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CHAPTER 10

Bioaccumulation of *p,p'*-DDE and PCB 1254 by a Flatfish Bioindicator from Highly Contaminated Marine Sediments of Southern California

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INTRODUCTION

Bottom sediments are a major reservoir for residues of the pesticide DDT and polychlorinated biphenyls (PCBs) released into aquatic environments. Fish consumption warnings or fishery closures in areas polluted by these chlorinated hydrocarbons are increasing. Thus, it is important to understand the processes by which such hydrophobic neutral synthetic organic compounds are incorporated into tissues of benthic seafood organisms. The fugacity model of bioaccumulation states that uptake is determined by the chemical fugacity differential between the organism and its environment. For benthic species this model most conveniently is tested by measuring residue concentrations in tissue (C[t]) and in the sediment (C[s]) to which the organism has been exposed. Here we describe such a test conducted through a field study of surficial sediments and a flatfish used successfully as a bioindicator for chlorinated hydrocarbon contamination in the Southern California Bight.

BACKGROUND

Numerous investigations over the last two decades showed that concentrations of DDT and PCB residues in sediments and organisms from the Southern California Bight were among the greatest reported for any coastal marine ecosystems.[1-10] The principal constituents of these residues have been identified, respectively, as *p,p'*-DDE and a PCB mixture most closely resembling Aroclor 1254.[11-12] Highest values occurred on the Palos Verdes Shelf, which received municipal wastewater discharges from the Joint Water Pollution Control Plant (JWPCP) submarine outfall system of the Los Angeles County Sanitation Districts. Bottom sediments on the shelf also contained relatively high concentrations of organic material and supported large populations of

certain benthic/epibenthic organisms, such as the Dover sole (*Microstomus pacificus*). During the 1970s this flatfish, which often is found partially buried in the surficial sediment layer, was severely affected by a fin erosion disease.[13] Distinct gradients of both the incidence of this disease[14] and tissue concentrations of DDTs and PCBs,[15] generally associated with the sediment contamination gradient away from the JWPCP outfall system, suggested that this flatfish was a potentially useful bioindicator of benthic pollution in the Southern California Bight.[16] Therefore, when an extensive survey of bottom sediments was conducted during 1977 along the 60-m isobath of the Southern California coast,[17-18] tissues from Dover sole specimens also were collected and analyzed from a number of sites both on the Palos Verdes Shelf, and from reference zones to the north and south of this highly contaminated area.

PROCEDURES

A synoptic collection of surficial bottom sediments was obtained during summer 1977 with a modified van Veen grab sampler from a water depth of 60 m at numerous stations along the southern California coast.[17-18] The positions (latitude and longitude) of these stations are listed by Word and Mearns.[18] A single grab sample was taken at each station, and the upper 2 cm was subsampled using a clean stainless-steel spatula. Specimens of the Dover sole were collected in bottom trawls conducted along transects near five of the sediment stations on the Palos Verdes Shelf (JWPCP Monitoring Program trawl transects T1-200′ through T5-200′), and near ten stations in the reference zone to the north and south of the shelf (Figure 10.1). In three cases, the trawls were made between sediment stations in the reference zone. Therefore, average values for each pair of sediment samples (from stations 19 and 21; 41 and 45; 45 and 49) were taken as estimates of the surficial sediment concentrations to which flatfish from these trawls were exposed. Generally, six specimens were taken from each station trawl for analysis. The samples of sediment (in precleaned glass bottles with Teflon-lined caps) and flatfish (in plastic bags) were returned to the laboratory and frozen on the day collected, pending processing for analysis. Using procedures described in Word and Mearns,[18] aliquots of the homogenized sediment samples were analyzed for several conventional sediment parameters including total volatile solids (TVS). The methods described by Young et al. were used for the analysis of *p,p′*-DDE and PCB 1254.[5,11] First, approximately 40 g of wet sediment were oven-dried at 60°C for 24 hr. The sample then was extracted with *n*-hexane and cleaned up on activated Florisil; one-half of the extract was saponified for PCB analysis. Measurements on these extracts were conducted as described below.

