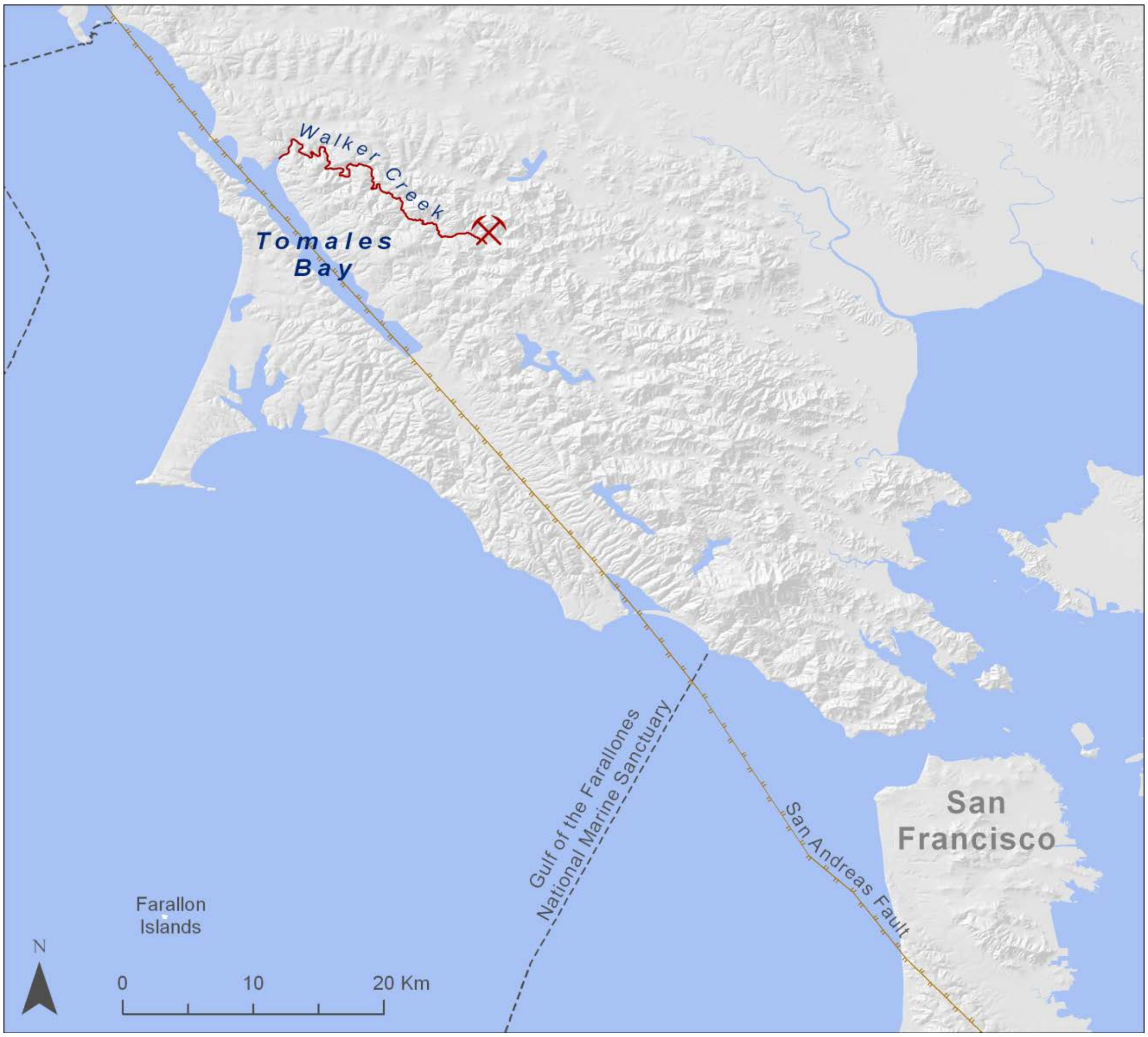
Total Maximum Daily Load (TMDL) for Mercury in Tomales Bay

Staff Report



California Regional Water Quality Control Board

San Francisco Bay Region

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San Francisco Bay Regional Water Quality Control Board

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Units of Measure

Mass (from lightest to heaviest)

- ng nanogram
- µg microgram
- mg milligram
- g gram
- kg kilogram
- MT million metric tonnes

Length (from shortest to longest)

- mm millimeter
- cm centimeter
- km kilometer

Volume

- l liter

Rate

- g/day gram per day
- µg/m2/yr microgram per square meter per year
- mg/m2/yr milligram per square meter per year

Concentration

- % percent
- ng/L nanograms per liter
- parts per million, billion and trillion (from largest to smallest concentrations):
  - ppm parts per million; usually mg/kg or µg/g
  - ppb parts per billion; usually µg/kg or ng/g
  - ppt parts per trillion; usually ng/kg

1. Introduction

Staff of the San Francisco Bay Regional Water Quality Control Board (Water Board) prepared this Staff Report to address the mercury impairment in all tidally-influenced areas of Tomales Bay (the Bay). Previously, the Water Board addressed upstream mercury impairment with the Walker Creek Watershed mercury total maximum daily load (TMDL) and implementation plan.[1] Where the Walker Creek Watershed mercury TMDL only addressed fresh waters in areas upstream of tidal influence, this Tomales Bay mercury TMDL addresses brackish, estuarine, and marine waters in all tidally influenced areas of Tomales Bay, including in large marsh areas at the mouths of Walker and Lagunitas creeks.

The Bay provides winter habitat for thousands of migratory waterfowl and is renowned for its fishery and oyster beds. The Bay is located approximately 64 km north of San Francisco (Figure 1-1) and is about 20 km long, with an average width of 1.4 km and highly variable bathymetry. A recent bathymetry map is available from the U.S. Geological Survey.[2]

The Clean Water Act requires California to adopt and enforce water quality standards to protect surface waters in the San Francisco Bay region. The San Francisco Bay Basin Water Quality Control Plan (Basin Plan) delineates those standards, which include beneficial uses of waters in the Region, numeric and narrative water quality objectives to protect those uses, and provisions to enhance and protect existing water quality (antidegradation). Section 303(d) of the Clean Water Act requires states to compile a list of "impaired" water bodies that do not meet water quality standards and to establish a TMDL for the pollutant that causes impairment.

The Bay has been designated as impaired (i.e., placed on the Clean Water Act 303(d) List) by mercury since 1996, due to concerns that drainage from the Gambonini Mercury Mine (the Mine) had probably contaminated wildlife and sport fish. The Mine drains to Walker Creek, where elevated aqueous mercury concentrations were found all the way downstream to the zone of tidal influence at the Bay. Subsequent observations found elevated mercury levels in biota. In 2004, Cal/EPA's Office of Environmental Health Hazard Assessment (OEHHA) issued an advisory against consuming any sharks from Tomales Bay, and limiting consumption of other sport fish and wild red rock crabs. Importantly, commercial filter-feeding shell fish (oysters, mussels, and clams) and wild clams have levels of methylmercury that are safe for human consumption. Some species of sport fish have levels of methylmercury that are safe for one to three meals per week.

This Tomales Bay mercury TMDL defines the allowable amount of mercury that can be discharged into the Bay while ensuring attainment of water quality standards. This TMDL is expressed as a suspended sediment mercury concentration because nearly all mercury is delivered to Tomales Bay in episodic, storm-driven pulses of sediment.

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[1] *Total Maximum Daily Load for Mercury In the Walker Creek Watershed, Staff Report*, April 4, 2008 Available from the Water Board at: www.waterboards.ca.gov/sanfranciscobay/water_issues/programs/TMDLs/

[2] *Interferometric Sidescan Bathymetry, Sediment and Foraminiferal Analyses; a New Look at Tomales Bay, California*; Open-File Report 2008–1237; Available from USGS at: http://pubs.usgs.gov/of/2008/1237/docs/of2008-1237.pdf

1.1. Regulatory process

This report explains that previously-completed cleanup of the Mine and previously-established regulatory requirements under the Tomales Bay pathogens and Walker Creek watershed mercury TMDLs are already addressing the mercury problem. Consequently, no further actions are required to address the mercury problem in the Bay.

This TMDL will be established by resolution rather than by amending the Basin Plan. The resolution, staff report, and associated materials will be made available to the public for a 30-day comment period. The Water Board will then consider adopting the tentative resolution during a public meeting. Subsequently, the U.S. Environmental Protection Agency will consider approving the adopted TMDL.

Previous and on-going actions make any further regulatory action (i.e., any “project”) unnecessary. Therefore, this action is not a “project” that requires compliance with the California Environmental Quality Act (California Public Resources Code § 21000 et seq.). The Water Board is not directly undertaking an activity, funding an activity, or issuing a permit or other entitlement for use (Public Resources Code § 21065; 14 Cal. Code of Regs. § 15378).

1.2. Report organization

This Staff Report is structured around the TMDL development process, which includes compiling and considering available data and information, conducting appropriate analyses relevant to defining the impairment problem, identifying sources, and establishing the TMDL and an implementation plan. For the Bay, no new regulatory actions will be necessary to control sources of mercury pollution as control measures are already in place.

Section 1 introduces the project, TMDL development process and Staff Report organization.

Section 2 presents the problem statement, which describes how mercury is impairing beneficial uses in the Bay.

Section 3 provides background information such as physical setting and geology, previous and current land uses including commercial fisheries and mercury mining, and a summary of mercury sampling data collected in the Bay.

Section 4 presents the derivation of the numeric targets and desired conditions for the Bay, and risks to human health and piscivorous birds.

Section 5 presents our understanding of the sources and loads of mercury to the Bay.

Section 6 presents the linkage analysis, which describes the relationship between mercury sources and the proposed targets. Our conceptual model describes the links between transport of mercury sources to the Bay, and chemical transformations that make mercury more toxic (i.e., methylation of mercury), and uptake into the food web (i.e., bioaccumulation)

Section 7 establishes the mercury loading capacity and TMDL, and sets allocations to sources.

Section 8 describes the implementation actions that have been completed to control mercury sources to the Bay, and our plans to monitor to determine whether targets have been achieved.

Section 9 presents the references section, which lists all the information sources cited and relied upon in preparation of this report.

2. Problem Statement

This section describes how mercury is impairing beneficial uses in the Bay.

2.1. Key points

- The Bay has been identified as an impaired water body due to the presence of mercury. Elevated mercury levels in biota has impaired the following existing beneficial uses in the Bay: commercial and sport fishing (COMM), estuarine habitat (EST), marine habitat (MAR), and wildlife habitat (WILD)
- Previously, mercury mines in the hills above Walker Creek discharged large amounts of mercury. However, the Bay's hydrodynamics confine mercury mining waste to the Walker Creek Delta. The only significant source of mine pollution was cleaned up in 2000.
- Sharks and bat rays are unsafe for human consumption due to high levels of methylmercury, however they may be getting exposed to mercury from areas outside of the Bay.
- Commercial filter-feeding shell fish (oysters, mussels, and clams) and wild clams have levels of methylmercury that are safe for human consumption. Some species of sport fish have levels of methylmercury that are safe for one to three meals per week.

2.2. Mercury problem

Mercury is a persistent and bioaccumulative toxic pollutant that occurs naturally in California's Coast Range. In 1996, the Bay was designated as impaired by mercury due to concerns that drainage from the Gambonini Mercury Mine had probably contaminated wildlife and sport fish. Water Board staff conducted intensive stormwater discharge monitoring at the Mine and downstream in Walker Creek during the 1998 wet El Niño winter (Water Board 1998, p. 6). Total mercury in water samples collected about 5 km downstream of the Mine ranged from 340–7,100 ng/L, with a mean of 3,300 ng/L. These high mercury levels persisted 25 km downstream of the Mine to the mouth of Walker Creek. There, total mercury ranged from 260–2,600 ng/L, with a mean of 830 ng/L. All of these measurements exceeded the water quality objective of 25 ng/L.

Subsequent observations found elevated mercury levels in biota (see Section 4, *Numeric Targets*). In 2004, OEHHA issued an advisory against consuming any sharks from Tomales Bay, and limiting consumption of other sport fish and wild red rock crabs (OEHHA 2004). Importantly, commercial filter-feeding shellfish (oysters, mussels, and clams) and wild clams have levels of methylmercury that are safe for human consumption. Some species of sport fish have levels of methylmercury that are safe for one to three meals per week.

2.3. Mercury mining in the Walker Creek Watershed

Mercury occurs naturally in cinnabar deposits in California's Coast Range, including in the hills above Walker Creek, the second-largest tributary to Tomales Bay (see Figure 2-1). Mercury was mined in the Walker Creek watershed in the 1960s and early 1970s. The Gambonini Mercury Mine (Mine) was the largest mercury mine in the Walker Creek watershed, and most of the ore from the other, smaller mines (i.e., the Franciscan, Cycle, and Chileno Valley mines) was processed at a single processing facility at the Mine. At the Mine, a mechanical separator and retort facility were used to extract the mercury from the ore. The mercury-laden waste material was then dumped in ravines and on the hill slopes below the processing facility.

Before Mine cleanup, storms would erode poorly-managed mining wastes (tailings) and transport them downstream. Local residents complained that discharges from the Mine caused Walker Creek to run red; cinnabar is a distinctive red color. A local rancher told Water Board staff that the red flow could even be seen flowing through the Bay towards the Pacific Ocean (McDonald 2007). In response, the California Department of Fish and Game required the Mine operators to better manage their tailings. Consequently, the operators constructed a small earthen tailings dam across a steep ravine just downslope of the Mine operations.

By 1972, all mining had ceased in the watershed. To close the Gambonini Mercury Mine, the operators buried some of the tailings, contoured the remaining waste pile, and planted grass seed (Johnson 2009). However, these non-engineered measures did not effectively contain the wastes.

In 1982, a 1-in-100 year storm event occurred. The Mine tailings dam failed catastrophically, sending tailings all the way through Walker Creek to its Delta in Tomales Bay. Although the quantity of sediment discharged during this event is unknown, the amount was sufficient to partially bury automobiles on the adjacent property (Johnson 2009). In subsequent winters, the tailings pile continued to erode, resulting in numerous rills, large gullies, and debris flow scars (see Figure 2-2).

2.4. Gambonini Mercury Mine cleanup

In 1998, Water Board staff conducted intensive stormwater discharge monitoring efforts during the wet El Niño winter. This study confirmed that episodic storms—infrequent storms with intense downpours—discharge the largest amounts of mercury and sediment, and that large loads of mercury were still being discharged. These measurements triggered emergency Superfund designation for the Mine.

In 2000, U.S. EPA cleaned up the Mine (Figure 2-2). The cleanup design goal was slope stabilization to control erosion, without offsite disposal, imported soils, or an impervious cap (Smelser 2001). Drainage from the re-contoured tailings pile is now collected in concrete drainage ditches and routed to a reconstructed creek channel at the toe of the waste pile slope, which sits on a gravity buttress made of cut material. Compost was incorporated into the outboard edge of the fill slope and an extensive native-plant revegetation program for surface erosion control has resulted in dense grass and bush coverage.

In 2005, Water Board staff again conducted intensive stormwater discharge monitoring. In contrast to 1998, the post-remediation monitoring took place during a relatively normal winter. Kirchner (2011) calculated that, holding rainfall constant, site cleanup reduced mercury loads by an environmentally significant amount: an estimated 92–93%.

2.5. Hydrodynamics prevent dispersal of mercury

The hydrodynamics of the Bay confine mercury mining waste to the Walker Creek Delta, rather than dispersing around the Bay. Flows from Walker Creek discharge to the Delta and out to the Pacific Ocean, as shown on Figure 2-3 (Stacey 2010, Johnson 2010). During a storm, nearly all sediment transported in Walker Creek either accumulates at the Delta or is transported out the mouth of the Bay into the Pacific Ocean; very little of the sediment transported by Walker Creek makes it up the Bay past Hog Island.

In essence, the Bay has two sections. The section closer to the ocean, from the mouth to Hog Island, has high tidal energy. Walker Creek discharges into this area. Water in the rest of the Bay moves much more slowly and acts as a physical barrier to water from Walker Creek. Incoming tides come up along the Bay bottom towards Lagunitas Creek, and stormwater from Lagunitas Creek, which is cooler, fresher, and less-dense, flows along the Bay's surface. A small portion of Walker Creek stormwater flows around Hog Island and up the Bay. There, it meets both stormwater from Lagunitas Creek, which is flowing out toward the ocean, and a physical barrier of calm water. These two physical processes divert the Walker Creek flows back towards the ocean. While this understanding of the dominant pattern of sediment flows reflects what we know about the Bay, this estuary is dynamic. Tides and wind slosh water and sediment, and cause some mixing, even transporting some sediment from Walker Creek up towards Lagunitas Creek.

Sediment mercury concentrations around the Bay perimeter support our understanding of hydrodynamics. The most widespread surface sediment sampling was conducted in 2009 as part of the *Impairment Assessment* (Ridolfi 2010). Elevated mercury concentrations in sediment are confined to the Walker Creek Delta, as can be seen in Figure 2-4.

2.6. Water quality standards

The water quality standards for the Bay include beneficial uses, numeric water quality objectives, and narrative water quality objectives. Beneficial uses, and their status with respect to mercury, are listed in Table 2-1.

Both Basin Plan and U.S. EPA California Toxics Rule (CTR) numeric mercury water quality objectives apply to the Bay. Basin Plan numeric mercury water quality objectives are the following (Table 3-3 of the Basin Plan):

- 0.025 µg/L; 4-day average for waters with salinity greater than 10 parts per thousand 95% of the time, and
- 2.1 µg/L; 1-hour average for waters with salinity greater than 1 part per thousand 95% of the time

These Basin Plan objectives are based on the U.S. EPA's *Ambient Water Quality Criteria for Mercury – 1984* (USEPA 1985). The 4-day average objective was intended to protect human health, and the 1-hour average objective to protect aquatic organisms and wildlife.

The CTR mercury criterion is a 30-day average of 0.051 µg/L and protects human health for consumption of aquatic organisms.

The Basin Plan's narrative bioaccumulation objective states:

> Many pollutants can accumulate on particles in sediment, or bioaccumulate in fish and other aquatic organisms. Controllable water-quality factors shall not cause a detrimental increase in concentrations of toxic substances found in bottom sediments or aquatic life. Effects on aquatic organisms, wildlife, and human health will be considered.

3. Background

This section presents the Bay watershed's physical setting and geology, discusses previous and current land uses including commercial fisheries and mercury mining, and describes the many previous scientific studies of the Bay.

3.1. Key points

- Europeans settled the area about 150 years ago. Their farming activities significantly increased erosion and reduced the Bay's length and depth despite rising sea levels.
- The Bay has a high ecological value largely because human population density has remained low and the area was never industrialized.
- This TMDL is supported by substantial previous study of the Bay, including evaluation of bathymetry, nutrient cycling, mercury pollution, and biota contaminant levels.

3.2. Watershed setting and history

Two major streams flow into the Bay: Walker Creek at the northern end, about 4 km from the Bay mouth, and Lagunitas Creek at the head (south end of the Bay) (Figure 2-1). Both creeks are major sources of sediment to the Bay. Mercury mining took place in the hills above Walker Creek in the 1960s and 1970s. The following history is adapted from Johnson (2009).

> Alterations in land use over the last 150 years have significantly increased sediment loading, reducing Tomales Bay in both length and depth despite rising sea levels. Hillside erosion within the [*Walker Creek*] watershed was likely accelerated by the introduction of non-native grasses and farming practices in the late-1800s, and the high density of dairy farms in the early-1900s. The Delta at the mouth of Walker Creek has undergone significant geomorphologic changes in the last 200 years. From 1852 to 1870, the town of Tomales, now 4 km upstream from the current Delta, was once part of the estuary and was utilized as a port where small barges and steamers docked to load local produce. By the late 1800s, accretion

> along the lower reaches of the watershed made the waterway to the town of Tomales impossible to navigate, forcing the port to move elsewhere.

The following descriptions are adapted from Ridolfi (2010).

> Tomales Bay is renowned for its wildlife, herring fishery, and commercial shellfish industry. It is included in four protected areas due to its ecological significance: the Gulf of the Farallones National Marine Sanctuary, Golden Gate National Recreation Area, Point Reyes National Seashore, and Tomales Bay State Park (see Figure 2-5). Nearly 2,000 species, including many threatened and endangered species, were recently recorded in a study of biodiversity within Tomales Bay. The Bay is also an important migratory stop along the Pacific Flyway and supports approximately 20,000 shorebirds and 22,000–25,000 waterfowl. Nearly half the bird species of North America have been spotted in this region.
>
> Tomales Bay occupies the rift zone of the San Andreas Fault, which separates the Pacific and North American plates. It is the most significant geologic feature of the area, and has shaped the topography and geology of the landscape since Tomales Bay was filled with glacial melt water 15,000 to 5,000 years ago at the end of the last ice. Evidence suggests that since the Cretaceous period, the Point Reyes Peninsula has been moving northward at an average rate of 1–5 cm per year. The 1906 earthquake, however, resulted in horizontal displacement on the peninsula of up to six meters in a matter of seconds, providing a reminder that much of the movement on the fault is punctuated rather than a gradual continuous creep.
>
> The San Andreas Fault serves as a dividing line between two distinct geologic regions of the watershed. The western side of the Bay is underlain by the late Cretaceous Inverness Ridge formation (granodiorite and granite) and Tomales Point (tonalite) and its soils are more coarse-grained and well drained. In contrast, the eastern shore has much finer-grained soils that originate from the Wilson Grove Formation (north of Walker Creek only) and the Franciscan Formation, where most of the mercury mining took place. Geologic and mining-related mercury sources therefore are present only in the eastern portion of the watershed. Both of these formations are highly erodible, and produce soils susceptible to landslides and gullies. This underlying geology and orientation to the Pacific Ocean results in the substrate of the northern portion of Tomales Bay (to just below Hog Island) being made up of sandy deposits, while the southern half is dominated by clay and silt.

