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Contaminant Trends in Lake Trout (*Salvelinus namaycush*) from the Upper Great Lakes

David S. DeVault,* Wayne A. Willford,† Robert J. Hesselberg,† David A. Nortrupt,†
Eric G. S. Rundberg,* Alwan K. Alwan,* and Cecilia Bautista*

*United States Environmental Protection Agency, Great Lakes National Program Office, 536 South Clark Street, Chicago, Illinois 60605 and †United States Fish and Wildlife Service, Great Lakes Fishery Laboratory, Ann Arbor, Michigan 48105

Abstract. Contaminant body burdens in lake trout (*Salvelinus namaycush*) from the Upper Great Lakes have been monitored since 1970 on Lake Michigan and since 1977 and 1978 on Lakes Superior and Huron by United States Environmental Protection Agency (USEPA), Great Lakes National Program Office and U.S. Fish and Wildlife Service (USFWS), Great Lakes Fishery Laboratory.

Analysis of the Lake Michigan data shows that mean polychlorobiphenyl (PCB) concentrations declined from a maximum of 22.91 mg/kg in 1974 to 5.63 mg/kg in 1982. Mean total DDT concentrations declined from 19.19 mg/kg in 1970 to 2.74 mg/kg in 1982. The decline in both contaminants closely followed first order loss kinetics. If the current declines continue, PCB concentrations will decline to the U.S. Food and Drug Administration (USFDA) tolerance of 2.0 mg/kg in 1988. Mean total DDT concentrations will fall to the International Joint Commission (IJC) objective of 1.0 mg/kg by 1991. Mean dieldrin concentrations increased significantly from 0.20 mg/kg in 1971 to 0.58 mg/kg in 1979 before declining to 0.21 mg/kg in 1982. The decline from 1979–1982 followed first order loss kinetics. As this decline is not reflected in other species (bloater chubs, smelt) it will require additional years of monitoring to determine if dieldrin concentrations between 1979 and 1982 truly represents a declining trend. The only statistically significant trend other than in Lake Michigan was for total DDT which declined significantly in Lake Superior lake trout. Large data variance and the short time frame covered (1977–1982) interfered with detection of trends on Lakes Superior and Huron.

The presence of toxic organic contaminants, many of which bioaccumulate in fish, is one of the most serious environmental problems in the Great Lakes. During the past decade, contaminants such as polychlorobiphenyls (PCBs), DDT, and dieldrin have been found in Great Lakes fish at concentrations exceeding U.S. Food and Drug Administration action levels. Bans and restrictions on the use of these compounds have been implemented in an effort to control these problems.

As part of a coordinated surveillance program, the U.S. Environmental Protection Agency's (USEPA), Great Lakes National Program Office (GLNPO) and the U.S. Fish and Wildlife Service (USFWS), Great Lakes Fishery Laboratory have cooperated in a program to monitor contaminant trends in fish collected from each of the Great Lakes and Lake St. Clair. This first report from the Cooperative Program presents the results of the analysis of lake trout (*Salvelinus namaycush*) collected in Lakes Michigan, Superior, and Huron in 1977–1982.

Methods

Lake Trout were collected by gillnet from one location on each of Lakes Michigan, Huron, and Superior (Figure 1) in the fall of the year by USFWS personnel. The sites were located at offshore fishing grounds away from point sources of contaminants. The fish (60 per collection site) were placed into plastic bags, frozen whole, and transported to the USFWS, Great Lakes Fishery Laboratory in Ann Arbor, Michigan where they were sorted into small (300–450 mm), medium (451–650 mm), and large (≥650 mm) size categories (20 fish per category) for each collection site and year. Within each size category the fish were grouped into four composite samples consisting of five fish each

Fig. 1. Sample collection sites

and homogenized in a Hobart™ vertical chopper/mixer. Subsamples of the homogenized tissue were weighed and stored frozen at < -30°C in solvent-washed glass jars with foil-lined screw caps until analyzed by the USEPA Central Regional Laboratory (CRL), Chicago, Illinois.

