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Mussel watch update: Long-term trends in selected contaminants from coastal California, 1977–2010

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ABSTRACT

This study examined trends in contaminants measured during three decades of "Mussel Watch" monitoring on the California coast. Chlorinated organic contaminants and butyltins declined the most rapidly, with tissue concentrations in 2010 that were up to 75% lower than during the 1980s. Silver and lead declined at about half of the stations statewide, but generally exhibited slower rates of decline relative to the organic compounds. In contrast, copper increased at many stations, and PAHs showed little evidence for declines. Mussels from San Francisco Bay and the Southern California Bight were historically the most contaminated and have had the steepest declines. Overall, these data show that the "Mussel Watch" approach to monitoring contaminants in California has provided some of the best evidence of the effectiveness of actions to improve water quality over the past 30 years. These datasets also highlight challenges that remain in managing PAHs and copper.

1. Introduction

Bivalve molluscs represent one of the most frequently used spatial and temporal trend indicators of contaminants in aquatic environments. Bivalves are important components of coastal habitats, are closely connected to changes in water quality, and reflect bioaccumulation near the base of the food web. Bivalves also offer advantages as indicator species because of their capacity to retain certain classes of chemicals. For example, PAHs do not accumulate in fish, but reach easily measurable concentrations in bivalves (Hellou et al., 2000; Mearns et al., 1999; Oros and Ross, 2005). The limited metabolism of most toxic compounds in bivalves (Walker and Livingstone, 1992) also makes them excellent monitoring tools for emerging contaminants.

Bivalves have been used as indicators of contaminants throughout the United States and around the world (Hunt and Slone, 2010; Lauenstein and Daskalakis, 1998; O'Connor and Lauenstein, 2006; Ramu et al., 2007; Roach and Runcie, 1998; Stephenson and Leonard, 1994; Stephenson et al., 1995). Resident and transplanted mussels have been used to assess patterns in bioaccumulation in California waters for over four decades, including some of the earliest work conducted anywhere using the "Mussel Watch" approach (Butler, 1973; Graham, 1972; Mearns et al., 1991; Wyland, 1975).

California initiated its statewide mussel watch program in 1977 to provide an indication of spatial and inter-annual trends in toxic contaminants, principally heavy metals, synthetic organic compounds, and pesticides (Martin, 1985). The California State Mussel Watch (CSMW) conducted annual monitoring of contaminants in resident and transplanted mussels until 2003. The CSMW yielded a wealth of useful information, including demonstration of rapid improvements in water quality resulting from bans on PCBs and several organochlorine pesticides, reductions in metals due to air pollution regulation and wastewater treatment, and bans on tributyltin hull coatings (Davis et al., 2006; Stephenson et al., 1995; Tetra Tech, 2008). However, the CSMW has been mostly discontinued due to a lack of funding. Since 2003, 20 sites have been monitored with limited funding resulting from an enforcement action settlement.

The most extensive mussel watch program in California has been the National Mussel Watch (NMW), part of the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Monitoring Program. The program began in 1986 and is one of the longest running, continuous coastal monitoring programs in the world. The NMW was designed to monitor the status and trends of chemical contamination of U.S. coastal waters (Lauenstein and

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Daskalakis, 1998). The NMW initially performed chemical analysis of resident oysters and mussels that spanned 145 stations. This number has steadily grown to 300 currently monitored stations throughout the United States. In California, 40 stations were sampled between 1986 and 2005, with consistent sampling performed at 35 sites. Sampling was conducted annually in the early years of the program, but was reduced to a biennial frequency in 1994. Note that the majority of NMW sites are in different locations than those of CSMW.

The CSMW and NMW have together yielded one of the longest running time series on statewide bioaccumulation patterns. The objectives of this paper are to describe spatial and temporal trends in mussel bioaccumulation using CSMW and NMW data in California. Long-term trends were examined in 10 contaminants at more than 50 stations along the California coastline.

2. Methods

2.1. California State Mussel Watch

Twenty-four CSMW stations along the California coastline have been consistently monitored for approximately 120 individual contaminants in the California mussel, *Mytilus californianus* (Fig. 1 and Table 1a). Stations sampled for ten years or more were included in our analyses. Applying this criterion, 10 stations were located north of Point Conception (northern California) and 14 stations were located in southern California. Resident mussels were sampled at about one-third of the stations, and transplanted mussels collected from relatively clean locations were deployed and analyzed at the other stations. CSMW transplant methods follow protocols described by Gunther et al. (1999).

Forty-five resident or transplanted (45-day deployment) mussels were analyzed per site. Sample collection and analytical techniques varied somewhat over time, and have been described previously in Stephenson et al. (1995). Procedures employed in 1992 have been consistent since that time.

Quality Assurance (QA) has been conducted by CSMW since the program began in 1977, including standard reference materials and method blanks. Inter-calibration exercises between CSMW and NMW were conducted in 1988 with mussels from the Gulf of Farallones and Channel Islands, and good agreement between labs was observed (Martin and Stephenson, 1990a, b). From 1986 to 1998, CSMW participated in inter-calibration with the National Research Council of Canada. Since 1996, CSMW has also participated in NOAA's inter-calibration program. Excellent agreement was achieved each year for the metals and organics included in this paper.

2.2. Regional Monitoring Program

The Regional Monitoring Program for Water Quality in the San Francisco Estuary (RMP) was initiated in 1993 and has extended the CSMW time series in San Francisco Bay. The RMP began monitoring 11 stations using transplanted mussels, oysters, and clams to cover the range of salinities found in the Estuary (SFEI, 1994). In each sampling year, approximately 300 individual contaminants were assessed. From 1994 to 2003, the number of stations fluctuated between 11 and 15 stations, based on design modifications (SFEI, 2010). The frequency of bivalve sampling was reduced from annual to biennial in 2006 (Melwani et al., 2008). Of the original 11 bivalve sampling stations in the RMP, six are historic CSMW sites where the long-term time series initiated by CSMW are still being extended (Gunther et al., 1999; SFEI, 2005). Three of these stations (Emeryville, Dumbarton Bridge, and San Mateo Bridge) also coincide with long term NMW monitoring stations in San Francisco Bay. Although detection of trends may be obscured to some extent by the use of different analytical laboratories and methods, the six CSMW/RMP stations represent the best dataset available on bioaccumulation trends in San Francisco Bay over the past 30 years (Fig. 1 and Table 1b). All CSMW/RMP stations were assessed by transplanting mussels from relatively clean locations to targeted stations for deployment and subsequent chemical analysis (Gunther et al., 1999). All RMP data included QA/QC samples that met the data quality objectives specified in the RMP's Quality Assurance Project Plan (Davis et al., 2001).

2.3. National Mussel Watch

Since 1986, the National Mussel Watch (NMW) has analyzed approximately 150 contaminants in resident bivalve species from stations across the United States (Kimbrough et al., 2008; O'Connor and Lauenstein, 2006). From 1986 to 2009, consistent sampling was performed at 35 sites in California (Fig. 1 and Table 1c), including seven stations that correspond to protected locations designated as Areas of Special Biological Significance (ASBS) by the State Water Resources Control Board. The majority of the NMW stations in California have results for more than 15 unique years.

Sample collections and analysis have followed NOAA's Standard Operating Procedure (e.g., Lauenstein and Cantillo, 1998; McDonald et al., 2006). *M. californianus* are collected from the open coast sites, while both *M. galloprovincialis* and *M. trossulus* are collected in enclosed waters. Analytical protocols and quality assurance follow methods approved for the NMW Program (Lauenstein and Cantillo, 1998). Since 2007, the California State Water Resources Control Board and the Southern California Coastal Water Research Project have collaborated with NOAA on expanding the spatial coverage and targeted analyte list for the NMW effort in the state.

