Spatial and temporal patterns of mercury concentrations in freshwater fish across the Western United States and Canada

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Abstract

Methylmercury contamination of fish is a global threat to environmental health. Mercury (Hg) monitoring programs are valuable for generating data that can be compiled for spatially broad syntheses to identify emergent ecosystem properties that influence fish Hg bioaccumulation. Fish total Hg (THg) concentrations were evaluated across the Western United States (US) and Canada, a region defined by extreme gradients in habitat structure and water management. A database was compiled with THg concentrations in 96,310 fish that comprised 206 species from 4262 locations, and used to evaluate the spatial distribution of fish THg across the region and effects of species, foraging guilds, habitats, and ecoregions. Areas of elevated THg exposure were identified by developing a relativized estimate of fish mercury concentrations at a watershed scale that accounted for the variability associated with fish species, fish size, and site effects. THg concentrations in fish muscle ranged between 0.001 and 28.4 (μg/g wet weight (ww)) with a geometric mean of 0.17. Overall, 30% of individual fish samples and 17% of means by location exceeded the 0.30 μg/g ww US EPA fish tissue criterion. Fish THg concentrations differed among habitat types, with riverine habitats consistently higher than lacustrine habitats. Importantly, fish THg concentrations were not correlated with sediment THg concentrations at a watershed scale, but were weakly correlated with sediment MeHg concentrations, suggesting that factors influencing MeHg production may be more important than inorganic Hg loading for determining fish MeHg exposure. There was large heterogeneity in fish THg concentrations across the landscape; THg concentrations were generally higher in semi-arid and arid regions such as the Great Basin and Desert Southwest, than in temperate forests. Results suggest that fish mercury exposure is widespread throughout Western US and Canada, and that species, habitat type, and region play an important role in influencing ecological risk of mercury in aquatic ecosystems.

1. Introduction

Mercury (Hg) contamination of aquatic ecosystems contributes to 80% of all fish consumption advisories in the United States and Canada (Environment_Canada, 2015, USEPA, 2011), and it negatively affects the beneficial uses and ecological health of aquatic resources globally (Selin, 2011). The global prevalence of Hg contamination is partially attributable to a 3-to-5 fold increase in atmospheric Hg concentrations over the past 150 years associated with fossil fuel combustion (Driscoll et al., 2013), as well as regional and local releases from mining and industrial applications (Amos et al., 2015, Beal et al., 2015, Eckley et al., 2013, Horowitz et al., 2014). However, the environmental threat posed by Hg is not fully ascribed to inorganic Hg loading. The microbially mediated conversion of inorganic Hg to methylmercury (MeHg) greatly increases its bioavailability and toxicity, as well as its biomagnification potential (Wiener et al., 2003). Methylmercury production is controlled by biogeochemical conditions in the environment that promote activity of the microbial groups that methylate Hg (Gilmour et al., 1992, Gilmour et al., 2013, Hall et al., 2008). Thus, Hg concentrations in fish and other components of aquatic food webs may not reflect background Hg loading if environmental conditions are not conducive to microbial Hg methylation. For example, Hg concentrations in fish and aquatic birds have displayed a range of trends in temporal variation since the 1960s, including increases (Drevnick et al., 2015, Vo et al., 2011), decreases (Champoux et al., 2015, Cross et al., 2015), and more variable and complex patterns, such as trend reversals (Gandhi et al., 2014, Monson, 2009), yet there has been approximately a 2–4 fold decrease in net atmospheric deposition since its peak at that time (Beal et al., 2015). Although more recent deposition trends are variable and even increasing in some cases (Weiss-Penzias et al., 2016-in this issue), the linkage with fish Hg concentrations is thus far equivocal. Although fish may not always reflect inorganic Hg inputs, they are useful and effective indicators of relative methylmercury availability within food webs across the landscape, as well as of toxicological risk to humans, wildlife, and fish themselves.

Application of fish to evaluate landscape-scale spatial and temporal variation of Hg availability within food webs is an effective bioassessment tool for scientists and resource managers. However,

interpretations and proper assessments can be complicated by the inherent variability in fish Hg concentrations associated with fish species and foraging guild differences, fish size, and tissue analyzed (Peterson et al., 2007, Walters et al., 2010). Differences in fish community composition among habitats and regions further confound efforts to make robust comparisons across large geographical scales. Several large-scale efforts have applied a variety of successful approaches for examining landscape variation in fish Hg concentrations in the northeastern US and Canada (Kamman et al., 2005), the Great Lakes region (Monson et al., 2011, Sandheinrich et al., 2011, Wiener et al., 2012), Canada (Depew et al., 2013a, Depew et al., 2013b), and the Canadian Arctic (Chetelat et al., 2015). A consistent conclusion among all of these studies was that even after controlling for sources of inherent variation, such as species and size, fish Hg concentrations showed substantial heterogeneity across the landscape. Additionally, landscape and habitat factors together influence fish Hg concentrations (Drenner et al., 2013, Shanley et al., 2012), thus gradients in those factors are likely important drivers of this heterogeneity.

The western region of North America is an expansive, ecologically diverse area (Eagles-Smith et al., 2016-in this issue) occupying 8.9 million km^2 and comprising 10 different level 1 ecoregions (Fig. S1). Major physiographic divisions include the Pacific Mountains and Valleys, Intermontane Basins and Plateaus, the Rocky Mountain System along the continental divide, and the Interior Plains. The ecological diversity is largely characterized by the broad gradient in precipitation associated with these physiographic divisions, with extremes ranging from average annual precipitation of < 13 cm in the arid southwestern deserts to > 254 cm in the Pacific coastal temperate rainforests. The climatological gradient and abundant large river systems of the region have also facilitated extensive modification and management of the hydrology through the construction of large dam and water transport networks for irrigation, water supply, flood control and hydroelectric power. These modifications have had profound impacts on ecological dynamics of the region (Herbert and Gelwick, 2003, Ligon et al., 1995, Martinez et al., 1994, Richter et al., 1997). Moreover, through their influence on the biogeochemistry of aquatic ecosystems, they could also influence Hg cycling (Kasper et al., 2012, Wang and Zhang, 2013). A unique aspect of western North America with respect to Hg dynamics is that compared to the central and eastern regions, the West is disproportionally impacted by the extensive legacy of gold, silver, and Hg mining activities (Davis et al., 2008, Domagalski, 2001, Hornberger et al., 1999, Rytuba, 2000, Singer et al., 2013). Additionally, 60% of the land area in the Western US is publicly owned and managed, and much of the land in Western Canada is Crown Land. Thus, it is valuable to understand the distribution and variability of fish Hg across the West in order to inform public land management agencies of potential risk from Hg contamination to trust resources, as well as to facilitate the development of predictive tools to help manage public resources in a way that reduces Hg threats to human and environmental health.

In this study, 96,310 individual fish THg records were compiled from 4262 unique locations across 15 States, 3 Canadian Provinces, and 2 Canadian Territories in Western US and Canada to evaluate the spatial and temporal variation in fish THg concentrations across the region. The primary goal of this assessment is to describe patterns in fish THg concentrations across the region to facilitate a clearer understanding of how broad ecological and habitat gradients influence THg concentrations. Such insights will ultimately provide a foundation to support a predictive framework for determining the factors that are most important in driving THg concentrations in fish. This assessment did not address marine environments or risk to human and wildlife health, but those assessments are available within this special issue (Ackerman et al., 2016, Davis et al., 2016, Jackson et al., 2016, Lepak et al., 2016).

2. Materials and methods

2.1 Data compilation

Original fish tissue THg concentration data were obtained and compiled from several Federal, State, and Provincial databases (Table S1). Data compilation was largely constrained to locations west of the Continental Divide, including Alaska, Yukon Territories, Northwest Territories, British Columbia, Washington, Oregon, Idaho, California, Nevada, Arizona, New Mexico, Colorado, Utah, Wyoming,

Montana, and Alberta. Additionally, data from Saskatchewan, North Dakota, and South Dakota were included to expand representation of the Great Plains ecoregion.

2.2 Data validation, assumptions, and standardization

Data from each source were examined for completeness and standardized such that all total mercury (THg) concentrations, fish lengths, and fish masses were converted from their reported units to µg/g, cm, and g, respectively. Because complete QA/QC reports were not available with all data sets, a detailed assessment of data quality was not feasible. However, because each agency dataset was originally stored and evaluated relative to Quality Assurance Project Plans (QAPPs), it was assumed that data were of sufficient quality to warrant inclusion in this analysis. Analytical detection limits varied considerably, and ranged from 0.001 to 0.1 µg/g ww among datasets. However, only 91 (0.09%) of the 96,310 data records equaled or were below reported detection limits. Therefore, those 91 values were included as reported and not adjusted further. After each dataset was appropriately standardized, all were merged into a single database, and the merged dataset was examined for gross errors in reported values. All georeferenced information also was standardized to a common datum and verified that all species names were consistent across datasets and followed similar conventions. In some cases there were duplicate fish data held in state and federal databases. Therefore, both automated and manual screenings were conducted to identify duplicate data entries based upon combinations of THg concentrations, species, fish lengths and weights, and sampling locations and dates. 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 standardized. Across datasets fish THg concentrations represented those from both individual fish and composites. For composite samples it was often unclear how many fish each composite represented. Therefore, the 4275 composite samples (4.4% of data) were not weighted differently than individuals, and were simply treated as individual records. Additionally, there were 20,392 data records that did not specify whether they were composite or individual samples, and were thus assumed to be individuals.

Total Hg concentrations in the original dataset were reported as skinless boneless fillet (76.8% of data rows), whole body (19.9% of data rows), or skin-on fillet (3.3% of data rows). All whole body concentrations were converted to skinless boneless fillet equivalents by dividing by 0.74, the average ratio of whole body to muscle concentration from studies where both tissue types were measured on the same individuals (Bevelhimer et al., 1997, Boalt et al., 2014, Goldstein et al., 1996). Skin-on fillet (Depew et al., 2013a) concentrations were not converted because the difference is typically small (< 10%; Dellinger et al., 1995, Zhang et al., 2013). Records of THg concentrations presented on a dry-weight basis were then converted to wet-weight values because 82.1% of the original THg data were reported as wet weight. Conversions from dry-weight to wet-weight concentrations used the original moisture content data where available (31.1% of dry-weight data rows); for the remaining data, dry-weight THg concentrations were converted to wet-weight THg concentrations using the mean tissue-specific moisture content (76%) derived from the 6594 fish that included moisture content measurements. All fish length measurements were standardized to fork length because 54.2% of the fish lengths were reported in fork length, whereas 33.7% and 12.1% of the fish lengths were reported as total length and standard length, respectively. Species-specific length conversion equations from FishBase (Froese and Pauly, 2003) were used to convert all fish length measurements to fork length. Equations from closely related species that had similar morphology were used for species that lacked published length conversion equations. For a small number of fish (< 1% of data) the type of length measurement reported 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.