At CRL, the samples were thawed, dried with Na_2SO_4, and lipids and contaminants were column extracted with two 50-ml aliquots of 10% ethyl acetate in petroleum ether (Hesselberg and Johnson 1972). Lipids were separated from the extract by gel permeating chromatography (Stalling *et al.* 1972). Pesticides and PCBs were separated on a column of silica gel (Snyder and Reinert 1971). Final separation and quantitation of pesticides and PCBs was by a Varian 3700 dual column gas chromatograph with Ni^{63} electron capture detectors (CRL 1982) under the following operating conditions:[1]

	Column A	Column B
Column length	2.4 m × 2 mm i.d.	2.4 m × 2 mm i.d.
Column materials	4% SE-30/6% SP2401 on Supelcoport 100–120 mesh	3% OV-101 on Supelcoport 80–100 mesh
Injection temperature	270°C	270°C
Oven temperature	200°C	200°C
Detector temperature	320°C	320°C
N_2 carrier gas flow	30 ml/min	30 ml/min
Range (amps/mv)	10^{-10} amp	10^{-10} amp

All results were computed on a mg/kg wet weight basis and were not corrected for extraction or recovery efficiency.

As initially designed, the sampling program was intended to compare contaminant levels in fish, both temporally and spatially, in the three size categories of fish collected from each location. The results were to be compared by analysis of covariance techniques. This approach requires that a significant correlation exist between contaminant concentrations and the size of fish. If differences are to be tested, the regressions of contaminant levels and fish size must also have similar slopes. The data for 1977–1979 from Lakes Michigan, Huron, and Superior were initially analyzed by using this approach. Since data for three years, 13 organic contaminants and three sites were included in the analysis, 117 regressions between contaminant concentration and fish size were possible. Of these, 63 were significant at the 95% confidence level. Among the significant regressions, several exhibited significant ($p < 0.05$) differences in slopes, either between years or collection sites. Thus, less than half of the available data met the requirements for the test. It was, therefore, decided that although the analysis of covariance technique may be worthy of further evaluation, the use of mean statistics with specific size ranges of fish was the most appropriate approach for analyzing available data and determining differences in contaminant concentrations between sites and within sites over time.

To achieve a comparable size range of fish, from 1977 through 1981 and into future years (current collections consist of fish 600–700 mm long), we used the following procedure. The mean length of all medium and large size group composite samples from each site and year was calculated. When this mean was below 620 mm or above 640 mm, the composite sample with the smallest or largest length was dropped and the mean recalculated. This procedure was repeated until we arrived at a mean

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[1] ™Mention of trade names or commercial products does not constitute endorsement or recommendation by USEPA or USFWS.

length within the 620 mm to 640 mm range. The composite samples selected were treated as environmental replicates and mean statistics were used to compare contaminant concentrations. Table 1 presents the fish size and lipid data for the samples thus developed.

For purposes of statistical analyses and graphical display, we calculated concentrations of contaminants detected, but below quantitation limits, as ½ of the limits given in Table 2.

Results

Polychlorobiphenyls (PCBs) were the dominant contaminant at all sites reflecting its once wide spread use (Durfee 1976), environmental persistence, and tendency to bioaccumulate (EPA 1983). The continued presence of DDT residues illustrates the duration of contaminant problems long after measures to ban manufacture and use are enacted. Lake Michigan lake trout generally contained the highest contaminant concentrations and Lake Superior the lowest. Clark *et al.* (1984) and DeVault and Weishaar (1984) found a similar pattern in the relative contaminant concentrations in coho salmon in 1980 and 1982. Studies by Frank *et al.* (1980, 1981) found the sediment concentrations of PCBs, chlordane plus heptachlor, and DDT to be similar in Lake Michigan and Huron and substantially lower in Lake Superior.

