

## ECOTOXICOLOGIC CHANGE AT A REMEDIATED SUPERFUND SITE IN SAN FRANCISCO, CALIFORNIA, USA

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**Abstract**—Lauritzen Channel is an industrial waterway adjacent to the former United Heckathorn facility in the inner Richmond Harbor area of San Francisco Bay, California, USA. Marine sediments at this Superfund site were dredged from late 1996 through early 1997 to remove the primary chemicals of concern: DDT, and dieldrin. This study assessed the Lauritzen Channel marine environment immediately before and approximately one year after the dredging of sediments. The study included chemical analysis of sediments, tissue concentrations of transplanted mussels, toxicity testing of sediment samples, and characterization of benthic community structure. Results indicated that sediment toxicity to bivalve larvae (*Mytilus galloprovincialis*) decreased in postremediation samples, but that toxicity to the amphipod *Eohaustorius estuarius* increased significantly. Assessment of benthos at this site suggested a transitional benthic community structure. In addition, postremediation sediments remained contaminated by a variety of organic chemical compounds, including DDT, dieldrin, chlordane, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls. Tissue concentrations of DDT and dieldrin in mussels (*M. galloprovincialis*) were lower than those in preremediation samples, indicating that although sediment concentrations of organochlorine pesticides remained high, concentrations of these chemicals in the water column were reduced after dredging. This study demonstrates that the components of the site assessment were useful in determining effectiveness of the remediation activities.

**Keywords**—Marine superfund Sediment remediation DDT Toxicity

## INTRODUCTION

Lauritzen Channel is an industrial waterway adjacent to the former United Heckathorn facility in the inner Richmond Harbor area of San Francisco Bay, California, USA. Various corporations operated pesticide-processing facilities in the upland area adjacent to this site from approximately 1945 to 1966. These activities resulted in soil and sediment contamination by chlorinated pesticides, primarily DDT and dieldrin, and the area was designated by the U.S. Environmental Protection Agency (U.S. EPA) as a Superfund site in 1990. Several actions were taken to clean the most contaminated areas of the site, including removal of contaminated soil from the upland areas and an embankment adjacent to the marine habitat.

The U.S. EPA completed an ecologic risk assessment for the marine environment at the site in 1992 that included measures of sediment chemistry and toxicity, bioaccumulation, and benthic community structure [1]. Results of this study confirmed that dieldrin and DDT were the primary chemicals of concern, and that concentrations of these compounds were sufficient to account for low amphipod survival in the sediment toxicity tests, degraded benthic community structure, and significant bioaccumulation of DDT in the resident and transplanted biota [1,2]. As part of the remediation activities in the marine habitat, approximately 100,000 metric tons of contam-

inated sediment were removed from Lauritzen Channel. In this process, the channel was dredged to remove all sediment above the relatively uncontaminated, older bay mud [3] and then capped with 15 to 46 cm of clean sand. These activities were completed in April 1997.

The present study was sponsored by the California Department of Fish and Game to assess the marine environment immediately before and approximately one year after the dredging of Lauritzen Channel. The study components included chemical analysis of sediments, tissue concentrations of transplanted mussels, toxicity testing of sediment samples, and characterization of benthic community structure.

Remediation of the United Heckathorn Superfund site provided an opportunity to use standard ecotoxicologic tools to evaluate the restoration of a pesticide-contaminated marine habitat. The objectives of this study were twofold: to assess whether remediation activities reduced concentrations of the chemicals of concern in the marine environment to levels that prevent risk of injury to marine biota, and to evaluate the effectiveness of the various monitoring components to describe ecotoxicologic change because of dredging and capping at the site. The results are intended to provide guidance for future projects involving remediation of contaminated marine sediments.

## METHODS

*Station locations and study dates*

For comparative purposes, stations were selected on the basis of those sampled during the ecologic risk assessment

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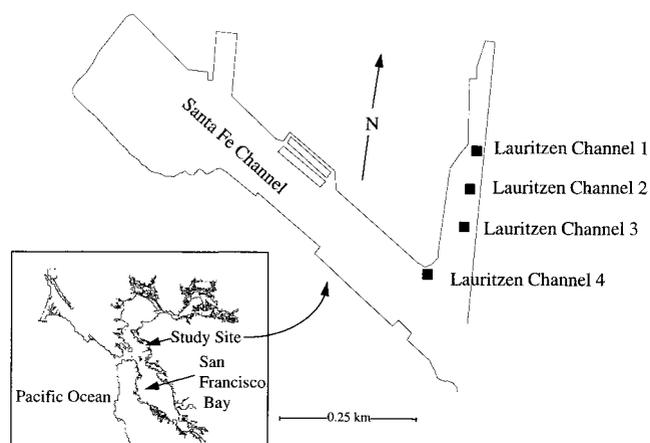


Fig. 1. Location of sediment sampling stations in Lauritzen Channel. Global Imaging System (GIS) coordinates (GIS latitude/GIS longitude) by station: station 1, 37.92261667/122.36701667; station 2, 37.92246667/122.36701667; station 3, 37.92178333/122.36703333; station 4, 37.92030000/122.36846667.

study described by Lee et al. [1] and Swartz et al. [2]. Stations designated as Lauritzen Channel 1 to 4 were sampled before and after remediation of channel sediments (Fig. 1). Lauritzen Channel 1 was identified in previous studies as the most contaminated station. Lee et al. [1] found that contamination decreased along a gradient leading out of the Channel toward Lauritzen Channel 4, which was located at the confluence of the Lauritzen and the Santa Fe Channels (Fig. 1). Note that Global Imaging System coordinates were not available for stations 1 to 4 reported in Lee et al. [1]; therefore, the locations for these stations in the present study are based on best approximations from the site maps of the previous study. Stations 1 to 4 in the present study are identified by Global Imaging System coordinates (Fig. 1). Samples for preremediation toxicity tests were collected during July 1996, and preremediation mussel bioaccumulation analyses were conducted from August through September 1996. Channel dredging began in late September 1996 and was completed in April 1997. Samples for postremediation toxicity tests and bulk-phase chemistry were collected during March 1998. Postremediation bioaccumulation in mussels was conducted for 125 days, ending in September 1997.