3.3. Land use and commercial fisheries

The following descriptions are adapted from Ridolfi (2010).

> Current major land uses in the Tomales Bay watershed are livestock grazing and dairy farming (55%) and park and open space (42%). There are six small, unincorporated communities (Point Reyes Station, Tomales,

Woodacre, Lagunitas-Forest Knolls, Inverness and Dillon Beach) with a combined population of about 11,000 people. An additional 2.5 million visitors visit the National Seashore annually. Given these land uses, nearby industrial and urban land uses are unlikely to be significant sources of mercury [*to Tomales Bay*].

The California Department of Fish and Game regulates the Pacific herring fishery and commercial shellfish farming operations in Tomales Bay. The Bay's oysters have been farmed since the early 1900s, and now occupy 463 acres in the north and central-eastern areas of the Bay. The aquaculture industry (which includes small quantities of mussels and clams, in addition to oysters) contributes an estimated $2.49 million annually to the local economy. Commercial production of the native oyster (Ostrea lurida) began around 1875; however the fishery is now mostly comprised of Pacific oysters (Crassostrea gigas) and Bay mussels (Mytilus edulis and M. galloprovincialis) due to their higher growth rate and size. In addition, there are small amounts of Eastern (O. virginica), European (O. edulis), and Kumamoto (C. gigas kumamoto) oysters and Manila clams (Tapes semidecussata) in production. Oysters grow in bags or in wooden trays placed on the substrate of intertidal areas in the Bay.

In winter, the Bay is a major spawning ground for Pacific herring (Clupea pallasi), which are almost exclusively harvested for their roe and exported to Japan. Records of spawning biomass (measured in tons of eggs deposited during the winter spawning season) are available starting in 1973. Based on these records, the greatest biomass was observed in 1979 at over 20,000 tons, but later dropped to nearly zero tons in 1989. This trend was similar in San Francisco Bay. Subsequently, the fishery was closed for three seasons. Since then strict quotas have been assigned, and are roughly equivalent to 15–20% of the estimated biomass for the coming year.

In addition to the commercial herring fishery and aquaculture industry, Tomales Bay supports a thriving recreational fishery, with halibut and clams being the two most popular organisms taken each year by anglers. Other fish commonly caught include Dungeness and rock crabs, jacksmelt, perches, sole, striped bass, sturgeon, sharks, and rays.

3.4. Previous studies

Tomales Bay has been the subject of numerous research and monitoring programs over the past few decades. The three key reports about mercury in Tomales Bay, which we relied upon in developing this TMDL, are:

Impairment Assessment for Mercury in Tomales Bay, CA (Ridolfi 2010) This report was prepared by the Aquatic Science Center for the Water Board to provide supporting information for this mercury TMDL. The *Impairment Assessment* report includes descriptions of Tomales Bay's environmental setting, calculations for numeric targets, and results of recent Bay-wide sediment, biota, and water sampling.

Mercury accumulation and attenuation at a rapidly forming Delta with a point source of mining waste (Johnson 2009)
This work was initiated by the Water Board. The report concluded that mercury from the Gambonini Mercury Mine had accumulated at the Walker Creek Delta, and that it was buried by cleaner sediments. Additionally, this paper provides a succinct summary of land use changes since European settlement, and how the changes influence sediment transport to and accumulation in the Bay.

Assessing water quality impacts and cleanup effectiveness in streams dominated by episodic mercury discharges (Whyte 2000)
This work was initiated and conducted by the Water Board. It identified that infrequent, large storms cause much worse soil erosion. Consequently, the bulk of mercury-laden mining waste is transported downstream to the Bay in episodic events. U.S. EPA responded promptly with site cleanup overseen by their superfund emergency response team.

3.5. Data summary

Several different agencies have conducted mercury sampling campaigns in the Bay since 1979. The following are highlights of findings from these sampling campaigns.

Bay-wide sediment

Total mercury in sediment is elevated at Walker Creek Delta compared to other locations in the Bay. Results from 1999-2000 are presented on Figure 3-1, in which Walker Creek Delta is indicated by a bracket extending across bars from “WC Delta” to Hamlet. The slightly elevated mercury at Preston Pt. is likely due to inputs directly from Walker Creek and/or transfer of mercury through the Delta. The unexpectedly elevated mercury at Millerton, located 13 km from Walker Creek, could be due to transport from the Mine via Walker Creek or pollution from towns near Millerton Point. These 1999–2000 results, together with 2009 results (Figure 2-4), confirm that pollution from the Mine is confined to the Delta area (see Section 2.5, *Hydrodynamics prevent dispersal of mercury*).

Nonetheless, a few sediment samples collected at locations far from the Walker Creek Delta had total mercury concentrations above 0.2 mg/kg (Water Board 2012). Specifically, two samples collected by Water Board staff in 1999 at Millerton (shown on Figure 2-4; located near “North Millerton” on Figure 2-5) had over 2 mg/kg total mercury, and one sample collected by the Aquatic Science Center in 2009 at Lagunitas Creek Delta had slightly elevated total mercury of 0.6 mg/kg. One explanation for these elevated mercury concentrations is that the hydrodynamic forces we described in Section 2.5 are an imperfect barrier towards preventing sediment transport up towards the head of the Bay. Another plausible explanation is that these mercury detections could have been due to pollution from nearby towns, such as mercury from a broken thermometer or fluorescent tube.

Sediment at Walker Creek Delta

Another key finding is that mining waste is being buried by cleaner sediments at Walker Creek Delta. Sediment cores were collected in 2003 from the Walker Creek Delta. All of the cores were deep enough to reach pre-mining depths. The cores were age-dated with Cesium 137 and analyzed for total mercury over 2-cm depth intervals. Location, age and mercury concentration was then used to calculate mass of mercury loads delivered to, and accumulated at, each core location over time (Figure 3-2).

Peak mercury loadings likely correspond to an extremely large storm in 1982 that burst the Mine tailings dam. Peak accumulation of greater than 50 mg/m2/yr occurred between 1975 and 1985 near the mouth of Walker Creek (cores #13, 15, and 24). Several cores at the eastern side of the Delta (cores #8, 10, 12, and 22) had highest mercury concentrations near the surface. This is likely the result of natural estuary sediment mixing processes, although recent increases in mercury loads, perhaps from mining waste released from depositional areas along Walker Creek, cannot be ruled out. Nonetheless, the overall trend is less mercury accumulating at the Delta since the mine closed.

Chileno Creek, a tributary to Walker Creek, is located in easily eroded geology with relatively low naturally occurring mercury concentrations. Land use changes since the arrival of Europeans have caused high rates of erosion, and consequently large loads of clean sediment. Prior to the Mine cleanup in 2000, sediment from Chileno Creek mixed with Mine waste that had higher concentrations of mercury, resulting in relatively high concentrations of mercury in sediment at the Walker Creek Delta. Since the Mine cleanup, cleaner sediment from Chileno Creek and other tributaries is burying the mining waste accumulated at the Delta (Figure 3-2).

Bay-wide prey fish

In contrast to sediment, methylmercury in prey fish from the Walker Creek Delta was not elevated compared to prey fish from other areas around the Bay (Figure 3-3). This 2009 data set is the only Bay-wide sampling of prey fish, which were collected from a boat with an otter trawl at the edge of the Bay. Perhaps this lack of a hot spot at the Delta is because the collection method targeted fish with low methylmercury because they feed on the pelagic (water column) food web rather than the more polluted benthic (sediment) food web. (In Section 6, we describe that the methylmercury bioaccumulation problem is from sediment, not water.) Alternatively, this lack of a hot spot at the Delta could be due to small sample size, although this explanation is less likely because fish of the same species and lengths have equal methylmercury concentrations at the Walker Creek and Lagunitas Creek deltas (Figure 3-4).

Prey fish at Walker Creek Delta

Prey fish within the vegetated marsh had higher mean methylmercury concentrations compared to those collected in the subtidal zone at the edge of the Bay. In 2010, prey fish were collected at both the edge of the Bay in the subtidal zone and from within the vegetated marsh at the Walker Creek Delta. These data are discussed in more detail in Section 4.8, *Risks to piscivorous birds*.

Comparison of prey fish to South San Francisco Bay

Tomales Bay prey fish have higher methylmercury than fish from an area of San Francisco Bay that is similarly polluted by an upstream mercury mine. Specifically, Long-jawed mudsucker fish collected from channels in the vegetated marsh at the Walker Creek Delta have higher methylmercury than the same species collected from similar habitat in South San Francisco Bay (Figure 3-5).

The mudsuckers were collected with minnow traps placed in marsh channels at both the Walker Creek Delta and South San Francisco Bay, which is polluted by New Almaden, the largest-producing mercury mines in North America. The mudsuckers were all within target length, and their methylmercury-to-length relationship appears to be similar (Figure 3-6). Seasonality could explain some of the difference in fish mercury concentrations, because fish were collected on different dates from different years in San Francisco Bay (between April and September 2006–2008) than in Walker (June 2010).

However, it appears that we found a hotspot of methylmercury production and bioaccumulation at Walker Creek Delta, which confirms that our 2010 sampling focus at the Delta was appropriate. (In Section 8.3, *Monitoring Overview,* we describe how we plan to continue keeping our sampling focused at the Delta, at least in the near term.).

Invertebrates at Walker Creek Delta

Invertebrates within the vegetated marsh had elevated methylmercury concentrations. Invertebrate prey was sampled twice from the marsh at Walker Creek Delta, in 2000 and 2010.

Some birds are not obligate piscivores; that is, their diet is not limited to fish. For example, the Least bittern, a California Species of Special Concern, consumes both aquatic invertebrates and fish. An estimated 60% of the Least bittern diet consists of trophic level 2 (TL2) aquatic invertebrates (Ridolfi 2009; Table 10), such as tadpoles, salamanders, leeches, slugs, and insects. Biologists at Aquatic Science Center calculated that methylmercury should not exceed 0.01 ug/g in these TL2 prey (Ridolfi 2009; Table 12).

In 2000, Water Board staff collected invertebrates from the Delta. Mean methylmercury was 0.03 ug/g in small invertebrates (clams, shrimps, and snails; Table A-3; $n = 10$), a factor of 3 higher than the level for Least bittern.

In 2000, mean methylmercury was 0.08 ug/g in shore crabs (Table A-3; $n = 3$). In 2010, mean methylmercury was 0.06 ug/g in shore crabs collected in minnow traps set in channels in the vegetated marsh, a factor of 6 higher than the level for Least bittern (Table A-3; $n = 31$). This 2010 marsh shore crab data set provides a robust baseline to monitor methylmercury trend in marsh prey (see Section 8.3, *Monitoring Overview*).

4. Numeric Targets

This section presents the derivation of the numeric targets for methylmercury and desired conditions for the Bay.

4.1. Key points

- The numeric target to protect human health is 0.2 mg methylmercury per kg fish tissue, average, wet weight, in a commonly consumed fish species. This target applies to the average, wet weight mercury concentration in skinless fillets of legal size halibut.
- Methylmercury in halibut has consistently met the target, and is safe for one meal per week.
- The numeric target to protect fish-eating (piscivorous) birds, which we refer to as the "wildlife prey fish" target, is 0.05 mg methylmercury per kg fish, average, wet weight in whole trophic level 3 fish 5–15 cm in length.
- Methylmercury in prey fish met the target on a Bay-wide average basis when it was measured in 2009, but was about twice the target at the Walker Creek Delta in 2010.

4.2. Definition of "targets"

Numeric "targets" are measurable conditions that demonstrate attainment of water quality standards. Targets are the maximum amount of mercury allowed in a certain amount of water, animal or plant tissue, or sediments. Desirable targets are those that are easy and cheap to measure and interpret. Although the wildlife target proposed herein (prey fish mercury levels) is easy (and inexpensive) to measure, the human health target proposed herein (halibut mercury levels) is more challenging to measure, because halibut are more challenging to collect; laboratory preparation and mercury analysis of target samples is fairly easy and inexpensive. Additionally, it is fairly straightforward to interpret target data to evaluate whether fish are safe for birds and humans to consume; this analysis often requires some statistical analysis for size comparisons and sometimes for seasonality adjustments.

4.3. Steps to calculate targets

Fish tissue targets are calculated from the following equation:

$$\text{Target (ug/g)} = \frac{\text{(Body Weight [kg])} \times \text{(Reference Dose [g/day])}}{\text{(Food Intake at Trophic Level [ug/g/day])}}$$

In general, the steps to calculate targets are to identify (1) consumers (humans, birds, or mammals), (2) their body weight, (3) their consumption rate of prey, categorized by prey trophic level [food intake at trophic level], and (4) the appropriate reference dose [i.e., the safe amount of methylmercury for daily consumption].

4.4. Human health target for Tomales Bay

The target to protect human health is 0.2 mg methylmercury per kg fish tissue, average wet weight concentration, measured as total mercury in skinless fillets of legal size halibut. This is the same methylmercury concentration (0.2 mg/kg) as was adopted in other recent TMDLs (e.g., Walker Creek, San Francisco Bay, Guadalupe River, Sacramento–San Joaquin Delta, and others), and is more stringent than the U.S. EPA methylmercury criterion of 0.3 mg/kg (USEPA 2001).

Body weight (70 kg) and reference dose (0.0001 mg methylmercury per kg body weight per day) used in the target equation (see Section 4.3) are from the U.S. EPA methylmercury criterion, but the food intake rate is from a sport fish consumption study for San Francisco Bay (CDHS & SFEI 2000). U.S. EPA used the 90th percentile national sport fish consumption (intake) of 17.5 g/day, which like all percentiles discussed herein, includes those who do and do not consume fish. U.S. EPA recommends that states use local consumption data. Surprisingly, however, the 90th percentile San Francisco Bay sport fish intake is 16 g/d, lower than the national consumption rate. Therefore, Water Board staff used the 95th percentile San Francisco Bay sport fish intake of 17.5 g/d. Substituting this fish consumption rate into the equation from Section 4.3 results in a fish tissue target of 0.2 mg/kg. Additional calculation details are available in the San Francisco Bay mercury TMDL 2006 staff report.[3]

Each of the previously-mentioned TMDLs has site-specific targets that reflect locally-available species and consumption preferences. Water Board staff recommend halibut for the Tomales Bay target because they are a commonly consumed species. Halibut were the second most frequently consumed species from San Francisco Bay (CDHS & SFEI 2000; Figure 33). According to the Tomales Bay Watershed Council, halibut attracts "the largest number of anglers to Tomales Bay; peaking during the summer months, over 200 boats/day have been counted while other anglers fish from the shoreline. In an effort to quantify the fishing effort and to begin to understand the bay's halibut populations, CDFG and a number of volunteers began in 1995 to conduct a volunteer creel census…."

Further, Water Board staff recommends halibut for the target because it is protective to set the target level in high trophic level predators. Halibut are trophic level 4 predatory fish (SFEI 1999; Table 1). Bioaccumulation causes increasing methylmercury concentrations with each increase in trophic level. Therefore, targets set at high trophic levels are stringent and protective.

As explained in Section 4.9, *Analytical methods and interpreting target data*, nearly all mercury in fish muscle (i.e., skinless fillet) is methylmercury. Therefore, it is appropriate to continue analyzing halibut skinless fillets for total mercury.

4.5. Wildlife prey fish target for Tomales Bay

The target to protect wildlife was calculated in Ridolfi, (2010) using the following steps. The target is the same as the wildlife target in the Walker Creek Watershed Mercury TMDL.

Step 1 Identify piscivorous wildlife that feed largely on aquatic prey from the Bay

Step 2 Gather data needed for equation: maternal wildlife (consumer) body weight, consumption rate, typical diet (percent, size, and trophic level of fish; similarly for other prey), reference dose (safe dose of methylmercury determined from long-term feeding studies)

[3] Mercury in San Francisco Bay: Proposed Basin Plan Amendment and Staff Report for Revised Total Maximum Daily Load (TMDL) and Proposed Mercury Water Quality Objectives
Available from the Water Board at:
www.waterboards.ca.gov/sanfranciscobay/water_issues/programs/TMDLs/

Step 3 Tabulate consumers and safe prey methylmercury concentration by prey trophic level

Step 4 Select appropriately protective targets, generally the lowest methylmercury concentration for each trophic level.

Substituting these results into the equation from Section 4.3, results in a fish tissue target to protect wildlife of 0.05 mg methylmercury per kg fish. This target applies to average wet weight trophic level 3 whole fish concentrations in 5–15 cm length fish. Jacksmelt, for example, are plentiful in the Bay, and birds prey on these trophic level 3 fish.

Ridolfi (2010) presents potential wildlife targets for the Bay, based on a literature review and interviews with local wildlife experts. Wildlife targets selected for this TMDL are based on the most sensitive piscivorous birds resident in the Bay during the bird breeding season (i.e., Belted kingfisher, Caspian tern, and Black-crowned night-heron). This target is protective of all piscivorous bird species, because it protects the most sensitive species in the season of greatest sensitivity to mercury (breeding season).

This target was not driven by a threatened or endangered species. Readers may be interested to note that the Bay lies north, hence outside, of the California least tern's range. Therefore, the least tern was not considered in derivation of the wildlife targets. This tern is the most mercury-sensitive bird in San Francisco Bay and its presence there resulted in a lower target (0.03 mg/kg) in smaller fish (3–5 cm) in that Bay.

The target is based on chronic maternal dietary methylmercury. In particular, the reference dose was determined from long-term feeding studies, namely a 3-generation study in mallards. Residence during the breeding season is important because the avian toxicological endpoint is reproductive impairment, which is largely related to maternal uptake of methylmercury immediately prior to egg formation and laying. Therefore, the target is for average methylmercury in the maternal diet, for birds resident and feeding in Tomales Bay before and during their breeding seasons.

4.6. Threatened and endangered species

This section describes more fully why the wildlife target was not driven by a threatened or endangered species. Biologists evaluated 33 piscivorous bird and mammal species resident in the watershed. "Wildlife currently thought to be most likely at risk from mercury in an aquatic environment are terrestrial species that are primarily or exclusively piscivorous, ingesting methylmercury that has bioaccumulated and biomagnified in their aquatic prey" (USFWS 2005). The biologists narrowed the list of species to those most sensitive to methylmercury, which were 12 species of piscivorous birds that feed from the Bay during their breeding seasons (Ridolfi 2010, pp. 27–28). None of these birds are listed as threatened or endangered. Although Least bitterns are a California species of special concern, other birds are more sensitive to methylmercury from fish, so the wildlife target was not driven by a special status species.

As mentioned in Section 4.5, readers may be interested to note that the Bay lies north, hence outside, of the California least tern's range. This species of tern was the most mercury-sensitive bird in San Francisco Bay, resulting in a lower target (0.03 mg/kg) in smaller fish (3–5 cm). Since the least tern is not found in Tomales Bay, it was not considered in derivation of the wildlife target.

Additionally, we note that a federally threatened fish species, Green sturgeon, has been observed in the Bay (NMFS 2009). California sport fishing rules prohibit take of Green sturgeon at any time (DFG 2011). Sturgeon eat benthic invertebrates including shrimp, mollusks, amphipods, and even small fish (NMFS 2009). However, the Bay is exempted from their critical habitat. "Juvenile Green sturgeon rear and feed in fresh and estuarine waters from 1 to 4 years prior to dispersing into marine waters as subadults…. Subadult male and female Green sturgeon spend at least approximately 6 and 10 years, respectively, at sea before reaching reproductive maturity and returning to freshwater to spawn for the first time" (NMFS 2009). Because sturgeon are not a terrestrial species, and because they feed in the ocean rather than in the Bay during their adult reproductive years, the wildlife target was not driven by Green sturgeon.

4.7. Risks to human health

Bay sport fish have been sampled for pollutants many times since 1998. Due to high methylmercury concentrations in several species, OEHHA issued a fish consumption advisory.[4]

Importantly, commercial filter-feeding shellfish (oysters, mussels, and clams) and wild clams have levels of methylmercury that are safe for human consumption. Total mercury—and thus methylmercury—in wild clams is significantly lower than the target, and OEHHA determined that "the concentrations of methylmercury in the [*wild*] clam samples were sufficiently low to preclude any concern for public health."

Some sport fish have levels of methylmercury that are safe for 1 to 3 meals per week (see Figure 4-1). Methylmercury in halibut has consistently met the target, and is safe for one meal per week.

