SWAMP
Surface Water Ambient Monitoring Program

MUSSEL WATCH MONITORING IN CALIFORNIA: LONG-TERM TRENDS IN COASTAL CONTAMINANTS AND RECOMMENDATIONS FOR FUTURE MONITORING

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Mussel Watch Monitoring in California: Long-term Trends in Coastal Contaminants and Recommendations for Future Monitoring

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EXECUTIVE SUMMARY

Bivalve molluscs are excellent water quality biosentinels. Long-term monitoring of contaminants in bivalves at coastal California locations has yielded excellent documentation of the significant declines in concentrations of some contaminants that have occurred, and has identified some areas where recovery is progressing more slowly.

This report presents previously unpublished data generated by the State Water Resources Control Board and California Department of Fish and Game (State Mussel Watch [SMW]), compares SMW findings to trends observed in monitoring by the National Oceanic and Atmospheric Administration's National Mussel Watch program (NMW), and evaluates the role of bivalve monitoring in addressing SWAMP objectives and management questions.

Data from three major bivalve monitoring programs in California are summarized in this report. From 1977-2010, the State Water Board sustained measurement of bioaccumulation of organic contaminants and trace metals in *Mytilus californianus* at more than 20 stations along the California coastline. In San Francisco Bay, six locations sampled by the SMW Program from 1977-1993 have been continued by the Regional Monitoring Program for Water Quality in the San Francisco Estuary (RMP). Since 1986, the NMW Program has analyzed organic contaminants and trace metals in numerous resident bivalve species from more than 280 stations across the United States. From 1986-2003 consistent sampling was performed at 35 sites in California. The number of sites sampled in California was increased to 62 during the sample period 2007-2009. The SMW and RMP sampling was largely done using transplanted mussels, while the NMW exclusively sampled residents.

These programs have provided clear evidence of significant regional declines in many contaminants that caused serious water quality problems in the 1960s, 1970s, and 1980s. Data from the SMW, RMP, and NMW have documented declines in concentrations of organochlorine pesticides (DDTs, chlordanes, dieldrin), PCBs, tributyltin, lead, and silver. These declines occurred in response to bans on the use of these chemicals, along with improved wastewater treatment and source control. Other contaminants whose emissions have not been reduced have not shown comparable declines. PAHs showed signs of increase in the NMW sampling. SMW/RMP data for copper indicate increasing concentrations at many locations, although NMW did not. The absence of significant positive trends for all contaminants except PAHs and copper is also an indication of the success of water quality regulations in California.

The sampling approaches employed by the SMW and NMW are generally equally effective, the primary difference being increased power to detect trends in the SMW due to longer-time series and annual sampling. Another design feature affecting trend detection is the inclusion of contaminated sites near sources in the SMW, and the avoidance of these sites in the NMW.

Passive sampling devices (PSDs) provide an alternative to bivalves in some applications. PSDs are simple, low cost devices that rely on diffusive mass transport to concentrate chemicals of interest. Over the past 20 years, PSDs have been designed and optimized to measure a wide variety of target analytes, including trace metals, organics, and organometallics. They offer potential for providing time-averaged concentrations, spatial and temporal trends, and source characterization in a more cost-effective manner and over a greater span of habitats than bivalves. Potential limitations of PSDs include biofouling, slow equilibration times, and lower ecological relevance than biosentinels. The development and validation of PSDs as a water quality monitoring tool should be considered in moving toward a comprehensive, flexible, and cost-effective approach to bioaccumulation monitoring in California waterways.

Bivalve monitoring is an effective means of addressing many of the objectives of the SWAMP bioaccumulation monitoring program. Their greatest value is as indicators of status and trends of the impact of algal toxins on the fishing and aquatic life beneficial uses (especially in marine environments), CEC exposure and risk to aquatic life at lower trophic levels and CEC trends that are relevant both for aquatic life and fishing, and tracking long-term trends in contaminants affecting both fishing and aquatic life at a regional scale and in specific locations. They are especially valuable for monitoring bioaccumulation of certain pollutants, such as PAHs, that are metabolized by species at higher trophic levels. These attributes make bivalve monitoring a valuable component of a statewide bioaccumulation monitoring program.

A considerable amount of the needed monitoring is being conducted by other programs, making coordination a key part of a SWAMP strategy on bivalve monitoring. SWAMP should seek to fill any significant gaps not addressed by other programs.

The following steps are recommended to improve bivalve monitoring in California.

- Conduct a workshop including leading biotoxin scientists to obtain guidance on designing statewide monitoring for biotoxins, including the role of bivalve monitoring. Assess gaps in sampling based on the workshop conclusions.
- Follow up as needed on the 2010 CEC pilot study, in collaboration with NOAA, the RMP, and SCCWRP.
- Develop a coordinated overall plan for bivalve monitoring in California. Establish a prioritized list of stations across programs and all contaminant categories. Initiate a sound archiving plan for key long-term bivalve monitoring stations across all programs.

INTRODUCTION

The California State Water Resources Control Board has established a Surface Water Ambient Monitoring Program (SWAMP). A statewide bioaccumulation monitoring program is one major component of the SWAMP, and was initiated in 2007 (Davis et al. 2008). The SWAMP has developed a set of monitoring objectives and assessment questions for evaluating the impacts of bioaccumulation on beneficial uses (Table 1). This assessment framework is consistent with frameworks developed for other components of SWAMP, and is intended to guide the bioaccumulation monitoring program over the long-term. The four objectives under each of the beneficial uses can be summarized as 1) status; 2) trends; 3) sources and pathways; and 4) effectiveness of management actions.

The primary focus of SWAMP bioaccumulation monitoring to date has been on assessment of the status of impact on the fishing beneficial use through measurement of contaminant concentrations in sport fish. However, California historically has also been a leader in monitoring using mussels and other bivalves. Bivalves, due to their sessile habit in their adult stage, along with other advantageous properties, are indicators of bioaccumulation that can complement sport fish in addressing the four SWAMP objectives. Specifically, since bivalves are better indicators of site-specific conditions, they can provide valuable information on long-term trends, sources and pathways, and effectiveness of management actions that complements the information obtained from sport fish monitoring.

The objectives of this report are to:

- report previously unpublished data generated by the State Water Resources Control Board and California Department of Fish and Game;
- compare data generated by the major mussel monitoring programs in California; and
- evaluate the role of mussel monitoring in addressing SWAMP objectives and management questions.

The information compiled in this report provides a basis for a recommended strategy for incorporating bivalve monitoring into the SWAMP bioaccumulation monitoring element.

The report begins with a general discussion of the advantages of bivalve monitoring and a review of past and present bivalve monitoring programs in California. The main body of the report then presents some previously unpublished data from long-term mussel monitoring sites, and places these data in context through comparison to data from other programs. A thorough graphical summary of long-term trends at the many State Mussel Watch and National Mussel Watch stations in California is presented, with the intent of providing a useful reference for managers and scientists. Finally, based on the compiled information, recommendations for future bivalve monitoring are presented.

The Value of Bivalve Monitoring

Bivalve molluscs are excellent water quality biosentinels. They are particularly valuable for evaluation of long-term trends and spatial patterns in contaminant concentrations in aquatic food webs. Long-term monitoring of contaminants in bivalves at coastal California locations has yielded excellent documentation of the significant declines that have occurred in many cases, and has identified some areas where recovery is progressing more slowly (Davis et al. 2006).

Phillips (1980, 1988) provided a thorough discussion of the attributes that make bivalves one of the best indicators of spatial and temporal trends in bioaccumulation. Filter-feeding bivalves are an indicator for concentrations of contaminants in the water column. Advantageous attributes of bivalves as contaminant indicators include:

- indicating the presence of contaminants that bioaccumulate and have the potential for transfer through the food web;
- accumulating contaminants to higher concentrations than are found in water;
- providing an integrated measure of contaminant abundance over time;
- the sessile habit of bivalves makes them valuable indicators of local conditions and broader-scale spatial variation when data from multiple locations are compared;
- relatively low rates of metabolism allow the detection of a broad array of organic contaminants;
- residents are easily sampled where colonies are established in appropriate areas;
- in locations of interest without resident mussel beds, clean mussels can be transplanted and analyzed for contaminant accumulation;
- it is typically feasible to obtain large numbers of individual bivalves in order to gain statistical power;
- they provide information on exposure of humans and wildlife consumers of shellfish; and
- they offer the potential for studies of effects of contaminants on the bivalves themselves.

While bivalves offer many advantages as sentinels, they also have some important limitations in regard to answering questions that are frequently addressed in ambient monitoring programs. Principal among these are:

- the distribution of resident bivalves is limited, so they are not always present in areas of interest;
- the use of transplants is somewhat resource-intensive and sometimes clean bivalves to use as transplants can be difficult to find (e.g., *Corbicula*);
- bivalves regulate or rapidly depurate some metals, limiting their usefulness as temporal integrators (a prominent example is the rapid depuration of methylmercury by *Corbicula* demonstrated by Foe et al. [2002]; another is zinc [Phillips 1988]);
- variation in feeding rates or seasonal variation in physiology (reproduction) may affect uptake (Gunther et al. 1999, Foe et al. 2002)
- uptake will vary with environmental conditions such as salinity, turbidity, and food availability (Gunther et al. 1999); and

- uptake of some contaminants of concern may be negligible or slow (e.g., Chapman et al. 2010).

Bivalve molluscs have been more frequently employed as spatial and temporal trend indicators of contaminants in aquatic environments than have species of any other family or phylum, and the available literature on their use for such purposes is considerable. Extensive studies of the uptake, sequestration, and excretion of contaminants in bivalves have provided a firm basis for the evaluation of the usefulness of bivalves as indicators of contamination in aquatic ecosystems. The blue mussel, *Mytilus edulis*, has been sampled extensively by various programs in California and is probably the species most widely used for bioaccumulation monitoring worldwide (Phillips 1988).

Two programs that have conducted extensive sampling in California, NOAA's National Mussel Watch Project (NMW- part of the National Status and Trends Program) and the California State Mussel Watch Program (Rasmussen 2000, Foe et al. 2002, He et al. 2006, Boer 2009) were instrumental in gaining widespread international acceptance of this technique as a monitoring tool in aquatic environments.

Mussel Watch Monitoring in California

Early Studies

Mussel monitoring has a long history in California, including some of the earliest work done anywhere using the mussel watch approach. Butler (1969, 1973) described the first national-scale monitoring, an eight-year investigation using several bivalve species to quantify pesticide contamination of coastal waters, including stations in California. Young and coworkers (Alexander and Young 1976, Miller et al. 2010) were also among the pioneers in applying this technique on the Southern California coast in the early 1970s. Transplanted bivalves were used in the Young et al. studies to monitor radionuclides, trace metals, and organochlorines. Some of the early mussel monitoring also was performed in San Francisco Bay, including studies of metal accumulation by Graham (1972), Wyland (1975), and Girvin et al. (1975). The latter study included a relatively extensive survey of six species at nine locations. Risebrough et al. (1978) performed an even more extensive survey of metals and organics in *Mvtilus edulis* at 28 sites in San Francisco Bay, with samples collected in April 1976.

The first statewide mussel monitoring in California was also performed beginning in 1976 as part of the original national mussel watch conducted by USEPA 8, (Goldberg et al. 1978, Farrington et al. 1983, Opperhuizen and Sijm 1990). The U.S. Mussel Watch Program had the overall aim of developing strategies for pollutant monitoring in coastal waters. Mussels and oysters were collected on the west, east, and Gulf coasts during 1976-1978 and were analyzed for trace metals, chlorinated hydrocarbons, petroleum hydrocarbons, and radionuclides. In California, 19 stations were sampled. This program provided a first glimpse of the statewide status of these contaminants in California coastal waters, a valuable frame of reference for comparison with later studies. Collectively, these early studies provided a compelling demonstration of the effectiveness of the mussel watch approach.

California initiated its own statewide mussel watch program in 1977 (Martin 1985). The State Mussel Watch Program (SMW) was initiated to provide the State and Regional Boards with an indication of the spatial and interannual trends in selected toxic pollutants, principally heavy metals and higher molecular weight synthetic organic compounds and pesticides, in the California coastal zone. The SMW continued to conduct annual monitoring until 2003. The Program yielded a wealth of useful information on water quality in California (Stephenson et al. 1995, Davis et al. 2006, Tetra Tech 2008). Many instances of severe contamination were identified, leading to cleanup actions to reduce exposure of humans and wildlife. In addition, many relatively uncontaminated areas were identified. SMW documented the successful management of many pollutants that posed serious threats to wildlife and human health in the 1970s and 1980s. The SMW was instituted just in time to document the rapid improvements in water quality that resulted from bans on PCBs and legacy pesticides, reductions in metals due to wastewater treatment, and other improvements. The SMW was discontinued when plans for the State Board's new statewide Surface Water Ambient Monitoring Program (SWAMP) for water quality began to take shape.

Current Programs

Endowment Monitoring

Although the SMW ended in 2003, monitoring has continued at some of the stations established by that program, providing some of the state's best time series for assessing long-term trends in ambient concentrations of bioaccumulative contaminants. One effort that has extended SMW time series is a joint effort of the State Water Resources Control Board (State Board) and California Department of Fish and Game (CDFG) that has used funds obtained from an endowment established as a result of a $14.4 million legal settlement (Consent Decree) in 1997 between Pacific Gas and Electric (PG&E), the Central Coast Regional Water Quality Control Board, and the U.S. Environmental Protection Agency. The basis for the Consent Decree was that PG&E was responsible for incomplete and misleading reporting regarding marine life mortality due to intakes at the Diablo Canyon Nuclear Power Plant. Part of the settlement funded an endowment ($2.5 million) for a mussel watch program. Funds emanating from this endowment have fluctuated over the years, and were greatly reduced in 2008 and 2009 due to the recession. The endowment monitoring will end in 2012 when the funds run out. Endowment funds have generally sustained continued monitoring at 20 or more stations since the SMW ended.

Regional Monitoring Program for Water Quality in the San Francisco Estuary

Another effort that has extended SMW time series is the Regional Monitoring Program for Water Quality in the San Francisco Estuary (RMP). The RMP was initiated in 1993, and from the beginning has included monitoring of bioaccumulation in transplanted bivalves. The RMP began in 1993 with monitoring of 11 stations using transplanted mussels, oysters, and clams to cover the range of salinities found in the Estuary (SFEI 1994). These early years of the RMP included both wet season and dry season deployments. The number of stations sampled increased to 15 in 1994. In 2000, the wet-season deployments were discontinued since long-term temporal trends in contaminant concentrations were more consistently observed in dry-season data than in wet-season data (SFEI 2008). In 2003 the RMP reevaluated the design of its Status and Trends Monitoring elements and reduced the number of stations from 15 to 11, including 9 sites where transplants were deployed and 2 sites at the landward end of the Estuary where resident freshwater clams are collected (SFEI 2010). Another round of design optimization occurred in 2006, and, based on the results of power analysis, the frequency of bivalve sampling was reduced from annual to biennial (Melwani et al. 2008). Of the remaining 11 bivalve sampling stations in the RMP, six are historic SMW sites where the long-term time series initiated by SMW are still being extended (SFEI 2010).

National Mussel Watch

The most extensive mussel watch program in California is the National Mussel Watch (NMW), part of the National Oceanic and Atmospheric Administration’s National Status and Trends Monitoring Program. NOAA’s NMW Program was designed to monitor the status and trends of chemical contamination of U.S. coastal waters, including the Great Lakes (Alexander and Young 1976). The Program began in 1986 and is one of the longest running, continuous coastal monitoring programs that is national in scope. The Program performs yearly collection and analysis of oysters and mussels. In recent years the NMW included nearly 300 monitoring sites. Many NMW sites in California are coincident with the 1976-1978 USEPA Mussel Watch sites. In California, 40 stations had been sampled between 1985 and 2005, but consistent sampling was performed at 36 of those sites. Sampling was performed annually in the early years of the program, but was reduced to a biennial frequency in 1994.

Since 1995 the NMW program has also included histopathology analyses. These results are used to assess population health and potential interactions between parasites and pathologies and chemical contamination. Histopathology information from the NMW Program, including samples from California, has been assessed from the 1995-1998 period to document parasites and pathologies in bivalves (Brodberg 2007).

Starting in 2007 the State Water Resources Control Board (State Water Board) and the Southern California Coastal Water Research Project (SCCWRP) initiated a collaborative program with NOAA for NMW in California. The State Water Board and SCCWRP agreed to perform sampling at existing and new sites in California, and NOAA agreed to provide analytical services at no cost to the state for all these sites. Other sampling collaborators included the Multi-Agency Rocky Intertidal Network and the Gulf of Farallones National Marine Sanctuary. The number of sites sampled in California was increased from 36 to 65 sites during 2007-2009.

In 2009, the NMW Program began moving in a new direction, with an emphasis on emerging contaminants and an initial focus on California. In March 2009, NOAA convened a workshop in Costa Mesa, California titled "Contaminants of Emerging Concern (CEC): Adapting NOAA Monitoring and Research to Address CEC Management in Coastal, Marine and Great Lakes Environments" with the goal of developing a strategy to coordinate existing regional and national monitoring, research, and assessment programs into a contaminant of emerging concern (CEC) early warning network. Among the key workshop outcomes was that the NMW Program be re-engineered to become NOAA's principal CEC monitoring capability, serving as an early warning sentinel for CECs nationwide. NMW managers then began the task of implementation of a reconfigured NMW that can measure CECs throughout the nation's coastal, marine, and Great Lakes Waters. Also among the workshop recommendations was the need for regionally-tailored monitoring. To that end, NMW managers chose California as a testing ground for the new effort. NMW managers, in consultation with State Water Board, SCCWRP, and SFEI, designed and implemented a CEC monitoring survey for approximately 80 locations in California, conducted in 2010. This pilot study sampled and analyzed (analysis in progress) the following chemical classes:

1. Flame retardants: polybrominated biphenyls (PBBs), polybrominated diphenyl ethers (PBDEs), and a suite of PBDE replacements and other current use compounds
2. Perfluorinated compounds
3. Current use pesticides
4. Alkylphenolethoxylates (e.g. nonylphenol)
5. Hormones
6. Over 100 pharmaceuticals and personal care products
7. Single walled carbon nanotubes

To maintain long-term time series, the pilot study also included measurement of the traditional NMW list of legacy pollutants at approximately 25 existing NMW sites.

CCLEAN

Another smaller scale mussel monitoring effort in the Monterey Bay area is conducted by the Central Coast Long-term Environmental Assessment Network (CCLEAN) (Butler 1973). CCLEAN measures accumulation of legacy organics, PBDEs, and pathogens in mussels annually in the wet season at five sites in the Monterey Bay area. Mussel sampling in this program began in 2002.

Algal Biotoxin Monitoring by the California Department of Public Health

One last significant program that should be mentioned is the Marine Biotoxin Monitoring Program administered by the California Department of Public Health (CDPH) (Gassel et al. 2008). California public health officials have had a long-standing concern for protecting the public from paralytic shellfish poisoning (PSP), driven by the occurrence of 542 reported illnesses and 39 deaths attributable to the PSP toxin (saxitoxin) since 1927 (Girvin et al. 1975). The saxitoxin is produced by the dinoflagellate *Alexandrium catenella*. *Alexandrium* is normally absent or constitutes a minor component of the marine phytoplankton community along the California coast. Under certain environmental conditions this dinoflagellate may undergo periods of rapid population growth, or may produce relatively large amounts of toxin. In the fall of 1991 a second major natural toxin of concern was identified along the California coastline. Domoic acid, a neurotoxin of lower potency than saxitoxin, has become of concern because the blooms of diatoms (*Pseudonitzschia*) that produce this toxin have been of greater frequency and longer duration than most PSP events over the past 10 years. In addition, domoic acid has had dramatic impacts on marine mammal and seabird populations along the coast, raising public awareness of marine biotoxins in general. Because PSP toxicity represents a serious ongoing public health threat that requires year-round attention, the CDPH has implemented a prevention program comprised of six basic elements:

1. a coastal shellfish monitoring program;
2. monitoring of commercial shellfish product;
3. an annual statewide quarantine on sport-harvested mussels (from May 1 through October 31);
4. mandatory reporting of disease cases;
5. public information and education activities; and
6. phytoplankton monitoring.

Monitoring of saxitoxin began in 1991. The phytoplankton monitoring effort, which began a bit later in 1993, was the first volunteer-based phytoplankton monitoring program in the U.S. The Marine Biotoxin Monitoring Program employs mussels as a primary indicator species for PSP toxins because of their ability to bioaccumulate these toxins at a faster rate than other bivalve species. Mussels and other species are collected in an event-based manner. In 2010, 1041 shellfish samples were collected across the state by commercial growers, county, state, and federal agencies, and others. Saxitoxin above the alert level was detected in one sample in 2010. Domoic acid concentrations above the alert level were detected in 50 samples.

METHODS

State Mussel Watch

From 1977-2010 through the SMW and Endowment monitoring programs, the State Water Board has sustained measurement of bioaccumulation of organic contaminants and trace metals in *Mytilus californianus* at more than 20 stations along the

California coastline (Figure 1). Generally, up to 10 stations were sampled north of Pt. Conception, and the remainder (up to 20 stations) in Southern California. Chemical data have now been collected at many of these stations in a cumulative total of ten or more years, providing a sound basis for evaluating long-term trends. Not all contaminants were measured in all site-year combinations. Stations with less than ten cumulative years of data were excluded from the analysis, in order to increase confidence that the statistically significant relationships identified may constitute a real trend (α = 5%). 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. Studies in San Francisco Bay comparing these two approaches found them to yield nearly identical results (Stephenson 1992, Hardin xx).

