Available online at www.sciencedirect.com
ScienceDirect
Environmental Research 105 (2007) 87–100
Environmental Research
www.elsevier.com/locate/envres

The slow recovery of San Francisco Bay from the legacy of organochlorine pesticides

Michael S. Connor[a,*], Jay A. Davis[a], Jon Leatherbarrow[a], Ben K. Greenfield[a], Andrew Gunther[b], Dane Hardin[b], Thomas Mumley[c], John J. Oram[a], Christine Werme[d]

[a]San Francisco Estuary Institute, 7770 Pardee Lane, Oakland, CA 94621, USA
[b]Applied Marine Sciences, 4749 Bennett Dr., Suite L, Livermore, CA 94551, USA
[c]Regional Water Quality Control Board, San Francisco Bay Region, 1515 Clay St., Suite 1400, Oakland, CA 94612, USA
[d]Christine Werme, Independent Consultant, USA

Received 14 September 2005; received in revised form 22 June 2006; accepted 5 July 2006
Available online 23 August 2006

Abstract

The use of organochlorine pesticides, including DDTs, chlordanes, and dieldrin, peaked in San Francisco Bay's watershed 30–40 years ago, yet residues of the pesticides remain high. Known as legacy pesticides for their persistence in the Bay decades after their uses ended, the compounds and their breakdown products occur at concentrations high enough to contribute to advisories against the consumption of sport fish from the Bay. Combined with other data sets, the long-term monitoring data collected by the San Francisco Estuary Regional Monitoring Program (RMP) for trace substances allow us to track recovery of the Bay from these inputs and predict its future improvement.

Legacy pesticides enter the water and sediment of San Francisco Bay from a variety of sources, including runoff from California's Central Valley and local watersheds, municipal and industrial wastewater, atmospheric deposition, erosion of historically contaminated sediment deposits, and dredging and disposal of dredged material. Runoff from small-urbanized tributaries may contribute as much or more to the loads than runoff from the agricultural Central Valley, even though 90 percent of the freshwater flow comes from the Central Valley via the Sacramento and San Joaquin rivers.

The fates of legacy pesticides in San Francisco Bay are controlled by their chemical properties, including their solubilities and partition coefficients. Degradation in the sediments, outflow through the Golden Gate, and volatilization—in that relative order—result in removal of pesticides from the Bay.

A contaminant fate model was used to estimate recovery times of the Bay under various scenarios. For example, under a scenario in which no new legacy pesticides entered the Bay, model predictions suggested that concentrations of pesticides in the water and the active sediment layer would reach risk-reduction goals within one to three decades. Under scenarios of continued inputs to the Bay, recovery time would be considerably longer or not reached at all. Long-term tissue monitoring corroborates model predictions of slow declines in DDT and chlordane concentrations. Field-transplanted bivalve samples indicate declines since 1980, and lipid-weight concentrations of pesticides have declined in fishes, but the declines are slow. The critical management question for the Bay is whether there are feasible management actions that would decrease concentrations in sport fish significantly faster than the existing slow progress that has been observed.

Keywords: DDT; Chlordane; Dieldrin; Fish; Residues

1. Introduction

Organochlorine pesticides, which were used for agriculture, structural pest control, and mosquito abatement, continue to be detected in the fish and sediments of San Francisco Bay decades after their sales were banned.

*Corresponding author. Fax: +1 510 746 7300.
E-mail address: mikec@sfei.org (M.S. Connor).

0013-9351/$ - see front matter © 2006 Elsevier Inc. All rights reserved.
doi:10.1016/j.envres.2006.07.001

Known as legacy pesticides, they include: DDTs—the *o*,*p*′- and *p*,*p*′-isomers of dichlorodiphenyltrichloroethane (DDT) and its breakdown products: dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD); chlordanes—including α-chlordane, γ-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide; and dieldrin—including aldrin and dieldrin.

DDT was used in residential and agricultural applications and for mosquito abatement beginning in the 1940s. It was declared a restricted material in California in 1963, and substantial agricultural uses ended in 1970 (Mischke et al., 1985). The US banned it for all but emergency public health uses in 1972. Until 1971, California did not require reporting of DDT use, so there are no records of application rates. Nationally, more than 500 million kg were sold over a 30-year period.

Beginning in the late 1940s, chlordane was used in residential and agricultural applications to control termites and other insect populations. Agricultural chlordane use was restricted in California in 1975 and throughout the US in 1978. Production and sales of chlordane for structural pest control ended in 1988. Peak annual production was 11 million kg in the 1960s, but use in California remained high until the 1988 ban (Shigenaka, 1990).

In 1950, dieldrin began to be used for a variety of applications, including control of termites and other soil-dwelling insects, as a wood preservative, for moth-proofing clothing and carpets, and as a pesticide on cotton, corn, and citrus crops. Agricultural use of dieldrin was restricted in 1974, and most uses were banned in 1985. Use for underground termite control continued until 1987. Peak annual production of aldrin and dieldrin was 9 million kg in the 1970s.

DDTs, chlordanes, and dieldrin are neurotoxins and classified by the US Environmental Protection Agency (USEPA) as probable human carcinogens. They are persistent in the environment, lipophilic, and subject to biomagnification in aquatic food webs.

This paper examines the current status of legacy pesticides in San Francisco Bay and addresses the questions of how long adverse effects of the pesticides are likely to persist and whether management actions to address those effects are warranted. We first summarize recent monitoring data on the levels of legacy pesticides in the water, sediments, and sport fish in the Bay. These data suggest that concentrations of the pesticides remain at levels high enough to pose some risk to marine life and human health.

We then review existing information on sources of legacy pesticides to the Bay and calculate inputs of legacy pesticides to the water column and surface active sediment layer, which is that part of the sediment that is biologically and chemically connected to the water through burrowing organisms. Sources include runoff from the Central Valley and smaller local watersheds, municipal and industrial wastewater discharges, atmospheric deposition, erosion of deeply buried sediments, and dredging and disposal of deep sediments. These estimates are subject to great uncertainty, but provide some indication of the magnitude of continued inputs to the Bay.

We present results from a contaminant fate model, which was used to examine losses from the water and surface active sediments by transport through the Golden Gate, degradation in the water and sediments, and volatilization to the atmosphere. The model was also used to predict the time periods over which recovery of the Bay may be expected to occur, assuming several predicted levels of continued loading. We compare those projected recovery times to long-term data on concentrations of pesticides in mussels and fish. Finally, we discuss the feasibility of management actions to alleviate adverse effects of legacy pesticides in the Bay.

2. Current conditions in San Francisco Bay

2.1. Water

The San Francisco Estuary Regional Monitoring Program (RMP) has monitored water quality since 1993. Through 2001, monitoring was conducted at 21 sites located throughout the Bay. In 2002, the RMP implemented a new monitoring design, designed to provide greater spatial coverage and include both shallow areas and deep channels. This new design resulted in sampling 33 stations, 28 of which were randomly selected and located within the major hydrographic regions of the estuary: Suisun Bay, San Pablo Bay, Central Bay, South Bay, and Lower South Bay. Additional stations were in the delta of the Sacramento and San Joaquin rivers, upstream from the Lower South Bay, and outside the Golden Gate. To remove variability caused by flushing during major rainstorms, the new design includes sampling only during the dry season.

The concentrations of legacy pesticides in water monitored by the RMP have consistently been much lower than federal and state standards for the protection of aquatic life but have at times exceeded those for the protection of human health. Region-wide, from 1993–2001, the RMP has measured exceedances of state water quality standards in 5–20 percent of DDTs, chlordanes, and dieldrin samples.

There are sufficient water-column data to compare the geographic regions of the Bay (Table 1). Highest concentrations of the pesticides were observed in the northern and southern regions of the Bay. Lowest concentrations were observed in the Central Bay and outside the Golden Gate.

Analysis of the data by year showed some apparent decreases in pesticide concentrations from 1993 through 2001. However, those years also included a transition from predominantly wet years (1995–1998) to dry years (1999–2001), and any apparent trends may be related to weather conditions rather than representing attenuation in sources. Preliminary data from the Guadalupe River

Table 1
Dry season total water column concentrations (pg/L, mean ± SE) by region, from Suisun Bay in the north to Lower South Bay in the south (1993–2003). Data from just outside the Golden Gate are included in the Central Bay

	Suisun Bay	San Pablo Bay	Central Bay	South Bay	Lower South Bay	Range of detection limits
DDTs	657 ± 184 (15)	533 ± 65 (38)	160 ± 13 (36)	162 ± 21 (35)	372 ± 57 (17)	0.67–9.2
Chlordanes	77 ± 17 (13)	108 ± 11(32)	62 ± 7 (32)	69 ± 13 (30)	136 ± 18 (14)	0.50–6.6
Dieldrin	67 ± 10 (16)	47 ± 4 (39)	28 ± 3 (35)	36 ± 4 (34)	59 ± 8 (16)	0.12–12.9

Non-detectable concentrations were treated as zeros. The number of samples is indicated in parenthesis.

suggest that loading of pesticides is greatest during severe storm events (McKee et al., 2004).

2.2. Sediment

Because DDTs, chlordanes, and dieldrin are sparingly soluble in water, most of the current mass of legacy pesticides resides in the sediments rather than the water column. Most of the mass resides in buried sediments in depositional areas. There are few measurements from these deep areas, so estimating the mass of legacy pesticides in those sediments is difficult. We used data from cores taken from depositional areas and deep enough to encompass the period in which legacy pesticides were used (Venkatesan et al., 1999). Cores from two sub-embayments, Richardson Bay and San Pablo Bay, were used to estimate inventories of total DDTs of 557 and 3453 ng/cm^2. Non-DDT pesticide inventories ranged from 1154 to 4069 ng/cm^2 in Richardson and San Pablo bays, respectively, with chlordanes comprising 10–18 percent of total pesticides in the cores and dieldrin comprising approximately 2–3 percent. Extrapolating to the entire area of the Bay (1.1×10^9 m^2) suggests a range of 6000–38,000 kg DDTs, 3400–8300 kg chlordanes, and 380–2500 kg dieldrin.

