

Trends of Chlorinated Organic Contaminants in Great Lakes Trout and Walleye from 1970 to 1998

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Abstract

Levels of chlorinated organic contaminants in predator fish have been monitored annually in each of the Great Lakes since about the 1970s. This paper updates earlier reports with data from 1991 to 1998 for lake trout (*Salvelinus namaycush*) and walleye (*Stizostedion vitreum vitreum*, Lake Erie only), providing a record that now extends to nearly 30 years. Whole fish were analyzed for a number of industrial contaminants and pesticides, including PCBs, DDT, dieldrin, toxaphene and mirex, and contaminant trends were quantified using multi-compartment models. As in the past, fish from Lakes Michigan, Ontario and Huron remain have the highest levels of PCBs, DDT and dieldrin; Superior has the highest levels of toxaphene, and Ontario has the highest levels of mirex. In the period following curtailment of chemical use, concentrations rapidly declined, represented by a short half-life (generally <2 yr), followed by a considerably more gradual decline, represented by a longer half-life (generally from 8 to 18 yr), and/or an irreducible (constant) concentration. While trends depend on contaminant and lake, in many cases the rate of decline has gradually decreasing. For dioxin-like PCBs, levels have not been declining over the most recent period (1994-8). In cases, year-to-year variation in contaminant levels is large, largely due to food-web dynamics. While this variation sometimes obscures long-term trends, the general pattern of a rapid decline followed by slowing or leveling-off of the downward trend is consistent across the Great Lakes, and future improvements of the magnitude seen in the 1970s and early 1980s will likely take much longer.

Keywords

Great Lakes, lake trout, pesticides, PCBs, DDT, walleye.

Introduction

It is well now recognized that levels of chlorinated hydrocarbons, metals and other contaminants in the Great Lakes basin pose significant concerns to human and ecosystem health. Two examples of the continuing challenges posed by the presence of contaminants in the basin are fish consumption advisories due to contaminant residues in fish that exceed guidelines for human consumption, and the disposal of contaminated sediments from channel dredging and site-cleanup activities. Information regarding the levels and trends of contaminants in the Great Lakes basin is needed to control the use and disposal of contaminants, to set water quality standards and fish consumption advisories, and to understand the transport and fate of pollutants, including the development of ecosystem models.

This paper provides updated information regarding the trends of chlorinated organic contaminants in predator fish in all five Great Lakes through 1998. It builds on earlier studies (De Vault et al. 1985; 1986; 1996), which provided monitoring results for 1970 through 1990 or 1992. With few exceptions, sampling and analysis protocols over this nearly 30 year-long period remained consistent, thus this time series record provides an unparalleled and unique opportunity to understand the dynamics of persistent contaminants, especially after the manufacture or use of the chemicals has been curtailed. Given the global distribution of these chemicals, this information is relevant to both the Great Lakes and to other ecosystems.

Background

Long-term efforts to track contaminant levels were initiated by the Great Lakes Science Center which began monitoring contaminants in fish from Lake Michigan in 1966. In 1977, the Center (then called the Great Lakes Fishery Laboratory) in cooperation with the US EPA Great Lakes National Program Office (GLNPO) expanded monitoring to include all of the Great Lakes in order to address basin-wide contamination problems. This monitoring program was continued through 2003 under the auspices of a USEPA/GLNPO and most recently, a US Geological Survey/GLSC Cooperative Agreement (USEPA/USGS 1997).

In the basin, levels of many contaminants tend to be lowest in fish from Lakes Superior and Erie and highest in Lakes Michigan and Ontario, due to the generally larger loadings from agricultural, municipal, industrial and airborne sources. There are a few notable exceptions to this generalization, e.g., toxaphene levels have been highest in Lake Superior. Large declines observed in levels of DDT, PCBs and other contaminants in

the 1970s and 1980s (DeVault et al. 1985, 1986, 1996) followed soon after restrictions or bans on chemical use and manufacture. DDT use was banned in 1969-70; PCBs were placed under voluntary control in 1971 and manufacturing banned in 1977; toxaphene use was banned in 1982; most uses of aldrin and dieldrin, which were widely used from the 1950s to the early 1970s, were banned in 1975 and these chemicals are no longer produced in or imported into the USA (EPA 1980); and commercial use of chlordane was banned in 1988. However, contaminant levels may no longer be decreasing, or may be decreasing so slowly that levels appear to be static (Borgmann and Whittle 1983, Sun et al. 1993, Hebert et al. 1994; De Vault et al. 1986). Concentration trends can significantly vary by site and contaminant, e.g., Borgmann and Whittle (1991) show marked gradients of some contaminants in Lake Ontario, thus site-specific data are needed. Also, since contaminant levels can change dramatically from year-to-year, lengthy records are needed to discern long-term trends.

Methods

Sampling and chemical analysis. Sampling locations and protocols for fish collection and methods for contaminant extraction from tissues are described in DeVault et al. (1985, 1986, 1996). In brief, 10 sites, two for each lake, were selected in 1977 to represent US-held open water areas of the five Great Lakes, all sites being in or near deep-water (Figure 1). We also report on data collected in 1991 and 1993 at a third Lake Michigan site (Charlevoix). Because of the lakes' size, sites were balanced between those with and without influences from major tributaries and industries. Beginning in 1982, sites were sampled in alternate years (Table 1). A top predator fish was sampled, specifically, lake trout (*Salvelinus namaycush*) from Lakes Superior, Michigan, Huron and Ontario, and walleye (*Stizostedion vitreum vitreum*) from Lake Erie. In most cases, 10 composite samples representing 50 fish from each site and each year that met length specifications were analyzed.

For analysis, a 10-gram sub-sample was taken from each homogenized composite of five whole fish, analytes were solvent-extracted from fish tissue, lipids were removed by gel permeation chromatography, and analytes were separated by column chromatography. Gas chromatography/mass spectroscopy (GC/MS) analyses were performed using negative chemical ionization and selective ion monitoring, an HP Model 5890 GC/MS, and a DB-5 30 m × 0.25 mm id × 0.25 µm thickness capillary column (Schmidt and Hesselberg 1992; Schmidt 1997). Blanks, spikes, recoveries, and duplicates were performed with each batch of 10 samples.

Target compounds were not detectable in blanks. Surrogate spike recoveries were ~90%, and duplicate sample analyses agreed within $\pm 15\%$. Extensive quality assurance/quality control, including analysis of audit samples, was used to guarantee data comparability through time and across laboratories. Method detection limits (MDLs) for 99% confidence, defined as 3 times the standard deviation of 7 replicates of a procedural blank spiked with a very low level of analyte (Keith 1991), are presented in Table 1.

Fish tissue was analyzed for the following contaminants: PCBs (both total and individual congeners), total DDT (p,p'-DDT, p,p'-DDE, and p,p'-DDD), chlordanes (*cis*-chlordane, *trans*-chlordane, oxychlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide B), aldrin, dieldrin, endrin, lindane, mirex, and toxaphene (polychlorinated camphene mixture). *Trans*-chlordane data were measured during 1991-1993, but not during 1994-1998. Total PCBs, total DDTs, total chlordane, and total toxaphene were calculated following Schmidt and Hesselberg (1992) and Schmidt (1997). Total chlordane did not include *trans*-chlordane, since it was only measured from 1991-1993. Total DDT did not include p,p'-DDD, because it was not measured in Lake Superior prior to 1993 and it was not measured at Port Austin (Lake Huron) in 1991. Total PCBs did not include congeners #81/87, 123 and 167 because they were only measured from 1994-1998. PCB congener numbering follows Ballschmiter and Zell (1980).

Data analysis. Analyses emphasized total PCBs, total dioxin-like PCBs, total DDTs, total chlordane (sum of *cis*-chlordane, oxychlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide B), dieldrin, and toxaphene. Dioxin-like PCB congeners were selected for analysis because they are the ones primarily associated with PCB toxicity. The toxic equivalent index (TEQ), which represents toxicity due to the binding of the Ah receptor, is expressed as a 2,3,7,8-tetrachlorodibenzo-p-dioxin equivalents for the PCB mixture by summing weighted concentrations of 11 congeners (77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 189) using the toxic equivalency factor (TEF) weights for humans from WHO (1997). Of these, 126 was the most toxic congener, and 105 was the most abundant congener. Congener 169, also considered dioxin like, was not measured. Because only the most recent (1994 – 1998) data includes congeners #81/87, 123 and 167, for trending purposes, dioxin-like PCBs excluding these congeners are also calculated, called TEQ(8).

