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Evaluation and Interconversion of Various Indicator PCB Schemes for ∑PCB and Dioxin-Like PCB Toxic Equivalent Levels in Fish

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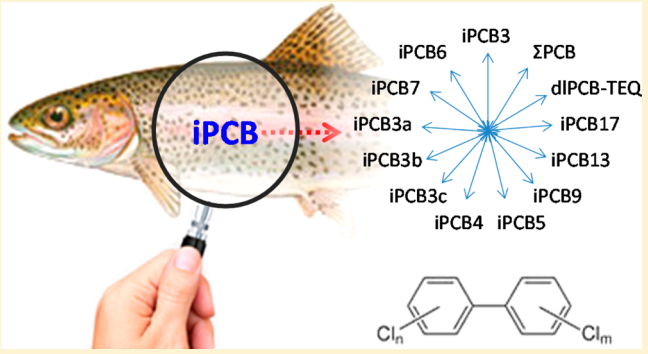
Supporting Information

ABSTRACT: Polychlorinated biphenyls (PCBs) remain chemicals of concern more than three decades after the ban on their production. Technical mixture-based total PCB measurements are unreliable due to weathering and degradation, while detailed full congener specific measurements can be time-consuming and costly for large studies. Measurements using a subset of indicator PCBs (iPCBs) have been considered appropriate; however, inclusion of different PCB congeners in various iPCB schemes makes it challenging to readily compare data. Here, using an extensive data set, we examine the performance of existing iPCB3 (PCB 138, 153, and 180), iPCB6 (iPCB3 plus 28, 52, and 101) and iPCB7 (iPCB6 plus 118) schemes, and new iPCB schemes in estimating total of PCB congeners (∑PCB) and dioxin-like PCB toxic equivalent (dlPCB-TEQ) concentrations in sport fish fillets and the whole body of juvenile fish. The coefficients of determination (R^2) for regressions conducted using logarithmically transformed data suggest that inclusion of an increased number of PCBs in an iPCB improves relationship with ∑PCB but not dlPCB-TEQs. Overall, novel iPCB3 (PCB 95, 118, and 153), iPCB4 (iPCB3 plus 138) and iPCB5 (iPCB4 plus 110) presented in this study and existing iPCB6 and iPCB7 are the most optimal indicators, while the current iPCB3 should be avoided. Measurement of ∑PCB based on a more detailed analysis (50+ congeners) is also overall a good approach for assessing PCB contamination and to track PCB origin in fish. Relationships among the existing and new iPCB schemes have been presented to facilitate their interconversion. The iPCB6 equiv levels for the 6.5 and 10 pg/g benchmarks of dlPCB-TEQ_{05} are about 50 and 120 ng/g ww, respectively, which are lower than the corresponding iPCB6 limits of 125 and 300 ng/g ww set by the European Union.



INTRODUCTION

Polychlorinated biphenyls (PCBs) are a class of persistent organic pollutants (POPs) and consist of 209 theoretical congeners that encompass a broad range of physicochemical properties and bioaccumulation potentials.[1,2] After initial commercial production in 1929, PCBs found their way in various industrial uses due to their high thermal stability and other properties.[1] However, these properties resulted in a widespread environmental PCB contamination in many parts of the world through both use and disposal.[3] Ecological and human health concerns from environmental and other exposures resulted in a ban on their manufacturing in North America in the late 1970s and in most of Europe in the 1980s.[2,4] However, their highly persistent, bioaccumulative and toxic nature has resulted in ongoing environment and related health concerns.[5,6]

PCBs used for commercial applications were produced as technical mixtures containing varying numbers and concentrations of congeners, and marketed under different trade names such as Aroclor, Clophen, Kanechlor, Phenoclor and Pyroclor.[7] Various analytical methods have been developed to measure levels of PCBs.[8,9] In general, the simplest methods are based on the approach of "technical mixture equivalent total PCB", which employs a commercial product or mixtures of products as a standard to quantitate the matching congeners contained in the product/mixture and sample.[9] An intermediate method aims to measure concentrations of various PCB homologues based on the number of chlorines in the congeners, while a more detailed approach targets various individual congeners with a goal of measuring as many of them as practically possible.[9] Another exhaustive method measures

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parts per trillion levels of the 12 dioxin-like PCB congeners (dlPCBs), some of which are among the most toxic PCBs.[10] As it can be discerned, detailed methods are more expensive due to greater analytical requirements.[11]

An extensive number of PCB studies have highlighted that only a subset of the theoretical 209 congeners are generally present in various matrices.[12] Hence, it is considered unpractical and unnecessary to analyze samples for all 209 PCB congeners, and summation of measurements of all major detected congener or ∑PCB is commonly used as a surrogate of total PCB. Further, numerous studies have also shown that total PCB composition is generally dominated by only a handful of congeners.[12−15] As a result, various monitoring and regulatory agencies have permitted measurements of a few selected PCB congeners, known as indicator PCBs (iPCBs), to fulfill risk assessment requirements and to develop health protection guidelines, for example, refs 5, 9, 16, and 17. Since measurements of PCBs have been reported in different ways, such as the sum of three congeners (PCB 138, 153, and 180), six congeners (PCB 28, 52, 101, 138, 153, 180) and seven congeners (sum of six indicator PCBs plus PCB 118), a direct comparison of data is challenging.[15]

In this study, we provide interconversion factors for various iPCB schemes and ∑PCB measurements in both fillets of sport fish and whole-body of juvenile fish to facilitate a comparison of reported iPCB data. In addition, the relationships of the iPCB schemes with dlPCB toxic equivalents (dlPCB-TEQ) have been presented for sport fish fillets. We also explore if inclusion of three different congeners or more congeners (up to 17) improves the estimation of ∑PCB and dlPCB-TEQ in fish. Any such improvements are also judged against augmented analytical demand.

MATERIALS AND METHODS

Data Source. This study considered fish PCB data collected by the Province of Ontario, Canada. Levels of various contaminants have been monitored in Ontario fish since the 1970s through Sport Fish Contaminant Monitoring Program of Ontario Ministry of the Environment and Climate Change (OMOECC).[18] The monitoring data generated by the program include 1151 fish samples measured for 56 PCB congeners. After excluding 112 samples with all major PCB congeners below their detection limits, the final data set included 572 skinless, boneless fillet (SBF) samples of 25 sport fish species from 51 locations, 22 whole body fish composite (WFC) samples of two forage fish species from one location, and 445 young-of-the-year whole body fish composite (YFC) samples of 11 fish species from 60 locations (Supporting Information (SI) Table S1a). Only 8 of 98 SBF location/species combinations were sampled over multiple years. There were more (29 of 112) location/species repeat measurements for YFC; however, most of the repeat measurements were conducted for only 2−4 times per location/species within a period of 3−5 years. As such, a major impact of the differential environmental weathering rates for the congeners on the results presented in this study is not expected. Since we had limited data for whole body forage fish, we provide a limited analysis and results, which should be viewed with caution. In addition, 470 SBF samples of 20 sport fish species (SI Table S1b) from 53 locations were measured for PCB congeners including dlPCBs. The samples were collected between 1996 and 2010, and only 6 of the 96 location/species were repeat measurements.

Analytical Details. Fish samples were analyzed for 56 PCB congeners using OMOECC method 3411.[19] Briefly, ∼5 g tissue samples were spiked with the surrogate decachlorobiphenyl and digested overnight with hydrochloric acid and then extracted with hexane/dichloromethane. Sample extracts were reduced in volume with a gentle stream of nitrogen and then cleaned using dry packed Florisil columns. The column was eluted with 20−25 mL of hexane and concentrated using a gentle stream of nitrogen. Extracts were analyzed for PCB congeners using an HP 6890 GC and ^{63}Ni electron capture detector (ECD) using a DB-1701 and DB-5 GC columns (20 m × 0.1 mm i.d. × 0.1 μm film thickness, J&W Scientific, Folsom, CA). The column head pressure was 66.8 PSI and the temperature program was 90 °C for 1 min, 90 to 160 °C at 35.5 °C/min; 150−200 °C at 71 °C/min; 200 to 275 °C at 5.3 °C/min then hold for 5 min. Each congener must be detected on both columns at its specific retention time. Samples were quantified with a 5-point calibration curve ranging from 1 to 500 pg with single point continuing calibration verification. Method blanks and matrix spikes were processed with every set of 20−30 samples. Method detection limits (MDLs) were 3 times standard deviation of the analytical measurements of the set and were statistically calculated using eight spiked blank matrix compounds. The MDLs ranged from 0.1 to 2 ng/g for individual congeners and 1−5 ng/g for ∑PCB. The MDL for ∑PCB was calculates as the root sum square of the individual MDLs and then rounded to the nearest 1, 2, or 5.

Twelve dlPCBs, along with 17 2,3,7,8-substituted polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), were analyzed using the OMOE method E3418[20] as previously described.[21] Homogenized ∼5 g wet weight tissue samples were fortified with ^{13}C-dlPCBs and ^{13}C-PCDD/Fs surrogates (Wellington Laboratories, Guelph, Ontario, Canada) then digested in hydrochloric acid and extracted with hexane. The extracts were reduced in volume, and then passed through an anhydrous sodium sulfate/sulfuric acid−silica column followed by a two-stage open column cleanup. The first column contained multilayered silica and was eluted with hexane. The second column consisted of Amoco PX21 carbon-activated silica and was eluted with 40 mL 25% dichloromethane/hexane (v/v) (Fraction 1; mono-ortho PCBs). The column was then inverted to reverse the flow and eluted with 160 mL toluene to isolate the coplanar PCBs and PCDD/Fs (Fraction 2). Analysis of the dlPCBs and PCDD/Fs (Fractions 1 and 2 in separate runs) was by gas chromatography-high resolution mass spectrometry (GC-HRMS) using a Waters Autospec HRMS coupled to a Hewlett-Packard HP6890 GC (Agilent Technologies, Willmington, DE) using a 40 m DB-5 column (0.18 mm i.d., 0.18 μm film thickness; J&W Scientific, Folsom, CA). Method blanks and matrix spikes using pretested clean Alaskan Pollock tissue were processed with every 10 samples. Congener-specific MDLs varied based on matrix effects of individual samples and ranged from 0.36 to 6 and 0.02−10 pg/g ww for dlPCBs and PCDD/Fs, respectively. Concentrations were reported if values exceeded five times the blank values for dlPCBs and PCDD/Fs.

The performance of the methods has been periodically monitored through laboratory intercalibration studies (the Northern Contaminants Program - NCP, and Quality Assurance of Information for Marine Environmental Monitoring in Europe - QUASIMEME).

Statistical Analysis. We considered 11 indicator PCB schemes: existing - iPCB3 (PCB 138, 153 and 180), iPCB6

(PCB 28, 52, 101, 138, 153, and 180) and iPCB7 (PCB 28, 52, 101, 118, 138, 153, and 180); new−iPCB3a (PCB 95, 118, and 153), iPCB3b (PCB 110, 138, and 153), iPCB3c (PCB 118, 138, and 153), iPCB4 (PCB 95, 118, 138, and 153), iPCB5 (PCB 95, 110, 118, 138, and 153), iPCB9 (PCB 28, 52, 101, 105, 118, 138, 153, 156, and 180), iPCB13 (PCB 28, 52, 95, 99, 101, 105, 118, 138, 153, 156, 170, 180, and 187), and iPCB17 (PCB 18, 28, 52, 74, 95, 99, 101, 105, 110, 118, 138, 149, 153, 156, 170, 180, and 187). The iPCB3 scheme has been used in reporting PCB data,[15] while the iPCB6 and iPCB7 schemes are used worldwide, especially in Europe, and are based on PCB congeners from different levels of chlorination and their greater abundance.[17] This is important considering that many lower chlorinated congeners are relatively rapidly degraded by fish and therefore do not bioaccumulate to the degree that the more highly chlorinated congeners do. The iPCB6 scheme has been adopted by the European Union to regulate PCB levels in foodstuff.[22] The iPCB3a, iPCB3b, iPCB3c, iPCB4, and iPCB5 schemes were formulated in this study based on the Partial Least Square (PLS) regression analysis as explained below. The iPCB9, iPCB13 and iPCB17 schemes were also created because they have potential for a practical use due to relatively greater abundance of those congeners. ∑PCB was calculated by summing all 56 individual PCB congeners measured. Values below the detection limits were treated as detection limits; however, negligible influence of such a treatment was confirmed by performing statistical analysis on values based on nondetects equal to zero.

Toxic equivalent (TEQ) concentrations of dlPCBs were calculated using World Health Organization (WHO) mammalian toxic equivalency factors presented in 1994 (dlPCB-TEQ_{94}), 1998 (dlPCB-TEQ_{98}) and 2005 (dlPCB-TEQ_{05}) (SI Table S2).[23] The dlPCB-TEQ concentrations were calculated by summing multiplications of individual dlPCB with the corresponding toxic equivalency factor (TEF).[24] For dlPCB measurements below the detection limits, the values were treated as half of the detection limits, which is a normal practice in environmental studies. The impact of such a treatment on relationships between iPCBs and dlPCB-TEQ is expected to be negligible.[11] Since the difference between dlPCB-TEQ_{94} and dlPCB-TEQ_{98} is negligible (1%),[23] we discuss results for dlPCB-TEQ_{94} and dlPCB-TEQ_{05} only. Since this study focuses on PCBs, the PCDD/F measurements, which are also included in the total-TEQ calculations, were not considered. It should be noted that although dlPCB-TEQ contribution to total-TEQ could be minimal to very high depending on the contamination history of a site, generally dlPCBs are the major (>50%) contributors to total-TEQ.

Based on information in published studies,[13,14,23] the relationship among different iPCBs, ∑PCB, and dlPCB-TEQ values were developed using linear regressions passing through the origin. Since iPCBs considered here are among the most dominating PCB congeners in fish, it is reasonable to assume that when iPCBs are zero, ∑PCB, and dlPCB-TEQ would be ∼0 as well. Most of the original data failed the Shapiro-Wilk normality test (mostly p-value <0.05; SI Table S3), and therefore the linear regressions were also performed on logarithmically transformed values to consolidate the robustness of the statistical analyses. Further, normality tests (SI Table S3) and regressions were also conducted on a revised PCB congener data set after removing distinct Lyons Creek data and a revised dlPCB data set by removing samples with most PCB congeners below the detection limits. The results for YFC should be viewed with caution because the logarithmic transformed and revised YFC data sets also failed the Shapiro-Wilk normality test (SI Table S3). Although most of the dlPCB-TEQ related data failed the Shapiro-Wilk normality test even after a log transformation, the log transformed values of filtered data after removing samples measuring most of congeners below detection had p-values of >0.05 for the Shapiro-Wilk normality test (SI Table S3c).

To examine if the current iPCB3, iPCB6, and iPCB7 schemes consider the most optimum set of congeners, a Partial Least Squares (PLS) regression analysis was conducted using the PCB congener and ∑PCB measurements for SBF. A PLS regression analysis is useful in parsing out relationships between numerous predictors and a response variable, especially when there is multicollinearity among the predictors. The analysis was carried using the plsr function and leave-one-out cross-validation in R (version 3.0.3). The first component explained 65% of the variance in ∑PCB, and the first four components explained 90% of the variance (SI Figure S1a). The loadings of the first component highlighted importance of five PCBs in the decreasing order: PCB 95, 153, 118, 101, and 138 (SI Table S4). Based on these results, we formulated and evaluated performance of iPCB3a (PCB 95, 118, and 153), iPCB3b (PCB 110, 138, and 153), iPCB3c (PCB 118, 138, and 153), iPCB4 (PCB 95, 118, 138, and 153), and iPCB5 (PCB 95, 110, 118, 138, and 153). The goodness-of-fit statistic used to draw inference in the study was the coefficient of determination (R^2), although we recognize that its values could be misleading at times. For instance, low R^2 values result when there is little variability in the observed data to predict, while high R^2 values can be obtained despite a strong systematic bias.[25] Further, because of the nature of the complexity problem tackled in our analysis, that is, comparison of same model structures (simple linear regression models) with predictors that vary with respect to their analytical error, the typical measures of fit that penalize model complexity (e.g., Akaike Information Criterion) could not be used efficiently. In this regard, one alternative approach could have been the development of "errors-in-variables" models that explicitly accommodate the uncertainty of the predictor variables, assuming that complete characterization of the associated error is feasible.[26]

RESULTS

Observed ∑PCB and dlPCB-TEQs, and iPCB Contributions. The data set represented a good range of PCB contamination with concentrations of ∑PCB ranging from 18 to 14 800 ng/g ww (25−75th percentile: 105−2630 ng/g ww) for the sport fish fillet samples, and from 17 to 185 000 ng/g ww (25−75th: 123−4770 ng/g ww) for juvenile fish. The concentrations of dlPCB-TEQ_{94} ranged from 0.01 to 379 pg/g ww (25−75th: 2−19 pg/g ww) and dlPCB-TEQ_{05} ranged from 0.01 to 223 (25−75th: 1.6−15) pg/g ww.

