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Polychlorinated Biphenyls in Salmon and Salmon Feed: Global Differences and Bioaccumulation

DANIEL L. CARLSON AND RONALD A. HITES*

School of Public and Environmental Affairs, Indiana University, Bloomington, Indiana 47405

Concentrations of 160 polychlorinated biphenyl (PCB) congeners or congener groups were determined in approximately 600 farmed Atlantic salmon from around the world and wild (ocean-caught) Pacific salmon from the Northeast Pacific. Concentrations and PCB congener profiles were analyzed to provide insight into the sources and uptake of PCBs in salmon as well as regional differences. Although total PCB concentrations in wild salmon appeared to be correlated to total lipid content, the increased proportion of total lipids in the farmed salmon could not account for the much greater PCB concentrations. We investigated the PCB congener patterns of hundreds of salmon samples using principal component analysis to further illuminate regional and species differences. Three major PCB patterns were observed, in most wild fish (except British Columbia and Oregon chinook), in farmed fish from the Atlantic, and in most farmed fish from the Pacific. The PCB congener profiles of farmed salmon often closely corresponded to a sample of commercial feed purchased in the same region, indicating that the feed is likely to be the major source of PCBs for farmed salmon. In such cases where PCB profiles in fish and feed were similar, a comparison of congener concentrations in fish and the feed showed that the majority of congeners, with some notable exceptions, were bioaccumulative to the same extent, irrespective of physical properties.

Introduction

The consumption of salmon in the United States and Europe has seen double-digit growth between 1987 and 1999 (*1*). This growth has come in concert with a 40-fold increase in the amount of low-cost, farm-raised salmon (mainly Atlantic salmon, *Salmo salar*) produced over the past 20 years (*2*). Consumption of salmon and other fish has also been promoted as a good source of omega-3 fatty acids, which are thought to protect against fatal myocardial infarction and sudden cardiac death (*3*). Clearly, salmon (and farmed Atlantic salmon in particular) are becoming more important in the diets of many people.

These trends prompted recent studies to evaluate the levels of environmental contaminants, including polychlorinated biphenyls (PCBs), and associated health risks in farmed salmon from around the world and in wild (used here to mean ocean-caught; some hatch in the wild, and some are initially raised in fish hatcheries) Pacific salmon (*4*−*7*). Results showed consistently higher concentrations of contaminants in farmed salmon compared to most wild salmon. For example, the results of the largest study (*4*) showed that total PCB concentrations were significantly higher in farmed salmon (median 41.5 ng/g wet weight) than those in wild salmon (median 3.2 ng/g wet weight). These PCB concentrations, together with those of dioxins, dieldrin, and toxaphene, resulted in a suggested limit of one or fewer meals per month for farmed Atlantic salmon using Environmental Protection Agency (EPA) calculation methods to limit the additional risk of cancer to 1:100 000 (*4*). This, of course, does not take into account potential health benefits of eating farmed salmon; however, similar health benefits can be obtained via other dietary sources with lower associated risks, including most wild Pacific salmon. Another solution is to reduce the contaminants in the farmed salmon while retaining the omega-3 fatty acids, thereby reducing the risk to the consumer while maintaining the benefits (*8*, *9*).

A wide range (4.9−76.2 ng/g wet weight) of total PCB concentrations was observed among the farmed salmon in the Hites et al. study (*4*), prompting further discussion on the need to identify the source of contaminants in farmed salmon (*10*, *11*). The exposure route for farmed salmon is thought to be the commercial feed (*4*−*7*); for example, regions of the world with higher PCB concentrations in farmed fish also have higher concentrations in the commercially available salmon feed (*4*). The PCB data reported in Hites et al. (*4*) included total PCBs only; however, 160 congeners or congener groups, including all 209 PCBs, were quantified. In this work, we investigate the behavior of PCBs in the same salmon and feed samples described previously but now on a congener-specific basis.

Two samples can have the same total PCB concentration but very different congener profiles; thus, using congener-specific data is a more precise way of identifying which samples are similar to each other. Using principal component analysis, we elucidate the differences in PCB congener profiles in salmon from different regions and compare the farmed salmon to the commercial salmon feed. Comparing the PCB profiles in fish and feed allows us to understand the behavior of PCB congeners and total PCBs in the Atlantic salmon aquaculture system; we hypothesized that in some cases the PCB profiles of salmon and the feed that they eat would be similar. Understanding the differences in PCB congener patterns between regions and species helps us determine possible explanations for the total PCB concentration differences among and between wild and farmed salmon. Being able to explain differences in total PCB concentrations will aid in developing ways to reduce those concentrations in farmed Atlantic salmon in the future.

