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Hg concentrations in fish from coastal waters of California and Western North America

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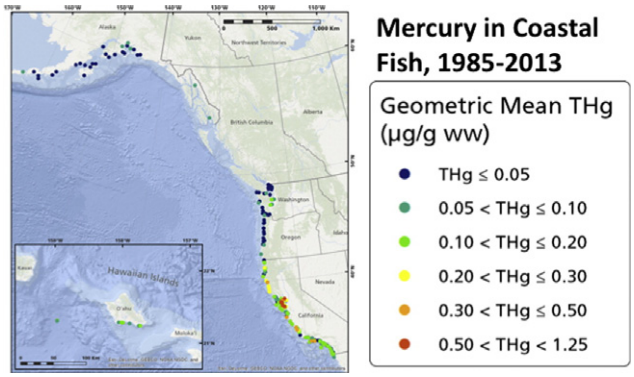
HIGHLIGHTS

- Mercury in Western US coastal fish is an exposure risk to fish consumers.
- High concentrations were observed in long-lived predators (e.g., rockfish, sharks).
- Data on mercury in coastal fish in publicly accessible databases are limited.
- Limited data on long-term trends indicate no change over the past 40 years.
- Systematic and consistent monitoring and data management would be of great value.

GRAPHICAL ABSTRACT



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ABSTRACT

The State of California conducted an extensive and systematic survey of mercury (Hg) in fish from the California coast in 2009 and 2010. The California survey sampled 3483 fish representing 46 species at 68 locations, and demonstrated that methylHg in fish presents a widespread exposure risk to fish consumers. Most of the locations sampled (37 of 68) had a species with an average concentration above 0.3 µg/g wet weight (ww), and 10 locations an average above 1.0 µg/g ww. The recent and robust dataset from California provided a basis for a broader examination of spatial and temporal patterns in fish Hg in coastal waters of Western North America. There is a striking lack of data in publicly accessible databases on Hg and other contaminants in coastal fish. An assessment of the raw data from these databases suggested the presence of relatively high concentrations along the California coast and in Puget Sound, and relatively low concentrations along the coasts of Alaska and Oregon, and the outer coast of Washington. The dataset suggests that Hg concentrations of public health concern can be observed at any location on the coast of Western North America where long-lived predator species are sampled. Output from a linear mixed-effects model resembled the spatial pattern observed for the raw data and suggested, based on the limited dataset, a lack of trend in fish Hg over the nearly 30-year period covered by the dataset. Expanded

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and continued monitoring, accompanied by rigorous data management procedures, would be of great value in characterizing methylHg exposure, and tracking changes in contamination of coastal fish in response to possible increases in atmospheric Hg emissions in Asia, climate change, and terrestrial Hg control efforts in coastal watersheds.

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1. Introduction

Recreational and commercial fishing are a vibrant part of the economy for California and other Western North American coastal states. In 2009, recreational anglers in California took $4.7 * 10^6$ fishing trips, including $3.6 * 10^6$ trips by shore-based anglers, $0.7 * 10^6$ trips in private boats, and $0.4 * 10^6$ trips by for-hire boats (NMFS, 2009). Together with sales of durable equipment, these trips generated 13,500 full and part-time jobs, and over $\$2 * 10^9$ in sales (NMFS, 2009). The commercial seafood industry in California generated 120,000 jobs and over $\$20 * 10^9$ in sales (NMFS, 2009). The species that were most often caught by recreational anglers in the Pacific region (California, Oregon, and Washington) were rockfishes and scorpionfishes ($2.7 * 10^6$ fish), mackerel ($2 * 10^6$ fish), barracuda, bass, and bonito ($1.6 * 10^6$ fish), and surfperches ($1.5 * 10^6$ fish). Most of the rockfishes and scorpionfishes in the Pacific region were caught in California.

Mercury (Hg) exists in the environment in many forms. MethylHg (MeHg) is the form that poses risks to the health of humans (Karagas et al., 2012) and wildlife (Scheuhammer et al., 2012) due to its toxicity and strong tendency to bioaccumulate in food webs. Although MeHg contamination of fish is well established as a hazard to the health of human and wildlife fish consumers across North America (Evers and Clair, 2005; Stahl et al., 2009; Evers et al., 2011; USEPA, 2013) and in the Pacific Ocean (Sunderland, 2007), little has been published on MeHg concentrations in fish (typically measured as "total Hg" because nearly all of the Hg in fish is in the form of MeHg) from the Pacific coast of Western North America. One notable exception is articles from the Regional Monitoring Program for Water Quality in San Francisco Bay (Davis et al., 2002; Greenfield et al., 2005; Greenfield et al., 2013). Other significant monitoring has been conducted, but published in grey literature technical reports (e.g., data from the Puget Sound Ambient Monitoring Program (West et al., 2001)). The limited information available from these studies indicates that Hg often reaches high concentrations, occasionally over the US Food and Drug Administration (USFDA) action level for commercial fish of 1 μg/g wet weight (ww) (USFDA, 2007), and very commonly above the US Environmental Protection Agency (USEPA) water quality criterion of 0.3 μg/g ww (USEPA, 2010).

In spite of the importance of coastal fisheries to the economy, and as a source of food for humans and wildlife, and studies indicating the potential for Hg to reach problematic concentrations in coastal fish, very little monitoring has been conducted to assess this threat to human and wildlife health. To address this information gap for California, the California State Water Resources Control Board's Surface Water Ambient Monitoring Program (SWAMP) conducted an extensive and systematic survey of Hg and other contaminants in fish from the California coast in 2009 and 2010. The objectives of this paper are to: 1) document the methods and results of the recent SWAMP survey, and 2) to assess the results of the SWAMP survey in the context of a compilation and synthesis of available data from other studies from the coast of Western North America from 1985 to the present.

2. Methods

2.1. The 2009–2010 California survey

California has over 840 miles of coastline that span diverse habitats and fish populations, and include dense human population centers with a multitude of popular fishing locations. The approach employed in this evaluation of potential human exposure to MeHg was to divide the California coast into 68 spatial units called "zones" (map provided in Fig. 1a–c, Supplemental material). The sampling focused on near-shore areas, including bays and estuaries, in waters not exceeding 200 m in depth, and mostly <60 m deep. These are the coastal waters where most of the sport fishing occurs. Within each zone, sample collection was directed toward the most popular fishing locations. Popular fishing areas were identified through published sources (e.g., Jones, 2004) and consultation with agency staff. The number of locations visited in each zone varied depending on fishing success.

Fish were collected from April through November in 2009 and 2010 using a variety of methods: hook and line; otter trawl, castnet, spear, gillnet, and beach seine. Details on methods, catch, and precise locations are provided in cruise reports for each sampling year (Supplemental material). Five species were sampled in each zone. Fish species were selected based on the following criteria, in order of importance: popularity for catch and consumption (PSMFC, 2009), sensitivity as indicators of contamination problems (accumulating relatively high concentrations of contaminants based on data from prior studies - e.g., Gassel et al. (2002); Greenfield et al. (2005)), wide distribution based on life history information from FishBase (www.fishbase.org), species representing safer choices for consumption (accumulation of relatively low concentrations of contaminants based on data from prior studies), representation of different exposure pathways (benthic versus pelagic) based on life history information from FishBase, and continuity with past sampling. A list of primary and secondary target species for each region (north, central, and south) and for two strata (outer coast, bays and harbors) was developed (Table 4 in the Sampling and Analysis Plan - Supplemental Information).

In general, composite samples consisting of five fish for each species were collected, following guidance from USEPA (2000). Size ranges for each species were based on a combination of sizes prevalent in past sampling efforts by the Regional Monitoring Program for Water Quality in San Francisco Bay (Greenfield et al., 2005) and the Coastal Fish Contamination Program (Gassel et al., 2002) and the 75% rule recommended by USEPA (2000). For each species, size ranges and compositing plans were also established prior to sampling (Table 6 in the Sampling and Analysis Plan - Supplemental Information). For selected species with relatively wide distributions on the coast and a tendency to accumulate elevated Hg concentrations (e.g., leopard shark, kelp bass, barred sand bass, olive rockfish, gopher rockfish, and black rockfish), individual fish were analyzed to support a more rigorous statistical assessment of spatial patterns by an analysis of covariance that adjusted observed concentrations to account for the correlation of Hg with length, following the approach described by Davis et al. (2008). In general, fish were processed without skin, and only the fillet muscle tissue was used for analysis (Table 1 in the Supplemental Information). Some species (e.g., shiner surfperch) were too small to be filleted and were processed whole, but with head, tail, and viscera removed.

In most cases, nearly all (>95%) of the Hg present in fish fillets and in whole fish is MeHg (Wiener et al., 2007; Greenfield and Jahn, 2010). Consequently, monitoring programs usually analyze total Hg (THg) as a proxy for MeHg, as was done in this study. USEPA (2000) recommends this approach, and the conservative assumption be made that all Hg is present as MeHg to be most protective of human health. Hg was analyzed according to EPA 7473, "Hg in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry" using a Direct Hg Analyzer. Quality assurance analyses for SWAMP Data Quality

Objectives (DQOs) (precision, accuracy, recovery, completeness, and sensitivity) were performed for each batch as required by the SWAMP BOG QAPP (Bonnema, 2009). All Hg analyses met measurement quality objectives for percent recovery in matrix spikes and matrix spike duplicates (75–125%), percent recovery in certified reference materials (DORM 3, 75–125%), and relative percent difference in matrix spike duplicates, lab duplicates, and field duplicates (25%). Hg results were reportable for 100% of the samples analyzed.

A complete set of documents for this study, including a technical report summarizing the study and its results (Davis et al., 2012a) and the dataset, are available on the web (http://www.waterboards.ca.gov/water_issues/programs/swamp/coast_study.shtml). Detailed information on the attributes of each sample is also available through the California Environmental Data Exchange Network (http://www.ceden.org/).

2.2. Western North America data synthesis

An extensive search and compilation was conducted of monitoring and research-based data on Hg concentrations in fishes from publicly accessible federal and state databases (Table 1 in Supplemental material). Data were combined into a single database with reference to the

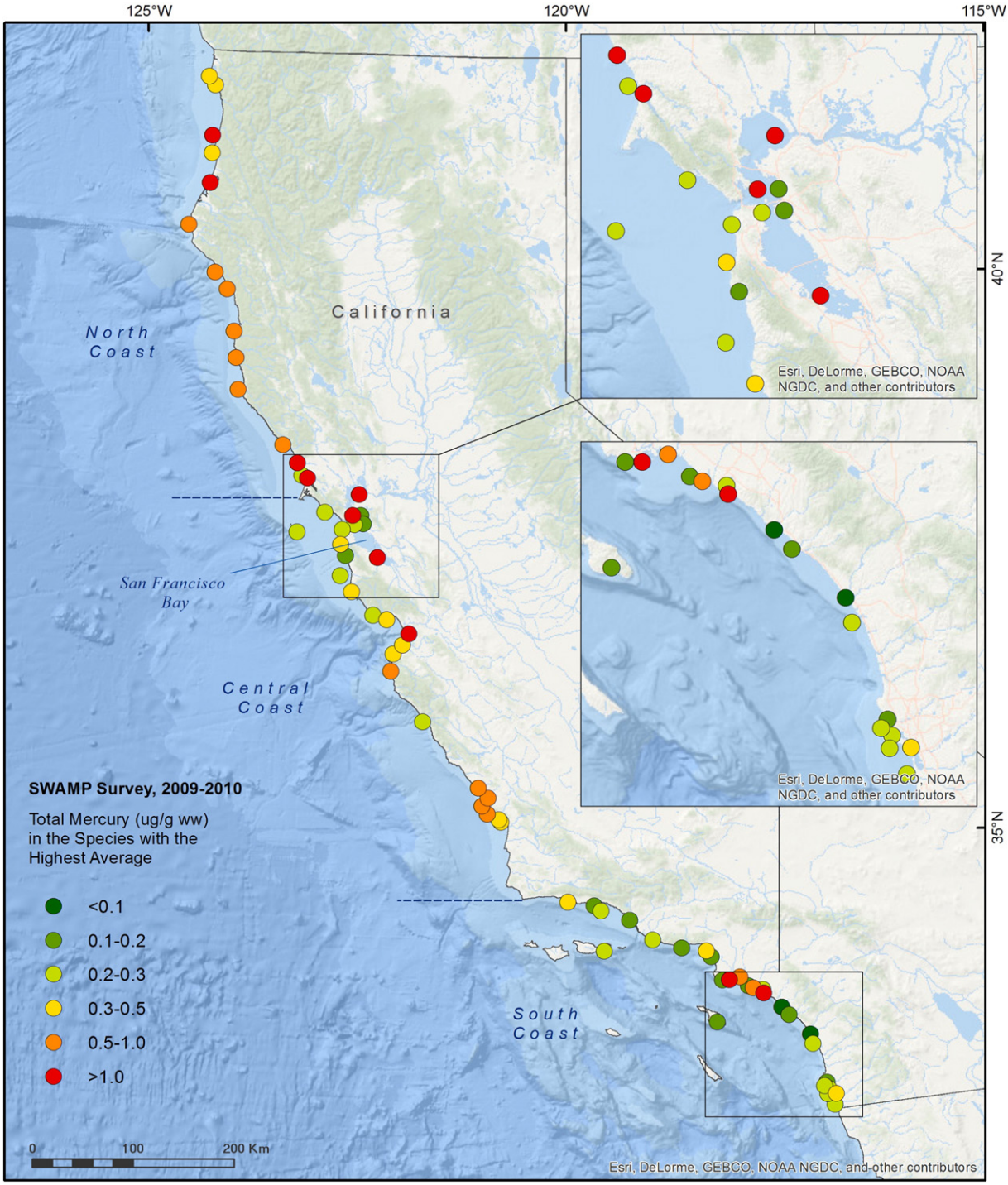

Fig. 1. Spatial patterns in total Hg concentrations (μg/g ww) among locations sampled in the California Coast Survey, 2009–2010. Each point represents the highest average Hg concentration among the species sampled at each location.

original data sources. Many of the data records that were accessible were incomplete. Parameters that were often missing included length and/or weight, sample type (composite or individual fish), tissue analyzed, and numbers of fish in a composite.