For skinless boneless sport fish fillets (SBF), approximate average contributions of iPCBs to ∑PCB are 18% for iPCB3, 29% for iPCB6, 35% for iPCB7, 20% for iPCB3a, 19% for iPCB3b, 20% for iPCB3c, 26% for iPCB4, 30% for iPCB5, 38% for iPCB9, 52% for iPCB13 and 65% for iPCB17 (Figure 1). The corresponding contributions in YOY fish (YFC) are lower at 6−10%, 26%, 30%, 19%, 11%, 11%, 24%, 27%, 33%, 49%, and 64%, respectively (Figure 1). Limited data indicate that the corresponding contribution in whole body levels of forage fish (WFC) are higher at 30%, 41%, 48%, 27%, 26%, 32%, 38%, 39%, 49%, 68%, and 79%, respectively (Figure 1). Contribu-

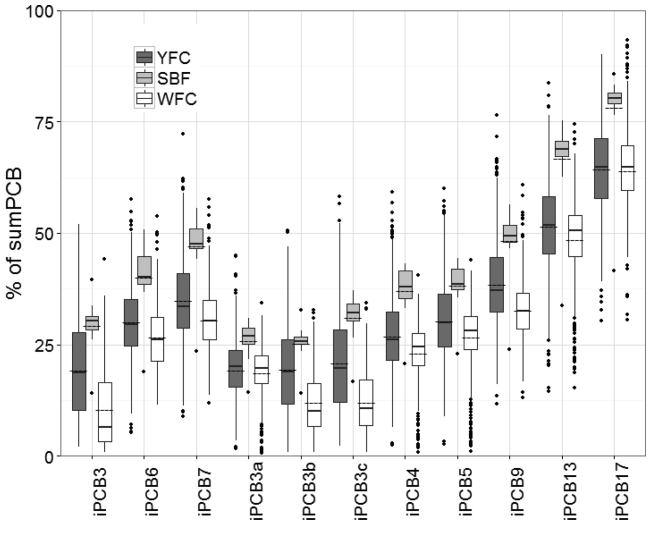

Figure 1. Contributions (as percentage) of various iPCBs to ΣPCB for skin-removed boneless fillets of sport fish (SBF, n = 572), whole fish composite offorage fish (WFC, limited data n = 22), and young-of-the-year fish composites (YFC, n = 445). The solid line in the box presents median, the dash line present mean, box represents 25−75 percentiles, and whiskers present nonoutlier and nonextreme values.

tions of iPCBs to ∑PCB differed by not only type of the tissue analyzed but also by fish species considered (SI Figure S2). However, these species-specific differences declined from iPCB3 to iPCB17 with increasing number of PCB congeners considered (SI Figure S2). The bimodal pattern characterized by two distinct peaks is evident in the relationships of iPCB3 to other iPCB schemes and ∑PCB, and is a result of relatively low levels of iPCB3 in fish samples collected from Lyons Creek (SI Figure S3), which is a historically PCB contaminated site near the Niagara River. A separate set of analyses was also performed on the data after excluding these Lyons Creek measurements.

Relationship Among iPCBs, ∑PCB, and dlPCB-TEQs. A strong relationship is evident between each iPCB scheme and ∑PCB, except for iPCB3 for SBF (Tables 1, SI S5; Figures S4, S5). The relationship of iPCBs with ∑PCB improves as more congeners are considered in an indicator scheme (Tables 1, SI S5; Figures S4, S5). Treatment of nondetects (i.e., ND = DL and ND = 0) had no appreciable impact on the relationships among iPCBs and ∑PCB (SI Tables S5a, S5b, S6).

Similarly, a decent, albeit relatively weak, relationship is also evident between each iPCB scheme and dlPCB-TEQs (Tables 2, SI S7; Figure S6). However, in contrast to those for ∑PCB for which variance explained by iPCB consistently increases from iPCB3 to iPCB17 (Table 1a), variance in dlPCB-TEQs explained by iPCBs generally remain constant between 0.67 and 0.70 for all data and 0.68−0.73 when major nondetects were removed (Table 2). The variance in dlPCB-TEQs explained by iPCBs is generally lower for the recent (2005) TEF scheme (dlPCB-TEQ_{05}) compared to the previous (1994 and 1998) TEF schemes (dlPCB-$TEQ_{94/98}$) (Table 3). Similar to ∑PCB, species-specific differences were evident in relationships between iPCBs and dlPCB-TEQs; however, it appears that these differences are driven by nondetects in iPCBs for which dlPCB-TEQs were highly variable (but mostly low) (SI Figures S7, S8). At these low (nondetect) levels of iPCBs, the dlPCB-TEQs appear to be relatively elevated in fatty, top-predatory fish (SI Figure S9).

iPCB Performance. For SBF and YFC, the R^2 values indicated that the iPCB regression models with a greater number of congeners performed better for both untransformed and natural logarithmically transformed data (Table 1 and SI Table S5). For WFC the differences in R^2 of various iPCBs were relatively minor (Table 1 and SI Table S5). Overall, the results suggest that adding more congeners to the iPCB−∑PCB regression model by the inclusion of up to 17 congeners markedly improves model fit, particularly for SBF and YFC measurements. Compared to the current iPCB3, the new iPCB3a containing PCB 95, 118, and 153 correlated much better with ∑PCB, especially for SBF and YFC (Table 1; SI Figures S4, S5). In contrast, the R^2 values suggested that the iPCB3b, iPCB6, iPCB7 and iPCB9 regression models are consistently best for estimating untransformed values of dlPCB-TEQs (data were available for SBF only) (SI Table S7). However, regression analyses using more appropriate logarithmically transformed dlPCB-TEQ values indicated minor differences in the performance of various iPCB schemes, with iPCB3a and iPCB4 being the best (Table 3b). These results indicate that the iPCB3a iPCB4, iPCB5, iPCB6, and iPCB7 are overall good enough to model ∑PCB and dlPCB-TEQs, and addition of more congeners in an iPCB scheme is not necessary.

DISCUSSION

Overall strong relationships of iPCBs with ∑PCB and dlPCB-TEQs suggest that iPCBs are good surrogates for these more expensive measurements. An order of magnitude variability observed in the concurrent measurements of iPCB3 and ∑PCB suggests that the current iPCB3 is a relatively unreliable indicator of ∑PCB and should be avoided. Relatively high abundance of iPCB17 (68% and 61−66% in sport fish and YOY, respectively) makes it a stronger indicator of ∑PCB. However, accounting for the strength of relationships of iPCBs with dlPCB-TEQs, it appears that newly formulated iPCB3a, iPCB4, and iPCB5, as well as the existing iPCB6 and iPCB7 are overall the best indicator schemes.

The novel iPCB3a presented in this study contain PCB 118 and 153, as well as PCB 95, which has not been considered in the current iPCB3/6/7. It should be noted that PCB 95 and 66 coelute, and can introduce an error in the estimates based on the iPCB3a scheme. The PCB congener method used for the data considered in this study did not look for PCB 66 and as such the coelution with PCB 95 was not a concern. Further, fish do not biotransform PCB 95 but terrestrial animals do. As such, caution should be taken while applying the iPCB3a presented in this study to other than fish.

It is believed that iPCB6 contributes about half of the nondioxin like PCBs in food and feed.[15] For fish considered in this study, ∑PCB is comprised of about 29% and 26% of iPCB6 in sport fish fillets and juvenile whole body, respectively. This is in agreement with increased bioaccumulation of iPCB6 congeners such as PCB 138, 153, and 180 with food web trophic levels, for example, refs 17, 27, and 28. Interestingly, variance in dlPCB-TEQs explained by the iPCB6 scheme, which does not contain any dioxin-like PCB congener, was comparable to iPCB9, iPCB13, and iPCB17. This finding is similar to that reported for skinless fillets of a variety of freshwater fish collected from Rhone River, France-Switzerland, as well as marine fish collected from the French coast and North-Eastern Atlantic Ocean.[14]

Table 1. Relationships (log$Y$ = m log$X$) of Indicator PCBs (iPCBs) and ∑PCB for a Variety of Fish Tissues[a]

	regression equation/coefficient of determination (R^2)											
↓Y X →	log(iPCB3)	log(iPCB6)	log(iPCB7)	log(iPCB3a)	log(iPCB3b)	log(iPCB3c)	log(iPCB4)	log(iPCB5)	log(iPCB9)	log(iPCB13)	log(iPCB17)	log(∑PCB)
(a) Skin-Removed Fillet of Sport Fish												
log(∑PCB)	1.34 ± 0 X	1.24 ± 0.01 X	1.21 ± 0.01 X	1.37 ± 0.02 X	1.33 ± 0.02 X	1.32 ± 0.02 X	1.27 ± 0.02 X	1.24 ± 0.01 X	1.18 ± 0.01 X	1.12 ± 0.01 X	1.08 ± 0 X	
log(iPCB17)	1.24 ± 0.02 X	1.15 ± 0.01 X	1.12 ± 0.01 X	1.27 ± 0.02 X	1.23 ± 0.01 X	1.22 ± 0.01 X	1.18 ± 0.01 X	1.15 ± 0.01 X	1.09 ± 0 X	1.04 ± 0 X		0.982
log(iPCB13)	1.2 ± 0.01 X	1.11 ± 0 X	1.07 ± 0 X	1.23 ± 0.01 X	1.19 ± 0.01 X	1.18 ± 0.01 X	1.14 ± 0.01 X	1.1 ± 0.01 X	1.05 ± 0 X		0.994	0.971
log(iPCB9)	1.14 ± 0.01 X	1.05 ± 0 X	1.02 ± 0 X	1.17 ± 0.01 X	1.13 ± 0.01 X	1.12 ± 0.01 X	1.08 ± 0.01 X	1.05 ± 0.01 X		0.996	0.988	0.965
log(iPCB5)	1.08 ± 0.01 X	1 ± 0.01 X	0.97 ± 0 X	1.11 ± 0.01 X	1.08 ± 0.01 X	1.07 ± 0.01 X	1.03 ± 0 X		0.973	0.977	0.967	0.934
log(iPCB4)	1.05 ± 0.01 X	0.97 ± 0.01 X	0.94 ± 0.01 X	1.08 ± 0.01 X	1.05 ± 0.01 X	1.04 ± 0 X		0.996	0.966	0.968	0.953	0.918
log(iPCB3c)	1.02 ± 0.01 X	0.93 ± 0.01 X	0.91 ± 0.01 X	1.04 ± 0.01 X	1.01 ± 0 X		0.983	0.975	0.951	0.941	0.923	0.885
log(iPCB3b)	1.01 ± 0.01 X	0.93 ± 0.01 X	0.9 ± 0.01 X	1.03 ± 0.01 X		0.985	0.968	0.969	0.940	0.932	0.919	0.889
log(iPCB3a)	0.97 ± 0.01 X	0.9 ± 0.01 X	0.87 ± 0.01 X		0.941	0.957	0.988	0.988	0.959	0.968	0.956	0.917
log(iPCB7)	1.12 ± 0.01 X	1.03 ± 0 X		0.968	0.965	0.969	0.979	0.984	0.992	0.989	0.980	0.955
log(iPCB6)	1.08 ± 0.01 X		0.996	0.954	0.970	0.964	0.969	0.974	0.985	0.980	0.971	0.949
log(iPCB3)		0.955	0.945	0.916	0.974	0.975	0.946	0.937	0.926	0.912	0.893	0.861
(b) Whole Body YOY												
log(∑PCB)	1.56 ± 0 X	1.22 ± 0.01 X	1.19 ± 0.01 X	1.25 ± 0.02 X	1.42 ± 0.02 X	1.42 ± 0.02 X	1.22 ± 0.02 X	1.19 ± 0.01 X	1.18 ± 0.01 X	1.1 ± 0.01 X	1.06 ± 0 X	
log(iPCB17)	1.48 ± 0.02 X	1.15 ± 0.01 X	1.12 ± 0 X	1.18 ± 0.02 X	1.34 ± 0.02 X	1.34 ± 0.02 X	1.15 ± 0.01 X	1.13 ± 0.01 X	1.11 ± 0 X	1.04 ± 0 X		0.996
log(iPCB13)	1.42 ± 0.02 X	1.11 ± 0 X	1.08 ± 0 X	1.14 ± 0.01 X	1.29 ± 0.02 X	1.29 ± 0.01 X	1.11 ± 0.01 X	1.09 ± 0.01 X	1.07 ± 0 X		0.997	0.992
log(iPCB9)	1.33 ± 0.02 X	1.04 ± 0 X	1.01 ± 0 X	1.07 ± 0.01 X	1.21 ± 0.01 X	1.21 ± 0.01 X	1.04 ± 0.01 X	1.02 ± 0.01 X		0.998	0.996	0.991
log(iPCB5)	1.31 ± 0.02 X	1.01 ± 0.01 X	0.99 ± 0.01 X	1.05 ± 0.01 X	1.19 ± 0.01 X	1.19 ± 0.01 X	1.02 ± 0 X		0.982	0.988	0.981	0.973
log(iPCB4)	1.28 ± 0.02 X	0.99 ± 0.01 X	0.96 ± 0.01 X	1.02 ± 0 X	1.16 ± 0.01 X	1.16 ± 0.01 X		0.999	0.983	0.988	0.980	0.973
log(iPCB3c)	1.1 ± 0.01 X	0.85 ± 0.01 X	0.83 ± 0.01 X	0.88 ± 0.01 X	1 ± 0.01 X		0.978	0.976	0.961	0.959	0.949	0.936
log(iPCB3b)	1.1 ± 0.01 X	0.85 ± 0.01 X	0.83 ± 0.01 X	0.88 ± 0.01 X		0.987	0.965	0.968	0.951	0.948	0.941	0.929
log(iPCB3a)	1.24 ± 0.02 X	0.96 ± 0.01 X	0.94 ± 0.01 X		0.950	0.964	0.997	0.997	0.976	0.985	0.976	0.970
log(iPCB7)	1.32 ± 0.02 X	1.02 ± 0 X		0.974	0.951	0.961	0.981	0.980	1.000	0.997	0.995	0.990
log(iPCB6)	1.29 ± 0.02 X		0.999	0.967	0.950	0.958	0.975	0.974	0.998	0.994	0.992	0.987
log(iPCB3)		0.897	0.894	0.878	0.962	0.968	0.904	0.899	0.892	0.881	0.871	0.855
(c) Whole Body Forage Fish												
log(∑PCB)	1.24 ± 0 X	1.17 ± 0.02 X	1.14 ± 0.02 X	1.28 ± 0.02 X	1.28 ± 0.02 X	1.23 ± 0.02 X	1.19 ± 0.02 X	1.18 ± 0.02 X	1.13 ± 0.02 X	1.07 ± 0.01 X	1.04 ± 0.01 X	
log(iPCB17)	1.2 ± 0.01 X	1.13 ± 0.01 X	1.09 ± 0.01 X	1.23 ± 0.01 X	1.23 ± 0.02 X	1.18 ± 0.01 X	1.14 ± 0.01 X	1.14 ± 0.01 X	1.09 ± 0.01 X	1.03 ± 0 X		0.931
log(iPCB13)	1.16 ± 0.01 X	1.09 ± 0.01 X	1.06 ± 0.01 X	1.19 ± 0.01 X	1.2 ± 0.02 X	1.15 ± 0.01 X	1.11 ± 0.01 X	1.1 ± 0.01 X	1.06 ± 0.01 X		0.998	0.915
log(iPCB9)	1.1 ± 0.01 X	1.03 ± 0.01 X	1 ± 0 X	1.13 ± 0.01 X	1.13 ± 0.01 X	1.09 ± 0.01 X	1.05 ± 0.01 X	1.04 ± 0.01 X		0.997	0.996	0.915
log(iPCB5)	1.05 ± 0.01 X	0.99 ± 0.01 X	0.96 ± 0.01 X	1.08 ± 0.01 X	1.08 ± 0.01 X	1.04 ± 0.01 X	1.01 ± 0 X		0.971	0.983	0.986	0.938
log(iPCB4)	1.05 ± 0.01 X	0.98 ± 0.01 X	0.96 ± 0.01 X	1.07 ± 0 X	1.08 ± 0.01 X	1.04 ± 0 X		0.994	0.977	0.989	0.987	0.918
log(iPCB3c)	1.01 ± 0.01 X	0.95 ± 0.01 X	0.92 ± 0.01 X	1.04 ± 0 X	1.04 ± 0.01 X		0.993	0.984	0.985	0.992	0.987	0.906
log(iPCB3b)	0.97 ± 0.01 X	0.91 ± 0.01 X	0.89 ± 0.01 X	1 ± 0.01 X		0.982	0.973	0.980	0.986	0.985	0.988	0.930
log(iPCB3a)	0.97 ± 0.01 X	0.92 ± 0.01 X	0.89 ± 0.01 X		0.975	0.996	0.996	0.986	0.983	0.992	0.988	0.910
log(iPCB7)	1.09 ± 0.01 X	1.03 ± 0.01 X		0.984	0.983	0.986	0.977	0.968	0.999	0.996	0.995	0.913
log(iPCB6)	1.06 ± 0 X		0.997	0.968	0.978	0.972	0.957	0.949	0.996	0.987	0.986	0.900
log(iPCB3)		0.994	0.994	0.976	0.984	0.985	0.965	0.955	0.993	0.987	0.983	0.890

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Table 1. continued

[a]The linear regression equations were prepared for the relationships passing through the origin using logarithmically transformed 861 concentrations (in ng/g wet weight) after removing distinct Lyons Creek related measurements. The equations present regression slopes ±95% confidence intervals for the slopes. Non-detects (ND) were set at the detection limits (DL). Similar relationships prepared using normal data are presented in SI Table S5b. The errors reported as "0" were < 0.01, and R^2 reported as 1 were > 0.999.

