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Exchange of Mirex between Lake Ontario and its Tributaries

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EXCHANGE OF MIREX BETWEEN LAKE ONTARIO AND ITS TRIBUTARIES

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INTRODUCTION

The 1974 discovery of mirex in Lake Ontario fish by Kaiser (1974) triggered a period of intensive study on the substance within the lake ecosystem. Two Lake Ontario tributaries were identified as sources of mirex. The Niagara River is the major source of mirex (366 kg) to Lake Ontario, while the Oswego River discharge (224 kg) has been attributed to the Armstrong Cork Company in Volney, NY (Holdrinet et al. 1978). Hooker Chemical and Plastics Corporation manufactured and processed mirex at its Niagara Falls, NY. plant from 1957 - 1976 (Task Force on Mirex [TFM] 1977). Peak discharge to the lake occurred in the 1960's and subsequently declined (Durham and Oliver 1983) as follows; 200 kg/yr from 1960 to 1962, 13.3 kg/yr in 1979 and 8 kg/yr in the period 1979 - 1981 (Warry and Chan 1981, Kuntz and Warry 1983, Halfon 1987). A single discharge (~1961) into the watershed of the Oswego River (Holdrinet et al. 1978) continues to supply mirex to the lake ecosystem (Scrudato and DelPrete 1983). Total mirex loading to Lake Ontario has been estimated at 688 kg (Holdrinet et al. 1978) of which half has been incorporated into the sediments (Pickett and Dossett 1979). Continuing losses from dump sites will augment existing mirex levels in Lake Ontario (Warry and Chan 1981, Scrudato and DelPrete 1982, Kuntz and Warry 1983, Halfon 1987).

Mirex bioaccumulates at all trophic levels in aquatic systems (TFM 1977). During the period of intensive monitoring from 1975 to 1981 (TFM 1977, Armstrong and Sloan 1980, Insalaco 1980, Norstrom et al. 1978) revealed detectable levels (usually > 5 ppb) of mirex in fish. The top predators (e.g.

salmonines) contained the highest concentrations of mirex which often exceeded the U.S. Food and Drug Administration's action level of 0.1 ppm. This knowledge prompted government agencies on both sides of the U.S.- Canadian border to issue health advisories on eating fish from Lake Ontario.

A high correlation between mirex levels and organic content of the sediments exists (Scrudato and DelPrete 1983). Availability of mirex to Lake Ontario biota ranges from 200 - 600 years before the contamination is buried by clean sediments (Halfon 1981, cited in Scrudato and DelPrete 1982). Since mirex is considered one of the most stable compounds ever evaluated (Metcalf et al. 1973), it could recycle within the lake ecosystem for many years via resuspension, uptake and bioaccumulation in the foodweb and sedimentation. Another potential mechanism of recycling, not generally considered, is the spawning migration of mirex laden fish. I report here an estimate of the amount of mirex available for recycling back to the Lake Ontario ecosystem by spawning migrations and on the contamination of resident fish in tributaries.

METHODS

To test the hypothesis that toxic substances are accumulating in stream resident fish due primarily to the spawning migration of Pacific salmon (*Oncorhynchus* spp.) from Lake Ontario, sampling sites were chosen in tributaries in Orleans County, NY. (Fig. 1). The control site (Oak Orchard Creek) was not accessible to migrating fish from Lake Ontario due to a dam, while the experimental site (Marsh Creek) is directly accessible to fish migrating from Lake Ontario (Fig. 1). Both sampling sites are above the direct influence of Lake Ontario and are typical of small upstate New York creeks.

Various fish species were captured from 4 November 1983 to 20 September 1984 with a backpack electroshocker. Fish were immediately packed in ice for transport to the laboratory. Length and weight data was recorded and samples were wrapped in foil and frozen until analysis.

