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Fate of Contaminants in Sediment of San Francisco Estuary: A Review of Literature and Data

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FINAL REPORT

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I. Introduction

The long-term fate of persistent contaminants, such as polychlorinated biphenyls (PCBs), mercury (and methylmercury), and organochlorine (OC) pesticides, in San Francisco Estuary is currently of primary concern due to the potential for bioaccumulation in the food web and deleterious effects on wildlife and human health. Although PCBs and OC pesticides have been restricted or banned for decades, they still persist in Bay sediment, as well as in soils that continue to enter San Francisco Bay from surrounding watersheds. Similarly, mercury contamination is widespread in the Bay and its watersheds due mainly to historic mercury and gold mining, although ongoing inputs through global and local atmospheric contamination and other pathways contribute mercury to an extent not seen for the legacy organic contaminants. A map of the San Francisco Estuary is presented on Figure 1.

Human health concerns have been a primary driver of regulatory action for these contaminants. In 1994, a survey of contaminant concentrations in sport fish caught in the Bay found that concentrations of several bioaccumulative contaminants (mercury, PCBs, dioxins, and organochlorine pesticides) were above screening values established to protect human health (Fairey et al., 1997). Consequently, the Office of Environmental Health Hazard Assessment (OEHHA) issued an interim seafood consumption advisory with detailed recommendations for limiting human consumption of Bay fish (OEHHA, 1994). The advisory ultimately led to inclusion of all segments of San Francisco Bay on the Clean Water Act 303 (d) list of impaired water bodies for these contaminants and the development of Total Maximum Daily Loads (TMDLs) for mercury and PCBs in the Bay.

To assist in understanding the long-term fate of persistent contaminants in the Bay, the RMP began developing mass budget models that delineate the processes by which persistent contaminants are introduced, distributed, and removed from the Bay and predict the rates of recovery under various management scenarios. Single-box mass budget models were applied to San Francisco Bay for predicting the fate of PCBs (Davis, 2002), PAHs (Greenfield and Davis, 2002) and OC pesticides (Leatherbarrow et al., 2003). Given that the most persistent of these organic contaminants, PCBs and OC pesticides, are highly associated with sediment and particulate material, Davis (2002) determined that the model was most sensitive to parameters that defined the existing mass of contaminants in sediment: the depth of sediment that actively mixes and exchanges with the water column, and the average concentration of the contaminants assumed to be uniformly distributed throughout sediments in the single box representing the entire Bay. Because of the high spatial variability in the actively mixed sediment layer and in concentrations of these contaminants in sediment, model behavior may be highly dependent on the data set selected as being representative of the Bay. Aside from the limitations inherent in using a simple single-box model, these parameters have not been adequately characterized and, therefore, further evaluation is needed to improve the accuracy and predictive capability of the mass budget models.

In addition to the PCB mass budget model of the Bay, a food web model was developed to predict concentrations in benthic and pelagic invertebrates and fish based on concentrations in water and sediment. Such a model is useful for illustrating the linkage between environmental contaminant concentrations and the ultimate reason of impairment of the Bay (for PCBs), concentrations in wildlife and fish detrimental to those organisms and human consumers. Consequences of management actions on ecosystem contaminant concentrations can thus be ultimately extrapolated to the beneficial uses of interest under different scenarios and assumptions.

Even for contaminants and ecosystems where such mechanistic models have not been developed yet, simple models of the linkage between biological and abiotic ecosystem components such as bioaccumulation factors (BAFs) and biota-sediment accumulation factors (BSAFs) are often used to back-calculate water and sediment target criteria from maximum concentrations in biota protective of wildlife and human health. Such methods may oversimplify the link between environmental contaminant concentrations and biological concentrations and effects, particularly for cases such as mercury where the complexity of mediating ecosystem processes raises the importance of considering other (non-contaminant) parameters for predicting environmental fate. However, simplistic approaches such as BAFs and BSAFs at least provide a starting point to which more complex models can be compared.

In response to the recommendations of a five-year external program review of the San Francisco Estuary Regional Monitoring Program (RMP), a Contaminant Fate Workgroup was developed with the objective of synthesizing existing information from various studies to provide a more comprehensive conceptual picture of contaminant transport and fate within San Francisco Bay. This literature review complements current efforts already underway to understand the dominant mechanisms that determine contaminant transfer processes between sediment and the benthic and pelagic food web in the Bay. The objectives of this literature review are to (1) provide a conceptual view of the long-term fate of particle-associated contaminants and their transfer between sediment and biota; (2) recommend future studies to address current gaps in information and data for San Francisco Bay.

This document provides an overview of the information on San Francisco Estuary and data from other regions addressing issues of sediment contaminants and their potential for impacts on biota that utilize this water body. The distribution of contaminants within estuary sediments, processes by which biota are exposed to these contaminants, and transport processes of contaminants will be discussed.

II. Sediment Contamination in San Francisco Bay

The distribution and fate of persistent contaminants in the Bay are governed by a complex array of physical, chemical and biological processes: hydrology and sediment dynamics, chemical and biological transformations of contaminants, and spatial and temporal (including historical) differences in sources and loading of contaminants to various regions of the Bay. Sources and pathways of contamination that contribute to loading or remobilization of particle-associated contaminants in Bay waters include

runoff from the Central Valley and local watersheds, point-source discharges, atmospheric deposition of particles, dredging, and erosion of historic contaminant deposits. Knowledge of past contaminant deposits in combination with ongoing inputs are needed in order to predict the fate of persistent contaminants of concern in the Bay.

Occurrence and Distribution of Sediment Contamination

Contamination by persistent particle-associated contaminants is widespread throughout the Estuary, but spatially heterogeneous on regional and local scales. Concentrations of PCBs, mercury, and OC pesticides in surface sediment and water samples are typically higher in the southern segments of the Bay compared to the northern reach (SFEI, 2003 "Pulse"). In addition, these contaminants have typically been higher in samples collected near the Bay margins compared to the deeper channels (Flegal et al., 1994; Hunt et al., 1998; SFEI, 2003).

Some of the highest concentrations have been found near the shorelines of urbanized areas in San Leandro Bay, Oakland, Richmond, San Francisco, and in sloughs of the Lower South Bay (see Table 1). Contaminant maps that display maximum concentrations of ΣPCBs, mercury, ΣDDT (Σ o,p'-isomers of DDT, DDE, and DDD), Σchlordanes (Σ alpha-chlordane, gamma-chlordane, cis-nonachlor, trans-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide), and ΣPAHs in sediment have been constructed from the results of several monitoring efforts conducted since 1991 (Figures 2-6). Contaminant concentrations in these studies have varied by 1 to 3 orders of magnitude between the margins and deeper channels for PCBs (<20-9,850 $\mu g\ kg^{-1}$), Hg (<0.5-11 $mg\ kg^{-1}$), DDT (<10-1000 $\mu g\ kg^{-1}$), chlordane (<5-290 $\mu g\ kg^{-1}$), and PAHs (<1000-230,000 $\mu g\ kg^{-1}$). It should be noted that sediment data from San Leandro Bay include sediment cores collected by Daum et al. (2000) at depths of 1, 2, and 3 feet (see Table 1). PCBs, DDT, chlordane, dieldrin, and mercury have also been measured at concentrations that have exceeded screening values in sport fish from near many of these shoreline locations (Fairey et al., 1997; Davis et al., 2002; Greenfield et al., 2002).

In addition to and within the regions noted above, there are more localized cases of contamination in the Bay and surrounding watersheds that may contribute to the cycling of persistent contaminants throughout the Bay. Two locations that are of particular concern and in direct contact with the Bay are currently on the EPA National Priority List (NPL): United Heckathorn in the Richmond Harbor, and Hunters Point Naval Shipyard in southeastern San Francisco (USEPA 2004). United Heckathorn and other companies historically used the site (20-acres, including approximately 15 acres of marine sediment) along the Lauritzen and Parr Canals in the Richmond Harbor to formulate, package, and ship pesticides such as DDT and dieldrin from 1947 to 1966. Despite cleanup efforts that occurred in the early to mid-1990s, concentrations of DDT were still as high as 30,100 $\mu g\ kg^{-1}$ in 1998.

The Hunters Point Naval Shipyard NPL site covers approximately 936 acres (493 on land and 443 in San Francisco Bay) and contains groundwater, sediment, soil, and surface water contaminated by pesticides, metals, and PCBs. Ghosh et al., (2003) measured

concentrations of PCBs in sediment of up to 9,900 ± 900 μg kg^{-1} in January, 2001 with PCB congener distributions similar to Aroclor 1260.

Concentrations of DDT and PCBs measured at these sites were above the high range for most other sites reported in monitoring studies of the Bay (Figures 2-6). Inclusion of these sites in estimating the load of contaminants present in the Bay impacts our evaluation of its current status and provides at least a conservative accounting of its potential impact. However, better characterization and understanding the fate of contaminants from such locations is ultimately essential for assessing their local and regional impacts on water quality in the Bay.

Other areas of localized sources of contamination have been identified through the development of TMDLs for PCBs (Hetzel, 2000) and mercury (Johnson and Looker, 2003) for the Bay. Hetzel (2000) reviewed PCB sediment concentrations reported in specific site investigations and dredged material analyses conducted in and around San Francisco Bay and determined that locations with PCB concentrations greater than 1000 μg kg^{-1} were typically found near the shoreline areas mentioned above. Other locations with elevated PCBs included Richmond Harbor, the Emeryville shoreline, Oakland Inner Channel, San Leandro Bay, Yosemite Creek and South Basin in San Francisco, Seaplane Lagoon on Alameda Island, and scattered locations in or near the South Bay, San Pablo Bay and southeastern San Francisco.

For mercury, one of the primary local sources has been identified as the Guadalupe River watershed, which contains the inoperative mines of New Almaden Mining District (Johnson and Looker, 2003). The mining district, once the largest producer of mercury in North America, has left its signature of contaminated mercury in the streambed of Guadalupe River (Thomas et al., 2002; Leatherbarrow et al., 2002) and in depositional areas of Lower South Bay (Conaway, 2003b). Until contaminant concentrations at these sites and surrounding areas are reduced to background levels, these localized regions of contamination will likely serve as long-term reservoirs of persistent contaminants in sediment that are available for uptake into biota.

Although monitoring has identified areas of concern for specific contaminants in the Bay, few studies have provided the spatial and temporal resolution necessary for conceptual or predictive modeling of how contaminated areas, on local and regional scales, impact the overall water quality of the Bay. Ultimately actions taken at contaminated sites must use site specific knowledge to adequately address their risks.

Recent RMP modeling efforts applied a simple one-box model of the Bay to predict the long-term fate of persistent organic contaminants based on Bay-wide parameterization of important characteristics (Davis, 2002; Greenfield and Davis, 2002; Leatherbarrow et al., 2003). In addition to information specific to highly contaminated sites, another element to improving our understanding of contaminant fate in different regions of the Estuary is to develop a multi-box model consisting of five segments that would better characterize the heterogeneous coupling of contaminant sources and loadings with sediment dynamics and fate.

Vertical Profiles of Sediment Contamination

In depositional areas, sediment can provide a historic record of loading and deposition of persistent contaminants brought about by human activities in San Francisco Bay. One of the few long-term records of contamination in the Bay is from sediment cores collected by the U.S. Geological Survey (USGS) in 1990-1992. Although sediment mixing and resuspension distorts the vertical profile of contamination, sediment cores have successfully captured records of peak erosional disturbances in the bay and surrounding watersheds, loading, and deposition of mercury and other trace metals (Hornberger et al., 1999), chlorinated hydrocarbons, including PCBs and OC pesticides (Venkatesan et al., 1999) and PAHs (Pereira et al. 1999). The order of earliest detectable appearance of contaminants caused by human impact was as follows: PAHs > mercury > silver, copper, lead, zinc > DDT, and PCBs (van Geen and Luoma, 1999).

Sediment cores collected in the Bay have shown the impact of historic mercury and gold mining in the Sierra Nevada and Coast Range mountains. In three cores collected in Grizzly Bay, San Pablo Bay, and Richardson Bay, Hornberger et al. (1999) determined that the earliest anthropogenic signal of trace metals was from mercury (0.3-0.4 mg kg^{-1}) deposited in the San Pablo Bay core. The sediment layer was consistent with the time period between 1850 and 1880 (Figure 7), indicating that the mercury-laden sediment originated from hydraulic gold mining in the Sierra Nevada and Coast Range mountains that border the Central Valley. The highest mercury concentration of 0.95 mg kg^{-1} in the Grizzly Bay core was almost 20 times greater than the background levels (0.06 ± 0.01 mg kg^{-1}). More recently deposited sediment in the upper layers of all three cores has decreased in mercury concentrations to approximately 0.2-0.4 mg kg^{-1}.

In a core collected from a tidal marsh in the southern reach of the Bay, Conaway et al. (2003b) measured maximum concentrations of 1.2 mg kg^{-1} at a depth of ~65 cm, which was approximately 15 times higher than baseline concentrations (0.08 ± 0.03 mg kg^{-1}). Background concentrations were similar to those measured in other cores offshore from San Mateo and Oyster Point (0.070 ± 0.01 mg kg^{-1}) and by Hornberger et al. (1999) in the northern and central reaches of the Estuary. Surface concentrations in the South Bay core (0.4 mg kg^{-1}) were also consistent with concentrations from Hornberger et al. (1999). These coring studies have shown periods of increased mercury concentrations and loading occurring coincidentally with peak mining in the Bay watersheds followed by more diffuse distributions of mercury throughout the Bay.

As with mercury, concentrations of OC pesticides (DDT and chlordane) in the San Pablo Bay core reflected the influence of agricultural sources in the watersheds of the Sacramento and San Joaquin Rivers (Venkatesan et al., 1999). Maximum concentrations of ΣDDT were over three times higher in the San Pablo Bay core (57 μg kg^{-1}) than in the Richardson Bay core (17 μg kg^{-1}) (Figure 8). Less distinct differences were observed for Σchlordane concentrations, possibly due to widespread use in urban areas of Bay Area watersheds, combined with inputs from the Central Valley agricultural regions (Figure 9). In both cores, concentrations of ΣDDT and Σchlordane were minimal in layers that deposited before the advent of pesticide use. Like most contaminants measured in the

sediment cores, ΣDDT and Σchlordane concentrations declined in more recently deposited sediment to approximately 5-8 μg kg^{-1} and 0.4-0.6 μg kg^{-1}, respectively, in surface sediment.

In contrast to the marked impact from mercury and OC pesticide loading from the Central Valley, similar patterns was not observed for PCBs and PAHs (Venkatesan et al., 1999; Pereira et al., 1999). Subsurface maximum concentrations of PCBs in both cores were approximately 32-34 μg kg^{-1}, which declined at the surface to approximately 13-14 μg kg^{-1} (Figure 10). Sharp declines in PCB concentrations were measured in sediment layers deposited concomitantly with bans on the production and sale of PCBs in 1979. Higher concentrations of PAHs were measured in Richardson Bay compared to San Pablo Bay and were attributed to transport of PAH-contaminated sediment from sources of combustion in the Central and South Bays (Figure 11). Additionally, the San Pablo Bay core was most likely diluted by riverine inputs of sediment (Pereira et al., 1999) with terrigenous sources of hydrocarbons from peat moss in the Delta (Hostettler et al., 1999). A notable similarity between the cores was a shift in hydrocarbon biomarker signatures that symbolized a change from biogenic to anthropogenic influences that coincided with the advent of industrial activity in the region (Hostettler et al., 1999).

Sediment cores have been examined at several site investigations in highly contaminated areas to determine the potential risk of exposure to biota from historically deposited contaminants. Sediment cores collected in various studies indicate that some regions of the Bay have subsurface contaminant concentrations that are orders of magnitude greater than surface concentrations. For example, sediment core collected from Stege Marsh (on the Richmond shoreline) had a maximum mercury concentration of 430 mg kg^{-1} at a depth of 2.5 feet that was 460 times greater than surface concentration of 0.93 mg kg^{-1} (URS, 2000a). Similarly, maximum concentrations of PCBs measured in cores collected from San Leandro Bay and Stege Marsh were at least 70 times greater than concentrations in surface sediments (Daum et al., 2000; URS, 2000a). At these particular sites, the removal of sediment to a depth of 2 to 3 feet would expose previously deposited sediment that is at least an order of magnitude greater in concentration.

Taking into account that areas currently undergoing erosion have increased in some regions of the Bay since periods of peak contaminant deposition (Jaffe et al., 1998; Capiella et al., 1999), sediment cores provide valuable information about areas with potential for increasing 'legacy' contaminant inputs that may pose a greater environmental risk to biota in the event of large scale erosion or dredging of historically deposited material. For this reason, sediment coring studies should continue to evaluate the link between historic contamination and potential future impacts on water quality in erosional areas throughout the Bay and in localized areas of contamination along urbanized shorelines.

Sediment methylmercury

Similar to other contaminants of concern in San Francisco Bay, the distribution and production of methylmercury in aquatic systems is influenced in part by contaminant sources and sediment dynamics. However, unlike other contaminants, *in situ* production

of methylmercury from inorganic mercury by sulfate-reducing bacteria in the presence of organic material is an important mechanism of methylmercury distribution (Gilmour et al., 1992; Benoit et al., 1998; Lawrence and Mason, 2001). In many cases, factors other than total mercury affecting net production drive methylmercury concentrations in the environment.

