Sources and Impacts of Chemical Contaminants in San Francisco Bay

A Report for the San Francisco Bay Subtidal Goals Project (Sept. 2008)

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This paper discusses some of the major chemical contaminants in San Francisco Bay, including aspects of their sources, loading and pathways, and their impacts on organisms and human health. Inputs of nutrients into the Bay, considered a pollutant in some contexts, are addressed in the accompanying paper on "Sources, Mechanisms and Impacts of Changes in Nutrient Inputs to San Francisco Bay" (Cohen 2008). Exotic species, which are classified as a biological pollutant under the Clean Water Act and have been confirmed as such by recent federal court decisions, are addressed under the stressor "Release Exotic Organisms."

A wide variety of contaminants have been the subject of regulation, monitoring or research in the Bay. With limited resources, this paper has relied substantially on recent review articles, which were available or more complete for some contaminants than for others. Accordingly, there is less text on some currently important contaminants than on certain contaminants of lesser or declining regulatory significance. Still, the stories of these "declining" contaminants—why they were once of greater concern, and why the perception of their significance has changed—provide important context for our current understanding of contaminants in the Bay.

In a recent review, Davis et al. (2007b) offered an assessment of the current state of contaminants in the Bay. They sorted contaminants and contaminant issues into several categories based on their status and prognosis (Table 1).[1] They classified mercury, polychlorinated biphenyl compounds (PCBs) and dioxins to be the most serious current problems among the chemical contaminants because water quality in all parts of the Bay is impaired by them, because the concentrations of these contaminants in water or biota are well above established levels of concern, and because their abundance and persistence in sediment and their slow rates of depletion mean that they are likely to remain a problem for decades. Other contaminants of substantial concern include

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[1] Organic waste or enrichment, nutrients and exotic species are included as pollutants both in Davis et al. (2007b)'s assessments and on the 303(d) list, and exotic species qualify as biological pollutants under the Clean Water Act, as confirmed by recent federal court decisions. These three types of pollutants are listed in Table 1 and Appendix A, but as noted above are not otherwise treated in this paper on chemical contaminants since they are classified as distinct stressors.

selenium, polycyclic aromatic hydrocarbons (PAHs), and the banned organochlorine pesticides DDT, chlordane and dieldrin.

The significance of some contaminants may also be inferred from their listing as causes of impairment for various portions of the tidal waters of San Francisco Bay, in the most recent Clean Water Act 303(d) list (see Appendix A), and by recent regulatory activities (Table 2, modified from Mumley 2007).

Table 1. **Current status and prognosis for main San Francisco Bay contaminant issues.** Based on Davis et al. (2007b).

Contaminant Issue	Status	Prognosis
Mercury, Exotic Species	Biggest impacts	Further deterioration is likely unless effective management actions can be implemented
PCBs, Dioxins	Biggest impacts	Trend is toward slow reduction, but unlikely to fall below risk thresholds within 20 years
Selenium, PAHs	Significant threats remain	Trend is unclear, could fall below risk thresholds within 20 years
Legacy Pesticides (DDTs, Chlordane, Dieldrin)	Significant threats remain	Trend is toward steady reduction, likely to fall below risk thresholds within 20 years
Organic Waste, Nutrients, Silver	Problems largely solved	Trend is toward further recovery
Nickel, Copper	Problems largely solved	Will be carefully watched
PBDEs, Pyrethroids, Sediment Toxicity, Pollutant Mixtures	Emerging issues	Concerns growing due to increases in inputs or knowledge

Table 2. **Some recent regulatory activities regarding contaminants in San Francisco Bay.** Modified from Mumley (2007).

Contaminant	Regulatory Activity
Copper	Removed from 303(d) list in 2002, site-specific objectives adopted for South Bay in 2002 and for rest of Bay in June 2007.
Cyanide	Site-specific objectives adopted in December 2006.
Diazinon	Removed from 303(d) list in 2006.
Dioxins/Furans	On 303(d) list, TMDL project plan being developed.
DDTs, Chlordane, Dieldrin	On 303(d) list, TMDL project plan being developed.
Mercury	On 303(d) list, initial TMDL in 2004, revised TMDL and site-specific objectives in 2006.
Nickel	In South S.F. Bay, removed from 303(d) list and site-specific objectives adopted in 2002. Attained California Toxics Rule objectives in rest of Bay, Water Board to consider delisting in early 2008.
PCBs	Water Board to consider adopting a TMDL by end of 2007.
Selenium	TMDL project started in 2007, Water Board consideration in 2008/2009.

Background

By the late 19th and early 20th centuries, observers were commenting on the polluted state of parts of San Francisco Bay. Untreated domestic and industrial wastes led to contamination of shoreline areas by fecal bacteria and created anaerobic conditions near sewage outfalls and in the waters at the southern end of the Bay (Krieger et al. 2007), which promoted the growth of bacteria that caused avian botulism and cholera. Oily discharges were also common from the many refineries on the Bay shore, starting with the construction of the Union Oil Refinery in 1896. In the 1940s, synthetic organic pesticides were developed and applied to Central Valley farms and began washing into the Bay (Davis et al. 1991). Even as cities took the first steps toward cleaning up their waste streams with primary treatment in the late 1940s and 1950s (Davis et al. 1991; Buck et al. 2007; Krieger et al. 2007; Table 3), continued urbanization and population growth increased the volume of waste discharges and added further pollutants in urban runoff. In these decades, San Francisco Bay suffered from periodic incidents of oxygen depletion, which were accompanied by foul (hydrogen-sulfide) odors and sometimes by fish kills (e.g. Nichols 1979; Luoma and Cloern 1982; Cloern and Oremland 1983; Krieger et al. 2007).

