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Mercury bioaccumulation in estuarine wetland fishes: Evaluating habitats and risk to coastal wildlife

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ABSTRACT

Estuaries are globally important areas for methylmercury bioaccumulation because of high methylmercury production rates and use by fish and wildlife. We measured total mercury (THg) concentrations in ten fish species from 32 wetland and open bay sites in San Francisco Bay Estuary (2005–2008). Fish THg concentrations (μg/g dry weight ± standard error) differed by up to 7.4× among estuary habitats. Concentrations were lowest in open bay (0.17 ± 0.02) and tidal wetlands (0.42 ± 0.02), and highest in managed seasonal saline wetlands (1.27 ± 0.05) and decommissioned high salinity salt ponds (1.14 ± 0.07). Mercury also differed among fishes, with Mississippi silversides (0.87 ± 0.03) having the highest and longjaw mudsuckers (0.37 ± 0.01) the lowest concentrations. Overall, 26% and 12% of fish exceeded toxicity benchmarks for fish (0.20 μg/g wet weight) and piscivorous bird (0.30 μg/g wet weight) health, respectively. Our results suggest that despite managed wetlands' limited abundance within estuaries, they may be disproportionately important habitats of Hg risk to coastal wildlife.

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1. Introduction

Estuaries are among the most important habitats globally for maintaining biological diversity (Gray, 1997), aquatic productivity (Armstrong et al., 2003), and ecosystem services (Costanza et al., 1997). In particular, estuarine wetlands are important nurseries for many fish species (Beck et al., 2001), and foraging and breeding habitats for waterbirds (Erwin, 1996; Granadeiro et al., 2007). However, estuaries also are among the most threatened ecosystems worldwide. More than 50% of the world's population lives within 50 km of these coastal habitats (Small and Nicholls, 2003), and wetland habitat loss has substantially reduced these ecologically-important environments (Kennish, 2002). Chemical pollution has reduced the quality of existing estuarine wetland habitats, further threatening the fish and wildlife that utilize them. This is particularly concerning for mercury (Hg) pollution, which is uniquely problematic in estuaries for two reasons. First, estuaries are depositional areas for inorganic Hg from upstream watersheds (Gehrke et al., 2011; Singer et al., 2013). Secondly, conditions within estuarine habitats enhance the production of methylmercury (MeHg), which is the most bioaccumulative and toxic form (Hall et al., 2008; Lambertsson and Nilsson, 2006). In particular, estuaries have pronounced gradients in salinity, organic carbon cycling, and geochemical parameters that are conducive to MeHg production (Rolfhus et al., 2003). Thus, the convergent threats of increasing reliance by fish and wildlife on a reduced availability of estuarine wetlands, coupled with elevated Hg contamination within those wetland habitats, is an important conservation issue.

In response to the global decline in estuarine wetlands, efforts to increase habitat by restoring lost, and enhancing existing wetlands have been widely implemented. However, restoration efforts often result in creating more highly managed wetlands as opposed to conversion to habitats with unregulated hydrologic regimes in order to meet competing ecological and anthropogenic demands. Managed estuarine wetlands are wetlands enclosed with earthen levees in order to isolate them from direct tidal influence. In becoming more highly managed, these environments increase the available habitat for fish and wildlife that would otherwise be lost, while also protecting coastal developments from flooding. Many impounded estuarine wetlands are managed specifically for wildlife, particularly waterbirds (Beauchard et al., 2013; Coates et al., 2012). Others are designed for purposes such as salt production, flood control (Cox et al., 2006; Zanuttigh, 2011), aquaculture (Bui et al., 2013), or mosquito abatement (Middleton et al., 2008), but these impounded wetlands are nonetheless used extensively by waterbirds and have become globally important habitats for

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wildlife (James-Pirri et al., 2012; Lehnen and Krementz, 2013; Takekawa et al., 2001; Underwood et al., 2013). For example, habitat use of several waterbird species in coastal Louisiana was higher in managed impoundments than in more natural unimpounded wetlands (Bolduc and Afton, 2004). Similarly, managed wetlands in coastal Texas supported more bird species and greater bird densities than nearby unmanaged wetlands (Fitzsimmons et al., 2012).

Importantly, managed wetlands often represent only a small portion of an estuary's landscape, yet they are often disproportionately important for wildlife (Rosa et al., 2003). For example, managed wetlands represent less than 15% of the inundated area within the San Francisco Bay Estuary, California, but they are more important than the more expansive open water zones for many breeding waterbird species (Ackerman et al., 2008; Hickey et al., 2007; Strong et al., 2004; Takekawa et al., 2002; Warnock and Takekawa, 1995). Although managed estuarine wetlands are particularly common in urbanized and industrialized estuaries, less populated estuaries also contain an abundance of these habitats. In fact, more than 20% of the 314 estuarine RAMSAR Wetlands of International Importance contain substantial areas of managed wetlands (http://www.ramsar.wetlands.org/). The value of impounded estuarine wetlands as wildlife habitat illustrates the importance in understanding Hg dynamics within them relative to other estuarine habitats.

