Pesticides and Total Polychlorinated Biphenyls in Chinook Salmon and Carp Harvested from the Great Lakes: Effects of Skin-on and Skin-off Processing and Selected Cooking Methods

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Pesticides and total PCBs were determined in raw skin-on and skin-off chinook salmon harvested from Lakes Huron and Michigan as well as in carp fillets harvested from Lakes Erie and Huron and after baking, charbroiling, and canning salmon as well as pan and deep fat frying carp. Raw skin-off fillets had an average of less than 50% of the residues found in raw skin-on fillets. Cooked skin-off fillets were also found to have significantly lower residues than the cooked skin-on fillets. Cooking significantly reduced the DDT complex, dieldrin, hexachlorobenzene, the chlordane complex, toxaphene, heptachlor epoxide, and total PCBs contents. Few significant differences were found among cooking methods. Canning did significantly reduce DDE in salmon. Average losses of pesticides and total PCBs from the chinook salmon ranged from 30 to 41%. Similar average percentage losses were found for carp, ranging from 30 to 35% for the DDT complex, chlordane complex, and total PCBs, while the losses of HCB and dieldrin were greater than 40%.

Keywords: *Pesticides; PCBs; salmon; carp*

INTRODUCTION

Fish are an excellent source of protein and omega fatty acids, yet the fear of contaminant levels influences the use of this Great Lakes resource. It is essential to know the level of contaminants in processed/cooked fish as eaten by sportsfishermen and their families at the dinner table as well as to develop procedures to reduce contaminant levels in fish. Tourism, the charter boat fishing industry, and Native American fisheries are all adversely affected by consumers' fear of contaminants in Great Lakes fish. Risk assessment should take into account residue levels as eaten. Species important to urban ethnic minorities as well as sportsfishermen should be studied.

The Environmental Protection Agency (1992) recently published data from a national study of chemical residues in fish. At more than half the sites, fish had detectable levels of *p,p*-DDE, biphenyl, mercury, total PCBs, *trans*-nonachlor, pentachloroanisole, *cis*- and *trans*-chlordane, dieldrin, α-BHC, and 1,2,4-trichlorobenzene. Many of the fish with the highest level of contamination were from sites in the Great Lakes basin.

Preparation and cooking techniques have resulted in varying loss of PCBs and other organic toxicants from certain species of fish (Zabik et al., 1978, 1979; Armbruster et al., 1987; Sanders and Hayes, 1988). Trotter et al. (1989) reported PCBs were an average of 27% lower in blue fish after cooking and removal of the skin and oil drippings. In a study by Zabik (1974) that evaluated PCB levels in chicken, stewing or pressure cooking chicken pieces reduced PCBs by 50–70%, while half of the organochlorine residues transferred to the cooking media. Zabik (1984) reviewed early studies on the effect of cooking/processing on residue levels in meat, fish, and poultry. Nevertheless, cooking was not effective in reducing PCBs from carp (Zabik et al., 1982).

To assess the potential reduction of environmental contaminants from several species of Great Lakes fish using sizes and locations for harvesting fish common to those of typical sportsfishermen, a comprehensive study was conducted to assess the level of contaminant consumption at the dinner table. This paper will present data related to the effect of processing and cooking on the levels of organochlorine pesticides and total PCBs in skin-on and skin-off chinook salmon *(Oncorhynchus tshawytscha)* harvested from Lakes Huron and Michigan as well as in carp (*Cyprinus carpio*) fillets harvested from Lakes Erie and Huron. Chinook salmon are at the high end of the food chain and are prized as sportsfish, while carp are typical bottom-feeders. Fillets were cooked skin-on and skin-off since chlorinated contaminants are fat soluble and found in higher levels in the fat associated with the lateral line and skin (Zabik et al., 1978; Hora, 1981; Sanders and Haynes, 1988).

EXPERIMENTAL PROCEDURES

All fish were procured for the study in the 1991 season. The source and size of fish were chosen on the basis of the mean Creel census data from sportsfisherman for 1990 (Rakoczy, 1991, 1992).

Processing of the Fish. Fish were processed according to recommendations to sportsfishermen for trimmed skin-on or skin-off fillets. Skin-on fillets had the belly flap trimmed off, while skin-off fillets had the belly flap as well as lateral line and associated fat tissue removed. Process data given in

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Table 1. Analytical Parameters for Pesticides and PCBs

eluate	analytes	GC column	N_2, mL/min	oven parameters
SG-1	HCB, mirex	1.5% OV-17/1.95% OV-210	20	198 °C isothermal
SG-3	*cis*-Nonachlor			
	oxychlordane			
	heptachlor epoxide			
SG-2	PCB	3% SE-30	30	5 min 160 °C, 17 min at 3 °C/min, 10 min final hold at 210 °C
	p,p'-DDE			
SG-3	*p,p'*-DDT	4% SE-30/6% OV-210	20	198 °C isothermal
SG-3	*p,p'*-DDE	3% SE-30	30	5 min 160 °C, 17 min at 3 °C/min, 10 min final hold at 210 °C
	B-BHC			
	toxaphene			
	chlordane			
	trans-nonachlor			

Table 2. Comparison of the Level of Selected Pesticides and PCBs Expressed as Parts per Million of Wet Tissue in Raw Skin-on and Skin-off Chinook Salmon and Carp Fillets Harvested from Lakes Erie, Huron, and Michigan to the Pesticides's Action Level

pesticide/PCB	fish	skin on/off	Lake Erie	Lake Huron	Lake Michigan	action level
chlordane complex[a]	chinook salmon carp	on		0.282	0.246	0.3
		off		0.144	0.147	
		on	0.065	0.031		
		off	0.023	0.019		
DDT complex[b]	chinook salmon carp	on		0.83	0.75	5
		off		0.35	0.41	
		on	0.24	0.18		
		off	0.09	0.07		
dieldrin	chinook salmon carp	on		0.06	0.118	0.3
		off		0.039	0.072	
		on	0.032	0.027		
		off	0.012	0.005		
toxaphene	chinook salmon carp	on		0.41	0.34	5
		off		0.23	0.22	
		on	ND[c]	ND		
		off	ND	ND		
total PCBs	chinook salmon carp	on		1.39	1.34	2
		off		0.6	1.03	
		on	2.51	1.27		
		off	0.75	0.48		

[a] α-Chlordane, γ-chlordane, oxychlordane, *cis*-nonachlor, and *trans*-nonachlor. [b] *p,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD. [c] ND, not detected.

the supplementary material include carcass yield, which is based on the deheaded and degutted weight of each fish species, as well as as-prepared (AP) yield.