Mussels (n=45) were collected at each station for analysis, and tissues were dissected and homogenized with a stainless steel Sorvall model 17105 Omni-mixer (1977-1978), Vertis 45 homogenizer (1979-1986), or Brinkman Polytron (1987-2004). Samples were placed in a solvent-rinsed glass container and frozen for later analysis. Extraction and analytical techniques varied somewhat over time, and have been described previously in Stephenson and Leonard (1994), Stephenson et al. (1995), and Gunther et al. (1999). Procedures employed in 1992 have been consistent since that time. In the SMW, total DDTs was defined as the sum of o,p'-DDD, p,p'-DDD, o,p'-DDE, p,p'-DDE, o,p'-DDT, p,p'-DDT, p,p'-DDMU, and p,p'-DDMS. Total PCBs was defined as the sum of Aroclors 1248, 1254 and 1260. Total chlordanes was defined as the sum of cis-chlordane, trans-chlordane, cis nonachlor, trans-nonachlor, and oxychlordane. Dieldrin was analyzed using the same methods described by Stephenson and Leonard (1994) for the other organics.

In San Francisco Bay, six locations sampled by the SMW Program from 1977-1993 were continued by the Regional Monitoring Program for Water Quality in the San Francisco Estuary (RMP) (Gunther et al. 1999, SFEI 2005). These stations represent the best dataset available on trends in organic contaminants in San Francisco Bay over the past 20 years. Although trend signals in the RMP data are obscured to some extent by the use of different analytical laboratories and methods, regression analysis of trends in DDTs and PCBs at these stations were performed for comparison. Statistical methods were the same as described for the SMW analysis. RMP analytical methods are described in Gunther et al. (1999) and SFEI (2005). In the RMP, total DDTs was defined as the sum of o,p'-DDD, p,p'-DDD, o,p'-DDE, p,p'-DDE, o,p'-DDT, and p,p'-DDT. Total PCBs was defined as the sum of 40 congeners (SFEI 2005). Total chlordanes was defined as the sum of cis-chlordane, trans-chlordane, cis nonachlor, trans-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide.

Quality Assurance (QA) programs were initiated by the SMW in 1977. QA parameters for metals consisted of standard reference materials (SRM). QA for organic contaminants consisted of method blanks, duplicates, a matrix spike and surrogate sample through 1998. An inter-calibration exercise was conducted in 1978 with the National Mussel Watch Program and good agreement was obtained (Stephenson et al. 1979). From 1986 to 1998, SMW participated in inter-calibration with the National Research

Council of Canada. Since 1996, SMW has also participated in NOAA's intercalibration program. Excellent agreement was achieved each year for the metals and organics assessed in this report. From 1998 to 2004, additional QA parameters were included in each organic and trace metal analysis such as spike recoveries, duplicates, spiked matrix duplicates, and standard reference materials. The analytical batches passed all QA checks for SRM and other QA parameters. The QA results are reported in the SMW report listed above.

Data for selected organic compounds (total chlordanes, total DDTs, dieldrin, and total PCBs) and trace metals (lead, copper, mercury, silver, and zinc) measured by SMW were examined for temporal trends. Locations that sampled 10 individual years or more were used in statistical analyses. Long-term trends were assessed using linear regression of concentrations versus year in R Statistical Software (version 2.10.0). Concentrations were log-transformed to achieve normally distributed error values and equal variances. Linear regressions were performed on lipid-normalized organic concentrations and dry weight metal concentrations. Censored data (below the detection limit) were substituted with zero values prior to statistical analysis. Log (x+1) was used in log-transformation of the zero values (for the sum of the components where applicable). This affected the slopes and trend detection for a few sites with a prevalence of non-detects. Sites with greater than 50% censored values were not evaluated for trends. Finally, at locations with declining slopes, half-lives (number of years for concentrations to decline by 50%) were calculated using the equation: $HL = \log_{10}(2)/slope$.

National Mussel Watch

Since 1986, the NOAA Status and Trends Mussel Watch Program (NMW) has analyzed organic contaminants and trace metals in numerous resident bivalve species from more than 280 stations across the United States (O'Connor and Lauenstein 2006, Kimbrough et al. 2008). From 1986-2009, consistent sampling has been performed at 35 sites in California. The majority of the original NMW stations have results for more than 15 years. The number of sites sampled in California was increased to 62 during the sample period 2007-2009. As of 2009, *Mytilus californianus* were collected at 37 stations, and *Mytilus trossulus* complex (species identified as *Mytilus edulis*, *M. trossulus* and *M. galloprovincialis*) from 25 stations (Figure 2). During the period 2007-2009, 28 stations were located north of Point Conception, with the remainder (n = 34) in Southern California.

Sample collection follows a Standard Operating Procedure (SOP) developed for the NMW. Up to about 180 mussels are collected from each site. Samples are collected by hand from three sub-locations (up to 60 mussels each) for each site and placed in plastic bags and stored on ice. Mussels are shipped to NOAA's contract labs for analysis of trace constituents (TDI Brooks in Texas) and histopathology (Rutgers University). Analytical protocols and Quality Assurance follow those approved by for the NMW Program (Lauenstein 1998). Xx surfaces cleaned? Depurated?

Analytical methods for the NMW are summarized in O'Connor and Lauenstein (2006). In the NMW (O'Connor and Lauenstein 2006), total DDTs was defined as the sum of o,p'-DDD, p,p'-DDD, o,p'-DDE, p,p'-DDE, o,p'-DDT, and p,p'-DDT. Total PCBs was defined as the sum of 18 congeners, with that sum multiplied by two. Total chlordanes was defined as the sum of cis-chlordane, trans-nonachlor, heptachlor, and heptachlor epoxide. Total PAHs included 24 compounds with 2-5 benzene rings (O'Connor and Lauenstein 2006). TBTs are expressed as the sum of concentrations of parent compound and metabolites monobutyltin, dibutyltin, and tributyltin (O'Connor and Lauenstein 2006).

Variation in the organics analyte lists among the three programs should be considered when concentrations are directly compared. The differences are most significant for the PCBs and chlordanes. For trend analysis within each program, however, these variations in analyte lists do not affect the results.

Long-term trends in trace organics and metals at the 35 most recently sampled sites were assessed based on linear regression. Censored data were substituted with zero. Analysis was conducted using log-transformed concentration data versus year. Due to issues with historic lipid data (discussed below) all concentrations were assessed on dry-weight basis. In addition to the contaminants assessed in the SMW dataset, sufficient data on total PAHs and total butyltins were available from NMW for evaluation of temporal trends. As described above for SMW, half lives were calculated for each contaminant and station with declining trends. Seven NMW stations correspond to protected locations designated as Areas of Special Biological Significance (ASBS) by the State Water Resources Control Board, and the remaining 28 were regular monitoring stations.

RESULTS AND DISCUSSION

Long-term Trends

DDTs

DDTs declined at nearly all State Mussel Watch (SMW) stations. Seventeen of 19 (90%) stations had statistically significant declines (Table 2a; Figure 3). In Northern California, initial DDT concentrations (used as a proxy for historic conditions) were generally low (100-500 ppb lw) but varied greatly among sites. Crescent City and Mad River Slough were originally SMW reference stations and thus were not that contaminated to begin with (Stephenson et al. 1995). Greater than 60% of samples at these stations were below detection. At Trinidad Head, DDTs have improved dramatically where concentrations peaked in 1977 at 730 ppb lw, declining to non-detectable in 1985. More than half of all concentrations at this site were also undetectable for DDTs.

Initial (pre-1980) concentrations at stations south of Bodega Bay were higher than in the remote Northern stations; often well above 1000 ppb lw. At Sandholdt Bridge in Moss Landing Harbor, DDTs have not changed much in more than 20 years, with concentrations in 2010 still above 15,000 ppb lw. Runoff from agricultural fields in the watershed that drains into Moss Landing Harbor was the likely historical source of contaminants at this station. In recent years, resuspension of contaminated sediment along the Salinas River and Atascadero Slough during high flow events may explain the continued presence of DDTs in bivalve tissues, decades after DDT use has ended. Whatever the cause, the data from this location demonstrate that in some situations DDT contamination of food webs can be extremely persistent. The remaining two stations in Northern California (J. Fitzgerald Marine Reserve and Pacific Grove) began with elevated concentrations (> 1000 ppb lw) and significantly declined with predicted half-lives of 9 and 14 years, respectively.

DDTs at all but one station in Southern California (13 of 14) significantly declined by 50% or more since the 1980s. The majority of locations in this region had elevated historic concentrations (> 1000 ppb lw). These trends suggest half-lives of 6-13 years. Locations in Huntington Harbor and Newport Bay had relatively high initial concentrations (> 10,000 ppb lw) because they were in close proximity to historic discharge points for DDTs (Stephenson et al. 1995). From 1947-1982, Montrose Chemical Corporation was the only manufacturer of DDTs in Southern California, and for much of that time, the largest facility in the United States (U.S. Department of Commerce et al. 2007). Declines over time at many of these southern stations can be attributed to the ban on these compounds and management of wastewater discharges in the region, along with microbial degradation and burial of contaminated sediment by cleaner material.

In San Francisco Bay, combining SMW with Regional Monitoring Program data indicated that DDTs have declined significantly at all six stations evaluated, and at similar rates to Southern California (Table 2b). Concentrations at San Francisco Bay stations declined from approximately 1000 ppb lw in the early 1980s to about 200 ppb lw in 2008 (Figure 4). The estimated half-lives at San Francisco Bay stations ranged from 6-11 years, indicating that recent concentrations are 75% lower or more than those of the 1980s. These rates are similar to Southern California locations where substantial historic concentrations existed. Stations in Los Angeles Harbor, Huntington Harbor, and Newport Bay had half-lives of 6-13 years.

Fewer significant trends were evident for NOAA's NMW program compared to SMW. DDTs indicated significant declines at 20 of 35 (57%) NMW stations (Table 2c). Similar to SMW, Northern California locations had the lowest historic concentrations (Appendix A1). Of the 35 stations, eight had initial concentrations (< 20 ppb dry weight [dw]) including the six northernmost locations. Twenty of the remaining sites (n = 27) with initial concentrations above 20 ppb dw significantly declined. One of the steepest trends was at Royal Palms, a site that is in close proximity to the Montrose Superfund Site, where DDT concentrations dropped from 1061 ppb dw in 1986 to 275 ppb dw in 2008. In addition, there were increases (but not statistically significant) at three stations: Elkhorn Slough, Moss Landing, and Emeryville. It should be noted that the highest concentration of DDT in the NMW data set was at Emeryville (2118 ppb dw in 1998) in San Francisco Bay.

Chlordanes

Declines in total chlordane concentrations were evident at the majority of SMW stations where they were evaluated and indicated similar half-lives to DDTs. Statistically significant declines were indicated at 14 of 22 (64%) stations across the state (Table 2a; Figure 5). Only one significant decline was evident in Northern California, where initial concentrations were generally low. Only Pacific Grove and Bodega Head exhibited initial concentrations above 200 ppb lw, and the former station significantly declined. Notably, chlordane concentrations at Sandholdt Bridge have not changed much from the relatively high historic concentrations (median = 32,027 ppb lw) during the 1980s, similar to the lack of decline observed for DDTs at this station.

All but one station (San Diego Bay/Harbor Island) in Southern California exhibited significant trends, with many having a half-life of 6 to 9 years. Many of the stations with significant declines in DDTs also indicated declining trends for chlordanes. Locations from Huntington Harbour to Newport Bay had the steepest declines ($p \leq 0.001$, $R^2 > 0.6$). At Newport Bay/Bay Island, where the steepest slope was indicated, chlordanes declined by nearly two orders of magnitude, from approximately 1800 ppb lw in 1981 to 28 ppb lw in 2003.

Chlordane concentrations declined at most of the NMW stations. Significant declines were observed at 16 of 25 (64%) sites (Table 2c). The largest decline was observed at Marina Del Rey, where chlordane concentrations dropped from approximately 160 ppb dw in 1987 to about 25 ppb dw in 2008 (Appendix A2). Of the 35 stations monitored, the data for 10 stations (Newport Beach; Long Beach; San Pedro Harbor; Catalina Island; Redondo Beach; Las Tunas in Santa Monica Bay; Santa Cruz Island; Point Santa Barbara; Point Conception; and San Simeon Point) were not comparable due to changes in chlordane analysis over time at one of the analysis laboratories. Half lives at NMW stations exhibiting significant trends commonly ranged from 10-20 years suggesting concentrations have not declined as quickly as at SMW stations where half lives typically ranged from 6 to 9 years. It is noteworthy that the highest concentration of chlordanes in the NMW data set was observed at Elkhorn Slough (169 ppb dw) in 2000, with no significant trend observed at this station.

Dieldrin

Dieldrin has not significantly declined at SMW stations as frequently as DDTs and chlordanes. Significant declines were indicated at only 9 of 22 (41%) stations (Table 3a; Figure 6). One reason for this is that many of the stations did not have mussels with elevated historic concentrations. Fourteen stations had initial concentrations below 500 ppb lw. All but one station (Oceanside) that showed a significant decline began with concentrations above 500 ppb lw.

Northern California stations exhibited relatively low concentrations with slower rates of decline than the southern portion of the state. However, three stations (Bodega

Head, Fort Baker, and Pacific Grove) did indicate significant declines. Sandholdt Bridge in Moss Landing Harbor had the highest concentrations in the 1980s (4800 ppb lw in 1985). Concentrations at this location were lower during the 1990s, but have since shown an apparent increase.

Significant trends in dieldrin concentrations were primarily detected at stations in Southern California with elevated historic concentrations. Linear regression indicated significant declines at 6 of 7 stations when initial concentrations were above 500 ppb lw. The remaining stations in Southern California had lower concentrations. Royal Palms, which had significant declines in DDTs and chlordanes, exhibited relatively low initial concentrations for dieldrin and have since shown an apparent increase. However, like the other legacy pesticides, stations from Huntington to Newport Bay exhibited declines from relatively high concentrations during the early 1980s. At these stations, three indicated significant trends that corresponded to half lives of 8 to 13 years. The steepest decline occurred at Huntington Harbor/Warner Ave, where concentrations were over 1800 ppb (lw) in 1983, declining to 150 ppb (lw) in 2010, a decline of more than 90%. Generally, stations showing non-significant trends and low initial concentrations (e.g., LA Harbor/National Steel, Anaheim Bay/Navy Marsh, San Diego Bay/Shelter Island) had longer half-lives than those with higher initial concentrations.

Dieldrin exhibited declining concentrations over time at most of the NMW stations. However, significant trends were only observed at 7 of 35 (20%) stations (Table 3b; Appendix A3). One site (SFB Emeryville) also indicated a non-significant increase, with the strength of the trend being relatively weak ($R^2 = 0.17$). Overall, NMW stations exhibited relatively low dieldrin concentrations. Dieldrin concentrations at the beginning of the NMW program in 1986 only exceeded 100 ppb dw at one station, Dumbarton Bridge in San Francisco Bay, where the largest decline was observed; dieldrin concentrations there dropped from 110 ppb dw in 1986 to below detection in 2009. The generally low dieldrin concentrations may be due the sampling of locations away from potential point sources, or the initiation of NMW monitoring after the significant declines had occurred in the early 1980s (Gunther et al. 1999). However, a high concentration at Emeryville in San Francisco Bay (881 ppb dw in 1998) was observed.

Summary of Trends in Organochlorine Pesticides

Organochlorine pesticide concentrations in mussels measured by the SMW, RMP, and NMW programs are continuing to show long-term declines. Our evaluation has shown that many stations that were highly contaminated with DDTs, chlordanes, and dieldrin during the 1980s (e.g., Huntington Harbor and Newport Bay), have continued to decline over time. However, concentrations of dieldrin did not tend to be as high, and thus suggested the least frequent number of declines relative to DDTs and chlordanes. These trends are generally consistent with a previous state-wide assessment of legacy pesticides in SMW mussels. From 1977-1992, Stephenson et al. (1995) showed that half of the stations evaluated had declined significantly in DDTs (15 of 32) and chlordanes (14 of 28). In the evaluation of trends performed in this study, the similarly high proportion of SMW stations showing declines, particularly in Southern California, suggests that these trends have continued. The ability to detect trends was improved in our evaluation by using data spanning a longer period of time. Similarly, the NMW stations have declined at many of the stations monitored through 2009 (e.g., 20 of 35, 57% DDTs). The declines in Southern California are most likely the combined result of cleanup of discharges from the Montrose facility, improved wastewater treatment processes, and gradual improvements in water quality after the use restrictions. Declines in legacy pesticides have also been observed in biological matrices other than mussels. Sport fish in Southern California have shown significant declines over the past 20 years, especially for DDTs (Schiff and Allen 2000, Davis et al. 2006).

PCBs

Significant trends in PCBs were observed at over half (11 of 18, 61%) of the SMW stations. The majority of stations with appreciable (> 2000 ppb lw) initial concentrations (Table 3a; Figure 7) declined significantly. As with DDTs and chlordanes, PCBs in the northern part of the state generally exhibited low initial concentrations and a prevalence of "non-detects". At the four northernmost locations (Crescent City, Trinidad Head, Mad River Slough, and Bodega Head) 50% or more of observed concentrations were below detection (all Aroclors were below detection). These sites were relatively unaffected by local PCB sources. However, considerable inter-annual variability was evident, largely due to a lower signal-to-noise ratio for concentrations near the limit of detection. The only SMW site in Northern California with high initial concentrations was Sandholdt Bridge. This was one of the most contaminated sites across the state and showed a weak decline.

PCBs have significantly declined at all six RMP stations. PCBs in San Francisco Bay have shown two distinct temporal patterns (Davis et al. 2007). 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 (Table 3b; Figure 4). For the southern Estuary locations (Treasure Island/Yerba Buena Island, Hunter's Point/Alameda, Redwood Creek, and 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 half-lives varying from 6 to 15 years.

Many long-term SMW monitoring sites in Southern California exhibited very high initial PCB concentrations (> 5000 ppb lw) followed by significant declines. Only one station in Southern California had low initial concentrations (Oceanside). Of the 14 stations in Southern California, 10 showed significant declines, with many having a half-life of 7-12 years. Overall, these data suggest large declines in PCBs during the past 20 years. However, current concentrations at some locations such as Newport Bay/Rhine Channel remain relatively high (median = 17,500 ppb lw).

While PCB*s* declined at most of the NMW stations, fewer significant trends were indicated in comparison to SMW (Table 3b). Of the 35 sites monitored, 8 had low initial concentrations (<10 ppb dw) and thus were not expected to show significant declines

(Appendix A4). Of the remaining 27 sites with appreciable PCB contamination (> 10 ppb dw), 20 showed no significant trend, and seven exhibited significant declines. Three of the sites with declines were in Northern California (Humboldt Bay and San Francisco Bay/San Mateo Bridge and Dumbarton Bridge). The largest statistically significant downward trend was at Mission Bay in Southern California, where PCB concentrations dropped from approximately 100 ppb dw in 1988 to about 20 ppb dw in 2008. The highest initial PCB concentration for all NMW sites was 650 ppb dw measured in 1988 at San Diego Bay/Harbor Island. In 1990 the concentration there had increased to 1008 ppb, and while levels are currently lower there has not been a significant decline.

The rate of decline of PCBs in transplanted mussels in San Francisco Bay was previously estimated at 50% every 20 years (Davis et al. 2007). In this study, 11 SMW stations across the state had appreciable historic concentrations that have significantly declined. At these stations, PCBs had half-lives of 8 to 30 years. The majority (9 of 11) of stations indicated declines of 50% within 10 years. In San Francisco Bay, 50% declines were predicted in 6 to 14 years. These time series suggest faster rates of decline than have been reported previously.

PCBs have generally shown fewer significant declines than organochlorine pesticides across the state. Several factors could explain this pattern. First, DDTs have not been in use for more than 30 years, while a considerable amount of PCBs remain in use today. In San Francisco Bay for example, 200,000 kg were reported to be in use in transformers from 1998-2001 (USEPA 2004). Continued releases of PCBs to the environment may therefore be one factor contributing to less frequent detection of declines in PCBs with the SMW dataset. Second, PCBs may have longer residence times than organochlorine pesticides due to slower degradation rates in either aerobic environments in watershed soils and sediments, or in anaerobic environments in aquatic ecosystems.

PAHs

Examination of trends in PAHs was only possible with the NMW dataset. Of the 35 stations evaluated, 25 had appreciable concentrations of total PAHs (> 20 ppb dw) and the remainder had low concentrations (Appendix A5). Twenty-three out of 35 sites had increasing trends, but only three of these increases were statistically significant: Humboldt Bay, Point Delgado/Shelter Cove, and Point Santa Barbara (Table 4). The highest PAH concentrations (48,000 ppb dw) were observed at Yerba Buena Island in San Francisco Bay in 2008, following the Cosco Busan oil spill, though insufficient data were available to assess the trend at this site.

Three NMW sites had statistically significant declines, including an additional eight stations that showed lower concentrations in recent years. The stations with significant declines indicated half lives of 9-17 years, which was generally slower compared to other organic contaminants, such as DDTs and PCBs.

PAH concentrations increased in California in the 1900s as historic sources were augmented by fossil-fuel combustion (Pereira et al. 1999). With human population and automobile use expected to increase in California (ABAG 2002), the extent of PAH contamination may also increase. A particular concern is chronic PAH loading from multiple sources in highly urbanized water bodies, such as San Francisco Bay and the Southern California Bight. However, the correlation analyses of O'Connor and Lauenstein (2006) indicated that PAHs have not changed significantly at many locations. For example, in San Francisco Bay where PAH contamination might have been expected to increase, initial concentrations in mussels were relatively high, but have not changed significantly. A lack of significant trends was also found for transplanted mussels at ten stations in San Francisco Bay from 1993-2001 (Oros and Ross 2005). Therefore, the significant increases observed by the NMW at sites like Humboldt Bay and Point Santa Barbara are inconsistent with trends in mussels reported previously in NMW and the RMP. PAH concentrations (Appendix A5) were quite variable over time; these results may therefore also be artifacts of this variability rather than real trends over time.

