality changes. Once the watersheds were developed for each reservoir, land use data were overlaid to allow for calculation of attributes related to Hg contamination of the catchment

area, such as number of mines, proportion of wetland area, and total watershed area (Supplementary Fig. S1, Table 2, and Supplementary Table S1). In particular, three primary datasets were used to represent habitat and mining information: CalVeg, National Wetlands Inventory (NWI), and Mine Resources Data System (MRDS). The metadata behind each of these datasets have been described (Melwani et al., 2007).

2.6. Length-normalized black bass mercury concentrations

Largemouth Bass exhibit a strong size: MeHg relationship when collected over a wide (spanning 150 mm or more) size range (Davis et al., 2008; Melwani et al., 2009). Wet-weight MeHg concentrations in 350 mm Largemouth Bass was selected by Davis et al. (2010) as a typical site-specific estimate of MeHg. The 350 mm value represents the median of the typical size distribution above the legal limit of 305 mm (12 in) for Largemouth Bass in California. The 350 mm concentration of Largemouth Bass was estimated for all reservoirs by employing a general linear model with maximum likelihood (Littell et al., 1996), except the two sampled for Largemouth Bass in 2009 and four sampled for other black bass species (discussed below). The approach was used to evaluate the “best” regression model to estimate MeHg concentrations in Largemouth Bass for those locations. Davis et al. (2010) provides further details of the general linear model.

To supplement the dataset with estimates for an additional six reservoirs, a simple linear regression approach was used to estimate size-normalized concentrations in Largemouth Bass (Big Bear Lake and Lake Hemet), Spotted Bass (Camp Far West and Lake Engelbright), and Smallmouth Bass (Lake Nacimiento and Lake Oroville). Consistent with Davis et al. (2010), the regression

Table 2
Description & summary statistics of variables evaluated in the study.

Variable	Unit	n	Mean	Median	SD	Min	Max
length normalized bass (350 mm)	mg·kg ⁻¹	21	0.51	0.51	0.33	0.06	1.31
<i>entire study period</i>							
total Hg in sediment, top 2 cm	ng·g ⁻¹	21	194	81	338	31	1569
total organic carbon in lake sediment	%	21	8.2	8.1	3.3	2.2	15
total Hg in surface water	ng·L ⁻¹	21	1.21	0.850	1.62	0.253	7.86
MeHg in surface water	ng·L ⁻¹	21	0.05	0.03	0.04	0.01	0.18
total Hg in hypolimnion	ng·L ⁻¹	18 ^a	1.41	0.87	1.86	0.30	8.55
MeHg in hypolimnion	ng·L ⁻¹	21	0.07	0.03	0.09	0.01	0.35
chlorophyll <i>a</i> in surface water	µg·L ⁻¹	21	4.79	1.91	9.01	0.44	42.6
sulfate in surface water	mg·L ⁻¹	21	37.8	7.85	65.7	2.10	232
dissolved organic carbon in surface water	mg·L ⁻¹	21	11.7	9.45	15.6	1.47	65.3
temperature of surface water	C	21	16.8	16.5	3.0	11.5	22.0
dissolved oxygen of surface water	mg·L ⁻¹	21	9.16	9.29	1.05	7.32	11.2
pH of surface water	–	21	7.65	7.95	0.823	5.95	8.81
specific conductivity of surface water	µS·cm ⁻²	21	404	160	617	37.0	2812
<i>summer conditions (May–October)</i>							
total Hg in surface water	ng·L ⁻¹	21	1.15	0.61	1.78	0.06	8.53
MeHg in surface water	ng·L ⁻¹	21	0.05	0.03	0.05	0.01	0.18
total Hg in hypolimnion/deep water	ng·L ⁻¹	18 ^a	1.51	0.73	2.76	0.30	12.4
MeHg in hypolimnion/deep water	ng·L ⁻¹	21	0.11	0.03	0.19	0.01	0.71
chlorophyll <i>a</i> in surface water	µg·L ⁻¹	21	6.12	2.25	10.8	0.04	50.5
sulfate in surface water	mg·L ⁻¹	21	33.1	7.80	51.2	3.45	181
dissolved organic carbon in surface water	mg·L ⁻¹	21	22.0	6.78	37.1	2.05	154
temperature of surface water	C	21	21.4	23.0	3.35	13.9	25.1
dissolved oxygen of surface water	mg·L ⁻¹	21	8.38	8.24	1.09	5.58	9.95
pH of surface water (<6 m)	–	21	7.67	8.01	1.08	5.58	9.95
specific conductivity of surface water	µS·cm ⁻²	21	408	164	646	32	2955
<i>winter conditions (November–April)</i>							
total Hg in surface water	ng·L ⁻¹	18 ^b	0.97	0.88	0.71	0.25	2.85
MeHg in surface water	ng·L ⁻¹	20 ^c	0.03	0.03	0.03	0.01	0.13
total Hg in hypolimnion/deep water	ng·L ⁻¹	7 ^d	1.84	1.1	1.67	0.44	4.71
MeHg in hypolimnion/deep water	ng·L ⁻¹	19 ^e	0.04	0.03	0.03	0.01	0.15
chlorophyll <i>a</i> in surface water	µg·L ⁻¹	20 ^c	3.82	1.59	7.92	0.46	36.7
sulfate in surface water	mg·L ⁻¹	20 ^c	42.7	7.72	76.5	1.3	270
dissolved organic carbon in surface water	mg·L ⁻¹	20 ^c	5.15	3.09	5.12	0.97	21.0
temperature of surface water	C	20 ^c	13.2	13.2	2.99	7.13	18.7
dissolved oxygen of surface water	mg·L ⁻¹	20 ^c	10.1	9.92	1.53	7.99	12.9
pH of surface water (<6 m)	–	20 ^c	7.76	7.81	0.60	6.43	8.90
specific conductivity of surface water	µS·cm ⁻²	20 ^c	411	175	603	35	2676
<i>watershed characteristics</i>							
total Hg content of soil	µg·g ⁻¹	14 ^f	0.05	0.05	0.03	0.01	0.1
forested area	%	17	48	59	36	0	100
wetland area	%	17	0.7	0.69	0.89	0	3.4
watershed area	km ²	17	1989	748	2567	26	9426
mines	count	17	510	10	846	0	2539
<i>lake morphometry and location</i>							
max volume of lake water storage	m ³	17	535904	99295	837839	9991	2515066
maximum depth of lake	m	17	47	24	46	5	162
surface area of lake	km ²	17	12.9	7.2	16.0	0.4	62.1
elevation of lake	m	17	415	240	454	39	2060
max annual depth change of lake	m	16 ^g	12	5	15	1	49
age of lake (in 2010)	yr	17	57	44	28	27	121
latitude	dms	17	36.8	37.5	2.2	33.7	39.5
longitude	dms	17	–120.2	–121.0	2.2	–123.2	–116.7