Right fillets were analyzed raw and left fillets were frozen in foil overwrapped with polyethylene films at −23 °C until cooked. Raw samples were ground with dry ice to pulverize the sample before being placed in glass bottles and frozen as above.

Cooking of the Fish. Chinook salmon were cooked by baking and charbroiling. In addition, skin-off chinook salmon fillets were canned. Since increasing surface area had been effective in reducing dioxins (Stachiw et al., 1988), both skin-on and skin-off chinook salmon fillets were also scored 1 cm × 1 cm × ~0.3 cm before charbroiling. Carp fillets were pan fried according to the procedure of Puffer and Gossett (1983). Baking and charbroiling salmon were carried out as described by Stachiw et al. (1988). Carp were deep fat fried as outlined in the study of Morehouse and Zabik (1989). Canning followed standard USDA (1988) procedures. All fish were cooked to an internal temperature of 80 °C. Cooked samples contained only muscle tissue for all cooking methods except deep fat frying. Deep fat fried skin-on fillets included the skin as well as the muscle tissue. All cooking loss data are included in the supplementary material. Cooked muscle tissues were homogenized with an Osterizer blender prior to being stored in a glass bottle at −23 °C.

Materials. Distilled-in-glass isooctane and ethyl ether (2% ethanol as preservative) were purchased from Burdick and Jackson. All other solvents were either Baker Analyzed or MCB OmniSolv reagents. Anhydrous, granular Na_2SO_4 (Mallinckrodt 8024), 60−100 mesh Florisil (PR grade, Floridin Co.), and 70−230 mesh silica gel 60 (E. Merck 7734) were activated for at least 24 h at 130 °C. The U.S. Environmental Protection Agency (Health Effects Research Laboratory, Research Triangle Park, NC) supplied the following analytical reference standards: Aroclor 1254 (PRL-FDA lot 1543), Aroclor 1260 (PRL-FDA lot 1544), β-BHC, α- and γ-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, heptachlor epoxide, dieldrin, HCB, mirex, and toxaphene. Chemical names of these compounds are given in *Analytical Reference Standards* (Watts, 1981). All GC columns were 183 cm × 2 mm i.d. glass packed as outlined in Table 1 and obtained from Applied Science Laboratories Inc., State College, PA.

Pesticides and Total PCB Analyses. Pesticides and total PCBs were determined using packed column electron capture gas chromatographic analyses as outlined by Price et al. (1986). After the fish sample was ground with anhydrous Na_2SO_4 and the mixture placed on a chromatography column, lipid was extracted with 200 mL of 50% ethyl ether/petroleum ether (v/v) at a flow rate of 3−5 mL/min and placed in a tared beaker to obtain the fat content of the fish. An aliquot (0.5 g) of the lipid was dissolved in 5 mL of hexane and cleaned up on a Florisil column using 200 mL of 6% ethyl ether/petroleum ether (v/v) at a flow rate of 5 mL/min. This was followed by an elution with 200 mL of 20% ethyl ether/petroleum ether (v/v). Both fractions were concentrated to less than 1 mL in a Kuderna-Danish concentrator.

This concentrate was placed on a silica gel 60 column. After three 1 mL hexane rinses followed by discard, the column was eluted with 15 mL of hexane, which was collected as fraction SG-1, followed by an additional 20 mL elution with hexane (fraction SG-2). The column was then eluted with 20 mL of benzene (fraction SG-3). Each fraction was reduced to ca. 0.5

Table 3. Pesticides and Total Polychlorinated Biphenyls (PCBs) Expressed as Micrograms per Fillet in Skin-on and Skin-off Raw, Baked, Charbroiled, Scored and Charbroiled, or Canned Fillets of Chinook Salmon Harvested from Lake Huron