In some cases there were duplicate fish data in state and federal databases. Therefore, both automated and manual screenings were performed to identify duplicate data entry based on combinations of THg concentrations, fish lengths and weights, and sampling locations and years. After several initial iterations to identify and remove duplicate entries, THg concentrations were standardized by tissue type and moisture content, and all length measurements were also standardized. Other monitoring data exist along the Pacific Coast of Western North America, but a lack of availability in state and federal databases precluded their incorporation into this analysis.

Total Hg concentrations in the original dataset were reported as skinless boneless fillet (56% of data rows), whole body (41% of data rows), or skin-on fillet (3% of data rows). All whole body concentrations were converted to skinless boneless fillet equivalents by dividing by 0.74 (41% of records converted), the average ratio of whole body to muscle concentration from studies where both tissue types were measured on the same individuals (Goldstein et al., 1996; Bevelhimer et al., 1997; Boalt et al., 2014). Skin-on fillet concentrations were not converted because the difference is typically small (<10%; Dellinger et al., 1995; Depew et al., 2013; Zhang et al., 2013), and very few comparative studies exist. Additionally, all THg concentrations were adjusted to a wet weight basis. Conversions from dry weight concentrations to wet weight concentrations used the original moisture content data where available. For the remaining data the mean tissue-specific moisture content from the fish for which moisture content measurements were available were used to convert dry weight THg concentrations to wet weight THg concentrations.

All fish length measurements were standardized to fork length. Species-specific fork length to total length or standard length ratios reported in FishBase (Froese and Pauly, 2003) were used to convert all fish length measurements to fork length. Ratios from closely related species that had similar morphology were used for species without conversions in FishBase. For a small number of fish the type of length measurement was unclear in the original dataset and could not be inferred from other data in that source. In these cases the original lengths were treated as fork length since it is intermediate between standard length and total length, thus minimizing any length assignment errors.

Geographic coordinate information associated with each data record was entered into a geographic information system (ArcGIS v10) in order to validate and standardize site information.

The resulting dataset included a total of 6539 individual records for fish Hg concentrations comprised of 109 different species sampled between 1985 and 2014 from a total of 322 unique sites. Table 2 in the Supplemental material summarizes sample counts and sizes for each species included in the database.

2.2.1. Statistical methods

A tiered statistical approach was used to evaluate the spatial and temporal variability in fish THg concentrations. The first tier of analysis was descriptive in nature, in order to illustrate the variation in fish THg concentrations in the raw data, without accounting for species or temporal effects. For this analysis site-specific geometric mean THg concentrations were calculated for each of the 322 unique sites. These geometric mean concentrations for each site included all species within a site, not size-adjusted for any of the THg concentrations.

In a second tier analysis, linear mixed-effects models were used to evaluate differences in fish THg concentrations among 100-by-100 km cells in a grid covering the coast while accounting for species, site, and

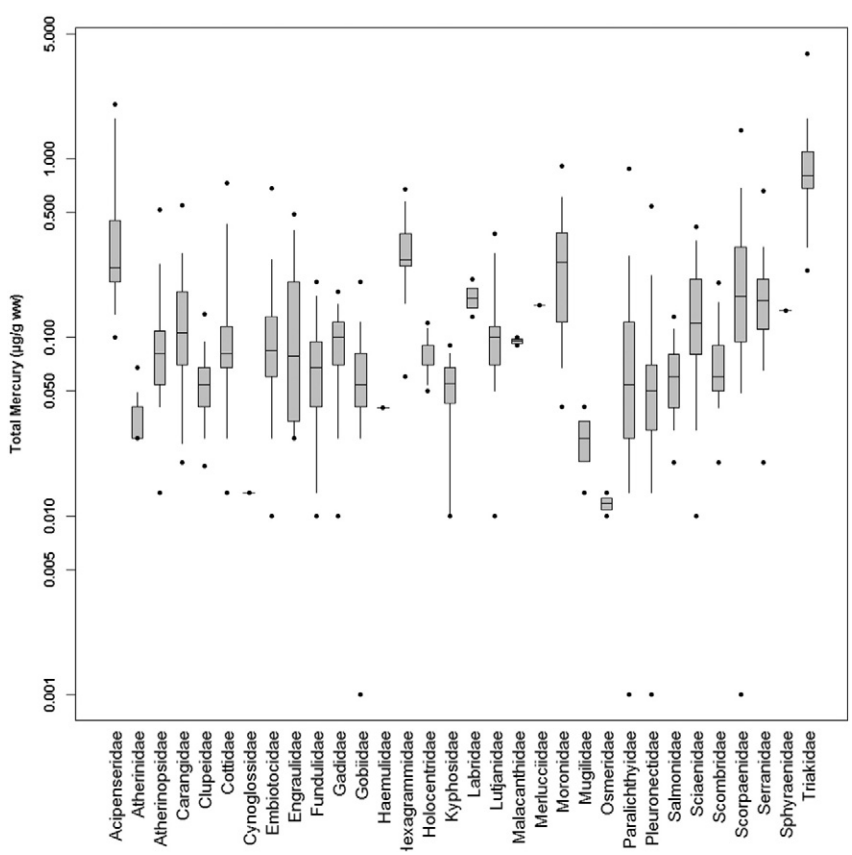

Fig. 2. Distribution of the raw data by family. Solid black line in the box represents the median. Box ends represent the 25th and 75th percentiles. The whiskers extend to the 5th and 95th percentiles. Points represent maxima and minima.

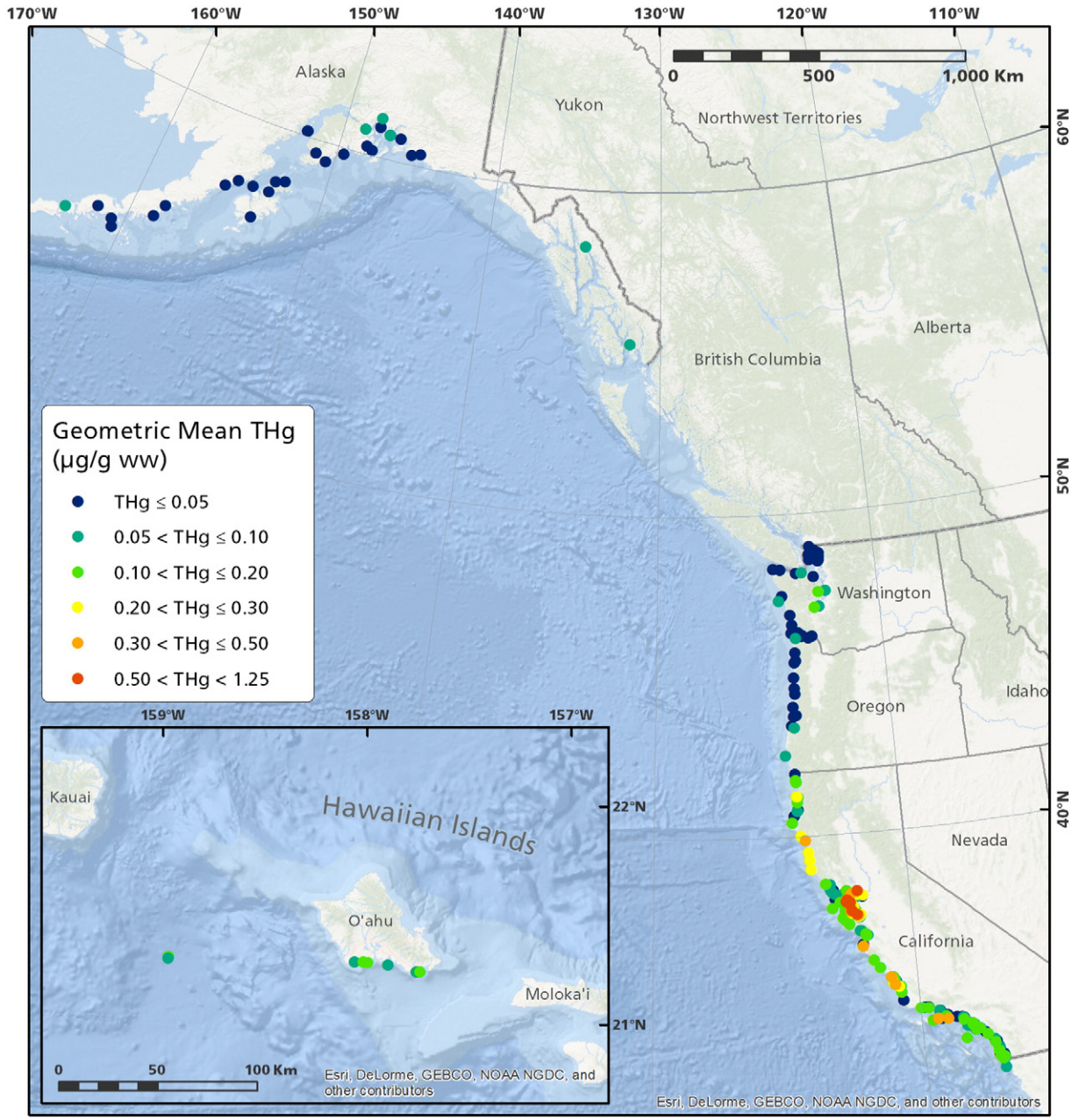

Fig. 3. Site-specific geometric mean total Hg concentrations (µg/g ww) in tissue of fishes in Western North America coastal waters. Geometric means represent raw data and do not control or account for effects of species, size, or year.

year. To minimize spatial distortion and maintain consistency of the grid-sizes over such a large region, an oblique Mercator projection was implemented with the line of tangency running from roughly Anchorage, AK to San Diego, CA. This is in contrast, for example, to the standard Mercator projection, with a line of tangency at the equator and steadily increasing in spatial distortion as a location deviates from that line towards the poles. With the standard Mercator projection, cell areas in Alaska were approximately 25% of the cell areas in Southern California. For Hawaii, a separate grid was created using Universal Transverse Mercator (UTM) Zone 4.

The statistical model was constructed to estimate spatial and temporal variation in fish THg concentrations for this highly heterogeneous dataset. Log_e-transformed THg concentration in wet weight was the dependent variable; species, site, and year were included as random effects; and tissue type and grid cell were included as fixed effects. This approach allowed for a comparison of the relative availability of THg in the coastal fish community even when there were different species assemblages across the geographical extent of the study. A second model was similar to the first model but also included trophic position as a fixed effect; this second model yielded nearly identical results to the simpler model. Unless otherwise specified, all THg concentrations were $\log_e$-transformed prior to analysis to meet assumptions of heteroscedasticity and normality of residuals. Model estimates were then back-transformed to linear space and standard errors were estimated using the Delta method (Seber, 1982). These analyses were performed using JMP version 11.2 (SAS Institute).