Lake Michigan

Table 3 presents the annual means and 95% confidence intervals for contaminants in lake trout off Saugatuck, Michigan in 1970–1982. Before 1977, individual fish were analyzed by the Great Lakes Fishery Laboratory by the methods described in Willford *et al.* (1976). Since 1977 five fish composite samples were analyzed by USEPA.

Mean PCB concentrations (mg/kg) increased from 12.86 in 1972 to 22.91 in 1974, and declined to 5.63 in 1982. The decline in PCB concentrations from 1974–1982 followed first order loss rate kinetics in the form of the following equation:

$$C_t = C_0 e^{-kT} \qquad (1)$$

where C_t = concentration at time, C_0 = initial concentration, K = rate constant, and T = Time

Figure 2 illustrates the loss trajectory based on the least squares fit ($r^2 = 89.6$, $P < 0.01$) to the mean yearly PCB concentrations. The rate constant is −0.18/yr.

Mean total DDT concentrations (mg/kg) declined from 19.19 in 1970 to 2.74 in 1982. The observed decline in mean total DDT concentrations also followed first order loss rate kinetics (equation 1). Figure 3 illustrates the loss trajectory from 1970 to 1982. The rate constant is −0.13/yr with $r^2 = 90.9$ ($P < 0.01$).

Table 1. Lake trout sample collection data

Lake and Year	N	Length (mm) mean SE	Weight (g) mean SE	Lipid % mean SE
Lake Michigan[a]				
Saugatuck				
1970	18	613 (9)	2084 (149)	25.63 (1.69)
1971	20	579 (8)	2314 (125)	18.03 (0.68)
1972	9	648 (15)	2576 (178)	18.52 (1.81)
1973	30	602 (10)	2353 (120)	16.00 (0.70)
1974	30	616 (10)	2516 (130)	16.50 (0.56)
1975	29	613 (8)	2581 (92)	19.10 (0.55)
1976	30	606 (9)	2337 (103)	16.80 (0.49)
1977	3	636 (53)	2888 (681)	16.59 (0.88)
1978	8	631 (27)	2447 (311)	17.50 (0.76)
1979	6	630 (40)	2844 (461)	16.78 (1.10)
1980	6	637 (18)	2843 (296)	17.68 (0.74)
1981	3	620 (37)	2646 (497)	17.96 (0.88)
1982	10	612 (4)	2466 (47)	16.53 (0.33)
Lake Huron				
Rockport				
1978	6	637 (18)	2522 (210)	19.19 (0.65)
1979	5	640 (21)	3017 (308)	21.30 (0.78)
1980	6	633 (18)	2674 (230)	19.03 (1.10)
1981	6	628 (28)	2775 (343)	18.88 (0.64)
1982	10	623 (7)	2527 (84)	18.10 (0.71)
Lake Superior				
Apostle Island				
1977	7	640 (27)	2927 (425)	16.46 (1.80)
1978	7	638 (27)	3080 (400)	21.14 (1.20)
1979	6	630 (24)	2797 (312)	21.80 (1.80)
1980	7	636 (24)	2868 (360)	15.10 (1.30)
1981	5	634 (29)	2762 (386)	15.92 (0.93)
1982	10	619 (7)	2479 (103)	15.67 (0.43)

[a] Data for 1970–76 from the U.S. Fish and Wildlife Service, Great Lakes Fishery Laboratory, collected before the Cooperative Program began, and are based on analyses of individual whole fish

Table 2. Polychlorobiphenyls (PCBs) and pesticide detection limits

Compound	Quantitation Limits mg/kg
o,p'-DDE	0.002
p,p'-DDE	0.002
o,p'-DDD	0.005
p,p'-DDD	0.005
o,p'-DDT	0.020
p,p'-DDT	0.002
Dieldrin	0.002
Total Aroclor® (as 1254)	0.030–0.050
Heptachlor	0.001
Oxychlordane	0.005