Tributyltins (TBT)

TBT was another contaminant where trends could only be assessed with the NMW data. TBT declined at 32 of 35 stations, and 11 (31%) of these declines were statistically significant (Table 4; Appendix A6). The majority of locations (22 of 35) exhibited initial concentrations that were well above 50 ppb dw.

Locations with the steepest declining trends were from San Francisco Bay (San Mateo Bridge and Dumbarton Bridge), Santa Cruz, Marina Del Rey, Anaheim Bay, Royal Palms, Mission Bay, and Pt. Loma Lighthouse. Many of these monitoring stations are located in harbors. Half-lives at these stations were estimated at 4 years, which was the quickest decline observed in any contaminant evaluated, representing a major success story. Notably, 16 other stations with non-significant declining concentrations, also revealed relatively low half lives (< 10 years). TBT was historically used in anti-fouling coating on boat hulls. The declining trends evident in the NMW data undoubtedly reflect the reductions in loads of TBT that occurred during the phaseout of TBT-based hull coatings in the 1990s and early 2000s.

Lead

Lead concentrations declined at many of the SMW stations with elevated concentrations (> 1 ppm dw). Significant declines were indicated at 11 of 21 (52%) stations (Table 5a; Figure 8). The steepest trends were observed at Pacific Grove, Los Angeles Harbour, Royal Palms, Huntington Harbour, Newport Bay, and Oceanside. None of the locations with initial concentrations below 1 ppm dw showed any trend in concentrations (e.g., Trinidad Head and Bodega Head). The majority of stations showing declines were located in Southern California (9 of 13), from Los Angeles Harbor to Oceanside. The steepest decline was at Royal Palms, where concentrations declined from 15 ppm dw in 1977 to approximately 1.8 ppm dw in 2010.

Fewer significant trends were evident at NMW stations compared to SMW. While lead declined at most of the NMW stations, significant declines were observed at 8 of 35 (23%) stations: San Diego Bay/Harbor Island, La Jolla, Anaheim Bay, San Pedro Harbor, Royal Palms, Marina Del Rey, Moss Landing, and Emeryville in San Francisco Bay (Table 5b; Appendix A7). Half-lives at these stations ranged from 9-28 years. 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.

Lead historically entered aquatic environments via leaded gasoline and urban runoff (Stephenson and Leonard 1994). 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 1971-1995, the combined mass emissions of lead from the four largest municipal wastewater treatment facilities in Southern California decreased by nearly 99% (Raco-Rands 1996). SMW stations Royal Palms to Oceanside are all located near these treatment facilities, and thus not surprisingly show the strongest trends. These results clearly indicate that reductions in mass emissions of lead from wastewater discharge, urban runoff, and atmospheric deposition have reduced lead contamination at the base of the food web.

Silver

Significant trends in silver were indicated at nearly half of SMW stations (10 of 21, 48%). Initial silver concentrations only exceeded 1 ppm dw at four stations (Table 5a; Figure 9). These historically contaminated stations indicated the steepest declines (> 0.01 ug/g/yr): San Francisco Bay/Fort Baker, Pacific Grove, Royal Palms, and Oceanside. Half lives at these stations ranged from 6-12 years. The majority of stations had relatively low initial concentrations of silver (0.02 – 0.6 ppm dw), longer half lives, and have not shown any consistent trend.

Silver declined at most of the NMW stations (Table 5b; Appendix A8). However, most of these trends were non-significant. Initial silver concentration only exceeded 1 ppm dw at less than one-third of stations (10 of 35). Out of these stations, significant declines were observed at 10 sites: Imperial Beach, Point Loma, La Jolla, Newport Beach, Royal Palms, Redondo Beach, Point Santa Barbara, San Simeon, Santa Cruz, and San Mateo Bridge in San Francisco Bay. With a few exceptions, half lives at the majority of these declining stations ranged from 4- 15 years. Eleven stations showed increasing concentrations, but none of these trends were significant. The largest statistically significant downward trend was at Point Loma, where silver concentrations dropped from a high of 34 ppm dw in 1991 to about 2 ppm dw in 2005.

Silver has been used in a variety of industries in California over the past 30 years, the most prominent being photography. During the late 1970s, silver concentrations in *M. californianus* and *M. edulis* from San Francisco Bay were equal to or higher than mussels from more than 60 estuaries and coastal locations across North America (Opperhuizen and Sijm 1990). Extraordinarily high concentrations of silver in coastal waters during this time have almost exclusively been attributed to municipal and industrial wastewater discharges (Luoma and Phillips 1988, Hornberger et al. 2000). 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 SMW 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 located 500 yards from an outfall that was terminated in 1980, which would explain the dramatic decrease. From 1971 to 1995, mass emissions of silver from large wastewater treatment facilities in Southern California decreased by 64% (Raco-Rands 1996). Decreases in emissions from large municipal wastewater facilities in this region, such as the Joint Water Pollution Control Plant near Royal Palms, explain the significant trends. The declining use of film-based photography has contributed to decreased silver emissions. The remaining Southern California stations have not indicated trends because they were never that contaminated to begin with. Only 3 of 10 Southern California stations had initial concentrations above 0.1 ppm.

Copper

Copper measurements at SMW stations suggest that concentrations are increasing across the state. All but two stations had an increasing trend in copper concentrations. Half of the stations (11 of 22) had initial concentrations above 10 ppm dw (Table 6a; Figure 10). Of these stations with already elevated concentrations, six indicated significant increases. Additionally, eight stations had lower initial concentrations (< 10 ppm dw), and two of these significantly increased. Three stations that exhibited increases in Southern California were in harbor areas. Furthermore, one station (Royal Palms) suggested that copper concentrations have significantly declined, but the trend was relatively slow (half-live > 100 years).

Trends in copper at NMW stations have been inconclusive. Eleven of 35 stations had initial concentrations above 10 ppm dw (Table 6b; Appendix A9). Ten stations exhibited statistically significant trends, comprising 4 increases and 6 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. Four stations exhibited significant increasing trends, and had initial concentrations below 10 ppm dw. In summary, the trends in copper from NMW stations were inconsistent from a statewide perspective, and did not suggest a general pattern of increasing concentrations as evident from the SMW dataset.

One primary source of copper in the marine environment is antifouling paints on boats and other water vessels. On a statewide basis, copper use has increased widely since the 1970s (Lauenstein et al. 1998) and new co-polymer anti-fouling paints on piers, marinas, or boats may be the source of increasing copper levels. There is an additional concern that copper may be increasing in urban runoff due to use in brake pads. Estimates in 2003 indicated that 240,000 kg of copper were released in the San Francisco Bay watershed due to human activity that year (Rosselot 2006). Of this total, 36% could be attributed to copper released from brake pads. Inputs of copper to San Francisco Bay from brake pads are a topic of ongoing study by the Brake Pad Partnership (http://www.suscon.org/bpp/index.php). However, neither the SMW nor NMW stations in San Francisco Bay have indicated increasing trends in copper. One long-term dataset going back to the mid-1970s documented substantial declines in copper in clams (*Macoma balthica*) in the vicinity of the Palo Alto wastewater treatment plant (Hornberger et al. 2000). Use as a wood preservative is another important source of copper, as is use as a pesticide in agricultural and residential applications.

Zinc

Few SMW stations had significant trends in zinc concentrations. Two northern stations and nine southern stations has initial concentrations above 200 ppm dw (Table 6a; Figure 11). Of these 11 stations with appreciable concentrations, four significantly declined (Crescent City, Sandholdt bridge, Royal Palms, and Oceanside), and one site significantly increased (San Diego Bay/Harbor Island). In all cases, the amount of variation in zinc concentrations explained by time was low (R^2= 0-0.5), suggesting that other variables significantly contributed to observed fluctuations.

Zinc concentrations declined slowly at most of the NMW stations (Table 6b; Appendix A10). Ten stations (29%) exhibited significant declines. Estimated half-lives were the highest of all the trace metals, commonly exceeding 30 years. The largest statistically significant downward trend was at San Francisco Bay's Yerba Buena Island (not one of the 35 sites with an extensive sampling record), where zinc concentrations dropped from approximately 220 ppm dw in 2003 to about 160 ppm dw in 2009. Five stations indicated increasing concentrations, but none of the trends were significant.

Previous evaluation of long-term trends in zinc concentrations in San Francisco Bay mussels indicated no temporal trend from 1980 - 1996 (SFEI 1998). The lack of trends in zinc at SMW stations may be due to the influence of sediment-water interactions on the bioavailability of zinc to mussels (Rivera-Duarte and Flegal 1997). Furthermore, other bivalve species (e.g., the fresh water clam *Corbula*) have been shown to regulate their body burdens of zinc in north San Francisco Bay (C. Brown, USGS, personal communication) and elsewhere (Phillips 1988), which may account for the large proportion of unexplained variability in long-term trends.

Mercury

Mercury concentrations declined significantly at the over half (9 of 16, 56%) of SMW stations (Table 7a; Figure 12). However, much of the variability in mercury concentrations was unexplained (maximum R^2= 0.49). The three significantly declining sites in Northern California (Bodega Head, Sandholdt Bridge, and Pacific Grove) had relatively high initial concentrations (~ 0.2 ppm dw). Similarly, most of the locations in Southern California that exhibited declines (e.g., LA Harbour/Consolidated Slip and Royal Palms) were also relatively high. Half-lives at stations with trends ranged from

15-36 years. Generally, mercury concentrations were low at the majority of stations not indicating trends and showed inconsistent patterns in concentrations over time.

Mercury concentrations have declined slowly at most of the NMW stations. Fifteen (43%) of these declines were statistically significant (Table 7b; Appendix A11). Only two of the 15 sites with declines were located in Northern California. The thirteen stations with significant declines in Southern California all exhibited slopes that equated to half-lives of > 80 years. The largest statistically significant decline was at San Diego Bay Coronado Bridge, where mercury concentrations dropped from 0.18 ppm dw in 1989 to about 0.07 ppm dw in 2007.

Comparison of the Mussel Watch Programs

Strengths and Weaknesses of the State Mussel Watch Program

Examination of the data generated through the State Mussel Watch Program has illustrated the utility of mussels for detection of temporal changes in contaminant concentrations at coastal locations across the state. Sampling of historic discharge points has revealed significant long-term trends in both organic contaminants (particularly, PCBs, DDTs and chlordanes) and trace elements (particularly lead and silver). In addition, the current set of SMW stations being sampled represents the longest running time series on bivalve tissue chemistry information available from anywhere in the state (1977-2010). Furthermore, the continuing time series generated by annual sampling of mussel watch stations has provided high statistical power for evaluating long-term trends. Recently, the power to detect 3.5% annual declines in PCBs and DDTs for RMP stations over the next 20 years with this dataset was shown to be greater than 99% (Melwani et al. 2008).

The sampling approach of SMW has been to target locations rather than species. To overcome the difficulty in obtaining adequate samples of the same species at specific locations of interest, transplanted bivalves have been used. This approach has been particularly effective for evaluating responses to management actions and progress of cleanups in Southern California. Furthermore, there have been no obvious differences in detection of trends with residents versus transplanted bivalves in this program, suggesting that the information they provide is essentially the same. Although transplants may have less relevance for evaluating ecological risks, they offer the significant advantage of being excellent indicators of contaminants that enter the food web over the 90-day deployment period.

Finally, one technical aspect of the SMW design should be highlighted as it may have inhibited detection of trends for PCBs. The SMW Program shifted its procedures for analyzing PCBs over time. PCBs measured as Aroclors were available for the most number of years, and therefore these data were used for our analyses. However, concentrations reported as Aroclor equivalents are less precise than data reported on a congener-specific basis. Trends in PCB concentrations would be more effectively evaluated using PCB congener data.

Strengths and Weaknesses of the National Mussel Watch Program

NOAA's Mussel Watch Program has produced valuable information on the distribution of and long term-trends in concentrations of organic contaminants and trace metals in California. The NMW approach has been to primarily sample representative coastal locations across the state, away from point sources of contamination. Effort was particularly made to avoid sampling near historic discharge points, such as outfalls from industrial or municipal wastewater treatment plants (O'Connor and Lauenstein 2006). Therefore, the information generated by the NMW has been different from the SMW. NMW data are useful for documenting background concentrations and contamination away from point sources of contamination. The SMW Program has targeted locations of interest with known historic contamination and reference areas for comparison, which has identified improvements in water quality over time, particularly near sources. As a result, the NMW Program has not documented occurrence of significant temporal trends to the same degree as SMW, particularly in Southern California.

The review of temporal trends at NMW stations across the state has identified certain limitations to their approach. The collection of different resident species is a concern due to lesser comparability across sites. Although a relatively large geographic distribution of locations can be examined by using *Mytilus* spp., there is concern that different bioaccumulation potential among *Mytilus* species may have obscured spatial comparisons. Considerable differences in accumulation of organics and metals are commonly found among bivalve species (Segar et al. 1971, O'Connor and Lauenstein 2006). In addition, the use of solely resident animals limits the ability of the NMW Program to target specific locations of interest. However, this is balanced by the benefit of higher ecological relevance through the collection of resident organisms.

The SMW data are somewhat better suited for evaluating long-term trends than NMW due to annual sampling of the same stations over time (1977-2010). Annual sampling provides substantially more power for trend detection than do more sites sampled less frequently (Melwani et al. 2008). NMW shifted from annual to biennial sampling in 1994 (O'Connor and Lauenstein 2006). Consequently, the NMW dataset presently has less power to detect trends than SMW.

Finally, certain limitations to evaluating long-term trends in the NMW Program were apparent, due to differing analytical procedures and changes in laboratories. First, NMW has analyzed a limited list of only 18 PCB congeners for the majority of years. The NMW congener list excluded two of the dominant congeners in bivalve tissues, PCB 101 and PCB 149. Second, in attempting to lipid-normalize organic contaminant concentrations for this study; changes in lipid determination methods were identified, which dramatically affected concentrations. From 1986-1989, Scientific Applications International Corporation (SAIC) analyzed the west coast samples. From 1990-1994, Battelle took over. Then, in 1995 Geochemical and Environmental Research Group

(GERG) at Texas A&M University began analyzing all the NMW samples from around the US. Much lower lipid values were observed beginning in 1995. Differences in the lipid extraction methods employed may explain these observations. For this reason, organic trends were not lipid normalized in the analyses conducted for this report.

Locations of Special Interest in California

San Francisco Bay

Data for other biological matrices in San Francisco Bay have indicated a lack of declines of organic contaminant and trace element concentrations. Organic contaminants in San Francisco Bay sport fish (e.g., white croaker, shiner surfperch, and California halibut) have shown no decline over time (Davis et al. 2011). Concentrations in water and sediments of the Bay have also been relatively constant (SFEI 2005). The results shown here for transplanted mussels indicate statistically significant declines in organics at numerous stations in the Bay. The NMW data also indicate significant declines in TBT and certain organic contaminants in San Francisco Bay: chlordanes significantly declined at all three Bay stations; TBT, DDTs, and dieldrin declined significantly at 2 of 3 sites; and PCBs declined significantly at one site. One possible explanation for these contrasting patterns is that the sport fish are consuming prey that receive their exposure in the margins of the Bay, where residence times for particle-associated contaminants are longer and recovery rates are slower, while the mussels are reflecting declines in water and sediment in the open Bay. RMP surveys of mercury and PCBs small fish in recent years (Greenfield et al. 2011) have found high concentrations that support this hypothesis.

Trace metal concentrations generally have not changed significantly in the Bay. SMW sampling only revealed declines in silver, which also declined significantly in most other areas of the state. NMW also did not show many significant trends with metals in the Bay, except that Emeryville declined significantly for silver and total mercury, and San Mateo Bridge declined in zinc. The lack of widespread, significant declines in metals is somewhat surprising since large improvements in effluent quality have occurred during the past 20 years of these monitoring programs (Hornberger et al. 2000). Therefore, these results may point to the importance of other pathways for trace metal loading to the coastal environment, such as urban runoff. Schiff et al. (2000) presented a summary of potential sources of pollutants to the Southern California Bight. As in San Francisco Bay, emissions from POTWs around the Bight have decreased over time due to improvements in controls and treatment processes. However, flows from urban runoff have remained constant or increased. The authors indicated that higher flows have occurred as a result of more impervious surface such as concrete being used in urban, municipal, and industrial areas along the coast. Furthermore, surface runoff to the Bight in 1995 was shown to discharge higher loads of trace metals, particularly, copper, lead, and zinc, than all other sources combined. This troublesome situation may also be a concern in San Francisco Bay, where urban runoff receives no treatment before being discharged, and thus may contribute to the lack of trends in copper, mercury, and zinc observed for San Francisco Bay.

Sandholdt Bridge

Over the many years of mussel monitoring in California, organic contaminants monitored at Sandholdt Bridge have been the highest of all stations in Northern California. For example, initial DDT concentrations in Northern California sites were below 800 ppb lw other than at Sandholdt Bridge, where concentrations exceeded 30,000 ppb lw. Concentrations of contaminants at Sandholdt Bridge were lower during the early 1990s, but otherwise they have not changed much from pre-1980 levels.

Sandholdt Bridge is located in Moss Landing Harbor at the downstream end of a large drainage area for agricultural fields and farms. The unusually high historic concentrations at this site likely originate from the application of organochlorine pesticides and subsequent runoff to the Salinas River and Atascadero Slough that drain into Moss Landing. Contamination of mussels at Sandholdt Bridge has continued through to recent times, possibly due to high flow events and resuspension of contaminated sediments in the rivers. High flow events move newly eroded sediment downstream and result in increased suspended sediments in the water column that are available for uptake by biota such as the filter feeding *Mytilus californianus*. The relatively low concentrations of organic contaminants during the early 1990s reported by Stephenson et al. (1995) may reflect low runoff years, and the relatively high levels 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 (Figure 13) corroborates this hypothesis.

Palos Verdes

Many of the southern locations in the state showed declines across numerous contaminants and trace metals (e.g., Royal Palms). The SMW and NMW 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 L.A. County Sanitation District outfall at White's Point that discharged wastes from the Montrose Chemical Corporation. Industrial waste produced by the Montrose Chemical Corporation, 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). The wastewater outfalls on the Palos Verdes Shelf (offshore of White's Point) were a principal source of releases of DDTs to this region and were one of several significant sources of PCBs, in addition to the other large wastewater discharge points, such as the Orange County Sanitation Districts (SCCWRP 1973). Therefore, the declines in organic contaminants and many trace metals evident at Royal Palms and other coastal locations in this region were undoubtedly influenced by reduced loads from POTW effluent discharges in the region. One of the metals in the NMW program that declined significantly at Royal Palms and two other neighboring stations (Marina Del Rey and San Pedro Harbor) was lead. This is a result of the phaseouts of leaded gasoline and lead-based paints that have occurred since the 1970s. Mussel monitoring in Southern

California has documented considerable improvement from the historic contamination of the late 1970s and early 1980s.

PASSIVE SAMPLING DEVICES (PSDs): AN ALTERNATIVE OR COMPLEMENT TO BIVALVE SAMPLING

In contrast to methodologies that require costly energy and instrumentation and long lead times to accurately determine ultratrace contaminant levels in ambient water, PSDs are simple, relatively rapid, low cost alternatives that rely on diffusive mass transport to concentrate chemicals of interest. Over the past 20 years, PSDs have been designed and optimized to measure a wide variety of target analytes, including trace metals (Blom et al. 2002), organics (Stuer-Lauridsen 2005) and organometallics (Aguilar-Martinez et al. 2008). Their potential for providing time-averaged concentrations, spatial and temporal trends, and source characterization in a more cost-effective manner has garnered attention from large collaborative monitoring networks, including the European Union's Water Framework Directive (Allan et al. 2006).

The most common PSD for metals utilizes diffusive gradients in thin films (DGTs) to selectively capture divalent cations (e.g., Cu^{2+}, Zn^{2+}, and Pb^{2+}) after diffusion through a thin membrane (Davison and Zhang 1994). Post-exposure acid extraction of the capture medium (typically an ion exchange resin) and subsequent analysis by atomic absorption, inductively couple plasma mass spectrometry, or other suitable detector is required. Aqueous concentrations of labile species are estimated from sampling rates pre-determined in lab calibration experiments. DGT samplers have been tested in natural waters including seawater (Twiss and Moffett 2002) as well as in urban runoff and stormwater (Aung et al. 2008). A recent study (Hintelmann et al. 2011) employed DGTs to evaluate spatial patterns in methylmercury throughout San Francisco Bay.

Similar to DGTs, samplers that employ a diffusion or protective membrane include lipid-filled semipermeable membrane devices (SPMDs) (Huckins et al. 1993) or those that contain sorptive disks (Kingston et al. 2000) to target and sequester hydrophobic organic compounds (HOCs). As its name suggests, the polar organic chemical integrative sampler (POCIS) is a recent adaptation of the SPMD designed for polar analytes (Petty et al. 2004), including several classes of emerging contaminants of concern. In contrast, samplers that rely on direct sorption of organic contaminants to a polymeric matrix utilize solid phase microextraction (SPME) fibers - glass capillaries tubes coated with a thin polymeric coating (Arthur and Pawliszyn 1990) - or thin polymer sheets, strips, or tubing composed of low density polyethylene (LDPE) (Adams et al. 2007), polysiloxane (silicone), or other polymeric material.

PSDs can be employed under kinetic or equilibrium conditions. Regardless of mode, laboratory pre-calibration is required to determine uptake rate constants or equilibrium distribution or partition coefficients for each analyte of interest (Mayer et al. 2003). DGT samplers can detect parts per trillion concentrations of certain metals whereas PSDs for organics are capable of substantially lower levels, particularly for very hydrophobic substances (Zeng et al. 2005, Adams et al. 2007). PSDs incorporating membranes are prone to fouling which attenuates transport and typically results in underestimation of concentrations (Prest et al. 1992, Webb and Keough 2002). Exposure times required for equilibrium samplers without membranes (e.g., SPME or LDPE) may exceed several weeks for specific analytes. Pre-loading of performance reference compounds within these devices may eliminate the need for lengthy equilibration times while improving measurement accuracy.

