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PCBs, PCDD/Fs, and Organochlorine Pesticides in Farmed Atlantic Salmon from Maine, Eastern Canada, and Norway, and Wild Salmon from Alaska

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Farmed Atlantic salmon (*Salmo salar*) from Maine and eastern Canada, wild Alaskan Chinook salmon (*Oncorhynchus tshawytscha*), and organically farmed Norwegian salmon samples were analyzed for the presence of polychlorinated biphenyls (PCBs), dioxin-like PCBs, polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzo-*p*-furans (PCDFs), and chlorinated pesticides. PCDD and PCDF congeners were not detected in >80% of the samples analyzed. Total PCB concentrations (7.2–29.5 ng/g, wet weight, ww) in the farmed salmon were significantly higher than those in the wild Alaskan Chinook samples (3.9–8.1 ng/g, ww). Concentrations of PCBs, WHO PCB TEQs, and chlorinated pesticides varied significantly by region. PCB and WHO PCB TEQ concentrations in farmed salmon from eastern Canada were lower than those reported in samples collected two years earlier, possibly reflecting recent industry efforts to lower contaminant concentrations in feed. Organically farmed Norwegian salmon had the highest concentrations of PCBs (mean: 27 ng/g, ww) and WHO PCB TEQs (2.85 pg/g,ww); their TEQ values are in the higher range of those reported in farmed salmon from around the world. Removal of skin from salmon fillets resulted in highly variable reductions of lipids and contaminants, and in some skin-off samples, contaminant levels were higher, suggesting that skin removal does not protect the consumer from health risks associated with consumption of farmed salmon.

Introduction

Between 1987 and 2000, consumption of salmon in the United States increased by more than 26% annually; currently, more than half of the salmon consumed globally is farmed (*1*). An estimated 23.1 million U.S. residents eat salmon (primarily Atlantic salmon, *Salmo salar*) more often than once a month, 1.3 million eat salmon at least once a week, and 180,000 eat salmon more often than twice a week (*2*). Over the past two decades, the annual production of farmed salmon has increased dramatically around the world. In the United States, production has largely centered in Maine, where the aquaculture industry tripled between 1990 and 2000 (*3*); Maine now supplies ~18% of U.S. domestic consumption of farmed salmon. Virtually 100% of Maine farmed salmon is marketed in New England along with farmed salmon from other regions but predominantly from Canada and Chile. In recent years, organically grown salmon has also become increasingly available at higher prices to consumers in this region.

Recent studies have shown that farmed salmon contains elevated concentrations of persistent organic pollutants (POPs) including polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), and furans (PCDFs), and chlorinated pesticides due to dietary input from fish oil and meals (*4*–*9*). Fish oil is a byproduct of the fish meal manufacturing industry and is derived from forage stocks (small pelagic fishes) in many different parts of the world. As lipophilic POPs bioaccumulate in ocean food chains, the oil extracted from fish caught in polluted waters can be contaminated with complex mixtures of POPs (*8*). Hites et al. (*5*) reported that POP concentrations were higher in farm-raised salmon than in most wild salmon, and concentrations in farmed salmon from northern Europe were higher than those in farmed salmon from North and South Americas. In view of the trend of rising farmed salmon consumption in the United States, its contribution of toxic POPs to human body burdens is of concern, especially for heavy consumers and for children, who may be more vulnerable to POP-related effects (*10*–*12*).

Many people eat salmon regularly because of the health and nutritional benefits associated with the intake of omega-3 fatty acids present in salmon. However, results of a quantitative benefit-risk analysis for farmed and wild salmon presented by Foran et al. (*13*) suggest that these benefits, primarily the possible prevention of sudden cardiac death, may be outweighed by health risks of consuming contaminated farmed salmon. This comprehensive analysis revealed that exposure to dioxins and dioxin-like compounds associated with even modest consumption of farmed salmon poses elevated cancer and noncancer health risks including neurodevelopmental, immune, and endocrine-disrupting effects that may occur at lower concentrations (*14*). Previously, Hites et al. (*5*) concluded that potential health risks (based on a quantitative cancer risk assessment) resulting from consumption of farmed salmon contaminated with PCBs, dioxins, toxaphene, and dieldrin exceeded risks of exposure to these contaminants in wild salmon, and triggered stringent consumption advice for farmed salmon. However, these approaches must be considered conservative estimates of human health risks, as they do not take into account the cumulative risks of exposure to complex mixtures of toxic POPs and inorganic compounds that may be present in farmed salmon.