Despite their worldwide importance and disproportionately heavy use by fish and wildlife, only limited research exists on Hg dynamics within managed estuarine wetlands. Instead, most estuarine Hg research has focused on the subtidal open waters, channels, and intertidal mudflats that tend to occupy the greatest spatial extent of estuaries. We identified 460 peer-reviewed studies published during the last decade (2002–2012) with keywords "estuary" and "mercury" (ISI Web of Knowledge) across 234 different estuaries and six continents. Of those studies where we could determine focal habitats ($n = 284$), open water (62%) and channel (36%) habitats were sampled most often, whereas less than 8% of studies included impounded wetland sites along an estuary's margin despite the fact that they existed in 35% of the estuaries studied. Although the open water and subtidal areas are important for understanding the distribution and cycling of Hg within estuaries, they may not be representative of the Hg risk faced by many estuarine wildlife. Importantly, managed estuarine wetlands may also represent the greatest potential for reducing MeHg in coastal wildlife because these wetlands typically have the infrastructure needed to apply hydrological controls (timing, duration, frequency, and depth of water inundation) that could potentially reduce MeHg production and bioaccumulation. Therefore, understanding how MeHg production and bioaccumulation vary among different types of estuarine wetlands is helpful for determining Hg sources to estuarine food webs.

We evaluated Hg bioaccumulation within fish communities of 32 impounded and intertidal wetlands, and open water sites of the San Francisco Bay Estuary, California, USA. The estuary is highly urbanized and has a well-documented legacy of Hg contamination from historic gold and Hg mining in its upper watersheds (Hornberger et al., 1999; Singer et al., 2013). The Estuary also is considered a site of hemispheric importance to waterbirds, supporting over a million wintering, migrating, and breeding birds annually (Page et al., 1999). Importantly, many of these birds rely on a limited number of managed wetlands, such as former salt evaporation ponds and muted-tidal marshes, for critical foraging and breeding habitat (Strong et al., 2004; Takekawa et al., 2001; Warnock et al., 2002). Additionally, recent work has indicated that the region is a hotspot for wildlife exposure to Hg, particularly for bird species that depend on the managed wetland impoundments adjacent to the Bay (Eagles-Smith et al., 2009). Our primary objectives were to determine whether these managed wetlands within estuaries represent enhanced risk for Hg exposure to fish and fish-eating birds relative to the more commonly studied open-bay habitats, and to evaluate potential Hg exposure among different classifications of wetland habitats. To do so, we: (1) compared Hg concentrations among 10 of the most common fish species that comprise the prey base for many fish-eating birds, (2) assessed the spatial variation in fish Hg concentrations among wetlands within the estuary, and (3) evaluated whether there were differences in fish Hg concentrations among different habitat types to facilitate wetland management decisions targeted to minimize risk of Hg to fish and fish-eating birds.

2. Methods

2.1. Study area

The San Francisco Bay Estuary has lost more than 90% of its historical tidal wetland area (Nichols et al., 1986). Much of the remaining habitat was diked and converted to managed wetlands such as salt-evaporation ponds, seasonally-inundated wetlands, and muted-tidal marshes (Williams and Orr, 2002). Although it still contains an array of wetland types, the Estuary is spatially dominated by subtidal, open water habitat and channelized sloughs, with smaller intertidal and managed wetlands along its perimeter. Mercury contamination within the estuary comes both from legacy upstream sources in the north and south Bay, as well as some likely atmospheric deposition. Although inorganic Hg loading from legacy sources has been an important long term contributor to the Hg profile within the estuary, little is known about the relative importance of different Hg sources to MeHg production in the wetlands along the Estuary's margins.

We sampled fish from 32 distinct locations throughout the San Francisco Bay between 2005 and 2008, which included diked former salt evaporation ponds, breached former salt ponds, diked seasonally-flooded saline wetlands, and unmanaged tidal sloughs, marshes, and open bay habitats (Fig. 1). The managed wetlands in this study included those within several managed complexes that comprise the Don Edwards San Francisco Bay National Wildlife Refuge in the southern portion of the Estuary, and the Napa Marsh Wildlife Area in the northern portion of the Estuary.

We identified eight distinct wetland regions or complexes and assigned individual wetlands within regions based upon their proximity to one another and their hydrologic connectivity (Fig. 1). The Moffet Region contained former salt ponds A1, A2W, A2E, AB1, and AB2. The West Alviso Region contained former salt ponds A5, and A7, and a seasonally-inundated former salt pond (A8). The Central Alviso Region contained former salt ponds A12, A13, A16, A17, A18, and an adjacent seasonally inundated wetland (New Chicago Marsh). The East Alviso Region contained former salt ponds A19, A20, and A21. The Mowry Region contained ponds M1 and M2, and the Newark Region contained former ponds N2A, N4A, N6, and N9. We also sampled tidal sloughs and marshes within the South Bay region, including Alviso Slough, Charleston Slough, and Coyote Creek, as well as several subtidal, open bay areas south of the Dumbarton Bridge. The North Bay Region consisted of open bay area within San Pablo Bay, and former salt ponds, which were all hydrologically connected with the open bay via either levee breaches or culverts.

2.2. Fish sampling

We sampled fish between March and August of each year using beach seines (3 m × 1.5 m or 6 m × 1.5 m, 3 mm mesh; salt ponds,