#### Sampling procedures

**Homogenized sediments.** Sediment sampling required collection of homogenized sediment for solid-phase chemistry and amphipod toxicity tests and of intact (i.e., unhomogenized) sediment cores for sediment-water interface toxicity tests. Sediments were collected using a Young-modified, Kynar<sup>®</sup>-coated (Elf Atochem, Paris, France), Van Veen grab sampler according to the procedures described by Fairey et al. [4]. Surficial sediments (top 5 cm) were collected from half of each grab sample using a Teflon<sup>®</sup> scoop; intact cores were taken from the second half of each grab (described later). Multiple grabs were deployed at each station to obtain enough sediment homogenate and intact sediment cores for chemical and physical analyses and for toxicity testing. Except for the intact cores, sediment samples were composited into 6-L polycarbonate tubs and covered with a Teflon sheet. The tubs were then purged with nitrogen gas, sealed, and placed in an ice chest for transport to the laboratory, where the sediments were thoroughly rehomogenized using a polycarbonate rod and ali-

quoted for solid-phase toxicity testing and chemistry as described by Fairey et al. [4].

**Intact sediment cores.** Intact sediment cores were collected for toxicity testing from the second half of each grab sample by pressing polycarbonate core tubes 5 cm into the sediment, sealing the bottom of the cores for removal from the sampler, and then removing the cores. Cores were quickly sealed with polyethylene caps, dried and tightly sealed with Parafilm<sup>®</sup> to prevent leakage, and then stored upright on ice for transport. Core integrity was confirmed by the presence of a shallow layer of overlying water atop the sediment.

**Benthics.** Benthic community structure was characterized in the sediment samples collected at each station after remediation had been completed. The methods used followed those described in Anderson et al. [5]. Cores for characterizing benthic community structure were collected from the Van Veen grab sampler at the same time that sediment samples were collected for chemistry and toxicity. The coring device was a polycarbonate cylinder with a diameter of 10 cm that enclosed an area of 0.0071 m<sup>2</sup> and sampled to an average depth of 10 cm. One sample was collected at each station, sieved through a 0.5-mm screen, fixed with formalin, and then transferred to isopropyl alcohol 3 d later. All samples were sorted, and infaunal organisms were identified to species (whenever possible) or to the next lowest taxonomic group.

**Toxicity testing procedures.** Sediment samples were held at 4°C until required for testing. All solid-phase sediment tests were initiated within 14 d of the sample collection date. Solid-phase sediment toxicity was assessed using the 10-d amphipod survival protocol for *Eohaustorius estuarius* [6]. Test containers were 1-L glass beakers containing 2 cm of sediment and filled to the 700-ml line with seawater adjusted to 20‰ using distilled water. Five laboratory replicates of each sample, including a negative sediment control consisting of five laboratory replicates of home sediment from the amphipod collection site, Yaquina Bay, Oregon, USA, were tested. After 10 d, the sediments were sieved through a 0.5-mm screen (Aquatic Eco-Systems, Apopka, FL, USA) to recover the test animals, and the number of survivors was recorded for each replicate. Overlying water-quality parameters, including ammonia, dissolved oxygen, pH, and salinity, were measured in one replicate test container from each sample. Interstitial sulfide and ammonia were also examined. Measurements were taken at the beginning and at the end of all tests. Positive control reference tests were conducted concurrently with each sediment test using cadmium chloride as a reference toxicant.

Sediment-water interface exposures were conducted according to the methods described by Anderson et al. [7]. Intact sediment cores were returned to the laboratory and prepared for testing by adding 300 ml of 28‰ overlying water. The cores were then allowed to equilibrate overnight with slow aeration. Before test initiation, 25- $\mu$ m mesh screen tubes were inserted into each sediment core and positioned 1 cm above the sediment.

Sediment toxicity was assessed using embryo-larval development of the mussel *Mytilus galloprovincialis* [8]. Approximately 200 mussel embryos were pipetted into the screen tubes and exposed for 48 h. Tests were terminated by removing the screen tube and then rinsing larvae into vials to be fixed with 5% formalin. All resulting larvae were counted in each test container at the end of the exposure to determine the percentage of embryos that developed into live, normal larvae. Five laboratory replicates of each sample were tested, with an

additional sacrificial replicate for water quality (i.e., overlying water dissolved oxygen, pH, salinity, and ammonia). A negative sediment control consisting of five laboratory replicates of Yaquina Bay home sediment was included as well. Positive control reference tests were conducted concurrently using cadmium chloride as a reference toxicant.

