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DISTRIBUTION OF POLYCHLORINATED BIPHENYL (PCB) CONGENERS AND OTHER HALOCARBONS IN FISH AND MUSCLE AMONG LAKE ONTARIO SALMONIDS

A.J. Niimi[2] and B.G. Oliver[1]

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[1]Environmental Contaminants Division
National Water Research Institute
Canada Centre for Inland Waters
Burlington, Ontario, Canada L7R 4A6

[2]Great Lakes Laboratory for Fisheries and Aquatic Sciences
Department of Fisheries and Oceans
Canada Centre for Inland Waters
Burlington, Ontario, Canada L7R 4A6

. Environment Canada

Distribution of polychlorinated biphenyl (PCB) congeners and other halocarbons in fish and muscle among Lake Ontario salmonids

MANAGEMENT PERSPECTIVE AND EXECUTIVE SUMMARY

Ninety-two monochloro- to decachlorobiphenyl congeners were monitored in brown trout (Salmo trutta), lake trout (Salvelinus namaycush), small and large rainbow trout (Salmo gairdneri), and small and large coho salmon (Oncorhynchus kisutch) from Lake Ontario. Concentrations of individual congeners ranged from nondetectable levels, primarily among the monochloro- to trichlorobiphenyl congeners, to 1 $mg.kg^{-1}$ for 2,2',4,4',5,5'-hexachlorobiphenyl. Congeners with the higher concentrations were generally the pentachloro- and hexachlorobiphenyl homologues. Total congener concentration ranged from 1-10 $mg.kg^{-1}$ in fish, and 0.3-4 $mg.kg^{-1}$ in muscle. Consistent values of congener composition were shown when individual congener levels were expressed as percent of total concentration. The 10 most common congeners represented about 52% of total congener content, this value does not appear to be influenced by species or by total congener concentration. Concentrations of 4 non-ortho chloro substituted congeners , including 3,3',4,4'-tetrachlorobiphenyl, were also monitored. These four toxic congeners represented about 1.2% of the total concentration. The PCB homologues monitored in fish and muscle contained an average content of 56% chlorine by weight. Comparative analyses on the composition of congeners observed in fish with that reported for PCB mixtures suggest the more persistent congeners behave as a homogeneous mixture rather than as individual congeners.

Levels of 12 other organochlorine compounds were also monitored to examine the influence of lipid on chemical distribution between fish and muscle. A good correlation was shown between the relative distributions of lipid and chemicals between muscle and fish for all species except brown trout. The distribution of chemicals between fish and muscle was also examined to determine if the measurement of one can be used to estimate the concentration of the other. No consistent relationship was demonstrated that would allow this extrapolation to be applied to all species with confidence.

Distribution des congénères de polychlorobiphényles (PCB) et d'autres hydrocarbures halogénés dans le poisson entier et le muscle des salmonidés du lac Ontario

PERSPECTIVES DE GESTION ET RÉSUMÉ

On a mesuré 92 congénères de polychlorobiphényles (monochloro-à décachloro-) chez la truite brune (*Salmo trutta*), le touladi (*Salvelinus namaycush*), chez la petite et la grande truite arc-en-ciel (*Salmo gairdneri*) et chez le petit et le gros saumon coho (*Oncorhynchus kisutch*) dans le lac Ontario. La concentration des congénères individuels varie : de non détectable (surtout chez les congénères monochloro- à trichlorobiphényles) à 1 mg/kg dans le cas du 2,2', 4,4', 5,5' -hexachlorobiphényle. Les congénères les plus concentrés sont en général les homologues pentachloro- et hexachlorobiphényles. La concentration totale des congénères varie de 1 à 10 mg/kg dans le poisson entier et de 0,3 à 4 mg/kg dans le muscle. La composition des congénères est constante lorsqu'on exprime les congénères individuels en pourcentage de la concentration totale. Les 10 congénères les plus courants représentent environ 52 % de la concentration totale de congénères, et cette valeur ne semble pas varier selon l'espèce ou la concentration totale de congénères. Nous avons également mesuré les concentrations de quatre congénères chlorés non substitués en position *ortho*, y compris le 3,3', 4,4', -tétrachlorobiphényle. Ces quatre congénères toxiques représentent environ 1,2 % de la concentration totale. Les homologues de PCB dosés dans le poisson

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entier et le muscle renferment en moyenne 56 % de chlore en poids. Une analyse comparée de la composition des congénères observés dans le poisson et de celle signalée pour les mélanges de PCB indique que les congénères les plus persistants se comportent comme un mélange homogène plutôt que comme des congénères individuels.

Nous avons également mesuré la concentration de 12 autres composés organochlorés dans le but d'étudier l'influence des lipides sur la répartition des composés chimiques entre le poisson entier et le muscle. La corrélation est bonne entre la répartition relative des lipides et celle des composés chimiques dans le muscle et le poisson entier dans le cas de toutes les espèces sauf la truite brune. Nous avons également mesuré la répartition des composés chimiques dans le poisson entier et le muscle pour déterminer si la mesure de l'une de ces concentrations peut être utilisée pour calculer l'autre. Aucun rapport conséquent n'a été démontré; il n'est donc pas possible d'appliquer aveuglément cette extrapolation à toutes les espèces.

Abstract

Ninety-two monochloro- to decachlorobiphenyl congeners were monitored in brown trout (Salmo trutta), lake trout (Salvelinus namaycush), small and large rainbow trout (Salmo gairdneri), and small and large coho salmon (Oncorhynchus kisutch) from Lake Ontario. Concentrations of individual congeners ranged from nondetectable levels, primarily among the monochloro- to trichlorobiphenyl congeners, to 1 $mg.kg^{-1}$ for 2,2′,4,4′,5,5′-hexachlorobiphenyl. Congeners with the higher concentrations were generally the pentachloro- and hexachlorobiphenyl homologues. Total congener concentration ranged from 1-10 $mg.kg^{-1}$ in fish, and 0.3-4 $mg.kg^{-1}$ in muscle. Consistent values of congener composition were shown when individual congener levels were expressed as percent of total concentration. The 10 most common congeners represented about 52% of total congener content, this value does not appear to be influenced by species or by total congener concentration. Concentrations of 4 non-ortho chloro substituted congeners , including 3,3′,4,4′-tetrachlorobiphenyl, were also monitored. These four toxic congeners represented about 1.2% of the total concentration. The PCB homologues monitored in fish and muscle contained an average content of 56% chlorine by weight. Comparative analyses on the composition of congeners observed in fish with that reported for PCB mixtures suggest the more persistent congeners behave as a homogeneous mixture rather than as individual congeners.

Levels of 12 other organochlorine compounds were also monitored to examine the influence of lipid on chemical distribution between fish and muscle. A good correlation was shown between the relative distributions of lipid and chemicals between muscle and fish for all species except brown trout. The distribution of chemicals between fish and muscle was also examined to

determine if the measurement of one can be used to estimate the concentration of the other. No consistent relationship was demonstrated to would allow this extrapolation to be applied to all species with confidence.

Résumé

On a mesuré 92 congénères de polychlorobiphényles (monochloro- à décachloro-) chez la truite brune (Salmo trutta), le touladi (Salvelinus namaycush), chez la petite et la grande truite arc-en-ciel (Salmo gairdneri) et chez le petit et le gros saumon coho (Oncorhynchus kisutch) dans le lac Ontario. La concentration des congénères individuels varie : de non détectable (surtout chez les congénères monochloro- à trichlorobiphényles) à 1 mg/kg dans le cas du 2,2', 4,4', 5,5' -hexachlorobiphényle. Les congénères les plus concentrés sont en général les homologues pentachloro- et hexachlorobiphényles. La concentration totale des congénères varie de 1 à 10 mg/kg dans le poisson entier et de 0,3 à 4 mg/kg dans le muscle. La composition des congénères est constante lorsqu'on exprime les congénères individuels en pourcentage de la concentration totale. Les 10 congénères les plus courants représentent environ 52 % de la concentration totale de congénères, et cette valeur ne semble pas varier selon l'espèce ou la concentration totale de congénères. Nous avons également mesuré les concentrations de quatre congénères chlorés non substitués en position ortho, y compris le 3,3', 4,4', -tétrachlorobiphényle. Ces quatre congénères toxiques représentent environ 1,2 % de la concentration totale. Les homologues de PCB dosés dans le poisson entier et le muscle renferment en moyenne 56 % de chlore en poids.

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Polychlorinated biphenyls (PCBs) have been used in different industrial applications since the 1930's, but were not recognized as environmental contaminants until 1966 (Jensen 1972). Nearly all quantitative analyses for PCBs are still based on chemical standards composed of mixtures and reported as total PCB concentrations, although some recent studies have reported congener specific analyses (e.g. Tuinstra et al. 1983). Analyses of PCB mixtures indicate measurable levels of 50-118 of the possible 209 congeners are present in some Aroclors (Albro et al. 1981, Mullin 1985). Studies that examined the composition of PCBs in environmental samples have reported the presence of up to 78 congeners in fish but no consistent relationships have been established among congeners or with total PCB content (Tanabe et al. 1980, Giesy et al. 1986). Laboratory studies have shown differences in elimination rates among congeners in fish (Niimi and Oliver 1983), and comparative studies on mammalian systems indicate some congeners are more toxic than others (Safe 1984). Additional congener specific studies are required before such information can be effectively applied to issues such as PCB kinetics in aquatic ecosystems, and the human health implications of PCBs in consumable fish products.