Comparative Studies of PSDs and Bivalves

Few studies have compared bivalve tissue levels with PSDs. For trace metals, DGT samplers were compared with tissue levels in mussels (*Mytilus galloprovincialis*) from two Australian harbors (Webb and Keough 2002). Whereas DGTs were able to replicate spatial differences in tissue concentrations of Cu, Pb and Zn, more subtle differences in tissue accumulation (e.g., seasonal variation) were not apparent in PSD measurements. In addition, tissue trends for Cd were not predicted by DGT and a correction for fouling that was proposed by the authors was not subject to validation. For PCBs and selected organochlorine pesticides, SPMDs replicated trends observed in freshwater clams (*Corbicula* spp.) but levels were underestimated and differences in accumulated homolog distributions were observed (Prest et al. 1992). In another study, total PAH in native mussels (*Mytilus trossulus*) and SPMDs deployed in an oiled intertidal environment were found to be correlated; however, the authors also reported differences in accumulated PAH profiles (Boehm et al. 2005). Moreover, blank contamination associated with pre-cleaned SPMDs precluded low-level assessment of 2-3 ring PAHs. Subsequently, low molecular weight PAH including naphthalenes have been detected in commercially available LDPE used for construction of polyethylene devices (PEDs), even after exhaustive solvent cleaning of the virgin material (W. Lao unpubl. data).

SCCWRP and collaborators are currently evaluating SPME and PEDs for quantifying the dissolved or “bioavailable” fraction of HOCs in situ. Little or no published data comparing bioaccumulation by bivalves with these types of PSDs are currently available. Thus, SCCWRP and the NOAA NS&T program deployed SPME water column samplers (Zeng et al. 2004) with a protective antifouling housing at 15 of the 34 sites established in Southern California during the 2007-08 collection season (Diehl et al. 2007). Roughly half of the mussel-SPME sites are located within embayments where contaminant levels are expected to be elevated compared to nearby open coastal sites. SPME concentrations for target PAH, PCBs, and organochlorine pesticides will be compared directly with corresponding mussel tissue when data become available. In another SCCWRP study, several pyrethroid insecticides were detected by PEDs deployed in Ballona Creek, a 303d-listed urban estuary in Los Angeles County. These detections were corroborated by results obtained from an *in situ* pumping system as well as from bedded sediments collected and analyzed using GC-ECD (W. Lao and K. Maruya unpubl. data).

PSD Recommendations

The promising performance and lower unit cost associated with PSDs underscore their utility as a supplemental or alternative sampling strategy for bioaccumulative chemicals. Whereas bioaccumulation is best monitored using target or sentinel organisms that are most relevant to the receptor of interest, other factors such as physiological anomalies, natural abundance, and suitability of habitat as well as cost need to be considered when designing monitoring studies. Clearly, sessile organisms such as bivalves are among the most widely utilized bioaccumulation sentinel due to their ability to reflect the local environment. Because native organisms clearly integrate contaminant exposure over the longest possible time duration, they best reflect ambient conditions over the long term. Transplanted mussels retain the biological significance associated with accumulation by native organisms; however, substantial differences in tissue levels with respect to native species have been reported, even after 2-3 months of exposure (Booij et al. 2002). PSDs represent an alternative to either type of biosentinel by providing quantitative levels for the dissolved or labile fractions of contaminants while remaining immune to biological and/or physiological confounding factors. PSDs may also be deployed in areas that are inhospitable to either native or transplanted mussels. Lastly, because of their lower per unit cost, PSDs may be used in conjunction with native or transplanted mussels to enhance spatial or temporal coverage. The development and validation of PSDs as a viable water quality monitoring alternative should thus be considered in moving toward a comprehensive, flexible, and cost-effective approach to bioaccumulation monitoring in coastal waterways.

RECOMMENDATIONS FOR FUTURE BIVALVE MONITORING IN CALIFORNIA

As described in the Introduction, the SWAMP bioaccumulation monitoring element was established to address objectives and assessment questions in four general topic areas (Table 1): status, trends, sources and pathways, and effectiveness of management actions. This section discusses the potential role of future bivalve monitoring in addressing these objectives.

Status

The primary importance of bivalves as status biosentinels stems from their value in monitoring biotoxins in coastal waters. CDPH has established an effective program (the Preharvest Shellfish Protection and Marine Biotoxin Monitoring Program: www.cdph.ca.gov/healthinfo/environhealth/water/Pages/Shellfish.aspx) for monitoring marine biotoxins that includes analysis of mussels, other shellfish species, and phytoplankton. Mussels are particularly valuable for this monitoring because they accumulate toxins at a faster rate than other bivalve species. A significant risk of human illness or death due to biotoxins is the impetus for an annual quarantine for sport-harvested mussels along the entire coastline, including bays and estuaries, from May 1 through October 31. In October 2011 the Department of Fish and Game issued a closure of the abalone fishery in Sonoma County due to an observed die off of abalone and other invertebrates in response to a harmful algal bloom. A biotoxin (microcystin) produced in freshwater coastal lakes and transported downstream has also been implicated in the recent deaths of 21 sea otters (Miller et al. 2010). Miller et al. (2010) also found that bivalves accumulate microcystin to concentrations 100 times greater than those in water and only slowly depurate these chemicals. Monitoring biotoxins in mussels is clearly a high priority for protection of human and wildlife health. The CDPH program is partially addressing this need. Additional monitoring may also be needed to adequately characterize biotoxin accumulation in coastal food webs and potential risks to humans and wildlife.

Bivalves also have some value as secondary indicators of status with regard to human exposure to toxic metals and organic chemicals. Fish are generally more valuable indicators of human exposure, however, due to the higher rates of fish consumption and the higher concentrations of pollutants that fish accumulate. Consumption advisories for shellfish due to chemical contamination are rare in California. One recent exception was in San Francisco Bay for a brief period after the Cosco Busan oil spill in 2007 (Brodberg 2007). Another instance of consumption guidelines for shellfish is for clams in the Delta region, where the most sensitive population is advised to eat no more than three servings a week (e.g., Gassel et al. 2008). Overall, the degree of human exposure and risk due to contaminants in shellfish is much lower than it is for sport fish due to lower concentrations of contaminants and lower consumption rates.

Bivalves have also been evaluated as indicators of effects on aquatic life, including the bivalves themselves. For example, in San Francisco Bay, selenium concentrations in clams (*Corbula amurensis*) have been studied as an indicator of exposure to higher trophic level species such as white sturgeon and diving ducks (Stewart et al. 2004). The SMW, NMW, RMP, and other studies have also evaluated the effects of contaminants on bivalves themselves. The extreme toxicity of tributyltin to bivalves was a primary impetus for the ban on the use of this compound as an anti-fouling agent. For other contaminants, however, effects on bivalves have not been sufficiently overt to drive regulatory action.

Bivalves are also valuable biosentinels of exposure and potential risk for lower trophic level species from contaminants of emerging concern. The limited metabolic activity of bivalves relative to vertebrate species makes them useful indicators of many emerging contaminants that are accumulated in lower trophic levels but not transferred up the food chain. Even though these contaminants generally do not accumulate in higher trophic level species, they can potentially cause endocrine disruption or other adverse effects to both lower and higher trophic level species.

Trends

As demonstrated in this report, bivalves have great value as indicators of long-term trends in bioaccumulative contaminants. Bivalve monitoring in California has documented distinct general statewide declines in many contaminants that have been subject to use restrictions or source control, including organochlorine pesticides, PCBs, TBT, lead, and silver. Bivalve monitoring has provided of the best available time series demonstrating the effectiveness of the management actions taken for these contaminants. Bivalve monitoring has also shown that contaminants with continuing emissions (e.g., PAHs and copper) are still accumulating in aquatic food webs across the state, and are on the increase in many locations.

For some contaminants, such as PAHs, bivalves are clearly superior to sport fish as biosentinels because of the greater capacity of fish to metabolize certain classes of chemicals. PAHs do not accumulate in fish, but reach easily measurable concentrations in bivalves. Invertebrates (including bivalves) represent the primary vector through which these compounds reach and affect vertebrates.

The more limited metabolism in bivalves also makes them excellent biosentinels for emerging contaminants. The utility of bivalves for CEC monitoring is being thoroughly tested in the NMW California pilot study. Bivalves appear to be an essential component in monitoring for early detection and management of CECs in the aquatic environment.

The existence of historical time series for contaminants of concern in bivalves is also of great value, especially for contaminants (e.g., PCBs) and locations where problems still persist. The existence of archived specimens is also valuable for retrospective studies of emerging contaminants or for obtaining more accurate data using improved analytical methods. Unfortunately, limited archives are available for samples collected prior to the late-2000s by the major programs in California (SMW, NMW, and RMP). In recent years the RMP (2010) and SWAMP (Bioaccumulation Oversight Group 2011) have begun systematic archiving of samples of bivalves, fish, and other tissues on a long-term basis.

Sources and Pathways

Bivalves have some value as indicators of sources and pathways of contaminants, primarily a function of their utility in tracking long-term trends. As described in this report, in some instances (e.g., mussel monitoring at Royal Palms and at Pacific Grove, and clam monitoring by USGS at Palo Alto) bivalve monitoring directly downstream of effluent discharges has provided compelling documentation of the reductions in ambient concentrations of contaminants in response to improvements in wastewater treatment and source control. The key to these and other successful examples of monitoring sources and pathways using bivalves is that it was possible to obtain residents or to deploy transplants in close proximity to the zone of influence of the discharges.

However, sampling of bivalves in close proximity to contaminant sources and pathways is not always possible. Resident bivalves have patchy distributions. Transplantation allows for broader spatial coverage, but estuarine and marine species are able to tolerate a limited range of habitat conditions (especially salinity). When species are subjected to conditions outside of their tolerance they may have abnormal physiology or behavior (e.g., feeding), and this interferes with their functioning as biosentinels (Gunther et al. 1999). The salinity tolerance of *Mytilus californianus* is 50-150% of natural seawater (Morris et al. 1980). This precludes the use of mussels in waters with salinities lower than approximately 17 ppt. Since effluent discharges and stormwater runoff have low salinity and are often discharged into freshwater tributaries to coastal waters, mussels frequently cannot be deployed close to these pathways where the signal of trends in contamination would be greatest. Combinations of bivalve species can be deployed to cover a wider range of habitats, but interspecific variation then becomes an obstacle to interpretation, and physiological stress can still be an important factor in habitats with varying salinity. As discussed in the previous section, passive sampling devices offer another alternative for sampling habitats that are close to contaminant sources but inhospitable to bivalve biosentinels. Other species of biota, such as small fish (Greenfield et al. 2011) or resident invertebrates, may also be an alternative in these situations.

Effectiveness of Management

In the habitats where they can be sampled, bivalves have proven to be powerful indicators of the effectiveness of management actions on a regional scale, again primarily due to their utility as indicators of long-term trends. The statewide impact of bans of many contaminants (DDTs, PCBs, dieldrin, chlordane, lead) has been best documented by bivalve monitoring. As described previously, bivalve monitoring has also demonstrated the effectiveness of management actions on a local spatial scale where sampling could be conducted within the zone of influence of wastewater outfalls (e.g., Royal Palms, Palo Alto).

Bivalve monitoring is of limited utility, however, in monitoring the effectiveness of management on a project scale due to the issues related to the distribution of residents and the physiological tolerances of transplants described above. Passive sampling devices are a more versatile alternative for project-scale evaluations. Other species of biota may also be an alternative in these situations. Another note of caution regarding bivalves as biosentinels of the effectiveness of management is that they may not always be representative of the entire food web or the portion of the food web of greatest concern. PCBs in San Francisco Bay appear to be an example of this, where long-term bivalve monitoring indicates declining concentrations, but sport fish do not appear to be showing similar declines and small fish remain quite elevated as well (Davis et al. 2007).
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Next Steps for Bivalve Monitoring in SWAMP

In summary, bivalve monitoring is an effective means of addressing many of the objectives of the SWAMP bioaccumulation monitoring program. The greatest value of bivalve monitoring is as a tool for evaluating status and trends in the impact of marine biotoxins on the fishing and aquatic life beneficial uses, CEC exposure and risk to aquatic life at lower trophic levels and CEC trends that are relevant both for aquatic life and fishing, and tracking long-term trends in contaminants affecting both fishing and aquatic life at a regional scale and in specific locations. They are especially valuable for monitoring bioaccumulation of certain pollutants, such as PAHs, that are metabolized by species at higher trophic levels. These attributes make bivalve monitoring a valuable component of a statewide bioaccumulation monitoring program.

A considerable amount of the needed monitoring is being conducted by other programs, making coordination a key part of a SWAMP strategy on bivalve monitoring, especially in the near-term with the large NMW effort on CECs still in progress. SWAMP should seek to fill any significant gaps not addressed by other programs. SWAMP plans for bivalve monitoring should be re-evaluated after the NMW CEC Pilot is completed.

Recommended Next Steps for Different Categories of Contaminants

Biotoxins

- Conduct a workshop including leading biotoxin scientists to obtain guidance on designing statewide monitoring for biotoxins, including the role of bivalve monitoring.
- Assess gaps in sampling based on the workshop conclusions. Consider use of existing stations or establishment of new ones. Consider needs for monitoring bivalves in freshwater.
- Coordinate with other programs to achieve the needed monitoring.

CECs

- Follow up as needed on the 2010 pilot study, in collaboration with NOAA, the RMP, and SCCWRP.
- Consider use of existing stations or establishment of new ones to address monitoring needs.

Recommended Next Steps Across All Contaminant Categories

- Develop a coordinated overall plan for bivalve monitoring in California
  - Establish prioritized list of stations across programs and all contaminant categories. Criteria should include: 1) strategic locations for biotoxins, 2) strategic locations for emerging contaminants, 3) strategic locations for tracking trends of traditional contaminants. The value of maintaining existing time series for traditional contaminants should be considered (including NMW sites if NMW does not continue). The value of usable archives for existing stations should also be considered (an inventory of existing archives should be developed). The list can be used to guide decisions under a variety of funding scenarios.
  - Initiate a sound archiving plan for all long-term bivalve monitoring stations (both resident and transplants) across all programs. The RMP archiving strategy can serve as a model.

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Table 1. The SWAMP bioaccumulation monitoring assessment framework for the fishing beneficial use.

1. Determine the status of the fishing beneficial use throughout the State with respect to bioaccumulation of toxic pollutants	
	1.1 What are the extent and location of water bodies with sufficient evidence to indicate that the fishing beneficial use is at risk due to pollutant bioaccumulation? 1.2 What are the extent and location of water bodies with some evidence indicating the fishing beneficial use is at risk due to pollutant bioaccumulation? 1.3 What are the extent and location of water bodies with no evidence indicating the fishing beneficial use is at risk due to pollutant bioaccumulation? 1.4 What are the proportions of water bodies in the State and each region falling within the three categories defined in questions 1.1, 1.2, and 1.3?
2. Assess trends in the impact of bioaccumulation on the fishing beneficial use throughout the State	
	2.1 Are water bodies improving or deteriorating with respect to the impact of bioaccumulation on the fishing beneficial use? 2.1.1 Have water bodies fully supporting the fishing beneficial use become impaired? 2.1.2 Has full support of the fishing beneficial use been restored for previously impaired water bodies? 2.2 What are the trends in proportions of water bodies falling within the three categories defined in questions 1.1, 1.2, and 1.3 regionally and statewide?
3. Evaluate sources and pathways of bioaccumulative pollutants impacting the fishing beneficial use	
	3.1 What are the magnitude and relative importance of pollutants that bioaccumulate and indirect causes of bioaccumulation throughout each Region and the state as a whole? 3.2 How is the relative importance of different sources and pathways of bioaccumulative pollutants that impact the fishing beneficial use changing over time on a regional and statewide basis?
4. Provide the monitoring information needed to evaluate the effectiveness of management actions in reducing the impact of bioaccumulation on the fishing beneficial use	
	4.1 What are the management actions that are being employed to reduce the impact of bioaccumulation on the fishing beneficial use regionally and statewide? 4.2 How has the impact of bioaccumulation on the fishing beneficial use been affected by management actions regionally and statewide?

Table 2a. Temporal trend results at SMW sites for Total DDTs and Total Chlordanes. Stations are organized from north to south. Green rows are resident mussel sites, remainder are transplants. Trend symbols: DL = greater than 50% values were non-detects, trend not evaluated; NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

Station Name	N (yrs)	Years	Total DDTs					Total Chlordanes				
			% ND	Trend	Half-life (yrs)	Slope	R^2	% ND	Trend	Half-life (yrs)	Slope	R^2
Crescent City	21	1983 - 2010	75%	DL	Not Determined			38%	NS	+	0.01	0.02
Trinidad Head	29	1977 - 2010	52%	DL	Not Determined			31%	NS	+	0.02	0.04
Mad River Slough	11	1982 - 2003	64%	DL	Not Determined			18%	NS	+	0.02	0.04
Bodega Head	29	1977 - 2009	8%	▼	8	-0.04	0.22	22%	NS	8	-0.04	0.12
San Francisco Bay/Fort Baker	15	1982 - 2010	0%	▼	13	-0.02	0.73	0%	NS	61	0	0.04
J. Fitzgerald	16	1977 - 2010	5%	▼	9	-0.03	0.38	19%	NS	13	-0.02	0.05
Sandholdt Bridge	24	1982 - 2010	0%	NS	41	-0.01	0.08	0%	NS	92	0	0.01
Pacific Grove	30	1977 - 2010	0%	▼	14	-0.02	0.38	4%	▼	9	-0.03	0.31
Royal Palms	29	1977 - 2010	0%	▼	12	-0.02	0.54	3%	▼	10	-0.03	0.23
LA Harbor/National Steel	22	1982 - 2010	0%	▼	13	-0.02	0.52	0%	▼	19	-0.02	0.24
LA Harbor/Consolidated Slip	27	1982 - 2010	0%	▼	11	-0.03	0.64	0%	▼	18	-0.02	0.42
Anaheim Bay/Navy Marsh	19	1983 - 2010	0%	▼	15	-0.02	0.32	0%	▼	16	-0.02	0.27
Huntington Harbour/Edinger Street	17	1983 - 2003	0%	▼	8	-0.04	0.67	0%	▼	8	-0.04	0.69
Huntington Harbour/Warner Ave	26	1983 - 2010	0%	▼	8	-0.04	0.86	0%	▼	7	-0.04	0.90
Newport Bay/Turning Basin	12	1986 - 2003	0%	▼	9	-0.03	0.76	0%	▼	7	-0.04	0.71
Newport Bay/Rhine Channel	16	1986 - 2002	0%	▼	10	-0.03	0.36	0%	▼	8	-0.04	0.55
Newport Bay/Crows Nest	27	1982 - 2010	0%	▼	8	-0.04	0.83	0%	▼	7	-0.04	0.84
Newport Bay/Highway 1 Bridge	25	1982 - 2010	0%	▼	8	-0.04	0.75	0%	▼	8	-0.04	0.77
Newport Bay/Bay Island	10	1982 - 2003	0%	▼	7	-0.05	0.67	0%	▼	6	-0.05	0.78
Oceanside	26	1977 - 2010	0%	▼	9	-0.03	0.49	0%	▼	9	-0.04	0.40
San Diego Bay/Harbor Island	14	1977 - 2010	0%	NS	51	-0.01	0.09	0%	NS	+	0.01	0.13
San Diego Bay/Shelter Island	17	1980 - 2010	0%	▼	6	-0.05	0.74	0%	▼	15	-0.02	0.38

Table 2b. Temporal trend results at RMP sites (including SMW data) for Total DDTs and Total PCBs. Stations are organized from north to south. All stations represent transplanted mussels. Trend symbol indicates a significant decline ($p < 0.05$).