	skin-on		skin-off	
compound	raw	cooked	raw	cooked
		Baked		
p,p'-DDT	36.1 ± 15.86	17.76 ± 0.20	10.39 ± 4.28	6.68 ± 3.23
p,p'-DDE	267.9 ± 142.7	175.3 ± 86.9	94.9 ± 27.7	50.2 ± 14.1
p,p'-DDD	24.67 ± 13.21	17.95 ± 6.86	6.43 ± 1.98	6.20 ± 3.96
α-chlordane	22.50 ± 15.40	15.02 ± 8.72	6.33 ± 3.02	3.10[a]
γ-chlordane	8.33 ± 5.85	5.57 ± 3.55	2.78 ± 1.07	1.59 ± 0.35
oxychlordane	10.35 ± 7.02	6.01 ± 2.65	3.41 ± 1.54	1.69 ± 0.66
cis-nonachlor	20.90 ± 8.02	14.51 ± 3.92	8.03 ± 3.51	4.37 ± 1.54
trans-nonachlor	75.89 ± 30.65	43.30 ± 13.31	22.96 ± 9.43	13.65 ± 6.07
HCB	3.15 ± 0.90	1.63 ± 0.44	1.06 ± 0.27	0.42 ± 0.16
dieldrin	31.40 ± 25.29	19.57 ± 16.86	9.41 ± 4.57	6.09 ± 1.92
heptachlor epoxide	6.58 ± 4.24	4.24 ± 3.24	2.01 ± 0.75	1.12 ± 0.15
toxaphene	233.1 ± 12.6	145.4 ± 5.2	67.48 ± 22.43	34.24 ± 10.54
total PCBs	691.3 ± 416.0	352.2 ± 89.0	185.7 ± 93.6	102.6 ± 42.1
		Charbroiled		
p,p'-DDT	41.22 ± 9.89	26.76 ± 6.65	17.34 ± 6.16	8.69 ± 1.27
p,p'-DDE	298.5 ± 90.1	175.2 ± 12.5	137.7 ± 41.4	53.8 ± 3.0
p,p'-DDD	26.49 ± 2.98	16.04 ± 3.10	12.00 ± 5.77	4.56 ± 0.27
α-chlordane	23.23 ± 0.85	12.91 ± 1.67	11.56 ± 4.14	4.33 ± 0.55
γ-chlordane	7.68 ± 0.64	4.76 ± 0.32	2.79 ± 0.21	1.46 ± 0.21
oxychlordane	10.82 ± 4.07	4.11 ± 2.71	4.03 ± 1.53	1.65 ± 0.14
cis-nonachlor	23.51 ± 5.38	12.93 ± 5.42	9.98 ± 4.50	3.85 ± 0.55
trans-nonachlor	60.50 ± 19.87	33.33 ± 15.94	36.75 ± 11.68	14.26 ± 2.49
HCB	2.83 ± 0.73	1.49 ± 0.57	1.13 ± 0.24	0.58 ± 0.13
dieldrin	32.38 ± 4.71	18.83 ± 2.41	14.55 ± 1.76	7.12 ± 1.42
heptachlor epoxide	7.28 ± 0.68	4.02 ± 0.57	2.50 ± 0.69	1.13 ± 0.12
toxaphene	202.56 ± 79.96	120.06 ± 43.58	89.60 ± 35.24	47.18 ± 25.95
total PCBs	704.2 ± 60.0	425.6 ± 69.8	305.6 ± 143.5	115.1 ± 16.6
		Scored and Charbroiled		
p,p'-DDT	59.29 ± 16.13	24.75 ± 10.69	14.08 ± 6.88	5.79 ± 1.63
p,p'-DDE	451.7 ± 152.9	186.1 ± 47.1	65.4 ± 24.1	31.8 ± 13.1
p,p'-DDD	40.40 ± 13.47	18.53 ± 4.29	6.43 ± 2.37	2.79 ± 0.28
α-chlordane	33.59 ± 15.47	12.36 ± 4.21	4.85 ± 2.67	2.24 ± 1.29[b]
oxychlordane	16.06 ± 5.93	8.23 ± 2.11	2.54 ± 1.01	1.26 ± 0.23
cis-nonachlor	33.02 ± 10.88	16.88 ± 5.49	6.43 ± 2.37	3.23 ± 0.95
trans-nonachlor	138.8 ± 32.2	50.67 ± 7.08	19.64 ± 8.46	9.07 ± 5.76
HCB	4.13 ± 2.00	2.06 ± 0.46	1.04 ± 0.34	0.56 ± 0.06
dieldrin	45.09 ± 17.80	17.17 ± 3.22	6.44 ± 1.66	3.69 ± 0.20
heptachlor epoxide	8.24 ± 3.04	3.58 ± 1.17	1.61 ± 0.50	0.71 ± 0.09
toxaphene	297.1 ± 89.3	130.1 ± 65.3	77.66 ± 31.00	40.01 ± 15.75
total PCBs	1106.7 ± 319.6	428.3 ± 97.3	152.7 ± 45.8	72.9 ± 24.7
		Canned		
p,p'-DDT			10.01 ± 2.84	1.96 ± 0.12
p,p'-DDE			75.6 ± 15.9	46.7 ± 13.0
p,p'-DDD			6.13 ± 2.41	6.59 ± 1.07
α-chlordane			6.28 ± 4.89	3.07 ± 1.02[b]
γ-chlordane			2.18 ± 0.84	1.21 ± 0.41
oxychlordane			2.29 ± 0.69	1.22 ± 0.17
cis-nonachlor			7.14 ± 2.77	3.88 ± 1.41
trans-nonachlor			22.06 ± 13.02	10.35 ± 3.18
HCB			0.67 ± 0.05	0.49 ± 0.03
dieldrin			7.53 ± 2.62	0.90 ± 0.11
toxaphene			54.41 ± 16.95	12.32 ± 0.82
total PCBs			146.8 ± 41.5	98.5 ± 35.1

[a] Only one fillet had value > minimum detectable level. [b] Two samples had values > minimum detectable level.

mL using a micro-Snyder column on a water bath and analyzed by electron capture gas chromatography according to the paramenters listed in Table 1. The gas chromatograph/data system was a Varian 3700 equipped with a constant current ^{63}Ni electron capture detector and Varian 8000 AutoSamplers. Pesticides and PCBs were expressed as parts per million of wet tissue.

Solids were determined using AOAC Method 24.002 (AOAC, 1984). The micrograms of contaminants were calculated from the sample weight of the cooking data times the ppm wet weight and used to determine the percent change. Values for pesticides which were below the reported detection limit, i.e. ND, were not included in the averages or standard deviations.

The following gives the levels of detection: *p,p'*-DDT, 0.005 ppm; *p,p'*-DDE, 0.003 ppm; *p,p'*-DDD, 0.005 ppm; α-chlordane, 0.003 ppm; γ-chlordane, 0.003 ppm; oxychlordane, 0.003 ppm; *cis*-nonachlor, 0.003 ppm; *trans*-nonachlor, 0.003 ppm; HCB, 0.001 ppm; dieldrin, 0.005 ppm; heptachlor epoxide, 0.003 ppm; toxaphene, 0.050 ppm; total PCBs, 0.025 ppm. Ten percent of the samples were run in duplicate. Variability of the pesticide and total PCB analyses was as follows: *p,p'*-DDT, 11.8 ± 12.0%; *p,p'*-DDE, 7.8 ± 10.5%; *p,p'*-DDD, 6.8 ± 6.5%; α-chlordane, 10.5 ± 9.6%; γ-chlordane, 13.6 ± 16.9%; oxychlordane, 5.5 ± 9.3%; *cis*-nonachlor, 4.5 ± 5.3%; *trans*-nonachlor, 14.4 ± 16.8%; HCB, 4.3 ± 8.7%; dieldrin, 4.6 ± 6.5%; heptachlor epoxide, 21.8 ± 49.8%; toxaphene, 6.5 ± 6.7%; total PCBs, 6.6 ± 11.3%. Data were analyzed for variance using a general linear model with SAS (1986). Tukey's test was used to sort out significant differences of individual means at $p < 0.05$.