^a Lake Pillsbury, Lake Mendocino, and Lake Sonoma not sampled.

^b Lake Pillsbury, Lake Nacimiento, and Lake San Antonio not sampled.

^c Lake Pillsbury not sampled.

^d Lake Pillsbury, Lake Mendocino, Lake Sonoma, Lake Nacimiento, Lake San Antonio, Thermalito Afterbay, Don Pedro Reservoir, Lake McSwain, Lake McClure, Big Bear Lake, Lake Irvine, Lake Elsinore, Lake Hemet, and Perris Reservoir not sampled.

^e Lake Pillsbury and Thermalito Afterbay not sampled.

^f Attribute not available for Big Bear Lake, Irvine Lake, Perris Reservoir.

^g Attribute not available for Big Bear Lake.

equations were used to estimate MeHg concentrations (mean and standard error) for each location at 350 mm total length. Summary statistics of all fish MeHg data are provided in [Table 1](#).

2.7. Statistical analysis

Pearson's correlation analysis was used to evaluate associations among environmental variables and tissue MeHg concentrations

using data from all three black species combined. Data were pooled across species based on preliminary analysis that revealed no significant difference in the results with the four reservoirs sampled for Spotted Bass and Smallmouth Bass excluded. Prior to correlation analysis, data were log-10 transformed to improve linear relationships, and ensure normally distributed values and equal variances. Water chemistry data were evaluated both by season (winter vs. summer) and grouped across seasons. Correlations were performed

using SPSS for Windows version 19.0 (SPSS Inc., Chicago, IL, USA). Due to the large number of variables evaluated, a more stringent p-value of 0.01 was selected for evaluation of statistical significance.

Subsequently, Partial Least Square regression models (also called Projection of Latent Structures or PLS) were used to assess the influence of 23 explanatory variables on 350 mm Largemouth Bass MeHg concentrations. Here, only the optimal seasonal combination of water chemistry variables (i.e. winter, summer, or average of both combined) was selected for each model iteration. Morphometric and land use data were not available for reservoirs where Largemouth Bass was not sampled, and thus those four reservoirs were excluded from the model. PLS analysis was conducted with predictor variables that were log-10 transformed and standardized (i.e., mean centered and scaled to the standard deviation).

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

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

In subsequent PLS model runs, variables were removed based on their regression coefficients, VIP, and relative contribution to the model. Although no defined limit exists for statistical significance in PLS models, VIP limits of 0.8 or 1.0 have often been used (Eriksson et al., 1995; Sonesten, 2003). In this study, significant predictor variables were defined as having a VIP >1.0. Predictors with VIP <1.0 were removed in the final model. Predictive ability of the different PLS models was estimated by cross-validation (Eriksson et al., 1995). The PLS models were developed using the R Statistical Software Package (<http://cran.r-project.org>).