Because contaminant trends in each lake differ from one another, analyses are performed individually by site. Differences between means, variances and trends between site pairs within a lake were evaluated.

Trends in contaminant concentrations in the earlier (1970-1991) data have been presented as a first-order decrease (DeVault et al. 1985, 1986, 1996). Here we extend this approach, modeling the downward trend over the extended (1970-1998) data record using a two compartment first-order model with an asymptote

$$C_t = C_0 + C_1 [f \exp(-k_1 t) + (1-f) \exp(-k_2 t)] \quad (1)$$

where C_t = concentration in fish (ng/g) at time t (year from start of the decline in the available record), C_0 = an irreducible or baseline concentration (ng/g), C_1 = the declining concentration (ng/g), f = fraction of C_1 attributable to the fast process ($1-f$ = fraction attributable to the slow process), and k_1 and k_2 = first-order rate constants for fast and slow processes, respectively (yr^{-1}). This model simulates up to three contaminant sources. Two sources decline at fast and slow first-order rates. Eventually, the slow process becomes dominant over the fast process. As these sources become depleted, concentrations eventually approach an irreducible level C_0 , due to the presence of a third, stable contaminant source. Similar models have been used by Stowe et al. (1994; 1995) for PCB data in Lake Michigan. Parameters C_0 , C_1 , f , k_1 and k_2 were estimated to best fit the observations using a quasi-Newton gradient search method so as to minimize deviations between predicted and observed concentrations. Due to the non-normal distribution of concentrations, data were log-transformed for optimization. Also, estimated parameters were constrained to be ≥ 0 , f was constrained between 0 and 1, and a single compartment model was forced if $k_1 \approx k_2$ or if f was near 1 or 0. Zero intercepts C_0 were allowed, thus eliminating the stable (third) source. Model results were plotted using a log-linear scale, which yields a straight line if a single compartment first-order model with no intercept applies.

Results

A summary of selected physical and chemical data for 1991-1998 GC/MS contaminant analyses of annual fall predator fish is given in Table 2. Not shown are several chemicals that are now below MDLs in most fish tissues samples, including aldrin, endrin and lindane.

Within lake differences. The 1991-8 data showed no statistically significant differences in variances among concentrations collected at the paired sites at each lake (F tests, $p > 0.05$). Two-sample t tests showed that mean concentrations may have differed between sites in two lakes for a few contaminants. In Lake Huron, dieldrin measurements were 33% higher at Port Austin as compared to Rockport ($p = 0.04$); mirex was 116% lower but this result had marginal significance ($p = 0.09$) and concentrations were low (3 ± 3 ng/g) and often

below MDLs; and toxaphene was 64% higher but again with marginal significance ($p = 0.06$). Notably, levels of dieldrin and toxaphene dramatically decreased during this time period and the site differences appear due simply to the staggered sampling. In Lake Michigan, Saugatuck tended to have higher concentrations than Sturgeon Bay for total PCBs ($p = 0.10$, 29% difference) and dieldrin ($p = 0.09$, 42% difference), but again the statistical significance is marginal. No other contaminants showed statistically significant or near-significant differences in mean concentrations within site pairs in these or other lakes. With some caveats, this simplifies the analysis as the data from pairs of sites can be pooled. It should be noted that spatial differences with a lake may be significant in the short-term, e.g., differences in PCB levels in fish collected from Sturgeon Bay and Saugatuck were found to be significantly different in 1994-5 (Madenjian 1989).

Initially, trends in each lake are analyzed separately by combining earlier measurements (De Vault et al. 1996) with the data collected here. Figures 2 to 6 show annual mean levels and standard deviations for five contaminants, along with long-term trends estimated using first-order models. Model parameters and statistics, e.g., fit (R^2) and half-life, are listed in Table 3. Then, results are discussed by contaminant class. Percentage changes in concentrations over the two most recent 4-year periods (1991-4 and 1995-8), are in Table 4.

Lake Michigan. Contaminant trends in lake trout in Lake Michigan are depicted in Figure 2. Lake Michigan fish have had and continue to have some of the highest concentrations of organochlorine contaminants in the basin, and the monitoring record is most complete for this lake. For these reasons and to discuss the modeling in more depth, results for this lake are discussed in some detail.

PCBs. PCB levels in Lake Michigan (along with Lake Ontario) are the highest in the Great Lakes. Concentrations declined from the maximum levels of 23000 ± 5000 ng/g observed in 1975 to 1800 ± 170 ng/g in 1998, and levels fell below the 2000 ng/g FDA action level (highest legally permitted residue level for a particular contaminant in food) in 1996. PCB levels continue to decline, though at a slower rate. The first-order model, which obtained a high fit ($R^2 = 0.96$), has rate constants of 0.35 and 0.051 yr^{-1} for fast and slow processes, respectively, equivalent to half-lives of 2.0 and 13.6 yrs, respectively (Table 3). The slow process, which is relevant for trends after about 1980, has a half-life of 13.6 yr. No irreducible or “background” level of PCBs was indicated (i.e., $C_0 = 0$). In comparison to an earlier using a 1-compartment model and data prior to 1992 where $t_{1/2} = 5.8 \text{ yr}$ and $R^2 = 0.86$ (De Vault et al. 1996), these results provide a higher fit and, more

importantly, quantify that the rate of PCB removal in Lake Michigan is declining, e.g., mean PCB levels will not drop to 1000 ng/g (half the FDA action level) until 2010. Of course, extrapolations can be faulty and lead to large errors, though the PCB trend in Figure 2 appears quite robust.

DDT. Total DDT levels in fish in Lake Michigan, the highest among the Great Lakes, follow trends similar to PCBs but with several nuances. First, the peak DDT levels likely occurred prior to 1970, before the monitoring record. Second, model results show a minor ($f = 0.11$) and very fast process ($t_{1/2} = 0.05$ yr), but most of the trend is controlled by the slow process ($t_{1/2} = 3.9$ yr) and, most importantly, is governed by an irreducible concentration ($C_0 = 864$ ng/g), meaning that future reductions will be both slow and small. For example, total DDT levels are predicted to decline by only 100 ng/g from 1998 levels to reach 880 ng/g in 2010, and after another 10 years levels will drop only to 866 ng/g, essentially reaching C_0 . No further reductions are predicted. While the model fit was very high ($R^2 = 0.97$), such extrapolations can be fraught with error. Still, the key result, a declining rate of DDT removal, is clear.

Pesticides. Oxychlorane levels peaked in 1979 or possibly earlier (data prior to 1977 is unavailable) and its current half-life is 6.2 yrs. Dieldrin levels also peaked in 1979 and the decline is slightly slower ($t_{1/2} = 8.6$ yrs). In most years (especially 1992 and 1995), dieldrin levels in Lake Michigan were much higher than levels measured in the other lakes. Toxaphene levels peaked in 1990 and while the decline was rapid, the short data span does not permit robust model estimates. These three compounds (or groups) show considerable fluctuations from year-to-year. Most mirex levels were below MDLs.

Lake Erie. In this lake, contaminant data come from walleye, a relatively lean fish (about half the fat content of the lake trout). Since sampling in the other lakes used lake trout, contaminant concentrations are not directly comparable, e.g., concentrations in walleye were generally much lower than those in the trout. Still, trends in walleye in Lake Erie follow the same general pattern as in Lake Michigan, although the variation around the trend line is greater, and most contaminants show an especially rapid (but possibly temporary) decline from a 1992 to 1995 (Figure 3). PCBs levels are gradually diminishing with a similar half-life ($t_{1/2} = 14$ yr) as observed in Lake Michigan. DDT levels are declining somewhat more rapidly, but fluctuations above (1986, 1988) and below (1982) the trend line are notable. Dieldrin also shows a comparable half-life ($t_{1/2} = 12$ yr), again with some large variations from the long-term trend, especially a high level in 1984 corresponding to

slightly larger and fattier fish (10% lipid) for that decade, and a low level in 1995 corresponding to the lowest (7.2%) lipid content observed. In contrast, oxychlordan levels are dropping more slowly in Lake Erie fish than in Lake Michigan, though data again are scarce.

