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Concentration of Selected Priority Organic Contaminants in Fish Maintained on Formulated Diets in Lake Ontario Waters

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Abstract.—Fish were grown in Lake Ontario water under conditions simulating commercial aquaculture and then analyzed for 10 priority organic contaminants. Black bullheads (*Ameiurus melas*) were grown in cages placed in a bay of Lake Ontario. Rainbow trout (*Oncorhynchus mykiss*) were grown in terrestrial raceways served with Lake Ontario water. Yearlings were reared on a commercial ration in these systems, which partially isolated them from the contaminant-laden food web and bottom sediments, to an average weight of 93 g for black bullheads (range, 31–220 g) and 213 g (29–558 g) for rainbow trout. Concentrations of contaminants in skinless fillets of both rainbow trout cultivated 6 months and black bullheads cultured 3.5 months in Lake Ontario waters were nondetectable or less than one-sixth the “action levels” defined by the U.S. Food and Drug Administration. Contaminant levels in rainbow trout were consistently less than concentrations observed in black bullheads. Of the 10 priority contaminants surveyed, 7 were nondetectable in rainbow trout and 3 were nondetectable in black bullheads. Concentrations of contaminants in both species were generally much lower than levels observed in wild fish from Lake Ontario. This investigation demonstrated that bioaccumulation of lipophilic contaminants by fish cultured under simulated commercial conditions in Lake Ontario was not significant. These findings have implications for commercial aquaculture, regulatory decisions, and fish consumers in the Great Lakes basin and elsewhere.

Commercial aquaculture in the Great Lakes is being explored and pursued. In several demonstration and production operations, fish are cultured in terrestrial flow-through raceways and open-water cages (Figure 1). Primarily cool- and coldwater fishes are cultured at various levels of intensity and with varying success, including rainbow trout (*Oncorhynchus mykiss*), Atlantic salmon (*Salmo salar*), yellow perch (*Perca flavescens*), walleye (*Stizostedion vitreum*), and bullheads (*Ameiurus* spp.).

Health advisories exist that restrict the consumption of wild conspecifics of cultured fish as well as many other fishes from the Great Lakes (e.g., Minnesota Department of Health 1991; Wisconsin Department of Natural Resources 1991; New York Sea Grant 1993; NYSDEC 1993; Ontario Ministry of the Environment and Energy 1993). Regulations designed for wild-caught fish

have been applied to cultured fish (Fong and Brooks 1989; National Fisheries Institute 1992). Additionally, consumer awareness, quality management programs, and industry needs require that nothing but a high-quality product be produced and marketed (e.g., Buttner 1988; DFO 1990; NMFS 1991; Anonymous 1992a, 1992b).

Under controlled conditions that simulated commercial raceway and cage culture, we previously demonstrated that skinless fillets of cultivated rainbow trout and black bullheads (*A. melas*) contained greatly reduced or nondetectable levels of mirex, perhaps the most persistent and prevalent organic contaminant in Lake Ontario (Makarewicz et al. 1993). Of the 50 fish examined, only 3 contained detectable levels of mirex and all were 95% below the action levels established by the U.S. Food and Drug Administration (FDA). Many priority contaminants are chlorinated hydrocarbons like mirex. They are likely assimilated and bioaccumulated in a similar manner; however, presumption is not proof. The present study quantified 10 priority organic contaminants in the edible portions of frozen fish carcasses retained from the mirex study (Makarewicz et al. 1993).

