Supporting document for

Forage fish mercury spatial patterns and predictors in San Francisco Bay

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Section I: Supplemental Text

Study hypotheses

The study design and site selection were intended to answer the four questions listed in the Introduction. Based on prior Bay studies [1-5], input from local natural resource managers, and current conceptual models regarding MeHg cycling and bioaccumulation in estuaries, we developed the following four hypotheses regarding factors that influence small fish Hg concentrations:

1. Mercury concentrations will increase with proximity to the Guadalupe River, in the Lower South Bay.
2. Concentrations will be higher in fully or partially enclosed areas, defined to include subembayments, natural or man-made channels, or estuarine creeks draining into the Bay (defined in this paper as "embayments"). These areas would tend to have low hydraulic mixing of subtidal water (i.e., locations with low water turnover rate), resulting in higher MeHg production.
3. Concentrations will be positively correlated with nearby wetland abundance.
4. Concentrations will be higher near mercury source areas, including urban or industrial watershed drainages, wastewater treatment plants (i.e., publicly operated treatment works, or POTWs), areas with historically Hg or MeHg contaminated sediments, and mine drainages.

Study parameters

Average total body length (mm) was based on field measurements of all individuals in a composite sample. Length is a widely reported correlate of fish Hg, and length correction is needed [6, 7], including for small fish evaluated in the Bay [2, 8]. Sampling was performed in three years: 2008, 2009, and 2010, which are treated as categorical variables (with 2010 as the base condition). There are also four categorical variables indicating different kinds of Hg source sites (Table S1), as well as randomly chosen sites that represent background conditions (statistically treated as the base condition).

Site selection description

Creeks draining Hg mines (N = 4) were chosen based on the priority scheme of Abu-Saba [9]. Sites were included based on connectivity to the Bay, evidence of mine waste discharging into State waters, and risk to fishery resources or other sensitive habitat areas. Three creeks were chosen based on drainage from mines meeting the inclusion criteria: Napa River (drainage from La Joya and Bella Oaks mines), American Canyon Creek (Borges Mine), and Guadalupe River (New Almaden Mining District). Due to the small number of candidate sites, all Hg mine drainage sites were sampled. To further evaluate the potential mine signal from La Joya mine, a freshwater site adjacent to and draining the mine (Dry Creek) was added and sampled in 2009.

POTW sites (N = 7 sites) were selected from among those Bay POTWs that are identified by the San Francisco Bay Regional Water Quality Control Board (Regional Board) as shallow-water discharge POTWs [10]. The selection of shallow water discharges was based on the expectation of greater potential impact of POTW discharge to the nearshore area biogeochemistry in shallow-water environments than deep-water or offshore environments. That is, shallow-water

POTW sites had a relatively high water volume ratio of discharge water to natural Bay water, potentially resulting in discharge water impacting Hg biogeochemistry. The original pool of sites was further reduced to only include sites having summer discharge because sampling occurred in the fall. The final POTW sites were City of American Canyon Wastewater Treatment Plant, Fairfield-Suisun, Hayward Pond 3B, Mountain View Sanitary District Peyton Slough, City of Palo Alto, City of Sunnyvale, and Sonoma Valley County Sanitary District Schell Creek. All seven POTW sites were sampled.

Candidate sites with relatively elevated sediment total Hg or MeHg (N = 35) were selected based on two prior sediment surveys from the Bay Protection and Toxic Cleanup Program [11] and a study funded by the Regional Board and the California Bay-Delta Authority program [3]. Both studies targeted areas known to be currently or historically industrial or otherwise suspected of having high contamination. Sites were included when sediment THg concentrations were greater than 700 ng g^{-1} or MeHg concentrations were greater than 2 ng g^{-1}. In the GRTS sample draw, 15 sites were sampled from this category.