3. Results

Length-normalized black bass (Largemouth, Smallmouth, and Spotted Bass) MeHg concentrations for 21 reservoirs in California ranged from 0.06 to 1.31 mg·kg⁻¹ (wet weight). Mean estimates of black bass MeHg in reservoirs of the Coast Range (N=5; 0.30–1.31 mg·kg⁻¹) and Sierra Nevada (N=11; 0.21–0.84 mg·kg⁻¹) were generally higher than for locations in southern California (N=5; 0.06–0.48 mg·kg⁻¹). The four reservoirs (Lake Oroville, Lake Nacimiento, Lake Engelbright, and Camp Far West Reservoir) sampled for Smallmouth Bass (0.51 and 1.24 mg·kg⁻¹) and Spotted Bass (0.52 and 0.84 mg·kg⁻¹), exhibited MeHg concentrations that were similar to or higher than the median estimate for Largemouth Bass MeHg (N=19; 0.47 mg·kg⁻¹).

To evaluate the influence of factors that may be associated with black bass MeHg concentrations, three sets of analyses are described below. Section 3.1 presents the best correlations among 22 variables using data from all reservoirs and black bass species combined. Section 3.2 presents a statistical model for reservoirs where Largemouth Bass MeHg was predicted using an optimized set of variables. Lastly, Section 3.3 compares the total Hg sediment concentrations at five reservoirs sampled in this study to the same locations sampled in 1987.

3.1. Significant factors correlated with MeHg in black bass

Twenty-two ecosystem factors, including sediment total Hg, surface/hypolimnion MeHg, chlorophyll *a*, organic carbon, and several reservoir characteristics were evaluated for correlation to black bass MeHg concentrations (Table 3), and amongst each other. With respect to black bass MeHg, five variables had a Pearson *r* coefficient >0.5 and p-value < 0.01; total Hg in sediment (*r* = 0.64, *p* = 0.002; Fig. 2), surface water total Hg during summer (*r* = 0.55, *p* = 0.009), surface water total Hg across seasons (*r* = 0.59, *p* = 0.005), surface water MeHg across seasons (*r* = 0.62, 0.002), and forested area (*r* = 0.63, *p* = 0.007). All five factors had similar degree of correlation (*r* = 0.55–0.64).

Sediment total Hg concentrations were significantly correlated with surface water and soil Hg concentrations. The highest correlation of sediment total Hg to surface water total Hg occurred during summer (*r* = 0.88; *p* < 0.001), while the highest correlation to surface water MeHg was evident during winter (*r* = 0.89;

Table 3

Pearson correlation coefficients between 350 mm length normalized black bass Hg concentrations and environmental variables from 21 reservoirs in California. **p* < 0.05; ***p* < 0.01

Variable	Length-normalized black bass	
	Pearson <i>r</i>	Significance
<i>entire study period</i>		
total Hg in lake sediment	0.64	**
total organic carbon in lake sediment (%)	-0.20	n.s.
total Hg in surface water	0.59	**
MeHg in surface water	0.62	**
chlorophyll <i>a</i> in surface water	-0.42	*
sulfate in surface water	-0.33	n.s.
dissolved organic carbon in surface water	-0.25	n.s.
temperature	0.26	n.s.
dissolved oxygen	-0.10	n.s.
pH	-0.23	n.s.
specific conductivity	-0.40	n.s.
<i>summer lake conditions (May-October)</i>		
total Hg in surface water	0.55	**
MeHg in surface water	0.49	*
chlorophyll <i>a</i> in surface water	-0.40	n.s.
sulfate in surface water	-0.38	n.s.
dissolved organic carbon in surface water	-0.31	n.s.
temperature	0.15	n.s.
dissolved oxygen	0.03	n.s.
pH	-0.07	n.s.
specific conductivity	-0.37	n.s.
<i>winter lake conditions (November-April)</i>		
total Hg in surface water	0.52	*
MeHg in surface water	0.48	*
chlorophyll <i>a</i> in surface water	-0.52	*
sulfate in surface water	-0.27	n.s.
dissolved organic carbon in surface water	-0.29	n.s.
temperature	0.48	*
dissolved oxygen	0.02	n.s.
pH	-0.36	n.s.
specific conductivity	-0.44	n.s.
<i>watershed characteristics</i>		
total Hg content of soil	0.53	*
forested area (%)	0.63	**
wetland area (%)	0.24	n.s.
watershed area	0.24	n.s.
mines	0.06	n.s.
<i>lake morphometry and location</i>		
max volume of lake water storage	0.29	n.s.
maximum depth of lake	0.32	n.s.
surface area of lake	0.10	n.s.
elevation of lake	-0.20	n.s.
max annual depth change of lake	0.46	n.s.
age of lake (in 2010)	-0.40	n.s.