Concentrations and net production of methylmercury vary throughout different regions of San Francisco Estuary (Conaway et al., 2003; Heim et al., 2004; Marvin-DiPasquale et al., 2003). Conaway et al. (2003) determined that methylmercury concentrations in sediment collected in the northern reach of the Estuary were positively correlated to total mercury, total organic carbon (TOC), and % clay. Conversely, methylmercury concentrations in the southern reach of the Bay (0.4 – 17 pmol g^{-1}; 0.09 – 3.7 μg kg^{-1}) were higher than those in the northern reach (<0.04 – 2.2 pmol g^{-1}; <0.009 – 0.75 μg kg^{-1}) and were only weakly correlated to mercury concentrations in sediment, TOC, or % clay. Methylmercury concentrations in the southern reach were therefore most likely a result of conditions that enhance methylation or external inputs of methylmercury (Conaway et al., 2003).

The southern reach also had the greatest net methylation evidenced by higher percentages of methylmercury to total mercury in sediment (>1% at Guadalupe River [BW15] and South Bay [BA21]). Conaway et al. (2003) attributed enhanced methylation in the southern reach of the Bay to a combination of nutrient inputs from wastewater, external supplies of methylmercury, and continuous mercury loading from areas impacted by historic mining, such as the Guadalupe River.

There were significant correlations between methylmercury and total mercury in the Delta, suggesting that the species are either co-deposited or that methylmercury production is a function of total mercury (Benoit et al., 1998; Heim et al., 2004). Similar to findings by Conaway et al. (2003) in the northern reach of the Estuary, methylmercury and methylmercury / mercury ratios were significantly correlated to organic carbon (reported as loss-of-ignition [LOI]) at numerous locations in Suisun Bay, the Delta, and tributaries.

Smaller scale spatial variability in methylmercury concentrations exists between wetland/marsh areas and open-water locations in the Bay. Marvin-DiPasquale et al. (2003) measured methyl mercury concentrations in sediment collected from a tidal marsh in San Pablo Bay (5.4 ± 3.5 μg kg^{-1}) that were over 10 times higher than concentrations measured offshore (0.45-0.75 μg kg^{-1}). In addition, methyl mercury production rates were at least five times higher in the marsh area (3.1 ng g^{-1} wet sediment d^{-1}) compared to open water locations (<0.06 ng g^{-1} wet sediment d^{-1}). Furthermore, the ratio of methylation to degradation rates was 25 times higher at the marsh site than ratios at other locations. Higher methylmercury concentrations and production rates in the marsh sediment were attributed to increased bacterial activity in anoxic sediment, high organic matter, and low pH (Marvin-DiPasquale et al., 2003). In wetland areas of the Delta, Heim et al. (2004) found that concentrations of methylmercury and sediment organic carbon (7-50%), as well as ratios of methlmercury /mercury, were greater in inner wetlands compared to

adjacent channels in the Delta. Greater mutilation production and concentrations were attributed to greater proximity to agricultural areas with higher nutrients and temperatures in the less-flushed areas of the inner wetlands.

Concentrations of methylmercury are typically higher in the summer in water and sediment of San Francisco Estuary (Conaway et al., 2003; Heim et al., 2004). In the Delta, Heim et al. (2004) measured twice-yearly peaks in methylmercury concentrations with the highest peak occurring in the summer. Similarly, Conaway et al. (2003) measured higher concentrations of methylmercury in the water column throughout the Estuary during summer sampling. These studies are consistent with other studies that measured increased methylmercury concentrations in spring and summer due to increased microbial activity and methylation in warmer temperatures, fresh supplies of organic matter from riverine inputs or spring phytoplankton blooms, and/or increased oxygen that solubilizes mercury sulfides and makes mercury available for methylation (Baeyens et al., 1998; Gilmour et al., 1998; Gill et al., 1999; Bloom et al., 1999).

In summary, concentrations of methylmercury in San Francisco Estuary vary spatially on regional and local scales, as well as seasonally. Concentrations in the Delta and South Bay are typically higher than in the northern reach of the Estuary. Additionally, wetland areas have higher concentrations and net production rates compared to well-flushed channels and open water locations. Seasonally, concentrations are typically higher during summer in the Delta and Bay. Due to the complex relationship between methylmercury loading, production and exposure to the food web, further empirical studies and trophic modeling are necessary for relating the distribution and production of methylmercury in sediment to contamination in fish and other top consumers.

III. Contaminant Transfer from Sediment to Biota

The primary health and management concerns over the existing pool of sediment contamination in San Francisco Bay are driven by the tendency of several persistent contaminants, particularly PCBs and methylmercury, to biomagnify in the food web to concentrations that may be harmful to wildlife and humans. As part of the PCB TMDL process, a food web model was developed to predict PCB concentrations in tissues of organisms at various trophic levels in the food web based on water and sediment concentrations in the Bay (Gobas and Wilcockson, 2002). This food web model was a first step in understanding how PCBs and other bioaccumulative contaminants biomagnify through the food web. In recognition that the food web structure of San Francisco Bay is highly complex and variable spatially and temporally, this review focuses on dominant mechanisms of contaminant transfer from sediment to invertebrate fauna that comprise large portions of diets of sport fish in the Bay.

Overview of Food Web Species of Concern

Key sport fish species have been identified and monitored every three years since 1994 to compare contaminant concentrations to screening values established to protect human health (Fairey et al., 1994; Davis et al., 1997; Greenfield et al., 2002). Fish species were selected for monitoring based on their popularity for sport fishing and included whiter

croaker, shiner surfperch, striped bass, jacksmelt, California halibut, white sturgeon, and leopard shark. According to a seafood consumption study that logged 1,700 interviews with Bay Area anglers between 1998 and 1999, the most popularly eaten fish were striped bass (> 50% of anglers), halibut (> 20%), jacksmelt, sturgeon, and white croaker (CDHS and SFEI, 2001).

To understand the dietary transfer of PCBs from the Bay to sport fish, the PCB food web modeling study investigated the gut contents of white croaker, jacksmelt, and shiner surfperch (Roberts et al., 2002). Invertebrates that were identified as large contributors to fish diets include major components of the benthic and pelagic food webs: phytoplankton, zooplankton, polychaetes, bivalves, amphipods, and crustaceans. In particular, the Asian clam *Potamacorbula amurensis*, which was introduced to the Bay in 1986 (Carlton et al., 1990), was an important dietary component in Redwood Creek samples of shiner surfperch (72.6% by weight) and jacksmelt (10.8%) and San Pablo Bay samples of jacksmelt (62.4%) and white croaker (24.6%). Other main components of shiner surfperch diets were the polychaete, *Neanthes succinea*, crustaceans, bivalves, and amphipods. Jacksmelt also fed on phytoplankton (> 85% by weight, in Redwood Creek and Oakland Inner Harbor) and *N. succinea* in San Pablo Bay. White croaker fed mostly on crangon shrimp and amphipods, including *Ampelisca abdita*, except in San Leandro Bay where zooplankton was the major portion of the diet (73%). In addition, Nobriega et al. (2003) analyzed gut contents of striped bass and largemouth bass in the Delta, which comprised approximately 88% of fish species caught in that study. Both bass species fed largely on crustaceans (*Corophium* spp., gammarid amphipods, opossum shrimp, and cladocerans) and smaller fish, such as inland silverside (*Menidia beryllina*) and yellowfin goby (*Acanthogobius flavimanus*). Largemouth bass also consumed damselflies (Zygoptea) as a large portion of their diet.

Pathways of Contaminant Exposure from Sediment

Accumulation of contaminants in benthic and pelagic food webs occurs through several pathways, including direct ingestion of particles, dietary uptake from food sources, and dissolved uptake from sediment pore water and the overlying water column. The mechanism by which invertebrates are exposed to contaminants is variable (e.g., habitat, feeding mode, etc.). In addition, an organism's ability to avoid or mitigate exposure to contamination is also variable (e.g., benthic dwelling polychaetes versus an aquatic daphnia). All of these pathways are influenced to a large extent by sediment processes and contamination and require further evaluation to improve our current ability in applying trophic-level modeling to predict how sediment contamination affects accumulation in both benthic and pelagic food web species of concern. Therefore, the following discussion focuses on processes involved for the first-order transfer between sediment and the benthic food web for mercury (via methylmercury), other toxic trace elements, as well as for organic contaminants.

Mercury and Methylmercury

Direct ingestion of particles, including phytoplankton, detritus, inorganic particles and sediment is a major source of exposure and uptake of mercury (primarily via methylmercury) into aquatic and benthic invertebrates. Bioaccumulation is typically

greater for methylmercury than for mercury in copepods (Lawson and Mason, 1998), mussels (Gagnon and Fisher, 1997), clams (Inza et al., 1997), polychaetes (Wang et al., 1998), and amphipods (Lawrence and Mason, 2001; Lawson and Mason, 1998). Studies have demonstrated that assimilation efficiencies (AEs) are typically higher for methylmercury uptake in polychaetes (*Nereis succinea*) and mussels (*Mytilus edulis*) from various sediment types and compositions (Wang et al., 1998; Gagnon et al., 1997) (Table 2). Using bioenergetic modeling, Wang et al., (1998) estimated that greater than 70% of mercury accumulation in *N. succinea* was derived from ingested sediment, while the relative importance of methylmercury uptake from sediment was more dependent on AE and partitioning (K_d) of methylmercury between the dissolved and particulate fractions. Furthermore, both mercury species were assimilated appreciably in anoxic sediments. Similar to findings for polychaetes and bivalves, biota-sediment accumulation factors (BSAFs) were approximately 10 times higher for methylmercury than for mercury in the amphipod, *Leptocheirus plumulosus*. Additionally, approximately 70-98% of methylmercury accumulation occurred from ingestion of food and/or sediment (Lawrence and Mason, 2001).

Although methylmercury typically comprises less than 1% of total mercury in coastal and estuarine sediments (*e.g.*, Conaway et al., 2003; Benoit et al., 1998), the contribution of methylmercury to total mercury accumulation has been estimated to be 5-17% in the polychaete, *N. succinea* (Wang et al., 1998), 3-50% in the polychaete, *N. diversicolor* (Muhaya et al., 1997) and ~10% in the amphipod, *L. plumulosus* (Lawrence and Mason, 2001). These studies indicate that ingestion of particle-associated methylmercury and mercury can be a major pathway of readily assimilable total mercury accumulation in benthic macroinvertebrates of San Francisco Bay.

The extent to which mercury and methylmercury are accumulated in benthic invertebrates is influenced by the presence of organic material in sediment. Several studies have shown that organic material in estuarine sediments may inhibit mercury and methylmercury accumulation (Muhaya et al., 1997; Mason and Lawrence, 1999; Lawrence et al., 1999; Lawrence and Mason, 2001). For example, the estuarine amphipod, *L. plumulosus*, accumulated less mercury and methylmercury in organic-rich water and sediment than in test media with lower organic carbon content (Lawrence and Mason, 2001).

In contrast to the negative correlation between organic material in sediment and bioavailability in some studies, there is evidence that organic material may enhance uptake and accumulation of methylmercury in some situations, while having little effect on mercury accumulation in polychaetes (Wang et al., 1998) and mussels (Gagnon and Fisher, 1997). Wang et al. (1998) calculated AEs of methylmercury in *N. succinea* that were higher in sediment with higher total organic carbon (TOC), while AEs from inorganic mercury were comparable over a range of TOC. Similarly, AEs of methylmercury in *M. edulis* increased with fulvic acid coatings on various types of sediment particles; however, AEs from mercury did not differ appreciably (Table 2) (Gagnon et al., 1997).

The character of sediment organic matter and quality of food are important factors that influence the extent to which some benthic invertebrates accumulate particle-associated mercury and methylmercury (Lawson and Mason, 1998; Lawrence and Mason, 2001). Lawrence and Mason (2001) exposed *L. plumulosus* to various conditions of mercury-spiked algae and sediment and found that the highest amphipod accumulation occurred in spiked sediment with added spiked algae. Less accumulation occurred in spiked sediment with unspiked algae, thus demonstrating the possibility of selective feeding on algae-rich sediment or greater bioavailability of mercury and methylmercury from algae. Additionally, while amphipods (*Hyalella azteca*) are able to assimilate phytoplankton that have been bacterially degraded for long periods of time (> 33 days), assimilation in copepods (*Eurytemora affinis*) decreases as phytoplankton degrades (Lawson and Mason, 1998). This suggests that suspension-feeding copepods actively feed on live algae and assimilate methylmercury quickly, while bottom-feeding amphipods can assimilate methylmercury from detritus and decaying matter in the sediment layer. Furthermore, the mussel, *M. edulis*, can rapidly assimilate methylmercury even from inorganic sediment particles with low nutritional value (Gagnon and Fisher, 1997). Therefore, addition of recent contaminated algal matter in sediment has been shown to improve the quality of food and enhance assimilation of methylmercury in selective-feeding invertebrates, but may not be necessary for appreciable assimilation in deposit and filter feeders, as well as detritivores.

Although sediment ingestion has been predicted to be a major pathway of mercury and methylmercury accumulation in benthic invertebrates, dissolved uptake from sediment porewaters is also an important mechanism of accumulation (Gagnon and Fisher, 1997; Wang et al., 1998; Lawrence and Mason, 2001). Wang et al., (1998) found that the proportion of methylmercury accumulation in *N. succinea* (concentrations of methylmercury ranged from 1.5 to 81.4 ng/L) increased linearly with the proportion of total dissolved mercury (concentrations of mercury ranged from 0.1 to 10 ug/L) that is methylated. Furthermore, dissolved uptake rates in *N. succinea* were highest for methylmercury compared to other trace metals with rates measured in decreasing order: silver > mercury > zinc > cadmium > cobalt > selenium (Wang et al., 1998; Wang et al., 1999b). For methylmercury, AE, dissolved uptake rates, and efflux rates are approximately 2 to 3.5 times higher than for mercury in *N. succinea* (Wang et al., 1998). Considering that dissolved methylmercury may comprise as much as 30% of total dissolved porewater mercury (C. Gagnon et al. 1996), exposure of benthic invertebrates to porewater may enhance methylmercury uptake.

Along with the influence of organic carbon and algae on bioaccumulation of mercury and methylmercury in aquatic and benthic invertebrates, accumulation of these mercury species is also dependent on numerous other geochemical, biological, and physiological factors. Gagnon and Fisher (1997) measured differences in K_d of approximately 2 to 3 orders of magnitude for mercury and methylmercury in various types of sediment with higher K_ds in organic-coated particles. While K_ds appeared to have little effect on mercury assimilation in *M. edulis*, methylmercury assimilation increased with increasing K_d (Gagnon and Fisher, 1997).

Biological factors that influence mercury assimilation include feeding behavior and physiological factors, such as feeding rate, gut passage times, and efflux rates. Wang et al., (1998) attributed greater than 70% of mercury uptake by sediment ingestion in *N. succinea* to the high ingestion rate of the worms. In other invertebrates (e.g., *M. edulis*) and larger fish, digestive processes and internal solubilization may regulate assimilation of mercury and methylmercury (Gagnon and Fisher, 1997; Leaner and Mason, 2002). Findings from these studies suggest that an accurate characterization of site-specific qualities of sediment (chemistry, composition and organic matter types), as well as the unique physiologies and feeding behaviors of various food web species, is required to accurately predict transfer of persistent contaminants from sediment to biota.

Other Trace Elements

The dominant accumulation mechanisms for mercury and methylmercury are similar to important pathways of accumulation of other toxic trace elements in benthic and aquatic invertebrates. Ingestion of particles as food or sediment is a major source of trace element assimilation in the benthic food web (Griscom et al., 2002; Lee and Luoma, 1998; Luoma et al., 1992; Gagnon and Fisher, 1997b). For trace elements that naturally exist in anionic form, such as arsenic and selenium, ingestion of food is the primary pathway of assimilation (Luoma et al., 1992; Wang et al., 1996; Wang and Fisher, 1998). Luoma et al. (1992) used kinetic modeling to predict that greater than 95% of selenium accumulation in the clam, *Macoma balthica*, is due to ingestion of food, which was attributed to a high AE and low dissolved uptake of selenite. Griscom et al. (2002) used similar modeling to estimate that filter-feeding clams would have greater accumulation of silver (>98%) and cadmium (>90%) from ingestion of food compared to deposit-feeding clams (49-93% for silver, 33-82% for cadmium). In a study of metal uptake in copepods, greater than 98% of selenium and 50% of zinc was derived from ingested food (Wang and Fisher, 1998). In addition, modeling of metal accumulation in the polychaete, *N. succinea*, showed that greater than 98% of accumulation of cadmium, cobalt, selenium, and zinc was due to sediment ingestion and was primarily a result of rapid ingestion rates, slow dissolved uptake rates, and relatively high AEs (Wang et al., 1999b).

