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DISTRIBUTION OF CHLORINATED HYDROCARBONS IN OVERLYING WATER, SEDIMENT, POLYCHAETE, AND HORNYHEAD TURBOT (*PLEURONICHTHYS VERTICALIS*) IN THE COASTAL OCEAN, SOUTHERN CALIFORNIA, USA

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Abstract—1,1,1-Trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT) and its primary metabolites (DDTs) and polychlorinated biphenyls (PCBs) are a major source of concern in the Southern California Bight (SCB), USA. The fate of DDTs and PCBs is a key element in assessing the effects imposed by these potential carcinogens on the marine ecosystem. We found that DDTs and PCBs remained widely distributed in the overlying water, sediment, polychaetes, and liver and muscle tissues of the hornyhead turbot (*Pleuronichthys verticalis*) collected from three nearshore locations of the SCB with different levels of contamination. Student's *t* tests indicated that the measured partition coefficients between the nonaqueous phases (sediment, polychaete, and fish) and overlying water at a heavily contaminated location were significantly greater than those predicted by the equilibrium partitioning theory (EPT). Measured partition coefficients between the nonaqueous phases and overlying water for a few DDT components at two other stations (moderate and low contamination) were also generally greater than the EPT predictions. On the other hand, DDTs and PCBs in polychaetes and fish tissues may be taken up from sediments via equilibrium partitioning or from food sources. These findings are suggestive of the possibility that contaminated sediments may have become an important source of contamination.

Keywords—1,1,1-Trichloro-2,2-bis(*p*-chlorophenyl)ethane Polychlorinated biphenyl Polychaete *Pleuronichthys verticalis* Palos Verdes Shelf

INTRODUCTION

Very large amounts of 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT) and its primary metabolites (DDTs) were discharged to the coastal waters of the Southern California Bight (SCB), USA, before the early 1970s, mainly via the submarine sewage outfall of the Joint Water Pollution Control Plant (JWPCP) operated by the County Sanitation Districts of Los Angeles County, California, USA. The main origin of these contaminants was industrial production–related DDT residues generated by the Montrose Chemical Corporation [1]. Approximately 20 metric tons of DDTs were discharged via the JWPCP outfall to the Palos Verdes Shelf (PVS; California, USA) in 1971 [2]. As a consequence, a sediment zone of more than 20 km^2 with highly elevated levels of DDTs has been present on the PVS [3]. Lower amounts of polychlorinated biphenyls (PCBs) also were discharged along with the DDT-containing wastes. Although inputs of DDTs and PCBs from the JWPCP outfall have reached virtually nondetectable levels [4], contaminated sediments on the PVS remain a potential source of contamination to the SCB.

The fate of historically deposited DDTs and PCBs is a key element in understanding the effects of these potential carcinogens on the marine ecosystem. Although virtually no DDTs and PCBs currently are discharged from the sewage outfall, DDTs and PCBs historically buried in sediments may continue to be amended via at least three mechanisms: biodegradation, remobilization and dispersion from the originally deposited area to other areas, and uptake by marine organisms. The first mechanism for modification of DDTs largely involves transformation of 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethene (DDE) to other metabolites, because DDTs found in coastal sediments of the SCB are constituted largely of DDEs (both *p*,*p*′- and *o*,*p*′-DDE) [5]. Transformation of DDE to 1-chloro-2,2-bis(*p*-chlorophenyl)ethene recently was demonstrated in a marine microsm study that used PVS sediments [6]; this transformation was recently investigated further [7]. On the other hand, biodegradation of PCBs in aquatic sediments has been well documented [8–11]. The second mechanism is supported by the widespread distributions of DDTs and PCBs in the water column of the PVS and in the nearshore and basin sediments of the SCB [5,12]. The third mechanism has been investigated extensively, yielding tremendous amounts of data; how this mechanism works may depend upon many internal and external factors.

The goal of this study was to investigate the fate of historically discharged DDTs and PCBs in the coastal environment of the SCB. The objectives were to evaluate how historically discharged DDTs and PCBs were distributed in the environmental compartments of different trophic levels, and to investigate the tendency of DDTs and PCBs to move among the food chain of hornyhead turbot (*Pleuronichthys verticalis*). To accomplish these objectives, we collected and analyzed samples of overlying water, sediment, polychaete, and hornyhead turbot from three nearshore locations with different levels of historical DDT and PCB contamination off Palos Verdes and Newport Beach (CA, USA) (Fig. 1). Bioaccumulation was assessed with the equilibrium partitioning theory (EPT) [13]. This theory deals with the concentration ratio of a specific analyte between two interacting phases. Any devi-

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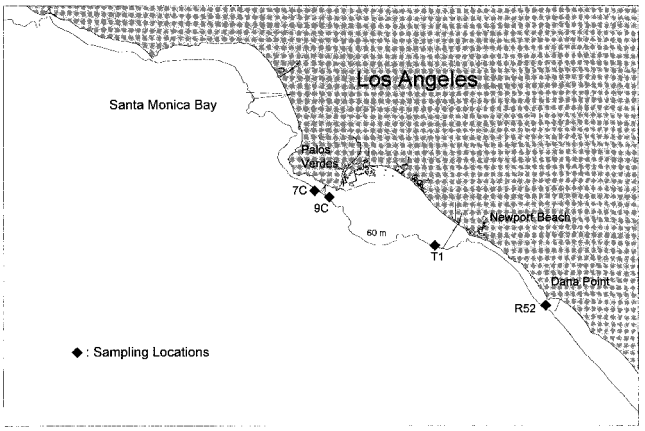

Fig. 1. A map showing the sampling stations off Palos Verdes, Newport Beach, and Dana Point (CA, USA).

ation from the EPT predictions may indicate a thermodynamic tendency for the analyte to move from one phase to another. A more detailed description of how EPT can be used in this study is given in the next section. Recently, the applicability of EPT as a screening tool for estimating bioaccumulation was verified with a large set of data covering a variety of sample types and analytes [14,15].