The decline in variance of dlPCB-TEQs explained by iPCBs from the TEF schemes of the 1990s to the recent (2005) TEF scheme (Table 2) can be explained by decreased contribution of a number of dlPCBs (e.g., PCB105, PCB118, and PCB156) to dlPCB-TEQ.[23] Since these congeners are generally more abundant than the most dominating dlPCB in dlPCB-TEQ (i.e., PCB126),[13] the relationships of iPCBs with dlPCB-TEQ have mostly deteriorated for the 2005 TEFs (Table 2). However, it should be noted that the uncertainties associated with estimation of dlPCB-TEQs from the correlations presented in this study are expected to be much lower (within a few fold; Table 2, SI Figure S6) compared to 10−40 fold due to the uncertainties in the TEFs.[29]

The contributions of various iPCBs to ∑PCB can vary by species (SI Figure S2) and potentially by location for each species and fish age. However, on a larger scale, the majority of samples had a relatively narrow band of iPCB contributions (Figure 1). Similarly, although it was highlighted that variation among sites in muscle tissue PCB composition of Belgian yellow eel was likely due to differences in sources of PCB, contribution of iPCB7 to ∑PCB of 30 congeners was relatively constant (average: 54%, 25−75th quartiles: 52−56%, locations = 48, n = 410).[5] Some species-specific differences were also noted in the relationships of iPCBs and dlPCB-TEQs in this study; however, iPCBs and dlPCB-TEQs appear to be strongly related on a large scale (SI Figures S7, S8). Likewise, although minor fish species-specific differences were noted for iPCBs and dlPCB relationships for freshwater fish collected from Rhone River, France-Switzerland, as well as marine fish collected from the French coast and North-Eastern Atlantic Ocean, overall differences were considered minor.[14]

The findings from this study and many other published studies[11,14,27,30−33] emphasize that the PCB pattern is more dependent on fish trophic level at a macro scale as well as the type of tissue being considered than the location of fish collection. In addition, studies conducted on other food items such as milk, eggs, poultry and beef have reported similar observations.[AFSSA, 2007 in14,34] This further highlights that fate, transport and accumulation of PCBs in biota result in a relatively consistent matrix specific pattern and inter-relationships of the PCB congeners (including dioxin-like PCBs). Therefore, correlation studies such as this can be helpful in revealing less resource intensive approaches for quantifying PCB levels and associated risk. It is recommended that periodically a small subset of samples be analyzed for a full suite of PCB congeners, including dlPCBs, to verify the assumptions made in using these schemes, and revise the relationships as needed. It should be noted that a significantly new PCB pattern in fish would change the weightings of individual PCB congeners in total PCB, and thus affect the set of iPCBs needed to estimate total PCBs. Using this approach, an effort must be made to demonstrate that the selection of iPCBs is still appropriate as new locations and especially new species are added to studies.

Regulatory benchmarks have been developed for PCBs in both ∑PCB and dlPCB-TEQ, as well as iPCB forms. The most relevant benchmarks for the data considered in this study would be for the protection of human health from fish consumption. The ∑PCB benchmarks have varied dramatically from one regulatory agency to another but can be generalized as approximately 100 ng/g ww for issuance of partially restrictive fish consumption advisories to ∼1000 ng/g ww for complete restriction.[18,35] Based on these results, partial

Table 2. Relationships (log$Y$ = m log$X$) of Indicator PCBs (iPCBs) and ∑PCB with dlPCB- TEQs in *Skin-Removed Fillets of Sport Fish*[a]

	regression equation			coefficient of determination (R^2)		
↓X Y →	log(dlPCB-TEQ94)	log(dlPCB-TEQ98)	log(dlPCB-TEQ05)	log(dlPCB-TEQ94)	log(dlPCB-TEQ98)	log(dlPCB-TEQ05)
(a) All Data						
log(∑PCB)	0.406 ± 0.025 X	0.405 ± 0.025 X	0.355 ± 0.026 X	0.716	0.716	0.689
log(iPCB17)	0.459 ± 0.024 X	0.457 ± 0.025 X	0.404 ± 0.026 X	0.725	0.725	0.696
log(iPCB13)	0.488 ± 0.024 X	0.486 ± 0.024 X	0.431 ± 0.026 X	0.719	0.719	0.688
log(iPCB9)	0.517 ± 0.025 X	0.515 ± 0.025 X	0.458 ± 0.027 X	0.714	0.714	0.683
log(iPCB5)	0.551 ± 0.024 X	0.55 ± 0.024 X	0.492 ± 0.026 X	0.714	0.714	0.683
log(iPCB4)	0.567 ± 0.024 X	0.566 ± 0.024 X	0.508 ± 0.026 X	0.715	0.715	0.684
log(iPCB3c)	0.59 ± 0.025 X	0.588 ± 0.025 X	0.53 ± 0.026 X	0.711	0.711	0.682
log(iPCB3b)	0.593 ± 0.025 X	0.592 ± 0.025 X	0.533 ± 0.027 X	0.701	0.701	0.672
log(iPCB3a)	0.611 ± 0.025 X	0.61 ± 0.025 X	0.55 ± 0.027 X	0.716	0.716	0.684
log(iPCB7)	0.529 ± 0.025 X	0.528 ± 0.025 X	0.47 ± 0.027 X	0.712	0.712	0.682
log(iPCB6)	0.546 ± 0.025 X	0.544 ± 0.026 X	0.485 ± 0.027 X	0.706	0.706	0.676
log(iPCB3)	0.598 ± 0.025 X	0.597 ± 0.025 X	0.537 ± 0.027 X	0.708	0.708	0.680
(b) After Removing Major Nondetects						
log(∑PCB)	0.429 ± 0.019 X	0.428 ± 0.019 X	0.379 ± 0.02 X	0.694	0.694	0.658
log(iPCB17)	0.467 ± 0.019 X	0.466 ± 0.019 X	0.413 ± 0.021 X	0.733	0.733	0.696
log(iPCB13)	0.488 ± 0.019 X	0.487 ± 0.019 X	0.432 ± 0.021 X	0.739	0.738	0.700
log(iPCB9)	0.516 ± 0.02 X	0.514 ± 0.02 X	0.457 ± 0.022 X	0.733	0.733	0.697
log(iPCB5)	0.548 ± 0.02 X	0.546 ± 0.02 X	0.486 ± 0.022 X	0.765	0.765	0.725
log(iPCB4)	0.564 ± 0.02 X	0.563 ± 0.02 X	0.502 ± 0.022 X	0.774	0.773	0.734
log(iPCB3c)	0.573 ± 0.025 X	0.572 ± 0.025 X	0.511 ± 0.026 X	0.504	0.504	0.492
log(iPCB3b)	0.593 ± 0.021 X	0.592 ± 0.021 X	0.528 ± 0.023 X	0.737	0.737	0.704
log(iPCB3a)	0.615 ± 0.02 X	0.614 ± 0.02 X	0.548 ± 0.023 X	0.775	0.775	0.734
log(iPCB7)	0.527 ± 0.02 X	0.525 ± 0.02 X	0.467 ± 0.023 X	0.739	0.739	0.702
log(iPCB6)	0.543 ± 0.021 X	0.541 ± 0.021 X	0.481 ± 0.023 X	0.722	0.722	0.685
log(iPCB3)	0.596 ± 0.021 X	0.594 ± 0.021 X	0.53 ± 0.024 X	0.741	0.742	0.713

[a]The linear regression equations were prepared for the relationships passing through the origin using logarithmically transformed a) all 470 measurements of iPCB in ng/g wet weight and dlPCB-TEQ in pg/g wet weight, and b) after removing 83 measurements that were mostly below detection for the PCB congeners. The equations present regression slopes ±95% confidence intervals for the slopes. Non-detects (ND) were set at the detection limits (DL) for the PCB congeners for iPCBs and at half of the detection limits for dlPCBs. Similar relationships prepared using normal data are presented in SI Table S7.

Table 3. iPCB Equivalent Values (ng/g ww) for Illustrative ∑PCB (ng/g ww) and dlPCB-TEQ$_{05}$ (pg/g ww) Benchmarks for Consumption of Skin-Removed Fish Fillets[a]

	∑PCB equivalent guidelines		dlPCB-TEQ$_{05}$ equivalent guidelines		
	100 ng/g ww	1000 ng/g ww	4 pg/g ww	6.5 pg/g ww	10 pg/g ww
iPCB3	31	312	14	34	77
iPCB6	41	406	18	49	120
iPCB7	46	456	20	55	139
iPCB3a	29	289	13	30	67
iPCB3b	32	320	14	35	78
iPCB3c	33	329	15	39	91
iPCB4	37	375	16	42	99
iPCB5	41	414	17	47	114
iPCB9	49	493	21	60	155
iPCB13	60	603	25	76	206
iPCB17	71	710	29	93	263

[a]The relationships presented in Tables 1a and 2b were utilized.

restriction on fish consumption (no more than once a week) should be advised when iPCB6/7 exceeds 40−45 ng/g ww (Table 3). Similarly, dlPCB-TEQ benchmarks have ranged from about 4 to 20 pg/g ww. For example, the European maximum tolerance level for dioxin-like PCBs has been set at 6.5 pg/g ww TEQ (including dioxins/furans) and 10 pg/g ww TEQ for eel based on the WHO 2005 TEFs.[22] Corresponding iPCB equivalent benchmarks based on relationships presented in this study have been summarized in Table 3. The iPCB6/7 equiv levels for 6.5 and 10 pg/g dlPCB-TEQ$_{05}$ are about 50−55 and 120−140 ng/g ww, respectively (Table 3). These values are lower than the corresponding iPCB6 limits of 125 and 300 ng/g ww set by the European Union.[22]

It has been recognized that a wide variety of approaches can be used for reliable measurements of PCB concentrations.[9] However, almost every method involves the following general steps: sample storage and handling, sample preparation, extraction and isolation, quantification and quality assurance procedures. Regardless of how many PCB congeners in a sample are being measured, all of the above-mentioned analytical steps, except quantification, are performed almost identically. At present, PCB quantification is almost exclusively performed using capillary GC instruments with either electron capture or mass spectrometry detection, and contributes about 20−40% to the PCB analysis cost. Depending on the number of congeners being measured, the quantification cost may increase by 20−35% for 50+ congeners and by 5−10% for 17 congeners (iPCB17) compared to three congeners (iPCB3). As such, overall PCB analytical cost of a sample may increase by about 10−20% for a full suite of congeners and by 5−10% for

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17 congeners (iPCB17) compared to three congeners (iPCB3). For ∑PCBs, the increased analysis costs associated with increasing the number of congeners are mainly due to the additional time needed to interpret chromatograms and quality control procedures. However, to accurately analyze dlPCBs with the required sensitivity and selectivity, a separate set of standards and analysis using HRMS is required along with additional cleanup steps (e.g., carbon column cleanup which is solvent intensive), which can add dramatically to the cost. Having the capability to translate iPCB metrics to dlPCBs would therefore provide considerable economic benefit.

In summary, we evaluated three existing and eight new iPCB schemes containing 3−17 congeners for relationships with the corresponding observed ∑PCB and dlPCB-TEQs. We also present relationships among the iPCB schemes to facilitate their interconversion. Inclusion of an increased number of congeners in an iPCB scheme enhances performance of the regression models for relationships with ∑PCB; however, all iPCB schemes considered in this study (except iPCB3c) are correlated more or less equally well with dlPCB-TEQs. The analytical cost among iPCBs (up to 17 congeners) and a more detailed analysis of 56 congeners differ marginally by 10−20%. Overall, it appears that iPCB3*a*/4/5/6/7 and a more detailed ∑PCB (based on 50+ congeners) are the best indicator options for PCB levels in fish. Since the European Union has already set the regulatory standards for PCBs using the iPCB6 scheme, it may be advisable to adopt this approach in other jurisdictions to harmonize the standards.

■ ASSOCIATED CONTENT

Supporting Information

List of 56 PCB congeners analyzed, additional 7 tables and 9 figures. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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Supporting Information

Evaluation and inter-conversion of various indicator PCB schemes for sum-PCB and dioxin-like PCB toxic equivalent levels in fish

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List of 56 PCB congeners analyzed using OMOECC method 3411 (OMOE 2010).

PCB018	PCB105	PCB170
PCB019	PCB110	PCB171
PCB022	PCB114	PCB177
PCB028	PCB118	PCB178
PCB033	PCB119	PCB180
PCB037	PCB123	PCB183
PCB044	PCB126	PCB187
PCB049	PCB128	PCB188
PCB052	PCB138	PCB189
PCB054	PCB149	PCB191
PCB070	PCB151	PCB194
PCB074	PCB153	PCB199
PCB077	PCB155	PCB201
PCB081	PCB156	PCB202
PCB087	PCB157	PCB205
PCB095	PCB158	PCB206
PCB099	PCB167	PCB208
PCB101	PCB168	PCB209
PCB104	PCB169	

Table S1a: Types of fish tissue, names of fish species and number of samples considered in exploring relationships among iPCBs and ΣPCB.

Common fish names	Scientific fish names	n
Fillet of sport fish		
Atlantic Salmon	*Salmo salar*	5
Black Crappie	*Pomoxis nigromaculatus*	11
Bluegill	*Lepomis macrochirus*	5
Bowfin	*Amia calva*	8
Brown Bullhead	*Ameiurus nebulosus*	20
Brown Trout	*Salmo trutta*	15
Channel Catfish	*Ictalurus punctatus*	29
Chinook Salmon	*Oncorhynchus tshawytscha*	36
Cisco(Lake Herring)	*Coregonus artedii*	3
Coho Salmon	*Oncorhynchus kisutch*	3
Common Carp	*Cyprinus carpio*	119
Humper (Banker) Lake Trout	*Salvelinus namaycush humper*	20
Lake Sturgeon	*Acipenser fulvescens*	10
Lake Trout	*Salvelinus namaycush*	97
Lake Whitefish	*Coregonus clupeaformis*	40
Largemouth Bass	*Micropterus salmoides*	21
Ling (Burbot)	*Lota lota*	5
Northern Pike	*Esox lucius*	1
Pink Salmon	*Oncorhynchus gorbuscha*	5
Pumpkinseed	*Lepomis gibbosus*	13
Rainbow Trout	*Oncorhynchus mykiss*	48
Rock Bass	*Ambloplites rupestris*	7
Siscowet	*Salvelinus namaycush siscowet*	19
Walleye	*Sander vitreus*	10
White Sucker	*Catostomus commersoni*	22
Whole body forage fish		
Alewife	*Alosa (Pomolobus) pseudoharengus*	11
Rainbow Smelt	*Osmerus mordax*	11
Young-of-the-year Juvenile		
Blacknose Dace	*Rhinichthys atratulus*	5
Bluntnose Minnow	*Pimephales notatus*	46
Common Shiner	*Luxilus cornutus*	32
Creek Chub	*Semotilus atromaculatus*	134
Fathead Minnow	*Pimephales promelas*	52
Golden Shiner	*Notemigonus crysoleucas*	33
Northern Redbelly Dace	*Chrosomus eos*	14
Spottail Shiner	*Notropis hudsonius*	10
Stickleback	*Gasterosteus aculeatus*	41

White Sucker	*Catostomus commersoni*	4
Yellow Perch	*Perca flavescens*	74
Total		1039

Table S1b. Names of fish species and number of samples considered in exploring relationships among iPCBs and dlPCB-TEQ measurements for skin removed boneless fillets.

Common fish names	Scientific fish names	n
Atlantic Salmon	*Salmo salar*	5
Brown Bullhead	*Ameiurus nebulosus*	18
Brown Trout	*Salmo trutta*	19
Channel Catfish	*Ictalurus punctatus*	29
Chinook Salmon	*Oncorhynchus tshawytscha*	36
Cisco(Lake Herring)	*Coregonus artedii*	5
Coho Salmon	*Oncorhynchus kisutch*	10
Common Carp	*Cyprinus carpio*	82
Humper (Banker) Lake Trout	*Salvelinus namaycush humper*	11
Lake Sturgeon	*Acipenser fulvescens*	10
Lake Trout	*Salvelinus namaycush*	82
Lake Whitefish	*Coregonus clupeaformis*	53
Ling (Burbot)	*Lota lota*	7
Northern Pike	*Esox lucius*	7
Pink Salmon	*Oncorhynchus gorbuscha*	5
Rainbow Trout	*Oncorhynchus mykiss*	39
Rock Bass	*Ambloplites rupestris*	8
Siscowet	*Salvelinus namaycush siscowet*	12
Walleye	*Sander vitreus*	19
White Sucker	*Catostomus commersoni*	13
Total		470

Table S2. Toxic Equivalency Factors (TEFs) used to calculate dlPCB-TEQ. The values were taken from for the 1994, 1998 and 2005 TEFs (Bhavsar and others 2008).

	1994	1998	2005
PCB 77	0.0005	0.0001	0.0001
PCB 81	0	0.0001	0.0003
PCB 105	0.0001	0.0001	0.00003
PCB 114	0.0005	0.0005	0.00003
PCB 118	0.0001	0.0001	0.00003
PCB 123	0.0001	0.0001	0.00003
PCB 126	0.1	0.1	0.1
PCB 156	0.0005	0.0005	0.00003
PCB 157	0.0005	0.0005	0.00003
PCB 167	0.00001	0.00001	0.00003
PCB 169	0.01	0.01	0.03
PCB 189	0.0001	0.0001	0.00003

Table S3: P-values for Shapiro-Wilk normality tests for normal as well as logarithmically transformed values (a) for all PCB congener related data, (b) for a subset with Lyons Creek related PCB conger data removed, and (c) dlPCB and PCB congener related measurements (all as well as major non-detects removed).