Table 4. Pesticides and Total Polychlorinated Biphenyls (PCBs) Expressed as Micrograms per Fillet in Skin-on and Skin-off Raw, Baked, Charbroiled, Scored and Charbroiled, or Canned Fillets of Chinook Salmon Harvested from Lake Michigan[a]

compound	skin-on raw	skin-on cooked	skin-off raw	skin-off cooked
		Baked		
p,p′-DDT	38.54 ± 20.63	29.68 ± 19.60	9.24 ± 2.79	6.88 ± 3.49
p,p′-DDE	266.3 ± 144.3	224.3 ± 127.4	71.5 ± 19.4	52.6 ± 22.5
p,p′-DDD	29.08 ± 12.75	26.13 ± 11.02	7.34 ± 2.87	5.12 ± 3.27
α-chlordane	20.53 ± 10.70	14.61 ± 6.32	5.58 ± 3.42	3.85 ± 2.82
γ-chlordane	8.32 ± 5.00	6.02 ± 3.14	2.14 ± 1.04	1.66 ± 1.13
oxychlordane	12.25 ± 6.38	11.03 ± 4.49	3.53 ± 1.47	2.25 ± 1.44
cis-nonachlor	22.19 ± 8.67	15.00 ± 3.99	5.45 ± 1.75	3.64 ± 2.27
trans-nonachlor	65.76 ± 32.44	47.14 ± 19.23	15.47 ± 4.28	11.11 ± 5.60
HCB	3.71 ± 1.86	2.45 ± 1.28	0.91 ± 0.48	0.66 ± 0.45
dieldrin	70.61 ± 37.86	56.07 ± 30.28	17.39 ± 7.54	13.00 ± 7.81
heptachlor epoxide	16.09 ± 8.81	13.83 ± 7.60	4.30 ± 2.30	2.91 ± 2.08
toxaphene	181.5 ± 69.4	168.7 ± 72.2	47.96 ± 25.41	37.18 ± 21.02
total PCBs	754.9 ± 358.4	566.2 ± 267.9	168.4 ± 53.6	119.4 ± 57.1
		Charbroiled		
p,p′-DDT	53.43 ± 23.01	27.87 ± 8.61	20.26 ± 5.56	15.67 ± 3.27
p,p′-DDE	598.8 ± 433.0	354.1 ± 334.6	145.6 ± 43.0	102.6 ± 27.8
p,p′-DDD	42.29 ± 24.00	22.12 ± 12.49	13.09 ± 2.82	10.47 ± 2.43
α-chlordane	30.07 ± 16.74	17.29 ± 9.75	10.60 ± 3.50	7.72 ± 1.67
γ-chlordane	12.53 ± 6.77	7.12 ± 3.12	4.58 ± 1.18	3.23 ± 0.67
oxychlordane	17.49 ± 10.02	9.37 ± 5.31	5.47 ± 1.18	4.30 ± 1.43
cis-nonachlor	32.32 ± 17.21	16.34 ± 8.66	10.97 ± 2.84	7.57 ± 1.57
trans-nonachlor	86.69 ± 52.43	49.02 ± 26.99	27.58 ± 6.26	21.85 ± 4.98
HCB	4.86 ± 2.96	2.29 ± 1.43	1.61 ± 0.30	0.97 ± 0.16
dieldrin	96.95 ± 59.71	58.69 ± 36.29	28.73 ± 4.62	25.23 ± 1.87
heptachlor epoxide	22.66 ± 13.77	11.89 ± 7.46	6.85 ± 0.94	4.87 ± 1.37
toxaphene	253.5 ± 130.9	169.2 ± 92.7	88.44 ± 29.37	74.61 ± 18.79
total PCBs	964.4 ± 566.1	542.7 ± 336.2	353.3 ± 39.0	238.3 ± 68.4
		Scored and Charbroiled		
p,p′-DDT	77.29 ± 35.42	35.68 ± 12.51	22.76 ± 11.77	12.59 ± 2.55
p,p′-DDE	429.3 ± 60.4	277.4 ± 56.4	149.2 ± 59.6	98.5 ± 13.3
p,p′-DDD	37.27 ± 9.29	24.75 ± 8.24	14.61 ± 5.53	8.44 ± 0.92
α-chlordane	30.30 ± 10.03	16.26 ± 2.18	11.45 ± 3.88	6.95 ± 0.20
γ-chlordane	14.18 ± 6.71	7.46 ± 1.83	4.50 ± 1.33	3.06 ± 0.70
oxychlordane	15.79 ± 2.93	10.35 ± 1.42	5.12 ± 2.04	3.41 ± 0.15
cis-nonachlor	34.48 ± 12.11	16.80 ± 7.38	11.24 ± 4.51	6.65 ± 0.64
trans-nonachlor	94.00 ± 17.95	59.12 ± 8.51	37.74 ± 20.84	21.04 ± 3.53
HCB	4.09 ± 0.44	2.82 ± 0.24	1.67 ± 0.47	0.95 ± 0.22
dieldrin	75.44 ± 27.08	43.38 ± 16.51	29.50 ± 8.94	17.47 ± 7.54
heptachlor epoxide	17.40 ± 1.82	10.11 ± 1.93	5.54 ± 1.76	3.80 ± 2.04
toxaphene	236.8 ± 47.1	148.9 ± 18.9	91.59 ± 34.09	71.11 ± 25.09
total PCBs	1020.1 ± 191.5	645.1 ± 180.5	490.0 ± 171.1	273.1 ± 68.3
		Canned		
p,p′-DDT			28.30 ± 4.34	68.16 ± 1.26
p,p′-DDE			209.6 ± 23.8	133.0 ± 22.9
p,p′-DDD			16.11 ± 1.14	21.52 ± 6.53
α-chlordane			14.18 ± 1.59	9.18 ± 2.00
γ-chlordane			5.96 ± 1.00	3.90 ± 0.73
oxychlordane			6.83 ± 0.82	4.80 ± 1.08
cis-nonachlor			13.70 ± 0.70	9.85 ± 2.41
trans-nonachlor			48.23 ± 8.48	27.45 ± 3.75
HCB			1.62 ± 0.16	1.09 ± 0.21
dieldrin			27.18 ± 8.72	15.58 ± 4.23
heptachlor epoxide			4.76 ± 0.99	3.42 ± 1.17
toxaphene			93.39 ± 3.19	26.35 ± 1.73
total PCBs			493.4 ± 55.3	303.2 ± 66.3

[a] $n = 3$.

RESULTS AND DISCUSSION

Lake Huron chinook salmon ranged in age from 3 to 5 years with a mean age of 3.8 ± 0.7 years. Chinook salmon from Lake Michigan ranged in age from 2 to 4 years and averaged 2.7 ± 0.6 years of age. Males were 67 and 63% of the Lake Huron and Lake Michigan chinook salmon, respectively.