As with mercury and methylmercury, organic carbon in sediment may reduce bioavailability of metals in some invertebrates (Decho and Luoma, 1994; Griscom et al. 2000), but has little influence on the deposit-feeding polychaete, *N. succinea* (Wang et al., 1999b). The lack of a correlation between organic carbon and bioavailability in the polychaete was consistent with mercury uptake (Wang et al., 1998) and was attributed to the facultative feeding characteristics of *N. succinea*. In the bivalves, *M. balthica* and *M. edulis*, Griscom et al. (2000) measured higher AEs of selenium and zinc from organic-poor particles in the bivalves, but lower AEs of silver and cadmium in the same particles (Table 3). Consistent with the findings for silver and cadmium, many invertebrates accumulate trace elements primarily by selectively feeding on organic-rich particles that are finer in size (Wang et al., 1999).

Differences associated with the effect of organic carbon on bioaccumulation of various metals depend on the nutritional quality and the type of organic matter associated with ingested particles. Improved nutritional quality or organic matter has increased

accumulation of metals in the bivalves, *M. balthica*, *P. amurensis* (Decho and Luoma, 1994; Lee and Luoma, 1998; Shlekat et al., 2000a), and *M. edulis* (Wang and Fisher, 1996; Gagnon and Fisher, 1997b) and the amphipod, *L. plumulosus* (Schlekat et al., 2000b). For example, the spring phytoplankton bloom in San Francisco Bay increased AEs of cadmium, zinc, and chromium in the clams, *P. amurensis* and *M. balthica* (Lee and Luoma, 1998). Furthermore, addition of phytoplankton to ingested particles increased selenium AEs by approximately 30% in *P. amurensis* (Schlekat et al., 2000a) and 66% in *M. balthica* (Luoma et al., 1992). This pattern is supported by study findings of higher metal AEs from phytoplankton in *M. edulis* and *M. balthica* compared to AEs from natural sediment (Wang and Fisher, 1996; Reinfelder et al., 1997; Griscom et al., 2002); however, this does not necessarily occur in copepods (Wang and Fisher, 1998) and the polychaete, *N. succinea* (Wang et al., 1999a).

One reason for enhanced accumulation in some species is the greater association of metals, such as cadmium, zinc, and selenium, for cytoplasm in algal matter (Lee and Luoma, 1998; Schlekat et al., 2002). Schlekat et al. (2002) determined that this pattern was supported by selenium accumulation from algal-rich particles in *M. balthica* and partially for *P. amurensis*, but not for the amphipod, *L. plumulosus*. Therefore, accumulation in some selective-feeding benthic invertebrates is well correlated to concentrations of cytosolic metals in algal matter, while other factors control accumulation in many nonselective-feeding amphipods, bivalves, and polychaetes.

Along with sediment ingestion, the relative importance of metal assimilation from the dissolved phase depends on physiological and biological characteristics, as well as geochemical factors that affect metal speciation. In mussels, dissolved uptake of metals (except for selenium), food ingestion and solute uptake can equally contribute to metal accumulation (Wang et al. 1996). In a study on dissolved metal influx rates in *M. balthica* and *P. amurensis*, Lee et al. (1998) calculated influx rates that were linearly correlated with dissolved metal concentrations. Similar correlations were found for the polychaete, *N. succinea* (Wang et al.,1999b). In addition, although most of the selenium and zinc accumulation in copepods occurs from food ingestion, greater than 50% of cadmium, cobalt, and silver accumulation is from dissolved uptake (Wang and Fisher, 1998). This was due to greater efflux rates following food ingestion than efflux rates following uptake from the dissolved phase (Wang and Fisher, 1998).

As noted previously, numerous geochemical factors and physiological differences control the varying extent to which metals are assimilated from all pathways. Bioavailability of metals may be reduced by binding to iron-oxyhydroxides, organic matter, and manganese oxides in oxidized sediment and to sulfides in anoxic sediment (Schlekat et al., 2000b; Wang et al., 1999b). For example, assimilation of silver and cadmium was about twice as high in *M. balthica* and *N. succinea* exposed to oxic sediments than in anoxic sediments (Table 4) (Wang et al., 1999; Griscom et al., 2000). A similar pattern was observed for silver assimilation in *M. edulis*, but not for selenium and zinc in any of the species. Nevertheless, bivalves and polychaetes were still able to assimilate sulfide-bound metals (Griscom et al., 2000; Lee et al., 2000; Wang et al., 1999), even when metal

concentrations were only a fraction of sulfide concentrations in anoxic sediment (Lee et al., 2000).

Feeding behavior and physiological factors, such as gut retention time, digestive processes, and efflux rates, also influence the extent of metal exposure and accumulation within the organism (Weston, 2000; Wang and Fisher, 1996; Wang et al., 1995; Decho and Luoma, 1994). Biphasic digestion, in which organic-coated particles may undergo relatively slow and intensive digestion compared to inorganic particles (Gagnon and Fisher, 1997), is evident in clams and mussels with greater AE values estimated from ingestion of organic-coated particles (Wang et al., 1995; Decho and Luoma, 1994). The time of food passage through the gut has been well correlated to assimilation (Wang and Fisher, 1996). Even speciation of metals within the organism can influence assimilation. Methylmercury, often associated with proteins in biota, has been shown to have the highest uptake rate constant in *N. succinea* (Wang et al., 1998). Other metals that tend to associate with proteins (e.g., silver, zinc, and cadmium) also display a higher dissolved uptake rate than other elements (Wang et al., 1999b).

These differences in physiology and feeding can cause species-specific differences in assimilation rates of metals in the benthic food web. Of particular interest for San Francisco Bay is the faster uptake rates and greater accumulation in *P. amurensis* than other species (Lee et al., 1998; Schlekat et al., 2002; Linville et al., 2002). For example, influx rates of cadmium, chromium, and zinc in *P. amurensis* were 4 to 5 times faster than in *M. balthica* (Lee et al., 1998). Faster uptake rates by *P. amurensis* were attributed to the large volumes of suspended particles that are filtered compared to *M. balthica*, which is predominately a deposit feeder (Lee et al., 2000). Furthermore, Schlekat et al. (2002) calculated higher AEs of selenium in *P. amurensis* (78-89%) and *M. balthica* (58-92%) than in the amphipod, *L. plumulosus* (32-70%). These species-specific differences, along with sediment chemistry and speciation, need to be accurately characterized in determining the extent to which toxic trace elements are introduced into the food web from sediment.

Organic contaminants

As with (methyl)mercury and other toxic trace elements, direct ingestion of sediment is an important mechanism by which benthic organisms assimilate organic contaminants such as PCBs and PAHs (Weston et al., 2000; Kaag et al., 1997; Timmerman and Anderson, 2003). In San Francisco Bay, Pereira et al. (1992) found similar distributions of alkanes and of hopane and sterane bioamarkers in sediment and *P. amurensis*, indicating that petroleum hydrocarbons associated with sediments are bioavailable to this bivalve. Similarly, deposit-feeding polychaetes in the Bay accumulate greater proportions of benzo(a)pyrene from ingested sediment than via other pathways (Weston et al., 2000). A comparison of biota-sediment accumulation factors (BSAFs; lipid-normalized concentration in tissue/organic carbon-normalized concentration in sediment) indicates that accumulation factors for the bioaccumulative contaminants PCBs (1.03-2.17) and OC pesticides (1.36-2.7) are higher than for PAHs (0.19-0.29) (Table 6). Therefore, the benthic food web is likely to readily assimilate the chlorinated hydrocarbons, PCBs and OC pesticides, from contaminated Bay sediments.

The high affinity of nonionic organic contaminants, such as PCBs and PAHs, to organic carbon in estuarine sediment may hinder their bioavailability and assimilation into benthic invertebrates (Meador et al., 1997, Maruya et al., 1997; Ewald et al., 1997). In an intertidal marsh of San Francisco Bay, BSAFs from PAHs in *P. amurensis* decreased with increasing percent fines in sediment and organic carbon (Maruya et al., 1997), suggesting that highly aromatic soot particles in depositional areas of the Bay may be important for controlling accumulation of PAHs in macroinvertebrates. Similar negative correlations between organic carbon and bioaccumulation of chlorinated hydrocarbons have been found for amphipods, polychaetes, and oligochaetes (Meador et al., 1997; Ewald et al., 1997).

Conversely, in some cases organic carbon on sediment may enhance bioaccumulation in benthic species by improving the nutritional quality of the ingested particles (Gunnarsson et al., 1999a; Gunnarsson et al., 1999b). For example, Gunnarsson et al., (1999a) exposed the echinoderm (*Amphiura filidformis*) to various forms of organic carbon, including green and brown macroalga, eelgrass, phytoplankton, and lignins to characterize uptake of PCBs from varying substrates. Greatest accumulation and growth occurred with uptake of the most labile organic carbon sources, while lowest accumulation occurred with the most refractory carbon sources. In a similar study, Gunnarsson et al. (1999b) found that PCB accumulation in the polychaete, *N. diversicolor*, increased with selective feeding on organic-rich particles. Considering that PCBs and PAHs in Bay sediment bind to various types of organic carbon with varying sorption affinities (Ghosh et al., 2003), characterizing the dominant sources of carbon within sediment matrices is essential for predicting the effect of organic carbon on bioaccumulation of these persistent organic contaminants in the benthic food web.

Assimilation of organic contaminants is also highly dependent on specific chemical properties that influence the extent to which they either bioaccumulate or remain associated with recalcitrant fractions of sediment. As previously noted, Tracey and Hansen (1996) compiled BSAF values from numerous studies and determined that BSAFs were much higher for PCBs and pesticides than for PAHs in various species (Table 5). When BSAFs were sorted by chemical class and individual species, mean BSAF values are almost an order of magnitude higher for PCBs (2.17) and pesticides (2.70) than for PAHs (0.29). Differences were attributed to differences in sorption affinities for sediment particles between contaminants (Tracey and Hansen, 1996). Significant variation in PCB accumulation also occurs due to difference in chemical properties of individual congeners. For example, Pruell et al. (1993) determined that sandworms and shrimp accumulated PCB 153 more than other PCB congeners, because they readily metabolized PCBs 52, 101, and 151, all of which have hydrogen atoms at the meta and para sites of the biphenyl molecule. These studies suggest that predictions of bioaccumulation need to take into consideration the varying chemical properties between contaminants, as well as between congeners of contaminant classes (e.g., PCBs and PAHs) to accurately reflect the transfer of organic contaminants from sediment to the benthic food web.

Similar to trace metal uptake, species-specific characteristics account for differing routes of exposure and capacities to accumulate organic contaminants. Maruya et al. (1997) calculated biota-sediment accumulation factors (BSAFs) for PAHs that were three orders of magnitude higher in *P. amurensis* than in polychaetes in an intertidal marsh of San Francisco Bay (Maruya et al., 1997). Accumulation of PCBs was also higher in clams (*M. nasuta*) and grass shrimp (*Palaemonetes pugio*) than for sandworms (*Nereis virens*) exposed to contaminated marine sediment (Pruell et al., 1993). Furthermore, in a comparison of accumulation between a non-deposit feeding amphipod and a nonselective deposit feeding polychaete, Meador et al. (1997) found that accumulation of (more hydrophilic) trichlorobiphenyls were similar between species; however, accumulation of more hydrophobic PCBs in polychaetes were 3 to 10 times higher than in the amphipod, which was attributed to sediment ingestion by the polychaete.

Implications for Food Web Modeling

The accumulation of persistent contaminants from sediment by benthic invertebrates is influenced by many complex factors. The modes and rates of assimilation in key benthic species in turn affect transfer through the food web to top consumers. Although the factors that influence accumulation in biota vary temporally, spatially, and even among individual organisms, there is a need to evaluate potential impacts on an aggregate basis. The better we are able to refine models of contaminant uptake and food web transfer in San Francisco Bay the better we can manage these pollutants in the environment without being either overly or insufficiently protective of wildlife and human health.

The complexities associated with the diverse geochemistry of Bay sediments highlight further information needs to improve or augment current food web modeling efforts. For example, the food web model developed for San Francisco Bay predicted actual concentrations in benthic invertebrates and fish reasonably well; however, greatest variation between observed and predicted values occurred for high-molecular weight PCBs. This was consistent with other modeling efforts that found highest variation for PCBs with relatively high K_{OW}s (Colombo et al., 1995; Bremle and Ewald, 1995; Thomann et al., 1992). The large variation was attributed to accumulation of high-molecular weight PCBs primarily from sediment, rather than uptake from water. Given that variable composition and characteristics of Bay sediment (e.g., labile versus refractory organic matter), as well as seasonal pulses of organic matter from runoff and algal blooms, greatly influences bioaccumulation and food web model predictions, further studies should attempt to characterize the distribution and types of particulate organic carbon in foraging areas for sport fish, wildlife, and humans.

In addition, species-specific routes of exposure influence food web model predictions due to differences in contaminant accumulation from sediment in various invertebrates of San Francisco Bay. In particular, *P. amurensis* has shown greater accumulation and faster uptake rates of trace elements and organic contaminants than other species. The high assimilation of contaminants in *P. amuren*sis may, in turn, enhance the assimilation in benthic predators and lead to greater accumulation through the benthic food web to top consumers, such as sturgeon and diving ducks (Baines et al., 2002). Furthermore,

abundance and types of invertebrate prey varies throughout the Bay, which results in large spatial differences in dietary composition for benthic predators.

Building on the simple food web model developed for PCBs in the Bay, improved models should include parameters for relating concentrations of contaminants of concern to wildlife. Because benthic invertebrates provide an important link between sediment and top consumers, both monitoring and food web models should address relationships between sediment contamination, benthic species abundance, and top consumer feeding patterns in the Bay.

IV. Sediment Processes

The impact of contaminated sediment on biota depends on complex sediment dynamics that govern accumulation and loss rates and post-depositional processes that rework the sediment through physical and biological mixing. Sediment processes that influence the rates of sediment contaminant transport and transfer to biota include (1) physical mixing and dispersion of contaminants through the active sediment layer to underlying buried sediment; (2) erosion and deposition of surface sediment; (3) molecular diffusion of soluble contaminants from porewater; and (4) bioturbation by benthic macrofauna.

Sediment Mixing

The depth of the sediment layer that actively mixes and interacts with the overlying water column is one of the most important characteristics influencing the long-term fate of contaminants in San Francisco Bay (Davis, 2002). The 'active sediment layer' depth varies due to site specific differences in physical and biological mixing, accumulation, and burial. For this reason, the depth of this layer is difficult to characterize on a bay-wide basis. However, some sediment coring and tracer studies have provided information on the depth and time scales of mixing and burial of sediment and associated contaminants at specific locations of the Bay (Leahy et al., 1976; Fuller, 1982; Fuller et al., 1999).

Residence times of particles in the water column influence time scales that suspended particles and associated contaminants are available for uptake from Bay water before settling into the bottom sediment. Fuller (1982) measured ^{234}Th activity in suspended particles in the Bay and estimated that residence times of particles in suspension and in the surface sediment zone that is easily resuspended were on the order of 0.3 to 8 days in water samples collected near the surface and 13 to 17 days in deep water samples. Shorter residence times of sediment in shallow waters were attributed to movement of sediment from shallow areas to deeper channels (Fuller, 1982). Geographically, residence times were approximately 3.5 ± 2.2 days in the South Bay and 5 ± 3 days in San Pablo Bay. Resuspension rates were also estimated to be an order of magnitude greater in deep regions of the South Bay (1.1 ± 1.4 g cm^{-2} yr^{-1}) than in the shallow regions (0.1 g cm^{-2} yr^{-1}). In addition, resuspension rates were approximately 2 to 5 times higher than net accumulation rates, suggesting that sediment is resuspended 2 to 5 times before depositing into the active sediment layer. Thus, sediment-associated contaminants are

available for biotic uptake or repartitioning with the liquid phase on the order of days in the Bay before resettling in the bottom sediment.

Once particles settle to the bottom, sediment mixing and accumulation rates determine the time scales in which sediment and persistent contaminants are buried and essentially removed from the Bay ecosystem. Consistent with particle residence time estimates, sediment accumulation rates are higher in the deeper channels compared to the shoal areas of the Bay (Figure 12). Also, accumulation rates were consistently higher in summer for all stations than in winter. Fuller (1982) estimated that mixing coefficients in the deep channel of the South Bay (32-320 cm^2 yr^{-1}) were at least an order of magnitude faster than rates in an adjacent shallow area (<3.2 cm^2 yr^{-1}). This was attributed in part to reworked sediment by live polychaetes in the deeper areas. In more recent cores collected in August 1992 from Richardson Bay, Fuller et al. (1999) estimated a mixing coefficient of 71 cm^2 yr^{-1} that fell within the range of the earlier estimates for deep South Bay.

Similar to mixing rates, accumulation rates were as much of an order of magnitude greater in deep zones of San Pablo Bay (0.7-1.0 g cm^{-2} yr^{-1}) compared to shallow areas (0.05-0.10 g cm^{-2} yr^{-1}). The accumulation rate of 0.825 g cm^{-2} yr^{-1} from the 1992 Richardson Bay core was also within the summer season range of accumulation rates found in Central Bay (Figure 12). Considering that concentrations of most contaminants of concern tend to be higher in the shallow margins of the Bay, slower sediment mixing and accumulation in these typically depositional areas could prolong time periods of burial (or removal) of persistent contaminants.