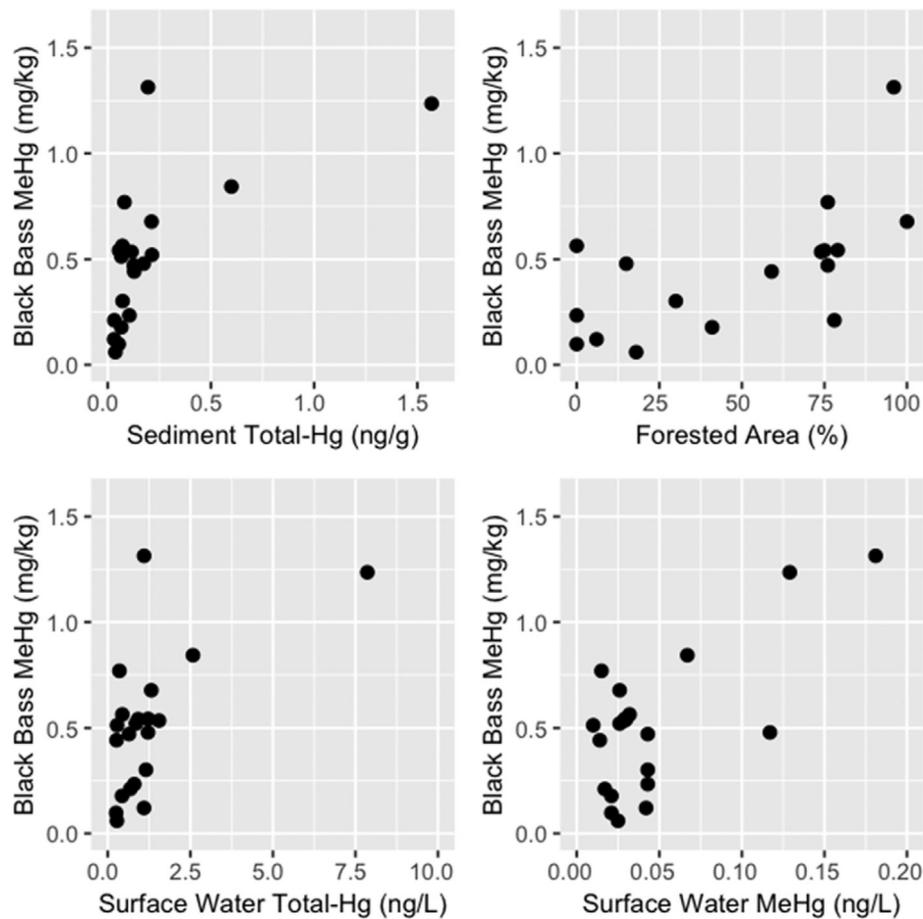


Fig. 2. X–Y scatter plot between sediment total Hg, forested area, surface water total Hg, and surface water MeHg, compared to black bass MeHg at 21 reservoirs in California. Data are untransformed.

$p < 0.001$). Total Hg in sediment was also correlated with average total Hg concentration in soil for the watershed ($r = 0.71$; $p = 0.006$).

Surface water total Hg and MeHg were significantly correlated between seasons, but less in the same season. The highest Pearson r coefficient ($r = 0.91$, $p < 0.001$) was observed between surface water total Hg during summer and surface water MeHg during winter. In contrast, correlations between total Hg and MeHg during summer ($r = 0.49$, $p = 0.026$) and winter ($r = 0.56$, $p = 0.015$) were less evident.

Of the seven general water quality variables (chlorophyll a , DOC, SO_4 , pH, temperature, specific conductivity, and dissolved oxygen) investigated for correlation to black bass MeHg, only surface water chlorophyll a during winter exhibited a Pearson $r > 0.5$ ($r = -0.52$, $p = 0.020$). Regardless of season, chlorophyll a was also highly correlated with pH ($r = 0.60$, $p = 0.005$), specific conductivity ($r = 0.80$; $p < 0.001$), sulfate ($r = 0.79$, $p < 0.001$), and DOC ($r = 0.81$, $p < 0.001$), but did not correlate with any of the surface water total Hg or MeHg variables.

Lastly, forested area (as % of the watershed) was the only land use variable that was significantly correlated with black bass MeHg, while none of the reservoir morphometric variables were. Forested area was also a significantly correlated with several of the water chemistry variables. Specifically, reservoirs with a higher proportion of forested area, had lower amounts of chlorophyll a , sulfate, and DOC. Neither surface water or hypolimnion total Hg or MeHg correlated with forested area, however.

3.2. Multivariate regression model

The influence of 22 ecosystem variables on Largemouth Bass MeHg from 17 of the study reservoirs was evaluated using several PLS models. The purpose was to evaluate multivariate relationships, as opposed to individual correlations, between explanatory variables (predictor) and Largemouth Bass MeHg (response). All four of the PLS model runs were generally consistent regarding the significant environmental factors explaining Largemouth Bass MeHg concentrations (Table 4 and Supplementary Table S3). The variables included in the final model were all relatively significant ($VIP > 1.0$), and contributed variance to the model (similar regression coefficients; Table 4).

