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CONTAMINANT MONITORING AND RESEARCH

California Bay - Delta Authority Fish Mercury Project

The Relationship between Landscape Features and Sport Fish Mercury in the Sacramento-San Joaquin Delta Watershed

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EXECUTIVE SUMMARY

The goal of this study was to quantify the relationship between landscape features and mercury in sport fish. We hypothesized that spatial variation in sport fish mercury would most strongly relate to major point sources associated with mining activity, habitat features (*e.g.,* abundance of wetlands), and watershed-scale patterns in mercury deposition from the atmosphere. Geographic information system (GIS) techniques were used to evaluate the influence of these features on the bioaccumulation of mercury in sport fish. In addition, we attempted to determine the most appropriate spatial scale for analysis of these influences.

Fish mercury data were selected from a state-wide historical bioaccumulation database, which also included data collected by the Fish Mercury Project (FMP) in 2005. Based on the geographic distribution of the sampling locations, three species common across the study area were chosen for analysis: largemouth bass, channel catfish, and white catfish. For the time interval examined (1990 – 2005), the mean wet weight concentration for each species at a given location was calculated.

By superimposing hydrologic flow paths with watershed delineations, we were able to determine flow direction and pathways from upstream sources to downstream habitats. "Fish watersheds" (watershed areas upstream of fish sampling locations) were created for a subset of locations in the CalFed Ecosystem Restoration Program Geographic Scope. Regions heavily influenced by mining within each watershed were also delineated. We examined landscape features in fish watersheds and within close proximity (circular buffers) of fish locations. Using a GIS database, several statistics were derived for each fish watershed area and buffer, including the percent covered by habitat features (vegetated vs. non-vegetated wetlands, permanent vs. temporary inundation), number of gold and mercury mines, total land area (km^2), and total length (km) of all streams and rivers.

The work presented in this report is the first attempt the authors are aware of to relate landscape features to fish mercury concentrations using GIS on a large regional scale. Fish mercury concentrations appeared to be influenced by many complex and interacting factors that obscured any obvious correlations with atmospheric deposition, historic mining districts, and wetlands at the watershed scale. Some features seemed to have an influence at the proximal-scale (buffer) analyses, namely vegetated wetland and temporarily inundated aquatic habitat. Though these tentative conclusions were based on small sample sizes, they contradicted the prevailing notion that wetlands generally increase methylmercury accumulation in the food web.

The lack of clear relationships in the dataset analyzed is probably due to shortcomings in the available data, rather than a true absence of influence of atmospheric deposition, mining, or wetlands on spatial patterns in mercury bioaccumulation. Future attempts to relate landscape features with mercury in the food web at the watershed scale should focus on watersheds with a stronger linkage between landscape features and mercury in fish and fewer confounding factors. Statewide monitoring of sport fish is currently being planned that should provide a better basis for conducting this type of analysis in the future.

INTRODUCTION

Mercury is a heavy metal that is highly toxic in its organic form methylmercury. It is known to bioaccumulate in food webs of the San Francisco Bay Estuary and Sacramento-San Joaquin Delta watershed (Bay-Delta). Studies conducted in this region over the last 15 years (*e.g.,* Fairey *et al.* 1997, Davis *et al.* 2000, Davis *et al.* 2002, Davis *et al.* 2003a, Greenfield *et al.* 2003, Davis *et al.* 2006b) have found mercury and other contaminants at concentrations of concern for human health in largemouth bass, white catfish, and other popular sport fish species. The Office of Environmental Health Hazard Assessment (OEHHA) has had an interim human health advisory for fish consumption in the Bay-Delta since 1994 (OEHHA 1994). OEHHA has also issued draft advisories for the lower Cosumnes River, lower Mokelumne River, lower Feather River and Putah Creek, and final advisories for Cache Creek, Bear Creek, Lake Natoma, and the lower American River, due to harmful levels of chemical contaminants, including mercury.

Mercury is a legacy contaminant that was mined historically from the California Coast Range and transported to the Sierra Nevada for use in gold extraction during the 19th century Gold Rush. Approximately 26 million pounds of mercury were transported from the Coast Ranges to the Sierra Nevada for gold extraction (Alpers and Hunerlach 2000). This mercury continues to enter the Bay-Delta from the Sacramento-San Joaquin Delta watershed. Runoff and weathering from the watershed continue to mobilize mercury from the landscape into the reservoirs, tributaries, and main rivers. However, mercury mobilization within the watershed is variable and highly driven by precipitation patterns (McKee *et al.* 2006). The Sacramento River is the dominant contributor of both fresh water and total mercury to the Delta. The Sacramento River is also the largest contributor of methylmercury to the Delta, followed by the Yolo Bypass and San Joaquin River (Wood *et al.* 2006).

The two main measures of mercury entering the Bay-Delta that are of interest to regulators are total mercury and methylmercury. Mercury sources include historic gold mining areas, wastewater, stormwater and other urban runoff, agricultural runoff, and direct atmospheric deposition. The contribution of each source varies across time and space. However, the most significant mercury source by mass is attributable to mining activity during the 1800s. Historical releases of mercury to the aquatic environment from gold mining areas were substantial (1.4 – 3.6 million kg; USGS 2000), and mercury continues to wash downstream from these areas today (Domagalski 1998). High mercury concentrations measured in present-day water and sediments downstream from mines suggest that hundreds to thousands of pounds of mercury remain in many Sierra watersheds. In addition to this continuing input of mercury to the Bay-Delta, there is also a large, historically-deposited reservoir of mercury in the sediments of water bodies within the region. Both new and historically-deposited mercury are available for transport and uptake into the aquatic food web.

Wetlands are habitats of concern for the production of methylmercury. Many studies have shown that the transformation from inorganic mercury (the form of mercury not significantly biologically available) to organic methylmercury (the form that is most toxic to biota and most readily enters food webs) can occur at high rates in wetlands. The concern is that wetlands may produce an abundance of methylmercury in regions of historic mercury contamination, which would then accumulate in local food webs or be exported to other habitats. Positive correlations between percentage of wetlands in an area and water methylmercury concentration have been observed (St Louis *et al.* 1994, Hurley *et al.* 1995, Krabbenhoft *et al.* 1999). Methylmercury is produced by methylation of inorganic mercury by sulfate-reducing bacteria that thrive in anoxic wetland soil and sediment (Gilmour and Henry 1991, Gilmour *et al.* 1992). Therefore, the potential exists to inadvertently increase the risk of mercury methylation and accumulation in fish and wildlife by increasing wetland habitat through restoration within the Bay-Delta. Mercury cycling is complex, and there are many properties of wetlands that contribute to the production of methylmercury, including high rates of microbial activity, high concentrations of dissolved organic carbon, and steep redox gradients. However, wetlands may also sequester mercury or be neither sources nor sinks of methylmercury – the full complexity of the relationship between wetlands and methylmercury is not completely understood (St Louis *et al.* 1996, Wiener and Shields 2000, Stephenson *et al.* 2006). Degradation of methylmercury is equally important, and also varies significantly in time and space. The balance of methlmercury production and degradation drives the movement of mercury into aquatic food webs and sport fish.

Objectives

The goal of this study was to quantify the relationship between landscape features and mercury in sport fish. Many factors could influence the variation in mercury concentrations in sport fish across the study area. We hypothesized that spatial variation in sport fish mercury would most strongly relate to major point sources associated with mining activity), habitat features (*e.g.,* abundance of wetlands), and watershed-scale patterns in mercury deposition from the atmosphere. Geographic information system (GIS) techniques were used to evaluate the influence of these sources on the bioaccumulation of mercury in sport fish of the Bay-Delta. In addition, we attempted to determine the most appropriate spatial scale of influence by conducting analyses on attributes from entire watersheds and from close proximity to fish sampling locations. The proximal (near-field influence) analyses were applied to mining and wetland features to test whether local sources correlated better with sport fish mercury concentrations than distant sources upstream. Therefore, each of the source hypotheses outlined below (H1 to H3) was combined with a hypothesis relating the scale of influence for the source(s) (H4 or H5). Both watershed-scale and proximal-scale analyses aimed to identify those landscape features that best explained mercury bioaccumulation in sport fish. In addition, this report will discuss other landscape attributes (unavailable for this analysis) that could explain mercury bioaccumulation in sport fish.

Hypotheses

The following three mercury source hypotheses (H1 to H3) were tested in combination with a predictive scale of exposure hypothesis (H4 or H5). For example, H2 and H4 examined the relationship between mining sources and fish mercury concentration on a watershed scale. However, the hypothesis that related biota mercury to atmospheric deposition (H1) was only considered at the watershed scale (H4). This was done because we did not have sufficient data to examine atmospheric effects at the local scale.

Relating mercury sources to fish mercury concentrations:

H1: Mercury in biota is primarily caused by mercury deposited from the atmosphere.
P1: Mercury in fish will be higher in watersheds with no mining sources.

H2: Mercury in biota is primarily caused by mercury that enters waterways from mining sources.
P2: Mercury in fish will be higher in watersheds with more mining activity.

H3: Mercury in biota is primarily caused by export of methylmercury from wetlands.
P3: Mercury in fish will be higher in watersheds with more wetlands.

Relating exposure scale of mercury sources to fish mercury concentrations:

H4: Methylmercury entering food webs is transported over a long distance.
P4: Mercury in fish will be related to the amount of mining and/or wetlands in the upstream watershed.

H5: Methylmercury entering food webs is derived from local sources.
P5: Mercury in fish will be related to proximal sources (mining and/or wetlands).

METHODS

Fish Mercury Data

The regional focus of this study was the area defined by the CalFed Ecosystem Restoration Program Geographic Scope (CalFed boundary area = 120,000 km^2), which encompasses the Sacramento-San Joaquin Delta and its watershed.

Fish mercury data were selected from a state-wide historical bioaccumulation database (SWAMP; Davis *et al.* 2006a), that also included data collected by the Fish Mercury Project (FMP) in 2005. These data were stored in Microsoft Access (2003) database tables. Locations within the CalFed boundary that were sampled between 1990 and 2005 were selected. Significant intra-annual trends have not been shown to be widespread in this region (Davis *et al.* 2003a, Grenier *et al.* 2006). Therefore, variation due to averaging across years was not considered to be a major issue with these data. Data represented total mercury in sport fish muscle tissue measured on a wet-weight basis. Table 1 summarizes the numerous authors and agencies whose data comprise this regional dataset. Based on the geographic distribution of the sampling locations (Map 1), three species common across the study area were chosen for analysis: largemouth bass, channel catfish, and white catfish. Largemouth bass was the most widely sampled species (Table 2); channel catfish was well distributed outside the Delta; and white catfish was well sampled within the Delta. All three species are of importance to human consumption, are known to bioaccumulate mercury, and are target species for the FMP. Size limits for each species were applied to limit the variation in mercury concentration due to fish length (Table 3). The size limits were chosen to include a large proportion of the available data and to follow USEPA guidance. USEPA guidance (U. S. EPA 2000) specifies that the smallest fish in a composite should be no less than 75% of the largest. Fork and total length measurements were both included in the analysis, because excluding either one would have drastically reduced the sample size. In the time interval examined (1990 – 2005), the mean wet weight concentration for each species at a given location was calculated. Largemouth bass data were used in analyses conducted both inside and outside of the Delta. The limited distributions of the catfish species, however, meant that channel catfish was used solely for outside-Delta analyses, whereas white catfish data were limited to the Delta analyses.

Mapping and GIS Analysis

Six primary datasets were used in mining and landscape analyses: the fish mercury bioaccumulation database, National Wetlands Inventory (NWI), National Hydrography Dataset (NHD), Calwater 2.2.1, Mine Resources Data System (MRDS), and the Delta Methylmercury TMDL Sub-Area Delineations. Table 4 summarizes the metadata for each dataset. Map 2 shows the fish sampling locations, NHD, NWI, and MRDS datasets used in analyses.

A Geographic Information System (GIS) was used to evaluate sources of mercury. We superimposed hydrologic flow lines from the NHD with Calwater 2.2.1 (hydrologic area delineations, *i.e.,* watersheds), and were able to determine flow direction and hydrologic pathways from upstream sources to downstream habitats. Calwater watersheds that were determined to be connected hydrologically to a fish location were merged (and sometimes reshaped for accuracy) to create a "fish watershed". These fish watershed areas were created for sampling locations on main stem and tributary river sites (Maps 3 and 4), as well as for lake and reservoir sites (Map 5). The manual nature of this process, however, may have led to minor errors, whereby an area that did not directly influence a fish sampling location was included.

Regions within each fish watershed area that were downstream of the main cluster of gold and mercury mining inside each fish watershed were identified. Hence, these were the areas where mercury emanating from historic mines would be present within the fish watershed. High concentrations of inorganic mercury have been observed in regions with substantial influence from historic mining, such as in the Cache Creek watershed (Domagalski 2001, Domagalski *et al.* 2004). In our analyses, these regions are referred to as "areas of mining influence". In all cases, over 90% of mines within a fish watershed were located within the area of mining influence. Mines were selected from the full MRDS dataset based on mine sites having either "gold" and/or "mercury" listed in the attribute table fields reflecting major commodities and/or ore minerals or materials. The MRDS was not completely standardized, being created from a variety of different sources that had varying levels of detail and consistency, both in positional and ancillary information. Additionally, this dataset did not distinguish high production from low production mines nor did it include years of peak production. Despite these limitations, the MRDS dataset represents the best GIS layer for mines presently available for California.

Fish watersheds could not be created for all locations monitored for the three sport fish species examined. These fish data were not ideally structured for this analysis, and the sample size of spatially distinct locations was limited. A number of locations were found to be within the same fish watershed, and, thus, were averaged. Others were located in regions with many indistinguishable hydrologic influences, or lacked the necessary GIS data to create a fish watershed. In addition, the clearest relationships between landscape features and fish mercury were hypothesized to occur at fish locations along tributaries. However, the number of sampling locations along tributaries was low. Tributaries were often sampled downstream of dams and/or reservoirs, where the influence of landscape features above the reservoirs on mercury is unknown. In general, we found that each tributary was only sampled at one or two locations. Despite these shortcomings, the data set analyzed is the most comprehensive dataset ever compiled for this region and provides the best opportunity for analysis on the landscape scale. Therefore, we have conducted the analyses that follow with the understanding that we may miss important relationships due to the data gaps and data structure problems (Type II error – failing to detect a relationship when it does exist), but we are less likely to conclude that important relationships exist when they truly do not (Type I error – see Methods: *Multiple Statistical Testing).*

Using a GIS database, several statistics were derived for each fish watershed area, including the percent covered by NWI polygon features (aquatic habitats, excluding streams and rivers), number of gold and mercury mines, total land area (km^2), and total length (km) of all NHD streams and rivers (excluding underground pipelines). Wherever possible, the high resolution NHD dataset (1:24000) was used. In geographic areas where this was not possible (*e.g.,* sub-basin 18040003), we used NHD medium resolution data (1:100000) instead. The NWI polygon features were further categorized by habitat type (vegetated wetland and non-vegetated aquatic habitat) and flooding frequency (permanently flooded area and temporarily inundated area). The specific landscape attributes from NWI that were used to characterize habitat and flooding frequency are listed in Tables 5 and 6. The choice of which group to place each NWI category into was based on the detailed description of that category in the NWI metadata.

The NWI coverage varied both regionally and across the State. Regions had variable coverage both in years of data collection and degree of detail. Density of polygons differed depending on the individual cartographers who performed the digitizing, and some regions completely lacked NWI coverage. To address these inconsistencies, NWI statistics were not computed for fish watersheds with more than 40% of the total area missing NWI data. For watersheds that fell under this threshold, the missing coverage was extrapolated from the NWI data available.

In addition to watershed-scale analyses, we examined landscape features within close proximity of fish locations, by creating circular regions ("buffers") of varying size around the locations. One- and 5-km-radius buffers were used to approximate the home range size of the species examined (Moyle 2002). Within each buffer, NWI polygonal area (aquatic habitat areas, excluding rivers and streams) was calculated using the same categories as for the watershed-scale analysis. The number of mines falling within each buffer was only enumerated outside of the Delta in 5 km buffers, as there were so few mines present within the Delta or within 1 km buffers outside the Delta. Where buffers overlapped more than 50% between different fish locations, mean values were calculated for mercury, NWI statistics, and mine totals.

The map figures were designed using ESRI ArcInfo 9.1 software in a California Teale Albers NAD 83 Projection. Two styles of GIS maps were produced for this report as follows:

1. Maps that show examples of the fish watershed areas and largemouth bass mean mercury concentrations from different types of sites (tributaries, main stem rivers, and lakes/reservoirs).
2. Maps that show results from the NWI and mine layers, within 1-km and 5-km-radius buffers of the fish locations. These buffer maps indicate mean mercury concentrations using a four-color graduated scheme: green, yellow, orange, red from low to high concentration.

Statistical Analysis

Comparing Mercury Concentration with and without Mining Influence

To examine sources of mercury that could only be attributed to atmospheric deposition, watersheds that lacked mining influence were identified. However, only four watersheds (all lakes or reservoirs) within the CalFed boundary were found. Locations outside of the CalFed boundary were therefore used to supplement the dataset. This approach added a further four locations (in southern California) that were also sampled for largemouth bass in lakes or reservoirs (Map 6). This approach was required as adequate atmospheric deposition data was not available for the Bay-Delta. A two-sample t-test was used to compare mercury concentrations between northern and southern California, due to possible confounding factors such as rainfall, temperature, etc. There was no significant difference between the northern and southern mercury concentrations, and these data were subsequently pooled (n = 8). A second two-sample t-test was then used to compare ($\log_{10}$-transformed) mercury concentrations in largemouth bass from the eight fish watersheds lacking mining influence to 16 lake and reservoir locations (within the CalFed boundary) that contained more than 10 mines each. A Bonferroni adjustment for multiple tests was applied. Significance in all statistical analyses was set to $\alpha = 0.05$.

Comparing Mercury Concentrations between Major Aquatic Habitat Types outside the Delta

Prior to performing mining and landscape-feature analyses outside of the Delta, mercury concentrations were compared between major aquatic habitat types. Fish locations were assigned to one of three types (main stem rivers, tributaries, or lakes/reservoirs). Both largemouth bass (n = 53) and channel catfish (n = 32) were distributed widely enough to examine these differences between habitat type. Largemouth bass mercury concentrations were compared between the three habitat types using a one-way analysis-of-variance (ANOVA), but for channel catfish there were insufficient samples to treat main stem and tributaries separately, hence a two-sample t-test was used.

Mining and Habitat Analyses outside the Delta

Linear regression models were used to test the relationship of mining and habitat features to fish mercury concentration at locations outside of the Delta. Note that sample sizes varied depending on coverage of GIS data (see Appendix Table 1). Based on a lack of observed differences in the

ANOVA result previously described, data from all major habitat types were pooled for both largemouth bass and channel catfish. Size limits were applied prior to analysis, to account for the variation of mercury concentration with fish length. There were insufficient data to adjust for fish length using an analysis of covariance model. Data were $\log_{10}$ or square-root transformed to improve normality and variance homoscedasticity of residuals. Regressions relating fish mercury to mining influence were based on the density of mines (number of mines per km^2), rather than absolute mine abundance. Unfortunately, there was an inadequate sample size of fish sampling locations in regions with a high density of mercury mines (eastern side of the Central Valley) to compare these to fish collected from gold mining watersheds in the Sierra Nevada. In regressions that included wetlands and other aquatic habitats as a factor, the wetlands inside the areas of mining influence, rather than from the full fish watersheds, were used to narrow the analysis to wetlands that were downstream from large mercury sources. Furthermore, the aquatic NWI features were converted to a percentage of the watershed area, or percentage of the buffer area, depending on the scale of the analysis. The habitat features consisted of total aquatic habitat, vegetated wetland, non-vegetated aquatic habitat, and temporarily flooded aquatic habitat. Both watershed and proximal-scale data were examined similarly.

Comparing Mercury Concentrations between Hydrologic Sub-areas in the Delta

Fish locations sampled for largemouth bass (n = 54) and white catfish (n = 37) within the Delta were initially grouped based on the Delta Methylmercury TMDL Sub-area Delineations (Wood *et al.* 2006). Group means of mercury concentration were compared using a one-way analysis of variance. Significant results were followed by a multiple comparison test to determine which means differed, using the conservative Tukey's Honestly Significant Difference (HSD).

Habitat Analyses in the Delta

Statistical analysis of habitat (*i.e.,* aquatic and wetland) features in relation to fish mercury concentration on the Delta hydrologic sub-area scale was performed using linear regression. Data from white catfish and largemouth bass were included. The regression models examined the relationship of habitat features (*i.e.,* % total aquatic habitat area, % vegetated wetland, % non-vegetated aquatic habitat, and % temporarily flooded aquatic habitat) to the average fish mercury concentration of a given species within each sub-area. Each data point (n = 6) in the regression therefore represented the percent of habitat feature and average mercury concentration for one of the six sub-areas.

Comparison of habitat features in relation to fish mercury concentrations using the radial buffer data required a modified approach. Since fish mercury concentrations varied significantly between the TMDL sub-areas of the Delta (ANOVA described above; Grenier *et al.* 2006), hydrologic sub-area was included as a factor in all these models of potential proximal mercury sources. These ANCOVA models used dummy variables, with backward, stepwise elimination, to determine differences in means and slopes between sub-areas (see Appendix Tables 2 and 3 for sample sizes). As with analyses outside of the Delta, fish mercury and habitat feature data were transformed to improve normality and variance homoscedasticity of residuals. However, in the 1-km buffer analyses, there was a high occurrence of zero values, violating the normality assumption. In this case, $\log(x+1)$ was the preferred transformation. Sub-areas with at least 5 samples and an approximate variation of at least 5% in the habitat feature being tested were included in the analysis. Due to the small sample size of locations in some sub-areas (*e.g.,* Sacramento and San Joaquin Rivers), outliers were removed if they significantly affected the outcome of the models or prompted a violation of the normality assumption. Interactions between sub-area and habitat features were tested first to determine whether a common-slope model was appropriate. In cases where no interaction was found, a traditional ANCOVA (constant slope) was performed.

