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Mercury concentrations in fish from a Sierra Nevada foothill reservoir located downstream from historic gold-mining operations

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Abstract This study examined mercury concentrations in whole fish from Camp Far West Reservoir, an 830-ha reservoir in northern California, USA, located downstream from lands mined for gold during and following the Gold Rush of 1848–1864. Total mercury (reported as dry weight concentrations) was highest in spotted bass (mean, 0.93 μg/g; range, 0.16–4.41 μg/g) and lower in bluegill (mean, 0.45 μg/g; range, 0.22–1.96 μg/g) and threadfin shad (0.44 μg/g; range, 0.21–1.34 μg/g). Spatial patterns for mercury in fish indicated high concentrations upstream in the Bear River arm and generally lower concentrations elsewhere, including downstream near the dam. These findings coincided with patterns exhibited by methylmercury in water and sediment, and suggested that mercury-laden inflows from the Bear River were largely responsible for contaminating the reservoir ecosystem. Maximum concentrations of mercury in all three fish species, but especially bass, were high enough to warrant concern about toxic effects in fish and consumers of fish.

Keywords Spotted bass · Bluegill · Threadfin shad · Mercury · Camp Far West Reservoir · Bear River · California

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Introduction

Mercury contamination from gold mining operations dating back to the California Gold Rush of 1848–1864 is believed to be widespread in many rivers, lakes, and reservoirs on the western slopes of the Sierra Nevada (Alpers et al. 2005). Miners used elemental mercury (quicksilver) to recover gold from placer (alluvial) mines, which used hydraulic, drift, and dredging methods, and from hardrock (lode) mines. At hydraulic mining operations, which began in 1852 and peaked in 1880 (Craig and Rimstidt 1998), placer ores were eroded with monitors (water cannons) and the resulting slurry was directed through sluices and drainage tunnels where the gold particles were combined with liquid mercury to form gold–mercury amalgam. Bowie (1905) estimated that 10–30% of the mercury used in this process

was lost each season, resulting in highly contaminated sediments downstream from the mines. According to Alpers et al. (2005), the annual loss of mercury from a typical sluice was likely several hundred kilograms during the operating season or roughly 1,400–4,000 metric tons statewide. Although mercury was used in drift mining operations, in dredging operations, and at hardrock mines, which grew in importance after hydraulic mining came under control of the California Debris Commission in 1893 and placer deposits were exhausted (Craig and Rimstidt 1998), the amounts lost to the environment by these sources have not been estimated.

Although mercury was used throughout the northwestern Sierra Nevada, highest average concentrations measured in fish tissues occur in the Bear River and South Yuba River watersheds (Slotton et al. 1997). Judging from limited sampling of aquatic biota mostly above and below selected foothill reservoirs, the reservoirs seemingly serve as traps for both sediment-associated inorganic mercury and biologically available mercury (Slotton et al. 1997). May et al. (2000) reported that mercury concentrations were highest in upper-trophic-level predators (largemouth bass, *Micropterus salmoides*; smallmouth bass, *M. dolomieui*; and spotted bass, *M. punctatus*) from Camp Far West Reservoir and Lake Combie on the Bear River and Lake Englebright on the South Yuba River, with lower concentrations occurring in benthic omnivores (channel catfish, *Ictalurus punctatus*) and intermediate-trophic-level predators (bluegill, *Lepomis macrochirus*; green sunfish, *L. cyanellus*; and black crappie, *Pomoxis nigromaculatus*). Moreover, within Camp Far West Reservoir, 14 spotted bass measuring 315–444 mm total length (TL) contained 0.58–1.5 μg Hg/g (wet weight basis) in skinless fillets (May et al. 2000). In addition, one largemouth bass measuring 387 mm TL contained 0.81 μg Hg/g, three channel catfish measuring 437–479 mm TL contained 0.51–0.75 μg Hg/g, and three bluegill measuring 159–175 mm TL contained 0.22–0.34 μg Hg/g, all in skinless fillets (May et al. 2000).

Our study was intended to verify the preliminary findings of May et al. (2000) and better understand the extent and severity of mercury contamination in fish from Camp Far West Reservoir. Specific objectives were as follows: (1) determine if mercury concentrations varied spatially and temporally in selected fish species and (2) determine if mercury concentrations exceeded toxic threshold levels or related criteria for piscivorous fish and wildlife, and human consumers. The results were also intended to contribute towards a multidisciplinary federal investigation of mercury uptake and cycling within the reservoir, and help to identify mercury "hot spots" in the Bear–Yuba river watersheds for possible future remedial action (e.g., Kuwabara et al. 2003; Alpers et al. 2008; Stewart et al. 2008).

Study area and methods

Camp Far West Reservoir is an 830-ha reservoir located about 52 km north of Sacramento and 31 km southeast of Yuba City in Nevada, Placer, and Yuba counties, California (Fig. 1). The reservoir, which was constructed in 1963, is used mostly for storage of irrigation water by the South Sutter Water District (http://cdec.water.ca.gov/cgi-progs/damMeta?dam_id=202, accessed 30 January 2009). Inflow into Camp Far West Reservoir originates from a 741 km^2 drainage basin, mostly from the Bear River with smaller inflows from Rock Creek and other sources. Surface-water characteristics measured at monthly or bi-monthly intervals from May 2002 to August 2003 varied as follows (values are minima–maxima): temperature, 7.3–28.9°C; dissolved oxygen, 4.5–12.3 mg/L; pH, 6.6–8.5; specific conductance, 0.02–0.69 μmhos/cm @ 25°C; and turbidity, 0–656 nephelometric turbidity units (NTUs; turbidities >90 NTUs were measured only during a period of extreme reservoir drawdown in October 2002; M.K. Saiki, unpublished data).