Methods

Culture of black bullheads.—Between 12 and 14 June 1990, four floating cages (1-m³ volume, 1-cm plastic mesh; Buttner 1992a, 1992b) were anchored in Braddock Bay, Lake Ontario (Figure 1). Each cage received 100 or 300 yearling black bullheads. Average weight of black bullhead yearlings was 28.2 g (range, 22.4–33.5 g). Yearlings were third- and fourth-generation fish obtained from parents spawned at the State University of New York, College at Brockport. Fish in cages were fed once each day (1–3% body weight/d) with Catfish Cage Chow (Ralston Purina 5144; use of this product does not constitute an endorsement; Buttner 1992b). Water temperature, dissolved oxygen, pH, alkalinity, and Secchi disk visibility were monitored weekly (Makarewicz et al. 1993) and consistently remained in ranges considered nonstressful for fish. Cages were harvested on 27 September

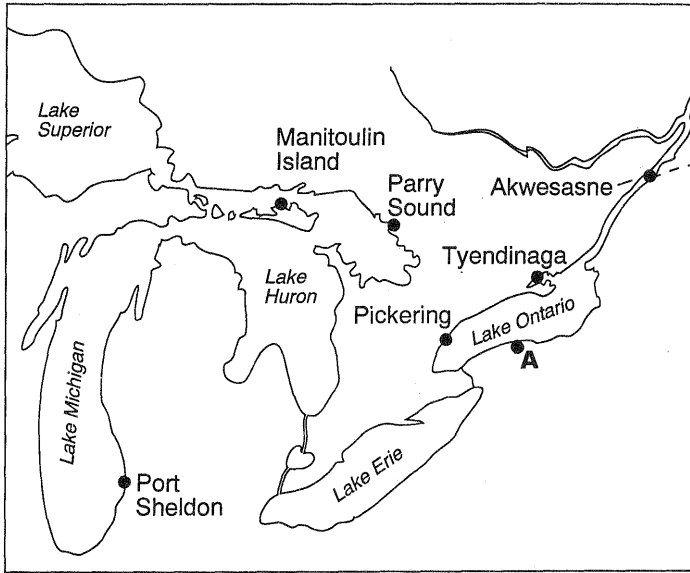


FIGURE 1.—Location of aquaculture operations in the Great Lakes. Fish are cultured in terrestrial flow-through raceways by Port Bay Aquaculture, Inc., Lake Michigan; Cool Water Farms, Ltd, Lake Ontario; and the Aquaculture Resource Center, Bay of Quinte, Lake Ontario. Cage culture is practiced by five operations in Georgian Bay (Lake Huron) and by the Mohawks of Akwesasne, St. Lawrence River. In this study, black bullheads were reared in cages in Braddock Bay and rainbow trout were cultivated in raceways at the Russell Station Power Generating Facility (A).

1990, after water temperature fell below 16°C and bullhead growth became negligible (Renaan 1990). Harvested fish were immediately weighed and frozen at -4°C.

Culture of rainbow trout.—On 18 April 1990, each of six flow-through raceways (1.8 × 0.6 × 0.3 m; 310 L) at the Russell Power Generation Station (Figure 1) received yearling rainbow trout. The Nashua (New Hampshire) strain trout (mean, 59 g; range, 23–123 g) were stocked at 0.32 yearlings/L. Yearlings were obtained from the Caledonia hatchery of the New York State Department of Environmental Conservation and were fed once daily to satiation with a 40% protein ration (Ralston Purina Trout Chow 5105 and 5106). Raceways were supplied solely with unfiltered Lake Ontario water either pumped directly from the lake or mixed with lake water that had passed through heat exchangers (residence time, 6 min) of the coal-fired plant. In April and May, variable amounts of heated lake water were blended with ambient lake water to maintain water temperatures above 10°C. By 15 May 1990, average daily water temperature exceeded 10°C and only ambient lake water was used for the remainder of the study. Between mid-July and late August, lake water temperature frequently exceeded 20°C and trout were

noticeably stressed. Routine maintenance included adjustment of water flow (maintained at 8–15 L/min), daily siphoning to remove sediments and feces, and alternate-day scrubbing and treatment with copper sulfate (0.4–0.8 mg/L) or formalin (45–65 µL/L) as a 30–40-min bath to control monogenic trematodes and a ciliated Protista (*Ichthyophthirius* sp.). Water temperature was measured daily and dissolved oxygen, pH, alkalinity, and total ammonia-nitrogen were monitored weekly (Makarewicz et al. 1993). Except for temperature, water quality consistently remained in ranges considered nonstressful for trout (Makarewicz et al. 1993). All rainbow trout were removed on 18 October 1990 and weighed; those used for analysis were immediately frozen.

