Mercury in San Francisco Bay forage fish

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Abstract

In the San Francisco Estuary, management actions including tidal marsh restoration could change fish mercury (Hg) concentrations. From 2005 to 2007, small forage fish were collected and analyzed to identify spatial and interannual variation in biotic methylmercury (MeHg) exposure. The average whole body total Hg concentration was 0.052 μg g^{-1} (wet weight) for 457 composite samples representing 13 fish species. MeHg constituted 94% of total Hg. At a given length, Hg concentrations were higher in wetland, mudflat, and slough species (*Clevelandia ios*, *Menidia audens*, and *Ilypnus gilberti*), compared to species that move offshore (e.g., *Atherinops affinis* and *Lepidogobius lepidus*). However, gut content analysis indicated similar diets between *Atherinops affinis* and *Menidia audens*. Hg concentrations were higher in sites closest to the Guadalupe River, which drains a watershed impacted by historic Hg mining. Results demonstrate that despite differences among years and fish species, nearshore forage fish exhibit consistent Hg spatial gradients.

Capsule: Total mercury in estuarine forage fish varies with species type, habitat, and proximity to a historic mercury mine

Keywords

mercury; forage fish; estuary; *Menidia audens; Atherinops affinis*

1. Introduction

Mercury (Hg) contamination is responsible for the vast majority of fish consumption advisories in North America (Wiener et al., 2007). Aquatic habitat managers often face difficult decisions, due to the association of wetlands and other productive aquatic habitats with increased production of methylmercury (MeHg) (St. Louis et al., 1994; Davis et al., 2003; Marvin-DiPasquale et al., 2003), the most bioaccumulative and toxic Hg form (Wiener et al., 2002). There is concern that restoration of wetland and aquatic habitats may increase Hg methylation, and consequent availability for food web uptake. Therefore, monitoring of Hg and other water quality measurements (Wiener et al., 2003) are needed for adaptive management of habitat restoration and impacts to Hg processes. Information is also needed on halogenated organic pollutants (e.g., PCBs, pesticides, PBDEs) in forage fish (i.e., small prey fish) to characterize potential risk to piscivorous wildlife resulting from dietary exposure to these pollutants (Jarvis et al., 2008).

Fish are well suited for Hg monitoring because they are relatively easy to capture and analyze, they strongly bioaccumulate MeHg, and they indicate exposure hazard to wildlife and human consumers (Wiener et al., 2002; 2003; 2007). Fish body length and mass correlate with many factors that influence Hg accumulation, including age, growth rate, and trophic position. Forage fish within small, specified length ranges are useful bioindicators of contaminant bioavailability, due to their short lifespan, relatively small range, and dietary proximity to the base of the food web (Wiener et al., 2007). Forage fish are particularly useful for understanding mechanisms influencing interannual and spatial variation in net MeHg production among aquatic ecosystems (Wiener et al., 1990;

Snodgrass et al., 2000; Greenfield et al., 2001; Essington and Houser, 2003). Despite these advantages, the majority of contaminant monitoring focuses on sport fish, because of concerns about human health risk due to dietary exposure (U. S. EPA, 2000). Few studies have compared Hg among multiple forage fish species to examine spatial and temporal consistency in exposure (Snodgrass et al., 2000; Swanson et al., 2006), particularly within estuaries and marine embayments (Paiva et al., 2008).

Estuaries, containing complex gradients in salinity, habitat, and water quality, represent a template for examination of spatial and temporal variation in food-web uptake of MeHg (Davis et al., 2003). Estuarine forage fish exhibit substantial differences in salinity tolerance and habitat affinity, which could affect their Hg exposure (Allen et al., 2006). For example, polyhaline species (occurring in brackish waters) may be restricted to localized backwaters having a freshwater source. Their affinity to a specific wetland or creek mouth could cause differences in measured Hg concentrations among sampling locations. In contrast, euhaline species (inhabiting marine waters) and pelagic schooling fish would move in open Bay waters among locations. These species may therefore be expected to have more consistent Hg among locations.

San Francisco Bay managers were required to develop a Total Maximum Daily Load (TMDL) regulatory program, to reduce Hg impacts on human fish consumption (SFBRWQCB, 2006). MeHg in sediments and water are highest in southernmost portions of San Francisco Bay, closest to where the Guadalupe River drains into the Bay (Conaway et al., 2003). The Guadalupe drains the New Almaden Mining District, which was the largest Hg mining district in North America (Thomas et al., 2002; Conaway et al., 2008).

We report on a three-year study to determine contaminant concentrations in small forage fish in San Francisco Bay. The study included annual Hg monitoring at fixed monitoring sites, from 2005 to 2007, with additional monitoring at other sites. The ultimate objective was to develop a monitoring program to evaluate long-term changes in Hg bioavailability in the Estuary, including response to wetland restoration and other management actions. Selected halogenated organic pollutants (PCBs, legacy pesticides, and PBDEs) were also monitored in one fish species at a subset of stations in 2007. Four questions were evaluated: 1. How do forage fish concentrations of Hg and organic pollutants compare to predator effects thresholds? 2. Do Hg concentrations vary with species life history attributes or habitats? 3. Do different forage fish species exhibit consistent interannual and spatial variation? and, 4. Do Hg concentrations in forage fish increase with proximity to the Guadalupe River watershed?

2. Materials and methods

2.1. Study design and target species

To cover the expected range of habitats in the Estuary, as well as to encompass differences in mobility and feeding habits, we sampled a variety of benthic (bottom-dwelling) and pelagic (free-swimming) species (Orsi, 1999; Goals Project, 2000; Moyle, 2002; Allen et al., 2006; Froese and Pauly, 2006). Benthic species were represented by five gobies, a sculpin, and a flatfish. These included four native marine species common on tidal flats: juvenile starry flounder (*Platichthys stellatus*), juvenile staghorn sculpin (*Leptocottus armatus*), arrow goby (*Clevelandia ios*), and cheekspot goby (*Ilypnus gilberti*). Additionally, yellowfin goby (*Acanthogobius flavimanus*) and shimofuri goby (*Tridentiger bifasciatus*), both non-native estuarine gobies with greater freshwater tolerance, were sampled. Finally, the native bay goby (*Lepidogobius lepidus*), a nearshore marine species that inhabits deeper channels in the Estuary, was sampled. The six pelagic species included two coastal pelagic species that tolerate (northern anchovy, *Engraulis mordax*) or require (Pacific herring, *Clupea pallasi*) estuarine habitat as juveniles; two marine species with very wide salinity tolerance (topsmelt, *Atherinops affinis*, and striped bass, *Morone saxatilis*), and two estuarine species more closely associated with fresh water (Mississippi silverside, *Menidia audens*, and rainwater killifish, *Lucania parva*) (Table 1). None of these species were captured at all sites in all years. We report average Hg concentrations for each species, and break the data down by total length, region, and other factors as sample coverage permitted.

The sampling design involves fixed sites, to facilitate analysis of trends in bioaccumulation of Hg over time in follow-up work. Fish were sampled at nearshore locations by beach seine at multiple sites (Fig. 1). Eight sites were selected for annual monitoring adjacent to natural wetlands or locations with anticipated wetland restoration activity planned for the future. These eight sites will allow annual monitoring and comparison of long-term Hg trends in natural versus restored wetlands. The natural wetland sites include Newark Slough, China Camp, and Benicia State Park. The sites with planned wetland restoration include Bird Island, Napa River, Eden Landing, Alviso

Slough, and Oakland Middle Harbor. Fish were also captured as available at 23 additional shoreline and midwater locations (Fig. 1; Supplemental Table S.1), including otter trawling in nine offshore locations (Orsi, 1999). As Alviso Slough drains directly from the Guadalupe River, distance from the Guadalupe River (km) was calculated based on linear distance from the Alviso Slough site (Supplemental Table S.1).

At each sampling event, four composites per species were targeted for total Hg analysis. Target number of individuals per composite was five to ten fish. As fish Hg concentration often increases with length (Huckabee et al., 1979; Wiener et al., 2002; Wiener et al., 2007), fish within a limited length range were targeted (Table 1). For most samples, total Hg was analyzed rather than MeHg, because the majority of Hg assimilated by fish is MeHg (Grieb et al., 1990; Wiener et al., 2002). For 39 composite samples collected in 2007, both total Hg and MeHg analyses were performed.