Multiple Statistical Testing

Adjustments for multiple statistical tests were not performed in this study (except the one Bonferroni correction mentioned previously). Due to the high occurrence of non-significant results and the limitations of the datasets available, we considered the risk of Type-I error (rejecting the null hypothesis when it is true) to be small. Instead, we used a weight-of-evidence approach to address the multiple statistical tests. Analyses pointing to the same trend in the data (*e.g.,* negative relationship between landscape feature and fish mercury concentration) were considered together to increase our confidence in the results.

RESULTS AND DISCUSSION

Atmospheric Deposition in Largemouth Bass outside the Delta (Hypothesis H1)

Four fish watersheds within the CalFed boundary (and eight across the State) that were sampled for largemouth bass contained no mines within their boundaries (Map 6). All of these fish were from lakes or reservoirs on the western side of the Central Valley (n = 4) or near Malibu in southern California (n = 4). There was no significant difference (p = 0.15) between largemouth bass mercury concentrations in southern and northern California. The mercury concentrations for these eight locations without mines (mean = 0.43 µg/g) were compared to 16 lake and reservoir sampling sites in the CalFed boundary (mean = 0.40 µg/g) whose watersheds contained 10 or more mines (Figure 1), and there was no significant difference between the groups (p = 0.75). In the few fish watersheds that lacked mining influence, mercury levels remained sufficiently high to be indistinguishable from fish watersheds that contained an abundance of mines, suggesting that atmospheric deposition may contribute to the observed mercury concentrations in sport fish.

The lack of significant difference between mine-impacted fish watersheds and those lacking mines requires further investigation. The sample size available for this analysis was too small to draw any definitive conclusions. Atmospheric deposition is considered to be a principal source of inorganic mercury to most aquatic systems in the United States. Few atmospheric deposition datasets have been collected in California, however, with only a handful of monitoring stations gathering long-term data (*e.g.,* San Jose, CA; SFEI 2001). It is generally considered that both wet and dry deposition are relatively low in the State (*e.g.,* Steding and Flegal 2002, NADP 2004, Wood *et al.* 2006). Given that the current research suggests that the relative inputs of atmospherically deposited mercury to aquatic systems in California are small, other factors may explain the results observed. Inputs from agricultural and urban runoff, for example, were not examined. The National Land Cover Dataset (NLCD) was considered during the early conception of this work to test hypotheses that would correlate density of agricultural and urban landscape features to mercury levels in fish. However, as with atmospheric deposition data, the NLCD contained large data gaps that prohibited its use for this study. On the other hand, atmospherically deposited mercury may be more bioavailable than mercury that has been on the landscape for decades, in which case even the relatively small atmospheric inputs could be the source of observed fish mercury (Wiener *et al.* 2003). Future work on non-point source inputs to aquatic systems is needed to ascertain their contributions to fish mercury in the region.

Comparing Mercury Concentrations between Major Aquatic Habitat Types outside the Delta

Largemouth bass data from 121 locations in rivers, lakes, and reservoirs sampled between 1990 and 2005 were evaluated in this analysis (Table 2). Fifty-seven locations were selected to compare mercury concentrations between fish collected from three major habitat types (main stem rivers, tributaries, and lakes/reservoirs). The main stem sampling sites were located along the Sacramento and San Joaquin Rivers (n = 15). The average mercury concentration ranged from 0.35 µg/g (Sacramento River at Rio Vista) to 0.98 µg/g (San Joaquin River at Howard Road), with a mean of 0.65 µg/g (sd = 0.20 µg/g). Tributary locations (n = 14) consisted of samples from the Tuolumne, Mokelumne, Merced, Stanislaus, American and Feather Rivers.

Mercury concentrations at these locations were somewhat lower, ranging from 0.19 µg/g (Feather River at Gridley) to 0.67 µg/g (Feather River at Nicolaus), with a mean of 0.47 µg/g (sd = 0.20 µg/g). The lakes and reservoirs (n = 24) were distributed across the CalFed region, and exhibited average mercury concentrations that ranged from 0.06 µg/g (Antelope Lake) to 0.85 µg/g (Lake Combie), with a mean of 0.42 µg/g (s.d. = 0.25 µg/g). However, there was no significant difference (p = 0.24) between the mercury concentration in bass collected from the different habitat types.

White and channel catfish data from 56 and 52 locations, respectively, collected between 1990 and 2005, were also evaluated (Table 2). However, none of the white catfish were collected from reservoirs, and the majority (39 of 56, 70%) were from the Delta or its tributaries. A comparison of fish mercury from the different habitat types was therefore not possible, and this species was used only to address hypotheses related to the variation among Delta sub-areas. On the other hand, channel catfish were rare in the Delta but common in both reservoirs and rivers, though not as frequent along the main stems. Thirty-two channel catfish locations were selected to compare mercury concentrations between rivers and lakes/reservoirs. The average channel catfish mercury at river locations (including both main stems and tributaries) ranged from 0.1 µg/g (San Joaquin River at Fremont Ford) to 1.07 µg/g (Stanislaus River), with a mean of 0.37 µg/g (s.d. = 0.27 µg/g). In lakes and reservoirs, mercury concentrations appeared to be slightly lower and less variable, ranging from 0.1 µg/g (Lake Chabot) to 0.63 µg/g (Camp Far West Reservoir), with a mean of 0.28 µg/g (s.d. = 0.14 µg/g). However, as with largemouth bass, there was no significant difference between mercury concentrations of catfish from rivers compared to lake and reservoirs (p = 0.31). Therefore, data from all major habitat types were pooled within each species for subsequent analyses.

Fish Mercury and Mining outside the Delta (Hypothesis H2)

Mercury associated with mining activity has contributed to the contamination of water bodies in the Central Valley for over 150 years. Map 2 (top left) illustrates the extent of mining influence in this region. Mercury concentrations in largemouth bass and channel catfish at locations close to mines (May *et al.* 2000) and within mine-impacted watersheds (Saiki *et al.* 2004, May *et al.* 2005) have been reported at concentrations well above 0.3 µg/g. However, high fish mercury concentrations in the Central Valley region have also been shown in watersheds with minimal or no mining influence (*e.g.,* Indian Valley reservoirs; Boles 2004). These regional datasets were included in the bioaccumulation database to evaluate the relationship of mine influence to fish mercury concentrations, which has not been previously examined on such a broad scale as the Delta watershed.

Regression analyses on data outside the Delta did not indicate a relationship between upstream mining sources and fish mercury contamination. Neither the density of mines in fish watersheds nor the abundance of mines in 5-km-radius buffers were significantly related to mercury concentration in largemouth bass or channel catfish (p > 0.05; Appendix Table 1). Maps 3 and 4 illustrate how variable mine density in fish watersheds and largemouth bass mercury concentrations were in eight tributary watersheds and two main stem watersheds. These maps suggest that locations with relatively low fish mercury concentrations (*e.g.,* Map 3c: Feather River at Gridley, mine density = 0.10 mines/km^2) may have similar mine density upstream as those locations with much higher fish mercury concentrations (*e.g.,* Map 4: Sacramento River at Butte City, mine density = 0.04 mines/ km^2). Similarly, Map 3d illustrates two locations on nearby tributaries that exhibit very different concentrations. Specifically, largemouth bass from Tuolumne River at Shiloh Road were exposed to fewer mines at half the density (0.15 mines/km^2) relative to fish from Stanislaus River at Caswell State Park (0.33 mines/km^2), yet the mercury concentration at the Tuolumne River site was twice as high. These observations indicate that factors other than mine density are contributing significantly to the mercury concentrations found in these species.

Proximal analyses also indicated that mine density within a 5-km-radius buffer does not relate to fish mercury concentrations. Maps 7 and 8 again illustrate that fish mercury concentrations were high regardless of mine density nearby. For example, New Melones Reservoir (Map 7) contains more than 200 mines within a 5-km-radius, yet largemouth bass had a much lower concentration (0.26 µg/g) than at locations with minimal mining influence (*e.g.*, Map 7: Mokelumne River between Beaver and Hog Slough, 0.55 µg/g). Thus, either the mining data lack sufficient detail to understand how they relate to fish mercury (*e.g.,* more data is needed on production quantity, years of operation, remediation actions, etc.) or no significant relationship exists because of the specific water body conditions. For example, the selected locations may not be conducive to net methylation and food web accumulation. Microbial mercury methylation rate is considered to exert a significant influence on the rate of bioaccumulation in fish (*e.g.,* Allen-Gil *et al.* 1995). Conditions that favor microbial methylation may be very important (*e.g.,* pH, redox potential, organic content of sediments, and temperature). Thus, point sources of inorganic mercury would not explain fully the concentrations in fish. Mercury bioaccumulation is not dependent solely on the amount of mercury that enters an aquatic system, but also on the tendency of the system to convert inorganic mercury to methylmercury.

The relationship of mine density to fish mercury was clouded in our analyses by the lack of detail in the MRDS layer (see *Methods:Mapping and GIS Analysis*). The mine data did not include quantities produced, years of peak production, mercury mass loading information, or other statistics that would relate to the degree of impact from specific mining districts. Therefore, we were unable to account for a single high production mine having a stronger influence on a fish location, compared to hundreds to thousands of smaller mines spread across an entire watershed. We consider the lack of detail in the mining dataset to be a major factor contributing to the result of no relationship between density of mines and fish mercury concentration. Again, local variation in biogeochemical processes governing mercury bioaccumulation is also likely important.

Fish Mercury and Habitat Features outside the Delta (Hypothesis H3)

Recently, the potential of mercury to accumulate in food webs close to wetlands has attracted considerable attention. Numerous studies have identified wetlands (St Louis *et al.* 1994) and their environmental conditions (*e.g.,* anoxia and temporary inundation) as potentially enhancing net mercury methylation rates (Zillioux *et al.* 1993). This issue is of particular concern due to the large-scale restoration efforts proposed for the Bay-Delta to restore currently dry areas to wetland habitat (Davis *et al.* 2003b). We separately evaluated whether aquatic and wetland habitat extent could be used to predict fish mercury concentrations both inside and outside of the Delta. These analyses were separated due to the high variability in fish mercury concentrations reported in the Delta (Davis *et al.* 2003a, Grenier *et al.* 2006), and the complex water regimes in this region (Wood *et al.* 2006).

Extent of aquatic and wetland habitats in fish watersheds was not correlated with fish mercury concentration. Outside the Delta, there was no relationship between aquatic and wetland habitat extent and either largemouth bass or channel catfish mercury concentration ($p > 0.05$; Appendix 1). Length of NHD river and streams (results not presented) were similarly unrelated. The non-significant results may be strongly influenced by fish mercury concentrations throughout the region that vary irrespective of habitat extent (Maps 3 – 5). Additionally, the complex and interacting factors that would be acting at this scale may obscure relationships with large scale habitat features, such as percent vegetated wetland. Concentrations of methylmercury entering affected waterways and the base of the food web reflect an integrative signal derived from a combination of biogeochemical and hydrologic processes, including soil adsorption and erosion, overland runoff, drainage, and microbial methylation and degradation. Furthermore, some of the previous results that have indicated relationships of vegetated wetland area to fish mercury have done so in wetlands of much smaller area (*e.g.,* < 10 km^2; St Louis *et al.* 1996). In smaller wetland areas, variability in the integrative signal due to other factors may be reduced, leading to clearer relationships to wetlands at small sample sizes.

In contrast, on the proximal scale we found a relationship between vegetated wetland extent and mercury concentration in fish, but in the opposite direction from what previous researchers observed (St Louis *et al.* 1996, St Louis *et al.* 2004, Castro *et al.* 2007). We observed that channel catfish mercury concentration was significantly related ($p = 0.04$, $R^2 = 0.37$) to vegetated wetland within a 5-km-radius buffer (Figure 2). Channel catfish mercury concentration decreased with increasing wetland extent. This finding is consistent with other recent studies suggesting that some wetlands in the Bay-Delta watershed are not necessarily net exporters of methylmercury, but rather mercury sinks (Stephenson *et al.* 2006). Our results on this subject should be viewed with caution, due to the small sample size ($n = 12$), the single species, and considering that wetland extent is simply one gross attribute of wetlands. Furthermore, all locations that were selected for the catfish analysis were lakes and reservoirs. It is therefore conceivable that the observed relationship may only occur in such locations with similar hydrology and biogeochemical processes. A larger sample size of locations that are more hydrodynamic (*e.g.,* main stem rivers) would show if this trend persists across habitat types (see largemouth bass results below).

The absence of a similar relationship between fish mercury and vegetated wetland extent in largemouth bass may reflect different exposure scales or dietary habits compared to channel catfish. Largemouth bass are considered to have a home range of less than a few kilometers (CDFG unpublished tagging data), whereas Moyle (2002) suggests that channel catfish make daily movements between habitats. Channel catfish are benthic feeders that prefer highly oxygenated waters, and seek river cuts, log jams, and dark pools during the day, and riffles at night (Moyle 2002). Becker (1983) also suggested that channel catfish make long migrations up and downstream of rivers. Largemouth bass are non-migratory, remaining close to rivers-edge for most of the season, feeding on crayfish and small fishes. Recently, Marvin Di-Pasquale

(2005) suggested that methylmercury bioaccumulation is predominantly governed by water chemistry. Therefore, regional differences in feeding behavior and food web structure may play secondary roles to the quantity of methylmercury entering the base of the food web (Cabana and Rasmussen 1994, Kidd *et al.* 1995). Given that most of the locations monitored for channel catfish were also sampled for largemouth bass, there must be some underlying difference between their exposures to explain non-significant results in one species relative to the other.

Comparing Mercury Concentrations between Hydrologic Sub-areas in the Delta

Largemouth bass and white catfish mercury concentrations varied significantly between regions defined by the Delta Methylmercury TMDL Hydrologic Sub-area delineations (Figures 3 and 4). Largemouth bass mercury concentrations in the Cosumnes/Mokelumne region were significantly higher than in other sub-areas of the Delta. There were inadequate samples to statistically differentiate this sub-area from the others for white catfish, though the single sample present was relatively high. In both species, the Central Delta clearly exhibited the lowest concentrations (Grenier *et al.* 2006), with the West Delta a close second. These results indicate some structure to the variation in fish mercury concentrations between sub-areas. Thus, we included this term in the models relating wetland and aquatic habitat extent to fish mercury.

Fish Mercury and Habitat Features in the Delta (Hypothesis H3)

In general, habitat features were not related to fish mercury concentration when the habitat characteristics were averaged across Delta sub-areas (Figures 5 and 6; Appendix Table 2). This was despite obvious differences in vegetation (Map 9) and flooding frequency (Map 10) among sub-areas. The greatest extent of aquatic habitat occurred in the West Delta (32%) where average mercury concentrations were low. Notably, the sub-area that showed the highest mercury concentrations (Figure 3; Cosumnes/Mokelumne) had the highest ratios of both vegetated wetland to non-vegetated aquatic habitat (10%:3%) and of temporarily inundated to permanently flooded habitat (8%:4%). Vegetated wetlands and temporarily inundated areas have been hypothesized to relate to higher methylmercury entering aquatic food webs (St Louis *et al.* 1996, Snodgrass *et al.* 2000). However, on a sub-area scale, only non-vegetated aquatic habitat extent was significantly related to largemouth bass mercury (Figure 5c; $p = 0.05$, $R^2 = 0.67$). White catfish concentrations did not relate to any habitat features at the sub-area scale (Figure 6). Except for the largemouth bass result, these analyses suggest that sub-area differences in the habitat features did not significantly influence fish mercury concentration.

None of the habitat features in 1-km- and 5-km-radius buffers correlated to largemouth bass mercury concentration (Map 11; Figures 7 - 10; Appendix Table 3). The significant relationship between non-vegetated aquatic habitat extent and fish mercury evident at the sub-area scale was not repeated at these proximal scales. Map 11 depicts San Joaquin River at Howard Road, where less than 5% of the buffer consists of aquatic habitat, yet the mercury concentration approached 1.0 µg/g. These significant main effects for hydrologic sub-area may be related to differences in water source, or other factors, with a coincidental correlation with percent non-vegetated aquatic habitat (see below for further discussion).

Habitat features did not relate to white catfish concentrations either. However, white catfish (1-km scale) results did indicate that the relationship between fish mercury and aquatic habitat extent varied between sub-areas (Figures 11 - 14). Significant interaction terms between both vegetated wetland area and temporarily flooded aquatic habitat (Figures 12 and 14; Map 12) and hydrologic sub-area indicated that these features were dependent on the sub-area (Appendix Table 3). Figures 12 and 14 illustrate that the interaction was most likely driven by the strong negative correlation in Sacramento River, which differed from the weakly positive correlation in San Joaquin River and Yolo Bypass, and the absence of a correlation in the Central Delta.

It is unclear which factors may be varying significantly between sub-areas to explain the presence and absence of correlations observed in our habitat analyses. Mercury supply and geochemistry, and local wetland features could explain the strong correlations in the Sacramento sub-area. Sources of mercury to the Delta vary depending on flow conditions, and tributary inputs (Wood *et al.* 2006), and conditions reported at the margins of the Delta (Sacramento River at Hood and San Joaquin River at Vernalis) suggest that binding of mercury to sediment would be different along these inputs compared to the Central Delta (Stepanauskas *et al.* 2005). However, wetland features comprising the vegetated wetland and temporarily inundated aquatic habitat (Tables 5 and 6) in the Sacramento sub-area did not differ significantly compared to other sub-areas, but it may be that the differences are simply not captured by the categories of features (vegetated and temporarily inundated) analyzed in this study. The small sample size of locations in some of the sub-areas may have led to the presence of correlations in the Sacramento sub-area that are not evident in the others. If such factors are varying significantly between sub-areas, the trends in Sacramento should be supported or refuted in other regions of the Delta. Therefore, the negative correlations in Sacramento, without further evidence, are considered only suggestive of a possible correlation with these features.

Conclusions

The work presented in this report is the first the authors are aware of to relate landscape features in the Bay-Delta watershed to fish mercury concentrations using GIS on a large regional scale. Fish mercury concentrations appeared to be influenced by many complex and interacting factors that hid any obvious correlations with atmospheric deposition, historic mining districts, and wetlands at the watershed scale. Some relationships were suggested by the proximal analyses, namely vegetated wetland and temporarily inundated habitat. Though based on small sample sizes, these results were surprisingly contrary to the prevailing notion that wetlands generally increase methylmercury accumulation in the food web.

The lack of clear relationships in the available dataset is probably due to shortcomings in the data, rather than a true absence of influence of atmospheric deposition, mining, or wetlands, on spatial patterns in mercury accumulation. The state-wide historical bioaccumulation database contained few samples from outside the Delta watershed where a clear hydrologic connectivity to sources could be made, without reservoirs or other barriers. Inside the Delta, many areas had too few sampling locations to yield confident results. Additionally, important data gaps exist for key parameters of habitat and mining features that may affect mercury cycling and bioaccumulation in fish. NWI had many areas of missing or incomplete coverage. The MRDS lacked information on quantities mined and loadings to the aquatic environment.

Future attempts to relate landscape features with mercury in the food web at the watershed scale should focus on watersheds with a stronger linkage between landscape features and mercury in fish and fewer confounding factors. The best areas for this would be at or above the lakes and reservoirs. The State Water Resources Control Board's Surface Water Ambient Monitoring Program is currently planning a statewide survey of mercury and other pollutants in sport fish of California lakes and reservoirs. Approximately 300 lakes and reservoirs will be sampled in 2007 and 2008. This effort will yield a dataset that should provide a much better basis for conducting a future assessment of the influence of atmospheric deposition, mines, wetlands, and perhaps other landscape-scale factors on mercury accumulation in sport fish. The data compilation and analytical framework developed in the present study would provide a foundation for such a future assessment.

Acknowledgements

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Table 1. Studies included in the SWAMP and FMP 2005 review datasets.

Short Name	*Agency*	*Contact*	*Recent Report*
CalFed	CalFed	Jay Davis	Davis, J.A., B.K. Greenfield, G. Ichikawa, and M. Stephenson. 2004. Mercury in Sport Fish from the Delta Region (Task 2A). Final Report submitted to the CALFED Bay-Delta Program for the Project: An Assessment of the Ecological and Human Health Impacts of Mercury in the Bay-Delta Watershed. 63 pp.
CFCP	OEHHA	Del Rasmussen	Gassel, M., Brodberg, R.K., and Roberts, S. 2002. The Coastal Fish Contamination Program: Monitoring of Coastal Water Quality and Chemical Contamination in Fish and Shellfish in California in California and the World Ocean '02: Revisiting and Revising California's Ocean Agenda. 977-990.
DWR Reservoir	DWR	Glen Pearson	Boles, J. 2004. Mercury Contamination in Fish from Northern California Lakes and Reservoirs. Department of Water Resources.
NFTS	EPA	Michael Walsh	CSC Environmental. 2005. Quality Assurance Report for the National Study of Chemical Residues in Lake Fish Tissue: Analytical Data for Years 1 through 4. US EPA. 57 pp.
RMP	SFEI	Jay Davis	Greenfield, B.K., Davis, J.A., Fairey, R., Roberts, C., Crane, D., and Ichikawa, G. 2005. Seasonal, inter-annual, and long-term variation in sport fish contamination, San Francisco Bay. Science of the Total Environment 336:25-43.
SRWP	SFEI	Jay Davis	LWA, 2004. Sacramento River Watershed Program. Annual Monitoring Report 2002-3.
TSMP	SWRCB	Del Rasmussen	Crane, D.B. et al. 2004. Toxic Substances Monitoring Report, 2000-01 Data Report.
UCDavis1	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, J.E. Reuter, C.R. Goldman. 1999. Lower Putah Creek 1997-1998 Mercury Biological Distribution Study. Dept. of Environmental Science and Policy, University of California, Davis.
UCDavis2	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, J.E. Reuter, and C.R. Goldman. 1997. Cache Creek Watershed Preliminary Mercury Assessment, Using Benthic Macro-Invertebrates. Division of Environmental Studies, University of California, Davis Final Report.
UCDavis4	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, J.E. Reuter, and C.R. Goldman. 1997. Gold mining impacts on food chain mercury in northwestern Sierra Nevada streams (1997 revision). In Sacramento River Mercury Control Planning Project. Larry Walker and Associates (editors). Final project report prepared for Sacramento Regional County Sanitation District. Davis, CA.
UCDavis5	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, T.H. Suchanek, R.D. Weyand, and A.M. Liston. 2002. Mercury Bioaccumulation and Trophic Transfer in the Cache Creek Watershed, California, in Relation to Diverse Aqueous Mercury Exposure Conditions. CALFED Mercury Program Draft Final Project Report.