3. Results and discussion

3.1. Patterns in indicators of elevated risk to humans based on the recent data from California

The 2009–2010 California sport fish survey demonstrated that MeHg in fish represents a largely unrecognized and widespread exposure risk to

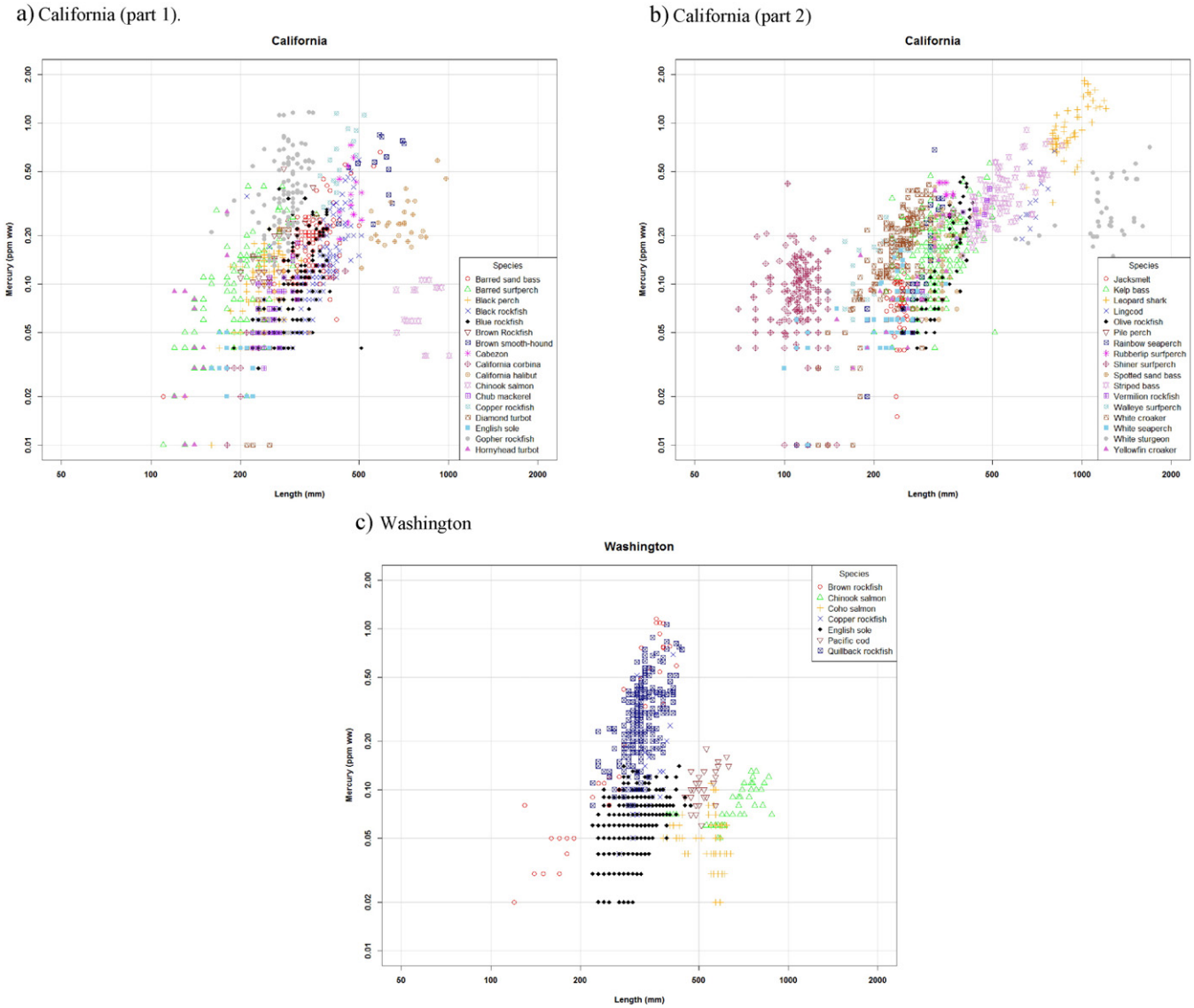

Fig. 4. Scatterplots of Hg versus length for species with at least 10 samples per state. Muscle data only. Data are for individual fish. a) California (part 1). b) California (part 2). c) Washington.

fish consumers. In this two-year study, 3483 fish representing 46 species were collected from 68 locations on the California coast. Most of the locations sampled (37 of 68, or 54%) had a species with an average concentration above the 0.3 μg/g ww USEPA water quality criterion. Many locations (24 of 68, or 35%) had a species with an average concentration above 0.5 μg/g ww, a range in which the State of California considers a recommendation of no consumption for children and women of childbearing age (Klasing and Brodberg, 2008). Several locations (10 of 68, or 15%) had a species with an average concentration above the 1.0 μg/g ww FDA action level.

Regional differences in the occurrence of species with high average concentrations were observed, with a higher prevalence in waters of the north and central coast of California relative to the south coast of California (Fig. 1, Tables 3–5 in the Supplemental material). These regional patterns were partially driven by regional variation in the distribution of species. The more common occurrence of species that tend to accumulate high MeHg concentrations, based on past studies and life history characteristics (Table 6 in the Supplemental material), including high trophic position (TP) and relatively older age (copper rockfish - TP = 4.1 and estimated age at collection 9–20 years; gopher rockfish - TP = 3.6 and age = 7 years; China rockfish - TP = 3.8 and age = 11 years; and leopard shark - TP = 3.7 and age = 16 years), was a primary reason for the relatively high concentrations observed in the north and central coast regions. The assemblage of species in the south coast region is distinctly different from the north and central coasts (Tables 3–5 in the Supplemental material). The species most commonly sampled at locations on the outer North and Central coasts were the rockfish species mentioned above, cabezon, kelp greenling, lingcod, rainbow surfperch, and barred surfperch. In contrast, the species most commonly sampled on the outer south coast were kelp bass (TP = 3.9, age = 5–7 years), barred sand bass (TP = 3.5, age = 7 years), and chub mackerel (TP = 3.1, age = 1 year). The kelp bass collected for the study had a relatively high trophic position and age that were comparable to gopher rockfish, but the kelp bass had lower concentrations at several locations where these two species were sampled together (data not shown). Brown smooth-hound sharks, with a moderately high TP (3.6) and high age at collection (over 15 years), had high concentrations in the south coast bays and harbors where they were collected (Table 5 in the Supplemental material). Gopher rockfish were collected at several south coast locations and had concentrations that were similar to those observed on the north and central coasts (data not shown). This survey targeted and collected the species

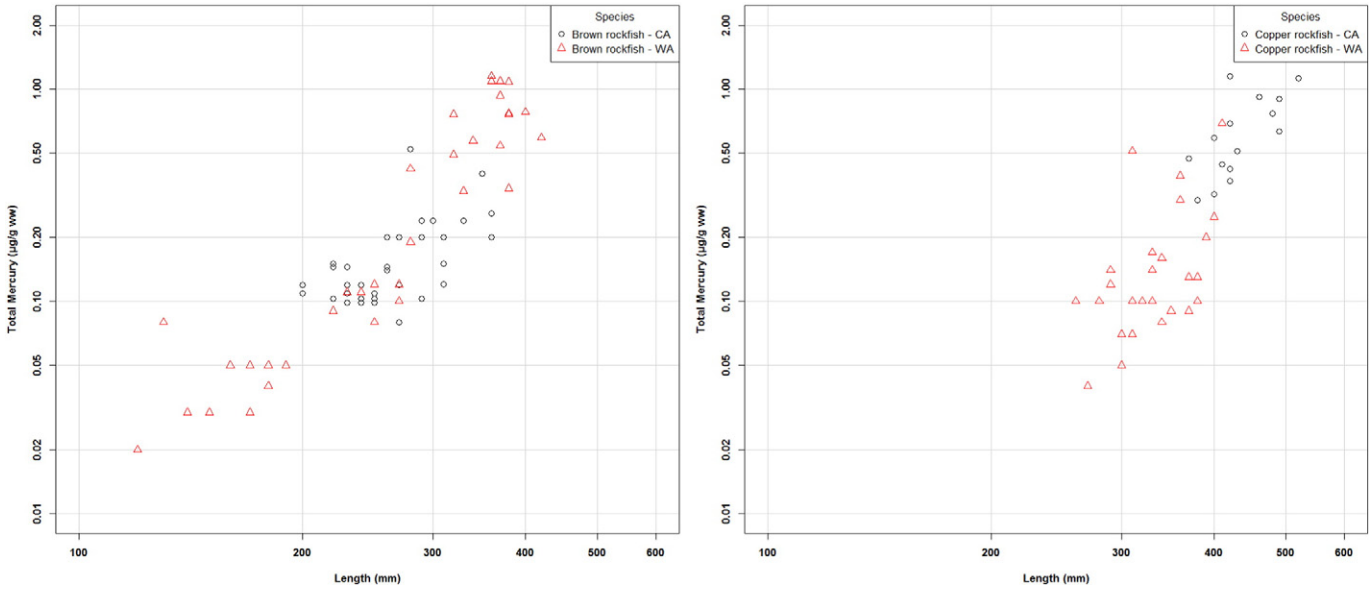

Fig. 5. Scatterplots of THg versus length for species common to CA and WA. Muscle data, individual fish. Left: brown rockfish; Right: copper rockfish.

that are most commonly caught and consumed on the California coast. Aside from brown smooth-hound and gopher rockfish, species with the combination of high trophic position and age are not common on the south coast.

Many coastal locations had species with high or very high mean concentrations. Species with location means above 0.5 µg/g ww included several rockfish species, lingcod, cabezon, bat ray, and several shark species. Copper rockfish and three shark species (leopard shark, spiny dogfish, and brown smooth-hound) had location means above 1.0 µg/g ww.

3.2. Patterns in indicators of elevated risk to humans based on the dataset for the Western US

The recent and robust dataset from California provided a basis for a more thorough examination of spatial and temporal patterns in fish Hg in coastal waters of Western North America.

There is a striking lack of data on Hg and other contaminants in coastal fish in publicly accessible state, federal, and provincial databases (Fig. 2 in the Supplemental material). A search of these databases identified 14 programs that have generated publicly accessible data on Hg in

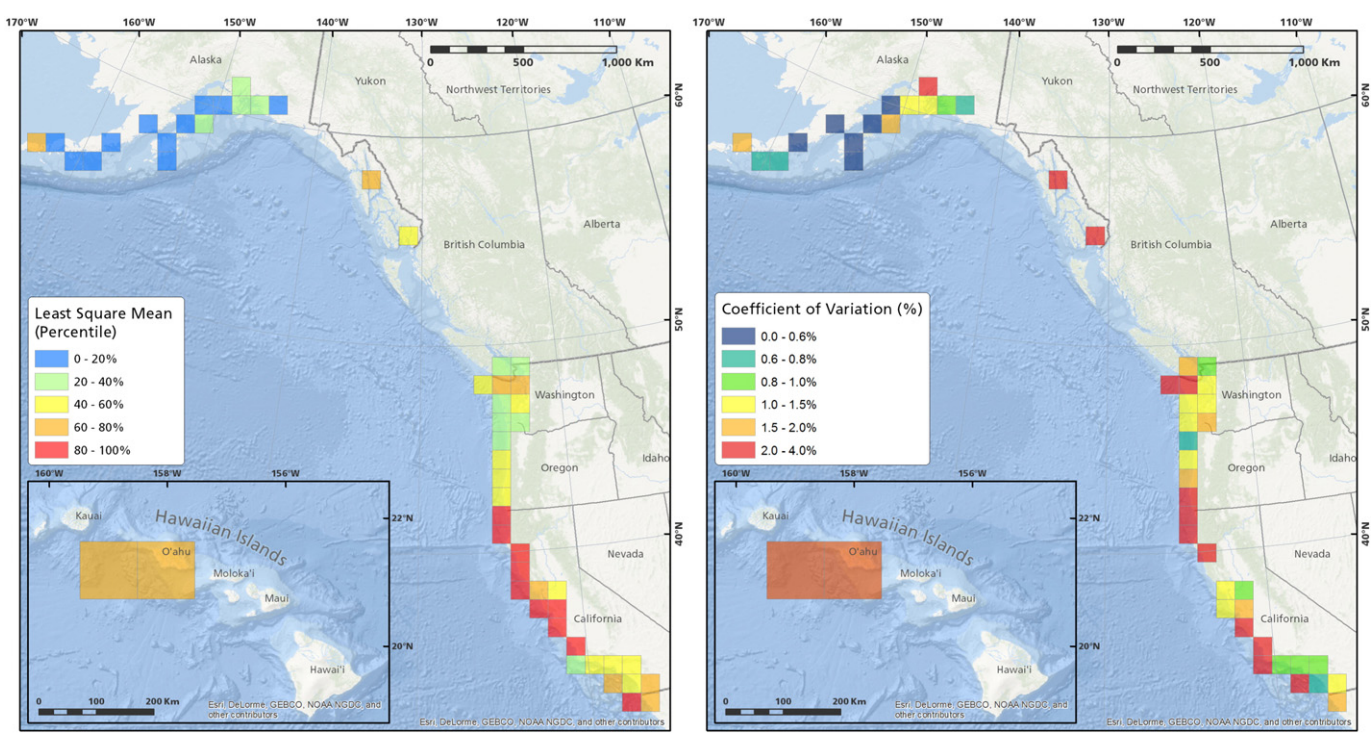

Fig. 6. Distribution of least square mean THg concentrations (left) and coefficients of variation (right) in fish muscle tissue (µg/g ww) for Western North American coastal grid cells. Least square mean values account for the effects of species, size, site, and year.