Rates of mixing, accumulation, and burial influence the magnitude of the active sediment layer depth, which has been estimated using isotopic profiles, tracer studies, and from anecdotal evidence, such as existence of burrowing organisms (discussed later in this report) (Fuller et al., 1999; Fuller, 1982; Leahy et al., 1976). For example, Fuller et al. (1999) estimated that the active sediment layer was approximately 33 cm in the 1992 Richardson Bay sediment core based on mixing and accumulation rates. In addition, Fuller (1982) estimated that the upper layers of sediment (15 cm) in deeper regions of the Bay are overturned by physical and biological mixing on the order of 100 days. In 1974, the U.S. Army Corps of Engineers used iridium-labeled sediment (^{192}Ir half-life = 74.37 days) to track the movement and fate of 1,500,000 m^3 of dredged material disposed in San Pablo Bay (Leahy et al., 1976). Within two months of disposal, tagged dredge material was found throughout San Pablo Bay, Carquinez Strait, and Suisun Bay in cores collected at depths of at least 13-23 cm. This time scale (~ 60 days) for overturning sediment to depths greater than 10 to 20 cm is consistent with the time scales estimated by Fuller (1982). Six months after disposal, a sample collected as far south as San Bruno Shoals in the South Bay had tagged sediment at a depth of 13-23 cm, while a sample collected from near-shore Emeryville had measurable amounts of tagged sediment in the core layer at a depth of 23-33 cm. Leahy et al. (1976) also measured considerable variation in the depth of actively mixed sediment on monthly time scales at several locations throughout the northern reaches of the Bay (Figure 13). Depths ranged from minimum depths of 3-13 cm to maximum depths of 23-52 cm. These studies reveal that

sediment is not only widely distributed throughout the Bay, but also actively mixed to depths of 30 cm or more over the course of just 2 to 3 months in some areas.

Models of sediment mixing and accumulation have also been used to demonstrate spatial and temporal variability in contaminant fluxes and accumulation in the Bay (Fuller et al., 1999; Hornberger et al., 1999). Fuller et al. (1999) developed a mixing model from radioisotopic analyses in the 1992 Richardson Bay core (from a highly depositional area) to predict that sediment mixing was responsible for delayed burial of contaminants on the order of decades. A one-year pulse of contaminant would have approximately 95% of its initial mass remaining in the mixed layer after 12 years, 82% after 20 years, 50% after 33 years, and 25% after 50 years. Contaminant fluxes were also estimated for Richardson Bay from that mixing model. For example, the flux of total DDT to Richardson Bay was approximately three times higher from 1952 to 1971 (410 ng cm^{-2}) compared to 1972 to 1992 (140 ng cm^{-2}). Mercury fluxes were at least twice as high from 1952 to 1971 (5.9 ng cm^{-2}) than from 1972 to 1992 (2.6 ng cm^{-2}).

To understand the spatial variability in metal deposition to the Bay, Hornberger et al. (1999) estimated metal fluxes in Grizzly Bay, San Pablo Bay, and Richardson Bay (Figure 14). Fluxes (in µg cm^{-2} yr^{-1}) were greater in San Pablo Bay for all metals, except for mercury, for which the highest flux was calculated in the Grizzly Bay core. As previously noted, the sediment core from Grizzly Bay had the highest subsurface maximum concentration of mercury (0.95 mg kg^{-1}) (Hornberger et al., 1999). High fluxes of other metals in San Pablo Bay could have been due to faster sedimentation rates at San Pablo Bay (4.5 ± 1.5 cm yr^{-1}) compared to Richardson Bay (0.8 cm yr^{-1}) (Fuller et al., 1999), and possibly Grizzly Bay.

Erosion and Deposition

Localized processes of erosion and deposition in Bay sediment pose a concern over the potential for exposing previously buried historic deposits of 'legacy' contaminants in areas. Bathymetric studies conducted by the USGS have provided the most recent and detailed information on these processes in San Pablo Bay (Jaffe et al., 1998) and Suisun Bay (including Grizzly Bay, Honker Bay, and Carquinez Strait) (Capiella et al., 1999), and South Bay (Foxgrover et al., 2003). Between 1856 and 1887, hydraulic mining in the Sierra Nevada foothills deposited approximately 250 million m^3 of sediment into San Pablo Bay (Jaffe et al., 1998). In Suisun Bay, approximately 61 million m^3 was deposited from 1867 to 1887 (Capiella et al., 1999). After cessation of hydraulic mining, Suisun Bay has been net erosional at a rate of 1-2 million m^3 yr-1 from 1887 to 1990 (Capiella et al., 1999). Over that time, Suisun Bay has lost approximately 40 acres of tidal mudflats from erosion and constructed dikes. Unlike Suisun Bay, San Pablo Bay gradually decreased in sediment supply and only began to be erosional in the 1950s (Jaffe et al., 1998). San Pablo Bay lost approximately 7 million m^3 of sediment between 1951 and 1983 with about 90 acres a year of mudflats disappearing (Jaffe et al., 1998).

The occurrence of net erosion in both embayments since the cessation of hydraulic mining in the Sierra Nevada foothills creates the possibility of erosion of historic deposits of mercury and other contaminants that are more concentrated than contemporary surface

deposits in sediment. Subsurface maxima of mercury in Grizzly Bay and San Pablo Bay (Hornberger et al., 1999) indicate that erosion of certain areas within these embayments may ultimately expose higher concentrations. In addition, the erosion of mudflats and tidal marshes in these areas may introduce contaminants from previously depositional areas or areas of enhanced mercury methylation (Marvin-DiPasquale et al., 2003). Furthermore, concentrations of other 'legacy' contaminants, such as PCBs and DDT, may have been deposited in areas of the Bay that are now undergoing erosion.

As discussed previously, subsurface concentrations in the Bay can range up to 2 orders of magnitude greater than surface concentrations. In terms of predicting future impacts of contaminants of concern, knowledge of subsurface concentrations, especially at potential erosional areas with suspected highly contaminated deposits, must be linked to bathymetric changes in the Bay. Because the most recent bathymetric surveys were conducted over a decade ago in the late 1980s and early 1990s, more updated information is required for accurately assessing where erosional areas of the Bay may coincide with potential exposure of very contaminated subsurface sediment.

Bioturbation

In San Francisco Bay, there is an abundance of burrowing macroinvertebrates that bioturbate and/or irrigate bottom sediment and potentially introduce sediment-associated contaminants to the water column through enhanced diffusive fluxes, particle resuspension, erosion, or porewater and particle advection (Korosec, 1979; Fuller, 1982; Nolan and Fuller, 1986; Fuller et al., 1999; Hammond et al., 1985; Caffrey et al., 1996; Kuwabara et al., 1999; Rivera-Duarte and Flegal, 1997; Gunnarsson et al., 1999b; Madsen et al., 1997; Ciarelli et al., 1999). Even capped sediments can be disturbed by organisms, resulting in increased contaminant fluxes from the underlying sediment (Simpson et al., 2002). There is also evidence that bioturbation may enhance the uptake and accumulation of contaminants in benthic macroinvertebrates (Ciarelli et al., 1999; Ciarelli et al., 2000).

Numerous studies conducted in San Francisco Bay have found burrow depths of approximately 5 to 80 cm in depth (Figure 15), while some sampling locations have been absent of burrows altogether (e.g., Korosec, 1979). Caffrey et al. (1996) found burrows in South Bay sediment from the tube-dwelling polychaetes, *Asychis elongate* and *Heteromastus filiformis*, which typically have burrow densities of approximately 0.1 to 0.7 burrows cm^{-2} (Hammond et al., 1985). In San Leandro Bay, Nolan and Fuller (1986) found evidence of small wormholes at a depth of 12 cm and a maximum of 22 cm. From San Pablo Bay to the South Bay, Korosec (1979) used x-radiographic techniques to determine that tube-dwelling polychaetes burrowed to depths of 7 to 40 cm with burrow densities of 0.13 to 0.62 burrows cm^{-2} on the surface and 0.08 to 0.70 burrows cm^{-2} at depth. Using modeling techniques, Korosec (1979) concluded that bioturbation enhanced dissolved nutrient fluxes, increased sediment porosity and mass transfer coefficients for dissolved species within interstitial waters, and exchanged water, sediment, and fecal material from burrows with overlying aerated water.

In some Bay sediment cores, the depth of radioisotope activity has not been adequately explained solely by rapid accumulation of sediment, but also by physical or biological mixing of sediment (Fuller, 1982; Fuller et al., 1999). Fuller (1982) determined that sediment cores from stations in the deeper channels of the South Bay had significant excess ^{234}Th activity at depths of 15 cm in cores that also had numerous live polychaetes (*Heteromastus filiformis*) as deep as 60 cm. Profiles of ^{137}Cs in sediment further supported greater mixing in the deeper channels compared to the shoals brought on by rapid reworking of sediment by bioturbation (Fuller, 1982).

More recently collected cores continue to support the abundant activity of burrowing organisms in mixing sediment (Fuller et al., 1999; Allison et al., 2003). Fuller et al. (1999) found cemented polychaete worm burrows in eight cores collected from Richardson Bay in 1992 extending to 12-15 cm. Live burrowing shrimp (*Callianassa sp*) were also observed at depths of 30 cm in some cores. Cores collected as recently as March 2000 further indicate that burrowing organisms are widespread throughout the northern reach of the Estuary at least to depths of 15 cm (Allison et al., 2003). The effect of burrowing organisms must be considered both in their mixing of sediment and porewater and in their role as a pathway for contaminants in the food web.

Benthic Fluxes

The geochemistry and biological activity (bioturbation/irrigation) in Bay sediments influence the benthic flux of dissolved constituents (including contaminants) across the sediment-water interface. The importance of benthic fluxes to San Francisco Bay has been demonstrated in several studies on nutrients and trace metals (Hammond et al., 1985; Kuwabara et al., 1999; Rivera-Duarte and Flegal, 1994; Rivera-Duarte and Flegal, 1997; Topping et al., 2001). Hammond et al. (1985) estimated that approximately 90% of the annual production of carbon and 65% of silica is exchanged between the water column and the benthos. The study further estimated that benthic fluxes in Bay shoals replace water column masses of ammonia in 2 to 6 days and silica in 17 to 34 days. In terms of mass budgets of trace metals in the Bay, benthic fluxes of dissolved lead and silver may be greater than fluvial inputs (Rivera-Duarte and Flegal, 1994; Rivera-Duarte and Flegal, 1997), while dissolved nickel fluxes are of the same magnitude as inputs from freshwater sources (Topping et al., 2001, 2003).

Magnitudes of dissolved contaminants fluxes are typically dependent on the gradient between porewater concentrations and overlying water column; therefore, more contaminated areas may have benthic fluxes out of the sediment, while less contaminated areas may have fluxes into the sediment (Rivera-Duarte and Flegal, 1997; Gill et al., 1999; Gill, 2001). Gill (2001) estimated methyl mercury fluxes in the range of –10 to 62 ng m^{-2} d^{-1} at several locations within the Delta and nearby tributaries (Table 6). The highest flux was estimated from a sediment core collected from Consumnes River, which had the highest porewater concentrations measured in the study (10.88 ng L^{-1} in May, 2000; 12.57 ng L^{-1} in September, 2000). Fluxes of mercury and methyl mercury estimated by Gill (2001) were within the range of reported fluxes from other studies. One note of interest is that methylmercury fluxes in the estuarine Lavaca Bay ranged approximately three orders of magnitude (0.2-1500 ng m^{-2} d^{-1}; Gill et al., 1999).

There is also considerable spatial and temporal variability in the magnitudes of benthic fluxes (Hammond et al., 1985; Topping et al., 2001; Gill et al., 1999). Hammond et al. (1985) measured differences in dissolved fluxes of 30% in distances of just a few meters to tens of meters in San Francisco Bay. Dissolved fluxes of silica and radon were greater on the shoals than in the deeper channels because of greater bioturbation and/or irrigation (Hammond et al., 1995). Furthermore, dissolved fluxes of oxygen, carbon dioxide, ammonia, and silica were greatest following the spring bloom. Gill et al. (1999) also measured seasonal differences in methylmercury fluxes in Lavaca Bay with highest fluxes occurring in late winter to early spring, which coincided with high production rates and inverse relationships with partitioning (Kd). In addition, diurnal variation in methylmercury fluxes caused higher fluxes during dark periods that coincided with increased nutrient fluxes and decreasing dissolved oxygen in the water column. Similar to previously discussed patterns of increased methylmercury concentrations and production, Gill et al. (1999) measured the highest fluxes in intertidal mudflats and grass flats.

Diffusive fluxes of trace metals have been shown to contribute significant proportions of total inputs to the Bay; studies are being conducted to characterize the relative contributions of mercury and methylmercury fluxes from sediment in the context of overall mass budgets for these contaminants. Several studies have measured potentially significant magnitudes of fluxes of both mercury species in other coastal and estuarine systems. Therefore, understanding the transfer of mercury and methylmercury in Bay sediment to the food web also requires accurate measurements of diffusive fluxes from sediment to porewaters and the overlying water column coupled with measurements uptake and assimilation rates.

V. Conclusion

Summary

Sediment Contamination in the Bay

- Contamination by persistent particle-associated contaminants is widespread throughout the Estuary, but spatially heterogeneous both on regional and localized scales. Ambient concentrations of PCBs, mercury, and OC pesticides are typically higher in southern segments of the Bay than in the northern reaches. Furthermore, contaminant concentrations may be at least 3 orders of magnitude higher in the margins of the Bay and areas of past contaminant use than in deeper channels. Areas of relatively high sediment contamination have also coincided with areas of high concentrations in sport fish and top consumers, such as piscivorous birds.

- Sediment cores collected in the Bay have provided an integrative record of deposition of sediment and associated contaminants. Relatively few cores have been collected for analyses of contaminants of concern; however, data indicate that concentrations of 'legacy'contaminants, such as mercury, PCBs, and organochlorine pesticides are highest at depths where sediment was deposited during time periods of peak usage. Contemporary concentrations of 'legacy' contaminants indicate more widespread distribution of lower concentrations throughout the Bay. As a result, subsurface concentrations of mercury and PCBs are up to 2 orders of magnitude higher at depths of 2 to 3 feet in some areas. Subsurface sediment in such areas could pose potential problems for water quality and food web uptake in the event of large-scale erosion or dredging of buried sediment.

- Methylmercury, like other persistent contaminants, is influenced by sediment dynamics and fate, but is also produced in sediment *in situ* by sulfate-reducing bacteria. Conditions that favor methylation of inorganic mercury (anoxic sediment, high organic matter and nutrient supplies, and warm temperatures) influenced spatial variation in concentrations throughout the Estuary. Methylmercury concentrations in the Delta and southern segments of the Bay were higher than concentrations measured in the northern reaches. Additionally, concentrations in marsh/wetland areas were approximately 10 times higher than in deeper channels, while production rates were 5 times higher. Seasonal increases in spring and summer of methylmercury concentrations have also occurred in the Bay and Delta due to conditions that enhance the production of methylmercury. Further studies are required to understand the coupling of methylmercury in sediment to sensitive food web species.

Contaminant Transfer from Sediment to Biota

- The major pathway responsible for uptake and accumulation of persistent trace elements and organic contaminants into many deposit-feeding invertebrates is through sediment and/or food ingestion. In particular, methylmercury is assimilated 2 to 10 times more efficiently than mercury in deposit-feeding polychaetes (*Nereis succinea*) and amphipods (*Leptocheirus plumulosus*). For selenium, ingestion of food

and sediment may contribute greater than 90% of accumulation in Bay resident clams (*Macoma balthica*), as well as in other filter- and deposit-feeding clams, copepods, and polychaetes. The relative contribution of accumulation of other trace elements from either ingested sediment or dissolved uptake is heavily influenced by partitioning between dissolved and particulate phases, as well as by differences among species in feeding behavior and physiology.

- Organic carbon may inhibit the accumulation of contaminants through chemical binding to organic carbon that renders contaminants relatively unavailable for uptake. This is especially important for hydrophobic nonionic organic contaminants, such as PCBs and PAHs, due to their differential affinities for various types of organic matter. Due to binding of organic contaminants to recalcitrant fractions of organic matter in sediment, negative correlations have been found between organic carbon content and bioaccumulation in the Asiatic clam, *Potamocorbula amurensis*, a resident of San Francisco Bay, as well as species of amphipods, polychaetes, and oligochaetes.

- Organic carbon may also enhance the uptake and accumulation of contaminants into biota. Assimilation of methylmercury in Bay resident polychaetes (*N. succinea*) and mussels (*M. edulis*) increased when exposed to organic-rich sediment. Similarly, the echinoderm (*Amphiura filiformis*) and polychaete (*Nereis diversicolor*) assimilated greater amounts of PCBs in the presence of organic-rich particles. Contrary to these observed patterns, assimilation of mercury and other trace elements (not including methylmercury) by the polychaete, *N. succinea*, and presumably other deposit-feeders that rapidly ingest large amounts of sediment may be unaffected by the presence of organic carbon on particles. This lack of a correlation was also observed in for mercury accumulation in the Bay resident mussel, *Mytilus edulis*.