This study monitored the presence of 92 monochloro- to decachlorobiphenyl congeners, and 12 other organochlorine chemicals, in fish and muscle of four species of Lake Ontario salmonids. The objectives were to examine the quantitative composition of the more common congeners present in fish, compare their composition in fish with that reported in PCB mixtures, corroborate the influence of lipid on the distribution of chemicals in fish, and examine the relationship between congener distribution in fish and muscle.

Materials and Methods

Ten brown trout (Salmo trutta) averaging 1430±360 g (mean±SD), 8 "small" rainbow trout (Salmo gairdneri) of 1140±120 g, and 10 "small" coho salmon (Oncorhynchus kisutch) of 1190±190 g were collected off Vineland, Ontario. Ten lake trout (Salvelinus namaycush) weighing 2410±770 g were taken off Port Credit, and 12 "large" rainbow trout of 3380±870 g, and 9 "large" coho salmon of 3330±520 g were taken from the Credit River. Eggs from gravid females in the last three groups were removed, and all fish were frozen at -20°C until analyses. Fish were prepared for analyses according to procedures described previously (Niimi and Oliver 1983). Tissue samples were taken that would be representative of chemical levels in muscle and whole fish.

All samples were analyzed for 92 monochloro- to decachlorobiphenyl congeners (Table 1) that are identified according to the International Union of Pure and Applied Chemistry (IUPAC) system (Ballschmiter and Zell 1980). Samples were also analyzed for p,p'-DDT, p,p'-DDD, p,p'-DDE, mirex, photo-mirex, 1,2,3,4-tetrachlorobenzene (TECB), pentachlorobenzene (QCB), hexachlorobenzene (HCB), α-hexachlorocyclohexane (α-BHC), γ-hexachlorocyclohexane (lindane), γ-chlordane, and octachlorostyrene (OCS). In addition, the 1,2-, 1,3-, and 1,4-dichloro-, 1,2,3-, 1,2,4-, and 1,3,5-trichloro-, the 1,2,3,5- and 1,2,4,5-tetrachlorobenzenes; 2,3,6- and 2,4,5-trichloro- and 2,3,4,5,6-pentachlorotoluenes; and hexachlorobutadiene were analyzed, but not detected, in any of the samples. Lipid content was determined according to Bligh and Dyer (1959). Chemical and lipid concentrations are reported on a wet weight basis.

Sample clean-up procedures for chromatographic analyses has been described (Oliver and Nicol 1982). Chemical analyses were done by gas chromatography using dual fused silica capillary columns (30 m x 0.22 mm ID Durabond-5 and Durabond-17 with 0.25 μm film thickness) and electron capture detectors.

Detector temperature was 350°C. Carrier gas was helium with a linear velocity flow of 20 cm.sec^{-1}. Temperature program was 50 to 250°C at a rate of 1°C per minute. Some congeners coeluted even with this slow temperature program. These included the trichloro- (24+27) and (28+31), tetrachloro- (40+71), (70+76), and (56+60), pentachloro- (87+97), heptachloro- (170+190) and (182+187), and octachloro- congeners (196+203). Detection limits for most chemicals were in the 1-10 $\mu g.kg^{-1}$ range, except for the monochlorobiphenyls with detection limits of 50 $\mu g.kg^{-1}$.

The procedure described above was not suitable to monitor the levels of many non-ortho chloro substituted congeners because of their low concentrations and potential coelution with the more common congeners of higher chlorine content (Mullin et al. 1984). To resolve these factors, equal volumes of sample extracts from each fish within each group were combined and reduced in volume to increase instrument sensitivity. The composite sample for each group was analyzed using a Hewlett-Packard mass selective detector (MSD) set in the selected ion monitoring mode. Detection limit was about 2 $\mu g.kg^{-1}$.

Congener identification and quantitation were based on a mixture of Aroclors 1221, 1016, 1254, and 1262 in a ratio of 10:5:3.5:3. These Aroclors were obtained from the U.S. EPA Repository for Toxic and Hazardous Materials (Research Triangle Park, NC). Composition of this mixture was determined from congener specific analyses (Mullin et al. 1984, Mullin 1985). Confirmation of peak identification and quantitative analyses were made from 50 standards that were obtained from commercial sources, and 51 congeners from the National Research Council of Canada (Atlantic Research Laboratory, Halifax, NS). Some samples were also analyzed by gas chromatography/mass spectrometry for further confirmation. OCS was obtained as a gift from Dow Chemical Chemical, Walnut

Creek, CA. Photomirex was synthesized according to Chau and Thomson (1979). Standards for the other organochlorine chemicals were obtained from commercial sources.

Chemical concentrations in $\mu g.kg^{-1}$ were reported for all individual congeners observed in fish, and their sum as total congener concentration. Percent chlorine content of total congener concentration was estimated by multiplying the total concentration of each homologue group by the percent molecular weight represented by chlorine for each homologue. Results shown for most of the parameters measured represent the mean±standard deviation for each group of 8-12 values depending on species. Paired t-tests were used to compare differences between fish and corresponding muscle values within each group. Analysis of variance (ANOVA) was used to test for statistically significant differences among groups. Differences among linear regression relationships were determined by analysis of covariance (ANCOVA).

Results

PCB analyses on four salmonid species indicated an average of 54-68 congeners were present in fish and 33-62 in muscle, including the non-_ortho_ chloro substituted congeners which were analyzed as a composite sample, among the 92 congeners monitored (Table 2). Lake trout contained the highest number of congeners in fish and muscle among the species. There was a large range in mean congener concentrations, and deviation about the means, in fish and muscle among species (Table 2). Most congeners observed were in the 10-200 $\mu g.kg^{-1}$ range, and differences in mean concentrations exceeded six-fold among species. Many trichloro-, and some tetrachloro-, and heptachloro- to decachlorobiphenyl congeners monitored were not detected in some species. Monochloro- congeners 1 and 3; dichloro- 4, 5, 6, 7, 8, 10, 12, and 13;

trichloro- 19 and 25; tetrachloro- 45; and hexachloro- congener 169 were not detected in any species. Total concentration of all congeners monitored ranged from 1450 $\mu g.kg^{-1}$ in small rainbow trout to 9970 $\mu g.kg^{-1}$ in lake trout, and 288 to 3880 $\mu g.kg^{-1}$ in muscle for the respective species.

Congener levels were reported according to decreasing concentrations in Table 2. This procedure does demonstrate a trend of decreasing values that is consistent for fish and muscle, within homologue groups, as well as among species. Individual concentrations varied widely within species although a consistent relationship among species can be demonstrated when individual levels are expressed as a percent of total congener concentration. Values shown for the more commonly occurring congeners indicate their mean percent contribution are similar for fish and muscle among different species even though total concentrations vary widely (Table 3). Deviation about the mean is also greatly reduced. This same response is also demonstrated when total percent contribution by each homologue group is calculated. The pentachloro- and hexachloro- homologues represented about 37 and 29% of the total congener concentrations. Trichloro-, octachloro-, nonachloro-, and decachloro- homologues were not major contributors, their cumulative values represented less than 5% of total concentration in fish and muscle among all species.

Contributions by individual congeners indicated 2,2′,4,4′,5,5′-hexachlorobiphenyl (153) was the most common averaging 9.0% of total congener concentration in fish, and 10.1% in muscle, followed by 2,2′,4,5,5′-pentachlorobiphenyl (101) which accounted for 6.4% in fish and muscle (Table 3). Consistency among congener content expressed as percent of total concentration is further demonstrated by the cumulative percentages of the more common congeners. The ten most common were pentachloro- congeners 84, (87+97), 101, 110, and 118, hexachloro- congeners 138, 149, and 153, and the heptachloro- congener 180. These ten accounted for 49.3±3.2, 52.2±0.9, 53.0±1.7, 52.8±1.8, 51.7±1.0, and 54.4±0.6% of the in brown, lake, small and large rainbow trout, and small and large coho salmon respectively. Corresponding values in muscle were 52.3±2.4, 52.8±0.7, 60.3±5.5, 53.7±1.2, 53.7±1.2, and 55.1±0.6% for the respective species. The ten next most common congeners were the tetrachloro- 66 and (70+76), pentachloro- 95 and 105, hexachloro- 146, and heptachloro- congeners (182+187) and (170+190) whose individual contributions ranged from 1.8-3.9% in fish. Cumulative contributions for the 20 most common congeners were 68.5±3.4, 70.1±1.3, 72.4±0.9, 71.0±0.9, 70.9±1.5, and 73.0±1.0% in fish, and 71.5±2.4, 70.8±1.3, 81.0±5.8, 72.0±1.1, 73.0±1.3, and 73.5±0.7% in muscle for the respective species. Many of the other congeners observed contributed less than one percent of total concentration.