2.2. Fish collection and sample preparation

Fish were collected between 2005 and 2007, with the majority (360 of 457 composite samples) collected between September 5 and November 15. Beach seines were used to collect fish from intertidal and subtidal sites around margins of the San Francisco Estuary. Bay goby, northern anchovy, and Pacific herring were captured by benthic trawling in the main channel and shoal areas from sites throughout the San Francisco Estuary (Orsi, 1999). Total length was measured for each individual fish, fish were rinsed with deionized water, and each composite was placed in a separate freezer weight Ziploc ® bag. Bags were stored in the field on ice, and then transferred in the lab to conventional (- 20º C) freezers. At each collection site, latitude and longitude were collected with a Garmin GPS III Plus (Olathe, Kansas).

2.3. Chemical analysis

Samples were shipped overnight on ice to the Department of Biology/River Studies Center at the University of Wisconsin – La Crosse. Upon receipt at the laboratory, whole fish carcasses were thawed, weighed (nearest 0.001 g), re-frozen and stored in a conventional freezer. Frozen whole-body carcasses were lyophilized to a constant dry-weight in a Virtis DBT Benchtop 7.0 Freeze Dryer for a minimum of seven days at ≤ -85º C and ≤ 100 mtorr. To assess constant dry-weight, 10% of the samples were weighed after a minimum of seven days, dried overnight, and re-weighed.

In preparation for analysis of total mercury (THg), dried carcasses from each composite sample were digested whole or homogenized prior to digestion in a stainless-steel blender. Either the entire composite (samples with few, very small fish) or a subsample, approximately 0.1 g, of the homogenized composite was digested following a modification of EPA Method 1631. Samples and subsamples were digested for 3 h at 90 - 95^{o} C in a solution of H_2SO_4 and HNO_3 followed by digestion with BrCl for 8 h at 40^{o} C. Each digestate was analyzed by flow injection cold-vapor atomic fluorescence spectroscopy with a Leeman Labs Hydra AF Gold Plus Mercury Analyzer. For MeHg analysis, tissue samples were dissolved in 8 ml of 20% KOH in methanol at 47 C for 24 h. MeHg measurements were performed with gas chromatography and inductively coupled plasma mass spectrometry, in the Hintelmann laboratory (Trent University, Peterborough-Ontario). Mercury concentrations in composite samples were determined on a dry-weight basis, with wet-weight concentrations calculated based on dry weights and tissue percent moisture.

The accuracy of Hg determinations for each batch of fish samples was verified by the concomitant analyses of (1) certified reference materials from the National Research Council of Canada (NRCC) and the U.S. National Institute of Standards and Technology (NIST), (2) triplicate subsamples of homogenized fish, (3) spiked (before digestion) subsamples of homogenized fish, and (4) blanks and standards taken through the digestion procedures. Quality control criteria and quality assurance results for determinations of THg in composite samples conformed to requirements of a Quality Assurance Plan (Lowe et al., 1999). Concentrations in all fish samples analyzed exceeded the estimated limit of quantification (Clesceri et al., 1998) of 0.0097 $\mu g\ g^{-1}$ Hg dry-weight.

Organic contaminant analyses included organochlorine pesticides (including six DDT isomers and metabolites, seven chlordane compounds, and dieldrin), eight PBDE compounds, and 46 PCB congeners. Organic analyses were performed on separate composite samples of topsmelt collected from six stations (Fig. 1): Candlestick Point, Benicia State Park, Alviso Slough, Steinberger Slough, Newark Slough, and Point Isabel. Each composite contained ten fish, with composite average total length ranging from 81 to 96 mm. Samples were spiked with ^{13}C labeled surrogate standards, dried with sodium sulphate, Soxhlet extracted with dichloromethane, and analyzed for organochlorine pesticides, PCBs (USEPA Method 1668A), and PBDEs (USEPA Draft Method 1614). All analyses were performed by isotope dilution high resolution gas chromatography/high resolution mass spectrometry, by AXYS Analytical Services (Sidney, BC, Canada).

2.4. Dietary analysis

Dietary analysis was performed on topsmelt and Mississippi silverside. Ten individuals of each species were examined from each of three sampling sites (China Camp, Newark Slough, and Eden Landing). All samples were from 2006 except for China Camp topsmelt, which were taken in 2007. Whole fish were fixed with formalin, and then transferred to alcohol after a 24-hr leaching. Prey items from the entire digestive tract were identified following Carlton (2007). Data were reduced to general taxonomic category, as described elsewhere (Jahn, 2008). Relative volume of prey consumed was estimated in two ways: 1. the % volume, averaged per fish; and 2. weighted average %, obtained by multiplying % volume for each individual fish by total gut content mass for that fish, summing by food category, and dividing by the total mass consumed by that species.

2.5. Statistical analysis and comparison to thresholds

Contaminant residues were compared to effects thresholds. Hg was compared to a 0.03 $\mu g\ g^{-1}$ wet-weight target for wildlife piscivores. This threshold was established in the Hg TMDL to be protective of California least tern and other piscivorous wildlife that forage in San Francisco Bay (SFBRWQCB, 2006). Hg was also compared to a 0.2 $\mu g\ g^{-1}$ wet-weight tissue threshold for biological effects to fish, including growth, reproduction, development, and behavior, developed by Beckvar et al. (2006). DDTs were compared to a 14 ng g^{-1} prey tissue residue guideline developed by Environment Canada for protection of wildlife consumers (Canadian Council of Ministers of the Environment, 1999).

Evaluation of normal-scores plots, histograms, and residuals plots indicated that tissue Hg concentrations were square-root normally distributed in this study. Therefore, unless indicated otherwise, Hg data were square-root transformed, to achieve normally distributed data, prior to parametric statistical analyses. To compare the impact of total length, sampling year, and collection location for individual species, general linear models (GLM) were fit to the data. Parameter addition was based on F-ratio and p value, with parameters included only when still significant ($p < 0.05$) with more influential parameters already in the model. Partial R^2 was the increase in R^2 when that parameter was added to the full model. In the text, capitalized words indicate parameter estimates; e.g., LENGTH is the slope estimate for the effect of length on Hg, and YEAR and SITE indicate vectors of parameter estimates for the categorical variables year and site. Analyses were performed on incomplete designs, with site-year combinations missing in some cases; therefore, there is some risk of confounding site versus year effects. When substantial data were missing for certain years, those years were excluded from analysis. For bay goby, yellowfin goby, staghorn sculpin, and northern anchovy, year effects were not examined due to missing year-site combinations. For shimofuri goby, site effects were not examined due to small sample sizes ($n < 3$) at two of three sites. Statistical analyses were performed using SYSTAT (version 11) and SAS 9.1.

3. Results

3.1. Mercury

The average wet-weight Hg concentration of the 457 samples analyzed was 0.052 μg g^{-1} and the average total length was 59.5 mm. Eight of the samples exceeded the 0.2 μg g^{-1} tissue effects threshold, and 293 of the samples (64%) exceeded the 0.03 μg g^{-1} wildlife effects threshold (Fig. 2). Supplemental Table S.2 presents all sample results.

In 39 fish collected from five sites in 2007, THg and MeHg were significantly correlated ($R^2 = 0.96$; $p < 0.0001$; both dry weight). The linear relationship was MeHg = 0.943 × THg + 0.006. The intercept term was not significantly different from zero. The standard error of the slope estimate was 0.030, and the 95% confidence interval of the slope was 0.882 to 1.003. These results indicated that in the measured fish in San Francisco Bay, MeHg constituted between 88% and 100% of THg, with the best estimate being 94%.

