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SPATIOTEMPORAL TRENDS IN FISH MERCURY FROM A MINE-DOMINATED ECOSYSTEM: CLEAR LAKE, CALIFORNIA

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Abstract. Clear Lake, California, USA, receives acid mine drainage and mercury (Hg) from the Sulphur Bank Mercury Mine, a U.S. Environmental Protection Agency (U.S. EPA) Superfund Site that was active intermittently from 1873 to 1957 and partially remediated in 1992. Mercury concentrations were analyzed primarily in four species of Clear Lake fishes: inland silversides (*Menidia beryllina*, planktivore), common carp (*Cyprinus carpio*, benthic scavenger/omnivore), channel catfish (*Ictalurus punctatus*, benthic omnivorous predator), and largemouth bass (*Micropterus salmoides*, piscivorous top predator). These data represent one of the largest fish Hg data sets for a single site, especially in California.

Spatially, total Hg (TotHg) in silversides and bass declined with distance from the mine, indicating that the mine site represents a point source for Hg loading to Clear Lake. Temporally, fish Hg has not declined significantly over 12 years since mine site remediation. Mercury concentrations were variable throughout the study period, with no monotonic trends of increase or decrease, except those correlated with boom and bust cycles of an introduced fish, threadfin shad (*Dorosoma petenense*). However, stochastic events such as storms also influence juvenile largemouth bass Hg as evidenced during an acid mine drainage overflow event in 1995.

Compared to other sites regionally and nationally, most fish in Clear Lake exhibit Hg concentrations similar to other Hg-contaminated sites, up to ~2.0 mg/kg wet mass (WM) TotHg in largemouth bass. However, even these elevated concentrations are less than would be anticipated from such high inorganic Hg loading to the lake. Mercury in some Clear Lake largemouth bass exceeded all human health fish consumption guidelines established over the past 25 years by the U.S. Food and Drug Administration (1.0 mg/kg WM), the National Academy of Sciences (0.5 mg/kg WM), and the U.S. EPA (0.3 mg/kg WM). Mercury in higher trophic level fishes exceeds ecotoxicological risk assessment estimates for concentrations that would be safe for wildlife, specifically the nonlisted Common Merganser and the recently delisted Bald Eagle.

Fish populations of 11 out of 18 species surveyed exhibited a significant decrease in abundance with increasing proximity to the mine; this decrease is correlated with increasing water and sediment Hg. These trends may be related to Hg or other lake-wide gradients such as distribution of submerged aquatic vegetation.

Key words: bioaccumulation; Clear Lake, California, USA; consumption guidelines; fish; mercury; mining; remediation; wildlife risk assessment.

Introduction

The American Heart Association recommends that healthy adults eat at least two fish meals per week. However, because of its well-documented neurotoxicological effects, especially on humans, mercury (Hg) in fish tissues is a growing concern throughout the United States and the world (Wiener et al. 2003, Mergler et al. 2007). Furthermore, recent data suggest that human Hg levels may be much higher than previously recognized for some groups, further increasing concern (Hightower and Moore 2003). In 2004, 44 states, two tribes, and one territory had issued over 3200 fish consumption advisories based on Hg, with California issuing the highest number within the western states (U.S. EPA 2005). The most widely recognized source for Hg

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contamination in fishes derives from sites where the Hg source is atmospheric deposition (Keeler et al. 1995, Lockhart et al. 1995). However, certain regions of the United States and the world have elevated Hg in fishes associated with industrial pollution such as chloralkali plants, Hg mining, or the use of Hg for gold or silver mining (Parks and Hamilton 1987, Lacerda and Salomons 1999, Turner and Southworth 1999, Alpers et al. 2005, Suchanek et al. 2008*c*, Wiener and Suchanek 2008). Northern California, and specifically the Sacramento River Basin, has been identified as one of five regions within the United States with the highest Hg concentrations in freshwater fishes (Brumbaugh et al. 2001), and this is almost certainly related to the legacy of Hg, gold, and silver mining that began in the 1800s (Churchill 2000, Alpers et al. 2005). Furthermore, nearly 75% of almost 300 sites sampled in California from 1998 to 2003 had fish with median Hg concentrations that resulted in recommendations for no consumption or significantly limited human consumption (i.e., no more than one meal per month; Davis et al. 2007).

As of 2006, the California Office of Environmental Health Hazard Assessment (OEHHA) lists fish consumption advisories for about 25 water bodies within the state based on elevated levels of Hg in fish tissue; Clear Lake is one of those sites (Alpers et al. 2005; OEHHA, *available online*).[11] On its shoreline is an abandoned Hg mine (the Sulphur Bank Mercury Mine, active intermittently from 1872 to 1957), which is one of nearly 300 abandoned Hg mines or prospects within the California Coast Range (Churchill 2000). The mine, also a U.S. Environmental Protection Agency (U.S. EPA) Superfund Site since 1990, is located on the eastern shoreline of the Oaks Arm of the Lake. It has been estimated that >100 Mg of Hg have been released into the Clear Lake aquatic ecosystem as a result of Hg mining from this single site (Chamberlin et al. 1990). Details of the mine, Hg concentrations in sediments, water, and lower trophic species and Hg loading from various sources into the Lake are described in Suchanek et al. (2008*a*, *b*, *c*).

Clear Lake is a shallow, eutrophic, alkaline lake that supports a complex aquatic ecosystem containing at least 21 species of fishes dominated primarily by nonnative centrarchids (largemouth bass [*Micropterus salmoides*] and bluegill [*Lepomis macrochirus*]) and inland silversides (*Menidia beryllina*) (Suchanek et al. 2003). The fish community has also changed dramatically since the late 1800s, with >75% of the current fish species having been introduced (Moyle 2002). As the largest natural lake completely contained within California, Clear Lake also supports the only commercial fisheries (Sacramento blackfish [*Orthodon microlepidotus*] and common carp [*Cyprinus carpio*]) on a lake in California and is a popular sport-fishing destination, with many largemouth bass and catfish tournaments held annually.

Clear Lake fishes were first analyzed for Hg in 1970 by the California Department of Health Services, with just two composite samples of largemouth bass and white catfish (*Ameiurus catus*) (CVRWQCB 1985). Subsequently, the U.S. Food and Drug Administration (U.S. FDA) analyzed additional samples in 1976 (CVRWQCB 1985) and the Toxic Substances Monitoring Program of the State Water Resources Control Board analyzed samples from 1980 to 1983 (Rasmussen 1993). A summary of data collected through 1985 was provided in a report by the California Central Valley Regional Water Quality Control Board (CVRWQCB 1985). Based on these Hg concentrations in several species of fishes, the California Department of Health Services issued the first fish consumption advisory for Clear Lake in 1986 (Stratton et al. 1987). And, in 1988, Clear Lake was placed on the Clean Water Act's 303(d) List of Impaired Water Bodies due to Hg contamination (see CVRWQCB 2002 and Suchanek et al. 2008*c*).

Here we document Hg concentrations in many species of Clear Lake fishes. Specifically, our objectives were: (1) to determine whether there were any significant relationships between Hg concentrations in different fish species and proximity to the Sulphur Bank Mercury Mine, (2) to identify any temporal trends in fish Hg concentrations over the 30+ years of data collection in Clear Lake, (3) because different ethnic groups consume a variety of fish tissues, sometimes the entire fish, determine how Hg is distributed amongst various fish tissue and organ systems, (4) to evaluate whether there is any relationship between the abundance of different fish species and proximity to the mine, and (5) to compare fish Hg concentrations in Clear Lake to human health consumption guidelines and wildlife protection criteria.

METHODS

We focused our studies primarily on four species of fishes representing four trophic guilds from sites within Clear Lake, although we also report data for several other species. As adults, these species represent different trophic guilds: inland silversides (*Menidia beryllina*, a planktivore), common carp (*Cyprinus carpio*, a benthic scavenger/omnivore), channel catfish (*Ictalurus punctatus*, a benthic omnivorous predator), and largemouth bass (*Micropterus salmoides*, a piscivorous top predator) (see also Suchanek et al. 2003, Eagles-Smith et al. 2008*a*). However, most other species in Clear Lake were analyzed for Hg as well. Mercury data for Clear Lake fishes were compiled from several sources. Historical data (prior to 1992) were obtained from the California Department of Fish and Game (CDFG 1983, 1984*a*, *b*, *c*, *d*), Department of Health Services (Stratton et al. 1987), and Lake County Vector Control (A. E. Colwell, *unpublished data*). Additional data were collected during the 10-year Clear Lake Environmental Research Center (CLERC) Monitoring Program (Su-

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[11] ⟨http://www.oehha.ca.gov/fish.html⟩ and ⟨http://www.oehha.ca.gov/fish/pdf/Adv_Map_2006.pdf⟩

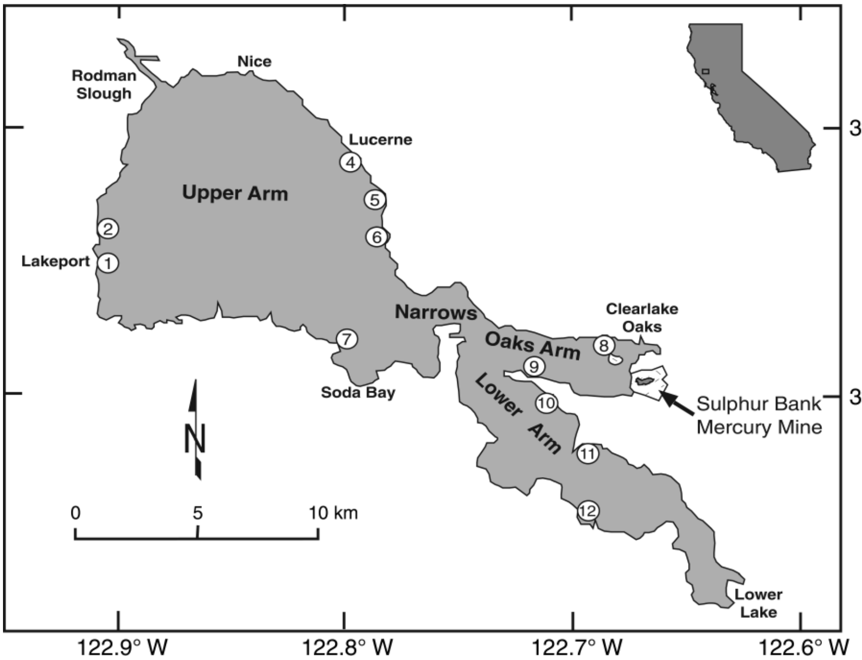

FIG. 1. Map of Clear Lake, California, USA, indicating shoreline collection sites for near-shore fish population surveys.

chanek et al. 2008*c*) from 1992 to 2001 and additional collections through 2006. Collections were made in all three arms of Clear Lake (Oaks Arm, Upper Arm, and Lower Arm) plus some additional sites (Narrows [at the junction between the Oaks Arm, Upper Arm, and Lower Arm] and Rodman Slough at a primary inflow site to the Lake; Fig. 1).