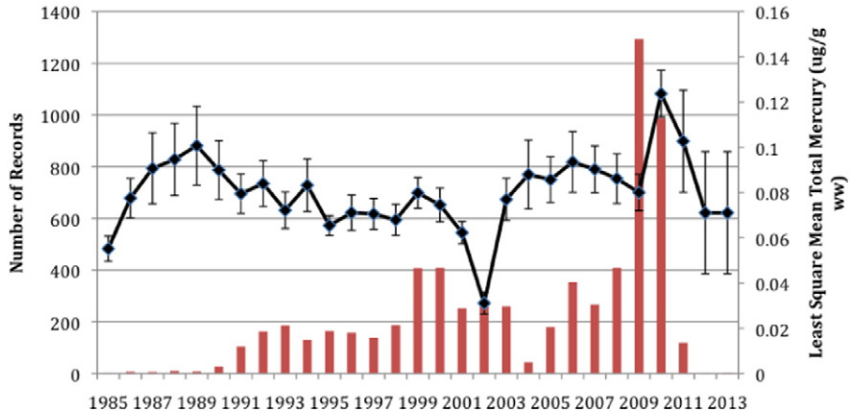

Fig. 7. Number of records for each year (columns) and annual mean of least square mean fish muscle total Hg (THg) concentrations (μg/g ww) for the entire coastal fish dataset (line). Least square mean concentrations account for the effects of site, size, species, and year. Error bars represent one standard error.

coastal fish of the Western US (Table 1 in the Supplemental material). California has the most extensive dataset and is the only state to generate substantive datasets after 2002. Most of the available data have been generated by five significant programs: in Washington, the Puget Sound Ambient Monitoring Program; in California, the Surface Water Ambient Monitoring Program (discussed in Section 3.1), the statewide Coastal Fish Contamination Program, Orange County Sanitation District's outfall monitoring, the Regional Monitoring Program for Water Quality in San Francisco Bay; and USEPA's Environmental Monitoring and Assessment Program. Only the three California programs have generated data in the past 10 years, and only the RMP sport fish and Orange County programs are conducting continued monitoring of long-term trends. Very few data were found in publicly accessible databases for Alaska, Oregon, and Hawaii, and none were found for the coastal waters of British Columbia. The Alaska Department of Environmental Conservation has recently been conducting extensive marine fish contaminant research and monitoring, but publicly accessible data are currently limited because that study is still in progress. Many of the data records that were accessible were incomplete, as discussed in the Methods section. Parameters that were often missing included length and/or weight, sample type (composite or individual fish), tissue analyzed, and numbers of fish in a composite. The dataset included a mixture of muscle and whole body analyses.

The assembled dataset included data from 109 species (Table 2 in the Supplemental material) from 32 families (Fig. 2) and 322 locations (Fig. 3). Several species included in monitoring by the California programs had the most extensive spatial coverage, including white croaker (64 locations), shiner surfperch (57 locations), and topsmelt (40 locations).Starry flounder (37 locations) and English sole (35 locations) also had relatively extensive coverage, with distributions spanning multiple states.

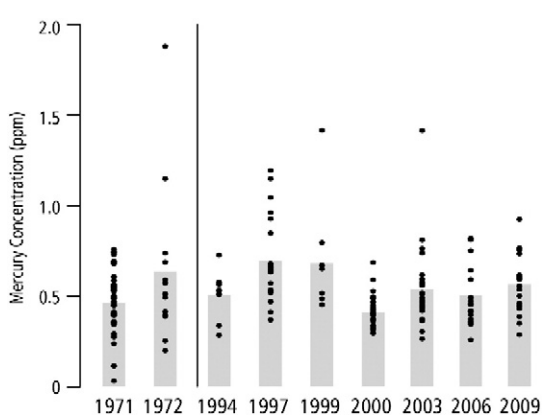

Fig. 8. Hg concentrations (μg/g) in striped bass from San Francisco Bay, 1971–2009. Bars indicate average concentrations. Points represent individual fish. Data from the RMP (1994–2009) and an earlier study (1971–1972). To correct for variation in fish length, all plotted data were calculated for a 60-cm fish using the residuals of a length vs. log(Hg) relationship.

Geometric means of the raw data for each site ranged over two orders of magnitude, from 0.01 to 1.24 μg/g ww. The raw data suggest relatively high concentrations along the California coast and in Puget Sound, and relatively low concentrations along the coasts of Alaska and Oregon, and the outer coast of Washington (Fig. 3). San Francisco Bay had particularly high concentrations, with several site means exceeding 0.5 μg/g ww. The north coast of California was another region with relatively high location means, generally in excess of 0.2 μg/g ww. Alaska, Washington, and Oregon had a majority of location means below 0.05 μg/g ww, while only a few locations in California had means in this range.

3.2.1. The influence of species, age, and trophic position

As observed in the recent California survey, the species collected played a large role in determining the observed concentrations. The species collected at each location was a function of the design of each program (i.e., species targeted and sampling methods) and spatial variation in the distribution of fish species and assemblages. Grouping the species by family (Fig. 2), the medians spanned almost two orders of magnitude, from 0.01 μg/g ww for the Osmeridae (including eulachon, a small smelt) to 0.80 μg/g ww for the Triakidae (sharks including leopard shark and brown smooth-hound). The Scorpaenidae is a diverse family (including 19 species represented in the database) that displayed a wide range of concentrations. Locations where sharks and the rockfish species that are high accumulators were collected (e.g., San Francisco Bay, the north coast of California, and Puget Sound) were the locations where high Hg concentrations were observed. Low concentrations were generally observed in Alaska and Oregon because small specimens of species that accumulate lower concentrations (arrowtooth flounder in Alaska, and primarily starry flounder, English sole, and Pacific sanddab in Oregon) were targeted in those areas.

Age and trophic position are two factors that have a large influence on Hg concentrations in fish (Wiener et al., 2007). The dataset assembled for this study suggests that Hg concentrations of public health concern can be observed at any location on the coast of Western North America where long-lived predator species are sampled. Age data were not commonly gathered in these studies, but fish size measured as length or weight provides an index of age. Even length data were missing for many records in the dataset, but were available for some of the key datasets. Graphical examination of Hg versus length provides many valuable insights into the dataset, including variation among species and regions (Fig. 4a,b,c). This analysis was limited to the two states

with extensive datasets for muscle concentrations. A general trend is evident of smaller fish having lower THg concentrations. Trophic position accounts for some of the residual variation in this general length: THg relationship. A prime example of this is the contrast between gopher rockfish and blue rockfish (Fig. 4a), which had a similar size and age and were often co-located, but had distinctly different trophic positions (3.6 for gopher rockfish and 2.8 for blue rockfish). Hg concentrations in blue rockfish were consistently very low, with an average of 0.09 μg/g ww. Growth rate is another important factor, highlighted, for example, by the contrast between copper rockfish and black rockfish, which had similar size and trophic position (4.1 and 4.4, respectively), but copper rockfish have a much slower growth rate (estimated age at collection of 9–20 years versus 5 years for black rockfish) and much higher THg. Fast growth probably also contributed to the low THg in chinook salmon, which reach a relatively large size at a relatively young age (maximum reported age of 9 years), but use of offshore habitat may also be a contributing factor. The data suggest that size, age, and trophic position are all factors that can be important for managers to consider in their efforts to limit exposure and public health risks.

Species that frequently accumulated high concentrations are of greatest interest from a public health perspective. Samples that exhibited the highest concentrations (above 0.5 μg/g ww) were generally for relatively old individuals in species with high trophic positions. Based on published growth curves and information in Fishbase (Table 2 in Supplemental material), the estimated average ages and trophic positions for these species were 7 years and 3.6 for gopher rockfish, 9–20 years and 4.1 for copper rockfish, 5.5 years and 4.3 for lingcod, >15 years and 3.6 for brown smooth-hound, 6 years and 4.5 for striped bass, 16 years and 3.7 for leopard shark, 22 years (based on actual age measurements for the Puget Sound data by West et al. (2001)) and 4.0 for brown rockfish (in Washington where they reached high concentrations - Fig. 4c), and 14 years (West et al., 2001) and 3.8 for quillback rockfish.

These long-lived, high trophic position species accumulated high Hg concentrations wherever they occurred. The limited spatial distributions of species on the Pacific coast and the limited monitoring that has been performed leave few cases where regional comparisons can be made for particular species. Shark species had high concentrations throughout California, even on the south coast where observed concentrations in general were lower than on the north and central coasts. Brown rockfish and copper rockfish are species where direct comparisons between California and Puget Sound in Washington are possible (Fig. 5). The length-Hg plots show remarkably consistent distributions between these two regions, and suggest that high Hg concentrations can be expected more broadly at locations where larger individuals (at or above 400 mm) of these species are analyzed.

Examination of the relationship between trophic position (based on the information compiled in Table 5 in the Supplemental material) and median THg concentration (Fig. 3 in the Supplemental material) suggests that species with a trophic position <3.5 typically have concentrations in the range of 0.1 μg/g or lower.

3.3. Spatial and temporal patterns in MeHg availability to coastal fish in Western North America

In spite of the limitations of the dataset noted previously, statistical modeling was performed to evaluate spatial patterns in MeHg availability to coastal fish. The model controlled for species, site, grid cell, year, and tissue type. Another version of the model that also controlled for trophic position yielded nearly identical results (one grid cell had a different color), so results for the simpler model are presented. Similar to the raw data (Fig. 3), the highest THg concentrations (expressed as least square means for each grid cell) were observed on the north and central coasts of California (Fig. 6), suggesting that not only are species with life history characteristics that favor high accumulation more prevalent in this region, but that more MeHg is available for bioaccumulation in this region. Also similar to the pattern for the raw data, Alaska had relatively low least square mean concentrations. The spatial pattern of least square means differed, however, in a few areas. Two grid cells in Alaska and a few on the south coast of California had relatively high least square means paired with relatively low raw data concentrations, but these means were often based on very few data points (Fig. 2 in the Supplemental material).

The cause of the apparent increased availability of MeHg on the north and central coast of California is unclear. Two general categories of factors to consider are sources and ecosystem sensitivity.

There is clear evidence of the influence of elevated THg loading in one portion of the central coast of California: San Francisco Bay (reviewed by Davis et al., 2012b). Extensive historic Hg and gold mining in the San Francisco Bay watershed created a persistent and distinct legacy of Hg contamination. Striped bass from San Francisco Bay have the highest average THg concentration measured for this species in US estuaries. Studies of spatial gradients in concentrations of THg and Hg isotopes in Bay biota and sediment have linked food web contamination to inputs from the largest historic Hg-mining district in North America (the New Almaden mining district) (Gehrke et al., 2011a, 2011b; Greenfield et al., 2013). It is possible that the elevated THg in San Francisco Bay is being exported to the outer coast and contributing to the higher MeHg observed on the central coast. It is also possible that Hg mines and geologic Hg sources in the Hg mineral belt of the California coast range (Rytuba, 2000), which coincides in extent with the central coast region delineated in this study, are supplying THg in this region. The mineral belt and associated mining activity, however, extend only into the southern tip of the north coast region, and do not appear to be a plausible explanation of the higher MeHg availability in that region. Upwelling of THg or MeHg is another possible source on the coast of California, but the upwelling associated with the California Current System extends along the entire coast from southern British Columbia to southern Baja California (Bakun et al., 2015).

Other studies have indicated that variation in ecosystem sensitivity can drive variation in MeHg availability to the food web (e.g., Wiener et al., 2006; Chalmers et al., 2014, Eagles-Smith et al., 2016). Whether the north and central coasts of California have characteristics that make them more prone to net methylation and availability of MeHg to the food web is unknown.

Output from the same model was used to construct a time series of annual means based on the least square means for each grid in each year (Fig. 7). Although the weaknesses of the dataset must be kept in mind, the time series suggests that MeHg availability to fish on the coast of Western North America has been relatively constant over the nearly 30-year period covered by the dataset.

Another dataset that is more robust but with a limited geographic scope is available for striped bass in San Francisco Bay (Fig. 8). A relatively extensive historical dataset exists for striped bass in the Bay, allowing evaluation of trends over 39 years from 1971 to 2009. Extending the analysis presented by Greenfield et al. (2005), the data are presented as estimated concentrations of each individual fish at a standard length of 60 cm in order to remove bias that might occur from sampling different sized fish in different years. Overall, intra-annual variance has been high, but average concentrations in recent years are not significantly different from those measured in the early 1970s.

3.4. Management implications

The dataset assembled in this study suggests that MeHg contamination is ubiquitous in the coastal food webs of the coast of Western North America. The data suggest that enough Hg is supplied to the entire coast to cause problematic bioaccumulation wherever long-lived predatory fish species are caught and consumed.

This Hg is derived from a variety of sources, including global emissions to the atmosphere followed by deposition to ocean surface waters; upwelling of organic matter from the deep ocean; natural terrestrial

geologic sources; historic Hg, gold, and silver mining; and urban and industrial wastewater and stormwater. The relative importance of the different sources is an important information gap, but the ubiquitous contamination observed suggests that atmospheric deposition of Hg from global sources may be a primary contributor. Peterson et al. (2002) reached a similar conclusion based on the broad distribution of MeHg bioaccumulation in Oregon streams. The north and central coast regions of California, which are relatively free of urban and industrial sources of Hg, had more locations with high concentrations than the areas near the urban centers of San Francisco, Los Angeles, and San Diego. Locations that are even more remote like the Farallon Islands (27 miles west of the San Francisco Peninsula) and island locations off the south coast of California still had moderate contamination. Other sources to coastal waters such as geological sources (volcanoes and vents) and the upwelling transport pathway may also play a role. The California Current System, which stretches from southern British Columbia to southern Baja California, is characterized by substantial coastal upwelling (Bakun et al., 2015). Terrestrial runoff is another likely contributor, though apparently generally a minor one. San Francisco Bay provides an example where local sources play a distinct and significant role (Davis et al., 2012b).

Statewide coastal consumption advisories for Hg in fish are in place for Alaska and Hawaii, but not for California, Oregon, or Washington (USEPA, 2013). British Columbia also does not have a coastal consumption advisory. Given the likely importance of sources that are challenging or impossible to control (global atmospheric emissions and upwelling), raising awareness of options to reduce MeHg exposure is an especially important means of minimizing risk for people who catch and eat fish from coastal waters.

