Mercury in the San Francisco Estuary

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I Introduction

A Purpose and Scope

Ever since the recognition of mercury as an environmental problem, San Francisco Estuary has been an active area of mercury research. It is little wonder that this is so: the estuary is in the middle of a region of mercury mineralization and historic mercury mining, and it is downstream of an area of historic gold mining where millions

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of kilograms of mercury were used. It is also a heavily urbanized area that once featured chloralkali facilities and numerous shipyards potentially contaminated with mercury-based paints. In addition, it is a drainage area for rich agricultural regions that may have seen substantial environmental applications of mercury insecticides and fungicides. In this review, we present a survey of literature on mercury contamination and biogeochemistry focusing on San Francisco Estuary. Our intent is to stimulate scientific questions addressing mercury contamination in this and other estuarine systems, as well as to describe the restoration and management efforts that accompany mercury-contaminated sites.

B Overview of the Problem

Before presenting work specific to mercury contamination in San Francisco Estuary, an overview of the environmental mercury problem and mercury chemistry is appropriate. There are many valuable reviews on this wider topic, with focuses on toxicological (Clarkson and Magos 2006), biogeochemical (Benoit et al. 2003; Fitzgerald and Lamborg 2003; Fitzgerald et al. 2007; Ravichandran 2004; Ullrich et al. 2001), ecological (Wiener et al. 2003), and microbiological aspects (Barkay et al. 2003).

Mercury is an environmental and human health concern largely because of the formation of methylmercury, particularly monomethylmercury (MMHg), which is bioaccumulated and biomagnified to toxic concentrations in higher trophic level organisms, including birds (Schwarzbach et al. 2006) and mammals (Wiener et al. 2003). It is a neurotoxin for humans, and effects have been noted in populations consuming fish (Clarkson and Magos 2006). In estuarine systems, sediments are a primary area of MMHg production (Mason et al. 2006). Sulfate-reducing bacteria are thought to be the principal methylators of mercury in anoxic estuarine sediment (Compeau and Bartha 1985), although iron-reducing bacteria have recently also begun receiving scrutiny (Kerin et al. 2006). The production of MMHg is, therefore, controlled by factors influencing the distribution of mercury between abiotic and biotic compartments, such as sulfur chemistry and organic matter, and by factors that control microbial activity, such as temperature and the availability of suitable organic matter for cellular respiration (Gilmour and Henry 1991; Hammerschmidt and Fitzgerald 2004; Heyes et al. 2006; King et al. 2001). Methylmercury produced in sediment that is exported to the water column can be bioaccumulated by phytoplankton or other organisms (Pickhardt and Fisher 2007) and biomagnified to higher trophic levels (Lawson and Mason 1998).

C Environmental Setting of San Francisco Estuary

An understanding of the setting of San Francisco Estuary is essential as a backdrop for this review. The monograph *San Francisco Bay: The Urbanized Estuary* is an older, but excellent description (Conomos 1979), as is the more recent *San Francisco*

Bay: The Ecosystem (Hollibaugh 1996). In addition, articles are available on the characteristics and circulation patterns in the estuary (Conomos et al. 1985), temporal fluctuation and time scales of variability of estuarine parameters (Cloern and Nichols 1985; Thomson-Becker and Luoma 1985), and anthropogenic modification of the estuary over time (Nichols et al. 1986). Some recent studies have covered water circulation, salinity, and nutrients (Kimmerer 2002; Monismith et al. 2002; Smith and Hollibaugh 2006); suspended sediment (Ganju et al. 2005; McKee et al. 2006; Ruhl et al. 2001; Schoellhamer 2002); organic carbon (Lesen 2006; Murrell and Hollibaugh 2000; Stepanauskas et al. 2005); marsh formation (Watson 2004); and sedimentation (Foxgrover et al. 2004; Jaffe and Foxgrover 2006; Jaffe et al. 1998).

San Francisco Estuary is a truly unique setting (Fig. 1). It is a natural, semienclosed body of water created by right-lateral movement on the San Andreas fault system (Hedgpeth 1979). It is the largest estuary on the California coast and is heavily urbanized (Nichols et al. 1986). Its circulation is controlled by tidal currents and freshwater flow, which is dominated by the distinctly Mediterranean climate in the region—dry summers and wet winters (Kimmerer 2002). San Francisco Estuary can be divided into two geochemically distinct subestuaries, the northern and southern reaches, which join in the Central Bay and connect to the Pacific Ocean via the Golden Gate (Flegal et al. 1991). The system has further been divided into six hydrographically distinct regions: Tributaries, Southern Sloughs, South Bay, Central Bay, Northern Estuary, including San Pablo Bay and Suisun Bay, and River-Delta (Conaway et al. 2007). Ninety percent of the annual freshwater inflow to the estuary enters via the northern reach through the delta formed by the convergence of the Sacramento-San Joaquin drainage basins, which includes most of the Coast Ranges, the Central Valley of California, and the western Sierra Nevada (Conomos et al. 1985). The Napa and Petaluma Rivers, which also drain to the northern reach, provide local drainage from the Coast Ranges, but their discharges are relatively small in comparison. In contrast, the southern reach receives only a small amount of freshwater input (<10% of the total freshwater input to the estuary), mostly from the Guadalupe River, Coyote Creek, and other small tributaries that locally drain the Coast Ranges and the Santa Clara Valley. Onto this physically and chemically complex system is superimposed an ecologically and biogeochemically complex mercury contamination issue, which has been the focus of many studies reviewed here.

II Issues Related to Mercury Contamination in San Francisco Estuary

Concerns about mercury in San Francisco Estuary center on human health and ecological effects on birds. The San Francisco Bay Regional Water Quality Control Board (SFRWQCB), which is tasked with the preservation of beneficial uses of the estuary, has determined that the estuary is impaired for mercury, in part because of the reported concentrations of mercury in fish tissue and bird eggs (SFRWQCB 2006). Studies on fish and ecotoxicological effects on birds both support this regulatory statement and highlight concerns of mercury toxicity.