Methylmercury concentrations in sharks and bat rays are much higher than the OEHHA guidelines so no consumption is advised (see Figure 4-1). However, humans prefer to consume other sport fish. This is supported by a comprehensive consumption survey of California anglers fishing in San Francisco Bay (CDHS & SFEI 2000, pp. 51–53). This survey indicated that fewer than 10% of anglers consumed sharks, and so few anglers consumed rays that they were not one of the top 14 species consumed (CDHS & SFEI 2000, pp. 51–53 and Appendix E, *Questionnaire*, question 14d).

Leopard sharks and bat rays feed from the benthic food web in shallow mudflat areas in estuaries, so could accumulate methylmercury at the Walker Creek Delta. Studies in central and northern California bays and estuaries have shown that leopard shark movements are tidally influenced. They move into shallow mudflat areas to forage during high tides, and retreat to deeper water as the tide goes out." (Smith 2007, p. 14-4). Ackerman et al. (2000) confirmed these leopard shark tidal movements in the Bay. "Leopard sharks are opportunistic feeders, feeding on a wide variety of primarily benthic prey. Their diet is known to vary by location, season and shark size. Large adults are mostly piscivorous … while smaller adults and juveniles consume greater proportions of crustaceans, clam siphons, innkeeper worms (Urechis caupo), and fish eggs." (Smith

[4] *Safe Eating Guidelines for Fish and Shellfish from Tomales Bay (Marin County)*,
Available from Office of Environmental Health Hazard Assessment (OEHHA), Cal/EPA, at: www.oehha.ca.gov/fish/so_cal/tomales.html#tomales

2007, p. 14-5). Webber (1998) studied leopard shark diets at two locations in the Bay, and found one predominant food item at each location. Crabs from the *cancer,* not *grapsid*, family were the predominant prey at the south side of Hog Island in an eel grass bed, whereas innkeeper worms were the predominant prey at Indian Beach, near Heart's Desire Beach.

However, we do not know what proportion of methylmercury sharks and rays bioaccumulate from the Bay, because they spend a portion of their lives outside of the Bay. Leopard sharks "are seasonally abundant in central and northern California bays and estuaries, but leave for the open coast in the winter months…" (Smith 2007, p. 14-4). Similar to leopard sharks, bat rays travel into bays and estuaries in the spring and summer months, but leave in the winter (Meloni 2002, p. 462).

Nonetheless, at least a portion of the methylmercury threat that sharks and rays pose to human health may not be from the Bay, because these species may be consuming mercury-contaminated prey from areas outside the Bay. Consequently, we do not know whether the actions taken to solve the mercury problem in the Bay will reduce shark and ray methylmercury concentrations. However, as described below, this TMDL includes monitoring to evaluate this question. Additional action may be considered in the future if shark and ray methylmercury concentrations do not meet human health standards.

OEHHA and the Water Board's Surface Water Ambient Monitoring Program (SWAMP) measure mercury concentrations in a variety of coastal sport fish, such as sharks, surf perch, and jack smelt. They plan to sample coastal locations, including the Bay, at 10-year intervals. This information will be used to assess overall trends and human health risks.

4.8. Risks to piscivorous birds

Prey fish were sampled Bay-wide just once, in June 2009, and mean methylmercury was 0.05 mg/kg, equal to the target (see Figure 4-2). These fish were collected in the subtidal zone, using an otter trawl from a boat at the edge of the Bay.

Since prey fish were only sampled bay-wide once, we researched the scientific literature to evaluate how prey fish methylmercury concentrations might vary from March through August. This period extends from a couple of months preceding through the bird breeding seasons, which last from about May through August, and is when birds are most vulnerable to methylmercury (Ridolfi 2010, p. 45). We conclude that methylmercury in prey fish could have been equal to the target on average, based on review of the following two studies of prey fish in a nearby estuary.

In one study from San Francisco Bay, methylmercury in some prey fish species increased linearly with length (see Figure 4-3); although none of these same species were caught in Tomales Bay in 2009. It is reasonable to assume that the prey fish caught in Tomales Bay in late June were growing from March through August. If the linear increases in methlymercury concentrations observed in San Francisco Bay also occur in Tomales Bay, then methylmercury in prey fish could have been lower than the target from March through May, and could have been higher than the target in July and August, and still equal to the target on average from March through August.

In another study from San Francisco Bay, methylmercury concentrations in Threespine sticklebacks increased from early March and peaked in mid-May, but then declined through August (see Figure 4-3). These fish accumulated methylmercury as they grew between early March and mid-May. Biodiution is one possible explanation for the decline in fish methylmercury after mid-May, which occurs when the growth rate at the bottom of the food web exceeds the rate of methylmercury production (Eagles-Smith 2009, p. 8661). If the same accumulation and biodilution pattern occurred in Tomales Bay as in San Francisco Bay, then methylmercury in prey fish could have been higher than the target over a short period of time from early May through mid-June, and still equal to the target on average from March through August.

Generally, the Bay-wide mean reflects average methylmercury in the maternal diet, as these birds are highly mobile and forage from different areas around the Bay. However, piscivorous birds may forage over a smaller area when they are nesting. We are concerned that some piscivorous birds might feed mainly at the Walker Creek Delta when they are nesting, and if so, their offspring could be at increased risk from methylmercury. In particular, if egg-laying lasts for some time during nesting, such as multiple clutches, then there could be greater maternal uptake of methylmercury from reliance on prey at the Walker Creek Delta immediately prior to egg formation and laying, compared to their typical foraging from different areas around the Bay.

Prey fish at Walker Creek Delta

Prey fish were sampled again in May and June 2010 at only the Walker Creek Delta, and mean methylmercury was 0.10 mg/kg (Water Board 2012), and exceeded the target by a factor of 2. These fish were collected from throughout this delta in (a) the subtidal zone, using an otter trawl from a boat at the edge of the Bay, (b) main channel of Walker Creek, using an otter trawl from a boat, and (c) channels within the vegetated marsh, using minnow traps.

Prey fish methylmercury concentrations were lowest in the subtidal zone at the edge of the Bay, and highest in channels in the vegetated marsh (Figure 4-4). Concentrations were very similar in 2009 and 2010 in fish from the edge of the bay, although the variance was much higher in 2010.

Some birds are not obligate piscivores; that is, their diet is not limited to fish. We are concerned that if they feed heavily on invertebrates at the Walker Creek Delta, they could be at increased risk from methylmercury.

Invertebrates at Walker Creek Delta

Invertebrates were sampled at Walker Creek Delta in both 2000 and 2010 (see Section 3.5, *Invertebrates at Walker Creek Delta*). In 2010, mean methylmercury was 0.06 ug/g in grapsid shore crabs collected in minnow traps set in channels in the vegetated marsh, a factor of 6 higher than the safe level biologists calculated for non-obligate piscivorous birds. This 2010 shore crab data set provides a robust baseline to monitor methylmercury trend in marsh prey, which is expected to decline as sediment mercury concentrations decline (see Section 8.3, *Monitoring Overview*).

Marshes are sites of high methylmercury bioaccumulation, and therefore consumption of marsh prey is a potential risk to wildlife health (Ridolfi 2010 addendum). However, as described in the following paragraphs, we do not know enough about which species and what sizes of marsh prey best represent the methylmercury risk to wildlife, and therefore we do not recommend they be adopted as targets at this time. Instead, we recommend that shore crabs (marsh-dwelling invertebrates) be used to measure methylmercury time trends in marsh invertebrates.

Biologists calculated the following safe methylmercury concentration: 0.01 mg methylmercury per kg, wet weight, edible tissue of trophic level 2 (TL2) marsh-dwelling invertebrates (Ridolfi 2010, Table 12). Two of the 12 piscivorous birds in Tomales Bay (*ibid*, Table 12) require this low concentration: Black-crowned night-herons and Least bitterns; the five other invertevore birds on the list can safely tolerate consuming invertebrates with somewhat higher methylmercury concentrations.

The invertebrate portion of these two birds' diets is as follows. Black-crowned night-herons and Least bitterns consume invertebrates for approximately 35% and 60% of their diets, respectively (Ridolfi 2010, Table 12). These night-herons are opportunistic foragers "with a varied diet … ranging from 100% fish to 100% large aquatic insect larvae to 100% mammals, with other combinations…" (Davis 1983). They eat larger fish than other herons, around 10 cm length (Post 2008). However, little information is available on the size of the night-herons' invertebrate prey or the proportion of crabs in their diet. Least bitterns mainly eat small fish and insects, but, similar to night-herons, little information is available on the size of their invertebrate prey, especially the size and proportion of crabs in their diet (Poole 2009).

It is unclear that crabs well represent "trophic level 2 invertebrates" for several reasons. Invertebrates comprise a wide range of creatures, including aquatic and terrestrial insects, worms, tadpoles, leeches, slugs, prawns and crayfish, clams, mussels, squid, and eggs. Crabs are probably higher than trophic level 2 (herbivores) because they are omnivores and detritovores. Opportunistic foragers will consume plentiful and easy-to-catch prey. In 2010, the field crew found that crabs are plentiful during the day at Walker Creek Delta, although night-herons are, as their name implies, night feeders. The crabs were easy for humans to catch in minnow traps, so presumably, they are also easy for birds to catch. However, the difference in when crabs are present and when night herons forage means that these birds may well consume invertebrates other than the species of shore crabs caught in minnow traps. Finally, there may be other plentiful and easy-to-catch prey that better represent trophic level 2 invertebrate methylmercury risk to piscivorous birds.

We recommend using shore crabs to measure methylmercury time trends in marsh prey, but not as targets. That is because of the robust baseline data set from 2010 (see Section 3.5, *Invertebrates at Walker Creek Delta*), and the uncertainty in the portion of the diet of sensitive bird species made up by shore crabs. We anticipate a decrease in methylmercury in the marsh-dwelling invertebrates that piscivorous birds consume, as total mercury in sediment declines, as discussed in Section 6, *Linkage Analysis*.

4.9. Analytical methods and interpreting target data

This section explains why biota data, such as that shown on figures and tables in this report, is presented as "mercury" but compared to "methylmercury" thresholds. Biota methylmercury concentrations are more frequently analyzed as total mercury than as methylmercury. As long as measured total mercury concentrations are well below methylmercury targets, or percent methylmercury is known, then total mercury is a conservative (protective) measure for reporting methylmercury levels in biota. For example, total mercury, and hence methylmercury, in clams from Tomales Bay is so far below levels of concern for human health that clams do not even appear on the consumption advisory.[5] Therefore, it would be appropriate to analyze the edible portion of clams for total mercury, should there be any reason to evaluate clam mercury levels in the future. Additionally, nearly all mercury in fish muscle (i.e., skinless fillet) is methylmercury. Therefore, it is appropriate to continue analyzing halibut skinless fillets for total mercury.

The wildlife prey fish target presents a different situation than sport fish because predators consume the entire fish. Some animals may ingest significant volumes of sediment, and sediment at Walker Creek Delta has elevated inorganic mercury. Consequently, total mercury analysis of these prey could potentially overstate the methylmercury concentrations. Predators do not bioaccumulate inorganic mercury from their prey.

Therefore, in 2009, tests were conducted on Bay prey fish to measure whether there were significant amounts of inorganic mercury in their digestive tracts. Aquatic Science Center staff measured both total and methylmercury in prey fish from the Bay (specifically, from Shiner perch, Speckled sanddab, and Staghorn sculpin), and found that nearly all mercury in whole body prey fish is methylmercury (Ridolfi 2010, p. 44 and Table 23). These fish were all caught with an otter trawl at the Bay edge, not caught with minnow traps in marsh channels. Therefore, it is appropriate to continue analyzing whole prey fish caught at the Bay edge for total mercury.

However, marsh-dwelling prey fish, especially those from the mercury-polluted Walker Creek Delta, may have excessive amounts of inorganic mercury in their digestive tracts, and therefore methylmercury could be significantly lower than total mercury. These are the prey fish caught with minnow traps in marsh channels. Therefore, it may be necessary in the future to determine the proportion of methylmercury in marsh-dwelling prey fish by measuring both total and methylmercury in prey fish caught with minnow traps in marsh channels. In the short term (that is, for the first two rounds of sampling, see Section 8.3, *Monitoring Overview*), it is appropriate to continue analyzing whole prey fish caught in marsh channels for total mercury.

5. Source Analysis

This section presents our understanding of the sources and loads of mercury to the Bay.

5.1. Key points

- There are three sources of mercury to the Bay: mercury mining, watershed loading (i.e., mercury-enriched geology), and global atmospheric deposition

[5] *Safe Eating Guidelines for Fish and Shellfish from Tomales Bay (Marin County)*, Available from Office of Environmental Health Hazard Assessment (OEHHA), Cal/EPA, at: www.oehha.ca.gov/fish/so_cal/tomales.html

- Past mercury mining is by far the largest source of mercury to the Bay (see Table 5-1)
- The Gambonini Mercury Mine was cleaned up in 2000; clean-up was effective. It reduced mercury loads discharged from the mine site by greater than 90%

5.2. Soil erosion process

Landscapes are constantly evolving from natural processes, and one mechanism of landscape change is soil erosion. Precipitation is the main cause of soil erosion and subsequent transport of sediment through creeks, rivers, and bays. In the Bay watershed, infrequent, large winter storms, meaning the storms with greatest precipitation intensity and duration, are the "episodic" storms that cause the greatest soil erosion and transport the largest sediment loads (Whyte 2000). Additionally, site conditions control how readily soil can be eroded. Site conditions are both natural (i.e., geology and topography) and affected by human activity (e.g., land uses such as timber harvesting, grazing, and mining, all of which took place in the Bay watershed).

Mercury occurs in soils and largely remains sediment-bound even once it reaches surface waters, because it has very low solubility. Kirchner (2011) describes how mercury from mines is transported downstream:

> Abandoned mines are [significant] sources of water pollution in [California]. Precipitation can trigger episodic erosion of tailings piles and [contaminated] soil, leading to downstream transport of pollutants in stormwaters. Many sediment-bound metals, such as [mercury], are toxic pollutants of great concern. Delivery of these sediment-bound metals to downstream waters [depends] on storm intensity, duration, and frequency, and also on mine site conditions that control how readily sediment can be eroded from the surface or mobilized from in-channel deposits. In the California Coast Range and the Sierra Nevada, [mercury]-laden mine waste continues to be transported downstream.

Mining waste (mercury-laden sediment) from the Gambonini Mercury Mine was transported in episodic, storm-driven pulses through a single, ephemeral drainage and into Walker Creek. Most of this creek is a transport zone. That is, it serves as a conduit for sediment, where there is little change in the volume of accumulated sediment over the long term. However, Walker Creek does have a few depositional areas that accumulate sediment, including mercury-laden sediment (For more information on Walker Creek sediment transport and mercury concentrations, see *Walker Creek Watershed mercury TMDL*, "watershed TMDL", Water Board 2008.). However, this local sediment accumulation is a temporary phenomenon—when a large storm hits, it is likely to scour depositional areas and transport that previously accumulated material downstream. In any one storm, it is possible for a large pulse of newly-eroded sediment to be transported from the surrounding landscape, through the full 21-km length of Walker Creek, and out the mouth of the Bay. It is unlikely that the entire pulse will make it out of the Bay mouth. In that case, the rest of that pulse will travel a shorter distance and accumulate in depositional areas in Walker Creek or the Delta.

Similarly, smaller storms also transport sediment. In any single storm, some sediment might be scoured from a depositional area and be transported part of the way down Walker Creek. Overall, the range of storms that occur each year will typically transport some of the sediment load through Walker Creek and out the mouth of the Bay.

Precipitation erodes and stormwater runoff transports both clean sediment from non-mined areas and mining waste (mercury-laden sediment) from the Mine (Water Board 2008). Mine site runoff discharges via a single, ephemeral drainage at the top of Walker Creek, whereas the watershed is a dispersed source of clean sediment. Chileno Creek is a particularly significant source of clean sediment because it has a comparatively high sediment load. Consequently, the proportion of clean sediment in Walker Creek increases with distance from the Mine. Mean total mercury decreases from 320 mg/kg in the Mine tailings pile (Whyte 2000) to 1.4 mg/kg in 2003 in a depositional area of Walker Creek just downstream of Chileno Creek (Water Board 2008, Table 4.2).

Mining waste pulsed downstream from the Mine until 2000 when the Mine was cleaned up. Over time, mining waste accumulated in depositional areas will be pulsed out of Walker Creek, and suspended sediment mercury concentrations will approach the allocation of 0.5 mg/kg assigned to areas downstream of the Mine by the watershed TMDL (Water Board 2008). Suspended sediment mercury concentrations may even decrease to 0.2 mg/kg, the background concentration found in Chileno Creek, and the allocation assigned to the remainder of the Walker Creek watershed by the watershed TMDL (Water Board 2008). Some of these suspended sediments accumulate at the Delta. Therefore, eventually, these same low mercury concentrations will be reached at the Walker Creek Delta, too.

5.3. Mercury loads from the Gambonini Mercury Mine

Mercury loads from all sources are summarized in Table 5-1, including both pre-cleanup and post-cleanup mercury loads from mining. We expect continued decline in mining-related mercury loads and accumulation at Walker Creek Delta, because of effective cleanup at the Gambonini Mercury Mine.

Mining loads were measured both as discharges from the Mine and as accumulated at the Delta, as follows. In early 1998 before cleanup, Water Board staff measured both suspended sediment and mercury loads from the Mine over two months in a wet, El Niño winter. Kirchner (2011) calculated that over just this two-month period, 135 kg mercury and 2,300 MT sediment were discharged from the Mine site.

Five years after site cleanup, in early 2005, Water Board staff again measured both suspended sediment and mercury loads from the Mine. Kirchner calculated that 0.13 kg mercury and 13 MT sediment were discharged over two months in this average rainfall year. Additionally, Kirchner calculated that *if the same storm conditions had occurred in 2005 as had occurred in the 1998 El Niño winter*, much *higher* loads would have been discharged over a two-month period; he estimated suspended loads of 9.3 kg mercury and 950 MT sediment. Comparing 1998 actual to 2005 estimates, and holding rainfall constant, Mine site cleanup resulted in 92–93% mercury load reduction, and 55–60% sediment load reduction. This is greater than 90% pollutant load reduction, which indicates effective site cleanup.

We also have estimates of mining loads from sediment cores at the Walker Creek Delta (see Figure 3-2). Although estuaries are dynamic mixing zones, deltas are depositional areas. Dr. Johnson (2009) was able to age-date the cores with Cesium 137, and, together with mercury concentration data, determined mercury accumulation patterns. The peak mining-related mercury annual accumulation rate at Walker Creek Delta was nearly 45 kg, corresponding to the 100-year storm event in 1982 (Johnson 2009; Figure 6a). The second-highest peak mining-related Hg accumulation rate at the Walker Creek Delta was about 32 kg/yr, corresponding to the El Niño winter event in 1998 (Johnson 2009; Figure 6a). These results correspond to our understanding of sediment pulses from episodic storms.

As mentioned previously, we expect continued decline in mining-related mercury accumulation because of effective Mine site cleanup. Additionally, we anticipate stream bank stabilization projects will be undertaken along Walker Creek to comply with both the Walker Creek watershed mercury and the Tomales Bay pathogens TMDLs (see Section 8). Stream bank stabilization will slow sediment mercury transport from Mine-polluted mainstem Walker Creek, but we expect continued high loads of clean sediment from Chileno Creek. Therefore, mercury concentration in sediment that accumulates at the Walker Creek Delta might approach the pre-mining, background, concentration as discussed in the next section.

5.4. Mercury loads from watershed soils

Soils in the Bay watershed are naturally enriched in mercury because of their location in the California Coast Range, which is one of the world’s five most productive mercury mineral belts (Rytuba 2003). Also, mercury emitted from global industrial activities deposits on the landscape surface, increasing surface mercury concentrations.

Watershed mercury comes from naturally-occurring mercury in local geology, from global industrial emissions to the atmosphere, and other potential sources to the landscape or creeks (e.g., broken thermometers). Global mercury emissions have been increasing since the industrial age began. Mercury emitted into the atmosphere is later deposited across vast areas of the earth’s surface. Global mercury emissions have been increasing since the industrial age began, they contaminate the Bay environment (Sanders 2008, Figure 2a; Hornberger 1999, Figure 4), and this mercury is bioaccumulating in piscivores (Vo 2011).

Land use changes, such as timber harvesting and grazing, have greatly increased sediment delivery to the Bay over pre-European conditions (Johnson 2009). However, these land use changes do not change mercury concentrations in surface soil or sediment. In contrast, mercury loading from global industrial emissions has increased mercury concentrations in surface soil and sediment over pre-European conditions (Johnson 2009; Hornberger 1999).

Similar to loads for mining, we have estimates of watershed mercury loads, exclusive of mining contributions, from sediment cores at the Walker Creek Delta. The peak watershed mercury annual accumulation rate in the Delta was nearly 20 kg in 1982, corresponding to a 100-year storm event (Johnson 2009; Figure 6a). The next-highest peak accumulation rate was over 15 kg/yr in 1998, corresponding to an El Niño winter (ibid.). These are the peak loads for this one delta within Tomales Bay; next, let us examine average loads for the entire Bay.