Using a metal scalpel with a carbon-steel blade, the flatfish specimens were dissected while still semifrozen to minimize contamination of the muscle and liver tissue samples by mucous or visceral fluids. Approximately 5-10 g of wet muscle and the entire liver (typically weighing 1-5 g) were taken for DDE and

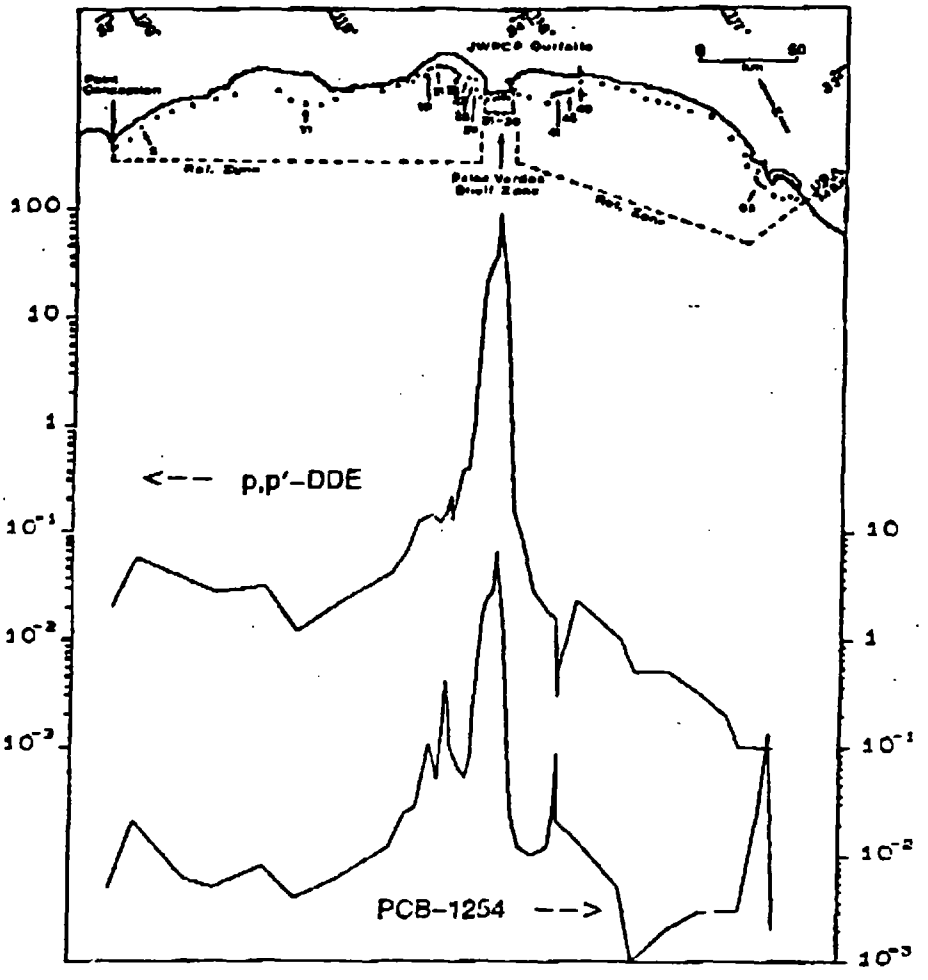

Figure 10.1. Southern California Bight station locations for the 1977 60-m surficial sediment collections, and corresponding concentrations (mg/kg dry wt.) of p,p'-DDE and PCB 1254. Station numbers for sites of Word and Mearns[18] included in this study, and associated reference and Palos Verdes Shelf zones, are indicated.