Station Name	N (yrs)	Years	Total DDTs				Total PCBs			
			Trend	Half-life (yrs)	Slope	R^2	Trend	Half-life (yrs)	Slope	R^2
Pinole Point	24	1981 – 2010	▼	11	-0.03	0.68	▼	11	-0.03	0.68
Richmond Bridge/Red Rock	20	1980 – 2010	▼	6	-0.05	0.80	▼	6	-0.05	0.69
Treasure Island/Yerba Buena Island	24	1980 – 2010	▼	8	-0.04	0.84	▼	9	-0.03	0.77
Hunters Point/Alameda	19	1981 – 2010	▼	8	-0.04	0.77	▼	8	-0.04	0.49
Redwood Creek	21	1981 – 2010	▼	10	-0.03	0.59	▼	15	-0.02	0.31
Dumbarton Bridge	21	1981 - 2010	▼	11	-0.03	0.67	▼	12	-0.02	0.45

Table 2c. Temporal trends at NMW sites for Total DDTs and Total Chlordanes. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

			Total DDTs				Total Chlordanes			
Station Name	N (yrs)	YEARS	Trend	Half-life (yrs)	Slope	R^2	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	15	1986 - 2009	▼	11	-0.03	0.62	▼	25	-0.01	0.34
Eureka Samoa Bridge	11	1990 - 2009	NS	17	-0.02	0.34	▼	16	-0.02	0.40
Humboldt Bay Jetty	15	1986 - 2009	▼	11	-0.03	0.47	NS	+	0	0.00
Pt. Delgado Shelter Cove	16	1986 - 2009	NS	22	-0.01	0.67	NS	+	0.01	0.08
Pt. Arena Lighthouse	16	1986 - 2009	NS	41	-0.01	0.03	NS	> 100	0	0.00
Tomales Bay Spenger's Res.	16	1986 - 2009	NS	29	-0.01	0.11	NS	24	-0.01	0.09
SFB Emeryville	12	1987 - 2009	NS	+	0.02	0.10	▼	12	-0.02	0.21
SFB San Mateo Bridge	16	1986 - 2009	▼	11	-0.03	0.42	▼	9	-0.04	0.49
SFB Dumbarton Bridge	16	1986 - 2009	▼	10	-0.03	0.43	▼	8	-0.04	0.45
Monterey Bay Pt. Santa Cruz	16	1986 - 2009	NS	42	-0.01	0.03	NS	29	-0.01	0.16
Monterey Bay Elkhorn Slough	9	1994- 2005	NS	+	0.06	0.17	NS	+	0.02	0.04
Monterey Bay Moss Landing	14	1990 - 2009	NS	+	0.01	0.03	NS	47	-0.01	0.02
Pacific Grove Lovers Pt.	16	1986 - 2009	NS	17	-0.02	0.22	NS	21	-0.01	0.24
San Simeon Pt.	14	1986 - 2009	▼	16	-0.02	0.58				
San Luis Obispo Bay	16	1986 - 2008	▼	16	-0.02	0.43	▼	14	-0.02	0.50
Pt. Conception	13	1986 - 2004	NS	37	-0.01	0.08				
Pt. Santa Barbara	15	1986 - 2008	▼	6	-0.05	0.68				
Santa Cruz Is. Fraser Pt.	13	1986 - 2008	NS	16	0.02	0.25				
Pt. Dume	15	1986 - 2008	▼	20	-0.02	0.30	▼	22	-0.01	0.28
Santa Monica Bay Las Tunas Beach	11	1990 - 2008	▼	12	-0.02	0.27				
Marina Del Rey South Jetty	15	1986 - 2008	▼	15	-0.02	0.29	▼	11	-0.03	0.58
Redondo Beach Municipal Jetty	11	1990 - 2008	NS	17	-0.02	0.05				
Royal Palms	15	1986 - 2008	▼	10	-0.03	0.69	▼	12	-0.03	0.36
San Pedro Harbor	14	1986 - 2008	▼	13	-0.02	0.70				
Long Beach Breakwater	11	1990 - 2008	▼	13	-0.02	0.51				
Anaheim Bay West Jetty	15	1986 - 2008	▼	10	-0.03	0.66	▼	9	-0.03	0.69
Newport Beach West Jetty	14	1986 - 2008	NS	18	-0.02	0.32				
South Catalina Is. Bird Rock	13	1986 - 2008	▼	9	-0.03	0.48				
Oceanside Municipal Jetty	15	1986 - 2008	▼	17	-0.02	0.14	NS	29	-0.01	0.08
La Jolla	13	1986 - 2008	▼	10	-0.03	0.42	▼	21	-0.01	0.27
Mission Bay Ventura Bridge	15	1986 - 2008	▼	8	-0.04	0.88	▼	10	-0.03	0.66
SD Bay Harbor Island	15	1986 - 2008	NS	29	-0.01	0.20	▼	17	-0.02	0.51
Pt. Loma Lighthouse	12	1986 - 2005	▼	9	-0.03	0.59	▼	13	-0.02	0.61
SD Bay Coronado Bridge	13	1989 - 2008	NS	> 100	0	0.00	▼	16	-0.02	0.54
Imperial Beach North Jetty	15	1986 - 2008	▼	7	-0.04	0.86	▼	9	-0.03	0.84

Table 3a. Temporal trend results at SMW sites for Dieldrin and Total PCBs. Stations are organized from north to south. Green rows are resident mussel sites, remainder are transplants. Trend symbols: DL = greater than 50% values were non-detects, trend not evaluated; NS = no significant trend (p > 0.05); ▼= declining trend (p < 0.05. Sites with "+" have a positive but not statistically significant slope, half life not calculated.

Station Name	N (yrs)	Years	Dieldrin					Total PCBs				
			% ND	Trend	Half-life (yrs)	Slope	R^2	% ND	Trend	Half-life (yrs)	Slope	R^2
Crescent City	21	1983 - 2010	4%	NS	37	-0.01	0.02	67%	DL	Not determined		
Trinidad Head	29	1977 - 2010	7%	NS	+	0.01	0.05	58%	DL	Not determined		
Mad River Slough	11	1982 - 2003	9%	NS	+	0.01	0.02	55%	DL	Not determined		
Bodega Head	29	1977 - 2009	0%	▼	14	-0.02	0.37	61%	DL	Not determined		
San Francisco Bay/Fort Baker	15	1982 - 2010	0%	▼	27	-0.01	0.29	13%	NS	7	-0.04	0.18
J. Fitzgerald	16	1977 - 2010	0%	NS	>100	0	0.00	40%	NS	11	-0.03	0.06
Sandholdt Bridge	24	1982 - 2010	0%	NS	+	0	0.00	4%	▼	12	-0.01	0.33
Pacific Grove	30	1977 - 2010	0%	▼	20	-0.01	0.24	44%	NS	>100	0	0.00
Royal Palms	29	1977 - 2010	7%	NS	+	0.01	0.05	3%	▼	13	-0.02	0.52
LA Harbor/National Steel	22	1982 - 2010	0%	NS	>100	0	0.00	4%	▼	12	-0.03	0.46
LA Harbor/Consolidated Slip	27	1982 - 2010	0%	NS	92	0	0.02	4%	▼	28	-0.01	0.24
Anaheim Bay/Navy Marsh	19	1983 - 2010	0%	NS	31	-0.01	0.06	0%	NS	10	-0.03	0.08
Huntington Harbour/Edinger Street	17	1983 - 2003	0%	▼	11	-0.03	0.43	0%	▼	8	-0.04	0.76
Huntington Harbour/Warner Ave	26	1983 - 2010	0%	▼	8	-0.04	0.82	4%	▼	9	-0.03	0.75
Newport Bay/Turning Basin	12	1986 - 2003	0%	▼	9	-0.03	0.52	0%	▼	9	-0.04	0.50
Newport Bay/Rhine Channel	16	1986 - 2002	0%	NS	15	-0.02	0.19	0%	NS	48	-0.01	0.01
Newport Bay/Crows Nest	27	1982 - 2010	0%	▼	9	-0.03	0.74	3%	▼	9	-0.03	0.73
Newport Bay/Highway 1 Bridge	25	1982 - 2010	0%	▼	9	-0.03	0.64	4%	▼	12	-0.03	0.46
Newport Bay/Bay Island	10	1982 - 2003	0%	▼	13	-0.02	0.60	0%	▼	10	-0.03	0.61
Oceanside	26	1977 - 2010	13%	NS	14	-0.02	0.07	18%	NS	8	-0.04	0.10
San Diego Bay/Harbor Island	14	1977 - 2010	8%	NS	25	-0.01	0.01	7%	▼	7	-0.04	0.54
San Diego Bay/Shelter Island	17	1980 - 2010	0%	NS	31	-0.01	0.17	6%	NS	23	-0.01	0.20

Note – 2010 PCB data not included in trend analysis

Table 3b. Temporal trends at NMW sites for Dieldrin and Total PCBs. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

		Dieldrin					Total PCBs				
Station Name	YEARS	N	Trend	Half-life (yrs)	Slope	R^2	N	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	1986 - 2009	15	NS	81	0	0.04	12	NS	25	-0.01	0.03
Eureka Samoa Bridge	1990 - 2009	11	NS	74	0	0.01	11	NS	53	-0.01	0.03
Humboldt Bay Jetty	1986 - 2009	15	NS	+	0.01	0.21	13	▼	7	-0.04	0.41
Pt. Delgado Shelter Cove	1986 - 2009	16	NS	+	0	0.01	14	NS	+	0.01	0.14
Pt. Arena Lighthouse	1986 - 2009	16	▼	23	-0.01	0.27	13	NS	+	0.01	0.04
Tomales Bay	1986 - 2009	16	NS	22	-0.01	0.11	13	NS	65	-0.01	0.01
Spenger's Res. SFB	1987 - 2009	13	NS	+	0.04	0.17	12	NS	44	-0.01	0.07
Emeryville SFB	1986 - 2009	16	▼	6	-0.05	0.65	13	▼	11	-0.03	0.48
San Mateo Bridge SFB Dumbarton Bridge	1986 - 2009	16	▼	6	-0.05	0.68	14	▼	16	-0.02	0.30
Monterey Bay Pt. Santa Cruz	1986 - 2009	16	NS	62	0	0.02	13	NS	+	0	0.00
Monterey Bay Elkhorn Slough	1994- 2005	9	NS	6	0.05	0.14	9	NS	7	-0.04	0.37
Monterey Bay Moss Landing	1990 - 2009	14	NS	+	0.02	0.20	14	NS	+	0.01	0.04
Pacific Grove Lovers Pt.	1986 - 2009	16	NS	56	-0.01	0.01	13	NS	69	0	0.02
San Simeon Pt.	1986 - 2009	14	NS	81	0	0.03	13	NS	73	0	0.02
San Luis Obispo Bay	1986 - 2008	16	▼	17	-0.02	0.29	13	NS	23	-0.01	0.08
Pt. Conception	1986 - 2004	13	NS	40	-0.01	0.03	11	NS	+	0.02	0.16
Pt. Santa Barbara	1986 - 2008	15	NS	32	-0.01	0.02	11	NS	+	0.02	0.42
Santa Cruz Is. Fraser Pt.	1986 - 2008	13	NS	+	0.01	0.06	11	NS	+	0.02	0.11
Pt. Dume	1986 - 2008	15	NS	35	-0.01	0.05	11	NS	45	-0.01	0.01
Santa Monica Bay Las Tunas Beach	1990 - 2008	11	NS	59	-0.01	0.03	11	NS	51	-0.01	0.03
Marina Del Rey South Jetty	1986 - 2008	15	NS	+	0	0.01	13	NS	22	-0.01	0.31
Redondo Beach Municipal Jetty	1990 - 2008	11	NS	94	0	0.01	11	NS	+	0	0.00
Royal Palms	1986 - 2008	15	▼	13	-0.02	0.32	11	▼	15	-0.02	0.58
San Pedro Harbor	1986 - 2008	14	NS	14	-0.02	0.14	10	▼	13	-0.02	0.75
Long Beach Breakwater	1990 - 2008	11	NS	39	-0.01	0.04	11	NS	35	-0.01	0.11
Anaheim Bay West Jetty	1986 - 2008	15	▼	8	-0.04	0.51	13	▼	13	-0.02	0.60
Newport Beach West Jetty	1986 - 2008	14	NS	+	0	0.00	12	NS	31	-0.01	0.12
South Catalina Is. Bird Rock	1986 - 2008	13	NS	30	-0.01	0.15	11	NS	> 100	0	0.00
Oceanside Municipal Jetty	1986 - 2008	15	NS	+	0.01	0.02	13	NS	87	0	0.01
La Jolla	1986 - 2008	15	NS	> 100	0	0.00	12	NS	+	0.01	0.04
Mission Bay Ventura Bridge	1986 - 2008	15	NS	30	-0.01	0.17	13	▼	13	-0.02	0.75
SD Bay Harbor Island	1986 - 2008	15	NS	+	0.01	0.03	13	NS	+	0.003	0.04
Pt. Loma Lighthouse	1986 - 2005	13	NS	+	0.01	0.06	10	▼	31	-0.01	0.22
SD Bay Coronado Bridge	1989 - 2008	13	NS	16	-0.02	0.15	13	NS	+	0.01	0.20
Imperial Beach North Jetty	1986 - 2008	15	▼	13	-0.02	0.23	12	NS	36	-0.01	0.07

Table 4. Temporal trends at NMW sites for Total PAHs and Total Butyltins. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$); and ▲= increasing trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

		Total PAHs					Total Butyltins				
Station Name	YEARS	N	Trend	Half-life (yrs)	Slope	R^2	N	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	1986 - 2009	12	NS	+	0.02	0.26	11	NS	8	-0.04	0.17
Eureka Samoa Bridge	1990 - 2009	11	NS	+	0.01	0.07	10	▼	6	-0.05	0.73
Humboldt Bay Jetty	1986 - 2009	12	▲	+	0.02	0.40	11	NS	8	-0.04	0.14
Pt. Delgado Shelter Cove	1986 - 2009	13	▲	+	0.03	0.48	12	NS	6	-0.05	0.57
Pt. Arena Lighthouse	1986 - 2009	13	NS	+	0.03	0.29	12	NS	7	-0.04	0.29
Tomales Bay Spenger's Res.	1986 - 2009	13	NS	+	0.01	0.02	12	NS	6	-0.05	0.12
SFB Emeryville	1987 - 2009	11	NS	67	-0.01	0.00	10	NS	7	-0.04	0.04
SFB San Mateo Bridge	1986 - 2009	13	NS	32	-0.01	0.08	12	▼	4	-0.08	0.66
SFB Dumbarton Bridge	1986 - 2009	13	NS	18	-0.02	0.24	12	▼	4	-0.07	0.73
Monterey Bay Pt. Santa Cruz	1986 - 2009	12	NS	+	0	0.03	12	▼	4	-0.08	0.71
Monterey Bay Elkhorn Slough	1994- 2005	9	NS	+	0	0.00	9	NS	5	-0.06	0.37
Monterey Bay Moss Landing	1990 - 2009	14	NS	+	0.03	0.13	13	NS	5	-0.06	0.26
Pacific Grove Lovers Pt.	1986 - 2009	13	NS	+	0.01	0.05	12	NS	7	-0.04	0.25
San Simeon Pt.	1986 - 2009	12	NS	+	0.05	0.32	10	NS	7	-0.04	0.24
San Luis Obispo Bay	1986 - 2008	13	NS	20	-0.02	0.07	11	NS	5	-0.06	0.27
Pt. Conception	1986 - 2004	10	NS	+	0.01	0.07	10	NS	20	-0.02	0.05
Pt. Santa Barbara	1986 - 2008	12	▲	+	0.05	0.17	11	NS	6	-0.05	0.62
Santa Cruz Is. Fraser Pt.	1986 - 2008	10	NS	+	0.03	0.21	9	NS	+	0.02	0.07
Pt. Dume	1986 - 2008	12	NS	+	0.05	0.29	11	NS	23	-0.01	0.02
Santa Monica Bay Las Tunas Beach	1990 - 2008	11	NS	+	0.01	0.01	10	NS	12	-0.03	0.06
Marina Del Rey South Jetty	1986 - 2008	12	NS	31	-0.01	0.11	11	▼	4	-0.08	0.57
Redondo Beach Municipal Jetty	1990 - 2008	11	NS	17	-0.02	0.09	9	▼	5	-0.06	0.46
Royal Palms	1986 - 2008	12	NS	+	0.03	0.08	11	▼	5	-0.07	0.50
San Pedro Harbor	1986 - 2008	11	NS	+	0.05	0.21	10	NS	6	-0.05	0.08
Long Beach Breakwater	1990 - 2008	11	▼	9	-0.03	0.40	10	NS	7	-0.04	0.63
Anaheim Bay West Jetty	1986 - 2008	12	NS	+	0.03	0.23	11	▼	4	-0.07	0.79
Newport Beach West Jetty	1986 - 2008	11	NS	+	0.04	0.12	10	▼	5	-0.06	0.59
South Catalina Is. Bird Rock	1986 - 2008	10	NS	+	0.01	0.04	9	NS	+	0.02	0.02
Oceanside Municipal Jetty	1986 - 2008	12	NS	+	0.05	0.24	11	NS	12	-0.03	0.06
La Jolla	1986 - 2008	12	NS	+	0.03	0.33	11	NS	26	-0.01	0.01
Mission Bay Ventura Bridge	1986 - 2008	12	NS	12	-0.03	0.29	11	▼	4	-0.08	0.79
SD Bay Harbor Island	1986 - 2008	12	▼	17	-0.02	0.69	10	NS	7	-0.04	0.06
Pt. Loma Lighthouse	1986 - 2005	10	NS	58	-0.01	0.02	10	▼	4	-0.08	0.73
SD Bay Coronado Bridge	1989 - 2008	13	▼	15	-0.02	0.45	12	NS	5	-0.06	0.25
Imperial Beach North Jetty	1986 - 2008	12	NS	> 100	0	0.01	10	NS	+	0	0.00

Table 5a. Temporal trend results at SMW sites for lead and silver. Stations are organized from north to south. Green rows are resident mussel sites, remainder are transplants. Trend symbols: NA = not analyzed; NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

Station Name	N (yrs)	Years	Lead				Silver			
			Trend	Half-life (yrs)	Slope	R^2	Trend	Half-life (yrs)	Slope	R^2
Crescent City	20	1983 - 2010	NS	>100	-0.001	0.02	▼	10	-0.030	0.36
Trinidad Head	27	1977 - 2010	NS	>100	-0.001	0.01	▼	19	-0.015	0.36
Mad River Slough	11	1982 - 2003	NS	>100	-0.001	0.00	NS	26	-0.011	0.20
Samoa Bridge/West	10	1980 - 2003	NS	51	-0.006	0.09	▼	8	-0.036	0.57
Bodega Head	32	1977 - 2009	NS	63	-0.005	0.09	NS	32	-0.009	0.12
Tomales Bay	11	1980 - 2010	NA	Not Analyzed			▼	13	-0.023	0.32
San Francisco Bay/Fort Baker	14	1981 - 2010	NS	+	0.003	0.09	▼	7	-0.043	0.66
J. Fitzgerald	13	1977 - 2010	▼	39	-0.008	0.45	▼	13	-0.023	0.36
Sandholdt Bridge	19	1989 - 2010	NS	73	-0.004	0.03	NS	27	-0.011	0.05
Pacific Grove	30	1977 - 2010	▼	16	-0.019	0.50	▼	7	-0.041	0.73
Royal Palms	27	1977 - 2010	▼	10	-0.031	0.81	▼	12	-0.025	0.58
LA Harbor/National Steel	20	1983 - 2010	▼	23	-0.013	0.29	NS	78	-0.004	0.01
LA Harbor/Consolidated Slip	22	1982 - 2010	▼	17	-0.018	0.47	NS	+	0.002	0.01
Anaheim Bay/Navy Marsh	18	1983 - 2010	▼	40	-0.008	0.30	NS	33	-0.009	0.03
Huntington Harbour/Edinger Street	16	1983 - 2003	▼	10	-0.031	0.72	NS	29	-0.010	0.10
Huntington Harbour/Warner Ave	25	1983 - 2010	▼	11	-0.026	0.81	NS	23	-0.013	0.12
Newport Bay/Crows Nest	26	1982 - 2010	▼	18	-0.017	0.26	NS	94	-0.003	0.01
Newport Bay/Highway 1 Bridge	24	1982 - 2010	▼	36	-0.008	0.32	NS	49	-0.006	0.04
Oceanside	26	1977 - 2009	▼	15	-0.020	0.70	▼	6	-0.046	0.71
San Diego Bay/Harbor Island	11	1984 - 2010	NS	+	0.004	0.04	▼	24	-0.012	0.38
San Diego Bay/Shelter Island	18	1980 - 2010	NS	+	0.001	0.02	▼	14	-0.022	0.34

Table 5b. Temporal trends at NMW sites for lead and silver. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$. Sites with "+" have a positive but not statistically significant slope, half life not calculated.