Contaminant analysis.—Skinless fillets of four black bullheads from each cage ($N = 16$) and four rainbow trout from each of four raceways ($N = 16$) were examined for 10 priority organic contaminants (Table 1). Tissue was ground and homogenized in a food processor. Five grams of tissue were mixed with 20 g of anhydrous sodium sulfate and extracted overnight (Soxhlet; 200 cycles minimum) with 75 ml of methylene chloride : hexane (20:80 volume : volume). A 30-mL aliquot was evaporated to 1 mL under nitrogen, then cleaned

TABLE 1.—Detection limit, FDA action level, and mean (SE) concentration observed for organic contaminants in skinless fillets of rainbow trout and black bullheads cultivated in waters of Lake Ontario. Action levels are not available for endosulfan I and II (NA), and several contaminants were not detected (ND); $DDT_{total} = p,p'$ -DDT + p,p' -DDE + p,p' -DDD.

Contaminant	Action level (ng/g)	Detection level (ng/g)	Concentration (ng/g)	
			Trout	Bullhead
Chlordane (technical)	300	9	ND	45 (45)
DDT _{total}	5,000	12	140 (14)	149 (78)
Endosulfan I	NA	5	2 (2)	ND
Endosulfan II	NA	8	ND	34 (34)
Endrin	300	2	ND	ND
Heptachlor	300	2	ND	6 (6)
Heptachlor epoxide ^a	300	3	ND	38 (11)
Lindane	300	4	ND	31 (16)
Mirex	100	2	ND	ND
PCB _{total}	2,000	20	25 (7)	88 (9)

^a A technical chlordane component may coelute with heptachlor epoxide.

up through a 5-g Florisil[®] column as described by Insalaco et al. (1980) and Mills et al. (1972). The 5-g Florisil column was eluted with 40 mL of the 20:80 solvent followed by 40 mL of methylene chloride : hexane : acetonitrile (50:49.6:0.35 volumetric). The eluant was concentrated under nitrogen to a final volume of 1 mL. Analysis (1- μ L injection volume) was performed on a Hewlett Packard 5890A gas chromatograph with a 30 m \times 0.25-mm PTE5 (Supelco, Inc.) capillary column and HP 3396A integrator. The injector and ⁶³Ni electron capture detector temperatures were 220°C and 300°C, respectively. The oven temperature was programmed from 80°C to 275°C at a rate of 5°C/min. The sample was split 50:1. Standards were obtained from Supelco, Inc. Recovery efficiencies were estimated for all compounds analyzed. The mean recovery efficiency was 98.4% (range, 76.2–113.1%). Detection limits were based on the procedure employed; that is, detection limits were calculated based on the minimum peak detection and sample concentration during extraction and cleanup (Table 1). One blank and one replicate were examined per raceway or cage. Blanks were procedural blanks carried through the entire extraction and cleanup. Spiked recoveries were based on a final concentration of 0.33 mg/L. The coefficient of variation (SD/mean) of replicates of the spiked recoveries averaged 26% and ranged from 20% for mirex to 32% for total DDT.

Results

Concentrations of 10 priority contaminants in 16 skinless fillets of both rainbow trout and black bullhead cultivated for 6 and 3.5 months, respectively, in Lake Ontario waters were nondetectable or no more (usually much less) than 15%

of FDA action levels (Table 1). Contaminant levels in rainbow trout were consistently less than concentrations in black bullheads (Table 1). Of the 10 priority contaminants surveyed, seven were nondetectable in rainbow trout but only three were nondetectable in black bullheads.

Weight gains for trout and bullheads averaged 264% and 230%, respectively. Rainbow trout fed a commercial ration for 6 months grew to an average weight of 213 g (range, 29–558 g). Black bullheads maintained 3.5 months on a commercial ration attained an average weight of 93 g (31–220 g). Many rainbow trout attained a marketable size (>250 g), but few black bullheads reached preferred sizes (200 g and larger: Michael O'Hora, restaurant owner, Auburn, New York, and Kip Palmer, fish processor, Rochester, New York, personal communications).