Aquatic Science Center staff estimated that the watershed contributes 21 kg/yr, on average, to Tomales Bay (Ridolfi 2010). To calculate this number, they multiplied suspended sediment loads by background mercury sediment concentration, 0.2 mg/kg (Water Board 2008). The suspended sediment loads data is from USGS gages on Lagunitas Creek at Samuel P. Taylor State Park (11460400) and Walker Creek near the Town of Marshall (11460750).

5.5. Mercury loads from global atmospheric deposition

The "atmospheric deposition" mercury source is mercury from global industrial emissions to the atmosphere that later deposits onto the earth's surface, including onto the water surface of the Bay (Atmospheric deposition onto the land surface surrounding the Bay is considered in the previous section, *Mercury loads from watershed soils*). There are no significant local mercury emissions sources to the atmosphere, because this is a rural, non-industrial area.

Our estimate of the mercury load from global atmospheric deposition was calculated following the approach used in the San Francisco Bay mercury TMDL staff report (Water Board 2004). Tsai and Hoenicke (2001) measured both dry and wet mercury mass deposited to the surface of San Francisco Bay, and estimated a deposition rate of 4.2 $\mu g/m^2/yr$. This estimate is similar to other studies of mercury deposition rates in California, which had a range of 1.7 to 5.3 $\mu g/m^2/yr$ (mean; Gill 2008, Table 3.1). Additionally, Gill (2008) measured both dry and wet mercury deposition at Point Reyes Bird Observatory near the town of Bolinas, which is much closer to Tomales Bay than San Francisco Bay. He estimated that atmospheric deposition is higher, 5.9 $\mu g/m^2/yr$ (Gill 2008, Table 3.3).

Annual load estimates range from 0.4–0.7 kg/yr, and are calculated by multiplying the deposition rate by the water surface area of Tomales Bay. Based on the rate for San Francisco Bay, the estimated load of mercury that deposits on the surface of Tomales Bay is 0.4–0.5 kg/yr (Tsai & Hoenicke 2001, cited in Ridolfi 2010). Based on the higher observed rate at Point Reyes (Gill 2008), the estimated load is 0.5–0.7 kg/yr.

5.6. Other potential sources and loads of mercury

There are few other sources or loads of mercury to the Bay, because it is located in west Marin County, which has very low human population density, and very little development, commercial, or industrial activities (see Section 3, *Background*). Additionally, the Water Board's Basin Plan prohibits direct discharge of wastes to the Bay. Consequently, no industrial wastewater or sewage from residences is discharged to the Bay.

Sewage and urban stormwater are minor sources of mercury to the Bay. Most residences at the Bay's edge had relied on on-site sewage disposal systems (septic systems). Due to the Tomales Bay pathogens TMDL, many older septic systems at the Bay's edge are being replaced with small wastewater treatment facilities. These facilities provide more effective treatment than septic systems, and hence we expect a reduction in mercury loads from sewage.

Marin County's stormwater pollution prevention program (MCSTOPPP) already covers west Marin County, even though it is a rural area. MCSTOPPP implements best management practices to reduce mercury discharges. For example, they provide outreach and education about proper disposal of mercury-containing fluorescent tubes. More information about MCSTOPPP's on-going effective pollution prevention work is available on their website (http://mcstoppp.org/).

6. Linkage Analysis

This section presents the linkage analysis, which describes the relationship between mercury sources and the proposed targets (i.e., the proposed allowable methylmercury concentrations in biota).

6.1. Key points

- Past mercury mining is by far the largest source of mercury to the Bay. The vast majority of this source was sediment-bound, inorganic mercury, and is moving downstream and entering the Bay via sediment transport processes. As such, this TMDL focuses on reducing suspended sediment mercury loads entering the Bay.
- In the aquatic environment, inorganic mercury can be converted to methylmercury and bioaccumulate in the food web. The geochemical conditions found in estuarine sediments are often highly conducive for producing methylmercury. This linkage assumes a 1:1 relationship between surface sediment mercury concentrations to mercury in prey fish.
- In the Bay, sediment mercury concentrations are elevated in the Walker Creek Delta compared to the rest of the Bay, but are expected to decrease by a factor of 2 in the Walker Creek Delta. Similarly, small prey fish mercury concentrations are elevated in the Walker Creek Delta compared to the rest of the Bay. In the Walker Creek Delta, prey fish mercury concentrations need to be reduced by about a factor of 2 to meet the wildlife target.

6.2. Linkage

The main purpose of this linkage analysis is to describe the links between sources of mercury and the proposed targets (methylmercury concentrations in biota). In other words, this analysis describes mercury behavior in the Bay (see Figure 6-1). This includes transport of mercury from sources to water bodies, chemical transformations that make mercury more toxic (i.e., methylation of mercury), and uptake into the food web (i.e., bioaccumulation). This analysis is the basis for our estimate of the assimilative (loading) capacity of mercury in Tomales Bay, our estimate of the mercury TMDL for the Bay, and allocations of loads to sources.

6.3. Conceptual model

We have a simple conceptual model for mercury in the Bay, which is supported by current scientific literature (see Merritt and Amirbahman (2009) for technical details of mercury cycling in estuaries). Our model describes the links between mercury sources to the Bay and elevated methylmercury in biota. Our conceptual model is simply that there are three necessary conditions to have a mercury problem: (a) source(s) of mercury, (b) methylating conditions, and (c) a food web.

We know from Section 5, *Source Analysis*, that there are several sources of mercury to the Bay. Nearly all this mercury is sediment-bound, inorganic mercury. In the aquatic environment, under certain chemical conditions, inorganic mercury can be converted to a more toxic form, "methylmercury." Inorganic mercury is "methylated" by a biological process. In low-oxygen conditions in aquatic environments, sulfate-reducing bacteria convert sulfate to sulfides for energy, and in an incidental process these bacteria methylate mercury.

"Estuaries are…critical areas of [*mercury*] methylation, due to geochemical conditions in sediments that are conducive to [*methylmercury*] production, such as anoxia and periodic wetting and drying from tidal flux" (Chen 2009). In Long Island Sound, for example, Hammerschmidt and Fitzgerald (2006) found that "methylmercury in biota of coastal marine ecosystems is related to its production in underlying sediments." Accordingly, Tomales Bay is likely a critical area of methylmercury production. In Chesapeake Bay, Gilmour (2010) found that methylmercury production and accumulation in marsh sediments has "a potentially significant negative impact on marsh-utilizing organisms."

Methylmercury, the most bioavailable form of mercury, biomagnifies as it moves through the food web. Namely, methylmercury concentrations increase from algae to herbivores to small prey and to predators. We assume a one-to-one relationship between changes in methylmercury in prey fish and changes in surface sediment total mercury concentrations.

6.4. Water transports inorganic mercury

In the Bay, the main role that water plays for mercury is transport. Prior to the Gambonini Mercury Mine cleanup, Whyte (2000) found that particulate mercury represented over 99.97% of the total mercury discharged from the Mine. The next largest mercury source is erosion of watershed soil. Water transports these strongly sediment-bound mercury sources down creeks, where they accumulate as surface sediment in delta areas in Tomales Bay, such as at Walker Creek Delta where mining waste is being buried by cleaner sediments (see Section 3.5, *Sediment at Walker Creek Delta*).

Total mercury concentrations in water samples from the Bay are relatively low, and appear to have declined since the Mine was cleaned up in 2000 (Figure 6-2). Field personnel used boats to collect water samples from two or more locations. One-day average total mercury concentrations did not exceed the most stringent water quality objective. Since the one-day sample average did not exceed either the 4-day or 30-day objectives, and the 1-hour objective is nearly 100 times higher than the 4-day objective, there is no need to collect and evaluate samples over a different averaging period.

Water samples were also analyzed for methylmercury several times, in 2000–2002 (Water Board 2012), 2007–2008 (Negrey 2011), and 2009 (Ridolfi 2011). Peak total (i.e., unfiltered) methylmercury was 0.408 ng/L, 0.225 ng/L, and 0.157 ng/L, respectively. Further, there were no correlations between fish methylmercury and water methylmercury, or between mercury in sediment and water in the comprehensive 2009 Tomales Bay data set and reported in the *Impairment Assessment* (Ridolfi 2010). This demonstrates that water plays a much smaller role than sediment in methylation and bioaccumulation.

6.5. Delta sediment improved since Mine cleanup

Inorganic (total) mercury concentrations in sediment at the Walker Creek Delta have declined since the Mine was cleaned up in 2000. This is a rare instance where downstream improvements have been observed in just a decade. Mean total mercury concentrations declined from 1.6 to 0.9 mg/kg over a decade, and the variance (scatter) was also greatly reduced in this decade (Figure 6-3). Lower variance provides greater confidence in this downward trend. Mine cleanup means that less mining waste, and a proportionately greater amount of sediment from other parts of the Walker Creek Watershed, at background level of 0.2 mg/kg, is accumulating at the Delta.

We predict that total mercury in surface sediment at the Walker Creek Delta will decrease by about a factor of 2 from 0.9 mg/kg to at least 0.5 mg/kg, which is the Walker Creek Watershed mercury TMDL (Water Board 2008).

We further predict that total mercury in Delta sediments will decrease to lower than 0.5 mg/kg, as mining waste loads decrease. Currently, some mining waste is still caught in depositional areas along Walker Creek. Infrequent large storms will wash these downstream where they may accumulate at the Delta, or be transported all the way to the ocean. These episodic, storm-driven pulses of mining waste will continue to decline, so that more and more of the storm-generated sediment load will be generated from non-mined and non-mercury-enriched areas. Therefore, we expect the total mercury concentration in the sediment will eventually decrease to the background concentration of 0.2 mg/kg.

In contrast to total mercury, no time trend was detectable in sediment methylmercury at the Walker Creek Delta between 2000 and 2009 (Figure 6-4). Sediment was sampled in 2000 and 2009, but the 2009 sediment sampling protocols were different (4-point composites from 0–5 cm depth) from the discrete depth-specific cores collected in 2000. These changes in sampling protocol preclude detection of a time trend.

6.6. Methylation and Bioaccumulation

Surface sediment methylmercury concentrations were higher in the Mine-polluted Delta than in other areas of the Bay in 2009 (Figure 6-5). Both inorganic (total) mercury and methylating conditions contribute to elevated methylmercury at the Delta. However, it is difficult to measure their relative importance, especially with small data sets. We attempted to hold methylating conditions constant by comparing only mudflat samples (and not samples from marsh or subtidal areas). That is, we compared samples taken from areas likely to be exposed to similar methylating conditions. Over the small data set, sample size of 8 (“n = 8”), we found that more methylmercury was produced in mudflats with higher concentrations of inorganic mercury, see Figure 6-6 (Water Board 2012).

Nonetheless, methylating conditions are an important factor. Both nitrogen and carbon are indicators of the organic matter content of sediment (Ridolfi 2010, p. 59), and organic matter (carbon) is necessary for methylation (Lambertsson 2006). Nitrogen and carbon provide a better explanation than total mercury (R-squared of about 40% for total mercury compared to 70% and 60%, respectively for nitrogen and carbon, Water Board 2012). These data, and current scientific understanding, are insufficient to calculate future sediment methylmercury concentrations at the Walker Creek Delta with statistical confidence. Consequently, we assume a conservative one-to-one relationship between changes in methylmercury in prey fish and changes in surface sediment inorganic (total) mercury concentrations.

Only recently have scientists begun to develop accurate techniques to identify the sources of methylmercury in prey fish. Gehrke (2011) and others developed a technique that uses mercury isotopes, and recently showed that in San Francisco Bay, mercury from mines is accumulated in prey fish. This work shows that inorganic mercury from mining waste is bioavailable and bioaccumulated.

In Tomales Bay, sediment mercury concentrations are elevated in the Walker Creek Delta compared to the rest of the Bay, but are expected to decrease by at least a factor of 2 in the Delta. Similarly, small prey fish mercury concentrations are elevated in the Delta compared to the rest of the Bay. In the Delta, prey fish mercury concentrations need to be reduced by about a factor of 2 to meet the wildlife target. This linkage is the basis for loading capacity.

7. Loading capacity, TMDL, and allocations

This section establishes the mercury loading capacity and Total Maximum Daily Load (TMDL) for the Bay and sets allocations to sources.

7.1. Key points

- The loading capacity is 0.2 mg mercury per kg suspended sediment (dry weight), bay-wide annual average
- The TMDL is equal to the loading capacity
- The allocations are:
    - 0.2 mg mercury per kg suspended sediment, annual average, discharged from Lagunitas Creek, which is calculated from current conditions in surface sediment at the Lagunitas Creek Delta
    - 0.5 mg mercury per kg suspended sediment, annual average, discharged from Walker Creek, which is equal to the Walker Creek Watershed mercury TMDL
    - 0.2 mg mercury per kg suspended sediment, annual average, discharged from all other tributaries to the Bay , which is calculated from current conditions in surface sediment collected from areas downstream of these tributaries
    - 0.7 kg mercury per year deposited directly on the Bay water surface from global atmospheric sources, equal to the current load

7.2. Loading capacity, TMDL, and allocations

Loading (assimilative) capacity is "[t]he greatest amount of loading that a water can receive without violating water quality standards" (Code of Federal Regulations, Title 40, §130.2[f]). TMDLs are "[t]he sum of the individual waste load allocations for point sources and load allocations for nonpoint sources and natural background. … TMDLs can be expressed in terms of either mass per time, toxicity, or other appropriate measure" (Code of Federal Regulations, Title 40, §130.2[i]). We are establishing concentration-based loading capacity, TMDLs, and allocations in accordance with this provision of the Clean Water Act because, as described in Section 6, *Linkage Analysis*, mercury levels in fish are linked to sediment mercury concentrations.

Loading capacity is 0.2 mg mercury per kg suspended sediment (dry weight), bay-wide annual average, and is based on our assessment of current conditions that both the human health and wildlife targets are met. As described in Section 4.7, the human health target is consistently met. In 2009, the prey fish target was met on a Bay-wide average basis (see Figure 4-2). At that time, sediment total mercury was 0.2 mg/kg, excluding the Mine-polluted Walker Creek Delta (rounded to one significant figure, see Figure 2-4). Therefore, the loading capacity is 0.2 mg/kg.

The TMDL is equal to the loading capacity, and is 0.2 mg mercury per kg suspended sediment (dry weight), bay-wide annual average.

The allocation to Walker Creek is 0.5 mg mercury per kg suspended sediment, annual average, discharged from Walker Creek. This allocation is calculated to ensure that the wildlife target is met at the Walker Creek Delta. Additionally, this allocation is equal to the Walker Creek Watershed mercury TMDL (Water Board 2008). As described in Section 6, sediment mercury is expected to decrease by at least a factor of 2 in the Walker Creek Delta to meet the upstream watershed TMDL of 0.5 mg/kg, and prey fish methylmercury concentrations need to be reduced by about a factor of 2 to meet the wildlife target in the Walker Creek Delta.

Allocations to Lagunitas Creek, and other all other tributaries that discharge to Tomales Bay, are 0.2 mg mercury per kg suspended sediment (dry weight), annual average. These allocations are calculated from current conditions in surface sediment collected from areas downstream of these tributaries, and based on our conclusion that the prey fish target was met on a Bay-wide average basis in 2009.

The allocation to atmospheric deposition is 0.7 kg mercury per year deposited directly on the Bay water surface from global atmospheric sources. This allocation is the current load and is based on our conclusion that the prey fish target was met on a Bay-wide average basis in 2009.

The allocations meet the TMDL as an area-weighted average mercury concentration, as follows. Calculate the area-weighted average mercury concentration by taking the sum of each allocation multiplied by its respective area of the Bay, dividing the sum by the total area of the Bay, and rounding to one significant figure (see Figure 7-1). The Bay will meet the TMDL when the allocation is met at Walker Creek Delta, because allocations are already met at the Lagunitas Creek Delta and other areas in the Bay.

7.3. Margin of safety

TMDL analyses must incorporate a margin of safety to address potential uncertainties. The margin of safety is intended to account for any lack of knowledge concerning the relationship between load and wasteload allocations and water quality. The margin of safety can be derived either explicitly or implicitly. An implicit margin of safety involves using conservative assumptions (assumptions more likely to be over-protective than under-protective) in the analysis. Alternatively, an explicit margin of safety involves reserving a specific mercury load allocation for the margin of safety. This TMDL incorporates an implicit margin of safety.

The TMDL is based on a loading capacity that is derived using conservative assumptions. The TMDL is a bay-wide annual average of 0.2 mg mercury per kg suspended sediment (dry weight). This is equal to the loading capacity based on the current (2009) bay-wide average concentration of total mercury in surface sediments, excluding the Mine-polluted Walker Creek Delta. Our assessment of current conditions is that both the TMDL's human health and wildlife numeric targets are met on a Bay-wide basis. That means the Bay currently has the capacity to assimilate the load from the Walker Creek Delta that was excluded from the loading capacity calculation.

In addition, levels of mercury in the Walker Creek Delta surface sediments have already decreased significantly since the Gambonini Mercury Mine has been cleaned up and will continue to decrease. Assuming a conservative one-to-one relationship between changes in methylmercury in prey fish and changes in surface sediment inorganic (total) mercury concentrations, further expected reduction in mercury levels in the Walker Creek Delta will result in lower levels in fish. Consequently, future Bay-wide average levels are expected to be less than both the TMDL's human health and wildlife numeric targets that are currently met on a Bay-wide basis and prey fish in the Walker Creek Delta are also expected to meet the numeric targets.

7.4. Seasonal variations and critical conditions

The TMDL must describe how seasonal variations and critical conditions were considered. Critical conditions are a description of when and under what conditions impairment occurs. Specifically, the evaluation of temporal patterns in water quality data can provide substantial insight into the impairment because the analysis identifies the times of greatest impairment and because many of the factors affecting critical conditions exhibit seasonal variations (e.g., flow and weather conditions and source activity).

We considered both of these concepts in our source analysis. In the California Coast Range, almost all erosion and sediment delivery to channels occurs during the wet season, from October through May. The Gambonini Mercury Mine is located in the California Coast Range. Additionally, critical conditions occur during infrequent, large storms, because they transport the largest loads of mercury-laden sediment from this Mine (Whyte 2000). However, Mine site cleanup reduced mercury loads by 90% (Kirchner 2011).

Additionally, we considered both of these concepts for bioaccumulation and risk to wildlife. Specifically, we evaluated prey fish methylmercury concentrations during the bird breeding season, when birds are most sensitive to methylmercury.

7.5. Water quality standards attainment

The mercury TMDL and allocations herein will result in attainment of the Basin Plan narrative objective for bioaccumulation. Sport fish already meet the human health target (Figure 4-1). Prey fish already meet the wildlife target on a Bay-wide average basis (Figure 4-2). Additionally, the allocation for Walker Creek will ensure that prey fish will meet the wildlife target at the Walker Creek Delta. Mercury in the Bay already meets numeric water quality objectives in both the Basin Plan and in U.S. EPA's California Toxics Rule (Figure 6-2).

8. Implementation and monitoring

This section describes the implementation actions that have been completed to control mercury sources to the Bay and our plans to monitor to determine whether targets are achieved.

8.1. Key points

- The Water Board does not foresee a need for any actions in addition to the work listed below to address mercury in the Bay.
- Actions already required by the Walker Creek Watershed mercury TMDL, particularly a grazing waiver (also required by the Tomales Bay pathogens TMDL), will be sufficient to attain targets and achieve allocations.
- Mercury concentrations at the Walker Creek Delta in the Bay have already decreased as a result of cleanup of the Gambonini Mercury Mine in the Walker Creek watershed in 2000.
- We plan to monitor mercury in the Bay to determine whether mercury concentrations continue to decrease.

8.2. Implementation

No new implementation actions are required by this mercury TMDL for Tomales Bay, because previous actions addressed the main source (i.e., the Gambonini MercuryMine) and mining waste that accumulated downstream.

As explained in Section 5 (*Source Analysis*), the largest mercury source, the Gambonini Mercury Mine, was cleaned up in 2000. In 2008, via the Walker Creek Watershed Mercury TMDL, the Water Board imposed requirements on all creekside property owners downstream of the Mine and Soulajule Reservoir (requirements were not imposed on property owners upstream of the Reservoir because no mining took place there). Subsequently, the Water Board adopted a *Conditional Waiver of Waste Discharge Requirements for Grazing Lands in the Tomales Bay Watershed* that required grazing operations of at least 50 acres to implement best management practices that minimize mercury discharges and methylmercury production. This waiver also implements the Tomales Bay pathogens TMDL.

As explained in Section 6 (*Linkage Analysis*), mining waste at the Walker Creek Delta is being buried by cleaner sediments. Previous regulatory actions—Gambonini Mercury cleanup and the waiver—are sufficient to implement this Tomales Bay mercury TMDL.