- Increased assimilation through ingestion of organic-rich particles may be due to improved nutritional quality of particles. Ingestion of algal-rich particles increased assimilation of methylmercury in selective-feeding amphipods (*L. plumulosus*) and copepods (*Eurytemora affinis*), but had little effect on the filter-feeding *M. edulis*. Selective feeding by copepods, however, caused assimilation to decrease as phytoplankton began to degrade. A similar preference for algal-rich particles was observed for accumulation of trace elements in the bivalves, *P. amurensis*, *M. balthica*, and *M. edulis*, and the amphipod, *L. plumulosus*, but not for the polychaete *N. succinea*.

- Dissolved uptake from sediment porewaters and the overlying water column is also an important pathway of contaminant assimilation for some benthic invertebrates. Methylmercury can comprise as much as 80% of all mercury in porewaters of estuarine sediment, thus increasing the potential for assimilation in the benthic food web. In addition, methylmercury uptake rates are faster compared to other trace elements in *N. succinea*. Linear correlations have been determined between dissolved concentrations of trace elements in the water column in the clams, *M. balthica* and *P. amurensis*, and the polychaete, *N. succinea* suggesting that partitioning of trace

elements between sediment and porewaters is an important control on their dissolved uptake.

- Along with organic carbon and nutritional quality of ingested particles, there are numerous geochemical, physiological, and biological factors that influence the assimilation of contaminants into the benthic food web. Geochemical factors may include partitioning of contaminants between dissolved and particulate phases or differential binding of organic contaminants to various types of organic matter. Species-specific differences account for different routes of exposure and capacities for assimilation of contaminants between benthic invertebrates. In particular, greater accumulation of contaminants has been measured in *P. amurensis* than in other macroinvertebrates of San Francisco Bay.

Implications for Food Web Modeling

- The ability of benthic invertebrates to readily accumulate persistent contaminants from sediment underscores the need for further understanding of the extent to which contaminants in Bay sediment are assimilated in key benthic species and transferred through the food web to top consumers. Food web modeling facilitates understanding these processes of contaminant transfer and could be refined to relate contaminant concentrations in sediment to potentially harmful concentrations in wildlife and humans. Additional studies or information that could be incorporated into current food web modeling are spatial and seasonal characterizations of types of organic matter in sediment and the relationship between sediment contamination, benthic species abundance, and feeding patterns of top consumers.

Sediment Processes that affect Contaminant Fate

- Rates of sediment mixing were an order of magnitude lower in shallow areas of San Pablo Bay compared to open water locations. Similarly, accumulation rates were approximately an order of magnitude lower in shallow regions of the South Bay compared to the deep channels. Considering that concentrations of most contaminants of concern tend to be much higher in the shallow margins of the Bay, slower sediment mixing and accumulation in these typically depositional areas could slow the burial (or removal) of persistent contaminants and prolong the problem of contamination in the food web.

- Sediment mixing, accumulation, and burial rates control the depth of the actively mixed sediment layer and greatly influence the long-term fate of persistent contaminants in the Bay. The active sediment layer is highly variable throughout the Estuary in the range of approximately 3 to 50 cm. In addition, time scales for overturning sediment in the top 15 to 25 cm have been measured on the order of 60 to 100 days.

- Contaminant fluxes and accumulation are delayed on the order of decades by physical and biological mixing of sediments. Highest contaminant fluxes have occurred at locations that are either in close proximity to contaminant sources or that have higher sedimentation rates.

- Bathymetric studies indicate that areas of San Pablo, Suisun, and South Bay are gradually eroding. Sediment cores collected in these embayments had subsurface concentrations of mercury, OC pesticides, and PCBs that were 2 to 7 times greater than surface concentrations. In addition, cores collected in more contaminated areas of the Bay had concentrations of mercury and PCBs that were at least 1 to 2 orders of magnitude greater at depths of 2 to 3 feet compared to surface concentrations. As previously depositional areas of the Bay continue to erode, these areas of high subsurface maximum concentrations may prolong water quality problems and exposure of contaminants to sensitive food web components.

- An abundance of burrowing organisms in San Francisco Bay significantly alters bottom sediment through bioturbation/irrigation to depths in the range of 5 to 80 cm. These burrowing activities typically increase diffusive fluxes of solutes, resuspension and erosion of particles, and advection of porewater and particles from burrows. Therefore, the presence of burrowing organisms have a significant effect on the depth of the active sediment layer, and thus, the long-term fate of persistent contaminants, as well as the potential for increasing contaminant exposure to the Bay food web.

- Diffusive fluxes of dissolved nutrients and trace metals from Bay sediment are significant contributors to the cycling of these solutes within the Bay and in the overall context of mass budgets. However, no studies have measured fluxes of mercury and methylmercury from Bay sediment. Several studies have measured potentially significant magnitudes of fluxes of both mercury species in other coastal and estuarine systems, as well as significant spatial, seasonal, and diurnal variability in fluxes. Therefore, understanding the ultimate fate of mercury and methylmercury in Bay sediment also requires accurate measurements of diffusive fluxes from sediment and their effect on uptake rates and assimilation into the benthic and pelagic food webs.

Next Steps

This literature review identified several key areas that require further study or evaluation to improve our ability to understand and predict the fate of persistent contaminants in sediment and their uptake into the benthic and pelagic food webs of San Francisco Bay. Suggested studies for further investigation include empirical evaluations of sediment cores and bed sediment dynamics, as well as numerical modeling of contaminant fate in Bay sediment and water and its relation to accumulation in the food web.

1. Sediment Coring Studies – The best record of historical contamination in the Bay is from sediment cores collected by USGS in the early 1990s; however, only 3 cores were analyzed for mercury concentrations and 2 for organic contaminants. Additional sediment cores have been collected in more intensive site investigations, but with limited focus and suitability for providing information on the impact of localized sites on water quality and the food web in the Bay. For example, a Clean Estuary Partnership project that collected cores from near shore areas addressed the potential for these areas to supply contaminated sediments to the Bay but did not explore questions of fluxes or uptake. Therefore, collection of sediment cores in erosional areas of the Bay could provide valuable information on:

    a. Regional and smaller-scale spatial variability in contaminant distribution and fate of contaminants in ambient locations of the Bay and in more contaminated areas along shorelines or areas of past contaminant use.
    b. Vertical profiles of contamination in areas that are eroding to predict potential future areas of concern (coupled with current bathymetric studies; see #3).
    c. Sediment mixing, accumulation, and burial rates to assist in characterizing regional variation in active sediment layer depth and residence time scales of sediment and persistent contaminants before burial or removal from the Bay. .
    d. Diffusive flux measurements of mercury and methylmercury to understand benthic inputs of these mercury species in the context of loading from other pathways and effects on exposure to benthic and pelagic food webs.

2. Bed Sediment Dynamics – As part of the RMP, the USGS has collected continuous data on suspended sediment in the water column at several locations in the Bay; however, little is known about bed sediment dynamics and transport. Studies that evaluate bed sediment dynamics and transport will further our understanding on cycling and residence times of sediment and persistent contaminants in the Bay. Data on sediment mixing from radiotracers (Fuller et al., 1999) and introduced chemical tracers (Leahy et al., 1976) can be used to confirm or refine model predictions.

3. Bathymetric Surveys - The most recent bathymetric studies in San Pablo and Suisun Bays were completed by the USGS in the late 1980s and early 1990s. In addition, a bathymetric survey is being done for South Bay that will provide information on erosion/deposition in that region. Given that erosional areas in San Pablo and Suisun

Bays have continued to increase and subsurface concentrations of mercury and other contaminants of concern in these areas may be 2 to 7 times greater than surface concentrations, these areas may contribute to future impacts on water quality and food web contamination. Therefore, updated bathymetric studies in all segments of the Bay, coupled with sediment coring studies, will identify and describe areas of potential concern, especially in areas inhabited by sensitive species.

4. Multi-Box Modeling – The RMP has begun developing a multi-box model in cooperation with USGS to improve on the spatial resolution of the simple one box-model previously used to predict the long-term fate of PCBs, PAHs, and OC pesticides. Given that contamination in the Bay is heterogeneous on regional scales, the multi-box model, which consists of five boxes to represent major segments of the Bay, will improve our understanding of persistence and fate of PCBs in different regions of the Bay.

   Additional studies that would improve or augment multi-box modeling include:

   a. Greater spatial coverage and characterization of important characteristics of Bay sediment, such as the active sediment layer and surface sediment concentrations of persistent contaminants, especially in more contaminated areas.
   b. Characterization of sedimentary organic matter composition and types (e.g., labile and refractory components) and their effect on partitioning behavior of organic contaminants to sediment.

5. Hot-Spot Modeling- Smaller scale models of sediment and contaminant transport will be useful in decision-making for remediation of highly contaminated sites, as the risk of contaminant exposure and distribution will be very site-specific and would not be adequately characterized by a multi-segment Bay-wide model. A model by URS undertaken to predict the impacts of the San Francisco Airport expansion project contained over one million nodes; although such a detailed modeling effort may not be needed, it illustrates the level of complexity that is possible.

6. Food Web Modeling – The ability of benthic invertebrates to readily accumulate persistent contaminants from sediment highlights the need for development of food web modeling to extrapolate easily measurable concentrations in sediment to potentially harmful concentrations in ecological receptors, such as piscivorous birds and marine mammals. A simplified food web model has been applied to San Francisco Bay to predict the trophic transfer of PCBs in benthic invertebrates and fish based on concentrations in water and sediment; however, improvements in food web modeling are vital in understanding the fate of sediment-associated contaminants and their patterns of exposure to wildlife and humans.

   Additional studies that would improve or augment food web modeling include:

a. Evaluation of the effect of various types of organic matter, including algal blooms, on bioaccumulation of contaminants in various trophic levels.
b. Methylmercury-specific issues that tie spatial and temporal variability in sediment concentrations and production to accumulation in the food web.

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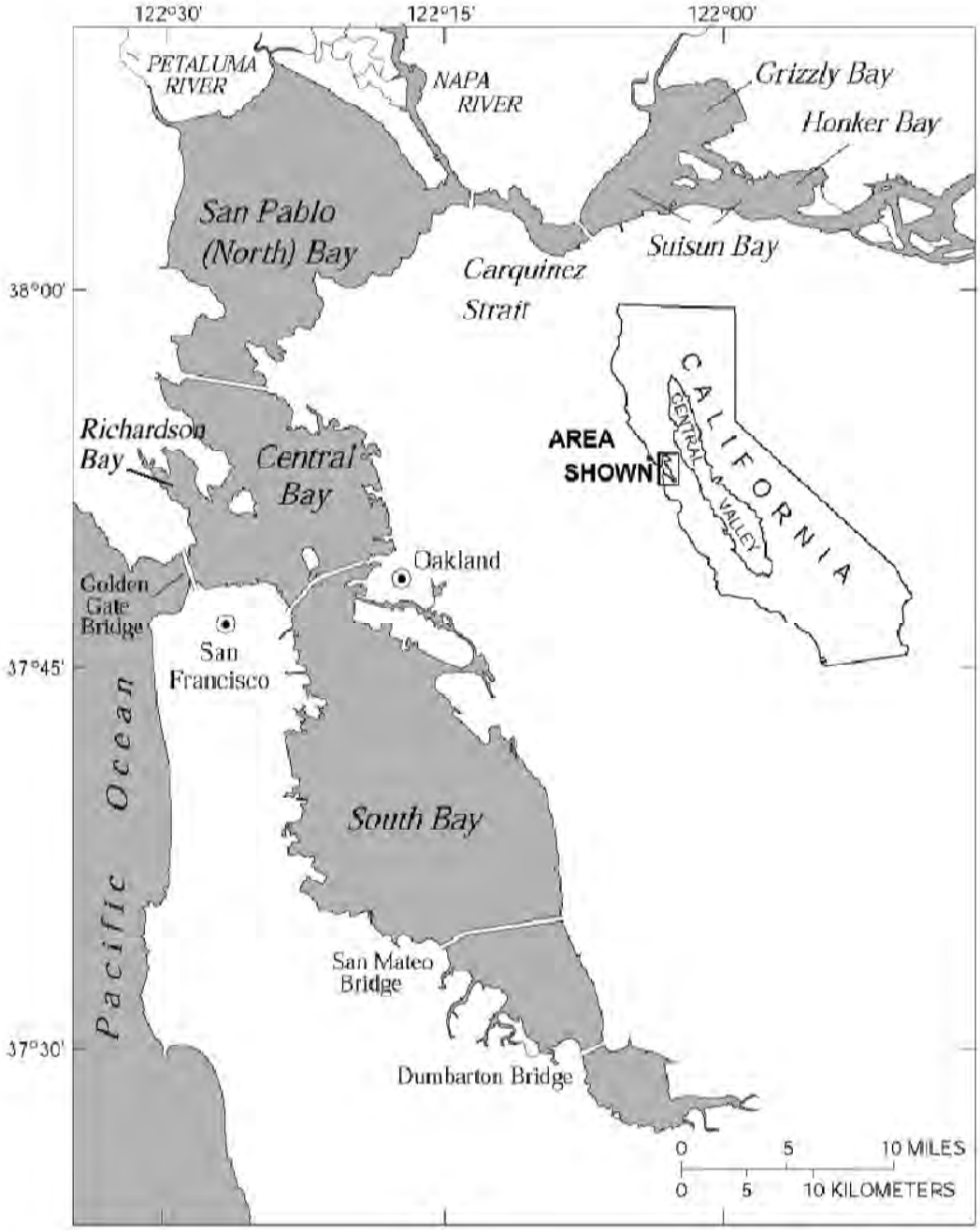

Figure 1. Map of San Francisco Estuary.

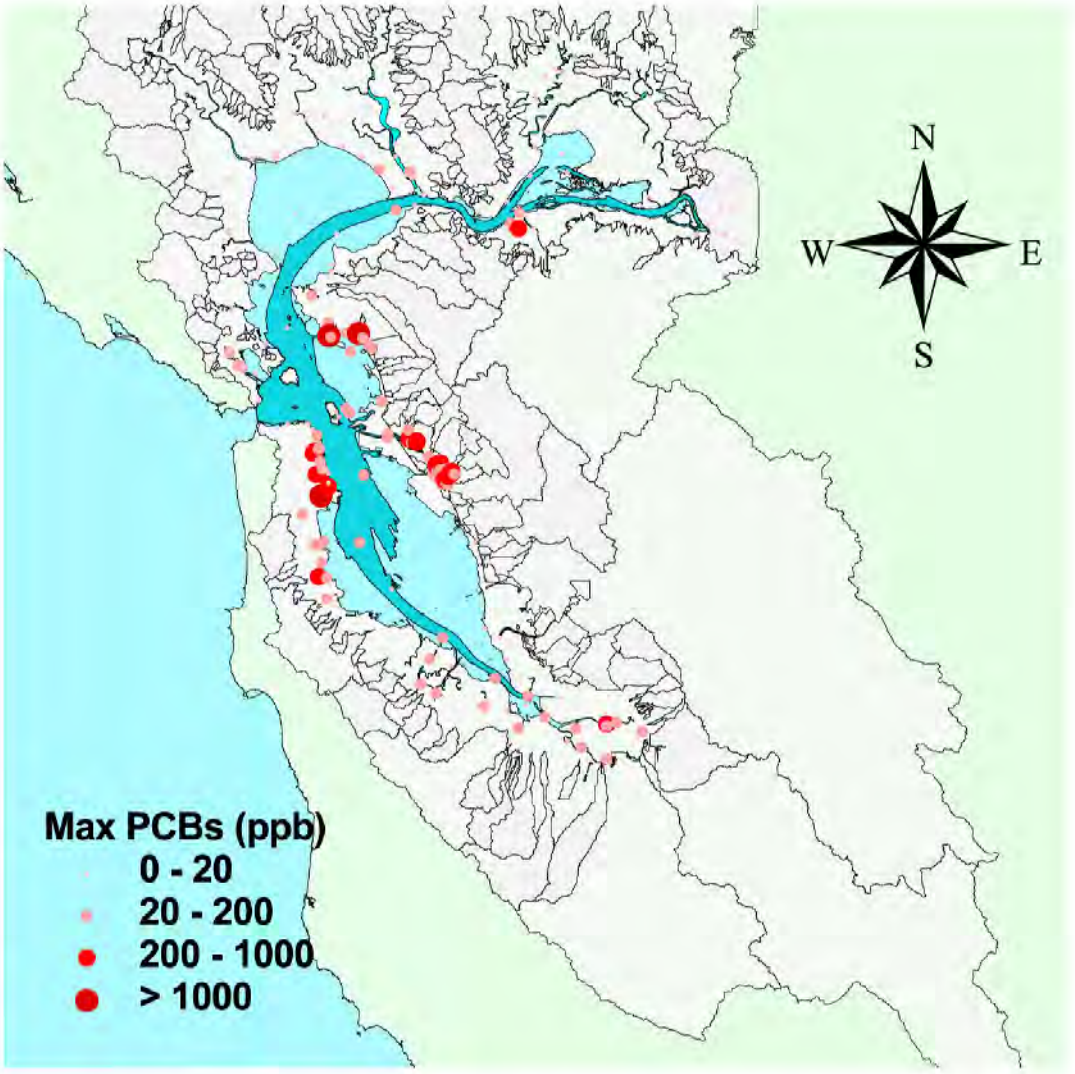

Figure 2. PCB concentrations in Bay sediment from 1991-1999. Maximum concentrations shown. Data from PRMP 1991-1992 (Flegal et al., 1994), RMP 1993-1999 (SFEI, 2003), BPTCP 1994-1997 (Hunt et al., 1998), and Daum et al., 2000.