The primary source of mercury to Camp Far West Reservoir is suspected to be transport of contaminated sediments from upstream reaches of the Bear River, especially during high-flow events (Kuwabara et al. 2003; Alpers et al. 2008). Historically, hundreds of gold mining operations occurred in the Bear River watershed upstream

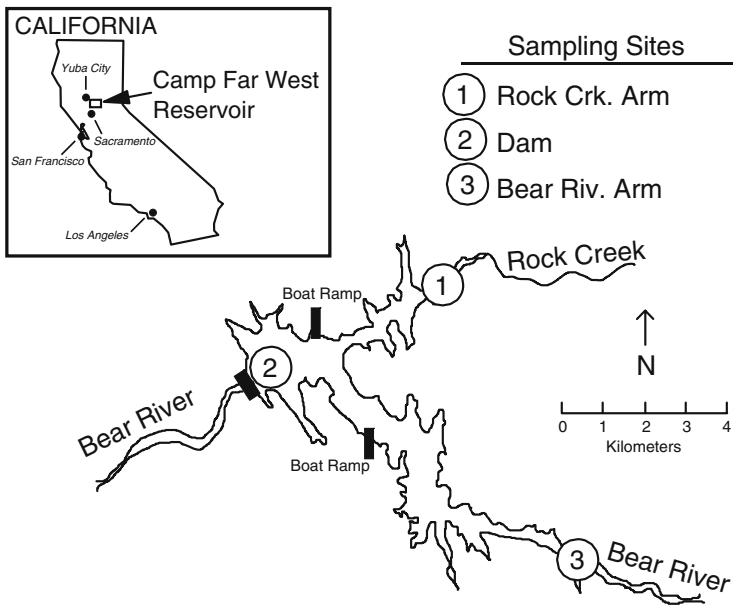

Fig. 1 Map of the study area

from the reservoir, each contributing to mercury loss during the gold-recovery process. Although data for the Rock Creek watershed are not available, its relatively small size was sufficient to support only a few gold-mining operations, probably leading to much lower inputs of mercury into the reservoir.

Fish collections

Spotted bass, bluegill, and threadfin shad (*Dorosoma petenense*) were sampled from Camp Far West Reservoir at three localities as follows: the Bear River arm, the Rock Creek arm, and the lower reservoir adjacent to the dam (Fig. 1). Sampling efforts focused on shallow shoreline areas of the reservoir where fish were thought to be most numerous. All sampling occurred during August 2002 and August 2003.

Fish were collected with a boat-mounted electroshocker. Sampling effort was not recorded because the goal was to fulfill specified quotas (either numbers of individuals or total biomass) of fish.

Captured fish were measured for TL and weight, then individually wrapped and bagged in plastic and chilled on wet ice. Within 12 h after returning from the field, the gastrointestinal tracts (esophagus to pyloric sphincter; however, only esophagus in threadfin shad) were opened by dissection to remove gut contents (food items), then most fish samples were rewrapped and bagged in plastic, and frozen (−10°C). Gut contents were removed to reduce this potential source of variation in whole-fish mercury measurements and to characterize the forage items consumed by the fishes (fish gut contents are reported by Stewart et al. 2008). Bluegill less than 53 mm TL and threadfin shad less than 60 mm TL were grouped into composite samples (two to four fish per composite) to yield sufficient biomass for determinations of mercury and other measurements (e.g., moisture content and stable isotope ratios; stable isotope ratios are reported by Stewart et al. 2008).

In 2002, 20 spotted bass and 15 bluegill were selected at random for dissection and removal of skinless and boneless fillets from both sides of each fish. After weighing, the fillets were wrapped and bagged in plastic, then frozen. The remaining carcass (including the dissected gastrointestinal tract) of each fish was weighed, then wrapped and bagged in plastic and frozen. Separate measurements of mercury in fish fillets and their carcasses were used to generate predictive equations for converting mercury concentrations in whole fish to mercury concentrations in fillets.

Determination of moisture content, total mercury, and methylmercury

All samples of fish were analyzed for moisture percentage and total mercury concentration. In addition, 15 whole-body samples of spotted bass and ten whole-body samples each of bluegill and threadfin shad were randomly selected from fish collected in 2002 for analysis of methylmercury concentration. Frozen samples were shipped overnight from the field station to analytical facilities at the Columbia Environmental Research Center (CERC), Columbia, Missouri, for further sample preparation and chemical analysis. Partially thawed fish samples were initially either minced with a titanium meat cleaver (samples weighing <100 g) or chopped with a meat cleaver and ground with a Hobart or Kitchenaid meat grinder (samples weighing >100 g), then lyophilized with a Virtis Genesis 35EL freeze dryer for moisture determination. After lyophilization, each dried fish sample was cryogenically ground to a fine powder with a Spex 6850 Freezer/Mill, then relyophilized overnight to remove residual moisture accumulated during cryogrinding. Dried samples were stored in glass vials in a desiccator while awaiting further processing.