The hornyhead turbot was selected as a prototype in the present study for the following reasons. First, the hornyhead turbot lies partially buried in sediments and feeds primarily on bivalves and sedentary tube-dwelling polychaetes such as *Diopatra ornata*, *Pista alata*, and *Paraprionospio pinnata*. Such a living and feeding habit favors a nearly equilibrated transport of contaminants between the prey and predator. Second, the hornyhead turbot occurs in areas around the 60-m depth contour, where major wastewater outfalls are situated in the SCB [16]. This ensured that sufficient individuals of the fish could be collected. Third, lipid contents in hornyhead turbot are generally less variable than in other species because of the active productivity of hornyhead turbot all year around [17], which allows more consistent comparison of contaminant concentrations among hornyhead turbot individuals. These reasons also are why a number of national and local environmental agencies have used hornyhead turbot (designated as fish thereafter) to monitor temporal trends for bioaccumulation of trace metals and chlorinated hydrocarbons in southern California, USA [1,18–20].

MATERIALS AND METHODS

Sample collection and extraction

Samples of overlying water, surface sediment, polychaete, and fish were collected from stations 7C and 9C near the JWPCP outfall (heavily contaminated), station T1 near the Orange County Sanitation District outfall (moderately contaminated), and station R52 off Dana Point (lightly contaminated) (Fig. 1). Sampling was conducted in two seasons: October–November 1995 (designated as 1995 thereafter) and April–May 1996 (designated as 1996 thereafter). The sampling information is summarized in Table 1.

Water samples were collected at approximately 1 m from the sea floor with an Infiltrex 100 water sampler (Axys Environmental Systems, Sidney, BC, Canada). Suspended particles were filtered with glass fiber filters (142 mm GF/F filters [Whatman International, Maidstone, UK] with 0.7-μm pore size), whereas dissolved organics were retained on XAD-II (Alltech Associates, Deerfield, IL, USA) resins packed inside a Teflon® column. Detailed operational procedures were described previously [12]. Sediment samples were collected, after water samples were taken, with a modified 0.1-m^2 chain-rigged Van Veen grab [21]. The top 2 cm of sediments were collected and transferred into precleaned glass jars. Polychaetes, including *Diopatra ornata*, *Pista alata*, and *Paraprionospio pinnata*, were sampled by passing sediments through a 1.0-mm screen. The polychaetes were washed thoroughly with seawater to remove attached sediment before being transferred to precleaned glass jars. Sediment and polychaete sample jars were kept frozen at −20°C until analysis.

Table 1. Sampling locations and sample types[a]

Station	October–November 1995	April–May 1996
7C	w, s, p, h	w, s, p, h
9C	s, p, h	s, p, h
T1	w, s, p, h	w, s, p, h
R52	s, h	w, s, p, h

[a] w = overlying water; s = surface sediment; p = polychaete; h = hornyhead turbot (including liver and muscle tissues).

Fish were collected opportunistically by trawling at the 60-m depth with 7.6-m headrope semiballoon otter trawls with 1.25-cm cod end mesh. Trawls were towed for 10 min (bottom time) at 0.8 to 1.2 m/s (1.5–2.4 knots). Once on board, the fish were wrapped in aluminum foil and placed in plastic bags. All samples were chilled in an ice chest during transportation to the laboratory and were dissected within 24 h. A total of six fish individuals from each station were dissected for chemical analyses. If possible, sexually mature fish were selected and the numbers of males and females were equal. Muscle and liver tissues from each individual fish were separately stored in glass vials and kept frozen at −20°C until analysis.

Teflon columns retaining dissolved organics were processed by using a procedure described elsewhere [5]. Samples of sediment, polychaete, and fish tissue were extracted three times and extracts were combined, cleaned up, and fractionated by using a previously described protocol [22]. Lipid content in each polychaete and fish extract was measured three times [22] and an average value was used.

Instrumental analysis

Total organic carbon was measured by a protocol described elsewhere [23]. Three replicates were analyzed for each sediment sample. Measurements of PCBs and DDTs were conducted by using the procedures given previously [5]. A total of 18 chlorinated pesticides and 122 PCB congeners were included in the analyses. Individual congeners were detected and quantified by a congener-specific method including gas chromatography–mass spectrometry confirmation of peak identification [22].

Data analysis

Although our list of analytes included 18 chlorinated pesticides (aldrin, γ-benzene hexachloride, α-chlordane, dieldrin, endosulfan I, endosulfan II, endrin, heptachlor, heptachlor epoxide, hexachlorobenzene, mirex, *trans*-nonachlor, *o,p′*-DDT, *p,p′*-DDT, 1,1-dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethane [*o,p′*-DDD], *p,p′*-DDD, *o,p′*-DDE, and *p,p′*-DDE) and

122 PCB congeners, smaller numbers of the target analytes were detected in the samples. Therefore, partitioning coefficients are reported only for detectable target analytes. An average of two replicate measurements was obtained for each value of overlying water concentration (ng/L) and polychaete concentration (ng/g wet wt). Sediment concentration (ng/g dry wt) was presented as an average of three measurements. For fish, the number of detectable PCB congeners varied substantially among individuals from the same sampling location. Therefore, liver and muscle tissue concentrations (ng/g wet wt) in one of the six samples from each sampling location with the largest number of detectable PCBs were used. The octanol–water partition coefficient (K_{OW}) values of DDTs and PCB congeners were taken from Gossett et al. [24] and Hawker and Connell [25], respectively.