PCB analysis. In addition, aliquots of these tissue samples obtained from the six specimens collected at two of the Palos Verdes Shelf stations (33 and 34) and at three reference zone stations (41/45, 45/49, and 65) were analyzed for lipid content according to the chloroform/methanol extraction procedure of Bligh and Dyer.[19]

Tissue samples to be analyzed for *p,p'*-DDE and PCB 1254 residues were extracted successively in acetonitrile and *n*-hexane. These extracts then were reduced in volume and cleaned up on activated Florisil. Analysis was conducted by electron capture gas chromatography using packed columns (1.5% OV-17 and 1.95% QF-1 on Gas Chrom Q). Quantitation of *p,p'*-DDE was

Table 10.1. Median Values (and Their Ratios) for Station Concentrations of Sediment Total Organic Carbon, *p,p'*-DDE, and PCB 1254 in the Palos Verdes Shelf (n = 5) and Reference (n = 10) Zones

Zone	TOC (% dry wt.)	p,p'-DDE (μg/g dry wt.)	PCB 1254 (μg/g dry wt.)
Shelf:			
Median	7.6	27	2.3
Range	(4.3–12)	(20–92)	(1.4–6.6)
Reference:			
Median	0.62	0.09	0.06
Range	(0.26–3.7)	(0.001–1.1)	(0.004–0.10)
Ratio of Zone Mds.	12	300	58

Note: Sediment total organic carbon calculated from TVS values using the regression:[22]
% TOC = 0.484 (% TVS − 1.86)

accomplished by direct comparison of its peak height with that of a standard obtained from U.S. EPA. The logarithm (base 10) of the octanol:water partition coefficient (log K_{ow}) for this compound is 5.8.[20] PCB 1254 was quantified against a corresponding Aroclor 1254 standard. The major IUPAC congener in the chromatograph profile chosen for this quantitation has been tentatively identified (by coauthor R. W. Gossett) as congener #110, which has a log K_{ow} of approximately 6.5.[21] All sediment and tissue concentrations were corrected for procedural blank and recovery values.

RESULTS AND DISCUSSION

The distributions of *p,p'*-DDE and PCB 1254 in the 1977 collections of surficial sediment from the 60-m isobath of the Southern California Bight are illustrated in Figure 10.1 and are summarized in Table 10.1. The data indicate that median sediment concentrations of these residues on the Palos Verdes Shelf were 38 to 300 times greater than those in the reference zone. In addition, TVS values quantifying organic content of the sediments ranged from 11 to 27% on the shelf, compared to values below 10% in the reference zone. Thus, for the purposes of this analysis the stations were classified into two groups or zones: a high-contamination shelf zone off the Palos Verdes Peninsula (stations 31–35) and a reference zone containing the other ten stations.

Total organic carbon (TOC) was not measured in these sediment samples. However, Mitchell and Schafer obtained the following regression ($r^2 = 0.986$; $p < 0.001$) between surficial sediment concentrations of TVS and TOC in 1974 samples obtained over a zone extending 16 km from the sludge outfall of Los Angeles City's Hyperion Municipal Wastewater Treatment Plant in Santa Monica Bay:[22]

$$\% \text{ TOC} = 0.484\ (\% \text{ TVS} - 1.86) \qquad (10.1)$$

Table 10.2. Median Muscle or Liver Tissue Concentrations of *p,p'*-DDE and PCB 1254 for Dover Sole Specimens from Each Shelf Zone Station and Corresponding Overall Median Values (and Ranges of Station Medians) for the Reference Zone

Sediment Station	Trawl Station	Muscle (μg/g wet wt.)			Liver (μg/g wet wt.)		
		n	*p,p'*-DDE	PCB 1254	n	*p,p'*-DDE	PCB 1254
Shelf Zone							
31	T1	2	12	0.51	—	—	—
32	T2	6	16	1.1	6	210	10
33	T3	6	22	1.4	6	240	17
34	T4	6	19	1.2	6	160	12
35	T5	5	8.0	0.21	—	—	—
Median			16	1.1	—	210	12
Reference Zone							
Median			0.24	0.11		0.80	1.5
Range			0.02–2.5	0.01–0.36		0.2–6.7	0.2–5.6
No. Stations			10	10		6	6
Ratio of Zone Mds.			67	10		260	8

Note. Median of individual station median tissue concentrations rounded to two significant figures.