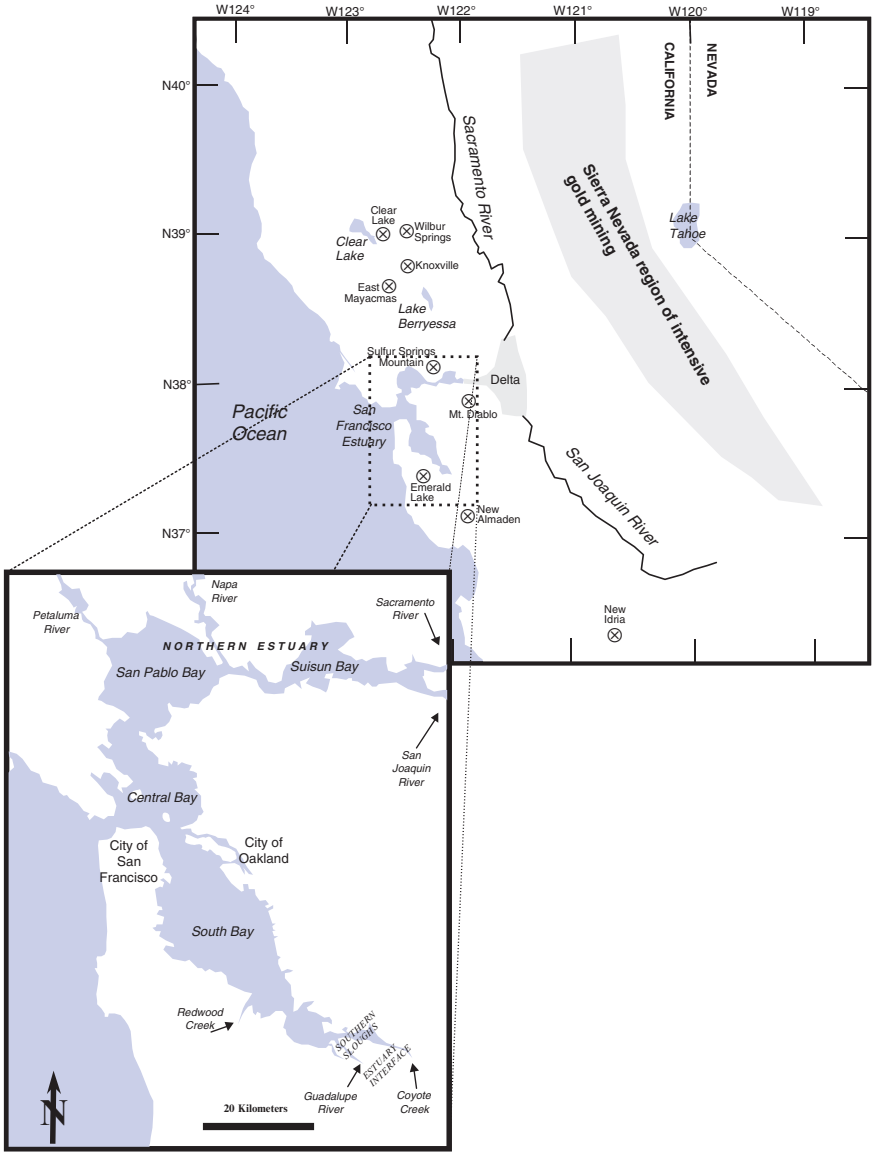

Fig. 1 Regional map of San Francisco Estuary, California, with inset detail. Regional map shows area where intensive gold mining in the foothills of the Sierra Nevada occurred. Locations of large mercury mining districts in the San Francisco Estuary watershed are shown with an "⊗" symbol. Inset shows the estuary and its larger tributaries. Distinct hydrographic regions are Rivers (the confluence of the Sacramento and San Joaquin), Northern Estuary, Central Bay, South Bay, the Southern Sloughs, and Estuary Interface

A Human Health

Consumption of mercury-contaminated fish from the estuary is the issue most relevant to human health. Accordingly, recent studies of mercury in fish in San Francisco Estuary (Davis et al. 2002; Fairey et al. 1997; Greenfield et al. 2005) have focused on concentrations and spatial and temporal trends in those concentrations in various fish species. The range of concentrations in several species are summarized in Table 1. Fish mercury concentrations can exceed regulatory standards in leopard shark, striped

Table 1 Survey of total mercury (Hg_T) and methylmercury (MeHg) concentrations (μg g^{-1}) in organisms from San Francisco Estuary

Species	Tissue	Hg_T (μg g^{-1}), range (mean), wet weight	MeHg, (μg g^{-1}), wet weight	Notes
Fish[a,b]				
California halibut (*Paralichthys californicus*)	Muscle	0.20–0.36		
Jacksmelt (*Atherinopsis californiensis*)	Muscle	0.068–0.17 (0.09)		
Leopard shark (*Triiakis semifasciata*)	Muscle	0.28–1.3		
Shiner surfperch (*Cymmatogaster aggregata*)	Muscle	0.068–0.42		
Striped bass (*Morone saxatilis*)	Muscle	0.15–0.55		
Sturgeon (*Acipenser transmontanus*)	Muscle	0.25–0.30		
White croaker (*Genyonemus lineatus*)	Muscle	0.069–0.41		
Birds				
California clapper rails *Rallus longirostris obsoletus*)[c]	Egg[d]	0.11–2.5		MeHg averaged 95% of total in subset analyzed
Canvasbacks (*Aythya valisineria*)[e]	Liver	ND–9.4[f]		
Greater scaup (*Aythya marila*)	Liver	1.8–20[f]		
Lesser scaup (*Aythya affinis*)	Liver	1.1–9.9[f]		
Surf scoters (*Melanitta perspicillata*)	Liver	5–21[f]		
Ruddy ducks (*Oxyura jamaicensis*)	Liver	2–7[f]		