Whole fish were homogenized using a Virtis tissue homogenizer. A 7.5 gram subsample was extracted using Soxhlet apparatus with methylene chloride / hexanes (20:80 v/v) a minimum of 200 cycles. When an individual specimen did not weigh 7.5 grams, multiple specimens from the same area and of the same species were pooled and homogenized together. Salmonine species (whole fillet) and their sex products were analyzed using a 5.0 gram subsample of homogenized tissue. Extractant cleanup consisted of lipid removal using a standard florisil column, PCB nitration following Norstrom *et al.* (1980) with modifications by Insalaco *et al.* (1980) and Kent (1983). Several modifications were made in analysis of non-salmonine species. Seventy-five (vs 25%) percent of the original extracted volume was taken and after nitration 75% (vs 50%) percent of that volume was carried on to cleanup. These modifications allowed quantification of extremely low levels of mirex. Mirex analysis was performed on a HP 5750-B research gas chromatograph equipped with a ^{63}Ni electron capture detector.

RESULTS

OAK ORCHARD CREEK

No fish samples from the Oak Orchard creek control sites contained detectable levels of mirex. Seventeen analyses were performed on twenty seven fish from Oak Orchard Creek (Table 1). Six creek chubs (*Semotilus atromaculatus*) ranging from 14.6 g (3 fish combined) to 55.6 g (mean = 32.1 g) were analyzed. Because of their small size (mean = 6.1 cm.) six bluntnose minnows (*Pimephales notatus*) were combined for a single mirex analysis (weight - 10.6 g). Six white suckers (*Catostomus commersoni*) were used for three analyses [10.8 g (4 fish) to 184.1 g (mean = 72.4 g]. Smallmouth bass (*Micropterus dolomieui*) (13.4 g - 212.2 g; mean = 84.6 g, n = 6) and brown bullhead (*Ictalurus nebulosus*) (28.0 g - 225.9 g; mean = 94.7 g, n = 3) were analyzed individually for mirex.

MARSH CREEK

Thirty fish from Marsh Creek were used to perform twenty three analyses for mirex. Detectable mirex levels were found in 12 of the 23 samples (Table 2). Fish species containing mirex were creek chub, smallmouth bass, and bluntnose minnow. White suckers from Marsh Creek did not contain detectable levels of mirex. Mirex concentrations of creek chubs [10.3 g - 167.9 g (mean = 36.0 g, n = 8)] ranged from N.D. to 0.025 mg/kg (mean = 0.008 mg/kg). Ten bluntnose minnows were combined into four samples (11.3 g - 13.1 g; mean = 12.0 g).

Mirex concentrations ranged from 0.003 to 0.008 mg/kg (mean = 0.006 mg/kg). Concentration of mirex of four smallmouth bass combined into three samples [8.3 g (2 fish) to 30.3 g; mean = 18.2 g] ranged from N.D. to 0.009 mg/kg (mean = 0.003 mg/kg). No detectable mirex was found in any of the eight white sucker samples (9.3 g - 37.1 g; mean = 24.3 g) analyzed.

Four 1983 fall spawning run Pacific salmon were analyzed. The two coho salmon (*Oncorhynchus kisutch*) (3.8 and 4.6 kg) had mirex concentrations of 0.104 and 0.142 mg/kg. The two chinook salmon (*Oncorhynchus tshawytscha*) (5.0 and 14.1 kg) had mirex concentrations 0.071 and 0.171. Eggs from the larger female chinook salmon had a mirex concentration of 0.217 ± 0.042 (mean ± S.E.; n = 2) mg/kg. Milt from the male chinook salmon contained no detectable mirex.

DISCUSSION

The study sites were chosen to provide one major difference, exposure to mirex from Lake Ontario biota. Oak Orchard and Marsh creeks have no known sources of mirex within their watersheds (Kaiser 1978). Marsh Creek, the experimental site, receives spawning migrations of salmonines, including coho and chinook salmon from Lake Ontario. The upper portion of Oak Orchard Creek, the control site, cannot receive spawning migrations of Lake Ontario fish due to a dam at Waterport, NY. (N.Y. Conservation Department 1940) (Figure 1).