Table 1. continued

Short Name	*Agency*	*Contact*	*Recent Report*
UCDavis6	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, and J.E. Reuter. 1996. Marsh Creek Watershed - 1995 Mercury Assessment Project. Study and report conducted for Contral Costa County, California. Final report.
UCDavis7	UC Davis	Darell Slotton	Slotton, D.G., S.M. Ayers, and J.E. Reuter. 1998. Marsh Creek Watershed - Mercury Assessment Project: Third Year (1997) Baseline Report with 3-Yr Review of Selected Data. Study and report conducted for Contral Costa County, California.
UCD Clear Lake	UC Davis	Darell Slotton	No reference available.
USGS Natoma	USGS	Michael Saiki	Saiki, M.K., Slotton, D.G., May, T.W., Ayers, S.M., and Alpers, C.N. 2004. Summary of Total Mercury Concentrations in Fillets of Selected Sport Fishes Collected during 2000-2003 from Lake Natoma, Sacramento County, California: USGS Data Series 103. 21 p.
USGS Trinity	USGS	Jason May	May, J.T., Hothem, R.L., and Alpers, C.N., 2005, Mercury concentrations in fishes from select water bodies in Trinity County, California, 2000-2002: U.S. Geological Survey Open-File Report 2005-1321.
USGS1	USGS	Jason May	May, J.T., Hothem, R.L., Alpers, C.N., and Law, M.A. 2000. Mercury Bioaccumulation in Fish in a Region Affected by Historical Gold Mining: The South Yuba River, Deer Creek, and Bear River Watersheds, California, 1999: USGS Open-File Report 00-367. 30 p.
FMP 2005	SFEI	Letitia Grenier	Grenier, L.G., Melwani, A., Hunt, J.A., Bezalel, S.N., Davis, J.A., Ichikawa, G., Jakl, B., Heim, W., Bonnema, A., and M. Gassel. 2006. Final Technical Report California Bay-Delta Authority Fish Mercury Project Year 1 Annual Report. Oakland, CA.

Table 2. Summary of fish mercury dataset. Sample size, range and median mercury concentrations (wet weight) and sampling years are given for each species. Limits were applied to account for known variation in fish mercury by size. Data included in this report were collected from within the CalFed boundary between 1991 and 2005.

Species	*Number of Stations*	*Number of Samples*	*Min Hg (ug/g)*	*Max Hg (ug/g)*	*Median Hg (ug/g)*	*From Year*	*To Year*
Channel Catfish	52	112	0.028	1.610	0.236	1991	2005
Largemouth Bass	121	535	0.060	2.080	0.354	1992	2005
White Catfish	56	249	0.039	1.211	0.246	1991	2005

Table 3. Size limits for the three fish species analyzed. Limits were applied to account for known variation in fish mercury by size. Size limits were determined by applying the 75% rule (USEPA 2000) to all historical fish samples obtained from the SWAMP Bioaccumulation Database (SFEI, 2006) that fell within the CALFED Geographic Scope boundary. Data collected by the Fish Mercury Project in 2005/6 were also included.

Common Name	*Scientific Name*	*Size Limit (mm)*
White Catfish	*Ameiurus catus*	244 - 325
Largemouth Bass	*Micropterus salmoides*	273 - 364
Channel Catfish	*Ictalurus punctatus*	390 - 519

Table 4. Datasets used for analysis.

Name	*Data Collection and Processing Dates*	*Publication Date*	*Source*	*URL (if available)*
National Wetlands Inventory (NWI)	1977–2004	10/2006	U.S. Fish and Wildlife Service, 200610, CONUS_wet_poly: Classification of Wetlands and Deepwater Habitats of the United States. U.S. Department of the Interior, Fish and Wildlife Service, Washington, DC. FWS/OBS-79/31., U.S. Fish and Wildlife Service, Branch of Habitat Assessment, Washington, D.C.	http://www.fws.gov/nwi/
National Hydrography Dataset (NHD)	1978–2005	1999–2005	U.S. Geological Survey in cooperation with U.S. Environmental Protection Agency, USDA Forest Service, and other Federal , State and local partners, 2005 National Hydrography Dataset (NHD) – High Resolution, U.S. Geological Survey, Reston, Virginia.	http://nhd.usgs.gov/
Mineral Resources Data System (MRDS)	1968–2005	2005	U.S. Geological Survey, 2005, Mineral Resources Data System. U.S. Geological Survey, Reston, Virginia.	http://tin.er.usgs.gov/mrds/
Calwater 2.2.1	1973–1999	2004	California Interagency Watershed Mapping Committee, California Department of Forestry and Fire Protection, Tierra Data Systems, California, Department of Water Resources, State Water Resources Control Board, California Department of Fish and Game, and State of California Stephen P. Teale Data Center GIS Solutions Group, 20041118.	http://cain.nbii.gov/calwater/
Delta MeHg TMDL Sub-areas	NA	2006	Sacramento-San Joaquin Delta Estuary TMDL for Methylmercury, Draft Report for Scientific Review	http://www.waterboards.ca.gov/centralvalley/programs/tmdl/deltahg.html
Fish Mercury Bioaccumulation Database (SWAMP)	1990–2005	In prep.	SWAMP Bioaccumulation Database and FMP 2005 Report. See Table 1 for list of studies.	Email: jdavis@sfei.org or aroon@sfei.org

Table 5. Classification of vegetated and non-vegetated areas

Vegetation Types	*Non-vegetated Areas*
Emergent	Unconsolidated Bottom
Scrub-Shrub	Unconsolidated Shore
Forested	
Aquatic Bed	

Table 6. Classification of permanently flooded versus temporarily inundated areas

Permanently Flooded	*Temporarily Inundated*
Regularly Flooded	Seasonal-Tidal
Semi-permanently Flooded	Temporary-Tidal
Permanently Flooded	Intermittently Flooded
Semi-permanent-Tidal	Temporarily Flooded
Permanent-Tidal	Artificially Flooded
Intermittently Exposed	Saturated
	Seasonally Flooded / Saturated
	Seasonally Flooded/ Well-drained
	Seasonally Flooded

Figure 1. Mercury concentrations in largemouth bass from watersheds with and without mine influence (gold or mercury mines). Box ends represents the 25th and 75th percentiles, the horizontal line within the box represents the median value, and the whiskers show the minimum and maximum values. Black circles represent samples from the CalFed area and "X" represents samples from southern California.

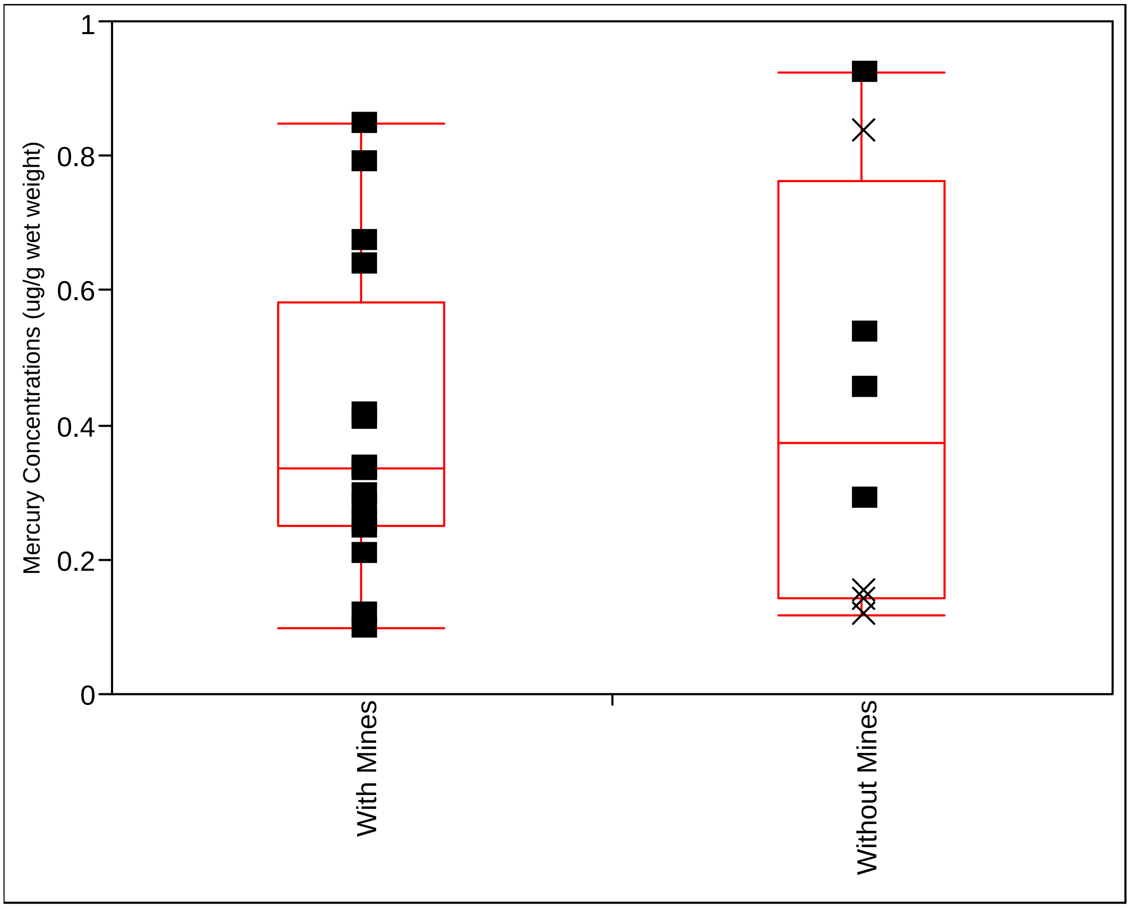


Figure 2. Relationship between mercury concentrations in channel catfish from lakes and reservoirs to percent total vegetated wetland within 5km radius. Size limits were applied (see Table 3). Data were log transformed for analysis and are presented on a linear scale.

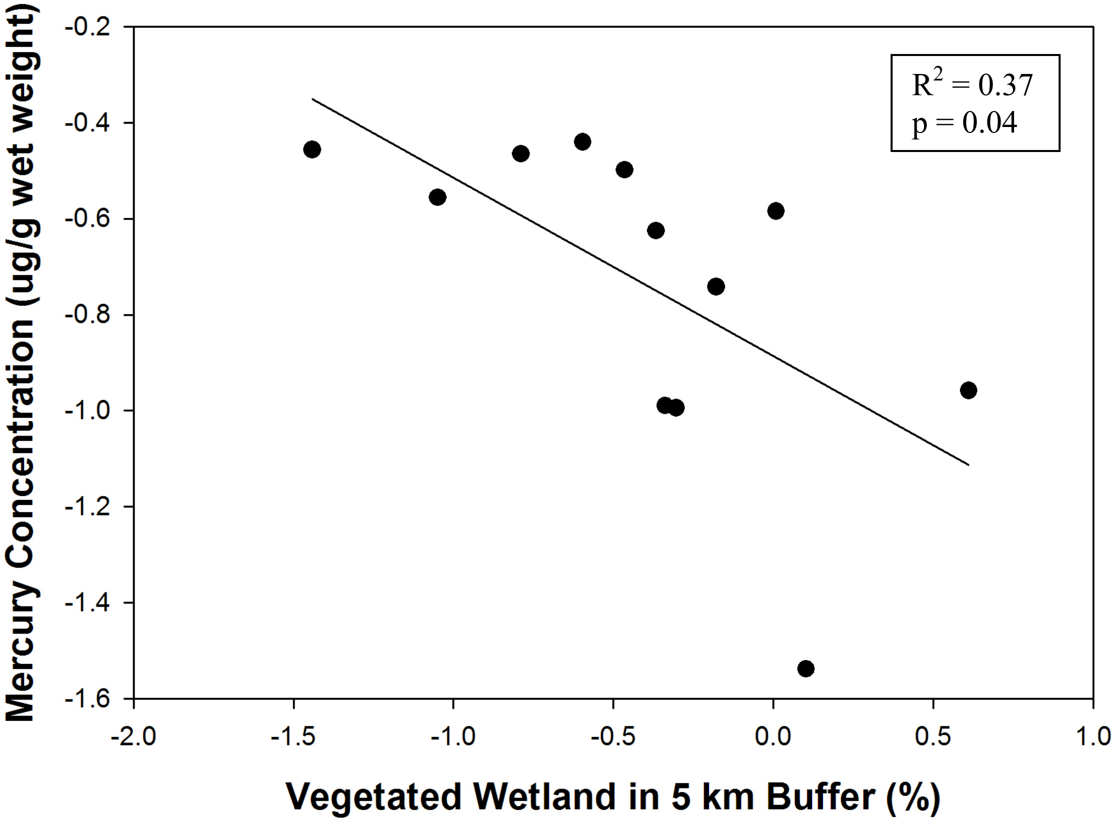


Figure 3. Mercury concentrations in largemouth bass in six sub-areas of the Delta. Box ends represents the 25th and 75th percentiles, the horizontal line represents the median value, and the whiskers indicate values within 1.5 interquartile ranges from the quartiles. Points beyond the whiskers are possible outliers in the data set. Sites not sharing the same letter are significantly different.

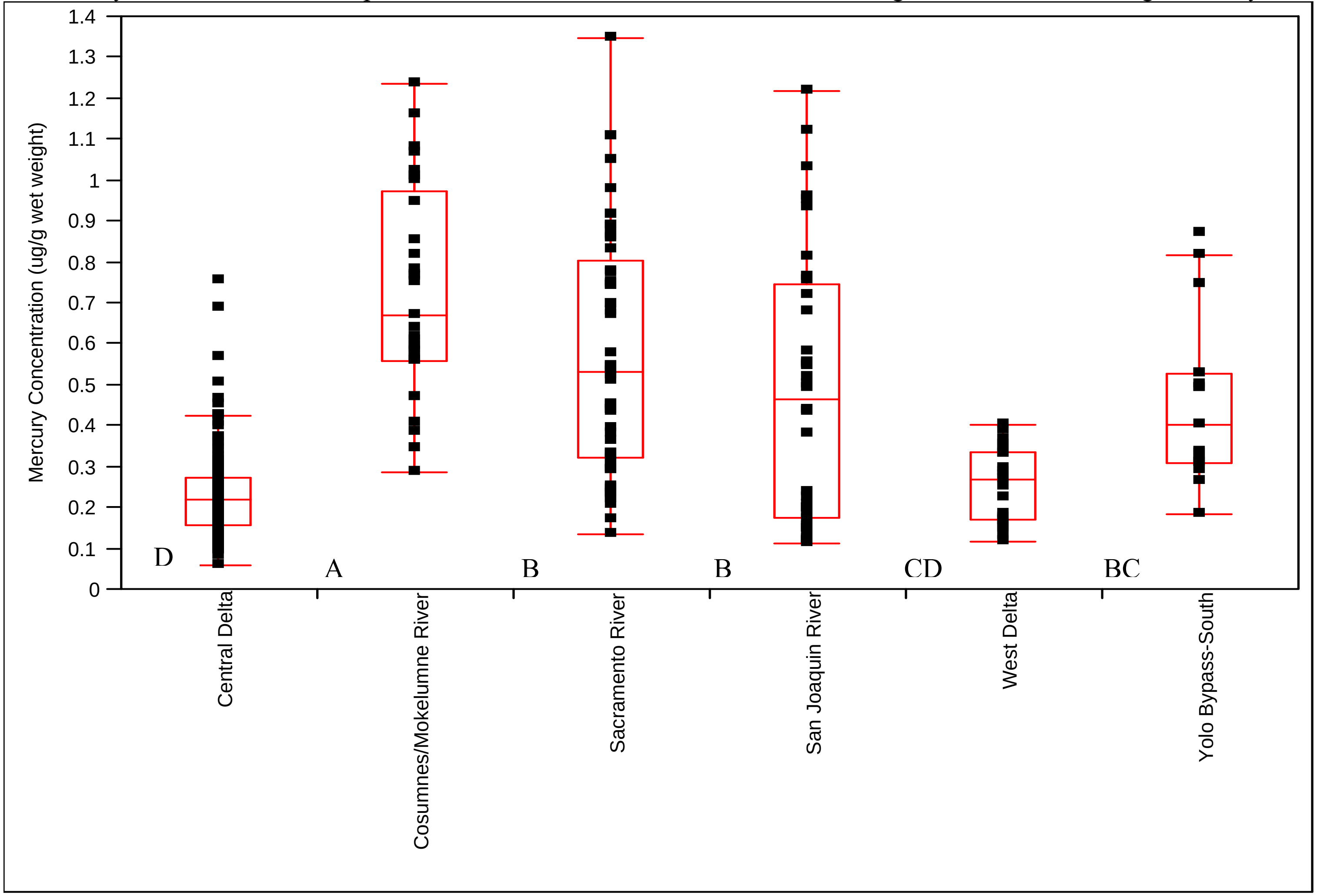


Figure 4. Mercury concentrations in white catfish in six sub-areas of the Delta. Box ends represents the 25th and 75th percentiles, the horizontal line represents the median value, and the whiskers indicate values within 1.5 interquartile ranges from the quartiles. Points beyond the whiskers are possible outliers in the data set. Sites not sharing the same letter are significantly different.

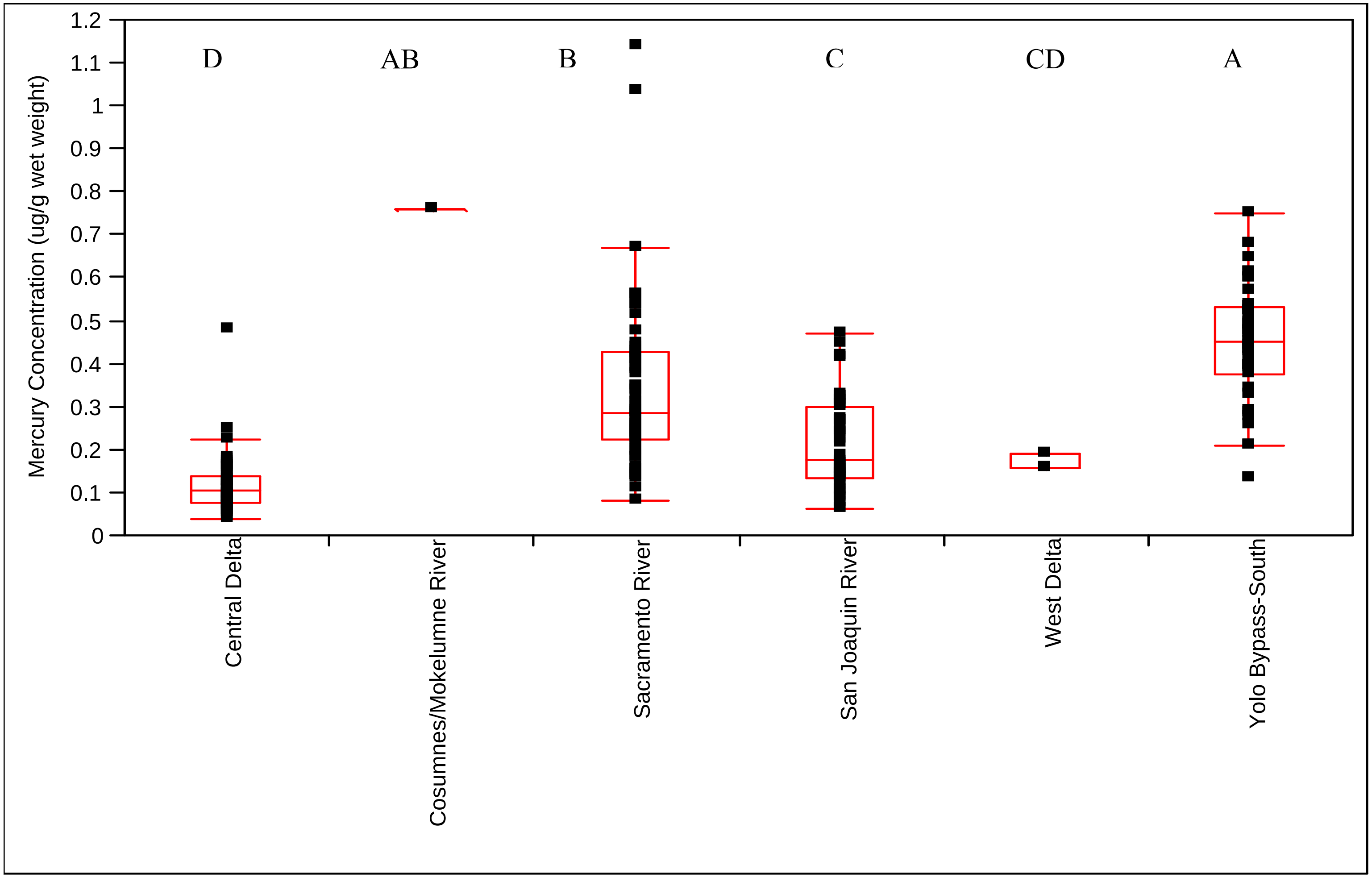


Figure 5. Wetland and aquatic habitat extent vs. mercury concentrations for largemouth bass in six sub-areas of the Delta: A) percent total aquatic habitat, B) percent vegetated wetland habitat, C) percent non-vegetated aquatic habitat, and D) percent temporarily inundated aquatic habitat. Only percent non-vegetated habitat had a statistically significant relationship with mercury for this species. Data were log transformed for analysis but are presented on a linear scale.