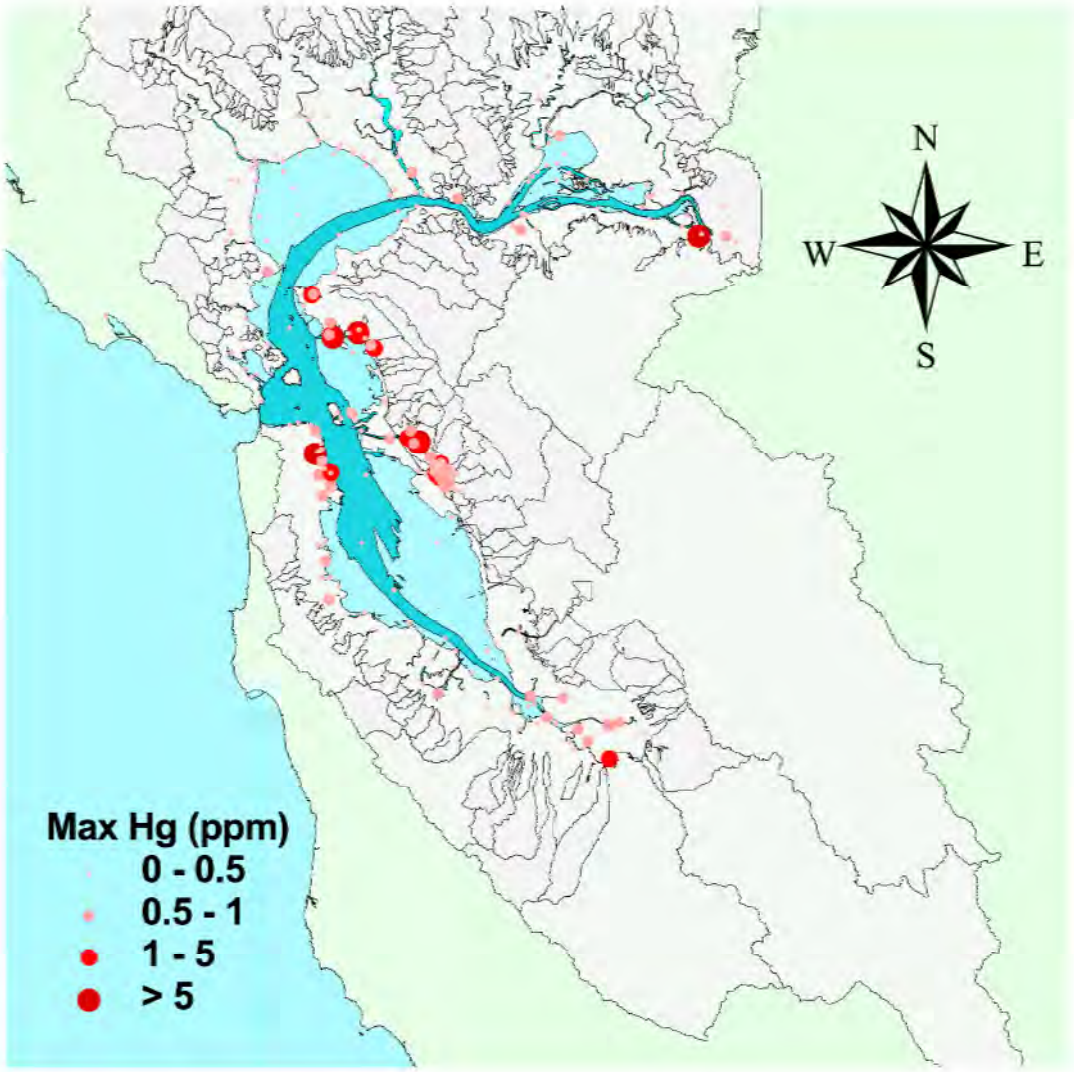

Figure 3. Mercury concentrations in Bay sediment from 1991-1999. Maximum concentrations shown. Data from CALFED 1999 (REFERENCE), PRMP 1991-1992 (Flegal et al., 1994), RMP 1993-1999 (SFEI, 2003), BPTCP 1994-1997 (Hunt et al., 1998), and Daum et al., 2000.

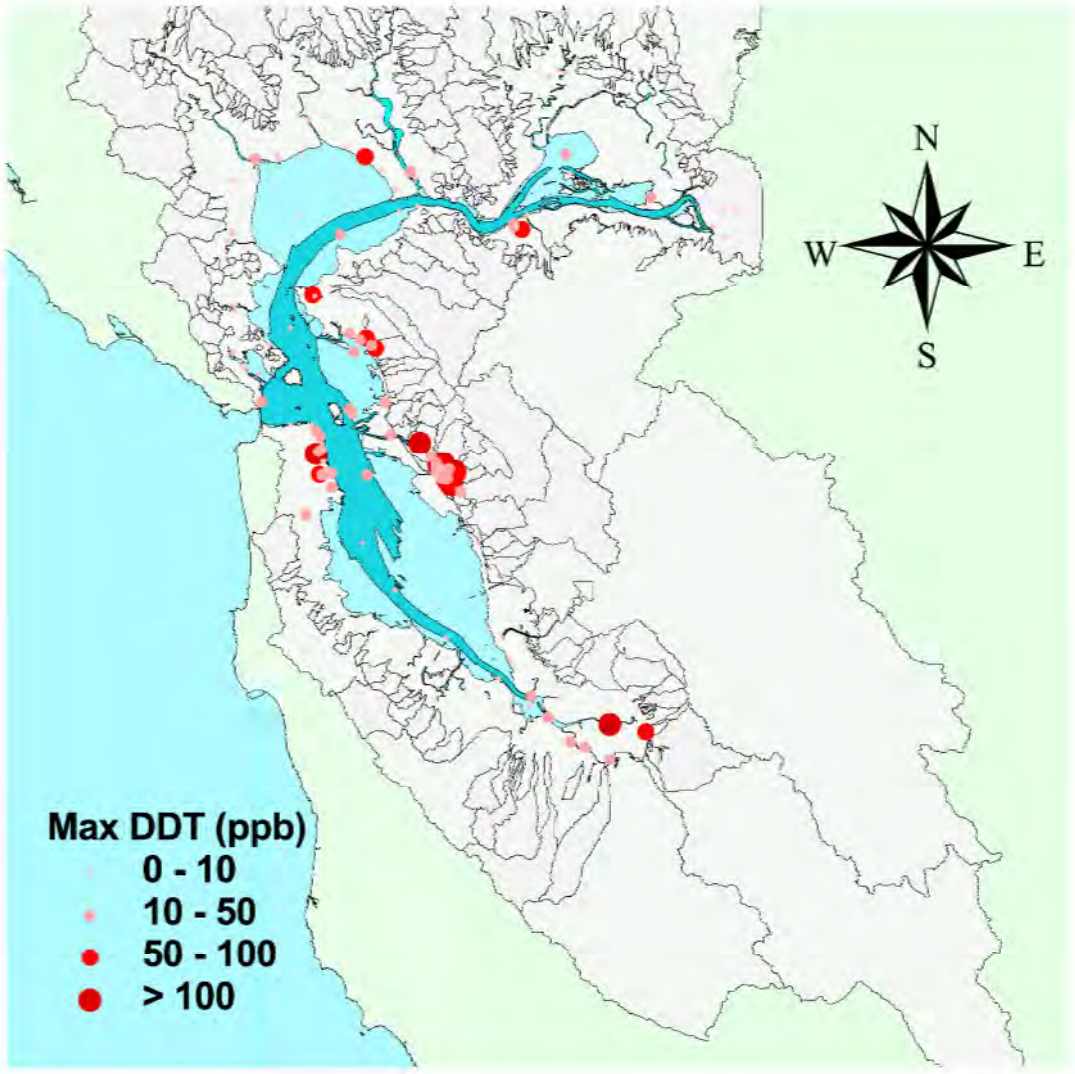

Figure 4. DDT concentrations in Bay sediment from 1991-1999. Maximum concentrations shown. Data from PRMP 1991-1992 (Flegal et al., 1994), RMP 1993-1999 (SFEI, 2003), BPTCP 1994-1997 (Hunt et al., 1998), and Daum et al., 2000.

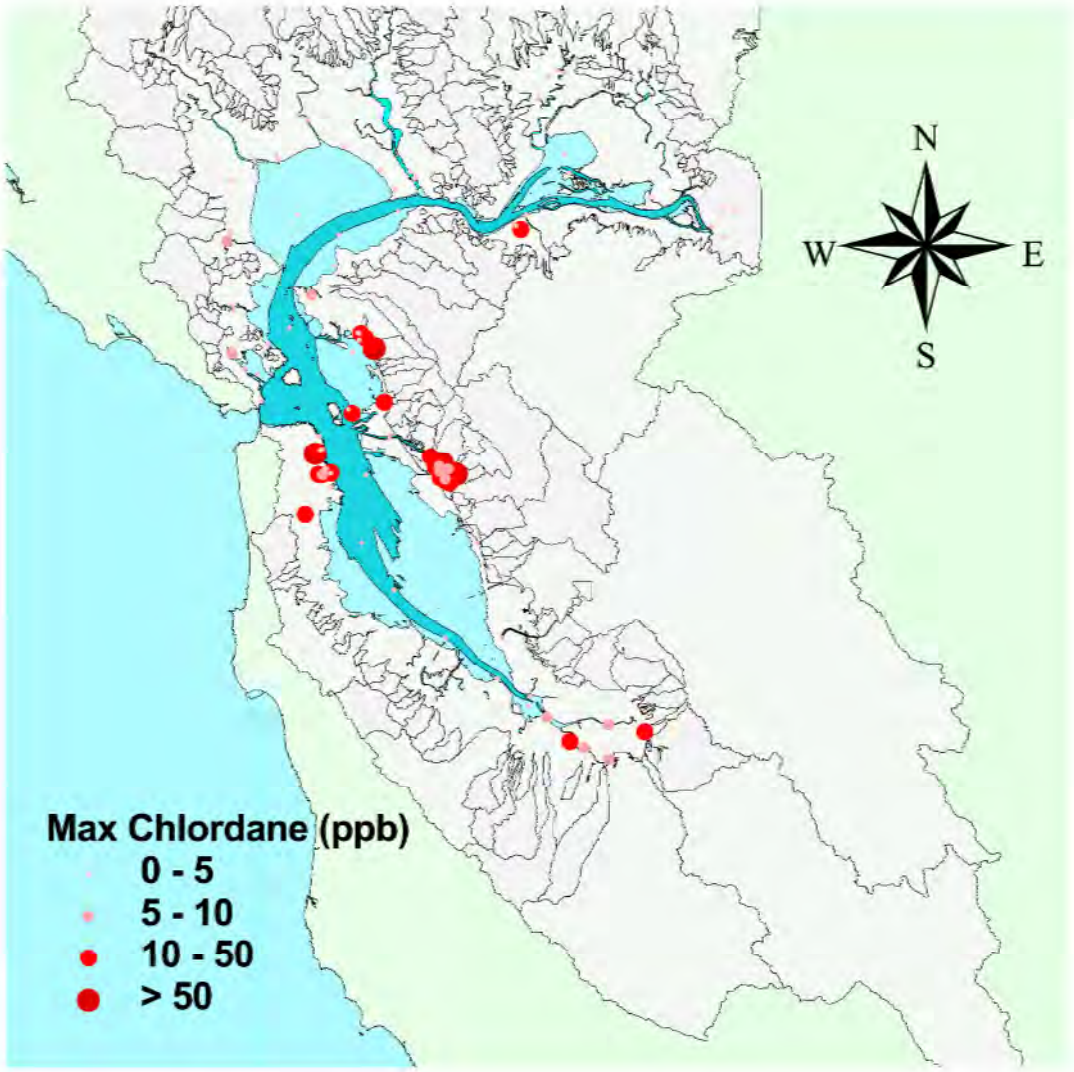

Figure 5. Chlordane concentrations in Bay sediment from 1991-1999. Maximum concentrations shown. Data from PRMP 1991-1992 (Flegal et al., 1994), RMP 1993-1999 (SFEI, 2003), BPTCP 1994-1997 (Hunt et al., 1998), and Daum et al., 2000.

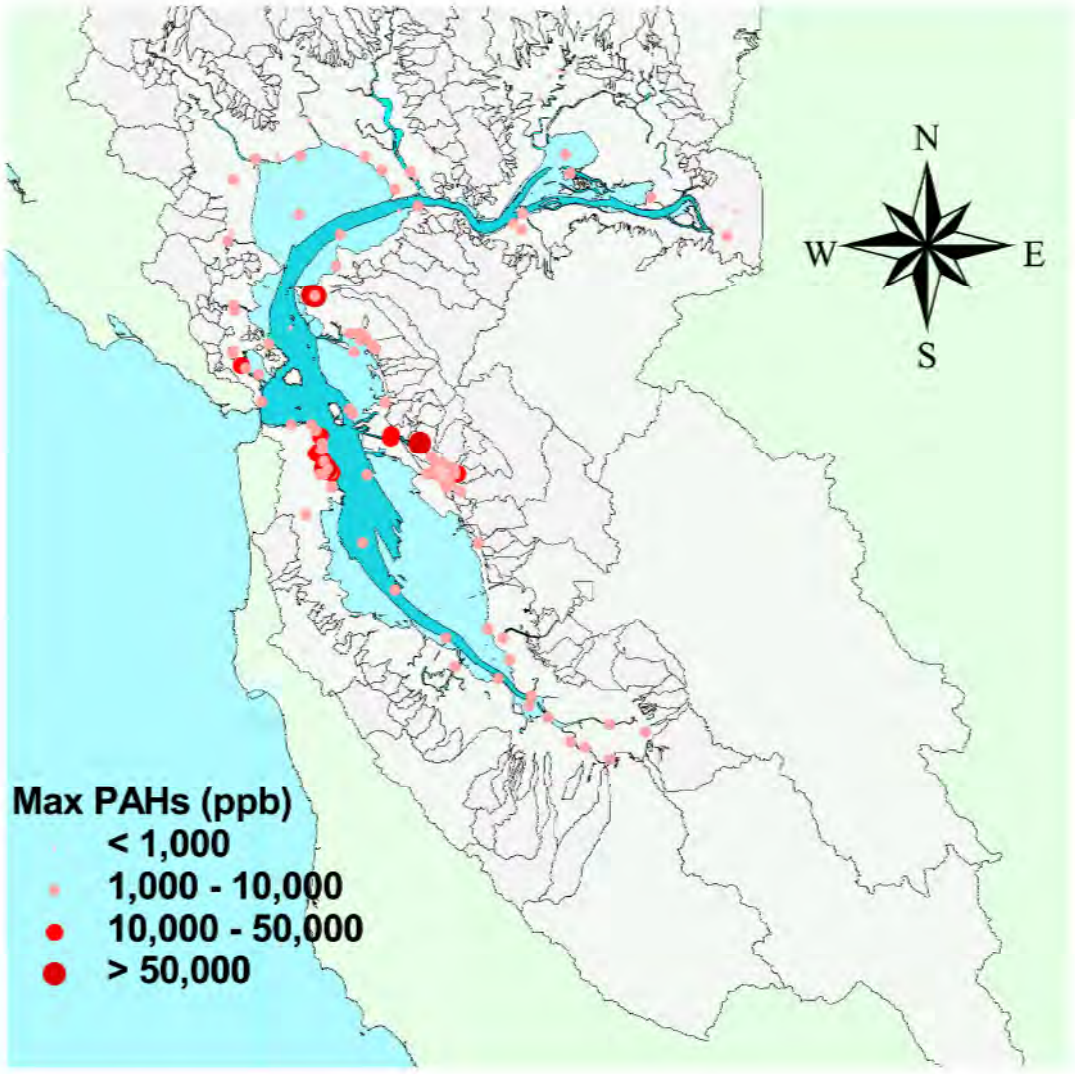

Figure 6. PAH concentrations in Bay sediment from 1991-1999. Maximum concentrations shown. Data from PRMP 1991-1992 (Flegal et al., 1994), RMP 1993-1999 (SFEI, 2003), BPTCP 1994-1997 (Hunt et al., 1998), and Daum et al., 2000.