Lake Huron. Trends of most contaminants in lake trout showed considerable short-term variation superimposed on a long term decline (Figure 4). The first-order models provided good fits for total DDT and dieldrin trends ($R^2 > 0.88$). PCBs and DDT levels slightly increased from 1995-1998, corresponding with a dramatic increase in lipid content (from 8 to 16%) in fish collected over this period. Since 1986, trends of PCB levels have been more or less constant, resulting in a very long half-life ($t_{1/2} = 26$ yr) rather than an irreducible level (i.e., $C_0 = 0$ ng/g). However, essentially an identical fit ($R^2 = 0.69$) could be obtained by setting $C_0 = 1000$ ng/g and changing other parameters ($f = 0.554$, $K_1 = 0.229$ giving $t_{1/2} = 3.0$ yr, and $K_2 = 0.086$ giving $t_{1/2} = 8.1$ yr). Irreducible levels of DDT and toxaphene are indicated, but the significance of the latter is questionable due to the short record. DDT levels have been more or less stable from 1988 – 1998, and further declines beyond the irreducible level ($C_0 = 571$ ng/g) are not predicted.

Lake Ontario. As mentioned, lake trout in Lake Ontario (along with those in Lake Michigan) showed the highest PCB levels. The first-order models provided good fits to long-term trends, and concentrations appear to be declining at a steady first-order rate (Figure 5). Single compartment models with an irreducible level are indicated for oxychlordan and toxaphene, but the record for both contaminants is short and the intercept is small. The peak dieldrin level of 340 ± 76 ng/g measured in 1979 corresponds to the heaviest and fattiest (lipid content = 22%) fish sampled in this lake. Also, the 1977 – 1981 fish were smaller (0.50 – 0.56 m length) than those sampled subsequently (0.61 to 0.65 m), though the lipid content was generally comparable.

Mirex levels in Lake Ontario trout averaged 234 ± 71 ng/g over the 1991-8 period, and a gradual concentration decrease was observed, e.g., 38% reduction from 1991-4 to 1995-8 averages ($p = 0.03$, Table 4). Levels consistently exceeded the 100 ng/g FDA action level. Historically, these mirex levels are similar or slightly higher than the 1977-1993 average of 207 ng/g at Main Duck Island near Oswego (Huestis et al. 1996), possibly due to diet changes. In contrast, mirex was infrequently detected in the other lakes, and maximum concentrations (10 - 14 ng/g in Lake Superior in 1991-3) were much lower.

The discovery of elevated levels of mirex in Lake Ontario fish during the 1960s triggered lake-wide fish

consumption advisories some of which still exist today (US EPA 1998). The use and production of mirex, also known as dechlorane and chlordecone, are now banned in North America. Mirex has been identified as a Lakewide Management Plan (LaMP) critical pollutant because levels in some Lake Ontario fish continue to exceed human health standards. Most of the mirex originated from a production facility on the Niagara River, and most (~1.8 kg/yr) still enters Lake Ontario from this river. Smaller amounts (~0.9 kg/yr) enter via the Oswego River, and ~0.7 kg/yr leaves Lake Ontario via the St. Lawrence River (Huestis et al. 1996). No reliable estimates of atmospheric deposition or volatilization for mirex are available. Although most widely known for its use as a pesticide, ~75% of the mirex produced was used as a flame retardant in industrial, manufacturing and military applications. Available sales records suggest that more than 50,000 pounds of mirex were used for industrial and manufacturing flame retardant purposes in the Lake Ontario basin, and >75,000 pounds were used as a flame retardant in other Great Lakes basins (Huestis et al. 1996).

Lake Superior. Contaminant levels in lake trout in Lake Superior have varied tremendously over the years (Figure 6). Some of the variation may be attributable to the slightly longer and fattier fish collected in 1977 – 1981 (lipid content = 15– 22%), and the fattier fish collected in 1991-3 (lipid content = 21 – 25%). Most of the variation, however, is caused by changes in the diet and food web. In the late 1980s, the diet of lake trout in Lake Superior changed from relatively lean and small slimy sculpins (*Cottus cognatus*) and rainbow smelt (*Osmerus mordax*) to larger, longer-lived and much more lipid-heavy lake herring (*Coregonus artedii*; Bronte and Hoff 1999; Gallinat and Bronte 1995), as confirmed by a stable-isotope study (Whittle et al. 1997). (These and other changes in the fish community in Lake Superior are discussed by Bronte et al. 2003). The herring diet dramatically increased contaminant intake and accumulation in lake trout. For example, compared to the 1986-9 average, levels in 1991-3 increased for total PCBs by 280%, total dioxins by 210%, dieldrin by 70%, oxychlorane by 340%, and toxaphene by 230%. These dietary changes appeared to occur earlier than 1993, as offered by Whittle et al. (1997) in explaining toxaphene trends. Both we and Swackhamer (1998; 1999) reexamined the original extracts and confirmed the elevation of all contaminants during 1991-1993. During 1993-4, the diet began to shift again to an even leaner diet than before, which included insects and particulate matter with only large lake herring and relatively few smelt available (Bronte and Hoff 1999; Gallinat and Bronte 1995). This dramatically dropped levels of lipids and contaminants in the lake trout, e.g., 1994-8

averages fell back to the 1986-9 averages, and even lower for dieldrin and toxaphene.

Due to the year-to-year fluctuations, contaminant levels in Lake Superior trout show the greatest divergence from the estimated long-term trends, and model fit was especially poor for total PCBs, dieldrin and oxychlordanes ($0.06 \leq R^2 < 0.41$). The first-order models indicate that trends in total PCBs, total DDT, and oxychlordanes have been flat since 1986. However, more recent data show statistically significant declines when 1991-3 (1994 data are not available) and 1995-8 data are compared (Table 4). As discussed above, much of this is due to the large drop in lipid content over this period (from $23 \pm 12\%$ to $12 \pm 3\%$ using 3- and 4-yr averages) and the dietary changes over this period.

As noted earlier, Lake Superior has had the highest toxaphene levels in the basin, especially in 1991 when levels reached 1250 ± 150 ng/g, and the greatest decrease during the late 1990s (Swackhamer et al. 1998; 1999). Unlike other hydrophobic organic contaminants, the atmospheric balance of toxaphene is dominated by gas exchange across the air-water. High concentrations of toxaphene in Lake Superior may result from the lake's low temperature, which reduces losses from volatilization, and its lower sedimentation rate, which reduces transport to sediment (Swackhamer et al. 1998; 1999). The first-order model for toxaphene in Lake Superior shows a half-life of 1.8 yr, comparable to that seen in Lake Michigan (Table 3). While these times are longer than those observed for toxaphene in the other lakes, the dietary changes and short data record precludes definitive analysis.

Persistence by chemical. Levels in the top fish predators in the basin continued to gradually decline for most contaminants, often with quite similar rates when expressed as a half-life. Still, there were important differences between lakes and chemicals.

PCBs. The half-life of total PCBs ranged from 9.3 (Lake Ontario) to 27 (Huron) yrs, excepting Superior which poorly fit the first-order model ($R^2 = 0.34$). Two compartment models best fit the data in all five lakes, and Lake Superior also showed a sizable irreducible term ($C_0 = 502$ ng/g). These results differ from previous analyses that showed little change in PCB concentrations in Lake Erie since 1982 and in Lakes Superior, Michigan, Huron and Ontario since 1986 or 1988 (De Vault et al. 1996). The first-order models and trend plots show that concentrations are declining, though at a decreasing rate, in Lakes Michigan and Ontario, and that levels remain relatively high (1820 and 1420 ng/g, respectively). The trends are much less definitive for Lakes

Huron, Erie and Superior over the late 1980s and 1990s, however, when concentrations fell below ~1500 ng/g and concentrations show sizable year-to-year variability. This variation reflects several influences, e.g., variation in fish range and diet (discussed earlier), and possibly sampling and analytical variation. We speculate that PCB concentrations in Lakes Michigan and Ontario will also increase in variability and possibly plateau as levels decrease to 1000 ng/g and below. Also, trends may vary for individual components of the PCB mixture (see below).

Comparing recent trends of PCBs using 4-year averages (Table 4), total PCB levels declined in all lakes, sometimes dramatically, although differences were not statistically significant for Lakes Erie and Michigan. It should be recognized that 8 years of data does not allow a statistically robust analysis of trends, e.g., 1994 peaks of certain PCBs and DDT at Lake Michigan highly influenced this analysis.

