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UPTAKE AND ELIMINATION OF PCBS IN FISH CONTAMINATED BY THE WAUKEGAN HARBOR

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Introduction

The sediments of the harbor of Waukegan, Illinois contain PCB mixtures similar to Aroclor 1242 and Aroclor 1248 at concentrations in excess of 1,000 µg/gm. Dissolution and resuspension of PCBs from such highly contaminated sediments may produce significant concentrations of PCBs in the water entering the Harbor from Lake Michigan and runoff for many years. The importance of the resulting concentrations of PCBs in the Harbor water can be measured in part by determining the PCBs residues which accumulate in fish that are exposed to the water in the Harbor and the extent to which man is exposed to the PCBs by eating the contaminated fish.

There were two major objectives concerning the measurement of PCB residue accumulation in this study. The first was the validation of laboratory experiments with PCBs which led EPA to expect that the concentrations of PCBs in the Waukegan Harbor water would produce PCB residues in fish much in excess of the 5 µg/gm Food and Drug Administration action level for PCBs. These laboratory experiments showed that fish bioconcentrate PCBs directly from the water being pumped over the gills. The concentration of PCBs in the fish is dependent on the size of the fish, the rate at which water is pumped over the gills, and the length of time the fish is exposed to the water which contains PCBs.

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The second objective concerning the PCB residues in fish was estimating the rate at which PCBs can be eliminated from fish that enter the Harbor, become contaminated with PCB residues, and then move back to Lake Michigan. If the time required to eliminate PCBs from the fish tissue were on the order of a few days, it is unlikely that residues bioconcentrated during movements to the Harbor would significantly increase the PCB residues in Lake Michigan fish populations in the vicinity of Waukegan. However, if weeks or months were required to eliminate PCB residues after a few days of exposure to contaminated water, the Waukegan Harbor could significantly increase the PCB residues in Lake Michigan fish through normal fish movement.

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Methods

The experimental approach used in this study consisted of two similar live-cage experiments to expose fish to Waukegan Harbor water in an elimination experiment in which exposed fish from one of the experiments were then held in Lake Michigan water for nearly three months. The first live-cage exposure was conducted in October-November, 1978, and the details of the experiment are found in the "Outboard Marine Corp Biological Studies Report" by the Central Regional Laboratory dated February, 1979. Briefly, uncontaminated bluegills and yellow perch were placed into separate live cages in Slip #3 of the Waukegan harbor. Composite samples of five fish were removed from each live-cage after 2, 4, 8, 14, 21, and 28 days of exposure to the water and analyzed for PCB residues by the Central Regional Laboratory.

The second live-cage experiment consisted of a similar exposure of bluegills and yellow perch to Slip #3. However, fish samples were not removed from the cages during the exposure period. After 30 days in Waukegan harbor, composite fish were collected to measure the residues in the groups of fish and the remaining fish were placed in tanks at the municipal waterworks which received Lake Michigan water. Composite samples were removed for PCB analysis after 14, 28, 42, 56, 70 and 84 days in clean water. Details of the procedures for this experiment can be found in the "Waukegan Harbor Bioconcentration and Depuration Study, June 19, 1979-October 10, 1979" by the Central Regional Laboratory, December, 1979. On completion of the test, the fish samples were shipped frozen to the Environmental Research Laboratory-Duluth for analysis.

The whole fish composite samples were homogenized while frozen and aliquots of tissue homogenate were weighed, mixed with anhydrous Na_2SO_4 and placed in all-glass Soxhlet extractor thimbles. The samples were extracted for 8 hours using a 1:1 mixture of hexane and methylene chloride. The extracts were concentrated to 20 ml. A 2 ml aliquot was pipetted into a tarred weighing pan for gravimetric lipid determination. The remaining 18 ml was placed on a 20 gm Florsil column and eluted with 250 ml hexane. The eluant was adjusted to an appropriate volume for GC analysis.

The samples were initially analyzed by packed-column GC. The packed-column analyses were conducted on a Hewlett-Packard 5730-A gas chromatograph fitted with an auto sampler, a Ni^{63} electron capture detector, and a Hewlett-Packard lab automation data system. The column was 1.5%/1.95% SP-2250/SP-2401 coated on 100/120 mesh Supelcoport. Approximately 40 ml/min of 5% methane in argon was used as a carrier in the 2m x 2.3mm column. The temperature was programmed with a 4 min hold at 160°C, and 2°C/min increase to 200°C, followed by a 16 min hold at 200°C. The inlet and detector temperatures were 250°C and 300°C, respectively.