(continued)

Table 1 (continued)

Species	Tissue	Hg_T (μg g^{-1}), range (mean), wet weight	MeHg, (μg g^{-1}), wet weight	Notes
Mammals				
Pacific harbor seal (*Phoca vitulina richardii*)[g]	Blood Hair Liver	0.015–1.4 0.41–93 0.15–160	0.068–2.9	Pups, juveniles, and adults from Central and Northern California, 2003–2005. Concentrations are typically highest in adults.
House mouse (*Mus musculus*)[h]	Liver	0.02–4.0		Small mammals collected from tidal salt marsh habitat dominated by pickleweed (*Salicornia virginica*).
Deer mouse (*Peromyscus maniculatus*)	Liver	0.05–1.1		
California vole (*Microtus caliJbrnicus*)	Liver	0.02–0.12		

ND, not detected.
[a] Guideline for human consumption is 0.23 μg g^{-1} (Davis et al., 2002).
[b] Davis et al. 2002; Fairey et al. 1997.
[c] Lonzarich et al. 1992; Schwarzbach et al. 2006.
[d] Lowest observed adverse effect concentrations (LOAEC) is 0.50 μg g^{-1} (Schwarzbach et al. 2006).
[e] Hoffman et al. 1998; Hothem et al. 1998; Takekawa et al. 2002.
[f] Dry weight.
[g] Brookens et al. 2007.
[h] Clark et al. 1992.

bass, and white sturgeon (Davis et al. 2002; Fairey et al. 1997; Greenfield et al. 2005). Concentrations in several species of fish are statistically higher in Oakland Harbor than in other locations in the estuary (Davis et al. 2002; Meador et al. 2005). Despite its drastic reduction in use over the past few decades, mercury concentrations in striped bass in the estuary show no long-term decrease since the 1970s (Greenfield et al. 2005).

To complement these studies on fish mercury concentration, a report on fish consumption in the San Francisco Bay area was conducted by the California Department of Health Services (SFEI 2000). Based on a survey of 1300 San Francisco Bay

anglers representing 150 fishing sites visits, the study found that the five most popular fish eaten by anglers, in order of preference, were striped bass, California halibut, jacksmelt, white sturgeon, and white croaker, and that about 1 in 10 of the anglers ate more than the amount recommended by the California Environmental Agency's Office of Environmental Health Hazard Assessment (~6.2 g sportfish/kg body weight/mon). Similarly, a fish consumption study on low-income persons in the Sacramento-San Joaquin Delta region found that 2% of women exceeded the Office of Environmental Health Hazard Assessment recommendation, and that 29% of women exceeded the FDA/EPA advisory limits (48.6 g/d commercial fish, 24.3 g/d sportfish) via a combination of sport and commercial fish consumption (Silver et al. 2007).

In addition to studies related to fish consumption, there has been one published study (Windham et al. 2006) suggesting a potential association between children with autism spectrum disorders and emission of hazardous air pollutants, including mercury, to ambient air in the San Francisco Bay area. However, this association, based on limited data, should be reexamined and confirmed with a more refined exposure assessment.

B Ecosystem Health

Birds

Studies of mercury in birds in the estuary have focused on measuring concentrations and detecting the ecological impacts of contamination (Hoffman et al. 1998; Hothem et al. 1995, 1998; Hui 1998; Hui et al. 2001; Lonzarich et al. 1992; Ohlendorf et al. 1991; Schwarzbach et al. 2006; Takekawa et al. 2002); these have demonstrated potentially toxic mercury concentrations in waterbirds around the estuary (see Table 1), with diving ducks typically showing the highest level of contamination. The relationship between mercury and selenium has also been investigated because the elements may offset each other's toxicity (Hothem et al. 1998; Hui et al. 2001). Based on elevated concentrations of mercury, other recent studies of birds in the estuary have sought to establish biomarkers of exposure, such as enzyme activity and reproductive success. Ratios of glutathione reductase to oxidized glutathione were used as a bioindicator to discriminate between mercury and selenium effects (Hoffman et al. 1998). Schwarzbach et al. (2006) linked decreased egg hatchability to mercury contamination in the California clapper rail (*Rallus longirostris obsoletus*).

Mammals

There have been a few published studies on mercury concentration and toxicity in mammals. A study of small mammals residing in pickleweed (*Salicornia virginica*)

habitats around the estuary showed relatively low mercury concentrations (<1 μg g^{-1} dry weight tissue) in salt marsh harvest mice (*Reithrodontomys raviventris*), house mice (*Mus musculus*), and deer mice (*Peromyscus maniculatus*), although concentrations did reach 4 μg g^{-1} in house mice at one site; these burdens had no observable effect on health of the mice (Clark et al. 1992). An investigation of mercury and MMHg in blood, hair, and liver of harbor seals (*Phoca vitulina richardii*) from central and northern California, including sites in the San Francisco Estuary area, showed concentrations that were considered toxic in other mammals (Brookens et al. 2007). However, the study did not have sufficient statistical power to resolve effects of sampling location on mercury concentrations (i.e., if seals from the estuary had higher concentrations than seals in adjacent coastal areas).

III Sources and History

The elevated concentrations of mercury in fish and birds in San Francisco Estuary are attributed to the widespread and persistent mercury contamination in the region (Greenfield et al. 2005; Schwarzbach et al. 2006). This contamination comes from both natural sources and anthropogenic activities, with origins ranging from local, to regional, to global scales. In this section, we review these sources and the transport of mercury to the estuary through water, sediment, and the atmosphere.

