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PREDICTORS OF MERCURY SPATIAL PATTERNS IN SAN FRANCISCO BAY FORAGE FISH

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Abstract: Pollution reduction efforts should be targeted toward those sources that result in the highest bioaccumulation. For mercury (Hg) in estuaries and other complex water bodies, carefully designed biosentinel monitoring programs can help identify predictors of bioaccumulation and inform management priorities for source reduction. This study employed a probabilistic forage fish Hg survey with hypothesis testing in San Francisco Bay (California, USA). The goal was to determine what pollution sources, regions, and landscape features were associated with elevated Hg bioaccumulation. Across 99 sites, whole-body Hg concentrations in Mississippi silversides (*Menidia audens*) and topsmelt (*Atherinops affinis*) followed a broad spatial gradient, declining with distance from the Guadalupe River (Pearson's $r = -0.69$ and -0.42, respectively), which drains historic mining areas. Site landscape attributes and local Hg sources had subtle effects, which differed between fish species. Topsmelt Hg increased in embayment sites (i.e., enclosed sites including channels, creek mouths, marinas, and coves) and sites with historic Hg-contaminated sediment, suggesting an influence of legacy industrial and mining contamination. In 2008, Mississippi silverside Hg was reduced at sites draining wastewater-treatment plants. Fish Hg was not related to abundance of surrounding wetland cover but was elevated in some watersheds draining from historic Hg-mining operations. Results indicated both regional and site-specific influences for Hg bioaccumulation in San Francisco Bay, including legacy contamination and proximity to treated wastewater discharge. *Environ Toxicol Chem* 2013;32:2728–2737. © 2013 SETAC

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INTRODUCTION

Mercury (Hg) is a global pollutant that adversely impacts ecosystems and human health. Global Hg concentrations are elevated due to widespread human use and inadvertent release, creating a need for coordinated efforts to curtail Hg release, transport, and exposure [1]. Anthropogenic Hg sources to estuarine and coastal ecosystems include runoff from Hg and gold mining operations, atmospheric emissions (e.g., coal combustion), and point sources associated with historic or current industrial activity [1–5]. Methylmercury (MeHg) is highly toxic and bioaccumulative [6], triggering reproductive effects in wildlife [7] and potential developmental and neurological effects in humans [8,9]. At the regional scale, carefully designed research and monitoring are needed to prioritize MeHg management actions in the presence of multiple spatially distributed sources.

Comparative studies of MeHg in forage fish (small, short-lived prey fish, consumed by piscivorous wildlife) aid in describing spatiotemporal patterns and explanatory variables for MeHg food-web accumulation [10–18]. Forage fish integrate exposure across a several-month time period and have limited ranges in age, diet, and movement area [17]. Thus, forage fish are often used to describe spatial patterns in food-web MeHg and the factors that contribute to elevated MeHg [10–13,18]. However, probabilistic spatial surveys and hypothesis-testing approaches are rarely employed to evaluate forage fish contamination within a single water body.

San Francisco Bay (California, USA) is influenced by Hg watershed loads and sediment deposits from historic mining operations and industrial sources, making it an important system for characterizing ecosystem MeHg exposure [3,19–21]. Local sources targeted for management reduction include Hg mines, stormwater runoff from urban and industrial watersheds, municipal publicly owned wastewater-treatment works (POTWs), drainage from the Central Valley watersheds, and industrial facilities [3,22]. Historic sediment contamination also contributes Hg to the water column and food web [23–25]. As in other estuaries, the spatiotemporal dynamics of MeHg concentrations, bioavailability, and bioaccumulation in San Francisco Bay are influenced by complex biogeochemical factors, including variable primary productivity and sulfate reduction, in addition to spatial differences in Hg loading [26–29]. Due to this biogeochemical and spatial complexity, the relative importance of different source categories for Hg bioaccumulation is poorly understood.

In addition to sources, several spatial factors may influence MeHg bioaccumulation within San Francisco Bay. Bay sediment and biota Hg are elevated in proximity to a historic Hg mining district (New Almaden mining district, California, USA), in salt ponds and other semienclosed embayments, and in interior wetlands [3,13,30–32]. Wetlands are frequently sites of MeHg production and consequently sources to adjacent ecosystems and biota [33–36]. Enclosed environments, channels, and freshwater tributaries are also frequently associated with increased MeHg in water, sediment, and biota [26,37–40], due to the combined effects of watershed Hg loading, legacy industrial sources, elevated organic carbon deposition, and spatial variation in biota diets [4,37,41–43]. Forage fish sampling could indicate whether proximity to wetlands or embayment areas (such as enclosed marinas, backwater sloughs,

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stream drainages, and natural coves) predicts differences in biotic MeHg exposure within an estuary.

We report Hg spatial patterns in San Francisco Bay forage fish collected from 99 sites between 2008 and 2010. Unlike many ambient monitoring programs, the study design was hypothesis-based. Monitoring strata were defined, selected, and randomly subsampled to identify what kinds of locations within San Francisco Bay exhibit elevated Hg concentrations in forage fish. Since MeHg is the predominant Hg form in these fish [13], analyses of total Hg (THg) are assumed to indicate MeHg. Four questions are addressed: 1) What are the spatial trends in forage fish Hg? 2) Are Hg concentrations elevated in embayments relative to open-water sites? 3) Does the extent of fringing wetland habitat correlate with Hg concentrations? and 4) Are concentrations elevated at potential Hg source sites relative to randomly selected sites? In addition to randomly selected sites, 4 types of source sites were evaluated: sites draining watersheds impacted by historic Hg mining (mine sites), sites draining urbanized and industrial watersheds (industrial watershed sites), sites receiving treated effluent from POTW sites, and sites known to have elevated sediment Hg (contaminated-sediment sites).