The relationship between homologue frequency and total congener concentration was examined using percent chlorine content. Chlorine composition were 55.74±0.51, 56.28±0.26, 55.97±0.77, 56.48±0.37, 55.74±0.14, and 56.42±0.21% for brown, lake, small and large rainbow trout, and small and large coho salmon respectively. Values in muscle were 55.87±0.42, 56.33±0.28, 56.75±0.82, 56.65±0.34, 55.95±0.17, and 56.51±0.19% for the respective species. ANOVA indicated there were significant differences ($P<0.01$) in percent chlorine content in fish within and among species. Further analyses suggested these differences may be attributed to fish size. No significant differences could be shown among the chlorine percentages when comparisons were made among lake, large rainbow trout, and large coho salmon, and for chlorine percentages of brown and small rainbow trout, and small coho salmon. Paired t-tests between fish and muscle for each sample within each group showed significantly higher chlorine percentages ($P\leq0.05$) in muscle than fish in all species, except lake trout.

Analyses for 12 other chemicals observed in fish indicated DDE was present at the highest concentrations among those monitored with lake trout again containing the highest concentrations (Table 4). A listing of chemicals by decreasing concentrations also demonstrated a consistent trend among species similar to that noted for PCB congeners.

Lipid levels varied widely among species. Values in fish were 14.3±1.6, 17.4±3.0, 6.0±2.4, 13.0±1.7, 8.4±1.8, and 4.5±0.8% for brown, lake, small and large rainbow trout, and small and large coho salmon respectively. Corresponding values in muscle were 4.8±1.1, 7.3±2.2, 1.3±0.5, 4.9±1.1, 3.4±0.7, 2.3±0.4% for the respective species. Regression analyses among samples indicated percent lipid in fish increased significantly ($P<0.05$) with weight in lake trout and small coho salmon, but not the other groups. Lipid levels in muscle increased significantly ($P<0.01$) with weight only in lake trout.

Discussion

General Behavior of PCB Congeners and Comparisons with Other Studies

These results show that consistent PCB composition relationships are found among salmonids when values are expressed as a percent of total congener concentration. These relationships appear to be independent of total concentration, and applicable to fish and muscle. The only inconsistent results were the cumulative percentages of the 10 and 20 most common congeners observed in muscle of small rainbow trout which were higher than those of other species. This response could be due to the presence of only 33 congeners in the muscle of this group compared to 52-62 in muscle of the other species, and 54-68 in fish of all species (Table 2). Many of the less abundant congeners are probably present at concentrations below detection limit in this group which had the lowest total congener concentration.

There is good agreement among the few studies that have reported congener specific measurements in fish. This study indicated congeners 101, 110, 138, 153, and 180 were among the 10 most common observed in Lake Ontario salmonids (Table 3). The most common congeners that have been reported in fish from other waters are 138, 149, 153, and 180 among the 66 congeners monitored in cod (*Gadus* sp.) reported by Zell et al. (1978), and 138, 153, and 180 among 17 congeners reported in eel (*Anguilla* sp.) by Tuinstra et al. (1983). Boon and Duinker (1986) observed 101, 110, 138, and 153 among 36 congeners in plaice, and Weigelt (1986) reported 101, 138, 153, and 180 among 31 congeners in cod. Other studies have reported values from which percent contributions for each congener can be determined. Tanabe et al. (1980) reported congeners 153 and 138 were the most common, representing 11.9%, and 10.6% of total concentration among the 59 congeners reported in pomfret (*Brama* sp.). In contrast, Giesy et al. (1986) reported levels of 78 congeners in chinook salmon (*Oncorhynchus tshawytscha*) eggs where congener 91 represented 9.4%, and 153 about 4.4% of total concentration.

The bioaccumulation of PCBs in aquatic ecosystems has been reported (Thomann 1981). Studies using mass fragmentation techniques have indicated percentages of higher chlorinated biphenyls increases with trophic levels in the marine ecosystem (Tanabe et al. 1984). Congener specific analyses could allow a more detailed examination of these processes, and the conclusions would be more relevant if the measurements are representative of most of the principal PCB congeners present in aquatic ecosystems. These criteria have been met in this study as shown by comparisons between the congeners observed and those reported in PCB mixtures. The 92 congeners monitored in this study accounted for about 82, 80, 97, and 93% of total congener composition of Aroclor 1254 (Clophen A50) reported by Zell et al. (1977), Albro et al. (1981), Capel et al. (1985), and Mullin (1985) respectively. Similar comparisons with Aroclors 1242 and 1248 indicated slightly higher percentages, while those with Aroclor 1260 (Clophen A60) were slightly lower. Thus the congeners monitored in this study provide a good account of the PCB composition present in Lake Ontario salmonids since all the major PCB congeners in the Aroclors were monitored.

Percent chlorine content of the PCBs averaged 56.1% among the salmonids examined, and exhibited a narrow range among species. This may be due to the similar trophic status of these species in Lake Ontario. Recalculation of data reported by Tanabe et al. (1980) indicate a PCB chlorine content of 55.7% in pomfret captured off Chile, and 54.7% in chinook salmon eggs from Lake Michigan (Giesy et al. 1986). The significance of these values cannot be examined from an environmental perspective at this time because no comparative information is available on PCB dynamics in these ecosystems.

An analysis of PCB congener composition monitored in fish with that reported in PCB mixtures, and probable composition of PCBs from contaminated ecosystems, does suggest a hypothesis on PCB kinetics among species at this trophic level. Congener composition of Lake Ontario salmonids was compared with Aroclor 1254 which has been used as a reference standard for PCB analysis (e.g. Bache et al. 1972). Comparisons among the percentage contribution of the more common congeners observed in fish and those reported in Aroclor 1254 mixtures generally indicate higher frequencies of congeners 153 and 180, and lower frequency of 110 in fish (Table 5). This analysis could suggest some congeners are selectively retained and others eliminated, and the kinetics of PCBs in fish would be determined by the net response of individual congeners. It was previously reported that most penta- and higher chlorinated congeners have biological half-lives in excess of several hundred days in trout, therefore this apparent response would not be due to differences in their clearance rates (Niimi and Oliver 1983).

The PCB congeners in Lake Ontario salmonids averaged 56% chlorine. A PCB mixture containing 56% chlorine by weight can be obtained by mixing Aroclors 1254 and 1260 in a 2:1 ratio (Table 5). A comparison of percent frequency of the more common congeners monitored in fish with the frequency of a 56% chlorine Aroclor mixture indicates similarities among values (Table 5). The average value of 9.0% for congener 153 in fish is higher than values for the Aroclor mixtures, although there is good agreement with the 7.7% value for Mullin (1985). The 5.6% value for congener 84 is also higher in fish although there is reasonably good agreement with the Aroclor mixture for Mullin. Percent frequency analyses using information adjusted for chlorine content would suggest the more persistent congeners in Lake Ontario salmonids respond more as a homogeneous mixture rather than as individuals.

Environmental and Health Implications of Congener Specific Analyses

Application of the results of this study should be useful in the areas of environmental monitoring of PCBs and evaluation of PCB levels in consumable fish products. Congener specific analyses for monitoring PCBs has been proposed (Tuinstra et al. 1983, Boon and Duinker 1986). The six proposed by Tuinstra, which includes 28, 52, 101, 138, 153, and 180, were selected on the basis of their frequencies in Aroclors 1242, 1254, and 1260. Comparative analyses on the content of these six congeners in fish indicate they account for 38% of total PCB in eel reported by Tuinstra et al. (1983), an average of 28% in fish from this study, about 34% in pomfret muscle (Tanabe et al. 1980), and 55% in salmon eggs (Giesy et al. 1986). These six congeners represent about 25% of the composition of Aroclor 1254 (Capel et al. 1985, Mullin 1985). Boon and Duinker (1986) proposed 14 congeners for environmental monitoring based on their occurrence in environmental samples and chromatographic properties. These include the tetrachloro- to heptachloro- congeners 44, 52, 84, 99, 101, 118, 128, 138, 149, 153, 170, 180, 183, and 187. Their cumulative contribution is about 51% for the six data groups in this study with 7 of the 10 most common observed accounting for 42%, about 48% in pomfret without congeners 149 and 170, and 30% in salmon eggs. The 14 congeners account for 44% of the composition of Aroclor 1254 (Mullin 1985), and 45% without congener 84 (Capel et al. 1985).

Based on current information, it is difficult to propose specific congeners that could be routinely monitored in fish for assessment of environmental quality and for comparative analyses among fish from different water bodies. Most congener specific studies generally agree 101, 138, 153, and 180 are present at the highest concentrations in fish, but other congeners would likely need to be monitored to provide a more comprehensive assessment. The comparative information on congener composition cited above tend to represent species at the higher trophic levels. Further studies would be required to examine the relationship between the percent contribution of specific congeners to total PCBs levels in fish from different trophic levels, and fish from different waters.