The PCB mixtures in the fish extracts produced packed-column chromatograms with approximately 16 major peaks. A computer program was prepared which measured the areas of 8 of the major peaks and calculated a mean concentration for the 8 peaks compared to Aroclor standards. Since the chromatograms indicated that the fish accumulated mixtures of PCBs similar to a mixture of Aroclor 1242 and Aroclor 1248 and PCB usage in Waukegan included a mixture of 1:2 Aroclor 1242/1248, standards used in this study were 1:1 and 1:2 mixtures of Aroclor 1242 and Aroclor 1248. Separate estimates based on each standard and expressed as total PCBs were made for each sample.

Results and Discussion

Bioconcentration is the result of competing processes in which the chemical is absorbed through the fish gills and eliminated from the body either by excretion or metabolism of the chemical. The rate of uptake is dependent on the rate at which water is pumped over the gills (i.e. species dependent) and the concentration of the chemical in the water. The rate at which a foreign chemical is eliminated from the body is often directly related to the concentration in the fish. Therefore, when fish contain small concentrations of a chemical, the elimination rate is small and the rate increases with increasing fish tissue concentration. When the bioconcentration process is measured by periodically measuring the fish tissue concentration, the concentration is expected to increase rapidly at first, with the rate of increase slowing and the tissue residue becoming larger. At the point where the tissue residue is large enough so that the elimination rate equals the rate of uptake, the tissue concentration remains constant. The fish is then considered to be at steady-state with respect to the chemical in the water.

The ratio of the concentration of a chemical in fish to the concentration in the water under steady-state conditions is called the bioconcentration factor for that chemical. The bioconcentration factor is a proportionality factor which relates the water concentration to the concentration of chemical in the fish. Therefore, if the concentration in the water is doubled, the residue in the fish can be expected to double also. For chemicals such as PCBs, which are very slowly eliminated from fish, the tissue concentration must be very large to reach a steady-state condition. The bioconcentration factor is likewise very large. Bioconcentration factors for PCBs may range from about 30,000 to over 100,000 depending on the PCB mixture and the species of fish tested. A bioconcentration factor of 30,000 means that fish exposed to 1 ppb (µg/l) PCB in the water can be expected to accumulate 30,000 ppb, or 30 ppm (µg/gm), in the fish tissue.

The initial live-cage study presented in the "Outboard Marine Corp Biological Studies Report" shows that the PCBs in Waukegan Harbor bioconcentrate in fish in a manner consistent with the above discussion. The study fish contained about 0.2 µg/gm PCBs when placed in the Harbor water. After two days in the water, the residues increased 6-10 times over the initial concentration. After three weeks the yellow perch contained an average of 19 µg/gm and the bluegills contained 16.9 µg/gm PCBs. After four weeks the yellow perch and bluegills contained 29.9 µg/gm and 19.7 µg/gm, respectively. The data also show that a steady-state condition was not reached in this study because the residues were still increasing with time at the end of the study. The study does not permit a direct measure of how great the PCB concentration can be expected to be in Waukegan Harbor; however, the data do demonstrate that the PCBs in Harbor water are biologically available and will bioconcentrate in fish which enter these waters.

The results of the live-cage and PCB elimination study are presented in Table 1. For comparison purposes, the results of the study will be discussed with respect to PCB estimates using a 1:1 mixture of Aroclor 1242/1248 as a standard. This will be followed by a discussion of the other estimates of total PCBs.

The data show that the bluegills initially contained 0.11 µg/gm PCBs and the yellow perch contained an average of 0.16 µg/gm PCBs. After the 30-day uptake

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phase in Waukegan Harbor water, the two species of fish contained averages of 20.1 µg/gm and 12.0 µg/gm PCBs, respectively. The PCB concentrations in the fish decreased throughout the elimination phase of the study (indicated by the dotted line in Table 1) to approximately 8 µg/gm in both species after 84 days in Lake Michigan water. The rate of elimination of PCBs from bluegills, which lost 59 percent of the residue in the 84 days, was faster than that of yellow perch which lost abou 32 percent in the 84 days.

As discussed previously, the absolute rate of elimination of many chemicals is dependent on the concentration in the fish. This is termed a first-order process. Evidence of a first-order process is obtained by plotting the concentration on a logarithmic scale versus time and determine if a linear or straight line is obtained. This plot for the elimination results is presented in Figure 1. Within the experimental variability, the data indicate that the approximation of elimination of PCBs being a first-order process is reasonable. The plot also gives some estimate how long it would take for the fish which migrate to Lake Michigan to reach the 5 µg/gm FDA action level, assuming the fish diet in Lake Michigan contained no PCBs. The data indicate that nearly 4.5 months would be required for the bluegills to reach 5 µg/gm and about six months for the yellow perch.

Implicit in the use of the first-order model for elimination of PCBs from fish is that the residue has a constant half-life in the fish under similar conditions. The half-life is the time required for the residue concentration to decrease by a factor of two. Therefore, if a fish contained 10 µg/gm PCBs, it would take one half-life for the residue to decrease to 5 µg/gm after the fish was placed in clean water, two half-lives to decrease to 2.5 µg/gm, and three half-lives to reach 1.25 µg/gm. Evidence for the constant half-life is shown in Figure 1.