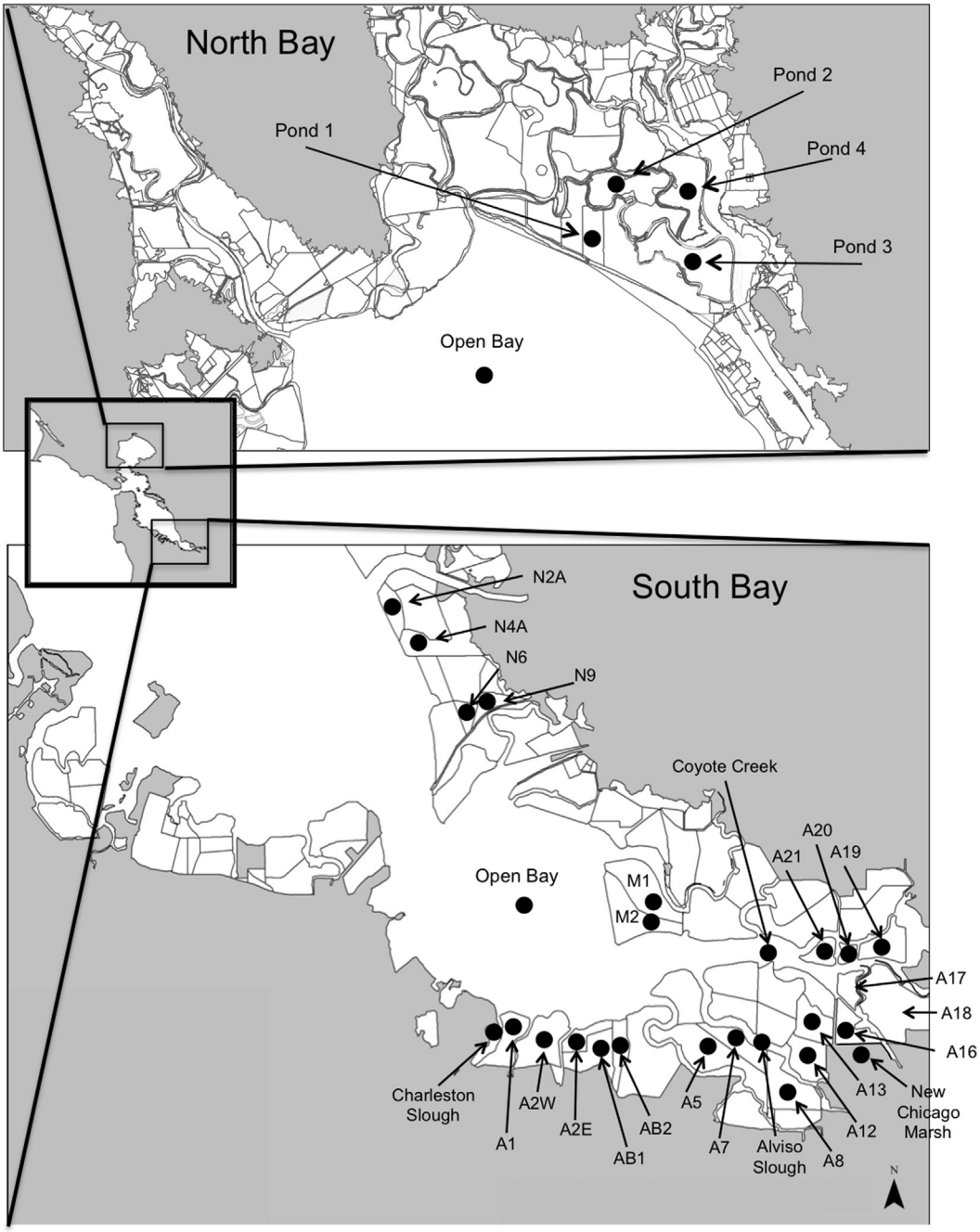

Fig. 1. Map of study area in San Francisco Bay, California. Impounded wetlands and salt ponds are denoted with a letter and number sequence.

seasonally-flooded wetlands, tidal sloughs), minnow traps (salt ponds, seasonally flooded wetlands), variable mesh-gill nets (salt ponds), and benthic and midwater-trawls (open bay, tidal sloughs). To link prey fish Hg concentrations with avian Hg risk, fish sampling sites were located within population home ranges of pre-breeding and breeding fish-eating waterbirds (Ackerman et al., 2008; Eagles-Smith and Ackerman, 2009). Each fish was identified to species, placed into labeled polyethylene bags, and stored on wet ice until return to the laboratory within 8 h. Upon return to the laboratory, fish were stored at −20° C until processing for Hg analysis. Across all habitats and locations, we sampled 10 different species of fish, including: longjaw mudsucker (*Gillichthys mirabilis*), Mississippi silverside (*Menidia audens*), northern anchovy (*Engraulis mordax*), prickly sculpin (*Cottus asper*), rainwater killifish (*Lucania parva*), shiner surfperch (*Cymatogaster aggregata*), staghorn sculpin (*Laptocottus armatus*), topsmelt (*Atherinops affinis*), threespine stickleback (*Gasterosteus aculeatus*), and yellowfin goby (*Acanthogobius flavimanus*).

2.3. Sample processing and chemical determination

Prior to THg analysis, we washed all samples in deionized water, patted them dry with lint-free wipes, and processed them as described in Eagles-Smith and Ackerman (2009). We analyzed all samples for THg concentrations because 94% of the THg in prey fish from San Francisco Bay is in the MeHg form (Ackerman et al., 2013). Total Hg concentrations were determined following Eagles-Smith and Ackerman (2009) on a Milestone DMA-80 Direct Mercury

Analyzer (Milestone, Monroe, CT, USA). Quality assurance measures included analysis of two certified reference materials, two system and method blanks, two duplicates, one matrix spike, and one matrix spike duplicate per batch of 28 samples. Recoveries averaged 107.0 ± 0.6% ($N = 112$) and 99.4 ± 0.4% ($N = 208$) for certified reference materials and calibration checks, respectively. Matrix spike recoveries averaged 103.9 ± 1.0% ($N = 146$). Absolute relative percent difference (RPD) for duplicates averaged 4.5 ± 0.4%, and absolute RPD for matrix spike duplicates averaged 2.6 ± 0.4%.