The use of congener specific analyses to examine potential health hazards of consuming fish products containing PCB residues presents an even more difficult task. Safe et al. (1983) have identified 16 congeners which are aryl hydrocarbon hydroxylase (AAH) inducers. These include tetrachloro- 77 and 81, pentachloro- 105, 114 (2,3,4,4',5), 118, 123 (2',3,4,4',5), and 126, hexachloro- 128, 138, 156, 157 (2,3,3',4,4',5'), 158 (2,3,3',4,4',6), 166 (2,3,4,4',5,6), and 169, and heptachloro- congeners 170 and 189 (2,3,3',4,4',5,5'). Ten of these congeners were monitored in this study of which only 105, 118, and 138 contributed more than 2% each to total concentration. Most of the other congeners proposed would likely contribute less than 1%. Further comparisons with the other studies cited indicate 8 of these congeners were reported in pomfret, and 7 in salmon eggs. Only 4 of these congeners were reported in Aroclor 1254 by Capel et al. (1985), while Mullin (1985) reported 12 congeners which accounted for about 18% of its composition of which 105, 118, 128, and 138 represented 13.5%.

The non-*ortho* chloro substituted congeners, with substitutions at both *para* and at two or more *meta* positions, are the most toxic although their environmental concentrations may be low (Safe et al. 1985). These are represented by congeners 77, 81, 126, and 169, and are nearly isosteric with 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). Capel et al. (1985) did not find these in Aroclor 1254. Mullin (1985) reported values of 0.12 and 0.37% for congeners 77 and 81, and Albro et al. (1981) values of 0.12 and 0.16% of total composition for 77 and 126, respectively, in this Aroclor. Congeners 77, 81, and 126 were detected in Lake Ontario salmonids at concentrations in the low $\mu g.kg^{-1}$ range (Table 2). These levels represent about 0.2% for congener 77 and 0.8% for 81 of total congener concentration (Table 3). The value for congener 77 in salmonids is comparable to the estimate of 0.3% for pomfret (Tanabe et al. 1980) and 0.5% in salmon eggs (Giesy et al. 1986). No comparative values were reported for congener 81.

Regulatory guidelines on PCB levels in consumable fish products are currently based on total concentration. The use of congener specific analyses for regulatory purposes would be more appropriate from a human health perspective, but developmental studies are still required before its application would be feasible. The non-ortho chloro substituted congeners occur at concentrations that approach or are below detection limits of most current routine analytical procedures. Chemical analysis for congener 77 using electron capture gas chromatography is difficult because of coelution with other congeners including the prevalent 110 on most capillary columns (Mullin et al. 1984, Schwartz et al. 1987). Coelution problems could be minimized by using a secondary cleanup procedure with a carbon/glass fiber packed column (Smith 1981), and quantitation made more reliable using a mass selective detector.

Studies on sublethal effects of PCBs on fishes inhabiting contaminated ecosystems could be more definitive if specific congeners rather than total PCBs were monitored. Emphasis should focus on the relation of the concentration of a congener and its toxicity. The concentration of congener 153 is about 45-fold higher than 77 in Lake Ontario salmonids. Data from other studies indicate congener 153 is 38-fold higher than 77 in pomfret (Tanabe et al, 1980), and 18-fold higher in salmon eggs (Giesy et al. 1986). Comparative studies on mammalian systems suggest congener 77 could be over 10 to 41-fold more toxic than 153 (McKinney et al. 1985, Safe et al. 1985). Thus, the concentrations of congener 77 in Lake Ontario salmonids could be as important as 153 from a toxicological perspective. This factor is more obvious among the chlorinated dioxins where 2,3,7,8-TCDD is many orders of magnitude more toxic than most other dioxins (e.g. Kociba and Cabey 1985). Therefore, chemical potency, in addition to chemical concentration, is an important consideration for chemical groups with many homologues when issues such as human health concerns and sublethal effects of chemicals on fishes are being examined.

Lipid-Chemical Distribution and Chemical Partitioning in Fish and Muscle

Lipid content has been suggested to play an important role in the accumulation of persistent chemicals by fish. Evidence to support this view has largely been from field studies. Positive correlations have been reported between pesticide levels and lipid content in salmon and trout (Anderson and Fenderson 1970, Reinert 1970). A reexamination of data reported for pike (Esox lucius) showed significant increases (P<0.01) in pesticides and PCB levels in tissue with increasing levels of extractable lipid (Olsson and Jensen 1975). In contrast, some studies reported no significant correlations between PCB content and lipid levels in fish although annual concentrations of both varied widely (Olssen et al. 1978). These lipid-chemical relationships were further examined in this study where comparisons were made among species as well as within species. The latter was of particular interest because large rainbow trout contained 13% lipid and small fish contained 6%, while large coho salmon contained 4% lipid and small fish 8%. General comparisons were made among data groups rather than individuals because fish of similar sizes were used.

Olsson in refs.

Concentrations of 12 other organochlorine chemicals in addition to PCBs were monitored to examine the influence of lipid on chemical distribution in fish. Analysis was based on the correlation of relative chemical and lipid distributions in fish and muscle. Lipid distribution was estimated as the ratio of lipid in muscle/fish. The ratios for lipids were 0.332±0.055 for brown trout, 0.412 ± 0.081 for lake trout, 0.220 ± 0.024 and 0.378 ± 0.064 for small and large rainbow trout, and 0.407 ± 0.034 and 0.524 ± 0.077 for small and large coho salmon. Corresponding muscle/fish ratios for chemicals were estimated for each group. Ratios for the 12 chemicals and total PCBs varied among chemicals as well as among species. Mean ratios ranged from 0.376-0.522 in brown trout, 0.322-0.440 in lake trout, 0.206-0.332 small rainbow trout, 0.335-0.463 in large rainbow trout, 0.381-0.510 in small coho salmon, and 0.397-0.569 in large coho salmon.

ANOVA indicated the lipid ratio in brown trout was significantly lower ($P \leq 0.05$) than ratios of all chemicals except OCS. In contrast, ANOVA could demonstrate no significant differences between the lipid ratio and chemical ratios for each chemical within each of the other species. A mean of means of the chemical ratios was calculated for each group, values ranged from 0.442 for brown trout excluding OCS, 0.393 for lake trout, 0.243 and 0.380 for small and large rainbow trout, and 0.426 and 0.522 for small and large coho salmon. Regression analysis of the chemical and lipid ratios for five groups, except brown trout, indicated the relationship ($F = 153.58$, $P<0.01$):

$$Y = 0.039 + 0.911(\pm 0.230)X$$

where X is the lipid ratio, and Y the chemical ratio. The regression coefficient (b) does not differ significantly from 1.0 as indicated by its 95% confidence limits.

The chemical or lipid distribution pattern is not the same for brown trout as for the other salmonids (Fig. 1). It cannot be established at this time whether this deviation is due to the chemical or the lipid distribution. It also cannot be determined if this response is species specific, or seasonal.

The data for small and large sizes of rainbow trout and coho salmon would indicate corresponding changes occur in lipid and chemical levels where the ratios follow along the regression line. Chinook salmon is another species that is present in Lake Ontario. Analysis of a 7.8 kg fish as part of this study indicated a total congener content of 5400 $\mu g.kg^{-1}$, and a distribution pattern that was similar to the other species. Its lipid ratio was 0.474 and chemical ratio was 0.515 which would suggest a distribution pattern that is consistent with the other species. These results would strongly suggest lipid is a major factor that influence the distribution of persistent chemicals in fish, and the ratios of 5 of the 6 data groups examined indicates a highly consistent relationship.

Chemical distribution in fish is of interest to regulatory agencies because environmental monitoring programs tend to use whole fish while those concerned with human health use the edible or muscle portion. Very little information is available which allows residue levels between fish and muscle to be compared because only one measurement is generally reported. The results of this study would suggest a good qualitative extrapolation of chemical composition between fish and muscle can be made because chemical distribution among the more persistent compounds apears to be a nonselective process. These results were also examined to determine if quantitative extrapolations among the six groups examined would be feasible.

Regression analyses on total congener concentrations indicated levels in muscle increased significantly ($P \leq 0.05$) with increasing concentrations in fish for brown, lake, large rainbow trout, and small coho salmon, but not for small rainbow trout or large coho salmon. ~~ANCOVA~~ among the first four groups indicated there were no significant differences among their regression equations, the common regression equation ($F = 109.86$, $P<0.01$) being: ANOVA 2

$$Y = 21.6 + 0.389(\pm 0.059)X$$

where X is congener concentration ($\mu g.kg^{-1}$) in muscle, and Y the concentration in fish. This relationship suggests that chemical levels in fish can influence those in muscle, but the response is not always statistically significant.