Table 3. Some of the early primary treatment plants that handled wastewater discharged to San Francisco Bay. From Krieger et al. (2007).

Year	Community or Agency
1934	Palo Atlo
1938	Petaluma
1948	Central Contra Costa Sanitary District
1950	Ora Loma Sanitary District
1951	San Francisco (North Point Treatment Plant) East Bay Municipal Utility District Mountain View
1952	San Francisco (Southeast Treatment Plant)
1954	Hayward
1956	San Jose/Santa Clara Sunnyvale
1957	Los Altos

Things began to change in the late 1960s and 1970s with the passage of state and federal water pollution laws, rising public concern about the state of the environment, and other developments (Table 4). The federal Clean Water Act provided over a billion dollars to upgrade Bay Area wastewater plants; the federal government banned the manufacture and use of PCBs, DDT, dieldrin and chlordane; many Bay Area military bases and industrial plants have closed; and a great deal of regulatory attention, research, and public agency and industry effort have been applied to reducing

contaminant discharges. As a result, several water quality problems that were of great concern in the 1970s have largely been resolved (Mumley 2007; Davis et al. 2007b). With the construction of secondary treatment facilities for municipal wastewater in the 1970s and 1980s, the discharge of suspended solids and biological oxygen demand (BOD) in wastewater effluent dropped sharply (Davis et al. 1991; Krieger et al. 2007), and hypoxic events have become rare in San Francisco Bay (Nichols et al. 1986; Cloern et al. 2003) The concentrations of several important contaminants in effluents, Bay water or sediments have also declined substantially, including a more than 95% reduction in total trace metal loadings into the Bay from municipal wastewater plants, from 943 tons/year in 1960 to 46 tons/year in 1999 (Davis et al. 1991; Van Geen and Luoma 1999; Squire et al. 2002; Davis et al. 2007b; Krieger et al. 2007). Copper and the organophosphate pesticide diazinon, which were listed as causes of impairment of the Bay's water quality in the 1990s, have been delisted;[2] nickel has been delisted for southern San Francisco Bay and delisting is under consideration for other segments.

Despite notable progress in understanding and managing many important contaminants in the Bay, there are reasons to be concerned for the future. Freshwater diversions have reduced the Bay's flushing capacity by about 30%, and the regional consequences of global climate change may reduce it further (Flegal et al. 2007). Large-scale tidal marsh restoration that is now underway may alter aspects of biogeochemical cycling, which could potentially include making some important contaminants more available to the biota (Davis et al. 2007b). Other concerns include an unexplained pattern of sediment toxicity in the Bay, the potential for increased erosion of Bay sediments to expose legacy contaminants, a dearth of information about possible synergistic effects between contaminants, and the problem of emerging contaminants. These issues are discussed briefly below, followed by discussions of a few of the current contaminants of concern in the Bay (mercury, PCBs, DDT, chlordane, dieldrin and PAHs), and two contaminants for which the level of concern has subsided (copper and nickel).

Sediment Toxicity

Sediment toxicity has been an unresolved puzzle in the Bay since it was first documented in the mid-1980s (Anderson et al. 2007). Different studies have found evidence of toxicity in Bay sediments, sediment elutriates, and sediment pore waters, using a variety of tests involving amphipods and the embryos of mussels and sea urchins (Anderson et al. 2007). Initial screenings at 127 sites by the Bay Protection and Toxic Cleanup Program (BPTCP) found that 21% of sediment samples were toxic to amphipod and 31% of pore water samples were toxic to sea urchin embryos (Anderson et al. 2007). In every seasonal sampling period since RMP testing began in 1993, at least one-third of the sediment samples were toxic to one or more test species, and in 1997-2001 at least 63% of the sediment samples were toxic to at least one test organism. A few stations at the north and south ends of the Bay were consistently toxic to bivalve embryos, while some but not all of these plus other stations were consistently toxic to amphipods (Anderson et al. 2007).

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[2] Site-specific water quality objectives for copper were adopted in 2007 in segments north of the Dumbarton Bridge (Mumley 2007); see discussion of copper below.

Table 4. Some key events in the management of contaminants in San Francisco Bay. Sources: Leatherbarrow et al. 2006; Buck et al. 2007; Davis et al. 2007a; Yee et al. 2007; Mumley 2007.