Industry efforts are under development to reduce POP levels in reformulated fish feed using vegetable oils, or a combination of vegetable and fish oils, thereby lowering the levels in farmed salmon and diminishing health risks while retaining the nutritional benefits of omega-3 fatty acids (*15, 16*). Another approach is the application of organic methods in salmon farming. Organically farmed salmon are typically

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fed diets containing lower levels of fish oil and are assumed to have lower POP concentrations than conventionally farmed salmon. Since lipophilic contaminants are stored in fat, removal of skin and subcutaneous fat from the edible portion of the flesh and some cooking procedures have also been suggested as a means to reduce POP exposure, although studies to date (*17*−*20*) have generated inconsistent results. Foran et al. (*13*) recently examined available data on this issue and found that contaminant reduction is highly variable within species, among species, and among contaminants.

Here we provide results on the analysis of PCBs, dioxin-like PCBs, PCDD/Fs, dichlorodiphenyl trichloroethane (DDT) and its metabolites, chlordane-related compounds (CHLs), hexachlorocyclohexane isomers (HCHs), and hexachlorobenzene (HCB) in farmed and wild salmon marketed to consumers in the northeastern United States, including conventionally farmed Atlantic salmon from Maine and eastern Canada, organically farmed salmon from Norway, and wild-caught Chinook salmon (*Oncorhynchus tshawytscha*) from the Aleutian Islands, Alaska. Whereas other studies have compared contaminant loads in farmed salmon by region, this study examined intra-regional differences in salmon from individual farms as well as differences between conventionally farmed and organically farmed salmon. In addition, we analyzed skin-on and skin-off samples to determine to what extent skin removal contributes to reductions in contaminant concentrations and health risks.

Experimental Section

Samples. A total of 70 farmed and wild salmon were collected from wholesale and retail outlets in Maine between August 2003 and May 2004. Suppliers provided information on the origin (region and farm) of the fish. The farmed salmon represented six locations in three regions, including two farms in eastern Maine (ME1 and 2), three in eastern Canada (CAN1, 2, and 3), and a farm in Norway (NOR). Wild-caught Chinook salmon from the Aleutian Islands, Alaska (AK), were purchased from a wholesale supplier. Ten whole salmon were obtained from each farm, nine of which were randomly pooled into three composite samples of three fish each from the seven locations for a total of 21 composites. Whole body weights of the fish ranged from 4 to 5 kg and lengths ranged from 75 to 80 cm, with the exception of salmon from the ME2 which were smaller than the others (average weight and length 1.2 kg and 56 cm, respectively) (Table S1). All farmed salmon were 2.5−3 years old; the ages of the wild salmon were unknown. The whole fish were thawed, weighed, measured, and filleted to yield two boneless fillets per fish. One fillet from each fish was left intact; from the other, we removed the skin and associated fat, belly flap, and lateral line, resulting in 42 composite samples (21 skin-on, 21 skin-off). Preparation methods followed procedures used by retail suppliers to ensure that samples were replicas of those sold to consumers. The fillets from the composites were then homogenized in a high-speed processor and subdivided into smaller replicate portions of 100 g and frozen at −20 °C in borosilicate glass containers until analysis.