This study showed that lipid is an important factor in the distribution of chemicals among salmonids. The influence of lipid on total congener distribution between fish and muscle was examined by expressing congener concentrations on a per g lipid basis. Regression analysis of lipid-adjusted congener concentrations in fish and muscle indicated a similar response to that shown for nonadjusted values. Significant increases ($P \leq 0.05$) in muscle levels with increasing levels in fish were observed for brown, lake, large rainbow trout, and small coho salmon, but not small rainbow trout or large coho salmon. ANCOVA indicated the relationship for brown trout was significantly different ($P \leq 0.05$) from large rainbow trout and small coho salmon, but not from lake trout. There was no significant differences among lake, large rainbow trout, and small coho salmon, whose common regression equation ($F = 102.72$, $P < 0.01$) is:

$$Y = 2.45 + 0.913(\pm 0.190)X$$

where X is ~~mgs~~ burden of total congeners ~~per g~~ lipid in muscle in mg/g, and Y the same in fish. These analyses would suggest that quantitative extrapolations of chemical levels in fish and muscle based on actual or lipid-adjusted values cannot be described by a simple relationship that would be applicable to all species.

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References

Albro, P.W., J.T. Corbett, and J.L. Schroder. 1981. Quantitative characterization of polychlorinated biphenyl mixtures (Aroclors 1248, 1254 and 1260) by gas chromatography using capillary columns. J. Chromatogr. 205: 103-111.

Anderson, R.B., and O.C. Fenderson. 1970. An analysis of variation of insecticide residues in landlocked Atlantic salmon (Salmo salar). J. Fish. Res. Board Can. 27: 1-11.

Bache, C.A., J.W. Serum, W.D. Youngs, and D.J. Lisk. 1972. Polychlorinated biphenyl residues: accumulation in Cayuga lake trout with age. Science 177: 1191-1192.

Ballschmiter, K., and M. Zell. 1980. Analysis of polychlorinated biphenyls by capillary gas chromatography. Fres. Z. Anal. Chem. 302: 20-31.

Bligh, H.G., and W.J. Dyer. 1959. A rapid method of total lipid extraction and purification. Can. J. Biochem. Physiol. 37: 911-917.

Boon, J.P., and J.C. Duinker. 1986. Monitoring of cyclic organochlorines in the marine environment. Environ. Monitor. Assess. 7: 189-208.

Capel, P.W., R.A. Rapaport, S.J. Eisenreich, and B.B. Looney. 1985. PCBQ: computerized quantification of total PCB and congeners in environmental samples. Chemosphere 14: 439-450.

Chau, A.S.Y., and R.T. Thomson. 1979. Analytical reference materials. I. Preparation and purification of photomirex. J. Assoc. Off. Anal. Chem. 62: 1302-1308.

Giesy, J.P., J. Newsted, and D.L. Garling. 1986. Relationships between chlorinated hydrocarbon concentrations and rearing mortality of chinook salmon (Oncorhynchus tshawytscha) eggs from Lake Michigan. J. Great Lakes Res. 12: 82-98.

Jensen, S. 1972. The PCB story. Ambio 1: 123-131.

Kociba, R.J., and O. Cabey. 1985. Comparative toxicity and biologic activity of chlorinated dibenzo-p-dioxins and furans relative to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD). Chemosphere 14: 649-660.

McKinney, J.D., K. Chae, E.E. McConnell, and L.S. Birnbaum. 1985. Structure-induction versus structure-toxicity relationships for polychlorinated biphenyls and related aromatic hydrocarbons. Environ. Health Perspect. 60: 57-68.

Mullin, M.D. 1985. Congener Specific PCB Workshop. U.S. EPA, Grosse Ile, MI.

Mullin, M.D., C.M. Pochini, S. McCrindle, M. Romkes, S.H. Safe, and L.M. Safe. 1984. High-resolution PCB analysis: synthesis and chromatographic properties of all 209 PCB congeners. Environ. Sci. Technol. 18: 468-476.

Niimi, A.J., and B.G. Oliver. 1983. Biological half-lives of polychlorinated biphenyl (PCB) congeners in whole fish and muscle of rainbow trout (<u>Salmo gairdneri</u>). Can. J. Fish. Aquat. Sci. 40: 1388-1394.

Oliver, B.G., and K.D. Nicol. 1982. Gas chromatographic determination of chlorobenzenes and other chlorinated hydrocarbons in environmental samples using fused silica capillary columns. Chromatographia 16: 336-340.

Olsson, M., and S. Jensen. 1975. Pike as the test organism for mercury, DDT and PCB pollution. A study of the contamination in the Stockholm archipelago. Inter. Freshw. Res. Drott. 54: 83-106.

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Olsson, M., S. Jensen, and L. Reutergard. 1978. Seasonal variation of PCB levels in fish - an important factor in planning aquatic monitoring programs. Ambio 7: 66-69.

Reinert, R.E. 1970. Pesticide concentrations in Great Lakes fish. Pest. Monitor. J. 3: 233-240.

Safe, S. 1984. Polychlorinated biphenyls (PCBs) and polybrominated biphenyls

(PBBs): biochemistry, toxicology, and mechanism of action. CRC Crit. Rev. Toxicol. 13: 319-393.

Safe, S., S. Bandiera, T. Sawyer, L. Robertson, L. Safe, A. Parkinson, P.E. Thomas, D.E. Ryan, L.M. Reik, W. Levin, M.A. Denomme, and T. Fujita. 1985. PCBs: structure-function relationships and mechanism of action. Environ. Health Perspect. 60: 47-56.

Safe, S., M. Mullin, L. Safe, C. Pochini, S. McCrindle, and M. Romkes. 1983. High resolution PCB analysis, p. 1-13. In D. Mackay, S. Paterson, S.J. Eisenreich, and M.S. Simmons [ed.] Physical behavior of PCBs in the Great Lakes, Ann Arbor Sci. MI

Schwartz, T.R., D.L. Stalling, and C.L. Rice. 1987. Are polychlorinated biphenyl residues adequately described by Aroclor mixture equivalents? Isomer-specific principal components analysis of such residues in fish and turtles. Environ. Sci. Technol. 21: 72-76.

Smith, L.M. 1981. Carbon dispersed in glass fibers as an absorbent for contaminant enrichment and fractionation. Anal. Chem. 53: 2152-2154.

Tanabe, S., H. Tanaka, and R. Tatsukawa. 1984. Polychlorobiphenyls, Σ DDT, and hexachlorocyclohexane isomers in the Western North Pacific ecosystem. Arch. Environ. Contam. Toxicol. 13: 731-738.

Tanabe, S., H. Tanaka, R. Tatsukawa, and I. Nakamura. 1980. Organochlorine compound residues in several species of fishes caught off Chile. Bull. Jap. Soc. Sci. Fish. 46: 763-769.

Thomann, R.V. 1981. Equilibrium model of fate of microcontaminants in diverse aquatic food chains. Can. J. Fish. Aquat. Sci. 38: 280-296.

Tuinstra, L.G.M., J.J.M. Driessen, H.J. Keukens, T.J. van Munsteren, A.H. Roos, and W.A. Traag. 1983. Quantitative determination of specific chlorobiphenyls in fish with capillary gas chromatography and its use for monitoring and tolerance purposes. Inter. J. Anal. Chem. 14: 147-157.

Weigelt, V. 1986. Kapillargaschromatographische PCB-musteranalyse mariner spezies - betrachtungen zwischen anreicherung und chlorsubstitution ausgewahlter PCB-komponenten in marinen organismen aus der deutschen bucht. Chemosphere 15: 289-299.

Zell, M., H.J. Neu, and K. Ballschmiter. 1977. Identifizierung der PCB-komponenten durch retentionsindexvergleich nach kapillar-gaschromatographie. I. Zusammensetzung technischer PCB-Gemische. Chemosphere 2/3: 69-76.

Zell, M., H.J. Neu, and K. Ballschmiter. 1978. Single component analysis of polychlorinated biphenyl (PCB)- and chlorinated pesticides residues in marine fish samples. Fres. Z. Anal. Chem. 292: 97-107.

Captions for Tables 1-5 and Figure 1

Table 1. IUPAC number and structure of 92 chlorinated biphenyl congeners monitored in Lake Ontario salmonids.

Table 2. Number of congeners observed in Lake Ontario salmonids, excluding the 4 non-ortho chloro substituted congeners, and their individual and total concentrations ($\mu g.kg^{-1}$) in fish and muscle. The values shown represents the mean±SD for each group. ND indicates nondetectable levels. Concentrations of the non-ortho chloro substituted congeners based on a composite sample.

Table 3. Percent of total PCB concentration for the more common congeners, and the four non-ortho chloro substituted congeners, observed in fish and muscle The total percent contribution of all congeners in each homologue group is also shown.

Table 4. Concentrations ($\mu g.kg^{-1}$) of other chemicals monitored in Lake Ontario salmonids.

Table 5. Percent composition of 10 congeners in Aroclors 1254 (Clophen A50) and 1260 (Clophen A60) reported by Zell et al. (1977), Albro et al. (1980), Capel et al. (1985), and Mullin (1985), and their mean composition observed in Lake Ontario fish. The "56%" equivalents were estimated from the compositions reported for Aroclors 1254 and 1260.