Carp harvested from Lake Erie were aged as 3.5 ± 1.3 years with a range in age from 3 to 5 years. Lake Huron carp had a greater age range of 2−7 years, and the mean age was 3.2 ± 1.3 years. Males and females were equally distributed in carp from both lakes.

The length and weight of the chinook salmon harvested from Lake Huron [80.8 cm; 5.7 kg (12.5 lb)] were significantly greater than those from Lake Michigan [76.0 cm; 5.0 kg (11 lb)]. Thus, these salmon were the same or very slightly greater in weight than those in Creel data (Rakoczy, 1991, 1992). Carp from Lake Erie were significantly longer (51.8 cm) than those from Lake Huron (46.6 cm). Nevertheless, there were no significant differences in carp weight. The average carp

Table 5. Comparison of the Level of Pesticides and Total PCBs Expressed as Parts per Million of Solids in Raw and Cooked Chinook Salmon

component	raw	cooked
p,p'-DDT	0.240	0.136
p,p'-DDE	1.714	1.099
p,p'-DDD	0.150	0.113
α-chlordane	0.120	0.071
γ-chlordane	0.048	0.032
oxychlordane	0.062	0.040
cis-nonachlor	0.130	0.027
trans-nonachlor	0.418	0.148
HCB	0.017	0.011
dieldrin	0.251	0.171
heptachlor epoxide	0.054	0.037
toxaphene	1.116	0.700
total PCBs	2.934	1.989

Table 6. Comparison of the Level of Pesticides and Total PCBs Expressed as Parts per Million of Wet Tissue in Skin-on and Skin-off Cooked Chinook Salmon

component	skin-on	skin-off
p,p'-DDT	0.062	0.032
p,p'-DDE	0.489	0.272
p,p'-DDD	0.046	0.031
α-chlordane	0.028	0.018
γ-chlordane	0.013	0.009
oxychlordane	0.018	0.010
cis-nonachlor	0.035	0.020
trans-nonachlor	0.106	0.062
HCB	0.005	0.003
dieldrin	0.074	0.044
heptachlor epoxide	0.017	0.009
toxaphene	0.325	0.163
total PCBs	1.072	0.624

ranged in weight from 4 lb for Lake Erie to 3.5 lb for Lake Huron and so were comparable to Creel data.

PCB and Pesticide Analyses. The DDT complex (*p,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD), dieldrin, hexachlorobenzene (HCB), α- and γ-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, toxaphene, heptachlor epoxide, and total PCBs (expressed as Aroclor 1254) were found at above the minimum level of detection for both of the species studied.

Table 2 presents the average levels of the DDT complex found in the raw skin-on and skin-off chinook salmon and carp fillets as compared to the action level for DDT. The average level of the DDT complex for all skin-off fillets was less than 0.5 ppm, while most values for skin-on fillets were also below 0.5 ppm or less than 10% of the action level. Total DDT compounds in skin-on chinook salmon were between 0.5 and 1 ppm. Skin-on carp from both Lake Erie and Lake Huron had 2–3 times the level of the DDT complex of the skin-off carp fillets but were still less than 5% of the action level. DDE was the major constituent in the DDT complex.

Values of the chlordane complex (α-chlordane, γ-chlordane, oxychlordane, *cis*-nonachlor, and *trans*-nonachlor) found in the raw fish are also shown in Table 2. Skin-on chinook salmon from Lakes Huron and Michigan had individual fish that exceeded the action level. Removing the skin brought the total level of the chlordane complex below the FDA action level. The FDA indicated that *cis*-nonachlor and *trans*-nonachlor should be included in the total chlordane value when the quantitation is based on individual isomers. *trans*-Nonachlor was the major constituent of the chlordane complex even though *cis*-nonachlor and *trans*-nonachlor are minor constituents of technical chlordane.

Dieldrin was found in all fish (Table 2). Skin-on chinook salmon and carp had dieldrin levels at least twice those in skin-off chinook salmon and carp. Only skin-on chinook salmon from Lake Michigan had an average dieldrin level greater than 0.1 ppm. Skin-on chinook salmon from Lake Huron and skin-off chinook salmon from Lake Michigan had dieldrin levels between 0.05 and 0.1 ppm, while the average dieldrin levels in the other fish fillets were below 0.05 ppm.

Low levels of toxaphene were found in all chinook salmon, but the level of toxaphene in the carp was below the limit of detection. Total PCBs in raw fish fillets are show in Table 2. The average level of PCBs in skin-on carp fillets from Lake Erie exceeded the 2 ppm action level. Three of the six fish had PCBs above 2 ppm, one as high as 6 ppm. The latter was the largest fish, but the other two fish with high PCBs were average in weight. Skin-on carp fillets from Lake Huron and skin-on chinook salmon from both Lakes Michigan and Huron had average PCB levels that exceeded 1 ppm.

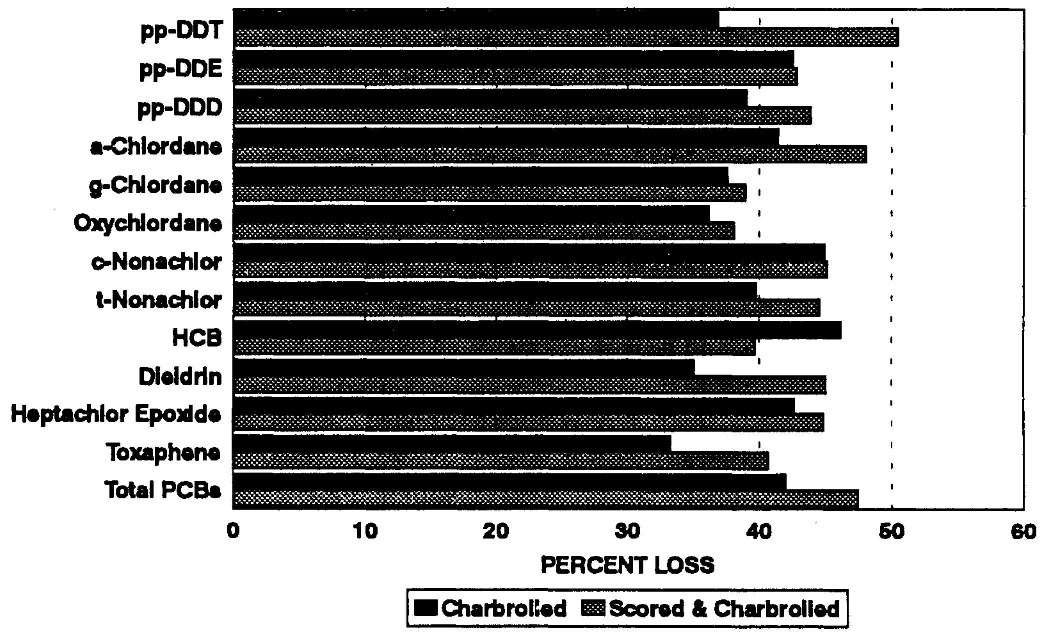

Figure 1. Effect of scoring on loss of pesticides and total PCBs from chinook salmon.