2.4. Statistical analysis

Data for selected organic compounds (total chlordanes, total DDTs, dieldrin, and total PCBs) and trace metals (lead, copper, silver, and zinc) measured by both CSMW and NMW were examined for temporal trends. Analysis of RMP data focused on DDTs and PCBs only, which have been two contaminants of concern in San Francisco Bay. In addition, sufficient NMW data were collected on total PAHs and total butyltins for evaluation of statewide trends. Summing of organic contaminants in each dataset followed standard protocols (SFEI, 2010) and was the same for each dataset, except for PCBs (Table S1). In the case of PCBs, CSMW and RMP data represent the summation of PCBs as Aroclors, while NMW represent sums of PCB congeners. This difference was due to CSMW only using Aroclors during the early years of the Program, while the NMW analyzed congener data.

Temporal trends were assessed using linear regression analysis of concentrations versus year in R Statistical Software (version 2.10.0). Concentrations were log-transformed to achieve normally distributed residuals and equal variances, because mussel bioaccumulation monitoring data do not fit a normal distribution. Censored data (below the detection limit) were substituted with zero values prior to all statistical analyses. In CSMW and RMP analyses, linear regressions were performed on lipid-normalized organic concentrations and dry weight metal concentrations. All concentrations in the NMW dataset were assessed on dry-weight basis. This was due to the use of inconsistent extraction methods to determine lipid concentrations in historic NMW monitoring. Finally, at each location with a declining slope (b), the number of years for concentrations to decline by 50% (i.e., half the initial concentration; HIC) were calculated by HIC = $\log_{10}(2)/b$.

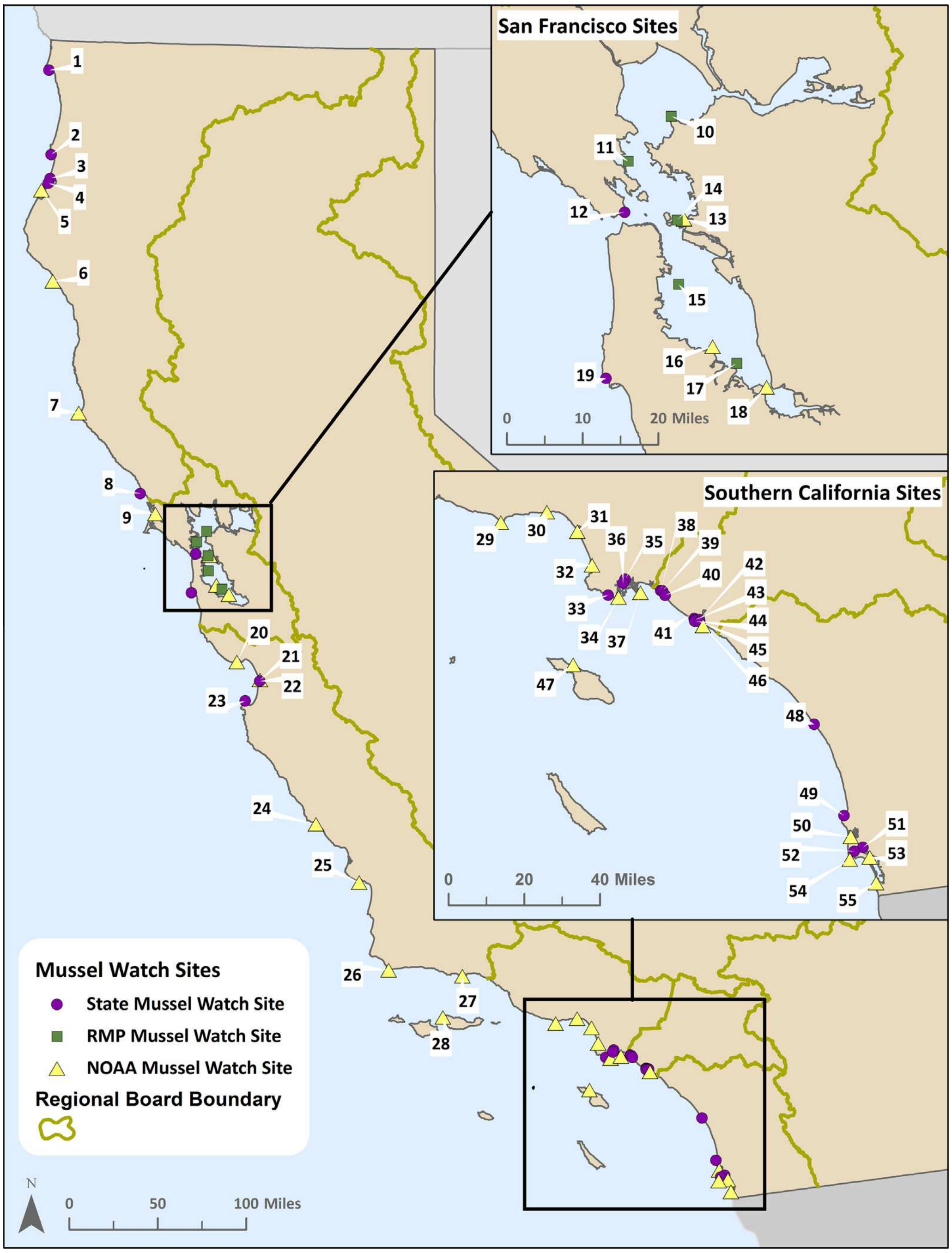

Fig. 1. Stations in California sampled across the three major mussel watch monitoring programs evaluated in this paper. Location names are provided in Tables 1a–c.

2.5. Data Presentation

Trends in 10 contaminants from three major mussel watch monitoring programs in California are summarized in this paper (Table 2). Data from 24 stations monitored by the California State Mussel Watch (CSMW) from 1977 to 2010; six stations that were originally CSMW stations (1980–1992) until the Regional Monitoring Program (RMP) took over (1993–2010); and 35 stations monitored by NOAA's National Mussel Watch (1986–2009). DDTs, PCBs, PAHs, butyltins, silver, lead, and copper have been selected for detailed interpretation below. The entire dataset and graphical analysis for individual stations and contaminants not presented are available by contacting the authors. In the results summary, two aspects of the concentrations should be noted: (1) organic contaminant units differ between programs, and when in lipid weight and dry weight are referred to by the notation 'lw' and 'dw', respectively; and (2) concentrations referred to as 'low' or 'high' reflect the rounded value from the 25th or 75th percentile of the data distribution for each contaminant and dataset assessed, respectively.

3. Results

3.1. DDTs

DDTs in mussels have declined at nearly all CSMW stations. Ninety-percent of stations (17 of 19) had statistically significant

Table 1a
California State Mussel Watch monitoring stations sampled from 1977 to 2010. Stations marked with * are the 10 longest running consistent time series available (all stations were not sampled in every year). Station numbers correspond to Fig. 1. Geographic locations refer to NC: North Coast; SF = San Francisco; CC: Central Coast; LA: Los Angeles; NB: Newport Beach; and SD: San Diego.