The recent extensive monitoring in California suggests that MeHg accumulation may be widespread in coastal fish. Comparable monitoring in the rest of the region may help to characterize the actual extent of this public and ecological health hazard. Region-wide monitoring would also facilitate a better understanding of any potential for increases in this hazard in response to increasing atmospheric emissions from Asia and in response to climate change. The indications of a lack of trend from 1971 to 2010 in the present study are in contrast to the findings of higher concentrations in the water column of the offshore waters of the Pacific Ocean in 2006 relative to previous studies in 2002 and 1987 (Sunderland et al., 2009). Sunderland et al. suggest that this observed increase may be attributable to increased atmospheric emissions in Asia. Sustained and expanded monitoring would be of great value in tracking changes in Hg contamination of coastal fish in response to global-scale changes and terrestrial Hg control efforts in coastal watersheds. The monitoring that is conducted should be accompanied by rigorous data management procedures to ensure that the datasets are complete and accessible for future assessments.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.03.093.

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Supplemental Material

Table 1. Data sources for fish mercury concentrations.

State	Data Source	Years of Record	Number of Sites	Number of Hg Records
Alaska	FWS AK	1991, 1993	2	9
Washington	PSAMP	1990-2001	5	1052
Oregon	OR	1999-2000	3	9
California	SWAMP (present study)	2009-2010	68	1281
	CA MISC	1989, 1990, 1992, 1998, 2003, 2005, 2007, 2008	12	66
	CCAMP	2004	3	3
	CFCP	1999-2000	55	213
	FWS CA	1991, 1993, 1996	5	85
	Orange County	1998, 2000-2007, 2009	8	859
	Region 4 Water Board	2005	1	1
	Region 8 Water Board	2004-2005	1	6
	RMP Prey Fish	2005-2011	36	1823
	RMP Sport Fish	1994, 1997, 2000, 2003, 2006, 2009	24	605
	TSMP	1985-1995, 1997, 2002	39	96
Hawaii	HONOLULU	2004-2013	8	72
National	EMAP	1999-2000, 2002-2003	105	359

Abbreviations:
FWS - US Fish and Wildlife Service
PSAMP - Puget Sound Ambient Monitoring Program
SWAMP - Surface Water Ambient Monitoring Program
CCAMP - Central Coast Ambient Monitoring Program
CFCP - Coastal Fish Contamination Program
RMP - Regional Monitoring Program for Water Quaity in San Francisco Bay
TSMP - Toxic Substances Monitoring Program
EMAP - Environmental Monitoring and Assessment Program

Supplemental Material

Table 2. Scientific and common names of fish species collected in the California survey, the number of locations in which they were sampled, summary statistics for length and THg, and whether samples were analyzed as composites or individuals.

Family	Common Name	Scientific Name	Number Of Locations Sampled	Min Total Length (cm)	Max Total Length (cm)	Median Total Length (cm)	Min THg (µg/g ww)	Max THg (µg/g ww)	Mean THg (µg/g ww)	sd THg (µg/g ww)	n	Analyzed as Composites	Analyzed as Individuals
Triakidae	Brown smooth-hound shark	*Mustelus henlei*	8	43	71	61	0.235	3.880	0.786	0.914	14		x
Triakidae	Leopard shark	*Triakis semifasciata*	13	46	121	88	0.283	1.840	0.943	0.353	58		x
Sphyraenidae	Pacific barracuda	*Sphyraena argentea*	1	50	50	50	0.140	0.140	0.140	NA	1	x	
Serranidae	Barred sand bass	*Paralabrax nebulifer*	13	11	59	35	0.020	0.660	0.194	0.106	106	x	x
Serranidae	Kelp bass	*Paralabrax clathratus*	16	20	51	32	0.040	0.560	0.159	0.083	201	x	x
Serranidae	Spotted sand bass	*Paralabrax maculatofasciatus*	6	20	43	33	0.060	0.400	0.172	0.069	68	x	x
Scorpaenidae	Black rockfish	*Sebastes melanops*	13	21	51	38	0.040	0.590	0.157	0.107	120	x	x
Scorpaenidae	Black-and-yellow rockfish	*Sebastes chrysomelas*	2	27	27	27	0.320	0.460	0.390	0.099	2	x	
Scorpaenidae	Blue rockfish	*Sebastes mystinus*	22	19	51	30	0.020	0.390	0.100	0.071	170	x	x
Scorpaenidae	Brown rockfish	*Sebastes auriculatus*	16	12	42	26	0.020	1.150	0.254	0.281	79	x	x
Scorpaenidae	China rockfish	*Sebastes nebulosus*	5	26	35	34	0.110	0.740	0.518	0.239	5	x	
Scorpaenidae	Copper rockfish	*Sebastes caurinus*	9	26	52	37	0.040	1.150	0.326	0.302	44	x	x
Scorpaenidae	Gopher rockfish	*Sebastes carnatus*	21	16	35	29	0.050	1.170	0.427	0.250	99	x	x
Scorpaenidae	Greenstriped rockfish	*Sebastes elongatus*	1	23	23	23	0.170	0.170	0.170	NA	1	x	
Scorpaenidae	Kelp rockfish	*Sebastes atrovirens*	3	27	32	29	0.050	0.240	0.103	0.064	7	x	
Scorpaenidae	Olive rockfish	*Sebastes serranoides*	10	22	43	32	0.040	0.460	0.141	0.110	75		x
Scorpaenidae	Quillback rockfish	*Sebastes maliger*	5	22	44	32	0.001	1.060	0.297	0.170	234	x	x
Scorpaenidae	Rasphead rockfish	*Sebastes ruberrimus*	2	61	75	68	0.930	1.440	1.185	0.361	2		x
Scorpaenidae	Rosethorn rockfish	*Sebastes helvomaculatus*	2	22	26	24	0.300	0.410	0.355	0.078	2	x	
Scorpaenidae	Rosy rockfish	*Sebastes rosaceus*	3	19	23	21	0.180	0.680	0.487	0.269	3	x	
Scorpaenidae	Splitnose rockfish	*Sebastes diploproa*	1	25	25	25	0.670	0.670	0.670	NA	1	x	
Scorpaenidae	Spotted scorpionfish	*Scorpaena plumieri*	3	15	30	24	0.040	0.220	0.147	0.095	3	x	
Scorpaenidae	Vermilion rockfish	*Sebastes miniatus*	10	29	49	45	0.080	0.390	0.239	0.098	10	x	
Scorpaenidae	Yellowtail rockfish	*Sebastes flavidus*	2	31	32	32	0.080	0.130	0.105	0.035	2	x	
Scombridae	Chub mackerel	*Scomber japonicus*	16	22	31	24	0.020	0.200	0.072	0.040	57	x	
Sciaenidae	Black croaker	*Cheilotrema saturnum*	2	25	25	25	0.030	0.410	0.220	0.269	2	x	
Sciaenidae	California corbina	*Menticirrhus undulatus*	8	18	45	20	0.010	0.120	0.047	0.037	10	x	
Sciaenidae	Orangemouth corvina	*Cynoscion xanthulus*	1				0.050	0.050	0.050	NA	1		x
Sciaenidae	Queenfish	*Seriphus politus*	8	12	18	16	0.027	0.090	0.056	0.024	8	x	
Sciaenidae	Spotfin croaker	*Roncador stearnsii*	1	35	35	35	0.050	0.050	0.050	NA	1	x	
Sciaenidae	White croaker	*Genyonemus lineatus*	64	13	32	24	0.010	0.414	0.156	0.090	242	x	x
Sciaenidae	Yellowfin croaker	*Umbrina roncador*	14	15	35	26	0.030	0.380	0.106	0.076	21	x	
Salmonidae	Chinook salmon	*Oncorhynchus tshawytscha*	7	39	101	71	0.036	0.130	0.081	0.023	57	x	
Salmonidae	Coho salmon	*Oncorhynchus kisutch*	3	38	64	56	0.020	0.110	0.049	0.019	63	x	
Pleuronectidae	Arrowtooth flounder	*Atheresthes stomias*	19	17	39	24	0.010	0.122	0.024	0.022	28		x
Pleuronectidae	Butter sole	*Isopsetta isolepis*	4	31	31	31	0.010	0.081	0.043	0.032	5	x	x
Pleuronectidae	Diamond turbot	*Hypsopsetta guttulata*	13	21	29	24	0.010	0.218	0.076	0.067	26	x	
Pleuronectidae	Dover sole	*Microstomus pacificus*	6	24	33	25	0.027	0.081	0.044	0.021	8	x	x
Pleuronectidae	English sole	*Pleuronectes vetulus*	35	6	47	28	0.001	0.300	0.059	0.034	702	x	x
Pleuronectidae	Flathead sole	*Hippoglossoides elassodon*	19				0.010	0.081	0.029	0.018	27	x	x
Pleuronectidae	Hornyhead turbot	*Pleuronichthys verticalis*	6	12	18	14	0.001	0.540	0.095	0.111	418	x	x
Pleuronectidae	Pacific halibut	*Hippoglossus stenolepis*	1				0.027	0.027	0.027	NA	1		
Pleuronectidae	Rex sole	*Glyptocephalus zachirus*	6				0.010	0.257	0.052	0.091	7	x	x
Pleuronectidae	Rock sole	*Lepidopsetta bilineata*	14	16	27	22	0.010	0.081	0.028	0.023	15	x	x
Pleuronectidae	Sand sole	*Psettichthys melanostictus*	2	10	13	12	0.027	0.041	0.032	0.008	3	x	
Pleuronectidae	Slender sole	*Lyopsetta exilis*	1	14	15	15	0.041	0.054	0.047	0.010	2	x	
Pleuronectidae	Spotted turbot	*Pleuronichthys ritteri*	3	21	24	22	0.010	0.050	0.038	0.015	6	x	

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Family	Common Name	Scientific Name	Number Of Locations Sampled	Min Total Length (cm)	Max Total Length (cm)	Median Total Length (cm)	Min THg (µg/g ww)	Max THg (µg/g ww)	Mean THg (µg/g ww)	sd THg (µg/g ww)	n	Analyzed as Composites	Analyzed as Individuals
Pleuronectidae	Starry flounder	*Platichthys stellatus*	37	6	45	12	0.014	0.257	0.048	0.040	65	x	x
Pleuronectidae	Yellowfin sole	*Limanda aspera*	9				0.010	0.054	0.030	0.019	9	x	x
Paralichthyidae	Bigmouth sole	*Hippoglossina stomata*	2				0.010	0.320	0.070	0.052	61	x	
Paralichthyidae	California halibut	*Paralichthys californicus*	22	18	98	55	0.014	0.586	0.167	0.118	58	x	x
Paralichthyidae	Fantail sole	*Xystreurys liolepis*	2	16	20	18	0.020	0.020	0.020	0.000	2	x	
Paralichthyidae	Longfin sanddab	*Citharichthys xanthostigma*	2				0.001	0.878	0.152	0.135	81	x	
Paralichthyidae	Pacific sanddab	*Citharichthys sordidus*	12	11	29	24	0.014	0.527	0.064	0.079	174	x	x
Paralichthyidae	Speckled sanddab	*Citharichthys stigmaeus*	18	7	29	10	0.010	0.068	0.037	0.018	31	x	
Osmeridae	Eulachon	*Thaleichthys pacificus*	2				0.010	0.014	0.012	0.002	2		x
Mugilidae	Striped mullet	*Mugil cephalus*	1				0.014	0.041	0.027	0.019	2	x	x
Moronidae	Striped bass	*Morone saxatilis*	21	4	86	48	0.041	0.906	0.281	0.178	142	x	x
Merlucciidae	North Pacfic hake	*Merluccius productus*	1	49	49	49	0.150	0.150	0.150	NA	1	x	
Malacanthidae	Ocean whitefish	*Caulolatilus princeps*	2	28	38	33	0.090	0.100	0.095	0.007	2	x	
Lutjanidae	Yellow and blue seaperch	*Lutjanus kasmira*	8				0.010	0.380	0.114	0.080	31	x	
Labridae	California sheephead	*Semicossyphus pulchere*	4	26	34	30	0.130	0.210	0.168	0.035	4	x	
Kyphosidae	Halfmoon	*Medialuna californiensis*	1	17	28	23	0.040	0.050	0.045	0.007	2	x	
Kyphosidae	Opaleye	*Girella nigricans*	6	14	34	22	0.010	0.090	0.053	0.029	8	x	
Holocentridae	Bigscale soldierfish	*Myripristis berndti*	2				0.050	0.120	0.083	0.021	9	x	
Hexagrammidae	Kelp greenling	*Hexagrammos decagrammus*	5	24	38	36	0.060	0.380	0.247	0.125	6	x	
Hexagrammidae	Lingcod	*Ophiodon elongatus*	13	62	81	67	0.190	0.670	0.341	0.135	14	x	
Haemulidae	Sargo	*Anisotremus davidsonii*	1	26	26	26	0.040	0.040	0.040	NA	1	x	
Gobiidae	Arrow goby	*Clevelandia ios*	3	2	5	3	0.014	0.203	0.077	0.034	182	x	x
Gobiidae	Bay goby	*Lepidogobius lepidus*	4				0.014	0.068	0.033	0.017	26	x	x
Gobiidae	Cheekspot goby	*Ilypnus gilbert*	1				0.041	0.149	0.057	0.020	61	x	x
Gobiidae	Gobies	*Gobiidae*	1				0.027	0.027	0.027	NA	1		x
Gobiidae	Longjaw mudsucker	*Gillichthys mirabilis*	11	6	14	8	0.001	0.122	0.046	0.027	47	x	x
Gobiidae	Shimofuri goby	*Tridentiger bifasciatus*	1	4	5	5	0.041	0.108	0.060	0.018	15	x	x
Gobiidae	Yellowfin goby	*Acanthogobius flavimanus*	6	7	12	11	0.027	0.095	0.054	0.016	37	x	x
Gadidae	Pacific cod	*Gadus macrocephalus*	4	45	63	50	0.041	0.180	0.105	0.031	31	x	
Gadidae	Pacific tomcod	*Microgadus proximus*	4	17	17	17	0.027	0.122	0.054	0.045	4	x	
Gadidae	Walleye pollock	*Theragra chalcogramma*	2				0.010	0.068	0.039	0.041	2		x
Fundulidae	California killifish	*Fundulus parvipinnis*	9	5	35	6	0.010	0.095	0.037	0.024	20	x	
Fundulidae	Rainwater killifish	*Lucania parva*	4	3	4	3	0.027	0.203	0.099	0.044	31	x	x
Engraulidae	Anchovy	*Engraulidae*	5	8	9	9	0.049	0.078	0.064	0.012	5	x	
Engraulidae	Northern anchovy	*Engraulis mordax*	7	6	10	8	0.027	0.486	0.145	0.126	38	x	x
Embiotocidae	Barred surfperch	*Amphistichus argenteus*	29	11	36	19	0.010	0.405	0.116	0.087	100	x	x
Embiotocidae	Black perch	*Embiotoca jacksoni*	17	13	32	24	0.010	0.220	0.098	0.042	114	x	x
Embiotocidae	Pile perch	*Rhacochilus vacca*	3	28	38	34	0.060	0.280	0.158	0.089	12	x	x
Embiotocidae	Rainbow seaperch	*Hypsurus caryi*	9	11	34	27	0.010	0.680	0.149	0.138	32	x	x
Embiotocidae	Redtail surfperch	*Amphistichus rhodoterus*	1	30	30	30	0.110	0.210	0.153	0.051	3	x	
Embiotocidae	Rubberlip Surfperch	*Rhacochilus toxotes*	1	32	37	35	0.268	0.427	0.353	0.059	12	x	
Embiotocidae	Shiner surfperch	*Cymatogaster aggregata*	57	5	15	11	0.010	0.420	0.086	0.048	181	x	x
Embiotocidae	Silver surfperch	*Hyperprosopon ellipticum*	2	21	22	22	0.120	0.180	0.150	0.042	2	x	
Embiotocidae	Spotfin surfperch	*Hyperprosopon anale*	1	10	10	10	0.040	0.040	0.040	NA	1	x	
Embiotocidae	Striped seaperch	*Embiotoca lateralis*	1	24	24	24	0.068	0.068	0.068	NA	1	x	
Embiotocidae	Walleye surfperch	*Hyperprosopon argenteum*	14	14	32	21	0.014	0.260	0.113	0.065	34	x	x
Embiotocidae	White seaperch	*Phanerodon furcatus*	13	10	35	22	0.010	0.162	0.067	0.032	49	x	x