Surface sediment samples from San Francisco Bay have been collected and analyzed by the RMP (e.g., SFEI, 2002) and the Bay Protection and Toxic Cleanup Program (Hunt et al., 1998). These programs observed higher concentrations of pesticides in the shallower areas at the urbanized edges of the Bay (Fig. 1). Of the total mass of legacy pesticides available to the biota (the water and the active layer of the sediments), 97–99 percent resides in the sediments. There is an estimated 350 kg DDTs, 45 kg chlordanes, and 12 kg dieldrin in the surface layer (0–15 cm).

2.3. Sport fish

San Francisco Bay is listed as impaired by legacy pesticides pursuant to §303(d) of the US Clean Water Act because of an interim fish consumption advisory developed by the California Office of Environmental Health Hazard Assessment (OEHHA) in 1994. The advisory is based on a 1994 study (San Francisco Regional Water Quality Control Board et al., 1995) of fish from 13 locations chosen to represent all areas of the Bay, including areas suspected of low or high contamination and locations known to be popular for sport fishing. Since then, the RMP has monitored contaminants in sport fish from the Bay on 3-year intervals (Davis et al., 1999, 2002; Greenfield et al., 2003, 2005).

The program has sampled jacksmelt (*Atherinopsis californiensis*), shiner surfperch (*Cymatogaster aggregata*), white croaker (*Genyonemus lineatus*), striped bass (*Morone saxatilis*), California halibut (*Paralichthys californicus*), leopard shark (*Triakis semifasciata*), and white sturgeon (*Acipenser transmontanus*). Fish fillets are prepared for analysis using methods commonly used to prepare, cook, and consume each species (SFEI, 2000). For each species, samples are composited for analysis.

Concentrations of DDTs, chlordanes, and dieldrin have been typically highest in fishes with high lipid content, such as shiner surfperch and white croaker. Striped bass, the most frequently taken and consumed sport fish in the Bay (SFEI, 2000), has lower lipid content and lower concentrations of legacy pesticides. In the 2003 sampling, two white sturgeon samples had unusually high lipid concentrations and correspondingly higher pesticide levels (Fig. 2).

Fish concentrations are compared to screening values, which are calculated using USEPA (2000) guidance. OEHHA is currently developing guidance for calculating screening values in California, but there are no "official" values. Using screening values that assume a risk level of one in 100,000 (10^{-5}), consumption of 32 g of fish per day (approximately one meal per week), and cancer slope factors as adopted by the California Toxics Rule (CTR; 40 CFR Part 131), concentrations of pesticides exceed the screening values in some samples. In comparison, PCB screening values are exceeded for almost every sample. These results indicate that the legacy pesticides do adversely affect San Francisco Bay; however, the effects are less than those from other contaminants.

3. Sources of legacy pesticides

There continue to be inputs of legacy pesticides to the water and the surface sediments of the Bay. Sources include runoff from the Central Valley and the local watersheds, municipal and industrial wastewater discharges, atmospheric deposition, erosion of sediments

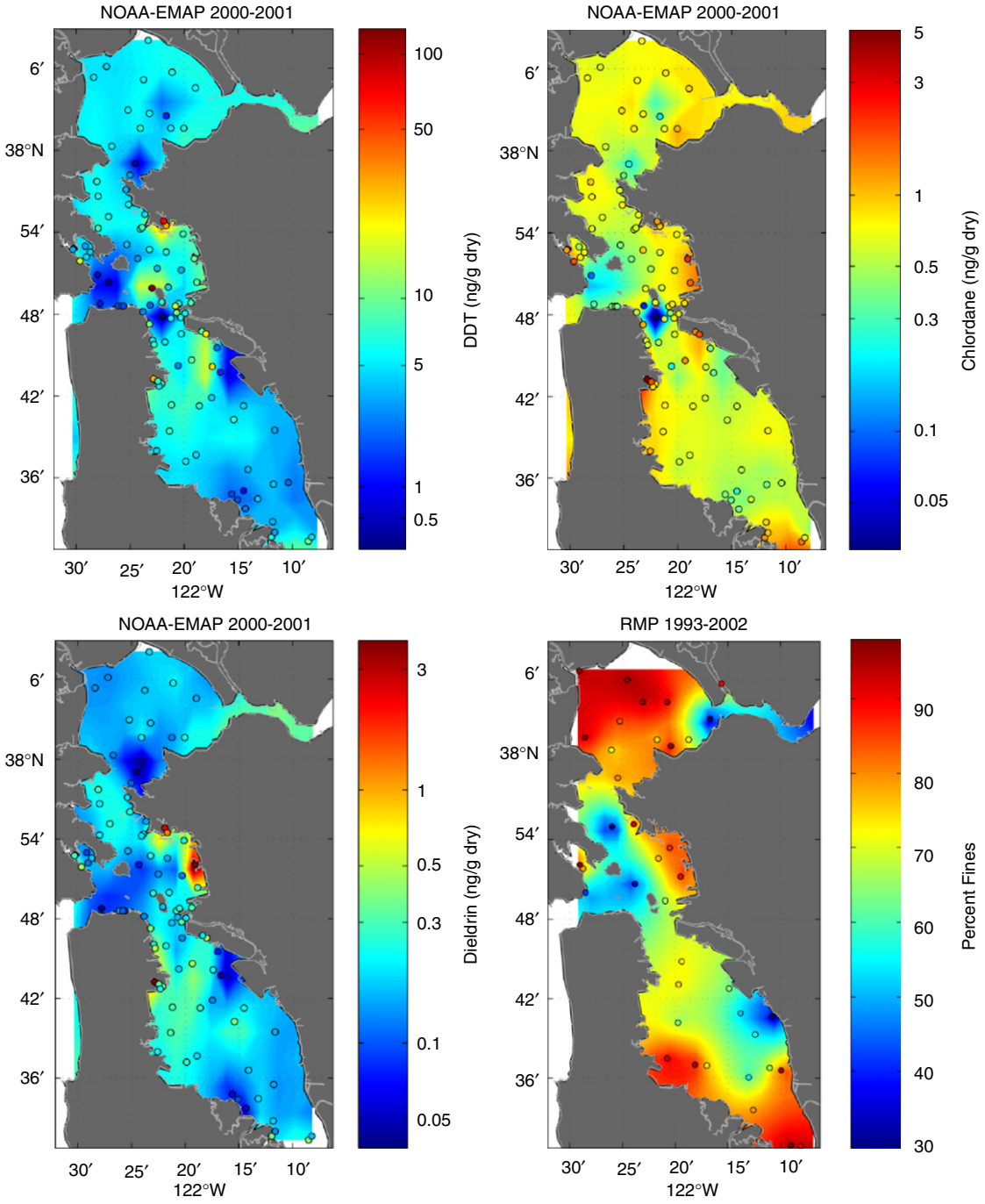

Fig. 1. Concentrations of legacy pesticides and percentage fines in San Francisco Bay sediments.

buried beneath the active layer, and dredging and disposal of deep sediments.

We used information from the literature to estimate loads from these sources (Table 2 and explained below). Our best estimates of total inputs are 60 kg/year DDTs, 30 kg/year chlordanes, and 10 kg/year dieldrin. These ongoing inputs are of varying importance in comparison to the reservoirs of pesticides estimated to be contained in the active sediment layer. Estimated annual inputs of DDTs are less than 20 percent of the amount estimated to be residing in the active sediment layer. Estimated inputs of chlordanes are about two-thirds the amount estimated in the active sediment layer. Estimated inputs of dieldrin are about the same as our estimate of the active sediment layer.

3.1. Central Valley

The intense agricultural activity in California's vast Central Valley left pesticide residues in the soils, stream