The final model equation was:

$$\begin{aligned} \text{Log}_{10}(\text{Largemouth Bass MeHg}) = & 0.15 + 0.010(\text{Lat}) + 0.012(\text{Lon}) + 0.021(\text{Hgt-Sed}) \\ & + 0.025(\text{MeHg-Surf-Water}) + 0.013(\text{Hgt-Soil}) \\ & + 0.010(\text{Forested area}) - 0.019(\text{Sp. Cond}) \end{aligned}$$

Results from the final model generally agreed with interpretations from the correlation structure, with several of the Hg variables comprising the model terms. The seven predictor variables in the final model explained 81% of the variance in Largemouth Bass MeHg. Exclusion of latitude and longitude from the final model reduced the model variance by approximately 5%. Therefore, despite latitude and longitude having relatively strong influence in the final model (indicated by 3rd and 4th highest VIP),

Table 4

Regression coefficients and variance in projection (VIP) for the final PLS model variables to predict length-normalized Largemouth Bass MeHg for 17 reservoirs in California.

Variable	Regression Coefficients		
	Component 1	Component 2	VIP
<i>entire study period</i>			
total Hg in sediment	0.015	0.021	1.69
specific conductivity	−0.017	−0.019	1.69
<i>summer conditions</i>			
MeHg in surface water	0.01	0.025	1.31
<i>watershed characteristics</i>			
total Hg content of soil	0.014	0.013	1.39
forested area	0.013	0.010	1.37
<i>lake location</i>			
latitude	0.014	0.010	1.45
longitude	0.015	0.012	1.49

the variables did not contribute much variance to the prediction of Largemouth Bass MeHg. This suggests that the vast majority of the variation due to spatial or regional differences was captured by other predictor variables.

Cross-validation was used to evaluate the predictive ability and stability of the PLS models. A final model R^2 of 0.81 and a cross-validated R^2 of 0.75 was the highest prediction R^2 of the models. The majority of mean predicted values of Largemouth Bass MeHg were within $0.25 \text{ mg} \cdot \text{kg}^{-1}$ of the observed mean concentration (Table 5). The majority of predicted values were higher than the observed concentration, suggesting that the model estimates are more conservative than the observed values. The least deviation from observed was evident at reservoirs in the low to moderate concentrations range ($0.1\text{--}0.6 \text{ mg} \cdot \text{kg}^{-1}$). This is likely because the majority of reservoirs used in the model corresponded to this range in concentration.

Overall, the majority of predicted values were reasonable given the limited sample size of reservoirs and number of variables employed in the final model. Of the variables included, land use may be the source of the highest degree of bias. Each of the reservoirs with the highest degree of bias (Thermalito Afterbay, Lake Hemet, and O'Neil Forebay) were outliers because their land use characteristics were anomalous. Thermalito Afterbay was biased by 1.1 times the observed concentrations because it had relatively high proportion of forested area (78%) in its catchment, but the observed concentration was not proportionally high. Similarly, Lake Hemet

was biased by 0.83 times the observed concentrations because it had a relatively low observed Largemouth Bass MeHg, but not the lowest values of total Hg in sediment. Therefore, this reservoir was predicted to have a higher Largemouth Bass MeHg than was observed. Another potential reason behind Thermalito Afterbay and O'Neil Forebay being outliers is the occurrence of frequent water transfers and water level fluctuations that were not accounted for by variables in the statistical model.

3.3. Historical reservoir sediment Total Hg

A supplementary analysis was conducted to compare historical reservoir sediment total Hg to the current study. There was no significant difference in total Hg in sediments from five reservoirs sampled in the current study relative to previous reservoir sediment total Hg from 1987. Total Hg at Camp Far West, Folsom Lake, Lake Engelbright, Lake Oroville, and Lake McClure sampled in 2009 exhibited an average concentration of $0.22 \pm 0.22 \text{ mg} \cdot \text{kg}^{-1}$, compared to sediments collected in 1987 that had a similar average of $0.21 \pm 0.25 \text{ mg} \cdot \text{kg}^{-1}$. Unsurprisingly, the paired *t*-test was not significantly different ($t_{2,4} = 0.63$, $p = 0.56$). These results suggest that total Hg of reservoir sediments has not changed significantly over the past two decades.

4. Discussion

4.1. Total Hg in sediment and catchment soils

In two-thirds of U.S. inland waters, the accumulation of MeHg in fish populations has been linked to atmospheric Hg (natural and anthropogenic; Hammerschmidt and Fitzgerald, 2006). Moreover, background atmospheric fluxes of Hg in California are greater than what is typically seen worldwide (Drevnick et al., 2010; Sanders et al., 2008). However, the majority of reservoirs included in the current study are in regions where past mining activities occurred or are areas rich in naturally occurring deposits of Hg (cinnabar). These reservoirs can act as "traps" for Hg contaminated sediments and soils washed down from the surrounding watershed (Minear and Kondolf, 2009).