The TVS values used to obtain this regression ranged from 3 to 52%, which encompassed the range (3 to 27%) obtained in the 1977 60-m sediment survey. Thus, this regression equation was used to estimate TOC concentrations from the sediment TVS concentrations.

The sediment concentrations (on a dry weight basis) obtained for the two study zones show that median concentrations of sediment TOC, *p,p'*-DDE, and PCB 1254 for the shelf zone were higher than those for the reference zone by factors of 12, 300, and 38, respectively (Table 10.1). Further, the percent TOC ranges for the two zones (4.3–12 vs 0.26–3.7) did not overlap, and the lower limit of the ppm DDE range (20) for the shelf zone was 18 times the upper limit of the range (1.1) for the reference zone. Similarly, the lower limit of the ppm PCB range (1.4) for the shelf zone was 14 times the upper limit of the range (0.10) for the reference zone. The fact that the surficial sediments of these two zones were so different in these parameters provided a good opportunity to test, from field data, the utility of the fugacity model of benthos:sediment bioaccumulation for a common marine flatfish of the northeastern Pacific.

In this analysis we have elected to use the median tissue concentration as the measure of central tendency for fish muscle or liver contamination at each station. Therefore, in Table 10.2 we list the median concentration (on a wet weight basis) of *p,p'*-DDE or PCB 1254 in Dover sole muscle or liver tissue obtained for individual shelf zone stations. Also shown are the reference zone overall median values (and ranges) obtained from the median specimen concentrations of DDE or PCB for each station in this zone. Liver tissue was analyzed in specimens collected at only three of the five shelf zone stations and

six of the ten reference zone stations. The ratios of the zone median values indicate that levels of DDE and PCB contamination in the tissues of specimens from the shelf zone were 67 to 260 and 8 to 10 times greater than those in specimens from the reference zone, respectively.

Previous studies had shown that the lipid content of Dover sole specimens collected from the Palos Verdes Shelf was substantially higher than that measured in specimens collected elsewhere off southern California.[15,23] We obtained similar results. Median (wet weight) muscle tissue concentrations of extractable lipid for the shelf and reference zone specimens analyzed (n = 12 and 18, respectively) were 2.36 and 1.34%; corresponding values for liver tissue from all of these specimens were 24.8 and 13.2%. Thus, our best estimate is that muscle and liver tissue for Dover sole specimens collected during 1977 from the Palos Verdes Shelf each contained approximately 80% more extractable lipid than did corresponding specimens from the reference zone.

The very large zonal differences in sediment and tissue concentrations of DDE and PCB first were used to evaluate the least complex form of the fugacity model.[24] This model is a simple partition coefficient commonly termed the *bioaccumulation factor* (BAF):

$$BAF = C_t/C_s \tag{10.2}$$

where C_t and C_s are a contaminant's concentrations in the specimen tissue and corresponding sediment samples, respectively. In our approach, median wet-tissue-to-dry-sediment ratios for *p,p'*-DDE and PCB 1254 in specimens from each trawl station were calculated by dividing the station median wet weight tissue concentrations by the corresponding dry weight sediment value (Table 10.3). For consistency of units, bioaccumulation factors typically are obtained from the concentration ratios of tissue and sediment each on a dry weight basis. However, percent moisture values for the flatfish tissues analyzed in this study were not available. Further, the fact that criteria for chlorinated hydrocarbon residues in seafood are promulgated on a wet weight basis, while sediment concentrations typically are reported on a dry weight basis, supports the utility of such a mixed-unit index (modified bioaccumulation factor, MBAF) for evaluating conditions leading to contamination of living resources.