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Family	Common Name	Scientific Name	Number Of Locations Sampled	Min Total Length (cm)	Max Total Length (cm)	Median Total Length (cm)	Min THg (µg/g ww)	Max THg (µg/g ww)	Mean THg (µg/g ww)	sd THg (µg/g ww)	n	Analyzed as Composites	Analyzed as Individuals
Cynoglossidae	California tonguefish	*Symphurus atricauda*	1	13	13	13	0.014	0.014	0.014	NA	1	x	
Cottidae	Cabezon	*Scorpaenichthys marmoratus*	12	43	51	47	0.190	0.730	0.387	0.160	13	x	
Cottidae	Pacific staghorn sculpin	*Leptocottus armatus*	24	3	15	11	0.014	0.432	0.081	0.053	78	x	x
Clupeidae	American shad	*Alosa sapidissima*	2	8	10	8	0.054	0.095	0.073	0.012	8	x	
Clupeidae	Baltic herring	*Clupea harengus*	1				0.027	0.050	0.037	0.010	4		x
Clupeidae	Pacific herring	*Clupea harengus*	5	7	21	17	0.019	0.135	0.053	0.022	122	x	x
Clupeidae	Pacific Sardine	*Sardinops sagax*	1	19	19	19			NA	NA	1	x	
Clupeidae	Threadfin shad	*Dorosoma petenense*	1				0.068	0.068	0.068	NA	2	x	
Carangidae	Bigeye scad	*Selar crumenophthalmus*	8				0.020	0.550	0.138	0.109	32	x	
Atherinopsidae	Inland silverside	*Menidia beryllina*	27	4	8	6	0.014	0.514	0.145	0.093	531	x	x
Atherinopsidae	Jacksmelt	*Atherinopsis californiensis*	20	20	29	25	0.015	0.257	0.083	0.048	67	x	x
Atherinidae	Topsmelt	*Atherinops affinis*	40	3	35	7	0.014	0.297	0.068	0.033	875	x	x
Acipenseridae	Green sturgeon	*Acipenser medirostris*	1	103	103	103	0.130	0.130	0.130	NA	1		x
Acipenseridae	Sturgeon (unspecified)	*Acipenseridae*	2	121	142	133	0.220	0.350	0.275	0.058	4		x
Acipenseridae	White Sturgeon	*Acipenser transmontanus*	8	60	169	122	0.100	0.707	0.298	0.138	38	x	x

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Table 3. Classification of average methylmercury concentrations for each species at each location: North Coast. Red: >0.44 ppm. Green: <0.07 ppm. Yellow: between 0.07 and 0.44 ppm.

			Coastal Species																													Bay and Harbor Species														
Regional Board	Zone	Station Name	Black Rockfish	Blue Rockfish	Quillback Rockfish	Olive Rockfish	Copper Rockfish	Gopher Rockfish	China Rockfish	Vermillion Rockfish	Yellowtail Rockfish	Rosy Rockfish	Brown Rockfish	Black and Yellow Rockfish	Kelp Rockfish	Cabezon	Kelp Greenling	Lingcod	Barred Sand Bass	Kelp Bass	Spotted Sand Bass	Chub Mackerel	Spotted Scorpionfish	Pacific Barracuda	Ocean Whitefish	Rainbow Surfperch	Barred Surfperch	Black Perch	White Croaker	Black Croaker	Bat Ray	California Halibut	Leopard Shark	Brown Smoothhound Shark	Gray Smoothhound Shark	Spiny Dogfish	Shiner Surfperch	Pile Surfperch	White Surfperch	Yellowfin Croaker	Spotfin Croaker	Topsmelt	Jacksmelt	Striped Bass	Opaleye	Cunnefish
1	68	Crescent City Coast																																												
1	67	Del Norte Coast																																												
1	66	North Humboldt County Coast Area																																												
1	65	Trinidad Area																																												
1	64	Humboldt Bay																																												
1	62	Cape Mendocino Area																																												
1	61	Shelter Cove Area																																												
1	60	North Mendocino County Coast Area																																												
1	59	Fort Bragg Area																																												
1	58	Mendocino Coast Area																																												
1	57	Point Arena Area																																												
1	55/56	South Sonoma Coast/North Sonoma Coast																																												
1	54	Bodega Harbor																																												
2	53	Northern Marin Coast																																												
2	52	Tomales Bay																																												

Supplemental Material

Table 4. Classification of average methylmercury concentrations for each species at each location: Central Coast. Red: >0.44 ppm. Green: <0.07 ppm. Yellow: between 0.07 and 0.44 ppm.

			Coastal Species																												Bay and Harbor Species														
Regional Board	Zone	Station Name	Black Rockfish	Blue Rockfish	Quillback Rockfish	Olive Rockfish	Copper Rockfish	Gopher Rockfish	China Rockfish	Vermilion Rockfish	Yelloweye Rockfish	Rosy Rockfish	Brown Rockfish	Black and Yellow Rockfish	Kelp Rockfish	Cabezon	Kelp Greenling	Lingcod	Barred Sand Bass	Kelp Bass	Spotted Sand Bass	Chub Mackerel	Spotted Scorpionfish	Pacific Barracuda	Ocean Whitefish	Rainbow Surfperch	Barred Surfperch	Black Perch	White Croaker	Black Croaker	Bat Ray	California Halibut	Leopard Shark	Brown Smoothhound Shark	Gray Smoothhound Shark	Spiny Dogfish	Shiner Surfperch	Pile Surfperch	White Surfperch	Yellowfin Croaker	Spotfin Croaker	Topsmelt	Jacksmelt	Striped Bass	Opaleye	Queenfish
2	51	Southern Marin Coast																																												
2	206SNPBLO	San Pablo Bay (5)																																												
2	203BRKLEY	Berkeley (4)																																												
2	203CENTRL	Central Bay (6)																																												
2	203SANFRN	San Francisco Waterfront (3)																																												
2	203OAKLND	Oakland (2)																																												
2	204STHBAY	South Bay (1)																																												
2	50	Farallon Islands																																												
2	49	San Francisco Coast																																												
2	48	Pacifica Coast																																												
2	47	Half Moon Bay Coast																																												
2	46	Pillar Point Harbor																																												
2	45	San Mateo Coast																																												
3	43	Santa Cruz Coast Area																																												
3	42	Santa Cruz Area Wharfs/ Beachs																																												
3	41	Elkhorn Slough																																												
3	40	Moss Landing/Marina Coast																																												
3	39	Monterey/Pacific Grove Coast																																												
3	38	Carmel Coast																																												
3	36/37	Southern Monterey County Coast/Big Sur Coast																																												
3	34/35	Cambria/Cayucos Coast/ Northern San Luis Obispo County Coast																																												
3	33	Morro Bay																																												
3	32	Morro Bay Coast																																												
3	31	Diablo Canyon Coast																																												
3	30	Port San Luis Area																																												
3	28/29	North Santa Barbara County Coast/Pismo Beach Area																																												

Supplemental Material

Table 5. Classification of average methylmercury concentrations for each species at each location: South Coast. Red: >0.44 ppm. Green: <0.07 ppm. Yellow: between 0.07 and 0.44 ppm.

			Coastal Species																												Bay and Harbor Species															
Regional Board	Zone	Station Name	Black Rockfish	Blue Rockfish	Quillback Rockfish	Olive Rockfish	Copper Rockfish	Gopher Rockfish	China Rockfish	Vermilion Rockfish	Yellowtail Rockfish	Rosy Rockfish	Brown Rockfish	Black and Yellow Rockfish	Kelp Rockfish	Cabezon	Kelp Greenling	Lingcod	Barred Sand Bass	Kelp Bass	Spotted Sand Bass	Chub Mackerel	Spotted Scorpionfish	Pacific Barracuda	Ocean Whitefish	Rainbow Surfperch	Barred Surfperch	Black Perch	White Croaker	Black Croaker	Bat Ray	California Halibut	Leopard Shark	Brown Smoothhound Shark	Gray Smoothhound Shark	Spiny Dogfish	Shiner Surfperch	Pile Surfperch	White Surfperch	Yellowfin Croaker	Spotfin Croaker	Topsmelt	Jacksmelt	Striped Bass	Opaleye	Queenfish
3	27	Goleta to Pt Conception																																												
3	25	Rincon to Goleta																																												
3	26	Santa Barbara Channel Oil Platform																																												
3	23	Northern Channel Islands																																												
4	24	Ventura to Rincon																																												
4	22	Pt Dume to Oxnard																																												
4	21	North Santa Monica Bay																																												
4	20	Middle Santa Monica Bay																																												
4	19	South Santa Monica Bay																																												
4	18	Palos Verdes																																												
4	16	San Pedro Bay																																												
4	15	Long Beach																																												
8	14	Orange County Oil Platforms																																												
8	13	Santa Ana River to Seal Beach																																												
8	12	Newport Bay																																												
8	11	Crystal Cove to Santa Ana River																																												
9	10	Dana Point Harbor																																												
9	9	San Onofre to Crystal Cove																																												
4	17	Catalina Island																																												
9	8	Oceanside Harbor																																												
9	7	La Jolla to San Onofre																																												
9	6	Mission Bay																																												
9	5	Pt Loma to La Jolla																																												
9	4	Pt Loma																																												
9	3	SD North Bay																																												
9	2	SD South Bay																																												
9	1	TJ to North Island																																												

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Table 6. Characteristics of the species sampled in the California survey. Information on trophic level, feeding position, habitat, and range from Fishbase (www.fishbase.org). Age at size sampled estimated by comparing the median size sampled in this survey to published growth curves.