Total mercury was determined with a direct mercury analyzer wherein a dried sample (usually 30–100 mg) was combusted in a stream of oxygen. All mercury in the sample was volatilized and trapped by amalgamation on a gold substrate, then thermally desorbed and quantified by atomic absorption spectrophotometry. The entire sequence was conducted with a Milestone DMA-80 analyzer equipped with an automated sample carousel. For spotted bass and bluegill, total mercury was usually determined from a single analysis of each sample. For threadfin shad, total mercury was usually determined from an average of two analyses of each sample.

After analysis for total mercury, samples designated for methylmercury analysis were shipped from CERC to analytical facilities at Brooks Rand LLC, Seattle, Washington. Typically, 100 mg of dried sample was digested by an alkaline KOH/ethanol procedure, with a final digestate volume of 2.5 mL. Thirty microliters of this digestate was then subjected to aqueous phase ethylation, purge and trap, followed by GC separation, isothermal decomposition, and analysis by atomic fluorescence detection.

Quality control for both total mercury and methylmercury included blanks, replicates, pre-combustion or predigestion spikes, and tissue reference materials. An independent calibration verification standard for total mercury was analyzed at the beginning and end of each instrumental run to confirm the calibration status of the Milestone DMA-80 analyzer system. With one exception, the percent errors were well within 10% for 37 measurements of reference solutions used to verify instrument calibration during analysis of total mercury; the exceptional measurement was 11.6%. Recoveries of reference solutions used for calibration during methylmercury determination varied from 79% to 113%, and averaged 96%. All analyses ($N = 30$) of National Research Council Canada dogfish muscle certified reference material (DORM-2) for total mercury were within the certified range of 4.64 ± 0.26 μg/g dry weight (hereinafter, unless indicated otherwise, all mercury concentrations are reported as dry weights). This same material (dogfish muscle) analyzed for methylmercury ($N = 10$) exhibited recoveries varying from 72% to 120%, and averaging 86%. Method precision for total mercury, determined as percent relative standard deviation (%RSD) from triplicate combustion, amalgamation, and analysis of fish tissue samples, was ≤10% except for one fish sample (20%). Method precision for methylmercury in fish varied from 1.3% to 11% relative percent difference (%RPD). Percent recovery of total mercury from pre-combustion tissue spikes ($N = 56$) varied from 77% to 117%, and averaged 99%. Recoveries of methylmercury from predigestion spikes varied from 78% to 105%, and averaged 88%. Total mercury blank equivalent concentrations were less than the method detection limits (MDLs) for 22 of 30 sample blocks or groups, but at or slightly above the MDLs for the remaining eight blocks. For total mercury, the MDLs varied from 0.0002 to 0.012 μg/g, and averaged 0.0025 μg/g, and the quantitation limits varied from 0.001 to 0.027 μg/g, and averaged 0.010 μg/g. For methylmercury, the MDLs averaged 1.5 ng/g, and the quantitation limits averaged

Table 1 Summary characteristics, moisture content, methylmercury, and total mercury concentration in three fish species sampled from Camp Far West Reservoir during August 2002 and 2003

Fish species	*N*	Total length (mm)	Weight (g)	Moisture (%)	Whole-fish methylmercury (μg/g dry wt.)[a]	Whole-fish total mercury (μg/g dry wt.)
Spotted bass	180	180 (49–443)	64.1 (0.97–1224.0)	73.6 (66.5–79.5)	0.72 (0.25–1.95)	0.93 (0.16–4.41)
Bluegill	120	100 (34–189)	16.1 (0.51–135.5)	73.9 (70.9–76.9)	0.39 (0.27–0.64)	0.45 (0.22–1.96)
Threadfin shad	104	68 (27–121)	2.54 (0.12–14.0)	77.7 (72.0–82.4)	0.36 (0.19–0.73)	0.44 (0.21–1.34)

Except for moisture, values are geometric means (ranges in *parentheses*). For moisture, values are back-transformed angular means (ranges in *parentheses*)
[a]Methylmercury determinations were based on a subset of samples used for total mercury determinations. For methylmercury determinations, sample sizes were as follows: spotted bass, $N = 15$; bluegill, $N = 10$; and threadfin shad, $N = 10$

Table 2 Relation between total length and weight of threadfin shad, bluegill, and spotted bass, and whole-fish measurements of moisture content, methylmercury concentration, and total mercury concentration

Fish species	Vital statistic	Moisture content (%)			Methylmercury (μg/g dry wt.)			Total mercury (μg/g dry wt.)		
		r	*P*	*N*	*r*	*P*	*N*	*r*	*P*	*N*
Spotted bass	Total length	−0.81*	< 0.0001	180	0.95*	< 0.0001	15	0.89*	< 0.0001	180
	Weight	−0.81*	< 0.0001	180	0.95*	< 0.0001	15	0.89*	< 0.0001	180
Bluegill	Total length	−0.28*	0.0021	120	−0.11	0.7552	10	0.45*	< 0.0001	120
	Weight	−0.28*	0.0020	120	−0.15	0.6860	10	0.44*	< 0.0001	120
Threadfin shad	Total length	−0.43*	< 0.0001	104	0.90*	0.0003	10	0.66*	< 0.0001	104
	Weight	−0.47*	< 0.0001	104	0.90*	0.0005	10	0.64*	< 0.0001	104

*Statistically significant according to the Bonferroni $P = 0.0028$ for 18 simultaneous comparisons

Table 3 Tests for homogeneity of slopes from regression equations that predict total mercury concentration as a function of total length in spotted bass, bluegill, and threadfin shad

Taxa	Source	*df*	*MS*	*F*	*P*
Spotted bass	Interaction	5	0.0162	0.97	0.4375
	Error	168	0.0167		
Bluegill	Interaction	5	0.0629	3.58	0.0049
	Error	108	0.0176		
Threadfin shad	Interaction	5	0.0275	3.17	0.0110
	Error	92	0.0087		

4.2 ng/g. Overall, these quality control results were within acceptable limits as specified by CERC.