The results listed in Table 10.3 suggest that use of the modified bioaccumulation factor may be misleading regarding the relative bioavailability of the contaminants in the two zones. Whereas the ratio of shelf-to-reference zone median concentrations (wet weight basis) for the four tissue-contaminant pairs ranged from 8 to 260 (Table 10.2), the corresponding ratios for the four modified bioaccumulation factors all were less than 1.0, ranging from 0.07 to 0.20. (We note that, assuming the muscle or liver tissue percent water values are similar for specimens from the two zones, the ratios of zone median BAFs also would be similar to those for the MBAFs given in Table 10.3.) Such MBAF (or BAF) values alone might be interpreted as

Table 10.3. Median *p,p'*-DDE and PCB 1254 Tissue/Sediment Modified Bioaccumulation Factors for Individual Shelf Zone Stations: Ratios of Concentrations in Tissues of Dover Sole Specimens Normalized to Corresponding Surficial Sediment Concentrations and Corresponding Overall Median Values (and Ranges of Station Medians) for the Reference Zone

Sediment Station	Trawl Station	Muscle			Liver		
		n	*p,p'*-DDE	PCB 1254	n	*p,p'*-DDE	PCB 1254
Shelf Zone							
31	T1	2	0.55	0.30	—	—	—
32	T2	6	0.57	0.50	6	7.7	4.4
33	T3	6	0.57	0.48	6	6.1	6.9
34	T4	6	0.20	0.18	6	1.8	1.8
35	T5	6	0.40	0.15	—	—	—
Median			0.55	0.30		6.1	4.4
Reference Zone							
Median			2.7	2.0		62	59
Range			0.40–32	0.35–5.1		12–410	9–120
No. Stations			10	10		6	6
Ratio of Zone Mds.			0.20	0.15		0.10	0.07

evidence that *p,p'*-DDE and PCB 1254 were less available for accumulation by Dover sole specimens from the Palos Verdes Shelf than by specimens from the reference zone.

Therefore, we examined the next level of the fugacity model. Here the approach utilizes a more complex partition coefficient obtained by normalizing C_t to tissue extractable lipid concentration L, and C_s to sediment TOC concentration.[24] The resultant ratio of normalized concentrations is termed the *accumulation factor* (AF):

$$AF = (C_t/L)/(C_s/TOC) \qquad (10.3)$$

These factors were calculated for *p,p'*-DDE and PCB 1254 concentrations obtained at each trawl station by dividing the station's median concentration for muscle or liver, normalized to the appropriate median lipid concentration value for a given zone and tissue, by the corresponding TOC-normalized sediment concentration (Table 10.4).

This application of the fugacity model of bioaccumulation, incorporating tissue lipid and sediment TOC normalizations, generally yielded good agreement between the degree of contaminant accumulation from bottom sediment (as characterized by station and zone median values) in Dover sole from the two study zones (Table 10.4). Median AF values for *p,p'*-DDE in muscle tissue of specimens from the shelf and reference zones were 1.7 and 1.8, respectively; corresponding AF values for the liver tissue were 2.0 and 3.4. Similar agreement was observed for PCB 1254. Median AF values for the two zones were 0.96 and 1.3 for muscle tissue, and 1.4 and 2.7 for liver tissue. Thus, despite the very large differences in median levels of sediment and tissue contamination between the two zones, this application of the

Table 10.4. Median Accumulation Factors for *p,p'*-DDE and PCB 1254 in Dover Sole Tissues Obtained for Specimens from Each Shelf Zone Station and Corresponding Overall Median Values (and Ranges of Station Medians) for the Reference Zone