Yearlings from the cohorts used in this study were analyzed for mirex before the experiment and no detectable levels (<2 ng/g) were observed; baseline analyses for the nine other priority contaminants were not conducted. Feed presented to fish was analyzed for organic contaminants. No detectable levels of mirex were observed, but a small and unquantified amount of contaminant was observed in the chromatographic region where PCBs are normally found. This was not unexpected, because commercial rations include a relatively large percentage of fish meal, which typically contains small amounts of PCBs (Sommer et al. 1982; Kadlec and Bush 1993).

Discussion

In this study, yearling rainbow trout attained marketable size in one growing season, but yearling black bullheads did not. Black bullheads are

TABLE 2.—Contaminant concentrations (ng/g wet weight) in fish from Lake Ontario, 1972–1988, and for commercially cultured channel catfish; ND = not detected.

Reference ^a or criterion	Fish species ^b	Mirex	Total PCBs	Total DDT	Chlordane	Lindane
1	Spottail shiner	7	116	34		
2	Rainbow smelt		2,650	270 ^c		
2	Slimy sculpin		630	30 ^c		
3	Lake trout	170	2,540	720 ^c	90	
4	Rainbow smelt		2,650	1,400		
4	Alewife		2,350	950		
4	Slimy sculpin		4,630	1,260		
5	White sucker	20	1,360	310	20	10
5	Rainbow trout	30	780	70		
5	Rainbow trout	110	1,480	110		
6	Rainbow trout	280	2,080	280 ^c		
7	Brown bullhead (wild)	37				
7	Rainbow trout (cultured)	ND	25	140		
7	Black bullhead (cultured)	1	88	149	45	31
8	Channel catfish		ND	220	100	10
	FDA action level	2	2,000	5,000	300	300

^a (1) Suns et al. (1991); (2) Borgmann and Whittle (1992); (3) Borgmann and Whittle (1991); (4) Haile et al. (1975); (5) NYSDEC (1981); (6) NYSDEC (1982); (7) Makarewicz et al. (1993); present study; (8) Nettleton et al. (1990).

^b Fish species not previously identified in text: spottail shiner (*Notropis hudsonius*), rainbow smelt (*Osmerus mordax*), slimy sculpin (*Cottus cognatus*), lake trout (*Salvelinus namaycush*), alewife (*Alosa pseudoharengus*), white sucker (*Catostomus commersoni*), brown bullhead (*Ameiurus nebulosus*), and channel catfish (*Ictalurus punctatus*).

^c Only DDE was reported.

coolwater fish that grow best between approximately 20 and 30°C (Renyaan 1990); rainbow trout are coldwater fish that grow best between approximately 10 and 20°C. Environmental conditions in Lake Ontario favor culture of rainbow trout. This hypothesis has been successfully tested at Cool Water Farms, Ltd., Pickering, Ontario, where rainbow trout are cultured commercially in large raceways, and at Akwesasne, where Mohawks grow modest numbers of rainbow trout, largely for personal consumption, in cages floated in the St. Lawrence River between April–May and November–December.

The Great Lakes are generally considered contaminated by a variety of chlorinated hydrocarbons and other chemicals (Holdrinet et al. 1978; Sport Fishing Institute 1986; USEPA 1992). Detectable and elevated levels of chlorinated hydrocarbons are found in the sediments, seston, and fish from these waters. Contaminants are particularly problematic in areas and societies where fish consumption is high, as it is in the Mohawk community of Akwesasne (NYSDEC 1990, 1992).