Total organic carbon– and lipid-based concentrations were used to assess the partitioning characteristics of DDT and PCB components. As stated by EPT, the fugacity (F) of a hydrophobic chemical is identical in all interacting phases under equilibrium conditions [26], that is

$$F_w = F_s = F_p = F_l = F_m \quad (1)$$

where the subscripts w, s, p, l, and m refer to overlying water (truly dissolved phase), sediment, polychaete, fish liver, and fish muscle, respectively. This relationship translates to the following equations for partitioning coefficients in the case of this study:

$$K_{iw} = C_i/C_{w,e} = K_{OW} \quad (2)$$

$$K_{ij} = C_i/C_j = 1 \quad (3)$$

where K_{iw} denotes the coefficient of partitioning between the overlying water and phases i; K_{ij} is the coefficient of partitioning between phases i and j; C_i or C_j is the total organic carbon– or lipid-normalized concentration of a specific component in phase i or phase j; $C_{w,e}$ is the dissolved phase concentration of a specific component at equilibrium; and i or j = s, p, l, and m. A Student's t test was applied to the examination of the correlations between measured $\log(C_i/C_w)$ or $\log(C_i/C_j)$ and $\log K_{OW}$ for all detectable DDT and PCB compounds. A 95% confidence interval was calculated for each slope or intercept examined, and p values of <0.01, between 0.01 and 0.05, and >0.05 were considered to be indicative of the differences being highly significant, significant, and not significant, respectively.

RESULTS

Contaminant distribution in environmental compartments

Among the 18 chlorinated pesticides quantified, only *o,p′*- and *p,p′*-DDE, DDD, and DDT were detected and the subsequent data analysis focused on these DDT components. The number of detectable PCB congeners varied with location and sample type and ranged from zero to 50. In general, samples collected from station 7C had the greatest number of detectable PCB congeners. On the other hand, samples collected from station R52 contained the smallest number of detectable PCB congeners. In terms of sample type, fish liver tissues contained the largest number of detectable PCB congeners among the samples from stations T1 and R52, whereas samples from station 7C (heavily contaminated) all had fairly large numbers of detectable PCB congeners. For example, among the 7C samples collected in 1996, the number of detectable PCB congeners was in the following order: sediment (50), overlying water filtrates (49), fish liver (43), polychaete (35), overlying water particles (28), and fish muscle (21). For T1 and R52 samples collected at the same time, the order was fish liver (20), polychaete (4), fish muscle (2), and overlying water filtrates and particles (zero) in the T1 samples and fish liver (8), fish muscle (5), and polychaete and overlying water filtrate and particle (zero) in the R52 samples. Because no PCB congeners were detected in the overlying water filtrates and particles from stations T1 and R52 in both the sampling seasons and from station 7C in the 1995 sampling, the assessment of contaminant partitioning between the overlying water and other compartments was mainly conducted with the 7C samples collected in 1996.

The measured concentrations of individual DDT components and total PCB exhibited certain patterns (Tables 2 and 3). First, DDT or PCB concentrations, when normalized to either lipid or total organic carbon, were highest in the fish liver samples and lowest in surface sediments (overlying water particle concentrations were excluded for comparison because total organic carbon was not measured in these samples). Conversely, fish muscle and polychaete samples contained similar amounts of total DDT and PCB. Within a specific compartment, DDT and PCB concentrations were higher at stations 7C and 9C than at stations T1 and R52. Second, *p,p′*-DDE was the most abundant compound among the six DDT compounds and generally constituted more than 60% of the total DDT concentration in a given sample. Third, the ratio of DDT to PCB concentrations was fair high (mostly ~10) with the exception of the sediment and fish muscle samples collected from station T1, which had DDT to PCB concentration ratios of about 1 to 2. Fourth, total DDT and PCB concentrations varied substantially with individual fish liver and muscle samples, as indicated by the large standard deviations from six replicates for each sample analysis. Finally, no significant seasonal difference was found in the DDT and PCB concentrations for comparable samples.

Statistical analysis of partitioning coefficients

Figure 2 shows the correlations between $\log(C_i/C_w)$ (i = s, p, l, and m) and $\log K_{OW}$ for detectable DDT and PCB components in samples collected from station 7C. Linear regression analysis was conducted on the samples collected in 1996 only, because insufficient DDT and PCB components were detected in the samples collected in 1995 (only *o,p′*- and *p,p′*-DDD and *o,p′*- and *p,p′*-DDE were detected in the overlying water filtrate samples). The dotted lines represent the $\log K_{OW}$–$\log K_{OW}$ correlation at which equilibrium partitioning is supposedly established as shown in Equation 2. Although no significant statistical analysis was possible for the 1995 samples, all the calculable concentration ratios are above the $\log K_{OW}$–$\log K_{OW}$ correlation line. In all the 1996 samples, the differences between the linear regressions and the $\log K_{OW}$–$\log K_{OW}$ correlation line are highly significant or significant (Table 4). Among the four correlations, $\log(C_p/C_w)$–$\log K_{OW}$ and $\log(C_m/C_w)$–$\log K_{OW}$ have higher intercept values (with $p < 0.001$) but smaller slopes than the other two correlations. On the other hand, the $\log(C_s/C_w)$–$\log K_{OW}$ and $\log(C_l/C_w)$–$\log K_{OW}$ correlations have slopes closest to unity (0.78 and 0.70, respectively). All but $\log(C_p/C_w)$ correlate moderately (r^2 = 0.34, 0.39, and 0.41) with $\log K_{OW}$.