		Lead					Silver				
Station Name	YEARS	N	Trend	Half-life (yrs)	Slope	R^2	N	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	1986 - 2009	15	NS	+	0	0.14	11	NS	>100	0	0.10
Eureka Samoa Bridge	1990 - 2009	11	NS	>100	0	0.09	11	NS	+	0	0.01
Humboldt Bay Jetty	1986 - 2009	15	NS	+	0	0.07	11	NS	+	0	0.02
Pt. Delgado Shelter Cove	1986 - 2009	16	NS	>100	0	0.04	12	NS	+	0.01	0.09
Pt. Arena Lighthouse	1986 - 2009	16	NS	+	0	0.05	12	NS	+	0.01	0.19
Tomales Bay Spenger's Res.	1986 - 2009	16	NS	+	0	0.05	12	NS	+	0.01	0.11
SFB Emeryville	1987 - 2009	13	▼	19	-0.01	0.50	10	NS	>100	0	0.30
SFB San Mateo Bridge	1986 - 2009	16	NS	+	0	0.00	12	▼	27	-0.01	0.56
SFB Dumbarton Bridge	1986 - 2009	16	NS	+	0	0.02	12	NS	>100	0	0.03
Monterey Bay Pt. Santa Cruz	1986 - 2009	16	NS	15	-0.02	0.22	13	▼	24	-0.01	0.34
Monterey Bay Elkhorn Slough	1994- 2005	9	NS	>100	0	0.00	9	NS	>100	0	0.00
Monterey Bay Moss Landing	1990 - 2009	14	▼	14	-0.02	0.38	14	NS	>100	0	0.01
Pacific Grove Lovers Pt.	1986 - 2009	16	NS	65	-0.01	0.04	12	NS	+	0	0.02
San Simeon Pt.	1986 - 2009	15	NS	+	0	0.01	11	▼	61	-0.01	0.40
San Luis Obispo Bay	1986 - 2008	16	NS	+	0.01	0.12	12	NS	+	0	0.02
Pt. Conception	1986 - 2004	13	NS	>100	0	0.01	9	NS	+	0.02	0.23
Pt. Santa Barbara	1986 - 2008	15	NS	92	0	0.03	11	▼	4	-0.07	0.75
Santa Cruz Is. Fraser Pt.	1986 - 2008	13	NS	+	0	0.02	10	NS	65	-0.01	0.17
Pt. Dume	1986 - 2008	15	NS	+	0	0.07	10	NS	>100	0	0.06
Santa Monica Bay Las Tunas Beach	1990 - 2008	11	NS	34	-0.01	0.08	11	NS	18	-0.02	0.16
Marina Del Rey South Jetty	1986 - 2008	15	▼	9	-0.03	0.57	11	NS	>100	0	0.01
Redondo Beach Municipal Jetty	1990 - 2008	11	NS	41	-0.01	0.15	11	▼	14	-0.02	0.65
Royal Palms	1986 - 2008	15	▼	21	-0.01	0.47	12	▼	14	-0.02	0.37
San Pedro Harbor	1986 - 2008	14	▼	24	-0.01	0.47	10	NS	+	0	0.00
Long Beach Breakwater	1990 - 2008	11	NS	48	-0.01	0.09	11	NS	+	0	0.00
Anaheim Bay West Jetty	1986 - 2008	15	▼	9	-0.03	0.55	11	NS	72	0	0.11
Newport Beach West Jetty	1986 - 2008	14	NS	22	-0.01	0.28	10	▼	15	-0.02	0.70
South Catalina Is. Bird Rock	1986 - 2008	13	NS	38	-0.01	0.26	10	NS	>100	0	0.03
Oceanside Municipal Jetty	1986 - 2008	13	NS	>100	0	0.06	11	NS	>100	0	0.00
La Jolla	1986 - 2008	13	▼	20	-0.02	0.35	10	▼	9	-0.03	0.51
Mission Bay Ventura Bridge	1986 - 2008	15	NS	130	0	0.04	11	NS	>100	0	0.00
SD Bay Harbor Island	1986 - 2008	15	▼	28	-0.01	0.37	11	NS	+	0	0.03
Pt. Loma Lighthouse	1986 - 2005	13	NS	>100	0	0.01	9	▼	13	-0.02	0.40
SD Bay Coronado Bridge	1989 - 2008	13	NS	25	-0.01	0.25	12	NS	>100	0	0.15
Imperial Beach North Jetty	1986 - 2008	15	NS	96	0	0.05	11	▼	12	-0.02	0.60

Table 6a. Temporal trend results at SMW sites for copper and zinc. Stations are organized from north to south. Green rows are resident mussel sites, remainder are transplants. Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$); and ▲= increasing trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

Station Name	N (yrs)	Years	Copper				Zinc			
			Trend	Half-life (yrs)	Slope	R^2	Trend	Half-life (yrs)	Slope	R^2
Crescent City	20	1983 - 2010	NS	+	0.001	0.02	▼	59	-0.005	0.36
Trinidad Head	27	1977 - 2010	▲	+	0.004	0.13	NS	+	0.002	0.04
Mad River Slough	11	1982 - 2003	NS	+	0.002	0.04	NS	>100	0	0.00
Samoa Bridge/West	10	1980 - 2003	NS	+	0.002	0.09	NS	>100	-0.003	0.04
Bodega Head	32	1977 - 2009	NS	+	0.005	0.09	NS	+	0.001	0.01
Tomales Bay	11	1980 - 2010	NS	+	0.003	0.10	NS	+	0.005	0.13
San Francisco Bay/Fort Baker	14	1981 - 2010	NS	+	0.001	0.03	NS	+	0.002	0.11
J. Fitzgerald	13	1977 - 2010	NS	+	0.001	0.05	NS	+	0.001	0.01
Sandholdt Bridge	19	1989 - 2010	NS	> 100	-0.002	0.04	▼	33	-0.009	0.24
Pacific Grove	30	1977 - 2010	NS	+	0.002	0.03	NS	>100	-0.003	0.12
Royal Palms	27	1977 - 2010	▼	> 100	-0.003	0.16	▼	43	-0.007	0.39
LA Harbor/National Steel	20	1983 - 2010	NS	+	0.005	0.06	NS	+	0.001	0.00
LA Harbor/Consolidated Slip	22	1982 - 2010	NS	+	0.007	0.16	NS	>100	-0.003	0.08
Anaheim Bay/Navy Marsh	18	1983 - 2010	▲	+	0.004	0.23	NS	+	0.001	0.01
Huntington Harbour/Edinger Street	16	1983 - 2003	▲	+	0.011	0.37	NS	+	0.006	0.18
Huntington Harbour/Warner Ave	25	1983 - 2010	▲	+	0.007	0.21	NS	+	0.003	0.09
Newport Bay/Crows Nest	26	1982 - 2010	▲	+	0.020	0.35	NS	+	0.004	0.03
Newport Bay/Highway 1 Bridge	24	1982 - 2010	▲	+	0.019	0.46	NS	+	0.005	0.14
Oceanside	26	1977 - 2009	NS	+	0.001	0.02	▼	44	-0.007	0.20
San Diego Bay/Harbor Island	11	1984 - 2010	▲	+	0.022	0.50	▲	+	0.010	0.51
San Diego Bay/Shelter Island	18	1980 - 2010	▲	+	0.011	0.44	NS	+	0.005	0.16

Table 6b. Temporal trends at NMW sites for copper and zinc. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$); and ▲= increasing trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

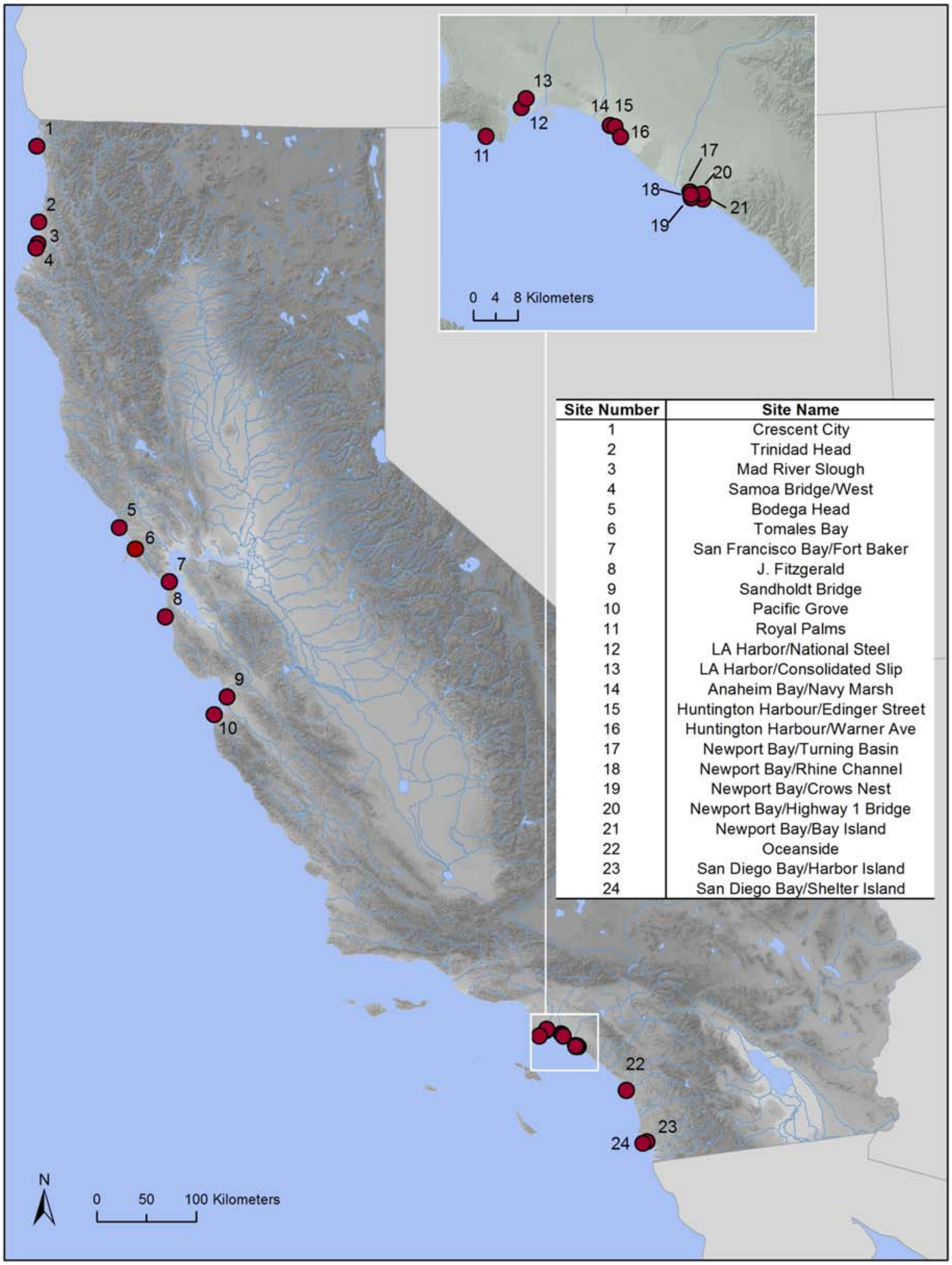
			Copper				Zinc			
Station Name	N (yrs)	YEARS	Trend	Half-life (yrs)	Slope	R^2	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	15	1986 - 2009	NS	+	0.009	0.20	NS	>100	-0.002	0.03
Eureka Samoa Bridge	11	1990 - 2009	NS	>100	-0.003	0.02	NS	+	0.001	0.00
Humboldt Bay Jetty	15	1986 - 2009	NS	+	0.001	0.00	NS	+	0.001	0.01
Pt. Delgado Shelter Cove	16	1986 - 2009	▲	+	0.003	0.14	NS	48	-0.006	0.11
Pt. Arena Lighthouse	16	1986 - 2009	▲	+	0.008	0.47	NS	>100	-0.003	0.05
Tomales Bay Spenger's Res.	16	1986 - 2009	NS	+	0.016	0.10	NS	+	0.017	0.09
SFB Emeryville	13	1987 - 2009	NS	>100	-0.002	0.02	▼	18	-0.017	0.50
SFB San Mateo Bridge	16	1986 - 2009	NS	>100	-0.001	0.01	NS	96	-0.003	0.05
SFB Dumbarton Bridge	16	1986 - 2009	NS	+	0.003	0.02	NS	+	0.003	0.05
Monterey Bay Pt. Santa Cruz	16	1986 - 2009	NS	12	-0.024	0.13	▼	17	-0.018	0.34
Monterey Bay Elkhorn Slough	9	1994- 2005	NS	40	-0.007	0.08	NS	32	-0.009	0.37
Monterey Bay Moss Landing	14	1990 - 2009	NS	80	-0.004	0.12	▼	38	-0.008	0.36
Pacific Grove Lovers Pt.	16	1986 - 2009	NS	+	0.004	0.10	NS	>100	0	0.00
San Simeon Pt.	15	1986 - 2009	▲	+	0.004	0.35	▼	55	-0.006	0.48
San Luis Obispo Bay	16	1986 - 2008	NS	>100	-0.002	0.07	▼	58	-0.005	0.35
Pt. Conception	13	1986 - 2004	NS	>100	-0.002	0.06	NS	>100	-0.001	0.02
Pt. Santa Barbara	15	1986 - 2008	NS	38	-0.008	0.33	NS	>100	-0.002	0.01
Santa Cruz Is. Fraser Pt.	13	1986 - 2008	NS	>100	-0.001	0.01	NS	+	0.003	0.20
Pt. Dume	15	1986 - 2008	▲	+	0.010	0.49	NS	92	-0.003	0.09
Santa Monica Bay Las Tunas Beach	11	1990 - 2008	NS	47	-0.006	0.27	NS	38	-0.008	0.16
Marina Del Rey South Jetty	15	1986 - 2008	▼	37	-0.008	0.34	▼	19	-0.015	0.53
Redondo Beach Municipal Jetty	11	1990 - 2008	NS	>100	-0.001	0.01	NS	39	-0.008	0.40
Royal Palms	15	1986 - 2008	▼	37	-0.008	0.37	NS	51	-0.006	0.13
San Pedro Harbor	14	1986 - 2008	▼	27	-0.011	0.28	▼	26	-0.012	0.32
Long Beach Breakwater	11	1990 - 2008	NS	+	0.002	0.03	▼	25	-0.012	0.47
Anaheim Bay West Jetty	15	1986 - 2008	▼	43	-0.007	0.34	NS	45	-0.007	0.12
Newport Beach West Jetty	13	1986 - 2008	NS	>100	-0.003	0.06	NS	50	-0.006	0.13
South Catalina Is. Bird Rock	13	1986 - 2008	NS	+	0.004	0.15	NS	86	-0.003	0.19
Oceanside Municipal Jetty	15	1986 - 2008	NS	+	0.001	0.00	NS	78	-0.004	0.05
La Jolla	13	1986 - 2008	NS	+	0.004	0.10	NS	>100	-0.002	0.01
Mission Bay Ventura Bridge	15	1986 - 2008	NS	+	0.004	0.10	NS	42	-0.007	0.17
SD Bay Harbor Island	15	1986 - 2008	NS	>100	-0.002	0.02	▼	24	-0.013	0.54
Pt. Loma Lighthouse	13	1986 - 2005	▼	52	-0.006	0.34	▼	24	-0.013	0.51
SD Bay Coronado Bridge	13	1989 - 2008	▼	19	-0.016	0.46	NS	28	-0.011	0.30
Imperial Beach North Jetty	15	1986 - 2008	NS	89	-0.003	0.08	NS	49	-0.006	0.09

Table 7a. Temporal trend results at SMW sites for total mercury. Stations are organized from north to south. Green rows are resident mussel sites, remainder are transplants. Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with “+” have a positive but not statistically significant slope, half life not calculated.

Station Name	N (yrs)	Years	Mercury			
			Trend	Half-life (yrs)	Slope	R^2
Trinidad Head	27	1977 - 2010	NS	>100	-0.002	0.01
Mad River Slough	11	1982 - 2003	NS	>100	0	0.00
Samoa Bridge/West	10	1980 - 2003	NS	30	-0.010	0.16
Bodega Head	32	1977 - 2009	▼	36	-0.008	0.22
Tomales Bay	11	1980 - 2010	NS	+	0.006	0.25
Sandholdt Bridge	19	1989 - 2010	▼	23	-0.013	0.46
Pacific Grove	30	1977 - 2010	▼	22	-0.014	0.27
Royal Palms	27	1977 - 2010	▼	17	-0.018	0.36
LA Harbor/National Steel	20	1983 - 2010	NS	>100	-0.001	0.00
LA Harbor/Consolidated Slip	22	1982 - 2010	▼	32	-0.009	0.22
Anaheim Bay/Navy Marsh	18	1983 - 2010	▼	22	-0.013	0.15
Huntington Harbour/Edinger Street	16	1983 - 2003	▼	15	-0.020	0.42
Huntington Harbour/Warner Ave	25	1983 - 2010	▼	15	-0.019	0.49
Newport Bay/Crows Nest	26	1982 - 2010	NS	59	-0.005	0.05
Newport Bay/Highway 1 Bridge	24	1982 - 2010	▼	34	-0.009	0.10
San Diego Bay/Shelter Island	18	1980 - 2010	NS	>100	-0.002	0.03

Table 7b. Temporal trends at NMW sites for total mercury. Stations are organized from north to south. Blue rows are Areas of Biological Significance (ASBS). Trend symbols: NS = no significant trend ($p > 0.05$); ▼= declining trend ($p < 0.05$). Sites with "+" have a positive but not statistically significant slope, half life not calculated.

			Mercury			
Station Name	N (yrs)	YEARS	Trend	Half-life (yrs)	Slope	R^2
Crescent Pt. St. George	15	1986 - 2009	NS	>100	-0.001	0.07
Eureka Samoa Bridge	11	1990 - 2009	NS	+	0.001	0.12
Humboldt Bay Jetty	15	1986 - 2009	NS	+	0	0.03
Pt. Delgado Shelter Cove	16	1986 - 2009	NS	>100	-0.001	0.28
Pt. Arena Lighthouse	16	1986 - 2009	NS	+	0.001	0.09
Tomales Bay Spenger's Res.	16	1986 - 2009	NS	>100	-0.001	0.03
SFB Emeryville	13	1987 - 2009	▼	54	-0.006	0.36
SFB San Mateo Bridge	16	1986 - 2009	NS	>100	-0.002	0.12
SFB Dumbarton Bridge	16	1986 - 2009	NS	>100	-0.002	0.11
Monterey Bay Pt. Santa Cruz	16	1986 - 2009	▼	>100	-0.002	0.27
Monterey Bay Elkhorn Slough	9	1994- 2005	NS	+	0	0.00
Monterey Bay Moss Landing	14	1990 - 2009	NS	+	0	0.00
Pacific Grove Lovers Pt.	16	1986 - 2009	NS	+	0.001	0.05
San Simeon Pt.	15	1986 - 2009	NS	>100	-0.001	0.02
San Luis Obispo Bay	16	1986 - 2008	NS	+	0	0.03
Pt. Conception	13	1986 - 2004	NS	+	0.001	0.21
Pt. Santa Barbara	15	1986 - 2008	▼	83	-0.004	0.21
Santa Cruz Is. Fraser Pt.	13	1986 - 2008	NS	>100	0	0.06
Pt. Dume	14	1986 - 2008	NS	>100	0	0.00
Santa Monica Bay Las Tunas Beach	11	1990 - 2008	▼	94	-0.003	0.59
Marina Del Rey South Jetty	15	1986 - 2008	▼	>100	-0.002	0.50
Redondo Beach Municipal Jetty	11	1990 - 2008	▼	>100	-0.002	0.48
Royal Palms	15	1986 - 2008	▼	>100	-0.002	0.28
San Pedro Harbor	14	1986 - 2008	▼	>100	-0.001	0.46
Long Beach Breakwater	11	1990 - 2008	NS	+	0	0.00
Anaheim Bay West Jetty	15	1986 - 2008	▼	>100	-0.001	0.43
Newport Beach West Jetty	13	1986 - 2008	▼	>100	-0.002	0.42
South Catalina Is. Bird Rock	13	1986 - 2008	NS	>100	-0.001	0.21
Oceanside Municipal Jetty	15	1986 - 2008	NS	+	0	0.01
La Jolla	13	1986 - 2008	▼	91	-0.003	0.54
Mission Bay Ventura Bridge	15	1986 - 2008	▼	>100	-0.001	0.39
SD Bay Harbor Island	15	1986 - 2008	NS	>100	-0.001	0.21
Pt. Loma Lighthouse	13	1986 - 2005	▼	>100	-0.001	0.33
SD Bay Coronado Bridge	13	1989 - 2008	▼	94	-0.003	0.50
Imperial Beach North Jetty	15	1986 - 2008	▼	>100	-0.001	0.57



Site Number	Site Name
1	Crescent City
2	Trinidad Head
3	Mad River Slough
4	Samoa Bridge/West
5	Bodega Head
6	Tomales Bay
7	San Francisco Bay/Fort Baker
8	J. Fitzgerald
9	Sandholdt Bridge
10	Pacific Grove
11	Royal Palms
12	LA Harbor/National Steel
13	LA Harbor/Consolidated Slip
14	Anaheim Bay/Navy Marsh
15	Huntington Harbour/Edinger Street
16	Huntington Harbour/Warner Ave
17	Newport Bay/Turning Basin
18	Newport Bay/Rhine Channel
19	Newport Bay/Crows Nest
20	Newport Bay/Highway 1 Bridge
21	Newport Bay/Bay Island
22	Oceanside
23	San Diego Bay/Harbor Island
24	San Diego Bay/Shelter Island

Figure 1. Location map of 24 State Mussel Watch stations examined for trends in this report.

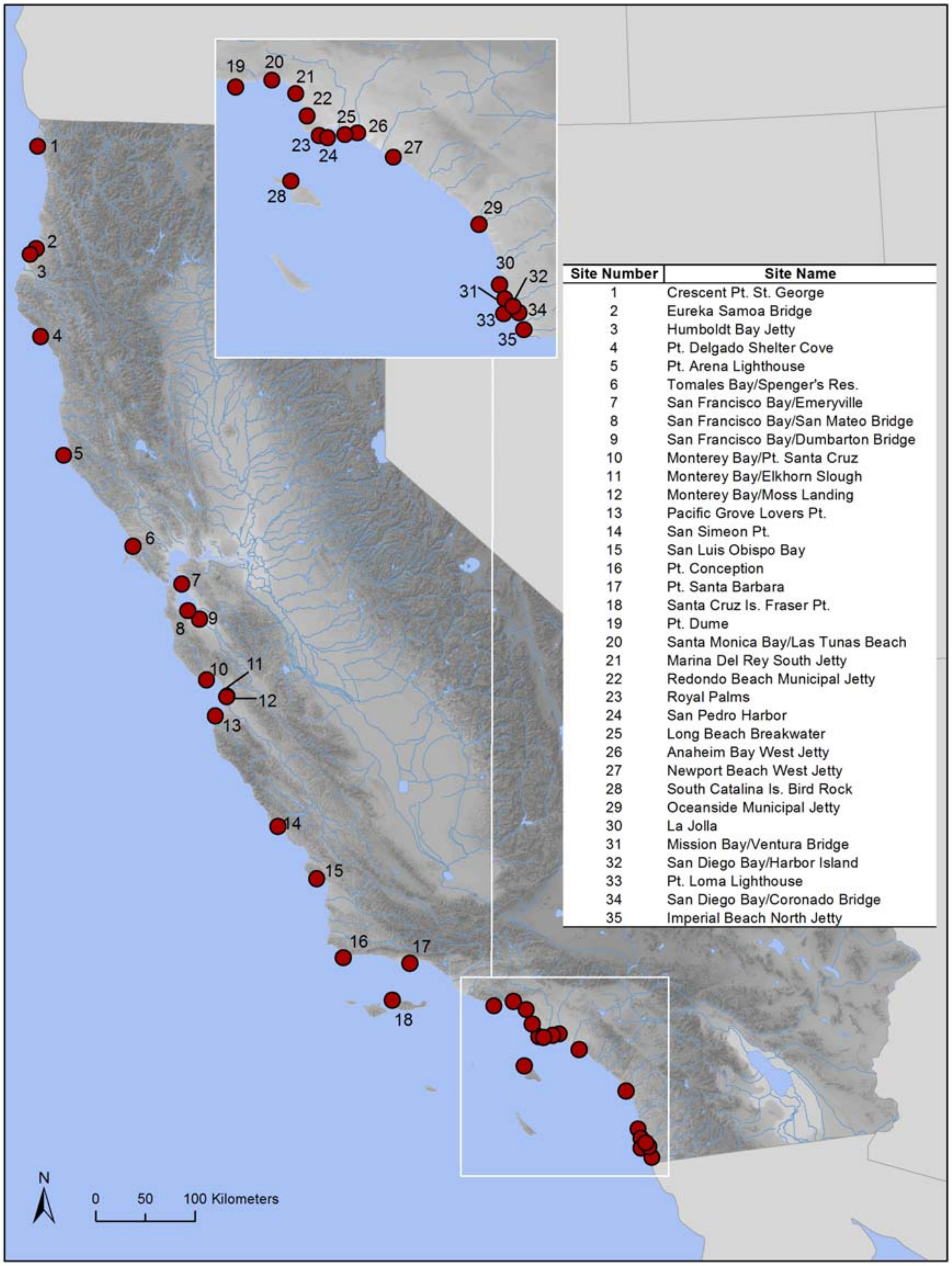


Figure 2. Location map of the 35 National Mussel Watch stations examined for trends in this report.