Mirex was observed in three of the four species of fish sampled in Marsh Creek, but in none of the fish sampled from Oak Orchard Creek. Mean mirex levels found in Marsh Creek fish were about 4 ppb (Figure 2). White suckers do migrate (Raney and Webster 1942, Werner 1979) and would be expected to be in contact with mirex in Lake Ontario. However, mirex was not detected (n = 8) in any of the white suckers sampled in Marsh Creek perhaps due to their relatively low lipid content (0.9%) (NYSDEC 1981) and the small size of the fish. Mirex was observed in decreasing concentrations in the creek chubs, bluntnose minnows, and smallmouth bass and may be related to their differing ages (Table 2). Concentration of lipophilic compounds in fish are related to the trophic status and the age and size of the individual (Insalaco et al. 1980, Kent 1983). Generally individuals higher in the trophic web contain higher body burdens than those lower in the trophic web. Intraspecifically, older larger fish tend to be higher in lipid content and thus possess higher concentrations of chlorinated hydrocarbons (Norstrom et al. 1978, Armstrong and Sloan 1980, Insalaco et al. 1982). In this case, all the fish were probably feeding at similar trophic levels (Kraatz 1928, Keast and Webb 1966, Newsome and Gee 1978). Therefore, the mirex concentration is related to length of exposure to lipophilic compounds in the water, or more likely, the ingestion of mirex-laden food.

There are two possible causes of contamination in Marsh Creek. Individual fish could migrate to the known source (Lake Ontario) and pick up mirex through the integument or by feeding, or mirex could be introduced into the stream ecosystem. The sampling sites on Marsh Creek are 7 - 10 kilometers upstream from Lake Ontario. It is unlikely any of the fish that contained mirex had migrated to or from the lake. Although chubs are found in small lakes they prefer small, clear streams (Barber and Minckley 1971, Scott and Crossman 1973) and travel of more than 7 kilometers is doubtful. Movement in adult smallmouth bass is usually limited to 0.8 - 8 kilometers (Scott and Crossman 1973), it is unlikely that young of the year or the 1 year old bass would make a trek of 7 kilometers. Bluntnose minnows have a body form that is "less maneuverable than other cyprinids" (Keast and Webb 1966) and show no tendency to travel the distance required to reach Lake Ontario (Scott and Crossman 1973).

It is unlikely that any of the stream fish that contained mirex had migrated to or from Lake Ontario. And there are no known sources of mirex in the watershed of Marsh Creek. The evidence suggests that mirex has been introduced into the stream ecosystem by some other mechanism that requires an expenditure of energy, i.e. the transport of the contaminant upstream from Lake Ontario. Generally the movement of toxics in the environment occurs from high to low as in a diffusion gradient such as the migration of leachate from a toxic dump into ground water then to a river or lake. Rarely do we observe toxics being transported against an energy gradient (e.g. the flow of a stream) in the environment.

An obvious source of contamination to Lake Ontario tributaries are fish that migrate into tributaries to spawn. There is no doubt that fish species in Lake Ontario are contaminated with mirex and a number of species utilize tributaries to spawn. The major source of mirex in Marsh Creek is probably Pacific salmon (*Oncorhynchus* spp.) (Table 3). These species normally die in the tributary soon after spawning. This process exposes the stream ecosystem to both contaminated sex products and the decomposing carcasses. Direct transfer to stream resident species could occur by ingesting portions of the dead carcass or the eggs. In fact, fish were observed eating a decomposing carcass in Marsh Creek during the 1983 fall spawning run. Low (1983) found that stream resident brown trout were contaminated with mirex from spawning Pacific salmon in eastern Lake Ontario tributaries. The uptake was attributed to the ingesting of salmon eggs which had an average mirex concentration of 0.049 ppm in the 1981 spawning run. Indirect transfer may occur due to release of mirex into the water or sediments and movement up the food chain.

A crude estimate of mirex loading into the stream ecosystem was calculated by counting the number of dead salmon after the fall 1983 run. Along 3.2 kilometers of sampling area on Marsh Creek 375 dead salmon littered the stream and its banks. Using the subsample of salmon that were taken for analysis an estimate of total mirex loading was calculated. The mean weight of the salmon sampled was 4.8 kilograms with a mean mirex concentration of 0.122 mg/kg or

0.586 milligrams of mirex per individual (Table 3). The mirex loading for the sampling area for fall 1983 was estimated to be 69 mg/km. Assuming a similar density of carcasses for all of Marsh Creek and the undammed portion of Oak Orchard Creek (34 km) the fall 1983 total mirex load to the Oak Orchard - Marsh Creek system was 2.3 grams. Thus, spawning salmon are contaminating stream ecosystems and their biota. There is some evidence that mirex may also enter the terrestrial food web. Johnson and Ringler (1979) and Low (1983) identified the blowfly larvae as another potential transporter of mirex in the upstream ecosystem. The blowfly larvae showed an affinity to bioaccumulate mirex to very high concentrations (mean = 0.150 ppm in 1981). Fisher (1981; cited in Low 1983) found that blowfly larvae are the only organism of consequence in the decomposition of salmon carcasses. In addition some of this mirex would eventually be recycled as erosive action of the streams carries sediment back to Lake Ontario.