### Chemical analyses

Because bulk-phase chemical concentrations at this site had been well characterized in several previous studies, including the extensive ecologic risk assessment completed by the U.S. EPA [1], we did not measure bulk-phase chemistry before remediation. Instead, the analyses conducted by U.S. EPA and reported in Swartz et al. [2] were used for comparison with postremediation analyses conducted as part of the current study. Trace organic compounds were measured in sediment homogenates collected during the postremediation sampling described earlier. Samples were analyzed for 24 polychlorinated biphenyl (PCB) congeners, 36 pesticides, and 24 polycyclic aromatic hydrocarbons (PAHs) using modifications of the methods described by Sloan et al. [9]. Sediment extracts were divided into two portions: one for chlorinated hydrocarbon analysis, and the other for PAH analysis. The chlorinated hydrocarbon portion was separated into two fractions on a silica/alumina column and then concentrated for analysis using a Hewlett-Packard (Palo Alto, CA, USA) 5890 Series II capillary gas chromatograph with an electron-capture detector. The PAH portion was eluted through a silica/alumina column with methylene chloride and then concentrated for analysis using gas chromatography/mass spectrometry in single ion monitoring mode. Standard quality-assurance procedures, including measurement of standard reference materials as well as quantification of surrogate recoveries and matrix spikes, were followed during all analyses; all chemical analyses met prescribed quality assurance guidelines.

Percent total organic carbon was determined using a Control Equipment Model 240-A elemental analyzer [10]. Sample grain size was determined using procedures described by Folk [11], incorporating both wet and dry sieve techniques, and was reported as percent fines.

### Bioaccumulation

**Preremediation.** Bags of mussels (*M. californianus*) were deployed at all four sampling locations approximately 1 year before dredging. Bags were suspended from pier pilings below the low tide level. Mussels remained in the field for 23 d (July 17–August 9, 1996), after which they were retrieved and taken to the California Department of Fish and Game Mussel Watch facility in Moss Landing, California, and stored frozen. Mussels were prepared and dissected under positive-pressure, clean-room conditions [12], and concentrations of selected trace organic compounds were analyzed according to the methods described by Sloan et al. [9].

**Postremediation.** Postremediation monitoring of bioaccumulation in deployed mussels was conducted by Battelle Marine Sciences Pacific Northwest Laboratory (Sequim, WA, USA) for U.S. EPA Region IX. Methods were comparable to those described earlier, except that mussels were deployed for a longer time period and were placed at two of the four preremediation monitoring stations (station 2 and 4). Postremediation mussels remained in the field for 125 d (September 3, 1997, to January 6, 1998). Methods of analysis followed those described by the U.S. EPA [13].

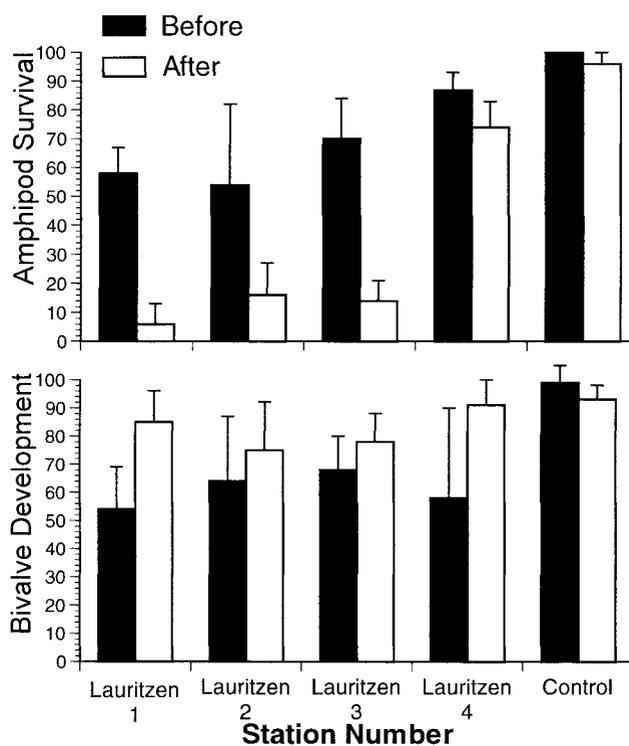


Fig. 2. Survival of amphipods in sediments and development of bivalve embryos exposed at the sediment-water interface in both pre- and postremediation samples from Lauritzen Channel.

## RESULTS

### Sediment toxicity

Amphipod survivals in preremediation sediment samples from Lauritzen Channel were comparable to those reported by Swartz et al. [2] for stations 1 through 4. Survival rates of *E. estuarius* were 58%, 54%, 70%, and 87%, respectively, at stations 1 through 4 (Fig. 2). Development of bivalves (*M. galloprovincialis*) exposed to preremediation sediments at the sediment-water interface was also inhibited at all four stations, with the lowest developmental rate (54%) occurring at station 1 and rates of 64%, 68%, and 58% occurring at stations 2, 3, and 4, respectively (Fig. 2).

Survival of amphipods was considerably lower in postremediation sediments, particularly at stations 1, 2, and 3. Postremediation amphipod survival rates were 6%, 16%, 14%, and 74% at stations 1 through 4, respectively (Fig. 2). Whereas amphipod survival declined, development of bivalve embryos exposed at the sediment-water interface improved in postremediation sediments. No significant toxicity to bivalve development was detected in any samples one year after the site was dredged and capped (Fig. 2).