General location	Station number	Station name	Mussel type	Total years monitored
NC	1	Crescent City	Residents	28
NC	2	Trinidad Head*	Residents	34
NC	3	Mad river Slough	Transplants	29
NC	4	Samoa Bridge/West	Residents	31
NC	8	Bodega Head*	Residents	34
SF	12	San Francisco Bay at Fort Baker	Transplants	30
SF	19	J. Fitzgerald	Residents	34
CC	22	Sandholdt Bridge*	Transplants	28
CC	23	Pacific Grove*	Residents	34
LA	33	Royal Palms*	Residents	34
LA	35	LA Harbor at Consolidated Slip*	Transplants	29
LA	36	LA Harbor at National Steel	Transplants	28
LA	38	Anaheim Bay at Navy Marsh	Transplants	28
LA	39	Huntington Harbor at Edinger Street	Transplants	28
LA	40	Huntington Harbor at Warner Ave*	Transplants	28
NB	41	Newport Bay at Turning Basin	Transplants	28
NB	42	Newport Bay at Highway 1 Bridge*	Transplants	29
NB	43	Newport Bay at Rhine Channel	Transplants	28
NB	44	Newport Bay at Crows Nest*	Transplants	29
NB	45	Newport Bay at Bay Island	Transplants	29
SD	48	Oceanside*	Residents	34
SD	51	San Diego Bay at Harbor Island	Transplants	28
SD	52	San Diego Bay at Shelter Island	Transplants	31

Table 1b
Stations sampled in the San Francisco Bay region from 1977 to 1992 by the California State Mussel Watch and from 1993 to 2010 by the Regional Monitoring Program (RMP). Transplanted mussels were collected at all stations. Station numbers correspond to Fig. 1.

General location	Station number	Station name	Total years monitored
SF	10	Pinole Point	30
SF	11	Richmond Bridge/Red Rock	31
SF	14	Treasure Island/Yerba Buena Island	31
SF	15	Hunters Point/Alameda	30
SF	17	Redwood Creek	30
SF	18	Dumbarton Bridge	30

declines (Tables 2 and S2), which included eight of the longest running stations (Fig. 2a). The majority of stations showing significant declines were located in southern California, where high concentrations (>9800 ppb lw) predominated during the early 1980s. Thirteen sites in southern California exhibited statistically significant trends. These stations exhibited a range in Half-Initial-Concentration (HIC) values of 6 to 13 years, which correspond to declines of 75% or more over 30 years. The two CSMW stations not showing a significant trend over time were located at Sandholdt Bridge (Moss Landing) and San Diego Bay at Harbor Island.

In San Francisco Bay, DDTs exhibited statistically significant declines at all six CSMW/RMP stations (Fig. 2b). Concentrations declined from approximately 1000 ppb lw in the early 1980s to about 200 ppb lw in 2008. HIC ranged from 6 to 11 years, indicating that 2010 concentrations in San Francisco Bay are also 75% lower or more than those of the 1980s (Table S3).

Fewer significant trends in DDTs were observed at the NMW stations. However, the majority of sites not showing declines had low initial concentrations (<20 ppb dw). Note that although these data are presented on dry weight, they would still be relatively low on a lipid basis (see Section 2). DDTs significantly declined at 20 of 35 (57%) NMW stations (Tables 2 and S4). Twenty-seven sites had initial concentrations >20 ppb dw, but only nine of these were considered high concentrations (>170 ppb dw). Many of the stations with significant trends had an HIC that fell within the range of 6–12 years, which was remarkably similar to the rates of decline found at CSMW and RMP stations. One of the steepest decreasing trends was at Royal Palms on the Palos Verdes Peninsula, a site that is close to the Montrose Superfund Site, and has also been monitored annually by CSMW. DDTs declined at this site from 1061 ppb dw in 1986 to 275 ppb dw in 2008.

3.2. PCBs

PCBs in mussels revealed statistically significant declines at over half of CSMW stations (11 of 18, 61%) (Tables 2 and S2), including six of the longest running stations (Fig. 3a). PCBs in the northern part of the state generally exhibited low initial concentrations (<300 ppb lw) and a prevalence of "non-detects". The only CSMW site in northern California with concentrations that consistently exceeded 300 ppb lw was Sandholdt Bridge in Moss Landing, which was also high in DDTs. This location was one of the most contaminated sites across the state with regard to PCBs and showed a slight decline. The majority of stations with high initial concentrations (>9900 ppb lw) were located in southern California. Of the 14 stations in southern California, 10 showed statistically significant declines. Many of these sites had HIC values that ranged from 7 to 12 years, which correspond to declines of 75% or more. However, current concentrations at a few southern locations, such as Newport Bay at Rhine Channel remain relatively high (median = 17,500 ppb lw).

PCBs exhibited statistically significant declines at all six CSMW/RMP stations. Mussels from San Francisco Bay have shown two distinct temporal patterns in PCBs. For the northern Estuary locations (Pinole Point, Richmond Bridge/Red Rock), concentrations declined from approximately 4000 ppb lw in 1982 to about 1000 ppb lw in 2010 (Fig. 3b). For the southern Estuary locations (Treasure Island/Yerba Buena Island, Hunter's Point/Alameda, Redwood Creek, and

Table 1c
National Mussel Watch (NMW) monitoring stations located in California sampled from 1986 to 2009. Resident mussels were collected at all stations. Station numbers correspond to Fig. 1. Site abbreviations correspond to NMW naming scheme. General locations refer to NC: North Coast; SF = San Francisco; CC: Central Coast; SCI: Southern California Islands; LA: Los Angeles; NB: Newport Beach; and SD: San Diego.

General location	Station number	Station name	Area of biological significance[a]	Total years monitored
NC	1	Crescent Pt. St. George (SGSG)	N	25
NC	4	Eureka Samoa Bridge (EUSB)	N	21
NC	5	Humboldt Bay Jetty (HMBJ)	N	25
NC	6	Pt. Delgado Shelter Cove (PDSC)	Y	25
NC	7	Pt. Arena Lighthouse (PALH)	N	25
NC	9	Tomales Bay Spenger's Residence (TMSR)	N	25
SF	13	SFB Emeryville (SFEM)	N	24
SF	16	SFB San Mateo Bridge (SFSM)	N	25
SF	18	SFB Dumbarton Bridge (SFDB)	N	25
CC	20	Monterey Bay Pt. Santa Cruz (MBSC)	N	25
CC	21	Monterey Bay Elkhorn Slough (MBES)	N	17
CC	22	Monterey Bay Moss Landing (MBML)	N	21
CC	23	Pacific Grove Lovers Point (PGLP)	Y	25
CC	24	San Simeon Point (SSSS)	N	25
CC	25	San Luis Obispo Bay (SLSL)	N	25
CC	26	Point Conception (PCPC)	N	25
CC	27	Point Santa Barbara (SBSB)	N	25
SCI	28	Santa Cruz Island Fraser Pt. (SCFP)	Y	25
LA	29	Point Dume (PDPD)	Y	25
LA	30	Santa Monica Bay Las Tunas Beach (TBSM)	N	21
LA	31	Marina Del Rey South Jetty (MDSJ)	N	25
LA	32	Redondo Beach Municipal Jetty (RBMJ)	N	21
LA	33	Royal Palms (PVRP)	N	25
LA	34	San Pedro Harbor (SPFP)	N	25
LA	37	Long Beach Breakwater (LBBW)	N	21
LA	38	Anaheim Bay West Jetty (ABWJ)	N	25
NB	46	Newport Beach West Jetty (NBWJ)	Y	25
SCI	47	South Catalina Island Bird Rock (SCBR)	Y	25
SD	48	Oceanside Municipal Jetty (OSBJ)	N	25
SD	49	La Jolla (LJLJ)	Y	25
SD	50	Mission Bay Ventura Bridge (MBVB)	N	25
SD	51	San Diego Bay Harbor Island (SDHI)	N	25
SD	53	San Diego Bay Coronado Bridge (SDCB)	N	22
SD	54	Point Loma Lighthouse (PLLH)	N	25
SD	55	Imperial Beach North Jetty (IBNJ)	N	25

[a] Designated by California State Water Resources Control Board.