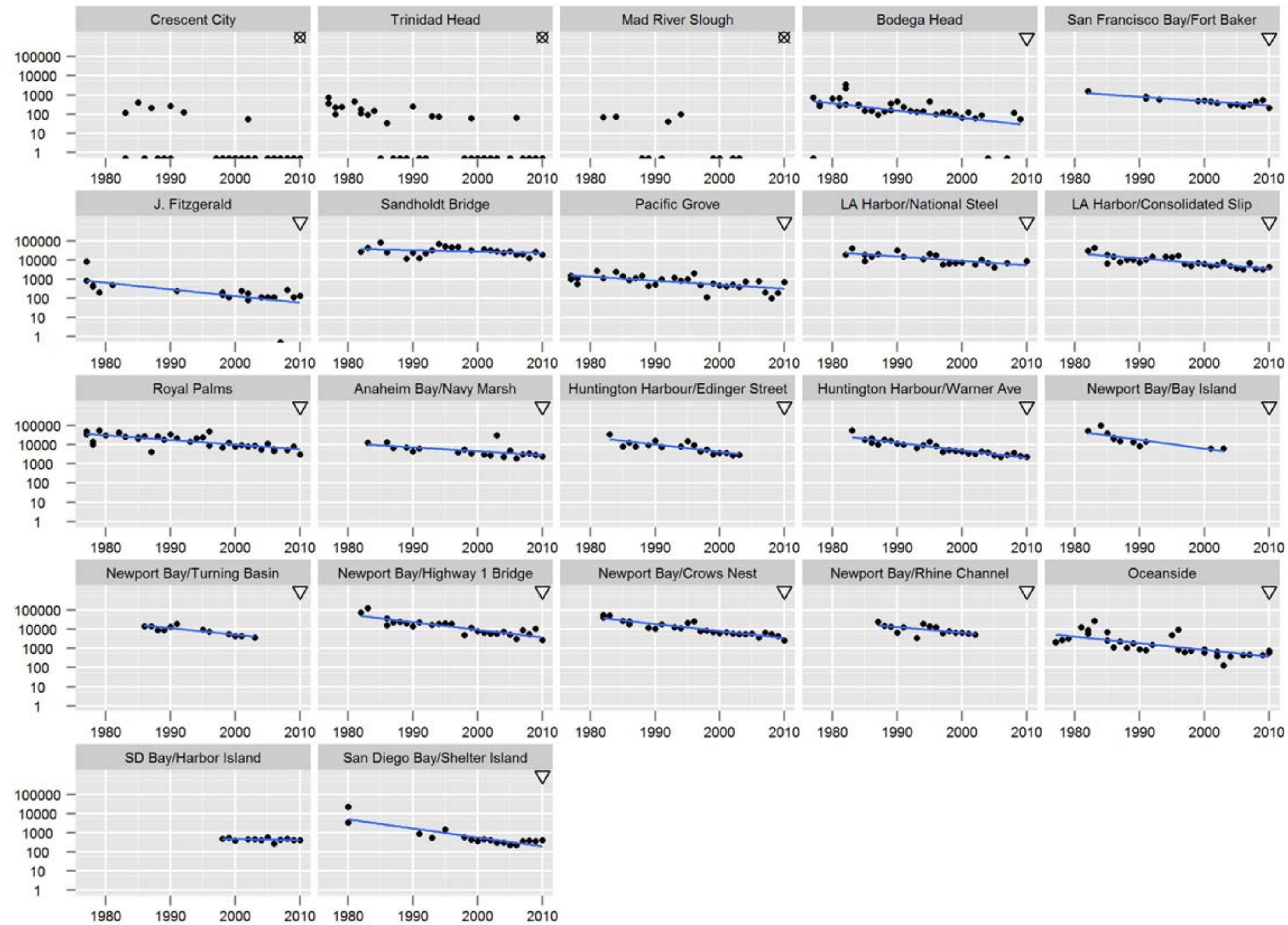


Figure 3. Total DDTs in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-billion, lipid weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (ND: > 50% results are non-detect, no trend determined; ▽: significant log-linear decline).

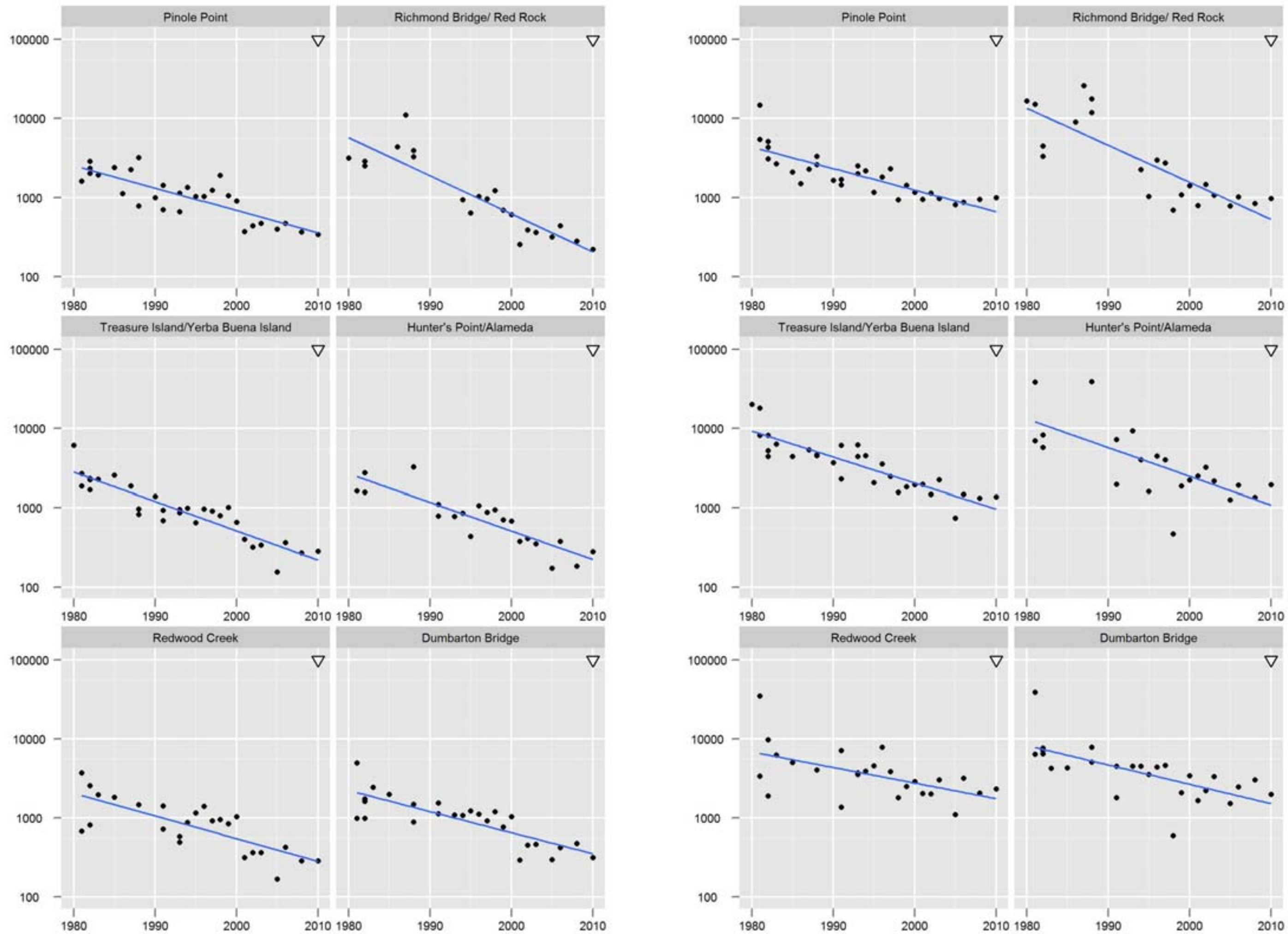


Figure 4. Total DDTs (left plot) and Total PCBs (right plot) in *Mytilus californianus* at State Mussel Watch / Regional Monitoring Program stations sampled from 1980 – 2010. Units are parts-per-billion, lipid weight. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

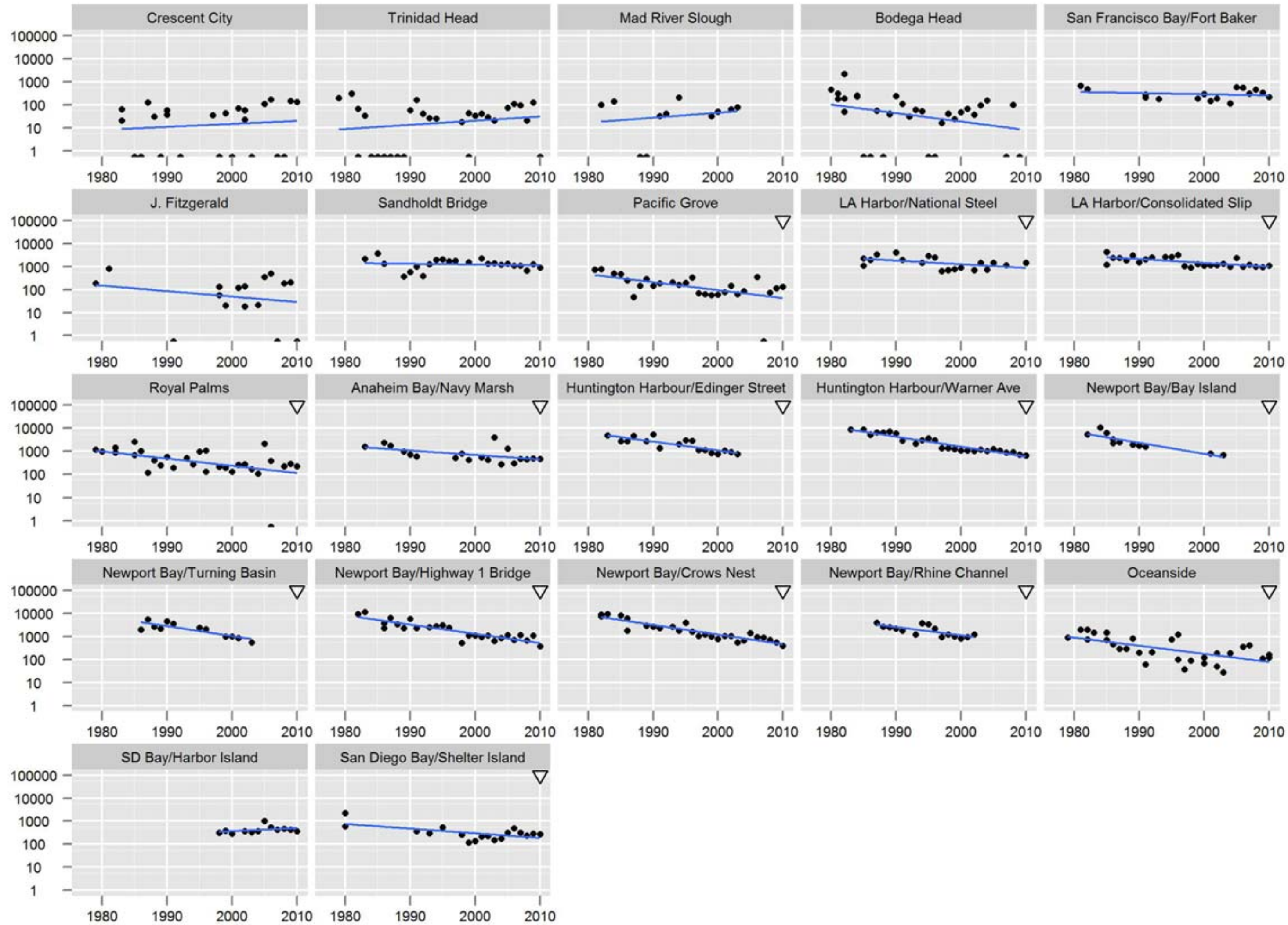


Figure 5. Total chlordanes in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-billion, lipid weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

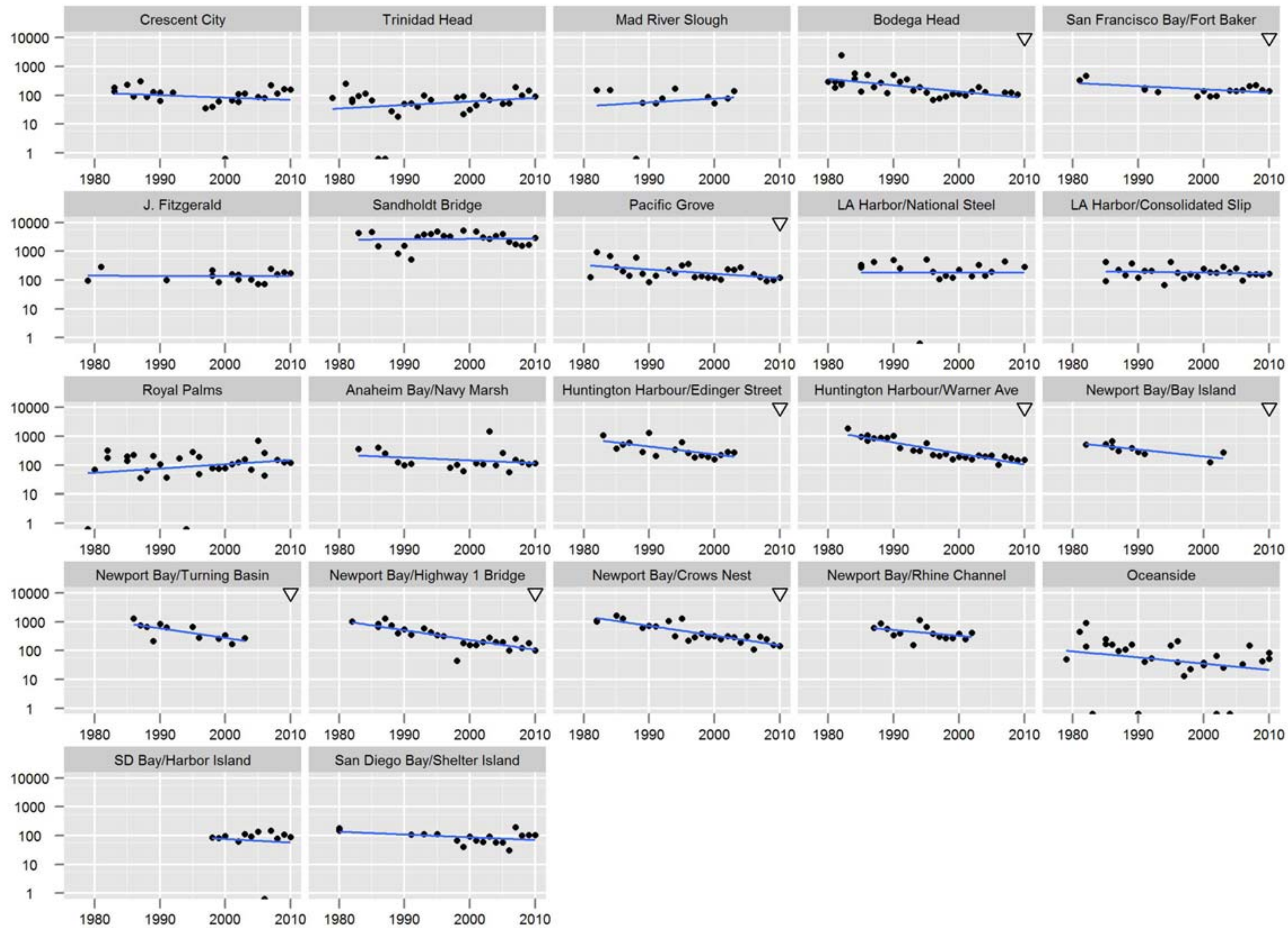


Figure 6. Dieldrin in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-billion, lipid weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

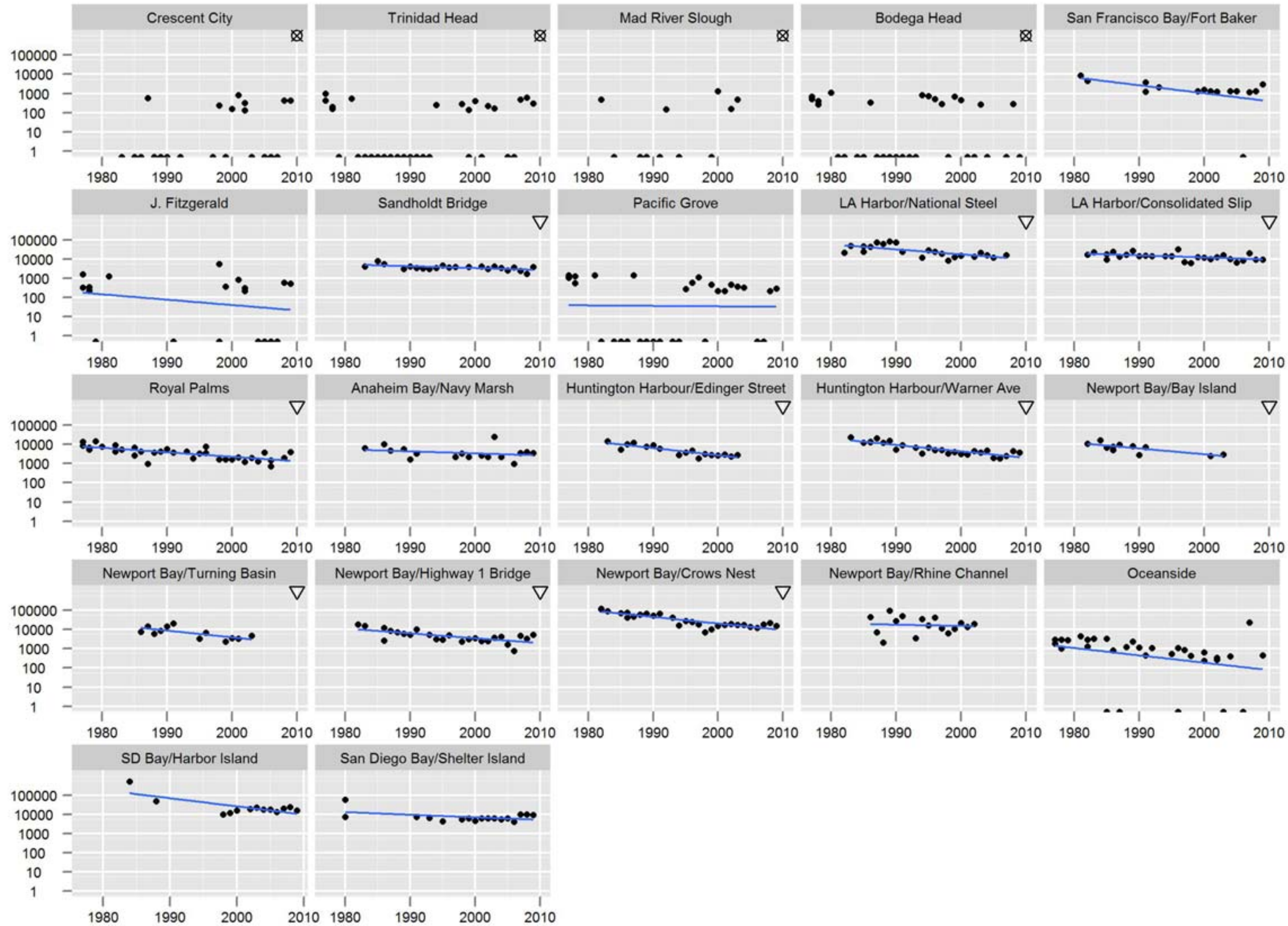


Figure 7. Total PCB Aroclors in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2009. Units are parts-per-billion, lipid weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (ND: > 50% results are non-detect, no trend determined; ▽: significant log-linear decline; $p < 0.05$).