Experimental Section

Details on the sampling methodology have been published elsewhere (*4*, *12*); an abbreviated description is provided here. PCBs were measured in about 600 farmed and wild salmon (totaling ∼2 metric tons) collected from around the world. Farmed Atlantic salmon (*Salmo salar*) were purchased from wholesale suppliers in the United States, the United Kingdom, Norway, and Canada between March and December, 2002. These suppliers provided farmed salmon from 51 different salmon farms in eight salmon farming regions: Norway, Chile, Scotland, British Columbia (Western Canada), Eastern Canada, the Faroe Islands, Maine (Eastern U.S.), and Washington (Western U.S.). Ten salmon were obtained from each farm, nine of which were randomly grouped into three composites of three fish each. Sizes were similar, with an

* Corresponding author phone: (812) 855-0193; fax: (812) 855-1076; e-mail: hitesr@indiana.edu.

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average length of 73.8 cm and a standard deviation of 3.1 cm. Each composite is referred to as a sample and was made up of homogenized whole fillets (including skin).

Between September 2001 and August 2002, other suppliers provided 135 ocean-caught fish representing five species of Pacific salmon that spent their adult years in the wild: chum (*Oncorhynchus keta*), coho (*O. kisutch*), chinook (*O. tshawytscha*), pink (*O. gorbuscha*), and sockeye (*O. nerka*). Individuals of each species were purchased from some, but not all, of the following geographic regions: Kodiak, Alaska, Southeast Alaska, British Columbia, and Oregon. Sizes were more variable than those of the farmed salmon; average lengths (with standard deviations) for each species were, for chum, 71.8 ± 3.4 cm; coho, 69.8 ± 2.6 cm; chinook, 78.7 ± 8.7 cm; pink, 55.5 ± 2.0 cm; and sockeye, 65.9 ± 3.4 cm. No attempt was made to distinguish salmon that hatched in the wild from those that spent their first months in fish hatcheries (where they consumed commercial feed) or to distinguish ocean-type (which migrate to the ocean in the first months of their lives and stay near the coast) from stream-type salmon (which migrate to the ocean in their second year or later and range over large portions of the North Pacific) in the case of chinook (*13*). Note that while all nonfarmed fish in this study are more properly called ocean-caught the colloquial term "wild" will be used here for consistency with prior work and commercial usage. Three composites of three fish for each species at each location resulted in a total of 45 samples for analysis.

Finally, 13 samples of salmon feed were purchased from the European, North American, and South American outlets of the two major feed companies, which together have ~80% of the global market for salmon feed (*14*). For the first company, two samples of feed, purchased 3−4 months apart, were obtained from facilities in Scotland, Eastern Canada, British Columbia, and Chile. For the second company, two samples of feed, purchased 3−4 months apart, were obtained from facilities in Scotland and British Columbia, and one sample was obtained from a facility in Chile. Two samples per location were purchased several months apart to account for possible seasonal variations in the feed formulation.

All samples were sent to the analytical laboratory (AXYS Analytical in Sidney, British Columbia) fresh or frozen on ice or gel-packs. PCBs were quantified using EPA method 1668A; this technique is an isotope-dilution, congener-specific method for the 12 dioxin-like congeners. An internal standard method was used for the remaining 197 congeners. Labeled surrogates were added before samples were extracted; data were recovery corrected for losses in extraction and cleanup, and analytes were quantified against similarly labeled analogues. The analysis was performed on a Micromass AutoSpec Ultima magnetic sector mass spectrometer equipped with a Hewlett-Packard 6890 gas chromatograph; the mass spectrometer was operated at a static resolution of 10 000. Chromatographic separation was achieved using a Supleco SPB−octyl column (30 m, 0.25 mm i.d., 0.25 μm film thickness). A total of 160 congeners and congener groups were separated and quantified (Table S1).

All analyses were conducted in accordance with AXYS' accredited quality assurance/quality control (QA/QC) program. Each analysis batch of nine samples also included a procedural blank, a "known" or laboratory control sample, and an analysis duplicate. The sample results were reviewed and evaluated in relation to the QA/QC samples worked up at the same time. The sample internal standard recoveries and detection limits, procedural blank data, and laboratory control sample data were evaluated against method criteria to ensure data quality. All instrument QA specifications for EPA methods were adhered to and applied to all analyses conducted for this study. All data met the QA/QC specifications. In general, duplicate measurements differed from each other by <15%. All blank measurements were near or below the detection limits; hence, blank values were not subtracted from the sample measurements.

Principal component analysis was carried out with 115 congeners or congener groups that had a high rate of detection (Table S1) using the SAS software program (Cary, NC) to examine similarities and differences of PCB congener profiles among farmed and wild salmon. The concentration of each congener was normalized to the sum of the 115 included congeners to give the fraction of the total for each congener. Because this value varied over nearly 2 orders of magnitude for a large number of low- and high-molecular-weight congeners, the logarithm of this fraction was taken. Of 24 265 measurements, 33 (0.14%) were below detection limits or could not be quantified. These were replaced either with a value of half the detection limit (in 12 cases) or with a value equal to the average for the same congener from the other two samples of salmon from the same farm or species and region (in 21 cases).