By estimating the number of salmon that spawn each fall, the amount of mirex being transported into tributaries and potentially being recycled back to the Lake Ontario ecosystem can be calculated. The number of dead carcasses per kilometer of Marsh Creek allowed the estimate of the actual salmon returns to the stream assuming an angler harvest of 50% (Keleher et al. 1985, Haynes et al. 1985, 1986) (Appendix 1). Salmon mature at different rates, consequently, an individual spawning run is comprised of fish from three different age classes. From salmon run data collected over 13 years, the mean age class composition of a spawning run was calculated (NYSDEC 1983, 1986, 1987) (Appendix 2). Using the stocking rates for the Oak Orchard - Marsh Creek system, (NYSDEC 1981, 1982, 1983, LeTendre 1987) the calculated age composition, and the maximum spawning returns, a percent survival to migration of 4.4% was estimated (Table 4). This compares favorably with an estimated survival rate of 5 - 10% provided by NYSDEC (LeTendre 1987).

The maximum spawning return for all tributaries for fall 1983 was calculated in a similar manner using the total number of salmon stocked in Lake Ontario (NYSDEC 1986, 1987) (Table 5). Assuming a 4.4% survival to migration and using body burdens of mirex in fish observed in this study, the amount of mirex transported into all Lake Ontario tributaries from fall 1983 spawning salmon was 53 gm/yr (Table 5) or 0.008% of the total mirex load in Lake Ontario. Migration to tributaries other than their "home" stream, or straying does occur in Lake Ontario salmonines (NYSDEC 1983). Individuals that stray would be considered fatalities in this survival estimate and would suggest that the 4.4% survival rate is conservative. If a 10% survival rate is assumed, the amount transported is 121 gm/yr or 0.018% of the total mirex load in Lake Ontario. Even with the 492% increase in salmon stocking comprising the 1987 fall spawning run, (NYSDEC 1986) the amount of transported mirex would increase only to 0.038% of the total Lake Ontario mirex load. Other lake species that utilize tributaries to spawn (i.e. brown trout, rainbow trout, white sucker, and rainbow smelt) are not considered, but would undoubtedly increase the mirex loading to the stream ecosystems.

These estimates are admittedly rough but do provide an idea of the range of magnitude of mirex that is transported annually into Lake Ontario tributaries. This value also provides an upper estimate, because it does not consider angler removal (as high as 90% in some tributaries, Wedge 1987), of the amount of mirex potentially recycled back to Lake Ontario. The input of mirex, from tributaries thought not to be sources of mirex, may need to be considered in models of mirex loading to the sediments of Lake Ontario (e.g. Halfon 1987).

