Accumulation and Elimination of Dieldrin in Muscle Tissue of Channel Catfish[1]

Larry R. Shannon
Iowa State University
11 Science Hall II
Ames, Iowa 50011

Abstract

Dieldrin accumulation and elimination in muscle tissue of channel catfish (Ictalurus punctatus) from water and food was determined in the laboratory. Twenty-eight-day exposure of fish 150-225 mm and 350-400 mm long to 75 parts per trillion (ng/liter) dieldrin resulted in the larger catfish consistently accumulating more dieldrin than the smaller fish. After 28 days of elimination, dieldrin levels in both size groups were nearly equal.

Catfish exposed to 2 ppm (mg/kg) dieldrin through their diets accumulated significantly more dieldrin in muscle than did fish exposed to 75 pptr in water. When fish were exposed to dieldrin both in food and water, dieldrin from both sources contributed to the total dieldrin load.

Large catfish accumulated more dieldrin from food and water than did smaller catfish. After 28 days of elimination, levels of dieldrin were not significantly different in muscles of 150-225 mm and 350-440 mm catfish exposed via both food and water.

Introduction

The pathway by which fish accumulate pesticides has been a matter for debate by researchers for some time. For example, Chadwick and Brocksen (1969) investigated the rates of accumulation of dieldrin from water and food by selected fish and fish food. They concluded that most dieldrin found in the fish tissues came from water. Murphy (1971) also found that guppies accumulated

1/ Journal Paper No. J-8373 of the Iowa Agriculture and Home Economics Experiment Station, Ames, Iowa. Project No. 1928. Financed by a grant from U.S. Department of Interior Office of Water Resources Research (Agreements 14-31-0001-3515, -3815, -4015) under Public Law 88-379 and made available through the Iowa State Water Resources Research Institute to the Iowa Cooperative Fishery Research Unit, which is sponsored by the Iowa State Conservation Commission, Iowa State University of Science and Technology, and the U.S. Fish and Wildlife Service.

Bulletin of Environmental Contamination & Toxicology,
Vol. 17, No. 6 © 1977 by Springer-Verlag New York Inc.

dieldrin in their tissues more efficiently from water than from food. In contrast, MACEK and KORN (1970) suggested that the food chain may be the major source of DDT residues in fish, whereas KLEINERT et al. (1968) stated that fish may pick up chlorinated hydrocarbon pesticides either by eating contaminated food or by direct absorption from water via the gills.

MURPHY (1971) sought to determine if body size played a role in rate and amount of DDT uptake. He found that small mosquito fish accumulated DDT from aqueous solutions faster than did large mosquito fish. According to MACEK and KORN (1970), DDT uptake from water by 150-mm brook trout was sufficiently slow to be of minor importance in accounting for the residues of DDT in great lakes salmonids. My study was conducted to determine if body size plays a role in the uptake from water and food and elimination of dieldrin by the edible portion of channel catfish (Ictalurus punctatus).

Materials and Methods

Channel catfish were obtained from two sources, but fish from only a single source were used in each experiment. Catfish of various sizes were collected from the Des Moines River below Fraser, Iowa, by means of baited hoop nets and electrofishing gear. Other fish were obtained from a commercial fish culturist. Fish were acclimated for two weeks in 380-liter tanks and fed an uncontaminated diet of Salmon Grower P49 obtained from Astra Pharmaceutical Products, Inc., Worcester, Mass. (SHANNON, 1974).

Water from the Iowa State University well was dechlorinated before it entered the tanks (SHANNON, 1974). Water temperature was maintained at $22 \pm 1^{\circ}C$.

Chemicals used for extractions contained less than 10 parts per trillion (pptr) total pesticides and therefore were suitable for pesticide determinations. All apparatus was washed and prerinsed with either petroleum ether or hexane. The technical dieldrin used was obtained from Shell Chemical Company and contained not less than 87% of 1, 2, 3, 4, 10, 10 hexachloro-6, 7-epoxy-1, 4, 4a, 5, 6, 7, 8, 8a-octahydro-1, 4-endo,exo-5, 8-dimethanonaphthalene (HEOD) and not more than 13% of insecticidally active related compounds (SHELL CHEMICAL, 1959). The aqueous solution of dieldrin was prepared by dissolving dieldrin in acetone before being mixed with water or food. Triton X-100, an organic solvent, was used in addition to acetone in the water-exposure experiments to reduce loss of dieldrin from solution. SEBA (1970) found that dieldrin crystallized out of solution and adhered to the walls of the container when tested in acetone alone but that effect was reduced in the presence of Triton X-100. The acetone and Triton X-100 levels (25 ppm and 1 ppm, respectively) were kept well below the lethal concentrations for fish.

