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Bioaccumulation of Chlorobiphenyls and Endrin from Food by Lobsters (*Homarus americanus*)

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Marine organisms concentrate organochlorine compounds to a lesser extent from contaminated food than from water. For example, concentration factors (CFF) for fish exposed to Aroclor 1254 in food ranged from 1 to 7 (MAYER et al. 1977; FREEMAN et al. 1978). CFFs for a crab exposed to endrin in food were 5 to 7 (PETROCELLI and ANDERSON 1975). With exposure to Aroclor 1254 in water, concentration factors (CFW) for fish were 61,000 and 100,000 (MAYER et al. 1977; VIETH et al. 1979), and for large and small Nereis virens at 24 h, they were 50 to 500 (derived from McLEESE et al., in press). With exposure to endrin in water, fish had a CFW of 1500 (NEELY et al. 1974) and oysters, Crassostrea virginica, had a CFW of 2700 (MASON and ROWE 1976). However, because of the low concentration of organic pollutants in sea water, BRYAN (1979) concluded that food is the main source of organochlorine residues in fish and crustaceans.

The disappearance of organochlorines from invertebrates following exposure to contaminated food has not been studied. McLEESE et al. (in press) found practically no loss of Aroclor 1254 from Nereis virens after exposure to contaminated water. The biological half-lives of endrin and dieldrin in oysters following exposure to contaminated water were 67 and 75 h, respectively (MASON and ROWE 1976).

The objectives of this paper are to determine the rates of dietary accumulation and clearance of two chlorobiphenyls and endrin in lobsters (Homarus americanus) fed contaminated mussels (Mytilus edulis). The chlorobiphenyls, representing intermediate and high degrees of chlorination, were chosen as model compounds to simulate the accumulation of PCB's. Endrin has been used extensively in eastern Canada and therefore it is of interest to determine its potential for accumulation within the trophic chain.

METHODS

Chlorobiphenyls

Lobsters (450 g) were maintained in individual compartments within larger tanks at 10°C. Before each feeding, 2,2',4,5' tetrachlorobiphenyl (TPCB) and 2,2',4,4',5,5' hexachlorobiphenyl (HPCB) in a 1:1 ratio dissolved in hexane were applied to shucked mussel

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tissue at nominal concentrations of 1.7 and 17 mg/kg for each chlorobiphenyl, and the hexane was allowed to evaporate. Each lobster was fed about 4.2 g of the treated tissue every other day for 6 wk. This was followed by 6 wk of feeding with uncontaminated mussel tissue. Two, or sometimes three, lobsters were removed at 2-wk intervals and hepatopancreas, tail and claw muscle samples as well as samples of food were frozen for later analysis.

Endrin

Endrin (6 mg), dissolved in methanol, was added to 2 L of sea water and shucked mussel tissue was soaked in the solution for 2 h. After draining, the mussel tissue was minced in a blender and divided into 2-g portions. Each portion was mixed with approximately 1 g of fish meal and frozen for use throughout the uptake phase of the test.

Lobsters (450 g) were maintained in individual compartments at 10°C and each was fed a portion of prepared food every other day for 2 wk. Each lobster was observed until the food was eaten; usually a period of less than 5 min. To follow excretion, the lobsters were held for an additional 4 wk and fed uncontaminated mussel tissue. Control lobsters were fed uncontaminated mussel tissue throughout the 6-wk period.

During the uptake phase, two or three lobsters were sampled 12-h after feedings 1, 2, 3, 5 and 7. The sampling delay allowed for incorporation of the ingested endrin. During the excretion phase, one or two lobsters were sampled at 1, 2 and 4 wk. The hepatopancreas and tail muscle of the lobsters were frozen for later analysis.

Chemical analysis

Mussel and lobster tissues were ground with anhydrous sodium sulfate and extracted with pesticide-grade hexane in a Soxhlet apparatus for 1 h. Extracts were cleaned-up on alumina by the method of ZITKO et al. (1974), evaporated to dryness, and re-dissolved in pesticide-grade iso-octane for analysis by gas chromatography.

Samples were analyzed on a Varian model 3700 gas chromatograph equipped with a ^{63}Ni electron capture detector. The 2 m x 2 mm I.D. glass column was packed with 3% OV-101 on 80/100 Chromosorb W-HP. Instrument temperatures were 190°, 210° and 300°C for the column, injector and detector, respectively.