For the CLERC Monitoring Program (1992–2001), collections were made using trawl, seine, or electrofishing. Fish were placed in plastic bags and kept cool on ice (for periods $\leq$24 h) or frozen (for longer periods) until processed. For inland silversides, composites of two to five fish per size class were analyzed for whole-body TotHg concentrations. (Hereafter, Hg concentrations will refer to TotHg unless otherwise stated.) In order to determine which body components contained the greatest Hg burden, carp, channel catfish, and largemouth bass were dissected in 1992, and Hg was analyzed from several tissues and organ systems including axial muscle, liver, kidney, brain, eggs, testes, skin, scales, fat, and bone. These tissues and organs were analyzed for both TotHg and methylmercury (MeHg). Ages of some fish species were determined by analysis of scale annulae. Because Hg in fish tissues typically increases as a function of body size or age (Wiener and Spry 1996), we also report the relationship between standard length and mass for all four species, as well as the Hg concentrations for the full range of body lengths for both spatial and temporal trends. For all other species, and unless otherwise noted, Hg was analyzed as TotHg from axial muscle (fillet) tissue in all species except inland silversides, threadfin shad (*Dorosoma petenense*), and young-of-year (YOY) largemouth bass, which were analyzed as whole-body composites.

Mercury in inland silversides from the Oaks Arm was not well documented before the CLERC Monitoring Program, which began in 1992. Before 1986, only a single historical datum was available to us from 1976. To obtain sufficient sample sizes of inland silversides for statistical analysis, data from several consecutive year-pairs were combined (see Fig. 5 and Table 5); year-pairs had similar distribution patterns of $\log_{10}$(Hg) vs. length. For YOY juvenile largemouth bass ($<$150 mm), we utilized frozen archived samples (1985–1991 from Lake County Vector Control) as well as freshly collected samples from the 1992–2004 period.

Spatial trends compared fish Hg data primarily from the three arms of Clear Lake, plus some additional data from the Narrows and Rodman Slough.

For temporal trends, the largest quantity of fish Hg data over the entire sampling period (1970–2004) was collected from the Oaks Arm. The mine is located on the eastern shore of the Oaks Arm, and any changes in fish Hg concentrations influenced by changes in Hg loading from the mine would be most easily tracked by analyzing fishes from that region of the lake. Thus, temporal trends in fish Hg from the Oaks Arm for the four primary fish species (inland silversides, carp, channel catfish, and largemouth bass) provide the best data for evaluation of any changes in Hg bioaccumulation that might have taken place in the lake over this time period.

Shoreline fish population surveys were conducted from 1987 to 2001 during the months of June–August (summer) and September–November (fall) at each of 11

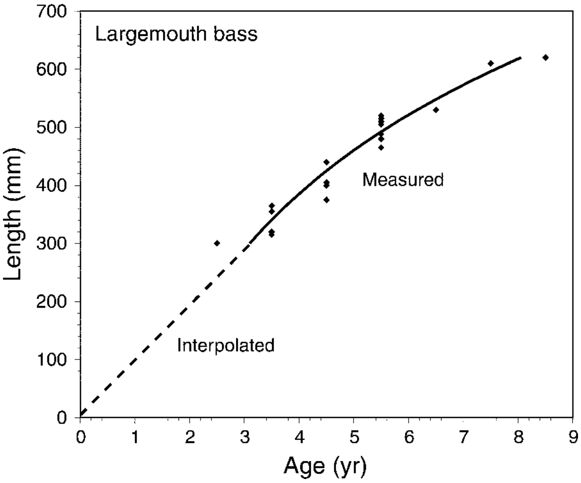

FIG. 2. Standard length vs. age extrapolations for largemouth bass (*Micropterus salmoides*) from Clear Lake collected in November 1992 based on scale annulae (L. Weeks, *unpublished data*). The fitted solid line is an exponential fit ($N = 21$); the interpolated dashed line is linear.

sites, with three transects (a, b, c) sampled at each site (Fig. 1): Upper Arm ($n = 6$), Oaks Arm ($n = 2$), and Lower Arm ($n = 3$). Stations were chosen to represent as consistent shoreline conditions as possible among all stations. A total of 929 seine hauls yielded 19 species (Table 6) for which abundance data were collected. Fish were sampled using a seine (9.1 m long × 1.2 m high) with an ace mesh (3.2 mm apertures); 83 m^2 were sampled at each station. All fish were identified to species and counted.

Analytical procedures

Total mercury was analyzed for fish collected through 1998 using standard cold-vapor atomic absorption (CVAA) methodology following EPA Method 245.6 (Roesijadi 1982, Slotton et al. 1995) at one of the following laboratories: Brooks Rand (Seattle, Washington, USA), Battelle Northwest Marine Sciences Laboratory (Sequim, Washington, USA), or the University of California Davis Environmental Mercury Laboratory (Davis, California, USA). Mercury in fish collected after 1998 were analyzed using a Milestone DMA-80 analyzer (Milestone, Shelton, Connecticut, USA) following EPA Method 7470, decomposition via combustion followed by gold amalgamation coupled with CVAA detection. Paired analyses using the different methodologies showed no statistical difference in results (Eagles-Smith 2006). Methylmercury analyses were performed by Brooks Rand or Battelle Northwest Marine Sciences Laboratory utilizing aqueous phase ethylation, followed by cryogenic gas chromatography with cold-vapor atomic fluorescence detection (Bloom 1989).

Following the convention of most fish Hg literature, all fish Hg data are reported as wet mass (WM) values. Whole-body Hg burdens were calculated using the mass percentages of each of the tissues/organs and their respective Hg concentrations.

Statistical analyses

Statistical analyses using the models and procedures described below were performed using JMP Statistical Software version 5.1 for the Macintosh (SAS Institute, Cary, North Carolina, USA). Models were fit and hypotheses were tested to assess both spatial and temporal trends of fish Hg in Clear Lake. Fish length was incorporated into models for assessing spatial and temporal variation. Specifically, $\log_{10}$[Hg] for Clear Lake locations (arms) and time (years or year groups) were adjusted for fish length using analysis of covariance (ANCOVA) models. Interaction effects between length and location (or time) were tested to determine whether a common slope model was appropriate.

In cases in which the null hypothesis of no interaction effect was accepted, an overall test for mean differences was conducted. This test was followed by multiple comparisons tests to determine which means differed using a more conservative method (Tukey's honest significant difference [HSD]) and a more liberal method (Fisher's least significant difference [LSD]). These multiple comparison methods result in a partial ordering of the least squares (model) means. That is, if two means are statistically indistinguishable, they are assigned an identical letter and cannot be ordered. Significance was set at $\alpha = 0.05$.

For population data, because shoreline fish abundance data included many zero values during any single collection date, presenting problems with normality, a very small number (0.167) was added to all abundance values for the entire data set (following the convention of Tukey [1977]). A linear regression model was then fit to $\log_{10}$(abundance) values vs. distance from the mine. A Poisson model was also fit, with comparable results. Significance was set at $\alpha = 0.05$.

RESULTS

By combining historical data (before 1992), data from the CLERC Monitoring Program (1992–2001), and additional collections through 2004, consistent relationships were observed between standard length vs. mass for largemouth bass, channel catfish, carp, and inland silversides (Eqs. 1, 2, 3, and 4, respectively). Based on scale annulae, Clear Lake largemouth bass collected in 1992 were estimated to be up to 8.5 yr old (Fig. 2).

The exponential curve fit equation ($R^2 = 0.907$) of standard length (in millimeters) vs. wet mass (in grams) in inland silversides for collections from the period 1976–2004 ($n = 331$) is

$$\text{wet mass(g)} = 0.049e^{[0.054 \times \text{length (mm)}]}. \quad (1)$$

The exponential curve fit equation ($R^2 = 0.978$) of standard length vs. wet mass in inland carp for collections from the period 1976–2000 ($n = 26$) is

TABLE 1. Total mercury (TotHg), methylmercury (MeHg), and percentage of methylmercury in fish tissues of carp, channel catfish, and largemouth bass (mg/kg wet mass), composited from $n = 3$ samples each for carp and largemouth bass and $n = 1$ for channel catfish.

	Common carp			Channel catfish			Largemouth bass		
Tissue	TotHg	MeHg	MeHg (%)	TotHg	MeHg	MeHg (%)	TotHg	MeHg	MeHg (%)
Liver	<0.10	0.048	>48	1.95	0.221	11.3	1.69	0.604	35.7
Muscle	0.24	0.227	94.6	0.89	0.824	92.6	1.04	1.010	97.1
Brain	···	0.054	···	···	0.169	···	···	0.214	···
Kidney	0.17	0.077	45.3	0.42	0.149	35.5	···	···	···
Spleen	<0.10	0.037	>37	0.21	0.112	53.3	···	···	···
Intestine	<0.10	0.033	>33	0.21	0.084	40.0	0.21	0.164	78.1
Eggs	<0.10	0.017	>17	···	···	···	<0.10	0.104	>10.4
Testes	···	···	···	0.26	0.084	32.3	···	···	···
Bone	<0.10	0.006	>6	<0.10	0.007	>0.7	<0.10	0.027	>2.7
Fat	<0.10	0.003	>3	<0.10	0.021	>2.1	<0.10	0.050	>5
Skin	<0.10	0.007	>7	<0.10	0.055	>5.5	<0.10	0.071	>7.1
Scales	<0.10	0.001	>1	···	···	···	<0.10	0.008	>0.8
Remainder	0.03	0.014	46.7	0.10	0.042	42.0	0.20	0.095	47.5

Notes: The "Remainder" concentrations were calculated values (see *Methods*). Individual tissues of inland silversides were not analyzed separately. The ranges of TotHg, MeHg, and percentage of MeHg for whole-body analyses of inland silversides were 0.02–0.41 ($n = 324$), 0.02–0.06 ($n = 4$), and 20–60% ($n = 4$), respectively. Ellipses indicate that no data were available.