Candidate sites draining industrial and urban watersheds (N = 21) were selected based on a combination of four attributes in the watersheds: the documented presence of historic mercury spills, density of historic industrial sites, density of railway lines, and density of car recyclers. Toxic Hg spills were identified using database queries from the CA Department of Toxic Substances Control, USEPA Superfund, and Toxic Release Inventory. Railway line density was considered as an indicator of historic industry, while auto recycler density indicated current industry and potentially an additional Hg source. All attributes were characterized using GIS layers developed from Lower South Bay to southern San Pablo Bay [12]. To increase geographical coverage, three sites were added from central San Pablo Bay and Suisun Bay (Kirker Creek, Corte Madera Creek, and Alhambra Creek). In the GRTS sample draw, 13 sites were sampled from this category.

In 2008, 12 sites adjacent to fringing wetlands were sampled to aid in evaluating the potential impact of wetland proximity on biosentinel Hg (Hypothesis 4). The wetland sites ranged in anticipated frequency of wetting and drying; wetlands with a high expected wetting and drying frequency were hypothesized to exhibit higher MeHg production and consequently higher Hg in biosentinels [13]. Results from 2008 indicated no apparent correspondence with fringing wetland abundance or type, and GIS indicated that a range of proximity to wetlands was achieved using the random samples in the GRTS design. Therefore, in 2009 and 2010 wetland site targeting was discontinued to increase sample sizes in the other strata. The final study analysis compared fish Hg based on percent wetlands at the site. The wetland sites were included in analysis of proximity to wetlands vs. small fish Hg (Question 3). To avoid confounding site categorization, the 2008 wetland sites were excluded from the comparison of embayment versus open sites (Question 2) and the comparison of random versus source sites (Question 4).

Section II: Supplemental Tables

Table S1. Parameters examined in study.

ID in Models	Description	Type	Units
HgWw	Hg concentration in composite fish sample, wet weight. Obtained from single species composite samples, $\log_{10}$ transformed for model inclusion	Numeric	$\mu g\ g^{-1}$
Total Length	Fish total body length, averaged across composite sample, centered before model inclusion	Numeric	mm
Site ID	Site Location ID. N = 99 stations; but only a subset for each species and analysis.	Categorical[a]	
Distance Guadalupe	Distance from Guadalupe River of collection site, centered before model inclusion	Numeric	km
Y2008	Indicates samples collected in 2008	Categorical	1=Yes
Y2009	Indicates samples collected in 2009. Default (i.e., baseline) year is 2010	Categorical	1=Yes
WetlandAbund	Surrounding wetland area, based on a 500 m buffer	Numeric	%
Embayment	Indicates sample collected from enclosed subembayment, rather than open Bay shoreline (Figure S1)	Categorical	1=Yes
SourcePOTW	Publicly operated treatment works (POTW) at station. This is influenced by wastewater treatment plant discharge. This and remaining source station categories are compared to randomly selected stations.	Categorical	1=Yes
SourceContamSed	Historic Hg contaminated sediment at station	Categorical	1=Yes
SourceWatershed	Station adjacent to industrial or urban watershed hypothesized to be high in Hg	Categorical	1=Yes
SourceMine	Station adjacent to watershed containing historic Hg mine	Categorical	1=Yes

a. All categorical variables are nominal, rather than ordinal

Model structures

Table S2. Silverside final model for embayment effects (N = 116). For this and remaining tables, likelihood ratio test is performed, and the difference in AIC (ΔAIC) is determined, between a model containing all parameters listed and a model with the current parameter removed. NA: since embayment was not significant, it is not included in the final model.

Fixed effects	**Value**	**SE**	**Likelihood ratio**	**p-value**	**ΔAIC**
(Intercept)	-1.20219	0.01802			
TLengthCen	0.00516	0.00190	39.5	<0.0001	37.5
DistGuadCen	-0.00345	0.00041	7.00	0.0082	5.0
Embayment	NA	NA	2.77	0.096 (NS)	0.8
Random effects:					
Formula: ~1 + TLengthCen | StationShort					

Table S3. Topsmelt final model for embayment effects (N = 133)

Fixed effects	**Value**	**SE**	**Likelihood ratio**	**p-value**	**ΔAIC**
(Intercept)	-1.47553	0.02565			
TLengthCen	0.00238	0.00082	8.50	0.0035	6.5
DistGuadCen	-0.00229	0.00068	11.1	0.0008	9.1
Y2009	0.06071	0.03098	4.18	0.041	2.2
Embayment	0.09145	0.03752	6.30	0.012	4.3
DistGuadCen:Embayment	0.00298	0.00119	17.1	0.0007	11.1
TLengthCen:Embayment	0.00308	0.00128	40.8	<0.0001	34.8
Random effects:					
Formula: ~1 | StationShort					

Table S4. Silverside final model for wetland effects (N = 278). NA = wetland was not significant, and was therefore not included in the final model.