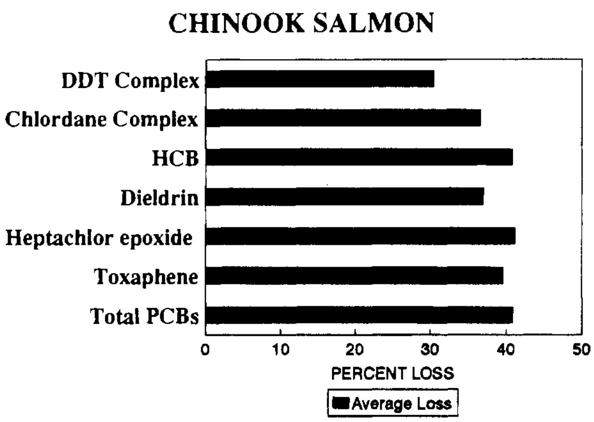

Figure 2. Average percentage losses of pesticides and total PCBs from chinook salmon from Lakes Huron and Michigan cooked by baking, charbroiling and canning.

Skin-off chinook salmon fillets from Lake Michigan also averaged 1 ppm of PCBs. These values are comparable to levels found in carp from the Michigan watersheds (0.8–2.7 ppm) in analyses of whole carp (EPA, 1992).

As had been reported in other studies (Hora, 1981; Sanders and Haynes, 1988), removing the skin and lateral line reduced the level of pesticides and total PCBs in both the raw chinook salmon and the carp as compared to skin-on fillets with only the belly-flap fat removed. This resulted in a significant difference in fat content as ANOVA established the fat content of the skin-on chinook salmon (7.8%) to be significantly higher than that of the skin-off (3.9%). Fat content of the skin-on carp also was greater than of the skin-off fillets (7.1 vs 2.6%). The as-prepared yield for these skin-off fillets was about 10% less than that of the skin-on fillets for both chinook salmon and carp.

Comparison of Levels Found in Raw and Cooked Fish. *Chinook Salmon.* Means and standard deviations of the pesticides and total PCBs expressed as micrograms per fillet for fillets from Lakes Huron and Michigan are in Tables 3 and 4, respectively. Means and standard deviations of the pesticides and total PCBs expressed as ppm wet or ppm solids for fillets from Lakes Huron and Michigan as well as the percentage loss of pesticides and total PCBs are in the supplementary material. Detectable levels of all pesticides shown in these tables were found in all three fish analyzed per cooking method.

Analyses of variance established that the raw salmon had significantly higher levels of all pesticides and total PCBs than did the cooked salmon when the values were expressed as ppm solids (Table 5), which was also true when expressed as micrograms per fillet.

DDT, α-chlordane, *cis*-nonachlor, *trans*-nonachlor, HCB, toxaphene, and total PCBs expressed as ppm wet tissue in raw fish fillets also was significantly higher than the levels in cooked fish fillets even though ppm of wet tissue does not reflect the change in weight in fillets as a result of cooking.

Skinning the fillet prior to cooking/processing also had a significant effect on the pesticide and total PCB residues when the residues were expressed as ppm wet tissue, ppm solids, or micrograms per fillet. Values for pesticides and total PCBs expressed as ppm wet tissue in the skin-on and skin-off cooked fillets are given in Table 6. These data confirm the studies of Hora (1981) and Sanders and Haynes (1988), who reported that skinning and fat removal reduced contaminant levels in raw fish. Our study establishes that the reduction is carried over to the cooked fillets. The data illustrate the need to enhance educational programs to ensure sports fisherman do skin and trim chinook fillets to reduce the amount of environmental contaminants ingested.

Several significant lake effects were found for the level of pesticides and total PCBs in the chinook salmon. Expressed as ppm of wet tissue, Lake Michigan chinook salmon had significantly higher levels of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, γ-chlordane, oxychlordane, HCB, dieldrin, heptachlor epoxide, and total PCBs than did Lake Huron chinook salmon. This was not due to size or age of the salmon since the Lake Michigan salmon were both lighter in weight and younger than the salmon caught in Lake Huron. Sportsfishermen catching fish from Lake Michigan need to take these higher levels into account in considering how much of and how often they eat their catch.

Although the Lake Huron chinook salmon were larger, they had less fat (4.2% in skin-on and 1.8% in skin-off) than Lake Michigan chinook salmon (11.6% in skin-on and 5.8% in skin-off). The as-prepared yield of the Lake Huron chinook salmon was 5–7% less than the as-prepared yield of the Lake Michigan chinook salmon, but the total cooking losses from chinook salmon from both lakes were similar.

Fewer significant differences were found for the effect of cooking method. For the DDT complex only significant differences due to cooking method occurred for DDT and DDE. Fish fillets which were scored and charbroiled had higher levels of DDT expressed as ppm solids than those that were baked. DDE expressed as micrograms per fillet was also higher in the chinook salmon that was scored and charbroiled than in those which were canned. This is a reflection of the higher level of DDT in the raw chinook salmon used for scoring.

Cooking methods did not significantly affect the percentage reduction of any of the chlordane complex (α-chlordane, γ-chlordane, oxychlordane, *cis*-nonachlor, and *trans*-nonachlor). Percentage reduction tended to be higher for skin-off chinook salmon fillets which were baked or charbroiled.