$$\text{wet mass(g)} = 167.5e^{[0.005 \times \text{length (mm)}]}. \qquad (2)$$

The power curve fit equation ($R^2 = 0.970$) of standard length vs. wet mass in channel catfish for collections from the period 1976–2004 ($n = 80$) is

$$\text{wet mass(g)} = e^{-(10.93)} \times [\text{length (mm)}]^{2.977}. \qquad (3)$$

The power curve fit equation ($R^2 = 0.990$) of standard length vs. wet mass in largemouth bass for collections from the period 1976–2004 ($n = 624$) is

$$\text{wet mass(g)} = e^{-(11.50)} \times [\text{length (mm)}]^{3.124}. \qquad (4)$$

Tissue concentrations and body burdens of mercury in fish tissues and organs

Total mercury and MeHg, including the percentage MeHg in each tissue, for the major body constituents of carp, channel catfish, and largemouth bass, as well as the range of Hg concentrations for whole-body inland silversides, are presented in Table 1. For TotHg, liver tissue contained the highest concentrations in both largemouth bass and channel catfish, at levels 38–54% greater than corresponding muscle tissue. Carp, however, had liver TotHg concentrations <50% of corresponding muscle. For catfish and largemouth bass, muscle tissue contained the second highest concentrations, with kidney exhibiting ~53% of muscle concentrations and spleen, intestine, and eggs/testes exhibiting even lower levels, generally ≤24% of corresponding muscle concentrations. Total mercury in eggs/testes, bone, fat, skin, and scales of all three species was below 0.10 mg/kg.

Methylmercury was highest in muscle tissue for all three species. Liver was next highest in channel catfish and largemouth bass, but substantially lower in carp. Brain and kidney were intermediate in MeHg concentrations, ~18–35% of muscle concentrations, whereas spleen, intestine, and eggs/testes were even lower at ~10–24% of muscle. Bone, fat, skin, and scales contained the lowest MeHg concentrations, generally from 1% to 7% of muscle.

Differential Hg body burdens for various tissues and organs are presented in Table 2. Although liver tissue in channel catfish and largemouth bass contained 1.6–2.2 times higher TotHg concentrations than muscle tissue, when weighted for mass contribution to overall body burdens, liver tissue contributed only ~1–5% of the TotHg burden and 0.5–1.5% of the MeHg burden, whereas muscle tissue contained the vast majority of the Hg body burden in all three species, with 87–90% of TotHg and 93–95% of MeHg.

Mercury in other Clear Lake fishes

Data from historical studies as well as the CLERC Monitoring Program and additional collections through 2004 were used to evaluate TotHg in the four species as well as the remaining common fish species that inhabit Clear Lake. Maximum TotHg concentrations in adult fishes from Clear Lake vary widely from 0.17 mg/kg WM for threadfin shad (an epi-pelagic planktivore) to 1.94 mg/kg WM for largemouth bass (a top predator; Table 3). See the Supplement for the cumulative data used in this study.

Spatial trends

Historical TotHg data from 1976–1988 obtained from the literature were combined with data from our study (1992–2004) for four species of fishes spanning a gradient of trophic guilds: inland silversides, common carp, channel catfish, and largemouth bass. However, for statistical analysis of spatial trends, we used data from the period 1992–1996, which represented those years with the most complete and consistent size distributions for inland silversides and largemouth bass.

TABLE 2. Total mercury (TotHg) and methylmercury (MeHg) body burden percentages for specific organs and tissues in common carp, channel catfish, and largemouth bass.

	Total mercury (%)			Methylmercury (%)		
Tissue	Common carp	Channel catfish	Largemouth bass	Common carp	Channel catfish	Largemouth bass
Muscle	89.71	87.14	86.85	94.33	95.39	92.77
Liver	1.04	5.26	3.85	0.58	0.70	1.51
Intestine	3.80	2.41	1.62	2.11	1.15	1.37
Kidney	0.61	0.37	0.38	0.31	0.16	0.35
Spleen	0.11	0.07	0.08	0.06	0.05	0.10
Gonads	0.15	0.28	0.19	0.08	0.11	0.10
Bone	0.86	0.25	0.82	0.48	0.15	0.45
Skin	0.40	1.19	0.86	0.22	0.70	0.47
Fat	0.22	0.38	0.76	0.12	0.22	0.42
Scales	0.04	···	0.07	0.02	···	0.04
Brain	0.08	0.06	0.06	0.04	0.04	0.04
Remainder	2.99	2.60	4.46	1.65	1.33	2.38

Note: Ellipses indicate that no data were available.

Preliminary screening yielded one significant interaction for largemouth bass due to the smaller size range of individuals in the Narrows collection compared with the Oaks, Upper, and Lower Arm sites. Further investigation of this interaction effect indicated that the existing data on fish [Hg] vs. length for the Narrows individuals was consistent with data from the other regions of the lake and was not biologically relevant. Therefore, we did not exclude these data from the analysis.

Inland silversides, a small (typically <5 g wet mass and <100 mm length) annual species that feeds on water column plankton and chironomid midge larvae (Moyle 2002, Eagles-Smith 2006, Eagles-Smith et al. 2008*a*), exhibited the lowest concentrations of TotHg (as measured on a whole-fish basis). Over 95% of those values fell below 0.14 mg/kg (WM), although one composite sample of five individuals collected from the Oaks Arm in 2000 reached as high as 0.41 mg/kg (Fig. 3,

TABLE 3. Minimum, mean, and maximum total mercury (TotHg) concentrations (WM, wet mass) for each fish species analyzed in Clear Lake from 1976 to 2004, with data on juvenile and adult trophic status, listed in increasing maximum Hg concentration.

				TotHg concentration (mg/kg WM)			
Common name	Species	*N*	Matrix	Minimum	Mean	Maximum	Juvenile/adult trophic status
Threadfin shad	*Dorosoma petenense*	23	whole body	0.03	0.07	0.17	epi-pelagic planktivore/ epi-pelagic planktivore
Prickly sculpin	*Leptocottus armatus*	6	whole body	0.08	0.14	0.18	benthic and planktonic invertivore/benthic invertivore
Green sunfish	*Lepomis cyanellus*	4	muscle	0.10	0.17	0.20	planktivore and invertivore/ littoral omnivore
Black bullhead	*Ictalurus melas*	6	muscle	0.12	0.22	0.37	benthic omnivore/benthic omnivore
Inland silversides	*Menidia beryllina*	324	whole body	0.02	0.08	0.41	littoral planktivore/planktivore
Bluegill	*Lepomis macrochirus*	56	muscle	0.04	0.15	0.47	planktivore/omnivore
Clear Lake hitch	*Lavinia exilicauda*	23	muscle	0.07	0.18	0.54	benthic and planktonic invertivore/pelagic planktivore and invertivore
Brown bullhead	*Ameiurus nebulosus*	36	muscle	0.07	0.27	0.58	benthic invertivore/benthic omnivore and piscivore
Sacramento blackfish	*Orthodon microlepidotus*	32	muscle	0.08	0.29	0.58	benthic and pelagic invertivore/ planktonic suspension feeder
Common carp	*Cyprinus carpio*	26	muscle	0.05	0.21	0.66	planktonic omnivore/benthic scavenger and omnivore
Black crappie	*Pomoxis nigromaculatus*	74	muscle	0.04	0.32	0.81	planktivore/omnivore
White catfish	*Ameiurus catus*	36	muscle	0.07	0.47	0.86	benthic invertivore/omnivore
Western mosquitofish	*Gambusia affinis*	11	whole body	0.02	0.32	1.11	omnivore/omnivore
White crappie	*Pomoxis annularis*	10	muscle	0.15	0.48	1.30	planktivore/omnivore
Channel catfish	*Ictalurus punctatus*	81	muscle	0.07	0.47	1.50	benthic invertivore/benthic omnivorous predator
Largemouth bass	*Micropterus salmoides*	625	muscle	0.03	0.29	1.94	benthic invertivore/invertivore and piscivore

Notes: Threadfin shad were analyzed as composites of six individuals per sample; inland silversides were analyzed as composites of five individuals per sample; the western mosquitofish were collected from wetlands surrounding Clear Lake.

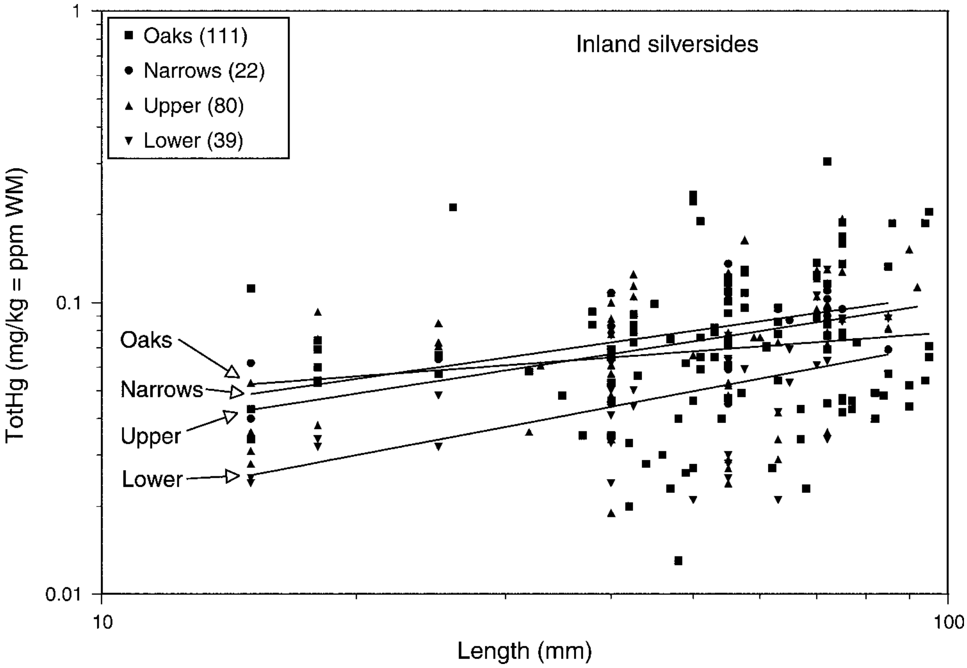

FIG. 3. Spatial variation of whole-body total mercury (TotHg; WM, wet mass) in inland silversides (*Menidia beryllina*) from Clear Lake for 1992–1996 collections. The values in parentheses in the key are sample sizes. The axes are log scale. The fitted lines are power curves: Oaks Arm, $R^2 = 0.026$; Narrows, $R^2 = 0.299$; Upper Arm, $R^2 = 0.167$; Lower Arm, $R^2 = 0.288$.