Analytical Methods. Organochlorine (OC) pesticides, PCBs and 2,3,7,8-substituted PCDDs (7 compounds), and PCDFs (10 compounds) were analyzed following the methods described elsewhere (*21*, *22*), with some modifications. Approximately 20 g of tissue was homogenized with anhydrous sodium sulfate and extracted in a Soxhlet apparatus for 16 h using dichloromethane and hexane (3:1; 400 mL). The extract was rotary evaporated at 40 °C to 11 mL, and an aliquot of 1 mL of the extract was used to determine lipid content by gravimetry. The remaining extract was spiked with ^{13}C-labeled PCB congeners 3, 15, 31, 52, 118, 153, 180, 194, 206, 209, (MBP-CG; Wellington Laboratories, Guelph, Ontario, Canada), ^{13}C-labeled coplanar PCB congeners 77, 81, 126 and 169 (MBP-MXF; Wellington Laboratories), and ^{13}C-labeled PCDD/F congeners (2378-TCDD/F, 12378PeCDD/F, 123678HxCDD/F, 1234678-HpCDD/F and OCDD) as internal standards (EDF-4053; Cambridge Isotope Laboratories, Andover, MA). PCB congener 30 (2,4,6-triCB) was spiked as a surrogate standard. Further details are provided in the Supporting Information.

For the determination of OC pesticides, an Agilent 6890N gas chromatograph equipped with ^{63}Ni-ECD was used. OC pesticide concentrations were calculated from the peak area of the sample to the corresponding external standard. For the determination of PCBs, extracts were injected into a gas chromatograph (Hewlett-Packed 6890) coupled with a mass selective detector (Hewlett-Packed, series 5973) operating in an electron impact (70 eV) selected ion monitoring (SIM) mode. Quantification of PCB congeners was based on external calibration standards containing known concentrations of di- through deca-CB congeners. Concentrations of individually resolved peaks of PCB isomers were summed to obtain total PCB concentrations. Quantification of 2,3,7,8-substituted PCDD/F congeners and four non-ortho coplanar PCBs (congeners 77, 81, 126, and 169) was carried out using a high-resolution GC coupled with a high-resolution MS (HRGC−HRMS; HP 6890GC with JEOL JMS-700D HRMS) and analyzed at a resolution of $R > 10\,000$ with SIM at the most intense ions of the molecular ion cluster.

PCDD and PCDF congeners were not found in >80% of the samples analyzed at a detection limit of 1 pg/g, ww, and therefore were excluded from this analysis. TEQs were calculated for seven dioxin-like PCBs (77, 81, 126, 169, 105, 118, and 189) using the most recent international toxic equivalent factors (WHO-TEFs) (*23*). Results are given as sum of the WHO PCB TEQs (pg/g, ww). In cases where congeners were reported as nondetects, TEQ values were calculated by treating the result as if half the detection limit.

Quality Assurance/Quality Control. QA/QC procedures are detailed in the Supporting Information.

Statistical Analysis. Statistical analyses were conducted with SPSS version 13.0 (Chicago, IL). Details are provided in the Supporting Information.

Results and Discussion

Differences in Concentrations. Total PCB concentrations were significantly higher in the farmed salmon samples (as a group) than in the wild Alaskan Chinook samples ($t_{19} = 2.8$, $p = 0.01$), with values ranging from 7.2 to 29.5 ng/g, ww, in the farmed salmon and 3.9−8.1 ng/g, ww, in the wild salmon. This difference in PCB concentrations between farmed and wild salmon is consistent with results of other recent studies (*5*, *6*, *8*, *24*). Figure 1a shows concentrations of total PCBs, DDTs, CHLs, HCHs, and HCB in farmed and wild salmon by region. POP concentrations in the Maine and Canadian samples were similar to each other, but significant differences were found in concentrations of total PCBs ($X^2 = 12.2$, $p < 0.01$), DDTs ($F_{3,20} = 7.6$, $p < 0.01$), and HCHs ($X^2 = 9.0$, $p < 0.05$) between the Norwegian samples, Maine and Canadian samples, and the Alaskan wild samples. The highest POP concentrations were found in organically grown salmon from Norway, with the exception of HCHs, which were lowest in the Norwegian samples. Alaskan salmon had the lowest concentrations of POPs with the exception that their HCH concentrations exceeded those in the samples from Norway. The higher concentrations of HCHs in the wild Alaskan Chinook salmon are likely a reflection of the long-range atmospheric transport of these compounds to polar regions and their ubiquity in Arctic marine food chains (*25*).