METHODS

Study design and site selection

The study employed a stratified sampling design intended to evaluate the 4 study questions based on a priori hypotheses (Supplemental Data). The sample design included 99 sites distributed along the entire shoreline of San Francisco Bay from Lower South Bay to Suisun Bay (Supplemental Data, Figure S1). Wetland channels and estuarine tributaries were included, but salt ponds and tidal lakes were excluded. Sites were probabilistically selected from this sample frame using a generalized random tessellation stratified spatially balanced sampling design [44]. Two sample draws were performed. The first was for random locations across the entire bay shoreline, and the second was from all identified points within the 4 source categories, treating each category as a stratum.

The random sample draw included 2 categories (i.e., strata): open-water sites ($n = 25$ sites) and embayment sites ($n = 23$ sites; Supplemental Data, Figure S2). The source sample draw included 4 categories: Hg-mine creeks ($n = 4$ sites), watersheds draining urban and industrial areas ($n = 13$), publicly owned wastewater-treatment drainages ($n = 7$), and areas with relatively elevated sediment THg or MeHg ($n = 15$). For each subcategory, appropriate sampling locations were identified using a geographic information system, literature, and unpublished data (further detailed in the Supplemental Data). Due to limited sample sizes for POTW and Hg-mine sites, all sites within these categories were sampled. To ensure sufficient coverage of wetland habitats, 12 additional sites adjacent to nearshore wetlands were sampled in 2008, including 6 sites fringing the South Bay and 6 sites fringing San Pablo and Suisun Bays (Supplemental Data). These wetland sites were included only in the analysis of fringing wetland habitat versus fish Hg.

Fish sampling

All fish sampling was performed by beach seine in 2008, 2009, and 2010. To minimize confounding seasonal variation with spatial variation, study analysis was restricted to the fall season (27 August–30 November of each year). The target species were topsmelt (*Atherinops affinis*, target total lengths of 60–100 mm) and Mississippi silverside (*Menidia audens*, target total lengths of 40–80 mm), both of which have been successfully employed in San Francisco Bay as Hg biosentinels [13,16,25]. Four composites of 5 individuals each per species were targeted for THg at each sampling event. Target composites each included similar-sized individuals, with the composites distributed in ascending 10-mm windows spanning the overall size range targeted for each species (i.e., for Mississippi silverside, composite 1, $n = 5$ at 40–50 mm, through composite 4, $n = 5$ at 70–80 mm).

For the Bella Oaks and Borges Hg-mine sites, target species were not available. At these 2 sites, prickly sculpin (*Cottus asper*, 52–100 mm), California roach (*Hesperoleucus symmetricus*, 54–82 mm), and three-spined stickleback (*Gasterosteus aculeatus*, 34–50 mm) were collected. Like the target species, these are all invertivores previously employed as Hg biosentinels in California [14,15,45–47].

Sample preparation and analysis

All fish collection and preparation followed protocols developed at the University of California Davis, with a slow-cooling euthanasia method certified by the University of California Davis Veterinary School's Institutional Animal Care and Use Committee. Fish were measured for total length; rinsed with site water; sorted into labeled, freezer-grade plastic bags as composites for analysis; field-frozen with air excluded and water surrounding, on dry ice; and subsequently transferred to a –20 °C laboratory freezer. Composite whole-body fish samples were subsequently thawed, weighed, dried to constant weight at 55 °C, and ground to a fine homogenous powder. Dry weight and percentages of solids were recorded. Samples were analyzed for THg at the University of California Davis. Analysis employed standard cold vapor atomic absorption spectrophotometry, using a dedicated Perkin Elmer Flow Injection Mercury System with an AS-90 autosampler, following two-stage digestion at 90 °C in a mixture of concentrated nitric and sulfuric acids with potassium permanganate. Routine analytical quality assurance/quality control included 20 quality assurance/quality control samples for every 30 analytical samples as well as blanks, aqueous standards, continuing control standards, standard reference materials with certified levels of Hg, laboratory split samples, matrix spike samples, and matrix spike duplicates. All results met the quality assurance protocols of the Regional Monitoring Program for Water Quality in San Francisco Bay and were well within laboratory control limits. All study Hg results are reported on a wet weight basis.

Geospatial data

Geospatial data were developed in ArcGIS v10. The shoreline was partitioned into open-water versus embayment site categories based on visual inspection of a shoreline vector file with depth data overlay and satellite imagery. Inclusion criteria were depth, degree of separation from the rest of San Francisco Bay, and presence of channels or sloughs. The embayment layer included habitats throughout the bay, with the largest areal coverage north of San Pablo and Suisun Bays (Supplemental Data, Figure S2).

Two numeric geospatial attributes were examined for association with fish Hg: percentage of surrounding wetland area and distance from the Guadalupe River. The percentage of surrounding wetland area was based on a 500-m buffer, using data from the Bay Area Aquatic Resource Inventory and Association of Bay Area Governments 2005 land-use polygons. Percentage of surrounding wetland was defined as the sum of the depressional, marsh, and tidal ditch land-cover categories.

Distance from the Guadalupe River, defined as the nautical distance from the westernmost tidal point of Coyote Creek, was negatively correlated with forage fish Hg at 22 sites sampled previously [13]. It was calculated following along the deep bay channel, extending from the starting point to the upstream study extent of Suisun Bay (Mallard Island, near the confluence of the Sacramento and San Joaquin Rivers). Distance from the Guadalupe River indicates how close the sites are to the Hg-contaminated New Almaden mining district, which drains into lower South Bay near the community of Alviso. However, distance from the Guadalupe River also indicates general position along the bay axis, with the most distant North Bay segments (Suisun Bay, San Pablo Bay) having potentially different net MeHg production and distribution from the progressively closer Central Bay, South Bay, and Lower South Bay [13,26].