Table 3). A linear regression analysis of $\log_{10}$[Hg] indicated that for all sites (Oaks Arm, Upper Arm, Lower Arm, and the Narrows), TotHg increased significantly as a function of body length (Table 4).

Analysis of covariance (both Tukey's HSD test and Fisher's LSD test; Table 4) indicated that location within Clear Lake plays a significant role in determining the TotHg concentration in inland silversides. More specifically, the Oaks Arm, Narrows, and Upper Arm exhibited the highest Hg concentrations, which were significantly indistinguishable from one another and significantly different from the lowest concentrations in the Lower Arm, as evidenced by both multiple comparisons tests (Fig. 3, Table 4).

As an adult, the common carp is an omnivorous benthic feeding species that roots in the contaminated bottom sediments of the lake (Moyle 2002). It lives to a maximum age of 12–15 yr (Moyle 2002), and, with TotHg concentrations in lake bed surficial sediments as high as 438 mg/kg (Suchanek et al. 2008*a*; Suchanek et al., *in press*), one might expect this species to exhibit significantly elevated Hg concentrations. However,

TABLE 4. Spatial analysis of mercury in inland silversides and largemouth bass from 1992 to 1996.

		Relationship with length		Differences between sites	
Location	*N*	*P*	R^2	Tukey's HSD test	Fisher's LSD test
Inland silversides					
Oaks Arm	68	0.0002	0.1890	A	A
Narrows	22	0.0109	0.2823	A	A
Upper Arm	80	0.0001	0.1774	A	A
Lower Arm	39	0.0001	0.3301	B	B
Largemouth bass					
Oaks Arm	90	<0.0001	0.5676	A	A
Narrows	30	<0.0001	0.5107	AB	AB
Upper Arm	85	<0.0001	0.6602	B	B
Lower Arm	36	<0.0001	0.4258	B	B

Notes: Relationship with length was determined by linear regression. Linear regression values indicate significance levels for the relationship between TotHg and standard length for each arm or region within Clear Lake. For multiple comparisons tests (Tukey's honest significant difference [HSD; more conservative] and Fisher's least significant difference [LSD; more liberal]) identical letters represent statistical equivalencies in Hg concentration distributions for individuals collected from each location.

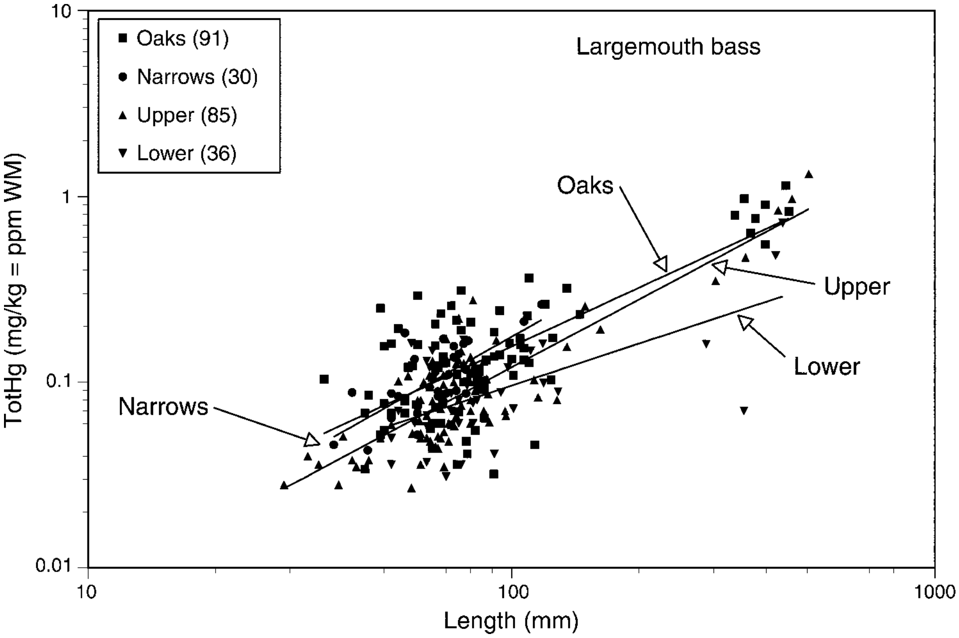

FIG. 4. Spatial variation of muscle total mercury (TotHg; WM, wet mass) as a function of length in largemouth bass (*Micropterus salmoides*) from several sites in Clear Lake from 1992–1996 collections. The values in parentheses in the key are sample sizes. The axes are log scale. The fitted lines are power curves: Oaks Arm, $R^2 = 0.541$; Narrows, $R^2 = 0.508$; Upper Arm, $R^2 = 0.709$; Lower Arm, $R^2 = 0.401$.

among the larger and older fish species analyzed in detail (carp, channel catfish, and largemouth bass), carp were consistently lowest in muscle TotHg, with a mean concentration of 0.213 mg/kg (Table 3). These results agree with the current dogma that trophic position is one of the primary factors influencing Hg concentrations in fish (Weiner et al. 2003). Because so few individuals were collected, it was not possible to determine whether there were significant spatial differences among sites within Clear Lake for carp Hg.

Adult channel catfish are benthic-dwelling omnivores, residing and feeding in a habitat similar to carp. This species, however, feeds on higher trophic level prey items, including other finfish and crayfish (Moyle 2002). Channel catfish were intermediate in muscle TotHg concentrations, with values reaching as high as 1.50 mg/kg (Table 3). As with carp, too few individuals were collected to conduct statistically meaningful spatial analyses for channel catfish Hg.

As adults, largemouth bass are voracious midwater top predators, feeding primarily on other fish species (Moyle 2002) as well as a heavy reliance on crayfish (Eagles-Smith et al. 2008*a*). Largemouth bass contained the highest concentrations of TotHg found in all four intensively studied fish species, with a high muscle Hg concentration of 1.94 mg/kg obtained from the Oaks Arm (Table 3, Fig. 4).

For all sites in Clear Lake, largemouth bass muscle Hg was positively correlated with fish length (Table 4). In addition, both Tukey's HSD and Fisher's LSD multiple comparisons tests indicate that largemouth bass in the Oaks Arm have significantly higher muscle Hg than the Upper and Lower Arm, which were indistinguishable from one another (Table 4). Mercury in largemouth bass from the Narrows is intermediate between the Oaks Arm and the Upper and Lower Arms.

For both inland silversides and largemouth bass, the greatest differences in fish Hg were observed between the Oaks Arm (high concentrations) and the Lower Arm (low concentrations) (Table 4). This pattern of contamination is consistent with the movement of Hg-contaminated particles originating from the mine and being transported first to the Narrows, then the Upper Arm, then finally down to the Lower Arm, as described by Rueda et al. (2008) and discussed in Suchanek et al. (2008*c*).

Temporal trends

For inland silversides, no striking or consistent long-term temporal patterns or trends were observed in whole-body TotHg between different year groups from 1986 to 2004, although there was considerable variability over that time period (Fig. 5, Table 5). Combined data from both types of ANCOVA multiple comparisons tests (Table 5) plus the temporal trends in Fig. 5 suggest that Hg concentrations in inland silversides were low in 1986/1987 and 1990/1991, whereas 2001/2002 and 2003/2004 were years when silversides had higher concentrations of Hg. These data are consistent with those found for juvenile largemouth bass (see below in this section).

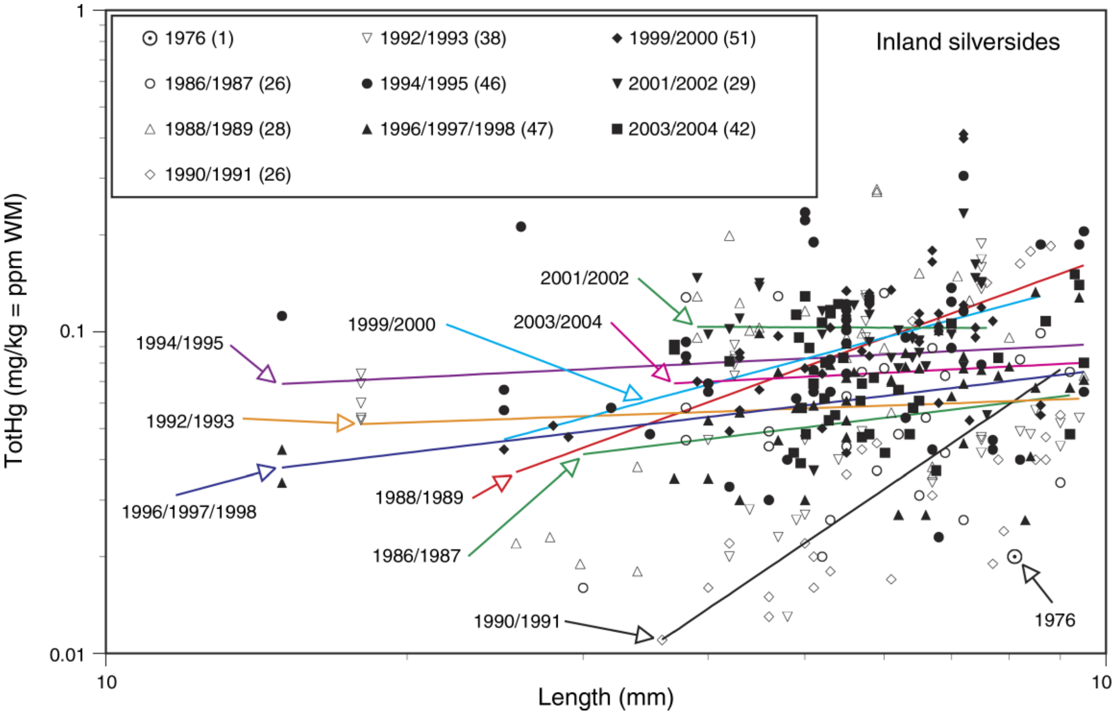

FIG. 5. Temporal variation of whole-body total mercury (TotHg; WM, wet mass) as a function of length in inland silversides (*Menidia beryllina*) from the Oaks Arm of Clear Lake from 1976 to 2004. The values in parentheses in the key are sample sizes. The fitted lines are power curves: 1986/1987, $R^2 = 0.035$; 1988/1989, $R^2 = 0.258$; 1990/1991, $R^2 = 0.484$; 1992/1993, $R^2 = 0.008$; 1994/1995, $R^2 = 0.010$; 1996/1997/1998, $R^2 = 0.111$; 1999/2000, $R^2 = 0.229$; 2001/2002, $R^2 = 0.001$; 2003/2004, $R^2 = 0.009$.