Fixed effects	**Value**	**SE**	**Likelihood ratio**	**p-value**	**ΔAIC**
(Intercept)	-1.07663	0.02652			
TLengthCen	0.00481	0.00103	18.7	<0.0001	16.7
Y2008	-0.10544	0.02671	14.6	<0.0001	12.6
Y2009	-0.03914	0.03234	1.59	0.21	-0.4
DistGuadCen	-0.00405	0.00046	46.9	<0.0001	44.9
Y2009:DistGuadCen	0.00169	0.00065	5.96	0.015	4.0
WetlandAbund	NA	NA	0.27	0.61 (NS)	-1.7
Random effects:					
Formula: ~1 + TLengthCen | StationShort					

Table S5. Topsmelt final model for wetland effects (N = 269). NA = wetland was not significant, and was therefore not included in the final model.

Fixed effects	Value	SE	Likelihood ratio	p-value	ΔAIC
(Intercept)	-1.39659	0.01656			
TLengthCen	0.00379	0.00051	38.7	<0.0001	36.7
DistGuadCen	-0.00229	0.00049	20.9	<0.0001	18.9
WetlandAbund	NA	NA	3.08	0.079 (NS)	1.1
Random effects:					
Formula: ~1 + TLengthCen | StationShort					

Table S6. Silverside final model for site effects (N = 237)

Fixed effects	Value	SE	Likelihood ratio	p-value	ΔAIC
(Intercept)	-1.10565	0.02760			
TLengthCen	0.00463	0.00122	12.7	0.0004	10.7
Y2008	-0.06108	0.03153	3.44	0.064	1.4
Y2009	-0.00657	0.03191	0.04	0.84	-2.0
DistGuadCen	-0.00414	0.00048	52.7	<0.0001	50.7
SourcePOTW	-0.09216	0.06358	2.15	0.14	0.2
Y2008:SourcePOTW	-0.19625	0.06058	10.4	0.0012	8.4
Y2009:DistGuadCen	0.00189	0.00063	60.8	<0.0001	54.8
Random effects:					
Formula: ~1 + TLengthCen | StationShort					

Table S7. Topsmelt final model for site effects (N = 231)

Fixed effects	Value	SE	Likelihood ratio	p-value	ΔAIC
(Intercept)	-1.43702	0.01879			
TLengthCen	0.00332	0.00047	45.4	<0.0001	43.4
Y2008	0.03358	0.02967	1.31	0.25	-0.7
DistGuadCen	-0.00228	0.00052	17.8	<0.0001	15.8
SourceContamSed	0.07831	0.03727	4.58	0.032	2.6
Y2008:DistGuadCen	0.00198	0.00087	5.24	0.022	3.2
Random effects:					
Formula: ~1 | StationShort					