### Sediment infauna

Sediment samples collected from all stations one year after remediation contained relatively few species and individuals and, for the most part, were dominated by polychaetes and oligochaetes (Table 1). No amphipods were present in any sample, and only one crustacean species was counted at each of two of the stations (*Cumacea*, *Nippoleucon hinumensis*). The number of species identified ranged from five to seven, but the number of polychaete species ranged from two to four (e.g., *Tharyx parvus*, *Eteone lighti*, *Dorvillea articulata*, *Capitella* spp.). In addition, all samples had at least one oligochaete

Table 1. Summary of benthic community structure at Lauritzen Channel stations 1–4 after remediation of channel sediments

|                         | Station<br>1<br>(n) | Station<br>2<br>(n) | Station<br>3<br>(n) | Station<br>4<br>(n) |
|-------------------------|---------------------|---------------------|---------------------|---------------------|
| Total individuals       | 77                  | 124                 | 205                 | 36                  |
| Total species           | 6                   | 5                   | 6                   | 7                   |
| Crustacean individuals  | 0                   | 2                   | 0                   | 6                   |
| Crustacean species      | 0                   | 1                   | 0                   | 1                   |
| Mollusk individuals     | 2                   | 3                   | 5                   | 3                   |
| Mollusk species         | 1                   | 1                   | 2                   | 1                   |
| Polychaete individuals  | 31                  | 6                   | 14                  | 12                  |
| Polychaete species      | 4                   | 2                   | 3                   | 4                   |
| Oligochaete individuals | 44                  | 113                 | 186                 | 15                  |

species. Stations 2 and 3 had relatively greater numbers of individuals because of high densities of oligochaetes, but no trends in either the number of species or individuals could be discerned among the four stations sampled.

#### Sediment chemistry

Bulk-phase analyses of postremediation sediment samples indicated that the site remains relatively contaminated by pesticides. Concentrations of the primary chemicals of concern identified at this site declined at all four stations, but  $\Sigma$ DDT and dieldrin concentrations remained relatively high (Table 2). This is particularly true when concentrations of these pesticides are expressed on an organic carbon (OC)-normalized basis. The concentration of OC-normalized  $\Sigma$ DDT remained particularly high at stations 1 and 2, which were the two most contaminated preremediation stations (Table 2). Postremediation concentrations of OC-normalized  $\Sigma$ DDT were 75%, 50%, 45%, and 28% of the preremediation concentrations at stations 1 through 4, respectively. Concentrations of OC-normalized dieldrin actually increased at stations 1 and 2. Concentration of OC decreased threefold in sediments from station 1 after remediation but increased in sediments from all other stations (Table 2). The relative proportion of DDT metabolites changed in postremediation sediments relative to those measured in preremediation sediments. In the postremediation sediments, 60% of the total was 4',4-DDT, whereas 22% was 4',4-dichlorodiphenyldichloroethane (DDD).

In addition to these pesticides, concentrations of PAH compounds were elevated in postremediation samples from all stations (Table 3). Total bulk-phase concentrations of low- and high-molecular-weight PAHs exceeded the effects range median (ERM) sediment-quality guideline values of Long et al. [14] at all stations. Concentrations of PAHs were particularly high at stations 2 and 3; in fact, the concentration of low-

molecular-weight PAHs at station 3 exceeded the ERM value by as much as 14-fold. The concentrations of total PAHs also exceeded the ERM value at stations 2 and 3. Concentrations of PCBs were higher in postremediation than in preremediation sediments as well. For example, the bulk-sediment concentrations of Arochlors® 1248, 1254, and 1260 (all from Monsanto, St. Louis, MO, USA) were 1400, 690, and 370 ng/g (dry wt), respectively, in the postremediation sediment sample from station 2. Bulk-phase concentrations of Arochlors 1254 and 1260 were 118 and 60 ng/g in preremediation sediment from station 2 [1].

#### Bivalve tissue chemistry

Concentrations of selected pesticides were elevated in caged mussels (*M. galloprovincialis*) deployed for 23 d at the four stations before the remediation of channel sediments (Table 4). Concentrations of total chlordane,  $\Sigma$ DDT, and dieldrin were greater at the stations with the most contaminated sediments (stations 1 and 2). Concentrations of  $\Sigma$ DDT and dieldrin in mussels deployed for 125 d at stations 2 and 4 were measured by the U.S. EPA as part of a postremediation monitoring program. Analysis of  $\Sigma$ DDT and dieldrin in postremediation mussel tissues indicated declines in  $\Sigma$ DDT and dieldrin at station 2 but increases at station 4. The  $\Sigma$ DDT declined by 77% in postremediation mussel tissue at station 2 and was 12% greater at station 4 during postremediation monitoring. Postremediation mussels that were compared to preremediation mussels from station 4, however, apparently were placed between preremediation stations 3 and 4, which could have resulted in elevated pesticide concentrations among these animals relative to those in the preremediation animals.

## DISCUSSION

The results of this study indicate that despite removal of approximately 100,000 metric tons of contaminated sediments and capping with clean San Francisco Bay sand, the sediments of Lauritzen Channel remain toxic to infaunal amphipods and are still polluted with organic chemicals. Toxicity of Lauritzen Channel sediments to the free-burrowing amphipod *E. estuarius* increased at all stations after remediation of channel sediments, particularly at stations 1 to 3, where the mean amphipod survival rate is now 12% (Fig. 2). Relative to sediments tested with amphipods from other U.S. coastal sites [2], these three stations are now among the most toxic of those measured nationwide.