(a)

	SBF		YFC		WFC	
	normal	log-transf	normal	log-transf	normal	log-transf
iPCB3	<0.001	<0.001	<0.001	<0.001	<0.01	0.65
iPCB6	<0.001	0.14	<0.001	<0.001	0.02	0.57
iPCB7	<0.001	0.25	<0.001	<0.001	0.02	0.46
iPCB3a	<0.001	0.56	<0.001	<0.001	0.01	0.5
iPCB3b	<0.001	<0.001	<0.001	<0.001	<0.01	0.17
iPCB3c	<0.001	<0.01	<0.001	<0.001	<0.01	0.42
iPCB4	<0.001	0.18	<0.001	<0.001	<0.01	0.35
iPCB5	<0.001	0.19	<0.001	<0.001	<0.01	0.23
iPCB9	<0.001	0.59	<0.001	<0.001	0.01	0.43
iPCB13	<0.001	0.84	<0.001	<0.001	0.01	0.37
iPCB17	<0.001	0.36	<0.001	<0.001	0.01	0.24
ΣPCB	<0.001	<0.01	<0.001	<0.001	<0.01	0.29

(b)

	SBF		YFC		WFC	
	normal	log-transf	normal	log-transf	normal	log-transf
iPCB3	<0.001	0.02	<0.001	<0.001	<0.01	0.65
iPCB6	<0.001	0.18	<0.001	<0.001	0.02	0.57
iPCB7	<0.001	0.21	<0.001	<0.001	0.02	0.46
iPCB3a	<0.001	0.21	<0.001	<0.001	0.01	0.5
iPCB3b	<0.001	<0.01	<0.001	<0.001	<0.01	0.17
iPCB3c	<0.001	0.1	<0.001	<0.001	<0.01	0.42
iPCB4	<0.001	0.1	<0.001	<0.001	<0.01	0.35
iPCB5	<0.001	0.18	<0.001	<0.001	<0.01	0.23
iPCB9	<0.001	0.49	<0.001	<0.001	0.01	0.43
iPCB13	<0.001	0.44	<0.001	<0.001	0.01	0.37
iPCB17	<0.001	0.07	<0.001	<0.001	0.01	0.24
ΣPCB	<0.001	<0.001	<0.001	<0.001	<0.01	0.29

(c)

	All data		Major ND removed	
	normal	log-transf	normal	log-transf
iPCB3	<0.001	<0.001	<0.001	0.2
iPCB6	<0.001	<0.001	<0.001	0.36
iPCB7	<0.001	<0.001	<0.001	0.48
iPCB3a	<0.001	<0.001	<0.001	0.34
iPCB3b	<0.001	<0.001	<0.001	0.15
iPCB3c	<0.001	<0.001	<0.001	0.46
iPCB4	<0.001	<0.001	<0.001	0.16
iPCB5	<0.001	<0.001	<0.001	0.15
iPCB9	<0.001	<0.001	<0.001	0.42
iPCB13	<0.001	<0.001	<0.001	0.51
iPCB17	<0.001	<0.001	<0.001	0.57
ΣPCB	<0.001	<0.001	<0.001	0.1
dlPCB-TEQ94	<0.001	<0.001	<0.001	0.03
dlPCB-TEQ98	<0.001	<0.001	<0.001	0.04
dlPCB-TEQ05	<0.001	<0.001	<0.001	<0.01

Table S4: Congener specific loadings of the first four components of the PLS analysis for relationships of PCB congeners with ΣPCB.

	Comp 1	Comp 2	Comp 3	Comp 4
PCB018	0.102	0.118		
PCB019				
PCB022				
PCB028	0.138	0.136		
PCB033	0.102			
PCB037	0.121	-0.15	0.156	
PCB044	0.133	-0.168	0.196	0.116
PCB049	0.166	-0.221	0.176	
PCB052	0.181	-0.244	0.238	
PCB054				
PCB070	0.122			
PCB074	0.175	-0.2		
PCB077	0.134			
PCB081				
PCB087				
PCB095	0.46	-0.473	0.161	
PCB099	0.238			
PCB101	0.31	-0.138	-0.126	
PCB104				
PCB105	0.148	-0.111		
PCB110	0.247	0.164		
PCB114				
PCB118	0.324	-0.263		
PCB119				
PCB123	0.224			
PCB126				
PCB128				
PCB138	0.306	0.434	-0.258	-0.178
PCB149	0.132	0.105	-0.203	0.215
PCB151				
PCB153	0.345	0.531	-0.174	-0.119
PCB155				
PCB156	0.106			
PCB157	0.107			
PCB158	0.17	-0.147		
PCB167	0.145	0.716	-0.55	

	Comp 1	Comp 2	Comp 3	Comp 4
PCB168	0.123	0.135		
PCB169				
PCB170	0.101			
PCB171				
PCB177				
PCB178	0.138	0.926		
PCB180	0.143	0.266	-0.112	0.101
PCB183				
PCB187	0.157	0.14		
PCB188	0.212			
PCB189				
PCB191				
PCB194				
PCB199				
PCB201	0.238	0.261		
PCB202				
PCB205				
PCB206				
PCB208				

Table S5a. Relationships (logY = *m* logX) of indicator PCBs (iPCBs) and ΣPCB for variety of fish tissues. The linear regression equations were prepared for the relationships passing through the origin using logarithmically transformed 1038 concentrations in ng/g wet weight. The equations present regression slopes ± 95% confidence intervals for the slopes. Non-detects (ND) were set at the detection limits (DL). Similar relationships prepared using normal data are presented in Table S5a. Separate equations were constructed by considering ND=0 and are presented in Table S6.

	Regression Equation / Coefficient of determination (R^2)											
↓Y X →	Log(iPCB3)	Log(iPCB6)	Log(iPCB7)	Log(iPCB3a)	Log(iPCB3b)	Log(iPCB3c)	Log(iPCB4)	Log(iPCB5)	Log(iPCB9)	Log(iPCB13)	Log(iPCB17)	Log(ΣPCB)
a) Skin-removed Fillet of sport fish												
Log(ΣPCB)	1.4±0 X	1.25±0.01 X	1.21±0.01 X	1.36±0.02 X	1.38±0.02 X	1.36±0.02 X	1.28±0.01 X	1.24±0.01 X	1.19±0.01 X	1.12±0.01 X	1.08±0 X	
Log(iPCB17)	1.3±0.02 X	1.16±0.01 X	1.13±0 X	1.27±0.01 X	1.28±0.02 X	1.27±0.01 X	1.19±0.01 X	1.15±0.01 X	1.1±0 X	1.04±0 X		0.983
Log(iPCB13)	1.25±0.02 X	1.12±0 X	1.08±0 X	1.22±0.01 X	1.23±0.01 X	1.22±0.01 X	1.14±0.01 X	1.11±0.01 X	1.06±0 X		0.994	0.973
Log(iPCB9)	1.18±0.01 X	1.05±0 X	1.02±0 X	1.15±0.01 X	1.16±0.01 X	1.15±0.01 X	1.08±0.01 X	1.05±0.01 X		0.992	0.984	0.965
Log(iPCB5)	1.13±0.01 X	1±0 X	0.97±0 X	1.1±0.01 X	1.11±0.01 X	1.1±0.01 X	1.03±0 X		0.973	0.977	0.968	0.939
Log(iPCB4)	1.1±0.01 X	0.98±0 X	0.95±0 X	1.07±0 X	1.08±0.01 X	1.07±0.01 X		0.995	0.967	0.970	0.956	0.923
Log(iPCB3c)	1.03±0.01 X	0.9±0.01 X	0.88±0.01 X	0.99±0.01 X	1.01±0 X		0.933	0.927	0.916	0.882	0.864	0.834
Log(iPCB3b)	1.01±0.01 X	0.89±0.01 X	0.86±0.01 X	0.97±0.01 X		0.975	0.905	0.913	0.894	0.862	0.850	0.829
Log(iPCB3a)	1.02±0.01 X	0.91±0.01 X	0.88±0.01 X		0.848	0.878	0.985	0.983	0.952	0.969	0.958	0.922
Log(iPCB7)	1.16±0.01 X	1.03±0 X		0.961	0.912	0.928	0.979	0.983	0.993	0.987	0.978	0.956
Log(iPCB6)	1.12±0.01 X		0.996	0.948	0.915	0.920	0.969	0.973	0.985	0.979	0.969	0.950
Log(iPCB3)		0.863	0.856	0.779	0.955	0.963	0.847	0.839	0.845	0.801	0.780	0.757
b) Whole body YOY												
Log(ΣPCB)	1.57±0 X	1.22±0.01 X	1.19±0.01 X	1.25±0.02 X	1.42±0.02 X	1.43±0.02 X	1.23±0.02 X	1.2±0.01 X	1.18±0.01 X	1.1±0.01 X	1.06±0 X	
Log(iPCB17)	1.48±0.02 X	1.15±0.01 X	1.12±0 X	1.18±0.02 X	1.34±0.02 X	1.35±0.02 X	1.16±0.01 X	1.13±0.01 X	1.11±0 X	1.04±0 X		0.996
Log(iPCB13)	1.43±0.02 X	1.11±0 X	1.08±0 X	1.14±0.01 X	1.3±0.02 X	1.3±0.01 X	1.12±0.01 X	1.09±0.01 X	1.07±0 X		0.997	0.992
Log(iPCB9)	1.34±0.02 X	1.04±0 X	1.01±0 X	1.07±0.01 X	1.21±0.01 X	1.22±0.01 X	1.05±0.01 X	1.02±0.01 X		0.998	0.996	0.991
Log(iPCB5)	1.31±0.02 X	1.01±0.01 X	0.99±0.01 X	1.05±0.01 X	1.19±0.01 X	1.19±0.01 X	1.03±0 X		0.981	0.987	0.979	0.970
Log(iPCB4)	1.28±0.02 X	0.99±0.01 X	0.96±0.01 X	1.02±0.01 X	1.16±0.01 X	1.16±0.01 X		0.999	0.981	0.987	0.978	0.970
Log(iPCB3c)	1.1±0.01 X	0.84±0.01 X	0.82±0.01 X	0.87±0.01 X	1±0 X		0.976	0.974	0.957	0.955	0.944	0.929
Log(iPCB3b)	1.1±0.01 X	0.84±0.01 X	0.82±0.01 X	0.87±0.01 X		0.987	0.964	0.967	0.948	0.945	0.937	0.924
Log(iPCB3a)	1.24±0.02 X	0.96±0.01 X	0.93±0.01 X		0.947	0.960	0.996	0.996	0.972	0.981	0.972	0.964
Log(iPCB7)	1.33±0.02 X	1.03±0 X		0.970	0.947	0.957	0.980	0.979	1.000	0.997	0.995	0.990
Log(iPCB6)	1.29±0.02 X		0.999	0.963	0.946	0.954	0.974	0.973	0.998	0.994	0.992	0.987
Log(iPCB3)		0.897	0.894	0.881	0.964	0.971	0.908	0.904	0.892	0.883	0.871	0.854
c) Whole body forage fish												
Log(ΣPCB)	1.24±0 X	1.17±0.02 X	1.14±0.02 X	1.28±0.02 X	1.28±0.02 X	1.23±0.02 X	1.19±0.02 X	1.18±0.02 X	1.13±0.02 X	1.07±0.01 X	1.04±0.01 X	
Log(iPCB17)	1.2±0.01 X	1.13±0.01 X	1.09±0.01 X	1.23±0.01 X	1.23±0.02 X	1.18±0.01 X	1.14±0.01 X	1.14±0.01 X	1.09±0.01 X	1.03±0 X		0.931
Log(iPCB13)	1.16±0.01 X	1.09±0.01 X	1.06±0.01 X	1.19±0.01 X	1.2±0.02 X	1.15±0.01 X	1.11±0.01 X	1.1±0.01 X	1.06±0.01 X		0.998	0.915
Log(iPCB9)	1.1±0.01 X	1.03±0.01 X	1±0 X	1.13±0.01 X	1.13±0.01 X	1.09±0.01 X	1.05±0.01 X	1.04±0.01 X		0.997	0.996	0.915
Log(iPCB5)	1.05±0.01 X	0.99±0.01 X	0.96±0.01 X	1.08±0.01 X	1.08±0.01 X	1.04±0.01 X	1.01±0 X		0.971	0.983	0.986	0.938
Log(iPCB4)	1.05±0.01 X	0.98±0.01 X	0.96±0.01 X	1.07±0 X	1.08±0.01 X	1.04±0 X		0.994	0.977	0.989	0.987	0.918
Log(iPCB3c)	1.01±0.01 X	0.95±0.01 X	0.92±0.01 X	1.04±0 X	1.04±0.01 X		0.993	0.984	0.985	0.992	0.987	0.906
Log(iPCB3b)	0.97±0.01 X	0.91±0.01 X	0.89±0.01 X	1±0.01 X		0.982	0.973	0.980	0.986	0.985	0.988	0.930
Log(iPCB3a)	0.97±0.01 X	0.92±0.01 X	0.89±0.01 X		0.975	0.996	0.996	0.986	0.983	0.992	0.988	0.910
Log(iPCB7)	1.09±0.01 X	1.03±0.01 X		0.984	0.983	0.986	0.977	0.968	0.999	0.996	0.995	0.913
Log(iPCB6)	1.06±0 X		0.997	0.968	0.978	0.972	0.957	0.949	0.996	0.987	0.986	0.900
Log(iPCB3)		0.994	0.994	0.976	0.984	0.985	0.965	0.955	0.993	0.987	0.983	0.890

Table S5b. Relationships (Y=*m*X) of indicator PCBs (iPCBs) and ΣPCB for a variety of fish tissues. The linear regression equations were prepared for the relationships passing through the origin using total 1038 concentrations in ng/g wet weight. The equations present regression slopes ± 95% confidence intervals for the slopes. **Non-detects (ND) were set at the detection limits (DL).** Similar relationships prepared using more appropriate logarithmically transformed data are presented in Table 1. Separate equations were constructed by removing distinct Lyons Creek data (Table S5b) and using ND=0 (Table S6).