For the analysis comparing salmon and feed congener profiles, the percent of total PCBs for each congener was calculated for a sample from one fish farm

$$P_{n,\mathrm{A}} = 100\frac{C_{n,\mathrm{A}}}{C_{\mathrm{tot,A}}} \tag{1}$$

where C_{tot} is the total PCB concentration (in pg/g wet weight), C_n is the concentration of congener n, and A denotes farmed Atlantic salmon. Likewise, the percent congener contributions were calculated for feed

$$P_{n,\mathrm{F}} = 100\frac{C_{n,\mathrm{F}}}{C_{\mathrm{tot,F}}} \tag{2}$$

where F denotes feed, and for wild Pacific salmon

$$P_{n,\mathrm{W}} = 100\frac{C_{n,\mathrm{W}}}{C_{\mathrm{tot,W}}} \tag{3}$$

where W denotes wild Pacific salmon. For Atlantic farmed salmon and wild Pacific salmon, $P_{n,\mathrm{A}}$ or $P_{n,\mathrm{W}}$ values were averaged for the three samples from the same fish farm or from the same species and location; these average values are denoted as $\overline{P_{n,\mathrm{A}}}$ or $\overline{P_{n,\mathrm{W}}}$. The P-values for the feed samples were not averaged together. Ratios of the percent congener contributions were also calculated; for example, the farmed Atlantic salmon/feed ratio for congener n is

$$R_{n,\mathrm{A/F}} = \frac{\overline{P_{n,\mathrm{A}}}}{P_{n,\mathrm{F}}} \tag{4}$$

We refer to these R-values as "percent congener ratios." The similarity of PCB congener profiles from different samples is described by calculating the variance of all percent congener ratios (approximately 140, depending on the number of nondetects) for one pair of samples. The ratio of congener concentrations, $B_{n,\mathrm{A/F}}$, was also calculated for certain fish/food pairs:

$$B_{n,\mathrm{A/F}} = \frac{C_{n,\mathrm{A}}}{C_{n,\mathrm{F}}} \tag{5}$$

Results and Discussion

Concentration Differences. Figure 1 shows the PCB concentrations by region for farmed salmon compared to wild salmon. Farmed Atlantic salmon raised on the eastern and western Atlantic coasts had higher PCB concentrations than

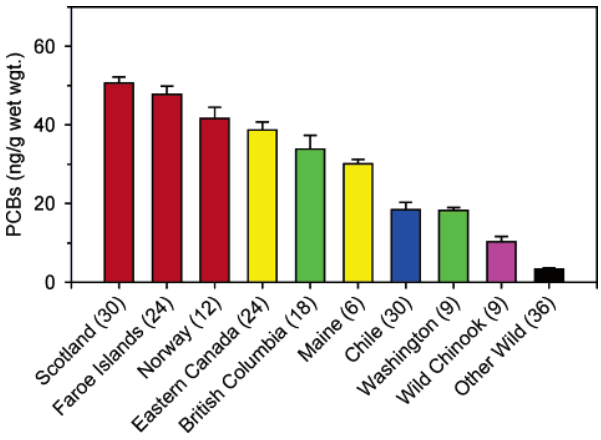

FIGURE 1. Average PCB concentrations (and standard errors) in farmed Atlantic salmon (by region East Atlantic, red; West Atlantic, yellow; North Pacific, green; South Pacific, blue) and in wild Pacific salmon. The number of composite samples for each group is given in parentheses.

those raised on the North American and Chilean Pacific coasts. On average, salmon caught in the wild (many of which likely had been initially raised in fish hatcheries) had lower PCB concentrations than farmed salmon from any location.

Chinook are reported separately from the other wild Pacific salmon because of their higher PCB concentrations. The higher concentrations are likely caused by chinook eating higher in the food web compared to sockeye, coho, and pink salmon; chum have a similar trophic level but very different diet (*15*−*17*). Differences in lifespan (total lifetime exposure to PCBs) and the efficiency of food conversion (how much food energy can be used for growth) should also affect PCB concentrations. The average concentrations for wild salmon, by region and species, are shown in Figure S1.

Unlike the other wild salmon species, there are two wild types of chinook: (a) ocean-type, which migrate to the ocean in the first months of their lives and stay near the coast, and (b) stream-type, which migrate to the ocean in their second year or later and range over large portions of the North Pacific (*13*). Most other wild Pacific salmon also have an offshore marine distribution, with the exception of many coho stocks that have coastal distributions (*16*). Chinook from Alaska are almost entirely stream-type, while chinook from northern British Columbia are mixed stream-type and ocean-type, and those from southern British Columbia and further south are mostly ocean-type (*13*). Whether a chinook is stream-type or ocean-type might affect PCB concentrations, because the ocean-type chinook spend their lives in waters with potentially greater anthropogenic contamination; also, ocean-type Chinook tend to be smaller. Indeed, our data show that chinook from Oregon and British Columbia (the likely ocean-type chinook) have higher PCB concentrations than those from Alaska (likely stream-type). Chinook from Oregon and British Colombia also have much higher concentrations of polybrominated diphenyl ethers (PBDEs) (*12*). Chinook from the Puget Sound in Washington (a region with areas of dense human population) had higher PCB (*18*) and PBDE (*19*) concentrations than even the farmed salmon in this study. However, further work in this area is necessary, including positively identifying stream- versus ocean-type chinook, before conclusions can be drawn; for instance, differences in trophic level could have a confounding effect. This work could be important for certain local communities. Although chinook typically only account for 1−2% of the Pacific catch and 50−70% of that is from Alaska (*2*), chinook likely represent an important exposure pathway in the diet of some people with subsistence lifestyles and with high salmon consumption