2.3 Spatial aggregation and GIS data layers

Geographic coordinate information associated with each data record was entered into a geographic information system (ArcGIS v10, ESRI) to validate and standardize site information. Near analyses were

performed for each sampling location to identify the closest flowline or water body feature in selected datasets, including USGS National Hydrography High Resolution Dataset (NHDHighRes; (USGS, 2014)) and Canadian National Hydro Network (NHN; (NRC, 2015)). Sites > 0.15 km from a feature were investigated and eliminated if site names did not match nearby water features. For lentic systems all fish locations were aggregated to the centroid of the water body so that each data row represented a site replicate, whereas for lotic systems fish locations were aggregated such that samples in the same water body within 10 lineal river km of each other were categorized as originating from the same location. The resulting dataset included a total of 96,310 individual records for fish THg concentrations comprised of 206 different species sampled between 1969 and 2014 from a total of 4262 unique sites. Point intersects were conducted for each sample location with relevant vector and raster datasets describing the geospatial setting of the site. Sites were assigned to one of four coarse habitats (lakes, pond and reservoirs; rivers and streams; wetlands, and canals) by cross checking the habitats designations in NHDHighRes, NHN, and the US National Wetlands Inventory (USFWS, 2014). Where habitats differed among these databases, satellite imagery was visually inspected for each site and appropriate designations were chosen based upon that visual assessment. Point intersects were then used to assign each site to a hydrologic unit code (HUC) to make more robust regional assessments that are less reliant on individual location data. Categorization was done at the HUC-8 scale because HUC-8 equivalents exist for both the US and Canada, allowing for HUC-based comparisons across both countries.

2.4 Length standardizing fish THg concentrations

Total Hg concentrations are often highly correlated with fish length (Scudder Eikenberry et al., 2015), and fish sizes in this dataset spanned a substantial range across and within species (Table S2). To facilitate spatial and temporal comparisons of fish THg concentrations, the THg concentrations of each fish were size-standardized to the median length of each respective species (Table S2). For each species group, a linear mixed-effect model was constructed with fork length as a fixed covariate, and site, species, and a species × fork length interaction as random effects to predict the THg concentration of each individual fish at the median fork length of their species. The residuals from the model were then added back to the predicted value to calculate the standardized THg concentration for each individual fish (Eagles-Smith and Ackerman, 2014). Total Hg concentrations were not length standardized for species where a likelihood ratio test suggested that the inclusion of length did not improve the model. In those 72 cases (Table S2) the raw, unadjusted THg concentrations were used in the models. For some species, the ability to size standardize THg concentrations was limited by sparse representation in the datasets. Therefore, related species with similar ecology and physiology (e.g. suckers) were grouped into aggregate species groups for size-correction (Table S2). However, THg concentrations were still standardized at the median length for each species, rather than the median length for a species group. Additionally, the inclusion of species and species × FL interactions as random effects accounted for variation among the species within a species group while pooling variance among species within a group to allow estimation of the length-THg relationship in underrepresented species.

Table 1. Distribution of Hydrologic Unit Codes (HUCs; level 8 equivalents) within Level 1 Ecoregions of western North America, and proportion of HUCs for which their least square mean fish THg concentrations fell within each data percentile category.

2.2 Statistical Analyses

A tiered statistical approach was applied to evaluate the spatial and temporal variability in fish THg concentrations, and to assess differences across broad habitat classifications. The first tier of analysis was descriptive, to illustrate the variation and distribution of fish THg concentrations from the raw data, without accounting for site, species, or temporal effects. In this initial analysis site-specific geometric mean THg concentrations were calculated for each of the 4262 sites. The geometric mean concentrations for each site included all species within a site, and did not include adjustments of any THg concentrations for fish size. An important caveat to this first tier analysis is that the results do not necessarily reflect the current state of exposure across the landscape because the data span more than a 45-year time period. Therefore, all subsequent statistical models include year as a factor in order to account for temporal variability, and a separate temporal analysis was conducted to investigate variation in fish THg concentrations over time.

In the second tier analysis linear mixed-effects models with size-standardized THg concentrations were used to evaluate differences among fish species, as well as the effect of habitat type and foraging guild on fish THg concentrations. The first model was constrained to include only those fish species with a sample size of at least 100 individuals to ensure a more robust interspecies comparison. Natural log-transformed length-standardized THg concentration was the dependent variable, species and habitat were fixed effects, and site and year were included as random effects. For the second model, each species was assigned to one of 5 different foraging guilds (piscivore [diet predominantly composed of fish], generalist [diet composed of both fish and invertebrate prey], generalist invertivore [diet composed of both benthic and planktonic invertebrates], benthivore [specialized foraging on benthic invertebrates], and planktivore [specialized foraging on planktonic invertebrates]; Table S2). Some species exhibit ontogenetic shifts in their feeding ecology and in those cases size thresholds at which fish most likely switch to a different guild were estimated using Mittlebach and Persson (1998) and FishBase (Froese and Pauly, 2003). Differences in fish THg concentration among four coarse habitat types (lakes, ponds, and reservoirs; rivers and streams; wetlands; and canals) were also assessed. The statistical model included habitat and foraging guild as fixed main factors, and species, site, and year as random effects. A habitat × foraging guild interaction was also included to assess whether relative habitat differences in fish THg concentrations differed among foraging guilds. For this test, records for any samples that lacked a measured fish length or defined habitat category were excluded (N = 18,559).

In the third tier analysis, linear mixed-effects models were used to develop relativized estimates of fish THg concentrations for comparison at the watershed scale. A relativized estimate of fish THg concentrations is defined here as the least squares mean fish THg concentration that statistically accounts for the effects of fish species and length. Thus, it allows for robust spatial comparisons of THg concentrations even when comparing among locations with different fish species. As such, it is reflective of the relative availability of mercury to the general fish community as opposed to an actual concentration within a given species. The relativized estimates were used to evaluate differences in fish THg concentrations among watersheds and along ecological gradients. Each site was categorized into their respective watersheds at the HUC-8 scale, as well as into level 1 and level 2 ecoregions (Omernik, 1987). Ecoregions are hierarchical geographical constructs of areas with similarity regarding patterns in the mosaic of biotic, abiotic, aquatic, and terrestrial ecosystem components (Omernik, 2004). Level 1 is the coarsest designation and there are 10 level 1 ecoregions in the Western US and Canada (Fig. S1), whereas level 2 ecoregions represent a finer scale of delineation and are nested within the level 1 ecoregions. The statistical model was constructed with ecoregion and hydrologic unit (nested within ecoregions) as fixed effects, and species, site, and year were statistically accounted for as random effects. In order to identify clusters of watersheds containing least squares mean fish THg concentrations that were higher or lower than would be expected by chance, a Getis-Ord Gi* hotspot analysis (Gettis and Ord, 1992) was conducted in ArcGIS (v10, ESRI). The analysis was conducted using HUC-8 least squares mean fish THg concentrations as the input feature class, and z-scores for each HUC were calculated using the polygoncontiguity (edges and corners) spatial relationship function.

The fourth tier of analysis focused on differences in size-standardized fish THg concentrations among habitats across ecoregions. The linear mixed effects model included level 1 ecoregion and habitat type as fixed effects; and species, site, and year as random effects; and a habitat × ecoregion interaction. Because wetlands and canal habitats were not represented in every ecoregion in our dataset, the analysis was constrained to include only data from lentic and lotic systems.

Finally, temporal variability in length-standardized fish THg concentrations was assessed with two approaches. The first incorporated all of the data into a single linear mixed effects model with year as a fixed categorical effect, and with species and site as random effects. The second evaluated temporal patterns separately in each ecoregion by running the same model as above in an ecoregion-specific analyses.

Unless otherwise specified, all THg concentrations were natural log 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 with the Delta method (Seber, 1982).

3. Results and discussion

3.1 Descriptive spatial distribution.

Across all fish species, the geometric mean THg concentration (± standard error) of 96,310 fish samples from 4262 unique locations was 0.170 ± 0.001 μg/g ww, and individual concentrations ranged from 0.001 to 28.54 μg/g ww. Thirty percent of fish muscle samples exceeded the US EPA Fish Tissue Residue Criterion for methylmercury (0.30 μg/g ww), which was established to protect the health of humans who eat noncommercial fish (Borum et al., 2001) and 3.9% exceeded the US Food and Drug Administration action level of 1.0 μg/g ww. Thirty-four percent of whole body samples exceeded the estimated threshold of 0.20 μg/g ww associated with potential impairment in fish (Beckvar et al., 2005). Although informative, an important caveat to these exceedence percentages is that sample sizes were unbalanced across locations and likely biased towards sites with higher mercury concentrations. Thus, these summary values are not unlikely to reflect the overall contamination across the landscape based upon a random sampling approach.

Site-specific geometric mean THg concentrations for all sites ranged from 0.006 to 2.98 μg/g ww, with a mean across all sites of 0.124 ± 0.002 μg/g ww (Fig. 1). Geometric mean muscle THg concentrations exceeded the US EPA Tissue Residue Criterion (0.30 μg/g ww) at 17% of sites, and site-specific whole body geometric mean THg concentrations exceeded the 0.20 μg/g ww fish health threshold at 20% of sites. Most sites were represented by numerous fish THg measurements, but some locations had smaller sample sizes. A second map is provided with only those sites that contained $n > 3$ fish to display only those sites where a mean and variance could be estimated (Fig. S2). Although geographic areas with aggregations of sites containing either elevated or low fish THg concentrations can be inferred from these maps, it is important to recognize that the raw summary statistics across taxa do not account for variation due to species, fish size, habitat, or year effects. Additionally, data from different fish species were unevenly distributed throughout the western US and Canada (Fig. S3). Thus, although it is valuable to examine the spatial distribution of the site-specific geometric mean fish THg concentrations, this approach is not appropriately representative of Hg availability to the fish communities across the western US and Canada.

Figure 1. Site-specific geometric mean fish total mercury (THg) concentrations (μg/g wet weight) across Western US and Canada. Concentrations represent geometric mean THg in muscle tissue across all fish species and years for each site. Data are not adjusted for fish size. Sample sizes varied considerably by location. See Fig. S2 for distribution of data from sites with at least 3 individuals.

3.2 Taxonomic effects

Fish species is a particularly important determinant of THg concentrations because of inter-specific differences in variables that influence bioaccumulation, such as trophic position (Lavoie et al., 2013), foraging habitat (Eagles-Smith et al., 2008a, Willacker et al., 2013), and bioenergetics (Lepak et al., 2012, Trudel and Rasmussen, 2006). Taxonomic variation in THg concentrations was assessed for a total of 206 fish species by aggregating ecologically, morphologically, and evolutionarily similar species into 32 different taxonomic groupings (Table S2), representing 5 distinct foraging guilds as described in *Methods*. Across the 32 taxonomic groups, unadjusted median THg concentrations ranged from 0.032 µg/g ww in cichlids to 0.497 µg/g ww in lamprey (Fig. S4). Among groups, unadjusted median muscle THg concentrations were at or above the estimated threshold associated with fish health impairment (0.20 µg/g ww) in goldeye (0.41 µg/g), gar (0.38 µg/g), walleye/sauger (0.32 µg/g), Morone bass (0.30 µg/g), black basses (0.29 µg/g), pike (0.24 µg/g), crappie (0.20 µg/g), and char (0.20 µg/g; Fig. S4). In addition to cichlids, fish groups with the lowest unadjusted median THg concentrations included sculpin (0.06 µg/g), anadromous salmonids (0.06 µg/g), Arctic grayling (0.07 µg/g), shad (0.075 µg/g), whitefish (0.075 µg/g), and minnows (0.078 µg/g; Fig. S4). However, THg concentrations varied substantially within many of the taxonomic groupings (Fig. S4). In fact, the lowest and highest THg concentrations differed by 100-fold or more in 24 of the 32 taxonomic groupings. Only gar had less than a 10-fold difference between the highest and lowest concentrations, but the dataset for gar was limited to 11 individuals from 5 sites. Silversides, stickleback, cichlids, Arctic grayling, freshwater drum, shad, and cisco had between a 25- and 61-fold difference between their lowest and highest THg concentrations. The difference between the highest and lowest THg concentrations spanned 4 orders of magnitude (~ 1000-fold) in Morone bass, black basses, minnows, trout, carp, walleye/sauger, whitefish, pike, char, killifish, and suckers. This substantial variation in unadjusted THg concentrations within taxonomic groups illustrates the substantial importance of spatial factors and fish size on THg concentrations in fish.