	Regression Equation / Coefficient of determination (R^2)											
↓Y X→	iPCB3	iPCB6	iPCB7	iPCB3a	iPCB3b	iPCB3c	iPCB4	iPCB5	iPCB9	iPCB13	iPCB17	ΣPCB
a) Skin-removed Fillet of sport fish												
ΣPCB	3.89±0.21 X	3.24±0.07 X	2.71±0.04 X	4.21±0.05 X	4.28±0.15 X	3.99±0.14 X	3.22±0.05 X	2.79±0.03 X	2.49±0.03 X	1.75±0.02 X	1.45±0.01 X	
iPCB17	2.65±0.14 X	2.22±0.05 X	1.86±0.03 X	2.9±0.02 X	2.93±0.1 X	2.73±0.1 X	2.22±0.03 X	1.92±0.02 X	1.71±0.02 X	1.21±0.01 X		0.986
iPCB13	2.25±0.11 X	1.86±0.03 X	1.55±0.02 X	2.4±0.02 X	2.46±0.08 X	2.3±0.07 X	1.84±0.02 X	1.59±0.01 X	1.43±0.01 X		0.996	0.985
iPCB9	1.66±0.06 X	1.31±0.01 X	1.09±0 X	1.65±0.03 X	1.78±0.04 X	1.66±0.04 X	1.29±0.01 X	1.11±0.01 X		0.986	0.971	0.965
iPCB5	1.44±0.07 X	1.17±0.02 X	0.97±0.01 X	1.5±0.02 X	1.56±0.04 X	1.46±0.04 X	1.16±0.01 X		0.989	0.994	0.988	0.976
iPCB4	1.27±0.05 X	1.01±0.01 X	0.84±0.01 X	1.28±0.02 X	1.36±0.03 X	1.28±0.03 X		0.995	0.994	0.988	0.974	0.965
iPCB3c	1.05±0.02 X	0.76±0.01 X	0.62±0.01 X	0.89±0.03 X	1.07±0.01 X		0.908	0.878	0.920	0.847	0.810	0.812
iPCB3b	0.98±0.02 X	0.7±0.01 X	0.58±0.01 X	0.83±0.03 X		0.993	0.895	0.870	0.910	0.837	0.803	0.804
iPCB3a	0.89±0.05 X	0.75±0.02 X	0.63±0.01 X		0.773	0.788	0.966	0.979	0.956	0.988	0.991	0.975
iPCB7	1.54±0.05 X	1.21±0.01 X		0.940	0.930	0.937	0.990	0.982	0.997	0.974	0.957	0.952
iPCB6	1.3±0.04 X		0.993	0.901	0.953	0.954	0.972	0.957	0.984	0.947	0.924	0.923
iPCB3		0.855	0.808	0.597	0.952	0.946	0.749	0.707	0.775	0.673	0.627	0.634
b) Whole body YOY												
ΣPCB	42.42±1.01 X	4.31±0.07 X	3.76±0.05 X	4.88±0.06 X	15.97±0.25 X	17.59±0.34 X	4.64±0.06 X	3.88±0.04 X	3.49±0.04 X	2.12±0.02 X	1.63±0.01 X	
iPCB17	26.06±0.6 X	2.66±0.04 X	2.32±0.02 X	2.99±0.04 X	9.81±0.14 X	10.79±0.21 X	2.85±0.04 X	2.38±0.03 X	2.15±0.02 X	1.31±0 X		0.994
iPCB13	19.96±0.46 X	2.04±0.03 X	1.77±0.02 X	2.29±0.03 X	7.5±0.11 X	8.27±0.16 X	2.18±0.03 X	1.82±0.02 X	1.65±0.01 X		0.999	0.992
iPCB9	11.97±0.31 X	1.24±0.01 X	1.08±0 X	1.37±0.03 X	4.53±0.07 X	4.94±0.12 X	1.3±0.03 X	1.09±0.02 X		0.991	0.992	0.980
iPCB5	10.95±0.22 X	1.09±0.03 X	0.95±0.02 X	1.26±0.01 X	4.09±0.06 X	4.56±0.06 X	1.2±0 X		0.958	0.986	0.985	0.983
iPCB4	9.11±0.19 X	0.9±0.02 X	0.79±0.02 X	1.05±0 X	3.39±0.06 X	3.8±0.05 X		0.998	0.947	0.981	0.978	0.977
iPCB3c	2.41±0.04 X	0.23±0.01 X	0.2±0.01 X	0.27±0 X	0.89±0.01 X		0.977	0.980	0.925	0.955	0.952	0.952
iPCB3b	2.65±0.05 X	0.26±0.01 X	0.23±0 X	0.3±0.01 X		0.963	0.956	0.972	0.965	0.972	0.974	0.969
iPCB3a	8.64±0.19 X	0.86±0.02 X	0.75±0.02 X		0.954	0.975	1.000	0.998	0.947	0.981	0.978	0.978
iPCB7	11.04±0.3 X	1.15±0.01 X		0.938	0.958	0.913	0.938	0.949	0.999	0.987	0.988	0.975
iPCB6	9.45±0.3 X		0.997	0.914	0.940	0.881	0.914	0.926	0.994	0.974	0.976	0.961
iPCB3		0.888	0.912	0.944	0.956	0.972	0.948	0.952	0.920	0.940	0.938	0.934
c) Whole body forage fish												
ΣPCB	3.28±0.17 X	2.38±0.12 X	2.07±0.09 X	3.93±0.2 X	3.8±0.15 X	3.24±0.15 X	2.74±0.12 X	2.62±0.09 X	2.02±0.08 X	1.5±0.06 X	1.27±0.04 X	
iPCB17	2.59±0.08 X	1.88±0.05 X	1.64±0.03 X	3.1±0.11 X	3±0.08 X	2.56±0.08 X	2.16±0.06 X	2.06±0.05 X	1.59±0.02 X	1.18±0.01 X		0.981
iPCB13	2.19±0.06 X	1.59±0.04 X	1.38±0.02 X	2.62±0.07 X	2.53±0.09 X	2.16±0.05 X	1.83±0.04 X	1.74±0.05 X	1.34±0.02 X		0.998	0.973
iPCB9	1.63±0.03 X	1.18±0.02 X	1.03±0.01 X	1.95±0.07 X	1.88±0.06 X	1.61±0.05 X	1.36±0.05 X	1.29±0.05 X		0.998	0.997	0.973
iPCB5	1.25±0.06 X	0.91±0.05 X	0.79±0.03 X	1.5±0.05 X	1.45±0.04 X	1.24±0.04 X	1.05±0.02 X		0.979	0.986	0.989	0.978
iPCB4	1.19±0.05 X	0.86±0.04 X	0.75±0.02 X	1.44±0.02 X	1.38±0.06 X	1.18±0.02 X		0.991	0.985	0.992	0.987	0.965
iPCB3c	1.01±0.03 X	0.73±0.03 X	0.64±0.02 X	1.21±0.02 X	1.17±0.05 X		0.997	0.984	0.988	0.991	0.985	0.961
iPCB3b	0.86±0.03 X	0.62±0.03 X	0.54±0.02 X	1.03±0.05 X		0.980	0.979	0.992	0.984	0.985	0.990	0.979
iPCB3a	0.83±0.03 X	0.6±0.03 X	0.53±0.02 X		0.969	0.997	0.995	0.977	0.986	0.990	0.981	0.954
iPCB7	1.58±0.03 X	1.15±0.01 X		0.989	0.977	0.989	0.985	0.973	0.999	0.997	0.993	0.967
iPCB6	1.38±0.03 X		0.998	0.981	0.972	0.981	0.975	0.963	0.997	0.992	0.989	0.962
iPCB3		0.995	0.996	0.988	0.979	0.992	0.984	0.970	0.994	0.991	0.986	0.959

Table S5c. Relationships (Y=*m*X) of indicator PCBs (iPCBs) and ΣPCB for a variety of fish tissues. The linear regression equations were prepared for the relationships passing through the origin using 861 concentrations (in ng/g wet weight) after removing distinct Lyons Creek related measurements.. The equations present regression slopes ± 95% confidence intervals for the slopes. **Non-detects (ND) were set at the detection limits (DL).** Similar relationships prepared using more appropriate logarithmically transformed data are presented in Table 2.

	Regression Equation / Coefficient of determination (R^2)											
↓Y X→	iPCB3	iPCB6	iPCB7	iPCB3a	iPCB3b	iPCB3c	iPCB4	iPCB5	iPCB9	iPCB13	iPCB17	ΣPCB
a) Skin-removed Fillet of sport fish												
ΣPCB	3.82±0.21 X	3.2±0.07 X	2.68±0.05 X	4.2±0.06 X	4.19±0.15 X	3.91±0.14 X	3.18±0.05 X	2.76±0.03 X	2.46±0.04 X	1.74±0.02 X	1.45±0.01 X	
iPCB17	2.6±0.15 X	2.2±0.05 X	1.84±0.03 X	2.91±0.02 X	2.87±0.1 X	2.68±0.1 X	2.19±0.03 X	1.9±0.02 X	1.69±0.02 X	1.2±0.01 X		0.985
iPCB13	2.22±0.12 X	1.84±0.04 X	1.54±0.02 X	2.41±0.02 X	2.42±0.08 X	2.26±0.07 X	1.83±0.02 X	1.58±0.01 X	1.41±0.01 X		0.996	0.985
iPCB9	1.64±0.07 X	1.32±0.01 X	1.09±0 X	1.67±0.03 X	1.75±0.04 X	1.63±0.03 X	1.29±0.01 X	1.11±0.01 X		0.987	0.973	0.968
iPCB5	1.42±0.07 X	1.17±0.02 X	0.97±0.01 X	1.51±0.02 X	1.54±0.04 X	1.44±0.04 X	1.16±0.01 X		0.991	0.996	0.990	0.977
iPCB4	1.25±0.05 X	1.01±0.01 X	0.84±0.01 X	1.3±0.02 X	1.34±0.03 X	1.26±0.03 X		0.996	0.995	0.988	0.975	0.966
iPCB3c	1.04±0.02 X	0.78±0.01 X	0.64±0.01 X	0.94±0.04 X	1.07±0.01 X		0.925	0.895	0.935	0.871	0.836	0.839
iPCB3b	0.97±0.02 X	0.73±0.01 X	0.59±0.01 X	0.86±0.04 X		0.994	0.913	0.885	0.926	0.860	0.828	0.831
iPCB3a	0.87±0.05 X	0.75±0.02 X	0.63±0.01 X		0.800	0.816	0.967	0.981	0.959	0.988	0.991	0.975
iPCB7	1.52±0.06 X	1.21±0.01 X		0.942	0.948	0.954	0.991	0.982	0.997	0.975	0.958	0.954
iPCB6	1.29±0.04 X		0.993	0.900	0.974	0.975	0.972	0.957	0.984	0.946	0.923	0.922
iPCB3		0.881	0.830	0.626	0.953	0.946	0.771	0.726	0.794	0.699	0.655	0.664
b) Whole body YOY												
ΣPCB	42.64±1.07 X	4.32±0.08 X	3.76±0.06 X	4.88±0.07 X	15.99±0.26 X	17.62±0.37 X	4.64±0.07 X	3.88±0.05 X	3.49±0.05 X	2.12±0.02 X	1.63±0.01 X	
iPCB17	26.2±0.64 X	2.67±0.04 X	2.32±0.02 X	2.99±0.04 X	9.83±0.15 X	10.81±0.23 X	2.85±0.04 X	2.38±0.03 X	2.15±0.02 X	1.31±0 X		0.994
iPCB13	20.06±0.48 X	2.04±0.03 X	1.78±0.02 X	2.29±0.03 X	7.52±0.12 X	8.28±0.17 X	2.18±0.03 X	1.82±0.02 X	1.65±0.01 X		0.999	0.992
iPCB9	12.03±0.34 X	1.24±0.01 X	1.08±0 X	1.37±0.03 X	4.53±0.08 X	4.94±0.13 X	1.3±0.03 X	1.09±0.02 X		0.991	0.992	0.980
iPCB5	11.01±0.23 X	1.09±0.03 X	0.95±0.02 X	1.26±0.01 X	4.1±0.06 X	4.57±0.06 X	1.2±0 X		0.958	0.986	0.984	0.983
iPCB4	9.16±0.2 X	0.9±0.03 X	0.79±0.02 X	1.05±0 X	3.4±0.07 X	3.8±0.05 X		0.998	0.947	0.981	0.978	0.977
iPCB3c	2.41±0.04 X	0.23±0.01 X	0.2±0.01 X	0.27±0 X	0.89±0.02 X		0.978	0.980	0.923	0.954	0.952	0.951
iPCB3b	2.66±0.05 X	0.26±0.01 X	0.23±0 X	0.3±0.01 X		0.963	0.957	0.972	0.965	0.972	0.974	0.969
iPCB3a	8.7±0.2 X	0.86±0.02 X	0.75±0.02 X		0.955	0.976	1.000	0.998	0.947	0.981	0.978	0.978
iPCB7	11.09±0.33 X	1.15±0.01 X		0.938	0.957	0.911	0.938	0.949	0.999	0.987	0.988	0.975
iPCB6	9.49±0.32 X		0.997	0.913	0.939	0.879	0.913	0.925	0.994	0.974	0.976	0.961
iPCB3		0.887	0.911	0.947	0.956	0.973	0.950	0.954	0.920	0.941	0.939	0.935
c) Whole body forage fish												
ΣPCB	3.28±0.17 X	2.38±0.12 X	2.07±0.09 X	3.93±0.2 X	3.8±0.15 X	3.24±0.15 X	2.74±0.12 X	2.62±0.09 X	2.02±0.08 X	1.5±0.06 X	1.27±0.04 X	
iPCB17	2.59±0.08 X	1.88±0.05 X	1.64±0.03 X	3.1±0.11 X	3±0.08 X	2.56±0.08 X	2.16±0.06 X	2.06±0.05 X	1.59±0.02 X	1.18±0.01 X		0.981
iPCB13	2.19±0.06 X	1.59±0.04 X	1.38±0.02 X	2.62±0.07 X	2.53±0.09 X	2.16±0.05 X	1.83±0.04 X	1.74±0.05 X	1.34±0.02 X		0.998	0.973
iPCB9	1.63±0.03 X	1.18±0.02 X	1.03±0.01 X	1.95±0.07 X	1.88±0.06 X	1.61±0.05 X	1.36±0.05 X	1.29±0.05 X		0.998	0.997	0.973
iPCB5	1.25±0.06 X	0.91±0.05 X	0.79±0.03 X	1.5±0.05 X	1.45±0.04 X	1.24±0.04 X	1.05±0.02 X		0.979	0.986	0.989	0.978
iPCB4	1.19±0.05 X	0.86±0.04 X	0.75±0.02 X	1.44±0.02 X	1.38±0.06 X	1.18±0.02 X		0.991	0.985	0.992	0.987	0.965
iPCB3c	1.01±0.03 X	0.73±0.03 X	0.64±0.02 X	1.21±0.02 X	1.17±0.05 X		0.997	0.984	0.988	0.991	0.985	0.961
iPCB3b	0.86±0.03 X	0.62±0.03 X	0.54±0.02 X	1.03±0.05 X		0.980	0.979	0.992	0.984	0.985	0.990	0.979
iPCB3a	0.83±0.03 X	0.6±0.03 X	0.53±0.02 X		0.969	0.997	0.995	0.977	0.986	0.990	0.981	0.954
iPCB7	1.58±0.03 X	1.15±0.01 X		0.989	0.977	0.989	0.985	0.973	0.999	0.997	0.993	0.967
iPCB6	1.38±0.03 X		0.998	0.981	0.972	0.981	0.975	0.963	0.997	0.992	0.989	0.962
iPCB3		0.995	0.996	0.988	0.979	0.992	0.984	0.970	0.994	0.991	0.986	0.959

Table S6a. Relationships (logY = *m* logX) of indicator PCBs (iPCBs) and ΣPCB for variety of fish tissues. The linear regression equations were prepared for the relationships passing through the origin using **logarithmically transformed** 1038 concentrations in ng/g wet weight. The equations present regression slopes ± 95% confidence intervals for the slopes. **Non-detects (ND) were set at the zero.** Similar relationships prepared using normal data are presented in Table S6b. Separate equations were constructed by ND=detection limit and are presented in Table 1.

	Regression Equation / Coefficient of determination (R2)											
↓Y X→	Log(iPCB3)	Log(iPCB6)	Log(iPCB7)	Log(iPCB3a)	Log(iPCB3b)	Log(iPCB3c)	Log(iPCB4)	Log(iPCB5)	Log(iPCB9)	Log(iPCB13)	Log(iPCB17)	Log(ΣPCB)
a) Skin-removed Fillet of sport fish												
Log(ΣPCB)	NA	NA	1.2±0.01 X	NA	NA	NA	NA	NA	1.18±0.01 X	1.11±0 X	1.07±0 X	
Log(iPCB17)	NA	NA	1.12±0 X	NA	NA	NA	NA	NA	1.1±0 X	1.04±0 X		0.984
Log(iPCB13)	NA	NA	1.08±0 X	NA	NA	NA	NA	NA	1.06±0 X		0.992	0.973
Log(iPCB9)	NA	NA	1.02±0 X	NA	NA	NA	NA	NA		0.992	0.983	0.964
Log(iPCB5)	NA	NA	NA	NA	NA	NA	NA		NA	NA	NA	NA
Log(iPCB4)	NA	NA	NA	NA	NA	NA		NA	NA	NA	NA	NA
Log(iPCB3c)	NA	NA	NA	NA	NA		NA	NA	NA	NA	NA	NA
Log(iPCB3b)	NA	NA	NA	NA		NA	NA	NA	NA	NA	NA	NA
Log(iPCB3a)	NA	NA	NA		NA	NA	NA	NA	NA	NA	NA	NA
Log(iPCB7)	NA	NA		NA	NA	NA	NA	NA	0.991	0.984	0.975	0.956
Log(iPCB6)	NA		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Log(iPCB3)		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
b) Whole body YOY												
Log(ΣPCB)	NA	1.22±0.01 X	1.19±0.01 X	NA	NA	NA	NA	NA	1.17±0.01 X	1.1±0 X	1.06±0 X	
Log(iPCB17)	NA	1.15±0.01 X	1.12±0 X	NA	NA	NA	NA	NA	1.11±0 X	1.04±0 X		0.997
Log(iPCB13)	NA	1.11±0 X	1.08±0 X	NA	NA	NA	NA	NA	1.07±0 X		0.996	0.994
Log(iPCB9)	NA	1.04±0 X	1.01±0 X	NA	NA	NA	NA	NA		0.998	0.995	0.992
Log(iPCB5)	NA	NA	NA	NA	NA	NA	NA		NA	NA	NA	NA
Log(iPCB4)	NA	NA	NA	NA	NA	NA		NA	NA	NA	NA	NA
Log(iPCB3c)	NA	NA	NA	NA	NA		NA	NA	NA	NA	NA	NA
Log(iPCB3b)	NA	NA	NA	NA		NA	NA	NA	NA	NA	NA	NA
Log(iPCB3a)	NA	NA	NA		NA	NA	NA	NA	NA	NA	NA	NA
Log(iPCB7)	NA	1.03±0 X		NA	NA	NA	NA	NA	1.000	0.997	0.994	0.991
Log(iPCB6)	NA		0.999	NA	NA	NA	NA	NA	0.998	0.993	0.991	0.988
Log(iPCB3)		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
c) Whole body forage fish												
Log(ΣPCB)	1.24±0 X	1.17±0.02 X	1.13±0.02 X	1.27±0.02 X	1.28±0.02 X	1.23±0.02 X	1.19±0.02 X	1.18±0.02 X	1.13±0.02 X	1.07±0.01 X	1.04±0.01 X	
Log(iPCB17)	1.2±0.01 X	1.13±0.01 X	1.09±0.01 X	1.23±0.01 X	1.23±0.02 X	1.18±0.01 X	1.14±0.01 X	1.14±0.01 X	1.09±0.01 X	1.03±0 X		0.931
Log(iPCB13)	1.16±0.01 X	1.09±0.01 X	1.06±0.01 X	1.19±0.01 X	1.2±0.02 X	1.15±0.01 X	1.11±0.01 X	1.1±0.01 X	1.06±0.01 X		0.998	0.915
Log(iPCB9)	1.1±0.01 X	1.03±0.01 X	1±0 X	1.13±0.01 X	1.13±0.01 X	1.09±0.01 X	1.05±0.01 X	1.04±0.01 X		0.997	0.996	0.914
Log(iPCB5)	1.05±0.01 X	0.99±0.01 X	0.96±0.01 X	1.08±0.01 X	1.08±0.01 X	1.04±0.01 X	1.01±0 X		0.971	0.983	0.986	0.938
Log(iPCB4)	1.05±0.01 X	0.98±0.01 X	0.96±0.01 X	1.07±0 X	1.08±0.01 X	1.04±0 X		0.994	0.977	0.989	0.987	0.917
Log(iPCB3c)	1.01±0.01 X	0.95±0.01 X	0.92±0.01 X	1.04±0 X	1.04±0.01 X		0.993	0.983	0.985	0.992	0.987	0.905
Log(iPCB3b)	0.97±0.01 X	0.91±0.01 X	0.89±0.01 X	1±0.01 X		0.982	0.973	0.980	0.986	0.984	0.988	0.929
Log(iPCB3a)	0.97±0.01 X	0.92±0.01 X	0.89±0.01 X		0.975	0.996	0.996	0.986	0.983	0.992	0.988	0.909
Log(iPCB7)	1.09±0.01 X	1.03±0.01 X		0.984	0.983	0.986	0.977	0.968	0.999	0.996	0.995	0.912
Log(iPCB6)	1.06±0 X		0.997	0.968	0.978	0.972	0.957	0.949	0.996	0.987	0.986	0.899
Log(iPCB3)		0.994	0.994	0.976	0.984	0.985	0.965	0.955	0.993	0.987	0.983	0.889

Table S6b. Relationships (Y=*m*X) of indicator PCBs (iPCBs) and ΣPCB for variety of fish tissues. The linear regression equations were prepared for the relationships passing through the origin using total 1038 concentrations in ng/g wet weight. The equations present regression slopes ± 95% confidence intervals for the slopes. **Non-detects (ND) were set at the zero**. Similar relationships prepared using more appropriate logarithmically transformed data are presented in Table S6a. Separate equations were constructed by ND=detection limit and are presented in Table S5a.