Toxicity to amphipods increased after remediation of channel sediments, but toxicity to bivalve embryos exposed at the sediment-water interface declined. No significant toxicity was detected using bivalve development at any station after dredg-

Table 2. Sediment  $\Sigma$ DDT and dieldrin concentrations before<sup>a</sup> and after site remediation of the Lauritzen Channel

| Chemical                                  | Station 1 |           | Station 2 |           | Station 3 |           | Station 4 |        |
|---|-----------|-----------|-----------|-----------|-----------|-----------|-----------|--------|
|   | Before    | After     | Before    | After     | Before    | After     | Before    | After  |
| $\Sigma$ DDT ( $\mu$ g/kg dry wt)         | 77,700.00 | 21,361.80 | 47,800.00 | 27,883.00 | 26,000.00 | 15,555.00 | 2,740.00  | 840.20 |
| $\Sigma$ DDT ( $\mu$ g/g OC) <sup>b</sup> | 3,500.00  | 2,637.26  | 2,710.00  | 1,366.81  | 1,520.00  | 691.33    | 189.00    | 53.18  |
| Dieldrin ( $\mu$ g/kg dry wt)             | 748.00    | 371.00    | 528.00    | 619.00    | 442.00    | 196.00    | 35.70     | 25.80  |
| Dieldrin ( $\mu$ g/g OC)                  | 35.20     | 45.80     | 28.70     | 30.34     | 25.80     | 8.71      | 2.46      | 1.63   |
| Fines (%)                                 | 92.30     | 23.50     | 85.20     | 91.59     | 85.90     | 94.45     | 89.50     | 92.58  |
| OC (%)                                    | 2.38      | 0.81      | 1.78      | 2.09      | 1.73      | 2.25      | 1.46      | 1.58   |

<sup>a</sup> [2].<sup>b</sup> OC = organic carbon.

Table 3. Concentrations ( $\mu\text{g}$  chemical/g organic carbon [OC]) of selected low-molecular-weight (LMW) and high-molecular-weight (HMW) polycyclic aromatic hydrocarbon (PAH) compounds in Lauritzen Channel sediments before and after remediation

| Compound                                    | Lauritzen Channel stations |                          |                        |                         |                       |
|---|----------------------------|--------------------------|------------------------|-------------------------|-----------------------|
|   | 1<br>After                 | 1<br>Before <sup>a</sup> | 2<br>After             | 3<br>After              | 4<br>After            |
| Acenaphthylene                              | 2.96                       | 8.20                     | 5.29                   | 11.38                   | 0.58                  |
| Acenaphthene                                | 71.60                      | 1.70                     | 166.67                 | 306.22                  | 25.82                 |
| Anthracene                                  | 71.11                      | 19.00                    | 126.96                 | 184.89                  | 28.73                 |
| Benzo(a)anthracene                          | 120.62                     | 25.00                    | 203.43                 | 276.89                  | 41.33                 |
| Benzo(a)pyrene                              | 125.93                     | 72.00                    | 177.94                 | 230.22                  | 40.06                 |
| Benzo(b)fluoranthrene                       | 180.25                     | 97.00                    | 270.10                 | 376.44                  | 55.06                 |
| Benzo(k)fluoranthrene                       | 61.85                      | 51.00                    | 92.65                  | 254.67                  | 19.43                 |
| Benzo(ghi)perylene                          | 53.46                      | NR                       | 66.18                  | 71.11                   | 21.90                 |
| Benzo(e)pyrene                              | 87.78                      | NR                       | 120.59                 | 157.33                  | 28.99                 |
| Biphenyl                                    | 11.72                      | NR                       | 26.96                  | 50.22                   | 4.04                  |
| Chrysene                                    | 132.10                     | NR                       | 196.08                 | 288.44                  | 50.70                 |
| Fluoranthrene                               | 337.04                     | 41.00                    | 612.75                 | 991.11                  | 139.24                |
| Fluorene                                    | 65.93                      | 4.00                     | 127.45                 | 209.33                  | 23.16                 |
| Naphthalene                                 | 39.51                      | 8.60                     | 58.33                  | 83.11                   | 8.23                  |
| Phenanthrene                                | 222.22                     | 16.00                    | 504.90                 | 840.00                  | 96.20                 |
| Perylene                                    | 29.88                      | NR                       | 42.55                  | 50.67                   | 11.39                 |
| Pyrene                                      | 250.62                     | 69.00                    | 416.67                 | 622.22                  | 108.86                |
| Total OC (%)                                | 0.81                       | 2.38                     | 2.04                   | 2.25                    | 1.58                  |
| LMW PAH ( $\mu\text{g}/\text{kg}$ dry wt)   | 4,664.90 <sup>b</sup>      | NR <sup>c</sup>          | 24,366.00 <sup>b</sup> | 44,734.00 <sup>b</sup>  | 3,367.60 <sup>b</sup> |
| LMW PAH ( $\mu\text{g}/\text{g}$ OC)        | 575.91                     |                          | 1,194.41               | 1,988.18                | 213.14                |
| HMW PAH ( $\mu\text{g}/\text{kg}$ dry wt)   | 11,828.00 <sup>b</sup>     | NR                       | 46,994.00 <sup>b</sup> | 77,356.00 <sup>b</sup>  | 8,613.00              |
| HMW PAH ( $\mu\text{g}/\text{g}$ OC)        | 1,460.25                   |                          | 2,303.63               | 3,438.04                | 545.13                |
| Total PAH ( $\mu\text{g}/\text{kg}$ dry wt) | 16,492.90                  | NR                       | 71,360.00 <sup>b</sup> | 122,090.00 <sup>b</sup> | 11,980.60             |
| Total PAH ( $\mu\text{g}/\text{g}$ OC)      | 2,036.16                   |                          | 3,498.04               | 5,426.22                | 758.27                |

<sup>a</sup> [2].<sup>b</sup> Exceeds the effects range median value.<sup>c</sup> NR = not reported.

ing and capping. Differences in response between these two toxicity test protocols likely results from variable sensitivity to contaminants present in postremediation sediments and variable routes of exposure. *Eohaustorius estuarius* likely is exposed to sediment contaminants via dermal uptake from pore water and consumption of particle-bound contaminants [15]. When exposed at the sediment-water interface, however, *M. galloprovincialis* embryos presumably are exposed to dissolved chemicals fluxed from the sediment into the overlying water [7]. To our knowledge, no published studies have compared the relative sensitivity of these protocols, though previous research using spiked water samples and contaminated field samples have indicated that these protocols have variable sensitivity to contaminants. Results of dose-response studies have demonstrated that embryo-larval development tests are particularly sensitive to metal toxicity but may be less sensitive

to some organic compounds [16,17]. Even so, *E. estuarius* is relatively sensitive to  $\Sigma\text{DDT}$  and dieldrin [2] and to PAH compounds [18,19]. Both protocols have shown variable sensitivity to contaminated sediments from San Francisco Bay [20,21].