A significant difference in Hg was observed among species (ANOVA; $f = 37.0$, $p < 0.0001$, $R^2 = 0.52$, $N = 457$). Among commonly captured species, concentrations were highest in Mississippi silverside, intermediate in topsmelt, cheekspot goby, arrow goby, yellowfin goby, and northern anchovy, and lowest in bay goby (Table 2). Three Pacific herring samples also exhibited relatively low concentrations. Examination of length versus Hg relationships (Fig. 2) indicated that, at a given length, concentrations were relatively high for arrow and cheekspot gobies and for Mississippi silverside. Concentrations were relatively low for topsmelt, bay goby, and northern anchovy.

Spatial and temporal variation was examined for those species having sufficient samples. For each species, a general linear model was developed including all sites that were analyzed over the three-year study (Table 3). For Mississippi silverside, topsmelt, and cheekspot goby, the model included significant LENGTH, SITE, and YEAR terms. For silverside, the majority of variation was explained by site, as well as a significant SITE versus YEAR interaction term. In contrast, length explained the largest amount of variation for topsmelt and cheekspot goby. A SITE effect was observed for northern anchovy, taken by trawl at open-water sites; this is likely an artifact resulting from confounding location with schooling behavior (see Discussion).

Variation among sampling years also differed among species in accordance with differences in body length. Across multiple stations, topsmelt were lowest in Hg (averaging 0.036 $\mu g\ g^{-1}$) and total length (averaging 57 mm) in 2005 and highest in Hg (0.053 $\mu g\ g^{-1}$) and length (80 mm) in 2007. Cheekspot goby were highest in Hg in 2006 (Fig. 3c), again related to higher total length in that year (33 mm) compared to other years (29 mm in both 2005 and 2007). In contrast, Mississippi silverside were highest in 2005 at Eden Landing and Newark Slough, and similar among sampling years at Alviso Slough, with no apparent association with changes in length among years.

The four most commonly captured monitoring species (Mississippi silverside, topsmelt, arrow goby, and cheekspot goby) were evaluated for spatial similarity among species and for the presence of a spatial gradient, after first adjusting for fish length and combining goby species, as explained below.

Given the significant effect of length on Hg (Table 3), length correction was performed prior to evaluating spatial patterns. For topsmelt and Mississippi silverside, Hg at each site was summarized based on parameter estimates from a general linear model (GLM). Specifically, for each site and year combination, square root Hg was estimated as SqrtHg = constant + SITE + LENGTH × RL + Average(RESIDUAL), with RL indicating a representative length (65 mm for silverside and 80 mm for topsmelt). In this equation, LENGTH is the slope effect for body length, SITE is the parameter estimate for the effect of sampling site, and RESIDUAL is the average of residuals from a particular site. A length versus site interaction was not significant in these analyses.

The multi-site comparison was facilitated by combining two species of native, shallow-water gobies (cheekspot and arrow), which tended to have complementary distributions among the sites. The small difference in mean Hg concentration between these two species (Table 2) was apparently due to mismatches between site and length (e.g., Fig. 4), because after correcting for total length, Hg concentrations were not significantly different between cheekspot and arrow goby. A GLM combining the data from both species indicated a highly significant length effect ($f = 195.2$; $p < 0.0001$; $R^2 = 0.63$; $N = 119$) but no significant effect of species or species versus length interaction ($p > 0.2$). Based on this finding and their similar taxonomy and life history (Brothers, 1975), these species were pooled for comparison with topsmelt and Mississippi silverside.

A GLM indicated a highly significant length versus site interaction for the arrow and cheekspot gobies ($f = 5.01$; $p < 0.0001$; $N = 119$), indicating some sites to have steeper length-Hg relationships than other sites (Fig. 4). To ensure that the length effect was properly accounted for, only gobies from the six sites having an adequate sample size ($N \geq 8$), and lengths spanning the average range (i.e., within 30 to 40 mm TL) were included. For these 85 samples, for each site and year combination, square root Hg was estimated as SqrtHg = constant + SITE + LENGTH × RL + SITE*LENGTH × RL + Average(RESIDUAL), with RL set at 35 mm, and SITE*LENGTH indicating the site versus length interaction effect.

After length-corrected Hg data were generated for all taxa, they were averaged for each year and sampling location. This resulted in a single data point for each combination of taxon, year, and sampling site. Using these data, statistical comparison among taxa indicated a strong positive correlation between native, shallow-water gobies and topsmelt (Pearson's $r = 0.79$), but weak positive correlations between silverside and the other taxa (Table 4).

Concentrations in all three taxa decreased with distance from the Alviso Slough site (Pearson's r from -0.58 to -0.63; Table 4), indicating that Hg concentrations were generally greater at stations closest to the Guadalupe River. Graphical evaluation of Hg concentrations also illustrated a general spatial gradient, with higher concentrations at stations closer to the Lower South Bay. For example, average concentrations in topsmelt were above 0.04 $\mu g\ g^{-1}$ in the four southernmost stations, between 0.03 and 0.04 $\mu g\ g^{-1}$ in most Central Bay stations, and below 0.02 $\mu g\ g^{-1}$ in Hamilton Army Airfield (San Pablo Bay) and Benicia State Park (east of San Pablo Bay; Fig. 1). Also, Mississippi silverside generally had lower Hg in Benicia State Park and China Camp (San Pablo Bay) than Newark and Alviso Sloughs (Lower South Bay) and Bird Island (South Bay; Fig. 3a). Finally, composite samples of arrow and cheekspot goby had higher Hg at a given length in Alviso and Newark Sloughs than in Pt. Isabel (Central Bay; Fig. 4).

3.2. Organic chemicals

Composite topsmelt samples collected from six locations in 2007 exhibited sum of DDT concentrations averaging 27 ng g^{-1} wet (SD = 6.8), with every sample exceeding the 14 ng g^{-1} Environment Canada threshold for protection of wildlife. The average sum of PCB congeners was 198 ng g^{-1} (SD = 122), sum of seven chlordanes was 6.4 ng g^{-1} (SD = 1.4), dieldrin was 1.3 ng g^{-1} (SD = 0.4), and lipid content was 3.7% (SD = 0.5). The average sum of nine PBDEs was 9.3 (SD = 3.0), with BDE 47 and BDE 100 comprising the highest and second highest proportion of the total (70% and 12.3%, respectively). All individual station measurements were within 95% CI of the mean except for DDTs at Benicia State Park (39.5 ng g^{-1}) and PCBs at Candlestick Point (445 ng g^{-1}).

3.3. Diet

Gut content analysis indicated topsmelt and Mississippi silverside had similar diets at the three sites analyzed. The percentage similarity index (PSI), which is the sum of the minimum weighted average percent over all food categories, was 81%. Both species fed principally on small epibenthic crustaceans (in particular, corophiid amphipods), with more limited utilization of insects and planktonic crustaceans (Table 5). Most of the insects eaten by both species were plant-hopper nymphs and adults (probably *Prokelisia* *marginata*), which infest cordgrass along the creek banks. Total length ranges and averages were also similar (topsmelt 28 to 101 mm TL, $\bar{x}$ = 58 mm, N = 30; Mississippi silverside 33 to 83 mm, $\bar{x}$ = 53, N = 30).

4. Discussion

4.1. Forage fish contaminant concentrations indicate potential hazards to wildlife

Our results suggest that the TMDL Hg target for wildlife is not currently met in San Francisco Bay (SFBRWQCB, 2006). A majority of fish samples exceeded the 0.03 $\mu g\ g^{-1}$ target for protection of California least tern and other piscivorous wildlife. Least terns are opportunistic piscivores (Elliott, 2005), and are likely to include most of the species and fish body sizes from this study in their diets. Least terns can consume fish up to about 9-15 mm in body depth, which corresponds to topsmelt in the range of 60 - 100 mm total length (Atwood and Kelly, 1984; Elliott et al., 2004; Elliott, 2005; Zuria and Mellink, 2005), the target length for topsmelt in this study. PCBs and DDTs were surprisingly high, with DDT residues consistently above the Environment Canada guideline, but similar to pelagic forage fish concentrations in the historically polluted Southern California Bight (Jarvis et al., 2008). Surprisingly, PCB residues were similar to concentrations in sport fish targeted in San Francisco Bay by human consumers (Greenfield et al., 2005). The elevated concentrations may be partially attributable to moderately high lipid content in the sampled fish (Kidd et al., 1998; Jarvis et al., 2008), but may also be related to proximity to contaminated sediments and historic sources. The highest PCB concentration (445 $ng\ g^{-1}$) was observed in the sample from Candlestick

Point, within 500 m of the Hunter's Point Naval Shipyard, a Superfund remediation site with historic PCB storage and use (Battelle et al., 2005).