Table 3. Mean contaminant concentrations in lake trout from Lake Michigan off Saugatuck, mg/kg wet weight (95% confidence intervals in parenthesis)

Year	PCBs[a]	DDT	Oxychlordane	Dieldrin	Number of samples[b]
1970	NA[c]	19.19 (15.90, 22.46)	NA[c]	0.27 (0.22, 0.32)	18
1971	NA	13.00 (11.24, 14.76)	NA	0.20 (0.17, 0.23)	20
1972	12.86 (8.11, 17.61	11.31 (8.05, 14.57)	NA	0.20 (0.14, 0.26)	9
1973	18.93 (16.85, 21.01)	9.96 (8.60, 11.32)	NA	0.27 (0.24, 0.30)	30
1974	22.91 (19.18, 26.64)	8.42 (6.68, 10.16)	NA	0.30 (0.27, 0.33)	30
1975	22.28 (19.38, 25.18)	7.50 (6.25, 8.75)	NA	0.35 (0.32, 0.38)	29
1976	18.68 (16.02, 21.34)	5.65 (4.61, 6.69)	NA	0.30 (0.28, 0.32)	30
1977	11.58 (−2.30, 25.4)	6.34 (−0.40, 13.10)	0.230 (−0.71, 0.531)	0.40 (0.35, 0.45)	3
1978	8.18 (5.50, 10.90)	4.58 (2.32, 6.83)	0.175 (0.072, 0.278)	0.44 (0.28, 0.61)	8
1979	8.82 (5.20, 12.50)	6.91 (2.80, 11.00)	0.240 (0.136, 0.344)	0.58 (0.25, 0.91)	6
1980	9.93 (2.00, 17.80)	4.74 (1.90, 7.50)	0.160 (0.039, 0.280)	0.335 (0.20, 0.47)	6
1981	6.49 (4.51, 8.46)	3.22 (2.11, 4.34)	0.061 (0.049, 0.073)	0.26 (0.24, 0.28)	3
1982	5.63 (5.36, 5.89)	2.74 (2.34, 3.14)	0.075 (0.057, 0.093)	0.21 (0.16, 0.28)	10

[a] PCBs from 1972–76 quantitated using 1:1:1 Aroclor® 1248, 1254, 1260 and beginning in 1977 only Aroclor® 1254

[b] From 1970–76 samples were individual fish; from 1977–1982 samples were composites of 5 fish each

[c] NA = Not Analyzed

Mean dieldrin concentrations (mg/kg) decreased from 0.27 in 1970 to 0.20 in 1971, then increased to 0.58 in 1979 after which concentrations declined to 0.22 in 1982. The increase in mean concentrations between 1972 and 1979 was highly significant ($P < 0.01$, $R^2 = 86.6$) following the linear equation:

$$C_t = 0.045\,(Y) - 3.02 \qquad (2)$$

where C_t = mean dieldrin concentration (mg/kg) in year t and Y = year (1900's) from 72–79

From 1979 to 1982 mean concentrations declined, following first order loss kinetics (equation 1), with a loss rate of −0.32/yr. The curve on Figure 4 illustrates the loss trajectory based on a least squares fit to the data ($r^2 = 93.2$, $P < 0.05$).

The chlordane metabolite, oxychlordane, increased from 1977 through 1979 and decreased from 1979 through 1982. The decrease (Figure 5) in mean concentration from 1979 to 1982 was significant ($P < 0.05$, $R^2 = 85.1$) following the linear equation.

$$C_t = 0.059\,(y) + 4.92 \qquad (3)$$

where C_t = mean oxychlordane concentration (mg/kg) in year t and y = year (1900's) from 79 to 82.