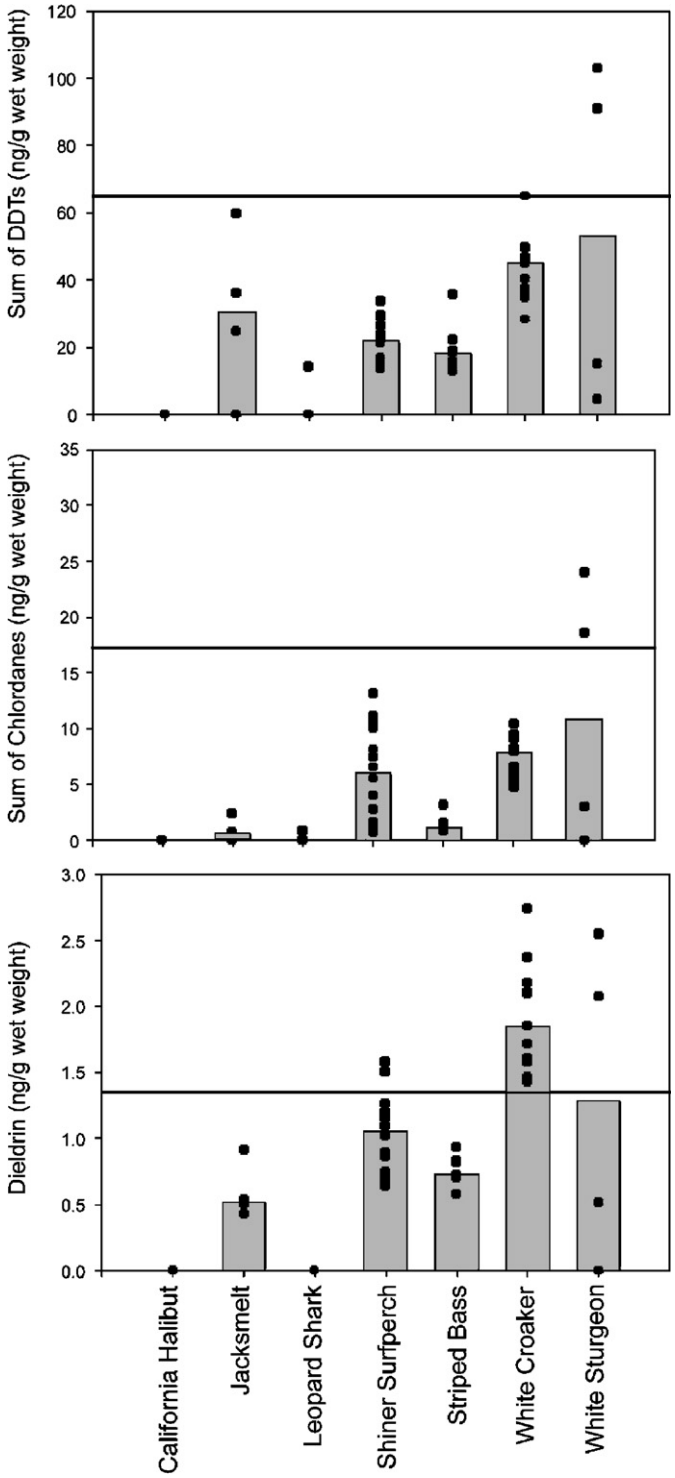

Fig. 2. Concentrations of legacy pesticides (ng/g wet weight) in fish collected by the RMP in 2003. Bar denotes CA screening values.

sediments, water, and biota (Mischke et al., 1985; Gilliom and Clifton, 1990; Pereira et al., 1996; Brown, 1997). Urban use of legacy pesticides within the Central Valley was also common and continued longer than agricultural use, as agricultural restrictions to the pesticides preceded their overall bans. In particular, total urban use of chlordanes may have exceeded agricultural use (Nowell et al., 1999). Consequently, while the contemporary occurrence and distribution of DDTs and dieldrin in the Central Valley are typically associated with historic agricultural applications, the presence of chlordanes is more likely related to historic use for termite and ant control in residential and commercial applications.

Table 2
Estimated loads (kg/year, best estimate and range) of legacy pesticides entering San Francisco Bay

Pathway	DDTs	Chlordanes	Dieldrin
Central Valley	15 (5–40)	2 (0.7–5)	5 (2–13)
Local watersheds	40 (9–190)	30 (7–160)	3 (0.7–15)
Municipal wastewater	0.2 (0.02–2)	0.1 (0.003–2)	0.06 (0.008–0.4)
Industrial wastewater	<0.2	<0.1	<0.06
Atmospheric deposition	1 (0.02–2)	0.9	1 (0.2–2)
Erosion of sediment deposits	9 (0.2–18)	2 (0–4)	0.2 (0–0.6)
Dredged material	−2 (−3 to −0.03)	−0.3 (−0.6–0)	−0.03 (−0.1–0)
Total best estimate	60 (10–250)	30 (10–170)	10 (3–30)

In 1985, concentrations of DDTs and dieldrin in sediments from the San Joaquin River watershed, the southern part of the Central Valley, were among the highest in the nation (Gilliom and Clifton, 1990). The stations with the highest concentrations were located in tributaries that primarily carried agricultural surface runoff. Pereira et al. (1996) observed a similar pattern, with high concentrations of DDTs and dieldrin in water, suspended sediments, sediments, and biota of a tributary bordered by apple orchards and field and row crops. In contrast, maximum chlordane concentrations in suspended sediments were measured in samples from a creek that received urban runoff from Modesto, one of the cities in the valley.

Monitoring data from a site located approximately 5 km downstream from the confluence of the two major rivers draining the Central Valley (Leatherbarrow et al., 2005) were used to estimate total pesticide loads entering the Bay from the Central Valley. Continuous turbidity data, collected on 15-min intervals by the US Geological Survey (USGS) since 1994, used in conjunction with regressions between suspended sediment and pesticide concentrations, allowed for extrapolation of continuous records of suspended sediment and pesticide loads (methods described in McKee et al., 2002; McKee and Foe, 2002).

Best estimate loads were derived from the median and ranges of annual loads estimated by Leatherbarrow et al. (2005) for the years 1995–2003. Median load estimates were 15 kg DDTs, 2 kg chlordanes, and 5 kg dieldrin per year (Table 2). Variability in outflow and sediment transport led to ranges of load estimates that spanned an order of magnitude. The maximum pesticide load, measured in 1995, occurred during a year of above average

outflow (52,000 Mm^3), approximately six times greater than flow in 2001 (8600 Mm^3).

3.2. Local watersheds

Inputs of legacy pesticides from the smaller, local watersheds that feed San Francisco Bay also reflect historic and current land use. Much of the area directly adjacent to the Bay was used for agriculture before the post-World War II period of rapid population growth and urbanization. Two studies conducted in the 1970s and 1980s found that DDT residues were ubiquitous and persistent in agricultural soils and tributary sediments throughout the region (Law and Goerlitz, 1974; Mischke et al., 1985). More recent monitoring conducted in 2001 found that concentrations of DDTs in sediments from urbanized regions of the watersheds were also high, with concentrations ranging as high as 4010 μg/kg (KLI, 2002).

The urban influence on chlordanes and dieldrin in the local watersheds has also been evident. Law and Goerlitz (1974) detected chlordanes in 92 percent of sediment samples from tributaries to the Bay. Recent measurements (KLI, 2002) found concentrations of chlordanes in sediments as high as 11,300 μg/kg in urban, industrial locations, with much lower concentrations in sediments from undeveloped areas. In the same study, dieldrin concentrations were as high as 70 μg/kg, with no dieldrin detected in samples from the undeveloped locations (KLI, 2002).

Estimating the total pesticide loads from the all of the local watersheds is inherently difficult, due to limited available data and insufficient techniques for extrapolating from existing data and accounting for different land uses, hydrology, and other watershed characteristics. San Francisco Bay Area storm water management agencies have used pesticide concentrations in bed sediments from storm water conveyance systems and the rational method, which computes peak discharge from an area based on rainfall intensity and a runoff coefficient, to derive preliminary estimates of annual DDT and chlordane loads (KLI, 2002; Salop et al., 2002). Best estimates (and ranges) were 9.2 (0.9–20) kg/year DDTs and 22 (19–102) kg/year chlordanes, with 98 percent of the total attributed to urban sources. There are considerable uncertainties associated with the estimates derived from the rational method (KLI, 2002). For example, the study focused on urban sources of the pesticides—no data exist to facilitate estimating loads from agricultural sources.

Pesticide loads from local watersheds were also estimated using data collected in 2003 for one tributary, the lower Guadalupe River watershed, and extrapolated to all local watersheds based on the overall sediment and water budgets in the Bay (McKee et al., 2004). The Guadalupe River watershed represents an area that was historically agricultural and converted to predominantly urban land uses during the period that the pesticides were used. Similar to Central Valley load estimates, estimated loads for local watersheds were derived using two types of data: linear regression between suspended sediment concentrations and pesticides; and flow-weighted mean concentrations of pesticides.

Linear relationships between total pesticide concentrations in water and suspended sediment concentrations in 22 Guadalupe River samples provided an estimate of pesticide concentrations associated with suspended particulate material entering the Bay from a local watershed. Slopes of the regressions resulted in approximate suspended sediment-normalized concentrations of 46 μg/kg DDTs, 41 μg/kg chlordanes, and 3.7 μg/kg dieldrin (McKee et al., 2004).

The best available estimates of annual sediment transport to the Bay range from approximately 0.56–1.0 million metric tons (McKee et al., 2003). Applying the suspended sediment concentration-normalized pesticide concentrations from Guadalupe River samples to the range of annual sediment loads from the combined local watershed area resulted in annual pesticide loads from the local watersheds of 26–46 kg DDTs, 23–41 kg chlordanes, and 2.0–3.5 kg dieldrin.

In the Guadalupe River samples, flow-weighted mean concentrations of total DDTs, total chlordanes, and dieldrin were 48, 40, and 3.7 ng/L, respectively. Annual freshwater flow from local watersheds ranges from approximately 180 Mm^3 in dry years to 3930 Mm^3 in wet years (McKee et al., 2003). Using an average annual flow of 920 Mm^3, annual pesticide loads were estimated to be approximately 44 kg DDTs, 37 kg chlordanes, and 3.4 kg dieldrin. These loads were consistent with the suspended sediment concentration-derived loads, while the range of local watershed pesticide loads reflects the variability expected between dry and wet years. Best estimates of loads were derived from the two methods of estimation.

Using the same methods, estimated chlordane loads were of similar magnitude to estimates calculated by the rational method (KLI, 2002). However, DDT loads were approximately an order of magnitude higher than rational method estimates. Lower DDT loads estimated by the rational method may be due to an underestimate of sediment loads by the model (McKee et al., 2003) and the fact that non-urban sites were not well characterized in the studies by KLI (2002) and Salop et al. (2002). Because chlordane was primarily associated with urban land uses, this discrepancy may not have greatly affected chlordane loads,

3.3. Municipal and industrial wastewater effluent

Estimates of pesticide loads from municipal wastewater were based on concentration ranges reported by Yee et al. (2001) and an estimated combined effluent discharge of 600 million gallons per day (MGD) to the Bay (Hetzel, 2004). Best estimates of loads were 0.2 kg/year DDTs, 0.1 kg/year chlordanes, and 0.06 kg/year dieldrin.