Another key aspect of PCB concentrations concerns the toxicity of the PCB mixture. Again comparing 4-yr periods, TEQs for PCBs declined in fish from Lakes Erie, Michigan and Superior, though only the latter was statistically significant, but TEQs increased in fish from Lakes Huron and Ontario, though changes were not statistically significant (Table 4). On average, 52% of the toxicity comes from 3,3',4,4',5-PeCB (PCB #126), which has the greatest weight (TEF), 15% comes from 2,3',4,4',5-PeCB (PCB #118), 13% from 2,3,3',4,4',5-HxCB (#156), and smaller amounts from other congeners. The TEQs are somewhat underestimated since one congener (3,3',4,4',5,5'-HxCB, #169) was not measured, and three congeners (3,4,4',5-TCB (#81), 2,3',4,4',5'-PeCB (#123), and 2,3',4,4',5,5'-HxCB (#167)) were not measured in 1991-3. However, these provide small contributions to TEQ and the exclusion of the latter did not alter results. Also, congener #126 could not be determined in most samples in 1993 due to a laboratory contamination issue. Recent trends in TEQs largely follow levels of congener #126 (Figure 8), in which increases (though not statistically significant) are observed in four lakes (Table 4). This congener is present at low levels (generally ≤ 2 ng/g), though well above MDLs (0.02 – 0.1 ng/g). Its measurements demonstrate substantial variation, both from year-to-year and within years (shown by the error bars on Figure 8). Other dioxin-like PCB congeners showed downward trends, except for PCB#81 (which was measured along with PCB#87). Thus, while total PCB concentrations appear to be declining in most lakes, PCB toxicity, which is driven largely but not exclusively by congener #126, is not decreasing. This demonstrates the importance of examining trends of individual components in mixtures such

as PCBs, especially for chemicals that are more persistent, bioavailable and/or bioaccumulative.

Aldrin and dieldrin: Aldrin and dieldrin are the common names of two insecticides that are closely related chemically with similar toxicities. Aldrin is readily converted to dieldrin in the environment, so these two closely related compounds are considered together by regulatory bodies. As mentioned, aldrin was below MDLs for all 1991-1998 samples. Levels of dieldrin, which probably peaked in the late 1970s in most lakes, had half-lives ranging from 8.5 yr (Michigan and Ontario) to 12 yr (Erie). (Lake Superior is excluded from this analysis due to poor model fit, $R^2 = 0.06$). Again, two compartment models best fit the data in all lakes (except Superior). Comparing 1991-4 and 1995-8 averages, dieldrin levels declined by 30 – 49% in four lakes, and by 86% in Lake Superior, changes which achieve or approach statistical significance in all cases (Table 4). The decline continues the trend reported by DeVault et al. (1996) who reported generally similar half-lives (from 6.3 yr for Erie to 11.6 yr for Michigan, again excluding Superior where the fit was very poor). The longer data record used here obtained higher model fits ($0.76 \leq R^2 \leq 0.92$, again excepting Superior) than those reported by De Vault et al. (1996).

Chlordane. Technical grade of chlordane is a mixture of ~50 compounds with the major constituents identified as *cis*- and *trans*-chlordane (38-48%), heptachlor (3-13%), *cis*- and *trans*-nonachlor (5-11%), and other chlordane isomers (17-25%) (USEPA 1998). Chlordane is primarily metabolized to oxychlordane. Heptachlor, which is also a separate pesticide, is metabolized to heptachlor epoxide. Chlordane is an unlikely source of exposure if heptachlor epoxide is found in the absence of either oxychlordane or *trans*-nonachlor. Based on the 1991-8 data at the five lakes, *trans*-nonachlor is the predominant component, accounting for $43 \pm 4\%$ of total chlordane. Of the other components, *cis*-nonachlor accounted for $22 \pm 4\%$, *cis*-chlordane for $17 \pm 5\%$, oxychlordane for $10 \pm 3\%$, and *trans*-chlordane for $3 \pm 3\%$. *Trans*-chlordane levels often dropped below the MDL, especially in recent years. Across the lakes, these fractions were consistent except that Lake Erie walleye had the highest levels of both *cis*-chlordane ($25 \pm 4\%$) and *trans*-chlordane ($5 \pm 5\%$), but the lowest levels of oxychlordane (5.4 ± 0.8). Also, Lake Superior trout had somewhat lower fractions of *cis*-chlordane ($12 \pm 2\%$) than the other lakes. Overall, this behavior represents a bioaccumulated and “weathered” technical chlordane mixture.

Over the available record (1977 – 1988 in most lakes), oxychlordane levels dropped rapidly, with half-

lives from 3.2 (Erie) to 6.2 (Michigan and Huron) yrs. Declines were slower in Lake Superior though the model fit was poor ($R^2 = 0.41$). Declines tend to be steeper for the constituents of technical chlordane. In the more recent period, the *cis*-chlordane fraction diminished, e.g., from $20 \pm 1\%$ in 1991 to 12 ± 1 in 1998 for lake trout in Lakes Huron, Michigan and Ontario, while *cis*-nonachlor and oxychlordane levels slightly increased.

In contrast to PCBs, dieldrin and the chlordanes, the trend models suggest that the declines in total DDT and toxaphene may have slowed, as discussed below.

DDT. Irreducible levels, specifically, 257 ng/g in Superior, 571 ng/g in Huron, and 864 ng/g in Michigan suggest are estimated. While no irreducible level was estimated for Erie and Ontario, DDT trends show long half-lives, 10 and 18 years, respectively. These results may occur due to contaminant cycling in the environment, and/or a large, essentially infinite reservoir of this contaminant. To examine recent (1991-8) trends in depth, DDT and metabolites DDE and DDD are shown in Figure 7. The typical composition of DDT was 80-90% p,p'-DDE, 8-15% p,p'-DDT, and 2-5% p,p'-DDD. Comparing 1991-4 and 1995-8 periods, total DDT levels increased by 37% in Lake Michigan ($p = 0.02$) and by 7% in Lake Huron ($p = 0.24$), while levels declined by 66% in Lake Superior ($p = 0.01$), 44% in Lake Erie ($p = 0.02$), and 19% in Lake Ontario ($p = 0.05$). Trends of metabolite p,p'-DDE, which comprises the bulk of total DDT, are similar. However, generally downward trends are seen for p,p'-DDT and p,p'-DDD.

Toxaphene. The ~670-member toxaphene group is made up of octachloro (~50% of the total), and hexa- and heptachloro homologues (~40%). The first-order models indicated irreducible levels of toxaphene in all five lakes, namely, specifically 13, 120, 152, 185 and 484 ng/g in Lakes Erie, Ontario, Michigan, Superior and Huron, respectively. These models are based on relatively few data points (e.g., $n = 6$ for Lake Erie), and there is considerable scatter around the trend line for all the lakes except Ontario. Therefore, the toxaphene trend models are considered to have little predictive ability. Considering the 1991-8 period, large (65 – 152%) and statistically significant reductions were seen for 4-yr averages in all five lakes, however (Table 4). Overall, these trends are similar to those reported by Glassmeyer et al. (1997) and Glassmeyer (1998).

Discussion

The nearly 30-yr record of contaminant levels in the Great Lakes shows both long- and short-term trends. For most contaminants in the basin, the long-term trend is clearly a decline in concentration following

restrictions or bans on chemical use. In many cases, however, the rate of decline is slowing. This was quantified using multi-compartment models that show in the period following curtailment of use, concentrations rapidly decline as represented by a short half-life, generally <2 yr, followed by a considerably more gradual decline, represented by a longer half-life, generally from 8 to 18 yr, and/or an irreducible (constant) concentration. Slowing declines also have been observed in fish in Canadian waters of the Great Lakes (Baumann and Whittle 1988). In cases, short-term contaminant fluctuations cause large year-to-year variations that may obscure long term trends, but these long-term trends are consistent across the basin.

Much of the short-term fluctuation in contaminant levels appears to be due to food-web dynamics and energetics. These processes play a large role in the uptake of PCBs and other persistent and lipophilic contaminants in predator fish such as lake trout and walleye. Biomagnification is the principal accumulation mechanism for species in the upper levels of the food web (Thomann and Connoly 1984; Oliver and Nimi 1988; Rasmussen et al. 1990; Madenjian et al. 1993, 1995). While prey species can have comparable lipid levels and contaminant loadings, the energetics of forage and consumption can vary among species. Thus, a switch between species can dramatically alter predator contaminant exposure (Stow et al. 1995a; 1995b). Dietary changes documented in Lake Superior lake trout, for example, caused rapid and increases followed by decreases (factor of 2 – 4) in contaminant levels in the space of a few years. However, these diet-based fluctuations are not indicative of long-term trends, and are not on the scale of the declines observed in the 1970s.