Cooking method also did not significantly affect the level of reduction of dieldrin in the chinook salmon. Although the percentage reduction was slightly higher for some of the skin-on fillets, it must be remembered that the level of dieldrin in the skin-on fillets was approximately 60% higher, so cooking chinook salmon with the skin-on should not be recommended. Cooking method did not affect the level of HCB or heptachlor epoxide, nor did cooking method affect the percentage reduction. These pesticides were found at very low levels in all fish.

Greater differences among cooking methods were found for levels of toxaphene. The ppm expressed on a wet tissue basis of toxaphene in the cooked fillets was significantly lower in the canned chinook than in the charbroiled. Expressed as ppm solids, the level of toxaphene in the cooked tissue after canning was significantly lower than in that cooked by any other method. In addition, the percent change in the canned chinook salmon (>70%) was significantly higher than by any other cooking method.

The percentage reduction of total PCBs in the chinook salmon cooked without the skin (42%) was slightly higher than that when cooked skin-on (38%). Cooking

Table 7. Pesticides and Total Polychlorinated Biphenyls (PCBs) Expressed as Micrograms per Fillet in Raw and Deep Fat Fried or Pan Fried Skin-on or Skin-off Carp Fillets Harvested from Lakes Erie and Huron[a]

	deep fat fried		pan fried	
compound	raw	cooked	raw	cooked
		Lake Erie, Skin-on Fillets		
p,p′-DDT	ND	ND	1.17[b]	ND
p,p′-DDE	22.08 ± 14.18	15.98 ± 10.06	47.93 ± 60.37	30.85 ± 40.83
p,p′-DDD	4.86 ± 2.76	3.42 ± 2.91	3.36 ± 1.71	1.56 ± 0.68
α-chlordane	2.55 ± 1.96	1.61 ± 1.52	3.40 ± 3.36	1.94 ± 2.11
γ-chlordane	1.12 ± 0.45	0.76 ± 0.68	1.67 ± 1.60	1.34 ± 1.05[c]
oxychlordane	ND	ND	0.92 ± 0.69[c]	0.57[b]
cis-nonachlor	1.04 ± 0.61	0.69 ± 0.39	1.86 ± 2.04	1.07 ± 1.20
trans-nonachlor	3.15 ± 1.50	4.85 ± 2.65	7.58 ± 7.88	7.07 ± 8.04
HCB	0.37 ± 0.19	0.32 ± 0.14	0.21 ± 0.12	0.50 ± 0.37
dieldrin	4.46 ± 1.67	2.12 ± 0.78	6.41 ± 5.16	4.67 ± 4.16
heptachlor epoxide	0.48[b]	ND	0.83 ± 0.50	0.76[b]
toxaphene	ND	ND	ND	ND
total PCBs	321.1 ± 180.1	268.9 ± 132.9	631.9 ± 798.0	494.2 ± 630.4
		Lake Erie, Skin-off Fillets		
p,p′-DDT	ND	ND	ND	ND
p,p′-DDE	14.23 ± 16.76	7.79 ± 8.90	4.09 ± 1.53	3.40 ± 1.11
p,p′-DDD	1.31 ± 0.64	0.85 ± 0.43	1.19 ± 0.74	0.71 ± 0.30
α-chlordane	1.30 ± 0.87	0.57 ± 0.34	0.47 ± 0.18	0.35 ± 0.14
γ-chlordane	0.70 ± 0.25[c]	0.41[b]	0.48[b]	ND
oxychlordane	ND	ND	ND	ND
cis-nonachlor	0.87*	0.41[b]	ND	ND
trans-nonachlor	2.15 ± 1.46	1.58 ± 1.49	0.99 ± 0.66	1.02 ± 0.33
HCB	0.219 ± 0.063	0.101[b]	ND	ND
dieldrin	2.11 ± 1.27	1.00 ± 0.31[c]	1.24 ± 0.59	1.00 ± 0.35
heptachlor epoxide	ND	ND	ND	ND
toxaphene	ND	ND	ND	ND
total PCBs	169.0 ± 167.5	114.9 ± 138.0	47.37 ± 19.31	38.51 ± 19.98
		Lake Huron, Skin-on Fillets		
p,p′-DDT	0.69[b]	ND	1.31[b]	ND
p,p′-DDE	31.67 ± 21.02	17.02 ± 12.81	9.32 ± 6.87	4.86 ± 3.23
p,p′-DDD	7.60 ± 4.41	5.28 ± 2.84	13.67 ± 14.97	8.52 ± 10.35
α-chlordane	1.16 ± 0.68	0.79 ± 0.60	0.92 ± 0.41	0.41 ± 0.13
γ-chlordane	0.58 ± 0.16[c]	0.41 ± 0.11[c]	0.48 ± 0.10[c]	0.24[b]
oxychlordane	0.55[b]	ND	ND	ND
cis-nonachlor	0.85 ± 0.30	0.39 ± 0.15	0.53 ± 0.17[c]	0.24[b]
trans-nonachlor	1.74 ± 0.40	1.97 ± 0.79	1.93 ± 1.09	1.25 ± 0.64
HCB	0.24 ± 0.10	0.16 ± 0.09	0.16 ± 0.05	0.13 ± 0.04
dieldrin	1.24 ± 0.55	0.70 ± 0.58[c]	4.98 ± 7.04	0.36 ± 0.06[c]
heptachlor epoxide	0.51 ± 0.06[c]	ND	ND	ND
toxaphene	ND	ND	ND	ND
total PCBs	269.9 ± 244.8	89.6 ± 71.1	83.2 ± 72.0	48.1 ± 30.3
		Lake Huron, Skin-off Fillets		
p,p′-DDT	ND	ND	ND	ND
p,p′-DDE	3.23 ± 1.35	1.96 ± 0.80	3.37 ± 1.66	1.68 ± 0.60
p,p′-DDD	4.91 ± 5.48	2.41 ± 2.58	4.09 ± 1.53	3.40 ± 1.11
α-chlordane	0.21[b]	0.14[b]	0.26[b]	0.17[b]
γ-chlordane	0.16[b]	ND	ND	ND
oxychlordane	ND	ND	ND	ND
cis-nonachlor	ND	ND	ND	ND
trans-nonachlor	0.52 ± 0.25	0.38 ± 0.23	0.56 ± 0.04[c]	0.34[b]
HCB	0.11 ± 0.00[c]	0.08 ± 0.01[c]	0.10 ± 0.04[c]	0.11[b]
dieldrin	ND	ND	0.33[b]	ND
heptachlor epoxide	ND	ND	ND	ND
toxaphene	ND	ND	ND	ND
total PCBs	27.79 ± 8.0	18.81 ± 5.09	31.31 ± 9.53	19.75 ± 2.04

[a] $n = 3$; cooked deep fat fried samples included skin. [b] Only one fillet had value > minimum detectable level. [c] Two samples had values > minimum detectable level.

method, however, did not result in any significant differences in the level of residue or in the percentage reduction.