4.2. Differences among species

We observed unique spatial and temporal patterns in Mississippi silverside Hg concentrations, compared to other species monitored. Potential explanations for different Hg concentrations among species include differences in habitat and differences in diet. Nevertheless, stomach contents of 30 topsmelt and 30 Mississippi silverside indicated similar reliance on predominantly epibenthic invertebrates, a result corroborated by a more intensive dietary examination at China Camp marsh creeks (Visintainer et al., 2006). That study also found that both topsmelt and Mississippi silverside ate mainly corophiid amphipods and cumaceans, with the difference of slightly higher percentages of planthoppers and copepods in the Mississippi silverside diet. Based on these findings, we suggest that prey choice does not explain the higher Hg concentrations in Mississippi silverside.

We hypothesize that the differences in Hg concentrations, spatial patterns, and interannual variation between Mississippi silverside and the other commonly captured species result from differences in movement and consequent dietary exposure to Hg. Topsmelt are marine migrants that move offshore as the tide recedes, and may move from shallows to Bay channels (Orsi, 1999; Allen et al., 2006). In contrast, Mississippi silverside are almost never collected in offshore portions of San Francisco Bay or in marine salinities; they are found exclusively along Bay margins (Orsi, 1999). Mississippi silverside remain within shoreline marshes, as described for the congeneric Atlantic silverside (*Menidia menidia*) (Bigelow and Schroeder, 1953; Griffin and Valiela, 2001), and are expected to move inshore, including source tributaries and wetland sloughs, especially in areas that are at least seasonally freshwater (Moyle, 2002). Thus, although Mississippi silverside and topsmelt apparently enter the fringing marshes to feed at high tides, topsmelt are more likely to be carried by the tide to deeper offshore locations. Bay goby are generally restricted to these deeper offshore locations, inhabiting higher salinity Bay channels (Orsi, 1999; Goals Project, 2000). For bay goby, relatively low Hg concentrations at a given length (Fig. 2) suggest that prey methylmercury concentrations may be particularly low in the deepwater offshore environments. Cheekspot and arrow gobies inhabit burrows in intertidal mudflats, and thus probably do not venture onto fringing marshes (Brothers, 1975). They also select generally smaller prey than topsmelt and Mississippi silverside (Barry et al., 1996). The spatial patterns of Hg in arrow and cheekspot gobies are similar to topsmelt, likely because of the greater reliance on dietary items captured in intertidal areas, as compared to silverside venturing into the marsh plain, and upstream brackish locations. Paiva et al. (2008) similarly found that lagoon-dwelling forage fish species had higher tissue Hg than marine species.

4.3. Spatial patterns

Our results support the use of forage fish as bioindicators of local spatial patterns such as impact of proximity to anthropogenic Hg sources or higher methylation areas (Suchanek et al., 2008). A number of factors may drive the higher Hg for fish captured in the South and Lower South Bays. Alviso Slough carries water from the Guadalupe River, and some of the other southern sites (Newark Slough and Steinberger Slough) are also relatively close to the Guadalupe River. The Guadalupe River is heavily impacted by historic Hg mining activity and an important Hg source to the Bay (Thomas et al., 2002; Conaway et al., 2003; SFBRWQCB, 2006; Conaway et al., 2008). Hg enters the North Bay via the Sacramento and San Joaquin Rivers (David et al., 2009) and is present there as in-bay sediment deposits from historic mining operations (Hornberger et al., 1999). Possible explanations for the relatively low concentrations in North Bay sites include high exposure to open water and consequent source dilution at the base of the food chain, faster turnover of water due to stronger freshwater flushing, and lower net MeHg production than at South Bay and Lower South Bay sites.

ANOVA results indicated that different species vary in ability to indicate spatial variation in Hg bioavailability. For Mississippi silverside and arrow goby, about half of the variability in Hg was explained by collection location (Table 3). This finding suggests that these species are likely restricted to foraging in locations relatively close to the site of capture, and thus could be useful biomonitoring tools for identifying "hotspots" of Hg bioavailability in San Francisco Bay and other waters. Arrow and cheekspot goby are burrow dwellers restricted to intertidal mudflats, and therefore would be expected to indicate Hg bioavailability in relatively small areas (Brothers, 1975).

In contrast to Mississippi silverside and arrow goby, bay goby exhibited no significant effect of collection site (Table 3). Bay goby are believed to migrate from nursery areas and concentrate as adults in Central Bay (K. Hieb, pers. comm., Orsi, 1999). Therefore, our trawl-captured samples are likely a mix of individuals coming from shoals in North, Central, and South Bays. Topsmelt represented an intermediate case; collection location was statistically significant, but only explained 14% of total variation in Hg (Table 3). Where shoals extend several km from shore, topsmelt are frequently taken in open water by purse seine and midwater trawls (Orsi, 1999, A. Jahn, unpublished data) and thus are likely to integrate local site differences while indicating broader regional spatial patterns, such as differences between estuary subembayments. Some of the species collected in smaller numbers indicated site effects that were less easy to reconcile with this conceptual model. In particular, yellowfin goby and northern anchovy, which range widely as adults, both exhibited highly significant effects of collection location (Table 3). In the case of yellowfin goby, this may be caused by our targeting juveniles, which presumably have had limited exposure to multiple areas of the Estuary. For northern anchovy, a schooling pelagic species with no expected local-scale site fidelity over time (Messersmith et al., 1969), it is likely that site was confounded by school, with all composites at each location collected from the same school. Our results suggest that separate juvenile anchovy schools may have unique exposure histories to MeHg.

5. Conclusions

Our results suggest a generalized geographic gradient in Hg concentration in forage fishes in San Francisco Bay that is consistent with patterns in MeHg in water, sediments, and other taxa, with higher concentrations in the southern reach of the Bay (Conaway et al., 2003; Ackerman et al., 2007). These findings indicate that small fish are useful indicators for regional spatial gradients in shoreline net MeHg exposure.

Forage fish are useful for long-term status and trends monitoring in estuaries. In particular, forage fish data allow for: 1) comparison of changes in Hg in time within and among sampling locations; 2) evaluation of success of Hg management efforts mitigating bioavailable Hg in the Estuary; and 3) assessment of the potential impact of regional restoration activities.

Acknowledgements

We thank Kathy Hieb, Steve Slater and Dave Crane (CDFG); Darell Slotton (UC-Davis); Marco Sigala (Moss Landing Marine Labs); Joel Baker (University of Maryland); Paul Salop (Applied Marine Science); Letitia Grenier and Jay Davis (SFEI) for guidance and feedback regarding project design and sampling protocols. Seth Shonkoff, April Robinson, Max Delaney, Aroon Melwani, Katie Harrold, Mami Odaya, Carrie Austin, Richard Looker, Bridget Mooney, Cindy Patty, Sarah Cohen, Steve Slater, and Tom Grenier provided valuable field assistance. Additional samples were provided by the USFWS-Stockton, the Interagency Ecological Program for the San Francisco Estuary and CDFG San Francisco Bay Study. Laboratory analyses were performed by Sean Bailey and Mark Sandheinrich (UW-La Crosse), Josh Ackerman and Collin Eagles-Smith (USGS), and AXYS Analytical Services. Elly Best (US ACE ERDC) and Holger Hintelmann (Trent University, Peterborough-Ontario) kindly provided MeHg in fish data. Site access and information were provided by Arthur Fong (California Department of Parks and Recreation); Carl Wilcox and John Krause (CDFG); and Joy Albertson, John Bradley, G. Mendel Stewart, Clyde Morris and Eric Mruz (SF Bay National Wildlife

Refuge). Lester McKee, Katie Harrold, Aroon Melwani, Jay Davis, Kat Ridolfi, Harry Ohlendorf, and Darell Slotton provided constructive comments on the manuscript. This project was funded by the Regional Monitoring Program for Water Quality, and is SFEI Publication # XXX.