Organochlorine contaminant levels in fish have not been consistently correlated with lipid contents, e.g., Stow et al. (1997) found size, age, diet, and environment to be more important than lipid content in lake trout. As shown in Lake Superior trout, the lipid content of fish may change with dietary changes, thus a correlation is expected. After correcting for the long term trend, we found that the correlation between fish lipid content was small. For example, using for Lake Michigan lake trout, the correlation between model residuals (observation – prediction) and lipid content was only 0.09 for total PCBs, -0.07 for total DDT, 0.43 for dieldrin, 0.04 for oxychlordan, and 0.00 for toxaphene. (The correlation for dieldrin drops to 0.26 when an outlier is removed.) Thus, lipid content alone accounts for only a few percent of the variation in contaminant concentrations.

Conclusions

Although concentrations of most contaminants in predator fish in the Great Lakes have continued to

decline during the 1990s, the rapid declines in concentrations observed through the 1970s and 1980s have slowed, and based on multi-compartment first-order models, controlling processes have half-lives on the order of a decade or two. Concentrations appear to have stabilized for several pollutants, including PCB #126, the most toxic dioxin-like PCB congener. Contaminant levels remaining in top predator fish remain a cause of concern, as reflected in the fish consumption advisories that may be expected to continue for some time. Short-term changes in contaminant levels are explained by chemical usage and dietary changes in the fish community. Long-term trends depend on chemical transport and fate processes at the basin, regional and even global scale, as well as bioavailability in the food web. Lengthy data records are needed to explain trends and predict future contaminant levels, thus continued monitoring is needed in each lake.

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Table 1 Method detection limits (MDLs) for target compounds.

PCBs - chlorination level		Pesticides and other organochlorines	
Compound	MDL (ng/g)	Compound	MDL (ng/g)
Tri-	4	Pentachlorobenzene	0.15
Tetra-	0.2-25	Hexachlorobenzene	1
Penta-	0.03-6	Octachlorostyrene	0.5
Hexa-	0.01-0.1	p,p-DDT	40
Hepta-	0.02-0.1	p,p-DDE	12
Octa-	0.04-0.3	P,p-DDD	70
Nona-	0.05-0.2	Heptachlor epoxyde	10
Deca-	0.04	oxychlordane	2
		A-chlordane	2
		Cis-nonachlor	0.5
		Trans-nonachlor	3
		Dieldrin	0.5
		Endrin	0.5
		Aldrin	3.5
		Lindane	1
		Mirex	2
		Total toxaphene	100

Table 2 Physical and contaminant measurements in Great Lakes lake trout and walleye during 1991-1998. Each sample was a composite of five whole fish, averages are shown.

Year	Site	Sample Size	Lipid (%)	Length (mm)	Tot. PCB (ng/g)	Non-ortho PCBs (ng/g)	PCB#126 (ng/g)	PCB TEQ (8) (pg/g)	PCB TEQ (11) (pg/g)	Total DDT (ng/g)	Total Chlordanes (ng/g)	Dieldrin (ng/g)	Mirex (ng/g)	Toxaphene (ng/g)
Erie														
1991	Dunkirk	10	12.5	492	945	67.7	0.4	54.8	54.8	138.1	109.0	31.4	0.0	470.0
1992	Middle Bass Isle	10	8.7	487	2190	120.1	0.0	18.9	18.9	123.6	81.7	32.4	0.0	446.3
1993	Dunkirk	5	10.9	523	1210	67.9	0.0	10.6	10.6	150.2	91.5	25.4	0.0	398.9
1994	Middle Bass Isle	4	9.0	474	625	10.6	0.0	2.7	3.6	88.0	39.9	17.9	0.0	50.1
1995	Dunkirk	9	7.2	429	400	9.3	0.2	21.4	22.6	77.2	30.6	11.7	0.0	25.1
1996	Middle Bass Isle	10	10.0	476	773	15.1	0.1	8.5	9.8	76.3	57.2	19.3	0.7	83.5
Huron														
1991	Port Austin	10	19.8	691	2475	373.9	1.0	147.0	147.0	652.0	248.0	77.3	2.6	1695.0
1992	Rockport	10	17.9	630	1575	206.8	0.1	39.9	39.9	519.8	253.4	60.4	2.4	1029.0
1993	Port Austin	10	18.6	668	2250	305.4	0.0	41.9	41.9	551.1	232.9	65.3	2.3	1319.0
1994	Rockport	10	17.9	611	1215	96.6	0.8	93.5	97.2	672.0	140.2	49.5	0.0	429.7
1995	Port Austin	9	8.4	634	985	66.2	0.8	92.2	96.1	521.3	130.5	60.8	0.0	525.5
1996	Rockport	10	12.2	638	1006	70.0	0.5	60.5	62.4	667.1	146.2	44.6	6.8	450.5
1997	Port Austin	10	16.2	631	1213	96.5	1.0	111.0	114.9	671.7	145.4	52.4	0.0	918.4
1998	Rockport	10	15.4	617	1251	90.9	1.8	192.1	197.5	703.8	107.8	28.2	9.4	376.3
Michigan														
1991A	Charlevoix	10	19.4	638	1580	198.5	0.8	104.8	104.8	663.3	259.6	162.1	0.3	1450.0
1991	Sturgeon Bay	10	16.0	640	2145	243.4	1.1	141.1	141.1	763.8	295.2	96.9	0.3	1230.0
1992	Sagatuck	9	17.9	635	3433	366.5	1.0	147.0	147.0	1096.4	446.2	193.3	5.3	1744.4
1993A	Charlevoix	10	18.9	631	1785	194.6	0.4	64.1	64.1	602.4	260.9	134.7	0.0	1131.3
1993	Sturgeon Bay	4	16.2	656	2900	343.1	0.0	46.3	46.3	967.1	312.0	101.3	0.0	1300.0
1994	Sagatuck	9	16.7	621	2717	321.4	3.4	388.0	394.0	1589.0	248.3	109.5	0.0	362.4
1995	Sturgeon Bay	10	16.4	651	1677	105.8	0.5	66.5	70.4	1193.7	340.3	143.4	0.0	447.1
1996	Sagatuck	10	17.8	648	1916	95.0	0.7	87.5	90.5	1065.7	282.8	110.2	0.0	335.5
1997	Sturgeon Bay	10	14.4	624	1651	82.2	0.6	72.7	76.6	971.7	158.8	70.0	0.0	268.2
1998	Sturgeon Bay	10	15.6	633	1821	141.5	0.8	97.9	104.2	1137.1	132.0	49.6	0.9	303.3
Ontario														
1991	N. Hamilton	10	17.3	632	3720	441.1	0.0	66.6	66.6	1118.5	291.4	93.0	360.0	1250.0
1992	Oswego	10	16.4	636	2630	322.7	0.0	46.7	46.7	839.1	214.5	79.8	272.0	964.4
1993	N. Hamilton	10	18.2	626	3100	359.0	0.0	49.8	49.8	905.7	202.7	64.8	281.0	786.5
1994	Oswego	9	16.7	631	2082	136.6	0.8	104.0	109.1	1135.3	166.7	62.4	200.9	270.4
1995	N. Hamilton	10	20.6	636	1687	76.4	0.3	39.3	41.2	798.8	150.6	58.2	205.6	183.6
1996	Oswego	10	19.7	633	1679	109.6	0.5	67.0	71.4	834.3	97.5	57.3	146.2	198.0
1997	N. Hamilton	10	17.7	636	1528	104.1	0.6	76.9	79.5	912.4	103.8	34.7	155.1	147.4
1998	Oswego	9	18.2	654	1421	64.8	1.7	184.1	187.9	745.0	134.6	43.7	248.1	152.3
Superior														
1991	Keewenaw Pt	10	23.9	540	1395	222.1	1.1	149.1	149.1	503.4	482.3	103.5	14.4	4935.0
1992	Apostle Isles	10	21.1	616	1083	148.1	0.6	83.6	83.6	357.2	371.4	78.0	9.5	3780.0
1993	Keewenaw Pt	10	24.8	614	1365	184.6	0.6	92.4	92.4	473.1	414.1	52.6	10.9	4480.0
1995	Keewenaw Pt	10	16.5	609	334	12.2	0.0	2.4	3.1	276.7	99.7	35.7	1.4	994.4
1996	Apostle Isles	10	12.2	651	401	17.9	0.0	3.1	3.9	219.0	164.3	36.3	0.0	1026.4
1997	Keewenaw Pt	9	11.9	650	354	12.5	0.0	2.6	3.5	256.0	127.4	24.3	0.0	669.4
1998	Apostle Isles	10	8.7	630	392	18.7	0.2	24.0	25.7	177.3	141.0	35.2	0.0	600.8

Table 3 Parameters of the first-order models for five contaminants in each lake.