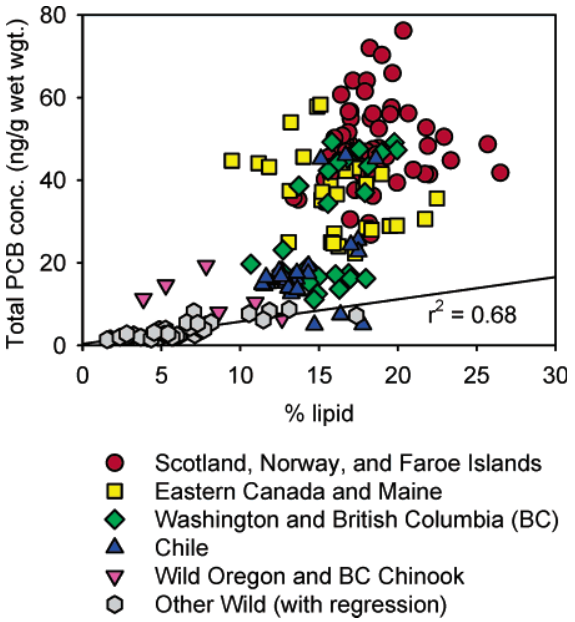

FIGURE 2. Total PCB concentration versus total lipid content in each salmon analyzed. The regression (black line) is for wild salmon only excluding chinook from Oregon and British Columbia.

rates. Such people include the Confederated Tribes of the Umatilla Indian Reservation in Oregon (*20*).

The differences in total PCB concentrations among all wild salmon species appeared to be related to the total lipid content of the salmon. (See the regression in Figure 2.) Prior work has found a strong relationship between average lipid content and average total PCB concentration across species for Lake Michigan salmonids, although only weak relationships are seen when total PCBs are plotted versus lipid content for individuals within a species (*21*, *22*). Indeed, no such relationship between PCB concentrations and total lipids can be seen within the farmed Atlantic salmon data set either. On the basis of the observed correlation between total PCBs and lipids for the wild salmon, though, one might infer that the greater PCB concentrations in the farmed salmon could be related to the higher lipid content in these fish. However, most of the PCB concentrations for the farmed salmon are well above (in some cases by almost an order of magnitude) the extrapolated regression line for wild salmon (Figure 2). When the data were transformed logarithmically, then a linear relationship was still observed for the wild salmon, and the farmed salmon data were also well above this extrapolated line (not shown). This indicates that the higher PCB content of the farmed salmon cannot be attributed to higher lipid content.

The increased lipid content in the farmed salmon may, in fact, indicate that one should expect farmed salmon to have lower contaminant concentrations than wild salmon, if all other factors (such as diet) were the same and the species differences in PCB uptake from the environment were negligible. Farmed salmon do not need to expend energy on migration or foraging; hence, a greater proportion of the food that they eat would be available for increasing body mass (as indicated by the high lipid content). A larger body mass, given the same intake of dietary PCBs, would lead to a lower PCB concentration in the fish tissue. Likewise, farmed salmon have a shortened life cycle for commercial purposes; a shorter life would lead to a lower lifetime PCB intake.

Differences in PCB Congener Profiles. Principal component analysis (PCA) was used to explore PCB congener profile differences of farmed and wild salmon in a systematic manner. The goal of PCA is to transform the variables (in our

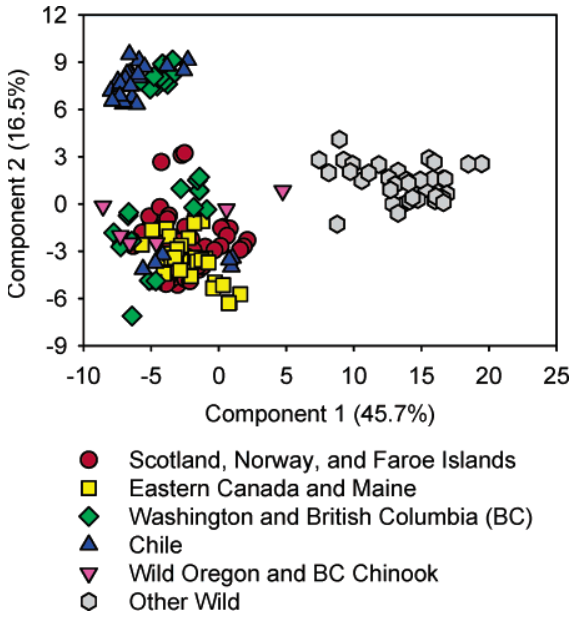

FIGURE 3. Plot of component 2 versus component 1 for all salmon samples. Plots of additional components can be seen in Figures S2–S4.