TABLE 5. Temporal analysis of mercury in inland silversides and largemouth bass from the Oaks Arm only.

	Relationship with length			Differences between years	
Sample years	*N*	*P*	R^2	Tukey's HSD test	Fisher's LSD test
Silversides					
1986/1987	26	0.4174	0.028	D	D
1988/1989	28	0.0287	0.171	ABC	AB
1990/1991	26	0.0001	0.473	E	E
1992/1993	38	0.4530	0.016	CD	D
1994/1995	46	0.3371	0.021	AB	AB
1996/1997/1998	47	0.0332	0.262	BCD	CD
1999/2000	44	0.0026	0.196	A	AB
2001/2002	29	0.9026	0.001	A	A
2003/2004	40	0.4118	0.018	ABCD	BC
Largemouth bass					
1982/1983	53	<0.0001	0.549	A	AB
1985/1986	16	<0.0001	0.793	ABC	ABC
1987/1988	16	0.1136	0.110	ABC	ABC
1989/1990	10	<0.0001	0.895	ABC	A
1991/1992	18	<0.0001	0.904	C	C
1993/1994	21	0.0086	0.275	ABC	ABC
1995/1996	56	0.0082	0.106	ABC	BC
1997/1998	11	0.0005	0.724	ABC	BC
1999/2000	65	<0.0001	0.780	BC	C
2001/2002	40	<0.0001	0.358	AB	B
2003/2004	60	<0.0001	0.475	A	AB

Notes: Relationship with length was determined by linear regression. Linear regression values indicate significance levels for the relationship between total mercury and standard length in each collection period. For multiple comparisons tests (Tukey's honest significant difference [HSD; more conservative] and Fisher's least significant difference [LSD; more liberal]) identical letters represent statistical equivalencies in Hg concentration distributions for individuals collected during each year group.

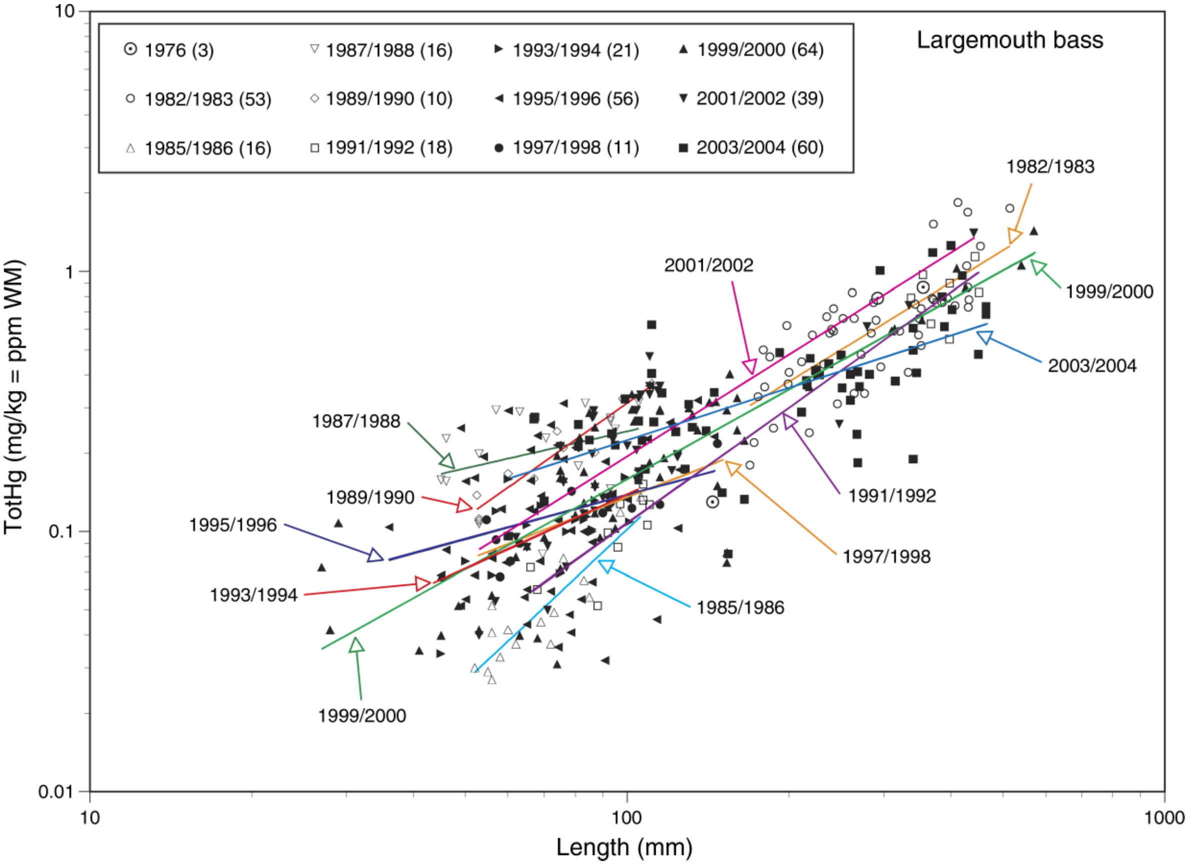

FIG. 6. Temporal variation of muscle total mercury (TotHg; WM, wet mass) as a function of length in largemouth bass (*Micropterus salmoides*) from the Oaks Arm of Clear Lake from 1976 to 2004. The values in parentheses in the key are sample sizes. The fitted lines are power curves: 1982/1983, $R^2 = 0.594$; 1985/1986, $R^2 = 0.784$; 1987/1988, $R^2 = 0.123$; 1989/1990, $R^2 = 0.899$; 1991/1992, $R^2 = 0.958$; 1993/1994, $R^2 = 0.386$; 1995/1996, $R^2 = 0.074$; 1997/1998, $R^2 = 0.690$; 1999/2000, $R^2 = 0.713$; 2001/2002, $R^2 = 0.640$; 2003/2004, $R^2 = 0.451$.

Although too few individuals of both carp and catfish were available from the Oaks Arm to conduct formal statistical analyses on temporal trends, our limited data do suggest that the most recent collections in 2000 and 2001 (compared with previous years) do not indicate an obvious reduction in fish Hg in recent years.

Our largest data set for temporal trend analysis was collected on largemouth bass ($N = 367$). As with inland silversides, several year groups were created to increase sample size for temporal analyses (Fig. 6, Table 5). Total mercury in largemouth bass exhibited considerable interannual variability, but no monotonic temporal trend was observed in the full range of sizes (Fig. 6). Linear regression analysis indicated a significant positive relationship between length and TotHg for nearly all the year groups in Oaks Arm largemouth bass (Table 5). Both Tukey's (more conservative) and Fisher's (more liberal) multiple comparisons tests indicated no consistent long-term temporal trend in TotHg concentrations from 1982/1983 to 2003/2004 (Table 5). However, the variable Hg fluctuations among years was similar to that exhibited by inland silversides.

Influence of alien species and stochastic events on mercury bioaccumulation

Alien species.—While adult largemouth bass do not change their Hg concentrations quickly, small individuals such as juvenile largemouth bass that are accumulating biomass (and contaminants) at a relatively rapid rate can reflect more rapid changes in the concentrations of Hg in the environment. Therefore, we also analyzed juvenile (YOY, <150 mm standard length) bass to better assess how potential changes in Hg loading to the lake might affect bass uptake and bioaccumulation on an annual basis. Dramatic fluctuations in TotHg concentrations were documented during this 20–30 year period, with peaks observed in 1989, 1995, and 2004 (Fig. 7). This suggests that fluctuations in juvenile bass Hg may be reflective of either: (1) changes in Hg concentrations in their environment or (2) changes in their dietary patterns. No long-term monotonic trends in the concentration of TotHg in sediments or water in Clear Lake have been documented between 1992 and 1998, other than a spike in water TotHg and MeHg in 1995 as a result of an overflow event at the mine (see below and

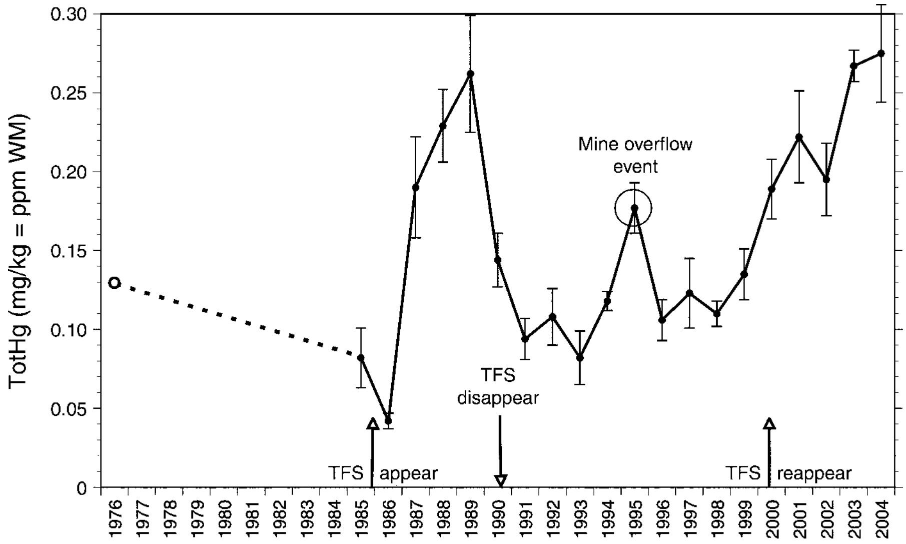

FIG. 7. Temporal variation of whole-body total mercury (TotHg; WM, wet mass; mean + SE) in juvenile (<150 mm) largemouth bass (*Micropterus salmoides*) from the Oaks Arm, with approximate dates of the invasions and population crashes of threadfin shad (TFS; *Dorosoma petenense*) (see Eagles-Smith et al. [2008*b*] for details). Only one datum point was available for 1976.