Data analysis

Data analyses were performed using the linear mixed effects model function in R, version 2.15 [48]. Data on Hg were $\log_{10}$-transformed to improve residual normality and variance homoskedasticity. Topsmelt and Mississippi silverside were analyzed separately. In line with the study questions, 4 separate linear models were built to examine the potential effect of spatial trend (distance from Guadalupe River), embayment category (embayment vs open water), surrounding wetland abundance, and site type (i.e., POTW, contaminated sediment, industrial watershed, and random sites) on fish Hg. Model evaluation was performed manually, using backward elimination of nonsignificant model terms. Parameter inclusion was based on the likelihood ratio test ($\alpha = 0.05$ to retain a parameter), as well as the Akaike information criterion and Bayesian information criterion [49,50]. Random effects were included to account for variability among sampling sites [50]. When significant, the slope for fish length was also allowed to vary by site. Four samples (3 Mississippi silverside and 1 topsmelt) were removed from the statistical analyses because their inclusion would have violated assumptions of residual normality and variance homoskedasticity. However, when analyses were performed with these samples included, the results were essentially unchanged. More details on the modeling approach and outlier removal are provided in the Supplemental Data.

Of the 4 mine sites, Mississippi silversides were present only at the Guadalupe River upstream of Alviso Slough, and topsmelt were present only at American Canyon Creek, draining Borges Mine. Since this was insufficient to statistically evaluate a mine site effect for these species, each mine site was compared with other data reported for additional species on an ad hoc basis. To provide context, data on additional species were compared with previously published Hg concentration data from mine sites [14,46,47] and unpublished data from reference (i.e., no known mine influence) sites. Unpublished data were obtained via queries performed on 23 March 2013 of the California Environmental Data Exchange Network (www.ceden.us), a collaboratively developed statewide environmental water quality database [51].

RESULTS

Graphical analysis indicated a spatial trend in average forage fish Hg concentrations, with the highest concentrations in and adjacent to Lower South Bay and concentrations progressively decreasing toward South, Central, San Pablo, and Suisun Bays (Figures 1 and 2). This spatial gradient was more pronounced for Mississippi silverside (Figure 1), whereas topsmelt exhibited more local-scale spatial heterogeneity, particularly within Central Bay (Figure 2).

As reported in a prior study [13], Hg concentrations were higher in Mississippi silversides (0.090 ± 0.058 μg/g, mean ± SD, $n = 237$) than topsmelt (0.041 ± 0.015 μg/g, $n = 239$). Total length was positively related to Hg and therefore included as a covariate in all models. Sampling year differences (treated as a categorical variable) were significant and included in some models (Table 1). Mixed models were needed to account for correlations among samples within a site, with site treated as a random effect. Additionally, a significant effect of site on the length covariate was observed for some models and thus incorporated as a random site effect on the length versus Hg slope (Table 1).

Distance from Guadalupe River

Distance from the Guadalupe River (question 1) was negatively related to Hg in Mississippi silverside ($r = -0.69$, $n = 237$; Figure 1) and topsmelt ($r = -0.42$, $n = 239$; Figure 2). Distance was also a significant predictor in mixed models accounting for site effect ($p < 0.0005$, Table 1) and therefore included as a covariate in models testing for other effects. The distance effect varied among sampling years. For silverside, in 2009, the decrease in Hg with distance from the Guadalupe River was weaker than other years (Distance Guadalupe × year 2009 interaction; Figure 3). Based on model predicted concentrations, in 2009, the closest site to the Guadalupe River (Coyote Creek upstream of Alviso Slough) exhibited 2-fold higher Hg concentrations than the farthest site (Kirker Creek near Pittsburg; 0.11 μg/g vs 0.053 μg/g), whereas in 2010, the predicted difference was 4-fold (0.16 μg/g vs 0.039 μg/g). For topsmelt, in 2008, the overall Hg decrease with distance from the Guadalupe River was weaker compared with other years (year 2008 × Distance Guadalupe).

Embayment and fringing wetland effects

Mercury concentrations in Mississippi silversides collected in embayment sites were not significantly different from those in Mississippi silversides collected in open-water sites (question 2; likelihood ratio test $p = 0.096$; Supplemental Data, Table S2). However, for topsmelt, Hg concentrations were significantly elevated in embayment sites compared to open-water sites ($p = 0.012$), and embayment site Hg significantly increased with distance from the Guadalupe River and with fish length (Table 1; Supplemental Data, Table S3). Embayment sites in Central and San Pablo Bays were more often elevated in topsmelt Hg versus adjacent open-water sites (Figures 2 and 4). For example, at the embayment site farthest from the Guadalupe River (the Petaluma River site), model predicted topsmelt Hg would be 0.043 μg/g, whereas an open site at the same distance would have a predicted Hg of 0.029 μg/g.

Percentage of surrounding wetlands (question 3) was not a significant predictor of Hg for Mississippi silverside or topsmelt. For both species, the final model included a significant increase with body length, a significant decrease with distance from the Guadalupe River, and no effect of wetlands (Table 1; Supplemental Data, Tables S4 and S5). For Mississippi silverside, the highest Hg wetland sites were in channels surrounding San Pablo and Suisun Bays and had lower Hg than Lower South Bay and South Bay sites (Figure 1).