To assure continuous exposure of the desired dieldrin level to the catfish, biweekly water samples were taken from the tanks. Samples of water from the test chambers were analyzed according to methods described by the U.S. ENVIRONMENTAL PROTECTION AGENCY (1970). Concentration of the dieldrin solution was checked by gas chromatographic analyses.

A dieldrin solution of 75 pptr was prepared for both experiments reported here. The desired dieldrin concentration was obtained by using a single diluter system to continuously meter the appropriate level of dieldrin into the test chamber. The diluter system was constructed of glass held together by silicon rubber.

The diet was prepared by thoroughly blending the commercial pellets with dieldrin dissolved in acetone and corn oil. Final dieldrin concentration in the food was 2 ppm. The acetone was allowed to evaporate from the food mixture, which was then frozen and fed to the fish once daily at the rate of 3% body weight. Food not treated with dieldrin was prepared and stored in the same manner. On the basis of field observations from the Des Moines River, these levels of dieldrin in food and water were considered the maximum concentrations to which channel catfish would be exposed in Iowa streams (BULKLEY et al. 1974). In each experiment, three samples of 2 small fish each and three samples of 1 large fish were taken each sampling period. Fish were fasted for 24 hr before each experiment.

Catfish dorsal muscle samples were analyzed according to the guidelines of the U.S. DEPARTMENT OF HEALTH, EDUCATION AND WELFARE (1970). For analysis, 10-40 g of dorsal muscle tissue were ground with 350 ml of 35% distilled water-acetonitrile solution in a Waring blender for 10 min. The samples were filtered through fluted no. 40 Whatman filter paper. The filtrate (260 ml) was transferred to a 1-liter separatory funnel, to which was added 100 ml of petroleum ether, and shaken vigorously for 2 min. Distilled water (600 ml) and a saturated saline solution (10 ml) were added to the samples and mixed thoroughly by vigorous tumbling action for 15 sec. The layers were allowed to separate, and the aqueous layer was discarded. The solvent layers were gently washed with two 100-ml portions of water. The washings were discarded, and the organic solvent layers each were filtered through a 50-mm column of anhydrous sodium sulfate into a 100-ml graduated cylinder. The volume of each was recorded.

The sample extracts were filtered through a 127-mm column of Florisil, which was topped by a 25-mm column of anhydrous sodium sulfate. Dieldrin was eluted from the column with 200 ml of 15% diethyl ether in petroleum ether following 200 ml of a 6% mixture of diethyl ether and petroleum ether. The two mixtures were collected in separate flasks and evaporated to 10 ml. The samples were then ready for injection into the chromatograph.

A Beckman GC-5 gas chromatograph was used to identify and quantify the dieldrin levels. An electron capture detector was used with a helium flow rate of 80 mm per min, a temperature of 180°C, and an attenuation of 2 x 10^{+3} on the 5% OV-210 column. A 4% SE-3/6/OF-1 column was used as a qualitative check with a gas flow rate of 120 mm per min, a temperature of 200 C, and an attenuation of 2 x 10^{+3}. Periodically throughout the study, known levels of aldrin were added to samples of fish muscle and water before chemical extraction was started. The portion of aldrin recovered in the extraction process ranged from 83 to 90%, with an average of 85%. Dieldrin extraction efficiency was assumed similar, even though the aldrin added was not incorporated within the muscle cells as was dieldrin.

Dieldrin extraction efficiency was also determined by water-exposing channel catfish to 10 ppb C^{14}-dieldrin for 6 hr (SHANNON, 1974). Stock meshes of catfish muscle containing dieldrin levels of 519±31 and 53.6±2.8 ppb were then prepared. Stock-mesh dieldrin levels were determined by direct counting of tissue samples as outlined by GAKSTATTER and WEISS (1967).

Three replicates of muscle tissue containing dieldrin levels of 5,190 ng, 1,072 ng, 107.2 ng, and 53.6 ng were prepared by varying sample sizes from the two stock meshes to cover the range of amounts in sample sizes of 10 to 40 g of tissue and concentrations encountered in this study. Extraction of dieldrin from the catfish tissue was performed in the same manner as was employed in the other experiments. True aliquots of these sample extracts were placed in scintillation vials containing 15 ml of BBOT scintillation cocktail for counting on a Packard Tri-Carb Scintillation Counter. Quenching was corrected for by internal standardization.