Total Hg concentrations differed among species after adjusting fish THg concentrations to standardized lengths for each species and statistically accounting for effects of habitat, site, and year (analysis constrained to species with $n > 100$; $F_{52,78,554} = 794.10$; $p < 0.0001$). Across all 53 species, least squares mean THg concentrations ranged 10.3-fold from 0.048 ± 0.001 µg/g ww to 0.493 ± 0.043 µg/g ww. Concentrations were highest in Sauger, Northern Pikeminnow, Walleye, White Bass, Striped Bass, Northern Pike, Lake Trout, Smallmouth Bass, Largemouth Bass, and Whiterock Bass (Fig. 2), and lowest in Broad Whitefish, Pumpkinseed, Dolly Varden, Mountain Whitefish, Tui Chub, Brook Trout, Coho Salmon, Redside Shiner, Rainbow Trout, and Slimy Sculpin (Fig. 2). In general, the species with the highest THg concentrations were piscivores, whereas those with lower concentrations were a mix of planktivores, invertivores, and generalists.

Figure 2. Least squares mean, size standardized muscle tissue total mercury (THg) concentrations (µg/g wet weight) in fish species across western US and Canada. Data represent species with a total sample size of > 100 individuals. Error bars represent 1 standard error. Least squares mean concentrations represent the mean THg concentration in each species after controlling for site and year effects. Fish THg concentrations were standardized to the respective median length for each species, shown in parentheses. ▯ indicates there weren't length-THg relationships and THg concentrations are not size-standardized.

3.3 Guild and habitat effects

Size-standardized fish THg concentrations differed among foraging guilds ($F_{4,477.7} = 4.13$, $p = 0.0027$) and among habitats ($F_{3,4866} = 28.91$, $p < 0.0001$), but the guild × habitat interaction ($F_{12,48,874} = 19.61$, $p < 0.0001$) indicated that habitat differences were not consistent among guilds after controlling for species, site, and year. Total Hg concentrations were higher in riverine habitats than in lakes for each of the five guilds ($F_{4,141.3} = 18.24$, $p < 0.0001$), but the relative concentrations in wetlands and canals were variable and showed no consistent patterns among guilds (Fig. 3). The magnitude of difference between lakes and riverine habitats ranged from 21% in piscivores to 61% in benthivores. Total Hg concentrations in piscivores were higher than the other 4 guilds in both lake ($p < 0.0001$) and riverine habitats ($p < 0.0001$). In lakes, piscivore least-squares mean THg concentrations were 2.2, 2.1, 2.4, and 3 times higher than those in benthivores, generalists, generalist invertivores, and planktivores, respectively. In riverine habitats,

piscivores were 1.6, 2, 2.4, and 2.9 times times higher than benthivores, generalists, generalist invertivores, and planktivores, respectively (Fig. 3). There were inconsistent patterns among habitats for the other guilds. Generalists had slightly higher concentrations than generalist invertivores across habitats, but the ifferences were not statistically significant. Planktivores had the lowest THg concentrations of all the guilds in canal, lakes, and riverine habitats, but were similar to piscivores in wetland and canal habitats (Fig. 3).

Figure 3. Fish muscle total mercury (THg) concentrations (µg/g wet weight) differ among habitat types and foraging guilds. Symbols represent size adjusted least squares (LS) mean THg concentrations controlling for the effects of species, site, and year. Error bars are one standard error.

Mercury concentrations in fish are known to differ among habitats (Ackerman and Eagles-Smith, 2010, Eagles-Smith and Ackerman, 2014, Eagles-Smith et al., 2008a) because of habitat-specific variability in biogeochemical drivers of Hg cycling and methylation (Heim et al., 2007, Marvin-Di Pasquale and Agee, 2003), as well differences in food web structure (Eagles-Smith et al., 2008b, Kidd et al., 1999, Swanson et al., 2006). However, few studies have conducted broad geographic comparisons in fish THg concentrations among coarse habitat designations. A comparison among waterbody types for 13 species in northeastern North America found that THg concentrations in White Sucker, Yellow Perch, and Largemouth Bass were higher in rivers than in lakes and/or reservoirs, whereas, four species of fish had higher THg concentrations in lakes or reservoirs than in riverine habitats, and six fish species showed no differences among waterbody types (Kamman et al., 2005). In the Great Lakes area of North America, THg concentrations in Largemouth Bass and Walleye were 10.6%–24.1% and 7.9%–10.7% lower, respectively, in riverine waterbodies than in lakes (Monson et al., 2011). The studies described above employed species-specific models for testing differences between habitat types, whereas a global model was used in this study that accounted for the effect of species in its parameter estimates. This approach evaluates the overall effect of habitats on fish THg across all taxa using pooled variance of the entire dataset. There are likely species-specific deviances from this global trend, but the goal of this analysis was to assess these differences across the entire fish assemblage to evaluate overall differences in Hg availability to the fish community. Additionally, natural lakes and reservoirs were not differentiated in this test because it was specifically focused on differences between lotic and lentic environments. However, fish THg concentrations are 3%–160% (mean = 44%) higher in reservoirs than in natural lakes in the western US and Canada (Willacker et al., 2016-in this issue). Thus, differences between rivers and reservoirs may be less pronounced, whereas those between rivers and natural lakes may be higher than what we detected with pooled data for reservoirs and lakes.

The mechanisms that may be driving the differences in fish THg concentrations between lakes and rivers are unclear, but factors could include differences in bioenergetic costs associated with residing in higher energy habitats (Crook and Robertson, 1999, Facey and Grossman, 1992, Tyler and Gilliam, 1995) higher rates of microbial MeHg production, more efficient entry of MeHg into the base of periphyton driven food webs relative to pelagic-based food webs (Cleckner et al., 1999, Jardine et al., 2012), or generally higher aqueous MeHg concentrations in riverine systems. Additionally, dams are a ubquitous feature across the western landscape, and many rivers in the western US contain multple dams or receive water from dammed tributaries. Fish THg concentrations are higher in reservoirs than natural lakes in the West, due in part to water management effects on MeHg production (Willacker et al., 2016-in this issue). Similarly, rivers downstream of dams may also receive elevated aqueous MeHg that can be rapidly incorporated into the base of riverine food webs. In fact, elevated MeHg signals have been detected as far as 250 km downstream from dams (Kasper et al., 2014, Schetagne et al., 2000).

3.4 Spatial variation in fish THg concentrations

After statistically accounting for species, site, and year effects, the spatial patterns of size-standardized fish THg concentrations across the landscape (Fig. 4; S5a–b) differed substantially from the descriptive analysis of geometric mean THg concentrations using non-standardized concentrations (Fig. S4). These differences were anticipated and highlight the importance of developing relativized estimates of fish THg concentrations when making spatial or temporal comparisons because sampling methods and varying fish assemblages can result in biased estimates of risk across the landscape. For example, comparing a sample of large piscivorous fishes from one area to a sample of smaller or lower trophic level fishes from another area would spuriously suggest that risk is higher in the area with piscivore samples even if mercury availability to the fish community is identical between the two areas. By size-standardizing each fish sample, and statistically accounting for the influence of species, this model allowed for making robust comparisons

of relative fish THg concentrations among locations. Another approach that can result in robust spatiotemporal comparisons and reduces model uncertainties is to constrain the dataset to only compare across locations with a single fish species (Monson et al., 2011). However, the habitat mosaic across western North America is sufficiently diverse and complex that few species are broadly distributed across multiple habitat types throughout the region (Fig. S2). Alternatively, derivation of a common indicator species can be accomplished by applying a statistical model that converts THg concentrations from multiple species into a single species with significant wildlife or human health implications (Depew et al., 2012), that can be applied across space and time (Depew et al., 2013b, Wente, 2004). Although effective, this approach carries a similar drawback as the single species comparisons across diverse ecological gradients in that there are few, if any, species that occur broadly across the expansive sub-continental-scale herein. Thus, the model would develop an estimate of fish THg concentrations for species that do not inhabit certain areas. From a strictly comparative perspective this is an unimportant drawback because the approach still provides a common indicator for making spatial comparisons across sites and through time. The approach in the present study is similar in that a linear model was used to account for the influence of species, but done within the framework of a mixed effects global model with species as a random effect, pooling variance across the entire data set and yielding relativized THg concentrations across species rather than selecting a reference species for comparison across the fish community.

Figure 4. Relativized total mercury (THg) concentrations (binned by quintiles of the data distribution) in fish across the western United States and Canada. Each bounded polygon represents a hydrologic unit at the HUC-8 scale. The categories represent the percentile of least squares mean THg concentration relative to the entire dataset, such that 20% of the data distribution occurs in each HUC. The least squares mean THg concentrations in the lowest category (0–20th percentile) ranged between 0.011 and 0.074 µg/g ww. The least squares mean THg concentrations in the 20th–40th percentile ranged between 0.075 and 0.126 µg/g ww. The least squares mean THg concentrations in the 40th–60th percentile category ranged between 0.127 and 0.178 µg/g ww. The least squares mean THg concentrations in the 60th–80th percentile category ranged between 0.179 and 0.248 µg/g ww, and the least squares mean THg concentrations in the highest category (80th–100th percentile) ranged between 0.249 and 1.45 µg/g ww. Least squares mean THg concentrations were estimated from a linear mixed effects model with level 1 ecoregion and HUC-8 (nested within ecoregion) as fixed effects, and species, site, and year as random effects. HUC-specific sample size and coefficient of variation are presented in Fig. S5.

The differences in model estimated fish THg concentrations among HUCs ($F_{1029,2377} = 2.16$, $p < 0.0001$) illustrates the spatial heterogeneity of fish THg concentrations across the western US and Canada. The least squares mean THg concentrations for fish muscle from individual HUCs ranged between 0.011 and 1.45 µg/g ww. To better illustrate the spatial distribution in HU*C*-specific fish THg concentrations that account for the effects of parameters listed above, each HUC was classified into 20th percentile categories (quintiles) such that each category contained 20% of the distribution in THg concentrations. The lowest category (0–20th percentile) contained 176 HUCs with least squares mean THg concentrations ranging between 0.011 and 0.074 µg/g ww. The 20th–40th percentile category contained 342 HUCs ranging between 0.075 and 0.126 µg/g ww. The 40th–60th percentile category contained 193 HUCs ranging between 0.127 and 0.178 µg/g ww. The 60th–80th percentile category contained 116 HUCs ranging between 0.179 and 0.248 µg/g ww, and the highest category (80th–100th percentile) was comprised of 62 HUCS ranging between 0.249 and 1.45 µg/g ww (Fig. 4).