Contaminant data from industrial dischargers were not readily available. However, the magnitude of industrial discharge is much lower than municipal discharge

(Hetzel, 2004; Johnson and Looker, 2003), and we would not expect significant concentrations of legacy pesticides in industrial discharges. Therefore, the loads from industrial discharges are simply assumed to be less than loads from municipal discharges.

3.4. Atmospheric deposition

There are no local data on atmospheric deposition of pesticides to San Francisco Bay. However, ranges of wet- and dry-depositional fluxes of legacy pesticides have been estimated for other water bodies, including the Great Lakes (Chan et al., 1994) and Galveston Bay, Texas (Park et al., 2001). Chan et al. (1994) estimated that wet depositional fluxes in the Great Lakes ranged from 0.02 to 1.3 g/km^2/year for DDE and 0.2–1.9 g/km^2/year for dieldrin. The magnitudes of these fluxes were consistent with total (wet + dry) fluxes estimated by Park et al. (2001) for Galveston Bay: 1.9 g/km^2/year for DDTs, 0.75 g/km^2/year for chlordanes, and 0.79 g/km^2/year for cyclodienes, including dieldrin. If the magnitudes of atmospheric flux were similar in San Francisco Bay, resulting atmospheric loads would be approximately 0.02–2 kg/year of DDTs, 0.9 kg/year of chlordanes, and 0.2–2 kg/year of dieldrin over the surface water area of the Bay (1.1×10^9 m^2).

3.5. Erosion of historic sediment deposits

Pesticide loads re-introduced to the biologically active water and sediments from erosion of buried sediment were estimated using methods and assumptions outlined by Johnson and Looker (2003) for mercury in San Francisco Bay. These estimates were based on bathymetric studies of sub-embayments within San Francisco Bay that were undergoing erosion: Suisun Bay (Capiella et al., 1999) and San Pablo Bay (Jaffe et al., 1998). Loading estimates from bed sediment were calculated using the several assumptions: there is an annual net loss of 1100 Mkg of sediment from Suisun and San Pablo bays; eroded sediment is 50 percent water and 50 percent sediment by weight and comprises 740 kg of dry sediment per cubic meter of wet volume; eroding material has approximately the same concentrations of pesticides as surface sediment monitored by the RMP; and eroded material remains within the Bay.

Similar to the Suisun and San Pablo bays, which are in the north, the South Bay underwent net erosion during 1956–1983 (Foxgrover et al., 2004). Over that time period, approximately 70 Mm^3 (962 Mkg) of sediment eroded (an annual average of approximately 2.6 Mm^3). The total estimate of sediment erosion in the Bay is approximately 2100 Mkg of sediment.

Pesticide loads from erosion of buried sediment were developed using the estimate of sediment erosion and a range of surface sediment pesticide concentrations measured at ambient water RMP stations from 1991 to 1999 (excluding stations in sloughs and tributaries). The best estimate and range of loads were based on average concentrations ± 1 SD. Average concentrations used to estimate loads of were 4.1 μg/kg DDTs, 0.71 μg/kg chlordanes, and 0.08 μg/kg dieldrin, resulting in loads of 9 kg/year DDTs, 2 kg/year chlordanes, and 0.2 kg/year dieldrin.

3.6. Dredged material

Pesticide loads to the Bay from dredging and disposal of formerly buried sediments were estimated based on methods and assumptions used for mercury in San Francisco Bay (Johnson and Looker, 2003) and on the assumption that dredged material disposed of in the Bay remains in the Bay. Other assumptions were the same as those used to estimate inputs from erosion.

Net loads of pesticides from dredged material disposal were estimated from average concentrations (±1 SD). Since more material is disposed of outside the Bay than inside, the best estimates indicated net losses rather than inputs, −2 kg/year DDTs, −0.3 kg/year chlordanes, and −0.03 kg/year dieldrin. While dredging and dredged material disposal may represent a loss process rather than an input, the dynamics associated with remobilizing or exposing contaminated sediments through dredging, the resulting magnitudes of pesticide loading to the Bay, and overall effects on water quality are unknown.

4. Loss of legacy pesticides from the Bay

The fate of legacy pesticides in the water, sediments, and biota of San Francisco Bay is dependent upon the physical, chemical, and biological traits of the pesticides and of the San Francisco Bay environment. Pesticides are lost from the water and active sediments of the Bay by transport through the Golden Gate to the ocean, degradation in water and the active sediment layer, and volatilization to the atmosphere. Deep burial in depositional areas of the Bay is another route of loss from the water column and active sediment layer. Historic burial has led to the reservoir of legacy pesticides found in deep cores from depositional areas. However, bathymetric studies have shown that in recent decades, there has been net erosion of sediments from the Bay (e.g., Foxgrover et al., 2004), so we have not accounted for burial as a loss pathway.

We used a contaminant fate model to evaluate long-term fate and important pathways of removal of pesticides from the Bay and estimate rates of recovery of the Bay under current and projected loading scenarios. The model was originally developed by Mackay et al. (1994) for Lake Ontario, and has been used to analyze the fate of PCBs (Davis, 2004) and polycyclic aromatic hydrocarbons (Greenfield and Davis, 2005) in the Bay. We have applied the same model equations and physical variables—including water flow rate, sediment deposition and transport, and depth of the active sediment layer—as were used in previous studies. We have also considered areas of

uncertainty and assessed the extent to which they limit the ability to quantify responses and rates. A more detailed discussion of the model results can be found in Leatherbarrow et al. (2006).

For the purposes of the model, the Bay is considered to include the water and the surface active sediment layer. The dynamics and depths of the active sediment layer are highly variable throughout the Bay and not well-characterized (Fuller et al., 1999; Davis, 2004). Recent studies have shown that the active layer is one of the most influential, yet least understood, factors that affect the long-term fate of contaminants in the Bay (Davis, 2004; Greenfield and Davis, 2005). The depth of the active layer can vary from as little as 3 cm to more than 50 cm (Leahy et al., 1976). For this study, the model used an estimate of 15 cm.

Inputs and losses to the water column and the active sediment layer were represented by two mass balance equations:

$$\Delta M_W/\Delta t = L + k_{SW}M_S - (k_V + k_O + k_{WR} + k_{WS})M_W,$$
$$\Delta M_S/\Delta t = k_{WS}M_W - (k_{SW} + k_{SR})M_S.$$

The two equations represent the change in water column contaminant mass (M_W) and sediment contaminant mass (M_S) over time. Each "k" is a daily rate constant, indicating the proportion of present contaminant mass transformed and moved by volatilization (k_V), outflow (k_O), degradation in water (k_{WR}), water to sediment transport (k_{WS}), sediment to water transport (k_{SW}), and degradation in sediment (k_{SR}). Inputs to the water column include loading (L; includes all external sources) and transport from the sediment layer.

4.1. Transport through the Golden Gate

The extent to which legacy pesticides are removed from the Bay by transport to the ocean is influenced by the distribution of pesticides between the water column and the sediments and on the complex processes of sediment transport and hydrodynamics. The rate of water outflow is a function of the rate at which freshwater flows into the Bay through the Sacramento-San Joaquin River Delta (Davis and Oram, 2005). Complexities associated with tidal exchange make it difficult to accurately estimate outflow rates (Davis, 2004). For the model, outflow of pesticides was approximated by analyzing the rate of water flow through the Golden Gate and the corresponding seaward gradient of pesticide concentrations in RMP water samples. Outflow is an important loss pathway for each of the modeled pesticides (Fig. 3), particularly for dieldrin.

4.2. Degradation in sediment

The long-term persistence of DDTs, chlordanes, and dieldrin in watershed soils is well-documented (e.g., Gilliom and Clifton, 1990; Mischke et al., 1985; Spencer et al., 1996; Stewart and Chisholm, 1971; Castro and Yoshida, 1971). Spencer et al. (1996) found that total DDT concentrations measured in the top 75 cm of agricultural soil samples collected in California in 1994 had declined to approximately 10–28 percent of the concentrations measured in 1971. Assuming first order reaction rates, the declines correspond to half-lives of approximately 7–13 years, resulting in approximately 2–11 percent of total DDT applied in 1965 remaining in watershed soils in 2003.

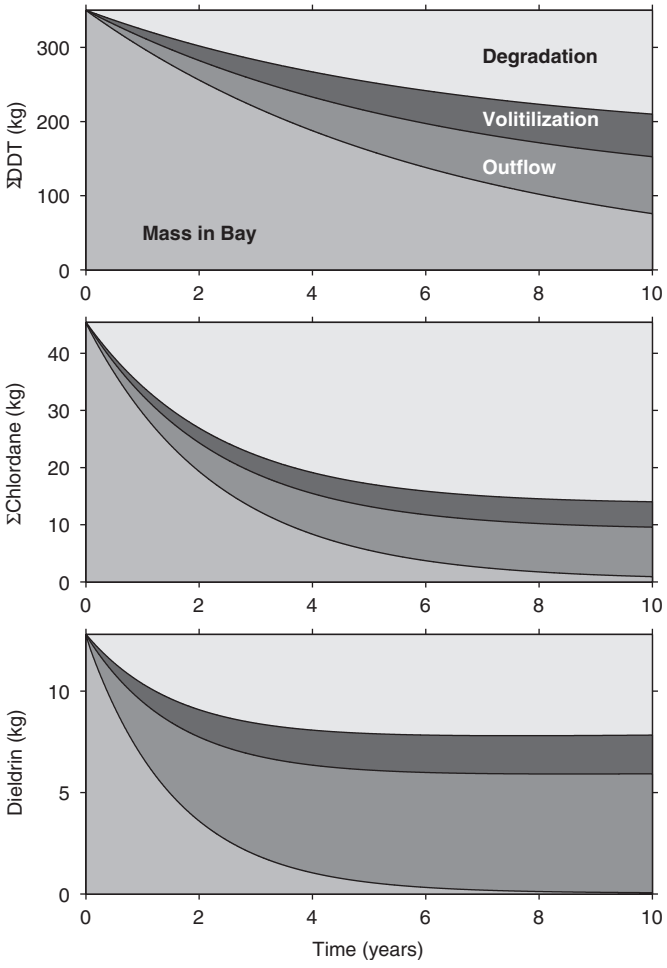

Fig. 3. Model results suggest that degradation in sediments is the most important loss pathway for legacy pesticides, followed by transport through the Golden Gate and volatilization.