Source-site type effects

Source-site effects (question 4) varied between Mississippi silverside and topsmelt. In 2008, Mississippi silverside Hg was

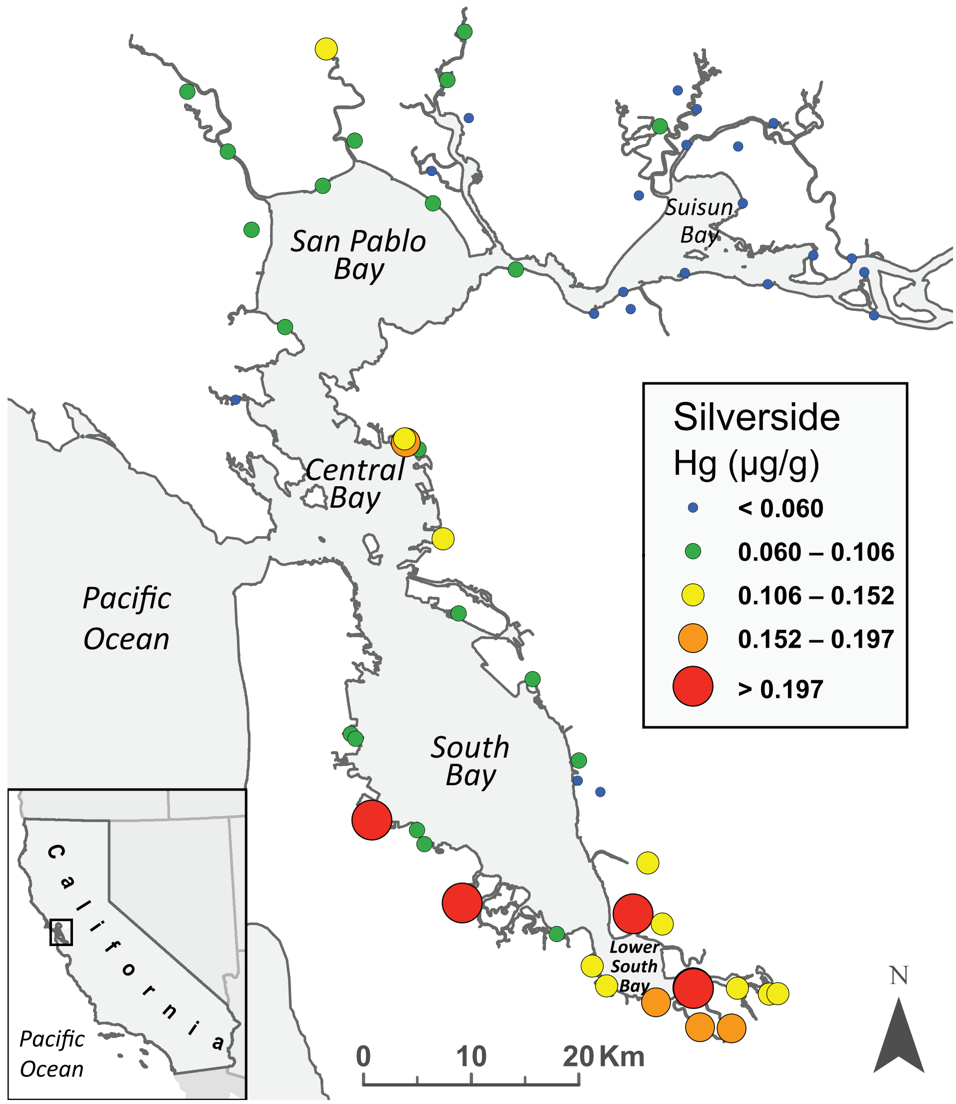

Figure 1. Site average Hg concentrations in Mississippi silverside.

lower at POTW sites than other site types (Source POTW × year 2008 interaction; Figure 3; Supplemental Data, Table S6). Based on model predictions for average-length fish, in 2008, POTW sites had approximately one-half of the Hg of non-POTW sites (0.035 µg/g vs 0.068 µg/g); in 2009 and 2010, POTW sites were predicted to be 0.015 µg/g lower than non-POTW sites. Graphical analysis indicated that POTW sites were lower than nearby sites in both 2008 and 2010 (Figure 3). In 2009, there was no apparent pattern of POTWs versus other sites. Topsmelt were obtained at only 1 POTW site, the Hayward wastewater-treatment plant discharge pond, monitored in 2010. Topsmelt Hg concentrations at that site (0.021 ± 0.0005 µg/g, $n = 4$) were less than half the concentrations at the nearest site measured in 2010, the Eden Landing Shoreline (0.045 ± 0.007 µg/g, $n = 4$).

In topsmelt, Hg was moderately elevated at contaminated-sediment sites ($p = 0.032$; Table 1; Supplemental Data, Table S7), which were present only in Lower South, South, and Central Bays (Figure 5). The model predicted topsmelt Hg at a contaminated-sediment site to be 1.2 times that predicted for another site type in the same location.

Fish species captured varied across the mining sites (Table 2), likely due to variable salinity conditions. The Guadalupe River upstream of Alviso Slough, which drains from the New Almaden Mine watershed, was elevated in Mississippi silverside Hg, consistent with the general spatial gradient observed in the present study and elsewhere [3,13,21,25]. Concentrations at this site were within the range of spatial variation observed in Lower South Bay (Figure 1) but higher than the baywide average and those reported for Mississippi silversides from Hg-contaminated Clear Lake [14]. The Guadalupe River site also had extremely high Hg in three-spined stickleback. In the Napa River below the Bella Oaks Mine watershed, prickly sculpin Hg was comparable to measurements from Clear Lake [14] and higher than the average of 8 sites from the Sacramento–San Joaquin Rivers Delta. Napa River California roach Hg was higher than the average of 8 California statewide sites lacking mine influence but well below the concentration previously measured by Slotton et al. [46] at Marsh Creek, a mine-dominated creek that drains into Suisun Bay. American Canyon Creek, which drains from the Borges Mine, was not elevated in topsmelt Hg relative to general baywide concentrations. Dry Creek also had relatively low Hg concentrations in prickly sculpin and California roach and unremarkable concentrations in three-spined stickleback, suggesting a lack of influence of the nearby La Joya Mine.