There were no consistent latitudinal or longitudinal geospatial patterns in least squares mean fish THg concentrations across the western US and Canada, but Getis-Ord Gi* analysis identified numerous distinct areas of high and low clustering and spatial autocorrelation (Fig. 5). Areas with the largest aggregations of HUCs containing least squares mean THg concentrations that were higher than predicted by chance include the Great Basin (northern Nevada, southeastern Oregon, and southwestern Idaho), San Francisco Bay-Delta Area and California Coast Range, central Saskatchewan, central Arizona, northeastern New Mexico, and central Wyoming, as well as several other HUCs scattered throughout the region (Fig. 5). Clusters of HUCs with lower THg concentrations than expected by chance were broadly distributed, and included southeastern California, central and eastern Washington and southern and north-central British Columbia, northern Saskatchewan, northwestern Alberta, and northern Yukon and Northwest Territories (Fig. 5). The substantial heterogeneity in fish THg concentrations across the landscape is

consistent with findings from other regions of North America; for example, large variation among individual water bodies was evident in the analyses of fish THg levels across the Great Lakes region (Sandheinrich et al., 2011), the northeastern US and eastern Canada (Kamman et al., 2005), and across Canada (Depew et al., 2013b). The variation in fish THg concentrations among water bodies elsewhere has been attributed to a number of catchment and lake-specific parameters, including wetland density (Burns et al., 2014, Burns et al., 2012), coniferous forest cover (Drenner et al., 2013, Eagles-Smith et al., 2016), pH (Clayden et al., 2014, Jardine et al., 2013), dissolved organic carbon concentrations (Driscoll et al., 1995, French et al., 2014, Rolfhus et al., 2011), and primary productivity (Chen and Folt, 2005), that influence the production of MeHg or its concentration at the base of aquatic food webs.

Figure 5. Analysis of fish total mercury concentration "hotspots" and "coldspots" at the HUC-8 watershed scale across western North America. Watersheds shaded red and blue represent least squares mean fish THg concentrations that are higher or lower, respectively, than expected by chance alone based upon adjacent watersheds. Different shades of red and blue represents different levels of statistical confidence. Spatial autocorrelation and statistical significance was determined using the Getis-Ord Gi statistic.

Site effects commonly account for much of the total variance in fish THg concentrations across broad geographical extents (Depew et al., 2013b, Kamman et al., 2005). Site accounted for 32% of the total variance in this study, exceeded only by species effects, which accounted for 40% of the variance in the data. The importance of site is related to the physicochemical and biogeochemical factors that characterize ecological processes and influence Hg transport, availability, and MeHg production. Food web structure and trophic transfer rates exert additional influence on fish THg concentrations, and also can vary substantially among sites. A major characteristic of the western North American landscape is its heterogeneity and the extreme gradients in attributes such as precipitation, vegetation structure, the abundance and cycling of organic carbon, and ecosystem processes that affect MeHg production and its concentrations at the base of aquatic food webs. Heterogeneity in these components and processes across the landscape likely results in similar variation in fish THg concentrations. This is evident by the fact that not only was there substantial variation at the HUC scale, but also among sites within HUCs as illustrated by the high coefficient of variation estimates for each HUC (Fig. S5b), even those with large sample sizes (Fig. S5a). Thus, waterbodies in relatively close proximity with one another often have fish with substantially different THg concentrations, associated with differences in site-specific characteristics.

Not only is western North America characterized by gradients in landscape scale factors that influence MeHg production, but it also contains considerable gradients in the type and magnitude of Hg sources. Wet deposition of atmospheric Hg is a major pathway for entry of Hg to watersheds (Selin et al., 2007), and because wet deposition largely mirrors precipitation gradients, deposition rates vary substantially across the west, with the Coastal and Cascade Ranges of the Pacific Northwest substantially higher than in the more arid portions of the region (National Atmospheric Deposition Program Mercury Deposition Network; http://nadp.isws.illinois.edu) where dry deposition may be of greater importance (Lyman et al., 2007). The west differs from the eastern portion of the continent in that it contains substantial geologic deposits of Hg. Historic Hg mining activities and the widespread legacy use of Hg in gold and silver mining operations released inorganic Hg to waterbodies throughout the West (Alpers et al., 2005), resulting in THg concentrations up to 303,255 μg/kg dw in surficial sediment, with a median of 91 μg/kg (Fleck et al., 2016). If mining-derived Hg contamination contributes substantially to MeHg exposure in fish, then the layering of these complex and diverse Hg sources with factors that influence MeHg production can complicate the interpretation of landscape-scale drivers of fish THg concentrations in the western US and Canada. Although some of the areas where we found elevated fish THg concentrations overlapped with regions of known mining influences, the distribution of mining impacts across the west is widespread and its overall importance on fish THg concentrations is unclear, and likely a matter of scale. Mining clearly can influence fish THg concentrations at localized watershed scales (Alpers et al., 2016-in this issue), but extrapolating to the subcontinental scale is less well defined. As a coarse assessment of the linkage between background Hg contamination and fish THg concentrations, the extent to which THg and MeHg concentrations in sediment were related to fish THg concentrations at the HUC-8 scale was assessed. Least squares mean concentrations of THg in sediment were not correlated with paired least squares mean fish THg concentrations at the HUC-8 scale ($p = 0.07$, $N = 418$; Fig. 6A), whereas least squares mean sediment MeHg concentrations were weakly correlated with fish THg concentrations ($p = 0.0005$, $N = 216$; Fig. 6B). The contrasting relation between fish THg concentrations and sediment THg and MeHg concentrations suggest that at the watershed scale

across the continental west, background inorganic Hg contamination may have less influence on fish THg concentrations than landscape processes that influence production of MeHg and its entry and bioaccumulation at the base of aquatic food webs. Importantly, the low r^2 value (0.06) of the relation between sediment MeHg and fish THg concentrations at the HUC-8 scale indicates that even sediment MeHg is a poor predictor of THg in fish at this scale. A more robust catchment-scale analysis that simultaneously models the relative importance of a range of factors on fish THg concentrations is needed to assess the key drivers of fish THg across the West.

Figure 6. Correlation between HUC-8 least squares mean fish total mercury (THg) concentrations (µg/g wet weight) and paired HUC-8 least squares mean (A) sediment THg concentrations and (B) sediment MeHg concentrations. Fish least squares mean concentrations were estimated using size standardized fish THg data in a linear mixed-effects model that included Level 1 ecoregion and HUC-8 (nested within ecoregion) as fixed effects, and site, species, and year as random effects. Sediment least squares mean concentrations were estimated from linear mixed effects models that included HUC-8 as a fixed effect, and site as a random effect (Fleck et al., in press).

3.5 Ecoregion differences in fish THg concentrations

Some of the major landscape gradients in western North America are captured in the ecoregion designations, which characterize broad areas of land based upon geographic, climatological, and ecological similarities. As a first-order understanding of the geographic and ecological variation in fish THg concentrations at the HUC-8 scale across the landscape, we evaluated the proportion of HUCs from each percentile category that occurred within each of ten level 1 ecoregions (Table 1). HUCs in the lowest category (0–20th percentile) comprised between 12% (Mediterranean California and Temperate Sierras) and 38% (Tundra) of all HUCs within each ecoregion. HUCs with least squares mean concentrations in the two highest (60th–80th and 80th–100th) percentile groups together accounted for < 20% of HUCs within 5 of the 10 ecoregions, whereas > 20% of HUCs fell in this category for the Great Plains (22% of HUCs), Mediterranean California (24% of HUCs), North American Deserts (25% of HUCs), Southern Semiarid Highlands (67% of HUCs), and Temperate Sierras (47% of HUCs). However, the Southern Semiarid Highland and Temperate Sierra ecoregions were represented by only 3 and 17 HUCs, respectively.

Despite the apparent heterogeneity in HUC-8 based least squares mean fish THg concentrations on the landscape, there were differences among level 1 ($F_{9,2659} = 7.91$, $p < 0.0001$) and level 2 ecoregions ($F_{20,3416} = 15.28$, $p < 0.0001$). Fish THg concentrations in the Southern Semiarid Highlands were higher than all other ecoregions except for the Temperate Sierras (Fig. 7). The Temperate Sierras and North American Deserts had higher least squares mean THg concentrations than the remaining level 1 ecoregions, except Mediterranean California and the Great Plains. Northern Forests, Tundra, and Taiga had the lowest THg concentrations of all the ecoregions. The variability in level 1 ecoregions was also reflected in level 2 ecoregions, though there were substantial differences among level 2 ecoregions within the Great Plains and North American Deserts (Fig. 7). Within the Great Plains, the Temperate Prairie and West-Central Semiarid Prairie Regions had fish THg concentrations that were between 40% and 80% higher than the Boreal Plain and South-Central Semiarid Prairie regions. The two dominant North America Desert regions differed by a factor 2.2, with Western Interior Basins and Ranges (cold deserts) being substantially higher than the Sonoran and Mohave Deserts (warm deserts; Fig. 7).

Figure 7. Least squares mean fish THg concentrations µg/g ww) across Level 1 (top panel) and Level 2 (bottom panel) ecoregions in the western US and Canada. Least squares mean THg concentrations were estimated using length-standardized fish muscle THg concentrations in a linear mixed effect model that included Level 1 and Level 2 (nested within Level 1) ecoregions, and HUC-8 (nested within ecoregion) as fixed effects, and site, species, and year as random effects. Error bars represent 1 standard error. Different letters represent statistical significance (alpha = 0.05) determined using Tukey's pairwise differences.

Next, consistence in habitat differences among ecoregions was tested. Because not all habitats were represented across all ecoregions, this analysis was constrained to a subset of data that included only riverine and lake habitats, which occurred across all level 1 ecoregions. Fish least squares mean THg concentrations again differed among ecoregions ($F_{9,3572} = 5.79$, $p < 0.0001$). There were no main effect differences between lake and riverine habitats ($F_{9,4421} = 1.40$, $p = 0.24$), but the significant habitat × ecoregion interaction

($F_{9,3433} = 3.51$, $p = 0.0002$) indicates that habitat differences varied among ecoregions. Riverine habitats had significantly higher fish THg levels than lake habitats in North American Deserts, Mediterranean California, and Northern Forests, whereas fish THg levels in lakes did not exceed those in riverine habitats in any of the ecoregions (Fig. 8). The highest least squares mean THg concentration in riverine habitats was in the Southern Semiarid Highlands, though it did not differ from any other ecoregion because of the large uncertainty in the least squares mean estimate. Fish THg concentrations were higher in riverine habitats of North American Deserts, Mediterranean California, and Temperate Sierras than those in the Northwest Forested Mountains and Marine West Coast Forests. Riverine habitats in the Tundra, Taiga, and Marine West Coast Forest were lowest of all ecoregions (Fig. 8). Least squares mean fish THg concentrations from lake habitats were highest in Southern Semiarid Highlands, and higher than all ecoregions except the Temperate Sierras (Fig. 8). Lake THg concentrations were lowest in Northern Forests, and were lower than lakes in all other ecoregions except Tundra.

Figure 8. Least squares mean fish muscle total mercury (THg) concentrations (µg/g wet weight) among Level 1 ecoregions for rivers and streams (black circles) and lakes, ponds, and reservoirs (white circles). Least squares mean THg concentrations were estimated using length-standardized THg concentrations in a linear mixed effects model that included habitat and level 1 ecoregion as fixed effects, and site, species, and year as random effects. Error bars represent standard error. □ indicates statistical differences (alpha = 0.05) between lentic and lotic habitats within ecoregions. Note the difference in y-axis scale for Southern Semiarid Highlands relative to the other ecoregions.