Year	Event
1934	A primary wastewater treatment plant was constructed at Palo Alto, probably the first in the Bay.
1948	The first Federal Water Pollution Control Act authorized the Surgeon General to prepare plans to reduce pollution.
1949	Dickey Water Pollution Act created the state and regional water boards.
1950s	Construction of primary wastewater treatment plants by most communities discharging into the Bay.
1960s	Construction of the first secondary wastewater treatment plants in the Bay.
1969	Porter-Cologne Water Quality Control Act.
1972	Clean Water Act (also known as the Federal Water Pollution Control Act) started a discharge permit system, required secondary treatment for municipal discharges, and provided grant funding to defray the costs of building and operating treatment plants. Federal ban on DDT.
1975	The first complete S.F. Bay Basin Plan designated the beneficial uses of the Bay; primarily addressed "conventional" pollutants (suspended solids, biological oxygen demand (BOD) and bacteria); and included a narrative water quality objective for toxic contaminants.
1979	Federal ban on sale and production of PCBs.
1986	S.F. Bay Basin Plan included water quality objectives and effluent limits for some metals in some parts of the Bay.
1988	Federal ban on chlordane.
1989	Bay Protection and Toxic Cleanup Program established. Federal ban on dieldrin.
1992	Regional Monitoring Program established.
1994	An interim fish consumption advisory was issued for all of San Francisco Bay, based primarily on high PCB and mercury concentrations in fish.
1995	The S.F. Bay Basin Plan's narrative water quality objective was amended to specify that wildlife and human health be protected against the bioaccumulation of toxic contaminants.
1996	The Bay was listed under Clean Water Act §303(d) as impaired by copper, mercury, nickel and selenium (modifying earlier listings of impairment by metals).
1998	The Bay was listed as impaired (under §303(d)) because of a Bay-wide fish consumption advisory based on elevated tissue concentrations of PCBs, dioxins, furans, chlordane, DDT and dieldrin; and listed as impaired by the organophosphate pesticide diazinon due to episodic toxicity in the Bay after storm runoff.
2000	The California Toxics Rule was adopted by the US EPA, which set water quality objectives for the Bay based on federal criteria, including a numerical objective for copper.
2002	Copper was removed from the Bay's 303(d) list, and nickel removed from the list for southern San Francisco Bay.
2006	Diazinon was removed from the Bay's 303(d) list. A revised TMDL for mercury was completed.
2007	A selenium TMDL project was started.

In the RMP samples, spatial patterns of toxicity to sediment and sediment elutriates did not match. The magnitude and frequency of sediment toxicity was greater during the winter wet season, and toxic sites were associated with Delta inflow in the northern Estuary and with urban creeks, suggesting that the toxicity may be derived from storm water inflows (Anderson et al. 2007). While sediment toxicity has been correlated to some degree with some trace metals, PAHs and organochlorine pesticides, the contaminant or contaminants causing the toxicity have not been identified. Since sediment quality objectives are now being developed for California (Anderson et al. 2007), these observations of unexplained sediment toxicity may end up driving regulatory decisions (Davis et al. 2007b).

Legacy Contaminants and Increased Erosion of Bay Sediments

"Legacy contaminants," whose concentrations in the Bay are due more to historic than to current inputs, include several trace metals (mercury, copper, nickel, silver) as well as some organochlorine compounds (PCBs, DDTs, chlordanes and dieldrin) that have not been commercially available for use in the Bay watershed for decades, but are still present at high enough levels to contribute to official warnings about the consumption of certain fish (Flegal et al. 2007; Davis et. al 2007a; Connor et al. 2007).

Bay sediments can be divided conceptually into two categories: active sediments that are at or near the surface and exchange with the water column by diffusion or by physical or biological mixing, and buried sediments that lie below the active layer and are isolated from the water column and organisms (Davis et. al 2007a). The depth of the active layer varies from place to place, and the general range is suggested by Bay organisms that turn over or irrigate sediments to depths of up to a few centimeters (snails, sea slugs, juvenile clams), up to 10-30 cm (lugworms, deeper burrowing clams), or up to 50-75 cm (bat rays, various polychaetes, and ghost shrimp) (Rubin and McCulloch 1979; Peterson 1979; Haderlie and Abbott 1980; Haig and Abbott 1980), and physical turnover due to wind waves or tidal currents that ranges from 2-5 cm depth in mud-clay sediments in Central and South Bays, to 40-100 cm depth in sand in the western part of the Central Bay (Rubin and McCulloch 1979; Hammond and Fuller 1979).[3]

Sediment flows to the Bay appear to be declining due to reduced water flows and the impoundment and retention of sediments in reservoirs (McKee et al. 2006; Jaffe et al. 2007), and recent assessments of bathymetric data suggest that the Bay as whole may now be losing sediment (Jaffe et al. 1998; Cappiella et al. 1999; Foxgrover et al. 2004; Schoellhamer et al. 2005). Planned floodplain and wetland restoration projects are likely to trap sediment and further reduce the sediment supply to the Bay (Davis et. al 2007a). All this suggests an increased and increasing rate of erosion of Bay bottom sediments, which can expose legacy contaminants in the buried layer that have been effectively isolated from the Bay. It is important to note, however, that the phenomenon of eroding bottom sediments is not new in the Bay; bathymetric analyses show that even in Bay

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[3] See the accompanying paper, "Impacts of the Removal or Disturbance of Sediments, Shells or Bedrock in San Francisco Bay", for further description of the turnover of Bay bottom sediments.

segments that are accumulating large amounts of sediment, substantial erosion may be occurring over parts of those segments during the same period (e.g. Cappiella et al. 1999).