Data analysis

Computerized databases were created as Excel spreadsheets. Raw data were analyzed by using SAS/STAT® and SAS/GRAPH® (SAS Institute Inc., Cary, NC, USA). Parametric techniques—e.g., Pearson product-moment correlation, analysis of variance (ANOVA), and analysis of covariance (ANCOVA)—were used to summarize and interpret statistically significant relations among variables such as mercury body burdens and total length or weight of fish, and temporal–spatial variations. For ANCOVA, we used fish length as the covariate when comparing mercury concentrations because length does not decrease over time whereas weight sometimes does (Huckabee et al. 1979). To ensure normality, data were routinely subjected to standard transformations (e.g., angular transformation for moisture percentage; logarithmic transformation for length, weight, and mercury concentration). However, exponential functions were used when fitting curves to scatter plots of fish length and mercury concentrations because they yielded the largest coefficient of determination (R^2) values. Prior to conducting ANCOVA, we tested the assumption that regression coefficients were similar (homogeneous) for the variables under comparison. If slopes were not homogeneous, we used a variation of ANCOVA referred to as "extra sums of squares." The "extra sums of squares" procedure measures the marginal reduction in the error sums of squares when one or several independent variables are added to the regression model, given that other independent variables are already in the model (Neter et al. 1990). Unless specified otherwise, the level of significance for all statistical tests was $P = 0.05$.

Results

A total of 404 whole-fish samples were measured for moisture content and total mercury concentration during this study (Table 1). In addition, some whole-fish samples (15 spotted bass, ten bluegill, and ten threadfin shad) were also measured for methylmercury. The whole-fish results included 35 measurements (20 spotted bass and 15 bluegill) that were estimated from analysis of skinless fillets and their corresponding carcasses (whole fish less skinless fillets). Although moisture content was variable, threadfin shad generally exhibited the highest moisture content, with lower percentages measured in spotted bass and bluegill (Table 1). In addition, moisture content was inversely associated with fish size (TL and weight) in all three species (Table 2). On average, methylmercury and total mercury concentrations were highest in spotted bass and lower in bluegill and threadfin

Table 4 Results of analysis of covariance (ANCOVA), as *F* values and significance levels, for total mercury concentrations (dry weight basis, adjusted for total length) in spotted bass collected from Camp Far West Reservoir during July 2002 and July 2003[a,b]

Source	*df*	*F*	*P*
Total length	1	729.61	< 0.0001
Site	2	4.63	0.0110
Year	1	0.98	0.3225
Site × year interaction	2	0.91	0.4030
Error MS	173	0.0167	–

[a]Before ANCOVA was conducted, regression equations were determined to exhibit homogeneous slopes (see Table 3)

[b]Bass used in total mercury determinations averaged 180 mm TL

shad (Table 1). Moreover, methylmercury and total mercury concentrations increased as TL and weight of spotted bass, bluegill (total mercury only), and threadfin shad increased (Table 2).

Methylmercury concentrations were strongly associated with total mercury concentrations in all three fish species (for spotted bass, $N = 15$, $r = 0.99$, $P < 0.0001$; for bluegill, $N = 10$, $r = 0.88$, $P = 0.0009$; for threadfin shad, $N = 10$, $r = 0.96$, $P < 0.0001$). Although linear regression equations describing the methylmercury:total mercury relations in each species exhibited similar slopes ($F_{2,29} = 1.15$, $P = 0.3321$), the intercepts were significantly different ($F_{2,31} = 5.31$, $P = 0.0104$). Moreover, the ratios of methylmercury to total mercury varied significantly among the three fish species ($F_{2,32} = 4.16$, $P = 0.0248$). This variation was due to relatively high ratios measured in bluegill (0.93, 0.84–1.04; values are geometric mean and 95% confidence interval) and lower ratios measured in threadfin shad (0.78, 0.71–0.86). The methylmercury:total mercury ratios for spotted bass (0.87, 0.81–0.93) overlapped those of bluegill and threadfin shad.

Spatial and temporal variations in total mercury concentrations

The significant associations between total mercury concentration and fish size (TL and weight) of spotted bass, bluegill, and threadfin shad required use of ANCOVA to assess variations in mercury concentrations over the three sites and 2 years encompassed by this study. Prior to conducting ANCOVA, slopes of the mercury vs. total length relationship for each species were tested for homogeneity (Table 3). According to this assessment, the standard ANCOVA procedure was applicable only to spotted bass (i.e., we accepted the hypothesis that slopes were the same for each regression line). Slopes were not homogeneous for bluegill and threadfin shad, necessitating use of an "extra sums of squares" version of ANCOVA.

Total mercury concentrations in spotted bass varied among sites but not years (Table 4). Mercury concentrations (computed as least-squares means for spotted bass averaging 180 mm TL) were highest in the Bear River arm (0.991 μg Hg/g), slightly lower at the dam (0.966 μg Hg/g, but the difference was not significantly different from the Bear River arm), and significantly lower in the Rock Creek arm (0.850 μg Hg/g).