Section III: Supplemental Figures

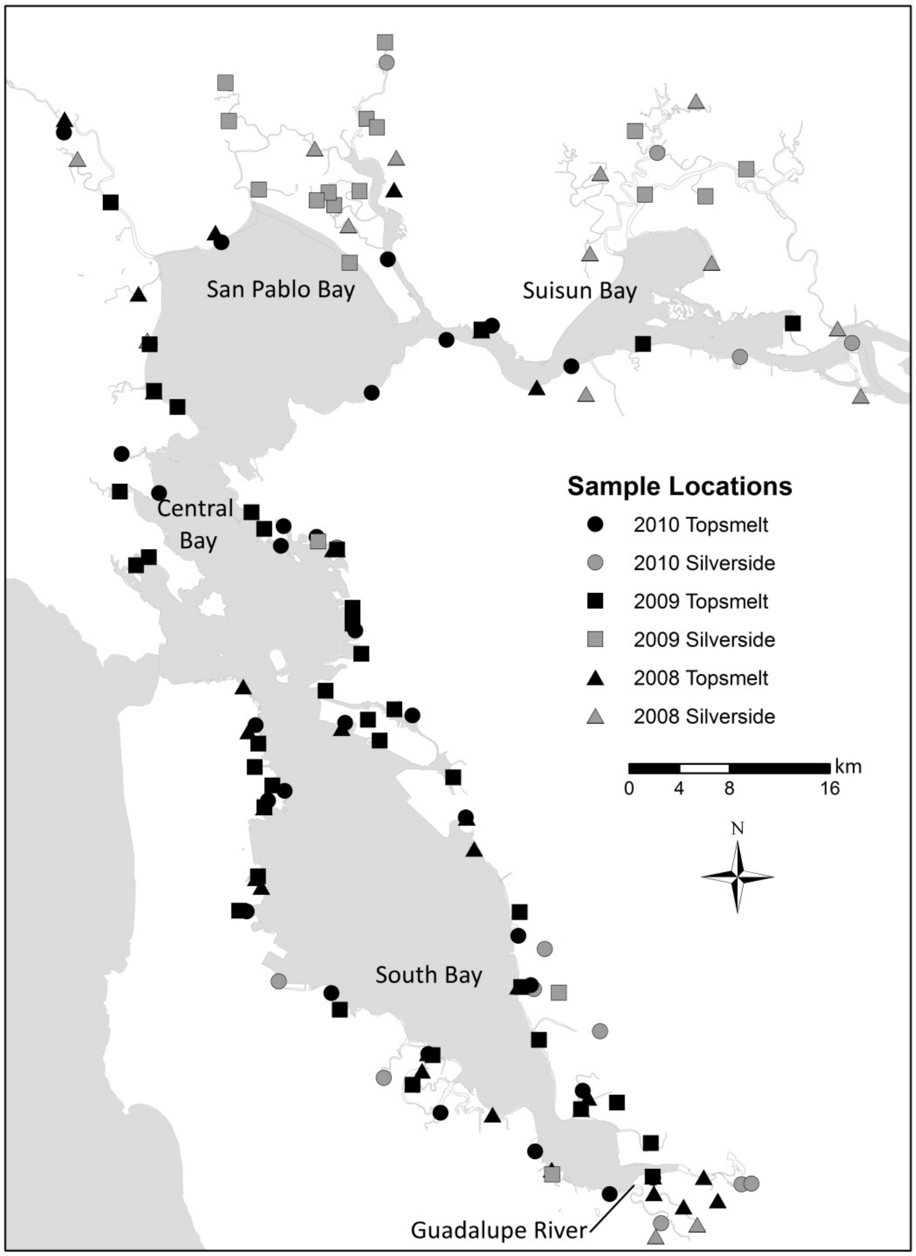

Figure S1. Study sample locations.