The degradation of DDT to DDD and DDE poses an added complexity in understanding the total degradation rates. DDT readily undergoes reductive dechlorination under anaerobic conditions, and the flooding of soils promotes the degradation of DDT to DDD (Castro and Yoshida, 1971). In aerobic environments, DDT is dehydrochlorinated to DDE. Both DDD and DDE are much more recalcitrant in aerobic and anaerobic soils (Castro and Yoshida, 1971; Strömpl and Thiele, 1997). As a result, in estuarine sediment, transformation rates of DDT compounds decrease in the order: DDT > DDD > DDE (Huang et al., 2001).

Degradation rates of organochlorine pesticides in marine and estuarine sediments have not been well-studied but are

known to increase with increasing moisture in soil (Spencer et al., 1996; Castro and Yoshida, 1971; Ghadiri et al., 1995) and marine sediment (Kale et al., 1999), suggesting that degradation rates are higher in the Bay than in watershed soils. Leatherbarrow et al. (2006) compiled literature estimates of degradation rates in soil and sediment to derive applicable best estimates of half-lives of pesticides in Bay sediments. Based on first-order decay rates, the corresponding half-lives due to degradation were 9 years for DDTs, 2.3 years for chlordanes, and 2.8 years for dieldrin. For each of the legacy pesticides, degradation in the sediments is the most important process contributing to loss from the Bay (Fig. 3). There was considerable uncertainty associated with the degradation rate estimates, which spanned at least an order of magnitude.

4.3. Degradation in water

In the water column, degradation occurs by direct and indirect photolysis and hydrolysis. Hydrolysis of legacy pesticides is not thought to be an important removal pathway (Mackay et al., 1997). While photolysis of DDT and DDD is not expected to be important (Callahan et al., 1979), rates of DDE photolysis high enough to essentially remove all DDE from a water body within one day have been reported (Zepp and Cline, 1977). The persistence of DDT compounds in the water column and sediment of San Francisco Bay indicate that rates of degradation are probably much slower than the reported values. Persistence of DDTs in other surface water bodies has been explained by its sorption to sediment (Zepp and Cline, 1977), which can decrease photolysis rates in the water column (Miller and Zepp, 1979; Oliver et al., 1979). Moreover, attenuation of sunlight in natural waters limits the photolysis rates of organic contaminants to the top few centimeters (Zepp and Cline, 1977).

Reported rates of degradation of legacy pesticides in water tend to be higher than those reported for soils or sediments (Leatherbarrow et al., 2006). However, only a small percentage of the total mass of DDTs, chlordanes, and dieldrin is in the water column, so degradation in water is not thought to be a major removal process (Fig. 3).

4.4. Volatilization

Volatilization of pesticides from the water column may be an important pathway of pesticide removal from the Bay; however, no data have been collected to directly study air–water exchange of legacy pesticides within the Bay. In this study, modeled volatilization rates were calculated as a function of the surface area-to-volume ratio of the Bay, the fraction of dissolved pesticides in the water column, and mass transfer coefficients. Mass transfer coefficients were estimated using a model commonly used to estimate organochlorine contaminant transport across the air–water interface (Davis, 2004; Mackay et al., 1994). The model results indicated that volatilization is not a major removal pathway for any of the legacy pesticides (Fig. 3).

5. Model prediction for recovery of the Bay

The model evaluated recovery of San Francisco Bay from legacy pesticide contamination by accounting for varying magnitudes of pesticide inputs, transfer of pesticides between the water and surface sediments, and loss by outflow, degradation, and volatilization. Significant uncertainties were introduced to the model by using Bay-wide estimates of spatially variable parameters, such as concentrations in sediments and the depth of the active sediment layer, and by the relatively large variability associated with loading and degradation estimates. The cumulative effect of these uncertainties has not been defined.

Under a scenario with no new pesticide loading, the model estimated that DDT was the most persistent of the legacy pesticides, with a half-life of about 4 years, compared to 1 year for chlordanes and 0.3 years for dieldrin (Fig. 4). Additionally, the model forecasted that 77 percent of the initial mass of DDTs would be removed from the Bay in 10 years, 98 percent of the initial mass of chlordanes would be removed in 10 years, and nearly all of the dieldrin mass was estimated to be removed within 10 years.

These results are uncertain, because the degradation rates were estimated from uncertain and highly variable data. Increasing the half-life of DDT over a plausible range of 2–16 years decreased the percentage of DDT mass removed from the Bay in 5 years from 100 to 46 percent. However, even using the slowest reported degradation rates, degradation was the most important removal pathway for DDTs and chlordanes. For more soluble dieldrin,

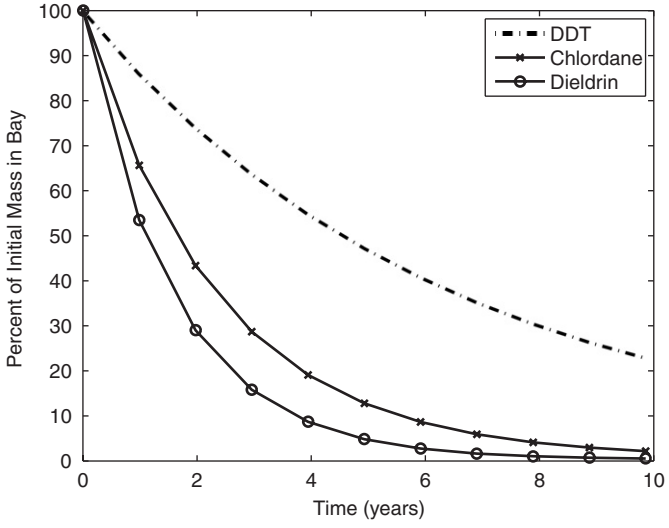

Fig. 4. Model predictions suggest that under conditions of no new loading, 72% of DDTs (model as *p,p′*-DDE), 98% of the chlordanes (modeled as alpha-chlordane), and all of the dieldrin would be lost in 10 years.

model results indicate that volatilization and outflow may be the more important removal pathways.

Based on best estimates of input parameters, the model estimated that, with no continued pesticide loading, San Francisco Bay would be cleared of legacy pesticides within one to three decades. In the absence of continued loading, 95 percent of the current mass DDTs, chlordanes, and dieldrin would be removed from the Bay within 25, 8, and 6 years, respectively.

Long-term model forecasts were also evaluated under scenarios of continued loading to the Bay (Fig. 5). For all scenarios, the model estimated that the Bay would eventually reach steady states in which rates of pesticide inputs equaled rates of pesticide outputs (Fig. 5). Annual loads of about 70 kg DDTs, 30 kg chlordanes, and 30 kg dieldrin would be sufficient to prevent any decrease in the current mass of pesticides in the Bay. The magnitudes of these loads are similar to the best estimates of current pesticide loads calculated for this report, 60 kg DDTs, 30 kg chlordanes, and 10 kg dieldrin (Table 2).

There are significant uncertainties in both the load estimates and the model outputs. For example, interannual variability in hydrology and sediment transport is high in California. Over time, loads may be expected to decrease, and the modeled output does not account for changing loads. However, degradation rates in the soil are slower than those in marine sediments (e.g., Kale et al., 1999). However, the modeled results highlight the question as to whether pesticide mass (and concentrations) in the Bay will decline.