Few studies of fish Hg concentrations have contained sufficient geographic breadth to make comparisons across numerous ecoregions. Across Canada, there was substantial variability among ecoregions with an apparent west-east gradient in median fish THg concentrations (Depew et al., 2013b) and the highest normalized concentrations in forested ecoregions. In contrast, this study found no evidence of apparent longitudinal gradients in fish THg concentrations, and forested ecoregions, Tundra, and Taiga generally contained the lowest relativized fish THg concentrations. The discrepancy between the two assessments is not clear, but is likely due in part to the differences in the longitudinal and latitudinal extents and associated climatological and disturbance gradients. The geographic scope of the national Canadian dataset has much greater longitudinal than latitudinal range, and the southern extent is 42°N, with much of the land area above 49°N. Eastern forested regions are known to have elevated MeHg production and fish THg concentrations because of high atmospheric deposition of THg, as well as sulfate, which stimulates microbial MeHg production (Coleman Wasik et al., 2012, Jeremiason et al., 2006) and acidifies poorly buffered waters that are often surrounded by abundant wetlands and with high DOC concentrations (Depew et al., 2013b). In contrast, the western focus of this study results in a more constrained longitudinal extent and an expanded latitudinal range down to the US border with Mexico. This limits the continental influence on atmospheric Hg and sulfate deposition patterns, but incorporates more temperate and arid latitudes with generally lower wet Hg deposition rates and more pervasive water management activities. Specifically, most of the major river systems contain extensive networks of dams and reservoirs with a wide array of management strategies that have the potential to influence MeHg cycling through patterns of wetting and drying (Willacker et al., 2016-in this issue), coupled with altered cycling of organic carbon (Tranvik et al., 2009). Additionally, dry Hg deposition may be important in western arid environments (Wright et al., 2013). These differences have important implications for understanding and managing Hg risks across western landscapes.

3.6 Temporal trends in fish mercury concentrations

After accounting for site, species, and ecoregion there was substantial interannual variation in fish THg concentrations across western North America between 1969 and 2014 ($F_{45,83,490} = 51.11$, $p < 0.0001$; Fig. S6). Least squares mean fish THg concentrations were highest between 1969 and 1977, and declined from 0.278 ± 0.028 µg/g ww in 1969 to 0.155 ± 0.010 µg/g ww in 1977. There were no discernable temporal trends between 1978 and 2012, but concentrations declined substantially in 2013 and 2014 to 0.060 ± 0.008 µg/g ww. However, the 2013 and 2014 data were representative of only a few sites in North American Desert and Northwest Forested Mountain ecoregions, and as a result may not reflect overall patterns across the West.

Although the annual least squares mean fish concentrations account for ecoregion in the model, data did not exist from all years in all ecoregions to test for an interaction between ecoregion and year. Therefore, as a second level temporal analysis, interannual variation in least squares mean fish tissue concentrations was

assessed separately for each ecoregion while accounting for site and species as random effects. Fish THg concentrations differed among years for all ecoregions (Great Plains: $F_{43,19,059} = 26.02$, $p < 0.0001$; Marine West Coast Forests: $F_{39,6836} = 15.05$, $p < 0.0001$; Mediterranean California: $F_{25,3605} = 4.40$, $p < 0.0001$; North American Deserts: $F_{42,6873} = 10.63$, $p < 0.0001$; Northern Forests: $F_{40,21,916} = 32.63$, $p < 0.0001$; Northwestern Forested Mountains: $F_{43,6870} = 13.54$, $p < 0.0001$; Southern Semiarid Highlands: $F_{8,81.1} = 12.85$, $p < 0.0001$; Temperate Sierras: $F_{11,306.1} = 2.82$, $p = 0.002$; Taiga: $F_{43,3996} = 11.69$, $p < 0.0001$; Tundra: $F_{15,114.6} = 2.03$, $p = 0.02$). However, the temporal patterns were not consistent across ecoregions (Fig. 9). Instead, fish THg concentrations showed marked interannual variability with very little directional trend, both over the extent of the time series as well as during the past two decades which contained a substantial proportion of the data. The overall reduction in fish THg concentrations during the early years of the time series is consistent with other long-term data records, and has been attributed to early controls on industrial point-source Hg releases (Wiener et al., 2003). More recently, negative trends in deposition have been reported in the US and Canada over the past two decades, but the data on fish THg concentrations in the West do not match the atmospheric deposition trends. This is consistent with the conceptual model that MeHg production and entry into the food web dominate ecological risk to Hg (Driscoll et al., 2013, Krabbenhoft and Sunderland, 2013), and that sufficient Hg already exists in the environment such that Hg may not be the limiting factor in MeHg production.

Figure 9. Least squares mean total mercury (THg) concentrations (μg/g ww) in size-standardized fish muscle tissue from 10 different level 1 ecoregions across western North America between 1969 and 2014. Least squares mean THg concentrations account for the effects of site and species. Error bars represent standard error.

4. Summary and conclusions

Widespread Hg contamination was evident in the fish communities throughout the Western US and Canada that reflected the broad gradient in Hg availability and cycling in the environment. These results suggest that there is a complicated and diverse suite of factors influencing Hg bioaccumulation, and the relative importance of these factors likely varies across this large geographic region. Further development of predictive estimates of landscape characteristics that influence THg concentrations in fish from western environments would be valuable for better understanding areas at risk and for identifying potential management approaches to mitigate Hg risks in a region dominated by publicly managed lands. The relativized watershed based assessment of fish THg concentrations applied here identified multiple hotspots of Hg contamination that were not always evident based upon the raw data, which did not account for species or size of fish. Additionally, the findings suggest that expanded monitoring of Hg in fish from waterbodies throughout the arid portions of the West could be important for evaluating risk to Hg in these sensitive environments. Finally, the difference between THg concentrations in riverine and lake habitats suggests that water management may play an important role in Hg cycling across the West. Quantifying a mechanistic understanding of those relationships will be important for addressing Hg contamination issues in the future.

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Appendix A. Supplementary data

Figure 1

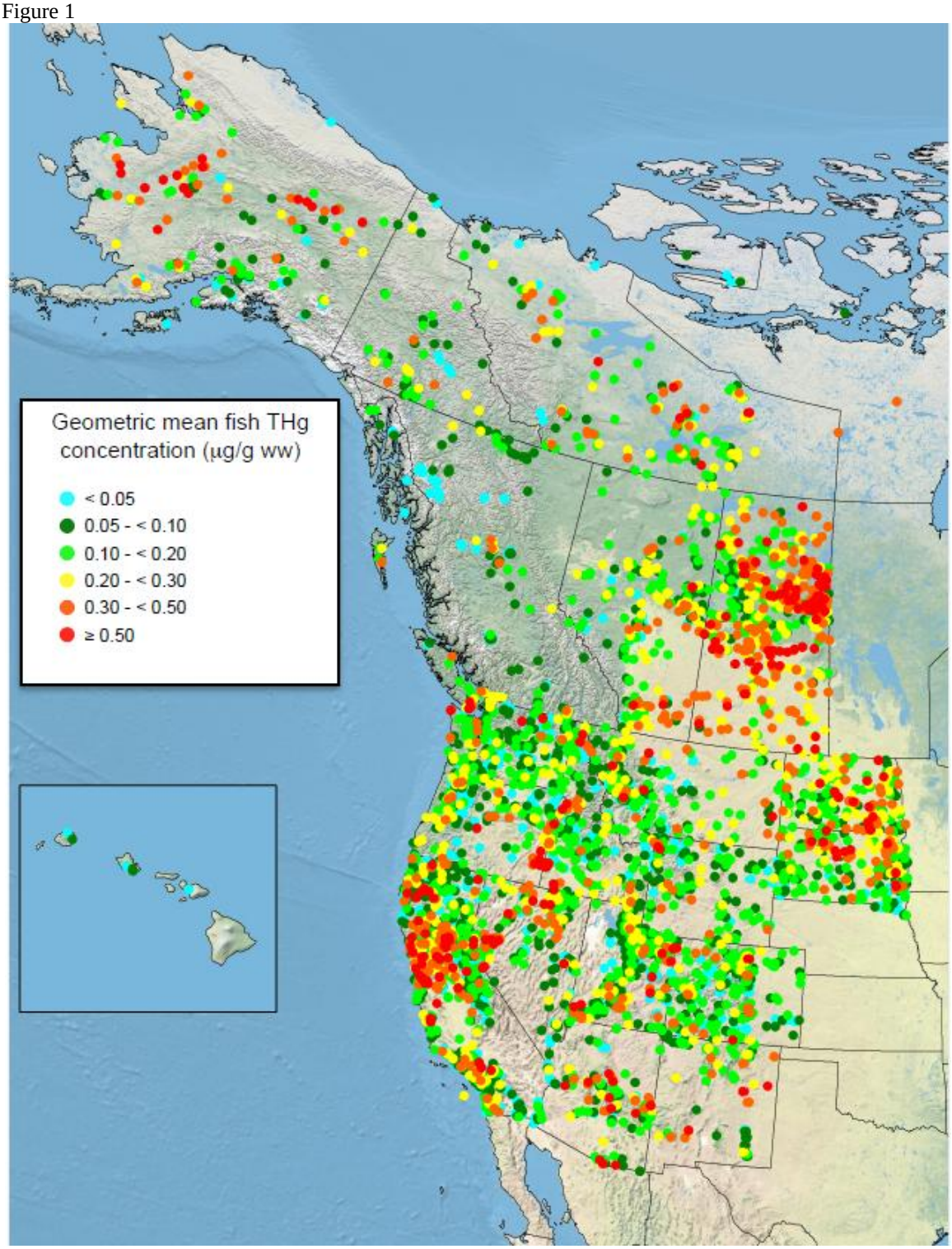

Geometric mean fish THg concentration (µg/g ww)
< 0.05
0.05 - < 0.10
0.10 - < 0.20
0.20 - < 0.30
0.30 - < 0.50
≥ 0.50