8.3. Monitoring overview

We plan to monitor total mercury in Walker Creek Delta sediment to determine whether mercury concentrations continue to decrease in Tomales Bay. Monitoring is anticipated to be conducted as part of the Water Board's Surface Water Ambient Monitoring Program (SWAMP), depending on the availability of funding.

The purpose of this monitoring is to track conditions at the Walker Creek Delta, to verify that less mercury mining waste is accumulating at the Delta, resulting in less methylmercury bioaccumulating into biota.

Total mercury levels in sediment at the Delta are anticipated to be monitored at 5-year intervals. If a measurable decrease in sediment total mercury concentration is observed, then subsequent monitoring may also include prey species (i.e., mercury in prey fish) It is expected that prey fish will meet the wildlife numeric target in the Walker Creek Delta within 20 years. Monitoring of shorecrabs may also be useful to assess trends in biota.

We also anticipate evaluating methylmercury in sport fish on a Bay-wide basis, to periodically evaluate the risk to human health. These data have been used in the past by the Office of Environmental Health Hazard Assessment in developing the Bay's fishing advisories. SWAMP anticipates monitoring coastal sport fish at about 10 year intervals. Sport fish would be collected according to the then-current SWAMP sampling plan; we anticipate that these programs will continue to collect and evaluate several sport fish species, such as shark, jack smelt and surfperch. For more information about SWAMP, including sampling plans, refer to the Water Board's website (www.waterboards.ca.gov). The next sampling is expected to occur in 2019.

The following description of monitoring at the Walker Creek Delta presents a possible approach to assessing improved water quality the Walker Creek Delta in the next 5-20 years.

The data set, sampling, and evaluation schedule would be the following:

<u>Baseline Data (2009 & 2010)</u>

- Walker Creek Delta (2009 & 2010)
  - Surface sediment Total Hg 0.9 mg/kg (mean, dry wt.)
  - Prey fish Total Hg 0.09 mg/kg (mean, whole fish, wet wt.)
  - Shore crabs Methyl Hg 0.06 mg/kg (mean, flesh, wet wt.)

<u>Year 0 (2012) TMDL adoption</u>

<u>Year 5 (2017) Monitoring</u>

- Surface sediment Total Hg at Walker Creek Delta
  - Evaluate for time trend and compare to allocation of 0.5 mg/kg

<u>Year 10 (2022) Monitoring</u>

- Surface sediment Total Hg at Walker Creek Delta
    - Evaluate for time trend and compare to allocation of 0.5 mg/kg
- If surface sediment mean Total Hg for either year 10, or both years 5 & 10, is less than baseline, consider monitoring prey fish and shore crabs
    - Evaluate for time trend and compare prey fish mean Total Hg to wildlife target of 0.05 mg/kg

As described above, we would evaluate the trend in mercury from baseline through the first 5-10 years. If necessary, in years 15 and 20, we would monitor total mercury in surface sediment at the Walker Creek Delta as well as prey fish, to evaluate attainment of the wildlife target. . If the numeric targets are not met, we could consider monitoring prey fish in the Lagunitas Creek Delta as a reference location to allow us to compare the two locations and determine the best achievable mercury concentrations in Tomales Bay.

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(Watershed Council 2003)	Tomales Bay Watershed Council. 2003. *The Tomales Bay Watershed Stewardship Plan: A Framework for Action*. July. 137 pp. www.tomalesbaywatershed.org/
(Webber 1998)	Webber, J.D., and J.J. Cech, Jr. 1998. *Nondestructive diet analysis of the leopard shark from two sites in Tomales Bay, California*. California Fish and Game Vol. 84, Issue 1, pp. 18–24.
(Whyte 2000)	Whyte, D. and. Kirchner, J. 2000. *Assessing water quality impacts and cleanup effectiveness in streams dominated by episodic mercury discharges*. Science of the Total Environment 260:1–9.

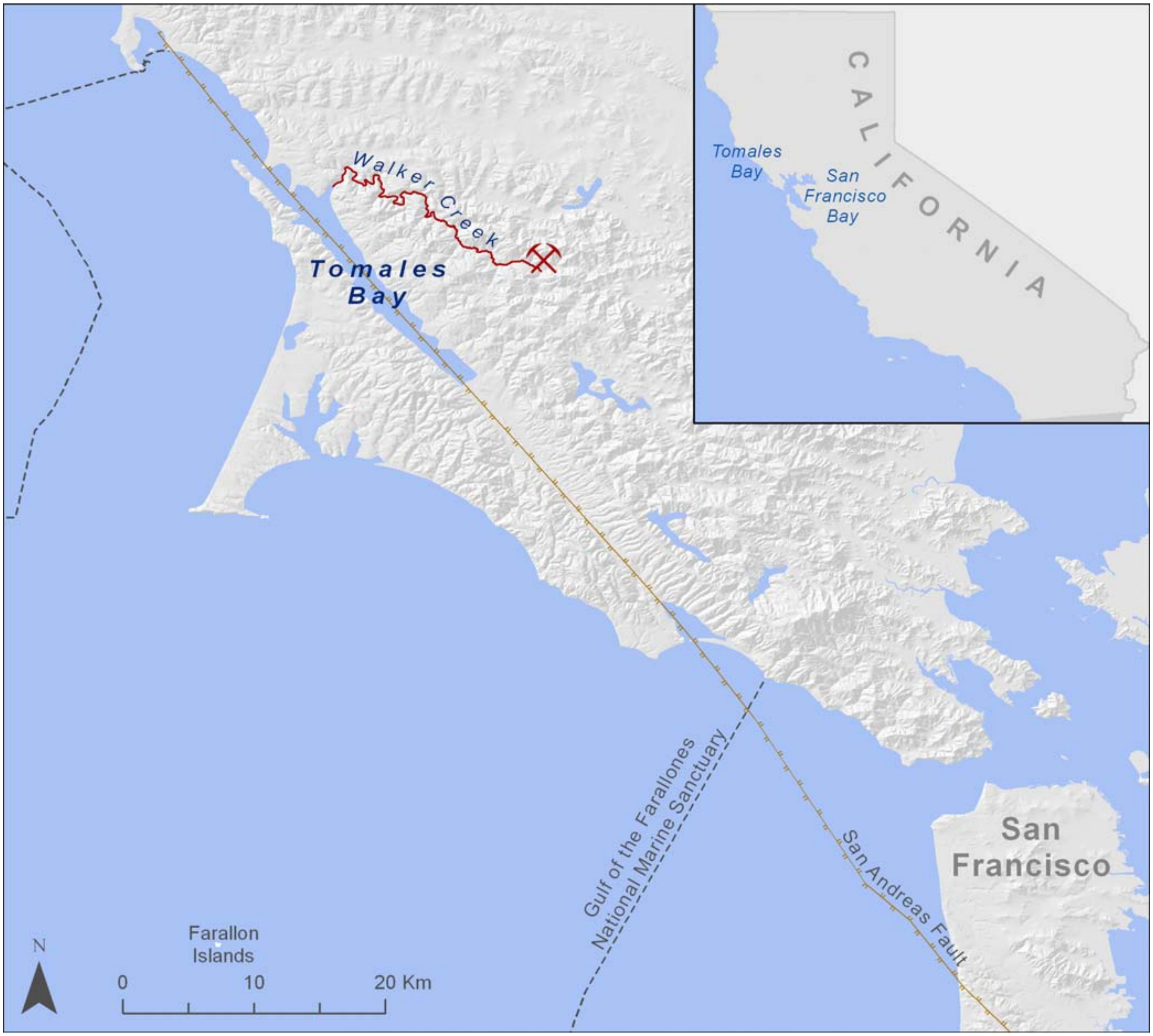


Figure 1-1 Location map

Tomales Bay is located north of San Francisco Bay, within a national marine sanctuary. The mine symbol indicates the Gambonini Mercury Mine.

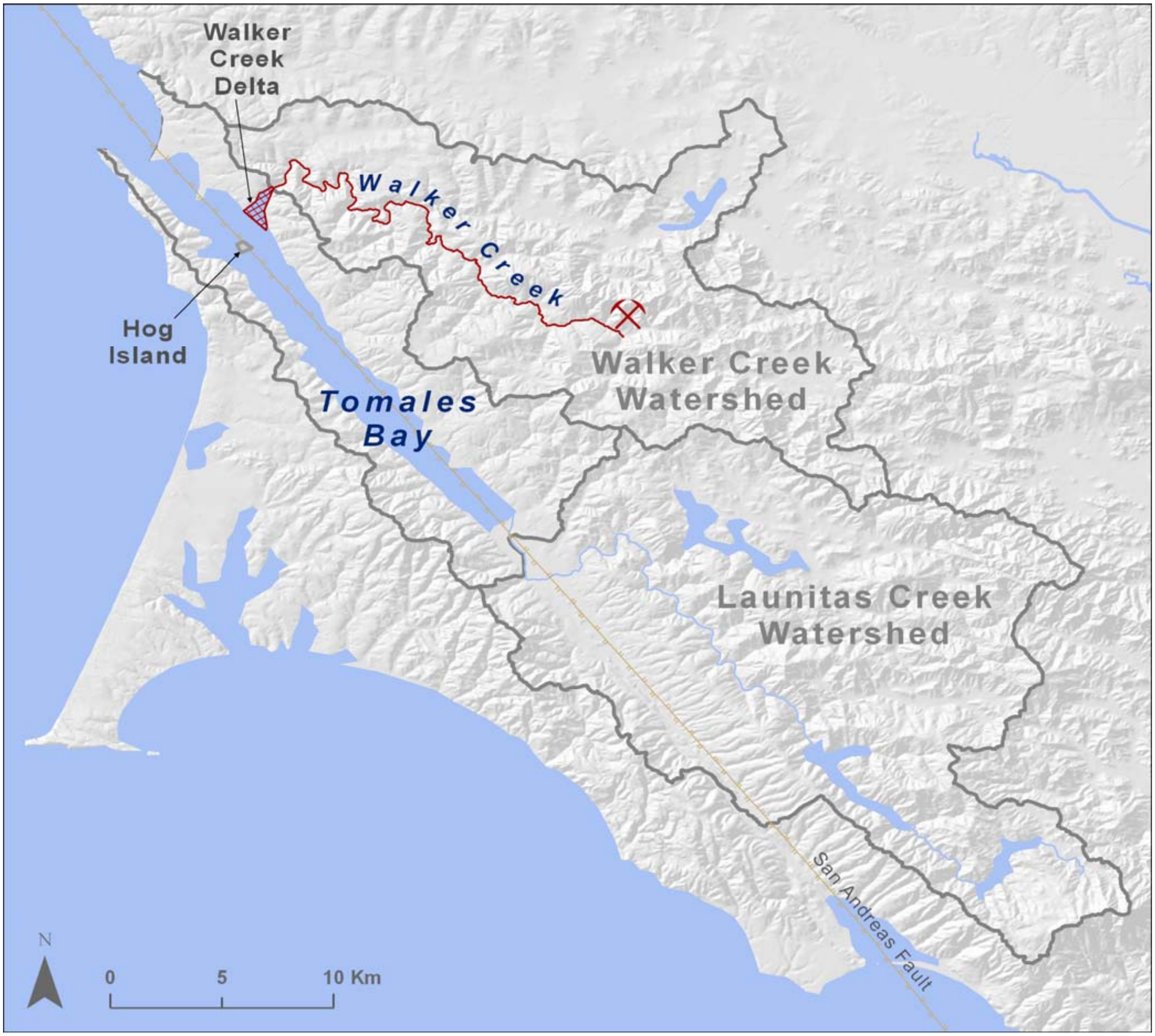


Figure 2-1 Watershed map

Lagunitas Creek, at the south (head) of Tomales Bay, is the largest subwatershed and contributes the highest volume of freshwater flows. Walker Creek, at the north (mouth) of the Bay, is the second largest subwatershed and contributes the second-highest freshwater flows. The mine symbol indicates the Gambonini Mercury Mine.

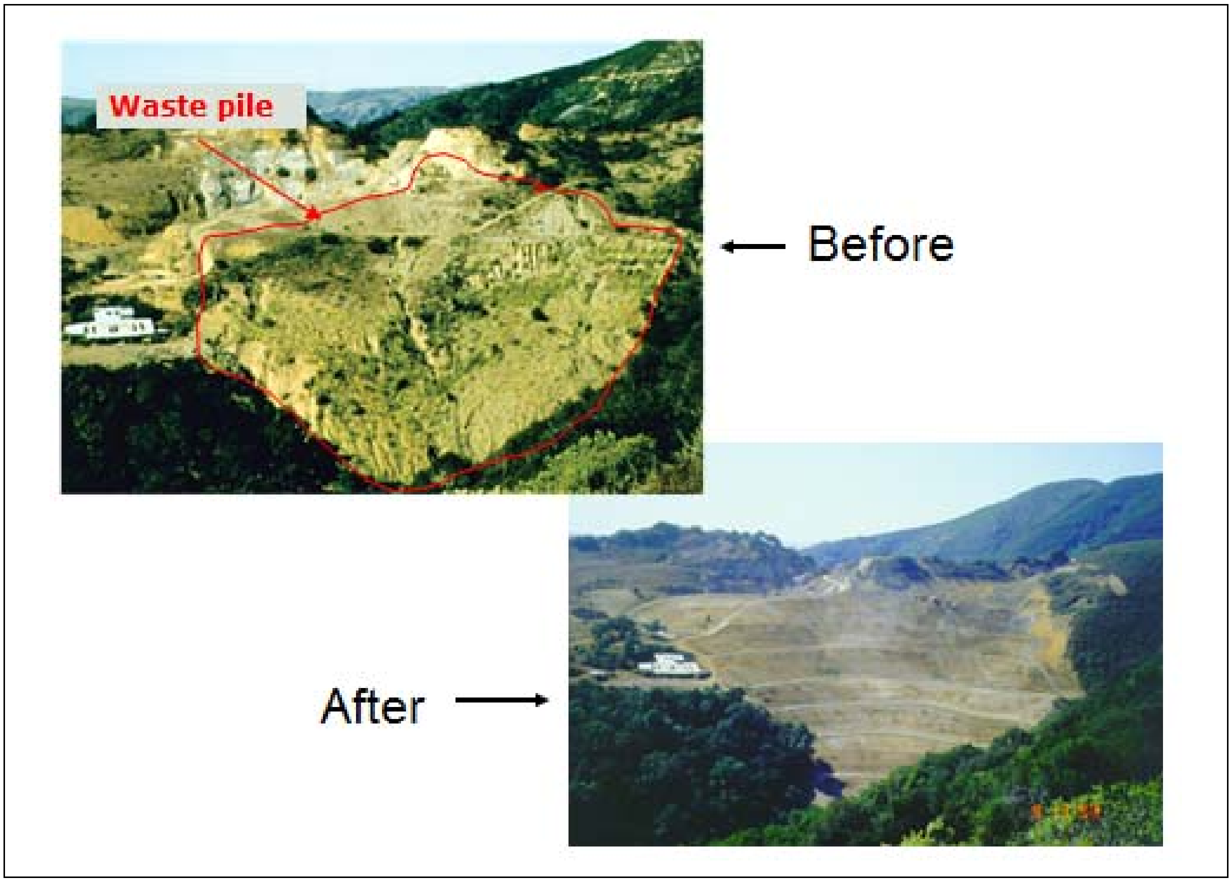


Figure 2-2. The Gambonini Mercury Mine before and after remediation

Upper left photo shows rills and gullies; not easily apparent at this scale is that the gullies were so deep the landowner had dumped cars in them. The lower right photo shows the terraced and compacted waste pile prior to revegetation.

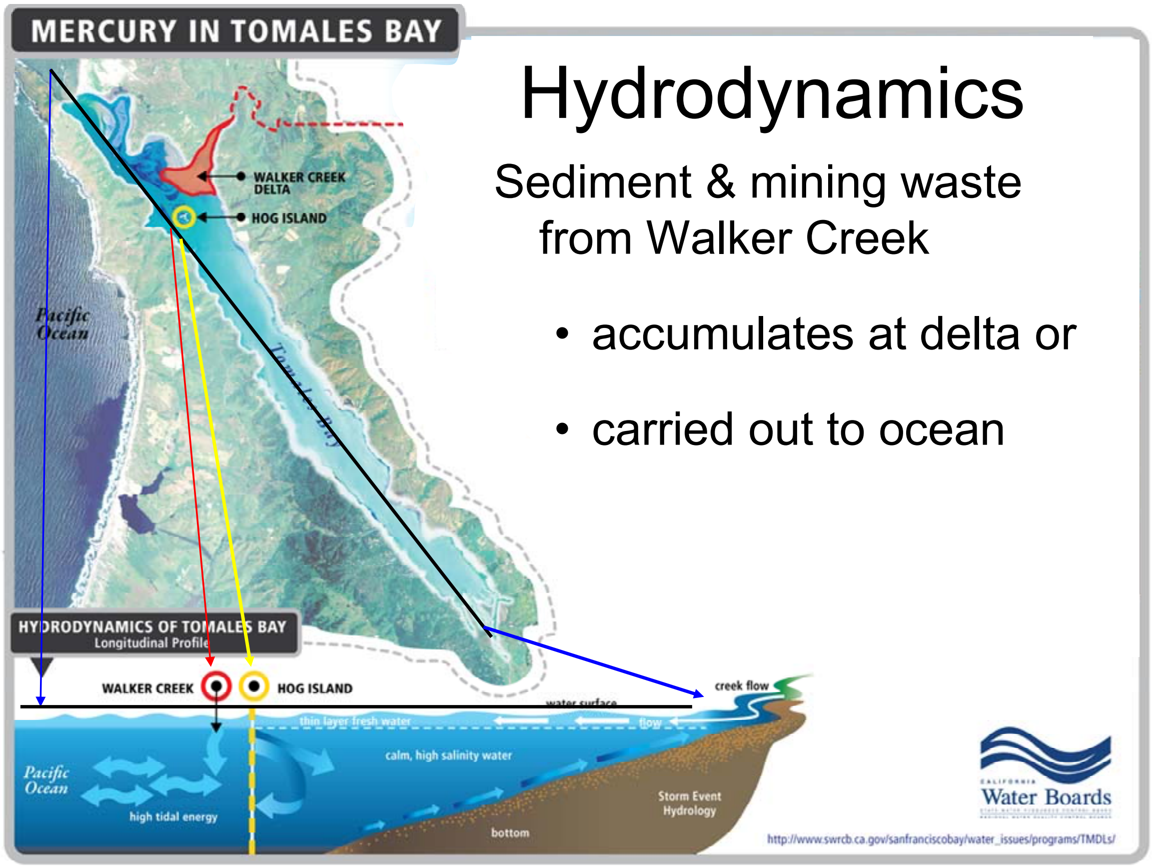


Figure 2-3. Hydrodynamics of Tomales Bay

This is a generalized representation of Tomales Bay hydrodynamics during a storm. It illustrates that mercury-laden sediment from the Gambonini Mercury Mine is either trapped at the Walker Creek Delta or transported out the mouth of the Bay to the Pacific Ocean. The upper figure is a satellite image of Tomales Bay. The lower figure is a cross section along the length of the Bay, and shows that most of the flow from Walker Creek is to the ocean. Very little of the Creek flow makes it up the Bay past Hog Island (yellow), where the Bay is very calm, and where differences in temperature and salinity prevent mixing with the Creek water.

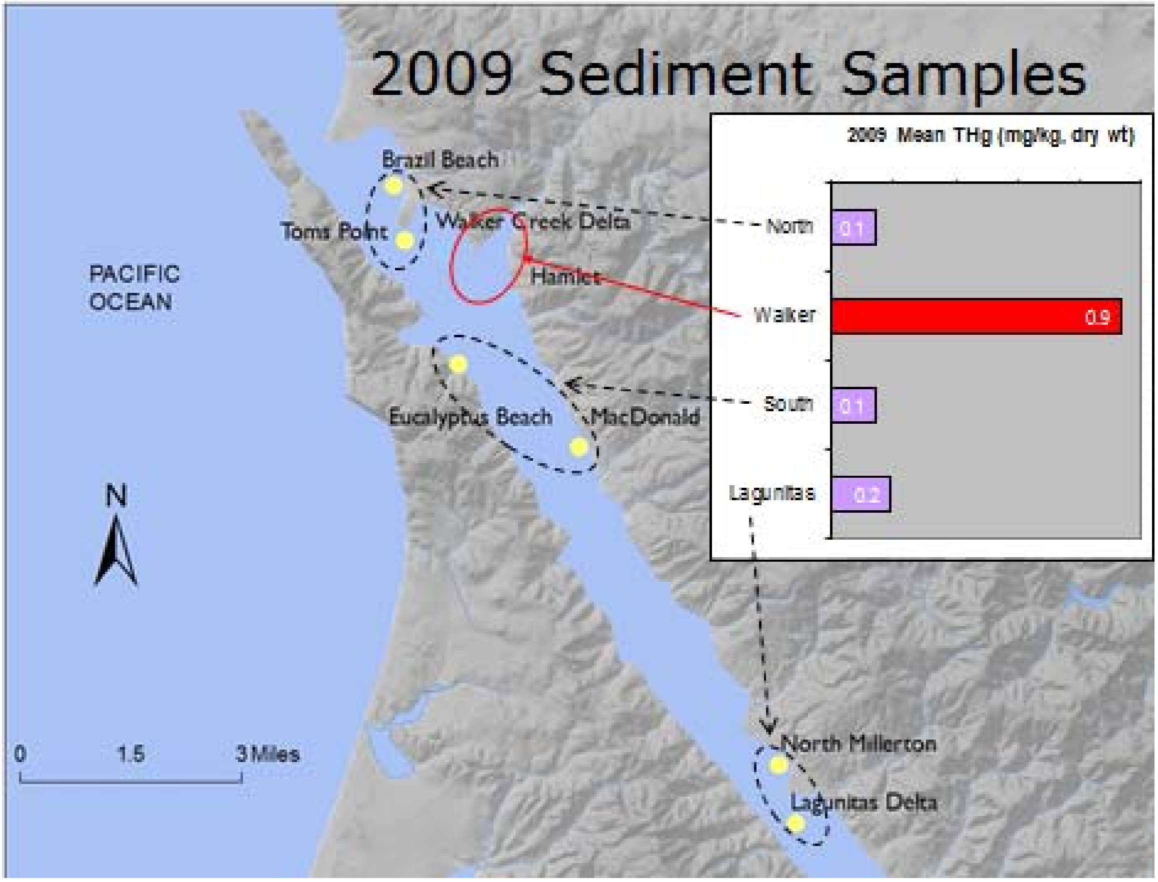


Figure 2-4-. Sediment Mercury Concentrations

Total mercury concentrations in surface soil samples (2009) from the edge of Tomales Bay. Elevated mercury concentrations are localized in the Walker Creek Delta.