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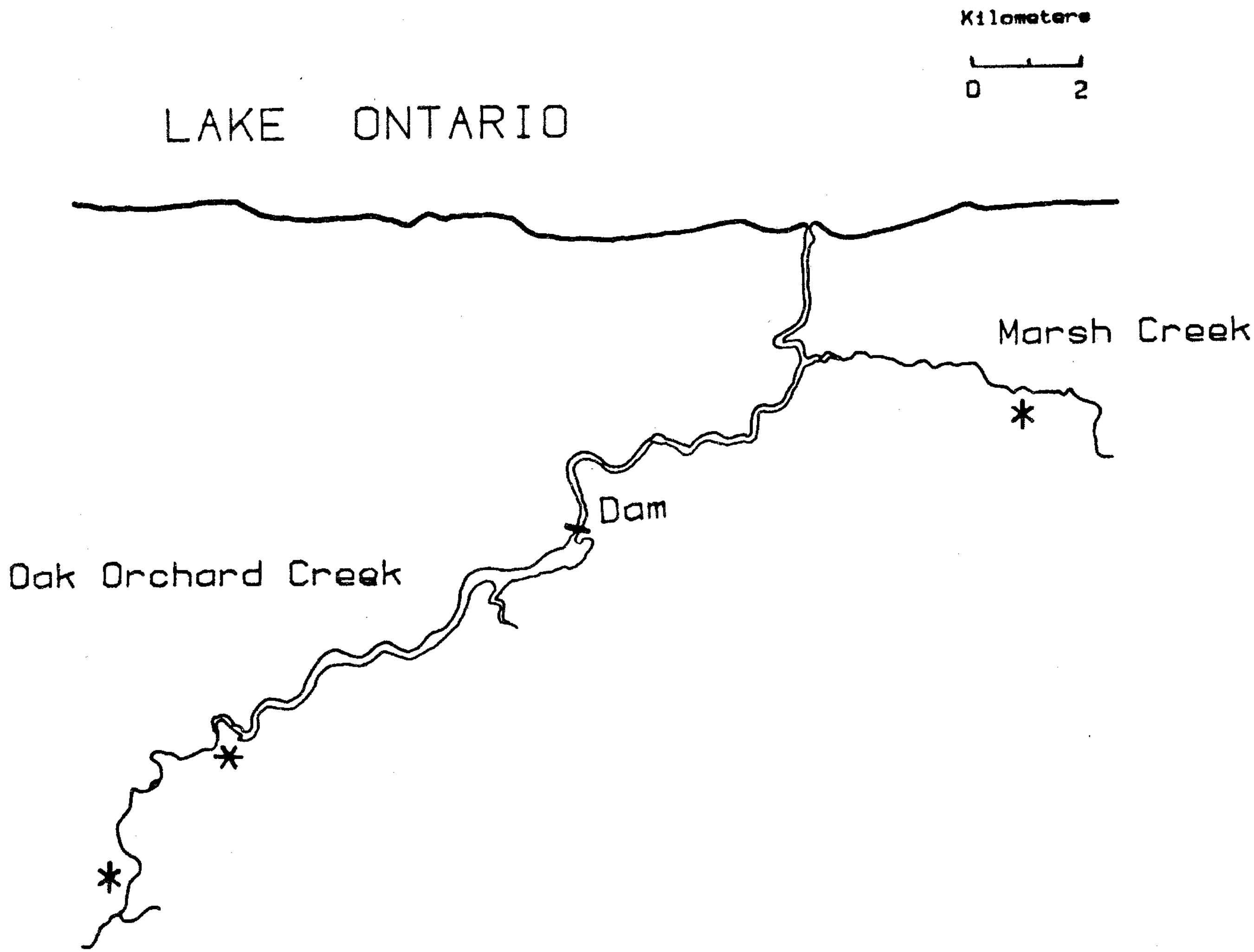

Figure 1. Map depicting the sampling sites (*) on Lake Ontario tributaries.