A Sources and Reservoir Size

Mineral Resources and Extraction

The estuary lies in the heart of the highly mineralized circum-Pacific mercury belt. The regional geology has been described in several sources, most notably the work of Bailey and Everhart (1964) and, more recently, Rytuba (2003). Within the California Coast Range part of this belt, major deposits are typically associated with serpentinites emplaced along fault zones and altered to an assemblage of silicate and carbonate minerals by carbon dioxide-rich hydrothermal fluids (Rytuba 2003). Background concentrations in the region are in the range of 10–100 μg g^{-1} (Conaway et al. 2004; Domagalski 2001; Hornberger et al. 1999; Kerin 2002; Thomas et al. 2002). Emissions of mercury vapor from natural background and mineralized areas in the region may contribute a sizeable amount of mercury to the atmosphere (Coolbaugh et al. 2002; Engle et al. 2006).

There are several large economic mercury deposits in the region, with locations and mining activities described in detail by Holmes (1965) and summarized by Cargill et al. (1980) and Rytuba (2000). The New Almaden mercury mining district, North America's largest, is situated 30 km south of the estuary and drains

through small tributaries into South San Francisco Bay. Other large mercury mining districts—Clear Lake, Knoxville, East Mayacmas, and Wilbur Springs—range up to 100 km north of the estuary and drain eventually to the northern reach (now via the Yolo Bypass). The New Idria district, North America's second largest mercury producer, is more than 100 km south of the estuary in the San Joaquin River watershed. Post-1945 production of mercury at many of these locations was by reworking surface tailings (Holmes 1965). The legacy of these mining activities has been presented in studies addressing mercury speciation, chemical weathering, and erosion (Conaway et al. 2004; Domagalski et al. 2004; Ganguli et al. 2000; Kim et al. 2004; Lowry et al. 2004; Rytuba 2000, 2003; Slowey et al. 2005a; Thomas et al. 2002). It is unclear if contamination from any but the largest of these districts, New Almaden, has a great influence on mercury concentrations in the estuary itself; and despite the size and proximity of New Almaden to the estuary, there is little to suggest that New Almaden-derived contamination is a pervasive and overwhelming mercury source in the estuary compared to industrial activities and gold mining.

Use of Mercury in the Region

The majority of mercury produced in California in the late 19th and early 20th centuries was used in gold mining (James 2005; Nriagu 1994), principally in hydraulic mining and dredging activities in the California Sierra Nevada (Nriagu and Wong 1997). Contamination from this mining activity has occurred principally by hydraulic mining debris transported through the watershed to the estuary (Conaway et al. 2003; Hornberger et al. 1999; Hunerlach et al. 1999; Jaffe et al. 1998; Marvin-DiPasquale and Agee 2003). An estimated 12 million kg mercury was used for gold recovery in California, and 4.5 million kg was lost to the environment in placer mining operations throughout California (Alpers et al. 2005; Churchill 2000). Although the bulk of the hydraulic mining sediment reached the estuary near the turn of the 20th century (Hornberger et al. 1999), studies on upstream geomorphology and geochemistry of hydraulic mining sediment show that this is still a pervasive and actively eroding source of contamination (Hunerlach et al. 1999; James 2005; Savage et al. 2000; Slowey et al. 2005b).

By the mid-20th century, the use of mercury in gold recovery fell drastically, and the major use of mercury became the incorporation into electrical devices and at chloralkali facilities (Nriagu 1987). Environmental uses, such as antifouling paint, pesticides, fungicides, and slimicides for wastewater treatment, also represent the use of tens of thousands of kilograms of mercury per year in the United States: the authors are unaware of specific data for California. Between 1945 and 1970, more than 100,000 kg/yr of mercury was used in agricultural applications in the U.S. (Nriagu 1987), mainly in seed treatment and foliar applications (D'Itri 1972); however, the State of California did not require reporting of pesticide use by type until 1970 (Federighi 2001), making estimates of mercury use in agriculture difficult at best. Other industrial uses and sources are presented in Table 2.

Table 2 Some potential local and regional sources of mercury contamination to the San Francisco Estuary from historical and modern human activity

Industry	Notes
Chloralkali[a]	Chlorine and caustic soda produced by the Castner-Kellner process (mercury cell) at facilities in Oakland, CA (1919–1957) and Pittsburgh, CA (1917–1973). Contamination occurred potentially via wastewater, spills, and air.
Petrochemical[b]	Major center of refineries 1900–present. Mean concentration of mercury in CA crude oil is 0.011 µg g^{-1}, but some reported values exceed 1 µg g^{-1}. Although historically CA was self-reli ant in petroleum supplies, recent years (1996–present) show an increase in foreign imports. Fate of mercury in refining process not well known.
Medical waste incineration[c]	Facility operated in East Oakland 1982–2001. Large source of mercury to atmosphere during that time (800 kg yr^{-1} in 1996).
Wastewater[d]	Mercury used as slimicide in wastewater treatment. Wastewater also contains mercury lost from hospitals, dental offices, and industrial waste. Total amount unknown.
Gold mining[e]	Gold mining began in mid-19th century and continued into the 20th century; 12 M kg mercury used in hydraulic-placer mining, ore-processing at hard rock mines, and drift mines and dredging operations in CA, with 4.5 M kg mercury lost to the environment from hydraulic placer mines, and 1.4 M kg lost at hard rock mines.
Other sources[f]	Mercury released to the atmosphere from cement manufacturing plants, carbon black production facilities, and waste burning. Mercury disposal in landfills of electrical devices (the principal use of mercury in the 20th century), including batteries, switches, and lamps, likely represents a primary repository of mercury in the region.
Antifouling paint[g]	Use of mercury in antifouling paints in the estuary dates at least to early 20th century. Between 1940 and 1970, approximately 0.04 M kg mercury per year used in antifouling paint in the U.S. California-specific data not available.
Agriculture[h]	Seed and foliar application of mercury as pesticides and fungicides. Between 1945 and 1970, 100,000 kg per year of mercury was used in agricultural applications in U.S. Potential contamination from both Central Valley and local agriculture.
Mercury mining districts[i]	Production data presented to show relative activity in districts. Loss to the environment unknown. Potential contamination occurs primarily through the weathering and erosion of tailings and other waste rock.