Table 2
Summary of trend analysis in California State Mussel Watch (CSMW) and National Mussel Watch (NMW) data, showing number of stations (N) and proportions of stations (%) with statistically significant trends. PAHs and butyltins (BTs) were not measured at CSMW stations.

Contaminant	CSMW[a]				NMW[b]			
	Increasing trends		Decreasing trends		Increasing trends		Decreasing trends	
	N	%	N	%	N	%	N	%
DDTs	0	0	17	90	0	0	20	57
Chlordanes	0	0	14	64	0	0	16	64
Dieldrin	0	0	9	41	0	0	7	20
PCBs	0	0	11	61	0	0	7	20
PAHs	–	–	–	–	3	9	3	9
BTs	–	–	–	–	0	0	11	31
Lead	0	0	11	52	0	0	8	23
Silver	0	0	11	52	0	0	10	29
Copper	8	38	1	<1	4	11	6	17
Zinc	1	<1	4	19	0	0	10	29

[a] Twenty-two stations assessed for Chlordanes and Dieldrin; $N = 19$ for DDTs; and $N = 18$ for PCBs. Twenty-one stations assessed for all metals.
[b] Thirty-five stations assessed for all contaminants, except for Chlordanes ($N = 25$).

Dumbarton Bridge), PCB concentrations were higher, and declined from approximately 6000 ppb lw in 1982 to about 2000 ppb lw in 2010. The rates of decline in PCBs were similar to DDTs at the same sites, with HIC values estimated to vary from 6 to 15 years (Table S3).

PCBs have declined at most of the NMW stations, but few stations exhibited statistically significant trends (Tables 2 and S4). Twenty-seven sites had high initial concentrations (>10 ppb dw), and seven of these exhibited statistically significant declines. Three of the sites with significant declines were in northern California (Humboldt Bay, San Francisco Bay at San Mateo Bridge and Dumbarton Bridge) and the other five were near Los Angeles and San Diego. The largest significant downward trend was at Mission Bay near San Diego, where PCB concentrations dropped from approximately 100 ppb dw in 1988 to about 20 ppb dw in 2008.

3.3. PAHs

Trends in PAH concentrations were inconsistent across the state. Twenty-three out of 35 sites exhibited increasing concentra-

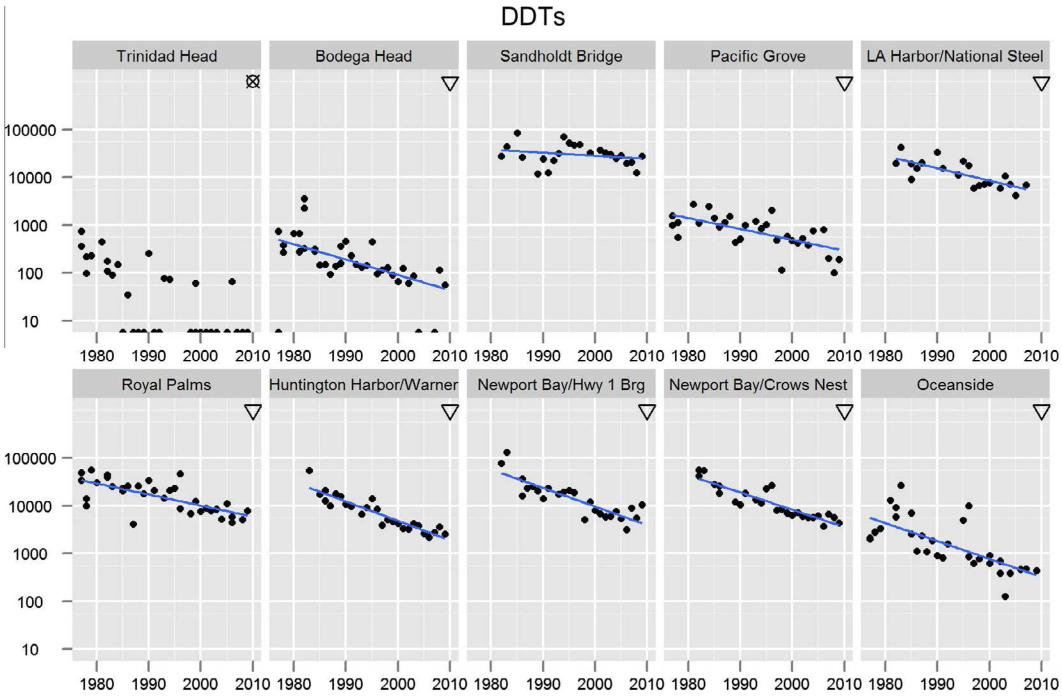

Fig. 2a. Total DDTs in *Mytilus californianus* at the 10 longest running California State Mussel Watch stations sampled from 1977 to 2010. Units are parts per billion, lipid weight. Values below detection are shown by half circles along *x*-axis. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline, $p < 0.05$; ⊗ = greater than 50% of results were below detection).

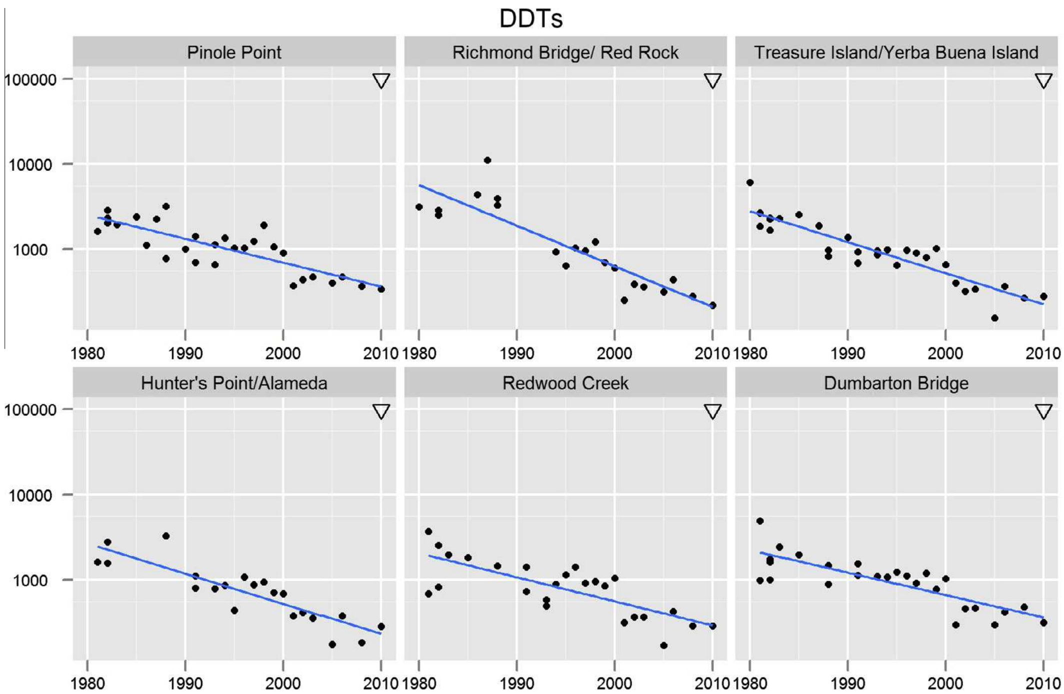

Fig. 2b. Total DDTs in *Mytilus californianus* at California State Mussel Watch/Regional Monitoring Program stations sampled from 1980 to 2010. Units are parts per billion, lipid weight. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline, $p < 0.05$).

tions, but only three (9%) of these increases were statistically significant: Humboldt Bay, Shelter Cove, and Point Santa Barbara (Tables 2 and S5). In addition, 12 sites had declining concentrations, with three (9%) being statistically significant: Long Beach, and San Diego Bay at Harbor Island and Coronado Bridge. These declining stations indicated HIC values that ranged from 9 to 17 years, which suggest generally slower declines compared to other organic contaminants, such as DDTs and PCBs.