Lake	Parameter	Starting Year	C ₀ (ng/g)	C ₁ (ng/g)	f -	K ₁ (yr ⁻¹)	K ₂ (yr ⁻¹)	R ² -	K ₁ half-life (yr)	K ₂ half-life (yr)
Michigan	Total PCB	1975	0	21671	0.735	0.345	0.051	0.96	2.01	13.62
	Total DDT	1970	864	18326	0.105	13.403	0.180	0.97	0.05	3.86
	Dieldrin	1979	0	580	0.377	22.000	0.081	0.86	0.03	8.57
	o-Chlordane	1977	0	230	0.216	13.378	0.111	0.83	0.05	6.23
	Toxaphene	1990	152	2040	1.000	0.376	-	0.77	1.84	-
Huron	Total PCB	1978	0	3693	0.455	0.254	0.027	0.69	2.73	25.88
	Total DDT	1978	571	2165	1.000	0.268	-	0.87	2.58	-
	Dieldrin	1978	0	175	0.000	0.074	-	0.88	9.33	-
	o-Chlordane	1978	8	54	1.000	0.111	-	0.73	6.23	-
	Toxaphene	1991	484	1266	1.000	0.668	-	0.70	1.04	-
Erie	Total PCB	1977	0	2660	0.260	0.794	0.048	0.62	0.87	14.49
	Total DDT	1977	0	430	0.472	0.443	0.072	0.70	1.56	9.69
	Dieldrin	1977	0	99	0.472	1.007	0.058	0.76	0.69	12.02
	o-Chlordane	1977	2	53	1.000	0.216	-	0.62	3.22	-
	Toxaphene	1991	13	719	0.973	0.841	-	0.66	0.82	-
Ontario	Total PCB	1977	0	8331	0.123	10.154	0.075	0.91	0.07	9.30
	Total DDT	1977	0	1791	0.017	0.650	0.039	0.74	1.07	17.89
	Dieldrin	1979	0	340	0.407	18.622	0.082	0.92	0.04	8.50
	o-Chlordane	1991	13	11	1.000	1.178	-	0.57	0.59	-
	Toxaphene	1991	120	1397	1.000	0.624	-	0.90	1.11	-
Superior	Total PCB	1977	502	1368	0.332	12.057	0.233	0.34	0.06	2.98
	Total DDT	1977	257	1165	1.000	0.357	-	0.79	1.94	-
	Dieldrin	1977	0	62	1.000	0.020	-	0.06	35.09	-
	o-Chlordane	1977	0	220	0.774	0.538	0.052	0.41	1.29	13.38
	Toxaphene	1991	185	5641	1.000	0.391	-	0.84	1.77	-

Table 4 Percentage decreases in fish lipid, length and contaminant concentrations, comparing 1991-4 and 1995-8 periods using 4-yr averages (%), and statistical significance based on 1-sided equal variance t-tests. Negative (-) change indicates increase in concentration. Statistically significant (p <0.05) changes in bold.

	Lipid	Length	Tot. PCB	Non-ortho PCBs	PCB#126	PCB TEQ (11)	Total DDT	Total Chlordanes	Dieldrin	Mirex	Toxaphene
Erie	17 (0.18)	9 (0.06)	64 (0.14)	112 (0.09)	-14 (0.47)	29 (0.38)	44 (0.04)	54 (0.10)	49 (0.06)	-300 (0.09)	117 (0.06)
Huron	35 (0.01)	3 (0.16)	51 (0.02)	101 (0.02)	-74 (0.09)	-36 (0.19)	-7 (0.24)	49 (0.01)	30 (0.06)	-76 (0.20)	65 (0.05)
Michigan	9 (0.07)	0 (0.38)	31 (0.05)	82 (0.00)	49 (0.24)	52 (0.18)	-14 (0.23)	27 (0.10)	34 (0.08)	110 (0.26)	101 (0.00)
Ontario	-11 (0.02)	-1 (0.08)	58 (0.01)	112 (0.01)	-116 (0.09)	-33 (0.24)	19 (0.04)	57 (0.01)	43 (0.01)	38 (0.03)	131 (0.01)
Superior	64 (0.00)	-7 (0.06)	120 (0.00)	193 (0.00)	200 (0.00)	192 (0.00)	66 (0.00)	113 (0.00)	86 (0.01)	218 (0.00)	152 (0.00)
Overall	17 (0.02)	-2 (0.29)	55 (0.00)	105 (0.00)	1 (0.49)	26 (0.19)	5 (0.39)	53 (0.00)	44 (0.01)	25 (0.35)	102 (0.00)

Table 5. 1991-8 average 2,3,7,8 TCDD toxic equivalents (TEQ) by lake, and apportionment by PCB congener.

	(ng/g)	Erie		Huron		Michigan		Ontario		Superior	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Coplanar PCBs - TEQ	(ng/g)	20	(18)	100	(54)	134	(111)	82	(48)	52	(57)
Apportionment of TEQ											
3,3',4,4'-TCB (PCB #77)	(%)	0.2	(0.3)	0.4	(0.3)	0.4	(0.2)	0.4	(0.4)	0.3	(0.6)
3,4,4',5-TCB (#81)	(%)	6.9	(9.4)	2.0	(1.7)	2.0	(2.5)	2.4	(2.3)	10.3	(11.2)
3,3',4,4',5-PeCB (#126)	(%)	36.5	(41.5)	64.6	(32.7)	66.2	(24.6)	48.2	(40.6)	42.3	(39.7)
3,3',4,4',5,5'-HxCB (#169)	(%)	-	-	-	-	-	-	-	-	-	-
2,3,3',4,4'-PeCB (#105)	(%)	8.3	(7.6)	7.2	(7.0)	6.8	(5.7)	9.5	(7.8)	6.8	(4.1)
2,3,4,4',5-PeCB (#114)	(%)	7.9	(9.7)	3.2	(2.3)	3.0	(1.8)	4.1	(2.5)	5.7	(5.0)
2,3',4,4',5-PeCB (#118)	(%)	16.4	(16.5)	14.0	(16.0)	12.9	(12.4)	20.2	(19.2)	12.5	(5.8)
2,3',4,4',5'-PeCB (#123)	(%)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)
2,3,3',4,4',5-HxCB (#156)	(%)	20.4	(12.9)	7.4	(6.9)	6.9	(4.2)	12.4	(9.9)	19.0	(14.6)
2,3,3',4,4',5'-HxCB (#157)	(%)	2.8	(3.0)	1.2	(1.7)	1.6	(1.8)	2.6	(3.9)	2.1	(1.6)
2,3',4,4',5,5'-HxCB (#167)	(%)	0.5	(0.8)	0.1	(0.1)	0.1	(0.1)	0.1	(0.2)	0.8	(1.0)
2,3,3',4,4',5,5'-HxCB (#189)	(%)	0.1	(0.1)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	0.1	(0.1)

Figure 1. Sampling sites for fall predator fish. Lake trout were sampled in most lakes, except for walleye in Lake Erie.