In bluegill, total mercury concentrations varied over both sites and years (Table 5). Mercury concentrations (computed as least-squares means for bluegill averaging 100 mm TL) were highest in the Bear River arm (0.552 μg Hg/g), intermediate at the dam (0.423 μg Hg/g), and lowest in the Rock Creek arm (0.353 μg Hg/g), with each mean value differing significantly from the other two mean values. In addition, mean mercury concentrations were significantly higher in 2002 (0.459 μg Hg/g) than in 2003 (0.413 μg Hg/g).

Table 5 Results of analysis of covariance (ANCOVA), as F values and significance levels, for total mercury concentrations (dry weight basis, adjusted for total length) in bluegill and threadfin shad collected from Camp Far West Reservoir during July 2002 and July 2003

Taxa	Source	df	F	P
Bluegill	Total length	6	9.71	< 0.0001
	Site	8	6.13	< 0.0001
	Year	6	3.67	0.0024
	Site × year interaction	4	0.65	0.6250
	Error SS	108	1.90	–
Threadfin shad	Total length	6	48.42	< 0.0001
	Site	8	12.53	< 0.0001
	Year	6	14.32	< 0.0001
	Site × year interaction	4	4.48	0.0024
	Error SS	92	0.80	–

Bluegill averaged 100 mm TL, whereas threadfin shad averaged 68 mm TL. The "extra sums of squares" procedure was used to compute ANCOVA results because regression equations exhibited unequal (non-homogeneous) slopes (see Table 3)

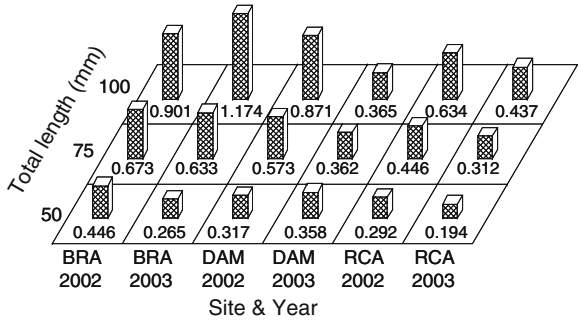

Fig. 2 Total mercury concentrations (μg/g, dry weight basis) in small (50 mm TL), medium (75 mm TL), and large (100 mm TL) threadfin shad captured from the Bear River arm (*BRA*), the Dam (*DAM*), and the Rock Creek arm (*RCA*) during August 2002 and August 2003. All values were computed as least-squares means

Total mercury concentrations in threadfin shad exhibited a significant site × year interaction (Table 5). Moreover, due to a significant total length × site × year interaction ($F_{2,92} = 6.25$, $P = 0.0029$) in the full ANCOVA model, the site × year interaction was not constant over the entire range of total length values (i.e., the relationship between mercury concentration and total length varied in shad captured from different sites and years). Thus, although mercury concentrations in all sizes of shad were generally highest in the Bear River arm, intermediate at the dam, and lowest in the Rock Creek arm, and higher in 2002 than in 2003, several exceptions were present. For example, in 2003, mercury concentrations were highest at the dam, intermediate in the Bear River arm, and lowest in the Rock Creek arm for small (50 mm TL) shad, and highest in the Bear River arm, intermediate in the Rock Creek arm, and lowest at the dam for large (100 mm TL) shad (Fig. 2). In addition, mercury concentrations were lower in 2002 than in 2003 for small shad at the dam and large shad in the Bear River arm (Fig. 2).

Total mercury concentrations in whole fish and fillets

To address human health concerns, 20 samples of spotted bass and 15 samples of bluegill were analyzed for total mercury as skinless fillets and carcasses (whole fish less the skinless fillets) to generate simple equations that predicted mercury concentrations in fillets when given whole-fish concentrations. For spotted bass, the relation

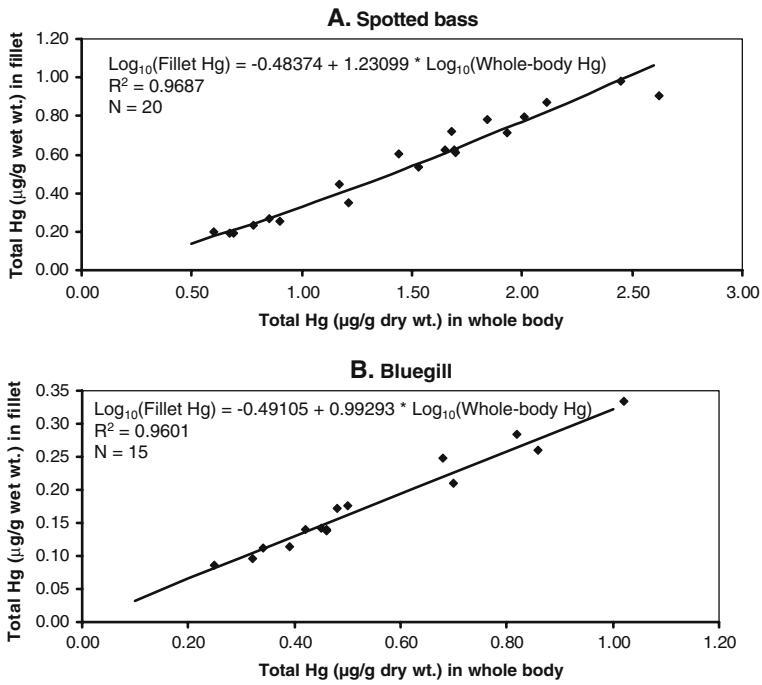

Fig. 3 Relation between total mercury concentrations in whole-fish samples and skinless fillets of spotted bass and bluegill sampled from Camp Far West Reservoir during August 2002