Analysis of benthic community structure in postremediation sediments indicates that this site has not recovered to what would be considered reference conditions for Central San Francisco Bay sediments [21]. During postremediation sampling, the infaunal communities at these stations were dominated by polychaetes and oligochaetes. Benthic community structure in San Francisco Bay is notoriously difficult to characterize [21], but the postremediation sediment infaunal communities in Lauritzen Channel were classified as being transitional by the benthic ecologists who analyzed the samples. This classification indicates a site with a benthic assemblage somewhere

Table 4. Concentrations (ng/g dry wt) of select organic compounds in mussel tissues (*Mytilus californianus*)<sup>a</sup>

| Station | Before                  |                  |                    |          | After <sup>b</sup> |          |
|---------|-------------------------|------------------|--------------------|----------|--------------------|----------|
|         | Total PCBs <sup>c</sup> | Total chlordanes | $\Sigma\text{DDT}$ | Dieldrin | $\Sigma\text{DDT}$ | Dieldrin |
| 1       | 522.70                  | 161.00           | 14,310.00          | 572.00   | NA                 | NA       |
| 2       | 376.80                  | 179.90           | 15,427.00          | 569.00   | 3,502.00           | 279.00   |
| 3       | 266.90                  | 52.30            | 4,853.00           | 224.00   | NA                 | NA       |
| 4       | 151.50                  | 26.68            | 1,283.00           | 90.60    | 1,448.00           | 165.00   |

<sup>a</sup> Before remediation, mussels were deployed for 23 d, from July 17–August 9, 1996. After remediation, mussels were deployed for 125 d, from September 3, 1997, to January 6, 1998.<sup>b</sup> [29].<sup>c</sup> PCBs = polychlorinated biphenyls.

between degraded and undegraded conditions and is based on the presence of negative indicator species (e.g., worms) and the absence of positive indicator species (e.g., mollusca, crustacea; P. Slattery, personal communication). Polychaete species in these samples included those that have previously been categorized as being contaminant tolerant (e.g., *Eteone* sp., *Capitella* sp., *Dorvillea* sp.) [21]. Sampling for benthic community structure consisted of only one replicate sample at each station; therefore, these data may underestimate species abundance at the site. Regardless of this limitation, only one crustacean species each was found in samples from stations 2 and 4 (Family Cumacea; data not shown), and no amphipod species were found in samples from any station. Swartz et al. [2] also found that sediments from Lauritzen Channel had few crustacean species and suggested that the amphipod species present in greatest numbers among preremediation samples (*Grandidierella japonica*) was one that may be capable of adapting to polluted conditions.

Interpretation of benthic community data at this site is confounded by several factors unrelated to chemical contamination. Because this site was completely dredged one year before the benthic sampling, the lack of a well-defined benthic community structure may have resulted from disturbance of the site and lack of adequate recovery time. In addition to the dredging of Lauritzen Channel as part of the remediation project reported here, the adjacent Santa Fe Channel had recently been dredged by the City of Richmond as part of a navigational maintenance project conducted before the postremediation sampling (A. Lincoff, personal communication). This probably disrupted adjacent infaunal communities that could have served as a recruitment source for Lauritzen Channel. Sediments at this site also continue to be disturbed by prop scour from tug boats and other shipping vessels. The combined influence of chemical contamination and disruption of benthos because of dredging and boat activities cannot be separated. Therefore, benthic community information may be less valuable than other ecotoxicologic indicators for assessing change at this site.

This study was not designed to investigate causes of toxicity, but results of previous studies associating bulk-phase chemical concentrations and impacts on amphipod survival may be applied to these results. Results of previous investigations regarding the toxicity of DDT, PAH compounds, and dieldrin indicate that concentrations of these chemicals in postremediation Lauritzen Channel sediments are sufficient to account for the observed amphipod mortality, particularly when these chemicals are considered as mixtures. Swartz et al. [2] reported that the threshold for 10-d sediment toxicity to amphipods was approximately 300  $\mu\text{g } \Sigma\text{DDT/g OC}$ , and that the *E. estuarius* 10-d LC50 for  $\Sigma\text{DDT}$  in Lauritzen Channel sediment was 2,500  $\mu\text{g } \Sigma\text{DDT/g OC}$ . The postremediation concentration of OC-normalized  $\Sigma\text{DDT}$  at station 1 (2,637.26  $\mu\text{g } \Sigma\text{DDT/g OC}$ ; Table 2) exceeded the 10-d LC50 for *E. estuarius*, and concentrations of  $\Sigma\text{DDT/g OC}$  were well above the threshold effect concentration for this species at stations 2 and 3. In addition to DDT, dieldrin concentrations in postremediation sediments remained at elevated concentrations and may have been partially responsible for amphipod mortality. Dieldrin concentrations in channel sediments were well below the amphipod 10-d LC50 (~1,955  $\mu\text{g dieldrin/g OC}$ ) [2] and probably contributed only incremental toxicity to *E. estuarius* (Table 2).