case, 115 of them) in a data set into a new set of variables (components), of which only a handful can explain a large portion of the variability in the original data set. Figure 3 shows a plot of components 1 (accounting for 45.7% of the variability) and 2 (accounting for an additional 16.5% of the variability) for all farmed and wild salmon. The greatest differences (as exhibited by component 1) were between most of the wild and farmed salmon. This is not surprising, considering the two groups differ dramatically not only in life history and diet but also in the genus and species sampled. We note the exceptions are the chinook from Oregon and British Columbia, which have component 1 scores similar to those of farmed salmon; these chinook are more likely ocean-type salmon with different life histories than most of the other wild salmon sampled. Component 2 generally reveals differences between farmed salmon raised in the Atlantic and those raised in the Pacific: Only 21 of 57 Pacific-raised salmon (including all 9 from Washington) have a component 2 score less than 5, while all Atlantic-raised salmon do.

Finer distinctions can be made with less important principal components; these components may not account for much overall variability, but they seem to account for substantial portions of the variability within subsets of the data. For example, farmed salmon from Chile, Washington, and British Columbia generally fall into three different clusters when components 3 and 4 are plotted against each other (Figure S2); the different regions in Northern Europe can also be separated from each other with components 5 and 8 (Figure S3); and components 6 and 7 can be used to reveal differences among wild salmon species (Figure S4).

The meaning of the differences shown in Figure 3 is revealed by the PCB principal component loadings (Figure 4). Positive loading values mean that above average values for the particular congener for a particular sample will contribute toward a positive principal component score, while negative loadings work in the opposite manner. Thus, Figure 4 shows that component 1 is related to the distribution between PCBs with four or fewer chlorines (positive loadings) and PCBs with six or more chlorines (negative loadings). A sample with a large proportion of PCBs with fewer than four chlorines will therefore have a positive score (such as the wild salmon), while a sample with a PCB profile with a large

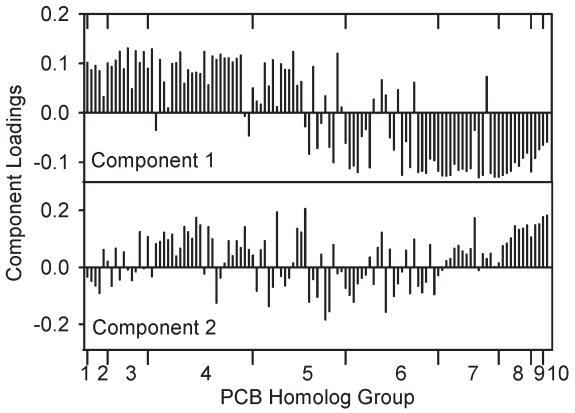

FIGURE 4. Component loadings for principal components 1 and 2.

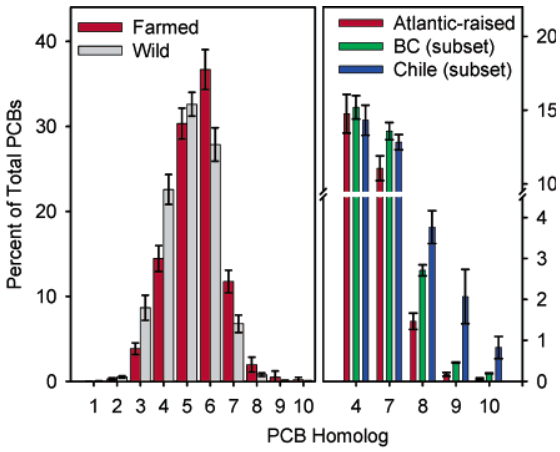

FIGURE 5. PCB homologue profiles for (left) all farmed Atlantic salmon and wild Pacific salmon excluding Oregon and British Columbia chinook and (right) all Atlantic-raised farmed Atlantic salmon, British Columbia farmed Atlantic salmon with component 2 score > 0 (12 of 18 samples), and Chile farmed Atlantic salmon with component 2 score > 0 (24 of 30 samples).

proportion of highly chlorinated PCBs will have a negative score. Figure 5 shows the average PCB homologue profiles for farmed and wild salmon; it is clear that the PCB distribution in farmed salmon is shifted to more chlorinated homologues compared to that of the wild salmon.

The component loadings can also explain the difference between the salmon farmed in the Atlantic and many of those farmed in the Pacific (Figure 3). Figure 4 shows a strong positive contribution to component 2 from the hepta- through the decachlorobiphenyls and the tetrachlorobiphenyls. Most Pacific-grown salmon in this study (with positive component 2 scores) have a congener profile more heavily weighted toward these PCBs. The difference in component 1 scores for the British Columbia and Chile salmon with similar high component 2 scores reflects a difference in the relative proportion of tetrachlorobiphenyls (with a positive contribution to component 1) and hepta- through decachlorobiphenyls (which have a negative contribution to component 1). Figure 5 shows this effect. Notice that the British Columbia and especially the Chilean salmon have elevated levels of the Cl_8 to Cl_{10} homologues relative to the Atlantic-raised salmon.