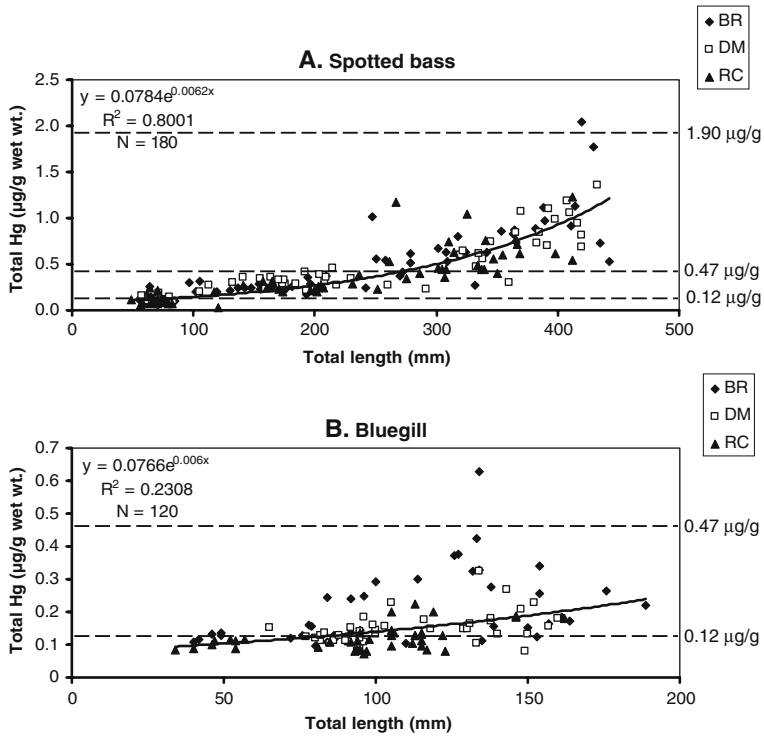

Fig. 4 Mercury concentrations in skinless fillets (estimated from whole-fish concentrations with equations given in Fig. 3) of spotted bass and bluegill from three localities (*BR*, Bear River arm; *DM*, dam; and *RC*, Rock Creek arm) in Camp Far West Reservoir. Also shown are fish tissue concentration thresholds (*horizontal dashed lines*) associated with a national advisory for noncommercial fish that recommended consumption limits of 4 meals/month (0.12 µg Hg/g), 1 meal/month (0.47 µg Hg/g), and 0 meals/month (1.90 µg Hg/g)

was described by the equation, $\mathrm{Log}_{10}(\mathrm{FC}) = -0.48374 + 1.23099 \times \mathrm{Log}_{10}(\mathrm{WC})$, where FC is fillet concentration (wet weight basis) and WC is whole-fish concentration (dry weight basis; see Fig. 3). For bluegill, the relation was described by the equation, $\mathrm{Log}_{10}(\mathrm{FC}) = -0.49105 + 0.99293 \times \mathrm{Log}_{10}(\mathrm{WC})$ (Fig. 3). According to these equations, the wet weight concentrations of mercury in fillets of spotted bass varied from 0.03 to 2.04 µg/g, whereas those of bluegill varied from 0.07 to 0.63 µg/g.

Discussion

Several investigators (Grieb et al. 1990; Bloom 1992; Wiener and Spry 1996) reported that approximately 95–99% of mercury in fish muscle tissues occurs as methylmercury, although at least one study (e.g., Mason et al. 2006) found methylmercury concentrations averaging only 28 ± 14% in axial muscle of planktivorous white perch (*Morone americana*) and 65 ± 22% in axial muscle of mostly piscivorous striped bass (*Morone saxatilis*) from tidal portions of Chesapeake Bay. Data from our study indicated that methylmercury averaged 78–93% of total mercury measured in whole fish, with threadfin shad containing the lowest percentages and spotted bass and bluegill containing higher percentages. Goldstein et al. (1996) reported that total mercury concentrations were higher in muscle tissue than in the corresponding whole body for common carp (*Cyprinus carpio*) and channel catfish (*Ictalurus punctatus*). These results suggest that many non-muscle tissues (e.g., bones, scales, skin) present in samples of whole fish may contain disproportionately low concentrations of methylmercury. However, as noted by Goldstein et al. (1996), the ratio of methylmercury:total mercury in internal organs such as liver and the whole fish can vary among fish species, which might reflect physiological differences among different trophic groups.

Although fish are exposed to mercury from both water and food, bioaccumulation of mercury through the food chain plays a more important role in determining mercury burdens (Hall et al. 1997; Wiener et al. 2003). Fish probably assimilate 65–80% or more of the methylmercury present in the food they eat and eliminate methylmer-

cury slowly relative to the rate of uptake (Wiener et al. 2003), resulting in a net increase in mercury burdens. This is one reason that mercury concentrations typically increase with size and age of the organism (Lange et al. 1993, 1994; Wiener et al. 2003). In Camp Far West Reservoir, mercury concentrations in spotted bass, bluegill, and threadfin shad also increased with TL and weight (Table 2; also Fig. 4).