Lake Huron

Unlike Lake Michigan, where comparable trend monitoring began as early as 1970, the Lake Huron data base began in 1978 (Table 4). Despite general declines in the mean concentrations of PCB and DDT from 1979 to 1982, no statistically significant

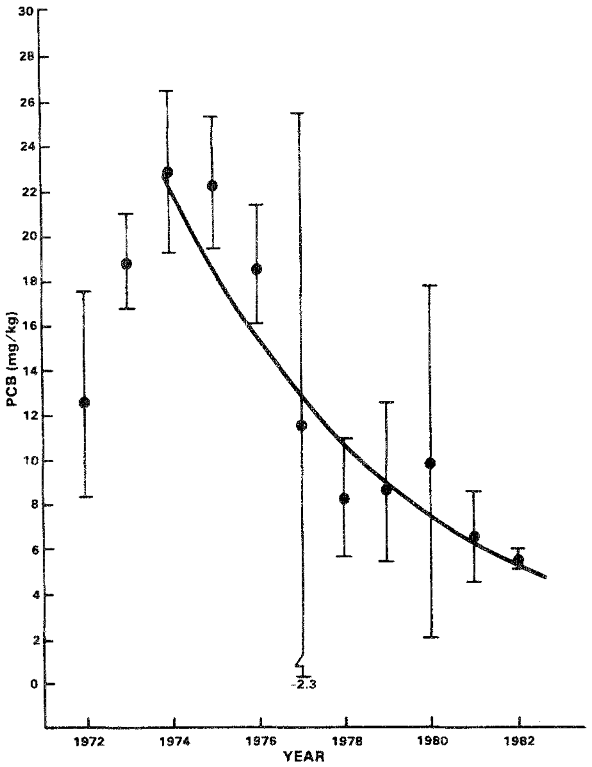

Fig. 2. Polychlorobiphenyl (PCB) concentrations in Lake Michigan trout. Mean and 95% confidence interval

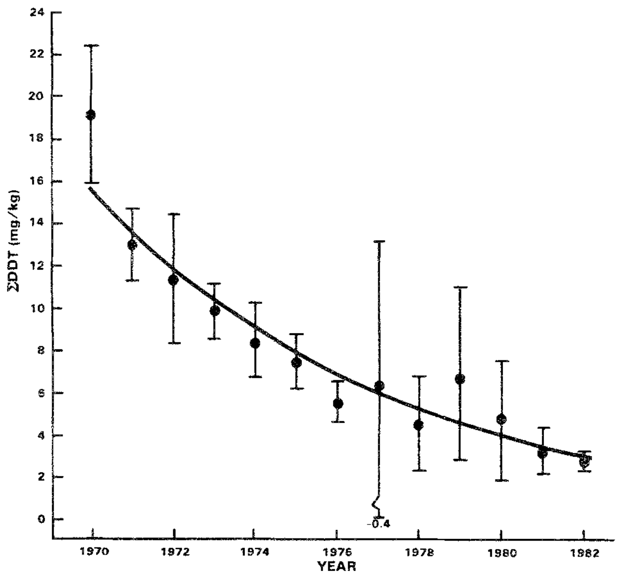

Fig. 3. Total DDT concentrations in Lake Michigan lake trout. Mean and 95% confidence interval

($P < 0.05$) trends were observed for any of the monitored compounds. Annual mean total DDT and PCB concentrations exceeded the IJC (1978) objectives (1.0 mg/kg and 0.1 mg/kg, respectively) in each year of the study.

Lake Superior

Contaminant concentrations in Lake Superior lake trout are given in Table 5. The relatively low levels of PCB, dieldrin, and oxychlordane did not exhibit trends nor statistically significant ($P < 0.05$) differences over the study period.

Total DDT concentrations in Lake Superior lake trout (Figure 6) declined significantly ($P < 0.05$, $r^2 = 78.9$) from 1977 to 1982, following the equation:

$$C_t = -0.162\,(y) + 13.7 \qquad (4)$$

where C_t = mean total DDT concentration (mg/kg) in year t and y = year (1900s).

The annual mean PCB concentration exceeded the IJC (1978) objective in each year of the study. The IJC (1978) total DDT objective was exceeded by the mean concentrations in 1977 through 1979, but not from 1980 to 1982.