3.4. Butyltins

Butyltins declined at 32 of 35 NMW stations, and 11 (31%) of these declines were statistically significant (Tables 2 and S5). The majority of locations (22 of 35) exhibited high initial concentrations (>320 ppb dw). Locations with the steepest declining trends were near San Francisco (San Mateo Bridge and Dumbarton Bridge), Los Angeles (Marina Del Rey, Anaheim Bay, and Royal

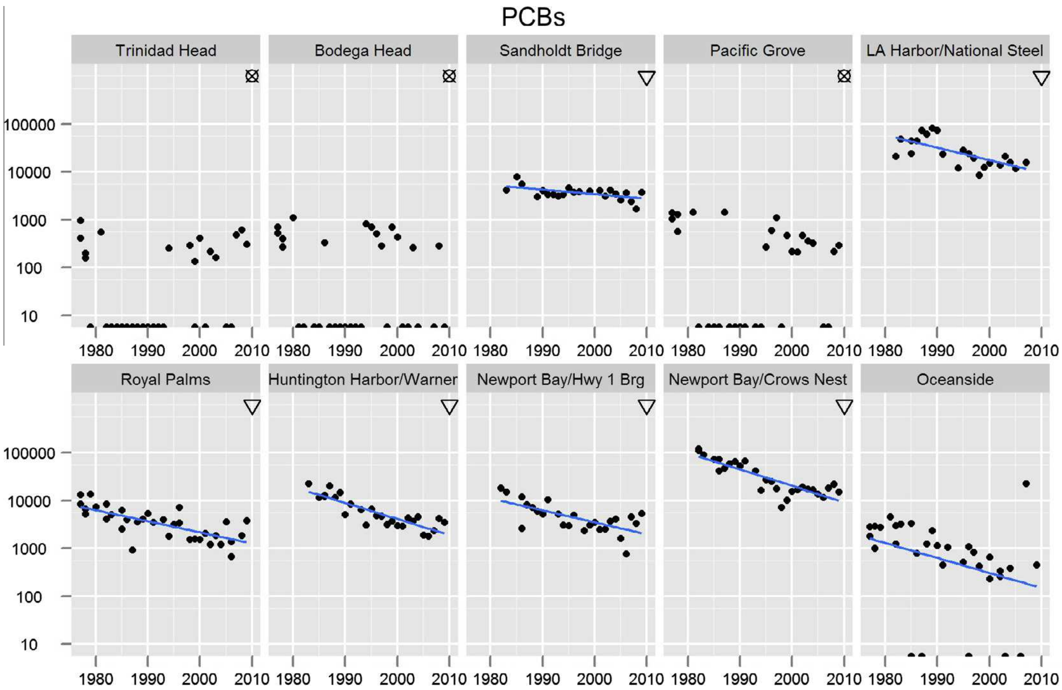

Fig. 3a. Total PCBs in *Mytilus californianus* at the 10 longest running California State Mussel Watch stations sampled from 1977 to 2010. Units are parts per billion, lipid weight. Values below detection are shown by half circles along *x*-axis. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline, $p < 0.05$; ⊗ = greater than 50% results were below detection).

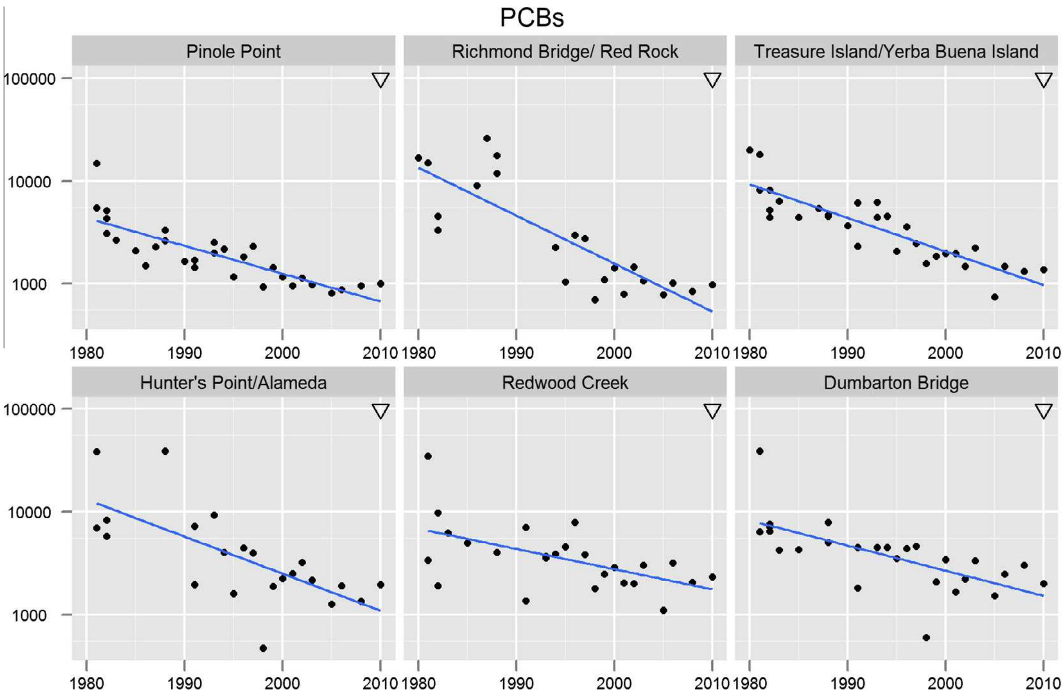

Fig. 3b. Total PCBs in *Mytilus californianus* at California State Mussel Watch/Regional Monitoring Program stations sampled from 1980 to 2010. Units are parts per billion, lipid weight. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline, $p < 0.05$).

Palms), and San Diego (Mission Bay and Pt. Loma Lighthouse). Many of these sites are located in harbors. HIC at these stations were estimated at 4 years, which was the quickest rate of decline observed in any contaminant evaluated. Sixteen other stations with non-significant declining concentrations also revealed relatively low HIC (<10 years).