Mining district name	Mercury produced (M kg)	Local watersheds upstream of estuary
Clear Lake (1870–1957)	4.4	Cache Creek
Wilbur Springs (1862–1961?)	1.9	Cache Creek
Knoxville (1862–1961?)	5.7	Cache Creek
East Mayacmas (1870–1956)	13	James and Putah Creeks, Lake Berryessa
Sulfur Springs (Vallejo) (1852–1943)	0.59	Blue Rock Springs Creek, Rindler Creek
Mt. Diablo (1863–1958)	0.38	Marsh Creek, San Joaquin River

(continued)

Table 2 (continued)

Mining district name	Mercury produced (M kg)	Local watersheds upstream of estuary
Emerald Lake (1955–1958)	0.09	Redwood Creek
New Almaden (1845–1975)	40	Guadalupe River
New Idria (1858–1972)	17	San Carlos Creek, San Joaquin River

[a] USEPA 2007b.
[b] Sheridan 2006; Wilhelm et al. 2007.
[c] CARB 2000.
[d] Nriagu 1987.
[e] Alpers et al. 2005; Churchill 2000; James 2005.
[f] CARB 2000; DTSC 2002; Nriagu 1987.
[g] Nriagu 1987; WHOI 1952.
[h] D'Itri 1972; Nriagu 1987.
[i] Cargill et al. 1980; Domagalski et al. 2004; Holmes 1965.

B Forces That Bring Mercury to the Estuary

Mercury contamination from historic mining and industrial sources reaches San Francisco Estuary via tributaries, wastewater input, and atmospheric deposition. The magnitude of flux from these various pathways was reviewed by MacLeod et al. (2005) and is presented in Table 3. Transport of suspended sediment by the Sacramento-San Joaquin Rivers into the northern reach is the primary input of mercury to the system (Domagalski 1998, 2001; Roth et al. 2001). This suspended sediment is dispersed throughout the estuary (Ruhl et al. 2001), but smaller tributaries, such as those draining the New Almaden Mining District, are important as well (Thomas et al. 2002). Preliminary data on the mercury isotopic composition of sediments, however, are insufficient to resolve different sources (Foucher and Hintelmann 2006). Mass balance calculations indicate that inputs of mercury from wastewater discharge are currently relatively small (Ellgas 2001; Hsu and Sedlak 2003), but may have been higher in the past before the advent of modern mercury removal technology from waste streams or when mercury compounds were used as slimicides in wastewater treatment (Nriagu 1987). Atmospheric deposition occurs through wet and dry deposition as a minor part of total flux to the estuary (Steding and Flegal 2002; Tsai and Hoenicke 2001). Mercury in precipitation in the region is higher than in adjacent coastal areas, which may represent scavenging of labile mercury from the atmosphere from local sources (Conaway et al. 2005; Steding and Flegal 2002). The relative bioavailability of the mercury from all these previously mentioned sources remains an important unknown.

IV Distribution, Speciation, and Transformation

The legacy of more than 150 years of mercury contamination has been distributed throughout the San Francisco Estuary, with current estuary surface sediment mercury concentrations roughly 5–15 times greater than background levels

Table 3 Estimate of recent or present-day fluxes of mercury (kg yr^{-1}) in San Francisco Estuary

	Mercury (kg yr^{-1})	Reference
Sources		
Atmospheric deposition		
Direct wet deposition	4.4–4.8	Steding and Flegal 2002; Tsai and Hoenicke 2001
Direct dry deposition	22.0	
Atmospheric emissions		
Stationary sources	244	CARB 2005
Areawide sources	1074	
On-road mobile	29	
Other mobile	83	
Gasoline combustion	0.7–13	Conaway et al. 2005
Watershed		
Central Valley	440–800	Domagalski 2001; McKee et al. 2005; SFRWQCB 2006; Thomas et al. 2002
Guadalupe River	4–116	
Wastewater	12	Ellgas 2001
Erosion of contaminated benthic sediments	460	SFRWQCB 2006
Stormwater runoff		
Urban	160	SFRWQCB 2006
Nonurban	25	
Sinks		
Ocean export	513	MacLeod et al. 2005
Burial	732	

(Conaway et al. 2004; Hornberger et al. 1999). The physical distribution and chemical speciation of this mercury in part determine the relationship between the sources and present-day human health and ecological effects described above. The distribution of MMHg and total mercury has been generally described by Choe and associates (Choe and Gill 2003; Choe et al. 2003), Heim et al. (2007), and Conaway et al. (2003). Focusing on the northern reach, the studies by Choe are distinguished by their detailed treatment of the surface water, including colloidal fractions, and are complemented by the study of Heim et al. which provides data on mercury speciation in sediment over an annual period. The study by Conaway et al. details total mercury and MMHg distribution in both water and sediment with a multiannual, multiseasonal statistical approach.

A general trend discernible from the data available is that water column concentrations of total mercury are higher in the rivers draining into the estuary than in the estuary itself. The northern reach, with large riverine inputs, has higher dissolved and total mercury concentrations than the southern reach, where so-called freshwater inputs are dominated by wastewater discharges. Total mercury concentrations in surficial sediments, averaging approximately 1 nmol g^{-1} (Choe et al. 2004; Conaway et al. 2003), are similar throughout the estuary as a result of mixing and resuspension, but decrease moving east into the delta (Heim et al. 2007). Water and sediment MMHg levels vary substantially both spatially and temporally, with highest

Table 4 Concentrations in water (ng L^{-1}), sediment (ng g^{-1}), and atmosphere (ng m^{-3}) in six hydrographic regions of San Francisco Estuary