Discussion

Of the data sets for the three upper lake monitoring sites reported here, only that for Lake Michigan was sufficient to allow rigorous statistical evaluation of trends with time. This is principally because USFWS had collected data for six years before the current US EPA/US FWS cooperative program was begun in 1977. As a result of these extra years of data, it was possible to demonstrate that mean PCB and mean total DDT concentrations in lake trout off Saugatuck declined in a manner that approximated first order loss kinetics. Rodgers and Swain (1983) reported that the decline of PCB concentrations in bloaters (*Coregonus hoyi*) from Lake Michigan (off Saugatuck) from 1972 to 1980 also approximated first order loss kinetics with a rate constant of −0.12/yr. They used the trend data for bloaters to hindcast loads and then used their loading estimates to forecast concentrations in other fish species. They estimated that PCB concentrations in Lake Michigan trout would reach 2 mg/kg in 1987. By solving equation (1) for t, we calculate that PCB concentrations will drop to 2 mg/kg in 1988. This estimate is in excellent agreement with the projections of Rodgers and Swain (1983), particularly when the difference in approach and the uncertainties of such projections are considered. These projections presuppose that PCB loading into Lake Michigan will continue to decrease at a rate at least equal to that before 1982. As atmospheric deposition now contributes well over 50% of the total load (Murphy and Rzesutko 1977), this decrease may not be affected.