Synergisms

Every Bay organism is exposed over its lifetime to hundreds of chemicals at various concentrations and in different, dynamically shifting combinations. However, almost nothing is known about the potential effects of such multi-chemical exposures, since nearly all studies of contaminant impacts on aquatic organisms have focused on one chemical at a time (Davis et al. 2007b). Especially where a contaminant is present in concentrations that are near threshold effect levels, there is concern that it could work in combination with other stresses (including other contaminants) to impair sensitive life-history processes such as reproduction, development, sexual differentiation, etc. (Davis et. al 2007a). The subtlety and potential complexity of such interactions means that the impacts of some contaminants could, in combination with others, be substantially greater than has been recognized. Currently unexplained phenomena — the widespread sediment toxicity in the Bay, pelagic organism decline, the relative scarcity of native oysters in the Bay — could possibly be the result of synergisms between contaminants (Davis et al. 2007b).

Emerging Contaminants

There are more than 7 million chemical compounds that are commercially available in the U.S. (Hoenicke et al. 2007), and the potential effects in the aquatic environment of most of these are largely or entirely unknown. Recent analyses have detected a wide variety of anthropogenic compounds in the water, sediments or tissue samples from San Francisco Bay, including flame retardants, pesticides and insecticide synergists, insect repellents, pharmaceuticals, ingredients of personal care products, plasticizers and non-ionic surfactants (Hoenicke et al 2007). Two groups of compounds that have recently been garnering attention are PBDEs and pyrethroid insecticides.

Polybrominated diphenyl ethers (PBDEs) are a type of flame retardant that has become very common in commercial goods since the early 1990s. PBDE concentrations in Bay Area wildlife and humans are among the highest reported in the world, and there is growing laboratory evidence of toxic effects from these chemicals. California banned the use of two types of PBDEs in 2006, and the US EPA is expected to establish a threshold for concern for PBDEs soon (Davis et al. 2007b).

As organophosphate pesticides have been phased out, the use of pyrethroid insecticides has increased in agriculture and for pest control around homes. Fish and aquatic invertebrates are sensitive to pyrethroids. They have often been linked to sediment toxicity and are suspected as a factor in pelagic organism decline (Davis et al. 2007b).

Mercury

Davis et al. (2007b) described mercury as "Bay water quality enemy number one." Recent studies have concluded that elevated mercury concentrations in fish tissue in the Bay pose a human health risk (Flegal et al. 2007), and this is a primary reason for the Bay-wide fish consumption advisory. Mercury may also be harming wildlife populations, including the endangered California clapper rail (Schwarzbach et al. 2006; Davis et al. 2007b).

Mercury contamination in the Bay is mainly derived from historic mercury and gold mining (Flegal et al. 2007). An estimated 12,000 metric tons of mercury was used in California, mainly in the Sierra Nevada, to separate gold from ore, with about 40% of this lost to the environment by placer mining operations (Conaway et al. 2007). Although Davis et al. (2006) reported that there has been no general decline in mercury concentrations in Estuary fish over the past 30 years, Conaway et al. (2007) found that RMP data from 1993-2001 showed a modest decrease in surface sediment concentrations of total mercury, which they attributed to burial of older, more contaminated sediments by relatively cleaner recent sediment the Central Valley watershed.

Mercury is present in the Bay in multiple chemical forms. Typically only about one percent of total mercury is present as methylmercury, but since methylmercury is biomagnified through the food web and is a neurotoxin that is especially harmful to early stages of human and animal development, it is the form of mercury in the Bay that is of greatest concern (Conaway et al. 2003; Davis et al. 2007b; Flegal et al. 2007).

Methylmercury concentrations often change substantially over short distances and short times, and do not correlate closely with concentrations of total mercury (Davis et al. 2007b; Flegal et al. 2007). The factors that control methylmercury concentrations in the Bay are not well understood (Davis et al. 2007b). Since wetlands appear to be sites of methylmercury production, the ongoing restoration of Bay wetlands raises concerns(Davis et al. 2006; Greenfield et al. 2006; Davis et al. 2007b). On the other hand, some recent studies suggest that some wetlands can trap methylmercury and render it unavailable for biotic uptake (Davis et al. 2007b).

Several studies have found elevated levels of mercury in Bay biota. Greenfield et al. (2006) reported that 40% of small fish sampled, representing seven species from eight locations in the Bay, had mercury concentrations that were higher than the 0.03 µg/g (wet-weight) TMDL target for prey fish tissues.

Most of the mercury in the Bay is bound to sediment particles and is distributed so widely in the Bay and its watershed that it will take many decades for the Bay's total mercury concentrations to decline significantly (Davis et al. 2007b). Any more rapid improvement in the status of mercury in the Bay will depend on identifying and implementing effective management actions to control methylmercury (Davis et al. 2007b).

PCBs

PCBs bind to sediment particles and are highly persistent in the environment. In the Bay they are distributed primarily around the shallow margins of the shore, associated primarily with industrial areas and with the mouths of creeks draining industrial areas (Davis et. al 2007a; Davis et al. 2007b). They bioaccumulate and can suppress growth, cause developmental abnormalities, disrupt endocrine pathways, harm immune systems and cause cancer in humans and in wildlife at the top of the food chain. PCBs may also harm young fish (Davis et al. 2007b).

The commercial use of PCBs was phased out during the 1970s, and the federal government banned their sale and production in 1979. Since then there has been a gradual decline in PCB concentrations in the Bay (Davis et. al 2007a). However, 25 years later, concentrations in some Bay sport fish are still more than 10 times the thresholds of concern for human health.