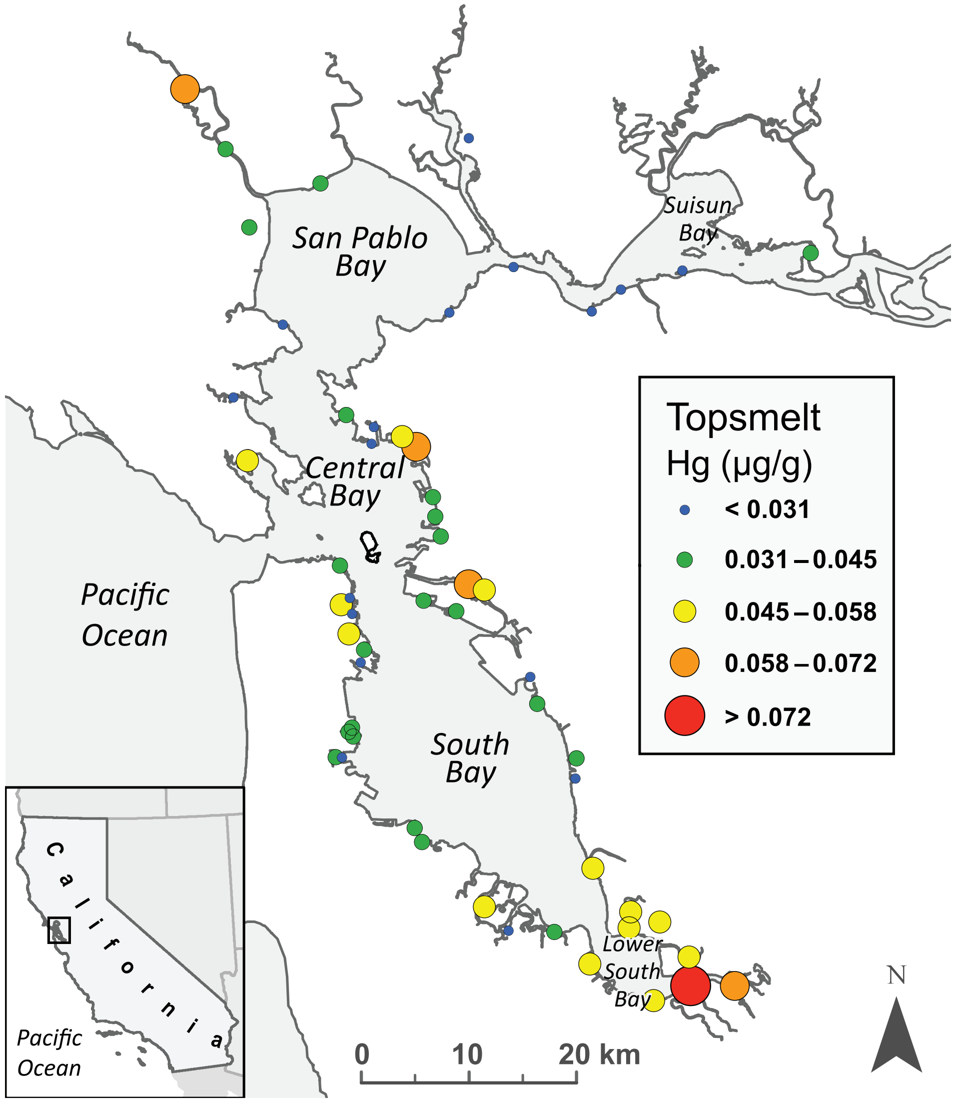

Figure 2. Site average Hg concentrations in topsmelt.

Three-spined stickleback were relatively low in Hg within the urbanized industrial watershed of Zone 4 Line A compared to mine sites and 4 other bay sites. The Zone 4 Line A sampling location is within a flood drainage channel, several kilometers above the San Francisco Bay shoreline [52].

DISCUSSION

We found that forage fish Hg concentrations were elevated in southern San Francisco Bay. This pattern has been noted previously [13], as well as elevated Hg or MeHg in water, sediment, and shorebirds of southern San Francisco Bay [3,26,53], suggesting that greater attention be dedicated to MeHg management in this region. The association between Hg and distance to the Guadalupe River indicates that Hg bioaccumulation in San Francisco Bay exhibits a spatial gradient across the relatively large distance of approximately 146 km from Coyote Creek upstream of Lower South Bay to the east side of Suisun Bay downstream of the Sacramento–San Joaquin Rivers Delta. A spatial gradient is also apparent in δ^{202}Hg stable isotope along San Francisco Bay for sediment and forage fish, suggesting increased bioaccumulation from Hg sources from the New Almaden mining district, draining into Lower South Bay [21,25]. The presence of tidal mixing and fluvial transport over time may create a relatively smooth gradient of both Hg source material and MeHg biogeochemistry. Results from the present study and previous studies suggest that in the western United States, fish Hg bioaccumulation exhibits regional spatial gradients, often following gradients of Hg mass in the sediments due to proximity to mines and other legacy Hg sources [5,11,14,54,55].

The higher Hg concentrations in Lower South Bay and South Bay likely result from multiple factors including historical Hg loading from the New Almaden mining district, South Bay hydrodynamics, and methylation in the bay and adjacent habitats. Relatively long water residence times in Lower South Bay and South Bay may result in reducing conditions that favor sediment and water column MeHg production [3], with additional elevated MeHg production in the extensive salt pond complexes adjacent to Lower South Bay [31,32] and periodic anoxia along Alviso Slough itself [16].