Calculations

Concentration factors were calculated as the concentration of the compound in the lobster tissue on a wet weight basis divided by the average concentration of the compound in the food on a wet weight basis. Uptake rates (K1), excretion rates (K2) and accumulation coefficients (K_B = K1/K2) were calculated according to the one-compartment model of BRANSON et al. (1975) using the computer program of BLAU and AGIN (1978).

Chlorobiphenyl feeding test

The concentrations of TPCB and HPCB in food, expressed as the mean of two analyses, were 0.60 and 0.55 mg/kg wet weight at the low dosage and 4.0 and 4.9 mg/kg wet weight at the high dosage treatment. Data on the uptake and excretion of TPCB and HPCB in the hepatopancreas (Fig. 1 and 2) indicate that concentrations of these compounds approach an equilibrium within 4 wk, except that HPCB concentrations in the high dosage tests increased throughout the uptake phase. Maximum concentration factors (concentration in hepatopancreas/ concentration in food) were 5.1 and 7.3 for TPCB and HPCB, respectively at the low dietary level and 1.0 and 2.9, respectively at the high dietary level.

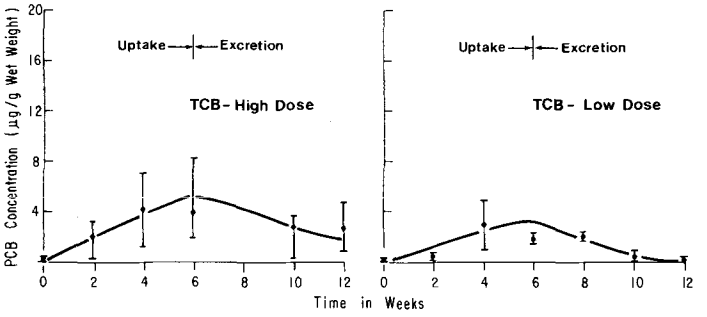

Fig. 1. Concentration of TPCB in lobster hepatopancreas. Diets during uptake were mussel tissues with high (4.0 mg/kg) and low (0.6 mg/kg) doses. Bars represent the range of concentrations in duplicate samples.

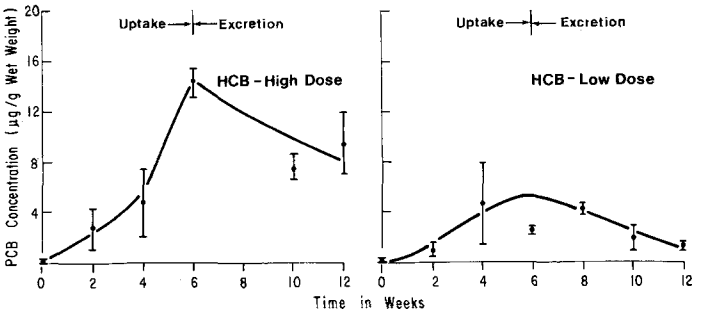

Fig. 2. Concentration of HPCB in lobster hepatopancreas. Diets during uptake were mussel tissues with high (4.9 mg/kg) and low (0.55 mg/kg) doses. Bars represent the range of concentrations in duplicate samples.

In all cases there was a decrease in the concentration of chlorobiphenyls in the hepatopancreas during the 6-wk clearance period. In the low dosage tests the concentrations approached control values after 6 wk clearance, and in the high dosage tests the concentrations of TPCB and HPCB declined by approximately 60 and 40%, respectively.

The lipid content of the hepatopancreas ranged from 10 to 28%. There was no discernible relationship between chlorobiphenyl concentration and lipid content in the hepatopancreas.

The maximum concentrations and maximum CFFs for TPCB and HPCB in tail and claw muscle (Table 1) are considerably lower than those in

TABLE 1

Measured chlorobiphenyl concentration and maximum concentration factors (CFF) in lobster tail and claw muscle during uptake (0-6 wk) and excretion (6-12 wk) phases. Diet during uptake was at the high dosage.

	Mean concentration (ng/g wet wt)			
	Tail muscle		Claw muscle	
Time (wk)	TPCB	HPCB	TPCB	HPCB
0	3	1	7	3
2	8	1	33	37
4	9	3	21	10
6	39	27	273	123
8	5	27	52	198
10	12	11	49	91
12	8	15	60	76
Maximum CFF	.01	.006	.068	.04

the hepatopancreas. By the end of the excretion phase, chlorobiphenyl concentrations in the muscle tissues were approaching control values; indicating that most of the residues had been eliminated.