Aquaculture offers a potentially safe alternative source of fish and use of these waters, provided the harvested product consistently meets regulatory guidelines. As analytically demonstrated by Makarewicz et al. (1993), it is possible to grow fish with no detectable or greatly reduced levels of mirex in Lake Ontario, which is characterized

by mirex-contaminated seston, sediments, and food chains. In the present study, we examined the carcasses of fish cultivated in a previous mirex study and determined that nine other priority organic contaminants were also greatly reduced (relative to concentrations in wild fish) or nondetectable both in normally pelagic rainbow trout and benthic black bullheads. Contaminant concentrations in cultured fish were much lower than FDA action levels (Table 1) and were generally much lower than or comparable to the lowest levels observed in wild fish from Lake Ontario (Table 2). Because the yearlings and their feed were “clean,” the reduction would be expected only if the food web were the major source of the contaminants. Clean fish were produced by simply following standard aquaculture practices that call for using clean fingerlings, feeding a high-quality commercial ration, and rearing fish in systems such as terrestrial raceways or cages that largely isolate them from contaminant-laden food chains and sediments.

Both the species cultured and the method of culture affected contaminant uptake. Black bullheads cultivated in cages floating in Braddock Bay accumulated a greater variety of contaminants at higher concentrations than did rainbow trout cultivated in raceways (Table 1). Because black bullheads contain considerably less fat (1.6 g/100 g) than do rainbow trout (11.4 g/100 g; Pennington

and Church 1985), differences in contaminant levels were probably related to the bullheads being cultured in cages floating in a natural environment near the contaminant-laden sediments of Braddock Bay and the trout being cultivated in raceways with reduced exposure to sediments. It is also probable that contaminated natural forage was more accessible to cage-cultured fish than to those grown in raceways.

The ratio of total DDT to total PCB was less than 1 for all wild-caught species of fish from Lake Ontario (Table 2), whereas the DDT:PCB ratio was greater than 1 in fish fed artificial diets in our study. We have no explanation for this other than that PCB levels in artificial feed (average, 44.6 ng/g wet weight, or 50 ng/g dry weight given a 10% feed moisture content; Kadlec and Bush 1993; Kadlec 1994) is much lower than PCB levels in naturally occurring benthic fauna (average, 341 ng/g dry weight; Haile et al. 1975) from Lake Ontario. Such a difference in concentration could account for the observed shift in the DDT:PCB ratio. Lindane was also observed in four black bullheads. This was somewhat surprising. However, NYSDEC (1981) also observed lindane in bottom-dwelling white suckers from Lake Ontario (Table 2). Polyaromatic hydrocarbons (PAHs), selenium, and phenols potentially associated with power plant waste water were not monitored.

Despite the presence of contaminants in Lake Ontario and the prevalence of low concentrations of contaminants (especially PCBs) in commercial feeds, fish that consistently met FDA guidelines and were much cleaner than their wild counterparts were cultivated in waters of Lake Ontario. Contaminant levels were much reduced or nondetectable and approximated concentrations observed in pond-cultured channel catfish monitored for toxic organic chemicals (Nettleton et al. 1990).

Our results on 10 priority pollutants have implications for aquaculture in the Great Lakes and other waters. The data indicate that blanket application of health advisories appropriately applied to recreationally and commercially captured fish is inappropriate for aquaculturally produced fish. Aquaculture can provide a legitimate alternative source of high-quality fish while creating new jobs and revenue in an environmentally friendly and socially acceptable fashion, as is being realized by a modest number of culturists. However, commercialization should proceed cautiously, and care should be given to site selection. "Hot spots," characterized by large amounts of the more water-soluble chlorinated PCB congeners dissolved in

the water and accumulated in the substratum, should be avoided because fish can quickly pick up these contaminants through the integument and bioconcentrate them in a few days to levels that exceed FDA guidelines (Kadlec and Bush 1993; Kadlec 1994). However, when dissolved levels are very low, as in most of the Great Lakes and their bays, bioconcentration is not a major route of uptake (Makarewicz et al. 1993). Biomagnification, which is the major route of uptake through the food web, can be interrupted by use of prepared rations in commercial production. To ensure that each batch of fish is indeed clean, it is desirable that monitoring protocols be established and followed.

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