PCBs are, along with mercury, a primary cause for the Bay-wide fish consumption advisory and for classifying the Bay as an impaired water body. To meet human health targets and protect Bay wildlife will require a greater than 90% reduction in contamination levels in Bay organisms. This is likely to take many decades since the concentrations of PCBs are currently far above the threshold for concern, they are distributed widely in sediments in the Bay and in the watershed, and concentrations in sediments decline slowly (Davis et. al 2007a; Davis et al. 2007b). Erosion of Bay bottom sediments may also expose older, more contaminated sediments lying underneath (Davis et. al 2007a).

Organochlorine Pesticides

Organochlorine pesticides (which include DDTs, chlordanes and dieldrin) came into use in the 1940s and 1950s as insecticides on farms and for pest control and mosquito abatement in urban areas (Leatherbarrow et al. 2006; Connor et al. 2007). They were used abundantly in California, and by the 1980s, the San Joaquin River had some of the highest concentrations of DDT of any U.S. river system (Gilliom and Clifton, 1990). Human health and environmental concerns led to restrictions on DDT use in California starting in 1963, most agricultural use was banned in the state by 1970, and DDT was banned by the federal government in 1972. Similarly, chlordane came into use in the 1940s to control termites and other insects, but agricultural use was banned in 1975 and in the U.S. in 1978, and using it to protect structures was banned in 1988. Dieldrin began to be used around 1950 for termite protection, for moth-proofing and to protect cotton, corn and citrus crops; most agricultural use was banned in 1974, most other uses in 1985, and use for termite control in 1987 (Leatherbarrow et al. 2006; Connor et al. 2007).

The organochlorine pesticides are neurotoxicants, they can also affect reproductive development, they are classified as probable carcinogens by the US EPA, and they may

also function as endocrine disruptors (Leatherbarrow et al. 2006). They persist in the environment and biomagnify in aquatic biota, accumulating in tissues that are high in lipids. Among fish, shiner surfperch and white croaker, and some white sturgeon, which have high lipid content, tend to have the highest tissue concentrations of organochlorine pesticides. For decades after the use of these pesticides was restricted and banned, concentrations in the Bay's water and fish tissues continued to exceed threshold concentrations of concern for human health (Gunther et al. 1999; Greenfield et al. 2002; Leatherbarrow et al. 2006). They continued to arrive in the Bay in flows from the Central Valley, some local tributaries and storm drains—presumably derived from deposits in soils—along with small amounts in atmospheric deposition and very minor quantities in wastewater discharges (Bergamaschi et al. 2001; McKee et al. 2004; Connor et al. 2007). Inflow of DDT to the Bay is estimated to total about 60 kg/yr, with about 71% coming from local watersheds and about 27% coming from the Central Valley. A substantial amount (9 kg) is also estimated to erode out of burial in Bay sediments each year. Inflow of chlordane is estimated at 30 kg/yr, with over 90% coming from local watersheds, and of dieldrin at 10 kg/yr, with 55% from the Central Valley and 33% from local watersheds (Connor et al. 2007). These pesticides can also disperse out of historic "hot spots" including a former pesticide packaging plant on the Lauritzen Canal in Richmond Harbor (Leatherbarrow et al. 2006). They are only weakly soluble in water, and most of the biologically accessible organochlorine pesticide in the Bay (≈97-99%) is in the upper, active layer of sediment rather than the water column (Connor et al. 2007). The presence of elevated concentrations of organochlorine pesticides in Bay fish tissues was one of the reasons for issuing a fish consumption advisory in 1994, and they are included as a cause of Bay-wide impairment in the Bay's current 303(d) list (Leatherbarrow et al. 2006; Davis et al. 2007b).

However, concentrations of these pesticides have continued to decline in sediment cores, bivalves and sport fish (Leatherbarrow et al. 2006). In recent sampling, very few sport fish samples exceeded concern thresholds for these pesticides, and as their concentrations are continuing to fall it is likely that within 20 years no sport fish tissue will be above thresholds (Davis et al. 2007b). The decline in bioavailable organochlorine pesticides in the Bay are mainly a result degradation of the compounds in the sediments, followed by ouflow in tidal currents through the Golden Gate and volatilization.

Polycyclic Aromatic Hydrocarbons (PAHs)

The concentrations of PAH in Bay water and sediments have remained relatively constant over the past 20 years (Oros et al. 2007; Davis et al. 2007b). The concentrations in Bay sediments may pose a risk to early life stages of fish (Davis et al. 2007b), and PAH concentration thresholds that have been recommended by NOAA to protect estuarine fish are frequently exceeded (Oros et al. 2007). PAHs are included on the 303(d) list as causing impairment at several Bay locations (Davis et al. 2007b).

In urban areas, PAHs are emitted into the atmosphere by a variety of processes including vehicle emissions and the burning of heating oils, wood and other biomass. From the air, PAHs can enter the water column by gaseous exchange across the air–water interface, by dry deposition of particulate matter, or by wet deposition in rainfall, and may enter the Bay directly or by runoff from streets and storm drains. Unburned fossil fuels can introduce PAHs into the Bay via street runoff or spills (Oros et al. 2007). The current maximum annual loading into the Bay is estimated at 10,700 kg/year, from the following sources: storm water runoff (51%), inflow from tributaries (28%), wastewater plant discharges (10%), atmospheric deposition (8%) and the disposal of dredge material (2%).