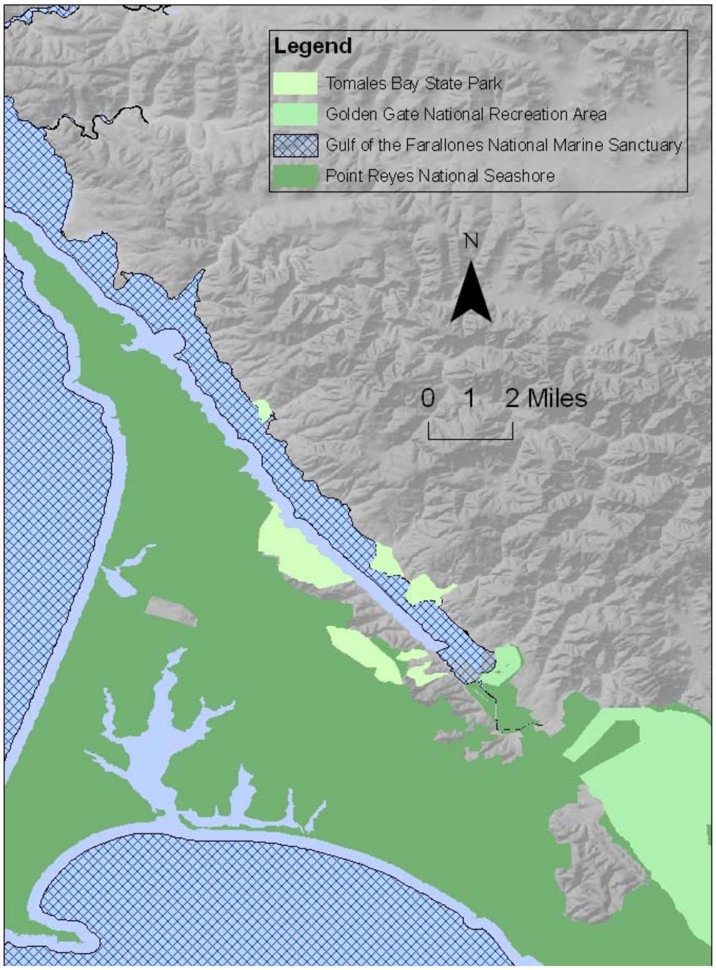


Figure 2-5. Protected areas around Tomales Bay

Tomales Bay lies within and is surrounded by protected areas, including the Gulf of the Farallones National Marine Sanctuary, which extends much further into the Pacific Ocean, see map at: http://sanctuaries.noaa.gov/pgallery/atlasmaps/images/gf_2000.jpg .

Citation: (Ridolfi 2010, Figure 7)

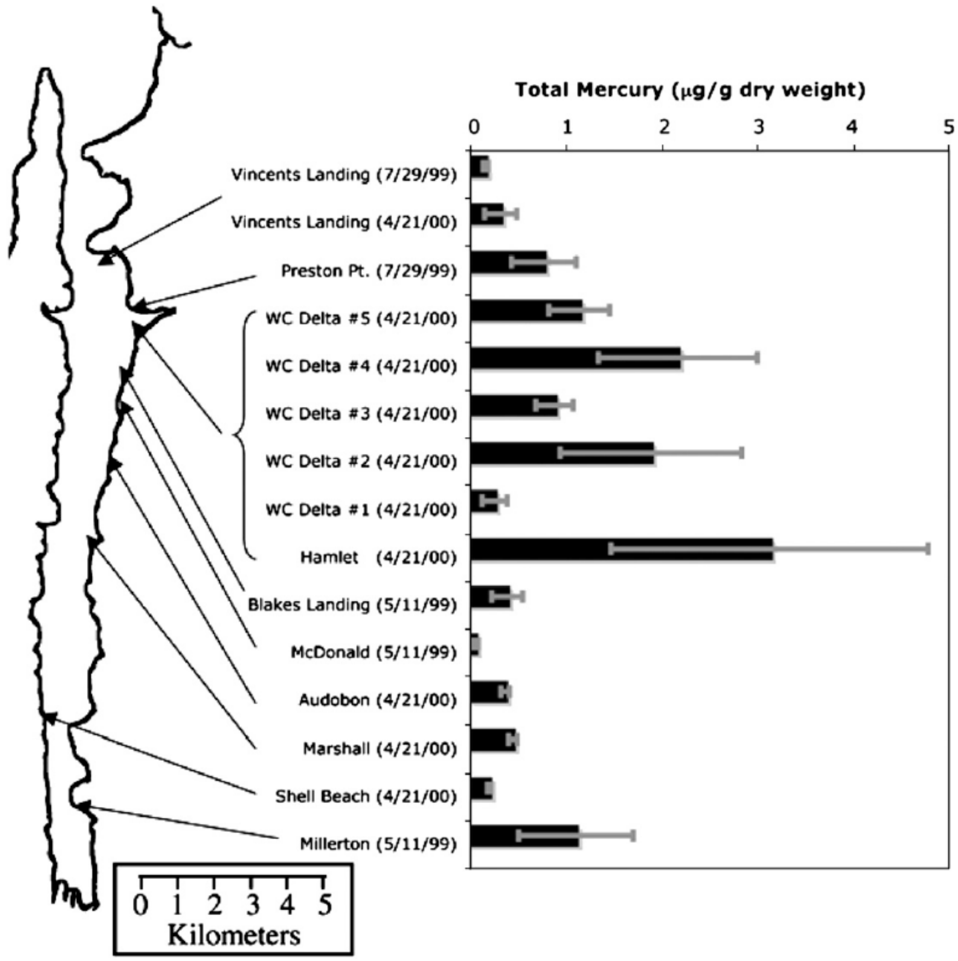


Figure 3-1. Total mercury in surface sediment (upper 5 cm; µg/g, dry wt)

Total mercury in surface sediment (1999–2000). The black bars are mean total mercury and the grey error bars are two times the standard deviation. There were 3 to 5 samples per location.

Mercury was elevated at the Walker Creek Delta (indicated by a bracket for bars from "WC Delta" to Hamlet) compared to other areas in Tomales Bay. The slightly elevated mercury at Preston Pt. is likely due to inputs directly from Walker Creek and/or transfer of mercury through the Delta.

The unexpectedly elevated mercury at Millerton, located 13 km from Walker Creek, could be due to transport from the Mine via Walker Creek or pollution from towns near Millerton Point (see Section 2, *Hydrodynamics prevent dispersal of mercury*, and Section 3.5, *Bay-wide sediment*).

Citation: (Johnson 2009, Figure 2)

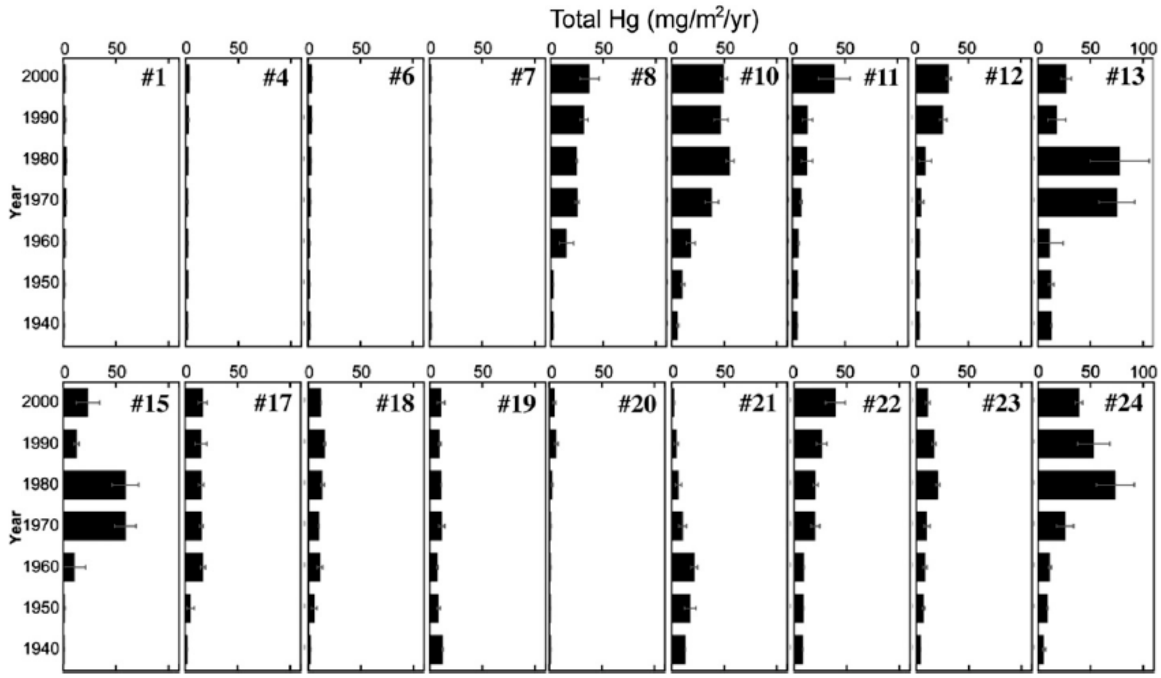


Figure 3-2. Mining waste is buried by cleaner sediments at Walker Creek Delta

Sediment cores (numbered) from the Walker Creek Delta were analyzed for total mercury concentration and age-dated with Cesium 137, yielding ± 5-year average mercury flux (accumulation) rates; the error bars represent one standard deviation. All of the cores were deep enough to reach pre-mining depths.

Peak mercury loadings likely correspond to an extremely large storm in 1982 that burst the Gambonini Mercury Mine tailings dam. Peak accumulation of greater than 50 mg/m2/yr occurred between 1975 and 1985 near the mouth of Walker Creek (cores #13, 15, and 24).

Several cores at the eastern side of the Delta (cores #8, 10, 12, and 22) had highest mercury concentrations near the surface. This is likely the result of natural estuary sediment mixing processes, although recent increases in mercury loads, perhaps from mining waste released from depositional areas along Walker Creek, cannot be ruled out. Nonetheless, the overall trend is less mercury accumulating at the Delta since the mine closed.

In other words, mining waste is slowly being buried by cleaner sediments at the Walker Creek Delta.

Citation: (Johnson 2009, Figure 4)

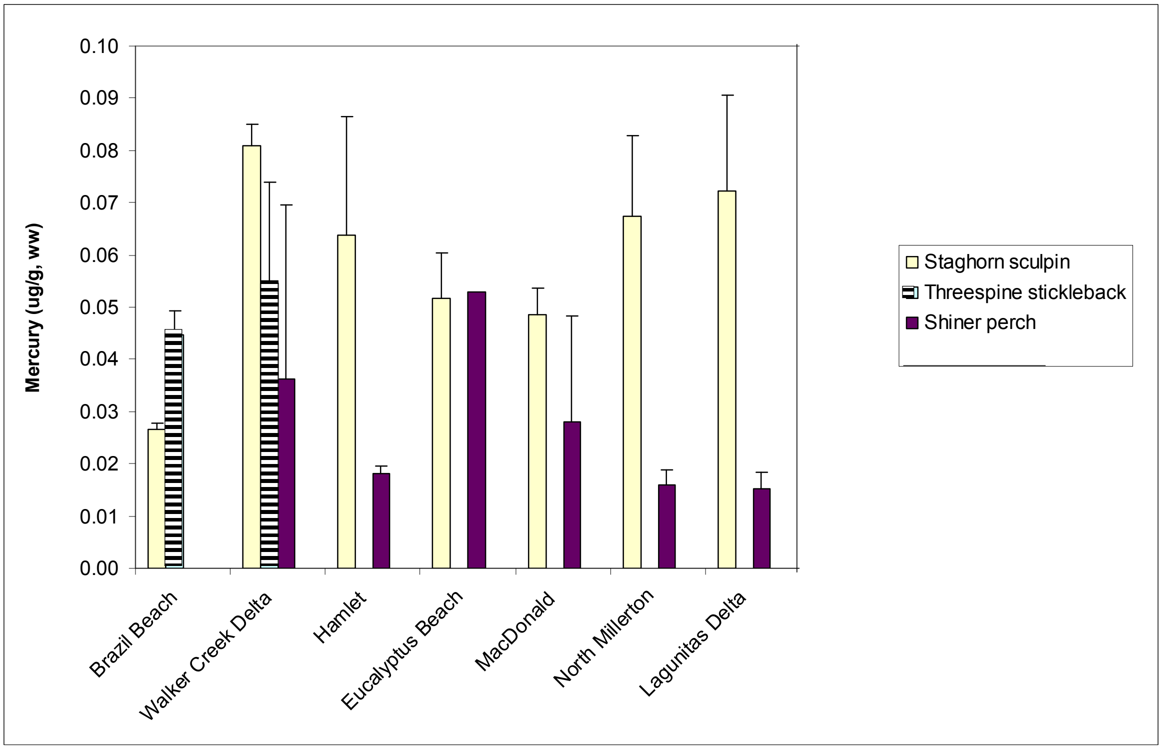


Figure 3-3. Prey fish methylmercury concentrations by species and site (2009)

There was no statistical difference in methylmercury concentrations in prey fish among sites in 2009, the only year with prey fish data from throughout Tomales Bay. This figure shows mercury (µg/g, wet wt, whole fish) concentrations in three frequently-caught species (Shiner perch, Staghorn sculpin, and Threespine stickleback). Fish were in target size range (5-15 cm), and collected with otter trawl at bay edge; *n* = 36 total. The bars represent the mean mercury concentration; error bars represent one standard deviation. (Nearly all mercury in prey fish is methylmercury, Section 3, Interpreting target data.)

Citation: (Ridolfi 2010, Figure 17)

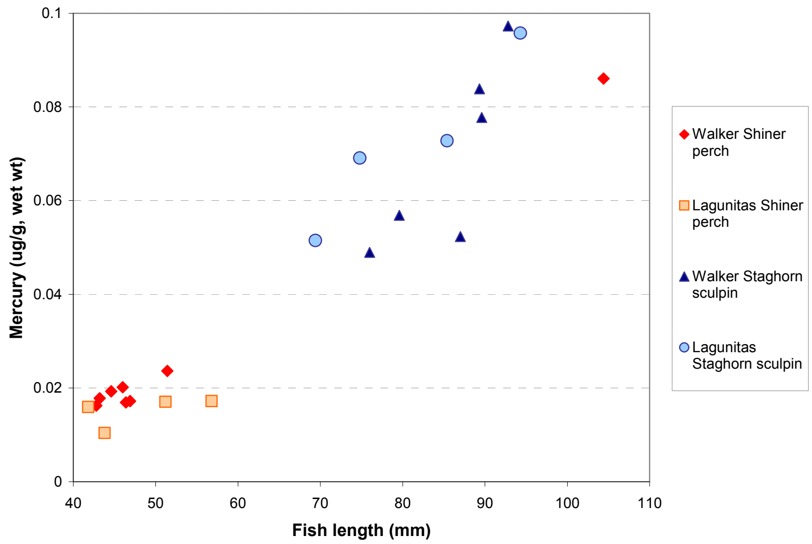


Figure 3-4. Prey fish methylmercury and length (2009)

This figure shows there is no difference among sites in prey fish methylmercury and length. The comparison is of fish collected from two ecologically similar locations (deltas) with very different sediment mercury concentrations.

Note: most of the collected Shiner perch were smaller than target size. The fish were collected with an otter trawl at the Bay edge. (Nearly all mercury in prey fish is methylmercury, see Section 3, *Interpreting target data*.)

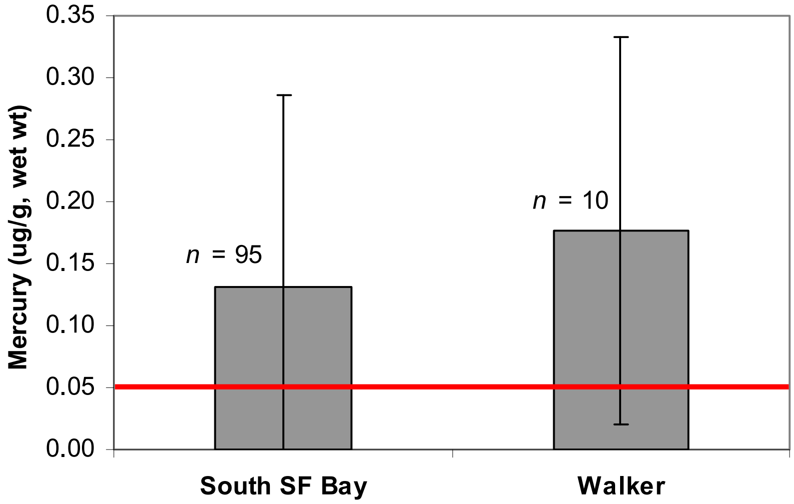


Figure 3-5. Comparison of prey fish methylmercury concentrations in mercury-mine polluted estuarine marshes

Methylmercury in Long-jawed mudsuckers from Walker Creek Delta is higher than in mudsuckers from South San Francisco Bay, which is also polluted by a mercury mine. The grey bars show mean methylmercury in target size (5-15 cm) whole fish (µg/g, wet wt); the red horizontal line is the wildlife target; the error bars are one standard deviation. Mudsuckers were collected with minnow traps in marsh channels. (Nearly all mercury in prey fish is methylmercury, see Section 3, *Interpreting target data*.).