Another reason that mercury concentrations often increase as fish get larger (older) is due to ontogenetic shifts in diet, especially in predatory species that initially feed on small lower-trophic-level invertebrates as larvae and juveniles, but switch to large higher-trophic-level fish as subadults and adults. For example, MacCrimmon et al. (1983) observed increased rates of mercury accumulation in lake trout (*Salvelinus namaycush*) when the young switched from a diet of invertebrates to forage fish. Wren and MacCrimmon (1986) also observed that piscivorous fish had higher concentrations of mercury than prey fish of comparable age. In Camp Far West Reservoir, top-level predators such as spotted bass had higher concentrations of mercury than did lower-trophic-level insectivores and planktivores such as bluegill and threadfin shad (Table 1). Moreover, smaller (younger) bass, which generally feed on zooplankton and small insects, contained lower concentrations of mercury than did larger (older) bass that feed primarily on large-bodied invertebrates (e.g., crayfish) and forage fish (Stewart et al. 2008). By comparison, mercury concentrations in bluegill and threadfin shad were weakly correlated with body size possibly because juveniles and adults of these species tend to forage on similar foods (e.g., phytoplankton, vascular plants, zooplankton, immature aquatic insects, and other benthic invertebrates; Stewart et al. 2008). Unlike adult spotted bass, bluegill and threadfin shad are seldom piscivorous.

The spatial patterns observed for mercury in spotted bass and bluegill, and to a lesser extent in threadfin shad, indicated highest concentrations in the Bear River arm of Camp Far West Reservoir, followed by the dam, and lastly by the Rock Creek arm. These patterns were consistent with methylmercury concentrations measured in water samples filtered through quartz fiber filters (pore size, 0.7 μm) on as many as eight occasions during 2001–2003, wherein the Bear River arm contained <0.04–0.32 ng/L, the dam contained <0.04–0.06 ng/L, and the Rock Creek arm contained <0.04–0.04 ng/L (Alpers et al. 2008). Methylmercury concentrations in particulate matter extracted from the water samples also showed highest concentrations in the Bear River arm (<0.029–0.39 ng/L), followed by the dam (<0.029–0.097 ng/L), and lastly by the Rock Creek arm (<0.029–0.051 ng/L; Alpers et al. 2008). Although data were not available from the Rock Creek arm, methylmercury concentrations measured during 2001–2003 in sediment samples from the Bear River arm (1.37–7.73 ng/g dry weight) were higher than from the dam (0.61–0.93 ng/g dry weight; M. Marvin-DiPasquale, U.S. Geological Survey, Menlo Park, California, unpublished data).

Mercury burdens in centrarchids (i.e., spotted bass and bluegill) from our study, which are known to exhibit restricted home ranges or site fidelity (Ball 1947; Fish and Savitz 1983; Horton 2000), probably reflect local exposure conditions wherein the primary source of mercury contamination was associated with inflows from the Bear River. However, we could not explain the higher mercury concentrations measured in bluegill during August 2002 than during August 2003 because too few samples of water and sediment were collected to assess annual variations in environmental exposure. Mercury burdens in threadfin shad were characterized by complex spatial and temporal patterns suggestive of highly variable exposure to mercury, such as might occur if the fish showed little or no site fidelity, instead roaming extensively throughout the reservoir. This contention is supported by limited information from other reservoirs that indicate shad are a highly mobile schooling species with patchy spatial distributions influenced diurnally and seasonally by water quality conditions (e.g., temperature and dissolved oxygen) and possibly by predators and prey (Allen and DeVries 1993; Schael et al. 1995).

Several investigators have attempted to establish toxic thresholds of mercury for fish life. According to Niimi and Kissoon (1994), overt effects on fish growth and survival occur at

relatively high concentrations of 10–20 μg Hg/g wet weight (roughly 40–80 μg/g dry weight, assuming 75% moisture) in whole fish, which is rarely encountered under natural conditions. Recently, however, Beckvar et al. (2005) evaluated several approaches for deriving protective (i.e., unlikely to have adverse effects) tissue residue-effect concentrations in fish by using published datasets, and determined that the tissue threshold-effect level (t-TEL) approach best represented available data. According to Beckvar et al. (2005), a whole-fish mercury t-TEL of 0.2 μg/g wet weight (roughly 0.8 μg/g dry weight, assuming 75% moisture) based largely on sublethal endpoints such as growth, reproduction, development, and behavior should protect juvenile and adult fish. During our study, total mercury concentrations exceeding 0.8 μg/g occurred in 60% (108 of 180) of spotted bass, 11% (13 of 120) of bluegill, and 11% (11 of 104) of threadfin shad, raising the possibility that some fish from Camp Far West Reservoir might contain sufficiently elevated body burdens of mercury to experience adverse ecotoxicological effects. Laboratory studies indicate that sublethal effects of mercury toxicity in fish can include suppression of sex hormones, altered reproductive behavior, and impaired reproduction, along with maternal transfer of potentially toxic doses of mercury to fish embryos during oogenesis (Scheuhammer et al. 2007).