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Figure legends

Fig. 1. Study sampling locations. ○ ● ● ⦿ = sites where topsmelt were captured; ⊗ = other sites. See figure for topsmelt concentrations.

Fig. 2. Length versus Hg for commonly captured species. Solid horizontal bar indicates fish effects threshold (Beckvar et al., 2006). Broken horizontal bar indicates target for local wildlife (SFBRWQCB, 2006). Note log scale.

Fig. 3. Spatial and interannual variation in Hg for species monitored at multiple station and year combinations. For each station, 2005, 2006, and 2007 from left to right are presented as adjacent box and whiskers plots. Boxes indicate the 25^{th}, 50^{th}, and 75^{th} percentiles, and dots indicate raw data. a. Mississippi silverside. b. Topsmelt. c. Cheekspot goby.

Fig. 4. Length versus Hg in gobies at three monitoring sites. ● = Alviso slough (crossed – cheekspot; others arrow goby), □ = Newark slough (crossed – cheekspot; others arrow goby). ◆ = Pt. Isabel (all arrow goby).

Table 1. Species captured for Hg analysis. Habitat affinity, salinity affinity, and movement are approximate, and based on published reviews and personal observations (Orsi, 1999; Goals Project, 2000; Moyle, 2002; Allen et al., 2006; Froese and Pauly, 2006, Kathy Hieb, pers. comm., and Andy Jahn, pers. obs.).

Common name	Scientific name	Family	Habitat affinity	Target total length (mm)
Pacific herring	*Clupea pallasi*	Clupeidae	Pelagic, coastal, spawns in San Francisco Bay, where juveniles remain for first summer	NA
Northern anchovy	*Engraulis mordax*	Engraulidae	Pelagic, coastal, common in nearshore waters; juveniles tolerant of estuarine salinity conditions	NA
Rainwater killifish	*Lucania parva*	Cyprinodontidae	Pelagic, freshwater and tidal creeks	NA
Mississippi silverside	*Menidia audens*	Atherinopsidae	Pelagic, shallow water; most typical of areas that are at least seasonally freshwater	50 – 80
Topsmelt	*Atherinops affinis*	Atherinopsidae	Pelagic, shallow water	60 – 100
Striped bass	*Morone saxatilis*	Percichthyidae	Pelagic, migratory along coast, anadromous; juveniles	NA

			estuarine	
Staghorn sculpin	*Leptocottus armatus*	Cottidae	Benthic, nearshore sandy habitats; juveniles tolerate fresh water	NA
Cheekspot goby	*Ilypnus gilberti*	Gobiidae	Benthic, shallow water, common on tidal flats	20 – 40
Arrow goby	*Clevelandia ios*	Gobiidae	Benthic, shallow water, common on tidal flats	20 – 50
Yellowfin goby	*Acanthogobius flavimanus*	Gobiidae	Benthic, shallow water	NA
Shimofuri goby	*Tridentiger bifasciatus*	Gobiidae	Benthic, shallow water	NA
Bay goby	*Lepidogobius lepidus*	Gobiidae	Benthic, offshore channels and shoals	20 – 40
Starry flounder	*Platichthys stellatus*	Pleuronectidae	Benthic, juveniles estuarine, common on tidal flats	NA

NA – target lengths not developed

Table 2. Summary statistics for Hg concentrations in fish captured in San Francisco Bay, 2005 - 2007. N = Number of composite fish samples analyzed. Sites = number of sites sampled.

Species	N	Length mean ± SD (mm)	Hg mean ± SD ($ng\ g^{-1}$ wet)	Sites
Rainwater killifish	4	29 ± 2	61 ± 6	1
Cheekspot goby	43	32 ± 6	30 ± 19	4
Arrow goby	76	34 ± 10	38 ± 26	13
Shimofuri goby	12	56 ± 13	40 ± 15	3
Mississippi silverside	92	62 ± 11	113 ± 63	12
Bay goby	29	64 ± 30	16 ± 8	7
Staghorn sculpin	9	65 ± 6	50 ± 9	3
Starry flounder	1	65	35	1
Pacific herring	3	69 ± 1	17 ± 1	1
Yellowfin goby	18	72 ± 17	33 ± 7	7
Topsmelt	153	73 ± 20	41 ± 15	22
Striped bass	3	86 ± 23	52 ± 6	1
Northern anchovy	14	97 ± 6	45 ± 21	5

Table 3. Model results for Hg concentrations as a function of collection site, total length and year. Sites = number of sites in complete block design. Partial sum of squares = effect sum of squares / total sum of squares. NS = not statistically significant ($p > 0.05$).

Species	Sites	N	Partial sum of squares			
			Site	Length	Year	Site*Year
Cheekspot goby	2	25	0.14	0.45	0.12	NS
Arrow goby	5[a]	40	0.47	0.37	NS	NS
Shimofuri goby	3[b]	12	-	0.52	NS	NS
Mississippi silverside	5	61	0.52	0.05	0.04	0.13
Bay goby	7	29	NS	0.89	-[d]	-[d]
Staghorn sculpin	3	9	NS	NS	-[d]	-[d]
Yellowfin goby	6[c]	18	0.66	NS	-[d]	-[d]
Topsmelt	5	50	0.14	0.27	0.16	NS
Northern anchovy	5	14	0.92	NS	-[d]	-[d]

a. sampling at 5 sites in 2006 and 2007 only. b. N < 3 at two sites. c. sampling haphazard among sites over study duration. d. not evaluated due to insufficient data.

Table 4. Pearson's correlation coefficients comparing three forage fish taxa and linear distance to the New Almaden Mine. Right side of table presents number of pairwise comparisons.

	Mississippi silverside	Topsmelt	Gobies	Distance to mine
Mississippi silverside		20	12	23
Topsmelt	0.32		16	39
Gobies	0.17	0.79		16
Distance to mine	-0.63	-0.58	-0.63	

Table 5. Dietary summary of two fish species.

	Topsmelt		Mississippi silverside	
Food Category	Avg. %	Wtd. Avg. %	Avg. %	Wtd. Avg. %
Diatom	0.1	0.2	1.6	7.8
Microplanktivores [a]	3.7	5.7	0.7	0.4
Copepods and ostracods [b]	34.2	9.3	27.9	18.4
Large Zooplankton [c]	4.9	4.8	6.4	4.0
Small crustacean [d]	15.9	7.6	18.4	8.0
Large crustacean [e]	30.4	55.7	32.7	52.5
Insect [f]	4.0	6.5	11.5	8.4
Polychaete	5.8	9.0	0.3	0.4
Bivalve	0.2	0.0	0.0	0.0
Unidentified animal	0.8	1.0	0.4	0.2

a. Foraminiferan, tintinnid, hydroid, or rotifera.

b. Planktonic and epibenthic crustaceans < 1 mm body length (BL); mainly Harpacticoid copepods.