6. Long-term trends in mussels and fish

Actual trends in pesticide concentrations in the Bay were evaluated using bivalve data collected by the California State Mussel Watch Program from 1980–1993 and the

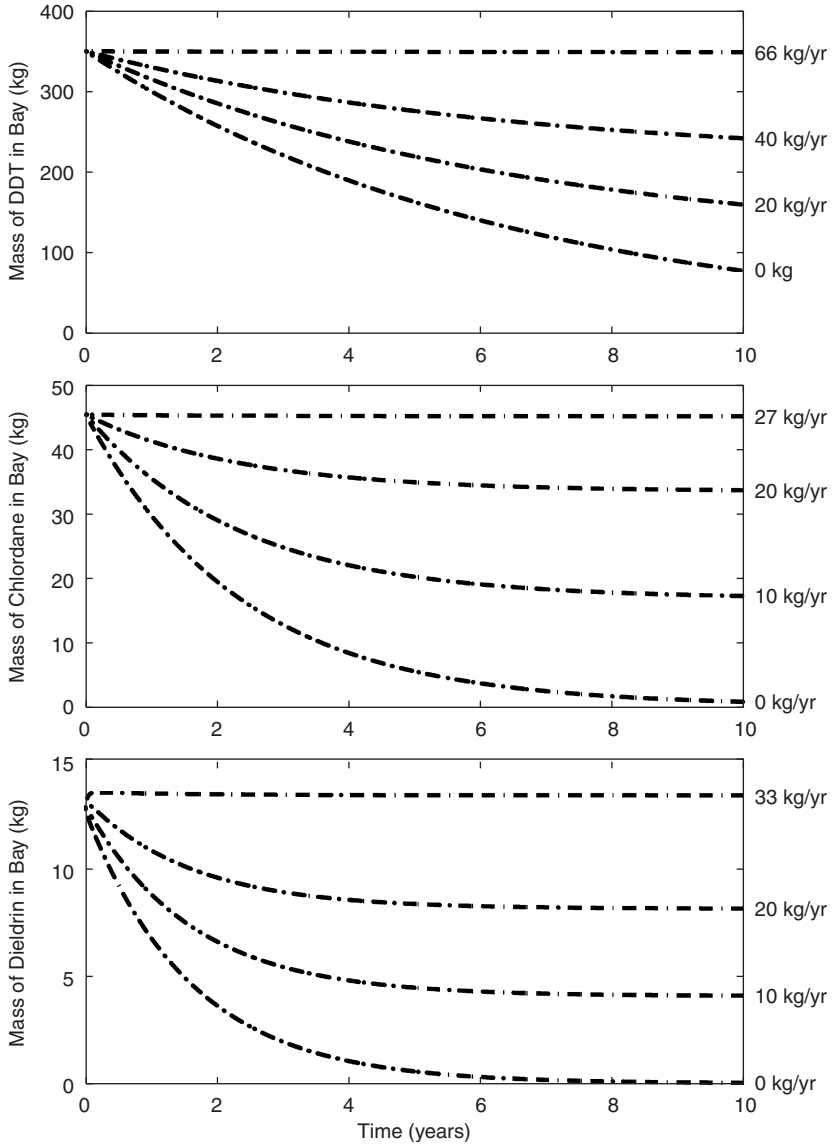

Fig. 5. Model predictions of continued pesticide load under varying continued inputs.

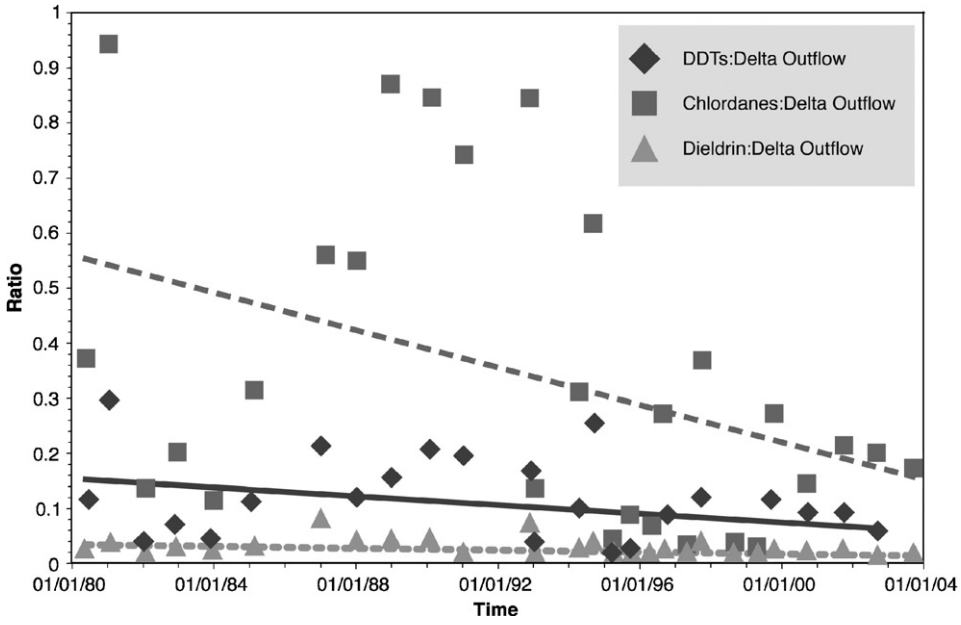

Fig. 6. Trends in ratios of legacy pesticides in mussels to mean daily Delta outflow during 100 days prior to mussel retrieval.

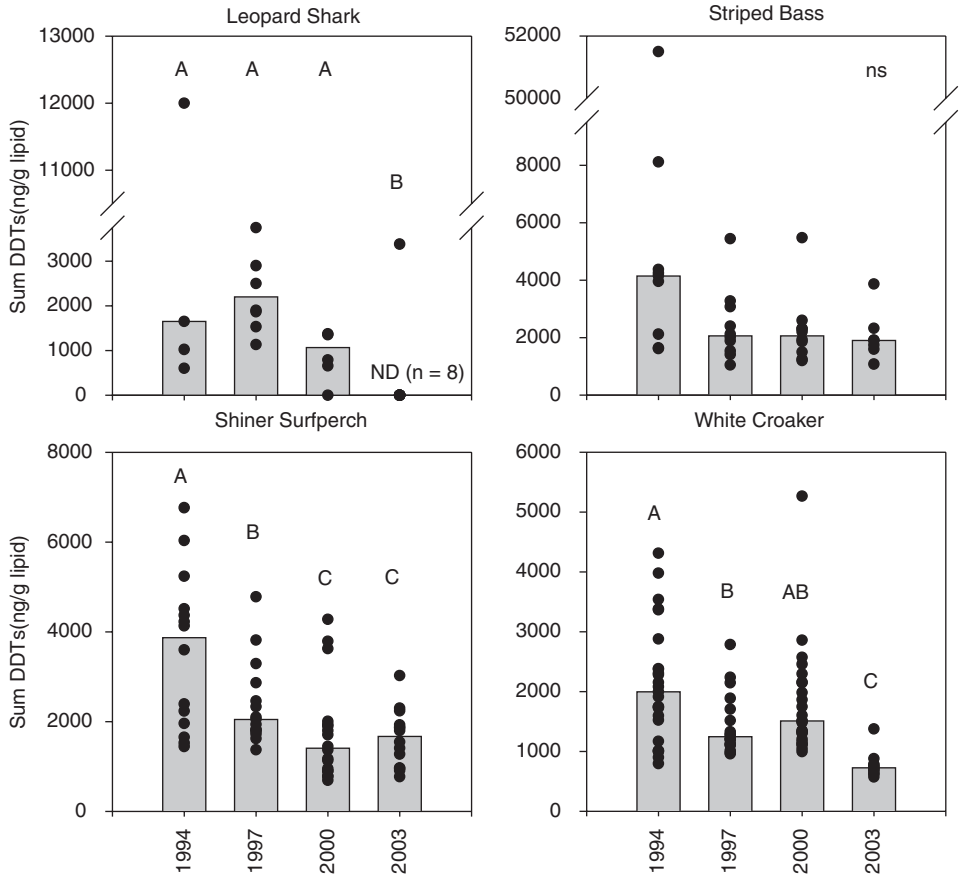

Fig. 7. Change in DDT (ng/g lipid weight) over consecutive RMP sampling periods for four fish species. Points are concentrations in each sample analyzed. Bars indicate median concentrations. ND = below detection limits. Within each panel, different letters indicate significant differences among sample years (*t*-test, LSD; $P<0.05$). ns = multiple year analysis of variance not significant ($P>0.05$).

RMP from 1993 to the present. For each program, mussels (*Mytilus californianus*) were collected from a relatively clean site and transplanted to the Bay for time intervals that, based on preliminary studies (Stephenson, 1992), were sufficient for concentrations to reach equilibrium for most substances (see Gunther et al., 1999 for methods).

For all three pesticides, there have been obvious declines in concentrations measured in transplanted mussels over time, but these declines are less apparent since the early 1990s (Gunther et al., 1999; Leatherbarrow et al., 2006). The observed patterns of declines suggest that higher concentrations occurred during years when mussels were deployed during California's rainy season. This pattern was investigated through stepwise multiple regressions with lipid-normalized concentrations (ng/g lipid) as the dependent variable. Rainfall at San Francisco International Airport during the 3 months prior to mussel retrieval (California Department of Water Resources, 2005) served as a surrogate for runoff from local watersheds. Runoff from the Central Valley was directly examined using three aspects of outflow from the Sacramento-San Joaquin River Delta: mean daily outflow, 95th percentile outflow, and maximum daily outflow in the 100 days prior to mussel retrieval. Retrieval date was also used as an independent variable.

Using log-transformed data to increase linearity, mean daily delta outflow and date were significant variables for all three legacy pesticides. Partial correlations suggested that date had a larger effect than outflow. Trends in the relationship between delta outflow and concentrations of legacy pesticides were examined by plotting the ratios of lipid-normalized pesticides to mean daily delta outflow over time (Fig. 6). Linear regressions of ratios versus time revealed that the decline in the ratio of pesticide concentration to delta outflow was significant for chlordanes ($P = 0.0247$), whereas it was marginally non-significant for DDTs ($P = 0.0507$) and dieldrin ($P = 0.0658$).

While the significant negative relationships between time and DDTs, chlordanes, and dieldrin indicate these legacy pesticides are declining with time, their significant positive relationship with delta outflow suggests the continued importance of runoff from the Central Valley. Declines in the ratios of legacy pesticides to delta outflow suggest attenuation of the upstream sources. However, continued inputs suggest half-lives of 16 and 20 years for DDTs and chlordanes in the Bay, longer than those predicted by the model when no new inputs were assumed.