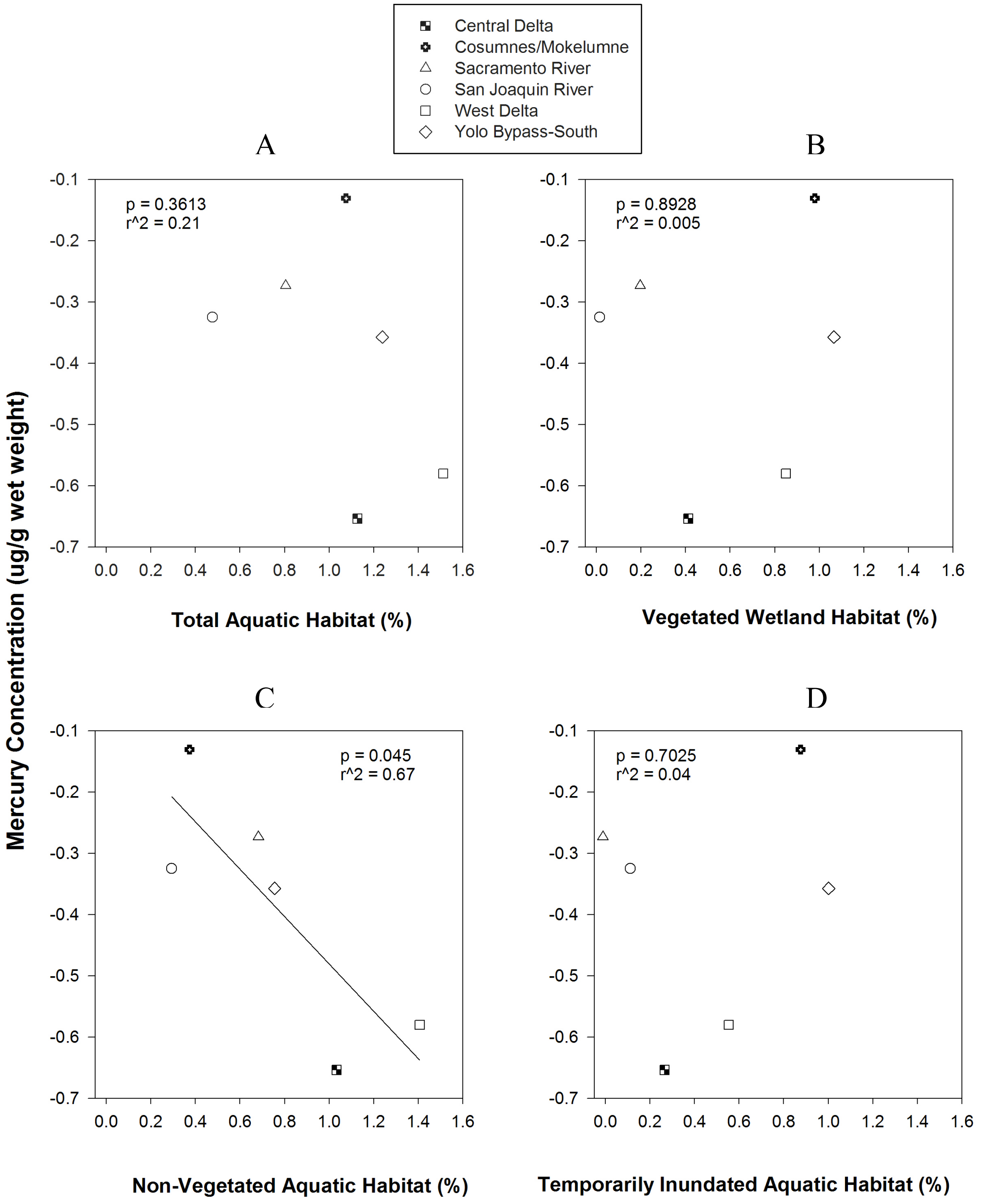


Figure 6. Wetland and aquatic habitat extent vs. mercury concentrations for white catfish in six sub-areas of the Delta: A) percent total aquatic habitat, B) percent vegetated wetland habitat, C) percent non-vegetated aquatic habitat, and D) percent temporarily inundated aquatic habitat. Only percent non-vegetated habitat had a statistically significant relationship with mercury for this species. Data were log transformed for analysis but are presented on a linear scale.

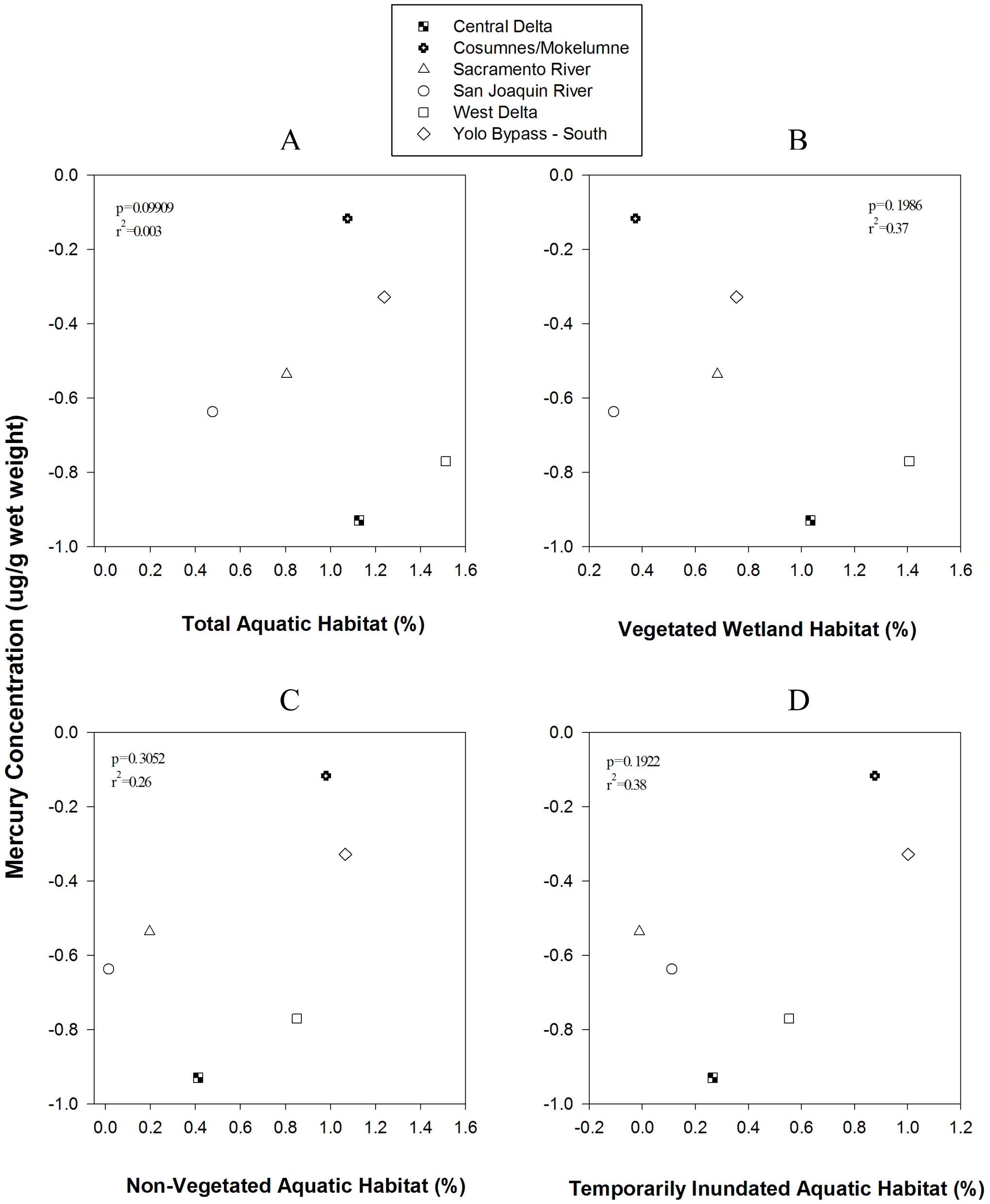


Figure 7 Relationship of percent total aquatic habitat in 1km radius buffer to largemouth bass mercury concentration for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

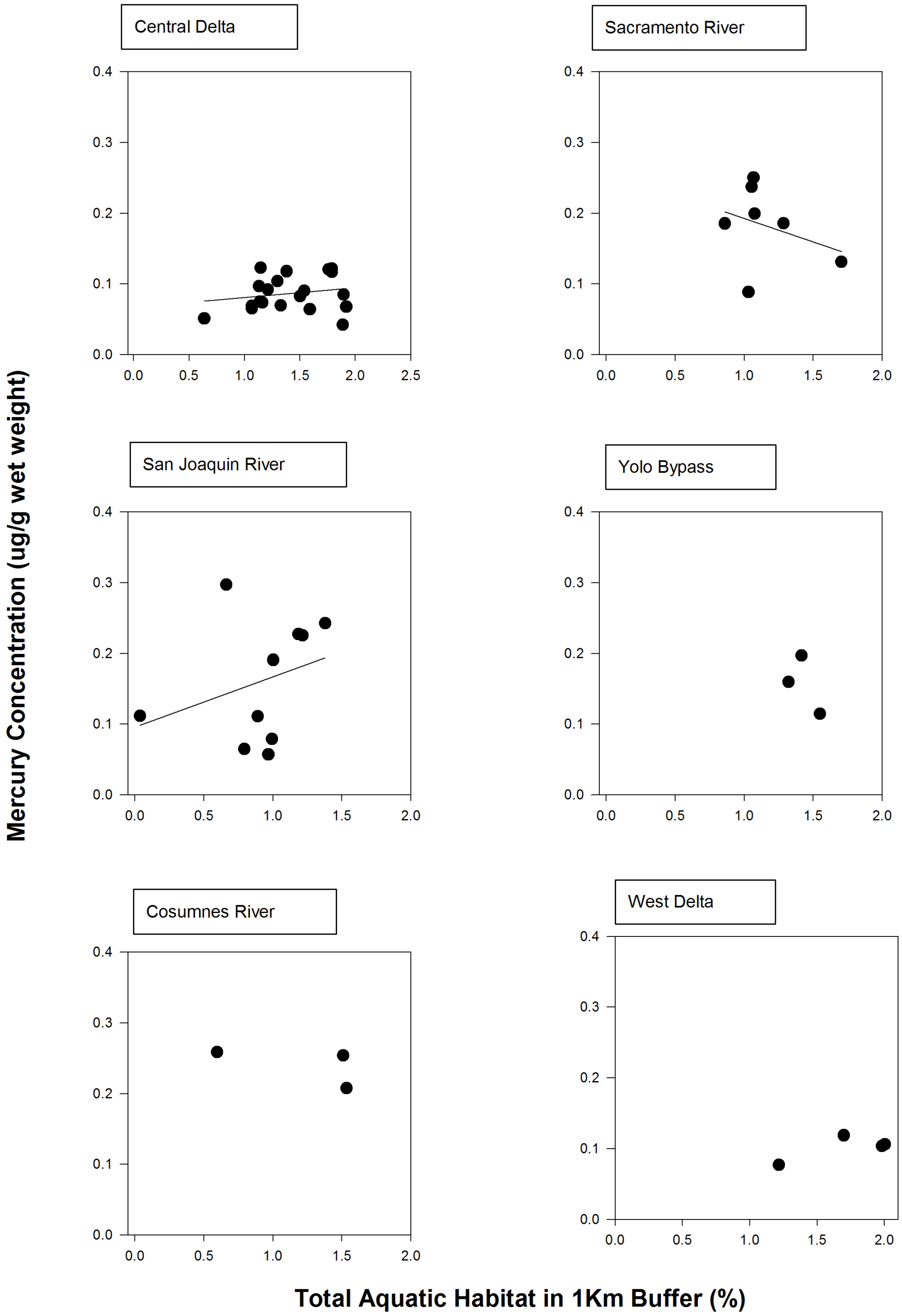


Figure 8. Relationship of percent vegetated wetland in 1km radius buffer to largemouth bass mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

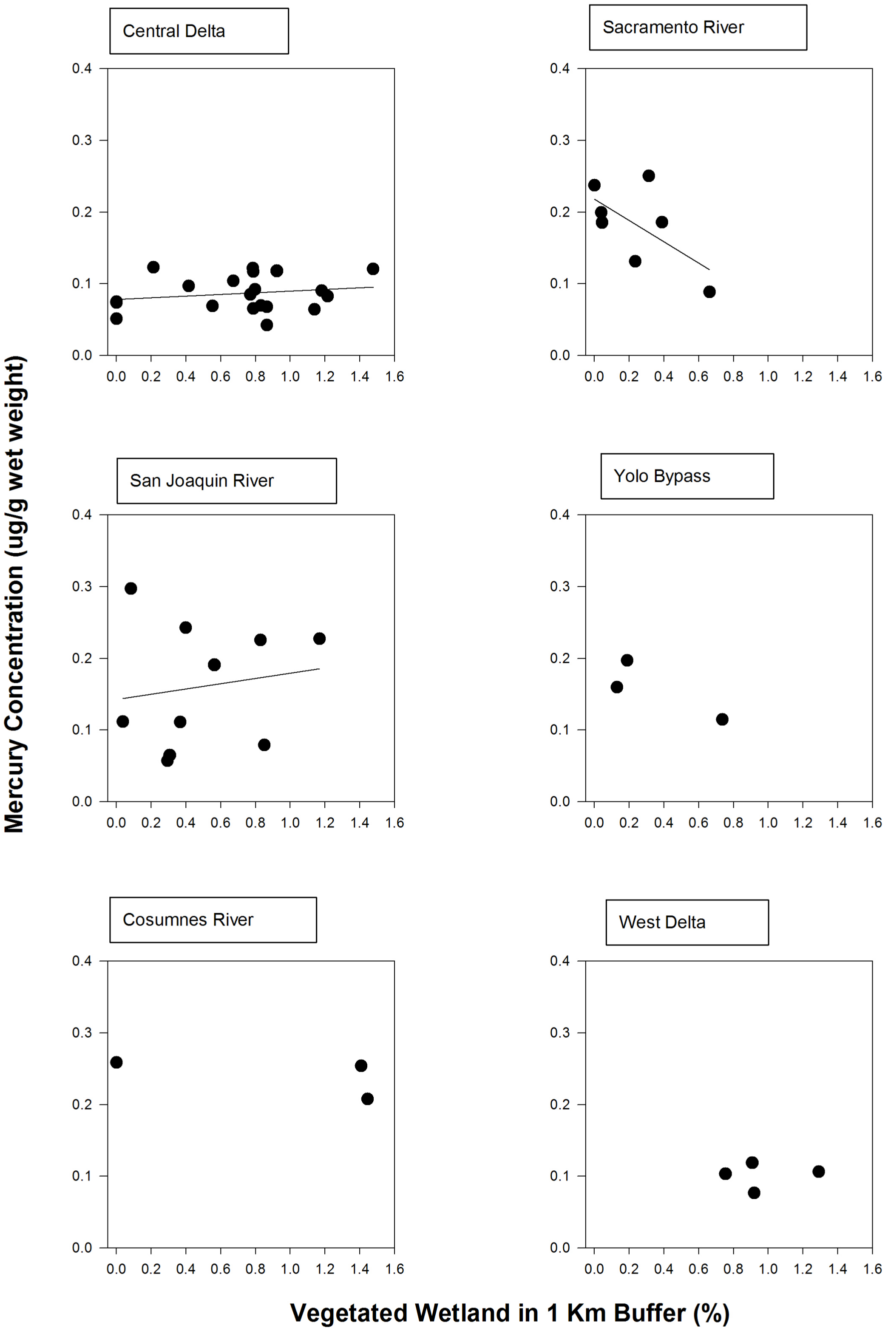


Figure 9. Relationship of percent non-vegetated aquatic habitat in 1km radius buffer to largemouth bass mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

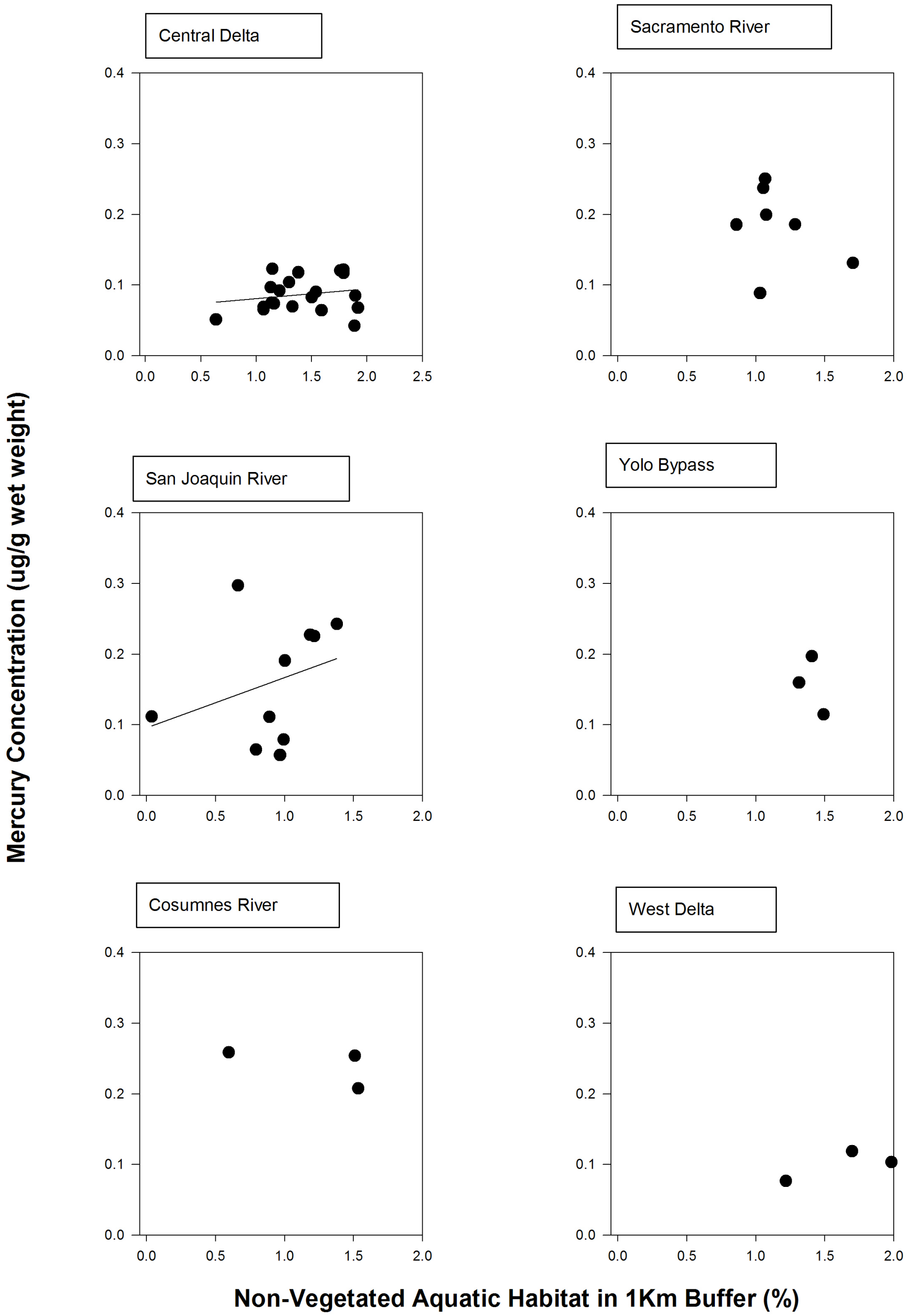


Figure 10. Relationship of percent temporarily inundated aquatic habitat in 1km radius buffer to largemouth bass mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

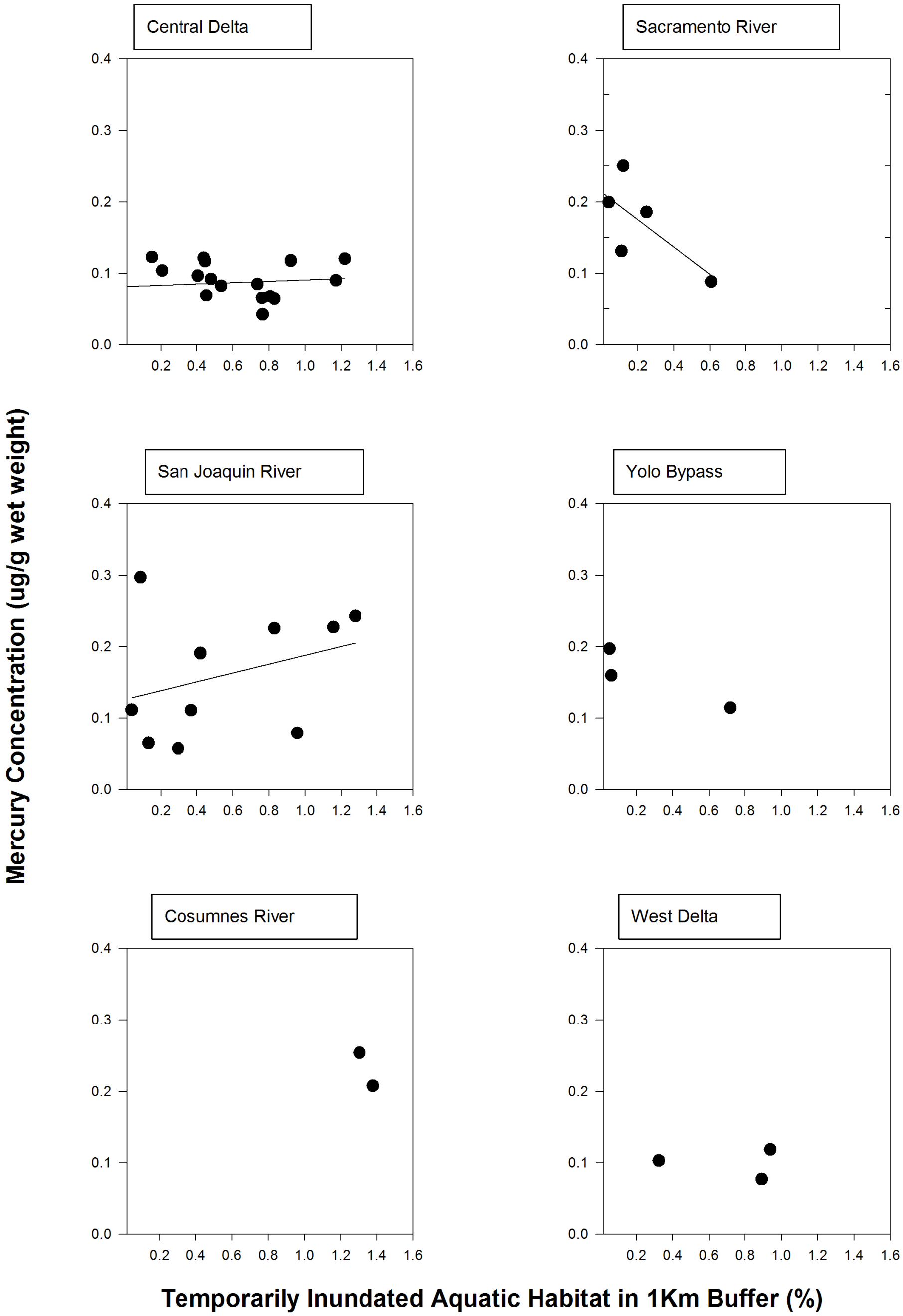


Figure 11. Relationship of percent total aquatic habitat in 1km radius buffer to white catfish mercury concentration for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