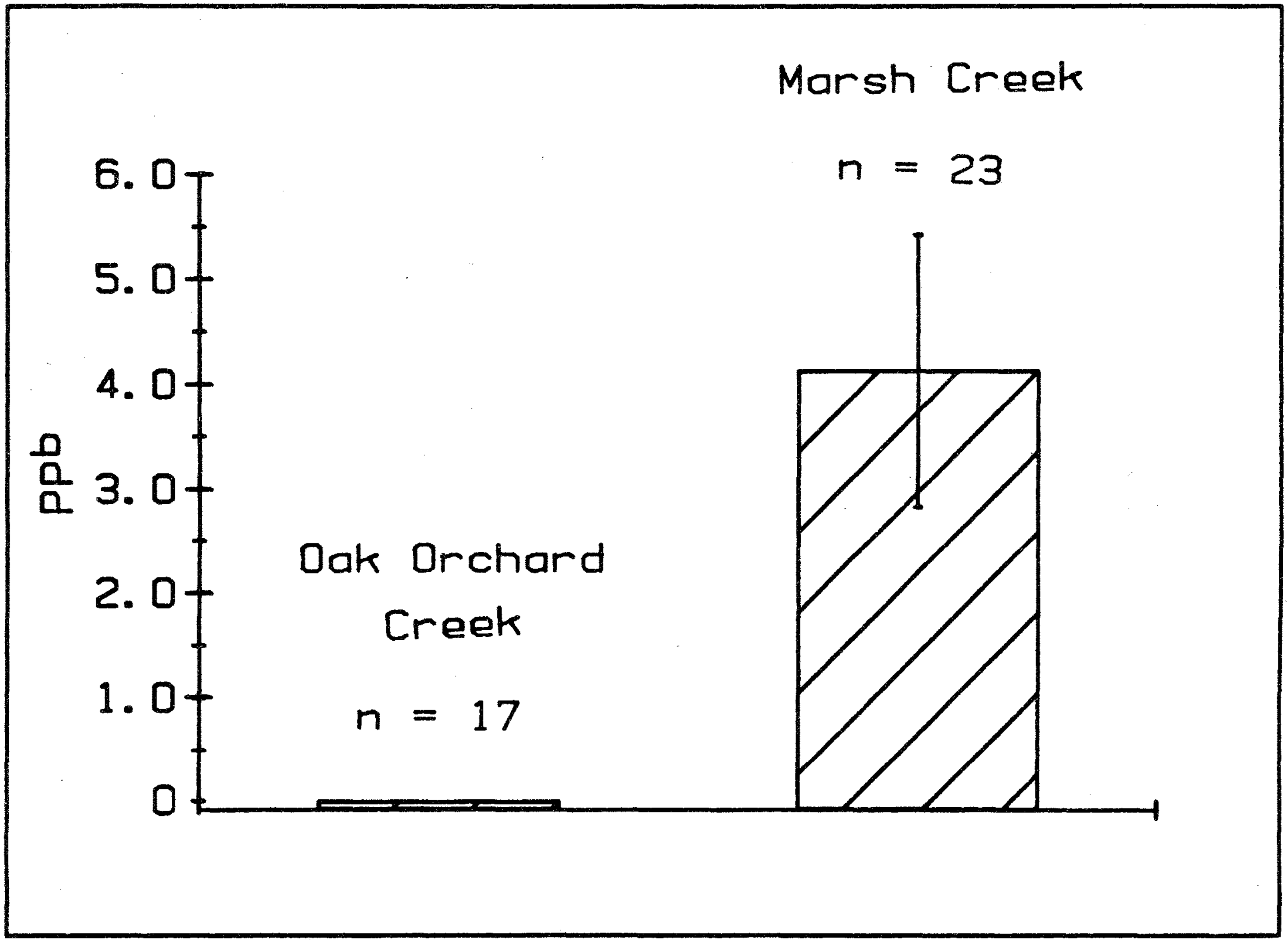

Figure 2. Mean (± S.E.) mirex concentrations of all samples analyzed from Oak Orchard Creek (control site) and Marsh Creek (experimental site).

Table 1. Biological data and mirex concentrations of fish collected from Oak Orchard Creek (control site) Orleans County, New York.

OAK ORCHARD CREEK

CREEK CHUB

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
55.59	17.4	0.000
33.92	14.1	0.000
24.50	13.0	0.000
14.57	3 FISH	0.000

WHITE SUCKER

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
184.06	24.9	0.000
129.00	22.4	0.000
10.79	4 FISH	0.000

BROWN BULLHEAD

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
225.92	25.8	0.000
29.65	13.4	0.000
28.60	12.6	0.000

SMALLMOUTH BASS

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
212.18	25.3	0.000
193.95	24.5	0.000
46.72	16.9	0.000
22.89	11.7	0.000
18.46	11.4	0.000
13.43	9.7	0.000

BLUNTNOSE MINNOW

NO. OF FISH	TOTAL WEIGHT (gm)	MIREX (mg/kg)
6	10.57	0.000

Table 2. Biological data and mirex concentrations of fish collected from Marsh Creek (experimental site) Orleans County, New York. Fish ages were estimated from length - age data (Westman 1938, Forney 1972, Beamish 1973, Scott and Crossman 1977, Powles _et al_. 1977).