Figure 1 also shows that the half-life for PCB elimination was greater for the perch than for the bluegills. One explanation of this is the half-life of residue elimination increases with increasing size of the fish. This may be due to the fact that the ratios of gill surface area/body weight and of metabolic rate/body weight decrease as the size of the fish increases (Hoar, W.S. and Randall, D. J., Fish Physiology, Academic Press 1970. p. 254.) This would be consistent with residues stored in large bodies having a smaller likelihood of being eliminated or metabolized than in small fish of the same species. If this principle were extrapolated to fish of the size eaten by fishermen, it would be expected that the half-life of PCBs in these larger fish would be substantially longer than measured for these small fish in this study. Lieb, et al (J. Agr. Food Chem. 22, 638-642, 1974) found no evidence of elimination or metabolism of Aroclor 1254 in rainbow trout during an eight-week elimination study which followed 32 weeks of feeding a PCB contaminated diet. Although the study by Lieb, et al (1974) cannot be directly extrapolated to this study, the data are consistent with the slower elimination of PCBs in larger fish.

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Table 1. Concentrations of PCBs in Fish from Bioconcentration and Depuration Study

Test Day	CRL-I.D.	mean weight (gm)[a]	mean length (cm)[a]	lipid content (percent)	Total PCB (µg/gm)-Mixed Std.-Packed Column Aroclor 1242/1248(1:1)	Aroclor 1242/1248(1:2)
			Bluegill - Composites of Exposed Fish			
0	BS-25-C01	1.7	4.5	8.53	0.11	0.10
0	BS-25-D01	1.7	4.4	8.39	0.11	0.11
30	GS-26-C01	3.7	5.7	8.31	18.3	19.8
30	GS-26-D01	3.5	5.1	7.85	21.9	23.3
44	GS-27-C01	2.6	5.0	7.25	18.0	19.2
58	GS-28-C01	8.3	5.2	6.55	14.6	15.3
58	GS-28-D01	6.6	4.8	4.76	14.0	14.4
72	GS-29-C01	2.5	5.0	5.63	12.3	12.8
86	GS-30-C01	4.1	5.1	5.44	10.2	10.6
86	GS-30-D01	3.8	5.1	6.34	11.6	12.1
100	GS-31-C01	3.1	5.7	5.28	9.3	9.6
114	GS-32-C01	3.6	5.6	5.49	8.3	8.4
114	GS-32-D01	4.8	5.4	5.41	8.0	8.1
			Bluegill - Composites of Control Fish			
30	BS-26-C01	3.7	5.3	7.72	0.32	0.32
30	BS-26-D01	4.3	5.1	6.30	0.34	0.37
58	BS-28-C01	5.6	6.1	6.62	0.50	0.51
58	BS-28-D01	6.9	5.6	5.80	0.44	0.47
86	BS-30-C01	3.9	5.8	7.02	0.56	0.67
114	BS-32-C01	6.1	5.9	6.78	0.60	0.62
114	BS-32-D01(A)	7.3	5.9	5.39	0.62	0.61
114	BS-32-D01(B)	7.3	5.9	8.59	0.69	0.70

[a] Taken from "Waukegan Harbor Bioconcentration and Depuration Study, June 19, 1979-October 10, 1979" by Central Regional Laboratory, U.S. EPA, December, 1979.

Table 1. Continued

Test Day	CRL-I.D.	mean weight (gm)[a]	mean length (cm)[a]	lipid content (percent)	Total PCB(µg/gm)-Mixed Std.-Packed Column Aroclor 1242/1248(1:1)	Aroclor 1242/1248(1:2)
				Yellow Perch - Composites of Exposed Fsh		
0	YS-25-C01	29.5	12.6	5.09, 5.30	0.14, 0.16[b]	0.12, 0.16
0	YS-25-D01	25.7	11.5	4.56, 5.10	0.13, 0.19	0.11, 0.18
30	PS-26-C01	19.0	11.6	3.75	10.6	11.2
30	PS-26-D01	24.4	11.2	3.39, 3.37	11.7, 13.6	12.2, 12.9
44	PS-27-C01	21.3	12.2	3.79, 3.89, 4.14	10.5, 11.6, 12.3	11.1, 11.8, 11.7
58	PS-28-C01	24.4	11.8	6.11, 5.30	11.0, 11.9	11.4, 11.2
72	PS-29-C01	19.5	11.9	4.76	9.1	9.4
86	PS-30-C01	17.4	11.3	3.25	9.2	9.4
100	PS-31-C01	15.5	11.2	3.87	8.0	8.1
114	PS-32-C01	15.0	10.6	4.13	8.3	8.6
				Yellow Perch - Composites of Control Fish		
30	YS-26-C01	25.0	11.8	3.94	0.26	0.23
30	YS-25-D01	24.3	11.3	5.20	0.31	0.29
58	YS-28-C01	29.9	12.7	4.54	0.38	0.39
58	YS-28-D01	26.7	11.5	4.81	0.49	0.51
86	YS-30-C01	21.0	11.9	4.73	0.63	0.64
114	YS-32-C01	28.7	12.8	3.66	0.42	0.42
114	YS-32-D01	22.5	19.9	2.84	0.50	0.50