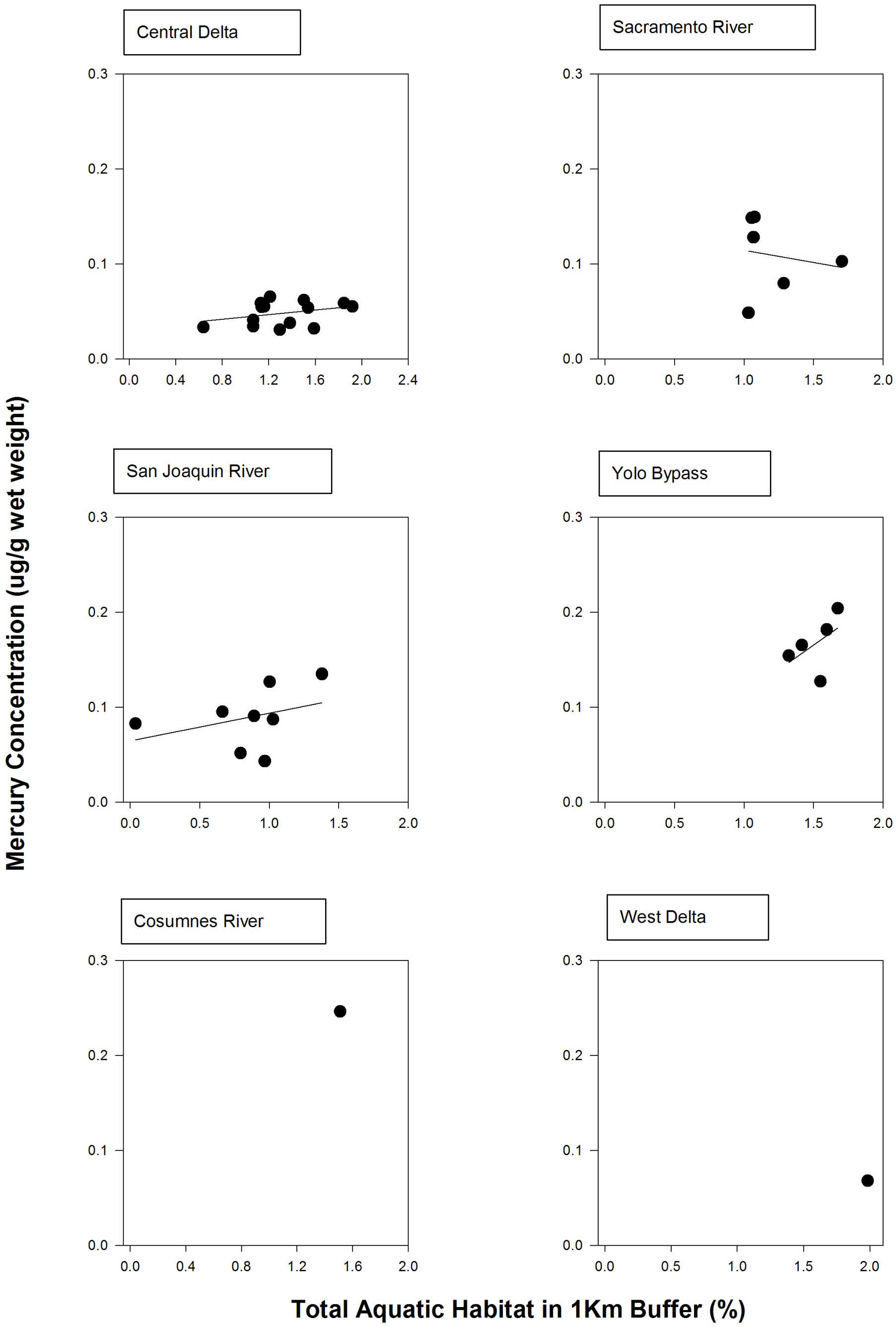


Figure 12. Relationship of percent vegetated wetland in 1km radius buffer to white catfish mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

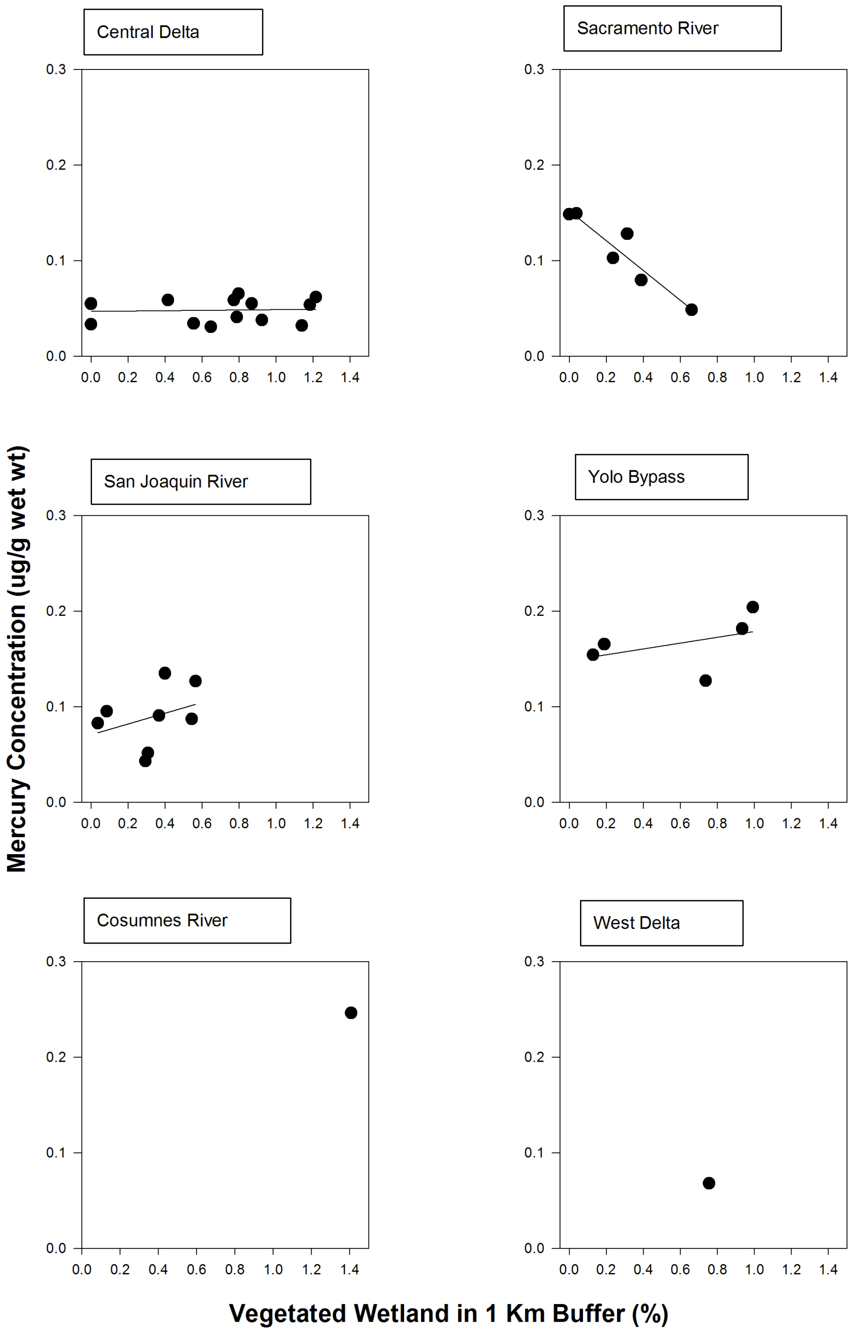


Figure 13. Relationship of percent non-vegetated aquatic habitat in 1km radius buffer to white catfish mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data were log transformed for analysis but are presented on a linear scale.

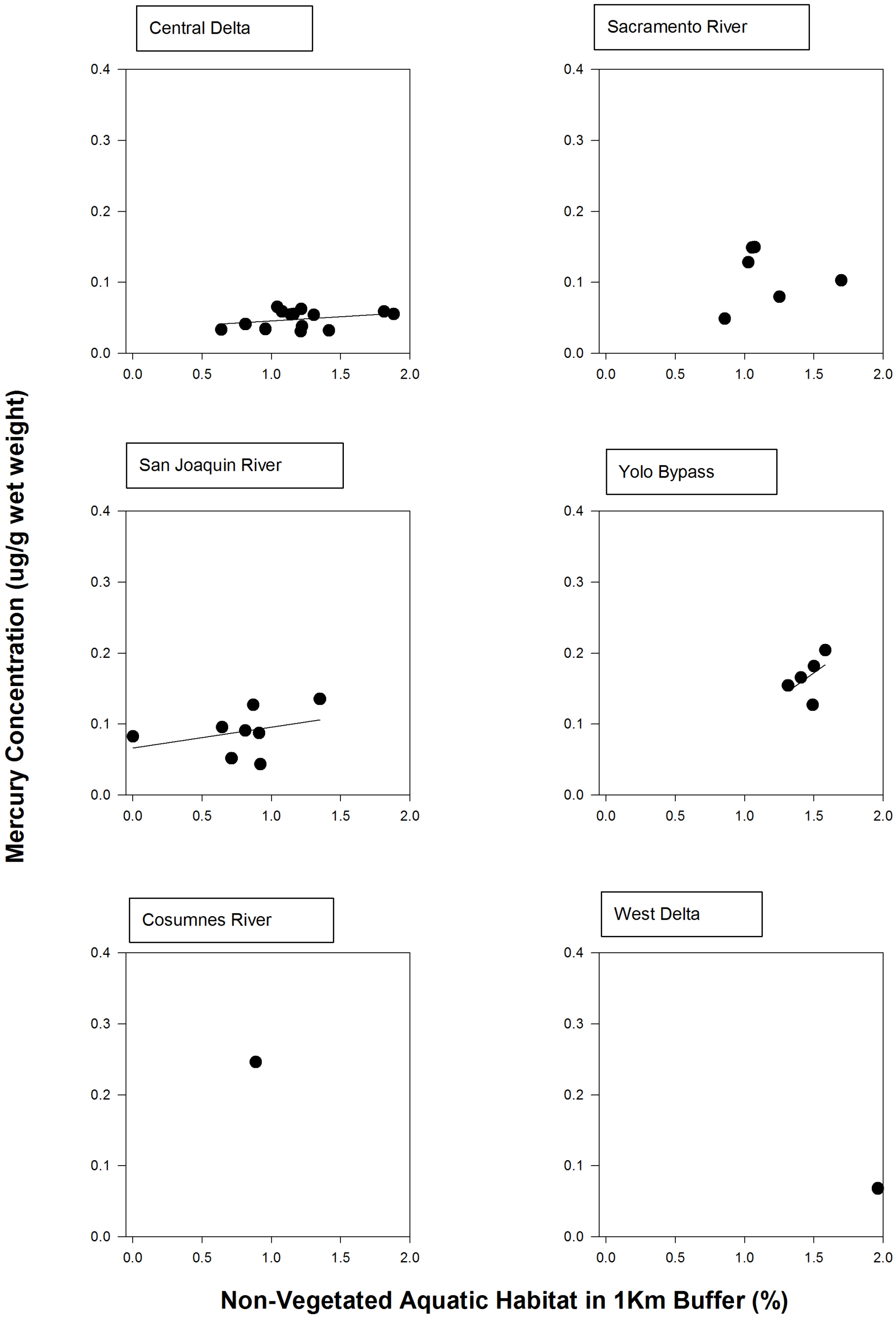
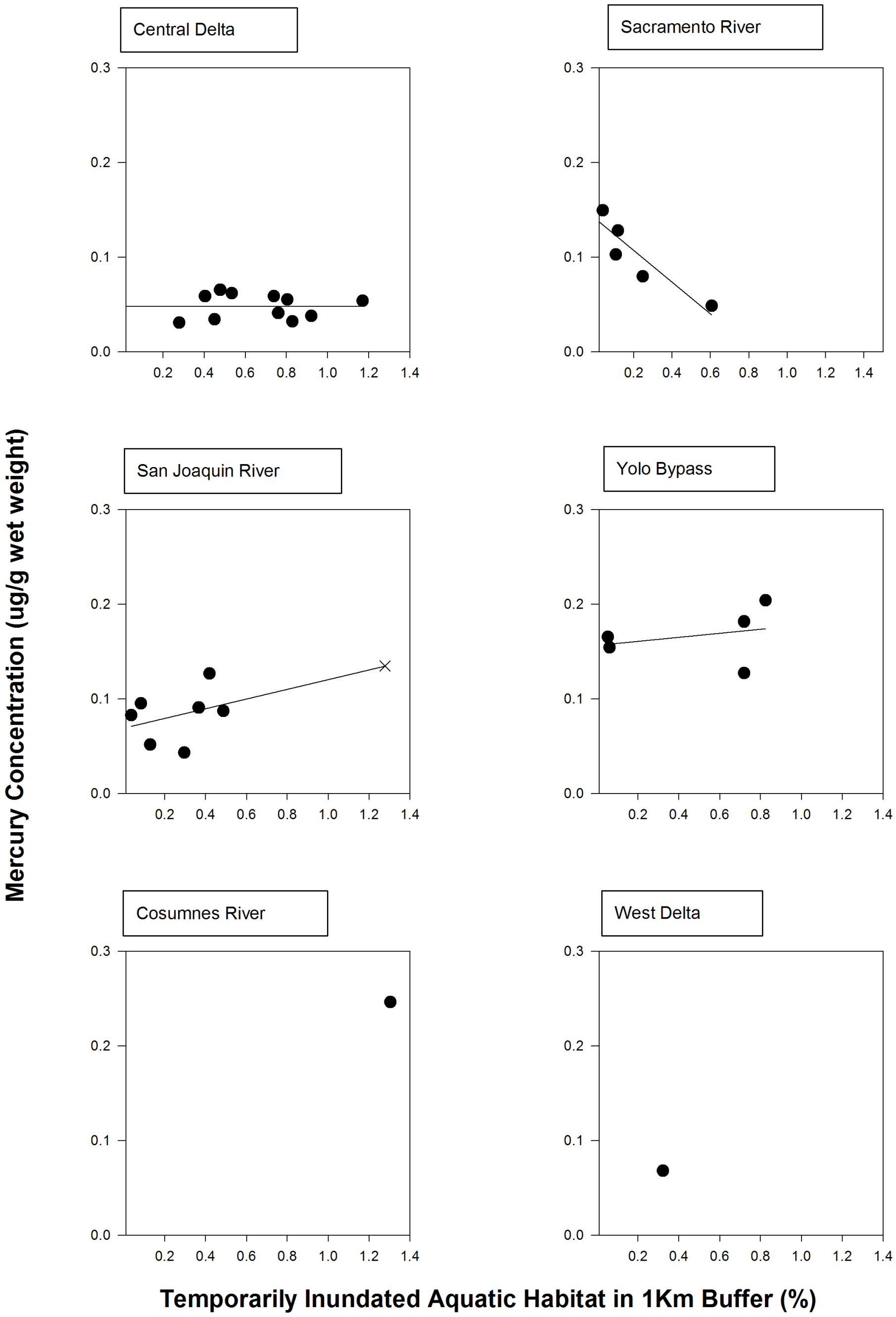


Figure 14. Relationship of percent temporarily inundated aquatic habitat in 1km radius buffer to white catfish mercury concentrations for each sub-area. Sub-areas showing trend lines were tested statistically: other sub-areas lacked sufficient data for statistical tests. Each data point represents the percent aquatic habitat and average mercury for a location within the designated sub-area. Data points marked by an "x" were not included in statistical tests. Data were log transformed for analysis but are presented on a linear scale.



MAP LEGENDS

Map 1. Sampling Locations of Largemouth Bass, Channel Catfish, and White Catfish within CalFed Boundary Study Area, 1990-2005. All historical sport fish sampling locations shown are within the CalFed geographic area.

Map 2. GIS and Fish Mercury Datasets within CalFed Boundary Study Area Used for Analysis. GIS layers with data for historical gold and mercury mines, wetlands, other surface waters (ponds, lakes, rivers, streams), and fish sampling locations that were included in analysis.

Map 3a. Fish Watersheds for Largemouth Bass Sampling Sites from Tributaries (American River at Discovery Park 1 and Merced River at Hatfield State Park). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 3b. Fish Watersheds for Largemouth Bass Sampling Sites from Tributaries (American River at Nimbus Dam and Mokelumne River between Beaver and Hog Slough). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). NWI coverage for wetlands and other aquatic habitats was not calculated in fish watersheds where more than 40% of the total area was missing NWI data. Mokelumne River between Beaver and Hog Slough provides an example of this situation, as 49.4% of the fish watershed is missing NWI data. Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 3c. Fish Watersheds for Largemouth Bass Sampling Sites from Tributaries (Feather River at Gridley and Feather River at Nicolaus). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 3d. Fish Watersheds for Largemouth Bass Sampling Sites from Tributaries (Stanislaus River at Caswell State Park and Tuolumne River at Shiloh Road). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 4. Fish Watersheds for Largemouth Bass Sampling Sites from Main Stem Rivers (Sacramento River at Butte City and San Joaquin River at Landers Avenue). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 5. Fish Watersheds for Largemouth Bass Sampling Sites from Lakes and Reservoirs (Lake Combie and New Melones Reservoir 2). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland (note that wetland data were not available for all areas). Purple dots designate gold and mercury mines. Dashed lines indicate the mining area of influence within these fish watersheds. Mines not included within the area of mining influence were excluded due to lower frequency of mines/unit area (i.e., they were sparse and outside the main mine cluster). Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 6a. Fish Watersheds with No Mines Present for Largemouth Bass Sampling Sites from Lakes and Reservoirs (Finnon Reservoir, Lafayette Reservoir, Lake Chabot, Anderson Reservoir). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland. Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 6b. Fish Watersheds with No Mines Present for Largemouth Bass Sampling Sites from Lakes in Southern California (Sherwood Lake, Westlake Lake, Malibu Lake, Lindero Lake). Red bars represent the average largemouth bass mercury concentration (µg/g wet wt) at each sampling location. Green regions represent areas of vegetated wetland. Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 7. Land Cover of Aquatic Habitats by Flooding Frequency within a 5-km-radius of Largemouth Bass Sampling Locations (Sacramento River at Butte City, New Melones Reservoir 2, and Mokelumne River between Beaver and Hog Slough). Colored circle (yellow, green, orange) in the center of the 5-km-radius buffer represents average largemouth bass mercury concentration (µg/g wet wt) at each site. Percent of permanently (blue) and temporarily (orange) flooded areas are noted graphically and in text for each sampling location. Mining locations are also noted graphically by dots and in text for each sampling location.

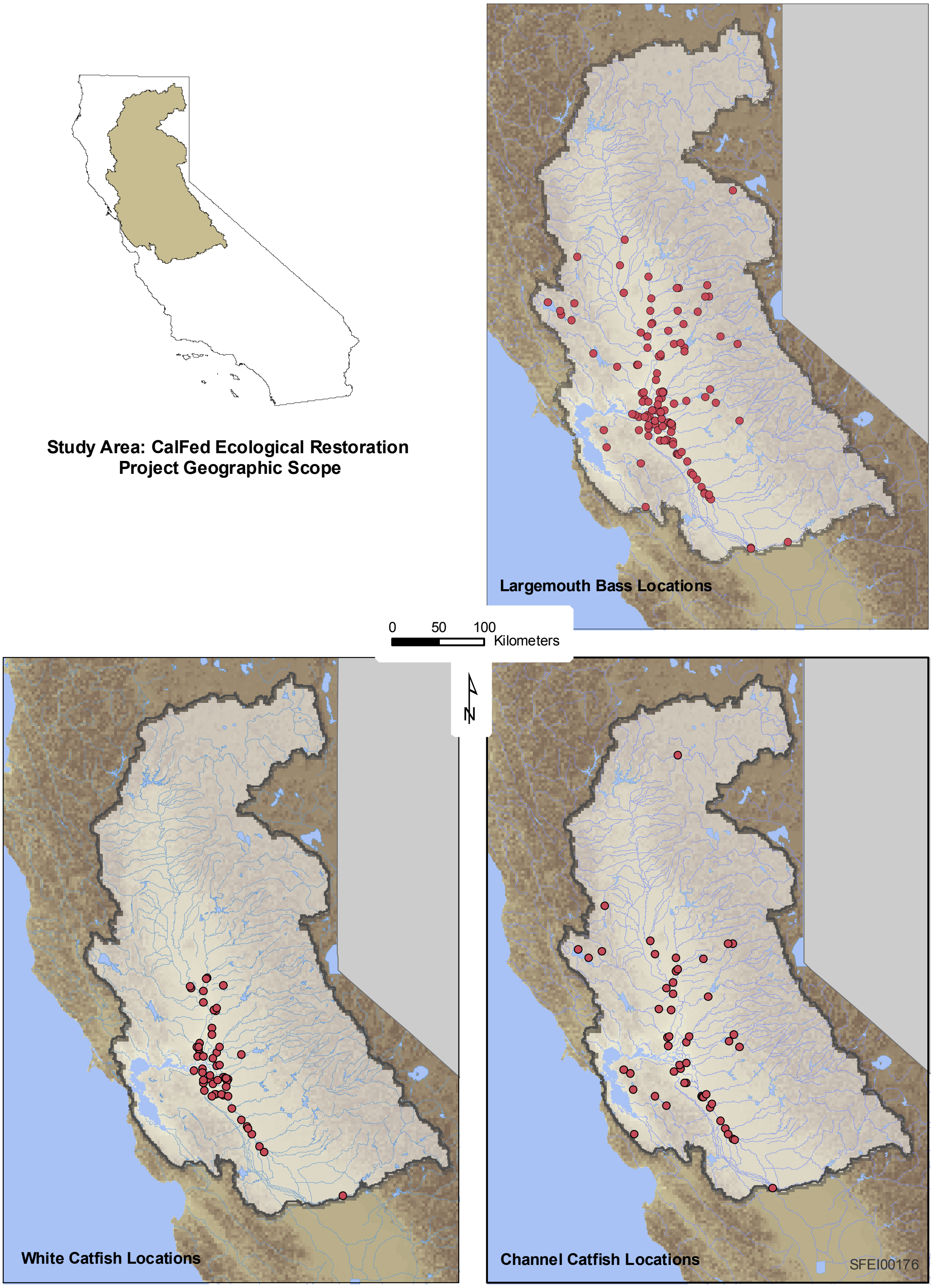
Map 8. Land Cover of Aquatic Habitats by Presence/Absence of Vegetation within a 5-km-radius of Channel Catfish Sampling Locations (Lake Britton near Highway 89, New Hogan Reservoir, and Stevens Creek Reservoir). Colored circle (yellow, green, orange) in the center of the 5-km-radius buffer represents average channel catfish mercury concentration (µg/g wet wt) at each site. Percent of permanently (blue) and temporarily (orange) flooded areas are noted graphically and in text for each sampling location. Mining locations are also noted graphically by dots and in text for each sampling location.