Sediment Station	Trawl Station	Muscle			Liver		
		n	*p,p'*-DDE	PCB 1254	n	*p,p'*-DDE	PCB 1254
Shelf Zone							
S1	T1	2	1.7	0.96	—	—	—
S2	T2	6	1.8	1.5	6	2.3	1.4
S3	T3	6	1.9	1.6	6	2.0	1.9
S4	T4	6	1.0	0.96	6	0.86	0.91
S5	T5	5	0.74	0.27	—	—	—
Median			1.7	0.96		2.0	1.4
Reference Zone							
Median			1.8	1.3		3.4	2.7
Range			(0.38–14)	(0.24–2.3)		(1.2–19)	(1.0–4.2)
No. Stations			10	10		6	6
Ratio of Zone Mds.			0.94	0.74		0.59	0.52

fugacity model of bioaccumulation yielded results for a given tissue and contaminant that each agreed within a factor of two. These results, based on field data obtained from a relatively small number of specimens (≤ 6) per station and of stations (5–10) per study area, indicate the potential usefulness of the approach in evaluating benthic contamination by major DDT and PCB residues on a regional basis.

In addition to providing comparable results for tissue:sediment ratios over a very large range of exposure, application of this fugacity model to the results of our survey yielded AF values that agree with the value predicted from independent laboratory experiments. McFarland[25] and McFarland and Clarke[26] analyzed results of separate experiments on partitioning of hydrophobic neutral trace organics between (1) sediment organic carbon and water and (2) fish and water. Assuming that octanol was a satisfactory surrogate for the total organic carbon pool to which an organism was exposed, they concluded that, under equilibrium conditions, the partition coefficient that is equivalent to the accumulation factor considered here should have a value of about 1.72. The results of our analysis for *p,p'*-DDE and PCB 1254 presented in Table 10.4 are in good agreement with this prediction. The shelf zone median AF value for these two hydrophobic neutral synthetic organics in flatfish muscle and liver range from 0.96 to 2.0, with a median value of 1.55. If the reference zone median AF values are included, the resultant median value is 1.75, similar to the equilibrium value (1.72) predicted by McFarland and Clarke.[26] Further, the lower (0.96) and upper (3.4) limits of the range of eight zonal median AF values agree with the predicted value within a factor of two.

SUMMARY AND CONCLUSIONS

A simple analysis of field-generated data on two hydrophobic (log $K_{ow} \cong 6$) neutral synthetic organic contaminants of the coastal marine ecosystem off southern California supports the fugacity model of bioaccumulation. Limitations of the study that might compromise the accuracy or precision of the results include estimation of sediment TOC from total volatile solids concentrations and extrapolation of tissue lipid median concentrations for each zone to nonanalyzed specimens. Also, the survey design included relatively few sediment ($n = 1$) and flatfish ($n \leq 6$) samples per station, and relatively few stations per study area (shelf zone: $n = 3$–5; reference zone: $n = 6$–10 for liver and muscle tissues, respectively). Further, the variability of sediment exposure experienced by the mobile flatfish specimens trawled near a given sediment station is unknown. Despite these limitations, the accumulation factors obtained for *p,p'*-DDE and PCB 1254 based on lipid normalization of flatfish muscle and liver tissue concentrations, and TOC normalization of surficial sediment concentrations, produced remarkably consistent results. Ratios of zonal median AFs, based on station median AFs for the two tissues and two contaminants, yielded values ranging from 0.52 to 0.94. These results indicated agreement between shelf and reference zone median AF values that was within a factor of two, despite the large range of sediment and tissue concentrations measured in the two study areas.

Finally, the four median AF values obtained for the shelf zone ranged from about 1.0 to 2.0 (median = 1.6). Corresponding results for the reference zone (where concentrations were lower and resultant uncertainties higher) ranged from 1.3 to 3.4 (median = 2.2). The overall median of these eight values was about 1.8, in good agreement with the value of 1.7 predicted for hydrophobic neutral trace organics from analysis of laboratory partitioning experiments. This provides further support for the reliability of the fugacity model of bioaccumulation, and its potential usefulness in predicting levels of such compounds expected in benthic organisms exposed to contaminated bottom sediments.

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