Fig. 1. Relationship between muscle/fish lipid ratio, and muscle/fish

chemical ratio for Lake Ontario salmonids. Regression line based on ratios of all groups except brown trout.

It would be interesting to see the error bars for the points as well!

Table 1

No.	Structure
Monochlorobiphenyls	
1	2
3	4
Dichlorobiphenyls	
4	2,2′
5	2,3
6	2,3′
7	2,4
8	2,4′
10	2,6
12	3,4
13	3,4′
Trichlorobiphenyls	
16	2,2′,3
17	2,2′,4
18	2,2′,5
19	2,2′,6
22	2,3,4′
24	2,3,6
25	2,3′,4
26	2,3′,5
27	2,3′,6
28	2,4,4′
31	2,4′,5
32	2,4′,6
33	2′,3,4

No.	Structure
Tetrachlorobiphenyls	
40	2,2′,3,3′
41	2,2′,3,4
42	2,2′3,4′
44	2,2′,3,5′
45	2,2′,3,6
46	2,2′,3,6′
47	2,2′,4,4′
48	2,2′,4,5
49	2,2′,4,5′
52	2,2′,5,5′
53	2,2′,5,6′
56	2,3,3′,4′
60	2,3,4,4′
64	2,3,4′,6
66	2,3′,4,4′
70	2,3′,4′,5
71	2,3′,4′,6
74	2,4,4′,5
76	2′,3,4,5
77	3,3′,4,4′
81	3,4,4′5

No.	Structure
Pentachlorobiphenyls	
82	2,2′,3,3′,4
84	2,2′,3,3′,6
85	2,2′,3,4,4′
87	2,2′,3,4,5′
91	2,2′,3,4′,6
92	2,2′,3,5,5′
95	2,2′,3,5′,6
97	2,2′,3′,4,5
99	2,2′,4,4′,5
101	2,2′,4,5,5′
105	2,3,3′,4,4′
110	2,3,3′,4′,6
118	2,3′,4,4′,5
126	3,3′,4,4′,5
Hexachlorobiphenyls	
128	2,2′,3,3′,4,4′
129	2,2′,3,3′,4,5
132	2,2′,3,3′,4,6′
136	2,2′,3,3′,6,6′
138	2,2′,3,4,4′,5′
141	2,2′,3,4,5,5′
146	2,2′,3,4′,5,5′
149	2,2′,3,4′,5′,6
151	2,2′,3,5,5′,6
153	2,2′,4,4′,5,5′
156	2,3,3′,4,4′,5
169	3,3′4,4′,5,5′

No.	Structure
Heptachlorobiphenyls	
170	2,2′,3,3′,4,4′,5
171	2,2′,3,3′,4,4′,6
173	2,2′,3,3′,4,5,6
174	2,2′,3,3′,4,5,6′
177	2,2′,3,3′,4′,5,6
178	2,2′,3,3′,5,5′,6
180	2,2′,3,4,4′,5,5′
182	2,2′,3,4,4′,5,6′
183	2,2′,3,4,4′,5′,6
185	2,2′,3,4,5,5′,6
187	2,2′,3,4′,5,5′,6
190	2,3,3′,4,4′,5,6
Octachlorobiphenyls	
194	2,2′,3,3′,4,4′,5,5′
195	2,2′,3,3′,4,4′,5,6
196	2,2′,3,3′,4,4′,5,6′
198	2,2′,3,3′,4,5,5′,6
201	2,2′,3,3′,4,5,5′,6′
203	2,2′,3,4,4′,5,5′,6
205	2,3,3′,4,4′,5,5′,6
Nonachlorobiphenyls	
206	2,2′,3,3′,4,4′,5,5′,6
207	2,2′,3,3′,4,4′,5,6,6′
Decachlorobiphenyl	
209	2,2′,3,3′,4,4′,5,5′,6,6′

Table 2

Congener	Brown trout (n=10)		Lake trout (n=10)		Rainbow trout				Coho salmon			
					Small (n=8)		Large (n=12)		Small (n=10)		Large (n=9)	
	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle
No. congeners	57±4	51±3	65±2	59±4	52±5	33±9	60±2	55±2	54±2	50±2	57±2	54±1
Trichlorobiphenyls												
(28+31)	36±15	15±7	72±20	27±12	15±5	1±1	42±13	14±3	22±4	8±2	30±10	15±3
18	ND	ND	19±12	6±6	ND	ND	7±7	ND	ND	ND	ND	ND
22	3±2	ND	6±2	2±2	ND	ND	1±2	ND	ND	ND	ND	ND
26	1±1	ND	2±4	ND	ND	ND	1±1	ND	ND	ND	ND	ND
16	1±2	ND	1±1	ND	ND	ND	ND	ND	ND	ND	ND	ND
33	1±3	ND	1±1	ND	ND	ND	ND	ND	ND	ND	ND	ND
17	ND	ND	2±2	ND	ND	ND	ND	ND	ND	ND	ND	ND
25	ND	ND	1±1	ND	ND	ND	ND	ND	ND	ND	ND	ND
(24+27)	ND	ND	1±1	ND	ND	ND	ND	ND	ND	ND	ND	ND
32	1±1	ND	1±1	ND	ND	ND	ND	ND	ND	ND	ND	ND
Tetrachlorobiphenyls												
66	95±51	43±30	353±141	138±77	64±26	12±7	191±52	73±19	84±21	34±12	174±53	87±24
(70+76)	96±48	43±30	320±108	124±63	56±23	11±7	149±49	63±18	82±22	32±12	164±56	78±18
(56+60)	50±22	21±11	184±56	69±32	24±11	1±1	80±19	27±7	35±8	13±4	69±24	32±7
52	43±18	18±10	148±57	61±33	24±9	5±3	72±17	26±5	31±7	13±5	54±15	28±6
47	30±11	16±11	140±50	56±26	22±10	1±1	60±15	24±6	28±8	11±5	54±16	30±6
44	31±10	13±5	99±34	37±18	21±8	3±3	53±10	20±4	27±6	10±4	40±10	21±5
74	24±13	10±7	91±24	33±18	14±5	2±2	45±13	16±4	17±4	6±2	34±9	17±4
49	22±10	10±6	72±47	25±15	12±5	1±2	38±12	14±4	17±5	6±3	24±10	14±4
64	15±12	7±6	64±22	24±14	11±5	2±2	35±9	12±4	16±6	5±3	29±9	13±4
42	9±3	4±2	22±6	8±4	4±2	1±1	12±3	5±1	6±1	2±1	9±3	5±1
48	1±1	ND	16±8	6±4	1±2	ND	2±2	ND	2±2	ND	2±2	ND
53	ND	ND	5±2	1±2	2±5	ND	1±1	ND	1±1	ND	ND	ND
40	ND	ND	4±5	1±1	ND	ND	1±2	ND	1±1	ND	2±2	ND
(41+71)	3±6	ND	1±5	ND	ND	ND	ND	ND	1±1	ND	ND	ND
46	ND	ND	3±4	ND	ND	ND	1±1	ND	ND	ND	ND	ND