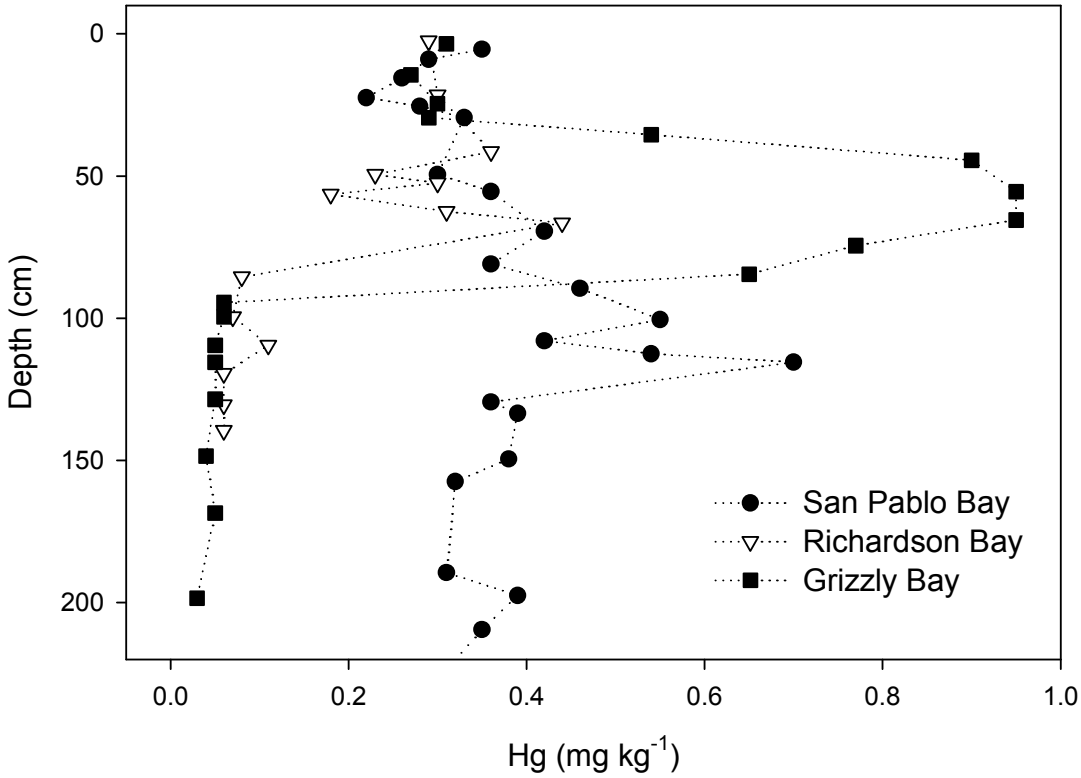

Figure 7. Mercury in sediment cores. Sediment cores were collected from Grizzly Bay, San Pablo Bay and Richardson Bay by the USGS in 1990, 1990, and 1992, respectively. Data from Hornberger et al. (1999).

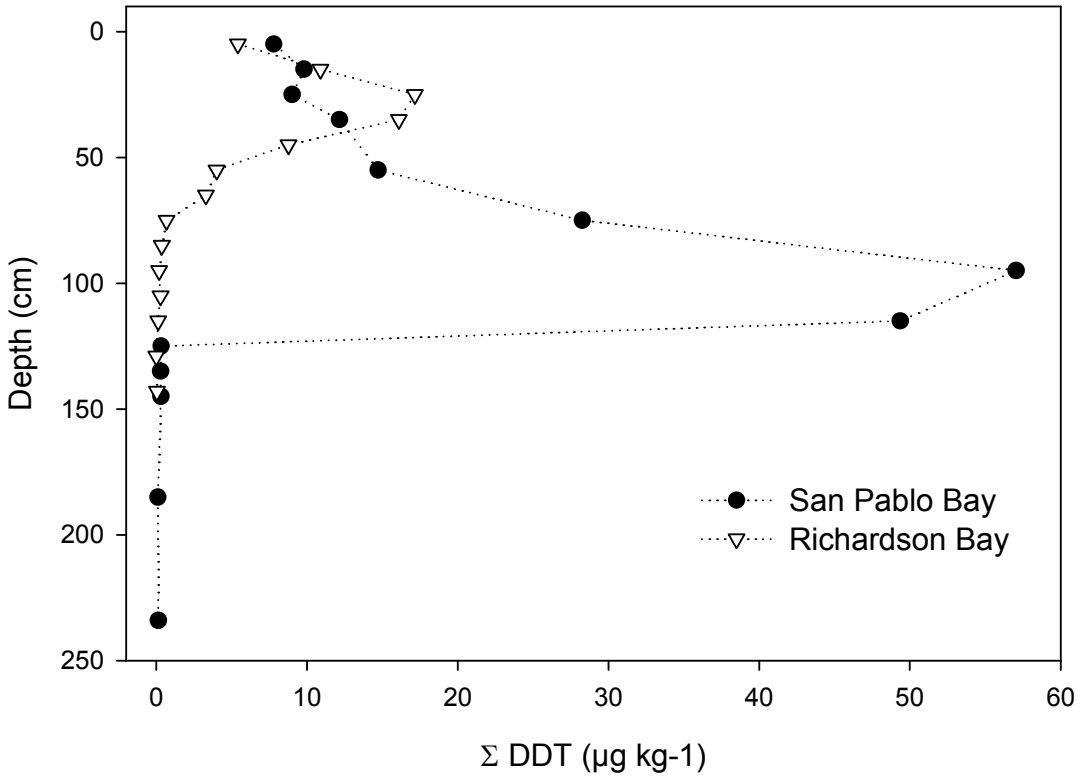

Figure 8. Total DDT in sediment cores. Sediment cores were collected from San Pablo Bay and Richardson Bay by USGS in 1990 and 1992, respectively. Data from Venkatesan et al. (1999).

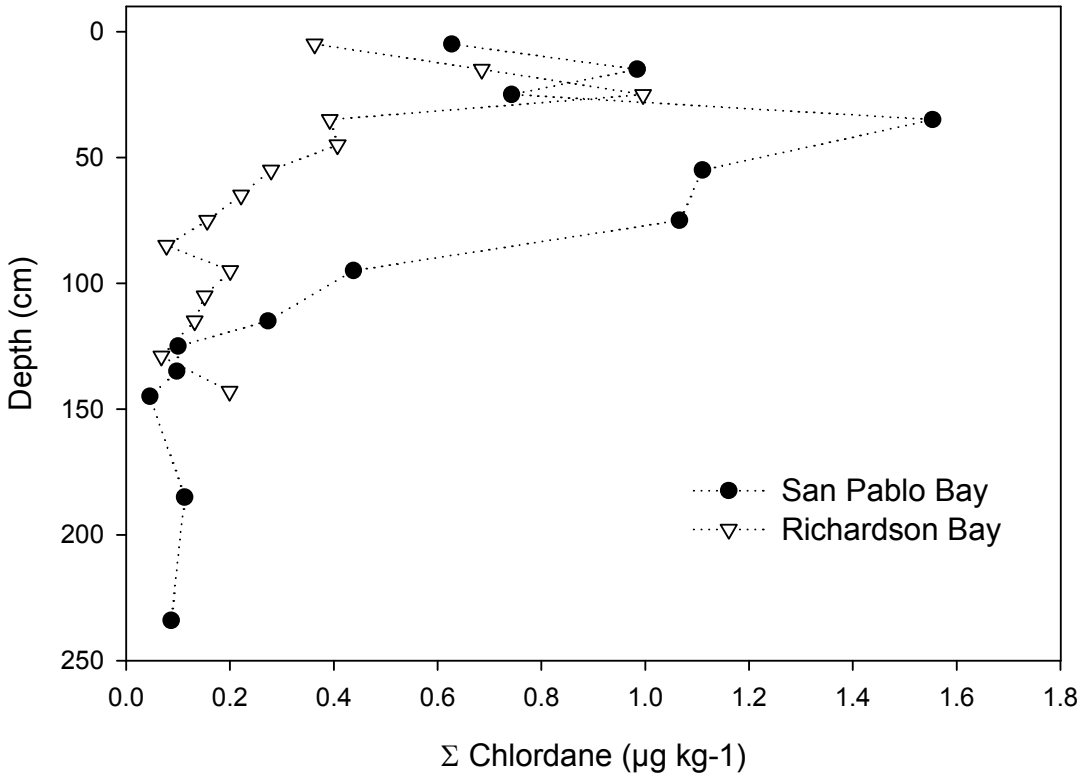

Figure 9. Total chlordane in sediment cores. Sediment cores were collected from San Pablo Bay and Richardson Bay by USGS in 1990 and 1992, respectively. Data from Venkatesan et al. (1999).

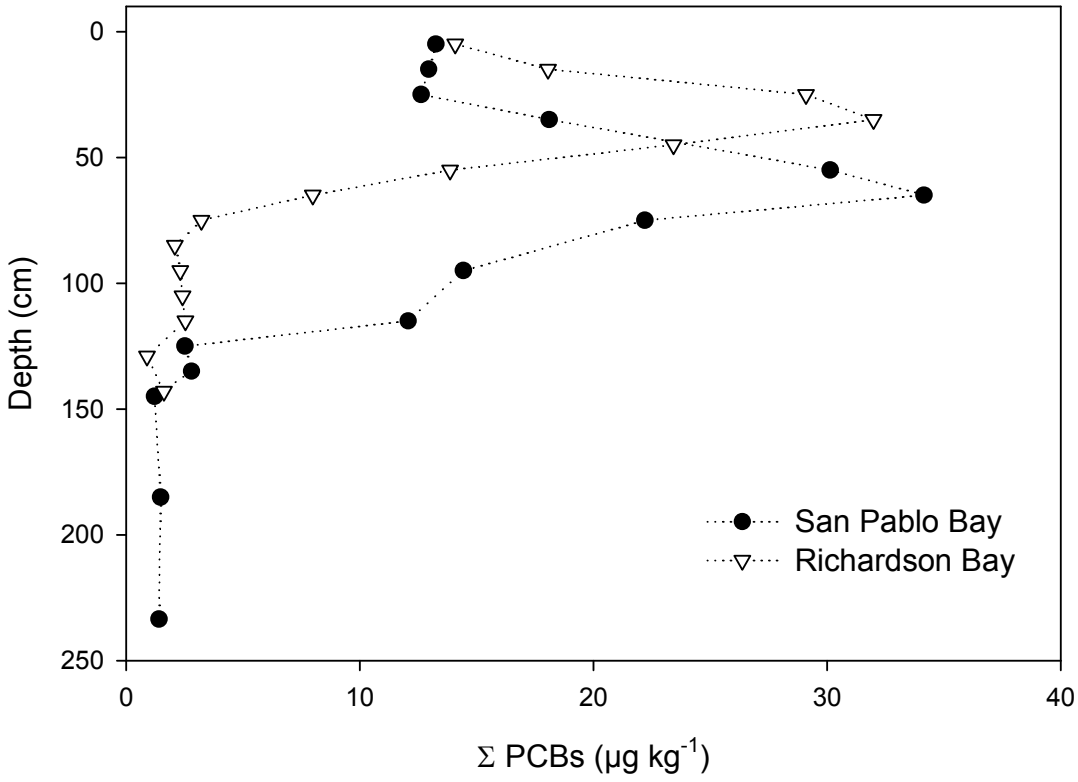

Figure 10. Total PCBs in sediment cores. Sediment cores were collected from San Pablo Bay and Richardson Bay by USGS in 1990 and 1992, respectively. Data from Venkatesan et al. (1999).

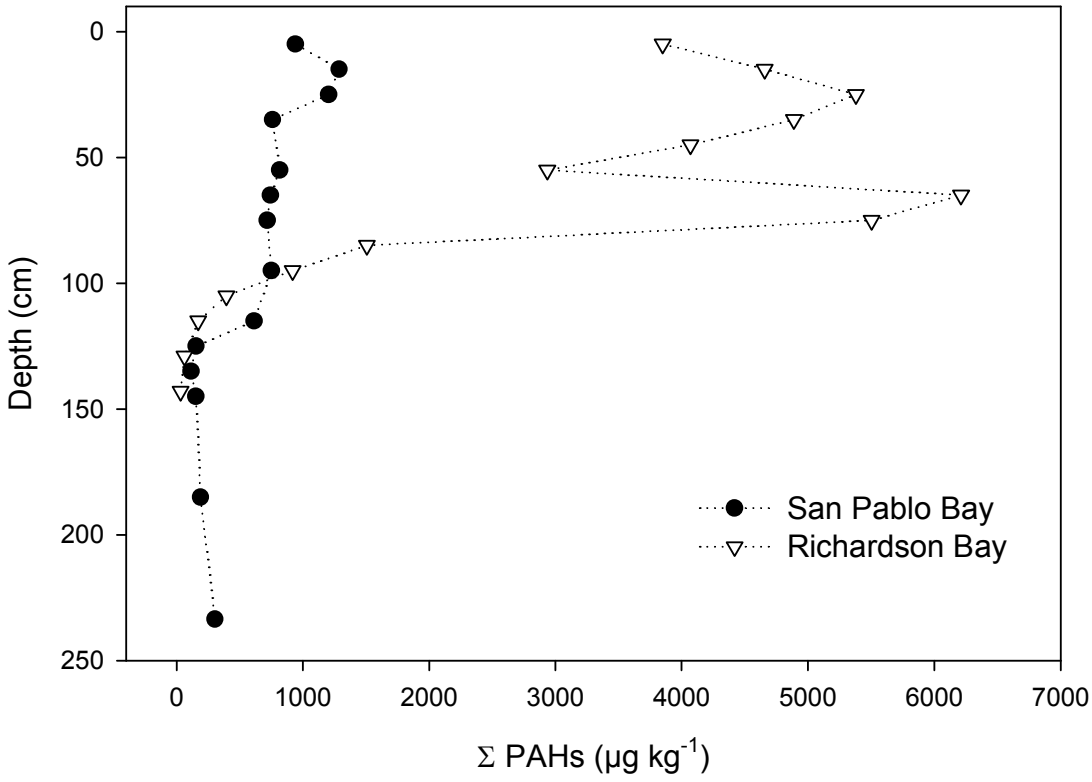

Figure 11. Total PAHs in sediment cores. Bulk sediment cores were collected from San Pablo Bay and Richardson Bay by USGS in 1990 and 1992. Data from Pereira et al. (1999).

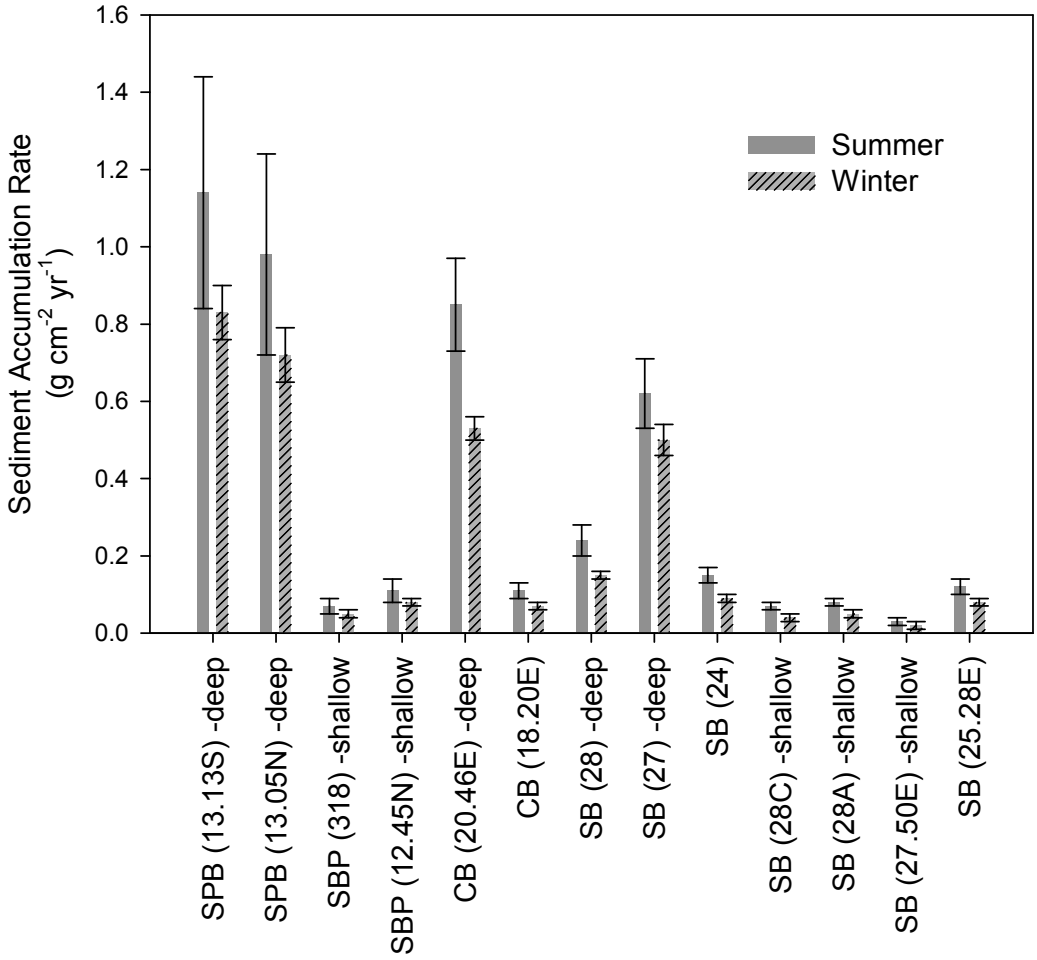

Figure 12. Sediment accumulation rates in San Francisco Bay. Sediment cores were collected from San Pablo Bay (SPB), Central Bay (CB), and South Bay (SB). Several stations were described as deep-water or shallow-water cores. Station codes are listed next to regional designations. Error bars represent standard deviations. Data from Fuller (1982).