However, as Figure 1 illustrates, POP distributions were highly variable among salmon from individual producers. Comparing the two Maine producers, salmon from ME1 had higher concentrations of total chlordanes ($t_4 = 3.1$, $p < 0.05$)

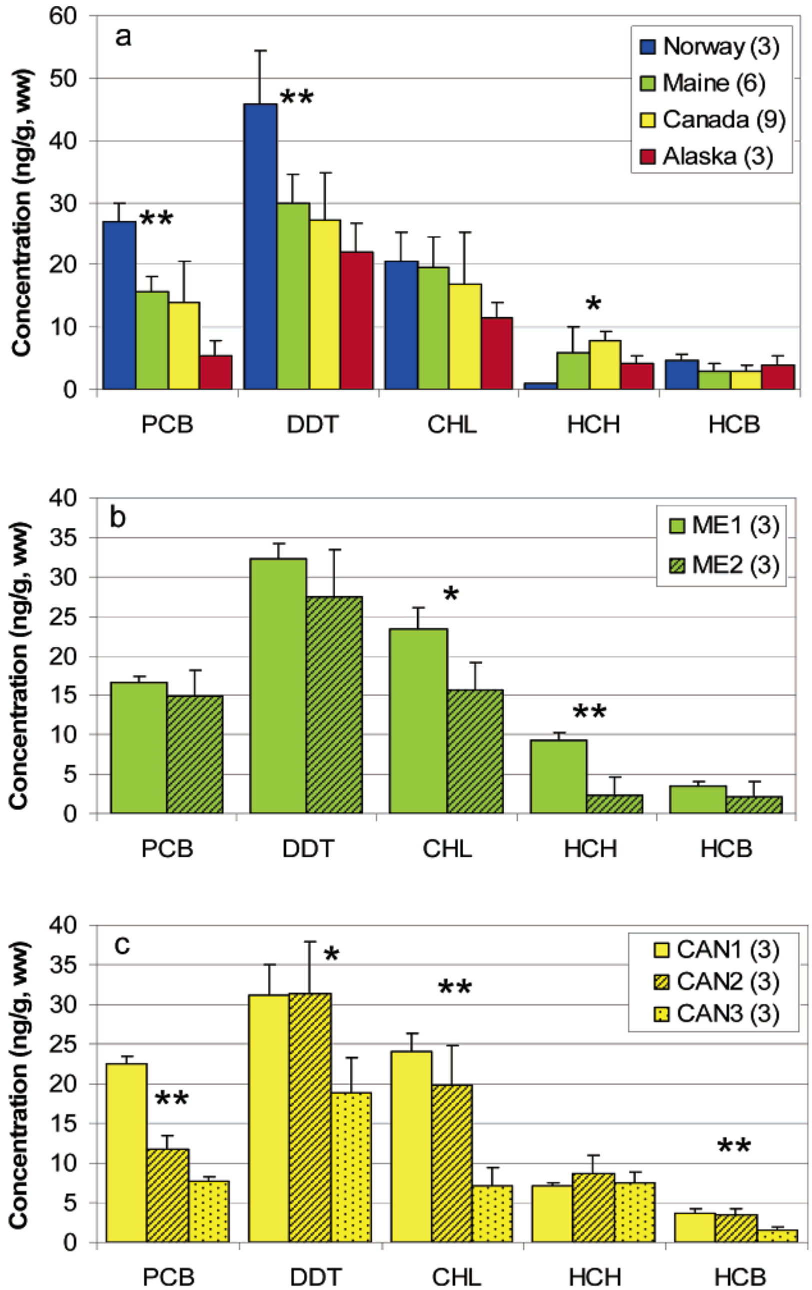

FIGURE 1. Skin-on concentrations of major POPs in farmed salmon from Norway, Maine, and Canada, and wild salmon from Alaska (a) and in farmed salmon from the Maine (b) and Canadian (c) producers. All error bars are one standard deviation. Significance of main effect is indicated by asterisks (* $p < 0.05$, ** $p < 0.01$).

and HCHs ($t_4 = 4.8$, $p = 0.01$) than those from ME2 (Figure 1b). Comparing the three Canadian producers, there were significant differences in DDTs ($F_{2,8} = 6.2$, $p < 0.05$), total PCBs ($F_{2,8} = 122$, $p < 0.001$), total chlordanes ($F_{2,8} = 20$, $p < 0.01$), and HCB ($F_{2,8} = 13$, $p < 0.01$) (Figure 1c). In general, salmon from the CAN1 and CAN2 farms had significantly higher POP concentrations than those from the CAN3 farm. Interestingly, salmon from the CAN1 and CAN3 farms were fed commercial feed from the same supplier.