One of the objectives of this study was to evaluate whether increasing the surface area by scoring could increase pesticide and total PCB loss. While there was not a statistically significant difference in the loss of these compounds from scored and charbroiled as compared to charbroiled chinook salmon, Figure 1 shows that the loss in scored and charbroiled salmon was consistently higher than by charbroiling alone. Average losses of pesticides and total PCBs from the chinook salmon cooked by all methods from both lakes ranged from 30 to 41% (Figure 2). The loss of the DDT complex was 30%, while the loss of all other compounds was greater than 35%.

Carp. Carp from Lakes Erie and Huron were pan fried or deep fat dried as skin-on and skin-off fillets. Since deep fat fried fish is frequently prepared with a breading or a batter, the skin was included in the cooked analyses for this cooking method. Means and standard deviations of pesticides and total PCBs expressed as micrograms per fillet are given in Table 7. Means and standard deviations of the pesticides and total PCBs

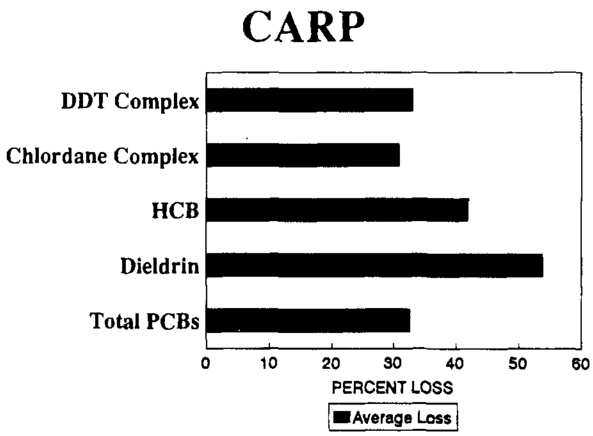

Figure 3. Average percentage losses of pesticides and total PCBs from carp from Lakes Erie and Huron cooked by pan frying and deep fat frying.

expressed as ppm wet tissue and ppm solids for fillets from Lakes Erie and Huron as well as the percentage losses are in the supplementary material.

Levels of *p,p*-DDT, γ-chlordane, oxychlordane, heptachlor epoxide, and toxaphene were below the limit of detection so often that the data were not analyzed statistically. In addition, the difference between the level of *trans*-nonachlor and dieldrin in the raw fillet did not differ significantly from that in the cooked, even though the average raw value was considerably higher. These statistics were affected by the large number of nondetectable values. Nevertheless, when expressed as ppm solids, *p,p'*-DDE, *p,p'*-DDD, α-chlordane, *cis*-nonachlor, HCB, and total PCBs were significantly lower in the cooked fillets than in the raw fillet.

Lake Erie carp had significantly more *p,p'*-DDD, α-chlordane, *trans*-nonachlor, and total PCBs when expressed as ppm solids than did Lake Huron carp. Size and fat content of the carp from both lakes were similar so these differences are due to differences in levels of contamination.

Whether the fillets were cooked skin-on or skin-off significantly affected the levels of several pesticides and total PCBs. Expressed as ppm of the cooked wet tissue, the skin-on fillets had approximately 3 times the levels of *p,p'*-DDE, *p,p'*-DDD, α-chlordane, *trans*-nonachlor, and total PCBs than did the skin-off fillets. Skin-on fillets did not have significantly greater values of *cis*-nonachlor, dieldrin, or HCB since there were many more values below the limit of detection in the skin-off fillet group.

Cooking method did not significantly affect the level of pesticide or PCBs in the carp nor did it affect the percentage of reduction during cooking. Either pan frying or deep fat frying can be used to cook skin-off fillets depending on the preference of the consumer. If a consumer has to cook carp skin-on, pan frying should be recommended over deep fat frying as it is easy to separate the muscle tissue from the skin and thus discard the skin and the associated fat. The percentage reduction in the DDT complex (*p,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD), the chlordane complex (α-chlordane, γ-chlordane, oxychlordane, *cis*-nonachlor, and *trans*-nonachlor), HCB, dieldrin, and total PCBs is illustrated in Figure 3. Average percentage losses ranged from 30 to 35% for the DDT complex, chlordane complex, and total PCBs, while the losses of HCB and dieldrin were greater than 40%.

Since cooking method did not significantly affect the level of residues, consumers can choose the method they prefer but they certainly should be encouraged to skin the carp fillets before cooking.

The average losses of the DDT complex of 30.4 and 33.1% for chinook salmon and carp, respectively, are less than the 64–72% which had been reported for lake trout by Reinert et al. (1972), but this early study used skin-on untrimmed fillets. Nevertheless, Smith et al. (1973) reported only minimum losses of DDT compounds (2–16%) during baking or poaching of chinook salmon. A later study of the effect of broiling, roasting, and cooking by microwave on DDT compound reduction from fat lake trout (siscowets) (Zabik et al., 1979) found losses of 30–57%. The average losses of dieldrin of 37 and 54% for chinook salmon and carp, respectively, agree with the loss of 25–57% from fat lake trout (Zabik et al., 1979). The current study is based on the analyses of 42 raw and cooked chinook salmon pairs and 24 raw and cooked carp pairs, whereas the early studies involved much smaller samples.

Sherer and Price (1993) used a mass basis to summarize PCB loss from a number of earlier studies. By this method they reported an average of 22% PCB loss by baking chinook salmon, lake trout, smallmouth bass, and bluefish; 27% loss from broiling lake trout and brown trout; 56% loss from frying smallmouth bass and white croaker; and 26% loss from microwaving lake trout. The current study had average losses of 41% PCBs from chinook salmon and 33% PCBs from carp.

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Supplementary Material Available: Description of procurement of fish and tables of processing data for fish (11 pages). Ordering information is given on any current masthead page.

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