The difference between the PCB profiles of farmed and most wild salmon may be explained by the composition of their diet. The farmed salmon in this study were fed formulations made from fish such as mackerel, herring, and anchovy (*23*). Several studies have shown that herring or anchovy have a greater proportion of the more highly

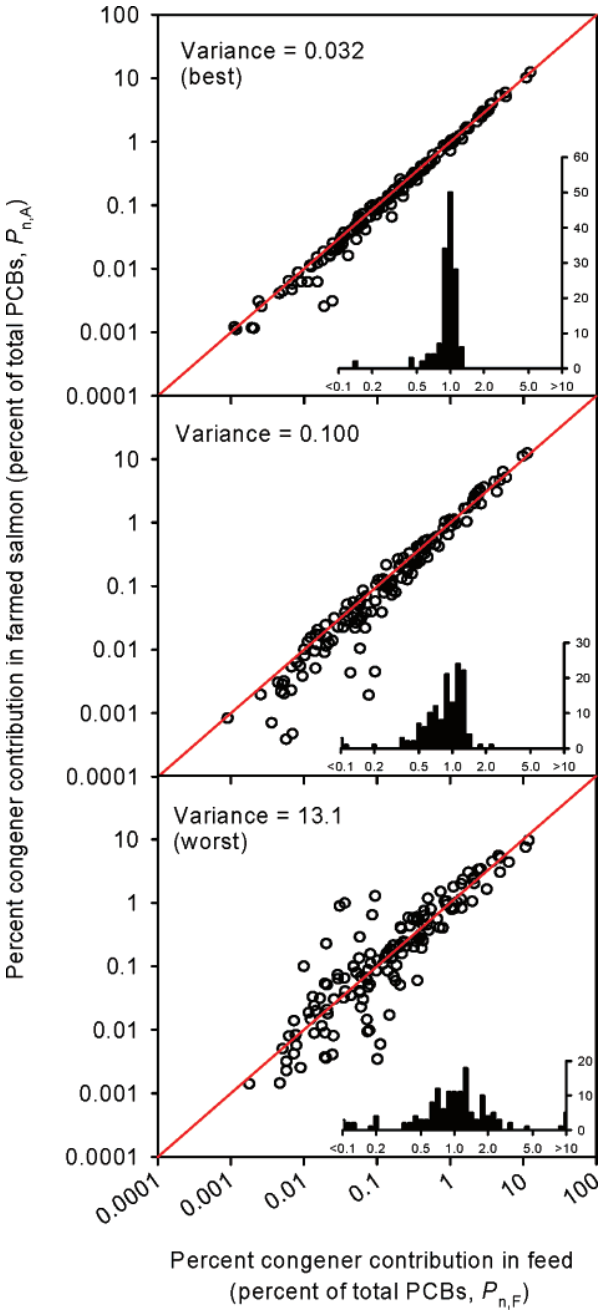

FIGURE 6. Percent congener contributions in farmed fish ($\overline{P_{n,A}}$) versus feed ($P_{n,F}$) for three farmed fish/feed pairs. The red line indicates a 1:1 relationship (not a regression) where all points would fall for a hypothetical perfect match between the PCB profiles of fish and feed. The ratio $R_{n,A/F}$ (ideally 1) was calculated for ~140 congeners with detections in each of the three cases, and the values of this ratio are plotted in the histograms shown as insets. (Note the logarithmic scale on the *x*-axes). The variance of $R_{n,A/F}$ in each of the three cases is included on the plot.

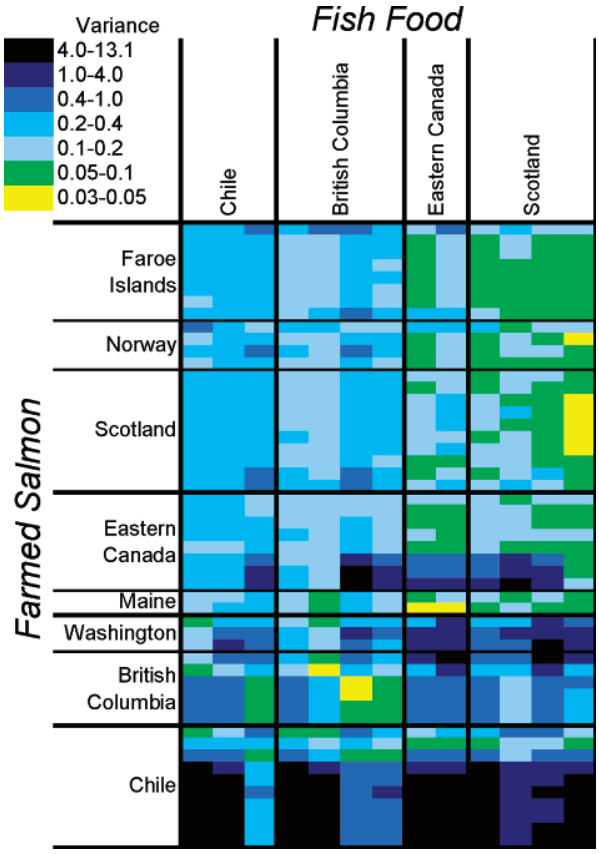

FIGURE 7. Variance of $R_{n,A/F}$ for each possible fish/food pairing. As a reference, the maximum variance of farmed salmon compared to salmon from the same farm is 0.05; the minimum variance of wild salmon compared to food they did not eat is 0.10. Variance for a perfect match would be zero.

chlorinated PCBs than zooplankton from the same waters, although in two of the three cases a greater proportion of a less chlorinated homologue group can also be seen (*24*−*26*). Wild salmon, however, do not feed exclusively on fish but also consume organisms from lower trophic levels such as euphausiids and crab larvae (*16*), which may explain the shift in PCB profiles toward less chlorinated congeners for most wild salmon compared to farmed salmon.