b Replicate analyses

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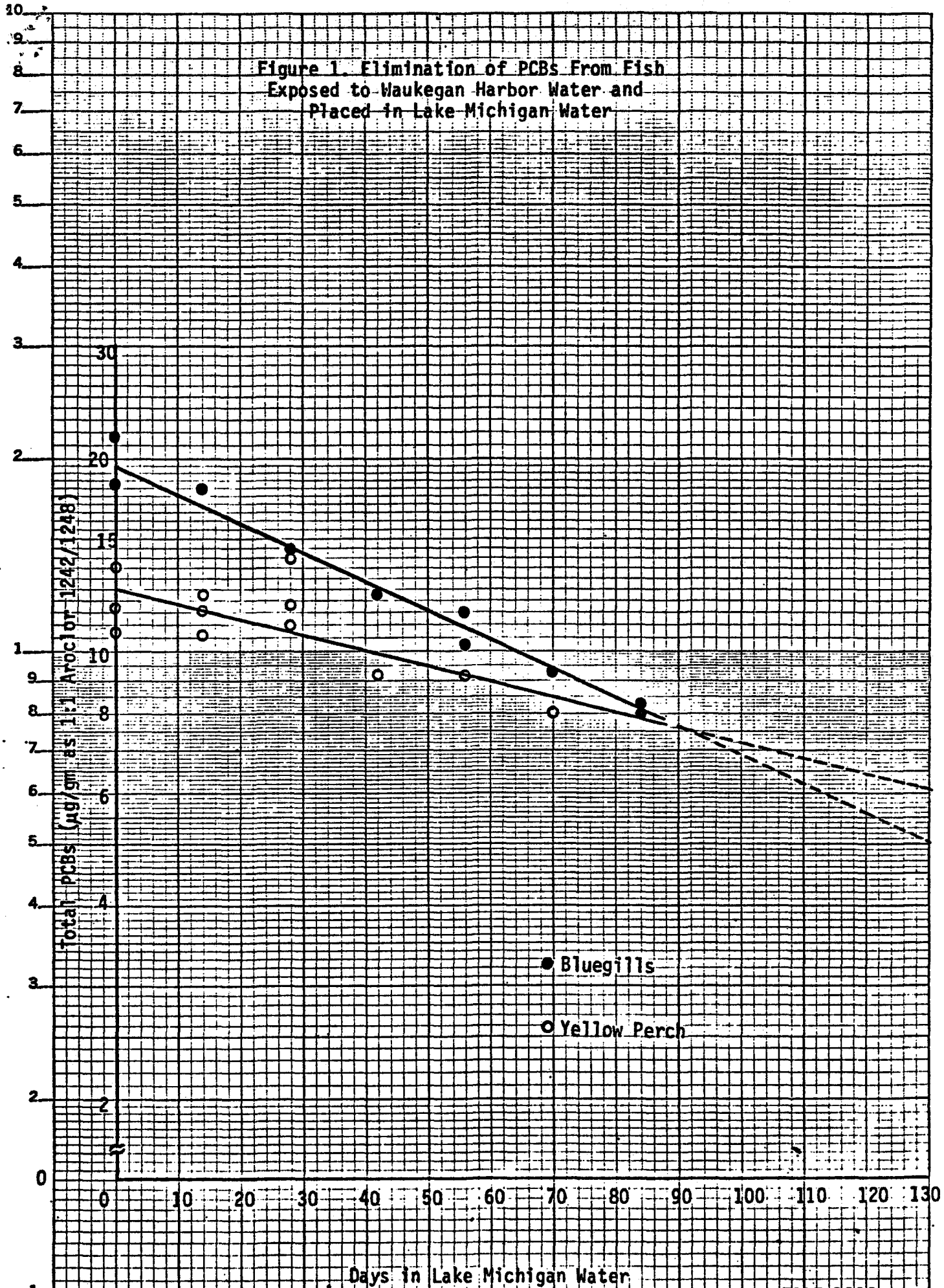


Figure 1. Elimination of PCBs From Fish Exposed to Waukegan Harbor Water and Placed in Lake Michigan Water

Semi-Logarithmic
2 Cycles x 10 to the Inch

EX P-3282
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