Citations: Citations: Walker data: (Ridolfi 2010 addendum); South San Francisco Bay data: (San Francisco Estuary Institute Regional Monitoring Program data, unpublished)

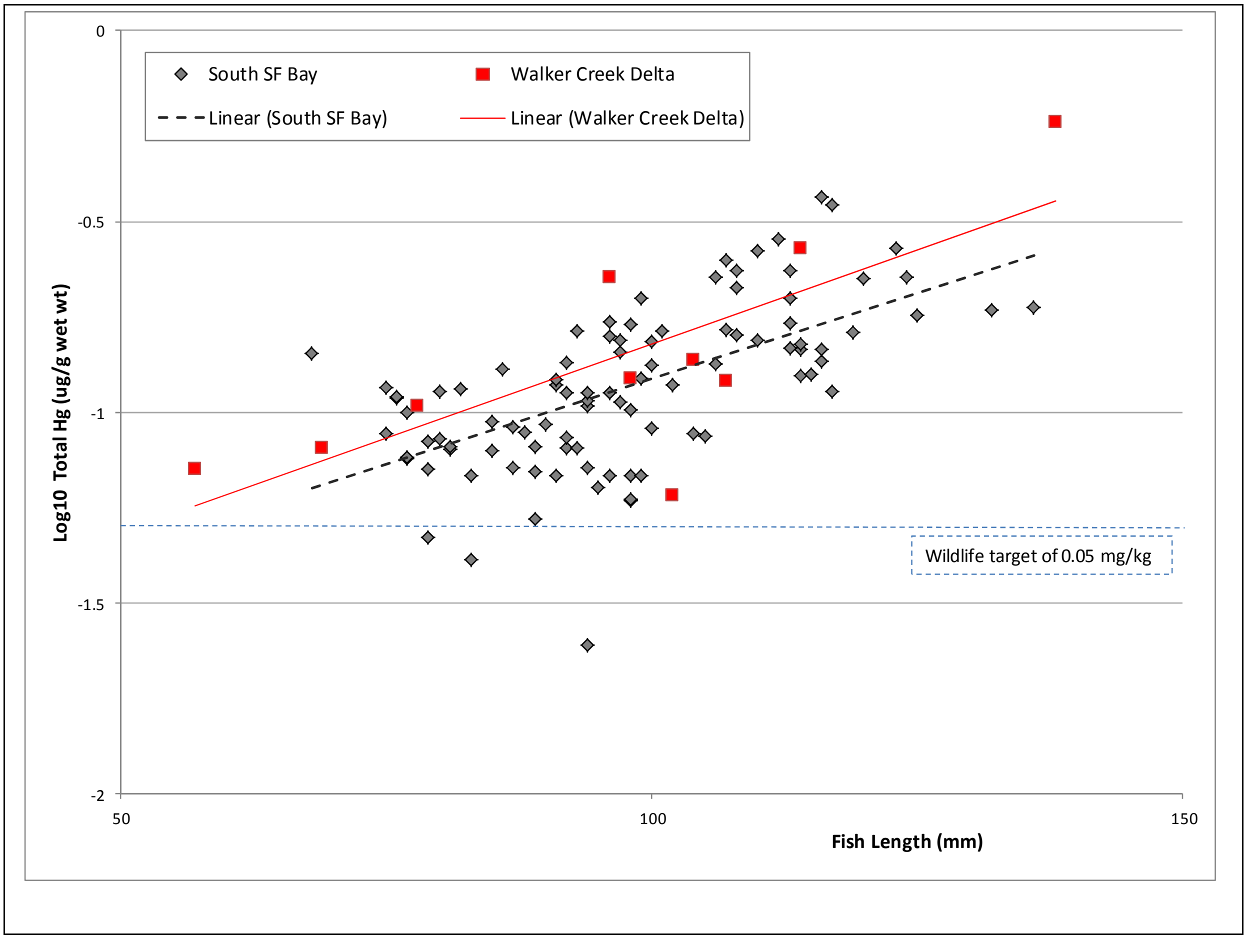


Figure 3-6. Prey fish methylmercury and length in mercury-mine polluted estuarine marshes

This figure illustrates higher prey fish methylmercury concentrations, accounting for fish length, in Long-jawed mudsuckers from Walker Creek Delta compared to South San Francisco Bay, which is also polluted by a mercury mine. Statistically significant linear regressions on Log (base 10) of fish methylmercury concentration is plotted against fish length; statistically significant at $p<0.001$ for San Francisco and $p<0.009$ for Walker, and r-squared values of 0.37 and 0.54, respectively; residual satisfy requirement of .

Citations: Walker data: (Ridolfi 2010 addendum); South San Francisco Bay data: (San Francisco Estuary Institute Regional Monitoring Program data, unpublished)

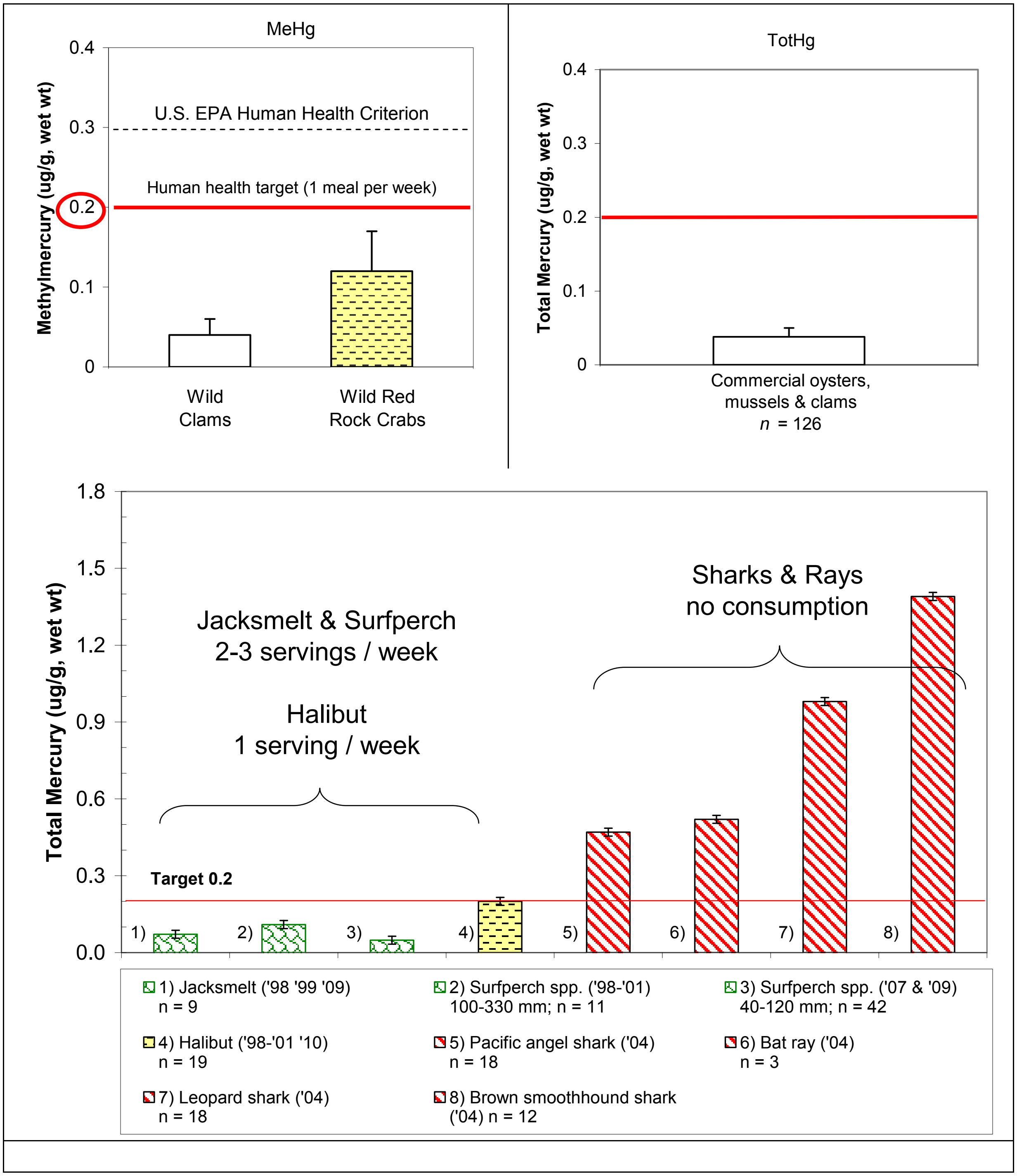


Figure 4-1. Shell and sport fish methylmercury concentrations and human health risk

Green, yellow, and red indicate low, medium and high risk (respectively) to human health. The color scheme is from the OEHHA fish consumption advisory. There are no consumption limits for wild clams or commercial shell fish. The upper charts show shell fish and the lower charts show sport fish mercury and methylmercury concentrations. The most popularly consumed shell fish (wild clams) and sport fish (halibut) are in the green and yellow categories.
(The human health target is for methylmercury, and nearly all mercury in edible portions of sport fish is methylmercury, see Section 3, *Interpreting target data*.)

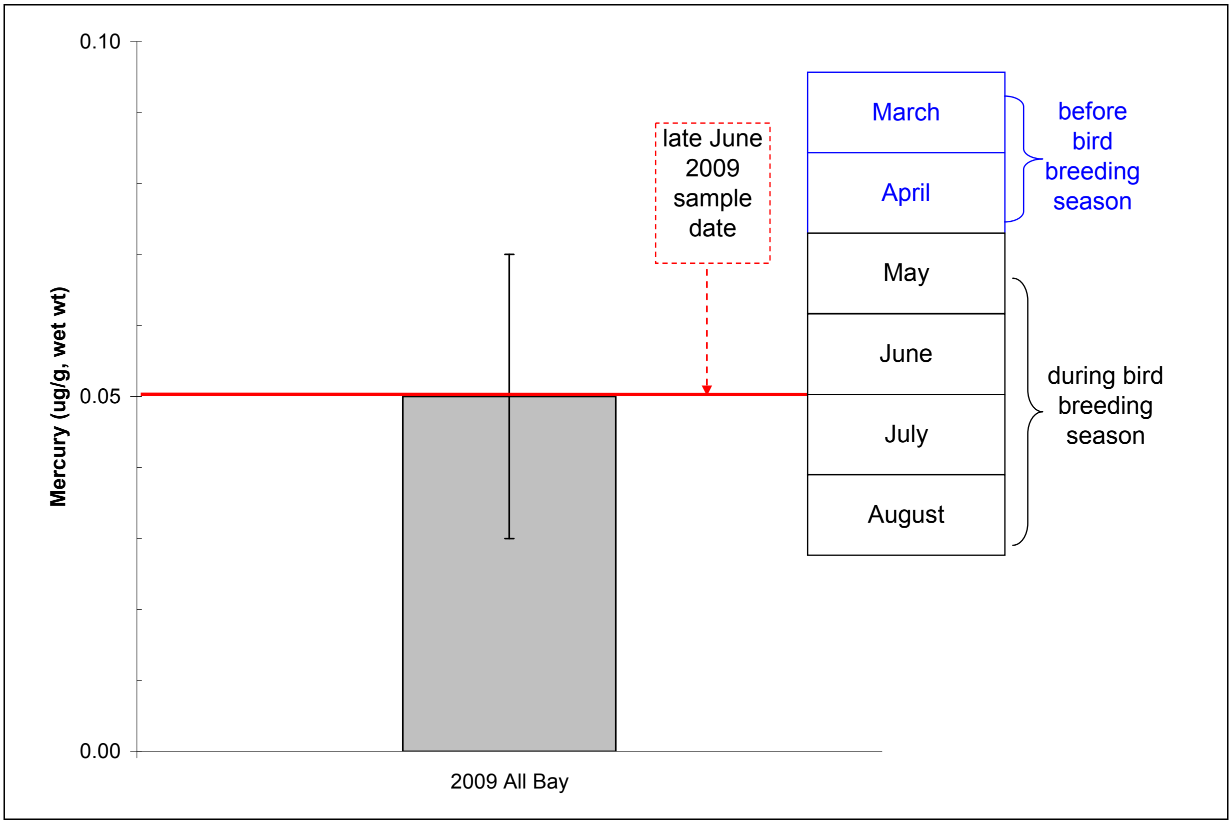
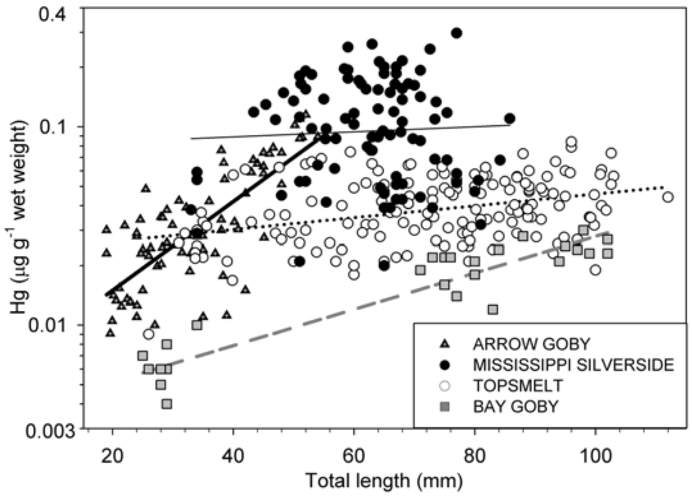


Figure 4-2. Methylmercury in prey fish (Tomales Bay, 2009)

The red line at 0.05 µg/g indicates the target concentration of methylmercury in prey fish. The grey bar is the measured Bay-wide mean, and error bars show the standard deviation. Bay-wide mean methylmercury in prey fish was 0.5 mg/kg in late June 2009, during the bird breeding season, equal to the target. Birds are most sensitive to methylmercury before and during their breeding season. (The wildlife target is for methylmercury, and nearly all mercury in prey fish is methylmercury, see Section 3, *Interpreting target data*.)



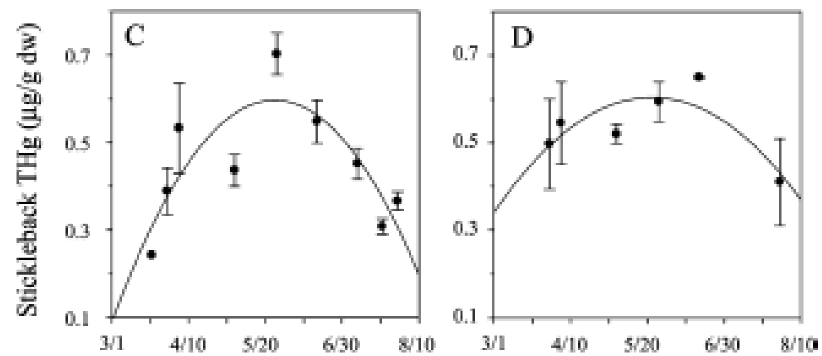


Figure 4-3. Methylmercury in San Francisco Bay prey fish

Left: methylmercury increases with prey fish length

(Both: Nearly all mercury in prey fish is methylmercury, see Section 3, *Interpreting target data*.)

Citation: (Greenfield 2010, Figure 2)

Right: short-term variation in methylmercury concentrations from March to August

Citation: (Eagles-Smith 2009, Figure 1c & 1d)

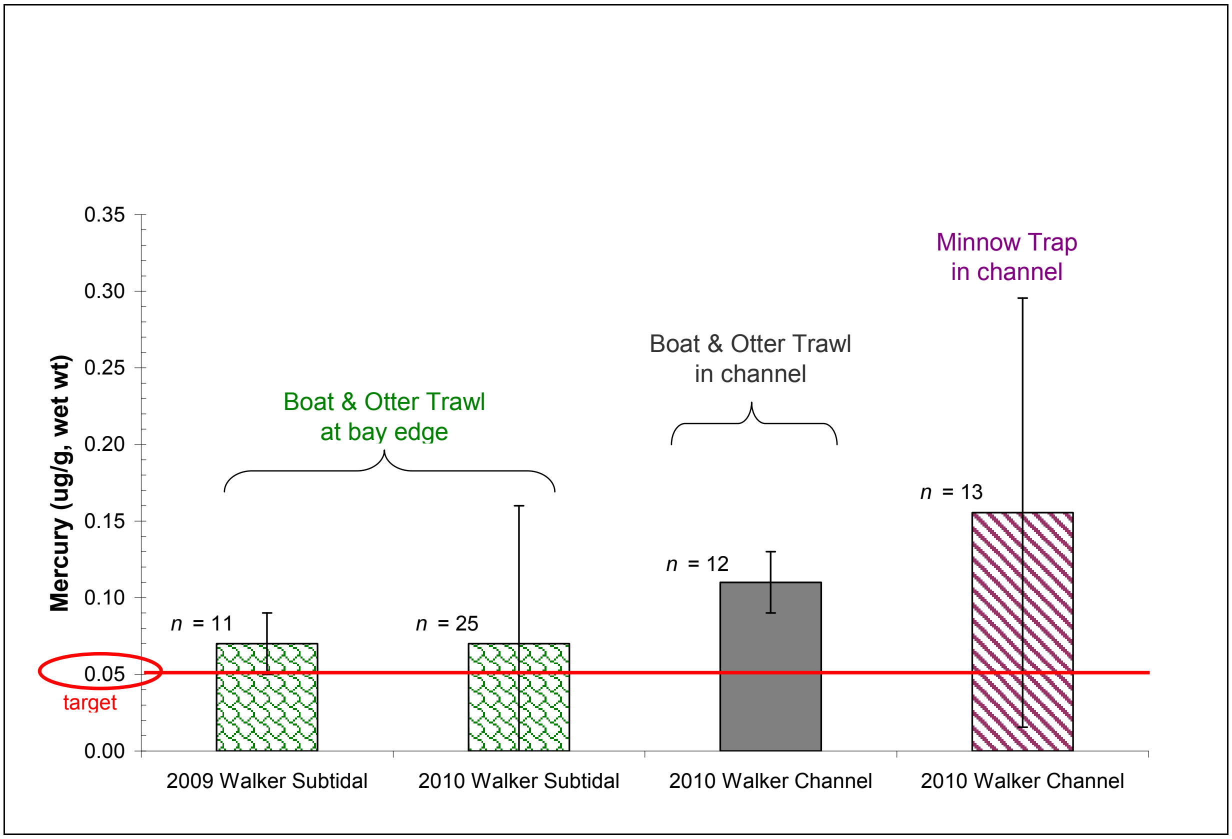


Figure 4-4. Methylmercury in prey fish at the Walker Creek Delta in 2009 & 2010

Methylmercury concentrations are lower in prey fish from the Bay edge (two left bars) than in prey resident in the vegetated marsh at the Walker Creek Delta (two right bars). The bars represent the mean concentration; error bars represent one standard deviation. Methylmercury concentrations were similar in 2009 and 2010 in prey fish from the Bay edge (two left bars), although the variance was much higher in 2010. Prey fish methylmercury concentrations increase farther into marsh habitat (two right bars). (Nearly all mercury in prey fish is methylmercury, see Section 3, *Interpreting target data*.)

A: The “before” (red) arrow indicates large amounts of mercury used to be discharged from the Gambonini Mercury. The “after” (blue) arrow indicates that since the Mine was cleaned up in 2000, much less mercury—90% less—is being discharged.

B: This satellite photo shows where mercury accumulates downstream of the Mine, at the Walker Creek Delta (red). We predict mercury concentrations at this Delta will continue to decline to background levels.

C: Hydrodynamics confine mercury from the Mine to the Walker Creek Delta, with eventual discharge to the Pacific Ocean. Nearly all of Walker Creek flows out the mouth of Tomales Bay into the Pacific Ocean. Very little of the Creek flow makes it up the Bay past Hog Island (yellow), where the Bay is very calm, and where differences in temperature and salinity prevent mixing with the Creek water.

D: Less mercury in the sediment at Walker Creek Delta means less of a mercury problem for wildlife and humans.