Suchanek et al. 2008*a*). However, shifts in diet may explain the dramatic cycles of Hg in juvenile bass as a result of the invasion of alien species (see Eagles-Smith et al. 2008*b*). Specifically, introductions and subsequent boom/bust cycles of threadfin shad, which compete with juvenile bass for planktonic food resources, may have forced juvenile bass to focus on other benthic prey that have higher Hg (documented in Suchanek et al. 2008*b*). Thus, population shifts in threadfin shad appear to be closely correlated with dramatic increases and decreases in juvenile bass whole-body TotHg concentrations (see Eagles-Smith 2006 and Eagles-Smith et al. 2008*b* for details).

Stochastic events.—Stochastic processes, such as unpredictable extreme weather events, can also have a dramatic influence on the bioaccumulation of Hg for some species. A striking example of this occurred when in January and March of 1995 two significant El Niño storm events caused large volumes of mine-derived water to (1) flow through the waste rock pile between a large open pit on the mine site (the Herman Pit, filled with pH 3 fluids) and the lake and (2) overflow the Herman Pit directly into the lake (Suchanek et al. 2000*a*, *b*, 2008*c*). These events significantly increased the concentration of dissolved Hg (both TotHg and MeHg) in the water column (Fig. 8A). Juvenile largemouth bass (<150 mm length) collected in all three arms of Clear Lake from 1994 to 1996 exhibited concomitant increases in Hg associated with this event during 1995, with subsequent decreases in 1996 (Figs. 7 and 8B), showing the immediate and close relationship between Hg loading from the Sulphur Bank Mercury Mine and bioaccumulation in fish tissues, especially in the Oaks Arm. The dramatic increase of Hg in individuals from the Oaks Arm and more subtle increases at other locations correlates with the increases in Hg observed in the water column near the mine shown in Fig. 8A, influenced primarily by the mine.

Fish population surveys

Data from the 929 seine hauls at 11 shoreline sites around Clear Lake from 1987 to 2001 suggest a strong negative relationship between fish abundance of some species and proximity to the mine and, by correlation, elevated concentrations of Hg in sediments, water, and prey close to the mine (Suchanek et al. 2008*a*, *b*). Eleven out of 16 fish species exhibited strikingly significant negative abundance trends with proximity to the mine (Table 6).

DISCUSSION AND CONCLUSIONS

Data presented here represent one of the largest and longest data sets of fish Hg from a single site yet reported, especially for California. However, a number of other regional studies produced fish Hg data that can be compared with the Clear Lake data set. (1) In a recent compilation, Davis et al. (2000, 2003) provided data for largemouth bass collected from the San Francisco Bay-Delta and the San Joaquin River system in fall 1998. For largemouth bass composite samples in the range of 310–480 mm standard length, TotHg ranged from 0.16 to 0.70 mg/kg WM, which is representative of mid-range values for Clear Lake bass. (2) In a similar sampling program from the Sacramento River watershed, Larry Walker Associates (2000) reported largemouth bass Hg ranging from ~0.25 to 1.16 mg/kg, which is still lower than the maximum values reported for Clear Lake. (3) May et al. (2000) reported largemouth bass and channel

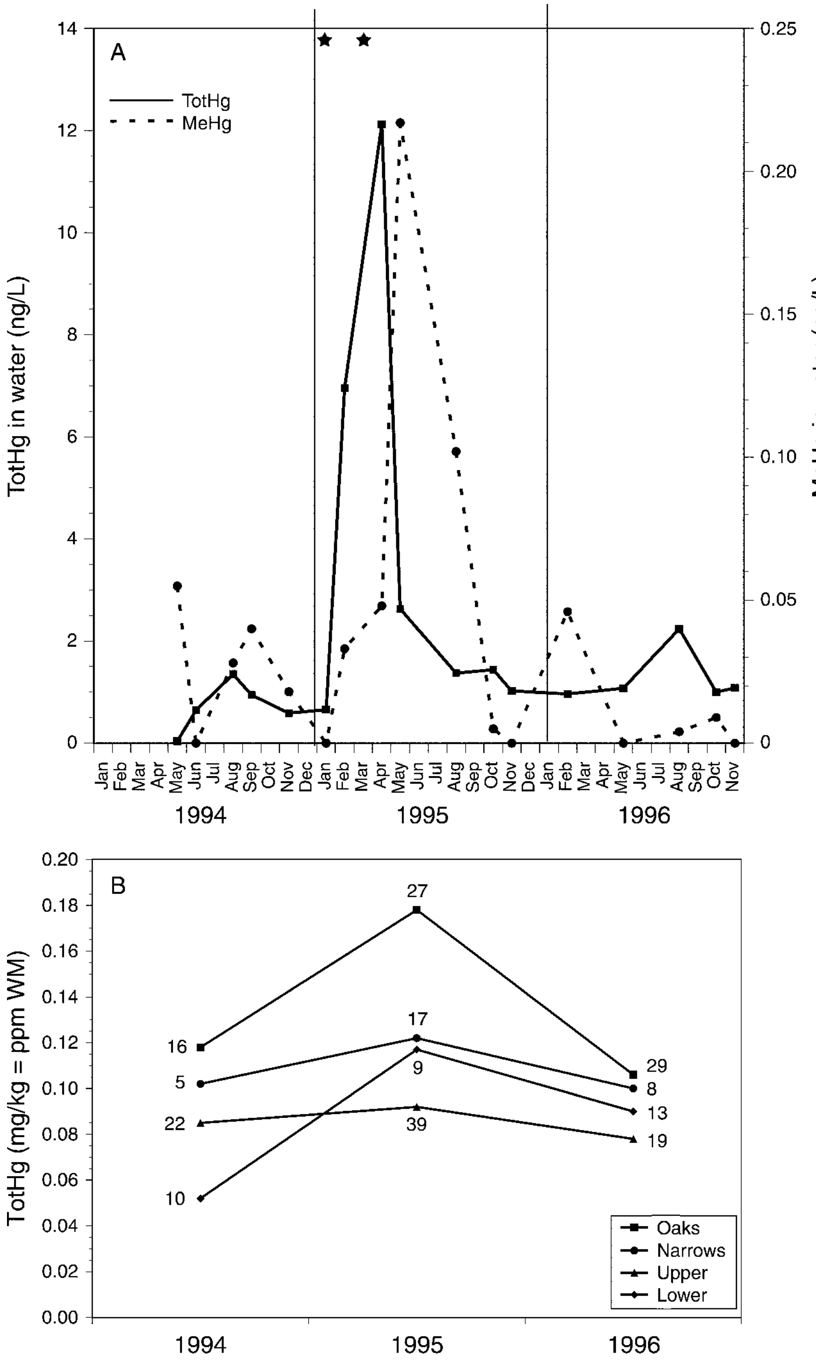

FIG. 8. (A) Total mercury (TotHg) and methylmercury (MeHg) in filtered (0.45 μm) bottom water at station OA-01 near the Sulphur Bank Mercury Mine from 1994 to 1996 (from Suchanek et al. [2008*a*]). Heavy rains in 1995 (dates shown as stars) resulted in dramatically increased TotHg and MeHg concentrations in bottom waters in the Oaks Arm. (B) Whole-body TotHg (WM, wet mass) in juvenile (<150 mm) largemouth bass (*Micropterus salmoides*) collected during the summers of 1994, 1995, and 1996 from Oaks Arm, Narrows, Upper Arm, and Lower Arm of Clear Lake. The values near data points are sample sizes for each collection.

catfish Hg data that ranged from 0.20 to 1.2 mg/kg WM for largemouth bass ($N = 26$) and from 0.16 to 0.75 mg/kg WM for channel catfish ($N = 16$) from gold-mining impacted reservoirs in the Sierra Nevada. (4) In northern California, May et al. (2005) reported largemouth bass data ($N = 33$), which ranged from 0.11 to 1.23 mg/kg from Trinity Lake (which is influenced by gold and Hg mining). (5) Brumbaugh et al. (2001) reported some Hg concentrations for largemouth bass within California and Nevada, with a sample size of 50, yielding a mean Hg concentration of 0.51 mg/kg WM (range 0.05–4.22 mg/kg); the extremely high values were

Table 6. Shoreline fish population survey results for summer and fall and both seasons pooled.

		Significance with distance from mine		
		"*R*" analyses		Linear regression
Common name	Species name	Summer	Fall	Pooled data
Black crappie	*Pomoxis nigromaculatus*	***	***	***
Bluegill	*Lepomis macrochirus*	***	***	***
Brown bullhead	*Ameiurus nebulosus*	***	***	***
Channel catfish	*Ictalurus punctatus*	ND	ND	NS
Clear Lake hitch	*Lavinia exilicauda*	***	NS	*
Common carp	*Cyprinus carpio*	***	***	***
Golden shiner	*Notemigonus crysoleucas*	ND	ND	NS
Goldfish	*Carassius auratus*	***	*	***
Green sunfish	*Lepomis cyanellus*	***	***	***
Inland silversides	*Menidia beryllina*	***	***	***
Largemouth bass	*Micropterus salmoides*	***	***	***
Mosquitofish	*Gambusia affinis*	NS	***	NS
Pikeminnow	*Ptychocheilus grandis*	ND	ND	NS
Prickly sculpin	*Leptocottus armatus*	***	***	**
Sacramento blackfish	*Orthodon microlepidotus*	***	***	NS
Sacramento sucker	*Catostomus occidentalis*	***	***	***
Threadfin shad	*Dorosoma petenense*	***	***	NS
Tule perch	*Hysterocarpus traski*	***	*	**
White crappie	*Pomoxis annularis*	NS	***	NS

* $P < 0.05$; ** $P < 0.001$; *** $P < 0.0001$; ND, insufficient data; NS, not significant.

samples from the highly contaminated Lahontan Reservoir, Nevada. Thus, Hg concentrations in the majority of Clear Lake fishes are very similar to those reported for comparable species at most other contaminated sites within California; but Lahontan Reservoir is considered one of the highest Hg-contaminated areas within the United States (Brumbaugh et al. 2001; also see Suchanek et al. 2008*a*).

Compared with other sites outside the region, Hg in Clear Lake fishes is comparable to sites that have Hg contamination from atmospheric deposition in acidic lakes (Wiener and Spry 1996), yet generally not as high as sites influenced by fungicides from golf courses (Koirtyohann et al. 1974), newly flooded reservoirs (Abernathy et al. 1975, cited in Wiener and Spry 1996), and chloralkali plants (Parks and Hamilton 1987).