	Water					Sediment		Air
Region	UHg_T (ng L^{-1})	FHg_T (ng L^{-1})	UMMHg (ng L^{-1})	FMMHg (ng L^{-1})	DGM (ng L^{-1})	Hg_T (ng g^{-1})	MMHg (ng g^{-1})	Hg^0 (ng m^{-3})
Rivers/Delta	2–10	0.4–2	0.04–0.3	0.02–0.08	0.04–0.2	20–500	0.02–0.08	
Northern Estuary	0.4–90	0.1–30	0.1	0.008–0.4	0.2–0.5	30–600	0.06–0.4	
Central Bay	0.3–10	0.08–0.6	0.02–0.06	0.02–0.1	0.01–0.04	10–400	0.0–0.7	2
South Bay	0.4–40	0.1–10	0.02–0.2	0.01–0.08	0.02–0.1	100–800	0.08–2	2
Southern Sloughs	6–70	0.1–4	0.08–0.5	0.3	0.1–0.2	70–800	0.2–2	
Estuary Interface	2–70	0.1–30	0.1–0.4	0.2	0.5–2	100–1000	0.6–3	

Total mercury in unfiltered water (UHg_T), total mercury in filtered water (FHg_T), monomethylmercury in unfiltered water (UMMHg), monomethylmercury in filtered water (FMMHg), dissolved gaseous mercury (DGM), total mercury in sediment (Hg_T), monomethylmercury in sediment (MMHg), and mercury vapor in air (Hg^0).

Sources: Data from Choe and Gill (2003), Choe et al. (2003), Conaway et al. (2003), and Conaway (2005).

concentrations found in and near wetlands (Choe et al. 2004; Marvin-DiPasquale et al. 2003) and in the central delta (Heim et al. 2007). Table 4 lists the concentration of mercury species in various matrices around the estuary.

Both Hg(II) and MMHg are highly particle reactive, with partition coefficients (K_d) of $10^{4.0}$–$10^{6.5}$ commonly measured in the estuary (Choe and Gill 2003; Choe et al. 2003; Conaway et al. 2003). As a result, much of the advective transport of mercury into and within the estuary occurs via particulate phases (Choe and Gill 2003; Choe et al. 2003; Conaway et al. 2003; Domagalski 2001; Roth et al. 2001) as mercury-sulfide minerals, adsorbed to particles, or associated with organic matter (Roth et al. 2001; Slowey et al. 2005b).

A Sediment: The Importance of Sediment Processes

A key area in linking mercury sources to ecological effects and human health is its biogeochemistry in estuarine sediments. Using samples from San Francisco Estuary, Olson and Cooper (1974, 1976) were the first to demonstrate that estuarine sediments were an important site for the methylation of Hg(II). Studies of microbial mercury methylation and demethylation potential in sediments from various environments throughout the estuary and delta (Marvin-DiPasquale and Agee 2003; Marvin-DiPasquale et al. 2003; Mehrotra and Sedlak 2005; Topping et al. 2004) have subsequently illustrated the role of wetlands as hotspots of mercury methylation as well as the importance of Hg(II) speciation and bioavailability, microbial

community, and respiration rate in controlling the rates of microbially mediated MMHg production and degradation in sediments of different environments in the estuary and delta.

Although there has been an apparent decrease in total concentrations in surface sediment through time in some parts of the estuary, there has been no observed corresponding decrease of mercury concentrations in sportfish (Conaway et al. 2007). This discrepancy is primarily attributed to MMHg being the dominant form in fish (Kuwabara et al. 2007), and that mercury methylation and uptake is controlled by a complex interaction of various biogeochemical factors. Nevertheless, because of active erosion and redistribution of sediment in the estuary (Cappiella et al. 1999; Foxgrover et al. 2004; Jaffe and Foxgrover 2006), buried sediments with relatively higher mercury concentrations (Conaway et al. 2004; Hornberger et al. 1999; Marvin-DiPasquale and Agee 2003) may be remobilized and increase levels of bioavailable mercury.

B Water: The Importance of Flux and Complexation

The majority of studies on water column mercury have so far focused on the distribution and transport of contaminant mercury to and within the estuary, making regional model and mass balance calculations possible (MacLeod et al. 2005). Water column measurements used to support flux calculations in the estuary have established the importance of investigating concentrations in suspended matter entering the estuary and also the dominance of sediment resuspension on water column mercury concentrations (Conaway et al. 2003; Domagalski 2001; McKee et al. 2006). Evasion of dissolved gas mercury from surface waters appears to be a small flux out of the estuary (MacLeod et al. 2005); however, high-temporal resolution studies with better spatial resolution are still required (Conaway 2005). In addition to flux calculations, studies on the relationship between salinity and freshwater flow on dissolved concentrations in the water column (Choe and Gill 2003; Choe et al. 2003; Conaway et al. 2003) are important in understanding the uptake of mercury by organisms, and water column measurements of mercury and MMHg have been used to show that sediments are an important source to overlying water (Choe et al. 2004; Topping et al. 2004).

Despite the many studies of the biogeochemical cycling of mercury in the estuary, there are few studies to date describing the element's complexation and speciation, which are relevant to its bioavailability. Detailed investigations of this type in the surface waters have been limited to studies of surface water in tributaries and wastewater effluent that showed the presence of strong, or nonlabile, Hg(II)-complexing ligands with conditional stability constants similar to those of reduced sulfur-containing ligands (Black et al. 2007; Hsu and Sedlak 2003). However, further studies of the complexation and chemical speciation of Hg(II) and MMHg in surface waters or pore waters of the estuary, as well as riverine inputs to the estuary, are needed.

V Bioaccumulation and Biomagnification

Although some organisms in San Francisco Estuary have relatively high and potentially toxic concentrations, their routes of exposure are not well known, because few investigations have focused on trophic transfer of mercury in the estuary (e.g., Pickhardt et al. 2006). It is assumed that the predominant form in biota is MMHg, which is readily bioaccumulated at the base of food chains and then biomagnified in higher trophic levels in the estuary, as elsewhere (Wiener et al. 2003).