Very few target analytes were detected in the overlying water filtrate samples collected from stations T1 and R52 in 1996 (Table 3), which did not warrant a significant statistical

Table 2. Measured concentrations of chlorinated hydrocarbons in various sample matrices acquired in October–November 1995[a]

Component	Filtrate avg. (n = 2) (ng/L)	Particle avg. (n = 2) (ng/g)	Sediment avg. (n = 3) (ng/g)	Polychaete avg. (n = 2) (ng/g wet wt)	Liver avg. (n = 6) (ng/g wet wt)	Muscle avg. (n = 6) (ng/g wet wt)
Station 7C						
o,p′-DDE	0.52 (0.02)[b]	0.21 (0.15)	984 (52)	886 (44)	18,526 (12,720)	185 (110)
p,p′-DDE	2.46 (0.73)	1.72 (1.21)	6,652 (166)	13,517 (864)	110,442 (89,621)	1,710 (1,164)
o,p′-DDD	0.17 (0.04)	0.05 (0.03)	259 (9.7)	135 (9)	2,005 (1,219)	14 (3)
p,p′-DDD	0.62 (0.20)	0.13 (0.08)	577 (68)	295 (31)	24,762 (40,797)	58 (50)
o,p′-DDT	<0.02	<0.02	<2.1	<2.3	31 (22)	<1.9
p,p′-DDT	0.03 (0.00)	0.06 (0.02)	473 (234)	<2.3	195 (107)	<1.9
ΣDDT	3.92 (1.03)	2.20 (1.48)	8,961 (531)	14,833 (948)	155,976 (144,490)	1,967 (1,328)
ΣPCB	<0.02	<0.02	935 (62)	1,043 (119)	15,059 (8,167)	253 (98)
Station 9C						
o,p′-DDE	NA	NA	165 (9.1)	261 (280)	5,807 (6,146)	120 (121)
p,p′-DDE	NA	NA	1,392 (451)	3,346 (723)	53,711 (56,652)	1,280 (955)
o,p′-DDD	NA	NA	48 (2.6)	60 (12)	1,023 (1,106)	13.3 (11.6)
p,p′-DDD	NA	NA	102 (6.7)	134 (23)	3,895 (3,632)	25.8 (20.3)
o,p′-DDT	NA	NA	<1.7	<1.7	13.9 (5)	<1.7
p,p′-DDT	NA	NA	74 (2.3)	<1.7	95 (84)	<1.7
ΣDDT	NA	NA	1,781 (472)	3,801 (1,039)	64,545 (67,626)	1,439 (1,108)
ΣPCB	NA	NA	<1.9	236 (11.5)	11,342 (7,034)	176 (60)
Station T1						
o,p′-DDE	<0.006	<0.006	<0.36	2.8 (1.0)	31.2 (18.3)	1.4 (n = 1)
p,p′-DDE	0.045 (n = 1)	0.009 (n = 1)	2.21 (0.38)	23.3 (11.0)	839 (621)	11.5 (6.9)
o,p′-DDD	<0.006	<0.006	<0.36	<1.1	63 (66)	<0.77
p,p′-DDD	<0.006	<0.006	<0.36	<1.1	22.1 (18.1)	<0.77
o,p′-DDT	<0.006	<0.006	<0.36	<1.1	<2.6	<0.77
p,p′-DDT	<0.006	<0.006	<0.36	<1.1	<2.6	<0.77
ΣDDT	0.045	0.009	2.21 (0.38)	26.1 (12.0)	956 (723)	12.9 (6.9)
ΣPCB	<0.006	<0.006	<0.33	21.7 (4.9)	413 (170)	5.25 (3.68)
Station DP						
o,p′-DDE	NA	NA	<0.41	NA	4.7 (0.5)	<0.86
p,p′-DDE	NA	NA	5.84 (0.28)	NA	130 (81)	5.9 (3.5)
o,p′-DDD	NA	NA	<0.41	NA	8.5 (n = 1)	<0.86
p,p′-DDD	NA	NA	<0.41	NA	<2.3	<0.86
o,p′-DDT	NA	NA	<0.41	NA	<2.3	<0.86
p,p′-DDT	NA	NA	<0.41	NA	<2.3	<0.86
ΣDDT	NA	NA	5.84 (0.28)	NA	143 (82)	5.9 (3.5)
ΣPCB	NA	NA	<0.40	NA	18.9 (n = 3) (15.6)	1.31 (n = 2) (0.24)

[a] DDE = 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethene; DDD = 1,1-dichloro-2,2-bis(*p*-clorophenyl)ethane; DDT = 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane; PCB = polychlorinated biphenyl; NA = not applicable; DP = Dana Point (CA, USA).
[b] The numbers in parentheses are standard deviations for replicate measurements.

analysis. However, all but one concentration ratio were near or above the log K_{OW}–log K_{OW} correlation line. The overlying water samples collected in 1995 contained no detectable DDT or PCB components (Table 2).

Regressions of log(C_i/C_j)–log K_{OW} were analyzed in all of the 7C samples (Fig. 3). The dotted lines represent the equilibrium state reached between the two interacting phases, as indicated in Equation 3. Results of the Student's t tests are summarized in Table 5. Among all the samples collected in 1995 and 1996, only the log(C_l/C_p)–log K_{OW} regression was significantly different from the equilibrium state in terms of both the slope and intercept ($p < 0.01$). The measured partitioning coefficients significantly increased in value with increasing K_{OW} (with the largest slopes among all the regressions). All the other correlations were not significantly different from the equilibrium state.

Partitioning coefficients for PCB and DDT compounds measured in the 9C samples are depicted in Figure 4 and the results of the Student's t tests are summarized in Table 5. Because very few DDT and PCB components were detected in the sediment samples, no statistical analysis was performed on partitioning coefficients involving sediment samples. Almost all the correlations were not significantly different from the equilibrium state. Only log(C_l/C_m)–log K_{OW} in the 1996 samples was moderately significantly different from the equilibrium state in terms of the intercept.

For the samples collected from station T1, only the log(C_l/C_p) data from the 1995 samples could be analyzed statistically (Table 5). The linear regression was not significantly different from the equilibrium partitioning state. All the other log-based partitioning coefficients (only DDD and DDE detected) ranged from −0.3 to 1.6. In the R52 samples collected in 1995, log(C_l/C_s), log(C_m/C_s), and log(C_l/C_m) for *p,p′*-DDE could be determined and were greater than zero. The R52 samples collected in 1996 had all partitioning coefficients available for *p,p′*-DDE and they were all greater than zero as well. Log(C_l/C_s) and log(C_l/C_m) for *p,p′*-DDD and *o,p′*-DDE were also obtained from the R52 samples; they were all greater than zero.