Figure 2

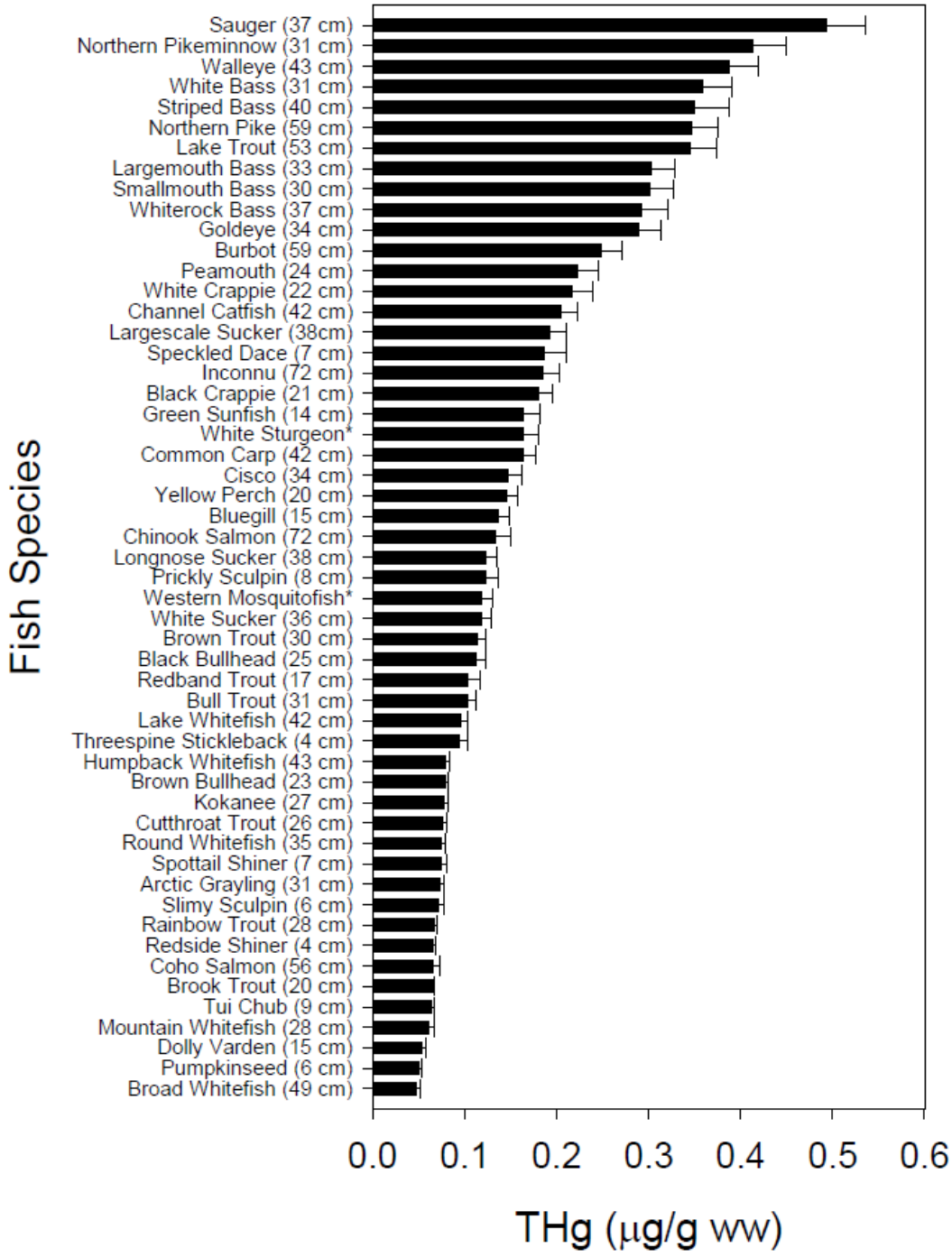

Sauger (37 cm)
Northern Pikeminnow (31 cm)
Walleye (43 cm)
White Bass (31 cm)
Striped Bass (40 cm)
Northern Pike (59 cm)
Lake Trout (53 cm)
Largemouth Bass (33 cm)
Smallmouth Bass (30 cm)
Whiterock Bass (37 cm)
Goldeye (34 cm)
Burbot (59 cm)
Peamouth (24 cm)
White Crappie (22 cm)
Channel Catfish (42 cm)
Largescale Sucker (38cm)
Speckled Dace (7 cm)
Inconnu (72 cm)
Black Crappie (21 cm)
Green Sunfish (14 cm)
White Sturgeon*
Common Carp (42 cm)
Cisco (34 cm)
Yellow Perch (20 cm)
Bluegill (15 cm)
Chinook Salmon (72 cm)
Longnose Sucker (38 cm)
Prickly Sculpin (8 cm)
Western Mosquitofish*
White Sucker (36 cm)
Brown Trout (30 cm)
Black Bullhead (25 cm)
Redband Trout (17 cm)
Bull Trout (31 cm)
Lake Whitefish (42 cm)
Threespine Stickleback (4 cm)
Humpback Whitefish (43 cm)
Brown Bullhead (23 cm)
Kokanee (27 cm)
Cutthroat Trout (26 cm)
Round Whitefish (35 cm)
Spottail Shiner (7 cm)
Arctic Grayling (31 cm)
Slimy Sculpin (6 cm)
Rainbow Trout (28 cm)
Redside Shiner (4 cm)
Coho Salmon (56 cm)
Brook Trout (20 cm)
Tui Chub (9 cm)
Mountain Whitefish (28 cm)
Dolly Varden (15 cm)
Pumpkinseed (6 cm)
Broad Whitefish (49 cm)
Fish Species
0.0
0.1
0.2
0.3
0.4
0.5
0.6
THg (µg/g ww)

Figure 3

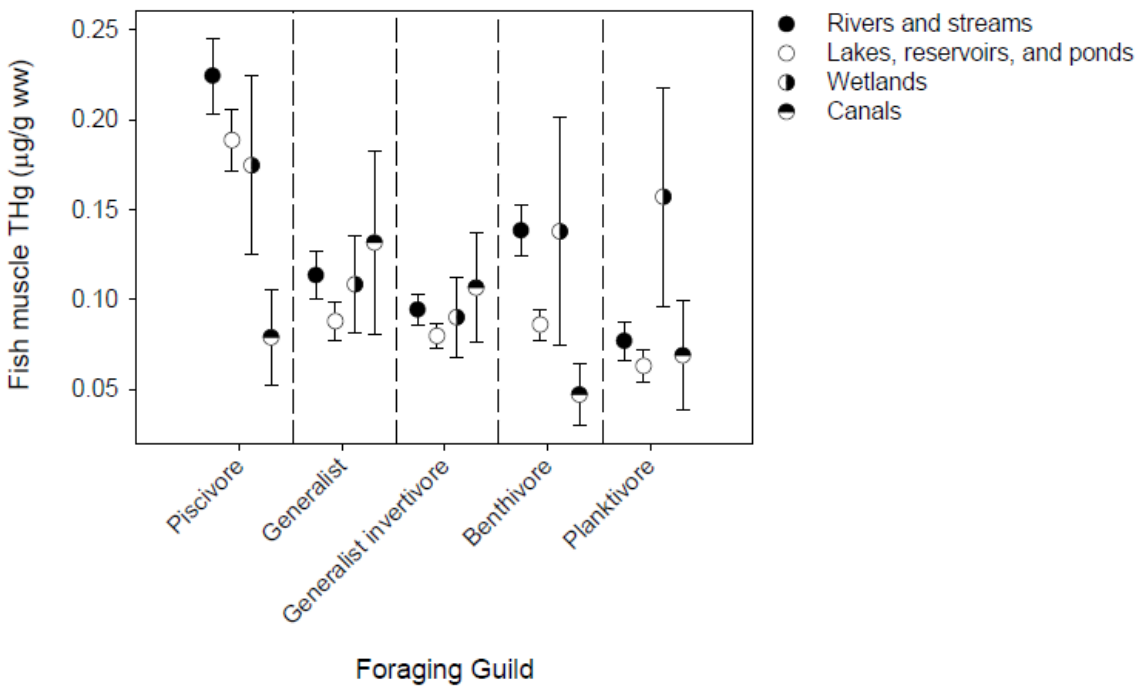

Fish muscle THg (µg/g ww)
0.25
0.20
0.15
0.10
0.05
Piscivore
Generalist
Generalist invertivore
Benthivore
Planktivore
Foraging Guild
Rivers and streams
Lakes, reservoirs, and ponds
Wetlands
Canals