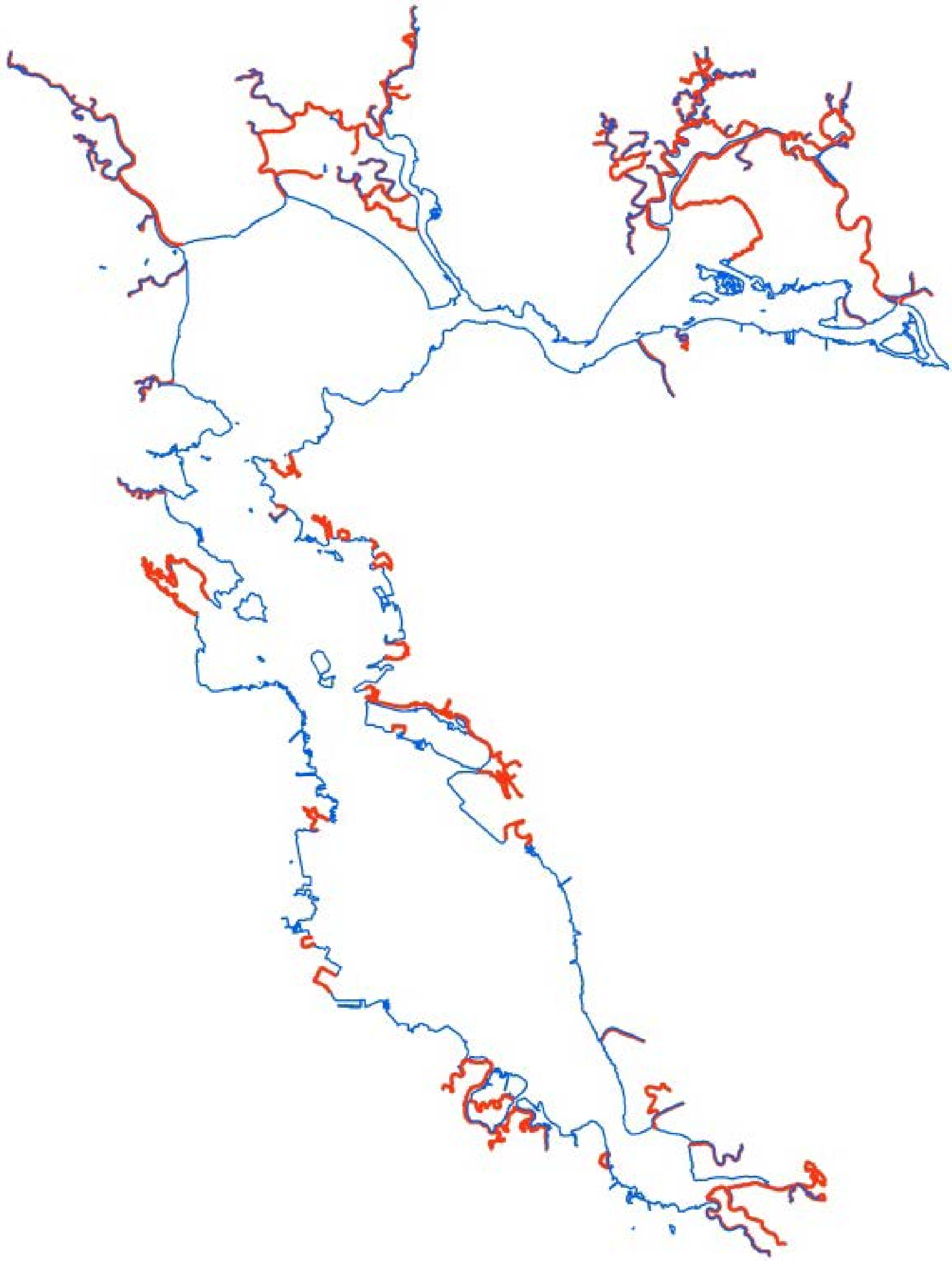

Figure S2. Final stratification of the Bay shoreline for the random sample draw. The red line represents the embayment stratum. Blue areas not parallel to a red line represent the open stratum.

Section IV: Description of Supplemental Data File

All forage fish Hg data collected in San Francisco Bay from 2008 to 2011 are provided as a comma separated (CSV) file (filename: SFForageFishHg.csv). Comprising 1260 composite samples, these data are more extensive than the samples analyzed for the present study. They include Bay forage fish Hg data analyzed for temporal patterns and published elsewhere [8], as well as samples collected and analyzed for Hg but falling outside the scope of both studies. Data on fish Hg and other contaminants are also presently available via the California Environmental Data Exchange Network (www.ceden.us) [14]

This file contains 30 data fields, including Hg concentrations, site and sample descriptive information, and ancillary information. Each row corresponds to a separate composite fish sample analyzed for Hg:

Sample ID Laboratory identifier indicating the composite fish sample analyzed.

Site ID Unique identifier for each collection site

Collection Code Unique identifier for each collection event (i.e., each site and sample date combination).