C^{14}-dieldrin recovery from catfish muscle ranged from 59.4% to 117.0%, with an overall mean of 84.2% recovery. An increase in percentage recovery was noted with utilization of larger gram-sample sizes containing greater dieldrin concentrations. Data presented in this study were not corrected for percentage recovery.

Three tanks were used to expose test fish. Fish in one tank were exposed to 75 pptr dieldrin in water; fish in a second tank were exposed to 2 ppm dieldrin in food, fish in a third tank were exposed to dieldrin in both food and water at concentrations of 2 ppm and 75 pptr, respectively. A similar tank contained control fish. Water flow through each tank replaced the volume every 12 hr.

Background samples were analyzed to determine the dieldrin concentrations in fish muscle before exposure. Test samples were then taken at the end of 7, 21, and 28 days during the uptake experiment, and for 7, 14, 21 and 28 days following 28 days of exposure during the elimination experiment.

Results and Discussion

Uptake from Water and Food

This experiment was designed to determine if dieldrin uptake would vary in catfish of different size exposed to dieldrin in food and water. Catfish of two length groups (150-225 mm and 350-400 mm) obtained from the Des Moines River were exposed to dieldrin in food, in water, and in food and water for 28 days. No dieldrin was detected in water of the tank in which fish were fed food containing 2 ppm dieldrin.

Average dieldrin content of muscle tissue before exposure was 14 ppb for small and 39 ppb for large catfish. After 28 days of exposure, the small catfish contained 544 ppb dieldrin resulting from food exposure, whereas catfish in the same size group exposed in water contained 175 ppb dieldrin (Table 1). The difference was significant at the 0.01 level of probability. Other catfish in this size group exposed to dieldrin both in food and water contained 898 ppb dieldrin, which was a significantly greater concentration (P=0.01) than attained by fish exposed to dieldrin either in food or in water alone.

Large catfish exhibited a pattern of dieldrin uptake similar to that of the smaller catfish. After 28 days of exposure, the large catfish contained 274 ppb dieldrin from water exposure, 1,243 ppb from food exposure, and 2,418 ppb from both food and water exposures. These differences among the large catfish were also significant at the 0.01 level of probability. In all three methods of exposure, large catfish accumulated greater concentrations (P=0.01) in muscle tissue than did small catfish. The reason larger catfish accumulated greater dieldrin concentrations could be explained by a study by BULKLEY et al. (1974). Their study indicated that larger fish have a greater capacity for accumulation and retention of dieldrin because of greater fat content.

Channel catfish accumulated dieldrin in muscle tissue both from water and from food exposure. At concentrations of dieldrin tested, more dieldrin was accumulated from food than from water. This relationship, under natural conditions, would undoubtedly change as the relative concentrations of dieldrin in food and water changed. When fish were exposed to dieldrin in both food and water, however, more dieldrin accumulated in muscle tissue than when fish were exposed to dieldrin in just food or water alone. These data agree with the suggestions of MACEK and KORN (1970), who claim that the food chain may be the major source of DDT in fish, and also with KLEINERT et al. (1968) who found that fish may accumulate chlorinated hydrocarbons from contaminated food or by absorption via gills.

TABLE 1.

Mean concentration (ppb) of dieldrin in dorsal muscle of channel catfish exposed to 75 pptr in water and 2 ppm in food. Standard deviation is in parentheses.

	Time (days) from beginning of exposure		
	7	21	28
Control			
150-225 mm	10(2)	12(4)	10(2)
350-400 mm	24(4)	18(4)	13(3)
Water			
150-225 mm	45(17)	81(10)	175(34)
350-400 mm	74(18)	179(24)	274(12)
Food			
150-225 mm	179(11)	506(28)	544(28)
350-400 mm	246(10)	796(37)	1243(103)
Food and water			
150-335 mm	303(26)	606(38)	898(93)
350-400 mm	280(5)	1199(35)	2418(374)

Elimination Following Exposure

Mean dieldrin contents before exposure were similar in small and large catfish (34 and 38 ppm, respectively). At the end of a 28-day exposure period, small fish contained 176 ppb dieldrin from water exposure, 287 ppb from food exposure, and 365 ppb from water plus food exposure (Table 2). Large catfish contained 185 ppb dieldrin after bath exposure, 302 ppb after food exposure, and 634 ppb from bath plus food exposure. Concentrations were not significantly different between large and small fish exposed to dieldrin in food and in water. By the end of the elimination period, differences were not significant between large and small fish from water, food or food-and-water exposure. Concentrations in water-exposed fish in both length groups had decreased to levels found in control fish. Thus, dieldrin concentrations after exposure in food and in water decreased at a similar rate among small and large fish; large fish exposed in food plus water, however, initially showed more rapid lowering of dieldrin concentrations than did small fish.