2.4. Statistical analyses

Statistical analyses were conducted using JMP V11.0 (SAS Institute, Cary, NC). We evaluated spatial and temporal trends in fish THg concentrations using a nested analysis of covariance (ANCOVA) model that included region (as outlined above), wetland site nested within region, collection year, and fish species as fixed categorical factors, standard length and date (day-of-year, standardized to the mean sampling data) as continuous covariates, and a species × length interaction. We evaluated non-linear temporal variation in fish THg concentrations by including date as both a linear and quadratic variable based upon past studies of seasonal variation in fish THg concentrations (Eagles-Smith and Ackerman, 2009). We could not adequately interpret the main effects of this model because of the significant species × length interaction (see Results), indicating that the relationships between THg concentrations and fish length differed among species. We therefore size-standardized each individual fish's THg concentration to the species-specific average length following Eagles-Smith and Ackerman (2009). Briefly, we evaluated the relationship between THg and fish length for each species while controlling for the other spatial and temporal factors (described above). Once we size-standardized each individual fish's THg concentrations, we re-conducted the above ANCOVA model but excluded size as a covariate since each fish was size-standardized.

In a second analysis, we examined the effects of habitat type on fish THg concentrations using a mixed-effects ANCOVA model. For this analysis we classified each wetland as a specific habitat type (high-salinity salt pond [60–80 ppt], medium-salinity salt pond [40–60 ppt], low-salinity salt pond [20–40 ppt], breached former salt pond, seasonal saline wetland, tidal slough, and open bay). We could not include habitat type in the first model used to test spatial effects because management changed in some of the wetlands during the course of the study, resulting in some sites being characterized as different habitat types depending upon sampling year. Therefore, to evaluate effects of habitat type on fish THg concentrations we included wetland site as a random effect nested with habitat type while accounting for the potential effects of species, date, and year (categorical).

Unless otherwise specified, we present our results as back-transformed least-squares means with standard errors estimated using the delta method (Williams et al., 2002). We present all tissue THg concentrations on a dry weight basis because Hg is associated with the protein matrix of tissues, variability in moisture content can add variance to the data, and there were clear differences in moisture content among species which would have affected the interpretation of the THg differences observed among species. However, to facilitate comparisons with other studies we provide species-specific estimates of moisture content (mean ± standard error) for conversion from dw to wet weight concentrations (shiner surfperch: 77.5 ± 1.3%; longjaw mudsucker: 75.8 ± 0.1%; prickly sculpin: 75.4 ± 0.9%; yellowfin goby: 75.2 ± 0.4%; staghorn sculpin: 74.2 ± 0.2%; topsmelt: 72.1 ± 0.2%; rainwater killifish: 71.8 ± 0.2%; northern anchovy: 71.7 ± 0.4%; Mississippi silverside: 71.1 ± 0.1%; threespine stickleback: 69.5 ± 0.1%).

3. Results

Between 2005 and 2008, we sampled 3098 individual fish across 10 species from 27 managed wetlands, 3 tidal sloughs and marshes, and 2 open bay regions of the San Francisco Bay. Across all sites, dates, and fish lengths, the geometric mean THg concentrations (±standard error; μg/g dw) in whole-body fish were 0.83 ± 0.02 in Mississippi silversides ($N = 486$), 0.55 ± 0.02 in topsmelt ($N = 365$), 0.51 ± 0.04 in northern anchovy ($N = 89$), 0.45 ± 0.01 in threespine stickleback ($N = 687$), 0.44 ± 0.02 in rainwater killifish ($N = 297$), 0.38 ± 0.03 in staghorn sculpin ($N = 54$), 0.36 ± 0.01 in longjaw mudsuckers ($N = 1000$), 0.36 ± 0.02 in yellowfin goby ($N = 74$), 0.35 ± 0.03 in prickly sculpin ($N = 20$), and 0.29 ± 0.04 in shiner surfperch ($N = 26$).

We could not adequately interpret the main effects of our initial global ANCOVA model because there was a significant species × fish length interaction ($F_{9,3042} = 48.80$, $p < 0.0001$), indicating that the relationship between THg concentration and fish length differed among species. We therefore conducted separate analyses for each species to evaluate the relationship between THg concentrations and fish length while accounting for variability associated with spatial and temporal factors in the global model. Even though we sampled a wide range of fish lengths for each species, THg concentrations did not always correlate strongly with fish length. In fact, we found no relationship between THg concentrations and fish length in northern anchovy, staghorn sculpin, or yellowfin goby, and very weak correlations in stickleback, and mudsucker (Fig. 2). Surfperch ($R^2 = 0.86$) and silverside ($R^2 = 0.55$) showed the strongest relationships between THg concentrations and fish length, followed by prickly sculpin ($R^2 = 0.45$), topsmelt ($R^2 = 0.36$), and killifish ($R^2 = 0.29$; Fig. 2). In those species with significant length-Hg correlations, we standardized each fish's THg concentration to the mean species-specific standard length for all further analyses (mudsuckers = 68 mm; silversides = 50 mm; prickly sculpin = 48 mm; killifish = 28 mm; surfperch = 87 mm; topsmelt = 85 mm; stickleback = 39 mm).

After accounting for wetland site, date, and year, the least square mean THg concentrations in length-standardized fish differed substantially among species ($F_{9,3052} = 128.39$, $p < 0.0001$; Fig. 3). There was a 2.4-fold difference between the species with the highest (silverside; 0.87 ± 0.03) and lowest (mudsucker (0.37 ± 0.01)) THg concentrations. We could not estimate least square means for prickly sculpin or shiner surfperch because they lacked sample size and appropriate spatial distribution to effectively account for sampling site and date effects. Therefore, they were excluded from the model, and instead report geometric mean concentrations, which were both lower than in mudsuckers. For the remaining eight species, we found a general pattern where THg concentrations were highest in the most pelagic-oriented species (Mississippi silverside and northern anchovy), lowest in the most benthic-oriented species (longjaw mudsucker and yellowfin goby), and intermediate in those species that generally exhibit less habitat specificity (threespine stickleback, rainwater killifish, topsmelt, and staghorn sculpin; Fig. 3).