Concentrations of PAH compounds were considerably

higher in postremediation sediments than those reported by Swartz et al. [2] (Table 3) for preremediation sediments. In addition, concentrations of low-molecular-weight, high-molecular-weight, and total PAH compounds were well above the ERM sediment quality guidelines values reported by Long et al. [14]. The ERMs are the bulk-phase PAH concentrations above which toxicity to amphipods is considered to be probable. When applied to the  $\Sigma\text{PAH}$  model described by Swartz et al. [18], the summed toxic units ( $\Sigma\text{TU}$ ) of the 13 PAH compounds for which this model applies were not sufficient to explain the observed toxicity in postremediation sediments ( $\Sigma =$  acenaphthylene, acenaphthene, anthracene, benzo[ $\alpha$ ]anthracene, benzo[ $\alpha$ ]pyrene, benzo[ $\beta$ ]fluoranthrene, benzo[ $\kappa$ ]fluoranthrene, chrysene, fluoranthrene, fluorene, naphthylene, phenanthrene, pyrene). The  $\Sigma\text{TUs}$  for the 13 PAH compounds in sediments collected from all four stations were always less than 0.20 (data not shown).

Swartz [19] recently proposed consensus sediment-quality guidelines for PAH mixtures using these 13 PAH compounds. These guidelines were based on the mean of the existing sediment-quality guidelines previously published for total PAH mixtures. The OC-normalized consensus guidelines proposed by Swartz [19] are separated into a threshold effect concentration (290  $\mu\text{g } \Sigma\text{PAH/g OC}$ ) below which no toxicity from PAHs is to be expected, a median effect concentration (1,800  $\mu\text{g } \Sigma\text{PAH/g OC}$ ) above which amphipod toxicity may occur, and an extreme effects concentration (10,000  $\mu\text{g } \Sigma\text{PAH/g OC}$ ) above which effects are expected. The sum of the 13 OC-normalized PAH compounds used by Swartz [19] were 1,853.3, 3,241.8, 5,096.9, and 692  $\mu\text{g } \Sigma\text{PAH/g OC}$  in sediments at stations 1 through 4, respectively. Total PAH concentrations at stations 1 to 3 exceeded the median effect concentration, and the  $\Sigma\text{PAH}$  exceeded the threshold effect concentration at station 4. The median effect concentration is considered to be a median effect value at which an approximately 50% probability of toxicity exists in samples where PAH contamination is the dominant ecotoxicologic factor [19]. We did not consider metals as a source of toxicity in this study, because metals measured in previous studies were not present at sufficient concentrations to be considered toxic [2].

Concentrations of selected pesticides as measured in the tissues of bagged mussels deployed before and after the site remediation showed a decline in  $\Sigma\text{DDT}$  and dieldrin at station 2 and an increase in these compounds at station 4. Comparisons can only be made for these two stations, however, because postremediation monitoring at the site did not include all four stations (Table 4). The increase in  $\Sigma\text{DDT}$  and dieldrin at station 4 may result from the exposure time for the postremediation mussel bioaccumulation monitoring (123 d) being almost four-fold as long as for those deployed before the site remediation (23 d). Preremediation mussels were removed early because of the initiation of channel dredging. In addition, postremediation mussels apparently were deployed on the east side of the channel and somewhat farther north of station 4 than the preremediation mussels. Because the concentrations of  $\Sigma\text{DDT}$  and dieldrin increased northward up the channel, this may have influenced bioaccumulation in the postremediation mussels from station 4.

These data indicate that bioaccumulatable concentrations of the two primary chemicals of concern declined at the most contaminated station (station 1). Bioaccumulation of  $\Sigma\text{DDT}$  and dieldrin was one of the principal ecologic concerns identified in the ecologic risk assessment conducted at this site by

Lee et al. [1]. Bioaccumulation using field-deployed mussels was included as the sole biologic component in U.S. EPA postremediation monitoring in Lauritzen Channel, because these data could be compared with a relatively large prerediation database extending over several years [3]. These data were intended to complement water column chemical measurements included in postremediation monitoring. Sediment toxicity testing data in this study augment such water column bioaccumulation data by providing information regarding the toxicologic effects of sediment contaminants. The additional ecotoxicologic measurements were included in the present study because, in addition to human health and ecologic concerns regarding elevated tissue concentrations of DDT and dieldrin to water column organisms, the California Department of Fish and Game was interested in determining whether the remediation practices removed chemicals that threatened sediment biota. Risk to sediment biota could only be assessed with appropriate toxicity tests and sediment chemical measurements.

Why postremediation sediments in Lauritzen Channel were so heavily contaminated is unclear. As discussed earlier, approximately 100,000 metric tons of contaminated sediments were dredged from this site. Extensive premeditation chemical characterization of the site showed that most chemical contamination was associated with the younger bay mud, and it was assumed that most of the chemical contamination would be removed after dredging to a depth of approximately 15 cm below the older bay mud horizon [3]. Possible sources of postremediation contamination may be separated into two categories: residual contamination, or new contamination. Residual contamination results from incomplete removal and might have occurred where dredging or capping was not effective, particularly at locations in the channel where the clamshell dredge could not operate. New contamination could occur via terrestrial surface runoff, storm water pipes or other discharge structures, the dredge material dewatering process, recontamination from the adjacent marine environment, and recontamination from polluted groundwater. All these sources were considered in the remediation feasibility study [3] and addressed (to varying degrees) in the final remediation workplan [22]. The source of PAHs in postremediation sediments is unknown, but industrial activities in the channel area include shipping operations and a variety of land-based businesses, including manufacturing, recycling, and construction. All are possible sources of PAHs. White et al. [23] reported relatively high concentrations of both low- and high-molecular-weight PAHs in composite (sample depth, 30.5 cm) samples collected from Lauritzen Channel sediments, particularly those collected near station 3 in the present study.