A similar projection of total DDT concentration

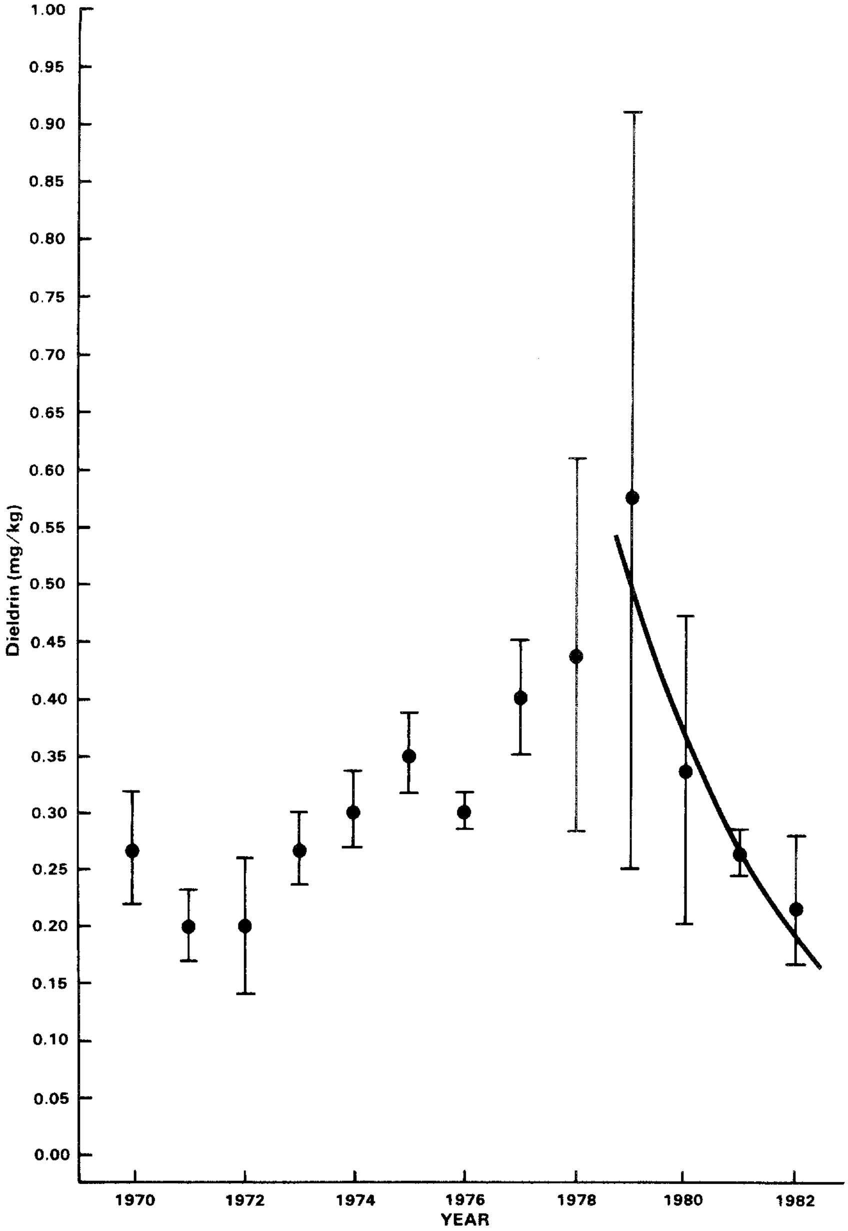

Fig. 4. Dieldrin concentrations in Lake Michigan lake trout. Mean and 95% confidence interval

suggests that mean concentrations will reach the IJC objective of 1.0 mg/kg in about 1991. Despite cancellation of most uses of aldrin and dieldrin in 1974 (EPA 1980), mean concentrations in lake trout in Lake Michigan off Saugatuck increased from 0.27 mg/kg in 1970 to 0.58 mg/kg in 1979. From 1979 to 1982 mean dieldrin concentrations declined in a manner approximating first order loss kinetics. However, other species such as bloater chubs and rainbow smelt have not exhibited the decline observed in lake trout. Dieldrin concentrations in bloaters in Lake Michigan increased from a mean of 0.27 (mg/kg) in 1970 to 0.43 in 1982 (Willford 1982), while rainbow smelt remained relatively constant (range 0.046–0.071 mg/kg) from 1977 to 1982 (GLNPO-unpublished data). Elevated dieldrin concentrations resulted in seizure of commercial catches of Lake Michigan bloaters by the U.S. Food and Drug Administration in 1984 (David Crockett, U.S. Food and Drug Administration, Chicago, Illinois, personal communications, 1984). Therefore, while the apparent decline in lake trout is encouraging, it will require additional years of monitoring to test this trend.

With the exception of DDT, which declined significantly in Lake Superior lake trout from 1977 to 1982, no significant changes in contaminant concentrations were observed in lake trout from Lake

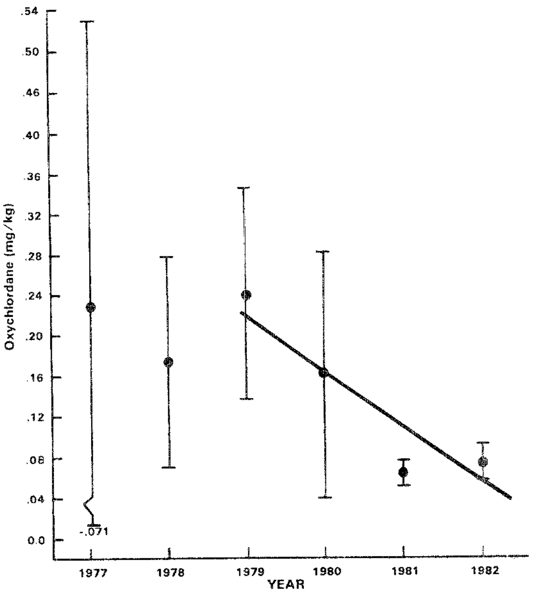

Fig. 5. Oxychlordane concentrations in Lake Michigan lake trout. Mean and 95% confidence interval