Concentrations of PAHs in Bay waters are highest in the Lower South Bay (120 ng/L), followed by the South Bay (49 ng/L), San Pablo and Suisun bays (29 ng/L), and lowest in the Central Bay (12 ng/L) (Ross and Oros 2004; Oros et al. 2007). Higher concentrations in the South Bay and especially the Lower South Bay may be due to a higher density of inputs from storm water discharge and atmospheric deposition, and perhaps longer residence times (Oros et al. 2007).

Although atmospheric concentrations of PAHs have declined in the Bay Area over the past ten years, increased motor vehicle use in the future could raise PAH levels in the Bay (Davis et al. 2007b). If, however, PAH emissions and inputs to the Bay can continue to be reduced, then concentrations in the Bay should drop relatively quickly (Davis et al. 2007b).

Copper

Copper is a micronutrient needed for phytoplankton growth, but when free copper ions are present in high concentrations they block the uptake of other micronutrients (manganese and zinc) (Buck et al. 2007). At free ion concentrations above 10^{-11} M (≈0.0006 µg/L), copper can be toxic to aquatic microorganisms (Brand et al. 1986; Buck et al. 2007). Copper in water has typically been measured in three categories, as *total dissolved copper*, as *exchangeable copper*, which is the copper associated with suspended sediments that could leach into the dissolved phase, and as *total dissolvable copper*, the sum of dissolved copper and exchangeable copper. In practice, dissolved copper is measured in a water sample that has been filtered through typically a 0.20- or 0.45-micron filter (over this range, the different filter sizes produce only minor differences in the results), and exchangeable copper is the amount of copper leached from filtered particles with a weak acid over a specified time (Buck et al. 2007).

Gordon (1980) made the first reliable measurements of dissolved copper concentrations in the Bay, using rigorous trace metal clean techniques (Buck et al. 2007). He measured copper concentrations in the Bay ranging from 0.4-2.8 µg/L, which was considerably higher than the 0.07-0.4 µg/L range that he measured in coastal waters (Buck et al. 2007). In the portion of the Bay between Suisun Bay and the Central Bay where he took his samples, Gordon (1980) found that copper concentrations were related to salinity

levels during periods of high Delta outflow, but that copper concentrations rose higher relative to salinity during low Delta outflows. Gordon (1980) suggested this could be due to anthropogenic inputs of copper into South Bay waters, which then infiltrated into and raised copper concentrations in the Central and northern Bay when freshwater inflows were low (Buck et al. 2007).

Kuwabara et al. (1989) measured total dissolved copper concentrations in the South Bay that that ranged from 1.8-4.2 µg/L and regularly exceeded 3.1 µg/L (=49 nM), a concentration guideline that was implemented by the US EPA as a national criterion continuous concentration in 1995 (Buck et al. 2007). Flegal et al. (1991) found that copper concentrations in the South Bay exceeded concentrations in the northern Bay when adjusted for salinity (Buck et al. 2007). Data from the Regional Monitoring Program for 1993-2001 showed total dissolved copper concentrations that were highest in the South Bay (2.0 µg/L) and the sloughs at the southern end of the South Bay (2.4 µg/L), lower in Suisun and San Pablo bays (1.8 µg/L), and lowest in the Central Bay (1.0 µg/L) (Buck et al. 2007). Along with nickel, mercury and selenium, copper was determined to be impairing beneficial uses in the lower South Bay, resulting in this section of the Bay being included on the Clean Water Act 303(d) list of impaired water bodies in 1998 (Yee et al. 2007).

These findings focused attention on the sources of copper in South Bay waters. Loadings from the largest municipal wastewater treatment plants in the area had been 343 kg/day in 1975, but were reduced to 52 kg/day by 1985 (Davis et al. 1991); total wastewater loadings of copper into the South Bay have since been reduced further, to 28-46 kg/day (Buck et al. 2007). The 1993-2001 RMP data, when adjusted for dissolved organic carbon (DOC) and Delta outflow, similarly showed that over this period copper concentrations in the Bay dropped by 44% in South Bay sloughs, by 29% in the South Bay, and by 17% in Suisun and San Pablo bays (Buck et al. 2007). Meanwhile, Gee and Bruland (2002) found that desorption of copper from sediment particles that were resuspended in the water column was probably a larger source of copper in the South Bay than wastewater discharges (Buck et al. 2007).

In the absence of organic material, at typical pH values for seawater, about 7% of total dissolved copper occurs as free copper ions (Buck et al. 2007). At dissolved copper concentrations of 2 µg/L (typical for the South Bay), the concentration of copper ions in the absence of organic material would be around 0.14 µg/L, which is a couple of orders of magnitude above the probable threshold level for toxic effects of about 0.0006 µg/L (10^{-11} nM) (Brand et al. 1986). Since large scale copper toxicity events have not been observed in San Francisco Bay in recent years, a large part of the dissolved copper in the Bay is probably bound up with organic molecules (Buck et al. 2007). This is consistent with Kuwabara et al.'s (1989) finding that dissolved copper concentrations in the South Bay correlated with concentrations of dissolved organic carbon (DOC), and with the 1993-2001 RMP data, in which dissolved copper concentrations correlate positively with DOC and negatively with Delta outflow (Buck et al. 2007)