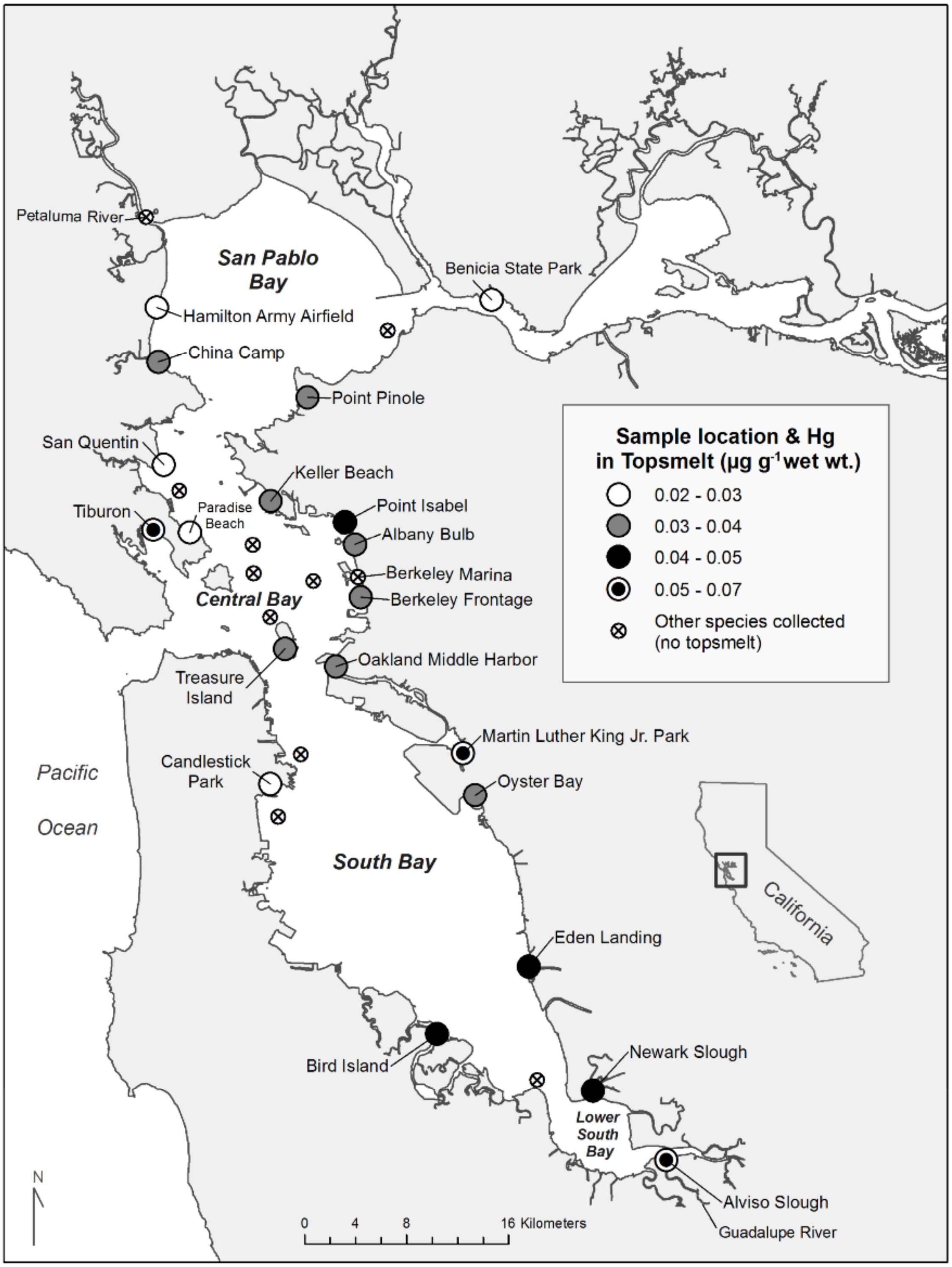
c. Planktonic crustaceans > 1 mm BL (calanoid copepods, Cyprid larva, *Neomysis* spp., and larval *Crangon* spp.).

d. Cumaceans (*Nipoleucon hinumensis*) and copepoda (*Coullana sp.*)

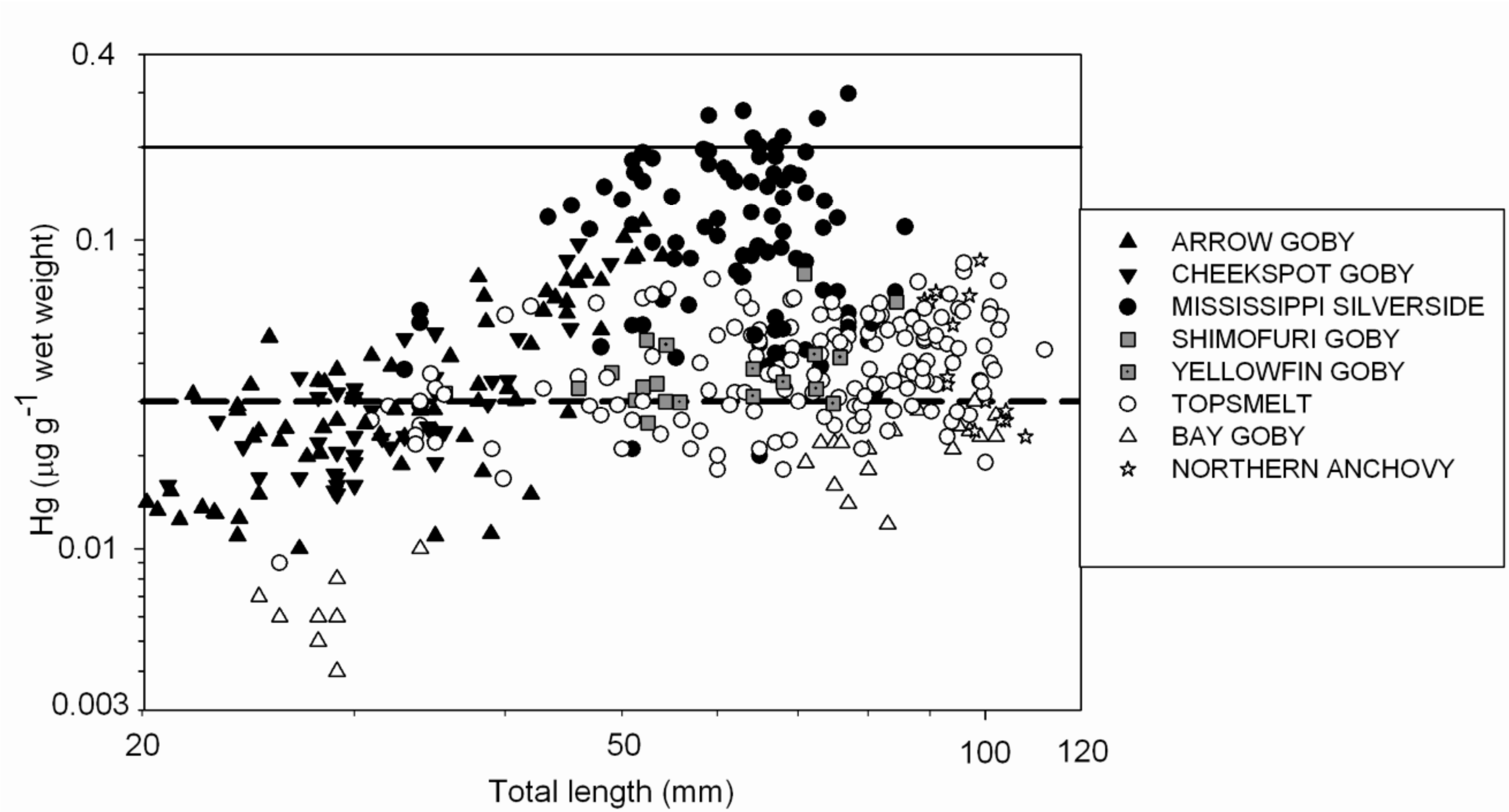
e. Amphipoda (e.g., *Corophium heteroceratum*), Tanaidacea (e.g., *Pancolus californiensis*), and Isopoda (e.g., *Synidotea harfordi*)

f. Hemiptera, Diptera, and Coleoptera.

Fig. 1.