Declines in reservoir sediment total Hg concentrations from the 1980s to present have been associated with controls on atmospheric Hg emissions across western North America (Drevnick et al., 2016, 2012; Marvin et al., 2004). However, despite having

Table 5

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 higher (+), lower (−), or equal (=) to the observed mean concentrations.

Lake Name	Observed $\text{mg} \cdot \text{kg}^{-1}$	Predicted $\text{mg} \cdot \text{kg}^{-1}$	Bias	Direction of Bias
Perris Reservoir	0.10	0.10	0.00	=
Big Bear Lake	0.18	0.17	0.06	−
Lake Natomas	0.54	0.47	0.06	−
Lake McSwain	0.53	0.56	0.11	+
Lake Mendocino	0.54	0.60	0.13	+
Lake Elsinore	0.12	0.10	0.17	−
Lake Pillsbury	1.31	1.00	0.20	−
Lake San Antonio	0.30	0.36	0.21	+
Folsom Lake	0.47	0.57	0.24	+
Lake McClure	0.77	0.52	0.25	−
Don Pedro Reservoir	0.44	0.55	0.28	+
Lake Sonoma	0.68	0.87	0.32	+
Irvine Lake	0.48	0.30	0.38	−
San Luis Reservoir	0.56	0.33	0.41	−
O'Neill Forebay	0.23	0.37	0.61	+
Lake Hemet	0.06	0.11	0.83	+
Thermalito Afterbay	0.21	0.44	1.10	+

the same Federal regulations on emissions and numerous controls on discharges at the state level (e.g., POTWs), there appears to be little change in sediment Hg concentrations in California reservoirs. One potential hypothesis for the lack of trend in sediment total Hg is the influence of soil erosion. Rose et al. (2012) found catchment soil erosion is the main transfer mechanism of total Hg to Scottish lakes. This, in turn, counteracts reductions in atmospheric deposition of Hg. In the current study, we found total Hg in catchment soils and reservoirs sediment to be important factors associated with higher black bass MeHg concentrations. Catchment soil Hg was correlated with total Hg in reservoir sediments as well, suggesting total Hg concentrations in these reservoirs are influenced by the surrounding watershed soils, and thus reductions in atmospheric sources may have potentially been negated.

4.2. Landscapes influences

Landscape and habitat use may also be critical to understanding higher Hg concentrations in bass at some of the study locations. Coast Range (e.g., Lake Sonoma and Lake Mendocino) and Sierra Nevada reservoirs (e.g., Folsom Lake, Lake Natomas, Don Pedro Reservoir, Lake McSwain, and Lake McClure) were associated with catchments with higher black bass MeHg, relatively high (>70%) forested area and total Hg in catchment soils. Previous studies of habitat influences on fish MeHg concentrations in lake and reservoir catchments have found significant correlations with land cover and habitat features of the watershed (e.g., Alpers et al., 2016; Chumchal et al., 2008; Hurley et al., 1995; Wiener et al., 2006). Furthermore, differences in Hg methylation efficiency have been reported on land use scales (e.g., agricultural, industrial, urban; Krabbenhoft et al., 1999), as well as habitat scales (e.g., wetland, forested, un-vegetated; Hurley et al., 1995; Wiener et al., 2006). Terrestrial soils have been estimated to contain more than 90% of Hg stored in aquatic environments (Corbitt et al., 2011).

New Melones, Crystal Lake, and Eastman Lake had black bass MeHg concentrations between 0.95 and 1.12 mg·kg⁻¹, and >75% forested area in their watersheds. However, in southern California, where catchments have relatively little forested area, no obvious pattern could be deduced, with some reservoirs having relatively high (>0.6 mg·kg⁻¹) or low (<0.3 mg·kg⁻¹) black bass MeHg concentrations. In addition, patterns of wetland area associated with elevated black bass MeHg were inconclusive. Overall, the results suggest a hypothesis for at least some reservoirs, where Hg may be leached more readily from forested areas than in catchments dominated by other habitat types, such as wetlands. However, the potential sources of Hg to these forested catchments still remains an open question, due to the lack of correlation to mine data, and the inability to examine atmospheric or other watershed-scale sources (e.g., POTWs) in the analysis.