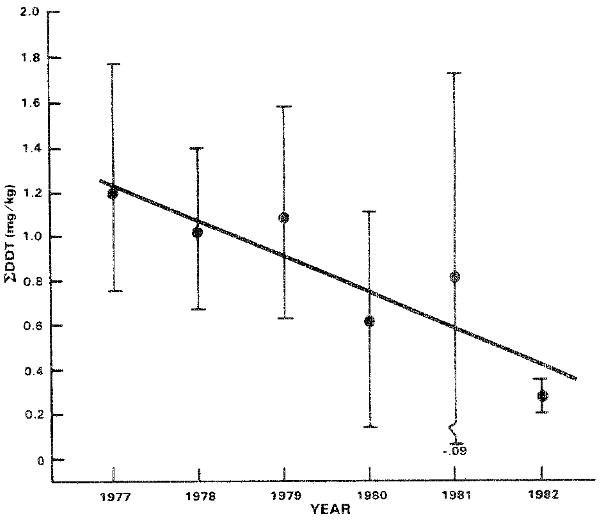

Fig. 6. Total DDT concentrations in Lake Superior lake trout. Mean and 95% confidence interval

Table 4. Mean contaminant concentrations in lake trout from Lake Huron off Rockport, mg/kg wet weight (95% confidence interval)

Year	PCBs[a]	DDT	Oxychlordane	Dieldrin	Number of samples
1978	2.92 (2.32, 3.52)	2.19 (2.05, 2.33)	0.038 (0.033, 0.043)	0.16 (0.14, 0.17)	6
1979	3.66 (1.57, 5.75)	2.85 (1.94, 3.76)	0.060 (0.024, 0.097)	0.19 (0.14, 0.24)	5
1980	3.44 (2.46, 4.42)	1.53 (1.10, 1.96)	0.056 (0.039, 0.073)	0.13 (0.10, 0.15)	6
1981	3.51 (2.31, 4.72)	1.79 (1.15, 2.43)	0.058 (0.040, 0.076)	0.14 (0.12, 0.16)	6
1982	2.10 (1.72, 2.49)	1.31 (1.13, 1.49)	0.059 (0.044, 0.074)	0.15 (0.11, 0.18)	10

[a] PCB quantitated as Aroclor® 1254

Table 5. Mean contaminant concentrations in lake trout from Lake Superior off the Apostle Islands, mg/kg wet weight (95% Confidence Interval)

Year	PCBs[a]	DDT	Oxychlordane	Dieldrin	Number of samples
1977	1.87 (0.95, 2.80)	1.2 (0.76, 1.80)	0.120 (0.059, 0.180)	0.05 (0.03, 0.06)	7
1978	0.93 (0.65, 1.21)	1.02 (0.66, 1.39)	0.402 (0.110, 0.700)	0.06 (0.05, 0.07)	7
1979	0.88 (0.59, 1.17)	1.095 (0.61, 1.57)	0.136 (0.102, 0.375)	0.07 (0.05, 0.08)	6
1980	1.89 (0.41, 3.36)	0.62 (0.13, 1.11)	0.026 (0.018, 0.035)	0.045 (0.02, 0.06)	7
1981	1.40 (0.37, 2.44)	0.81 (−0.09, 1.71)	0.056 (0.001, 0.111)	0.05 (0.03, 0.07)	5
1982	0.48 (0.36, 0.60)	0.29 (0.21, 0.47)	0.041 (0.036, 0.047)	0.05 (0.04, 0.05)	10

[a] PCB quantitated as Aroclor® 1254

Huron or Lake Superior. The combination of limited data sets, large data variance, and relatively lower contaminant concentrations interfered with the detection of trends. The ineffectiveness of the study design used from 1977 to 1981 (sampling three size ranges of fish), and the resulting requirements to select fish over a large size range and greatly reduce the number of yearly samples, was partly responsible for the large variance associated with the 1977 to 1981 data. Although the covariance approach will be further evaluated, the monitoring protocol used since 1982 for this cooperative program utilizes mean statistics on results from 10 composite samples of 5 fish each, all of which are 600 to 700 mm long. The reduced variance under this more stringent sampling protocol will greatly enhance the likelihood of detecting significant differences between any two sites or years when contaminant concentrations are changing.

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