Petaluma River
San Pablo Bay
Hamilton Army Airfield
China Camp
Benicia State Park
Point Pinole
San Quentin
Keller Beach
Tiburon
Paradise Beach
Point Isabel
Albany Bulb
Berkeley Marina
Central Bay
Berkeley Frontage
Treasure Island
Oakland Middle Harbor
Martin Luther King Jr. Park
Candlestick Park
Oyster Bay
Pacific Ocean
South Bay
Eden Landing
Bird Island
Newark Slough
Lower South Bay
Alviso Slough
Guadalupe River
N
0 4 8 16 Kilometers
California
Sample location & Hg in Topsmelt (µg g-1 wet wt.)
0.02 - 0.03
0.03 - 0.04
0.04 - 0.05
0.05 - 0.07
Other species collected (no topsmelt)

721 Fig. 2.



Hg (µg g^{-1} wet weight)
0.4
0.1
0.01
0.003
20
50
100
120
Total length (mm)
ARROW GOBY
CHEEKSPOT GOBY
MISSISSIPPI SILVERSIDE
SHIMOFURI GOBY
YELLOWFIN GOBY
TOPSMELT
BAY GOBY
NORTHERN ANCHOVY

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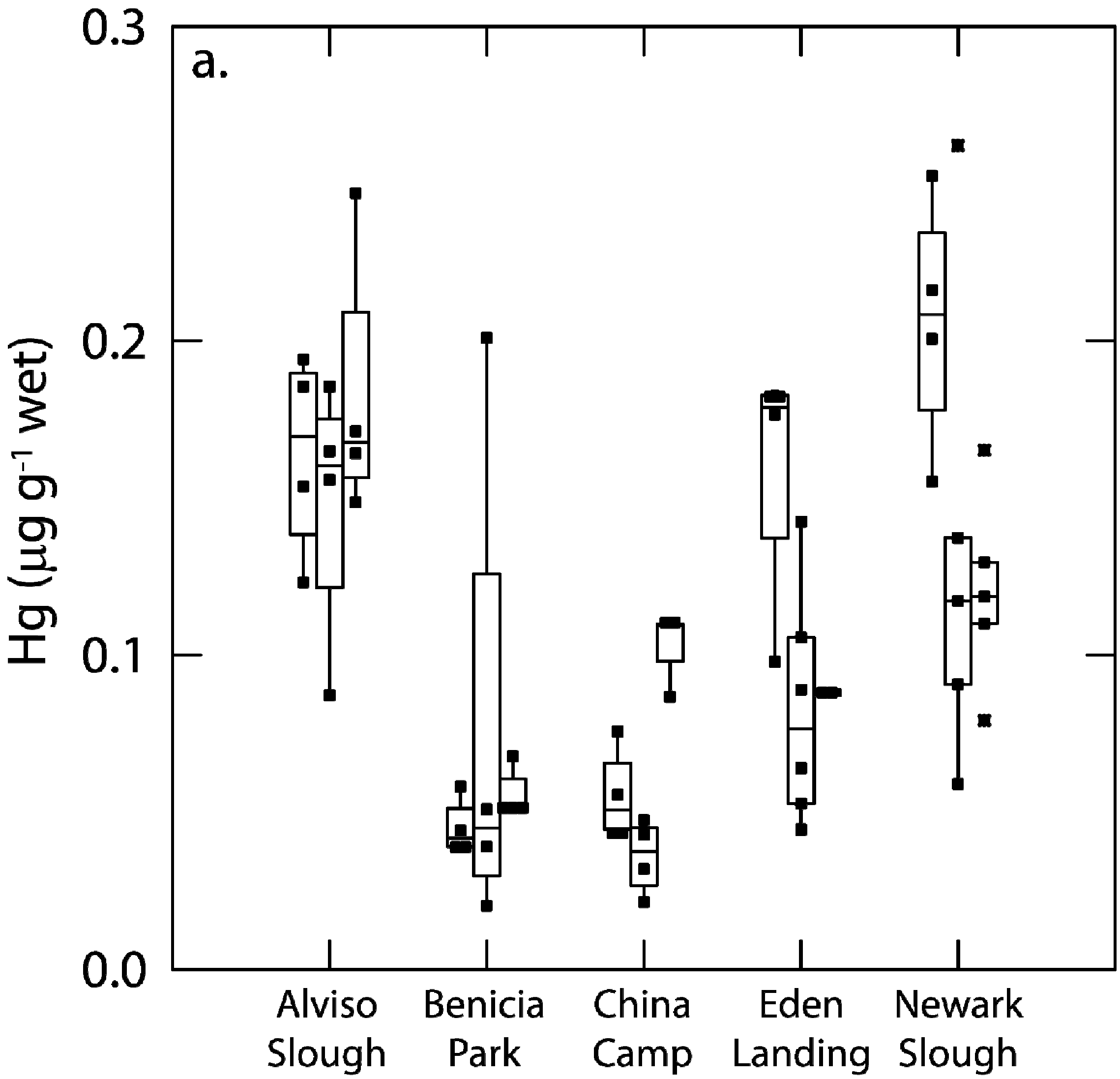
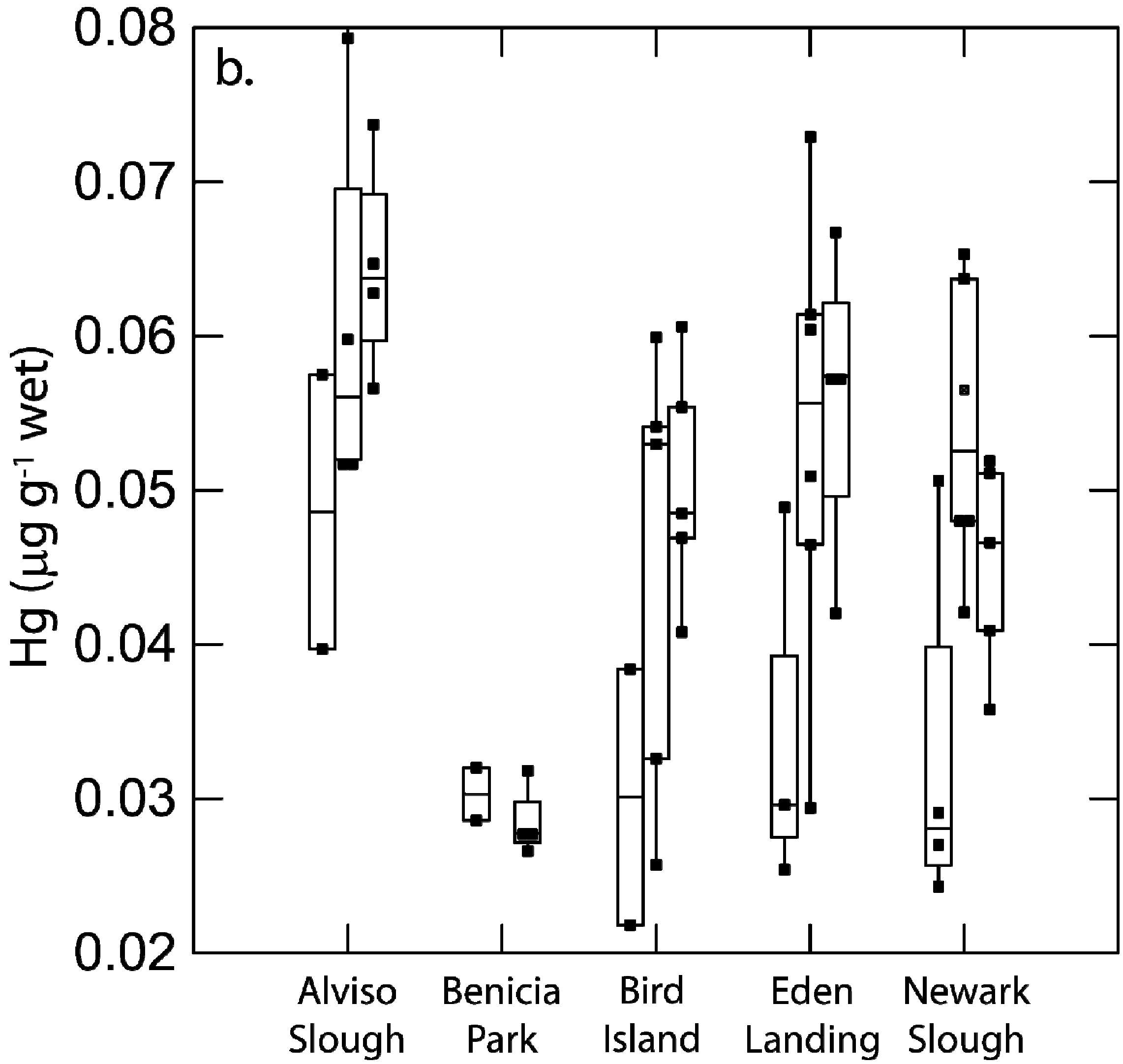
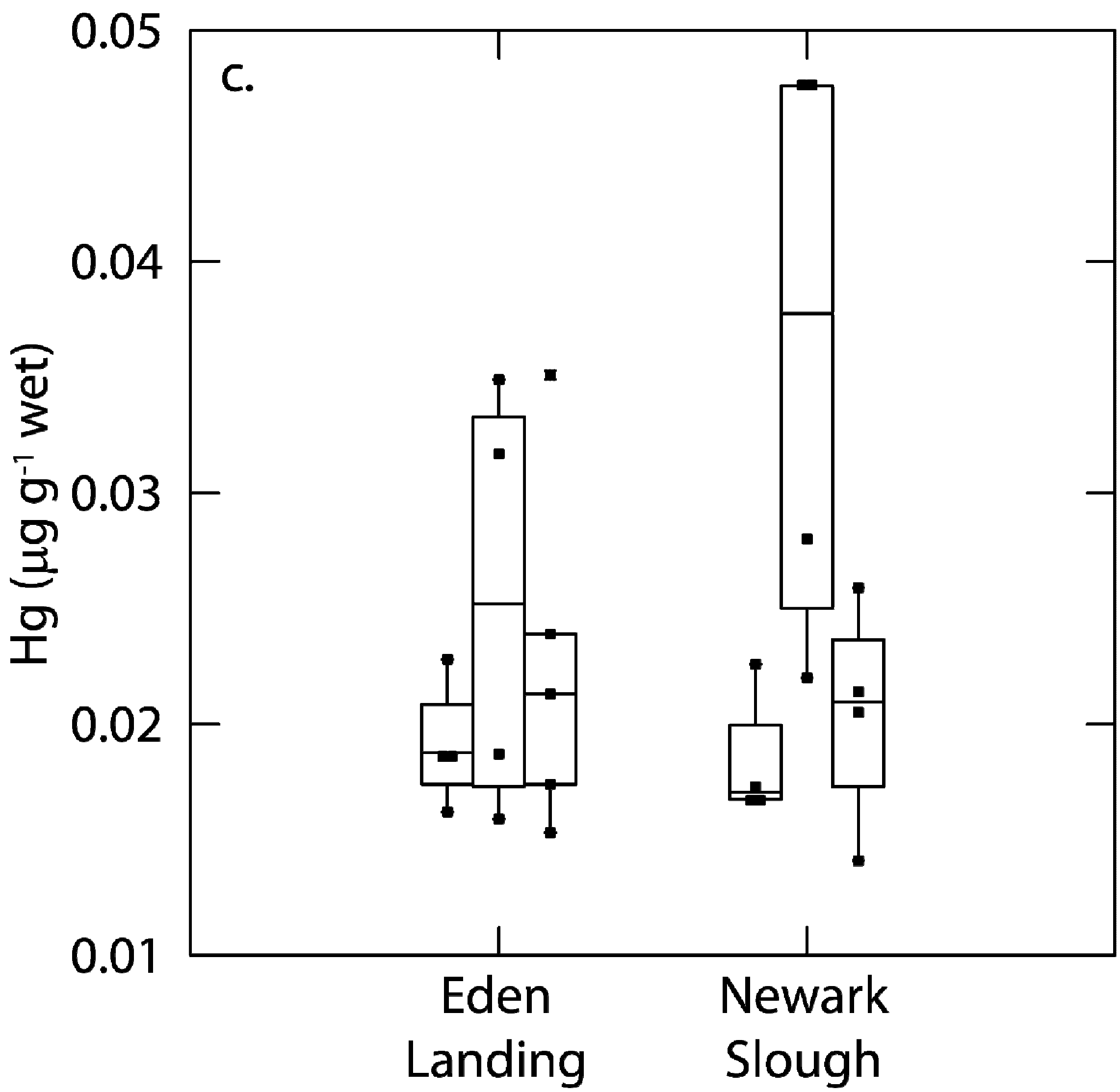
Fig. 3.