Studies of copper speciation in San Francisco Bay undertaken between 1994 and 2005 generally confirmed earlier studies in northeast Pacific coastal waters (Coale and Bruland 1988), which had found that >99% of total dissolved copper was tightly bound to organic molecules and unavailable for uptake by biota, and that the concentrations of the potentially toxic forms of copper were not consistently related to the concentrations of total dissolved copper (Buck et al. 2007). In the first of these studies in the Bay, 80-92% of dissolved copper at a South Bay site near the Dumbarton Bridge was bound to organic molecules, with the strongest-binding type of molecules (called L_1) present at concentrations sufficient to bind only 27% of the copper, the rest complexing with weaker binding molecules (L_2) (Donat et al. 1994). However, several studies conducted since 2000 consistently found that over 99.9% of the dissolved copper was bound to the L_1 or strongest-binding molecules, with a large excess of the weaker L_2-type molecules available to "back up" the L_1 molecules (Buck et al. 2007). In addition, it was found that most of the dissolved copper that enters the South Bay in wastewater effluent is probably already strongly bound to organic molecules in the discharge (Sedlak et al. 1997).

Since the analytical methods used in the earlier (1994) and later (2000-2005) studies of copper speciation in the Bay were similar (Buck et al. 2007), it's unclear why the results were different. In any event, organic molecules capable of strongly binding copper are now present in the Bay in sufficient concentrations to keep the concentration of free copper ions below 10^{-13} nM throughout the Bay, well below the probable threshold for aquatic toxicity of around 10^{-11} nM (Buck et al. 2007). The sources of these binding molecules, which have not been structurally characterized, are not known, but strongly binding molecules have been observed in the exudates from phytoplankton, especially cyanobacteria (which a recent studies have found to be flourishing in the South Bay), and weaker binder molecules in wastewater and runoff from soils (Buck et al. 2007).

In summary, copper was considered a major problem in San Francisco Bay when some of the early measurements found that total dissolved copper concentrations regularly approached or exceeded state and federal criteria (Flegal et al. 1991; Flegal et al. 2007; Buck et al. 2007). However, copper toxicity is due to the concentration of copper ions, rather than to total copper or total dissolved copper (Buck et al.2007). Subsequent studies of copper speciation showed that >99.9% of the copper in the Bay is tightly bound in organic molecules and is not readily available for biotic uptake (Flegal et al. 2007; Buck et al. 2007). Copper ion concentrations appear to be well below the threshold levels for aquatic toxicity, and there is a substantial excess of organic molecules that are capable of at least weakly binding copper, which should buffer any further additions of copper (Buck et al. 2007). As a result of these findings, copper was removed from the Bay's 303(d) listing in 2002, and new site-specific water quality objectives for copper were dadopted in June 2007 for Bay segments north of the Dumbarton Bridge (Davis et al. 2007b; Buck et al. 2007; Mumley 2007).

Nickel

The Coast Range rock formations that surround San Francisco Bay include serpentine and ultramafic rocks that contain high concentrations of nickel, and it is likely that the geologic composition of this watershed is the cause of relatively high nickel concentrations in the Bay (Yee et al. 2007). Nickel is also common in some industrial products, and enters the Bay through wastewater discharges and urban runoff (Yee et al. 2007), though the nickel in wastewater discharges may be relatively unavailable for biological uptake (Yee et al. 2007). Certain estuarine species, including some mollusks and crustaceans, are notably sensitive to nickel (Yee et al. 2007).

From measurements made by the RMP in 1993-2003 and the City of San Jose in 1997-2007, the Lower South Bay had the highest concentrations in the water column of both total nickel (13.7 µg/L) and dissolved nickel (4.0 µg/L). Suisun and San Pablo bays had the next highest concentrations (≈8 µg/L of total nickel), and Central Bay had the lowest (2.6 µg/L total and 1.3 µg/L dissolved nickel) (Yee et al. 2007). Concentrations in the sediments generally paralleled this, with higher concentrations in the South Bay, Suisun Bay and San Pablo Bay, and lower concentrations the Central Bay and the Delta — that is, with the higher concentrations in those parts of the system that receive a larger portion of their drainage from the Coast Range (Yee et al. 2007). In the northern part of the Estuary, the concentration of dissolved nickel increases downstream, presumably due to mobilization from the resuspension of these sediments (Yee et al. 2007).

In the mid-1990s, nickel concentrations in the water of the lower South Bay often exceeded water quality objectives; in contrast, concentrations measured in fish and bivalve tissues remained well below the recommended maximum tissue residue levels (Yee et al. 2007). Because of the high concentrations in water, nickel (along with copper, mercury and selenium) was determined to be impairing beneficial uses in the lower South Bay (Yee et al. 2007). As a result, this part of the Bay was included on the Clean Water Act 303(d) list of impaired water bodies in 1998 (Yee et al. 2007).

The primary sources of total nickel to the Bay as a whole are resuspended sediments (estimated at 67%) and Delta inflow (28%); for the lower South Bay, the main sources are resuspended sediments (≈80%), the inflow from local rivers (≈15%) and the effluent from wastewater treatment plants (4%) (Table 5). Total nickel is strongly correlated with suspended sediment concentrations, dissolved nickel less so (Yee et al. 2007). The primary sources of dissolved nickel in the Bay water are desorption from resuspended sediments, benthic diffusive fluxes, and wastewater discharges (Yee et al. 2007). Concentrations of total and dissolved nickel in the northern part of the Bay are higher in the wet season, reflecting the importance of the rivers as a source of nickel in that part of the Bay. Concentrations of dissolved nickel in the South Bay are higher in the dry season, reflecting the relatively greater importance of wastewater effluent, which dominates freshwater inflows in this part of the Bay during the dry season, as a source of dissolved nickel (Yee et al. 2007).