Data on concentrations of lipid normalized DDTs and chlordanes (there are no comparable dieldrin data) in some fishes indicate statistically significant declines in DDT and chlordane concentrations between 1994 and 2003, when the interim health advisory for consumption of sport fish was issued (Figs. 7 and 8). Using log or

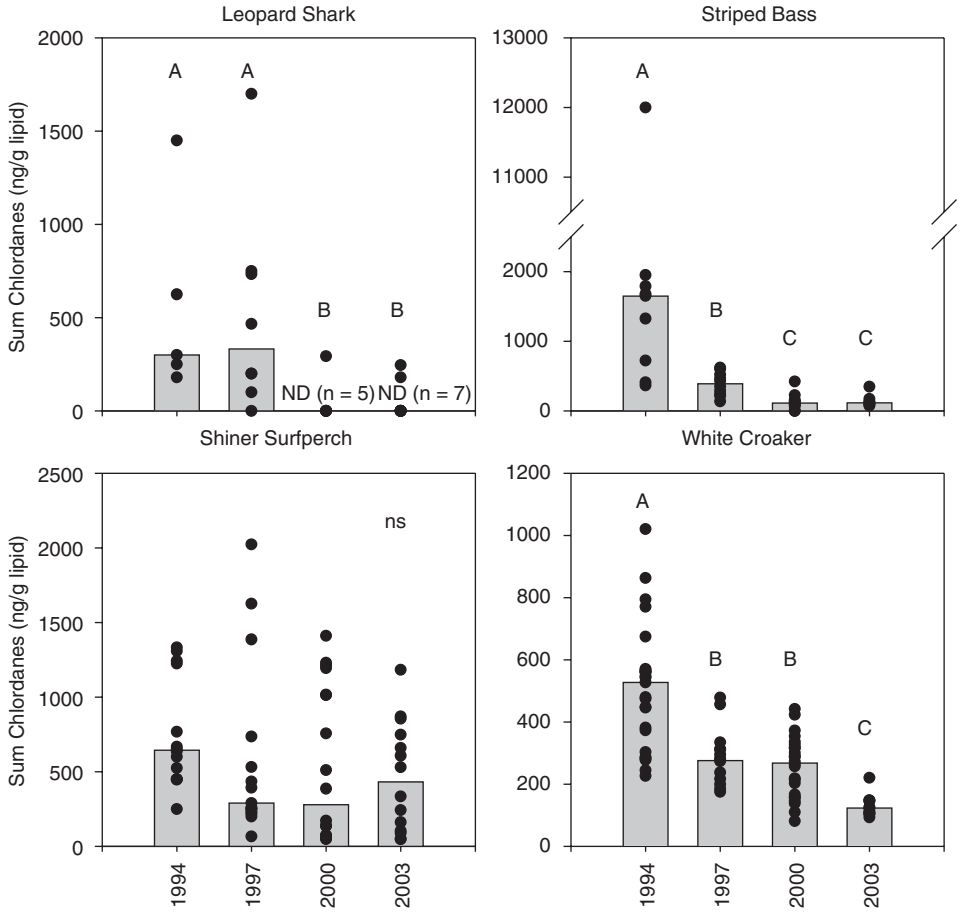

Fig. 8. Change in chlordanes (ng/g lipid weight) over consecutive RMP sampling periods for four fish species. Points are concentrations in each sample analyzed. Bars indicate median concentrations. ND = below detection limits. Within each panel, different letters indicate significant differences among sample years (*t*-test, LSD; $P<0.05$). ns = multiple year analysis of variance not significant ($P>0.05$).

square-root-transformed data to achieve normality, analysis of variance indicated significant differences among years ($P<0.001$) in DDTs and chlordanes (ng/g lipid weight) for three of the four fish species examined. Note that concentrations were evaluated as concentrations per gram lipid, based on the observation of significant effects of lipid content on contaminant concentrations in the Bay (Greenfield et al., 2005). Pairwise multiple comparison tests (Fisher's least-significant difference *t*-test) indicated that later years were significantly lower than earlier years ($P<0.05$; presented as different capital letters in Figs. 7 and 8).

7. Management

The most important management actions were taken when DDTs, chlordanes, and dieldrin were banned from use. A slow recovery of San Francisco Bay began with those regulatory actions. Since then, monitoring of water, sediments, and fish has shown that wildlife and public health continue to be adversely affected by legacy pesticides, although the degree of impact is probably low in comparison to other persistent compounds such as PCBs (Greenfield et al., 2005).

The critical management question for the Bay is whether there are feasible management actions that would decrease concentrations in sport fish significantly faster than the existing slow processes that have been observed. Options being considered include efforts to remediate hot spots with high levels of legacy pesticides in the Bay, in waterways or storm drains that discharge to the Bay, or on lands that drain to them. Some of those locations also have high levels of PCBs, so actions taken in response to PCB contamination will likely manage legacy pesticides as well. One DDT hot spot, Lauritzen Channel in Richmond Harbor, has undergone cleanup as a Superfund site, but remediation has not notably reduced DDT bioavailability at the site (Anderson et al., 2000; Weston et al., 2002).

The State of California is required to establish total maximum daily loads (TMDLs) for pollutants causing impairment of waterbodies. California has purposely delayed development of TMDLs for pesticides to enable the maximum benefit from actions taken to address PCB contamination and to determine what, if any, additional actions are appropriate.

Acknowledgment

This work was supported by the participants in the San Francisco Estuary Regional Monitoring Program.

References

Anderson, B.S., Hunt, J.W., Phillips, B.M., Stoelting, M., Becker, J., Fairey, R., Puckett, H.M., Stephenson, M., Tjeerdema, R., Martin, M., 2000. Ecotoxicologic change at a remediated Superfund site in San Francisco, California, USA. Environ. Toxicol. Chem. 19, 879–887.

Brown, L.R., 1997. Concentrations of chlorinated organic compounds in biota and bed sediment in streams of the San Joaquin Valley, California. Arch. Environ. Contam. Toxicol. 33, 357–368.

California Department of Water Resources, 2005. Detailed Monthly Precipitation Summary Reports, Previous. California Data Exchange Center. 〈http://cdec.water.ca.gov/cgi-progs/previous/PRECIPOUT〉

Callahan, M.A., Slimak, M.W., Gabel, N.W., May, I.P., Fowler, C.F., Freed, J.R., Jennings, P., Durfee, R.L., Witmore, F.C., Maestri, B., Mabey, W.R., Holt, B.R., Gould, C., 1979. Water-related environmental fate of 129 priority pollutants, vol. 1. USEPA 440/4-79-029a.

Capiella, K., Malzone, C., Smith, R., Jaffe, B., 1999. Historical bathymetric change in Suisun Bay: 1867–1990. US Geological Survey Open-File Report 99-563. Presented at the Fourth Biennial State of the Estuary Conference on 17–19 March 1999, San Francisco, CA.

Castro, T.F., Yoshida, T., 1971. Degradation of organochlorine insecticides in flooded soils in the Philippines. J. Agric. Food Chem. 19, 1168–1170.

Chan, C.H., Bruce, G., Harrison, B., 1994. Wet deposition of organochlorine pesticides and polychlorinated biphenyls to the Great Lakes. J. Great Lakes Res. 20, 546–560.

Davis, J.A., 2004. The long-term fate of polychlorinated biphenyls in San Francisco Bay (USA). Environ. Toxicol. Chem. 23, 2396–2409.

Davis, J.A., Oram, J.J., 2005. Comment on "the long-term fate of polychlorinated biphenyls in San Francisco Bay (USA)" letter to the editor, Environ, Toxicol. Chem 24 (10), 2397–2400.

Davis, J.A., May, M.D., Wainwright, S.E., Fairey, R., Roberts, C., Ichikawa, G., Tjeerdema, R., Stoelting, M., Becker, J., Petreas, M., Mok, M., McKinney, M., Taberski, K., 1999. Contaminant Concentrations in Fish from San Francisco Bay, 1997. San Francisco Estuary Institute, Richmond, CA.

Davis, J.A., May, M.D., Greenfield, B.K., Fairey, R., Roberts, C., Ichikawa, G., Stoelting, M.S., Becker, J.S., Tjeerdema, R.S., 2002. Contaminant concentrations in sport fish from San Francisco Bay, 1997. Mar. Pollut. Bull. 44, 1117–1129.

Foxgrover, A.C., Higgins, S.A., Ingraca, M.K., Jaffe, B.E., Smith, R.E., 2004. Deposition, erosion, and bathymetric changes in South San Francisco Bay: 1858–1983. US Geological Survey Open File Report 2004–1192, Santa Cruz, CA.

Fuller, C.C., van Green, A., Baskaran, M., Anima, R., 1999. Sediment chronology in San Franscisco Bay, California, defined by ^{210}Pb, ^{234}Th, ^{137}Cs, and ^{239,240}Pu. Mar. Chem. 64, 7–27.

Ghadiri, H., Rose, C.W., Connell, D.W., 1995. Degradation of organochlorine pesticides in soils under controlled environment and outdoor conditions. Environ. Manage. 43, 141–151.

Gilliom, R.J., Clifton, D.G., 1990. Organochlorine pesticide residues in bed sediments of the San Joaquin River, California. J. Am. Water Res. Assoc. 26, 11–24.

Greenfield, B.K., Davis, J.A., 2005. A PAH fate model for San Francisco Bay. Chemosphere 60, 515–530.

Greenfield, B.K., Davis, J.A., Fairey, R., Roberts, C., Crane, D.B., Ichikawa, G., Petreas, M., 2003. Contaminant concentrations in fish from San Francisco Bay, 2000. RMP Technical Report: SFEI Contribution 77, San Francisco Estuary Institute, Oakland, CA, 82pp.

Greenfield, B.K., Davis, J.A., Fairey, R., Roberts, C., Crane, D., Ichikawa, G., 2005. Seasonal, interannual, and long-term variation in sport fish contamination, San Francisco Bay. Sci. Total Environ. 336, 25–43.