Regional and inter-sample variability was observed for total WHO-TEQ values of dioxin-like PCBs in our farmed salmon samples. Total WHO−PCB TEQs were highest in the Norwegian organic samples, ranging from 1.2 to 5.9 (mean 2.85) pg/g, ww, and lowest in the wild Alaskan samples, ranging from 0.09 to 0.22 (mean 0.16) pg/g, ww. WHO−PCB TEQ values in the Maine and eastern Canadian samples were similar (mean 0.57 and 0.66 pg/g, ww, respectively). Among the Canadian farms, salmon from CAN1 consistently had the highest TEQ values. No differences were observed in concentrations between the two Maine farms. As expected, penta-CB 126 dominated the congener profiles, contributing 64−86% of the total TEQ. PCB 81 was not detected in >60% of our samples, and CB 169 was not detected in the wild Alaskan salmon.

Differences in PCB Congener Profiles. Principal component analysis (PCA) was used to further explore differences in the relative abundance of 45 PCB congeners between farmed and wild salmon and differences among the regions. Twelve components with eigenvalues greater than one were retained; together they accounted for 90.7% of the variability in the dataset. Figure 2 shows a plot of components 1 (accounting for 28.3% of the variability) and 2 (accounting for 11.9% of the variability) for all farmed and wild salmon. These two components separate farmed and wild salmon

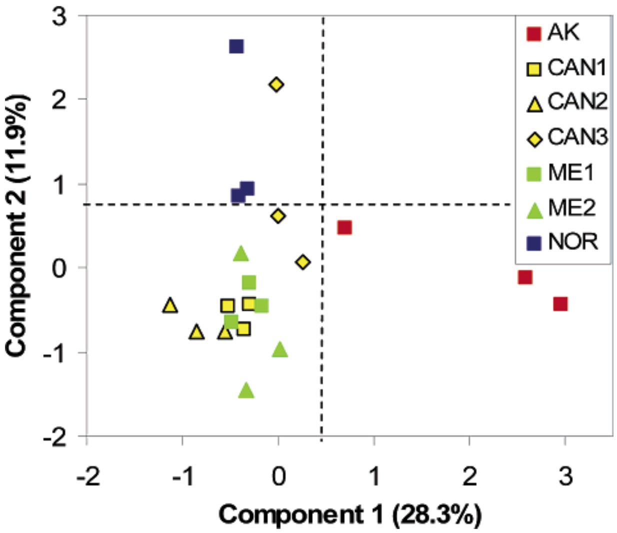

FIGURE 2. Plot of principal components 1 vs 2 for all skin-on salmon samples. The amount of variability accounted for by the components is in parentheses. AK = wild Alaskan Chinook salmon; CAN = farmed salmon from eastern Canada; ME = farmed salmon from Maine; NOR = organically farmed salmon from Norway.

and the regions quite well. The greatest differences (shown by component 1) are between the wild Alaskan Chinook salmon and the farmed salmon from all other regions. The wild salmon have positive scores while the farmed salmon have primarily negative scores for component 1. This variability would be expected, given the differences in the life history and diet between the two groups. Component 2 highlights differences between the farmed Norwegian samples and those from North America with the exception of a single CAN3 sample. Maine and eastern Canadian salmon had overlapping scores for all components.