DISCUSSION

Widespread distributions of DDTs and PCBs and the implications

Contamination by chlorinated hydrocarbons, particularly DDTs, has been the focal point of the research activities in

Table 3. Measured concentrations of chlorinated hydrocarbons in various sample matrices acquired in April–May 1996[a]

Component	Filtrate avg. (n = 2) (ng/L)	Particle avg. (n = 2) (ng/g)	Sediment avg. (n = 3) (ng/g)	Polychaete avg. (n = 2) (ng/g wet wt)	Liver avg. (n = 6) (ng/g wet wt)	Muscle avg. (n = 6) (ng/g wet wt)
Station 7C						
o,p′-DDE	0.89 (0.29)[b]	0.65 (0.08)	<0.55	1,428 (94)	14,988 (13,312)	82 (57)
p,p′-DDE	4.46 (2.37)	3.57 (0.16)	7,889 (1,019)	14,798 (837)	184,668 (200,490)	930 (879)
o,p′-DDD	0.31 (0.16)	0.17 (0.01)	216 (9.2)	191 (11)	1,069 (754)	35 (46)
p,p′-DDD	0.74 (0.39)	0.30 (0.01)	443 (83.9)	321 (18)	2,303 (2,209)	29 (14)
o,p′-DDT	<0.01	<0.01	<0.55	<2.1	<3.7	<1.1
p,p′-DDT	0.28 (0.11)	0.08 (0.01)	248 (142)	<2.1	788 (694)	<1.1
ΣDDT	6.80 (3.33)	4.78 (0.28)	9,923 (1,389)	16,738 (960)	203,845 (217,483)	1,077 (996)
ΣPCB	4.18 (1.32)	0.737 (0.039)	2,515 (207)	1,201 (66)	3,457 (2,091)	126 (71)
Station 9C						
o,p′-DDE	NA	NA	187 (15)	330 (2)	9,725 (7,245)	203 (346)
p,p′-DDE	NA	NA	1,246 (126)	5,298 (54)	104,145 (112,571)	2,506 (4,616)
o,p′-DDD	NA	NA	50 (2.2)	76 (6)	813 (651)	27 (43)
p,p′-DDD	NA	NA	237 (148)	134 (15)	2,129 (1,037)	54 (93)
o,p′-DDT	NA	NA	<0.41	<1.8	92 (n = 1)	<1.3
p,p′-DDT	NA	NA	364 (520)	<1.8	545 (470)	<1.3
ΣDDT	NA	NA	2,084 (811)	5,838 (78)	117,457 (121,977)	2,792 (5,098)
ΣPCB	NA	NA	4.69 (2.15)	476.0 (7.1)	2,779 (1,729)	204 (315)
Station T1						
o,p′-DDE	0.020 (0.009)	<0.006	<0.33	6.1 (0.25)	146 (244)	<1.6
p,p′-DDE	0.146 (0.084)	0.014 (0.003)	1.5 (0.26)	30.6 (5.95)	3,835 (7,732)	8.0 (3.2)
o,p′-DDD	0.009 (0.001)	<0.006	<0.33	<1.8	57 (9)	<1.6
p,p′-DDD	0.008 (0.003)	<0.006	0.39 (n = 1)	<1.8	75 (75)	<1.6
o,p′-DDT	<0.006	<0.006	<0.33	<1.8	<6.4	<1.6
p,p′-DDT	<0.006	<0.006	<0.33	<1.8	<6.4	<1.6
ΣDDT	0.183 (0.097)	0.014 (0.003)	1.89 (0.26)	36.6 (6.2)	4,113 (8,060)	8.0 (3.2)
ΣPCB	<0.006	<0.006	<0.33	21.4 (5.3)	245 (152)	8.9 (10.6)
Station DP						
o,p′-DDE	0.0085 (0.0008)	<0.0051	<0.41	<9.2	216 (358)	27.87 (n = 1)
p,p′-DDE	0.0847 (0.0112)	0.023 (0.013)	5.28 (0.67)	14.19 (n = 1)	7,209 (15,821)	116 (254)
o,p′-DDD	0.0054 (n = 1)	<0.0051	<0.41	15.12 (n = 1)	29.9 (34.1)	<1.0
p,p′-DDD	0.0067 (n = 1)	<0.0051	1.62 (0.85)	<9.2	33.2 (18.6)	<1.0
o,p′-DDT	<0.0051	<0.0051	<0.41	<9.2	<4.0	<1.0
p,p′-DDT	<0.0051	<0.0051	<0.41	<9.2	37.8 (n = 1)	<1.0
ΣDDT	0.175 (0.015)	0.023 (0.013)	6.90 (1.52)	29.31 (n = 1)	7,544 (16,232)	143.87 (254)
ΣPCB	<0.0051	<0.0051	<0.41	<9.2	118 (133)	8.9 (10.6)

[a] DDE = 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethene; DDD = 1,1-dichloro-2,2-bis(*p*-clorophenyl)ethane; DDT = 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane; PCB = polychlorinated biphenyl; NA = not applicable; DP = Dana Point (CA, USA).
[b] The numbers in parentheses are standard deviations for replicate measurements.

southern California. The ban on discharge of DDT-enriched wastes via sewer systems in 1970 has led to a steady decline in the concentrations of DDTs (as well as PCBs) in sediments and biota of the SCB. Conversely, DDTs and PCBs apparently have become widely distributed in the SCB, that is, concentrations of DDTs and PCBs have decreased drastically in historically contaminated areas but have remained steady in relatively clean areas [1]. A bight-wide survey in 1994 concluded that total DDT and PCB were detectable in 82 and 46%, respectively, of the sediments of the SCB [27]. In addition, virtually 100% of the Pacifica sanddab, longfin sanddab, and Dover sole populations in the SCB were contaminated with total DDT, and almost 100% of Pacifica sanddab and longfin sanddab and 17% of Dover sole populations were contaminated with total PCB [28].