Gunther, A.J., Davis, J.A., Hardin, D.D., Gold, J., Bell, D., Crick, J.R., Scelfo, G.M., Sericano, J., Stephenson, M., 1999. Long-term bioaccumulation monitoring with transplanted bivalves in the San Francisco Estuary. Mar. Pollut. Bull. 38, 170–180.

Hetzel, F., 2004. PCBs in San Francisco Bay: Total Maximum Daily Load Project Report. San Francisco Regional Water Quality Control Board, Oakland, CA.

Huang, H.-J., Liu, S.-M., Kuo, C.-E., 2001. Anaerobic degradation of DDT residues (DDT, DDD, and DDE) in estuarine sediment. J. Environ. Sci. Health B 36, 273–288.

Hunt, J.W., Anderson, B.S., Phillips, B.M., Newman, J., Tjeerdema, R.S., Taberski, K., Wilson, C.J., Stephenson, M., Puckett, H.M., Fairey, R., Oakden, J., 1998. Bay protection and toxic cleanup program final technical report: sediment quality and biological effects in San Francisco Bay. California State Water Resources Control Board, Sacramento, CA.

Jaffe, B.E., Smith, R.E., Torresan, L., 1998. Sedimentation and bathymetric change in San Pablo Bay, 1856–1983: US Geological Survey Open-File Report 98–759.

Johnson, B., Looker, R., 2003. Mercury in San Francisco Bay: Total Maximum Daily Load (TMDL) Proposed Basin Plan Amendment and Staff Report. California Regional Water Quality Control Board, San Francisco Bay Region.

Kale, S.P., Murthy, N.B.K., Raghu, K., Sherkhane, P.D., Carvahlo, F.P., 1999. Studies on degradation of 14C-DDT in the marine environment. Chemosphere 39, 959–968.

KLI, 2002. Joint Stormwater Agency Project to Study Urban Sources of Mercury, PCBs, and Organochlorine Pesticides. Kinnetic Laboratories Inc., Santa Cruz, CA.

Law, L.M., Goerlitz, D.F., 1974. Selected chlorinated hydrocarbons in bottom material from streams tributary to San Francisco Bay. Pestic. Monit. J. 8, 33–36.

Leahy, E.J., Lane, W.B., Tami, T.M., Inman, L.B., McLoud, W.R., Adams, N.J., 1976. Dredged Material Movement Tracing in San Francisco Bay Utilizing Neutron Activation. Weapons Effect Laboratory, US Army Engineer Waterways Experiment Station. Prepared for the US Army Engineer District, San Francisco, CA.

Leatherbarrow, J.E., McKee, L.J., Schoellhamer, D.H., Ganju, N.K., Flegal, A.R., 2005. Concentrations and Loads of organic contaminants and mercury associated with suspended sediment discharged to San Francisco Bay from the Sacramento-San Joaquin River Delta, California. RMP Technical Report, SFEI Contribution 405. San Francisco Estuary Institute, Oakland, CA.

Leatherbarrow, J.E., David, N., Greenfield, B.K., Oram, J.J., Davis, J.A., 2006. Organochlorine pesticide fate in San Francisco Bay. RMP Technical Report, SFEI Contribution 433. San Francisco Estuary Institute, Oakland, CA.

Mackay, D., Sang, S., Vlahos, P., Gobas, F., Diamond, M., Dolan, D., 1994. A rate constant model of chemical dynamics in a lake ecosystem: PCBs in Lake Ontario. J. Great Lakes Res. 20, 625–642.

Mackay, D., Shiu, W., Ma, K., 1997. Illustrated Handbook of Physical–Chemical Properties and Environmental Fate for Organic Chemicals. Volume V: Pesticide Chemicals. Lewis Publishers, Boca Raton, FL.

McKee, L.J., Foe, C., 2002. Estimation of total mercury fluxes entering San Francisco Bay from the Sacramento and San Joaquin river watersheds. Technical Memorandum Prepared for the San Francisco Bay Regional Water Quality Control Board, 23 December 2002. San Francisco Estuary Institute, Oakland, CA.

McKee, L.J., Ganju, N., Schoellhamer, D., Yee, D., Davis, J.A., Leatherbarrow, J.E., Hoenicke, R., 2002. Estimates of suspended-sediment flux entering San Francisco Bay from the Sacramento and San Joaquin Delta. RMP Technical Report, San Francisco Estuary Regional Monitoring Program, SFEI Contribution 65. San Francisco Estuary Institute, Oakland, CA.

McKee, L.J., Leatherbarrow, J.E., Pearce, S., Davis, J.A., 2003. A review of urban runoff processes in the San Francisco Bay Area: Existing knowledge, conceptual models, and monitoring recommendations. RMP Technical Report, San Francisco Estuary Regional Monitoring Program, SFEI Contribution 66. San Francisco Estuary Institute, Oakland, CA.

McKee, L.J., Leatherbarrow, J.E., Eads, R., Freeman, L., 2004. Concentrations and loads of PCBs, OC pesticides, and mercury associated with suspended particles in the lower Guadalupe River, San Jose, California. Prepared for the Clean Estuary Partnership. San Francisco Estuary Institute, Oakland, CA.

Miller, G.C., Zepp, R.G., 1979. Effect of suspended sediment on the photolysis of dissolved pollutants. Water Res. 13, 453–459.

Mischke, T., Brunetti, K., Acosta, V., Weaver, D., Brown, M., 1985. Agricultural sources of DDT residues in California's environment. A Report Prepared in Response to House Resolution No. 53 (1984), Prepared for the State Water Resources Control Board, Sacramento, CA.

Nowell, L.H., Capel, P.D., Lileanis, P.D., 1999. Pesticides in stream sediment and biota—distribution, trends, and governing factors. Pesticides in the Hydrologic System Series. CRC Press, Boca Raton, FL, 92p.

Oliver, B.G., Cosgrove, E.G., Carey, J.H., 1979. Effect of suspended sediment on the photolysis of organics in water. Environ. Sci. Technol. 13, 1075–1077.

Park, J.-S., Wade, T.L., Sweet, S., 2001. Atmospheric deposition of organochlorine contaminants to Galveston Bay, Texas. Atmos. Environ. 35, 3315–3324.

Pereira, W.E., Domagalski, J.L., Hostettler, F.D., 1996. Occurrence and accumulation of pesticides and organic contaminants in river sediment, water, and clam tissues from the San Joaquin River and tributaries, California. Environ. Toxicol. Chem. 15, 172–180.

Salop, P., Abu-Saba, K., Gunther, A., Feng, A., 2002. 2000–01 Alameda County Watershed Sampling Program: Two-Year Summary and Analysis. Prepared for the Alameda Countywide Clean Water Program, Hayward, CA.

San Francisco Estuary Institute (SFEI). 2000. San Francisco Bay Seafood Consumption Study, 84pp.

San Francisco Estuary Institute (SFEI). 2002. 2000 Annual Results: San Francisco Estuary Regional Monitoring Program for Trace Substances.

San Francisco Regional Water Quality Control Board, State Water Resource Control Board, and California Department of Fish and Game, 1995. Contaminant Levels in Fish Tissue from San Francisco Bay.

Shigenaka, G., 1990. Chlordane in the marine environment of the United States: review and results from the National Status and Trends Program. Technical Memorandum NOS OMA 55, NOAA, Seattle, WA.

Spencer, W.F., Singh, G., Taylor, C.D., LeMert, R.A., Cliath, M.M., Farmer, W.J., 1996. DDT persistence and volatility as affected by management practices after 23 years. J. Environ. Qual. 25, 815–821.

Stephenson, M., 1992. A report on bioaccumulation of trace metals and organics in bivalves in San Francisco Bay. Submitted to the California Regional Water Quality Control Board San Francisco Bay Region. California Department of Fish and Game, Moss Landing, CA.

Stewart, D.K.R., Chisholm, D., 1971. Long-term persistence of BHC, DDT, and chlordane in a sandy loam soil. Can. J. Soil Sci. 51, 379–383.

Strömpl, C., Thiele, J.H., 1997. Comparative fate of 1,1,-diphenylethylene (DPE), 1,1-dichloro-2,2,-bis(4-chlorophenyl)-ethylene (DDE), and pentachlorophenol (PCP) under alternating aerobic and anaerobic conditions. Arch. Environ. Contam. Toxicol. 33, 350–356.

USEPA. 2000. Guidance for assessing chemical contaminant data for use in fish advisories, vol. 1. Fish sampling and analysis, vol. 2. Risk Assessment and Fish Consumption Limits, third ed. US Environmental Protection Agency.

Venkatesan, M.I., de Leon, R.P., van Geen, A., Luoma, S.N., 1999. Chlorinated hydrocarbon pesticides and polychlorinated biphenyls in sediment cores from San Francisco Bay. Mar. Chem. 64, 85–97.

Weston, D.P., Jarman, W.M., Cabana, G., Bacon, C.E., Jacobson, L.A., 2002. An evaluation of the success of dredging as remediation at a DDT-contaminated site in San Francisco Bay, California, USA. Environ. Toxicol. Chem. 21, 2216–2224.

Yee, D., Leatherbarrow, J.E., Davis, J.A., 2001. South Bay/Fairfield-Suisun Trace Organic Contaminants in Effluent Study. Prepared for the San Jose/Santa Clara Water Pollution Control Plant, Sunnyvale Water Pollution Control Plant, Palo Alto Regional Water Quality Control Plant, and Fairfield-Suisun Sewer District. San Francisco Estuary Institute, Richmond, CA.

Zepp, R.G., Cline, D.M., 1977. Rates of direct photolysis in aquatic environment. Environ. Sci. Technol. 11, 359–366.