Site Description Descriptive name for each collection site

Site Type Indicates the site category each sample was included in. Table S8 (below) lists all possible categories

Date Collection date

Latitude In degrees and decimal degrees

Longitude In degrees and decimal degrees

Species Common name of the sampled species. Table S9 lists corresponding scientific names, as well as number of samples collected per species

HgDw Mercury concentration ($\mu g\ g^{-1}$ dry weight).

Moisture Tissue proportion moisture

HgWw Mercury concentration ($\mu g\ g^{-1}$ wet weight).

N Number of individuals in the composite sample

Total Length Average total length of individuals in the composite sample (mm)

Distance Guadalupe Distance from the Guadalupe River, following along the deep Bay channel (further described in Methods)

WetlandAbund Percent surrounding wetland area within a 500 m buffer of collection location

The remaining variables are binary categorical variables (1 = yes; 0 = no), indicating which study model evaluation (Table 1) or sampling strata a sample was included in.

Question1SpatialTrend Sample included in study Question 1: What are the spatial trends in forage fish Hg?

Question2Embayment Sample included in study Question 2: Are Hg concentrations elevated in embayments relative to open water sites?

Question3Wetlands Sample included in study Question 3: Does extent of fringing wetland habitat correlate with Hg concentrations?

Question4SiteType Sample included in study Question 4: Are Hg concentrations elevated at potential source sites, relative to randomly selected sites?

LongTerm Sample included in prior analysis of long term Hg trends [8]

Source Sample is in one of the four source site categories

Random Sample is in the random strata (either embayment or open) sampled from entire Bay shoreline (Table S2)

SourcePOTW Wastewater treatment plant source site sample

SourceMine Historic Hg mine source site sample

SourceContamSed Hg or MeHg contaminated sediment source site sample

SourceWatershed Urban or industrialized watershed source site sample

Embayment Site falls within the embayment stratum (1 = embayment; 0 = open; see also Figure S2)

OffSeason Sample collected outside the Fall season sampling window required for study inclusion. The study sampling window was August 27 to November 30

Wetlands Sample targeted to increase wetland coverage

Table S8. Site type descriptions.

Site type[a]	Description
Embayment	Random Bay shoreline site; embayment stratum (Figure S2)
Open Water	Random Bay shoreline site; open water stratum (Figure S2)
Source Contaminated Sediment	Source site with elevated sediment THg or MeHg
Source Wastewater Treatment Plant (POTW)	Source site draining wastewater treatment plant
Source Industrial Watershed	Source site draining urban and industrial watershed
Source Hg Mine	Source site draining historic Hg mine watershed
Wetlands	Site adjacent to nearshore wetland, targeted to increase wetland coverage, sampled 2008
Long Term Monitoring	Site monitored for annual variation from 2005 to 2010, analyzed elsewhere [8]
Seasonal/Long Term Monitoring	Site monitored for seasonal and annual variation, analyzed elsewhere [8]
Not in Hg Sampling Design	Site targeted for PCB study [15], analyzed for Hg but not included in Hg study analyses

a. Some sites are in more than one category

Table S9. Common and scientific names, and total number composite samples collected for each species.

Common Name	Scientific Name	Number samples
American shad	*Alosa sapidissima*	7
Arrow goby	*Clevelandia ios*	105
California roach	*Hesperoleucus symmetricus*	8
Diamond turbot	*Hypsopsetta guttulata*	6
Golden shiner	*Notemigonus crysoleucas*	3
Longjaw mudsucker	*Gillichthys mirabilis*	1
Mississippi silverside	*Menidia audens*	357
Northern anchovy	*Engraulis mordax*	24
Prickly sculpin	*Cottus asper*	8
Rainwater killifish	*Lucania parva*	27
Shimofuri goby	*Tridentiger bifasciatus*	3
Shiner perch	*Cymatogaster aggregata*	1
Staghorn sculpin	*Leptocottus armatus*	33
Striped Bass	*Morone saxatilis*	24
Threadfin shad	*Dorosoma petenense*	2
Three-spined stickleback	*Gasterosteus aculeatus*	24
Topsmelt	*Atherinops affinis*	610
Western mosquitofish	*Gambusia affinis*	7
Yellowfin goby	*Acanthogobius flavimanus*	10