Relationship between PCBs in Salmon and in Their Food. Figure 6 gives three example plots showing the percent of total PCBs for all congeners in salmon from one farm versus one food sample, $\overline{P_{n,A}}$ vs $P_{n,F}$. Each data point represents a given congener; note the logarithmic scales. The line indicates a one-to-one ratio; if the PCB congener profiles were exactly the same in the salmon and food samples, then all points would fall on this line. In some cases, the vast majority of the points fall very close to this line, so that we believe the salmon must have eaten that food or food very similar to it. Many other times there was a poor match between salmon and food.

To determine what constituted a good match between a salmon sample and a food sample, we looked at the congener percent ratio, $R_{n,A/F}$ (eq 4). This exercise resulted in ~140 congener percent ratios (depending on the number of nondetects) for each of 663 farmed fish/food pairs. For a perfect match between fish and food congener profiles, the congener percent ratio would be one for all congeners, and the variance of $R_{n,A/F}$ would be zero. The calculated variance for each of the farmed fish/food pairs ranged from 0.032 to 13.1; the fish/food pairs with these extreme values are shown in Figure 6, top and bottom panels.

For comparison, we looked at the congener percent ratios of wild Pacific salmon/food pairs as well, $R_{n,W/F}$. We know that the adult wild salmon did not eat the commercial food preparations, although a certain proportion of their diet may have consisted of the same types of fish used in the commercial food. Not only that, but when the PCA was repeated including feed samples, the feed samples had component 1 and 2 scores that were similar to those of the farmed Atlantic salmon but different from those of the wild Pacific salmon (except chinook from British Columbia and Oregon). The variance of $R_{n,W/F}$ ranged from just above 0.10 (for, not

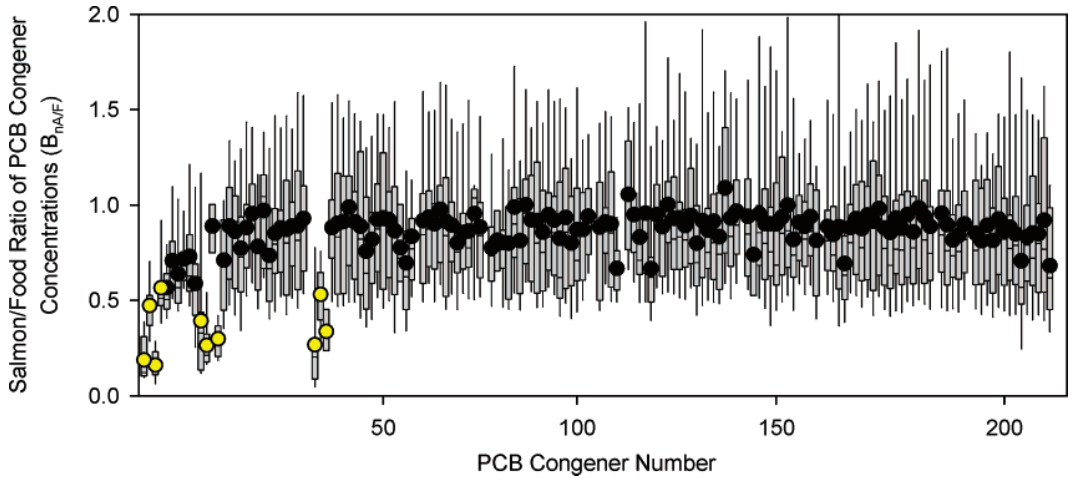

FIGURE 8. Ratios of PCB congener concentrations in salmon and commercial feed for 11 fish/feed pairs with matching PCB profiles. The range of values is indicated by 10th, 25th, 50th, 75th, and 90th percentiles; averages are shown by circles. Yellow circles indicate the 10 congeners with consistently lower values.

surprisingly, Oregon and British Columbia chinook and many of the foods) to 6.4. We therefore suggest that if the variance of the congener percent ratios for a farmed fish/food pair is greater than 0.10, then it should not be considered a good match. Figure 6 also shows the farmed fish/food pair with the variance of congener percent ratios closest to 0.10; from inspection, this pair indeed appears to have similar congener profiles, but it does not seem to be a very close match.