The lipid content of tail and claw muscle ranged from 0.11 to 0.44% and from 0.14 to 0.72%, respectively.

Endrin

The endrin content of spiked food averaged 4.7 mg/kg wet weight. Control lobsters did not contain detectable endrin residues at the start or end of the 6-wk period.

In the hepatopancreas, endrin reached a maximum concentration of 1.95 mg/kg wet weight (CFF = 0.41) after 2 wk of feeding and declined by about 65% after 4 wk excretion (Fig. 3). There was an indication

of an approaching equilibrium concentration during the uptake phase. Endrin in tail muscle reached a maximum concentration of .0047 mg/kg wet weight (CFF = 0.001) after 2 wk of feeding and declined by about 35% by the end of the excretion period (Fig. 4).

The lipid content of the lobster hepatopancreas and tail muscle ranged between 15 to 33% and 0.17 to 0.56%, respectively. There was no relationship between endrin concentration and lipid content in the hepatopancreas.

DISCUSSION

Endrin has a lower uptake rate constant (K1) than either of the chlorobiphenyls (Table 2). HPCB has a much lower excretion rate constant (K2) than the other compounds. Consequently, the accumulation coefficient (K_B) for HPCB is considerably higher. Estimates of equilibrium concentrations and times to 90% equilibrium indicate that HPCB reaches an equilibrium concentration of 49 mg/kg after 65 wk, a longer period than the other compounds. From a consideration of the excretion parameters (Table 2), the ranking for time to 50% clearance is endrin = TPCB < HPCB.

Accumulation coefficients (K_B) are similar to the range of CFFs reported by MAYER et al. (1972), FREEMAN et al. (1978) and PETROCELLI and ANDERSON (1975), for fish and invertebrates exposed to organochlorines in food, even though the dietary concentrations and feeding regimes vary between studies.

Direct comparison of the uptake of chlorobiphenyls and endrin cannot be made because of differences in the amount of contaminant provided at each feeding and in the length of the feeding phase. To resolve these difficulties, the data are expressed in terms of uptake efficiency; that is, the total amount of the compound accumulated by the hepatopancreas as a percentage of the total amount administered.

TABLE 2

Uptake (K1) and excretion (K2) rate constants, accumulation coefficients (K_B), equilibrium concentrations and times to equilibrium (uptake) and times to 50% clearance (excretion) for TPCB and HPCB at high dietary levels and for endrin.

Compound	K1 (wk^{-1})	K2 (wk^{-1})	K_B	Equil. conc. (µg/kg)	Time to 90% equil. (wk)	Time to 50% clearance (wk)
TPCB	0.32	0.16	2.0	8.1	15	4
HPCB	0.40	0.04	10.0	49.0	65	17
Endrin	0.17	0.16	1.1	5.2	15	4

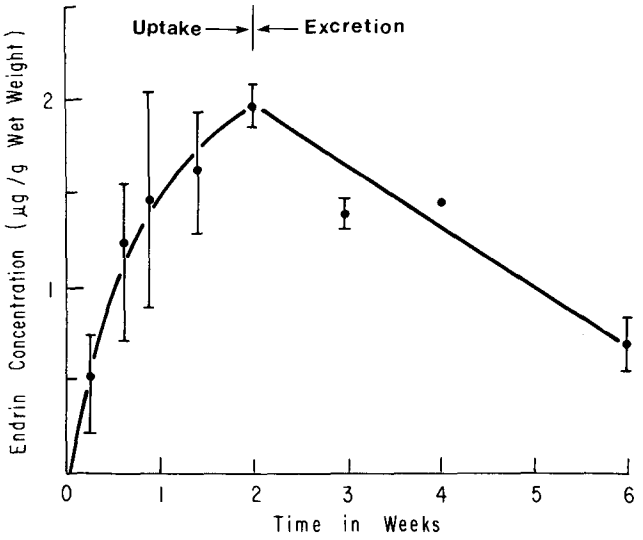

Fig. 3. Concentration of endrin in lobster hepatopancreas. Diet during uptake was mussel tissue with 4.7 mg/kg of endrin. Bars represent the range of concentrations in two or three samples.