Elevated Hg in topsmelt, but not Mississippi silverside, at embayment sites (e.g., marinas, creeks, and backwater sloughs) and no relationship between surrounding wetlands and fish Hg

Table 1. Results of study model evaluations

Test (questions)[a]	Species	*n*	Likelihood ratio	*p*	Final model: Fixed terms (effect sign)	Final model: Random terms
Distance from Guadalupe[b] (1)	Silverside	237	4.8	<0.0001	Distance (–)	Intercept, length[c]
Distance from Guadalupe (1)	Topsmelt	239	12.4	0.0004	Distance (–)	Intercept, length
Embayment (2)	Silverside	116	2.77	0.096 (NS)	Distance (–), length (+)	Intercept, length
Embayment (2)	Topsmelt	133	6.30	0.012	Distance (–), length (+), 2009 (+), embayment (+), embayment × distance (+), embayment × length (+)	Intercept
Embayment × length interaction (2)	Topsmelt	133	40.8	<0.0001	As above	Intercept
Embayment × distance from Guadalupe interaction (1, 2)	Topsmelt	133	17.1	0.0007	As above	Intercept
Wetland (3)	Silverside	278	0.27	0.61 (NS)	Distance (–), length (+), 2008 (–), 2009 (–), distance × 2009 (+)	Intercept, length
Wetland (3)	Topsmelt	269	3.08	0.079 (NS)	Distance (–), length (+)	Intercept, length
Source: POTW × 2008 interaction (4)	Silverside	237	10.4	0.0012	Distance (–), length (+), 2008 (–), 2009 (–), distance × 2009 (+), POTW (–), POTW × 2008 (–)	Intercept, length
Source: contaminated sediment (4)	Topsmelt	231	4.58	0.032	Distance (–), length (+), 2008 (+), contaminated sediment (+), 2008 × distance (+)	Intercept

[a]Likelihood ratio tests were employed to answer the 4 study questions on mixed models, which account for additional significant predictor variables: 1) What are the spatial trends in forage fish Hg? 2) Are Hg concentrations elevated in embayments relative to open-water sites? 3) Does the extent of fringing wetland habitat correlate with Hg concentrations? and 4) Are concentrations elevated at potential Hg source sites relative to randomly selected sites?
[b]Distance is always centered.
[c]Length = fish total length (centered).
NS = not significant; POTW = publicly owned wastewater-treatment works.

suggest a limited ability to predict biotic MeHg exposure based on natural landscape attributes. We hypothesized that surrounding wetland abundance would correlate with forage fish Hg based on the established role of freshwater wetlands as MeHg sources to adjacent waters [34], the consequent association between proximity to wetlands and freshwater fish Hg [12,18,35,36], and evidence of elevated MeHg production in estuarine wetland sediment [30,33,56]. However, for these forage fish that reside within the subtidal open waters of San Francisco Bay, MeHg concentrations were decoupled from fringing wetland abundance. This is in contrast to northern temperate lakes, which are dominated by atmospheric deposition and frequently exhibit at least a moderate effect of catchment wetland abundance and other landscape attributes on MeHg bioaccumulation [12,18,34,57]. This finding suggests that the extensive wetland-restoration activities planned for San Francisco Bay are not likely to adversely affect MeHg exposure for these subtidal forage fish or their predators. Nevertheless, San Francisco Bay and associated wetlands comprise a range of habitats, and changes in Hg exposure to organisms residing within the wetlands or ponds fringing the bay could still occur as a result of restoration or other management activities [32].

Our hypothesis that embayment status could predict increased MeHg exposure in forage fish was based on elevated Hg accumulation in San Francisco Bay forage fish species that heavily utilize intertidal and shoreline areas (e.g., Mississippi silverside) [13], elevated sediment and biota MeHg in proximity to freshwaters in San Francisco Bay and other estuaries [26,37–40], increased exposure to anthropogenic Hg pollution at embayment sites [4,31], and the possible importance of fringing wetlands, intertidal mudflat habitat, and shallow sediments for MeHg production at embayment sites [30,38,56]. The increase in topsmelt Hg from embayment sites was related to spatial location; differences were primarily observed in Central Bay, where Mississippi silversides were not readily available. We speculate that the embayment pattern for topsmelt largely stems from exposure to historic industrial contamination because topsmelt Hg was also increased near legacy contaminated sediment. Historic industrial activity was abundant along the Central Bay shoreline and is associated with elevated concentrations of polychlorinated biphenyls, another legacy and industrial pollutant, in sediment and forage fish [58,59]. This pattern suggests that regional priorities for minimizing MeHg production might focus on identifying and restoring those embayment sites with elevated sediment and biota MeHg.

Source-site type effects included higher topsmelt Hg near contaminated sediments, higher Hg near some historic mine drainages, and lower Hg adjacent to POTWs in 2008 and possibly 2010. Previous research suggests that San Francisco Bay forage fish Hg and polychlorinated biphenyls are sediment-derived [25,59], and we observed that topsmelt Hg, but not Mississippi silverside Hg, corresponded to contaminated sediment sites. Other studies have also exhibited variable relationships between fish and sediment Hg (or MeHg), with associations observed in Texas rivers (USA) [11], the Hudson River (New York and New Jersey, USA) [60], and the Willamette River (Oregon and Washington, USA) [5] but not in a survey of northeastern US freshwaters [57] or a Columbia River reservoir (Washington, USA) [61]. In the Gulf of Maine, biota Hg is generally elevated in regions with elevated sediment Hg, but the bioaccumulation factor is lower in more contaminated areas, due to elevated total organic carbon reducing bioavailability [43]. The complexity of Hg methylation and bioavailability, biota movement, and food-web structure all contribute to the weak and variable relationships between fish and sediment Hg [43,61,62].