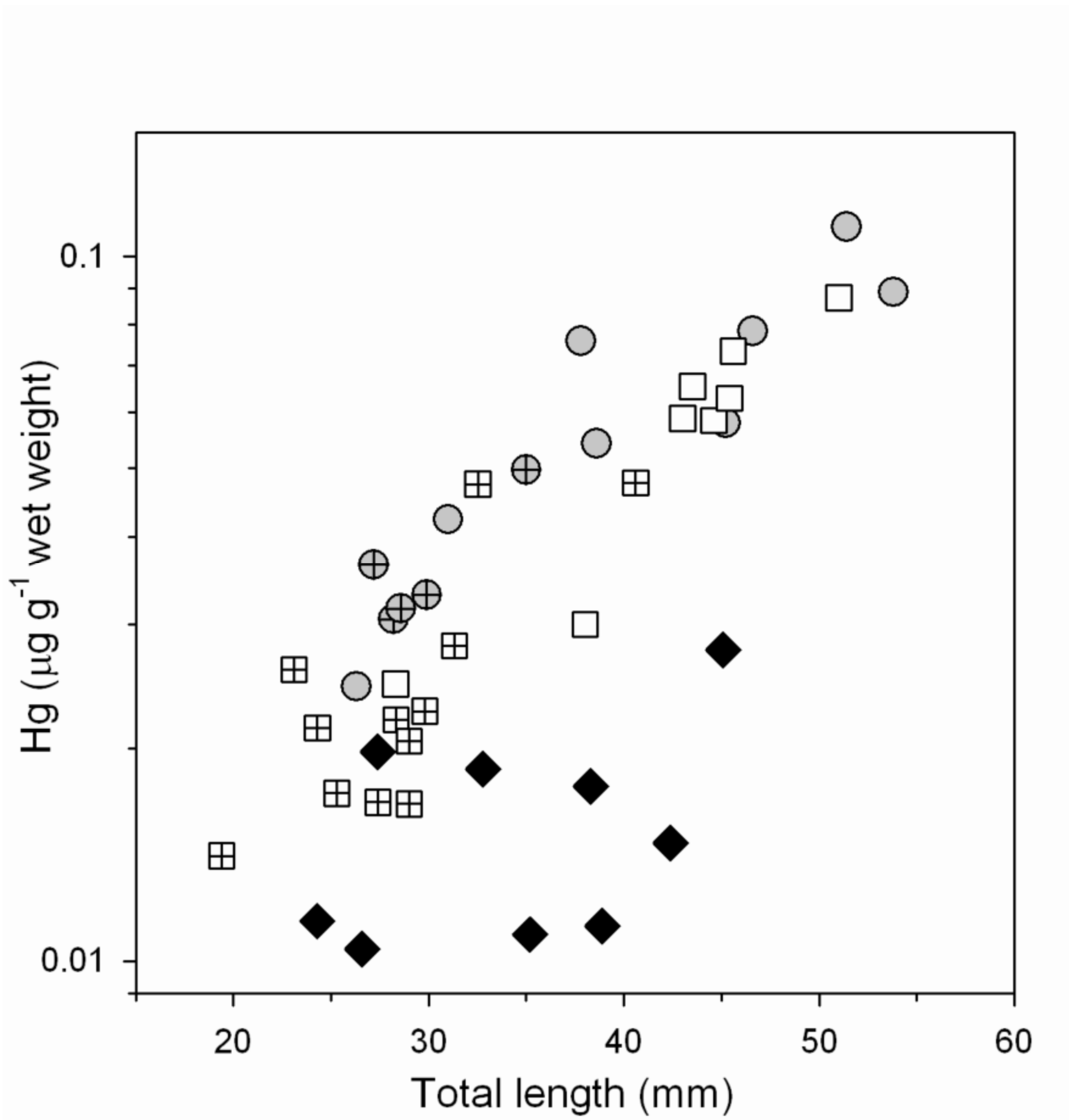

a.
0.3
0.2
0.1
0.0
Hg (µg g-1 wet)
Alviso Slough
Benicia Park
China Camp
Eden Landing
Newark Slough

725


b.
Hg (µg g-1 wet)
0.08
0.07
0.06
0.05
0.04
0.03
0.02
Alviso Slough
Benicia Park
Bird Island
Eden Landing
Newark Slough


c.
Hg (µg g-1 wet)
0.05
0.04
0.03
0.02
0.01
Eden
Landing
Newark
Slough

727 Fig. 4.



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