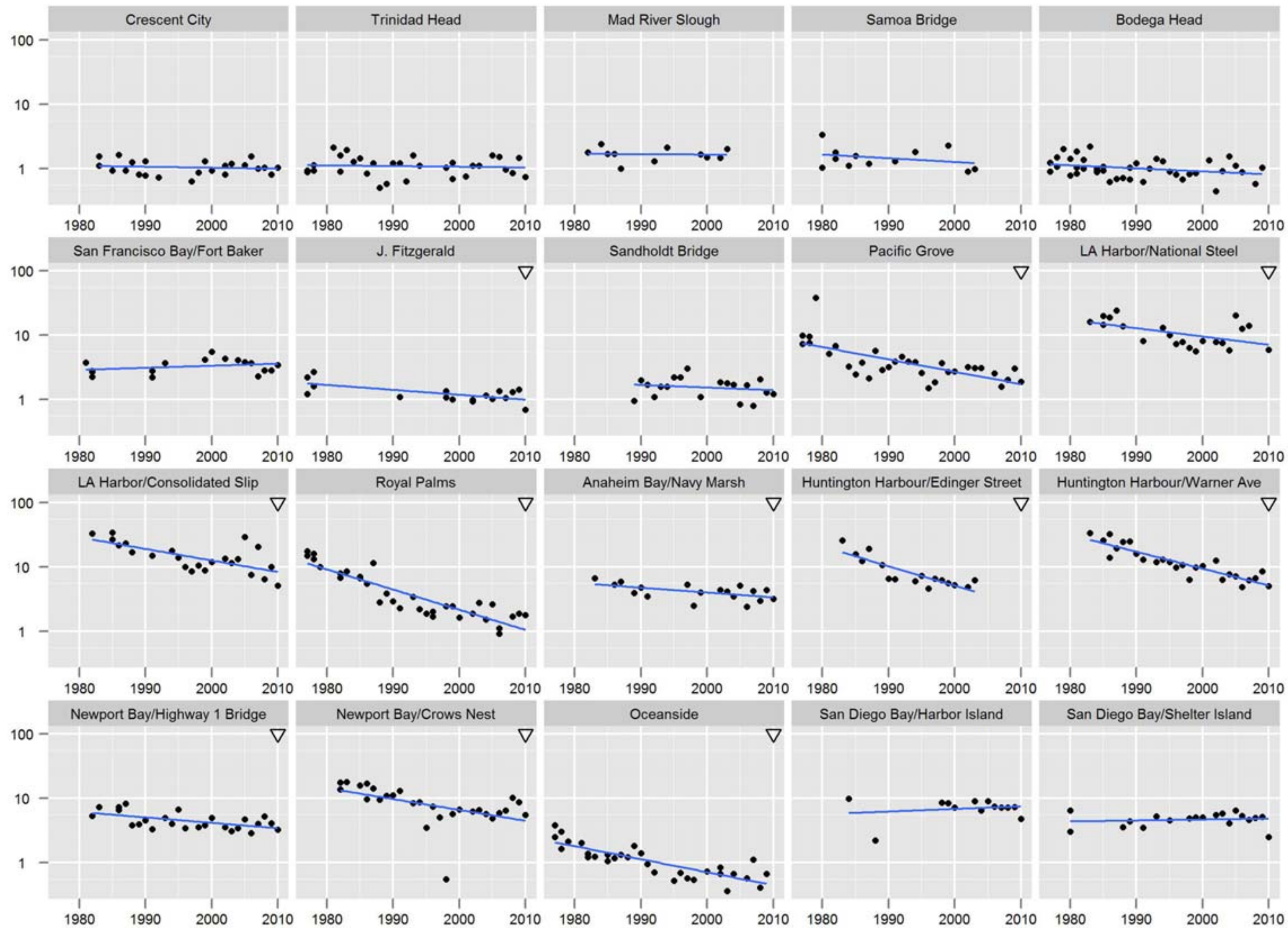


Figure 8. Lead in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-million, dry weight. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

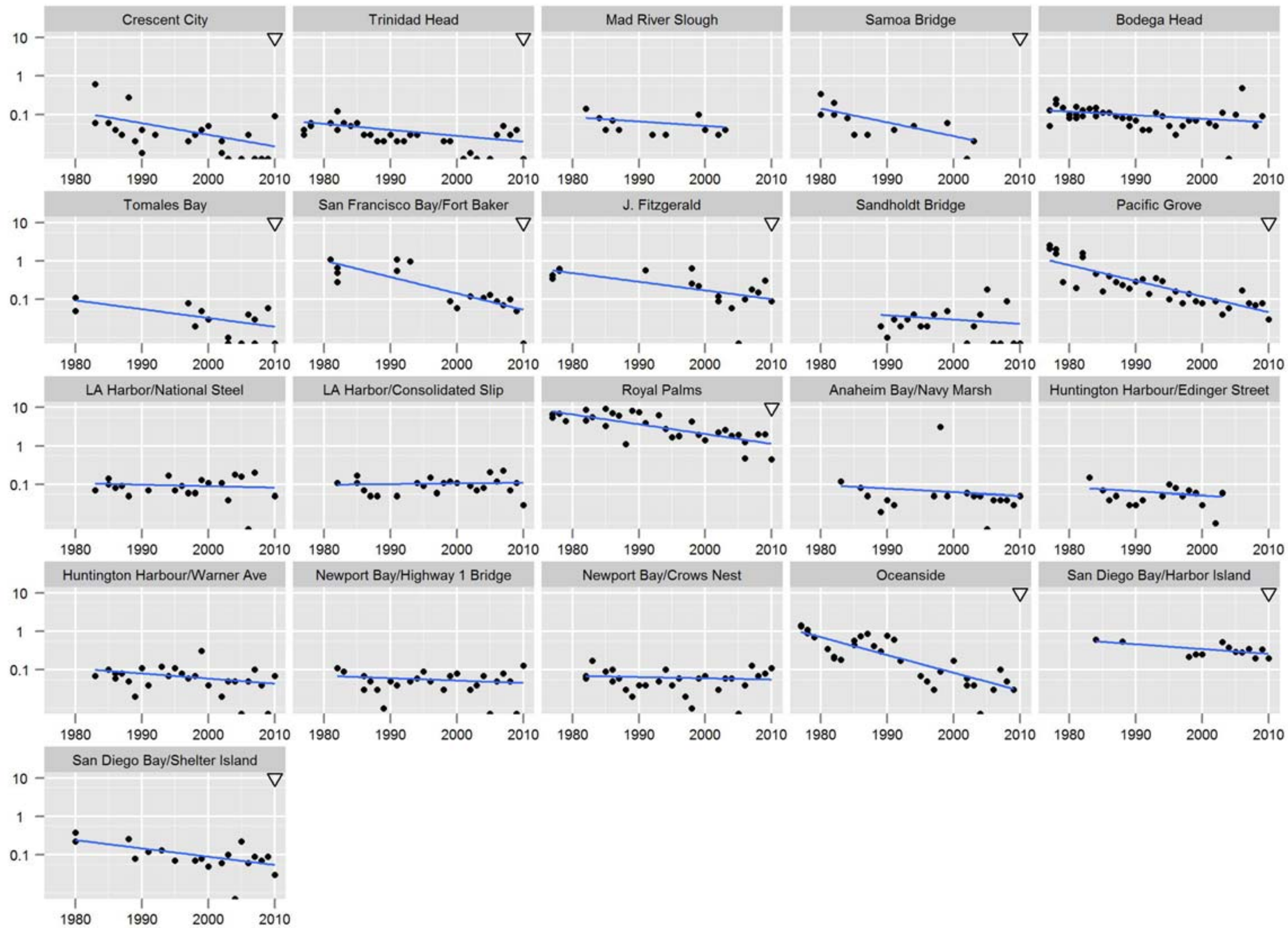


Figure 9. Silver in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-million, dry weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

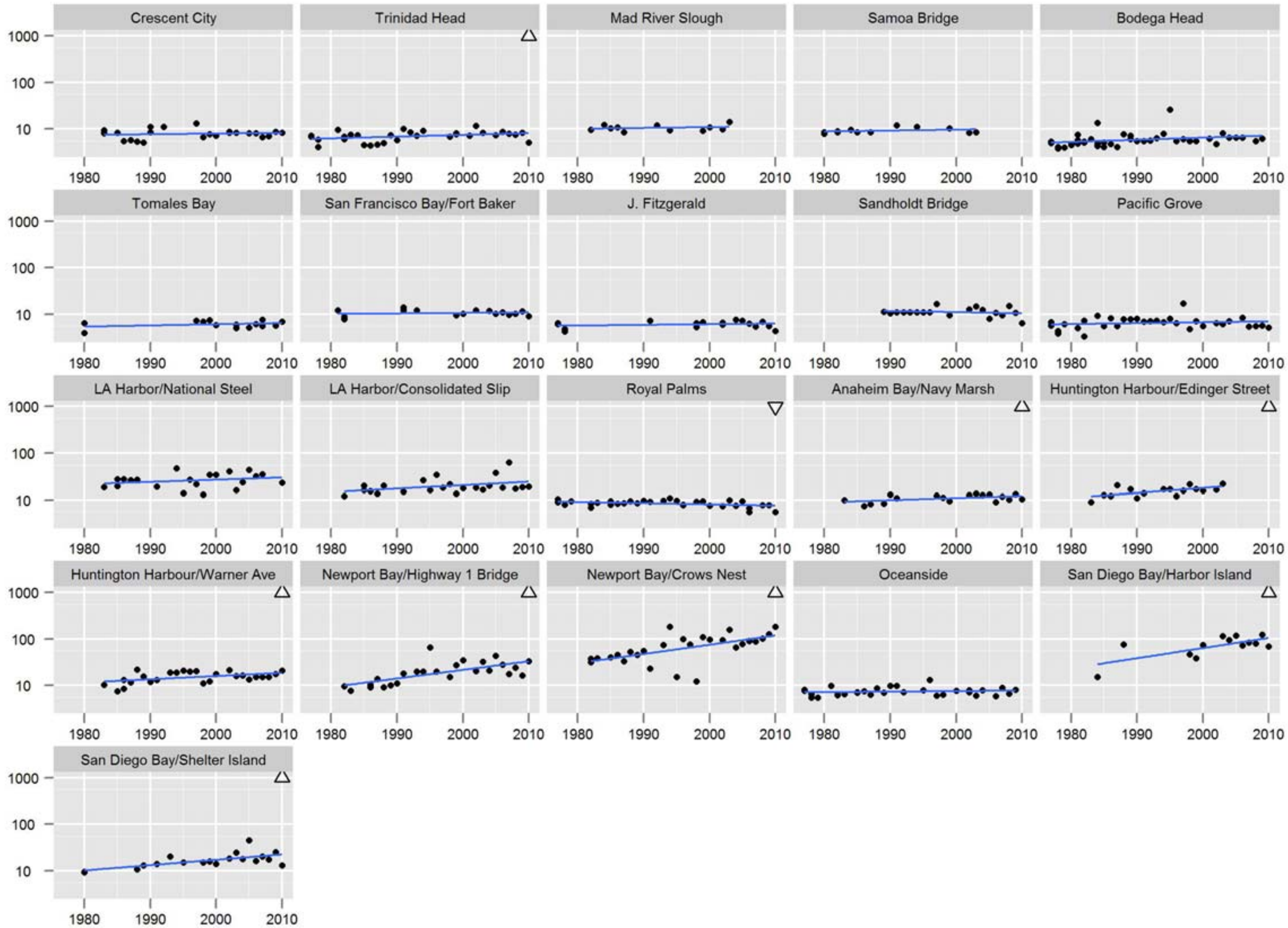


Figure 10. Copper in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-million, dry weight. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline; △: significant log-linear increase; $p < 0.05$).

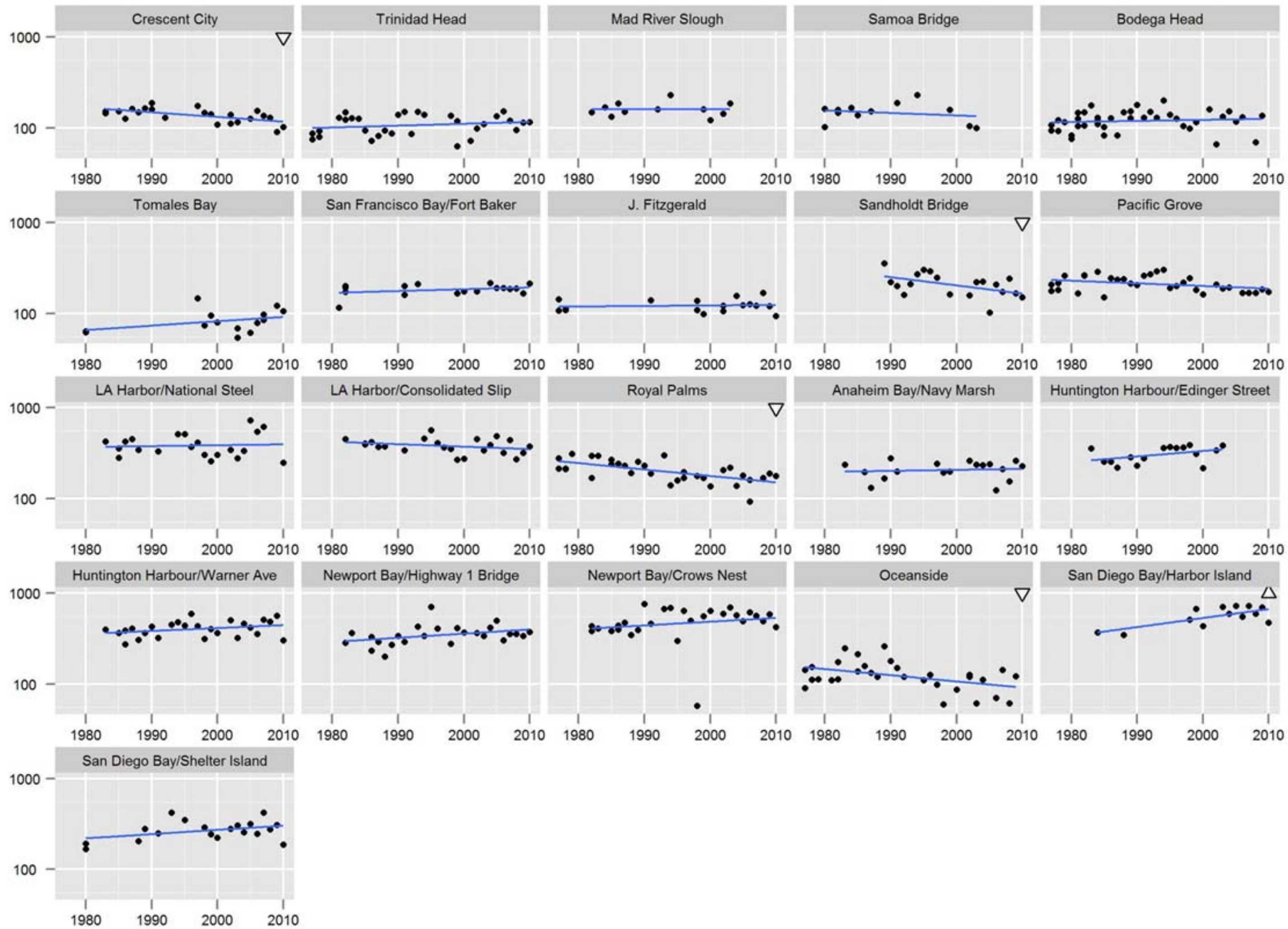


Figure 11. Zinc in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-million, dry weight. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline; △: significant log-linear increase;
$p < 0.05$).

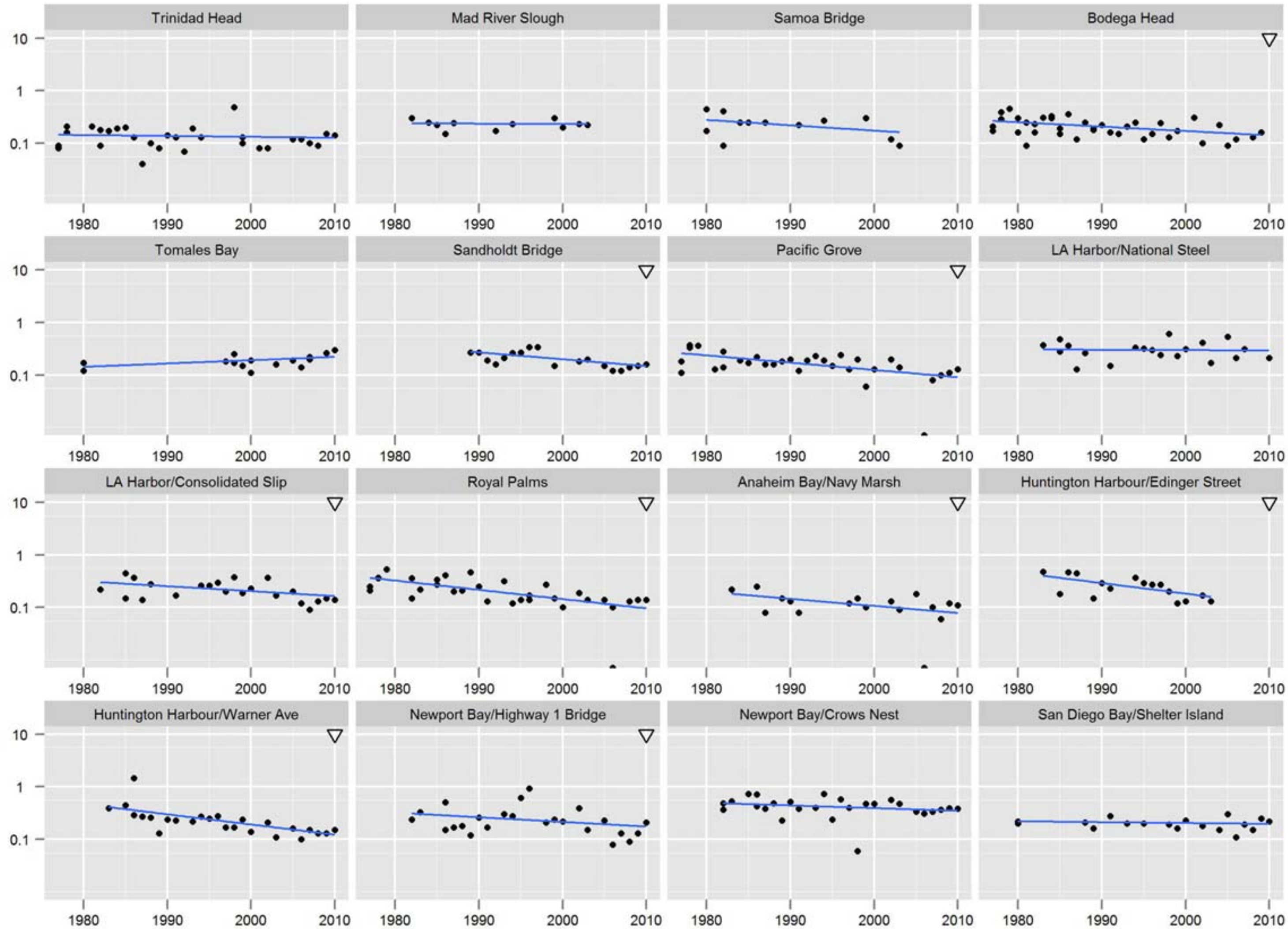


Figure 12. Total mercury in *Mytilus californianus* at State Mussel Watch stations sampled from 1977 – 2010. Units are parts-per-million, dry weight. Values below detection are shown by half circles along x-axis. Symbol on top right of each sub-plot indicates the result of log-linear trend analysis (▽: significant log-linear decline, $p < 0.05$).

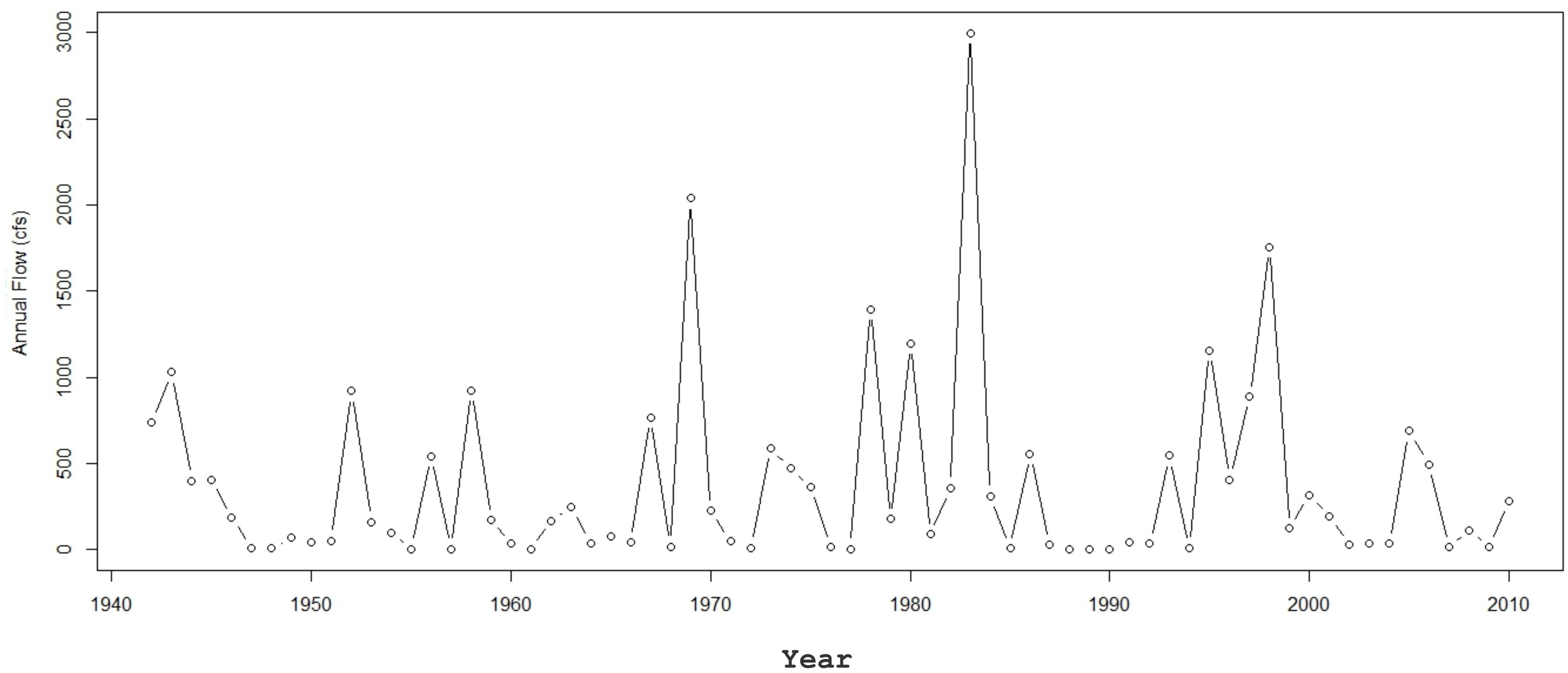


Figure 13. Annual mean flow at the USGS gauge for Salinas River at Spreckles.