The present study further revealed that DDTs and PCBs were widely spread in the environmental compartments of different trophic levels in the SCB (Tables 2 and 3). One example was the detection of DDTs in the water column samples collected at station R52 in April–May 1996 when sampling was enhanced (Table 3). This location is far from any known sources of DDTs (other than the sediments). In fact, the water concentrations of total DDT at station R52 were of the same order of magnitude as those found at station T1, which is subjected to the influence of outflows from Santa Ana River (Fountain Valley, CA, USA) and Newport Bay (Newport Beach, CA, USA), which are known to contain DDTs [29,30]. Additionally, fish livers collected from station R52 contained fairly high levels of DDTs (as well as substantially lower levels of PCBs), although the concentration varied greatly with sampling seasons and individual samples (Tables 2 and 3).

Thermodynamics for partitioning of DDTs and PCBs between nonaqueous phases and overlying water

Although the current inputs of DDTs and PCBs from the JWPCP outfall have reached virtually nondetectable levels [4], historically discharged DDTs and PCBs may be recycled into the water column via other sources. Particularly, the possibility that contaminated sediments may serve a potential source of contamination [31] should be considered carefully when monitoring, assessment, and remediation efforts are taken. An assessment of the movement of DDTs and PCBs between the overlying water and other environmental compartments was conducted based on EPT as follows.

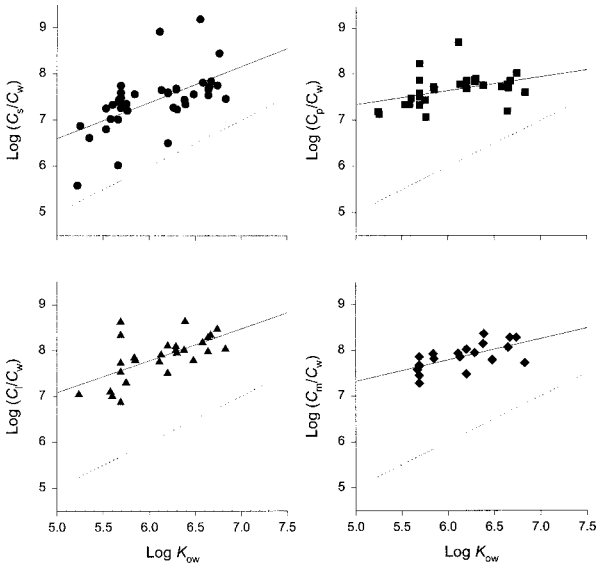

Fig. 2. Partitioning of 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT) and polychlorinated biphenyl (PCB) components between the overlying water and the other compartments of station 7C. The solid lines are linear regressions and the dotted lines represent the equilibrium partitioning state. All horizontal axes are log K_{OW}. C_i = total organic carbon–normalized or lipid-normalized concentration of a specific component in phase *i* where *i* is sediment (s), water (w), polychaete (p), fish liver (l), or fish muscle (m). K_{OW} = octanol–water partition coefficient.

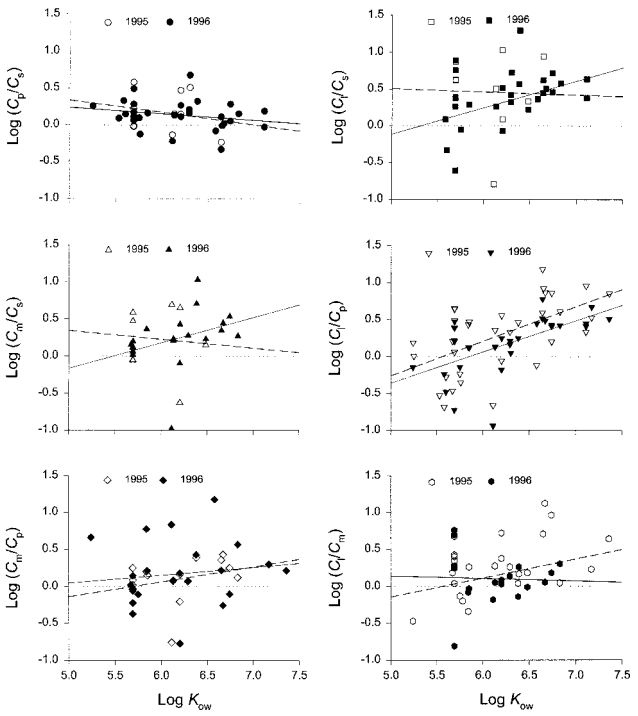

Fig. 3. Partitioning of 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT) and polychlorinated biphenyl (PCB) components among sediment, polychaete, or hornyhead turbot (liver and muscle tissues) collected from station 7C. The dotted lines represent the equilibrium partitioning state. All horizontal axes are log K_{OW}. C_i = total organic carbon–normalized or lipid-normalized concentration of a specific component in phase *i* where *i* is polychaete (p), sediment (s), fish liver (l), or fish muscle (m). K_{OW} = octanol–water partition coefficient.

Under strictly equilibrated conditions, the ratio of the concentrations of a specific chemical in a nonaqueous phase and overlying water is equal to K_{OW} (Eqn. 2). Any deviation from this equality corresponds to the thermodynamic potential for the chemical to move between the two phases. Therefore, this thermodynamic potential can be described by $\log(C_{w,e}/C_w) = \log(C_i/C_w) - \log K_{OW}$, where $C_{w,e}$ has been defined previously, C_w is the measured dissolved phase concentration, and *i* represents a phase interacting with the water phase. In this context, the thermodynamic potential for movement of a specific compound can be characterized by the difference between $\log(C_i/C_w)$ and log K_{OW}, which already was demonstrated for samples collected from station 7C (Fig. 2). The highly significant differences between any of the $\log(C_i/C_w)$–log K_{OW} regressions (Fig. 2) and the log K_{OW}–log K_{OW} correlation (Table 4) simply suggest that DDT and PCB compounds accumulated in sediment and biota (polychaete or fish) have a strong tendency to move into the overlying water at station 7C. Such a conclusion may not be drawn statistically from the samples collected from stations T1 and R52, although all but one $\log(C_i/C_w)$ were greater than log K_{OW}.