Map 9. Presence of Vegetation in Aquatic and Wetland Habitats in the Sacramento-San Joaquin Delta. Map shows areas of vegetated wetland (green) and non-vegetated aquatic habitat (orange) in the Delta hydrologic sub-areas (as defined by the Central Valley Regional Water Quality Control Board). Graph (inset) shows the percent of each type of habitat in each Delta sub-area. Wetland area has been augmented to enhance visibility (green regions). Actual percent of wetland area within the watershed is noted in the text box on the right side the map.

Map 10. Aquatic and Wetland Habitats by Flooding Frequency in the Sacramento-San Joaquin Delta. Map shows permanently flooded habitat (blue) and temporarily flooded habitat (orange) in the Delta hydrologic sub-areas (as defined by the Central Valley Regional Water Quality Control Board). Graph (inset) shows the percent of each type of habitat in each Delta sub-area.

Map 11. Land Cover of Aquatic Habitats by Presence/Absence of Vegetation within a 1-km-radius of Largemouth Bass Sampling Locations in Sacramento-San Joaquin Delta (Lost Slough 1, Mokelumne River d/s Cosumnes River 2, Franks Tract 3, and San Joaquin River/Howard Road). Colored circle (yellow, green and red) in the center of the 1-km-radus buffer represents average largemouth bass mercury concentration (µg/g wet wt). Percent of vegetated wetland (green) and non-vegetated aquatic habitat (orange) are noted graphically and in text.

Map 12. Land Cover of Aquatic Habitats by Flooding Frequency within a 1-km-radius of White Catfish Sampling Locations in Sacramento-San Joaquin Delta (Sacramento River/Hood, Cosumnes River 1, Old River/CV Pumps) Colored circle (yellow, green and red) in the center of the 1-km-radus buffer represents average white catfish mercury concentration (µg/g wet wt). Percent of permanently flooded (blue) and temporarily flooded (orange) habitats are noted graphically and in text.



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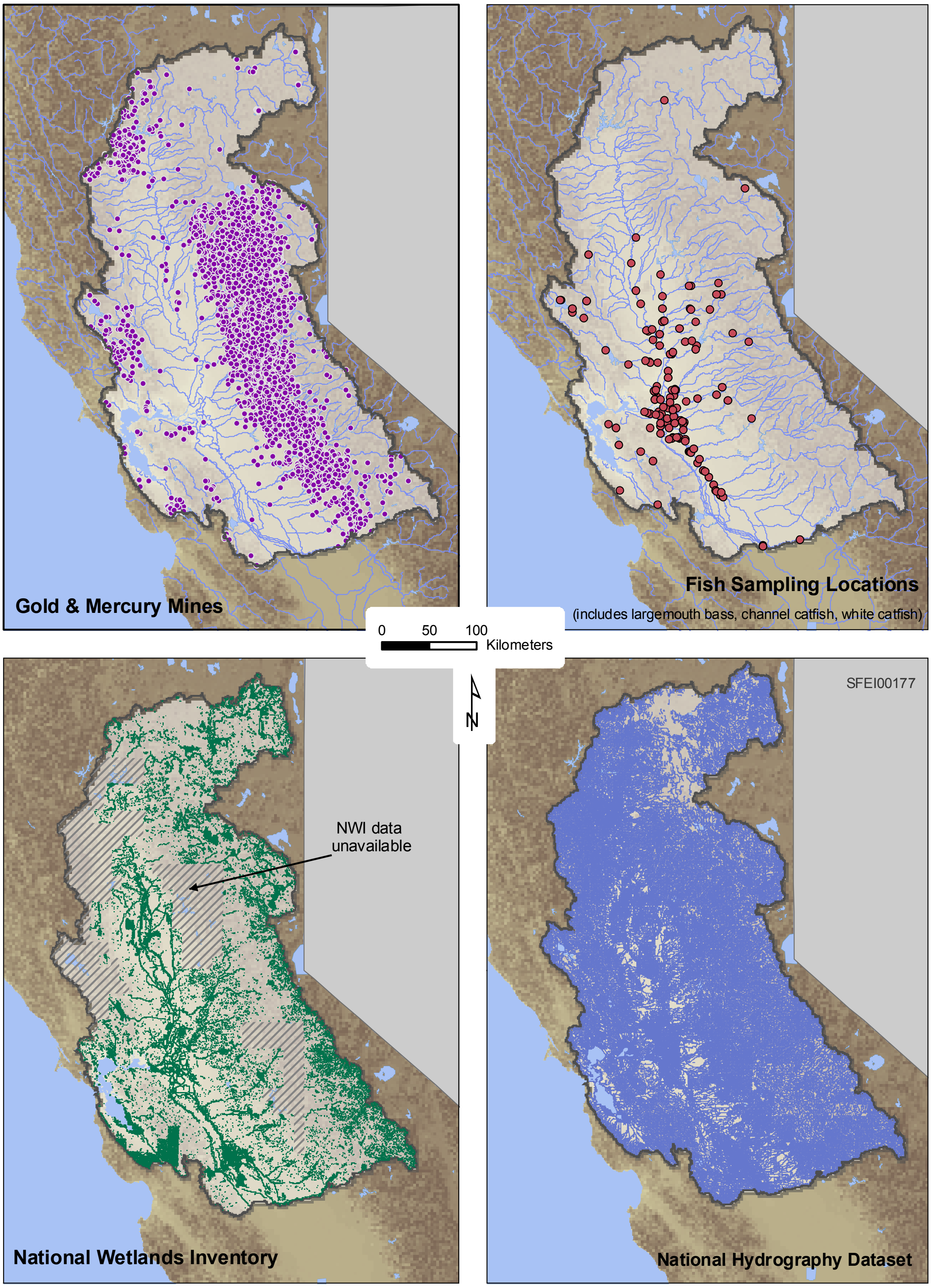

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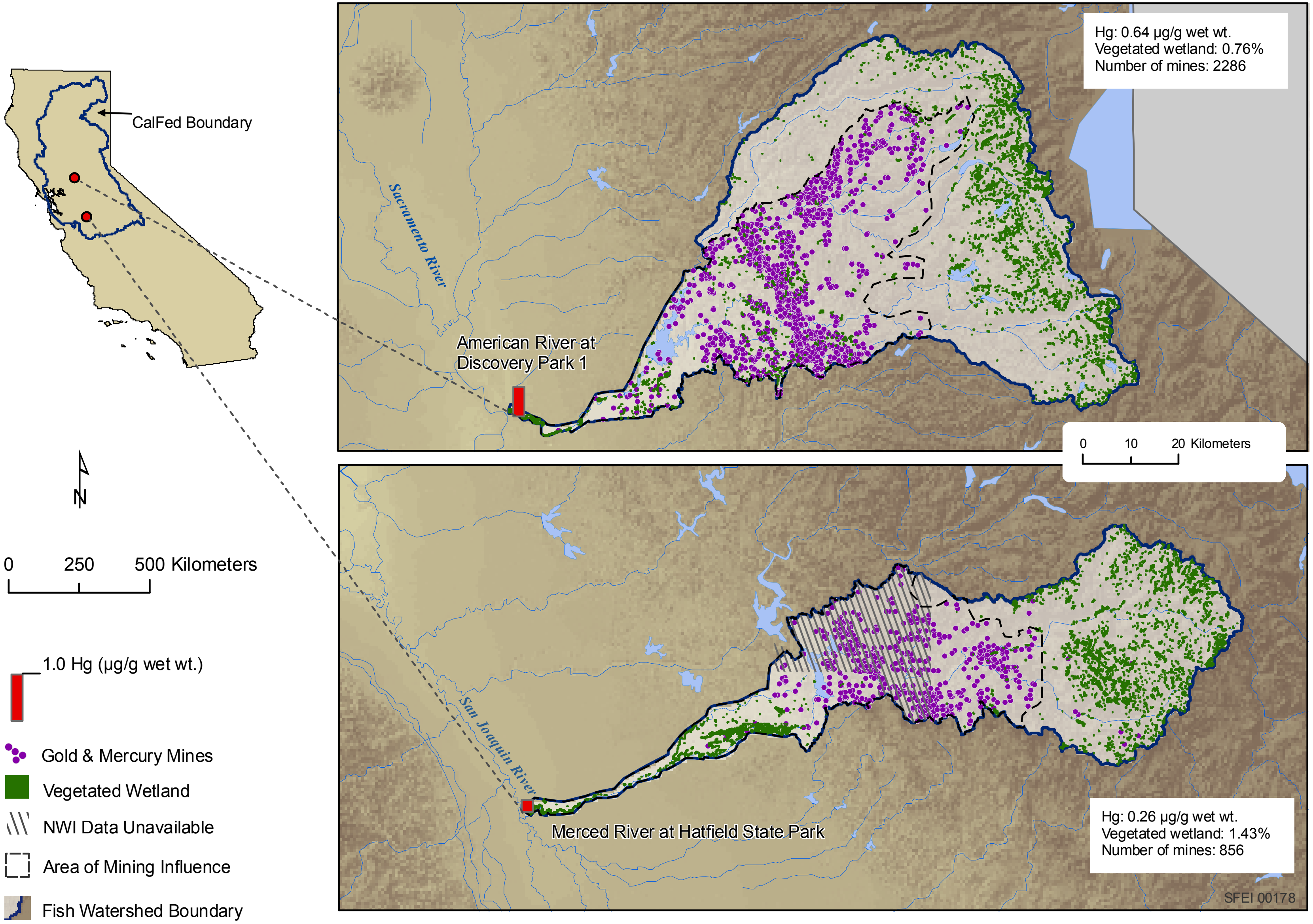

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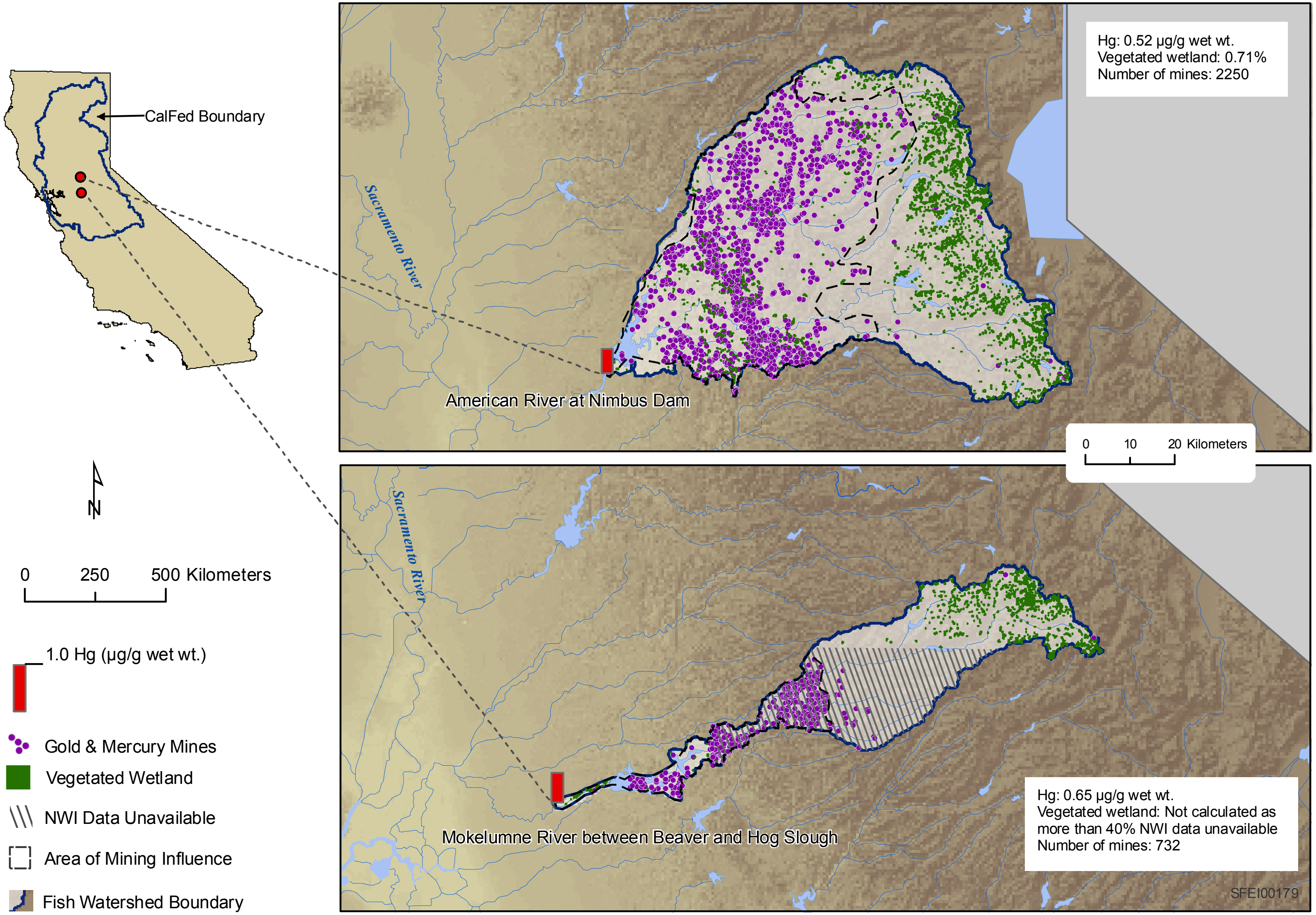

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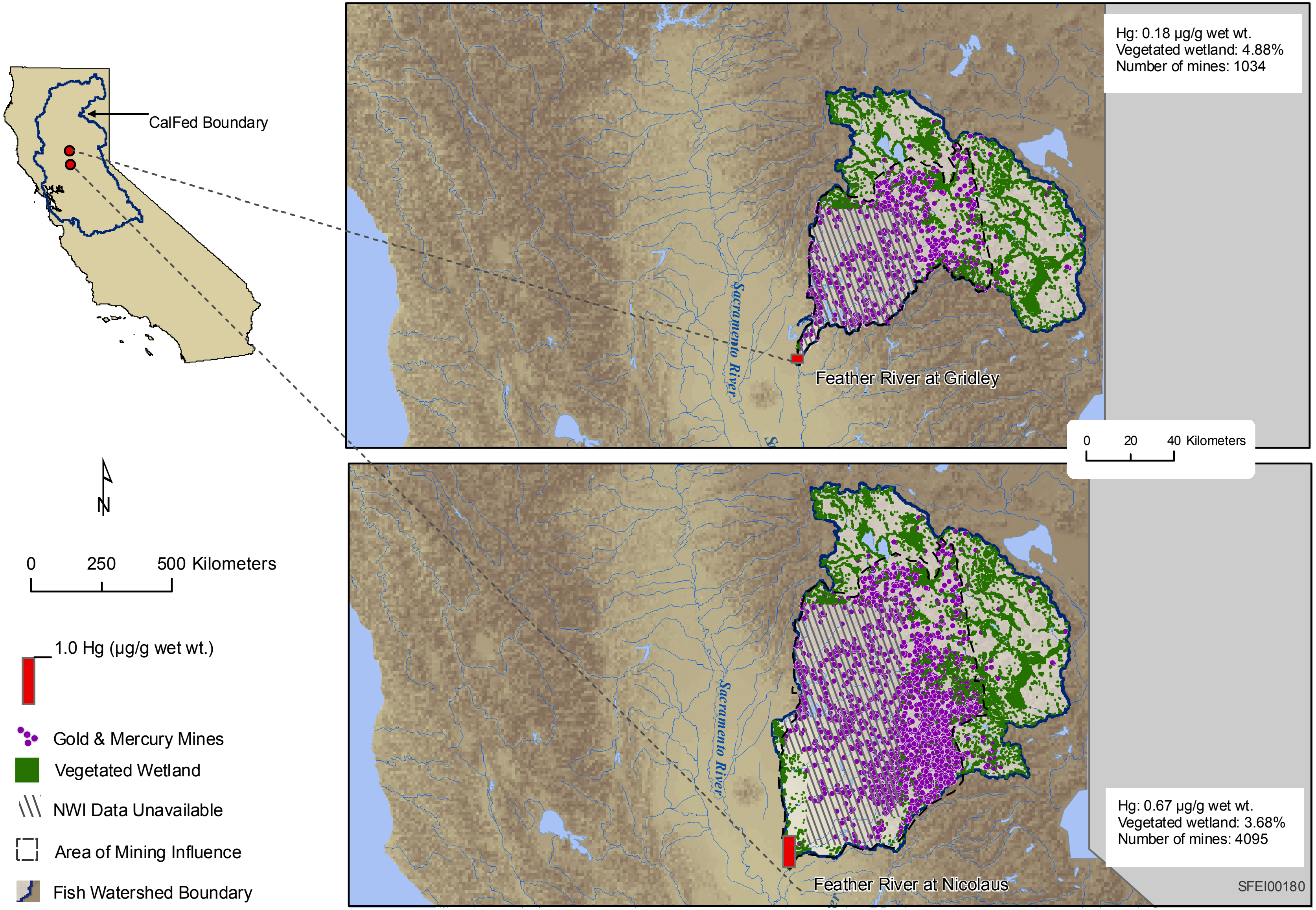

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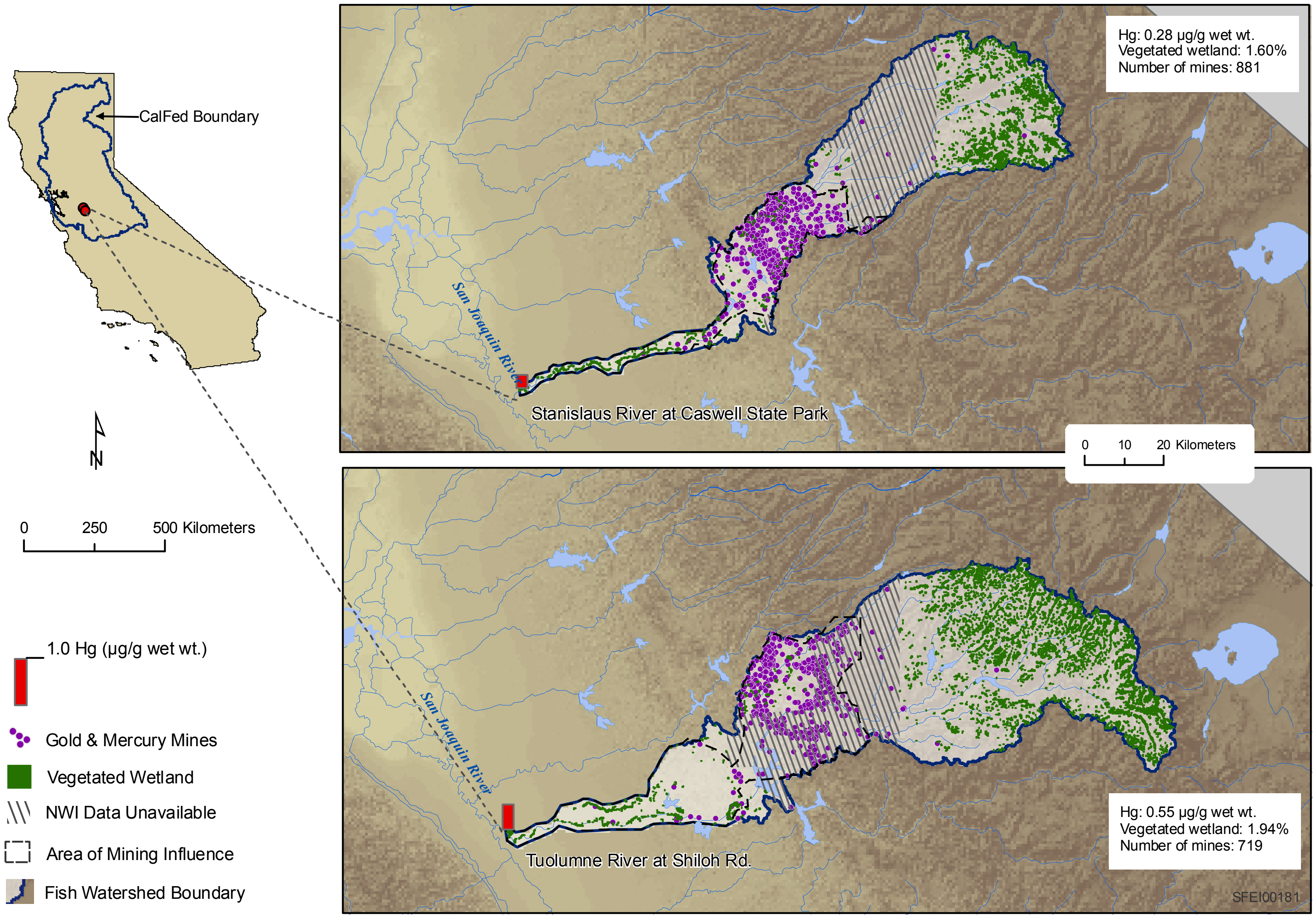