Fish THg concentrations exhibited substantial spatial variation, differing both among regions ($F_{7,3052} = 195.44$, $p < 0.0001$; Fig. 4a) and wetland sites nested within regions ($F_{24,3052} = 66.15$, $p < 0.0001$; Fig. 4b). Total Hg concentrations in fish within wetland regions differed by more than 4-fold. Among all wetland sites, THg concentrations varied considerably with nearly a 15-fold range in mean THg concentrations between the wetland sites with the highest and lowest THg concentrations. Even within regions, mean THg concentrations between wetland sites with the highest and lowest THg concentrations differed by up to 4-fold. In fact, wetland sites directly adjacent to one another and only separated by a

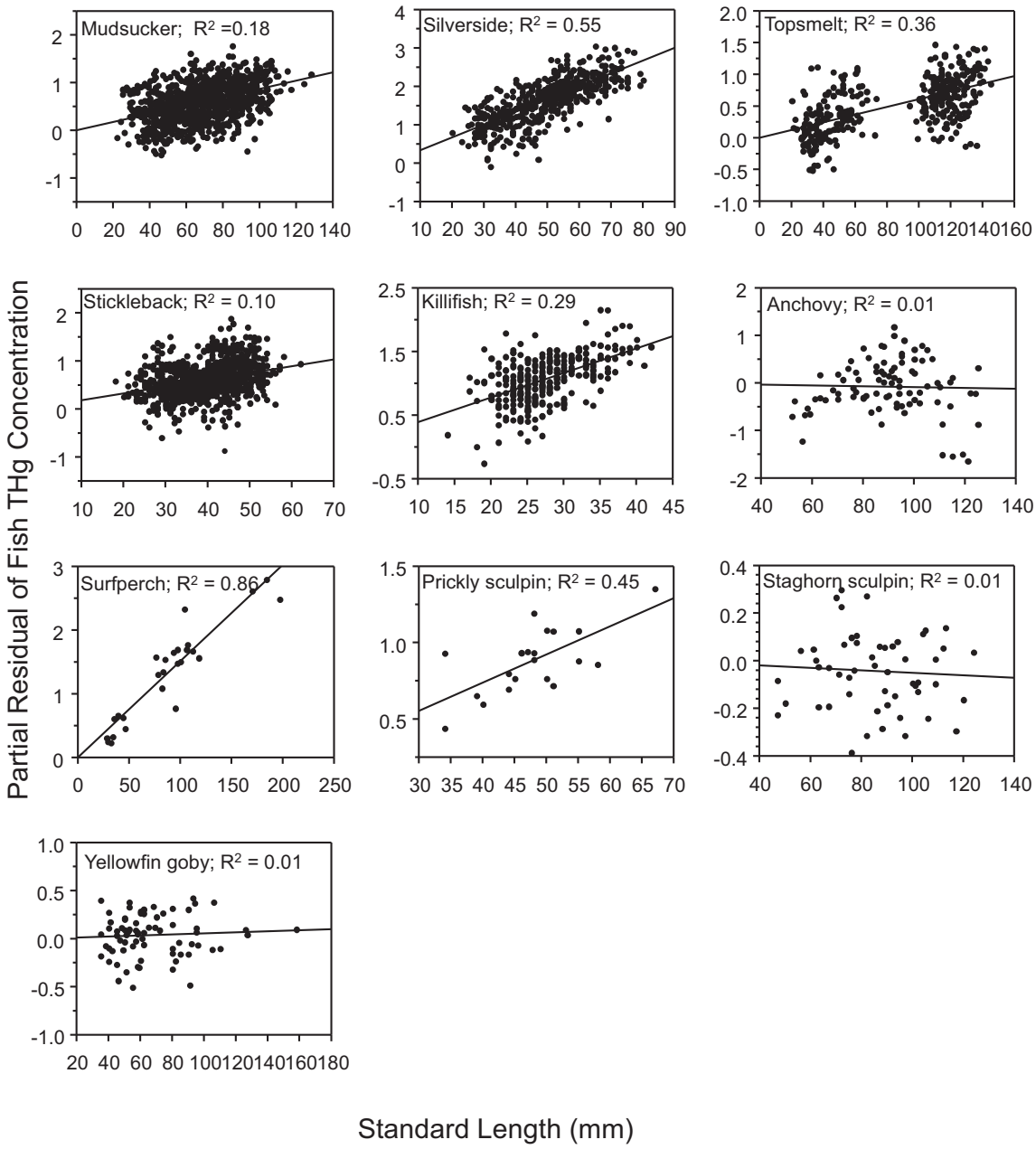

Fig. 2. Partial residual plot depicting whether total mercury (THg) concentrations were correlated with standard length in each species of fish. Partial residual plots accounted for the effects of site, date, and year.

narrow levee, such as ponds A5 and A8 in the Central Alviso Region, differed in fish THg concentrations by a factor of 4 (Fig. 4b).

Fish THg concentrations also differed temporally, both among years ($F_{3,3047} = 47.75$, $p < 0.0001$), and by date (date: $F_{1,3047} = 143.35$, date^2: $F_{1,3047} = 158.40$, $p < 0.0001$). Fish THg concentrations were higher in 2005 (0.55 ± 0.02) and 2007 (0.57 ± 0.03) than in 2006 (0.43 ± 0.01) or 2008 (0.40 ± 0.01). Fish THg concentrations also followed a nonlinear, quadratic relationship with date, where fish THg concentrations increased from spring to a maximum value in June, then declined again in fall.