To interpret the meaning of the components, we looked at the component loading scores by PCB homologue group (Figure 3). Component 1 is positively correlated with the tri-, tetra-, and octachlorobiphenyls and is negatively correlated with the heptachlorobiphenyls. Therefore, the wild Alaskan salmon samples (positive score for component 1) have relatively high proportions of lower chlorinated PCBs, while the farmed salmon samples (negative score for component 1) exhibit a PCB profile with a large proportion of higher chlorinated PCBs. A comparison of PCB homologue profiles for farmed and wild salmon (Figure S1) reiterates this pattern, reflecting differences in diet and exposure pathways between farmed and wild salmon. For farm-raised salmon, PCB contamination comes directly from fish oils and meals in feed derived from forage fish, while wild salmon are exposed to PCBs in their prey, which include not only fish but also lower trophic level organisms, reflecting atmospheric sources of contaminants that biomagnify through the food chain (*26*). Carlson and Hites (*4*) examined differences in PCB profiles between farmed and wild salmon from around the world, and reported that most wild salmon species including Alaskan Chinook have lower PCB burdens and greater proportions of the lower-chlorinated congeners than farmed salmon.

The congeners with the strongest loadings on component 2 are two coplanar PCBs, 77 and 126 (positive), and CB 99 (negative) (Figure 3). The Norwegian salmon and a sample from the CAN3 farm had higher component 2 scores than the other North American salmon. As would be expected, these samples have higher relative proportions of PCB 77 and 126 and lower relative proportions of CB 99 than the other salmon (Figure S2). Penta-CB 126, the most toxic dioxin-like congener, acts via *Ah* receptor-mediated mechanisms to elicit immune suppression, cancer, and other toxic effects in a wide range of species (*27, 28*); thus, the higher abundance of this congener in the Norwegian and CAN3 samples may reflect a greater proportion of dioxin toxic potency in these samples than in the other salmon in our study.

Effect of Skin Removal. This study examined the possible benefit to the consumer of skin and subdermal fat removal from the edible portion of the salmon. In skin-on samples, the lipid content varied significantly by region and among producers within the regions [$F_{6,14} = 23.7$, $p < 0.001$]. Lipids were highest (>18%) in salmon from Norway and the ME1 farm, and lowest (7−8%) in the wild Alaskan salmon and the smaller salmon from the ME2 farm. Removal of skin from these samples (as a group) significantly reduced the levels of lipids ($t_{20} = 6.3$, $p < 0.001$), total PCBs ($t_{20} = 4.1$, $p = 0.001$),

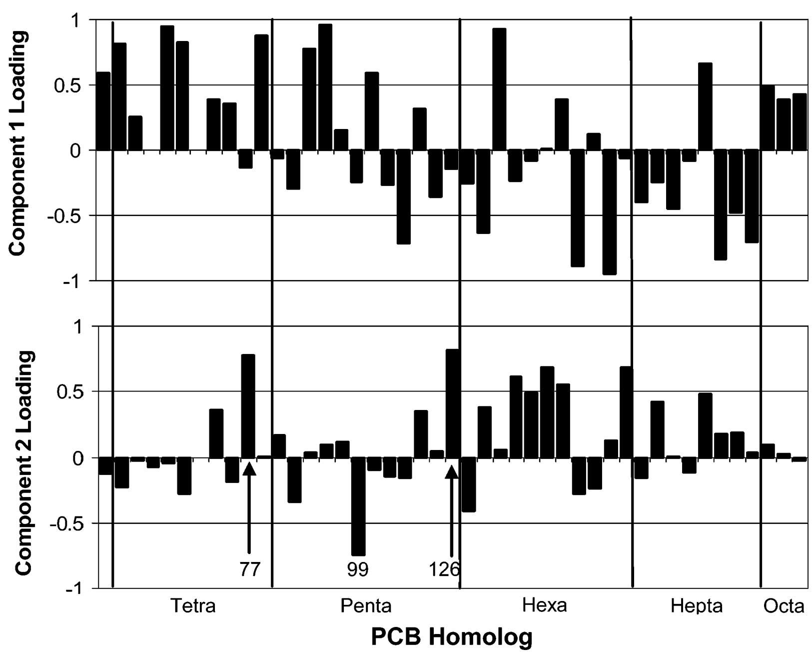

FIGURE 3. Component loading scores for principal components 1 and 2 from a principal components analysis of the relative abundance of 45 PCB congeners found in skin-on salmon samples.