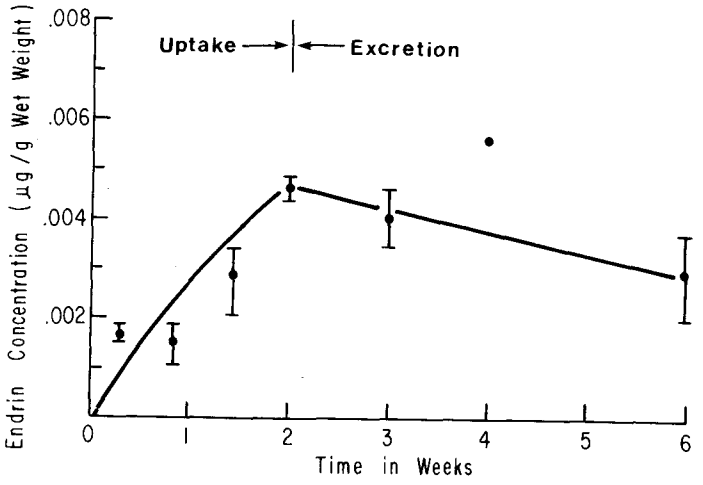

Fig. 4. Concentration of endrin in lobster tail muscle. Diet during uptake was mussel tissue with 4.7 mg/kg of endrin. Bars represent the range of concentrations in two or three samples.

The efficiency of uptake declines as the concentration in the hepatopancreas approaches an equilibrium, so that data from 2-wk uptake (Table 3) are best suited for comparisons of uptake efficiency. It was assumed for all calculations that the average wet weight of the hepatopancreas is 24 g.

TABLE 3

Efficiency of uptake of chlorobiphenyls and endrin in the hepatopancreas of lobsters at 2-wk uptake.

Compound	Amount administered per feeding (µg)	Efficiency of uptake (%)
TPCB	2.5	52
	9.4	45[a]
	16.8	41
HPCB	2.3	134
	9.4	75[a]
	20.6	40
Endrin	9.4	71

[a]Interpolated value.

The uptake efficiency for TPCB and HPCB decreases with increasing dietary dosage (Table 3). The efficiency for HPCB at the low dietary level was greater than 100%; presumably as a result of variations in the HPCB content of the food. The uptake efficiencies of chlorobiphenyls at a dose of 9.4 µg/feeding were calculated by interpolation from data at the higher and lower dosage tests, assuming a linear relationship between them. Uptake efficiencies at this common dietary level are ranked as TPCB < endrin ≤ HPCB. Correlations have been made between bioaccumulation of organic compounds and their water solubilities (LU and METCALF 1975; METCALF et al. 1975) or their octanol-water partition coefficients (CHIOU et al. 1977; METCALF et al. 1975; NEELY et al. 1974). The water solubility is 260 µg/L for endrin, 21 µg/L for TPCB and 2.1 µg/L for HPCB (WEIL et al. 1974). The ranking by uptake efficiency with TPCB before endrin does not agree with ranking by water solubility. There is general agreement between the ranking by uptake efficiency and by octanol-water partition coefficients. The log octanol-water partition coefficients are 5.7 for TPCB (calculated by the method of LEO et al. 1971), 5.8 for endrin (calculated by the method of CHIOU et al. 1977) and 6.7 for HPCB (CHIOU et al. 1977).

Chlorobiphenyls and endrin in food are accumulated to a greater degree in lobster hepatopancreas than in tail or claw muscle. The low CFFs for TPCB and HPCB in lobster tail muscle (Table 1) and low concentrations of endrin in tail muscle (Fig. 4) indicate that gross

contamination of muscle tissue is unlikely. Compared with the hepatopancreas, the lower concentrations of these compounds in muscle may be related to the lower lipid content of the muscle and to the role of the hepatopancreas in food incorporation.

The bioaccumulation of organochlorines by aquatic organisms from contaminated food may be several orders of magnitude less than from water (MAYER et al. 1977; VIETH et al. 1974). However, concentrations of organic contaminants in sea water are so low (DAWSON and RILEY 1977; PAVLOU and DEXTER 1979) that accumulation from food may represent the main source of residues for animals in the higher trophic levels (BRYAN 1979).

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