After categorizing each wetland site into their respective habitat categories, we evaluated the influence of habitat type on size-standardized fish THg concentrations. While statistically accounting for the effects of species, wetland site, date, and year, we found that habitat type substantially influenced fish THg concentrations ($F_{6,3050} = 156.66$, $p < 0.0001$). Fish THg concentrations were highest in seasonal saline wetlands and high-salinity salt ponds, followed by breached salt ponds and moderate-salinity salt ponds (Fig. 5). Total Hg concentrations in fish from seasonal saline wetlands and high-salinity salt ponds were more than 7 times higher than in subtidal, open bay habitats, and at least 2 times higher than any other wetland habitat. Total Hg concentrations in fish from subtidal, open bay sites were 2.7 times lower than any of the managed wetland habitats, and 2.4 times lower than tidal sloughs and marshes. Tidal sloughs and marshes had THg concentrations similar to low salinity salt ponds.

4. Discussion

Using a multi-species evaluation of Hg bioaccumulation across 32 different wetland and open water sites, we found strong spatial, temporal, and interspecific variation in THg concentrations of small

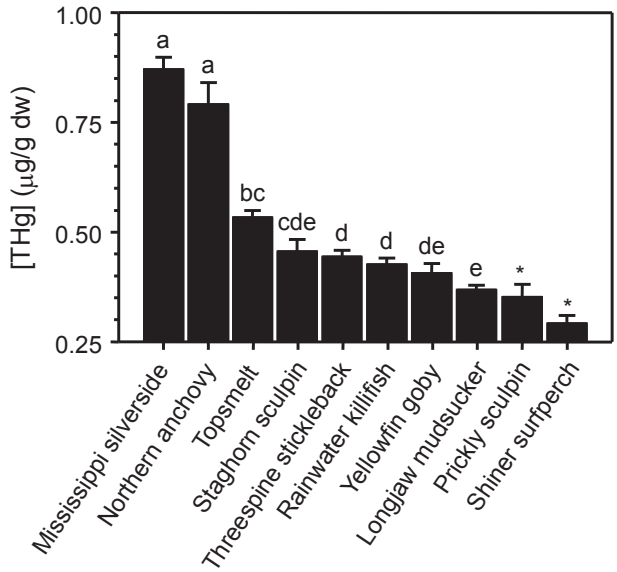

Fig. 3. Comparisons of total mercury (THg) concentrations (μg/g dry weight [dw]; size normalized least square means ± standard error) among fish species from San Francisco Bay, California wetlands. Least square means account for global model effects of region, wetland [region], date, and year. Concentrations are normalized to the species-specific mean standard length (silverside = 50 mm, anchovy = 90 mm, prickly sculpin = 48 mm, surfperch = 85 mm, topsmelt = 85 mm, staghorn sculpin = 85 mm, stickleback = 39 mm, killifish = 28 mm, yellowfin goby = 66 mm, mudsucker = 50 mm) (Tukey's Post-Hoc Pairwise Comparisons, $\alpha < 0.05$). Different letters indicate significant differences among groups. Asterisks indicate that geometric means are presented for prickly sculpin and surfperch, which did not occur at enough locations to adequately estimate representative least squared means.

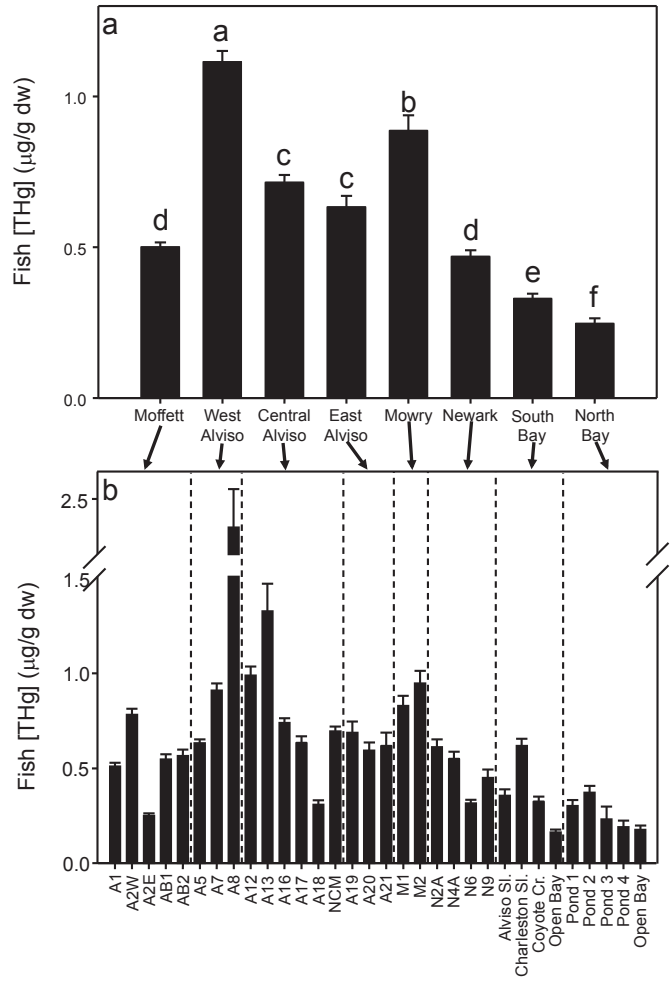

Fig. 4. Total mercury (THg) concentrations (μg/g dry weight [dw]; least square means ± standard error) in size-standardized fish from San Francisco Bay, California wetlands. Least square means account for global model effects of species, region, wetland [region], date, and year. 4a) THg concentrations differed among regions and different letters indicate significant differences ($\alpha < 0.05$, Tukey's Post-Hoc Pairwise Comparisons). 4b) THg concentrations differed among wetlands nested within regions.