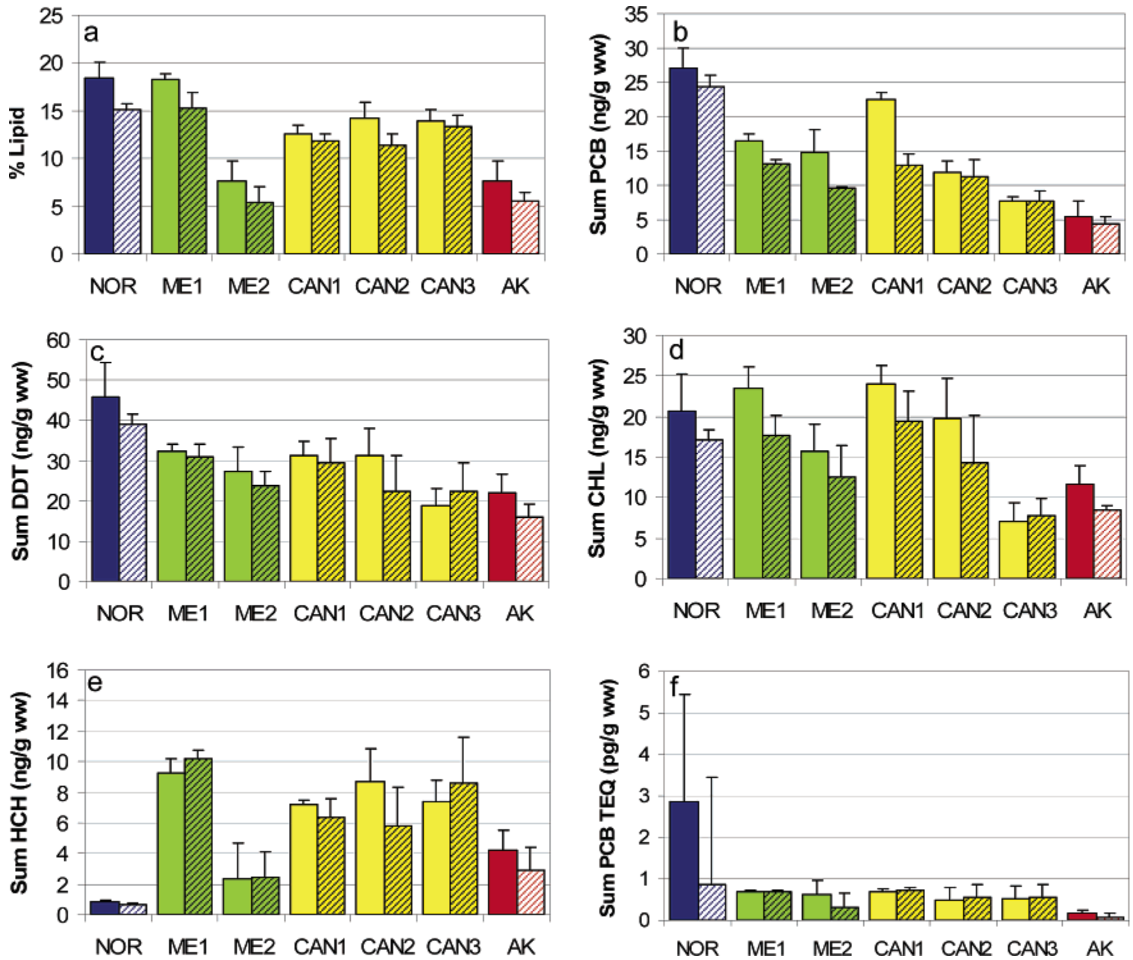

FIGURE 4. Comparison of mean concentrations of lipids (a), total PCBs (b), total DDTs (c), total chlordanes (d), total HCHs (e), and total PCB TEQ (f) in skin-on and skin-off samples of farmed and wild salmon by producer. AK = wild Alaskan Chinook salmon; CAN = farmed salmon from eastern Canada; ME = farmed salmon from Maine; NOR = organically farmed salmon from Norway. Solid bars are skin-on samples and striped bars are skin-off samples. All error bars are one standard deviation.

total DDTs ($t_{20} = 2.7$, $p < 0.05$), and total CHLs ($t_{20} = 5.0$, $p < 0.001$), but not HCHs or HCB. However, when compared by region and producer, the reductions of lipids and POPs were highly variable, and ranged from 26−30% for wild Alaskan salmon and the smaller (leaner) samples from the ME2 farm to ~5% for samples from two Canadian farms (Figure 4).