Table 2 – continued

Pentachlorobiphenyls												
101	144±67	62±31	609±228	237±130	101±31	21±8	366±103	133±37	128±35	52±20	271±69	136±30
84	127±81	60±43	636±294	254±153	84±34	19±9	323±99	129±42	108±32	46±18	276±84	142±34
118	133±77	60±41	634±230	242±133	80±35	16±8	310±105	115±40	100±30	39±13	271±89	136±24
110	135±80	61±45	475±185	183±99	85±33	18±7	309±88	113±33	114±32	45±17	256±79	125±25
(87+97)	111±63	36±54	390±117	156±70	76±30	15±9	245±92	94±32	135±46	54±25	258±93	117±35
105	55±38	24±19	253±101	101±60	34±16	6±4	138±44	50±15	48±15	19±7	121±40	56±13
95	50±24	22±13	170±58	68±38	32±11	8±3	112±27	41±10	42±9	17±5	75±20	38±11
85	28±13	12±7	143±44	54±26	16±7	4±2	74±20	27±8	24±5	9±4	62±19	31±6
92	15±7	6±4	164±69	64±37	14±7	3±3	63±28	24±13	22±10	9±5	41±12	21±6
82	20±12	7±6	51±19	19±9	10±4	1±2	46±13	15±6	16±5	5±2	33±10	16±3
91	18±10	5±3	58±20	20±11	10±5	1±1	42±11	14±3	16±4	6±3	32±10	15±3
99	12±5	4±2	43±14	15±7	7±3	1±1	24±6	8±2	12±3	4±2	21±7	10±3
Hexachlorobiphenyls												
153	182±125	92±66	1080±405	419±214	129±46	33±14	556±200	217±73	153±42	72±28	462±148	242±37
138	131±79	60±43	601±270	237±140	81±27	18±8	336±107	129±42	108±29	44±16	284±85	140±31
149	122±71	53±37	327±123	124±64	67±21	16±6	290±91	109±35	97±26	39±13	233±66	117±26
146	52±31	22±15	184±71	70±41	29±10	5±3	122±36	44±14	39±12	15±6	104±33	49±12
141	44±26	18±14	196±66	79±44	26±8	7±2	115±39	43±14	33±8	14±4	84±25	42±7
128	37±24	16±12	173±79	66±43	24±9	4±3	93±30	32±10	30±9	12±5	76±22	36±8
151	25±14	11±7	125±45	50±30	15±5	2±2	68±22	25±8	19±4	8±2	51±16	24±5
132	36±10	10±9	71±23	26±14	12±8	ND	48±12	16±5	27±7	8±4	41±12	18±7
156	17±11	8±6	81±41	31±25	9±3	1±1	47±20	18±6	13±5	5±2	39±16	18±4
136	21±13	2±6	60±20	21±13	6±8	2±5	48±10	13±8	17±7	ND	32±8	24±5
129	6±5	2±3	23±12	8±6	1±2	ND	10±9	2±3	6±3	1±2	11±9	1±1
Heptachlorobiphenyls												
180	104±67	49±32	471±237	195±122	62±20	17±7	272±94	111±37	78±20	35±13	218±64	118±29
(182+187)	70±37	31±20	285±83	110±51	41±15	16±4	167±50	67±22	52±13	29±13	143±44	73±13
(170+190)	42±30	20±15	206±106	82±57	26±7	7±3	111±40	46±16	32±10	14±5	87±26	45±14
183	42±26	18±12	164±65	67±39	21±8	5±3	93±39	36±14	28±9	13±5	75±22	37±8
177	20±14	9±7	65±22	25±18	12±3	3±2	58±21	24±8	15±6	6±2	46±19	22±6
174	16±8	10±7	65±29	23±14	11±3	3±2	50±17	19±7	12±3	7±2	40±12	20±5
178	18±7	5±5	77±27	30±16	10±3	1±1	38±14	15±6	11±3	4±3	32±10	16±3
171	15±8	5±4	64±26	27±16	8±2	ND	9±4	14±6	7±5	2±3	25±8	14±3
185	4±3	1±1	15±7	5±3	2±1	ND	9±4	3±2	2±1	ND	6±2	3±3
173	ND	ND	2±3	ND	ND	ND	1±1	ND	ND	ND	ND	ND

Table 2 - continued

Octachlorobiphenyls												
(196+203)	23±18	14±8	125±53	53±31	16±5	3±4	72±23	33±11	20±6	11±4	57±16	31±6
201	20±16	12±8	110±40	46±24	7±8	2±3	64±22	29±9	18±5	9±3	48±14	27±5
194	10±8	6±3	56±35	25±16	8±3	3±2	33±11	15±4	9±2	5±2	24±6	14±3
195	3±4	1±2	27±15	10±7	1±1	ND	14±5	5±3	1±2	ND	10±4	3±3
198	1±1	ND	7±4	2±2	ND	ND	3±3	ND	ND	ND	1±1	ND
205	1±2	ND	8±14	3±4	ND	ND	4±4	ND	ND	ND	ND	ND
Nonachlorobiphenyls												
206	5±4	2±3	24±16	11±8	2±3	ND	13±4	7±3	1±2	1±2	10±5	6±3
207	1±1	ND	9±4	3±3	ND	ND	5±4	1±1	ND	ND	1±1	ND
Decachlorobiphenyl												
209	2±4	ND	16±10	7±6	1±1	ND	8±4	2±3	ND	ND	4±4	1±1
Total congeners												
Mean	2379	1066	9966	3881	1449	288	5656	2132	1968	794	4646	2310
SD	1303	716	3570	2069	524	149	1692	642	541	292	1380	484
Non-*ortho* chloro substituted congeners												
Tetrachlorobiphenyls												
77	5	2	18	8	6	ND	11	4	8	3	10	5
81	24	10	90	38	13	ND	30	9	26	8	29	14
Pentachlorobiphenyl												
126	ND	ND	19	7	ND	ND	10	ND	ND	ND	ND	ND
Hexachlorobiphenyl												
169	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

Table 3

Congener	Brown trout		Lake trout		Rainbow trout				Coho salmon			
					Small		Large		Small		Large	
	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle
Trichlorobiphenyls												
Total	2.0±1.3	1.6±0.8	1.0±0.3	0.9±0.2	1.0±0.2	0.1±0.1	1.0±0.4	0.7±0.2	1.1±0.2	1.0±0.2	0.6±0.1	0.6±0.1
Tetrachlorobiphenyls												
66	4.0±0.4	4.1±0.4	3.6±0.4	3.6±0.5	4.3±0.9	4.4±0.5	3.5±0.5	3.5±0.4	4.3±0.2	4.3±0.2	3.8±0.4	3.7±0.5
(70+76)	4.1±0.4	4.0±0.5	3.3±0.8	3.3±0.8	3.8±0.8	4.2±0.4	3.5±0.5	3.0±0.4	4.2±0.2	4.0±0.2	3.5±0.4	3.4±0.3
(56+60)	2.1±0.3	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Total	18.4±2.9	17.9±2.6	15.4±1.4	15.3±1.4	17.0±3.9	11.5±5.3	13.9±2.0	13.5±1.9	17.9±0.8	17.1±0.8	14.2±1.0	14.1±1.0
Pentachlorobiphenyls												
101	6.3±0.7	6.3±0.9	6.1±0.6	6.0±0.6	7.2±0.8	7.7±1.2	6.5±0.4	6.3±0.5	6.5±0.7	6.5±0.8	5.9±0.4	5.9±0.3
84	5.1±0.7	5.5±0.5	6.3±0.7	6.4±0.5	5.6±1.0	6.6±1.2	5.7±0.3	6.0±0.4	5.4±0.4	5.8±0.3	5.9±0.5	6.1±0.4
118	5.5±0.6	5.5±0.6	6.3±0.4	6.2±0.5	5.3±0.8	5.4±0.6	5.4±0.5	5.3±0.4	5.0±0.3	5.0±0.3	5.8±0.4	5.9±0.4
110	5.6±0.3	5.6±0.3	4.8±0.9	4.9±0.8	5.8±0.3	6.3±0.8	5.5±0.2	5.3±0.3	5.8±0.2	5.7±0.2	5.5±0.4	5.4±0.5
(87+97)	4.6±0.5	6.0±0.7	4.1±1.0	4.3±0.9	5.3±0.8	5.6±1.6	4.3±0.5	4.4±0.6	6.8±0.6	6.7±0.6	5.4±0.7	5.0±0.6
105	2.2±0.3	2.1±0.3	2.5±0.1	2.6±0.2	2.2±0.8	2.4±0.2	2.4±0.1	2.3±0.1	2.4±0.2	2.4±0.1	2.6±0.2	2.4±0.2
95	2.2±0.3	2.1±0.3	1.7±0.1	1.7±0.1	2.2±0.2	2.7±0.5	2.0±0.2	2.0±0.2	2.2±0.4	2.2±0.3	1.6±0.2	1.6±0.2
85	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Total	35.5±1.8	36.4±1.6	36.5±1.2	36.6±0.8	37.5±3.2	38.2±4.9	36.4±0.8	35.9±1.1	38.7±1.2	38.3±1.3	36.9±1.1	36.4±0.8
Hexachlorobiphenyls												
153	7.3±0.9	8.5±0.9	10.8±1.3	10.9±0.9	9.1±1.3	11.8±1.4	9.7±1.0	10.1±0.7	7.8±0.4	9.0±0.9	9.9±1.1	10.6±0.6
138	5.4±0.4	5.5±0.3	5.9±0.7	6.0±0.6	5.7±0.5	6.4±1.1	5.9±0.2	6.0±0.3	5.5±0.2	5.6±0.1	6.1±0.2	6.0±0.2
149	5.1±0.4	4.9±0.3	3.4±1.0	3.4±1.0	4.8±0.6	5.7±0.9	5.1±0.2	5.1±0.2	4.9±0.2	4.9±0.2	5.0±0.2	5.0±0.2
146	2.1±0.3	2.0±0.3	1.8±0.2	1.8±0.1	2.0±0.2	2.1±0.5	2.2±0.2	2.0±0.1	2.0±0.1	1.9±0.1	2.2±0.1	2.1±0.1
141	<2	<2	<2	<2	<2	2.4+0.5	2.0+0.2	<2	<2	<2	<2	<2
Total	27.7±2.0	27.1±1.7	29.2±1.4	29.0±1.1	28.0±2.9	31.4±4.4	30.4±1.4	30.1±1.5	27.6±0.8	27.4±0.6	30.5±1.2	30.3±0.6