Fish species from different trophic levels exhibited a wide range of Hg concentrations. Of the four species studied in detail, inland silversides (the smallest and lowest trophic level species) exhibited the lowest Hg concentrations. Mercury concentrations increased progressively for the following species: inland silversides < carp < channel catfish < largemouth bass.

Mercury concentrations in fishes from Clear Lake vary widely, both spatially and temporally. Spatially, the Sulphur Bank Mercury Mine site, and the Oaks Arm into which it contributes ongoing acid mine drainage, represents the point source for present-day biotic Hg contamination in Clear Lake (see Suchanek et al. 2008*a*, *c*). This is substantiated by consistent patterns of elevated fish Hg concentrations in the Oaks Arm from multiple trophic level species, in historic (pre-1992) and recent (since 1992) collections. While fish Hg concentrations have fluctuated over time, they have not shown a monotonic decrease during the past 20–30 years, nor have they exhibited a significant decrease since the 1992 Sulphur Bank Mercury Mine site remediation conducted by U.S. EPA (see Suchanek et al. 2008*c*). Interestingly, Greenfield et al. (2005) also found no significant monotonic trends in Hg concentrations in several other species of fishes in this region (in San Francisco Bay) between 1970 and 2000, attributing the ongoing elevated concentrations to Hg from historical mining sources.

Using juvenile largemouth bass as a sentinel species for short-term changes, a dramatic increase in Hg bioaccumulation in this species was observed in the mid- to late 1980s and a second dramatic increase began in the mid-1990s with the highest values reported in the most recent samples collected for this study in 2004. This is especially significant because it was hoped that the 1992 remediation activities would lower Hg loading to Clear Lake, with subsequent reduction of fish Hg concentrations. To the contrary, Hg concentrations in juvenile bass have continued to increase over the 12 years since the remediation. These increases, however, may also be influenced by changes in the trophic structure of the fish community in the lake driven by an introduced alien species, threadfin shad (see Eagles-Smith et al. 2008*b*).

Because Clear Lake is an unusual site with extremely high inorganic Hg loading, it is useful to evaluate the proportions of TotHg and MeHg in relation to different biotic compartments, since this will influence the further biotransfer to higher trophic levels. Most tissues and organs (except liver and muscle) for each species studied in detail (carp, channel catfish, and largemouth bass) contained relatively little TotHg or MeHg (Table 1). In comparing absolute Hg concentrations, liver tissue contained the highest concentrations of TotHg in both largemouth bass and channel catfish, at levels 60–100%

higher than corresponding muscle tissues. Carp, however, exhibited liver Hg concentrations <50% of corresponding muscle tissue levels. Muscle tissue contained the next highest Hg concentrations, with kidney, spleen, intestine, testes, and brain exhibiting intermediate levels (generally ≤40% of corresponding muscle concentrations) and the other remaining organs/tissues (bone, skin, scales, fat, and eggs) falling below the operational detection level. The relatively high percentage of MeHg in the eggs of largemouth bass is of concern for maternal transfer and potential effects on juvenile bass (Wiener et al. 2003). From a body burden perspective, however, for both TotHg and MeHg, muscle tissue contained the vast majority (typically 85–95%) of the Hg body burden.

The relative proportions of TotHg and MeHg in the various organs and tissues of Clear Lake fishes is mostly representative of those previously reported in the literature. Goldstein et al. (1996) reported TotHg concentrations in muscle and liver tissue from common carp and channel catfish from Minnesota and North Dakota. In their study, carp livers contained one-third the Hg concentration of muscle tissue, whereas in channel catfish the concentrations were quite similar, implying a difference in the way Hg is either processed or stored in the liver between these two species. In our Clear Lake samples, Hg concentrations in carp livers were also approximately one-third of those in catfish, but in largemouth bass, the liver contained twice as much Hg as did muscle tissue. Cizdziel et al. (2003) analyzed channel catfish and largemouth bass from Lake Mead, USA. Their data show liver Hg ~166% and 62% of muscle tissue in catfish and bass, respectively. In our Clear Lake samples, Hg concentrations in liver tissue were ~25%, 219%, and 163% of muscle tissue for carp, catfish, and bass, respectively. Cizdziel et al. (2003) also observed that when muscle Hg concentrations were low (less than ~0.5 mg/kg WM), liver Hg was less than muscle Hg; however, when muscle Hg was high (more than ~1.0 mg/kg WM), liver Hg was much greater than muscle Hg. As observed above, Hg in carp is consistent with the observations of Cizdziel et al. (2003). For catfish and bass, with muscle Hg concentrations ~1.0 mg/kg WM, liver Hg was significantly higher. Has-Schön et al. (2006) analyzed individual tissues/organs in this same species of carp from Croatia, showing comparable results with liver Hg concentrations ~42% of those in muscle. Has-Schön et al. (2006) also found Hg in carp kidney was ~66% of that in muscle, about the same proportion (71%) as that in Clear Lake carp.

Reduction in fish mercury (or not)

With the intent of reducing Hg loading to Clear Lake in order to lower fish Hg concentrations, the U.S. EPA performed an emergency remediation in 1992 by reducing erosion of Hg-laden tailings and waste rock from the shoreline at the mine. In 1998 the agency also reduced the amount of rainwater that passes into the Herman Pit by redirecting surface flow directly into Clear Lake in order to reduce the amount of low-pH fluids that leach Hg from the waste rock dam and subsequently become transported to the lake. Despite these remediation actions at Clear Lake, no uniform reduction in fish Hg has been documented over the 12 years since remediation action was initiated in 1992. Rather, biological processes such as alien species introductions (Fig. 7 and Eagles-Smith et al. 2008*b*) and physical processes such as the El Niño flooding events in 1995 (Fig. 8) appear to be more significant drivers that control fish Hg concentrations on annual to decadal time scales.

A recent compilation of data evaluating the influence of Hg loading on fish Hg posed the question "Will efforts to reduce inorganic Hg inputs and mobility in the environment reduce fish Hg concentrations?" (Munthe et al. 2007:33). Results are varied and the ecosystem response to increases or decreases in Hg loading from point source Hg contamination is not always linear. Some investigators found direct relationships between increased inorganic Hg loading from a chlor-alkali plant and increased fish Hg (Parks and Hamilton 1987). A dramatic decline (~75%) in walleye muscle Hg concentrations was also observed at that site over a 13-year period after the closure of the plant, although the reduction from ~15 to 3.5 mg/kg WM still significantly exceeded human health consumption guidelines, and fish Hg further declined little over the next 15 years. Other remediated sites have also experienced similar declines in fish Hg by ~50–80% over the first decade after the Hg source was abated, with reductions slowing dramatically after that period (Borgmann and Whittle 1991, Lodenius 1991, Herut et al. 1996, Francesconi et al. 1997, Scheider et al. 1998, Lindeström 2001). However, at other Hg-contaminated sites, fish Hg reduction has been much slower or nonexistent following source removal (Southworth et al. 2000), sometimes even after 30 years (Effler 1996). This could be the result of continued high Hg loading, resuspension of contaminated sediments, high natural sulfate loading, high loading of labile carbon, and highly anoxic conditions in the hypolimnion (Munthe et al. 2007), most all of the conditions (within the exception of the anoxic hypolimnion) that exist at Clear Lake.

The lack of fish Hg reduction at Clear Lake 12 years after remediation at the Sulphur Bank Mercury Mine may also be related to the form of Hg that is being loaded to the lake and subsequently bioaccumulated. Mercury (in the form of cinnabar) from soil in steeply sloped waste rock piles was reduced significantly by the 1992 remediation (Suchanek et al. 2008*c*), but the ongoing leaching of dissolved Hg (see Suchanek et al. 2008*a*, *c*; Suchanek et al., *in press*) may have a much greater influence in maintaining elevated Hg concentrations in these fish species (Suchanek et al., *in press*). It is also becoming increasingly evident that at sites with extremely high inorganic Hg contamination, TotHg (as opposed to MeHg) may play an important, but unknown role in Hg bioaccumulation and toxicity and may influence the speed at which fish Hg declines after remediation (Munthe et al. 2007). Thus, further efforts should be made to investigate the origin and chemical form of ongoing Hg loading to Clear Lake with the aim of reducing fish Hg in this system.

Population trends

Eleven of the 18 fish species monitored between 1987 and 2001 exhibit a population decline with proximity to the mine. Based on data available at this time, it is impossible to know whether Hg plays a significant role in determining the abundance of these particular species. Mercury can impair reproduction in fishes by affecting gonadal development, spawning success, hatching success, or the health and survival of larval stages (Wiener and Spry 1996, Wiener et al. 2003, Scheuhammer et al. 2007). Whether these effects are manifested at Clear Lake is unknown at this time. However, at least one other data set suggests other possible explanations for the distribution of these species. A preliminary hydroacoustic survey conducted in Clear Lake in September 2002 (ReMetrix 2003) suggests that the Upper Arm and the Lower Arm might have greater biocover and biovolume of submerged aquatic vegetation than the Oaks Arm, where the mine is located. Further investigation is needed to determine whether aquatic vegetation, Hg concentrations, or other factors such as localized increased acidity or sulfate concentrations near the mine have the greatest influence on populations of these fish species.

Human health criteria

Human fish consumption advisories have been in place for Clear Lake since 1986 (Stratton et al. 1987). These include recommendations that no fish from Clear Lake be consumed by pregnant or nursing women, women anticipating becoming pregnant, or children under six years old. For all other groups of consumers, no more than 454 g (1 lb.) of fish fillet should be consumed per month of the listed species, including largemouth bass, channel catfish, and several other predatory species. Children 6–15 years of age should eat no more than one-half the amounts indicated for adults. As of 2006, the state of California issued fish consumption advisories for Hg within ~25 water bodies throughout the state (OEHHA, *available online*; see footnote 11).