Figure 4

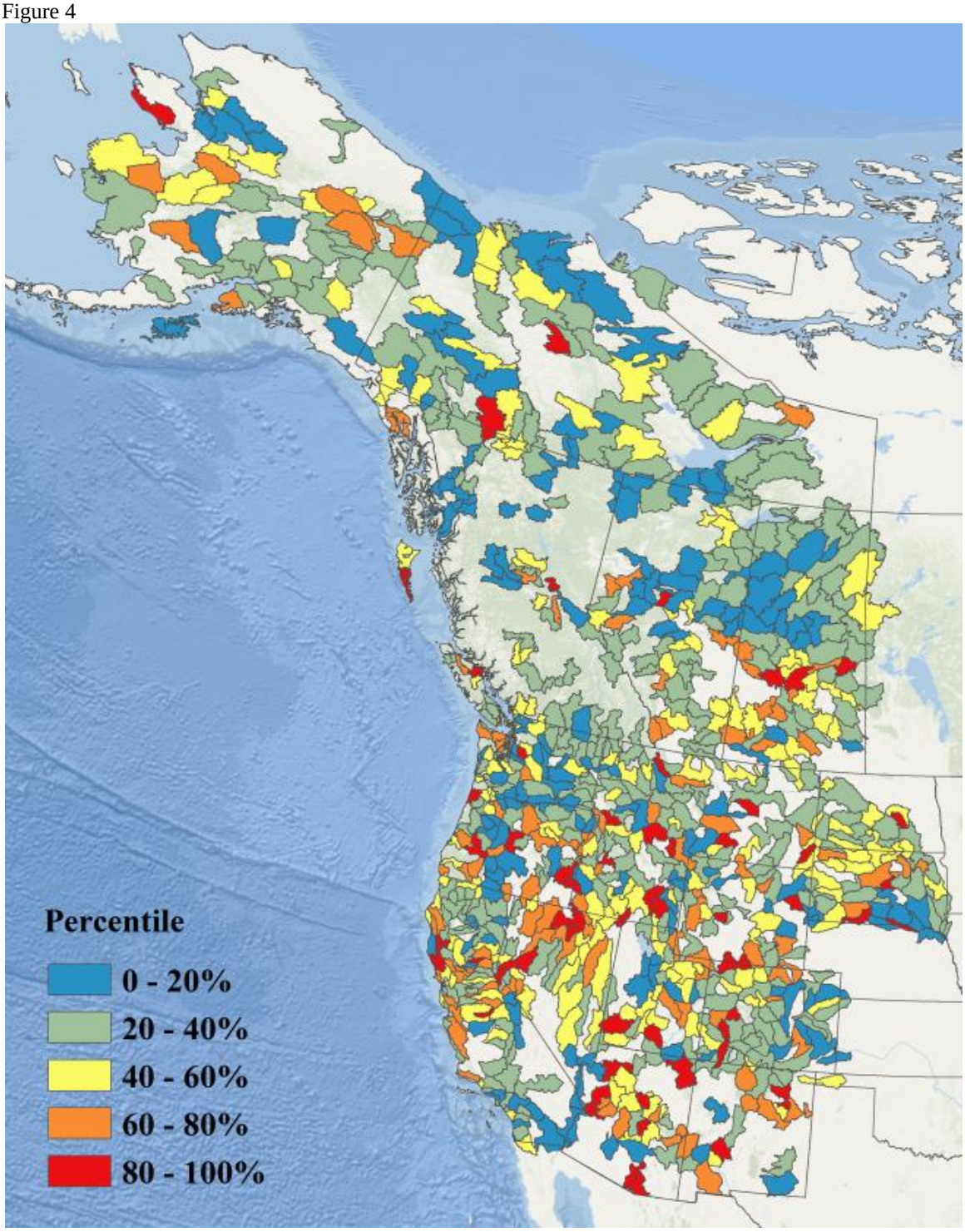


Figure 5

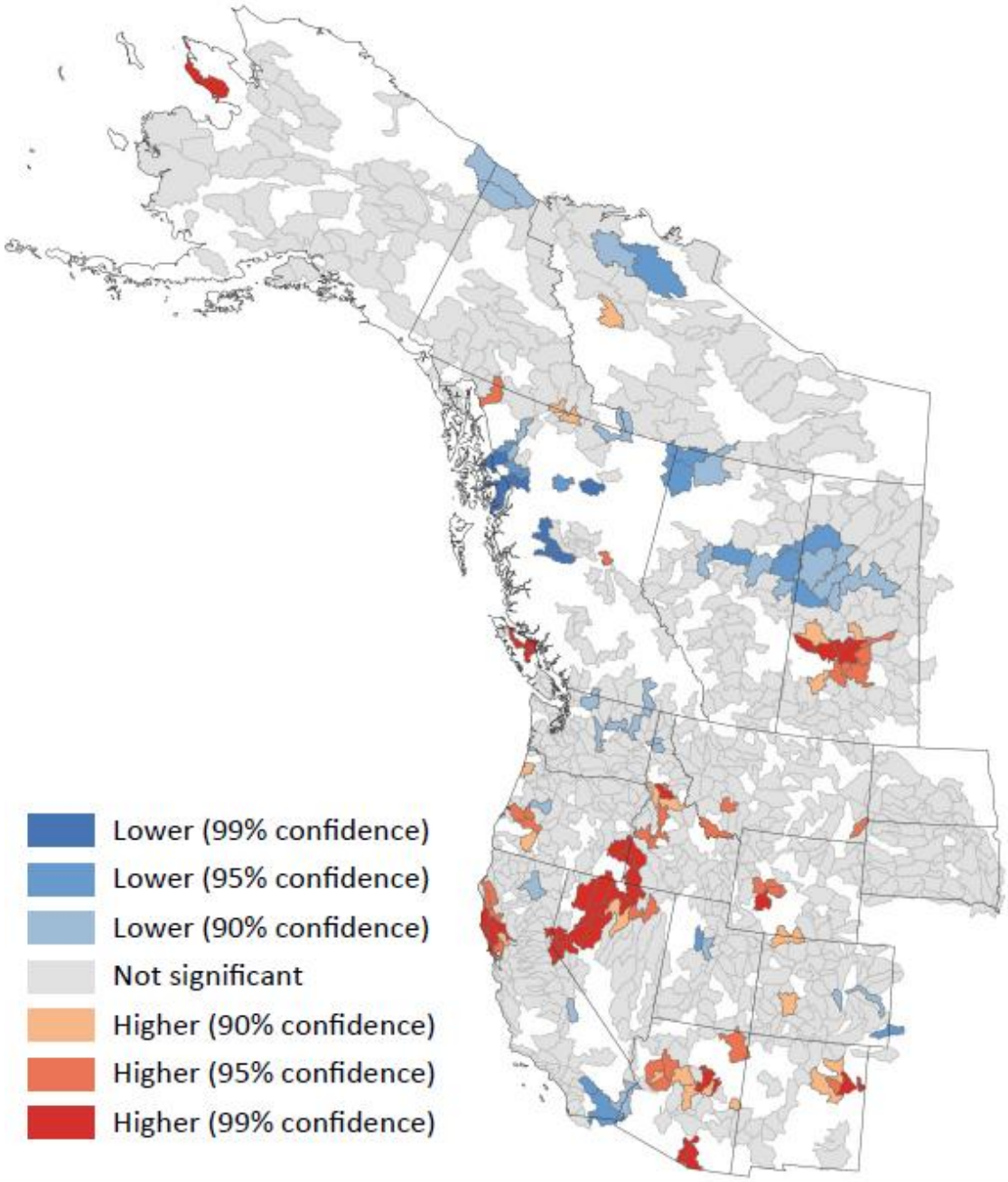
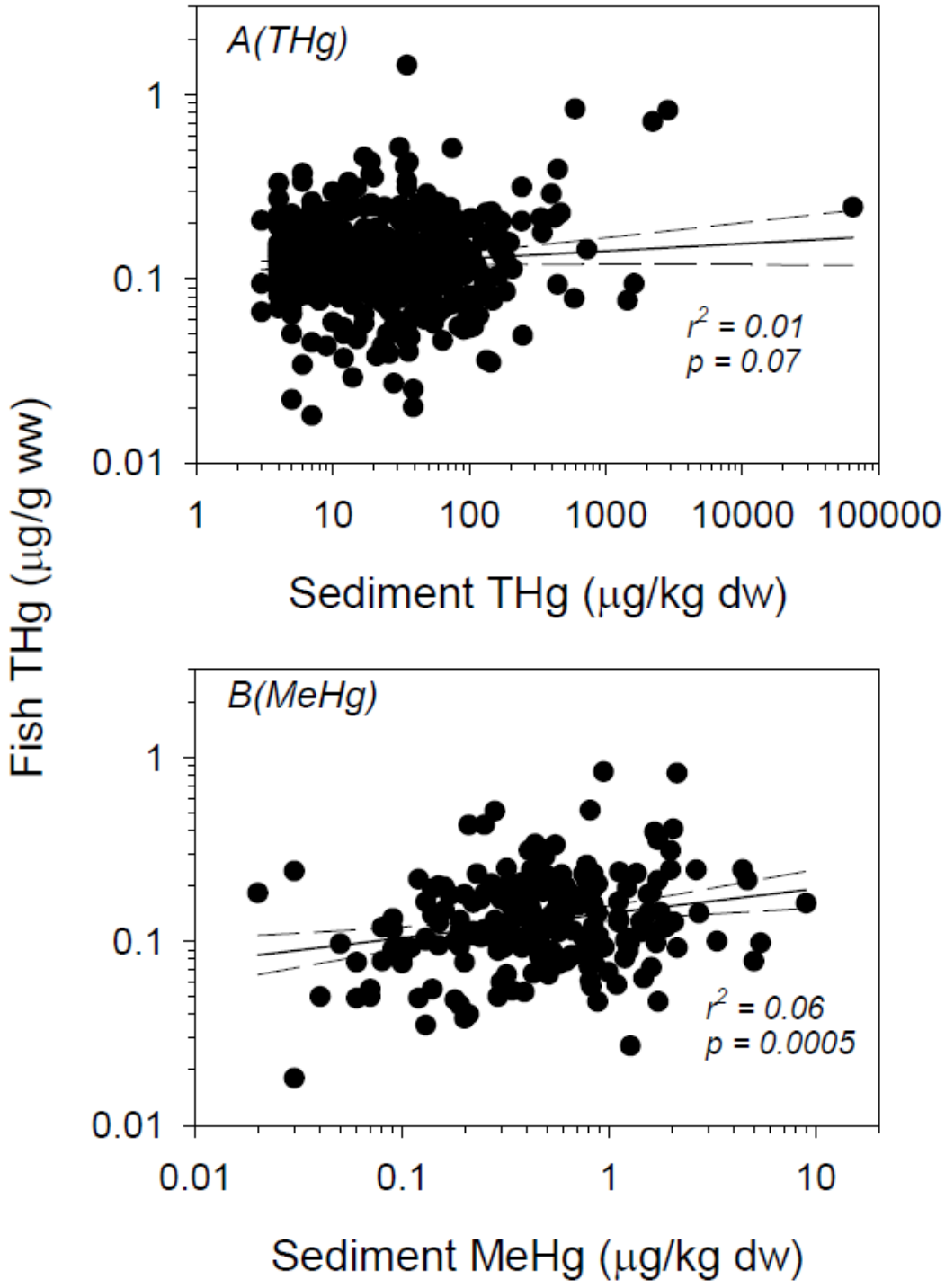

Lower (99% confidence)
Lower (95% confidence)
Lower (90% confidence)
Not significant
Higher (90% confidence)
Higher (95% confidence)
Higher (99% confidence)

Figure 6


A(THg)
1
0.1
0.01
1
10
100
1000
10000
100000
r2 = 0.01
p = 0.07
Sediment THg (µg/kg dw)
Fish THg (µg/g ww)
B(MeHg)
1
0.1
0.01
0.01
0.1
1
10
r2 = 0.06
p = 0.0005
Sediment MeHg (µg/kg dw)

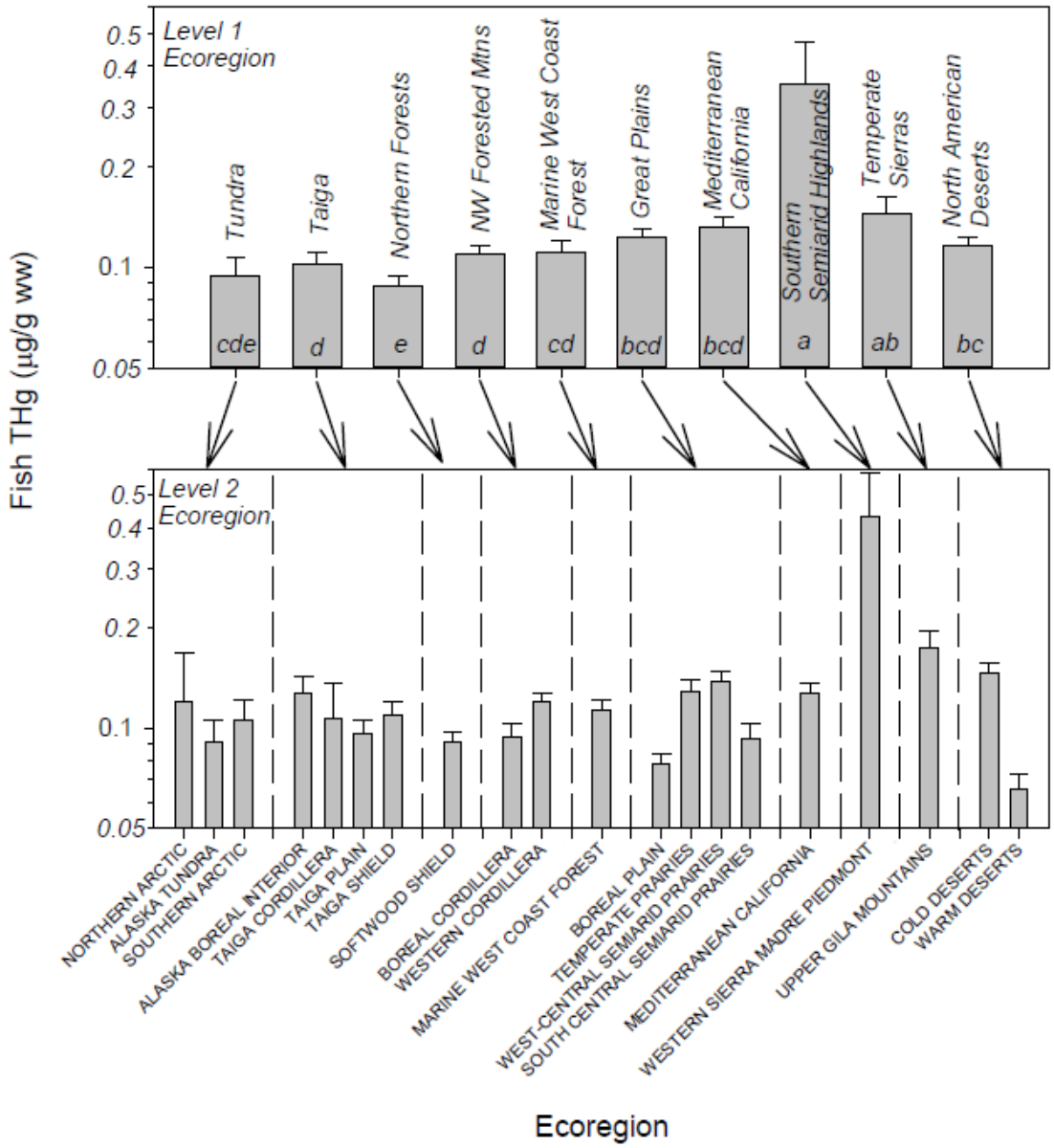

Level 1
Ecoregion
Tundra
Taiga
Northern Forests
NW Forested Mtns
Marine West Coast Forest
Great Plains
Mediterranean California
Southern Semiarid Highlands
Temperate Sierras
North American Deserts
cde
d
e
d
cd
bcd
bcd
a
ab
bc
Level 2
Ecoregion
0.5
0.4
0.3
0.2
0.1
0.05
Fish THg (µg/g ww)
NORTHERN ARCTIC
ALASKA TUNDRA
SOUTHERN ARCTIC
ALASKA BOREAL INTERIOR
TAIGA CORDILLERA
TAIGA PLAIN
TAIGA SHIELD
SOFTWOOD SHIELD
BOREAL CORDILLERA
WESTERN CORDILLERA
MARINE WEST COAST FOREST
BOREAL PLAIN
TEMPERATE PRAIRIES
WEST-CENTRAL SEMIARID PRAIRIES
SOUTH CENTRAL SEMIARID PRAIRIES
MEDITERRANEAN CALIFORNIA
WESTERN SIERRA MADRE PIEDMONT
UPPER GILA MOUNTAINS
COLD DESERTS
WARM DESERTS
Ecoregion