The negative effect of POTWs on forage fish Hg was unexpected, given that average total MeHg detected in discharge water from the 16 largest POTWs on San Francisco Bay was 0.37 ng/L versus 0.096 ng/L in bay ambient water [20]. We speculate that lower-than-expected forage fish Hg concentrations at some POTW sites may result from biodilution, in which increased primary and secondary production decreases Hg

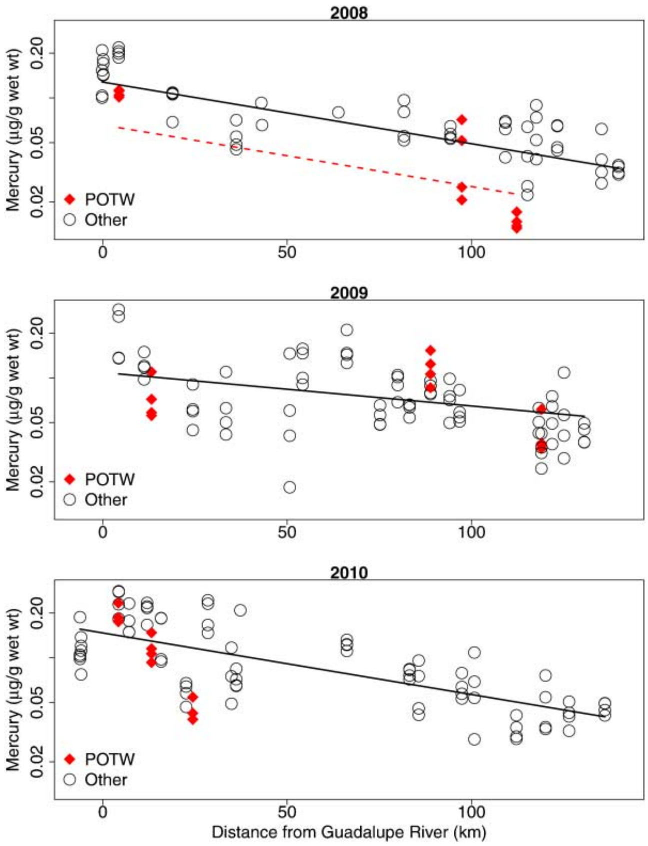

Figure 3. Mercury concentrations in Mississippi silverside as a function of distance from the Guadalupe River, sampling year, and site category. Each point represents a composite sample, and lines represent linear model fits to the associated data type for the given year. Note log-scale *y* axis. Solid diamonds (◆) represent publicly owned wastewater-treatment works (POTWs; i.e., draining wastewater-treatment plants); open circles (◯) represent all other sites.

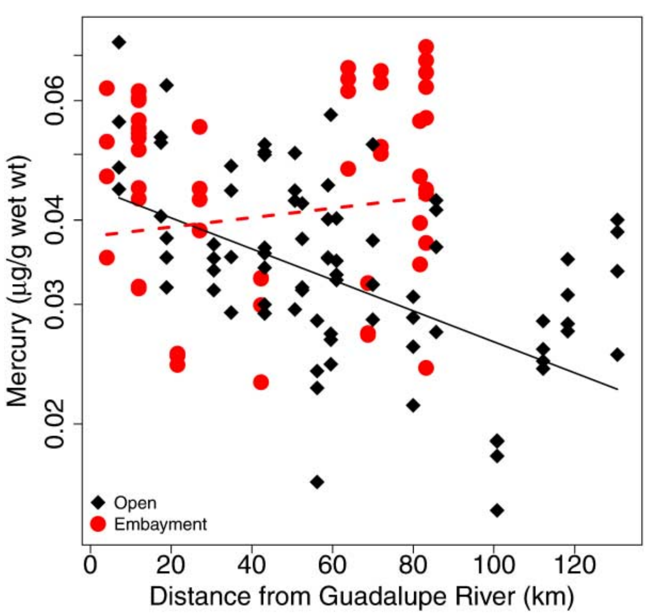

Figure 4. Mercury concentrations in topsmelt as a function of distance from the Guadalupe River and embayment category. Each point represents a composite sample, and lines represent linear model fits to the associated data type. Note log-scale *y* axis. Solid circles (●) represent embayment sites; solid diamonds (◆) represent open sites.

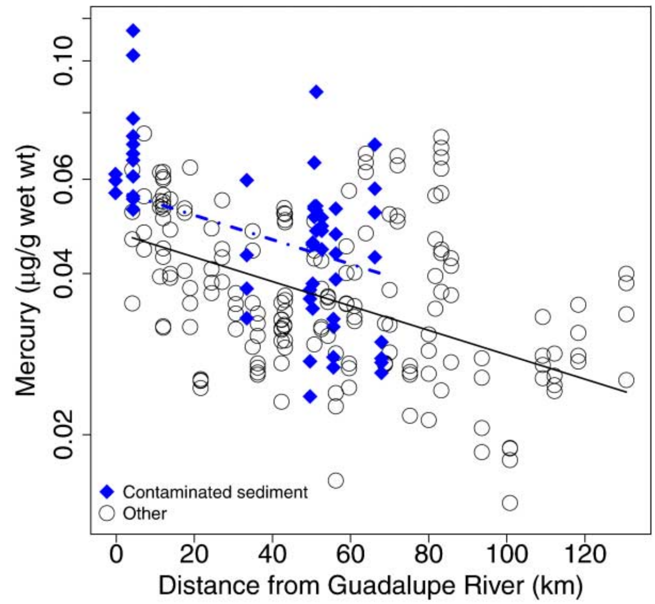

Figure 5. Mercury concentrations in topsmelt as a function of distance from the Guadalupe River and site category. Each point represents a composite sample, and lines represent linear model fits to the associated data type. Note log-scale *y* axis. (◆) represent contaminated sediment sites; open circles (◯) represent all other sites.

bioaccumulation and biomagnification [63,64]. The reduced Mississippi silverside Hg concentration at 4 South Bay POTW sites is associated with elevated discharge water ammonium concentrations compared with ambient bay conditions [65]. This may result in increased rates of primary production, higher densities of Mississippi silversides and their invertebrate prey, or more rapid growth rates, all resulting in decreased tissue Hg concentrations. Forage fish densities during collections at these sites were also observed to be substantially greater than typical for San Francisco Bay (D.G. Slotton, personal observation).