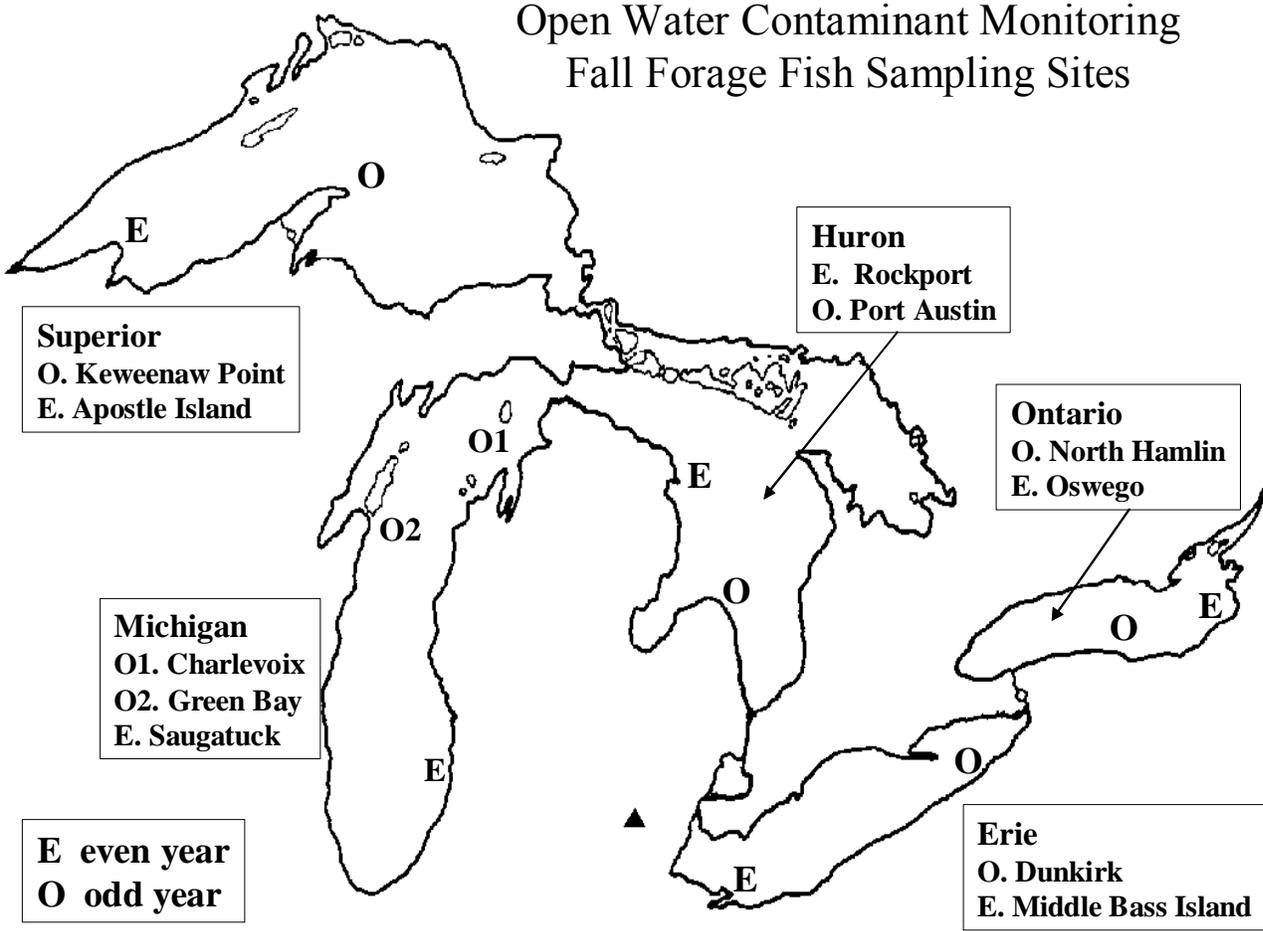


Figure 2. Long term trends of contaminants in Lake Michigan. Plots show mean concentration and standard deviation, trend lines from first-order models.

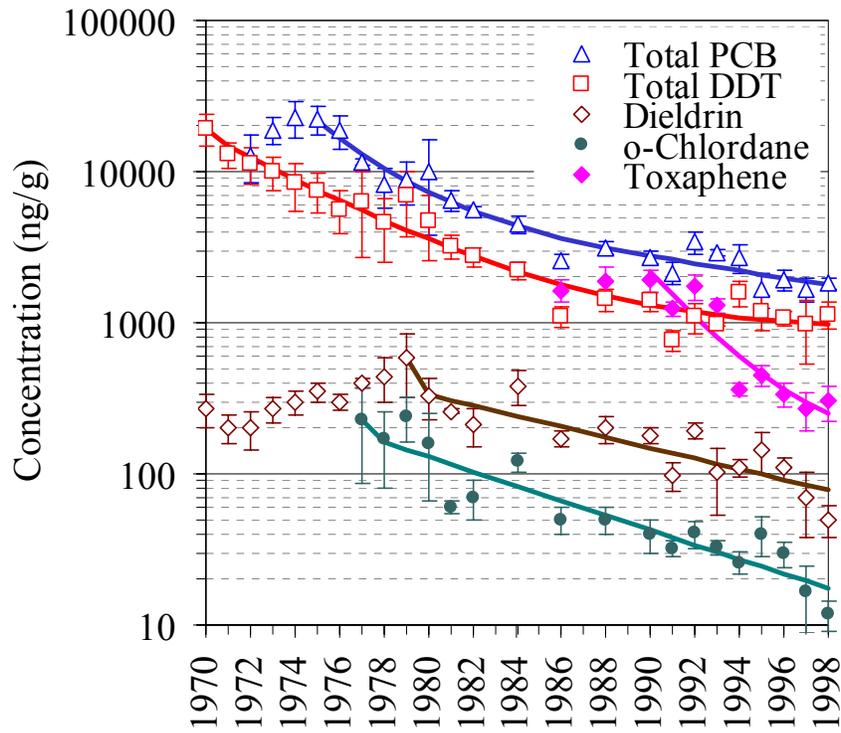


Figure 3. Long term trends of contaminant in Lake Erie. Otherwise as Figure 2.

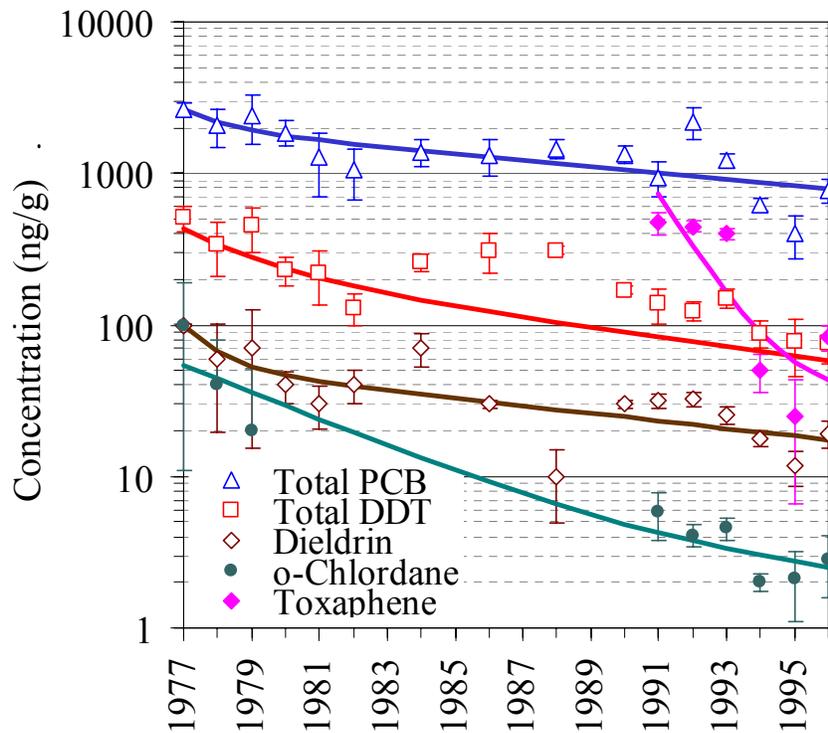


Figure 4. Long term trends of contaminants in Lake Huron. Otherwise as Figure 2.

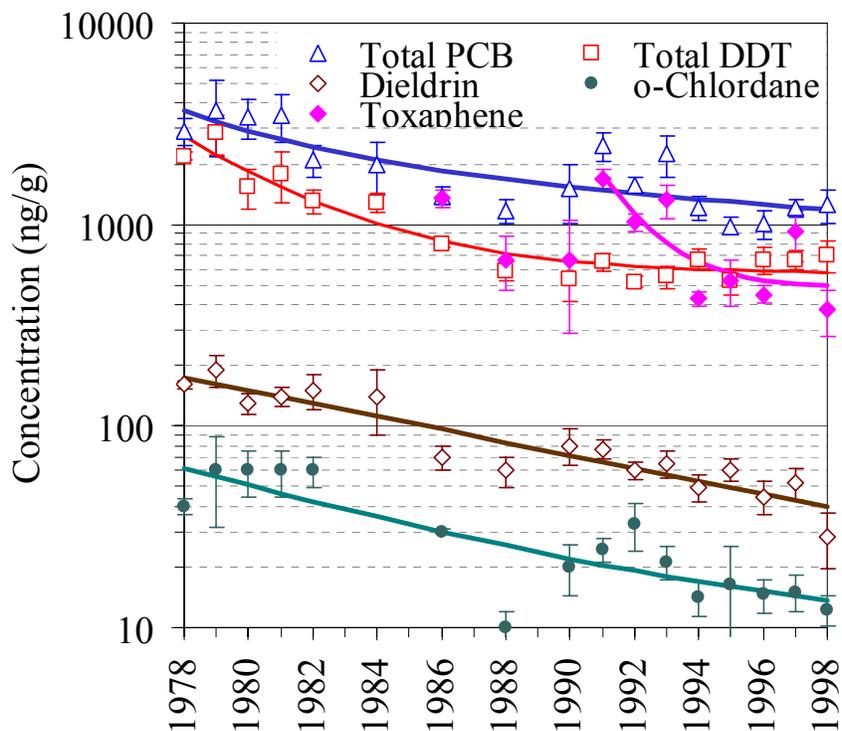


Figure 5. Long term trends of contaminants in Lake Ontario. Otherwise as Figure 2.