Table 3 - continued

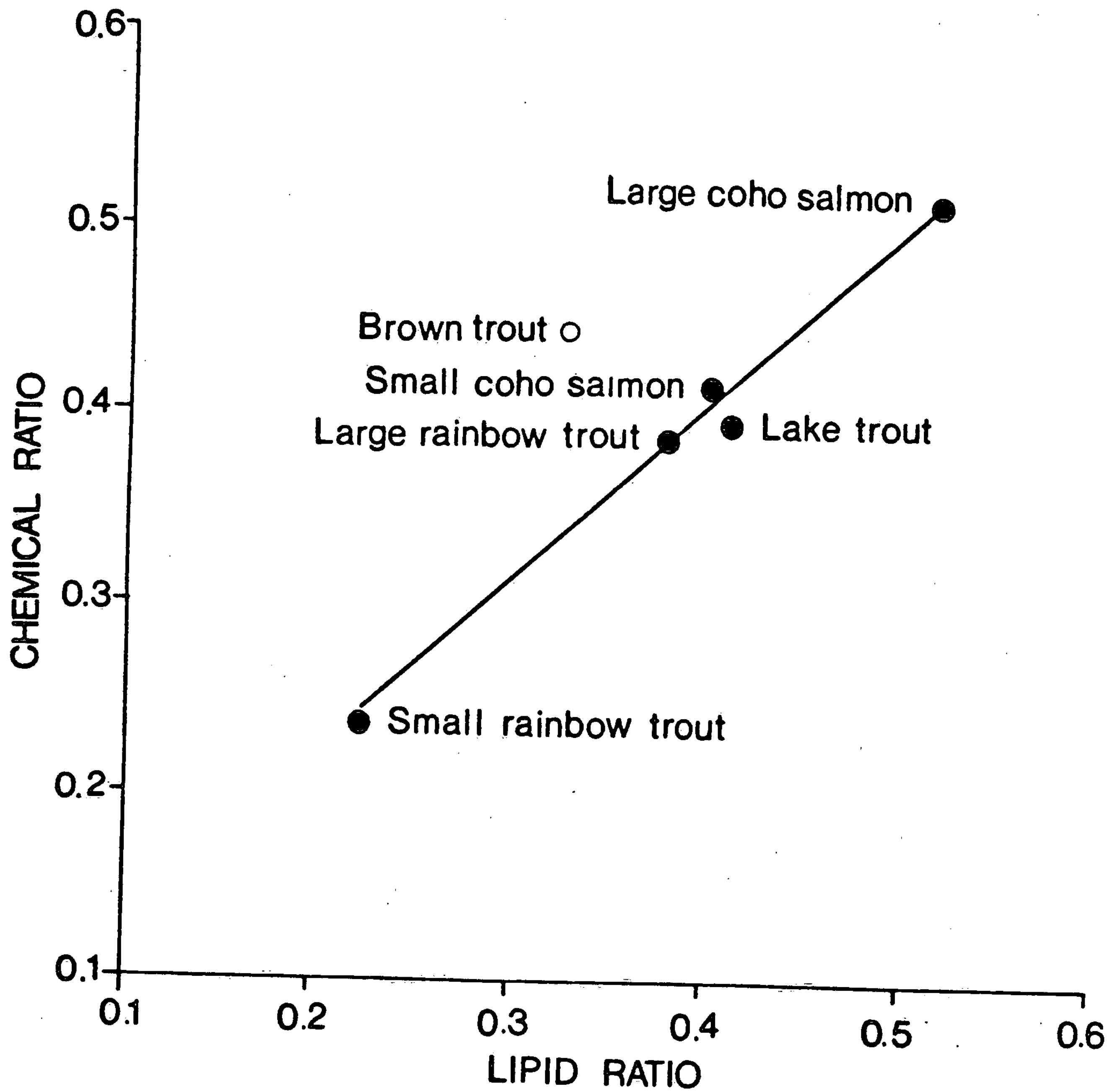
Heptachlorobiphenyls												
180	4.2±0.4	4.6±0.4	4.6±0.7	4.8±0.6	4.5±0.9	6.1±1.4	4.7±0.4	5.1±0.4	4.0±0.1	4.5±0.3	4.7±0.3	5.1±0.4
(182+187)	3.0±0.3	2.9±0.2	2.9±0.4	3.0±0.4	2.9±0.5	3.8±0.6	3.0±0.2	3.2±0.2	2.7±0.1	2.9±0.2	3.1±0.2	3.2±0.2
(170+190)	1.7±0.2	1.9±0.1	2.0±0.5	2.0±0.4	1.9±0.4	2.7±0.6	1.9±0.2	2.1±0.3	1.6±0.1	1.8±0.1	1.9±0.2	1.9±0.3
183	<2	<2	<2	<2	<2	2.2±1.0	<2	<2	<2	<2	<2	<2
Total	13.9±1.0	13.7±1.2	14.1±0.9	14.2±0.9	14.0±3.3	16.6±3.8	14.6±1.0	15.6±1.1	12.3±0.6	13.1±0.9	14.4±0.8	15.0±1.0
Octachlorobiphenyls												
Total	2.2±0.7	3.1±0.3	3.3±0.5	3.5±0.5	2.4±1.1	2.3±1.8	3.3±0.3	3.8±0.3	2.4±0.2	3.1±0.3	3.0±0.2	3.2±0.3
Nonachlorobiphenyls												
Total	0.2±0.1	0.1±0.1	0.3±0.1	0.3±0.2	0.1±0.1	0	0.3±0.1	0.3±0.1	0.1±0.1	0.1±0.1	0.2±0.1	0.3±0.1
Decachlorobiphenyl												
Total	0.1±0.1	0	0.2±0.1	0.2±0.1	0	0	0.1±0.1	0.1±0.1	0	0	0.1±0.1	0.1±0.1
Non-*ortho* chloro substituted congeners												
Tetrachlorobiphenyls												
77	0.2	0.2	0.2	0.2	0.4	0	0.2	0.2	0.4	0.4	0.2	0.2
81	1.0	0.9	1.0	1.0	0.9	0	0.5	0.4	1.3	1.0	0.6	0.6
Pentachlorobiphenyl												
126	0	0	0.2	0.2	0	0	0.2	0	0	0	0	0
Hexachlorobiphenyl												
169	0	0	0	0	0	0	0	0	0	0	0	0

Table 4

Chemical	Brown trout		Lake trout		Rainbow trout				Coho salmon			
					Small		Large		Small		Large	
	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle	Fish	Muscle
DDE	486±448	203±203	1982±1203	618±486	257±114	52±34	1073±504	372±107	391±124	154±58	964±315	456±125
DDD	31±16	15±12	218±114	83±56	31±17	8±6	125±44	42±11	25±6	12±4	70±32	27±14
DDT	59±24	26±12	160±94	64±49	35±11	8±2	87±33	38±8	58±14	24±9	79±25	36±10
Mirex	77±61	41±33	430±241	183±131	51±27	16±3	246±82	112±36	45±35	33±15	203±63	110±31
Photo-mirex	38±22	16±12	196±110	78±52	25±15	6±4	129±58	52±19	26±8	12±5	108±31	54±15
HCB	25±7	10±4	90±43	37±25	20±8	5±2	42±7	16±3	26±6	10±5	24±7	13±3
OCS	18±10	7±5	130±101	52±45	14±7	4±3	52±13	18±4	16±6	7±3	36±9	16±7
γ-Chlordane	3±1	1±1	48±30	19±15	10±2	2±1	24±3	8±2	13±3	5±2	15±4	8±2
α-BHC	25±5	11±2	22±7	8±5	11±5	3±2	12±4	5±1	16±2	6±2	4±1	1±1
Lindane	5±1	2±1	4±1	2±1	2±1	ND	2±1	1±1	4±1	2±1	ND	ND
QCB	7±5	3±2	10±4	4±2	2±1	ND	5±1	2±1	4±1	2±1	2±1	ND
TECB	5±7	2±3	4±2	2±3	1±1	ND	ND	ND	1±1	ND	ND	ND

Table 5

Congener	Zell			Albro			Capel			Mullin			Present study
	A50	A60	56%	1254	1260	56%	1254	1260	56%	1254	1260	56%	
153	3.3	10.4	5.7	3.3	8.2	5.0	3.1	1.3	2.5	6.2	10.6	7.7	9.0
101	8.1	6.6	7.6	7.0	5.0	6.3	8.4	4.0	7.0	7.1	2.5	5.6	6.4
138	4.8	12.4	7.3	–	–	–	9.1	1.3	6.5	5.9	5.2	5.7	5.8
118	0.5	0.6	0.5	8.1	2.0	6.1	11.5	0	7.7	6.1	0.6	4.3	5.6
149	5.8	–	–	3.6	9.5	5.6				5.9	8.7	6.8	4.7
84	2.5	0.4	1.8	1.7	0.7	1.4	–	–	–	4.2	0.2	2.9	5.6
110	1.1	0.1	0.8	8.6	3.6	6.9	10.3	1.6	7.4	9.6	1.4	6.9	5.5
180	0.4	6.2	2.3	0.8	7.2	2.9	0.5	6.5	2.5	1.2	12.1	4.8	4.4
(87+97)	7.0	1.5	5.2	6.4	1.7	4.8	6.4	0.5	4.4	5.5	0.4	3.8	5.2


Large coho salmon
Brown trout
Small coho salmon
Large rainbow trout
Lake trout
Small rainbow trout
CHEMICAL RATIO
LIPID RATIO
0.1
0.2
0.3
0.4
0.5
0.6