3.5. Lead

Lead declined at many of the CSMW stations with initially high concentrations (>7 ppm dw). Statistically significant declines were indicated at 11 of 21 (52%) stations (Tables 2 and S6). The majority of stations showing significant declines were located in southern

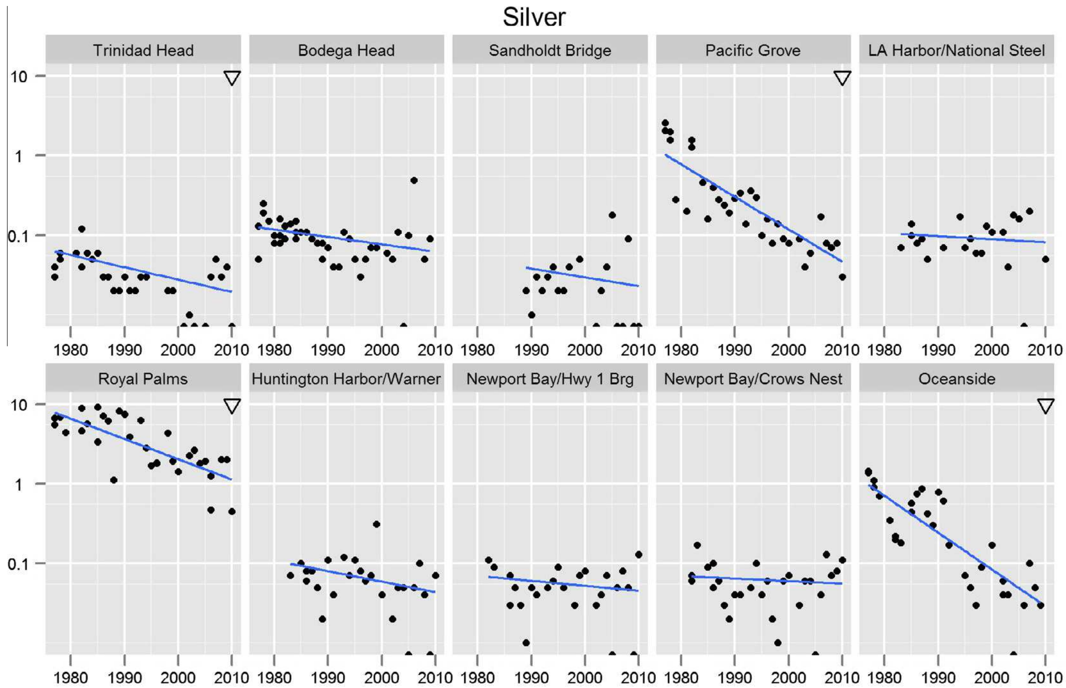

Fig. 4. Silver in *Mytilus californianus* at the 10 longest running California State Mussel Watch stations sampled from 1977 to 2010. Units are parts per million, dry weight. Values below detection are shown by half circles along *x*-axis. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline, $p < 0.05$).

California (9 of 11), located from Los Angeles Harbor to Oceanside. The steepest declines were observed at Pacific Grove, Los Angeles Harbor, Royal Palms, Huntington Harbor, Newport Bay, and Oceanside, where HICs generally ranged from 10 to 18 years. At Royal Palms, concentrations declined from 15 ppm dw in 1977 to 1.8 ppm dw in 2010.

Lead declined at most of the NMW stations, but statistically significant declines were only observed at 8 of 35 (23%) stations: Emeryville in San Francisco Bay, Moss Landing, Marina Del Rey, Royal Palms, San Pedro Harbor, Anaheim Bay, La Jolla, and San Diego Bay at Harbor Island (Tables 2 and S7). The largest statistically significant decline was at Marina Del Rey, where the lead concentration dropped from 35 ppm dw in 1987 to 2 ppm dw in 2008. Nine stations had low initial concentrations (<1 ppm dw), and did not show a significant trend.

3.6. Silver

Significant declining trends in silver were indicated at half of CSMW stations (11 of 21, 52%; Table 2). The most contaminated stations displayed the steepest declines; specifically, San Francisco Bay at Fort Baker, Pacific Grove, Royal Palms, and Oceanside (Table S6 and Fig. 4). HIC at these stations ranged from 6 to 12 years, corresponding to declines of 75% over 30 years. The remainder of stations had low initial concentrations (<0.05 ppm dw), higher HICs, and have not shown any consistent trend.

Silver concentrations have declined slowly at many of the NMW stations; however, similar to the CSMW stations, the majority of locations exhibited low (<0.05 ppm dw) initial concentrations (Table S7). Subsequently, most of the trends were non-significant. Significant declines were observed at 10 of 35 (29%) stations: San Mateo Bridge in San Francisco Bay, Santa Cruz, San Simeon, Point Santa Barbara, Redondo Beach, Royal Palms, Newport Beach, La Jolla, Point Loma, and Imperial Beach, all of which were among the mostly heavily contaminated stations. With a few exceptions, HIC at these stations ranged from 4 to 15 years. The largest statistically significant downward trend was at Point Loma, where silver concentrations dropped from 34 ppm dw in 1991 to about 2 ppm dw in 2005.

3.7. Copper

Copper increased at all but two CSMW stations. Eight of 22 (38%) stations indicated statistically significant increases over time (Tables 2 and S6), including four stations that have the longest running time series (Fig. 5). Many of the significant increases in southern California were at stations located in harbor areas, including Huntington Harbor at Warner Ave, Newport Bay at Crows Nest, and San Diego Bay at Harbor Island. Newport Bay at Crows Nest appears to be a particular recent hotspot, exhibiting copper concentrations above 100 ppm dw in each year since 2007. This is more than double the concentration detected at the same station during the 1980s. Only one station (Royal Palms) suggested a significant decline in copper, but the trend has been relatively slow (HIC ~100 years).

Trends in copper at NMW stations have been inconsistent, having increased at 14 stations and declined at the remaining 21 stations (Table S7). Ten stations exhibited statistically significant trends, with four statistically significant increases and six decreases. Three of four increasing trends were observed in northern California, and all the declining trends were in southern California. The largest statistically significant decline was at Coronado Bridge, where the copper concentration dropped from 35 ppm dw in 1992 to about 12 ppm dw in 2008 (HIC = 19 years). Trends in copper at NMW stations did not suggest a general pattern of increasing concentrations statewide as was evident from the CSMW dataset.

4. Discussion

4.1. Statewide trends

4.1.1. DDTs and PCBs

DDTs and PCBs have been two of the most prevalent legacy organic contaminants in California. They were used extensively until