fish in the San Francisco Bay Estuary. Our results highlight both the complexity of estuarine Hg cycling, as well as the utility of employing a multi-species approach across a diverse habitat mosaic. Specifically, we found that fish Hg concentrations were substantially more elevated in managed wetland habitats along the Estuary's margins than in the open bay habitat that occupies the majority of the area's aquatic environment. The differences in fish Hg concentrations among managed wetland habitat types, even those in close proximity to one another, emphasizes the overriding role that habitat characteristics can play in driving Hg risk to ecosystems. Not only are fish in wetlands along the Estuary's margins elevated in Hg, but these habitats are also the preferred foraging and breeding areas for the majority of waterbirds that use the estuary (Ackerman et al., 2008; Strong et al., 2004; Takekawa et al., 2002; Warnock and Takekawa, 1995). Despite representing only a small proportion of the Estuary's area, the combination of high fish Hg and high waterbird use make estuarine wetlands potential hotspots for Hg bioaccumulation. Yet, managed estuarine wetlands may also provide a unique opportunity to manage Hg risk due to infrastructure to manipulate water flow and water quality factors that influence Hg cycling. As such, integrating managed estuarine wetland habitats into assessments and monitoring of estuarine Hg risk could be particularly useful.

Mercury concentrations in San Francisco Bay fishes were elevated in comparison to forage fish from other locations around the world, and relative to toxicological risk thresholds. The mean THg concentration for all fishes in San Francisco Bay Estuary was 0.16 μg/g ww. This THg concentration is 3–6 times higher than ecologically similar prey fish species from Narraganset Bay Estuary, RI, USA (Szczebak and Taylor, 2011), 4 times higher than the Gulf of Maine (Chen et al., 2009), and 4 to 20 times higher than Barataria Bay Louisiana (Fry and Chumchal, 2012). Of the nearly 3100 fish that we sampled in the San Francisco Bay Estuary, 26% ($N = 796$) had THg concentrations exceeding 0.20 μg/g ww, a whole-body tissue benchmark associated with potential sublethal effects in fish (Beckvar et al., 2005). Additionally, 12% ($N = 386$) of all fish sampled exceeded 0.30 μg/g ww, which is a dietary benchmark developed for some fish-eating bird species that is associated with impaired reproduction (Albers et al., 2007; Burgess and Meyer, 2008; Shore et al., 2011). Importantly, most of the fish that exceeded these thresholds were collected from the managed wetlands and margins of the bay, as opposed to the open bay habitats. In fact, no fish sampled from the open bay habitats exceeded the 0.30 μg/g ww wildlife criterion and only 9% exceeded the fish health threshold in this habitat. Additionally, silversides are among the most commonly sampled fish in the estuary, and they comprise a large proportion of piscivorous bird diets. Considering only silversides, 36% of individuals exceeded the wildlife risk benchmark in managed wetlands, whereas no silversides sampled from the open bay habitat exceeded the benchmark in this study, nor was the benchmark exceeded in open bay habitats in prior studies San Francisco Bay (Greenfield and Jahn, 2010).

Habitat type is known to influence Hg cycling and bioaccumulation (Chen et al., 2009; Chetelat et al., 2011; Eagles-Smith

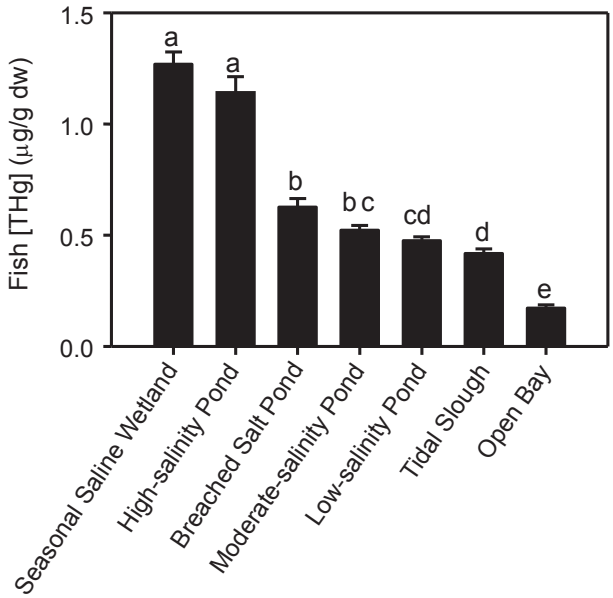

Fig. 5. Total mercury concentrations (THg μg/g dry weight [dw]; least square means ± standard error) across different habitat types in size-standardized fish from San Francisco Bay, California wetlands. Least square means account for global model effects of species, region, wetland [region], date, and year. Different letters indicate significant differences ($\alpha < 0.05$, Tukey's Post-Hoc Pairwise Comparisons).