Lauritzen Channel was capped with 15 to 46 cm of clean sand after dredging was completed in 1997, but postremediation grain-size distributions indicate that stations 2, 3, and 4 were dominated by fine-grained sediments (Table 2). Because chemicals are associated with finer-grained sediments, this may partially account for the continued contamination in the channel. The sand layer was relatively shallow, because it was intended to provide habitat for benthic fish species rather than to serve as a cap to prevent mobilization of chemicals in deeper layers. Finer sediments have settled into the channel, but the source of this sediment is unknown. Possibilities include resuspended material from the dredging in Santa Fe Channel, slumping of sediments from the undredged margins of Laur-

itzen Channel, and material deposited from the dredging vessels that are stored in the channel.

Approximately 60% of the  $\Sigma$ DDT in postremediation sediments was 4',4-DDT; the remainder was dominated by 4',4-DDD (22%). These ratios differed considerably from those reported by Lee et al. [1] for prerediation sediments from Lauritzen Channel. Those authors found a greater proportion of 4',4-DDD (50%) relative to 4',4-DDT (37%). Upland samples adjacent to the marine habitat had 80% 4',4-DDT, 13% 2',4-DDT, and 6% 4',4-DDD [1], which is similar to technical formulations of DDT [24]. The relative proportions of the different DDT metabolites in postremediation sediments therefore fall between those of prerediation sediments and those of the upland samples as reported by Lee et al. [1]. This suggests that postremediation contamination may have come either from an upland source or from one where the timing or conditions under which metabolic alteration of DDT differed from those of the prerediation sediments. Identifying the sources of postremediation contamination was beyond the scope of this study but should be addressed at this site before similar remediation plans are implemented. Chlordane and PAH concentrations were considerably higher in postremediation sediments relative to those concentrations as described by Lee et al. [1], but identification of the source (or sources) of these compounds in postremediation sediments was, again, beyond the scope of this study. Such identification should also be included in any future investigations at this site to minimize inputs and to prevent further habitat contamination.

To our knowledge, relatively few studies regarding the effects from remediation of organochlorine-contaminated sediments on ecotoxicologic endpoints have been reported. Bergen et al. [25] described the distribution of PCBs after New Bedford Harbor (NBH) sediments were dredged to remove the most contaminated hotspot. Approximately 7,600 m<sup>3</sup> of PCB-contaminated sediments from NBH were removed via suction dredging and disposed off-site. Congener measurements of PCB distributions were combined with statistical and graphic analyses to show that harborwide concentrations of total PCB congeners decreased in NBH sediments by more than four orders of magnitude in surficial sediments. Postremediation monitoring of NBH included toxicity tests with the amphipod *Ampelisca abdita*, characterization of benthic community structure, and bioaccumulation in mussel tissue. Postremediation monitoring conducted two years after the PCB hotspot in upper NBH was removed indicated that whereas no apparent change in benthic community structure had occurred, sediment toxicity had increased in the upper and lower harbors. Apparently, this resulted from resuspension of PCB-contaminated sediments during the dredging process ([26]; W. Nelson, personal communication). Bremle and Larson [27] measured concentrations of PCBs in water and fish tissue both upstream and downstream from Lake Järnsjön in southern Sweden after contaminated lake sediments were removed via suction dredging; concentrations of PCBs decreased by greater than 70% and 50%, respectively, in water and fish after removal of lake sediments. Removal of PCB-contaminated sediments was less successful, however, after dredging of the Shiawassee River, Michigan, USA. Rice and White [28] used caged fish and clams to show that postremediation concentrations of PCBs were not significantly less in river waters, because a considerable amount of PCBs remained in river sediments 6 months after remediation.

## CONCLUSIONS

The United Heckathorn Superfund site in Richmond, California, is one of a few marine habitats to date where sediment contamination by chlorinated pesticides has been considered to pose sufficient ecologic and human health risk to warrant remediation through dredging and off-site disposal. This study demonstrates the utility of including multiple ecotoxicologic measures in a weight-of-evidence approach for assessing the effectiveness of dredging as a remediation alternative. All measures employed in this study were useful for assessing change, but interpretation of the benthic community data was confounded because of sediment disturbance from shipping and dredging activities both at this site and in the adjacent area. Because the remediation activities were designed largely to minimize exposure of DDT and dieldrin to higher-trophic-level organisms, postremediation monitoring at this site emphasized water column concentrations of these chemicals and their bioaccumulation in mussel tissues because of ecologic and human health concerns [13]. These measures demonstrated that dredging reduced concentrations of DDT and dieldrin in Lauritzen Channel water and in the surrounding system [29]. Analysis of bulk-phase chemistry and toxicity indicated continued sediment contamination and toxicity at this site and demonstrated the applicability of these measurements for assessing ecotoxicologic change in this compartment of the system. These tools are relatively simple and provide no mechanistic information [30], but the combination of toxicity tests, analytic chemistry, and bioaccumulation measurements was sufficient to assess the effectiveness of remediation activities at this site. Concentrations of chlorinated pesticides have declined in mussels; however, insufficient data exist to determine whether the residual sediment contamination in Lauritzen Channel is affecting the larger system. Further analyses of these chemicals in the tissues of local fish populations, particularly in species that prey extensively on benthic fauna, would help to answer this question. The degree of contamination and toxicity at this site after extensive remediation of contaminated sediments is problematic, and it suggests that future remediation projects that rely on similar methodologies should incorporate greater consideration of possible sources for postremediation contamination to better achieve the project goals.

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