Toxic thresholds for mercury in diets of piscivorous birds and mammals have been proposed by several investigators. For example, the common loon (*Gavia immer*), which can occur in northern California during winter months (see http://www.mbr-pwrc.usgs.gov/bbs/htm96/cbc622/ra0070.html, accessed 30 January 2009), is especially susceptible to mercury toxicity. Adult loon diets containing >0.3 μg MeHg/g wet weight (about 1.2 μg/g dry weight, assuming 75% moisture) were associated with severely reduced reproductive success, mainly due to decreased egg laying and territorial fidelity in breeding adults (Barr 1986). Although loon chicks did not exhibit overt signs of toxicosis or significant reductions in growth or food-consumption rates when fed daily from hatch through day 105 on fish diets containing as much as 1.2 μg/g wet weight as methylmercury chloride (Kenow et al. 2003), there was evidence of reduced immune response and histological changes (central nervous system demyelination) in chicks receiving 0.4 μg MeHg/g wet weight, or roughly 1.6 μg/g dry weight (assumes 75% moisture; Meyer 2006, cited by Scheuhammer et al. 2007). In mammals such as American mink (*Mustela vison*) and river otter (*Lutra canadensis*), which are permanent residents of northern California (for mink distribution in California, see http://www.sibr.com/mammals/M158.html, accessed 30 January 2009; for otter distribution, see http://www.sibr.com/mammals/M163.html, accessed 30 January 2009), consumption of fish containing methylmercury ≥1 μg/g wet weight (about 4 μg/g dry weight, assuming 75% moisture) has been shown to cause neurotoxicity and death (Wiener et al. 2003 and references therein). These toxic concentrations of mercury were equaled or exceeded by at least some whole-body samples of spotted bass and bluegill (piscivorous birds only), but not threadfin shad, during our study. However, to our knowledge, no one has attempted to document adverse ecotoxicological responses in piscivorous birds and mammals from the Camp Far West Reservoir vicinity.

In 2004, a national advisory for human consumption of mercury-tainted noncommercial fish was jointly issued by the U.S. Food and Drug Administration (FDA) and the U.S. Environmental Protection Agency (EPA; US DHHS and US EPA 2004; for details on how the advisory was developed, see US EPA 2004). This advisory updated an earlier action level of 1.0 μg/g wet weight for methylmercury in fish muscle tissue that the FDA used to regulate the sale of commercially caught fish (US FDA 1994), and a fish tissue criterion of 0.3 μg methylmercury/g wet weight established by the EPA to avoid undesirable neurological abnormalities in human infants exposed *in utero* when pregnant women consume mercury-contaminated foods (US EPA 2001). The updated advisory included several thresholds for mercury concentrations linked to risk-based consumption limits of fish. For comparison with our study, we selected three arbitrary threshold concentrations (0.12, 0.47, and 1.90 μg/g wet weight) corresponding to 4, 1, and 0 meals of 227-g fish servings per

month. In other words, if fish contained 0.12 μg Hg/g, no more than four meals per month should be consumed to ensure a safe level of mercury exposure. However, if fish contained over 1.90 μg Hg/g, none should be consumed.

In Camp Far West Reservoir, spotted bass measuring 69 mm TL were predicted to contain 0.12 μg Hg/g wet weight in their fillets, whereas bass measuring 289 mm TL were predicted to contain 0.47 μg Hg/g wet weight (Fig. 4). Moreover, the largest bass captured during our study (443 mm TL) was predicted to contain 1.22 μg Hg/g wet weight in its fillets. However, at least four smaller fish exceeded this concentration, with one fish (a bass measuring 420 mm TL) containing as much as 2.04 μg Hg/g wet weight. By comparison, bluegill measuring 75 mm TL were expected to contain 0.12 μg/g wet weight in their fillets, with the largest fish (189 mm TL) predicted to contain 0.24 μg/g wet weight (Fig. 4). However, fillet concentrations in at least 15 bluegill exceeded 0.24 μg/g wet weight, with a fish measuring 134 mm TL exhibiting a maximum concentration of 0.63 μg/g wet weight. Nevertheless, our findings indicate that fish (especially bass) from Camp Far West Reservoir exhibit sufficiently elevated concentrations of mercury in their fillets to warrant fish consumption guidelines available in human health advisories. Mercury concentrations in threadfin shad were excluded from similar comparisons because this species is not routinely consumed by humans.

In conclusion, our results indicated that mercury concentrations in spotted bass, bluegill, and threadfin shad varied according to fish size, with higher concentrations occurring in larger (older) fish. Moreover, most of the mercury in whole-fish samples was present as methylmercury. Although variation was present, mercury concentrations were generally highest in fish sampled from the Bear River arm and lower elsewhere, possibly because this portion of the reservoir intercepted most of the bioavailable mercury transported from former gold-mining sites located upstream in the Bear River, along with trapping much of the sediment-associated inorganic load. Maximum concentrations of mercury measured in whole-fish samples of bass, but not bluegill or shad, fell within the lower ranges of concentrations that other investigators proposed as being potentially toxic to fish and to at least some fish-eating birds or mammals. Estimates of mercury concentrations in fillets of bass and bluegill were sufficiently elevated to warrant fish consumption guidelines available in a national human-health advisory. Collectively, these results confirmed the earlier findings of May et al. (2000) that fish from Camp Far West Reservoir were contaminated with undesirably high concentrations of mercury. Largely in response to data from May et al. (2000), the California Office of Environmental Health Hazard Assessment recommended no consumption of "black bass" by women of childbearing age and children 17 years and younger, and a maximum of two meals per month of "black bass" by women beyond childbearing years and men (Klasing and Brodberg 2003). These recommendations are also given in the 2008–2009 Freshwater Sport Fishing Regulations for the State of California (http://www.dfg.ca.gov/regulations/08-09-inland-fish-regs.pdf, accessed 30 January 2009).

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