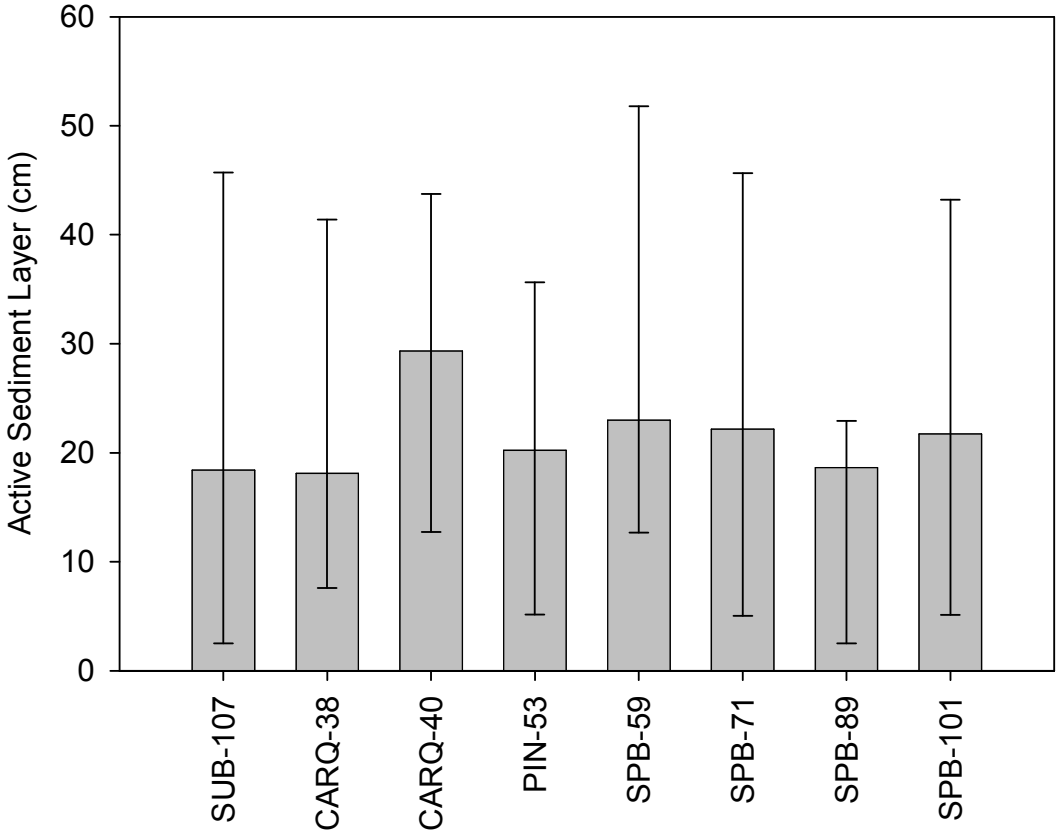

Figure 13. Depth of active sediment layer in sediment cores collected in northern reach of San Francisco Estuary from March to December 1974. Sediment samples collected from Suisun Bay (SUB), Carquinez Strait (CARQ), Pinole Shoals (PIN), and San Pablo Bay flats (SPB). Station codes are listed next to regional designations. Error bars represent minimum and maximum depths of actively mixed sediment layer. Data from Leahy et al., (1976).

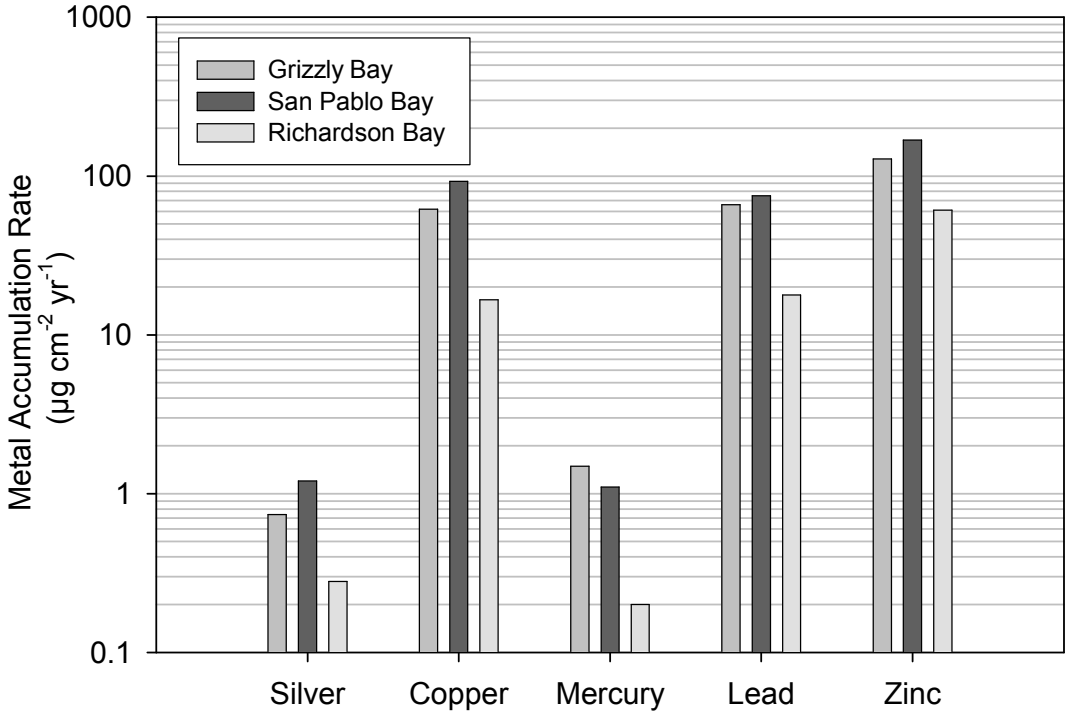

Figure 14. Estimated metal accumulation rates in San Francisco Bay. Estimates from Hornberger et al. (1999).

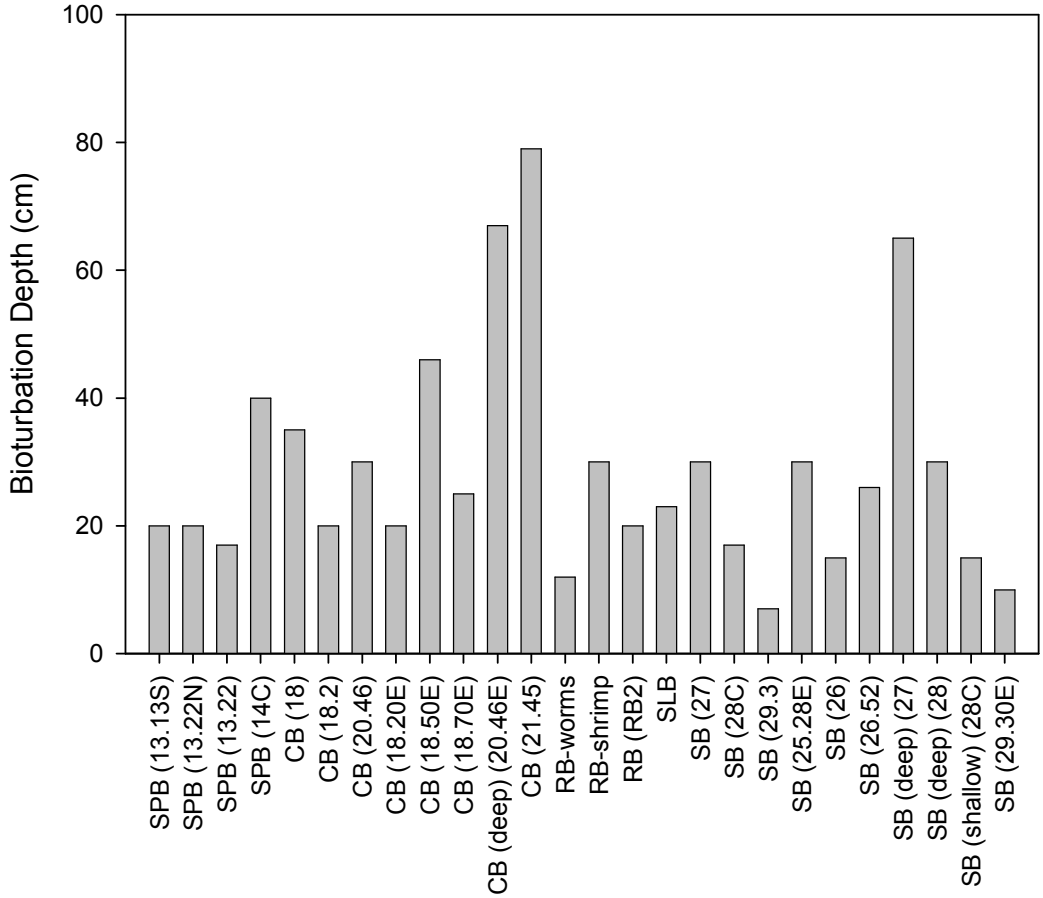

Figure 15. Depth of bioturbation in sediment cores of San Francisco Bay. Sediment cores collected from San Pablo Bay (SPB), Central Bay (CB), Richardson Bay (RB), and South Bay (SB). Station codes are in paranetheses. Stations 20.46E, 27, 28, and 28 C are described as deep-water or shallow-water cores. Data from Korosec (1979), Fuller (1982), Nolan and Fuller (1986), and Fuller et al. (1999).

Table 1. Contaminant concentrations in sediment cores collected in San Francisco Bay. Data include maximum concentrations (C_{MAX}) in the core, surface concentrations (C_{SURF}) and ratios of maximum to surface concentrations (C_{MAX}/C_{SURF}).

Contaminant	Location	C_{Max}	C_{SURF}	C_{MAX}/C_{SURF}	C_{MAX} Depth (cm) unless otherwise noted	Source
Mercury	Stege Marsh, Richmond	430	0.93	**462**	2.5 ft	URS, 2000a
	South San Francisco	6.2	0.25	**25**	2 - 3 ft	URS, 2000b
	San Leandro Bay	11	0.80	**14**	1 - 2 ft	Daum et al., 2000
	Grizzly Bay	1.0	0.31	**3.1**	55 - 56	Hornberger et al., 1999
	Lower South Bay	~ 1.2	~ 0.46	**2.6**	~ 65 - 70	Conaway et al., 2003b
	Treasure Island	0.91	0.36	**2.5**	305	US Navy, 1987
	San Pablo Bay	0.70	0.35	**2.0**	115 - 116	Hornberger et al., 1999
	Richmond Harbor (6/74)	0.65	0.38	**1.7**	152 - 229	San Francisco, 1981
	Hunters Point	0.65	0.41	**1.6**	152	US Navy, 1987
	Richardson Bay	0.44	0.29	**1.5**	66 - 67	Hornberger et al., 1999
	Richmond Harbor (3-4/74)	0.60	0.55	**1.1**	76 - 152	San Francisco, 1981
	Port of Richmond	7.5	7.5	**1.0**	0 - 18	Hart Crowser, Inc. 1993
PCBs	San Leandro Bay	2,660	35	**76**	2 - 3 ft	Daum et al., 2000
	Stege Marsh, Richmond	1,600,000	23,000	**70**	2 ft.	URS, 2000a
	San Pablo Bay	34	13	**2.6**	60 - 70	Venkatesan et al., 1999
	Richardson Bay	32	14	**2.3**	30 - 40	Venkatesan et al., 1999
	Port of Richmond	7,200	7,200	**1.0**	0 - 18	Hart Crowser, Inc. 1993
PAHs	San Leandro Bay	12,592	2,634	**4.8**	2 - 3 ft.	Daum et al., 2000
	Hunters Point	2,301	533	**4.3**	259	US Navy, 1987
	Treasure Island	3,588	1,101	**3.3**	91	US Navy, 1987
	Richardson Bay	6,212	3,850	**1.6**	60 - 70	Pereira et al., 1999
	San Pablo Bay	1,287	942	**1.4**	10 - 20	Pereira et al., 1999
DDT	San Leandro Bay	1,040	135	**7.7**	0 - 1 ft	Daum et al., 2000
	San Pablo Bay	57	7.8	**7.3**	90 - 100	Venkatesan et al., 1999
	Stege Marsh, Richmond	1,600,000	300,000	**5.3**	5 ft.	URS, 2000a (p,p'-DDD)
	Richardson Bay	17	5.4	**3.2**	20 - 30	Venkatesan et al., 1999
Chlordane	San Leandro Bay	293	91	**3.2**	0 - 1 ft	Daum et al., 2000
	Richardson Bay	1.0	0.36	**2.7**	20 - 30	Venkatesan et al., 1999
	San Pablo Bay	1.6	0.63	**2.5**	30 - 40	Venkatesan et al., 1999
	Treasure Island	302	131	**2.3**	274	US Navy, 1987

Table 2. Assimilation efficiencies (%) for mercury (Hg) and methylmercury (MeHg) in *Nereis succinea* and *Mytilus edulis*. Data for *Nereis succinea* from Wang et al. (1998); Data from *Mytilus Edulis* from Gagnon and Fisher (1997).

Species	Location	Hg	MeHg	Sediment Characteristics
Nereis succinea	Flax Pond salt marsh, NY	22-28	66-71	oxic, <500 µm
		17-19	70-73	anoxic, < 500 µm
		7-15	75-84	oxic, <63 µm
		8-14	70-76	oxic, 63-500 µm
	Verrazano Bridge, NY	21-31	43-49	oxic, < 500 µm
Mytilus edulis	Flax Pond salt marsh, NY	9 ± 6	27 ± 8	hydrous ferric oxide
		7 ± 6	87 ± 1	hydrous ferric oxide w/fulvic-acid
		8 ± 7	32 ± 6	hydrous manganese oxide
		3 ± 2	36 ± 19	hydrous manganese oxide w/fulvic-acid
		5 ± 2	5 ± 4	clay
		1 ± 2	49 ± 12	clay w/fulvic-acid
		.	37 ± 6	silica beads
		9 ± 6	81 ± 5	silica beads w/fulvic-acid
		2 ± 2	55 ± 5	natural sediment

Table 3. Assimilation efficiencies (%) of trace elements in bivalves, organic rich/poor sediment. Test sediment was organic-rich (Org) or organic-poor (Poor). Data from Griscom et al. (2000).

		Silver		Cadmium		Selenium		Zinc	
		Org	Poor	Org	Poor	Org	Poor	Org	Poor
Mussel	*M. edulis*	22	8.9	20	9.5	12.5	24	21	28.6
Clam	*M. balthica*	21	18	23	9.7	21.2	27	32	36

Table 4. Assimilation efficiencies (%) of trace elements in bivalves and polychaetes, oxic/anoxic sediment. Test sediment was oxic (Ox) or anoxic (An). Data from Griscom et al. (2000) and Wang et al. (1999).

		Silver		Cadmium		Selenium		Zinc	
		Ox	An	Ox	An	Ox	An	Ox	An
Mussel	*M. edulis1*	12.8	4.6	15.7	35.4	13.6	21.2	21.8	31.6
Polychaete	*N. succinea*	20-27	12	17-26	5-7	52-60	48-56	32-46	24-29
Clam	*M. balthica1*	20	11	21	9	.	.	.	.

Table 5. Biota-sediment accumulation factors (BSAFs) for PAHs, PCBs, and pesticides. Mean values were derived from data compiled by similar K_{OW}, species pooled by similar habitat and feeding mode (HABITAT/FEEDING), and individual species (SPECIES). Data from Tracey and Hansen (1996).

	K_{ow}	HABITAT/FEEDING	SPECIES
PAHs	0.34	0.19	0.29
PCBs	1.03	1.64	2.17
Pesticides	1.36	1.96	2.7

Table 6. Diffusive fluxes of mercury and methylmercury from sediment.

Metal	Location	Flux (ng m^{-2} d^{-1})	Source
MeHg	Lahonton Reservoir, NV	-9.9 - 32	Kuwabara et al., 2002
	Lavaca Bay, TX	0.2 - 1,500	Gill et al., 1999
	Sacramento-San Joaquin River Delta (and tribs)	-10 - 62	Gill, 2001
Hg	Patuxent River, MD	59 - 89	Benoit et al., 1998
	Baltimore Harbor, MD	130 - 169	Benoit et al., 1998
	Lahonton Reservoir, NV	-6,745 - 43,948	Kuwabara et al., 2002
	Lavaca Bay, TX	1.7 - 77	Gill et al., 1999
	Sacramento-San Joaquin River Delta (and tribs)	-25 - 29	Gill, 2001