Family	Species	Trophic Level±se[1]	Size Sampled (mm) range (median)	Estimated age at Size Sampled	Primary Prey	Feeding Position[2]	Habitat	Range	Depth
Hound Sharks (Triakidae)	Leopard Shark (*Triakis semifasciata*)	3.7±0.5	930-1410 (1238)	16 (Kusher et al. 1992)	nektonic and benthic fishes, crustaceans, octopi and clams	benthic	enclosed muddy bays, estuaries and lagoons	Oregon to Baja	0-91m
	Gray Smooth-hound (*Mustelus californicus*)	3.5±0.5	616-685 (630)	2 (Yudin and Cailliet 1990)	mostly crabs, ghost shrimp, and small fish	benthic	inshore and offshore soft bottom, entering shallow muddy bays	Northern CA to Baja	0-200m
	Brown Smooth-hound (*Mustelus henlei*)	3.6±0.5	826-1144 (978)	15+ (Yudin and Cailliet 1990)	crabs, shrimp and some fishes	benthic	offshore, soft bottom	Northern CA to Baja	0-200m
Dogfish Sharks (Squalidae)	Spiny Dogfish (*Squalus acantias*)	4.3±0.7	995-1140 (1011)		fishes, crustaceans, squid and octopi	benthic/ mid-water	Near bottom in enclosed bays and estuaries, also mid-water and near surface	Bering Sea to Chile	0-1460m

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2

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Table 6. Continued.

Barracudas (Sphyraenidae)	Pacific Barracuda (*Sphyraena argentea*)	4 (4.5±0.8)	450-590 (479)		small anchovies, smelt, squid, and other small, schooling fish	Mid-water	Usually near shore or near the surface; Pelagic spawners; Young enter bays	Alaska to southern Baja California; rare north of Pt. Concep. in California	0-18m
Basses (Serranidae)	Spotted Sand bass (*Paralabrax maculatofasciatus*)	4 (4.2±0.6)	195-430 (327)		small fishes and benthic crustaceans, clams	demersal	sand or mud bottom near rocks and eelgrass	Monterey, CA to Mexico	0-60m
	Kelp Bass (*Paralabrax clathratus*)	3.9±0.6	185-512 (316)	5 (Young 1963) 7 (Love et al. 1996)	Small fishes (including anchovies, sardines, surfperch), squid, octopus, crabs, shrimps, and amphipods	mid-water	in or near kelp beds, but may be associated with any structure	Washington to Baja	0-50m
	Barred Sand bass (*Paralabrax nebulifer*)	3.5±0.5	257-590 (346)	7 (Love et al. 1996)	fishes and crustaceans	demersal	sandy bottom among or near rocks	Santa Cruz, CA to Baja	0-183m

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Table 6. Continued.

Rockfish (Scorpaenidae)	Olive Rockfish (*Sebastes serranoides*)	3.9±0.6	208-425 (322)	4 (Lea et al. 1999)	fishes (particularly juvenile rockfishes), octopi, squid, copepods and crab larvae	mid-water	areas of reef or giant kelp, over hard, high relief	Northern CA to Baja (abundant SoCal to Mendocino County)	0-146m
	Yellowtail Rockfish (*Sebastes chrysomelas*)	3.5±0.5	290-350 (313)	5 (Lea et al. 1999)	Pelagic crustaceans, fish, krill, plankton	demersal	holes and crevices in rocky areas; Found in intertidal areas	Northern CA to Baja CA	0-37m
	Copper Rockfish (*Sebastes caurinus*)	4.1±0.7	340-522 (411)	9 (Lea et al. 1999) 20 (Love et al. 2002)	Shrimp, crab, octopi, small fish	demersal	shallow, protected bays and inlets, among rocks and kelp beds	Alaska to central Baja California	10-183m
	Vermilion Rockfish (*Sebastes miniatus*)	3.8±0.6	229-551 (437)	7 (Lea et al. 1999)	Shrimp, squid, octopi, fish (mainly smaller rockfish)	demersal	shallow to deep rocky reefs, less common on deep ones	BC Canada to central Baja CA	183-274m
	Rosy Rockfish (*Sebastes rosaceus*)	3.6±0.6	175-257 (215)	8 (Lea et al. 1999)	Squid, crustaceans, fish	demersal		Washington to central Baja CA	15-128m

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Table 6. Continued.

	Quillback Rockfish (*Sebastes maliger*)	3.8±0.6	423-439 (431)		shrimp, crab	demersal	rocky bottoms and reefs; never far from cover	Alaska to Central CA	0-274m
	Kelp Rockfish (*Sebastes atrovirens*)	3.4±0.5	269-335 (294)		shrimp, amphipods, small fish	demersal	found on or near bottom in kelp beds or rocky areas	Central CA to Baja CA	0-46m
	Gopher Rockfish (*Sebastes carnatus*)	3.6±0.5	147-371 (281)	7 (Lea et al. 1999)	crabs, brittle stars, mysids	demersal	Inhabit holes or crevices in rocky areas; territorial	Northern CA to central Baja CA	0-55m
	China Rockfish (*Sebastes nebulosus*)	3.8±0.6	245-385 (332)	11 (Lea et al. 1999)	brittle stars, shrimp, fish, other animals on the bottom	demersal	Inshore along rocks and reefs	Alaska to Redondo Beach, CA	3-128m
	Brown Rockfish (*Sebastes auriculatus*)	4.0±0.6	205-392 (302)	5 (Love and Johnson 1998)	small fish, crab, shrimp, isopods, and polychaetes	demersal	hard bottom; aggregate near rocks, oil platforms, sewer pipes	Alaska to Baja	0-128m
	Black Rockfish (*Sebastes melanops*)	4.4±0.8	213-511 (380)	5 (Wallace et al. 1999 in Love et al. 2002)	juvenile rockfish, euphausids and amphipods (upwelling), and invertebrates (non-upwelling)	mid-water	kelp beds	Alaska to SoCal	0-366m
	Black and Yellow Rockfish (*Sebastes chrysomelas*)	3.5±0.5	254-302 (270)	7 (Lea et al. 1999)	crustaceans, mollusks and fishes	demersal	holes and crevices in rocky areas; intertidal areas	Northern CA to central Baja CA	0-37m

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Table 6. Continued.

	Blue Rockfish (*Sebastes mystinus*)	2.8±0.3	51-395 (293)	7-11 (Miller and Geibel 1973) 8 (Love et al 2002)	tunicates, hydroids, jellyfishes, and larval and juvenile fishes	mid-water	deep rocky reefs or hard, flat substrates	Bering Sea to Baja	0-100m
	Spotted Scorpionfish (*Scorpaena guttata*)	4 (3.8±0.6)	200-322 (290)		crab, squid, octopus, fishes and shrimp	demersal	rocky areas of bays and along shore, especially in caves and crevices	Santa Cruz, CA to Baja CA	0-183m
Mackerels (Scombridae)	Pacific Chub Mackerel (*Scomber japonicus*)	3.1±0.4	199-335 (240)	1 (Hwang et al. 2008)	copepods, crustaceans, euphausids, small fishes and squids	mid-water	pelagic	Indo-Pacific	0-300m
Croaker (Sciaenidae)	Yellowfin Croaker (*Umbrina roncador*)	3.5±0.5	121-376 (195)		crustaceans and fishes	benthic	coastal waters and estuaries	Pt. Concep. To Gulf of CA (old records have as far north as SF)	0-45m
	White Croaker (*Genyonemus lineatus*)	3.4±0.5	164-300 (220)	7-8	polychaetes, small shrimps, crabs and mollusks	benthic	Over sandy bottoms	BC to Baja	0-183m
	Spotfin Croaker (*Roncador stearnsii*)	3.3±0.4	138-372 (221)		marine worms, clams, crabs and small crustaceans	demersal	sandy shores and bays, mostly in shallow surf zones	Pt. Concep. to south Baja CA	0-15m

Supplemental Material

Table 6. Continued.

	Queenfish (*Seriphus politus*)	3.7±0.6	156-174 (165)		small shrimps, marine worms and fishes	demersal	inshore, often over sandy bottoms. Common in bays and tidal sloughs, around pilings	Oregon to south Baja CA	1-21m
	Black Croaker (*Cheilotrema saturnum*)	3.6±0.6	234-261 (242)		Crabs,shrimp	demersal	near the bottom, often in caves and crevices of exposed coasts and open bays	Northern CA to Baja CA	0-46m
Sand Flounder (Paralichthyidae)	California Halibut (*Paralichthys californicus*)	4.5±0.6	266-810 (670)	7-9	fishes and squids	demersal	sandy bottoms, also in bays and estuaries	Northern WA to Baja	0-183m
Eagle and Manta Rays (Myliobatidae)	Bat Ray (*Myliobatis californica*)	3 (3.1±0.3)	176-921 (405)		bivalves, snails, polychaetes, shrimps, and crabs	demersal	sandy and muddy bays and sloughs, also on rocky bottom and in kelp beds	Oregon to Gulf of CA	0-46m
Temperate Basses (Moronidae)	Striped Bass (*Morone saxatilis*)	4.5±0.8	460-790 (600)	6 (Moyle 2002)	Zooplankton, invertebrates, fish	demersal	estuaries, bays, and coastal areas	British Columbia to northern Baja CA	0-30m

Supplemental Material

Table 6. Continued.

Tilefishes (Malacanthidae)	Ocean Whitefish (*Caulolatilus princeps*)	4 (3.9±0.6)	270-286 (279)	1-3	Worms, Shrimp, Crab, Octopi, Squid, small Fish	demersal	rocky bottoms; also found on soft sand and mud bottoms.	British Columbia to Peru	10-91m
Sea Chubs (Kyphosidae)	Opaleye (*Girella nigricans*)	2-3 (2.2±0.1)	194-230 (221)		Algae Sometimes eat Shrimp, Amphipods, Jellies	benthic	Intertidal tide pools; near or over rocks and in kelp beds	Oregon to south Baja CA	2-30m
Greenlings (Hexagrammidae)	Lingcod (*Ophiodon elongatus*)	4.3±0.7	551-932 (682)	5-6 (Miller and Geibel 1973)	mostly fishes but also crustaceans, octopi and squid	demersal	near rocks	Alaska to Baja	0-475m
	Kelp Greenling (*Hexagrammos decagrammus*)	3.6±0.6	220-422 (360)		crustaceans, polychaete worms, brittle stars, mollusks, and small fishes	demersal	rocky inshore areas, common on kelp beds, also on sand bottoms	Alaska to SoCal	0-46m
Surfperch (Embiotocidae)	White Surfperch (*Phanerodon furcatus*)	3.4±0.5	99-345 (202)	>7 (Eckmayer 1979)	Opportunistic Crabs, Worms, Amphipods	demersal	near piers, docks, in bays and sandy areas, but usually in quiet water and offshore areas near rocks	British Columbia to northern Baja CA	0-43m
	Shiner Surfperch (*Cymatogaster aggregata*)	3.0±0.3	50-199 (110)	2 (Eckmayer 1979)	calanoid copepods, crustaceans, mollusks	mid-water/ demersal	eelgrass beds, piers and pilings	Alaska to Baja	0-146m

Supplemental Material

Table 6. Continued.

	Rainbow Surfperch (*Hypsurus caryi*)	3.3±0.5	185-342 (280)		amphipods, crabs, worms, shrimp	demersal	rocky shores, often at edges of kelp beds, occasionally over sand but not in surf	Northern CA to northern Baja CA	0-40m
	Pile Surfperch (*Rhacochilus vacca*)	3xx	280-375 (340)		hard-shelled mollusks, crabs and barnacles	demersal	Rocky shore, kelp, pilings	Alaska to Baja CA	0-46m
	Barred surfperch (*Amphistichus argenteus*)	3.5±0.6	105-363 (186)	3 (Carlisle et al. 1960)	sand crabs, clams and other inverts	benthic	surf of sand beaches, also near rocks, pilings and other structures	Bodega Bay, CA to Baja	0-7m
	Black perch (*Embiotoca jacksoni*)	3.2±0.5	152-316 (232)		amphipods, crabs, worms	benthic	rocky areas near kelp, sand bottoms of coastal bays and around piers and pilings	Ft Bragg, CA to Baja	0-46m
Sculpins (Cottidae)	Cabezon (*Scorpaenichthys marmoratus*)	3.6±0.5	380-575 (467)	3-4 (O'Connell 1953)	crustaceans, fish and mollusks	demersal	rocky, sandy and muddy bottoms, kelp beds	Southeastern AK to Baja	0-200m
New World Silversides (Atherinopsidae)	Topsmelt (*Atherinops affinis*)	2.8±0.3	80-377 (128)		zooplankton, algae, benthic invertebrates (Lane and Hill 1975)	benthic/ mid-water	bays, muddy and rocky areas and kelp beds	Vancouver Island to Baja	0-26m

Supplemental Material

Table 6. Continued.

	Jacksmelt (*Atherinopsis californiensis*)	3.1±0.5	240-279 (265)	5-7	crustaceans, fish larvae	mid-water	inshore areas, including bays	Yaquina Bay, OR to Baja	0-29m

1. Trophic levels are the hierarchical strata of a food web characterized by organisms that are the same number of steps removed from the primary producers. The USEPA's 1997 Mercury Study Report to Congress used the following criteria to designate trophic levels based on an organism's feeding habits:
   Trophic level 1: Phytoplankton.
   Trophic level 2: Zooplankton and benthic invertebrates.
   Trophic level 3: Organisms that consume zooplankton, benthic invertebrates, and TL2 organisms.
   Trophic level 4: Organisms that consume trophic level 3 organisms.