Table 4. Results of Student's *t* tests on the correlations of $\log(C_i/C_w)$ with log K_{OW} in the station 7C samples collected in April–May 1996 (Fig. 2)[a]

Variable	r^2	Slope[b]	Intercept[b]	*p* value
$\log(C_s/C_w)$	0.34	0.78 (± 0.35)	2.7 (± 2.1)	0.017
$\log(C_p/C_w)$	0.17	0.31 (± 0.26)	5.8 (± 1.6)	<0.001
$\log(C_l/C_w)$	0.39	0.70 (± 0.34)	3.6 (± 2.1)	0.003
$\log(C_m/C_w)$	0.41	0.47 (± 0.26)	5.0 (± 1.6)	<0.001

[a] C_i = total organic carbon–normalized or lipid-normalized concentration of a specific component in phase *i* where *i* is overlying water (w), sediment (s), polychaete (p), fish liver (l), or fish muscle (m); K_{OW} = octanol–water partition coefficient.

[b] Values in parentheses are 1.96 times standard errors from the Student's *t* tests.

The thermodynamic potentials between the nonaqueous phases and overlying water obtained above were consistent with a previous assessment that sediments around station 6C (not far from 7C) remained a major source of DDT contamination to the water column of the PVS [12]. The present study seems to support the hypothesis that sediments as a depositing reservoir for historically discharged contaminants may have now become a new source of contamination. This appears to indicate the need to allocate future investigation and assessment efforts toward understanding the mechanisms with which historically deposited contaminants are being recycled into the surrounding environments.

Bioaccumulation of chlorinated hydrocarbons in the food web of hornyhead turbot

Bioaccumulation by marine species provides an alternative for disposal or redistribution of contaminants in the environment. Bioaccumulation is a general process for accumulation of chemicals in aquatic organisms from all possible routes of uptake [32]. If uptake of chemicals occurs with bioaccumulation of freely dissolved contaminants directly from water via passive diffusion [33,34], it is defined as bioconcentration [35]. Uptake via food consumption often leads to biomagnification [35]. A third mechanism of uptake is via equilibrium partitioning between the interacting phases, resulting in equal fugacity among all of the phases [26].

As discussed in the previous section, the thermodynamic potentials between the nonaqueous phases and overlying water

Table 5. Results of Student's *t* tests on the correlations of $\log(C_i/C_j)$ with $\log K_{OW}$ (Figs. 3 and 4)

	October–November 1995			April–May 1996		
Variable	r^2	Slope[b]	Intercept[b]	r^2	Slope[b]	Intercept[b]
Station 7C						
$\log(C_p/C_s)$	0.04	0.17 (± 0.58)	1.2 (± 3.5)	0.05	−0.09 (± 0.14)	0.67 (± 0.86)
p value		0.58	0.53		0.24	0.14
$\log(C_l/C_s)$	0.00	−0.04 (± 1.0)	0.71 (± 6.3)	0.13	0.36 (± 0.36)	−1.9 (± 2.3)
p value		0.94	0.83		0.11	0.063
$\log(C_m/C_s)$	0.01	−0.12 (± 1.1)	0.94 (± 6.3)	0.13	0.34 (± 0.43)	−1.9 (± 2.7)
p value		0.83	0.78		0.18	0.13
$\log(C_l/C_p)$	0.32	0.47 (± 0.24)	−2.6 (± 1.5)	0.36	0.42 (± 0.21)	−2.5 (± 1.3)
p value		<0.001	0.002		<0.001	<0.001
$\log(C_m/C_p)$	0.09	0.20 (± 0.32)	−1.1 (± 2.0)	0.02	0.10 (± 0.34)	−0.47 (± 2.1)
p value		0.24	0.28		0.55	0.66
$\log(C_l/C_m)$	0.05	0.25 (± 0.44)	−1.4 (± 2.7)	0.00	−0.03 (± 0.43)	0.31 (± 2.6)
p value		0.27	0.32		0.88	0.82
Station 9C						
$\log(C_l/C_p)$	0.11	0.27 (± 0.29)	−1.1 (± 1.9)	0.04	0.09 (± 0.14)	−0.13 (± 0.89)
p value		0.09	0.24		0.23	0.78
$\log(C_m/C_p)$	0.03	−0.29 (± 0.98)	−1.5 (± 6.0)	0.01	−0.05 (± 0.26)	0.38 (± 1.6)
p value		0.57	0.63		0.72	0.65
$\log(C_l/C_m)$	0.06	0.22 (± 0.45)	−0.98 (± 2.7)	0.00	0.002 (± 0.044)	0.33 (± 0.25)
p value		0.36	0.50		0.92	0.023
Station T1						
$\log(C_l/C_p)$	0.06	−0.21 (± 0.66)	1.5 (± 4.4)			
p value		0.59	0.55			

[a] C_i and C_j = total organic carbon–normalized or lipid-normalized concentration of a specific component in phase *i* or *j* where *i* or *j* are polychaete (p), sediment (s), fish liver (l), or fish muscle (m).
[b] Values in the parentheses are 1.96 times standard errors from the Student's *t* tests.