A Bioaccumulation

Measurements of mercury uptake by phytoplankton demonstrate the importance of mercury speciation in the estuary. Luengen (2007) reported a biodepletion of dissolved MMHg, but not the inorganic form, during a phytoplankton bloom in the southern reach of the estuary. This apparently selective uptake or scavenging of MMHg is consistent with mesocosm and lake studies showing an inverse correlation between algal abundance and MMHg concentrations in zooplankton and fish (Chen and Folt 2005; Pickhardt et al. 2002). Subsequent bloom decay in the estuary also appears to accelerate the formation of MMHg in suboxic benthic sediments (Luengen 2007).

In a phytoplankton culture experiment using two waters from the freshwater delta region of the estuary containing different concentrations of dissolved organic carbon (DOC), Pickhardt and Fisher (2007) showed greater bioaccumulation of added MMHg, which tended to accumulate in cytoplasm, than of added Hg(II), which accumulated in cell walls and membranes. Additional work using live and heat-killed cells suggested active uptake of MMHg in some phytoplankton. Pickhardt and Fisher also noted that bioaccumulation of MMHg was greater in high-DOC water, citing the possibilities of relatively higher neutral MMHg species in the higher-DOC water, or that the higher-DOC water enhanced phytoplankton membrane permeability to MMHg.

In contrast to this active uptake of MMHg by phytoplankton, some invertebrates in the estuary have been found to show relatively low bioaccumulation. Gunther et al. (1999) measured median accumulation factors of unity (0.9–1.3) for mercury in three species of filter-feeding bivalves (*Mytilus californianus*, *Crassostrea gigas*, and *Corbicula fluminea*) transplanted from relatively pristine sites in central California to contaminated sites within the estuary. This lack of bioaccumulation indicates that much of the mercury at the lower end of food chains in the estuary is not bioavailable (e.g., in inorganic forms) or is relatively dilute because of a bloom dilution effect. Similarly, concentrations measured in the tissues of a resident bivalve, *Macoma petalum*, at a tidal mudflat location in South San Francisco Bay are comparable to local sediment concentrations (Moon et al. 2005), demonstrating a lack of bioaccumulation.

Studies on trophic transfer of mercury and the effects of food web characteristics on its accumulation in organisms from San Francisco Estuary are few. Pickhardt et al.

(2006) contrasted the accumulation of additions of inorganic Hg(II) and MMHg from the dissolved phase and from invertebrate food by mosquitofish (*Gambusia affinis*) and redear sunfish (*Lepomis microlophus*) using water types collected from the delta with different DOC concentrations. Bioaccumulation and retention of MMHg in fish was substantially higher than that of inorganic mercury and differed by both fish species and water type. Based on biokinetic modeling of the observed fish concentrations, Pickhardt et al. concluded that high assimilation efficiencies and slow loss of MMHg from dietary sources are the principal determinants of mercury burdens, but that further research is needed to address the effects of DOC on its bioavailability at lower levels of the food web in aquatic ecosystems.

B Untangling Biomagnification: Food Webs and Environmental Effects

There are marked spatial and temporal variations in mercury concentrations in higher trophic level organisms in San Francisco Estuary, which confounds efforts to resolve processes governing bioaccumulation and biomagnification (Brookens et al. 2007; Greenfield et al. 2005). Interannual variation in sportfish in the estuary has been explained by factors ranging from variation in fish movement patterns, diet, and populations sampled, or, alternatively, variation in freshwater discharge causing increases of bioavailable mercury in the estuary (Greenfield et al. 2005).

Models of food chains need to be delineated within different regions and seasons in the estuary, and these modeling efforts are being complicated by ongoing stressors to the estuary (e.g., invasive species, climate change, water diversions and regulation, and wetland management and restoration). Although algal blooms have now been shown to have a bloom dilution effect on MMHg concentrations at the base of planktonic food chains in the estuary (Luengen 2007), what effect the new phenomenon of increasing phytoplankton blooms in the estuary (Cloern et al. 2006) will have on MMHg cycling in the estuary is unknown. These and other unknowns need to be addressed by first making accurate measurements of concentrations and speciation at different trophic levels and in different food chains within the estuary.

VI Management and Restoration

A History of Mercury Management in the Estuary

The history of mercury management in San Francisco Estuary begins with the widespread recognition of the environmental problem in the 1970s (D'Itri 1972). Measures resulting from litigation in the late 19th and early 20th century, such as the 1884 Sawyer Injunction and the 1893 Caminetti Act, sought to regulate the transport of

hydraulic mining debris and had an impact on the distribution of mercury-contaminated sediment (James 2005), but mercury itself was not specifically considered. In the early 1970s, concentrations in striped bass and catfish in the estuary were found to exceed the federal guidelines, and fish consumption advisories were issued by the State Department of Health (NRC 1978). Between 1970 and 1990, economic pressure and regulatory measures by the U.S. Environmental Protection Agency (EPA), such as the cancellation of product registration for mercury-containing pesticides, led to the rapid decline of its use in the region (Sznopek and Goonan 2000). These regulatory measures and a decline in economic viability resulted in the closure of most of the region's mercury mines in the early to mid-1970s (Cargill et al. 1980). The New Almaden mining district closed in 1975 and is listed on the EPA's Abandoned Mine Lands CERCLIS ("Superfund") inventory (USEPA 2007a).

B Recent Management and Restoration Efforts

The thread of recent management efforts in the estuary begins in 1994, when the State of California's Bay Protection and Toxic Cleanup Program measured mercury concentrations in fish that humans consume from San Francisco Estuary (Fairey et al. 1997). Based on the results, a health advisory for consuming fish from the estuary caused the San Francisco Bay Regional Water Quality Control Board (SFRWQCB) to formally list San Francisco Bay as impaired by mercury (Davis et al. 2002). Since 1999, the SFRWQCB has been developing a Total Maximum Daily Load (TMDL) to determine the load reductions necessary to attain the water quality standard for mercury (Johnson and Looker 2003; SFRWQCB 2006). The development of the TMDL is based on data from the Regional Monitoring Program (RMP) for Water Quality in San Francisco Estuary that identified the magnitude of mercury contamination as well as the temporal and spatial variability of concentrations in water, sediment, and biota (Conaway et al. 2003, 2007; Hoenicke et al. 2003; Thompson et al. 2000). The results of numerous other research projects from San Francisco Estuary, already described, and other mercury-contaminated locations have provided the current basic understanding of the processes affecting mercury biogeochemistry in the estuary.