	Regression Equation / Coefficient of determination (R^2)											
↓Y X→	iPCB3	iPCB6	iPCB7	iPCB3a	iPCB3b	iPCB3c	iPCB4	iPCB5	iPCB9	iPCB13	iPCB17	ΣPCB
a) Skin-removed Fillet of sport fish												
ΣPCB	3.88±0.21 X	3.24±0.07 X	2.71±0.04 X	4.2±0.05 X	4.27±0.15 X	3.98±0.14 X	3.21±0.05 X	2.78±0.03 X	2.49±0.03 X	1.75±0.02 X	1.45±0.01 X	
iPCB17	2.65±0.14 X	2.22±0.05 X	1.86±0.03 X	2.9±0.02 X	2.93±0.1 X	2.73±0.1 X	2.22±0.03 X	1.92±0.02 X	1.71±0.02 X	1.21±0.01 X		0.986
iPCB13	2.25±0.11 X	1.86±0.03 X	1.55±0.02 X	2.4±0.02 X	2.46±0.08 X	2.3±0.07 X	1.84±0.02 X	1.59±0.01 X	1.43±0.01 X		0.996	0.985
iPCB9	1.66±0.06 X	1.31±0.01 X	1.09±0 X	1.65±0.03 X	1.78±0.04 X	1.66±0.04 X	1.29±0.01 X	1.11±0.01 X		0.986	0.971	0.965
iPCB5	1.44±0.07 X	1.17±0.02 X	0.97±0.01 X	1.5±0.02 X	1.56±0.04 X	1.46±0.04 X	1.16±0.01 X		0.989	0.994	0.988	0.975
iPCB4	1.27±0.05 X	1.01±0.01 X	0.84±0.01 X	1.28±0.02 X	1.36±0.03 X	1.28±0.03 X		0.995	0.994	0.988	0.974	0.964
iPCB3c	1.05±0.02 X	0.76±0.01 X	0.62±0.01 X	0.89±0.03 X	1.07±0.01 X		0.908	0.878	0.919	0.847	0.810	0.811
iPCB3b	0.98±0.02 X	0.7±0.01 X	0.58±0.01 X	0.83±0.03 X		0.993	0.895	0.870	0.910	0.837	0.803	0.804
iPCB3a	0.89±0.05 X	0.75±0.02 X	0.63±0.01 X		0.773	0.788	0.966	0.979	0.956	0.988	0.991	0.975
iPCB7	1.54±0.05 X	1.21±0.01 X		0.940	0.930	0.937	0.990	0.982	0.997	0.974	0.957	0.952
iPCB6	1.3±0.04 X		0.993	0.901	0.953	0.954	0.972	0.957	0.984	0.947	0.924	0.923
iPCB3		0.854	0.808	0.597	0.952	0.946	0.749	0.707	0.775	0.673	0.627	0.634
b) Whole body YOY												
ΣPCB	42.42±1.01 X	4.31±0.07 X	3.76±0.05 X	4.88±0.06 X	15.96±0.25 X	17.59±0.34 X	4.64±0.06 X	3.88±0.04 X	3.49±0.04 X	2.12±0.02 X	1.63±0.01 X	
iPCB17	26.06±0.6 X	2.66±0.04 X	2.32±0.02 X	2.99±0.04 X	9.81±0.14 X	10.79±0.21 X	2.85±0.04 X	2.38±0.03 X	2.15±0.02 X	1.31±0 X		0.994
iPCB13	19.96±0.46 X	2.04±0.03 X	1.77±0.02 X	2.29±0.03 X	7.5±0.11 X	8.27±0.16 X	2.18±0.03 X	1.82±0.02 X	1.65±0.01 X		0.999	0.992
iPCB9	11.97±0.31 X	1.24±0.01 X	1.08±0 X	1.37±0.03 X	4.53±0.07 X	4.94±0.12 X	1.3±0.03 X	1.09±0.02 X		0.991	0.992	0.980
iPCB5	10.95±0.22 X	1.09±0.03 X	0.95±0.02 X	1.26±0.01 X	4.09±0.06 X	4.56±0.06 X	1.2±0 X		0.958	0.986	0.985	0.983
iPCB4	9.11±0.19 X	0.9±0.02 X	0.79±0.02 X	1.05±0 X	3.39±0.06 X	3.8±0.05 X		0.998	0.947	0.981	0.978	0.977
iPCB3c	2.41±0.04 X	0.23±0.01 X	0.2±0.01 X	0.27±0 X	0.89±0.01 X		0.977	0.980	0.925	0.955	0.952	0.952
iPCB3b	2.65±0.05 X	0.26±0.01 X	0.23±0 X	0.3±0.01 X		0.963	0.956	0.972	0.965	0.972	0.974	0.969
iPCB3a	8.64±0.19 X	0.86±0.02 X	0.75±0.02 X		0.954	0.975	1.000	0.998	0.947	0.981	0.978	0.978
iPCB7	11.04±0.3 X	1.15±0.01 X		0.938	0.958	0.913	0.938	0.949	0.999	0.987	0.988	0.975
iPCB6	9.45±0.3 X		0.997	0.914	0.940	0.881	0.914	0.926	0.994	0.974	0.976	0.961
iPCB3		0.888	0.912	0.944	0.956	0.972	0.948	0.952	0.920	0.940	0.938	0.934
c) Whole body forage fish												
ΣPCB	3.25±0.17 X	2.36±0.12 X	2.05±0.09 X	3.9±0.2 X	3.77±0.14 X	3.22±0.15 X	2.71±0.12 X	2.59±0.09 X	2±0.08 X	1.49±0.06 X	1.26±0.04 X	
iPCB17	2.59±0.08 X	1.88±0.05 X	1.64±0.03 X	3.1±0.11 X	3±0.08 X	2.56±0.08 X	2.16±0.06 X	2.06±0.05 X	1.59±0.02 X	1.18±0.01 X		0.981
iPCB13	2.19±0.06 X	1.59±0.04 X	1.38±0.02 X	2.62±0.07 X	2.53±0.09 X	2.16±0.05 X	1.83±0.04 X	1.74±0.05 X	1.34±0.02 X		0.998	0.973
iPCB9	1.63±0.03 X	1.18±0.02 X	1.03±0.01 X	1.95±0.07 X	1.88±0.06 X	1.61±0.05 X	1.36±0.05 X	1.29±0.05 X		0.998	0.997	0.973
iPCB5	1.25±0.06 X	0.91±0.05 X	0.79±0.03 X	1.5±0.05 X	1.45±0.04 X	1.24±0.04 X	1.05±0.02 X		0.979	0.986	0.989	0.978
iPCB4	1.19±0.05 X	0.86±0.04 X	0.75±0.02 X	1.44±0.02 X	1.38±0.06 X	1.18±0.02 X		0.991	0.985	0.992	0.987	0.965
iPCB3c	1.01±0.03 X	0.73±0.03 X	0.64±0.02 X	1.21±0.02 X	1.17±0.05 X		0.997	0.984	0.988	0.991	0.985	0.961
iPCB3b	0.86±0.03 X	0.62±0.03 X	0.54±0.02 X	1.03±0.05 X		0.980	0.979	0.992	0.984	0.985	0.990	0.979
iPCB3a	0.83±0.03 X	0.6±0.03 X	0.53±0.02 X		0.969	0.997	0.995	0.977	0.986	0.990	0.981	0.954
iPCB7	1.58±0.03 X	1.15±0.01 X		0.989	0.977	0.989	0.985	0.973	0.999	0.997	0.993	0.967
iPCB6	1.38±0.03 X		0.998	0.981	0.972	0.981	0.975	0.963	0.997	0.992	0.989	0.962
iPCB3		0.995	0.996	0.988	0.979	0.992	0.984	0.970	0.994	0.991	0.986	0.959

Table S7: Relationships (Y=*m*X) of indicator PCBs (iPCBs) and ΣPCB with dlPCB-TEQs in *skin-removed fillets of sport fish*. The linear regression equations were prepared for the relationships passing through the origin using a) all 470 measurements of iPCB in ng/g wet weight and dlPCB-TEQ in pg/g wet weight, and b) after removing 83 measurements that were mostly below detection for the PCB congeners. The equations present regression slopes ± 95% confidence intervals for the slopes. Non-detects (ND) were set at the detection limits (DL) for the PCB congeners for iPCBs and at half of the detection limits for dlPCBs. Similar relationships prepared using more appropriate logarithmically transformed data are presented in Table 3.

	Regression Equation			**Coefficient of determination (R^2)**		
↓X Y→	dlPCB-TEQ$_{94}$	dlPCB-TEQ$_{98}$	dlPCB-TEQ$_{05}$	dlPCB-TEQ$_{94}$	dlPCB-TEQ$_{98}$	dlPCB-TEQ$_{05}$
a) All data						
ΣPCB	0.033±0.002 X	0.032±0.001 X	0.023±0.001 X	0.746	0.744	0.655
iPCB17	0.048±0.002 X	0.048±0.002 X	0.034±0.002 X	0.731	0.729	0.631
iPCB13	0.058±0.003 X	0.058±0.003 X	0.041±0.002 X	0.758	0.756	0.664
iPCB9	0.087±0.003 X	0.086±0.003 X	0.062±0.003 X	0.804	0.803	0.729
iPCB5	0.095±0.004 X	0.094±0.004 X	0.067±0.004 X	0.775	0.773	0.686
iPCB4	0.11±0.005 X	0.109±0.005 X	0.078±0.004 X	0.792	0.791	0.712
iPCB3c	0.15±0.006 X	0.149±0.006 X	0.11±0.004 X	0.811	0.812	0.794
iPCB3b	0.164±0.006 X	0.164±0.006 X	0.121±0.005 X	0.807	0.808	0.796
iPCB3a	0.137±0.007 X	0.136±0.007 X	0.095±0.006 X	0.731	0.729	0.628
iPCB7	0.096±0.004 X	0.095±0.004 X	0.069±0.003 X	0.812	0.811	0.746
iPCB6	0.117±0.004 X	0.117±0.004 X	0.085±0.004 X	0.814	0.813	0.763
iPCB3	0.15±0.008 X	0.15±0.008 X	0.113±0.006 X	0.687	0.689	0.721
b) After removing major non-detects						
ΣPCB	0.033±0.002 X	0.032±0.002 X	0.023±0.001 X	0.734	0.732	0.639
iPCB17	0.048±0.003 X	0.048±0.003 X	0.034±0.002 X	0.719	0.716	0.614
iPCB13	0.058±0.003 X	0.058±0.003 X	0.041±0.003 X	0.747	0.745	0.649
iPCB9	0.087±0.004 X	0.086±0.004 X	0.062±0.003 X	0.794	0.793	0.716
iPCB5	0.095±0.004 X	0.094±0.004 X	0.067±0.004 X	0.765	0.763	0.672
iPCB4	0.11±0.005 X	0.109±0.005 X	0.078±0.004 X	0.783	0.781	0.699
iPCB3c	0.131±0.01 X	0.131±0.009 X	0.093±0.008 X	0.553	0.551	0.483
iPCB3b	0.164±0.007 X	0.164±0.007 X	0.121±0.005 X	0.797	0.798	0.785
iPCB3a	0.137±0.007 X	0.136±0.007 X	0.095±0.006 X	0.721	0.718	0.614
iPCB7	0.096±0.004 X	0.095±0.004 X	0.069±0.003 X	0.803	0.802	0.733
iPCB6	0.117±0.005 X	0.117±0.005 X	0.085±0.004 X	0.804	0.803	0.751
iPCB3	0.15±0.009 X	0.15±0.009 X	0.113±0.006 X	0.671	0.674	0.707

FIGURE S1: Estimated root mean squared error of prediction (RMSEP) as a function of the number of components from the PLS analysis on PCB congener data for SBF

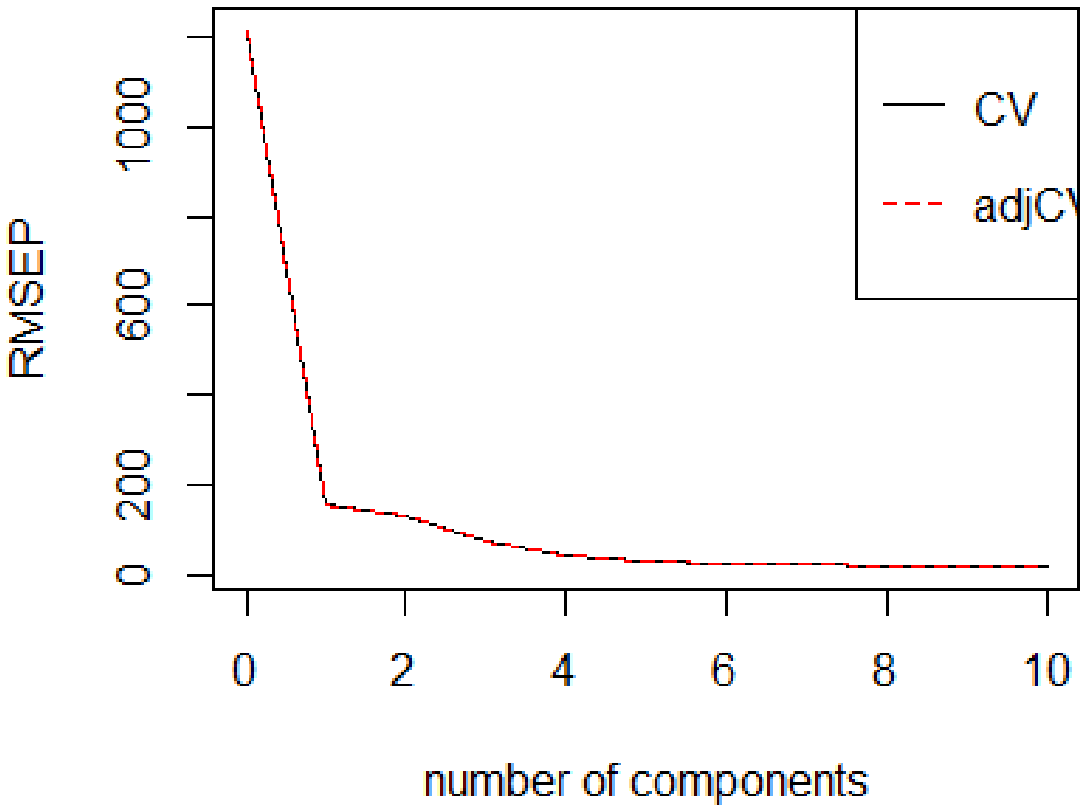
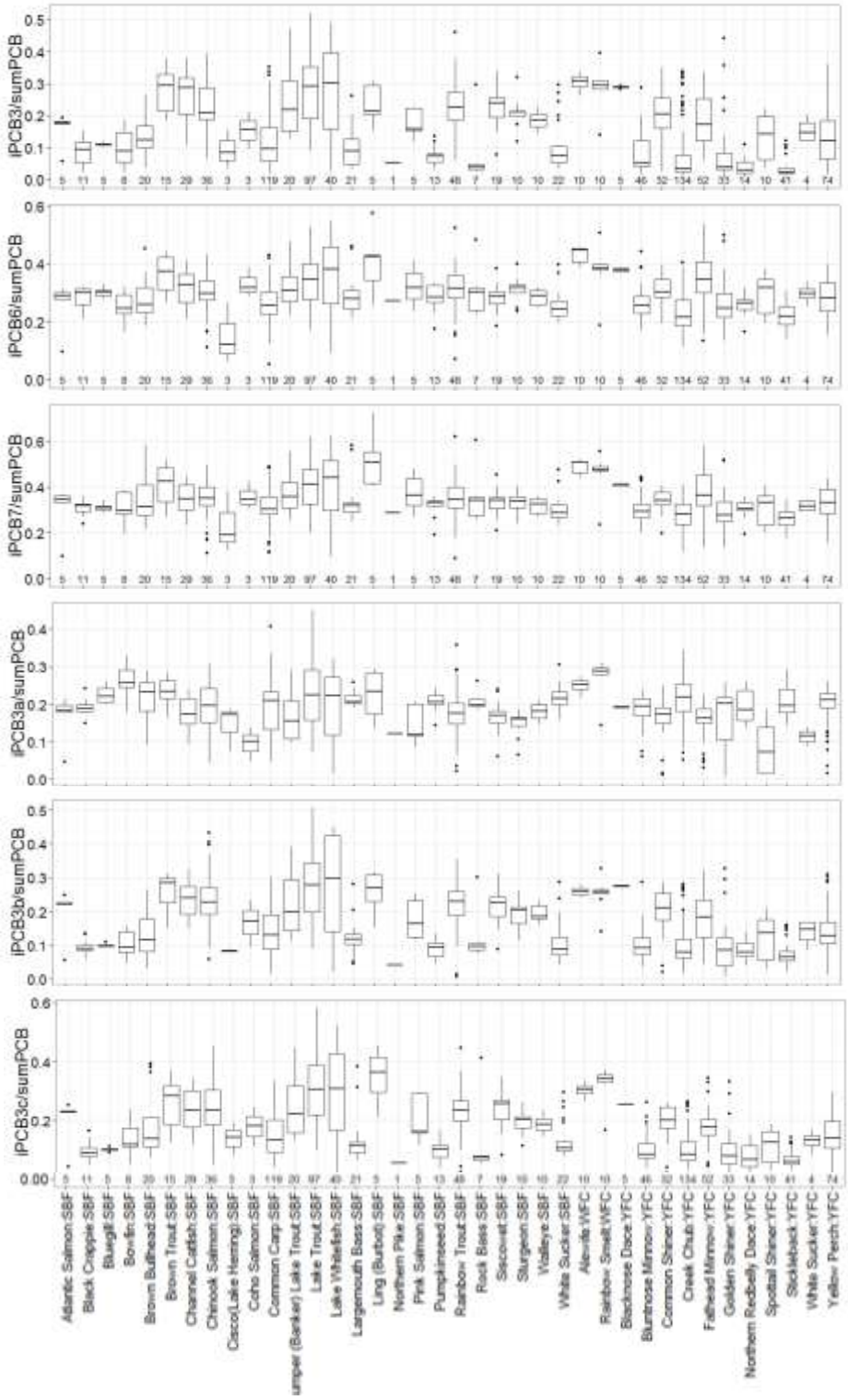