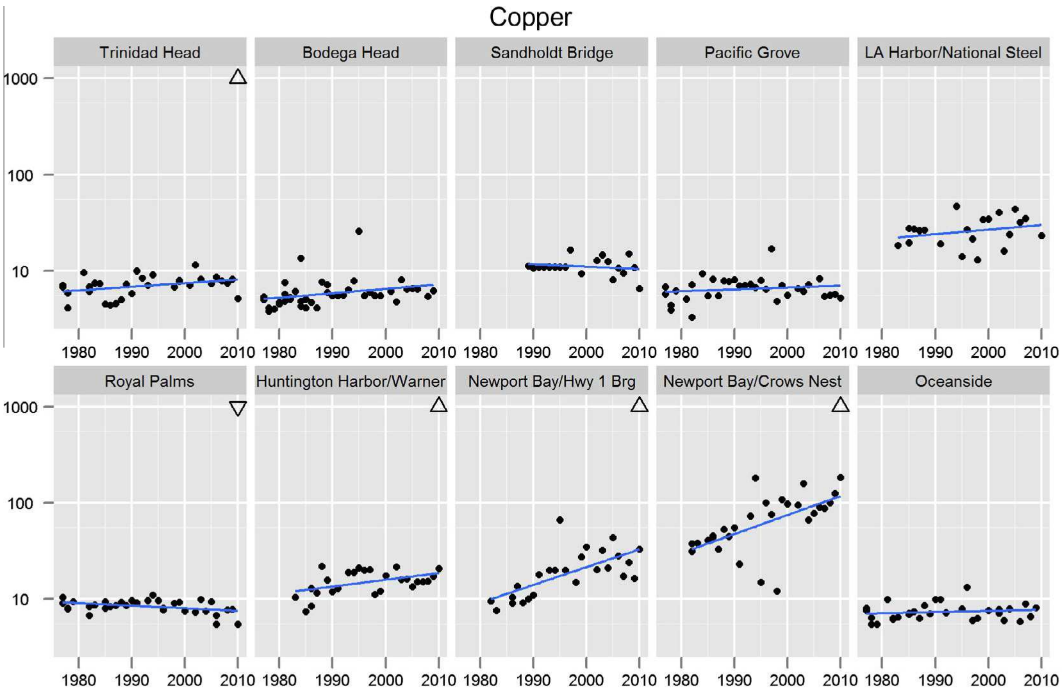

Fig. 5. Copper in *Mytilus californianus* at the 10 longest running California State Mussel Watch stations sampled from 1977 to 2010. Units are parts per million, dry weight. Symbol on top right of each sub-plot indicates if the trend was statistically significant (▽ = significant linear decline; △ = significant linear increase, $p < 0.05$).

banned in 1972. Consequently, temporal trends in bioaccumulation of these contaminants have regularly been assessed and documented at mussel watch stations in California since the 1990s (Kimbrough et al., 2008; Laurenstein and Daskalakis, 1998; O'Connor and Lauenstein, 2006; Stephenson et al., 1995). A previous assessment of CSMW by Stephenson et al. (1995) observed significant declines at about half the stations for DDTs (15 of 32) and approximately one-third of the stations assessed for PCBs (10 of 27). Our findings revealed declining trends at 17 of 19 (90%) stations and 11 of 18 (61%) stations, for DDTs and PCBs, respectively. Our study also found a similar number of NMW stations to have declining trends in DDTs and PCBs as the recent assessments by O'Connor and Lauenstein (2006) and Kimbrough et al. (2008). The slightly higher number of stations with distinct declining trends reported here may be attributable to both the availability of longer time series of declining concentrations (Kimbrough et al., 2008; O'Connor and Lauenstein, 2006), and the selection of a slightly different suite of stations for analysis.

Overall, the steep statewide declines in DDTs and PCBs can be attributed to reductions in sources of these chemicals to California's coastal waters. This inference is supported by observations of rates of decline (HIC) that were faster than would be expected by environmental degradation of these chemicals. The well documented use of organochlorine pesticides in agricultural areas and PCBs in industrial and urban areas, and the resulting contamination of estuarine and marine sediments were the most probable historical sources to the food web. Historical sport fish data also support this perception, as the highest concentrations were near agricultural or industrial sources of these chemicals, and these locations have since declined (Davis et al., 2006).

4.1.2. PAHs

Statewide trends in PAHs do not indicate a general decline or increase over time (O'Connor and Lauenstein, 2006). PAH contamination increased in California throughout the 1900s as natural sources (e.g., forest fires) were augmented by coal burning and fossil-fuel combustion (Pereira et al., 1999). With increases in human population and automobile use in California (ABAG, 2002), PAH inputs from some urbanized waterways have continued (Stein et al., 2006). Of highest concern are the water bodies near densely populated areas, such as San Francisco Bay and the Southern California Bight, which have loadings from multiple sources (e.g., petroleum refineries, creosote, motor vehicles, and wood-burning; Maruya and Schiff, 2009; Oros et al., 2007; Stein et al., 2006).

Although PAH contamination might be expected to increase over time in urbanized areas, historic concentrations in mussels have not significantly increased. Mussel watch stations near San Francisco and along the Southern California Bight did not reveal significant increases in this study or in the recent assessment by Kimbrough et al. (2008). A lack of significant trends was also found for transplanted mussels at ten stations in San Francisco Bay from 1993 to 2001 (Oros and Ross, 2005). Therefore, it is likely that the few significant increases that were observed (i.e., Humboldt Bay, Shelter Cove, and Point Santa Barbara) reflect localized sources of PAHs (O'Connor and Lauenstein, 2006). For example, petroleum seeps at Coal Oil Point in the Santa Barbara Channel may explain our observations of increasing PAH concentrations in that area (Hornafius et al., 1999). Similarly, urban runoff from industrial activities in Humboldt County, may relate to the other stations with increasing PAH concentrations (Humboldt County, 2012).

4.1.3. Silver

Significant declines in silver concentrations at many of the long term monitoring stations in California (e.g., Pacific Grove, Royal Palms, and Oceanside), represent a major success story. During the late 1970s, silver concentrations in *M. californianus* and *M. edulis* from San Francisco Bay were equal to or higher than concentrations in mussels from more than 60 estuaries and coastal locations across North America (Opperhuizen and Sijm, 1990). Accumulation of extraordinarily high concentrations (>100 ppm dw) of silver from coastal waters during this time have almost exclusively been attributed to municipal and industrial wastewater discharges (Hornberger et al., 2000; Luoma and Phillips, 1988). In recent years, with upgrades to wastewater treatment facilities and industrial source controls, decreases in silver have been achieved both in effluents and biota in receiving waters (summarized in Flegal et al., 2007). The steepest decline observed at a CSMW station was at Pacific Grove, where concentrations declined from approximately 2 ppm dw in 1977 to 0.1 ppm dw in 2004. This station is in an Area of Special Biological Significance and is located 500 yards from an outfall that was terminated in 1980 due to a waste discharge prohibition, explaining the dramatic decrease. Notably, NMW monitoring at Pacific Grove which began in 1986 has not shown a significant decline, likely due to missing the period of most significant change in mussel concentrations. The majority of declines in silver at both CSMW and NMW stations were observed in southern California. From 1971 to 1995, mass emissions of silver from large wastewater treatment facilities in the Southern California Bight decreased by 64% (Raco-Rands, 1996). Decreases in emissions from large municipal wastewater facilities in this region (e.g., the Joint Water Pollution Control Plant near Royal Palms) and improved source controls (e.g., photographic wastes) likely explain the significant trends.

4.1.4. Lead

Lead historically entered aquatic environments via leaded gasoline and urban runoff (Stephenson and Leonard, 1994). Similar to silver, large declines of this legacy contaminant represent a success story for environmental managers. Reductions in sources and loads of lead have occurred due to the phaseouts of leaded gasoline and lead-based paints that have occurred since the 1970s. From 1971 to 1995, the combined mass emissions of lead from the four largest municipal wastewater treatment facilities in southern California decreased by nearly 99% (Raco-Rands, 1996). Selected CSMW stations from Royal Palms to Oceanside are located near these treatment facilities, and thus not surprisingly, show the strongest declining trends. Similar trends in southern California were observed at National Mussel Watch stations by Kimbrough et al. (2008). The trends for lead and silver clearly indicate that reductions in mass emissions from wastewater discharge, urban runoff, and atmospheric deposition have resulted in reduced contamination at the base of the food web.

4.1.5. Copper

Copper use has increased widely in California over the past 30 years (Lauenstein and Daskalakis, 1998; Stephenson and Leonard, 1994). Consequently, higher concentrations have been detected at many locations in California during monitoring conducted in the past decade (O'Connor and Lauenstein, 2005).