Published studies on the effects of mitigation or remediation on environmental mercury are notably lacking for San Francisco Estuary. A laboratory study by Mehrotra and Sedlak (2005) used iron additions to decrease mercury methylation in wetland sediments from locations surrounding the estuary. In addition, phytoremediation of mercury-contaminated sediments using water hyacinth (*Eichhornia crassipes*) has been evaluated in the delta region of the estuary (Greenfield et al. 2007; Riddle et al. 2002). Nonetheless, there is a dearth of published papers on mercury experiments using constructed, managed, or remediated wetlands for the estuary. Compounding this lack of information, the scale of design and implementation of wetland restoration activity in San Francisco Estuary is changing from small restoration activities to large, landscape-scale projects, such as the 60 km^2 South Bay Salt Pond Restoration Project (Simenstad et al. 2006).

Although San Francisco Estuary is one of the most studied estuaries in the world (Flegal et al. 2005), large uncertainties still remain regarding the processes and factors controlling mercury methylation and bioaccumulation within the system. One initial step in addressing these uncertainties was the development of a conceptual model of mercury in San Francisco Estuary (Tetra Tech 2006) by the Clean Estuary Partnership, a consortium of stakeholders from industry and municipalities, environmental organizations, resource management agencies, and academic institutions. The conceptual model used existing data to develop an overview of mercury biogeochemistry and also identified the key management questions (Table 5) that must be answered to meet the needs of resource managers and other stakeholders.

Table 5 Management questions, data requirements, and technical approaches identified in conceptual model of mercury

Management question	Key data requirements	Technical approach
What is the relative bioavailability of mercury from different sources to San Francisco Bay?	Chemical and physical form of the mercury from natural and anthropogenic sources to prioritize remediation strategies	• Direct measurements of MeHg • Mesocosm experiments to quantify bioavailability under controlled conditions
At what locations are current methylation rates and methylmercury flux highest?	Characterization of existing methylmercury pool in sediments to determine whether hotspots are present	• Direct measurements of net methylation rates • Simultaneous measurement of factors affecting methylation rate: DO, TOC, SO_4, H_2S, chloride
Can existing wetlands be managed or new wetlands be designed to minimize net methylation rates, or limit exposure to methylmercury that is produced?	Quantification of the response in bioaccumulation to wetland characteristics that can be fully or partially controlled, e.g., nutrient salinity, depth, vegetation levels, type, hydroperiod	• Mesocosm experiments to quantify bioavailability under controlled conditions
Given various scenarios for management actions, when will we likely see improvements in sediment and tissue concentrations?	Characterization of local and bay-wide sediment mercury concentrations in response to localized interventions; quantification of the effects of sediment deposition and erosion on estimates of recovery	• Localized interventions to remove or cap high-mercury sediments • Measurement of responses to localized interventions and mass-loading reductions • Dated, deep-sediment cores to estimate effects of sediment erosion on recovery
How should we best monitor to detect changes in mercury concentrations in sediments and tissue?	Detection of statistically significant changes in reliable indicators	• Measurements of total and methylmercury concentrations in surficial sediments in shallow, depositional areas • Characterization of mercury concentrations in fish indicator species

Source: Tetra Tech (2006).

Some of the management questions identified by the conceptual model have highlighted additional field studies required to fill current gaps in our knowledge and understanding (Table 5). Although extensive monitoring programs in the estuary provide a synoptic view of conditions (Hoenicke et al. 2003; Thompson et al. 2000), detailed information on areas where methylation rates and MMHg flux are highest is not currently available. If these could be identified, then they could either be targeted for restoration directly or managed such that they do not expand. Additionally, this information is needed to guide and evaluate the relative effectiveness of alternative corrective actions. Some of this information is becoming available as several large multi-year field projects are nearing completion in the region (e.g., Marvin-DiPasquale et al. 2005; Schwarzbach et al. 2005; Yee et al. 2005).

The implementation plan for mercury management developed by the SFRWQCB (2006) must also address the anticipated changes in and around the estuary. For example, the planned conversion of salt ponds to wetlands surrounding the estuary will restore unique habitat for biota, particularly for wading birds, and nursery grounds for many species (Goals Project 1999). However, as wetlands are hotspots of mercury methylation, there is a trade-off between the beneficial functions of wetlands and the environmental risk of increased MMHg production and export to the estuary. It is estimated that the response time of concentrations to changes in mercury loading to the estuary is several decades (MacLeod et al. 2005). To determine whether restoration actions over this time frame are achieving any benefits, new and effective monitoring strategies are required to identify locations and processes where changes are anticipated to occur over much shorter time frames.

Summary

This review presents some of the published and other important literature on mercury contamination in San Francisco Estuary. Studies on human consumption of contaminated sportfish and on detecting ecological impacts of this contamination in wetland areas validate concerns regarding mercury's toxicity in this system. Mining, industrial, and environmental uses of mercury have occurred for more than a century, resulting in its large historic and continuing transport to the estuary. Consequently, there is a widespread distribution in the estuary, but more work is needed to show its relative chemical and biological availability from these sources. The uptake of mercury in the estuary has been shown in phytoplankton, but studies on biomagnification in local food webs have yet to draw a clear path to impairment in sportfish and waterbirds. In light of these concerns of impairment and the need for further information, large restoration activities planned for the estuary will require new technical approaches to solve important management questions, such as the location of key areas of methylmercury production.

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