Elimination of dieldrin was fastest from bath exposure. Slightly over 50% of the dieldrin was eliminated from catfish muscle after 14 days, and by the end of 28 days, the dieldrin level was comparable to that found in the control fish and those exposed via water. Small and large fish exposed via food contained more than four times the concentration of dieldrin at the

end of 28 days than did those fish exposed via water only. Catfish exposed to dieldrin through diet and bath retained more than six times the dieldrin retained by fish exposed via water, and approximately 50% more than those exposed via food.

TABLE 2.

Mean concentration (ppb) of dieldrin in dorsal muscle of channel catfish for 28 days after exposure to 75 pptr in water and 2 ppm in food. Standard deviation is in parentheses.

	Time (days) from end of exposure period				
	0	7	14	21	28
Control					
150-225 mm	34(3)	28(2)	28(4)	18(3)	21(1)
350-400 mm	38(6)	29(6)	35(5)	21(9)	28(6)
Water					
150-225 mm	176(12)	152(11)	78(5)	47(6)	24(4)
350-400 mm	186(6)	162(18)	84(6)	59(7)	28(4)
Food					
150-225 mm	287(19)	217(7)	168(13)	152(18)	108(23)
350-400 mm	302(19)	229(6)	184(11)	142(27)	109(19)
Food and water					
150-225 mm	365(19)	241(9)	217(8)	195(23)	172(28)
350-400 mm	634(130)	358(18)	221(18)	194(18)	170(9)

The results of this study agree with those of GRZENDA et al. (1971) who found that goldfish lost about 50% of the accumulated dieldrin within two weeks following exposure through their diet. In summary, large and small catfish are able to accumulate dieldrin from both food and water sources, and the rate of uptake is dependent upon the level and source of exposure. Furthermore, the rate of elimination is determined by the source of the dieldrin. Dieldrin accumulated via water is more easily eliminated than that accumulated via food.

References

BULKLEY, R. V., L. R. SHANNON, and R. L. KELLOGG: Contamination of channel catfish with dieldrin from agriculture runoff. Iowa State Water Resources Research Institute Completion Report 62. Iowa State Univ. Ames. 144pp. 1974.

CHADWICK, G. G., and R. W. BROCKSEN: J. Wildl. Manage. 33, 693 (1969).

GAKSTATTER, J., and C. WEISS: Trans. Am. Fish. Soc. 96(3), 301 (1967).

GRZENDA, A. R., W. J. TAYLOR, and D. F. PARIS: Trans. Am. Fish. Soc. 100, 215 (1971).

KLEINERT, S. J., P. E. DEGURSE, and T. L. WIRTH: Occurrence and significance of DDT and dieldrin residues in Wisconsin fish. Wis. Dep. Nat. Resour. Tech. Bull. 41. 43pp. 1968.

MACEK, K. J. and S. J. KORN: J. Fish. Res. Board Can. 27, 1496 (1970).

MURPHY, P. G.: Bull. Environ. Contam. Toxicol. 6(1), 20 (1970).

SEBA, D. B.:Ph.D. thesis. Univ. of Miami (Library Congress Card No. Mic.-70-18167). 104pp. Univ. Microfilms, Ann Arbor, Mich. (1970).

SHANNON, L. R.: Ph.D. thesis. Iowa State Univ.(Library Congress Card No. Mic.-74-23766). 82pp. Univ. Microfilms, Ann Arbor, Mich. (1974).

SHELL CHEMICAL CORPORATION: Handbook of aldrin, dieldrin, and endrin formulations. The Woodrow Press, Inc. New York. 195pp. 1959.

U. S. DEPARTMENT OF HEALTH, EDUCATION AND WELFARE: Food and Drug Administration. Pesticide analytical manual. 1(212.1. U.S. Government Printing Office, Washington, D.C. 1970.

U. S. ENVIRONMENTAL PROTECTION AGENCY: Methods for organic pesticides for water and wastewater. Nat. Environ. Res. Center, Cincinnati, Ohio. 38pp. 1970.