Nationally, several different human health guidelines have been developed over the past few decades for TotHg and MeHg concentrations in edible fish tissues. The U.S. Food and Drug Administration (U.S. FDA) first proposed a maximum allowable administrative guideline of 0.5 mg/kg WM for TotHg in fish and shellfish tissues in 1969. In 1972, the National Academy of Sciences (NAS) agreed with the U.S. FDA and established the same 0.5 mg/kg guideline (NAS 1973). After more detailed analysis of seafood consumption patterns prepared by the National Oceanic and Atmospheric Administration (NOAA), the U.S. FDA guideline was raised to 1.0 mg/kg Hg in 1979 (Federal Register 44, 3990, 19 January 1979). In 1984, this 1.0 mg/kg TotHg standard was converted to a standard based on MeHg (U.S. FDA 1984). Since then, the original more conservative guideline of 0.5 mg/kg Hg WM was also established in 1998 by Health Canada. As of this writing, the U.S. EPA established the most protective consumption advisory for fish tissues of 0.3 mg/kg MeHg WM (U.S. EPA 2001), which, for fish, is virtually equivalent to TotHg concentrations.

Wildlife health criteria

Methodologies for determining maximum Hg concentrations in fish tissues that would be protective of wildlife (especially threatened and endangered species), and especially as they relate to emerging total maximum daily load (TMDL) numeric targets for reducing Hg loading into various watersheds, have been developed by the U.S. Fish and Wildlife Service (Schwarzbach et al. 2001, CVRWQCB 2002, U.S. FWS 2003). Implications of fish tissue Hg concentrations for species that prey on fish in this region of California were considered for two non-listed species (Common Merganser, *Mergus merganser*, and Osprey, *Pandion haliaetus*) and one listed species (Bald Eagle, *Haliaeetus leucocephalus*). (At the time this manuscript was written, the bald eagle was listed as a Federally Threatened Species. On 8 August 2007 the Bald Eagle was officially delisted.)

Schwarzbach et al. (2001) used the Common Merganser to evaluate the effects of fish Hg concentrations on wildlife species. This species is a year-round resident of the Clear Lake and Cache Creek watershed and likely breeds there. Based on an ecotoxicological risk assessment, Schwarzbach et al. (2001) calculated a "safe mean fish concentration of Hg" that would be protective of the Common Merganser at 0.09 mg/kg WM.

As noted above, the U.S. EPA (2001) developed and recommended a new national human health consumption criterion for MeHg of 0.3 mg/kg WM in fish tissues. In order to fulfill consultation obligations under the federal Endangered Species Act (ESA; 16 U.S.C. 1531–1544, as amended) stemming from promulgation of the California Toxics Rule in 2000, the U.S. EPA agreed to propose this human health criterion in California, with the assurance that the criterion should be sufficient to protect federally listed wildlife species in California (U.S. FWS 2003). Two approaches were used by the U.S. FWS (2003) to evaluate the level of protection that the 0.3 mg/kg MeHg criterion would afford Bald Eagles, a federally listed threatened species that has been nesting at Clear Lake since the 1980s and reproducing there since the 1990s: (1) an "average concentration trophic level approach," which assumed the 0.3 mg/kg criterion represented the average concentration in the overall Bald Eagle diet, and (2) a "highest trophic level

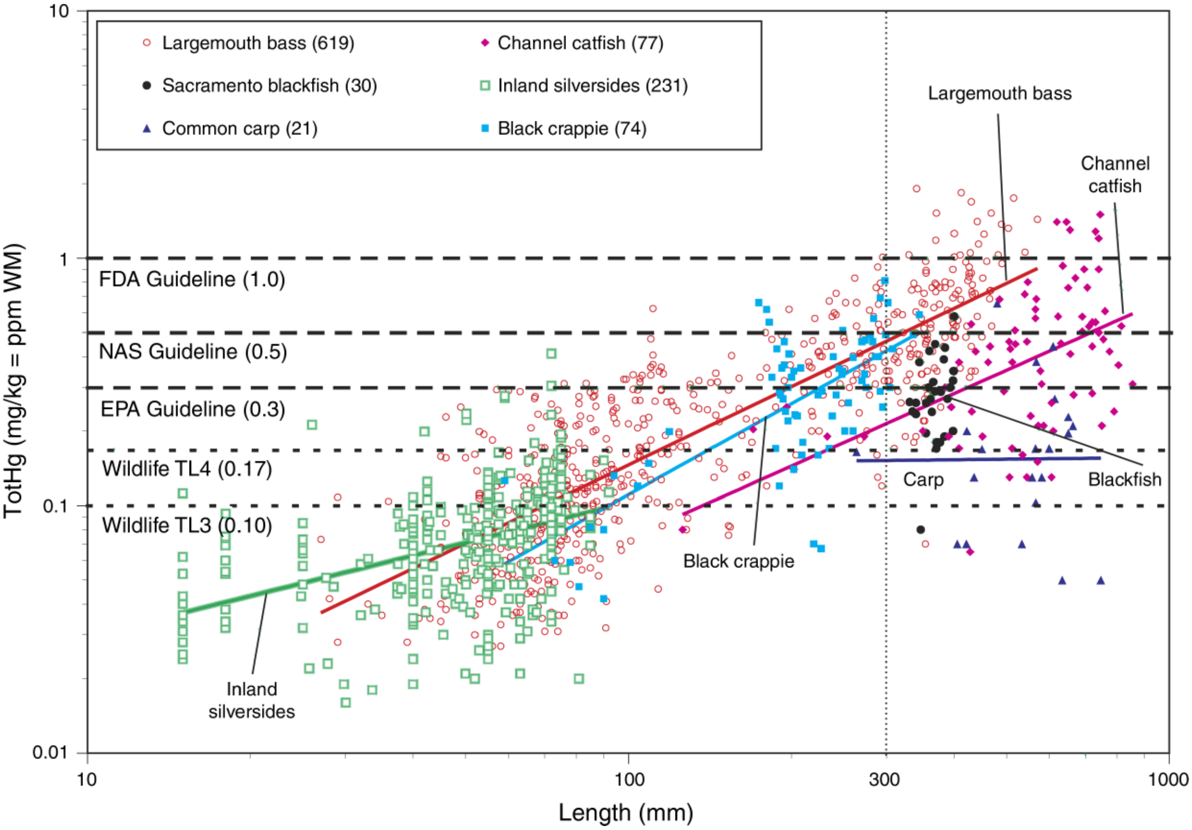

FIG. 9. Total mercury (TotHg) vs. length in six species of fishes from Clear Lake (data from the period 1976–2004) in relation to human health consumption guidelines and wildlife criteria (see *Discussion and conclusions: Human health criteria* and *Wildlife health criteria*). The values in parentheses in the key are sample sizes. The fitted lines are power curves. The fitted line for Sacramento blackfish is not shown because of a lack of sufficient data points over a sufficient size range. The vertical line at 300 mm indicates typical lower size limit for human consumption of largemouth bass and channel catfish. Abbreviations are: FDA, Federal Drug Administration; NAS, National Academy of Sciences; EPA, U.S. Environmental Protection Agency; Wildlife TL4, wildlife criterion for trophic level 4 fish; Wildlife TL3, wildlife criterion for trophic level 3 fish.

approach," which assumed the 0.3 mg/kg criterion represented the maximum concentration in trophic level 4 fish prey consumed by eagles. Both approaches assumed that Bald Eagles consume a variety of fish species from different trophic levels, with large-sized individuals of species such as largemouth bass and channel catfish occupying the top "trophic level 4" position. Based on these approaches, the U.S. FWS concluded that only by applying the human health criterion using the highest trophic level approach (i.e., 0.3 mg/kg in trophic level 4 fish) would the Bald Eagle be adequately protected. However, based on additional calculations from the derivation of wildlife targets for a TMDL in the nearby Cache Creek watershed, the U.S. FWS (2004) determined that MeHg concentrations in trophic level 3 and trophic level 4 fish (>150 mm length) would need to be 0.12 mg/kg and 0.20 mg/kg, respectively, in order to provide adequate protection for Bald Eagles. This conclusion resulted from both a refinement of the original methodology and site-specific data on fish tissue concentration relationships between trophic levels. In addition to these findings, MeHg concentrations that would be protective of Osprey, a common piscivorous bird species nesting and reproducing at Clear Lake (see Anderson et al. 2008), were calculated at 0.10 mg/kg and 0.17 mg/kg for trophic level 3 and trophic level 4 fish, respectively. Based on these calculations, nearly all of the largemouth bass and channel catfish at Clear Lake would exceed tissue Hg concentrations that would be protective of Bald Eagles and Osprey (see Fig. 9).

The big picture

A summary of Hg concentrations in six common fish species from Clear Lake (Fig. 9) constitutes the bulk of our data collection efforts from 1992 to 2004. This combines data from all arms over all years and provides a more global view of the range of TotHg values for each species in relation to the various human health consumption guidelines and wildlife criteria. It is clear that the bulk of the fish sampled from Clear Lake, including largemouth bass and channel catfish, fall below the historical U.S. FDA guideline of 1.0 mg/kg Hg. However, many largemouth bass and channel catfish, especially larger individuals over ~300 mm, exceed the NAS guideline of 0.5 mg/kg Hg. Furthermore, the bulk of the adult largemouth bass (i.e., >150 mm) and channel catfish and many black crappie and Sacramento blackfish exceed the more recently developed U.S. EPA guideline of 0.3 mg/kg Hg, which is also the screening guideline used by the State of California's Office of Environmental Health Hazard Assessment for fish consumption by humans. Only inland silversides (a planktivore) and carp (a detritivore/benthic omnivore) typically remain below the EPA guideline. Inland silversides are not known to be consumed by humans, but there is a commercial fishery for carp on Clear Lake that typically serves the San Francisco Asian market. In terms of the size that humans typically capture and eat, the lower legal fishing size limit for largemouth bass in California is 305 mm (Davis et al. 2000) and for both largemouth bass and channel catfish, most that are consumed are larger than ~300 mm. Thus, the vast majority of fish collected at Clear Lake that are typically consumed by humans exceed the U.S. EPA human health consumption guideline of 0.3 mg/kg WM. In addition, other edible Clear Lake fish species, such as the Sacramento blackfish and black crappie, often exceed the U.S. EPA guideline and sometimes exceed the NAS guideline. Only inland silversides and carp fall primarily below the EPA guideline for human consumption, but many of the inland silversides and carp exceed the wildlife criteria for trophic level 3 fish. While these Hg concentrations exceed many of the established fish consumption guidelines, they are also typical for other sites within the United States and specifically the California region, except for extremely contaminated sites. Based on ecotoxicological risk assessments, fish tissues in Clear Lake typically exceed Hg criteria that would be protective of human health and wildlife health.

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SUPPLEMENT

Total mercury and methylmercury concentrations for 16 species of fishes from Clear Lake, California, USA, during the years 1976–2004 (*Ecological Archives* A018-077-S1).