Section V: References

1. Conaway CH, Squire S, Mason RP, Flegal AR. 2003. Mercury speciation in the San Francisco Bay estuary. *Mar Chem* 80:199-225.
2. Greenfield BK, Jahn A. 2010. Mercury in San Francisco Bay forage fish. *Environ Pollut* 158:2716-2724.
3. Heim WA, Coale KH, Stephenson M, Choe K-Y, Gill GA, Foe C. 2007. Spatial and habitat-based variations in total and methyl mercury concentrations in surficial sediments in the San Francisco Bay-Delta. *Environ Sci Technol* 41:3501-3507.
4. Marvin-DiPasquale MC, Agee JL, Bouse RM, Jaffe BE. 2003. Microbial cycling of mercury in contaminated pelagic and wetland sediments of San Pablo Bay, California. *Environ Geol* 43:260-267.
5. Conaway CH, Black FJ, Grieb TM, Roy S, Flegal AR. 2008. Mercury in the San Francisco Estuary: A review. *Rev Environ Contam Toxicol* 194:29-54.
6. Tremblay G, Legendre P, Doyon J-F, Verdon R, Schetagne R. 1998. The use of polynomial regression analysis with indicator variables for interpretation of mercury in fish data. *Biogeochemistry* 40:189-201.
7. Wiener JG, Bodaly RA, Brown SS, Lucotte M, Newman MC, Porcella DB, Reash RJ, Swain EB. 2007. Monitoring and evaluating trends in methylmercury accumulation in aquatic biota. In Harris R, Krabbenhoft DP, Mason R, Murray MW, Reash R, Saltman T, eds, *Ecosystem Responses to Mercury Contamination: Indicators of Change*. CRC Press, Pensacola, FL, pp 87-122.
8. Greenfield BK, Melwani AR, Allen RM, Slotton DG, Ayers SM, Harrold KH, Ridolfi K, Jahn A, Grenier JL, Sandheinrich MB. 2013. Seasonal and annual trends in forage fish mercury concentrations, San Francisco Bay. *Sci Total Environ* 444:591-601.
9. Abu-Saba K. 2003. Managing inactive mercury mine sites in the San Francisco Bay Region. Applied Marine Sciences, Inc. http://www.bacwa.org/LinkClick.aspx?fileticket=RUiczIW5flU%3D&tabid=126&mid=572
10. California Regional Water Quality Control Board San Francisco Bay Region. 2011. San Francisco Bay Basin (Region 2) Water Quality Control Plan (Basin Plan). Web Document. Accessed October 11, 2012. http://www.waterboards.ca.gov/sanfranciscobay/basin_planning.shtml. http://www.waterboards.ca.gov/sanfranciscobay/basin_planning.shtml
11. Hunt JW, Anderson BS, Phillips BM, Newman J, Tjeerdema RS, Taberski K, Wilson CJ, Stephenson M, Puckett HM, Fairey R, Oakden J. 1998. Sediment quality and biological effects in San Francisco Bay: Bay Protection and Toxic Cleanup Program Final Technical Report. San Francisco Bay Regional Water Quality Control Board, Oakland, CA. http://www.swrcb.ca.gov/water_issues/programs/bptcp/docs/reg2report.pdf
12. SFEI. 2010. A BMP tool box for reducing polychlorinated biphenyls (PCBs) and mercury (Hg) in municipal stormwater. San Francisco Estuary Institute, Oakland, CA. http://www.sfei.org/documents/3813
13. Snodgrass JW, Jagoe CH, Bryan AL, Brant HA, Burger J. 2000. Effects of trophic status and wetland morphology, hydroperiod, and water chemistry on mercury concentrations in fish. *Can J Fish Aquat Sci* 57:171-180.
14. CEDEN. 2012. California Environmental Data Exchange Network General Information. Fact Sheet. http://www.ceden.org/docs/ceden_intro_010912.pdf

15. Greenfield BK, Allen RM. 2013. Polychlorinated biphenyl spatial patterns in San Francisco Bay forage fish. *Chemosphere* 90:1693-1703.