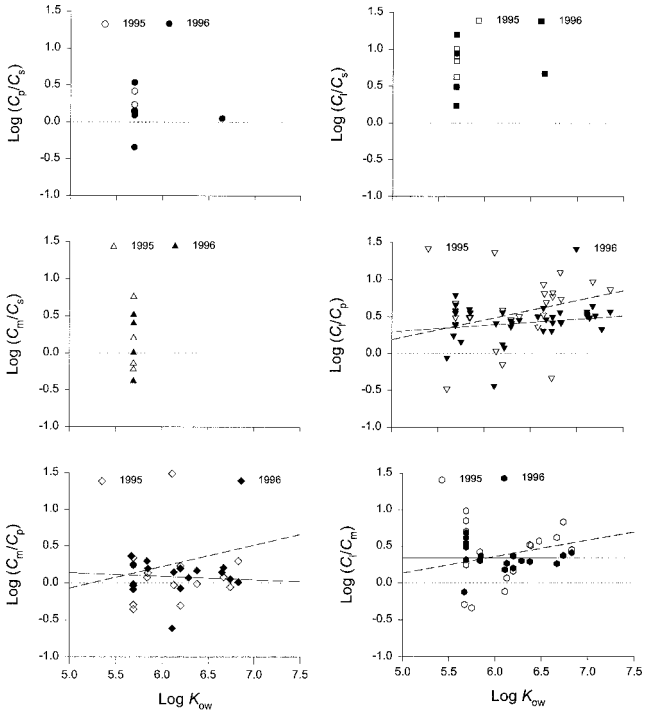

Fig. 4. Partitioning of 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT) and polychlorinated biphenyl (PCB) components amongst sediment, polychaete, or hornyhead turbot (liver and muscle tissues) collected from station 9C. The dotted lines represent the equilibrium partitioning state. All horizontal axes are $\log K_{OW}$. C_i = total organic carbon–normalized or lipid-normalized concentration of a specific component in phase *i* where *i* is polychaete (p), sediment (s), fish liver (l), or fish muscle (m). K_{OW} = octanol–water partition coefficient.

at station 7C indicated a strong tendency for DDTs and PCBs to move toward the overlying water (Fig. 2). Apparently, bioconcentration via the overlying water was unlikely to have been a significant mechanism leading to the enhanced accumulation of DDTs and PCBs in polychaete or fish.

For demersal fish such as hornyhead turbot occupying a higher trophic level than benthic organisms, food chain accumulation is expected to result in biomagnification. The contaminant-loaded particles may be a food source for filter-feeding or sediment-dwelling organisms. When the contaminant-containing lipids of the prey (e.g., polychaetes) are digested in the gut of the predator (e.g., hornyhead turbot), a higher chemical fugacity is established in the gut than in the rest of the predator's body. Because the lipid contents were higher in fish liver tissues than in muscle tissues and polychaetes, a net flow of chemicals from the gut into the liver would occur. The final outcome would be higher concentrations in the predator than those predicted by the EPT. High bioaccumulation levels in organisms occupying higher trophic positions, as compared to those in the lower end of the same food chain, were extensively investigated by many other researchers [33,36,37]. Oliver and Niimi [36] reported a stepwise increase in chemical concentrations of various PCB congeners in a benthic food chain consisting of phytoplankton, zooplankton, mysids, alewives, smelt, and salmonids in the Lake Ontario ecosystem. The total PCB concentrations in salmonids were nearly three orders of magnitude higher than those in their preys (alewives and smelt).

Our investigations indicated that transport of DDTs and PCBs from sediment to polychaete at station 7C was via equilibrium partitioning and no biomagnification existed, because the partitioning coefficients, $\log(C_p/C_s)$, are statistically identical to zero (Table 5). Transport between sediment and fish

(both liver and muscle tissues) also was an equilibrium process in the 7C samples. For the transport between fish liver and polychaete, a trend of increased $\log(C_l/C_p)$ with increasing log K_{OW} was evident in both the 7C and 9C samples (Figs. 3 and 4). Uptake via food sources likely is one of the important mechanisms for DDT and PCB enrichment in fish liver, resulting in a certain level of magnification. The liver contamination also was generally enriched relative to the muscle contamination.

Note that the dissolved phase in this study was operationally defined as containing materials that could pass through a 0.7-μm-pore-size filter and be retained by XAD-II resins. Obviously, some colloidal-bound or dissolved organic matter–bound organics might have been included in the dissolved phase. This would have resulted in overestimation of the measured dissolved-phase concentrations of DDTs and PCBs and, consequently, underestimation of the measured partition coefficients between nonaqueous phases and overlying water (Fig. 2). However, correcting the measured partition coefficients for the difference between the total dissolved phase and truly dissolved phase would not reverse the conclusions drawn in the present study. In contrast, such a correction would further support the conclusions because the measured partition coefficients would have been even greater than the EPT predictions.

CONCLUSIONS

Both DDTs and PCBs remain widely distributed in the SCB long after the ban on discharge of these chemicals via sewage outfalls. The detection of these potential carcinogens in the overlying water off Dana Point (California, USA), a presumably clean area, indicates the need for continuous research and monitoring. Thermodynamic potentials estimated based on EPT are suggestive of a strong tendency for these compounds to move from sediment or biota to overlying water. These findings favorably support the notion that the processes leading to the presence of DDTs and PCB in areas such as Dana Point away from the PVS, a historically contaminated area, occurred mostly in the past, that is, sediments containing historically discharged contaminants presently are recycling the contaminants to the water column.

Transport of DDTs and PCB among the food chain of hornyhead turbot (water, sediment, polychaete, and fish liver and muscle tissues) exhibited different characteristics at each trophic level. First, bioconcentration via passive diffusion was an unlikely mechanism contributing significantly to the DDT and PCB enrichment in polychaete and fish. Second, transport of DDTs and PCBs between sediment and polychaete or fish muscle tissue was dictated by equilibrium partitioning. Finally, uptake in fish liver tissue was mainly via food sources, resulting in moderate biomagnification.

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