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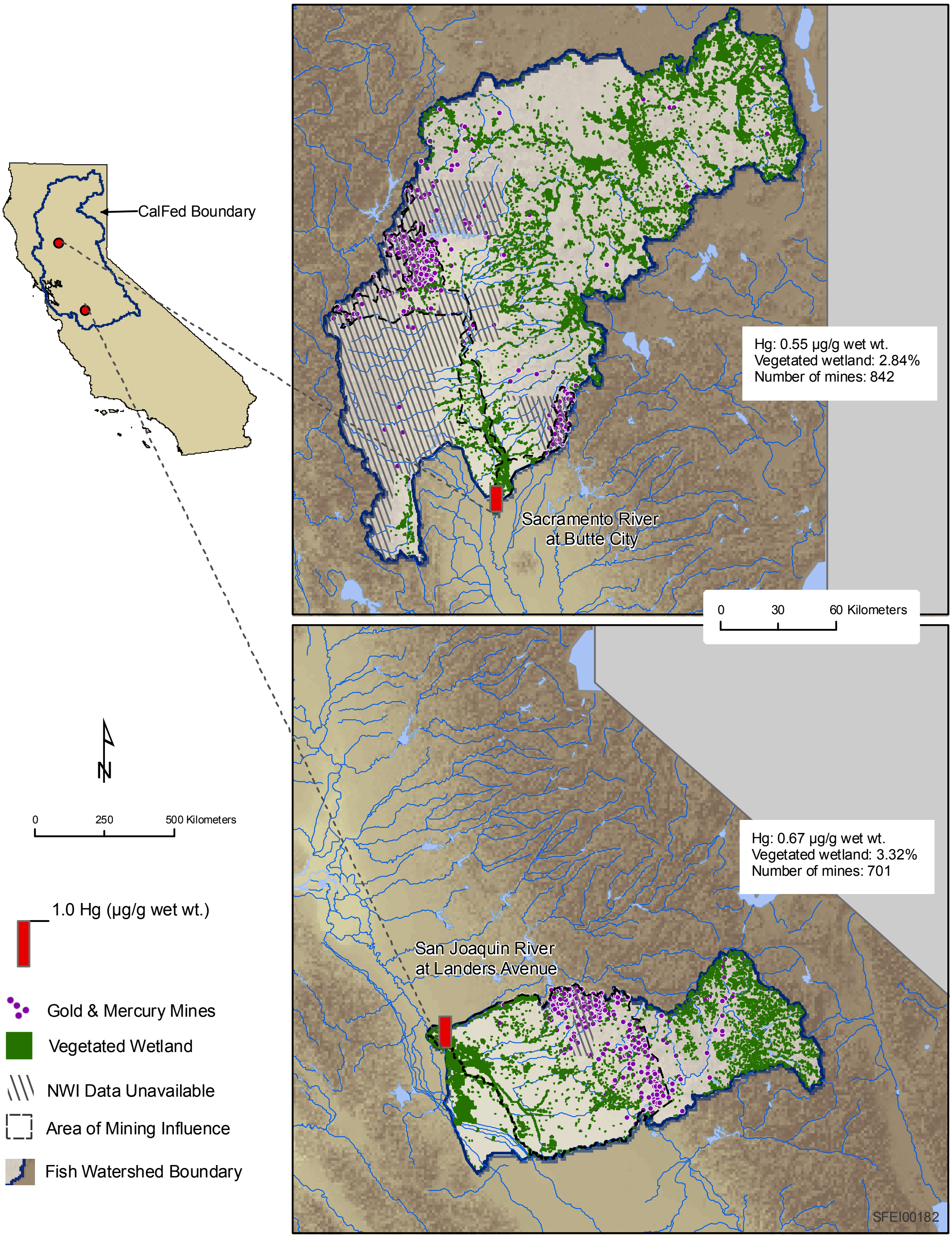

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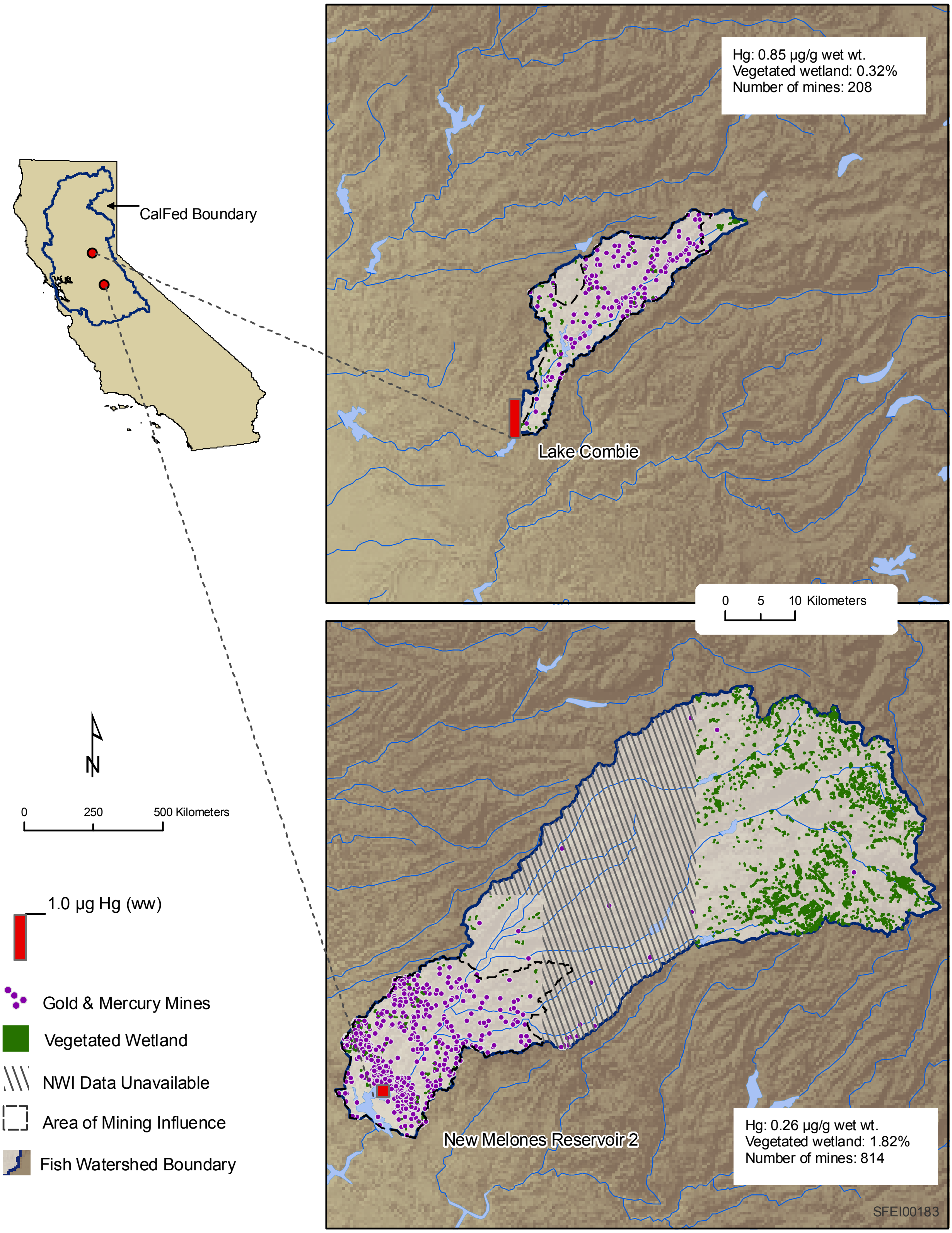

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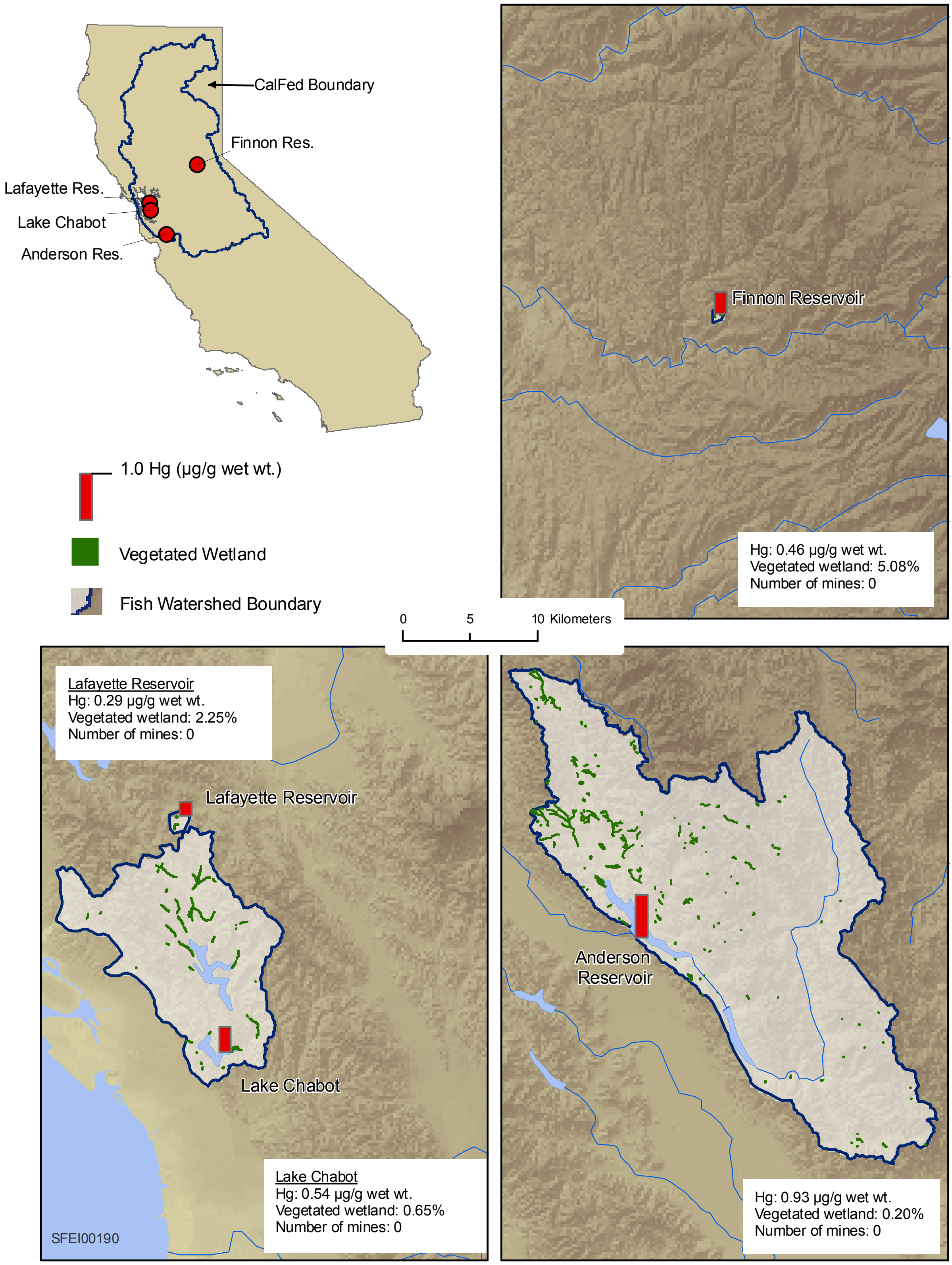

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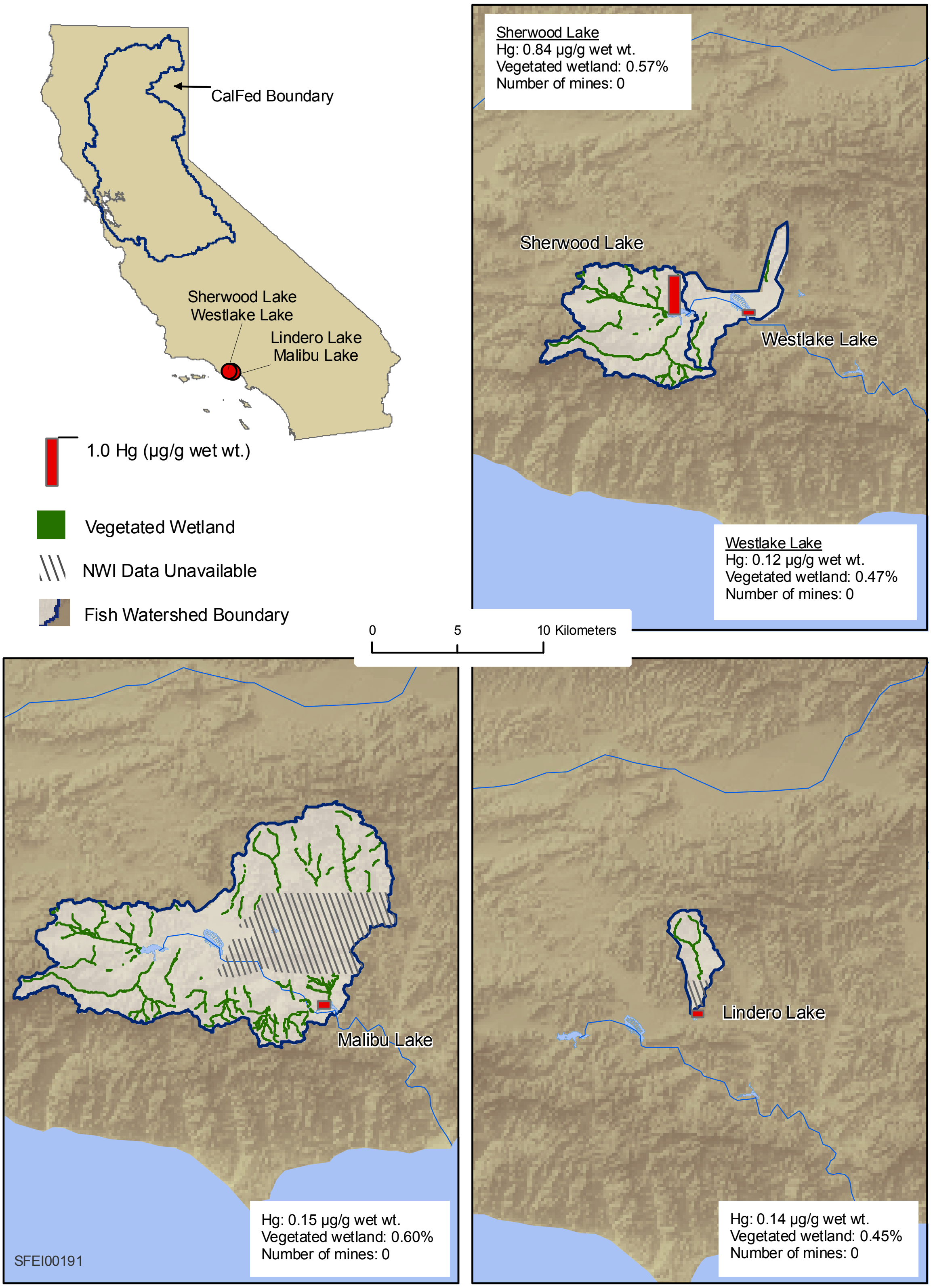

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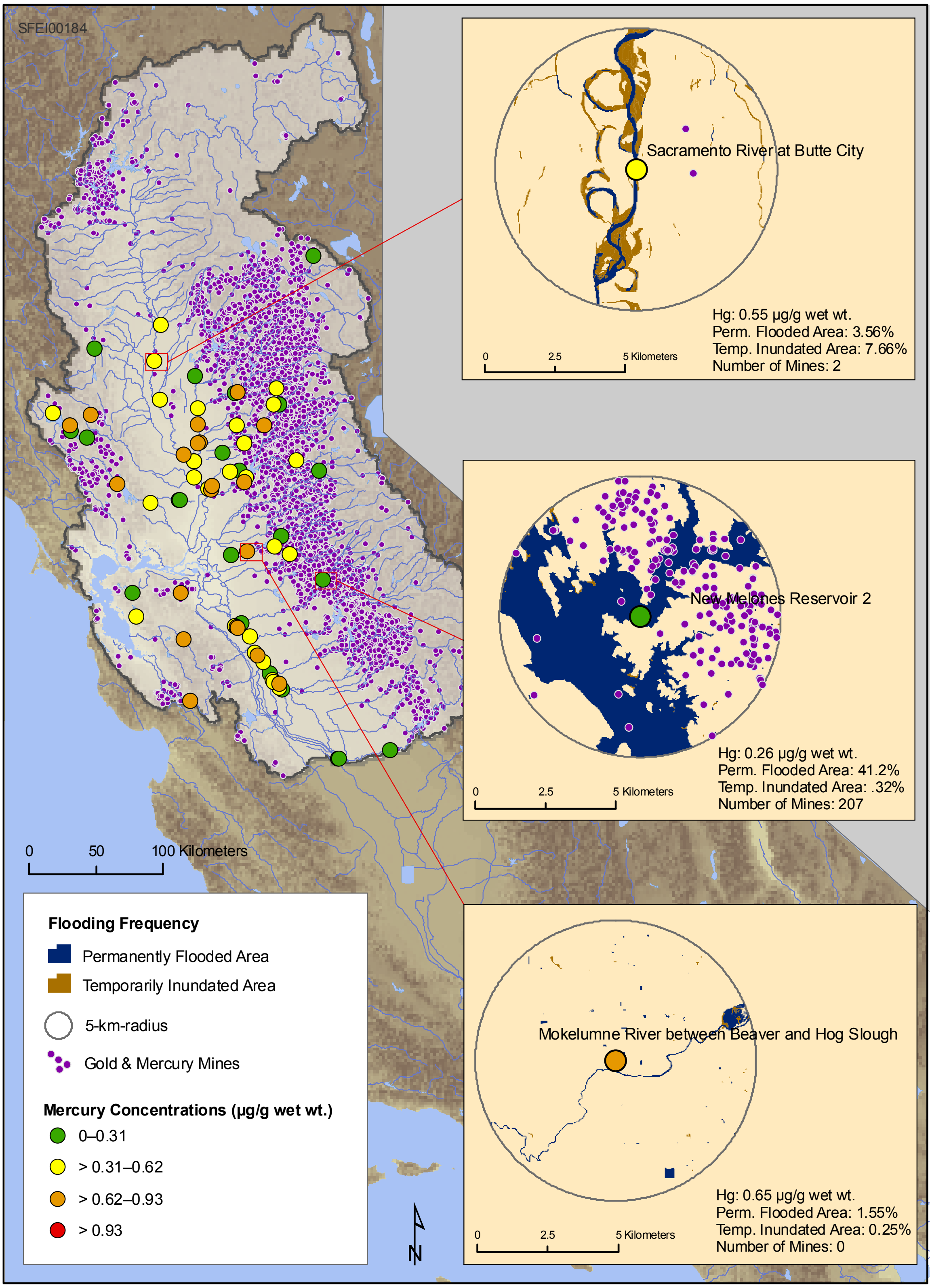

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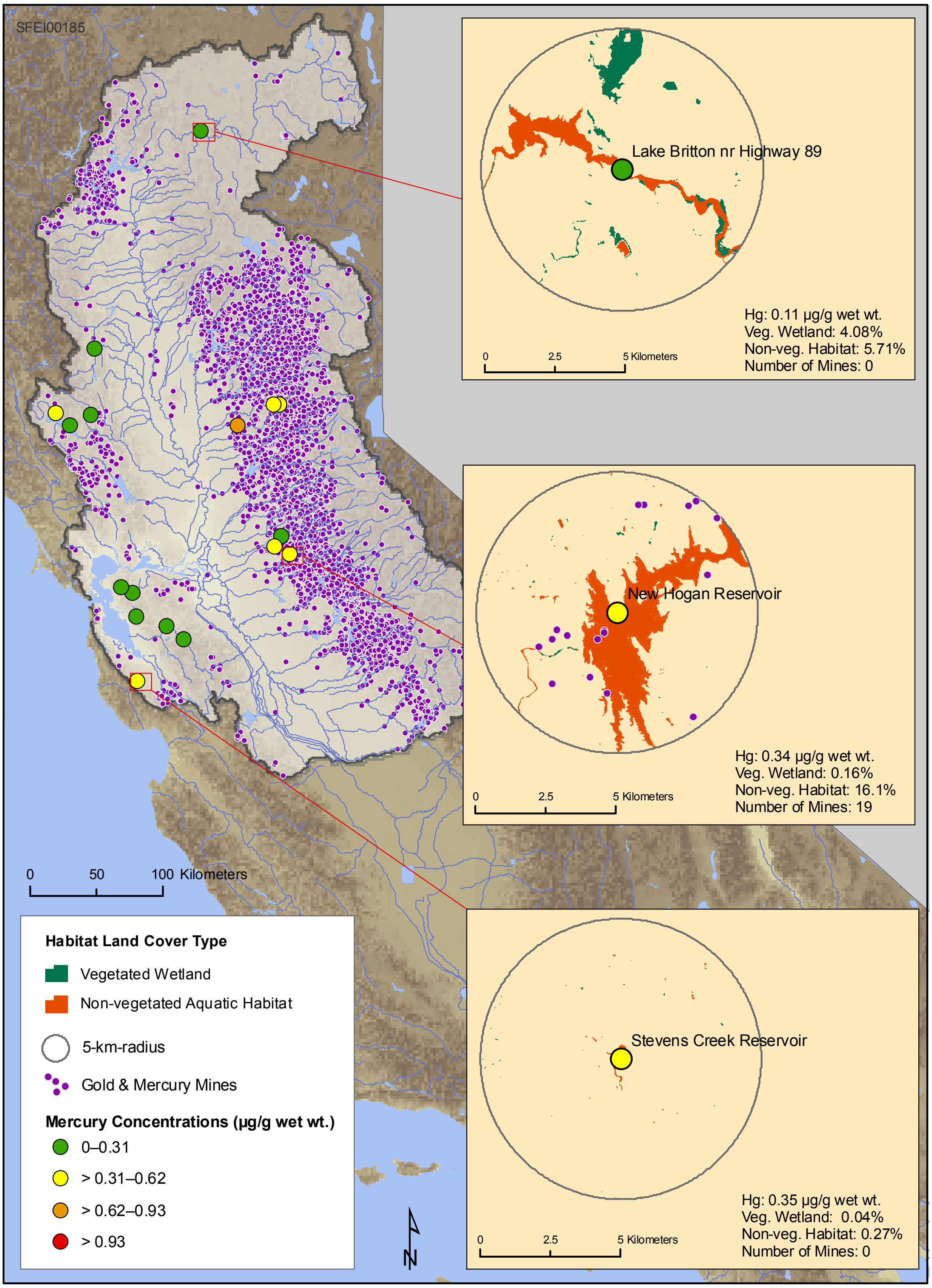