We also calculated the variance of congener percent ratios for each possible farmed fish/farmed fish pair within a farm ($R_{n,A/A}$), which we assume would have been fed the same food. If all of the fish were identical in every aspect of feeding, growth, and metabolism, and the sample preparation and analytical measurement were perfect, then the variance should be zero for all farmed fish/farmed fish pairs. What we found was an average variance of $R_{n,A/A}$ of 0.012, with a maximum of 0.049. That the values were not all zero indicates the degree of variability between fish from the same farm and slight inconsistencies and errors in processing and measurement.

On the basis of the variances of the farmed fish/farmed fish and wild salmon/food congener percent ratios, we can now categorize farmed fish/food pairs as having a good PCB congener profile match if the variance of congener percent ratios is less than 0.05 or having a poor match if it is more than 0.10. These divisions, along with additional divisions at natural breaks, were used to categorize the variances of farmed fish/food ratios for Figure 7. Many salmon/feed pairings showed poor agreements, which is not surprising considering most of the pairings relate fish raised in one part of the world to food purchased in another. However, most salmon farms had a congener profile similar to at least one feed sample, meaning a variance of congener percent ratios less than 0.10 but more than 0.05 (green in Figure 7), which falls between our well-defined zones. Ten had very good matches with one or two food samples (variance less than 0.05, yellow in Figure 7, 11 matches total); in all of these cases, both salmon and feed were from the same region.

The observation of a close correspondence between relative PCB concentrations in the food and salmon (as seen in Figure 6) implies that the farmed salmon are not sequestering significant amounts of PCBs from the ocean water relative to total PCBs. If that were the case, then the data would be offset or skewed from the 1:1 line because the water would have relatively higher concentrations of the less chlorinated (more water soluble) congeners than the feed. Even in those cases where there was not a close correspondence between the farmed salmon and any feed sample from the region (mainly the farmed salmon from Chile), we still did not see a pattern that we would have expected to result from substantial contamination from the water. In these cases, we believe it is more likely that we simply did not happen to sample the feed used at these particular Chilean salmon farms or these salmon were fed combinations of several feeds.

For the fish from the 11 farms with a variance of congener percent ratio <0.05 when compared to one or more feed samples, plots of PCB congener percents in fish versus food are shown in Figure S5. The ratio of congener *concentrations* ($B_{n,A/F}$) in the salmon compared to the food is plotted in Figure 8. We note that the $B_{n,A/F}$ is not necessarily the same as a bioaccumulation factor, because our samples did not include the whole fish. The concentrations in the whole fish may be more or less than the skin-on fillets that we analyzed. The total PCB concentrations in fillets without skin, for example, are about 80% of the concentration in the whole fish for tank-raised Atlantic salmon (*27*, *28*).

The vast majority of congeners have average $B_{n,A/F}$ values that fall within a narrow range of 0.8−1.0. This ratio does not increase with increasing congener number, although properties such as octanol/water partitioning coefficients are greater for the more highly chlorinated PCBs. Some congeners in our study, mostly with a lower molecular weight, have much lower ratios, often less than 0.5. The 10 congeners with the lowest ratio are shown in Figure 8 in yellow; they are PCB 1, 2, 3, 4, 11, 12/13, 15, 36, 37, and 38. These congeners have substantially different behavior. This could be explained by an enhanced elimination pathway (such as biodegradation), an inhibited uptake pathway, or sequestration in a part of the salmon not sampled.

The congener concentration ratios, $B_{n,A/F}$, shown in Figure 8 are similar to an assimilation efficiency factor. A gross assimilation efficiency can be calculated by multiplying this fish/food concentration ratio by the amount of mass that the salmon adds from a given amount of food (*29*), which in the case of 0−30-week-old farmed Atlantic salmon is about 1 kg of body mass produced for every 1.2 kg of feed consumed (*28*). Therefore, if whole body concentrations were similar to the skin-on fillets sampled in this study, then approximately 75% of PCBs consumed by the salmon remained in their bodies for most congeners. This is similar to the range calculated for tank-raised Atlantic salmon of approximately 80% for total PCBs (*27*). Lake Michigan coho and chinook salmon, for comparison, show a net trophic transfer efficiency of PCBs from prey to predator of around 40−60% (*29*, *30*). In both the Atlantic salmon study and a study of Lake

Michigan coho salmon, no clear differences in assimilation efficiencies were observed with increasing chlorination (*27*, *31*), in agreement with Figure 8. The 10 congeners shown in Figure 8 in yellow were not investigated in either study.

Implications for Aquaculture. Figure 8 shows that a large percentage of PCBs in salmon feed likely ends up in Atlantic salmon under farming conditions. Additionally, uptake from the ocean water does not appear to be significant. This means that PCB concentrations in the farmed Atlantic salmon can be decreased by reducing the concentrations in the feed. Because efforts by the aquaculture industry are already underway to reformulate the feed with this goal in mind (*9*, *28*, *32*), we expect concentrations of PCBs in farmed salmon and the associated health risks to the consumer should soon decrease significantly. Future studies will hopefully confirm the success of this strategy.

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Supporting Information Available

Congeners included in the analysis, plots of additional component scores, and a figure showing the 11 plots of PCB congener concentrations in salmon versus food in cases considered to be good matches. This material is available free of charge via the Internet at http://pubs.acs.org.

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