The primary source of copper to the marine environment has been through antifouling paints on boats and other water vessels. Because tributyltin was prohibited for use on vessels that are less than 25 m in length in the 1988 Organotin Antifouling Paint Control Act, copper became a preferred marine biocide. Copper may enter the water by both passive leaching as well as from hull cleaning (L.M. Candelaria, pers. comm.). Therefore, copper anti-fouling paints may be sources of increasing copper levels at California mussel watch stations. There is an additional concern that copper may be increasing in urban runoff due to use in brake pads (Rosselot, 2006). Many of the stations with increasing copper concentrations in mussels were observed in harbors and marinas. Other reports of increasing copper concentrations in bivalves have noted an association with enclosed embayments (O'Connor and Lauenstein, 2005; Stephenson and Leonard, 1994). These observations suggest that copper use in antifouling paints may be associated with the increasing copper concentrations in mussels. However, current levels of copper in mussels from most stations in California are relatively low compared to other National Mussel Watch stations across the United States (Kimbrough et al., 2008) and are currently below levels that have been associated with toxic effects (Jarvinen and Ankley, 1999). Exceptions are the few stations near Newport and San Diego that have concentrations during 2003–2010 that exceed 100 ppb dw.

4.2. Trends at specific locations

4.2.1. San Francisco Bay

Numerous matrices monitored for contaminants in San Francisco Bay have not shown a declining trend of organic contaminants and trace metal concentrations. PCBs in San Francisco sport fish (e.g., white croaker, shiner surfperch, and California halibut) have shown no decline over time (1994–2009) and a relatively high variance (Davis et al., 2011; Greenfield et al., 2005). Concentrations in water and sediments of the Bay have also been relatively constant (SFEI, 2005). The results shown here for transplanted mussels at CSMW/RMP stations indicate statistically significant declines in PCBs and DDTs at numerous stations in the Bay. The NMW data also indicate significant declines in butyltins and certain organic contaminants in San Francisco Bay: butyltins and DDTs, declined significantly at 2 of 3 sites; and PCBs declined significantly at one site. A possible explanation for these contrasting patterns is that the mussels are accumulating contaminants from different pathways. For example, contaminants in sport fish and mussels have been poorly correlated (Yujie Jin, SWRCB, unpublished data). Fish consume prey that receive their exposure from multiple locations on the margins of the Bay, where residence times for particle-associated contaminants are longer. In contrast, mussels are reflecting declines in water and sediment that occur at individual locations in the open Bay. RMP surveys of PCBs within small fish over the past few years (Greenfield et al., 2011) have found high concentrations at multiple locations of the Bay margins that support this hypothesis.

4.2.2. Salinas River mouth in Moss Landing Harbor

Sandholdt Bridge is located in Moss Landing Harbor at the mouth of the Salinas River. Mussels have been monitored for contaminants in this area by both CSMW and NMW. Over the many years of mussel monitoring in California, PCBs and DDTs in Moss Landing have been the highest of all stations in northern California. For example, DDT concentrations measured in the 1980s in northern California sites were all below 800 ppb lw other than in Moss Landing, where concentrations exceeded 30,000 ppb lw. Concentrations of DDTs at this site were lower during the early 1990s, but otherwise they have not changed much from pre-1980 levels.

The Salinas River has a large watershed with extensive irrigated agricultural fields. The unusually high historic concentrations of DDTs in Moss Landing Harbor likely originate from the application of organochlorine pesticides and subsequent runoff to the Salinas River, and its tributaries. DDT contamination of mussels in Moss Landing has continued through to recent times, possibly due to high flow events, resuspension, and transport of contaminated sediments in the rivers. The relatively low concentrations ($\sim$10,000 ppb lw) of DDTs during the early 1990s reported by Stephenson et al. (1995) may reflect low runoff years, and the relatively high levels (>20,000 ppb lw) during the early 1980s and later 1990s and 2000s reflect periods of increased flow and rainfall. Annual flow from the Salinas River during this period corroborates this hypothesis (Griffin, 2007). In addition, current sources of DDT to the Salinas River drainage may also exist. p,p′-DDE (a derivative of DDT) is a by-product in the insecticide Kelthane, and may be associated with the continued inputs of DDT that occur in the Salinas River watershed.

4.2.3. Royal Palms on the Palos Verdes Peninsula

Mussel watch sampling of Royal Palms represents one of longest-running time series in the country, including data from the earlier USEPA Mussel Watch program (Lauenstein and Daskalakis, 1998). This station is located near the outfall of the JWPCP at White's Point that discharged wastes from the Montrose Chemical Corporation. Industrial waste produced by Montrose, which manufactured DDT from 1947 to 1982 (Graham, 1972) was an immense source of DDT contamination to the coastal waters in this region. In addition, PCBs have been measured in sediments of these waters for more than 30 years, with peak inputs into the Southern California Bight from 1965 to 1970 (Mearns et al., 1991). Additional offshore outfalls contributed as principal sources of PCBs to the Bight, including those discharging treatment wastewater effluent from the Hyperion (City of Los Angeles) and Orange County Sanitation District wastewater treatment plants (SCCWRP, 1973). Therefore, the declines in organic contaminants and numerous trace metals evident at Royal Palms and other coastal locations in this region were undoubtedly influenced by reduced loads from the Montrose Chemical Corporation and POTW effluent discharges in the region (Raco-Rands, 1996). Mussel monitoring by CSMW and NMW throughout the coastal embayments of southern California has documented considerable improvement from the historic contamination of the late 1970s and early 1980s.

5. Conclusions

Bivalve monitoring in California has documented distinct regional declines in some contaminants that have been subject to use restrictions or source control, including organochlorine pesticides, PCBs, butyltins, lead, and silver. Statistical declines were more frequently detected at CSMW stations than NMW stations. This is most likely attributable to the earlier initiation of monitoring of the CSMW, and that many of those stations were in historically the most contaminated areas. In this study, statistically significant declines were found at more than 60% of CSMW stations for 3 of 4 organic contaminants examined (including DDTs and PCBs), and at about 50% of the CSMW stations for 2 of 4 trace metals (lead and silver). DDT also declined at more than 50% of the NMW stations. Overall, the contaminant trends noted from CSMW data were consistent with those compiled recently by the NMW program (Kimbrough et al., 2008; O'Connor and Lauenstein, 2006). Bivalve monitoring has provided crucial evidence of the effectiveness of the management actions taken to reduce these contaminants and the time for environmental breakdown to occur. Monitoring of contaminants with continuing emissions (e.g., PAHs and copper) has revealed that some pollutants are still accumulating in aquatic environment across the state, and are possibly on the increase in certain locations. Copper was the only contaminant that was indicated to have substantially increased. Generally, mussels from enclosed bay and harbor sites remain the greatest areas at risk from copper, due to being more heavily impacted by local inputs compared to open ocean sites.

The existence of historical time series for legacy contaminants of concern in bivalves has proven a valuable data resource in California. Bivalves can be ideal for addressing questions related to spatial trends and sources that affect coastal water quality, because they are stationary and integrate all forms of exposure that are occurring at the sampling location. The vast majority of stations showing declines occurred in regions of elevated historic contamination, such as locations near San Francisco, Los Angeles, and San Diego.

Bivalves are being considered as an important component in monitoring for emerging contaminants in the aquatic environment. The utility of marine bivalves for monitoring contaminants of emerging concern (CECs) is currently being tested in a NMW pilot study throughout California (Maruya et al., 2014). Bivalves are considered an essential component in monitoring for early detection and management of CECs. Therefore, initiation of time series for CECs, along with continued monitoring at a subset of stations for legacy contaminants where problems still persist, are likely to be important components of future mussel watch monitoring in California and other U.S. coastal waters.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2013.04.025.

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