2. Benthic – feeding on the bottom Demersal – feeding on or near bottom

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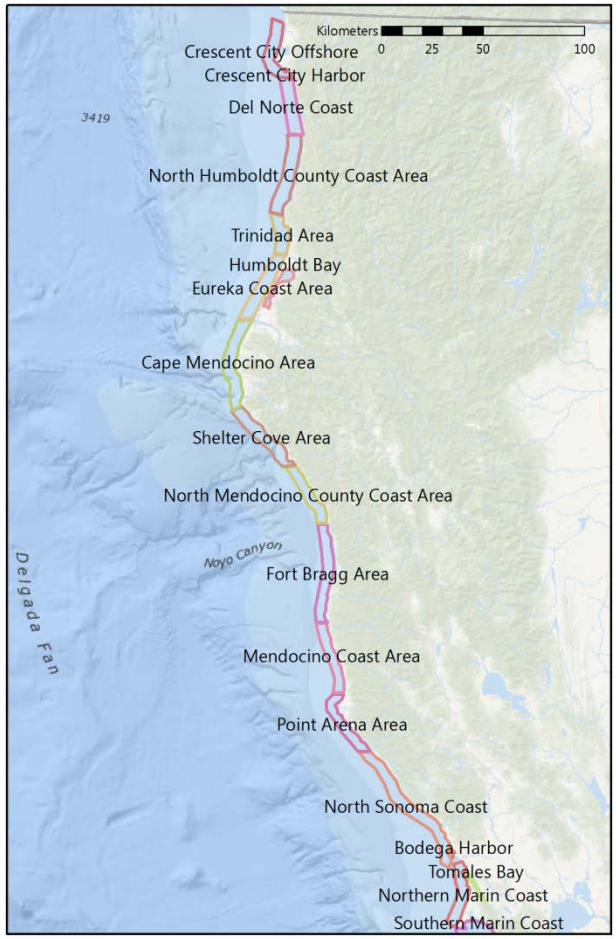
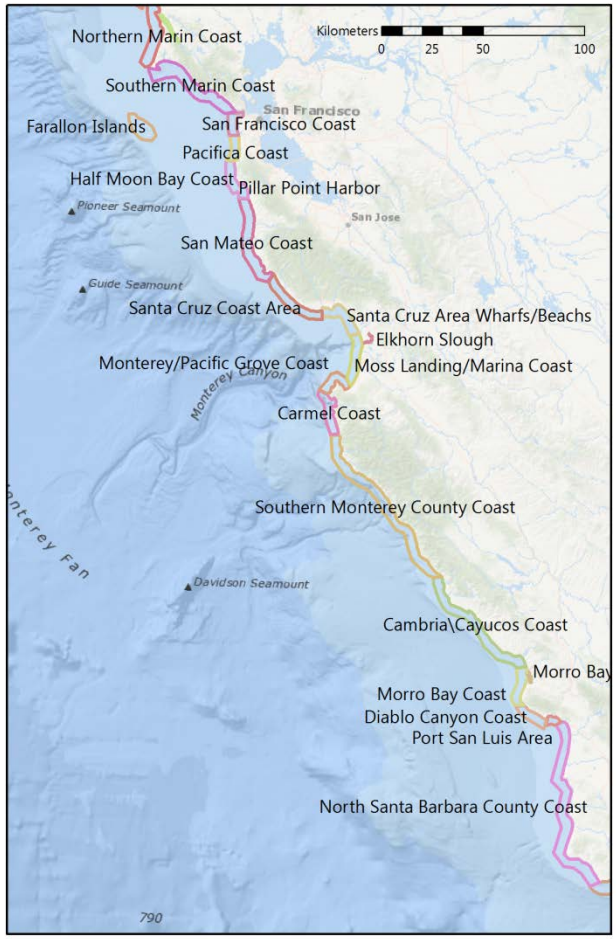
Supplemental Material

Figure 1a. Zones delineated for the California survey: north coast.



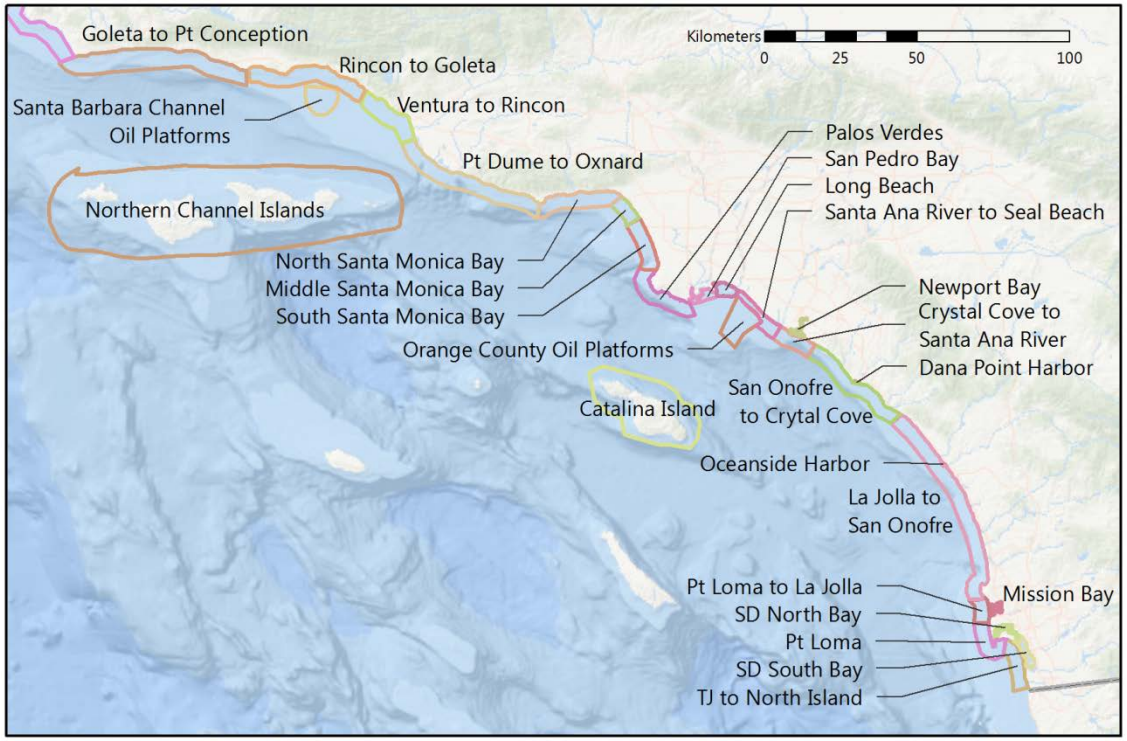
Supplemental Material

Figure 1b. Zones delineated for the California survey: central coast.



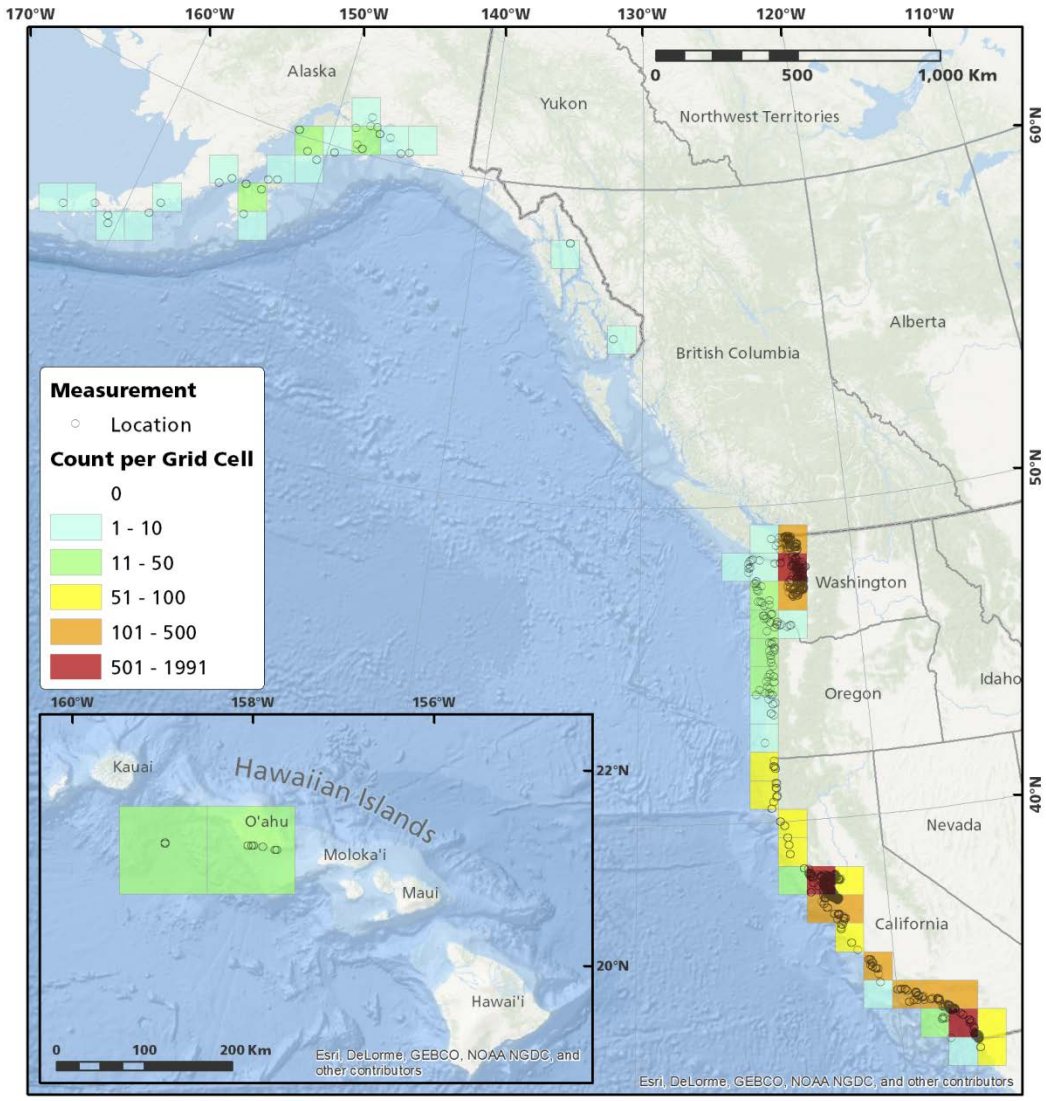
Supplemental Material

Figure 1c. Zones delineated for the California survey: south coast.



Supplemental Material

Figure 2. Distribution of number of THg concentration measurements for Western North America coastal grid cells.



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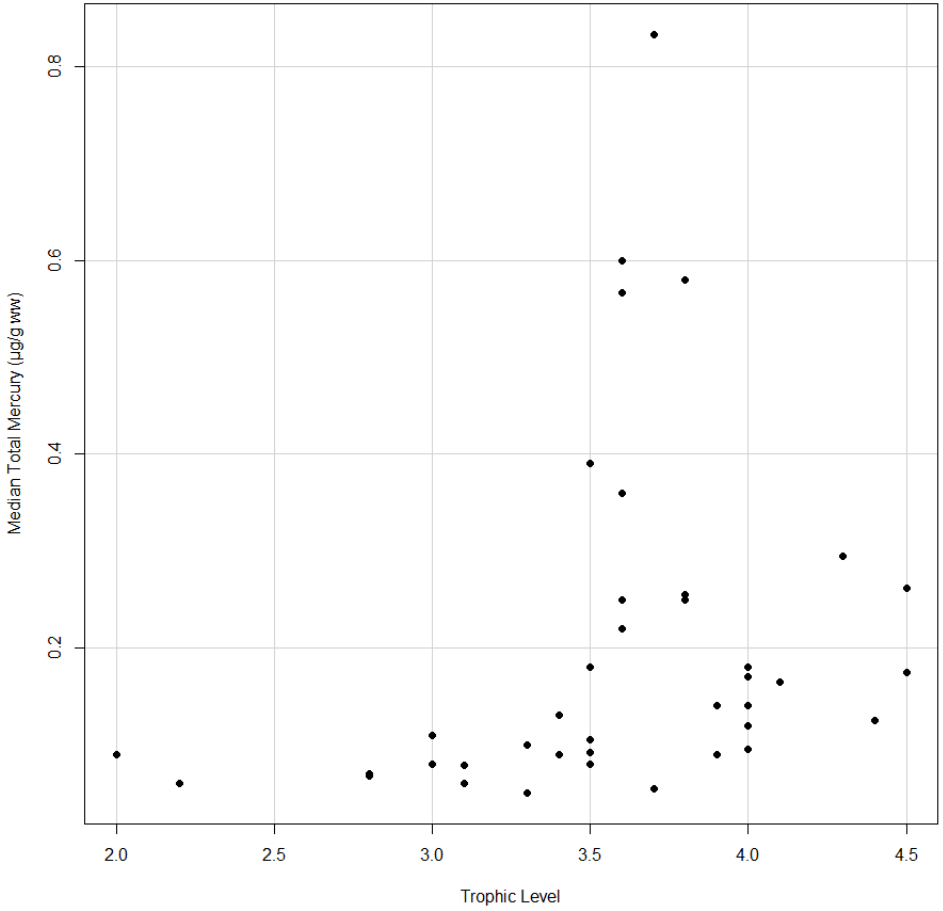
Figure 3. Trophic position (from Fishbase) versus median THg concentration for species in the WNAMS dataset.