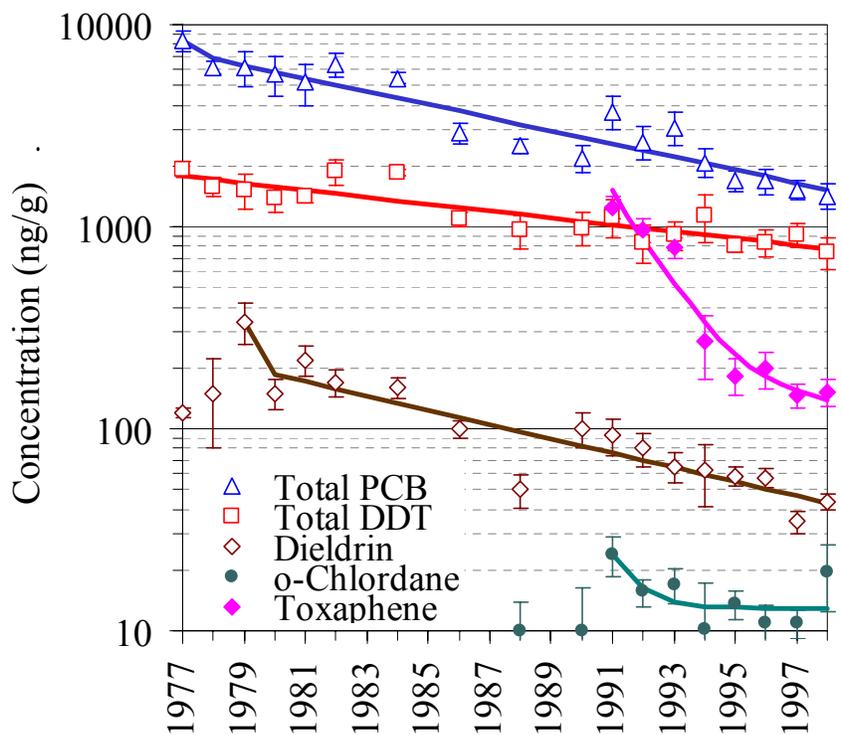


Figure 6. Long term trends of contaminants in Lake Superior. Otherwise as Figure 2.

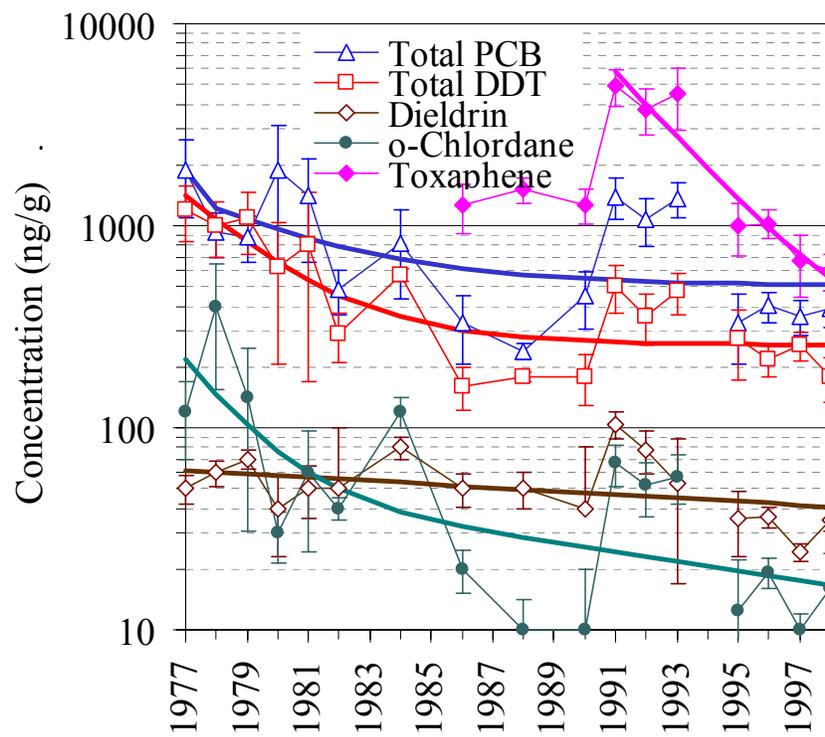


Figure 7. Recent trends of total DDT, DDE, DDD, and p,p'-DDT in the five Lakes.

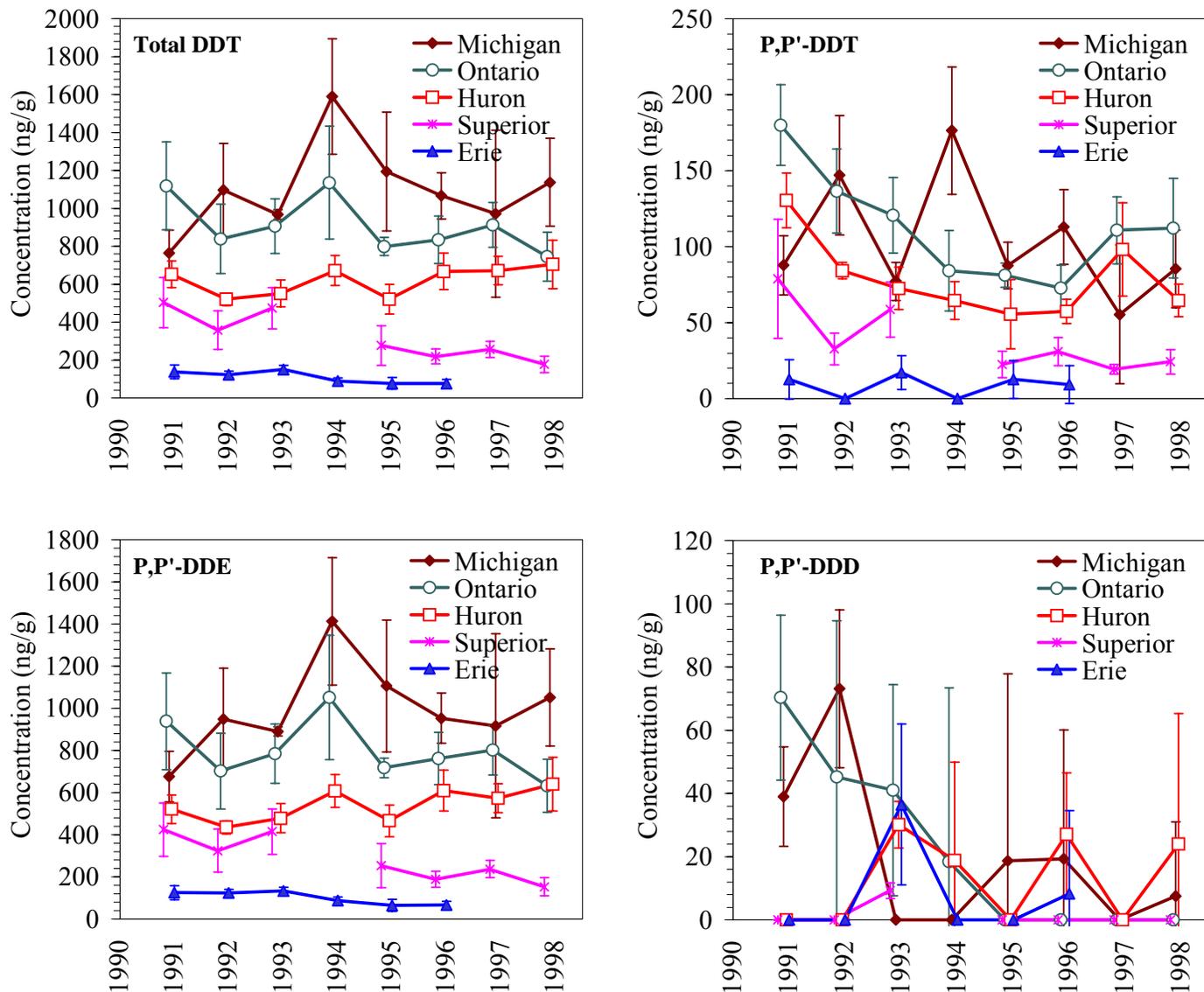


Figure 8. Recent trends of total PCBs, PCB TEQs, PCB#126 and PCB#118 in the five lakes.