FIGURE S2: Contributions (as fraction) of various iPCBs to ΣPCB (sumPCB) for skin-removed boneless fillets of sport fish (SBF, n=572), whole fish composite of forage fish (WFC, limited data, n=22), and young-of-the-year fish composites (YFC, n=445) of a variety of fish species. The line in the box presents median, box represents 25-75 percentiles, and whiskers present non-outlier or non-extreme values.



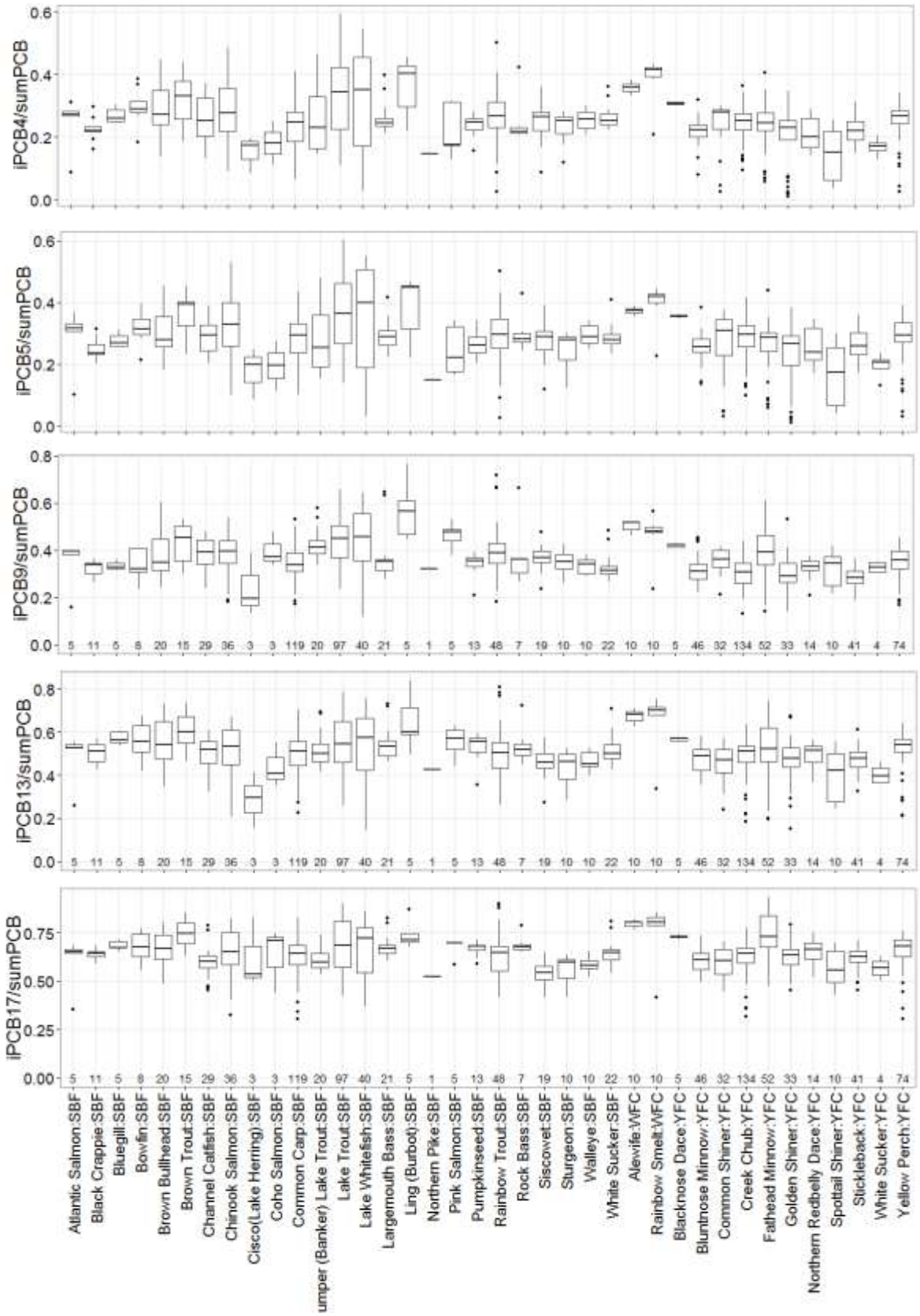

iPCB4/sumPCB
iPCB5/sumPCB
iPCB9/sumPCB
iPCB13/sumPCB
iPCB17/sumPCB
5 11 5 8 20 15 29 36 3 3 119 20 97 40 21 5 1 5 13 48 7 19 10 10 22 10 10 5 46 32 134 52 33 14 10 41 4 74
Atlantic Salmon:SBF
Black Crappie:SBF
Bluegill:SBF
Bowfin:SBF
Brown Bullhead:SBF
Brown Trout:SBF
Channel Catfish:SBF
Chinook Salmon:SBF
Cisco(Lake Herring):SBF
Coho Salmon:SBF
Common Carp:SBF
umper (Banker) Lake Trout:SBF
Lake Trout:SBF
Lake Whitefish:SBF
Largemouth Bass:SBF
Ling (Burbot):SBF
Northern Pike:SBF
Pink Salmon:SBF
Pumpkinseed:SBF
Rainbow Trout:SBF
Rock Bass:SBF
Siscowet:SBF
Sturgeon:SBF
Walleye:SBF
White Sucker:SBF
Alewife:WFC
Rainbow Smelt:WFC
Blacknose Dace:YFC
Bluntnose Minnow:YFC
Common Shiner:YFC
Creek Chub:YFC
Fathead Minnow:YFC
Golden Shiner:YFC
Northern Redbelly Dace:YFC
Spottail Shiner:YFC
Stickleback:YFC
White Sucker:YFC
Yellow Perch:YFC

FIGURE S3: Relationship of iPCB3 to ΣPCB (sumPCB) for skin-removed boneless fillets of sport fish (SBF, n=572) by a) fish species, and b,c) sampling location.

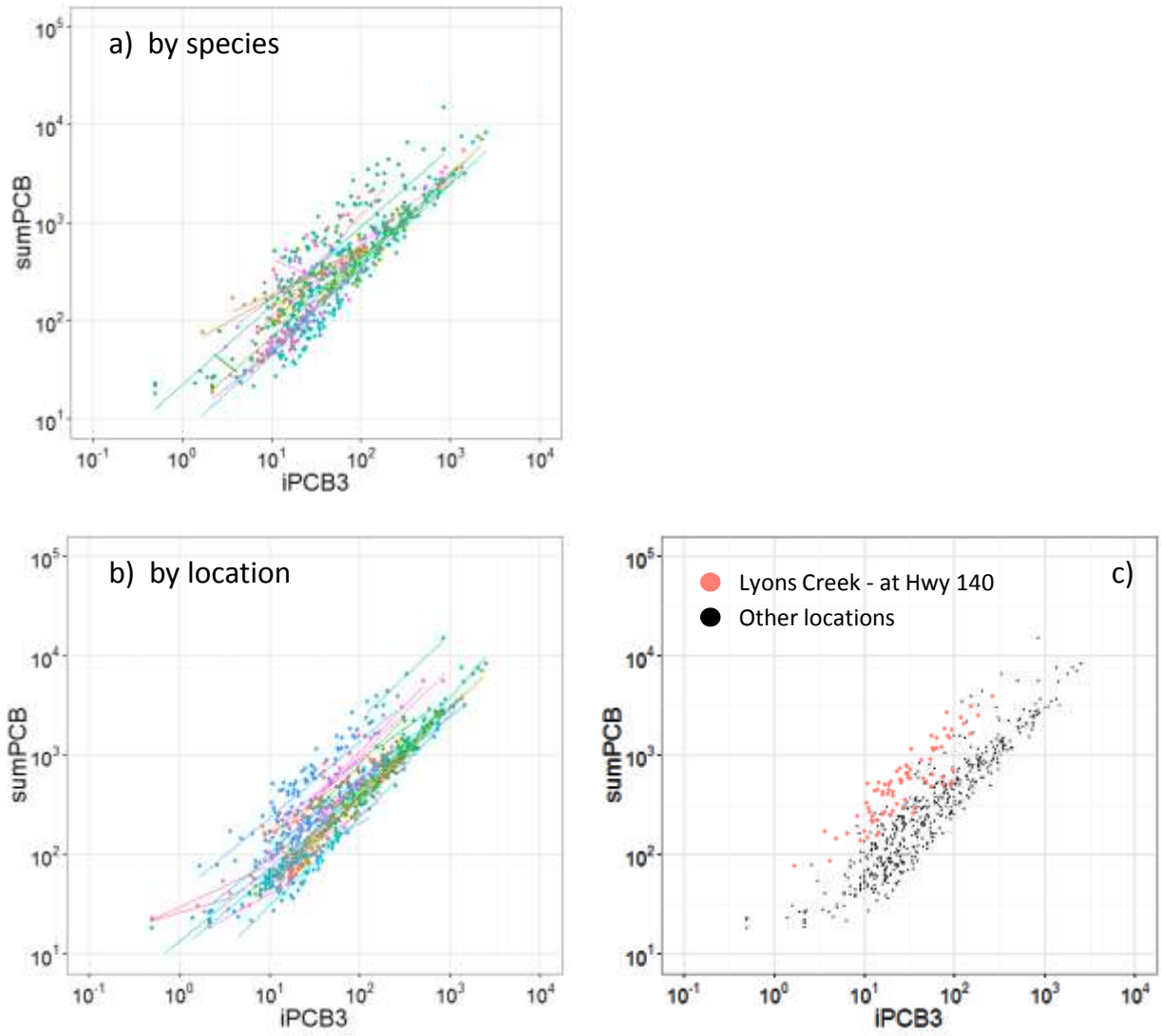


FIGURE S4: Relationships (logY = *m* logX) of indicator PCBs (iPCBs) and ΣPCB in *skin-removed fillets of sport fish*. The linear regression equations were prepared for the relationships passing through the origin using 572 concentrations in ng/g wet weight. R^2 is the coefficient of determination. Non-detects (ND) were considered at the detection limits (DL).

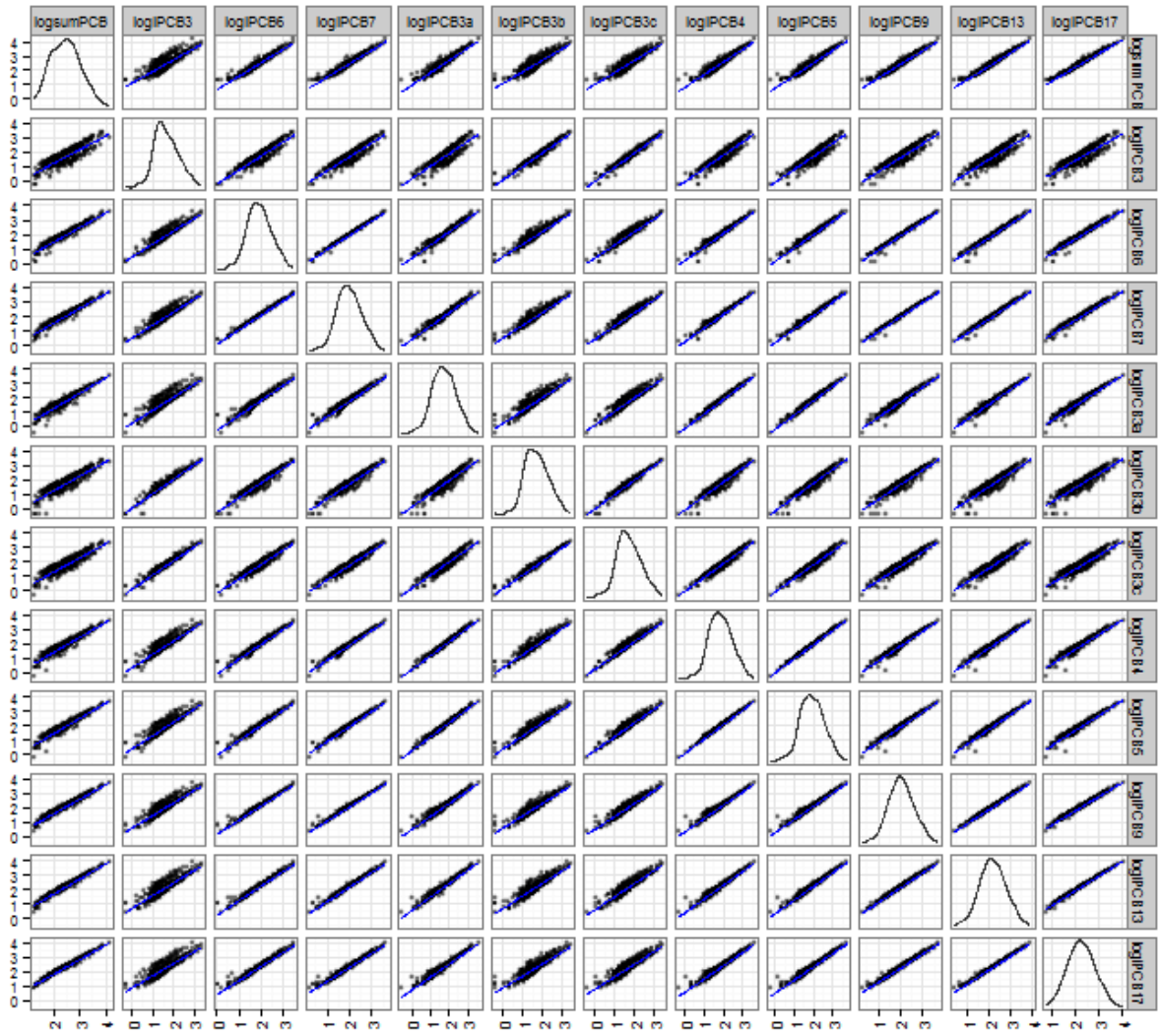


FIGURE S5: Relationships (logY = *m* logX) of indicator PCBs (iPCBs) and ΣPCB in *whole body young-of-the-year fish*. The linear regression equations were prepared for the relationships passing through the origin using 445 concentrations in ng/g wet weight. R^2 is the coefficient of determination. Non-detects (ND) were considered at the detection limits (DL).