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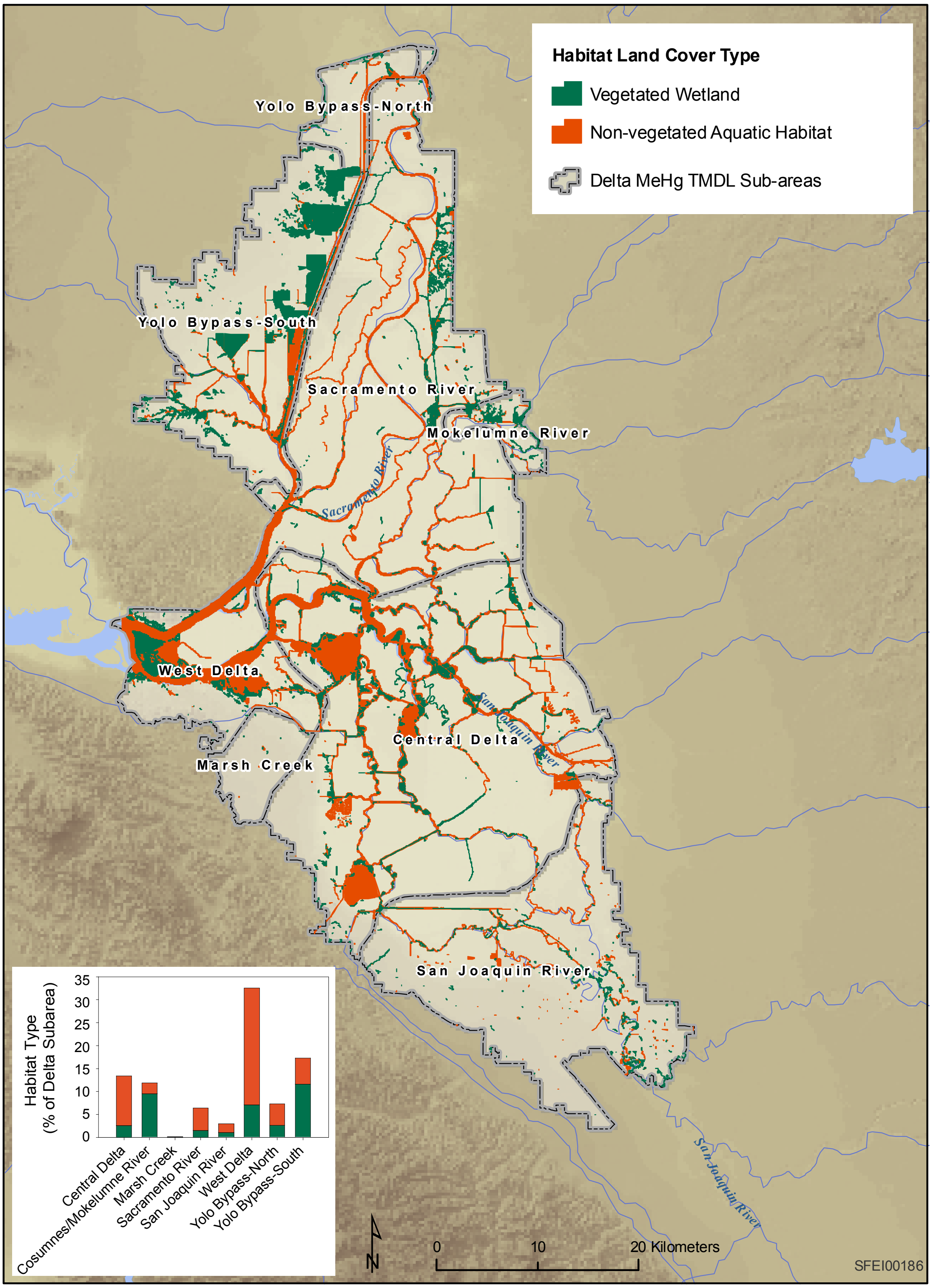

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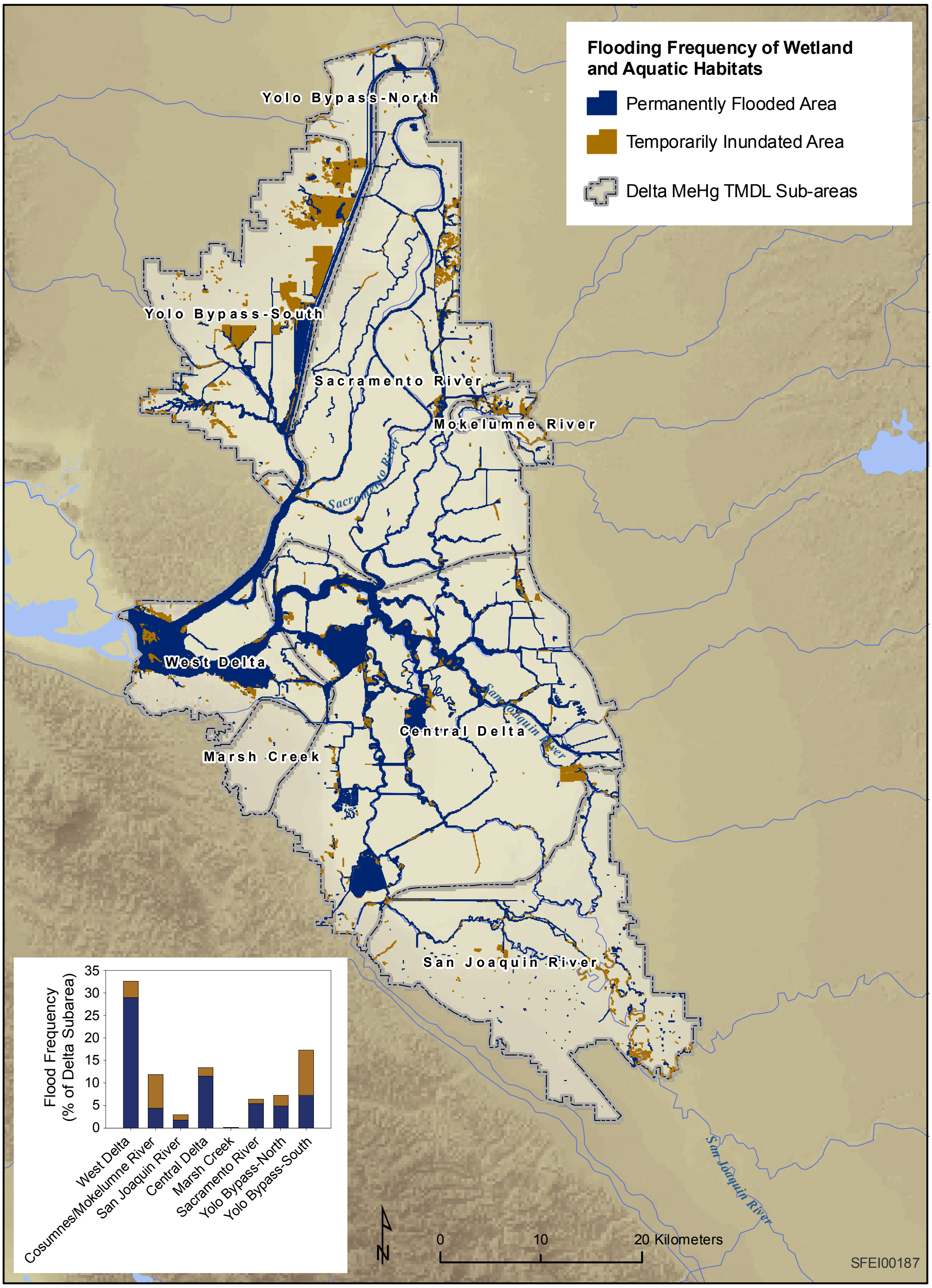

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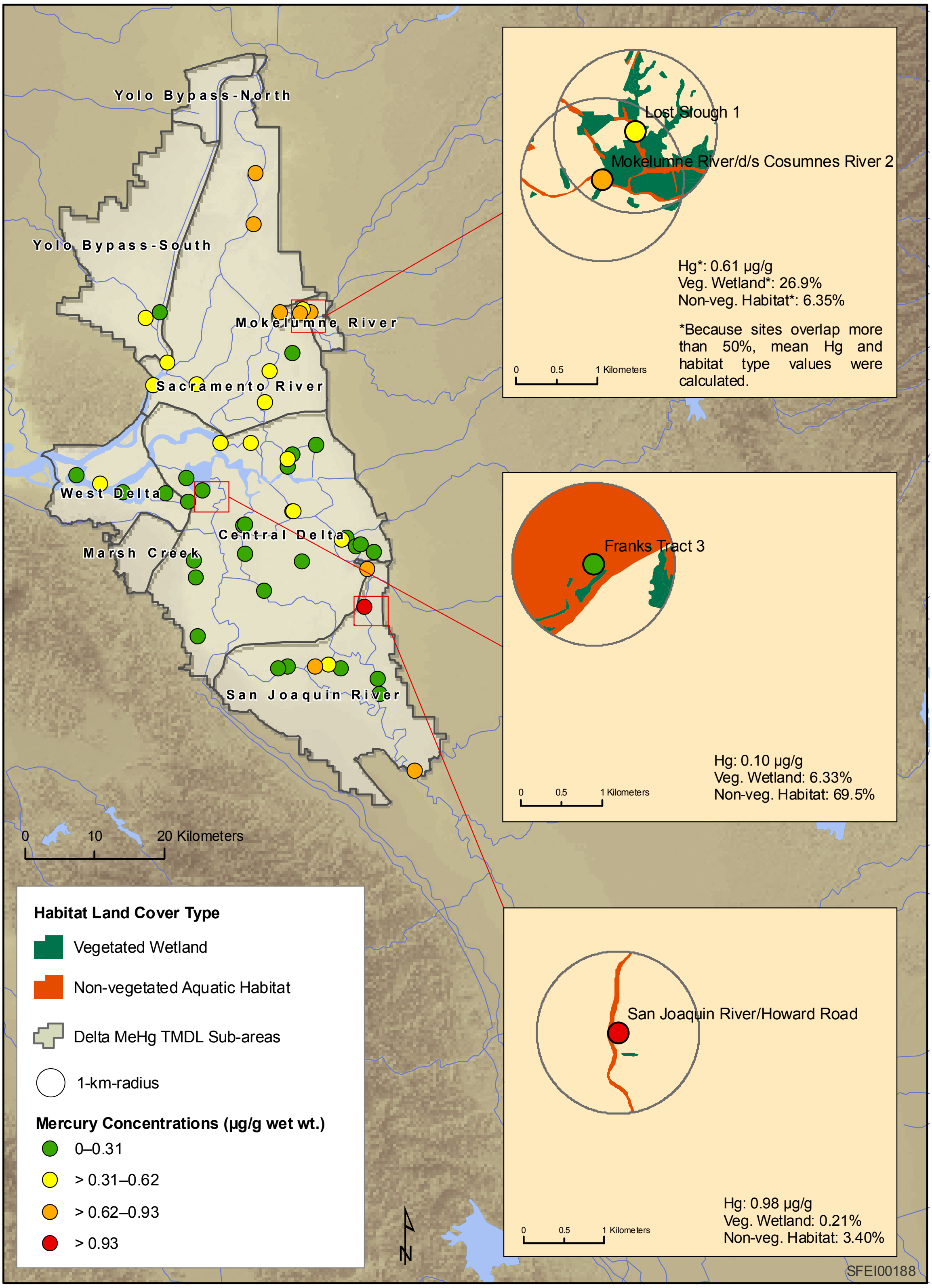

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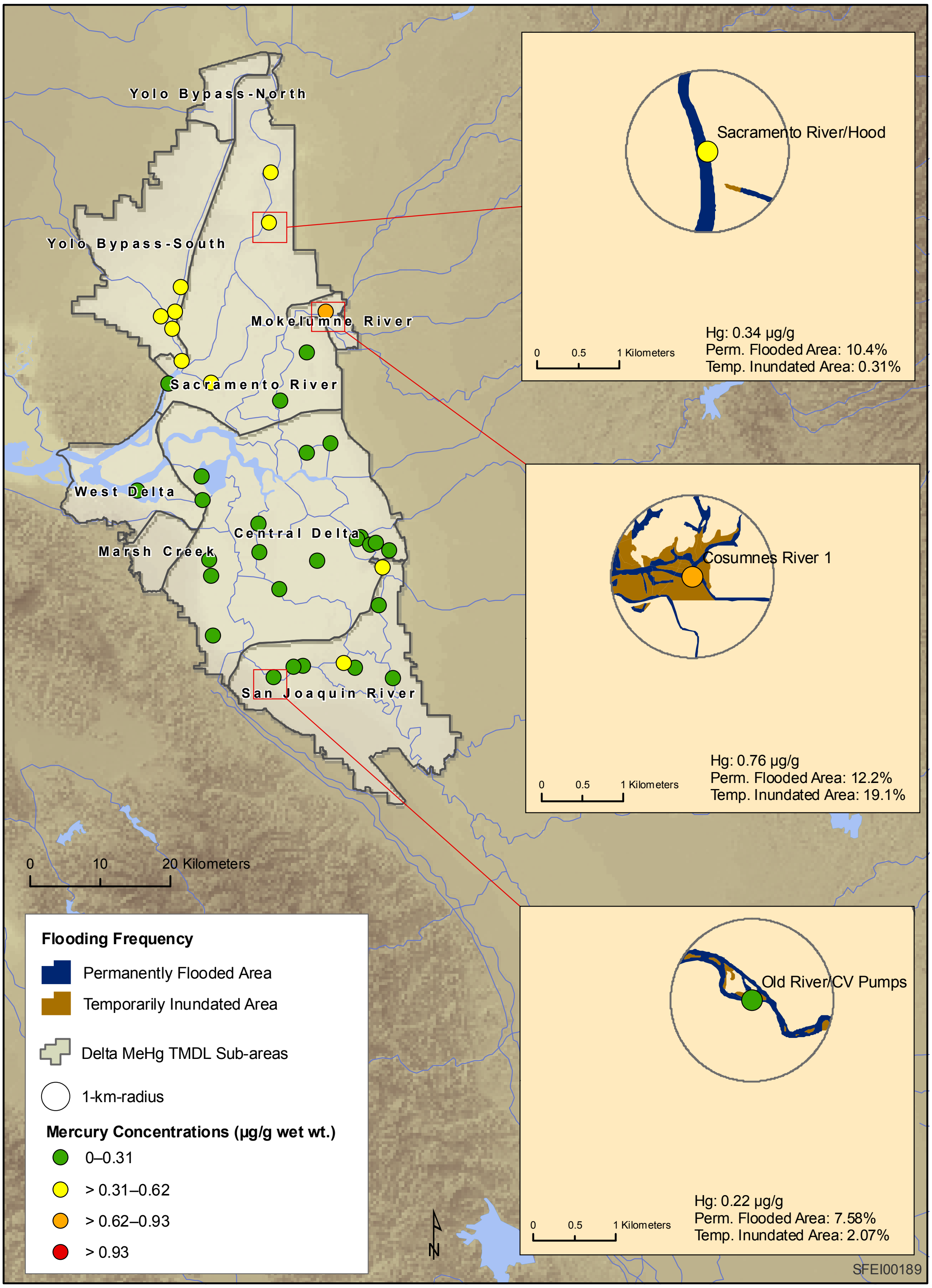

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Response to Reviewer Comments

In the appendix that follows, comments from three reviewers on the external draft of this report are provided. Many of the review comments suggested additional data analyses. However, based on recommendations from the Peer Review Panel, SFEI decided to not pursue further analyses for this report. Due to the non-significant relationships and inadequacies of certain datasets, effort to further develop the concepts and analyses suggested by these reviewers was not deemed an efficient use of Fish Mercury Project funds. Barbara Knuth's comments focused on typographical errors and clarification. Response to these comments resulted in changes to all sections of the report. The remaining reviewers' comments were more general, and did not result in any changes to the report. Future attempts to relate landscape features with mercury in the food web should consider these recommendations.

Comments on Final Draft, Melwani et al. 2/28/07: The Relationship between Landscape Features and Sport Fish Mercury in the Sacramento-San Joaquin Delta Watershed

Barbara A. Knuth
Member of Science Review Panel
Fish Mercury Project

Overall Comments:

The report is well-written and conveys its purpose, analysis, and conclusions clearly.

Specific Comments:

(1) Executive Summary (and throughout). It may be more straightforward to state explicitly that the major point sources equate with mining activity, rather than to refer to mining activity parenthetically (as in – i.e., mining activity). Unless there are other major point sources that were examined in this study, just call it "point sources associated with mining activity" clearly – so there is no question that perhaps you did include other types of point sources.
(2) Discussion of Hypotheses. It may be useful to state what you mean by "elevated mercury in biota" – elevated relative to what? To some standard? Relative to other locations in the watershed or among study sites?
(3) Hypotheses. You mention, as an example, that H2 and H4 were examined together to identify mining sources relative to landscape effects. It may be useful to include another example relating H1 to either H4 or H5. It's not intuitive how atmospheric deposition would be teased out on the local (H5) vs. landscape (H4) scale. A short example in the "narrative" regarding hypotheses might help the reader.
(4) Methods. Very clear discussion of your assumptions and Types of error.
(5) Statistical analysis. Explanation is very clear, as is justification for using a weight of evidence approach.
(6) Results and Discussion: In first paragraph, reference is made to "elevated" levels of mercury in fish watersheds with and without mines. This relates to point #2 above – be clear what "elevated" means in the context of this report.
(7) Page 13, line 18 – is "of" missing?
(8) Page 13, lines 34-36 – sentence seems to mix singular and plural (local variation … "are" likely?)
(9) Page 14, line 9. Why not present results for length, at least in summary form?
(10) Page 14, line 12 – delete comma
(11) Clear discussion of assumptions and limitations of the data, and suggestion of reasons for the results found.
(12) Page 16 – 17. Is if possible to provide any stronger recommendations re: what future sampling and analysis methods would be most likely to produce useful (more certain) results? The current report does this in a limited way. Is there any thing more you can suggest based on what you learned?

Comments on Final Draft, Melwani et al. 2/28/07: The Relationship between Landscape Features and Sport Fish Mercury in the Sacramento-San Joaquin Delta Watershed

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This study represents an interesting attempt to correlate landscape features such as historic gold mining and density of wetlands with mercury concentrations in fish. The study appears to have been competently carried out, using data available on watershed features and on mercury in three species of fish. The study was structured in a manner designed to test specific hypotheses potentially relating watershed characteristics to mercury in fish. The hypotheses appeared to be reasonable because they were testing ideas that are well known in the scientific literature on mercury cycling in the environment. Unfortunately, most of the relationships that were explored were not significant.

Some of the reasons for a general lack of significant relationships may be related to a lack of understanding about the role of historic and present landscape features how they affect mercury cycling. For example, we need to understand the conditions under which wetlands can be sources or sinks for methyl mercury and what factors will influence this. Also, we need a broader understanding of the effects of legacy mercury from historic gold mining and how it influences methyl mercury production in the present. However, I am in basic agreement with the authors' of the report that the lack of striking correlations are the result of limitations in the data used rather than the result of lack of landscape influences. As the authors' point out, the extent of mercury use at different mines could not be quantified, and it is therefore difficult to know what was actually being quantified by using the number of historic mines in a particular watershed. (However, the result confirms the general view that the conditions for mercury methylation may often be more important that the amount of mercury available for methylation in influencing the supply of methyl mercury to aquatic food chains.) Also, the particular characteristics of individual wetlands may have influenced their effect on mercury cycling.

I would suggest that if this kind of analysis was to be performed again in the future, there are some steps that could be taken that might help to uncover relationships between watershed characteristics and mercury in fish. First, I suggest that data for mercury in fish be standardized by size and/or age of fish. The authors were careful to include only fish within particular size ranges, but standardization by linear regression or polynomial regression may have given more precise measure of mercury in fish, uninfluenced by inter-site variation in average fish size or age. Second, I wonder if data on food chain length is available for the different sample sites and sport fish species sampled and whether including such data would make the analyses more precise. Sport fish are usually at or near the top of aquatic food chains and food chain length is known to influence mercury concentrations. If such data was available, it could be used to adjust mercury in fish to standard length food chains, i.e. make it more reflective of the input of methyl mercury to each local food chain. (If data on methyl mercury in water and/or sediments were available, it might be useful to look for direct relationships between watershed characteristics and methyl mercury in water. Methyl mercury in water and mercury in fish have been shown in other studies to be quite closely related.) Third, I would suggest that rather than using mean characteristics of watersheds or of “buffer zones” within watersheds, that a more dynamic approach to the watersheds might be tried. It is well known that lakes and reservoirs tend to be sinks for mercury and methyl mercury in watersheds. This means that lakes and reservoirs might be acting to “re-set” the supply of inorganic and methyl mercury for the watershed downstream of the lake or reservoir. Therefore, it might be useful to consider the active part of a watershed to be only the portion that is downstream of a sink such as a lake. It may not be easy, however, to decide on the criteria that would determine that a particular lake or reservoir was a sink for mercury, i.e. how deep, water renewal times, etc. A lake that was essentially a widening of a river would probably not be acting as a significant sink for mercury.

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Vicky Fry

1) In your variance analysis (ANOVA), did you or would you consider regression with the following parameters:

> water year type, temporal water level, datum or local tidal influence on water level, water surface area, and elevation (atmospheric deposition), slope or reach of river, areas of historic deposition (geomorphology), organic content of sediments, water temperature (profile by depth correlated to sample).

2) Could the Multiple Statistical Testing paragraph be translated into plain English please? I had a very hard time interpreting what it is trying to say.

3) Given the lack of correlation between mine sites and fish mercury concentrations, is there any conclusion to draw regarding mine site remediation? Could it be that where the mines sites were is no longer relevant because 1) mine site debris has scattered and re-deposited in downstream waterways, 2) naturally occurring mercury throughout the coastal range and atmospheric deposition buffer or obscure any correlation.

4) If you have time or get an opportunity, I would like to discuss your last paragraph before the Conclusions (p. 16). We are conducting a study looking at the importance of complexation of mercury to bioaccumulation. It seems that the complexity of the food web structure might also play a role.