et al., 2008a; Heim et al., 2007). Wetlands in particular facilitate MeHg production (Hall et al., 2008), and we found that fish THg concentrations were up to 6 times higher in impounded wetlands along the Estuary's margins than within the Bay itself. However, wetlands exist across a gradient of characteristics such as hydrology, biogeochemistry, and vegetation community structure (Day et al., 2012), which are important for Hg cycling (Guentzel, 2009; Mitchell and Gilmour, 2008; Schartup et al., 2013; Windham-Myers et al., 2009). Thus, some wetland habitats are more conducive to MeHg production and bioaccumulation than others. Indeed, after categorizing each wetland into one of five broad classifications based upon water permanence and salinity, we found substantial difference in fish THg concentrations among wetland habitats types, with seasonal saline wetlands and high-salinity former salt ponds having the highest fish THg concentrations. Moreover, the marked variability in Hg concentrations among individual wetlands in close proximity highlights the importance of factors other than inorganic Hg sources and loading in controlling MeHg bioaccumulation. Even for wetlands that were adjacent or hydrologically connected to one another through narrow gates and culverts, we documented more than 2-fold differences in fish Hg concentrations between neighboring wetlands. Nutrient dynamics, productivity rates, and temperature and salinity regimes of these wetlands also differ from one another substantially, highlighting the biogeochemical independence of them despite their close proximity to one another. Thus, the differences in pond-specific fish Hg concentrations may be linked to various pond-specific factors, such as MeHg production rates (Orihel et al., 2008; Wiener et al., 2006), food web structure (Eagles-Smith et al., 2008b; Kidd et al., 1999), and fish growth rates (Ward et al., 2010), but differences are unlikely to be caused simply by disparities in inorganic Hg loading. This is an important consideration for resource managers tasked with reducing MeHg risk to estuarine wildlife because it suggests that developing wetland management techniques that reduce MeHg bioaccumulation and production are likely more effective measures than trying to control inorganic Hg sources.

Although managed wetland impoundments represent a small proportion of the total area within estuaries, they serve as principal breeding and foraging habitats for sensitive wildlife, such as waterbirds (Strong et al., 2004; Warnock et al., 2002). Thus, relative to their availability in the environment these habitats can be proportionally more important for wildlife conservation than the more spatially expansive open bay and tidal environments. For example, within San Francisco Bay Estuary (south of Suisun Bay), managed wetland impoundments (20,165 ha) account for approximately 5 times less area than open bay and tidal habitats (115,123 ha), but are nonetheless habitats of primary use for shorebirds and fish-eating waterbirds that have been shown to have elevated Hg exposure (Ackerman et al., 2008; Eagles-Smith et al., 2009). Yet, most Hg research and monitoring in the estuary focuses on open bay and shoreline habitats that are more spatially expansive but do not necessarily provide as much value to waterbirds (Greenfield et al., 2013). This is also true in other estuaries, where the majority of studies focus on open water and channel habitats (see summary in Introduction). It is important to note that the majority of those studies were primarily designed to focus on important processes such as Hg transport, or sedimentary and aqueous parameters that control Hg speciation and cycling. Thus, it is reasonable that there exists no nexus in many of those efforts to wildlife habitat use and risk to Hg exposure. Still, over the past decade, only 3% of those Hg studies on estuarine fishes included managed wetlands, whereas 32% of Hg studies on estuarine waterbirds included sites in managed wetlands. This illustrates a potential disconnect in research on Hg exposure between fishes and estuarine wildlife, and that applications of non-avian bioindicators (e.g. forage fish) to infer avian Hg risk may be unsuitable due to the lack of appropriate habitat focus. This does not negate the value of monitoring fish Hg in larger estuarine environments in order to understand broader food web dynamics and risk to piscivorous fishes, but suggests that implementation of more refined designs that incorporate Hg risk to birds and other wildlife that utilize these important wetland habitats would be valuable.

The development of robust monitoring programs is important for evaluating changes in Hg cycling and bioaccumulation associated with habitat management and wetland restoration. Small fishes such as those sampled in this study represent a key conduit for Hg transport through estuarine food webs, thus they serve as useful bioindicators of Hg availability and cycling within ecosystems (Wiener et al., 2007). Importantly, the interpretation of environmental Hg risk is strongly influenced by the fish species used in monitoring programs. Mercury concentrations commonly differ among fish species within an ecosystem or waterbody (Campbell et al., 2008; Chumchal and Hambright, 2009), increasing along trophic gradients due to biomagnification (Lavoie et al., 2013). However, few studies focus on components of fish communities comprised primarily of lower-order consumers that occupy a narrower range of trophic positions, where foraging habitat might play an increasingly important role in inter-specific differences (Campbell et al., 2008; Greenfield and Jahn, 2010; Swanson et al., 2006). Even among low trophic level, invertebrate-foraging fish species, we found substantial differences in Hg concentrations. For example, THg concentrations in the two species with the highest values (Mississippi silversides and northern anchovy) were 1.6 and 1.5 times higher, respectively, than those with the next highest concentrations (topsmelt), and 2.4 and 2.2 times higher, than in the species with lowest THg concentrations (longjaw mudsucker). This has important implications for evaluating ecological risk of Hg bioaccumulation. It suggests that depending upon the species selected and the endpoint chosen for evaluating impacts, single-species monitoring could misinform ecological risk in complex systems such as estuaries unless the same fish species is sampled at

all sites and times. Thus, our findings underscore the importance of using the same fish species at all sites and time periods for studies evaluating spatial and temporal comparisons of Hg exposure. Alternatively, a multi-species design, as we have done in this study, is a very robust approach. However, an effective multi-species design requires substantially greater resource allocation to achieve spatial overlap among species across numerous sites, in order to reach the added sample size requirements needed across species to adequately generate model estimated (least-squared mean) fish THg concentrations. This multi-species design is a powerful approach that statistically accounts for the variance associated with different taxa and provides a more integrated assessment of overall exposure through the entire fish community and risk to wildlife.

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