Many types of habitat can exhibit conditions that are favorable for Hg methylation (e.g., Ackerman and Eagles-Smith, 2010; Eagles-Smith et al., 2016b; Zillioux et al., 1993). A recent study of 21 high-elevation lakes in the Pacific Northwest, Eagles-Smith et al. (2016a) found conifer density and several ecosystem variables, including lake size, SO₄, and DOC were significantly correlated with fish total Hg concentrations. Integrating Hg data from across the Western United States, Obrist et al. (2016) found that plant-derived Hg deposition was the predominant pathway for soil Hg distribution across the region, and that soil Hg concentrations were generally highest in forested ecoregions. Another relevant area of research has been to understand the activity of sulfur-reducing bacteria, which thrive in anoxic soils and sediment (Gilmour et al., 1992; Gilmour and Henry, 1991). Forested areas have been observed to transport the more reactive forms of Hg relative to other land use types (Chumchal et al., 2008), and sulfur-reducing bacteria are

primarily responsible for this transformation. Accordingly, positive correlations between forested area and MeHg concentration in water and bass have been observed previously (Chumchal et al., 2008; Hurley et al., 1995; St Louis et al., 1994). Watershed runoff has been indicated as a primary transfer pathway for Hg between waterbodies and connected forested watersheds (Kwon et al., 2015).

Reservoirs included in this study had relatively high black bass MeHg that coincided with watersheds with very few mines, as well as some with an abundance of mines. The lack of consistent correlation to number of gold and Hg mines suggests that other variables could be responsible for driving elevated black bass MeHg at the some of the reservoirs. For example, it has been suggested that total Hg in sediments and soil from mine sites are not as bioavailable as those from diffuse sources, such as atmospheric deposition or urban runoff (Krabbenhoft et al., 1999). Or it could be that some reservoir mine areas are simply not eroding Hg-contaminated sediments anymore. Some mines may have already washed the majority of their mined sediments downstream into rivers, which have subsequently been buried by cleaner material. Notably, Alpers et al. (2016) was successful in developing a statistical model of MeHg in fish associated with Sierra Nevada streams downstream of historic gold mining areas, thus supporting this hypothesis. One way to evaluate if mines continue to erode and affect the surrounding landscape would be to assess background reservoir sediment Hg concentrations through sediment coring.

4.3. Methylmercury in water

Total Hg and MeHg in surface waters of lakes and reservoirs are known to affect fish MeHg concentrations (Eagles-Smith et al., 2016b; Wiener et al., 2006). In two study reservoirs that had the highest black bass MeHg concentrations, Lake Pillsbury and Lake Nacimiento, surface water MeHg concentrations were the highest of all study locations. This suggest either water column methylation or increased methylation in bottom sediments as a result of anoxia (Eckley et al., 2005). MeHg contamination in fish has previously been attributed to MeHg in bottom water in other California reservoir areas, such as Davis Creek Reservoir (Slotton et al., 1995) and reservoirs in the Guadalupe Watershed near San Jose, California. There were a few reservoirs in this study (Lake San Antonio, Lake Hemet, and Lake Irvine) which had high hypolimnion MeHg concentrations during summer months and were consistently stratified suggesting, in these reservoirs, the source of MeHg is the hypolimnion.

Redox conditions associated with hypolimnetic water is often associated with enhancing sediment and/or water column methylation (Brito et al., 2017; Eckley and Hintelmann, 2006; Regnell et al., 2001, 1997) and fall turnover of reservoir waters can be a vector for MeHg to surface waters leading to increases in fish MeHg concentrations (Herrin et al., 1998; Slotton et al., 1995). Total Hg in reservoir sediment was significant and positively correlated to both surface water total Hg and MeHg in this study, suggesting the degree of MeHg exposure is related, in some part, to the quantity of total Hg present in reservoir sediments and transport into the water-column. McCord et al. (2016) found that deep water oxygenation in a California reservoir reduced hypolimnetic MeHg concentrations at the site of injection, which further supports the hypothesis that many of California reservoirs are subject to a bottom up process of MeHg exposure to biota. However, further evidence is needed to elucidate the exact role of stratification and hypolimnetic anoxia. We hypothesize that the hypolimnion may play a role in MeHg transport of some California reservoirs. The limited frequency of water sampling collected in this study makes it difficult to discern the long-term effect oxygen or thermal

stratification may have on MeHg production or sequestering Hg in surface or hypolimnion water. However, an anecdotal relationship showing a significant relationship between extent of oxycline (Delta-O2) and surface water MeHg in Sierra Nevada reservoirs ($R^2 = 0.68$; Supplementary Fig. S2) was observed in this study. It suggests that decreasing anoxia in the hypolimnion of reservoirs with substantial summer stratification (higher Delta-O2) could lead to lower surface water MeHg. This could be achieved through oxygenation of the hypolimnion and stimulation of primary production. This may also serve to reduce overall MeHg concentrations in fish via biodilution (a process that lowers fish MeHg concentrations when primary production is elevated). Based on limited observations from this study, moderation efforts may only need to be occur during the summer, prior to fall turnover.