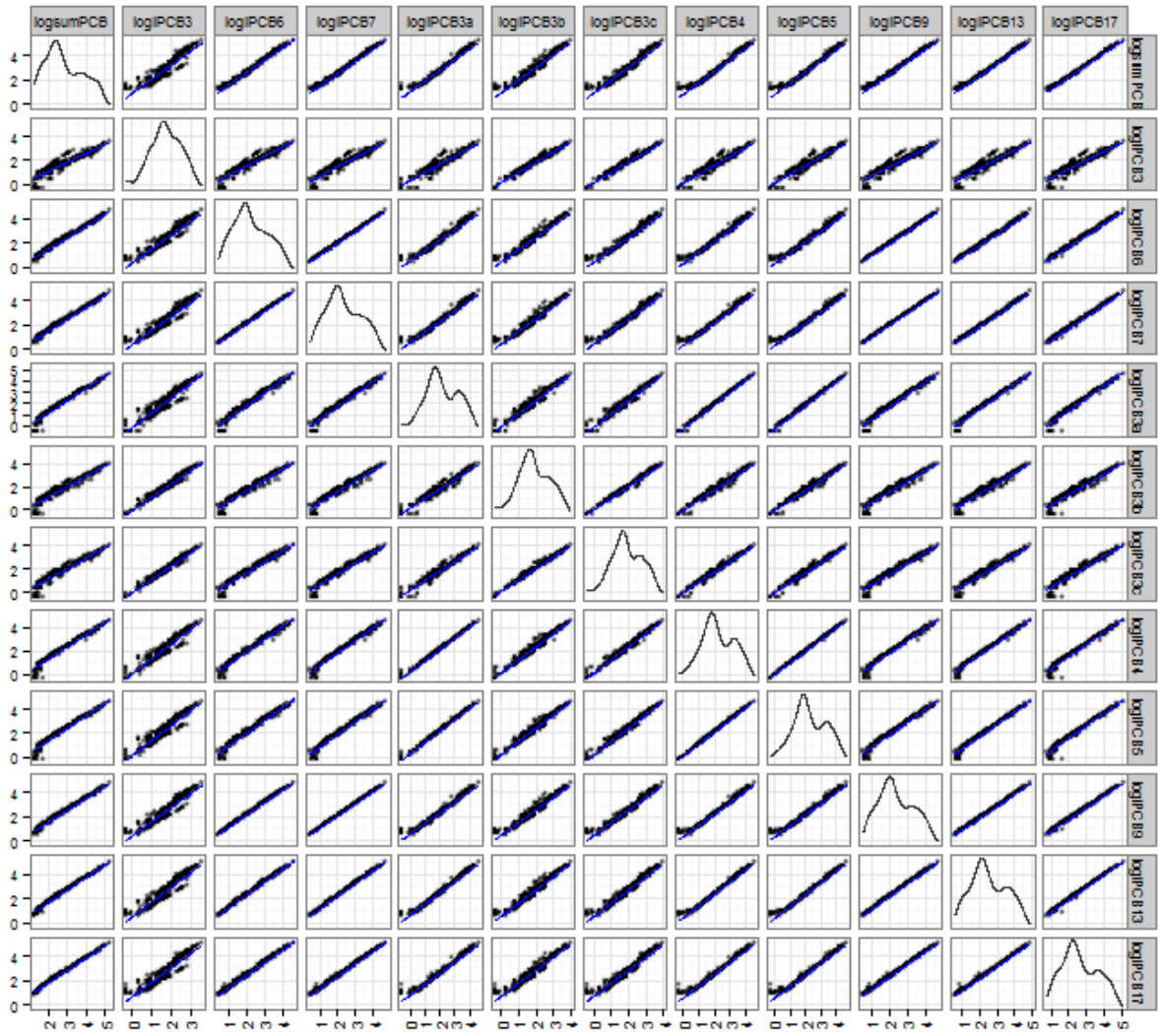


FIGURE S6: Relationships (Y=*m*X) of indicator PCBs (iPCBs) and ΣPCB (ng/g ww) with dlPCB-TEQs (pg/g ww) in *skin-removed fillets of sport fish*. The linear regression equations were prepared for the relationships passing through the origin using 470 measurements. R^2 is the coefficient of determination. Non-detects (ND) were considered at the detection limits (DL) for PCB congeners of iPCBs and half of the DLs for the dlPCBs of TEQs.

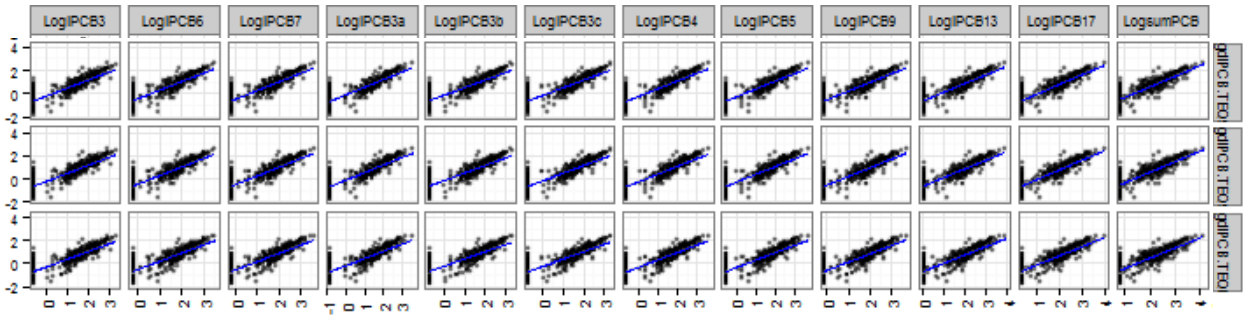


FIGURE S7: Relationships of indicator PCBs (iPCBs) and ΣPCB (sumPCB) with dlPCB-TEQs in *skin-removed fillets of various sport fish*. The species specific linear regressions are shown. Non-detects (ND) were considered at the detection limits (DL) for PCB congeners of iPCBs and half of the DLs for the dlPCBs of TEQs.

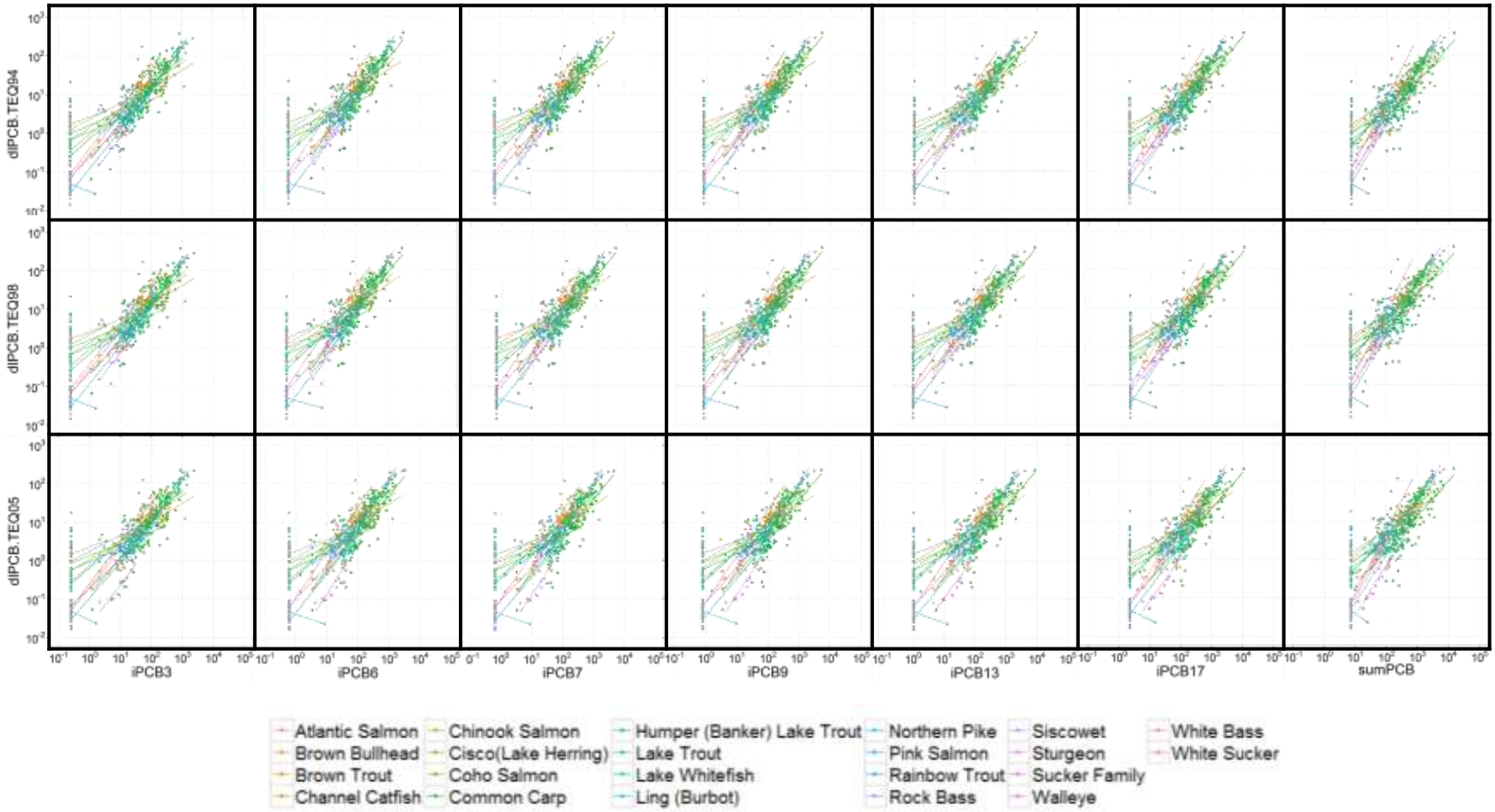


FIGURE S8: Relationships of ΣPCB (sumPCB) (≤7.15 ng/g removed) with dlPCB-TEQ_{05} in *skin-removed fillets of various sport fish*. The species specific linear regressions are shown. Non-detects (ND) were considered at the detection limits (DL) for PCB congeners of iPCBs and half of the DLs for the dlPCBs of TEQs.

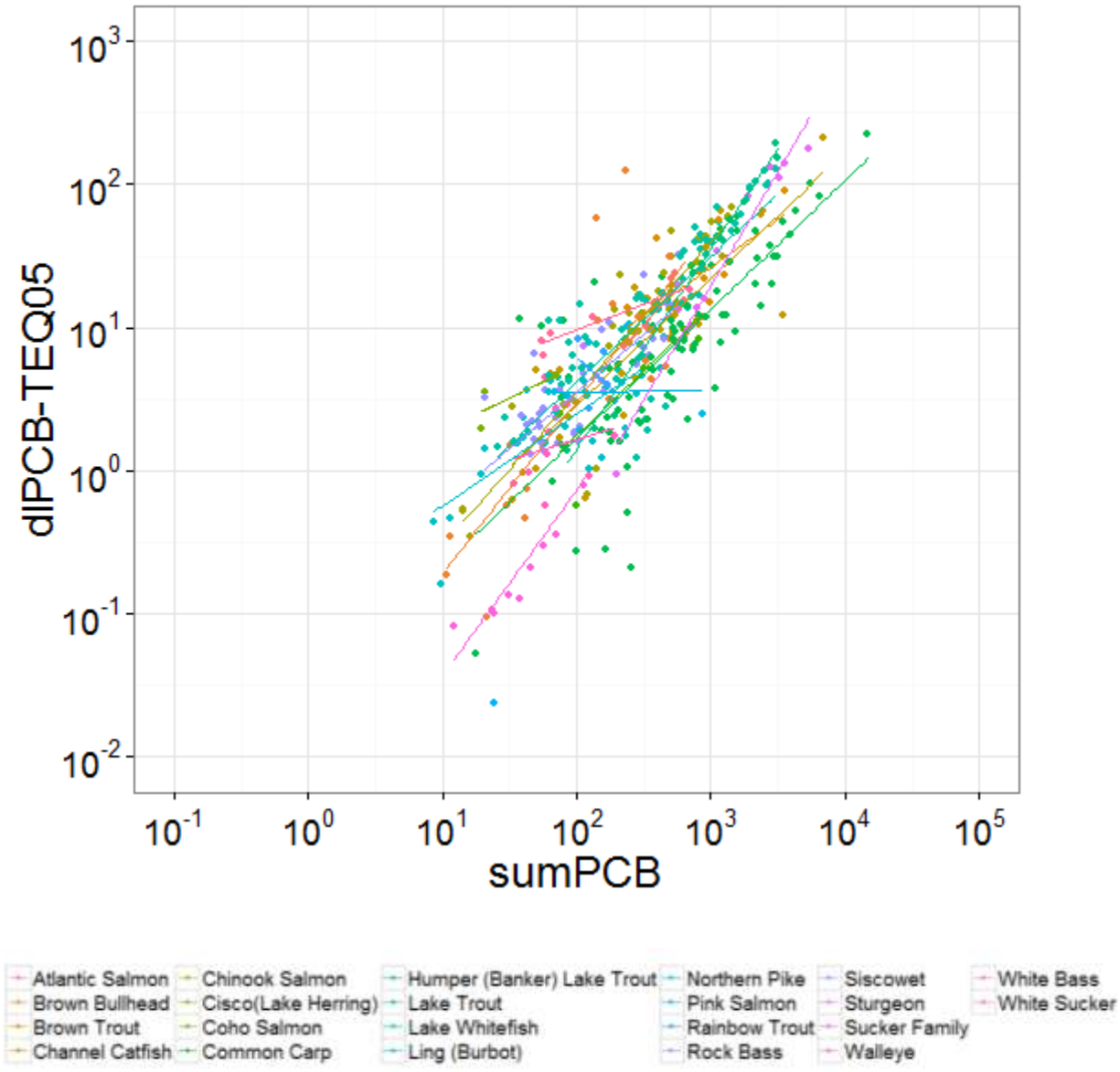
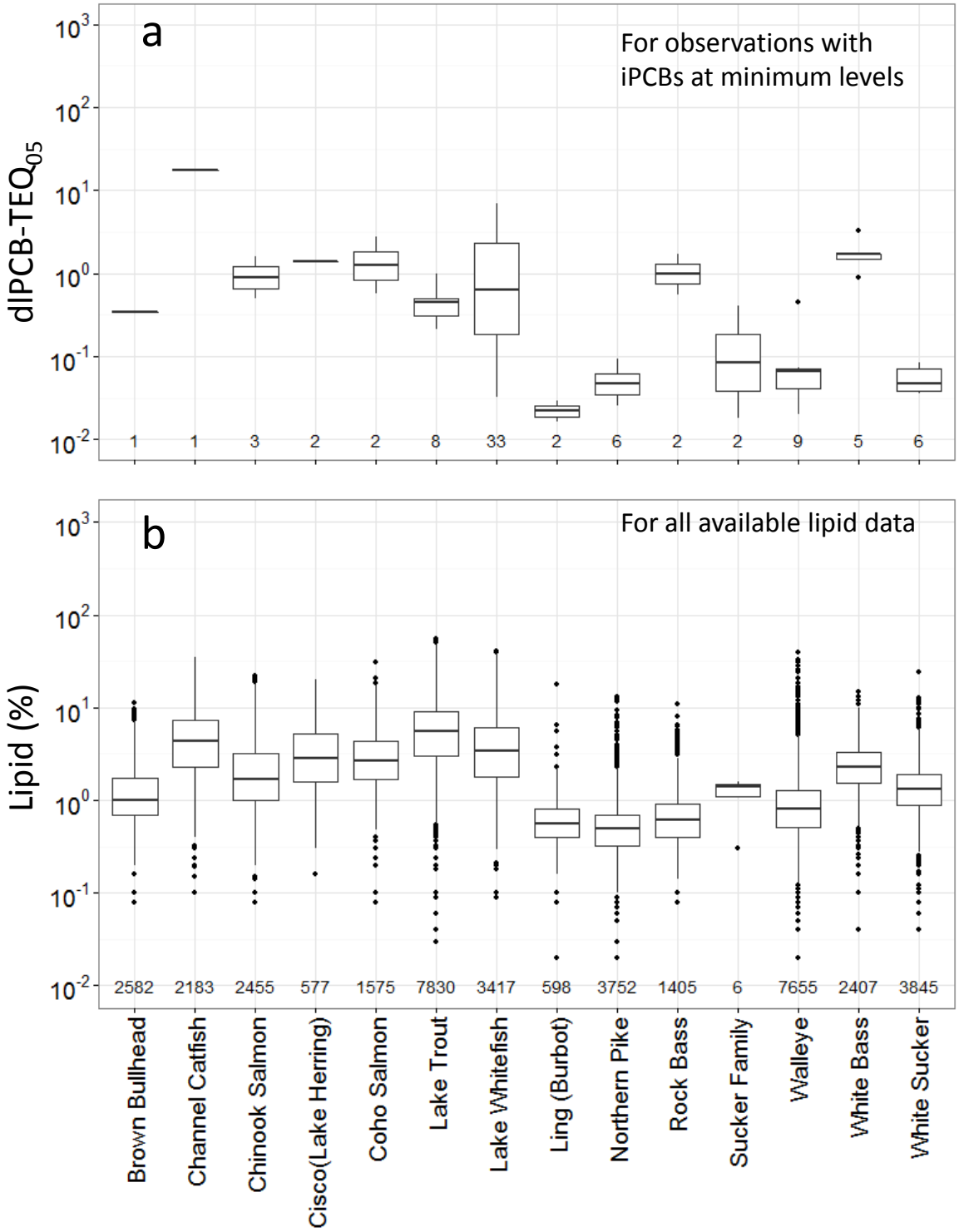


FIGURE S9: Boxplots of a) dlPCB-TEQ$_{05}$ (pg/g) by species for all observations with iPCB values at the minimum levels, and b) lipid content (%) by species for all available data. The values above the x-axes show sample sizes.



References

Bhavsar, S.P.; Reiner, E.J.; Hayton, A.; Fletcher, R.; MacPherson, K. Converting Toxic Equivalents (TEQ) of dioxins and dioxin-like compounds in fish from one Toxic Equivalency Factor (TEF) scheme to another. Environment International. 34:915-921; 2008

OMOE. The determination of polychlorinated biphenyl congeners (PCBc), in fish, clams and mussels by gas liquid chromatography-electron capture detection (GLC-ECD). Laboratory Services Branch Method PCBC-E3411. Toronto, Ontario, Canada: Ontario Ministry of the Environment; 2010