Figure 7

Figure 8

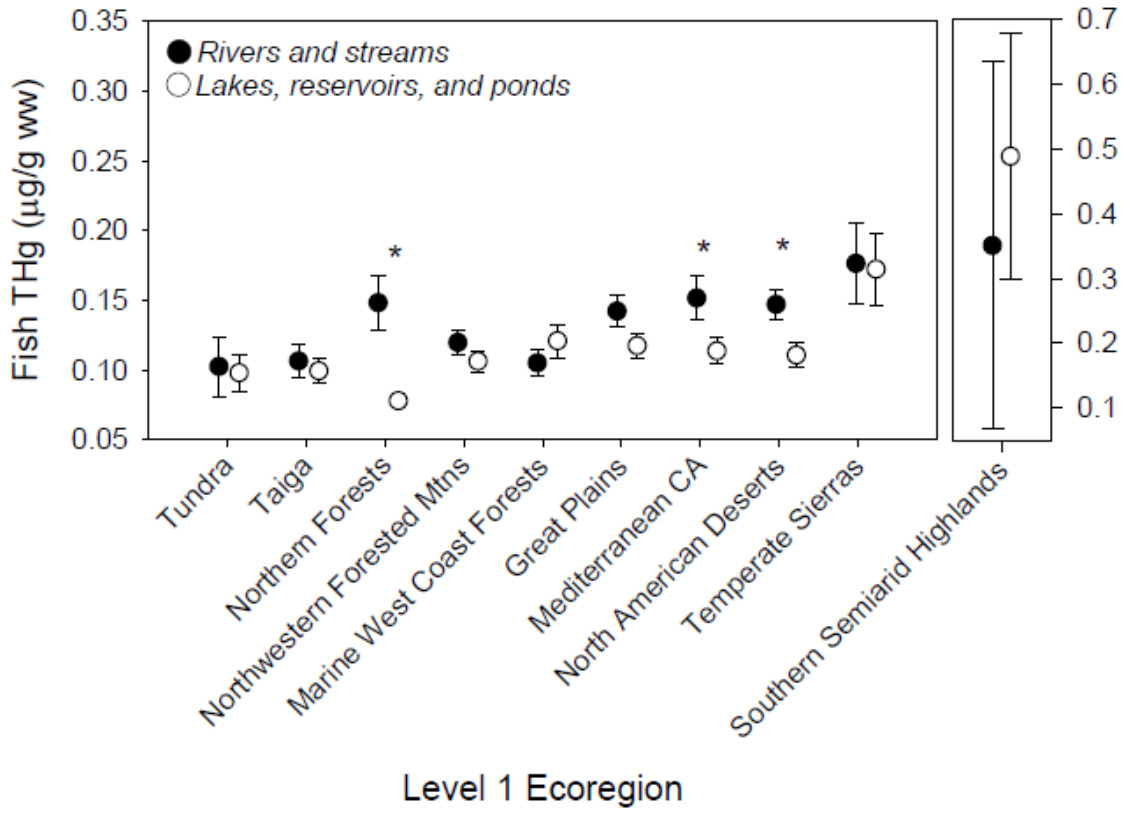

Rivers and streams
Lakes, reservoirs, and ponds
Fish THg (µg/g ww)
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.7
0.6
0.5
0.4
0.3
0.2
0.1
*
*
*
Tundra
Taiga
Northern Forests
Northwestern Forested Mtns
Marine West Coast Forests
Great Plains
Mediterranean CA
North American Deserts
Temperate Sierras
Southern Semiarid Highlands
Level 1 Ecoregion

Figure 9

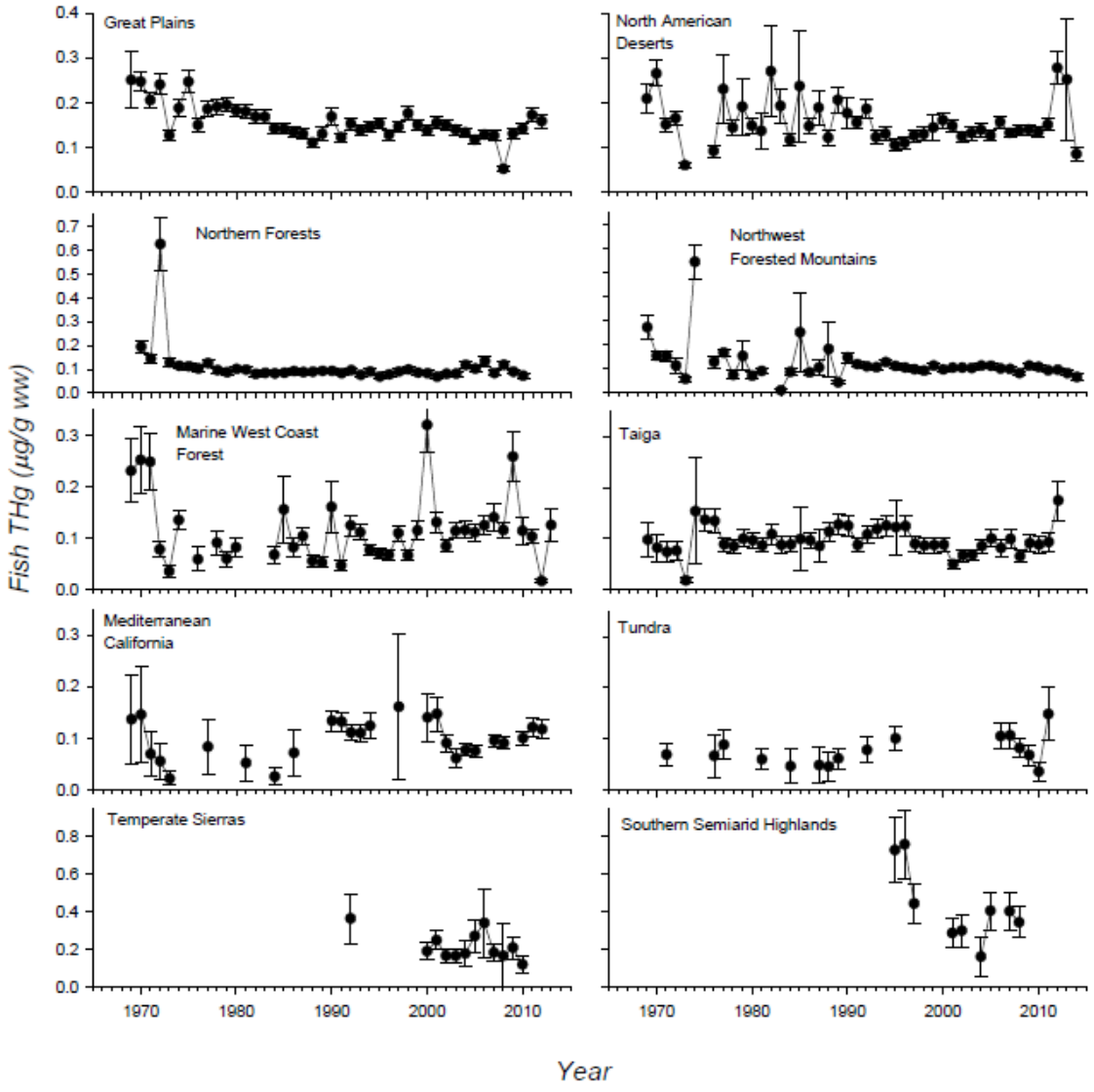
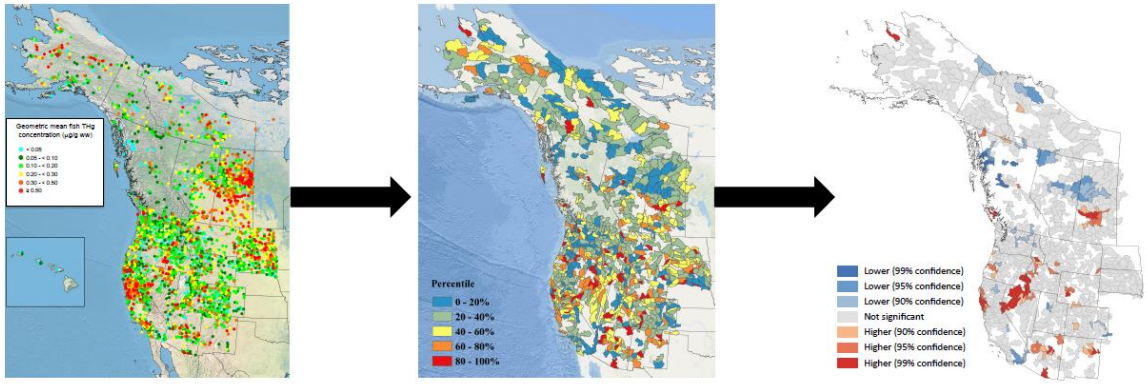

Great Plains
North American Deserts
Northern Forests
Northwest Forested Mountains
Marine West Coast Forest
Taiga
Mediterranean California
Tundra
Temperate Sierras
Southern Semiarid Highlands
Fish THg (µg/g ww)
Year
1970
1980
1990
2000
2010

Table 1. Distribution of Hydrologic Unit Codes (HUCs; level 8 equivalents) within Level 1 Ecoregions of western North America, and proportion of HUCs for which their least square mean fish THg concentrations fell within each data percentile category.

Level 1 Ecoregion	**Total HUCS (% of total)**	**0-20th percentile No. (%)**	**20-40th percentile No. (%)**	**40-60th percentile No. (%)**	**60-80th percentile No. (%)**	**80-100th percentile No. (%)**
Great Plains	152 (18%)	22 (15%)	58 (38%)	38 (25%)	23 (15%)	11 (7%)
Marine West Coast Forest	57 (6%)	14 (24%)	17 (30%)	18 (32%)	4 (7%)	4 (7%)
Mediterranean California	57 (6%)	7 (12%)	24 (42%)	12 (21%)	11 (19%)	3 (5%)
North American Deserts	163 (19%)	33 (20%)	49 (30%)	39 (24%)	22 (13%)	20 (12%)
Northern Forests	71 (8%)	23 (32%)	32 (45%)	9 (13%)	5 (7%)	2 (3%)
Northwest Forested Mountains	256 (30%)	50 (19%)	115 (45%)	45 (18%)	32 (12%)	14 (5%)
Southern Semiarid Highlands	3 (<1%)	0	1 (33%)	0	0	2 (67%)
Taiga	70 (8%)	17 (24%)	31 (44%)	16 (23%)	5 (7%)	1 (1%)
Temperate Sierras	17 (2%)	2 (12%)	3 (18%)	4 (23%)	5 (29%)	3 (18%)
Tundra	16 (2%)	6 (38%)	6 (38%)	2 (12%)	1 (6%)	1 (6%)



Graphical Abstract

Highlights

- Fish Hg concentrations were compared across Western US and Canada.
- Concentrations were heterogeneous across the landscape and differed among habitats.
- Mercury in sediment was a poor predictor of Hg in fish at the sub-continental scale.
- To manage environmental risk, knowledge of landscape scale drivers is important.