MARSH CREEK

CREEK CHUB

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
167.94	22.9	0.006
30.02	13.2	0.004
18.68	12.4	0.000
18.58	12.5	0.025
16.13	11.3	0.017
14.42	12.1	0.004
12.18	10.6	0.006
10.34	10.4	0.000
MEAN ± S.E.		0.008 ± 0.003

Age range: 1 - 5 yr

WHITE SUCKER

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
37.09	13.8	0.000
34.46	13.7	0.000
31.24	14.2	0.000
23.91	13.0	0.000
21.33	11.6	0.000
19.56	12.1	0.000
17.26	12.2	0.000
9.26	10.3	0.000
MEAN		0.000

Age range: 0+ - 4 yr

BLUNTNOSE MINNOW

NO. OF FISH	TOTAL WEIGHT (gm)	MIREX (mg/kg)
3	13.10	0.003
3	11.80	0.006
2	11.54	0.008
2	11.34	0.007
MEAN ± S.E.		0.006 ± 0.001

Age range: 1 - 2 yr

SMALLMOUTH BASS

WEIGHT (gm)	LENGTH (cm)	MIREX (mg/kg)
30.32	13.1	0.008
16.09	11.0	0.000
8.33	2 FISH	0.001
MEAN ± S.E.		0.003 ± 0.002

Age range: 0+ - 1 yr

Table 3. Biological data and mirex concentrations of salmon collected from the fall 1983 spawning run from Marsh Creek (experimental site) Orleans County, New York.

CHINOOK SALMON			COHO SALMON		
WEIGHT (kg)	LENGTH (cm)	MIREX (mg/kg)	WEIGHT (kg)	LENGTH (cm)	MIREX (mg/kg)
5.0	76.0	0.071	3.8	71.4	0.104
14.1	104.0	0.171	4.6	71.0	0.142

Table 4. Composition of the 1983 fall salmon spawning run for Oak Orchard Creek. Salmon stocking rates for Oak Orchard Creek are presented (NYSDEC 1981, 1982, 1983, LeTendre 1987). The calculation of percent survival to maturity is shown.

Stocking Year	Age	Chinook salmon stocked	Coho salmon stocked	Total salmon stocked	Percent of each age class (Appendix 1)	Maximum return
1982	1+	167,000	41,800	208,800	46.4	96,883
1981	2+	137,500	31,200	168,700	38.2	64,443
1980	3+	119,220		119,220	15.5	18,479
					Total	179,806

Estimate of returns (From Appendix 2)	7,956
Maximum returns	179,806
Survival rate to migration	4.4 %

Table 5. Composition of the 1983 fall salmon spawning run for all Lake Ontario tributaries. Salmon stocking rates for Lake Ontario are presented (NYSDEC 1986, 1987, LeTendre). The calculation of the amount of mirex available for recycling with a 4.4% surviyal rate is shown.

Stocking Year	Age	Chinook salmon stocked	Coho salmon stocked	Total salmon stocked	Percent of each age class (Appendix 1)	Maximum return
1982	1+	2,077,886	550,000	2,627,886	46.4	1,219,339
1981	2+	1,480,237	419,433	1,899,670	38.2	725,674
1980	3+	788,070		788,070	15.5	122,151
					Total	2,067,164

Percent survival rate (From Table 4)	4.4
Maximum returns	2,067,164
Total fall 1983 salmon run for Lake Ontario	90,955
Milligrams mirex per fish (mean wt = 4.8 kg) (mirex = 0.122 mg/kg)	0.585
Mirex available for recycling (grams)	53

Appendix 1. Sample calculation for estimating observed survival to migration for salmon in Oak Orchard Creek.

	375	Salmon observed in 3.2 km.
	X 2	Correct for angler harvest (Keleher et al. 1985, Haynes et al. 1985, 1986)
	750	Salmon per 3.2 km
	- OR -	
	234	Salmon per km
	34	Kilometers in Oak Orchard - Marsh creek system
Thus	7956	Estimated fall 1983 salmon run

Appendix 2. Salmon River chinook salmon abundance by age class for fall spawning run gill net sampling efforts (NYSDEC 1983, 1986, 1987).

	Number of spawning fish in each age class.				Percent age composition of fall spawning run.		
YEAR	1+	2+	3+		1+	2+	3+
1972	196	40	1		83	17	0
1973	223	105	1		68	32	0
1974	257	251	4		50	49	1
1975	79	161	31		29	59	11
1976	167	151	20		49	45	6
1977	54	134	32		25	61	15
1978	6	109	101		3	50	47
1979	13	9	39		21	15	64
1980	13	45	10		19	66	15
1981	160	13	12		86	7	6
1982	127	102	10		53	43	4
1985					55	28	17
1986					61	24	15
				MEAN	46.4%	38.2%	15.5%