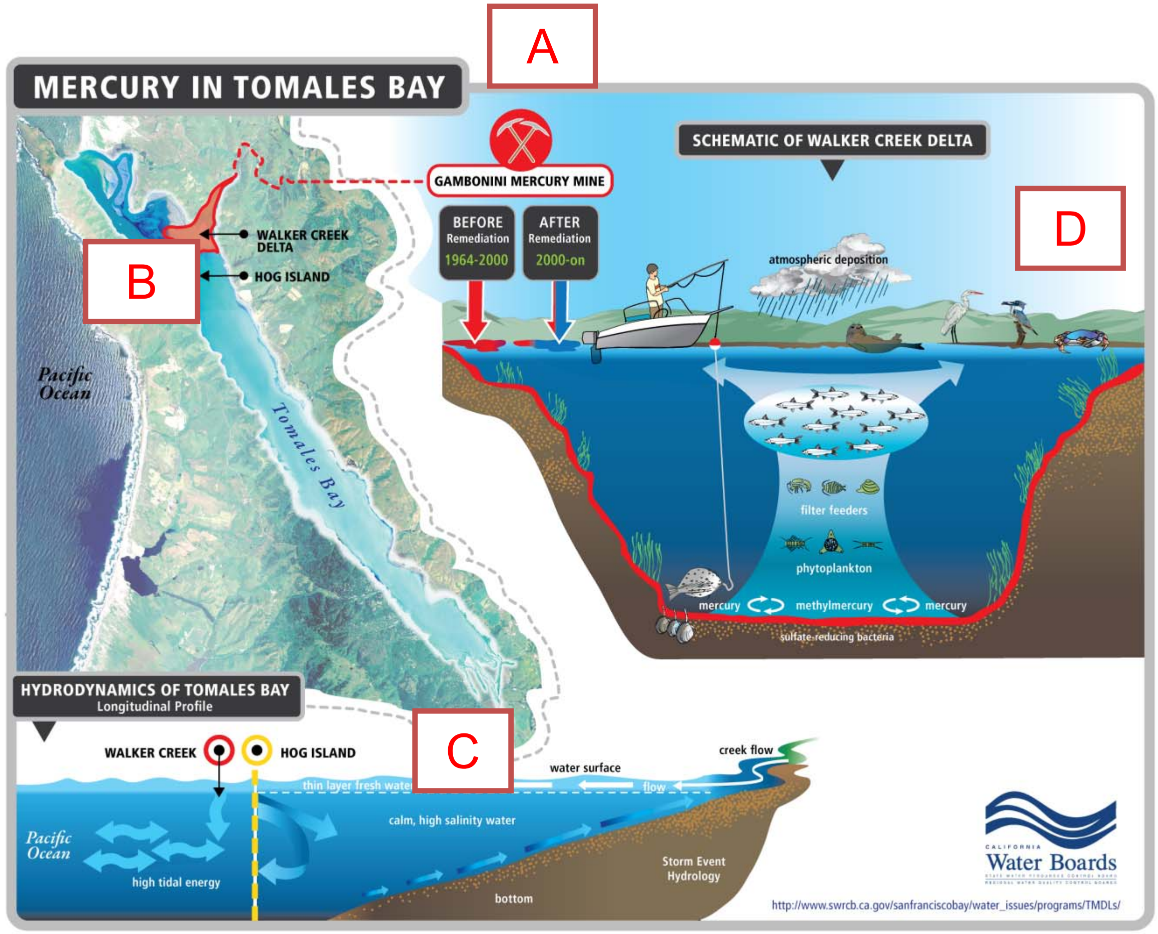


Figure 6-1. Mercury behavior in Tomales Bay

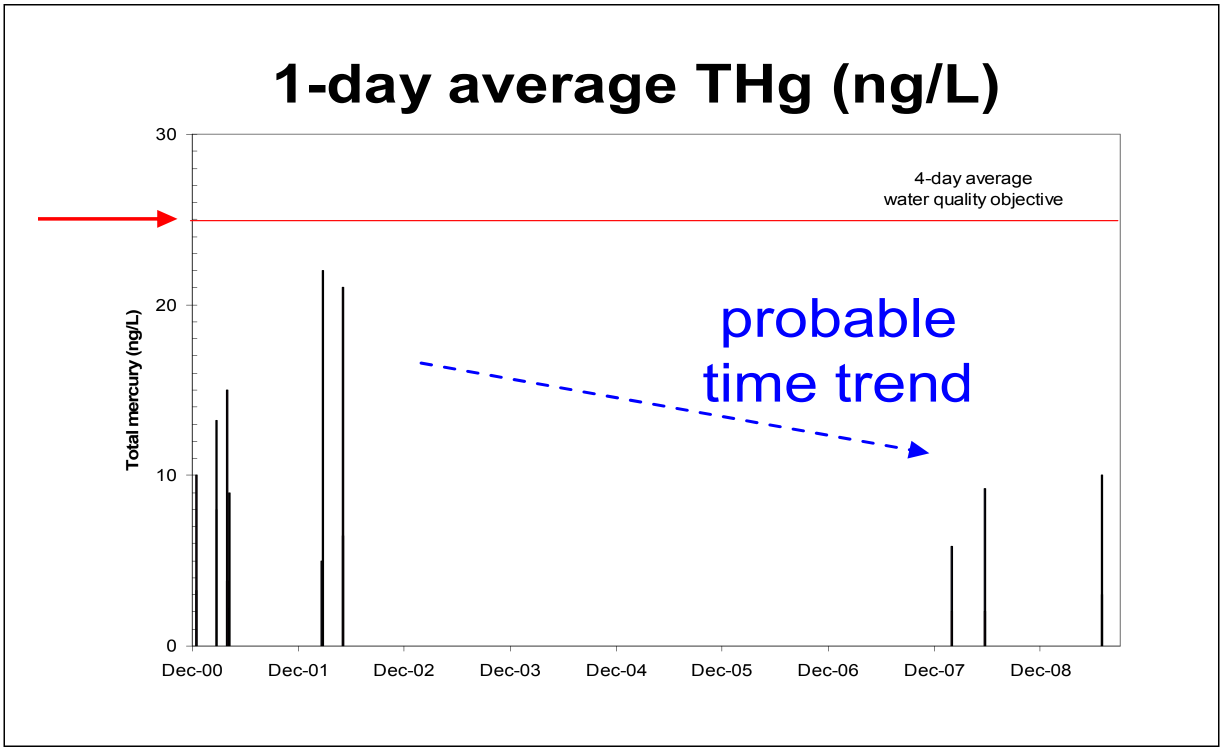


Figure 6-2. Total mercury in water

The graph shows one-day average total mercury concentrations in water samples collected from Tomales Bay. The samples did not exceed water quality objectives, which are: 25 ng/L, 4-day average; 51 ng/L, 30-day avg.; and 2,100 ng/L, 1-hour avg. The red line shows the 25 ng/L, 4-day average objective. Total mercury concentrations appear to have decreased from 2000 to 2009, corresponding to the Mine cleanup. Nine sampling events were completed during 2000–2001, 2007, and 2009. Each sampled 2 or more locations in the Bay, with 2 to 22 samples per sampling event.

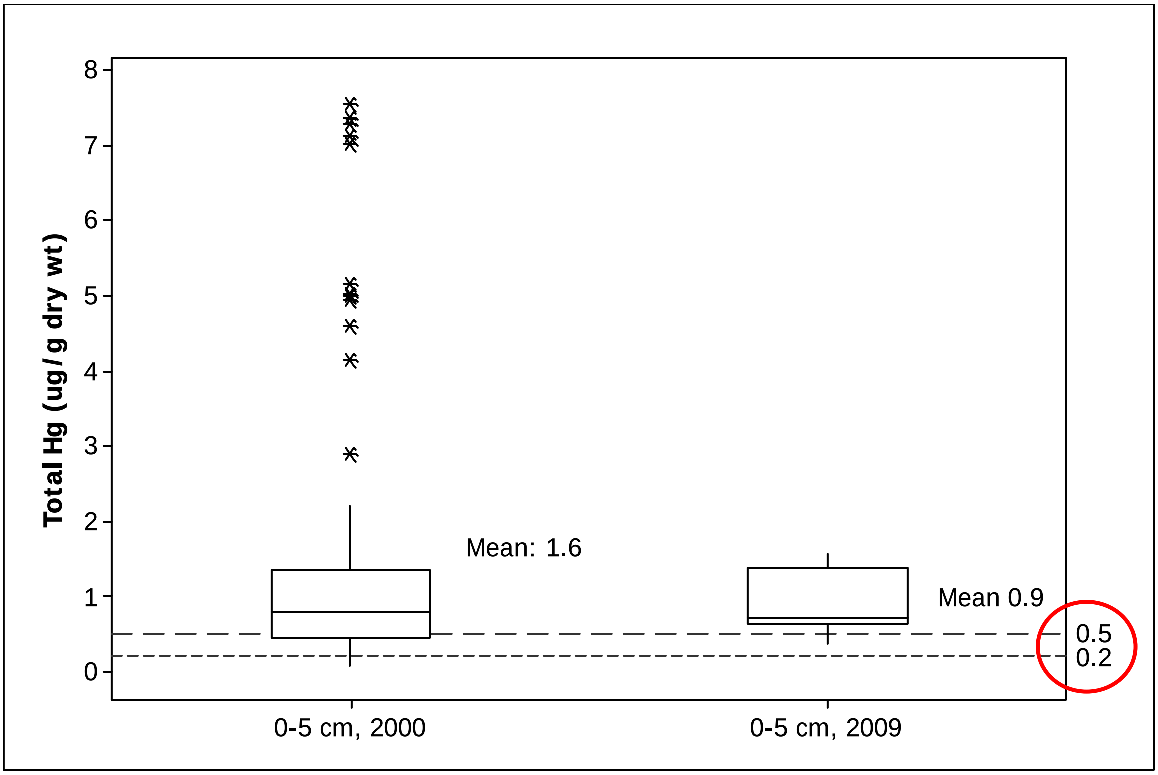


Figure 6-3. Time trend in sediment total mercury concentrations at Walker Creek Delta

Surface sediment total mercury concentrations decreased in the decade since the Gambonini Mercury Mine was cleaned up. The plot shows surface sediment total mercury concentrations, parts per million, in 2000 (left) and 2009 (right). The box represents the 25th – 75th percentiles, the midline is the median, whiskers extend through the full data set, and asterisks indicate data points above the 90th percentile. Additionally, the average (mean) is displayed outside the box.

The dashed line at 0.5 mg/kg indicates TMDL for Walker Creek, and allocation assigned to areas downstream of the Mine, by the Walker Creek Watershed mercury TMDL. The dashed line at 0.2 mg/kg indicates natural background mercury concentrations, and allocation assigned to areas of Walker Creek not influenced by the Mine, assigned by the Walker Creek Watershed mercury TMDL.

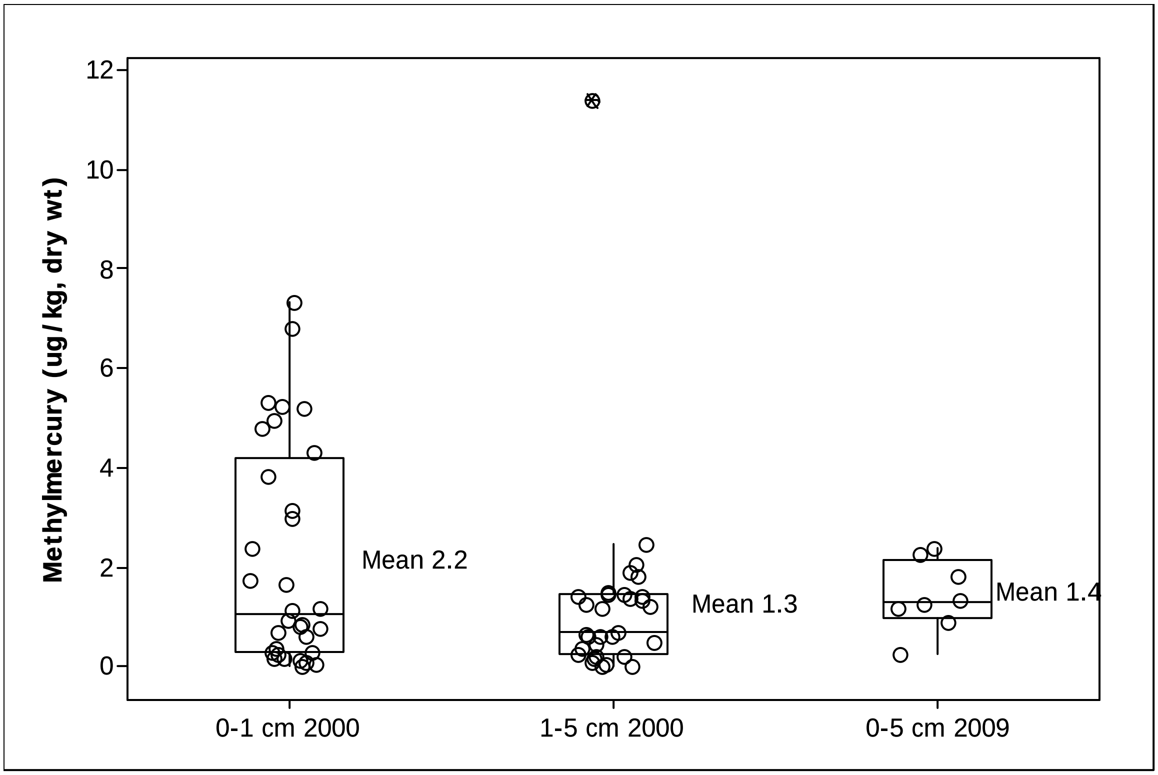


Figure 6-4. Sediment methylmercury concentrations at Walker Creek Delta

Surface sediment methylmercury concentrations in the decade since the Gambonini Mercury Mine was cleaned up. The plot shows surface sediment methylmercury concentrations, parts per *billion*, in 2000 (left and center) and 2009 (right). The box represents the 25th – 75th percentiles, the midline is the median, whiskers extend through the full data set, and asterisks indicate data points above the 90th percentile. Additionally, the average (mean) is displayed outside the box.

Total mercury concentrations decreased in the decade since the Gambonini Mercury Mine was cleaned up (see previous Figure 6-3). However, sediment methylmercury concentrations do not show the same trend (this Figure 6-4). Sediment methylmercury concentrations typically vary by depth. For example, in 2000, methylmercury in individual samples from 0–1 cm depth was higher than that in those from 1–5 cm depth at the Walker Creek Delta. However, the 2009 sediment sampling protocols were very different (4-point composites from 0–5 cm depth), as compared to discrete cores collected in 2000. These changes in sampling protocol preclude detection of a time trend.

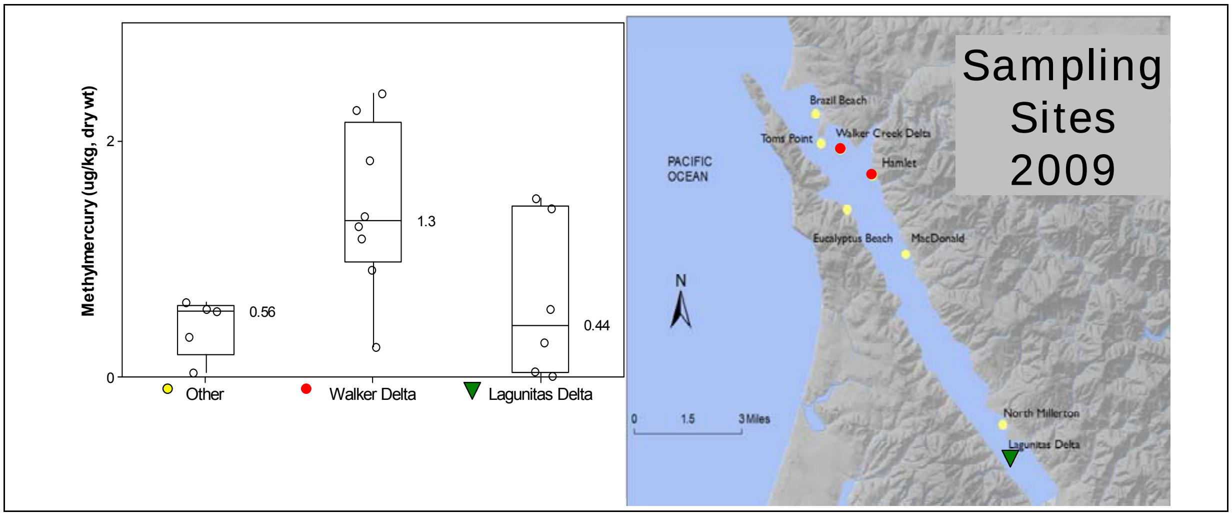


Figure 6-5. Methylmercury in surface and subtidal sediment (2009)

Methylmercury concentrations in 2009 were higher at the Walker Creek Delta than at the Lagunitas Creek Delta (combined mudflat and marsh samples; statistically significant at p = 0.0688). Each data point is indicated by an open circle, and is the mean of two 4-point field composite samples. The box represents the 25th –75th percentiles, the midline is the median (value is displayed), and whiskers extend through the full data set.

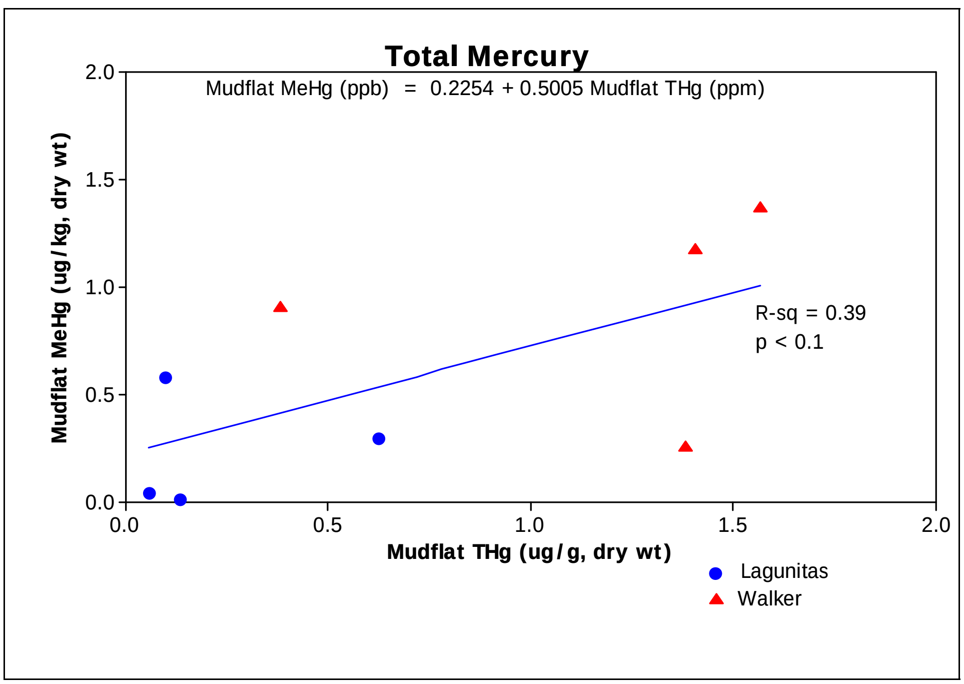


Figure 6-6. Total mercury contributes to methylmercury

This linear regression illustrates that more methylmercury was produced in Tomales Bay mudflats where more inorganic mercury was present. However, methylating conditions are also important for production of methylmercury.

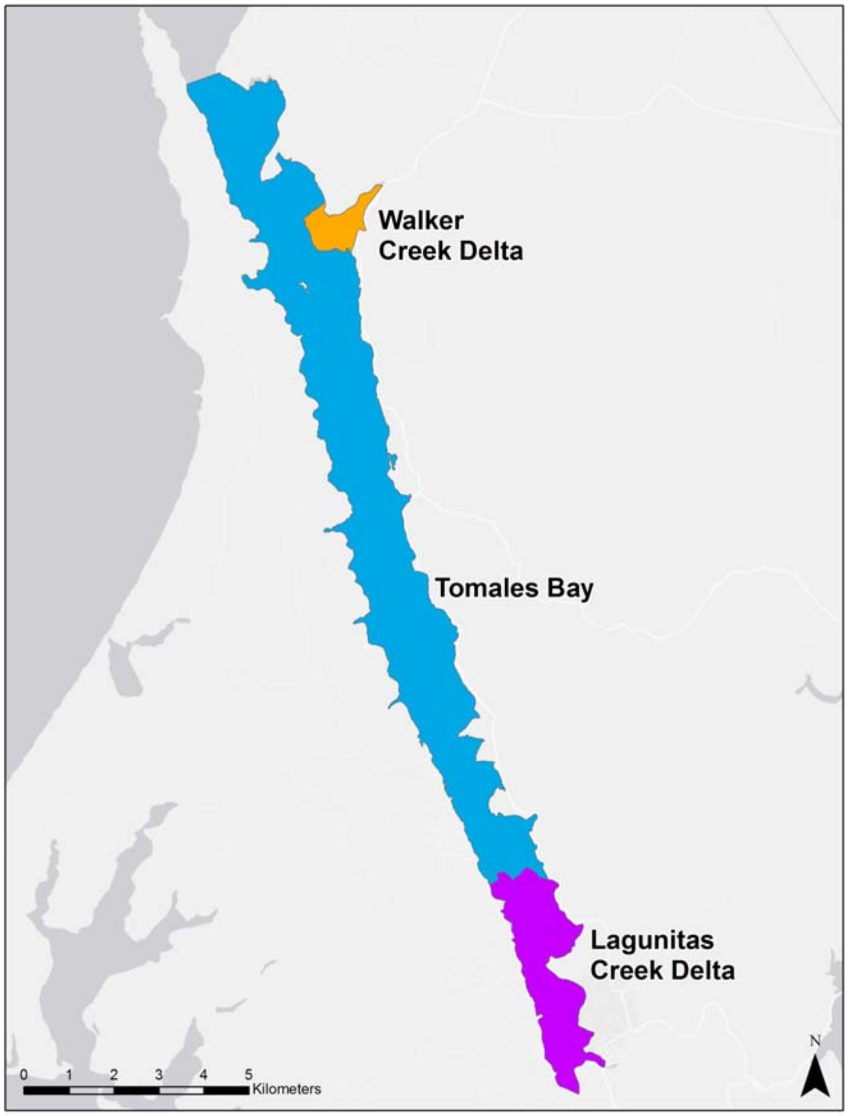


Figure 7-1. Percent Area of Tomales Bay

Area	Allocation (mg/kg)	% Area of Bay
Walker Creek Delta	0.5	3.4%
Lagunitas Creek Delta	0.2	15.4%
Other	0.2	81.2%

The area-weighted average mercury concentration will be 0.2 mg/kg, rounded to 1 significant figure, when the allocation is met at Walker Creek Delta (the allocations are already met at the Lagunitas Creek Delta and other areas in Tomales Bay). The area-weighted average mercury concentration is calculated by taking the sum of each allocation multiplied by its respective area of the Bay, and dividing the sum by the total area of the Bay.

Table 2-1 Beneficial uses in Tomales Bay

Beneficial uses	**Existing**	**Impaired by Mercury**
Commercial and sport fishing (COMM)	X	X
Estuarine habitat (EST)	X	X
Marine habitat (MAR)	X	X
Fish migration (MIGR)	X	
Navigation (NAV)	X	
Preservation of rare and endangered species (RARE)	X	
Contact water recreation (REC1)	X	
Noncontact water recreation (REC2)	X	
Shellfish harvesting (SHELL)	X	
Fish spawning (SPWN)	X	
Wildlife habitat (WILD)	X	X

Table 5-1 Mercury Loads

Source	**Estimated Annual Mercury Load (kg)**	**Comments**
Mining		
1964 – 2000	32–135	Range of peak loads for wet years, from El Niño to 100-year event, prior to mine site cleanup
Current loads (2000 – future)	0.26–19	Range of loads after mine site cleanup for average to wet (El Niño) years; mine cleanup resulted in >90% mercury load reduction
Watershed contribution (non-mining)	21	This source consists of both naturally-occurring mercury in local geology, and mercury from global atmospheric deposition
Atmospheric deposition (global)	0.4–0.7	Direct deposition onto surface of Tomales Bay
TOTAL (SINCE 2000)	**22-41**	

Note: citations and calculations are provided in Section 5 of the Staff Report.