Spikes in MeHg uptake have been shown to coincide with fall turnover (Slotton et al., 1995). Hypolimnion MeHg had the most positive correlations with near surface water variables during the winter (November–April) including total and MeHg and a negative correlation with chlorophyll *a*. This most likely is the result of late fall turnover when reservoirs became well mixed. This was also when chlorophyll *a* had the highest correlation to black bass MeHg, thus indicating it may have the strongest negative effect on uptake of MeHg during this period. Studies have found algal blooms reduce the uptake of MeHg in freshwater food webs (Pickhardt et al., 2002). It has also been shown to be negatively correlated to bass species and clams in the Sacramento-San Joaquin Bay Delta (Chris Foe, pers. comm.). Lake San Antonio had some of the highest levels of chlorophyll *a*, which may be acting to reduce the uptake of MeHg in this reservoir via biodilution.

4.4. Chlorophyll *a* & specific conductivity

Environmental processes occurring within reservoir waters can drive the abundance and bioavailability of MeHg (Wiener et al., 2006), and there are several physical, chemical, and ecological factors that control the potential for food web contamination. Lake characteristics and water chemistry has previous been shown to explain 87% of variation in fish MeHg concentrations in sub-alpine lakes (Eagles-Smith et al., 2016a). Here, we found that reservoirs that were high in chlorophyll *a* had lower bass MeHg concentrations. For example, Lake San Antonio had elevated total and MeHg in surface water but relatively low bass MeHg concentrations ($0.30 \text{ mg} \cdot \text{kg}^{-1}$). Increasing levels of algae (thereby chlorophyll *a*) can reduce MeHg concentrations in zooplankton 2–3 fold which decreases MeHg in higher trophic consumers (Pickhardt et al., 2005, 2002).

Specific conductivity can be considered an indication of reservoir acid neutralizing capability (Sorensen et al., 1990). An increase in pH as a result of acid neutralizing results in a lower potential for MeHg formation (Gilmour and Henry, 1991). MeHg in fish tissue has often been found to have a negative correlation with conductivity, hardness, and alkalinity (Hanten et al., 1998; Sorensen et al., 1990; Wren et al., 1991). Enhanced microbial production of MeHg has also been shown under low pH and hardness conditions (Xun et al., 1987). Therefore, it is plausible that the influence of conductivity on Largemouth Bass MeHg predictions relates to an underlying relationship to Hg methylation. A previous study of boreal lakes in Sweden also suggested that higher concentrations of ions such as Ca^{2+} and Mg^{2+} were related to lower MeHg in the food web (Sonesten, 2003). High Ca^{2+} levels have also been suggested as a potential inhibitor of MeHg production and direct water-borne uptake by fish (Hanten et al., 1998).

Specific conductivity was significantly positively correlated with chlorophyll *a*, DOC, and sulfate. At high concentrations, sulfate has been shown to inhibit bacteria responsible for converting inorganic

Hg to MeHg but at low concentrations can actually enhance methylation (Morel et al., 1998). DOC has been linked to increases in total Hg and MeHg in water but its effect on tissue MeHg may be negative due to the binding of MeHg to DOC (Watras et al., 1995). A few of the southern California reservoirs (e.g., Perris Reservoir) had relatively high DOC and SO_4 concentrations but lower bass MeHg concentrations. These reservoirs were high in total Hg in surface waters but low in MeHg, which suggests conditions may be less favorable for methylation.

5. Conclusions

This study investigated correlations between a variety of ecosystem factors and MeHg in black bass, and found total Hg in reservoir sediment to be the most important factor. Other important factors associated with reservoirs of higher black bass MeHg contamination was total Hg in surface water, total Hg in surrounding catchment soils, and forested area of the watershed. MeHg in summer surface water also correlated with black bass MeHg, but explained <5% of the variance in the Largemouth Bass MeHg model.

Research on reservoirs with atmospheric Hg as the primary vector for Hg contamination have put forth a top down paradigm for how fish ultimately accumulate MeHg (Hammerschmidt and Fitzgerald, 2006; Rudd, 1995; Trip and Allan, 2000; Wiener et al., 2006). In this model, atmospheric Hg is deposited on the water surface and surrounding flora. Under favorable conditions, the inorganic Hg is converted to MeHg in the water column (Eckley et al., 2005; Watras et al., 2005), and rapidly accumulated in biota (Harris et al., 2007). Based on findings from this study that show a significant correlation between total Hg sediment concentrations and black bass MeHg concentrations, an alternative bottom up paradigm for California reservoirs is indicated. While the exact processes linking total Hg in reservoir sediments to MeHg in fish was beyond the scope of this study, these results suggest that fish contamination originates from the sediment, possibly associated with sources of Hg from natural ore deposits and legacy mining operations. Subsequently, the trapped Hg contaminated sediment is exposed to conditions favorable for methylation. These conditions include the formation of anoxic hypolimnion from thermal stratification and inputs of organic carbon and sulfates from the surrounding watershed. At turnover, MeHg is mixed into the water column where it is then incorporated into phytoplankton and ultimately biomagnified in the food web. Future studies to test this hypothesis are needed to improve understanding of the conditions that may lead to lowering MeHg exposure to biota inhabiting California reservoirs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2019.05.019>.

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