Table 5. Estimated Sources of Nickel in San Francisco Bay water. From Yee et al. (2007), citing Davis et al. (2000) for San Francisco Bay and Tetra Tech (1999) for Lower South Bay.

Source	San Francisco Bay		Lower South Bay	
	kg/yr	% of total	kg/yr	% of total
Net particulate flux	975,000	67%	31,000-34,000	79-80%
Delta inflow	410,000	28%	0	0%
Non-delta tributaries	49,000	3%	6,040	14-15%
Benthic diffusion	21,600	1%	720	2%
Wastewater effluent	4,800	0.3%	1,740	4%
Atmospheric deposition	580	0.04%	30	0.07-0.08%
Total	1,460,980	100%	39,530-42,530	100%

With substantial regulatory focus on trace metals and the implementation of tertiary treatment, the overall trace metal loadings into the Bay from municipal wastewater treatment plants is estimated to have been reduced by >95% since 1960 (Squire et al. 2002). The estimated nickel loading into the lower South Bay from wastewater treatment plants has steadily declined, from around 12,000 kg/year around 1979, to 5,400 kg/year around 1989, to 1,700 kg/year around 1999 (Tetra Tech 1999, cited by Yee et al. 2007). Dissolved nickel is a higher fraction of total nickel in the South Bay than in the Central and northern Bay (Yee et al. 2007). The dissolved nickel delivered to the Bay is sometimes strongly bound up in stable molecules derived from soil or from industrial processes, though the amount of such complexing is highly variable (Yee et al. 2007).

The concentration of nickel in San Francisco Bay sediments (≈90 µg/L) is among the highest reported in U.S. estuaries (Yee et al. 2007). Concentrations did not vary significantly with depth in 2-meter deep sediment cores from the Central Bay and San Pablo Bay, indicating that historic anthropogenic changes have not caused significant changes in overall nickel inputs and confirming that natural occurrence in the watershed is the primary source of high nickel concentrations in the Bay (Hornberger et al. 1999). This is corroborated by the similarly high concentrations of nickel measured in the sediments of Tomales Bay, which has a similar surrounding geology but lacks anthropogenic sources of trace metals (Yee et al. 2007); and also by the lack of a detectable decline in sediment concentrations in the South Bay between the 1970s and 1998 despite dramatic reductions over that period in the total quantity of nickel in the wastewater discharged into the South Bay (≈86% reduction) (Hornberger et al. 2000).

There is evidence from laboratory studies and from observations of a 25% reduction in nickel concentration in the water column during a South Bay phytoplankton bloom (Luoma et al. 1998) that phytoplankton may take up significant quantities of nickel from the Bay, potentially making it available to higher trophic level organisms. Nickel concentrations in bivalve tissues were higher in the South Bay than in San Pablo Bay; however, concentrations in both bivalves and fish throughout the Bay were far below

harmful concentrations for the organisms or human consumers. Three species of bivalves used in RMP bioaccumulation studies in 1993-2003 had tissue concentrations that ranged from 0.9-113 µg/L dry weight (equivalent to about 0.2-28 µg/L wet weight).[4] Two species of fish collected by NOAA in 1984-88 had tissue concentrations that averaged below 1 µg/L dry weight (equivalent to ≈0.15 µg/L wet weight). These are all far below both the recommended maximum tissue residue levels of 220 µg/L wet weight and the concentrations (330-1460 µg/L dry weight) at which reduced survival rates have been reported in bivalves (Yee et al. 2007).

In summary, nickel concentrations in Bay sediments and water are quite high compared to concentrations in most other estuaries. Within the Bay, concentrations are highest in the South Bay, especially the Lower South Bay, followed by San Pablo and Suisun bays, and lowest in the Central Bay. Concern over these concentrations contributed to various sections of the Bay being listed as impaired on the Clean Water Act 303(d) list in the 1990s. The primary source of nickel in the Bay is sediments washed in from the Coast Range, whose rock formations are naturally high in nickel. Other important sources are Delta inflow (low concentrations but high volume), and in the South Bay, other tributaries. Wastewater inflows are a more significant contributor in the lower South Bay, but even there are responsible for only 4% of the total input. Consequently, substantial reductions in nickel inputs in wastewater discharges, on the order of 85%, have not resulted in detectable reductions in concentrations in the Bay. While there is evidence of uptake by phytoplankton, tissue concentrations in bivalves and fish have remained well below levels that would have impacts on these organisms or on people eating them. Accordingly, nickel has been de-listed as a cause of impairment in the South Bay and will be considered for de-listing elsewhere in the Bay (Krieger 2007).

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[4] The highest recorded value of 113 µg/L, from a Suisun Bay sample, is a statistical outlier that probably resulted from sediment contamination of the tissue. Typical concentrations averaged 10 µg/L dry weight (≈2.5 µg/L wet weight) or less in the different segments of the Bay (Yee et al. 2007).

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