We observed inconsistent impacts of local mining, especially compared with the broad spatial gradient across San Francisco Bay. Concentrations in proximity to mining-impacted sites varied widely. The Guadalupe River downstream of New Almaden mining district and the Napa River below the Bella Oaks Mine were elevated in fish Hg and at or above prior measurements of the same fish species in mine-influenced sites [14,46]. In contrast, American Canyon Creek and Dry Creek were not elevated, suggesting lower levels of residual contamination. In California roach monitored closer to the New Almaden mines (Guadalupe Creek at Meridian Avenue and Alamitos Creek at Harry Road), Hg concentrations were even greater versus other sites in the local Guadalupe River watershed [66], and Hg isotopes indicate a New Almaden mining source signal in sediment and forage fish [21,25]. In freshwater lakes and rivers, fish Hg concentrations are frequently elevated in sites impacted by mining waste versus reference sites and tend to decrease with increasing distance from mining sources [5,14,55,67,68]. Levels of Hg are elevated near mines, processing facilities, and waste tailings even in areas with naturally occurring Hg deposits and even with Hg mining completed several decades before fish collection. This indicates a remaining concern for mine Hg in the food web and a potential benefit of ongoing remediation focused on Hg-mining sources such as the New Almaden mining district [3].

Finally, sites adjacent to industrial watersheds hypothesized to be Hg-contaminated did not exhibit elevated forage fish Hg concentrations. This is consistent with the relatively small Hg

Table 2. Forage fish Hg at mine sites and selected comparison sites

Site[a]	Mine influence	Species	Hg concentration, mean ± standard deviation (*n*)
Guadalupe River upstream of Alviso Slough	New Almaden Mines	Mississippi silverside	0.16 ± 0.020 (4)
Study average (57 remaining sites)	Reference	Mississippi silverside	0.09 ± 0.059 (236)
Clear Lake [14]	Sulphur Bank Mine	Mississippi silverside	0.10 ± 0.055 (97)[b]
American Canyon Creek	Borges Mine	Topsmelt	0.030 ± 0.006 (4)
Study average (55 remaining sites)	Reference	Topsmelt	0.042 ± 0.020 (236)
Napa River	Bella Oaks Mine	Prickly sculpin	0.13 ± 0.009 (4)
Dry Creek	La Joya Mine	Prickly sculpin	0.068 ± 0.012 (4)
Clear Lake [14]	Sulphur Bank Mine	Prickly sculpin	0.13 ± 0.044 (5)
Sacramento–San Joaquin Rivers Delta[c]	Reference	Prickly sculpin	0.098 ± 0.062 (15)[d]
Napa River	Bella Oaks Mine	California roach	0.14 ± 0.003 (4)
Dry Creek	La Joya Mine	California roach	0.061 ± 0.003 (4)
Marsh Creek [46]	Mt. Diablo Mine	California roach	0.27 ± 0.21 (6)
California statewide[c]	Reference	California roach	0.084 ± 0.037 (45)[e]
Guadalupe River upstream of Alviso Slough	New Almaden Mines	Three-spined stickleback	0.30 ± 0.027 (4)
Dry Creek	La Joya Mine	Three-spined stickleback	0.099 ± 0.005 (2)
Zone 4 Line A	Industrial watershed	Three-spined stickleback	0.052 ± 0.004 (4)
Four additional San Francisco Bay sites	Reference	Three-spined stickleback	0.096 ± 0.034 (11)
Marsh Creek [46]	Mt. Diablo Mine	Three-spined stickleback	0.082 ± 0.021 (6)
Putah Creek, Central Valley[c]	Reference	Three-spined stickleback	0.065 ± 0.007 (2)[f]
Walker Creek [47]	Gambonini Mine	Three-spined stickleback	0.19 (1)[g]

[a]Data were from the present study or other referenced studies, where noted.
[b]Whole-body samples, collected 1999 to 2004.
[c]Data from www.ceden.us data query, 23 March 2013.
[d]Average of site averages for 8 sites, sampled by D.G. Slotton in 1998.
[e]Average of site averages for 8 sites, sampled 1995 to 1997.
[f]Composite samples, collected by D.G. Slotton in 1998.
[g]Single composite of 36 individuals, collected June 1992.

mass discharged from these industrial watersheds compared to other sources and bay sediment. The San Francisco Bay total maximum daily load staff report [22] estimates urban stormwater runoff to contribute 92 kg Hg/yr to the bay, which is only 7.5% of all sources (1222 kg/yr) [22], and Hg isotope studies found a significant relationship between sediments and forage fish, without any notable deviations adjacent to more industrial sites [25]. Even stickleback collected within a small industrial watershed (at the Zone 4 Line A, tributary site [52]) were lower than at other sites, suggesting that industrial watersheds are not locations of elevated MeHg bioaccumulation.

We demonstrated the use of biosentinel forage fish, combined with a stratified probabilistic survey design, to identify Hg bioaccumulation spatial patterns and sources in a large urbanized estuary. Both regional and local patterns were observed, reflecting the complex legacy Hg sources and system hydrology. Regionally, there was a clear spatial gradient with distance from a historic Hg-mining district. After accounting for that gradient, local differences among sites were subtle and varied between fish species. These findings suggest that forage fish Hg bioaccumulation predominantly exhibits broad regional variation and that sources varying at local scales, including POTW and legacy sediment Hg contamination, exhibit a secondary influence.

SUPPLEMENTAL DATA

Sections S1–S5. (353 KB DOC).
Table S1. (208 KB XLS).

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