Lipid content and distribution between and within tissues in fish vary from species to species and among individual fish, and are strongly influenced by diet (*29*). In Atlantic salmon, lipids are stored largely in the myosepta (connective tissue sheets at the termination of muscle fibers) of the flesh (white muscle layer) where the largest numbers of adipocytes are found. The exception is lean fish which may store a greater proportion of lipids subdermally. Thus, the variability in lipid reduction in our samples may reflect the fact that skin and subdermal fat only account for a small percentage of the total lipid content in farmed salmon. Reductions in PCBs were also variable, ranging from 1−3% in salmon from two Canadian farms to 34−43% in salmon from the ME2 and CAN1 farms, respectively. A reduction in PCB concentrations resulting from skin removal has been reported for commonly consumed fish including wild Chinook salmon (*17*), herring (*30*), carp (*18*), and brown trout (*19, 20*). However, in contrast to other studies (*20, 31*), skin removal from our samples did not consistently result in a reduction of all POPs, and the reductions of chlorinated pesticides and WHO PCB TEQs were highly variable (Figure 4). In some cases, reductions were negligible or concentrations were slightly higher in skin-off samples, particularly in salmon from Maine and Canadian farms. Whereas total WHO PCB TEQs in skin-off samples were 2- to 3-fold lower in the Norwegian and Alaskan salmon, no reductions were found in salmon from the ME1 and all Canadian farms.

These observations suggest that POPs may be preferentially bound to accumulated lipids in the flesh of salmon as a result of rapid growth farming practices using high fish oil diets, especially in older fish. It has been observed that immature fish are fed lower levels of oils, and tend to store lower levels of lipid in the flesh compared with mature fish (*17*). Moreover, there can be a marked variation in flesh lipid content within fish fed the same percentage of dietary oil, as certain individuals utilize high-energy diets but deposit little lipid in their flesh while others tend toward greater adiposity. Bell et al. (*29*) reported that the percent lipid in muscle of Scottish farmed salmon fed 28% oil varied from 3% to 17%. Earlier studies (*18*−*20*) found that both the lipid content and contaminant concentrations in the edible portion of the fish flesh increases with size and age. Therefore, skin removal in older, fattier fish results in a smaller reduction in percent lipid and concentrations of POPs. The ages of the farmed salmon in our study ranged from 2.5 to 3 years old, which is typical of salmon found on the market. It is possible that removal of skin in smaller, younger fish (<2 years) would have resulted in a greater and more consistent reduction of lipophilic POPs.

POP Concentrations and Lipid Content. The major POPs that were reduced by skin removal in our study were significantly correlated with total lipid content in the skin-on samples (Figure S3). Modest positive correlations were observed between lipid content of farmed and wild salmon (combined) and concentrations of total PCBs ($R^2 = 0.53$), DDTs ($R^2 = 0.56$), and CHLs ($R^2 = 0.45$), but no correlations were found with HCHs, HCB, or WHO PCB TEQs. Previous studies have reported similar inconsistencies in relationships between lipid content of salmonid fishes (with skin-on) and organochlorine contaminant levels. Stow et al. (*32*) found modest positive PCB/lipid relationships in Lake Michigan salmonids during spawning, but not among nonspawning fish, suggesting that the mobilization of lipids and contaminant accumulation in lipid during spawning may be responsible for the positive PCB/lipid association. In this study, the influence of spawning on PCB/lipid relationships could not be evaluated, as sex and reproductive status of the fish were unknown. Most of the previous work identifying the importance of lipids in contaminant uptake have been based on aggregate comparisons across species of fish, whereas no relationships were found among individuals within a species. Carlson and Hites (*4*) reported a strong positive PCB/lipid correlation among all wild salmon species but not among farmed Atlantic salmon from different regions. Thus, while lipid may be an important repository for POPs, factors other than lipid content (e.g., diet, age, sex, reproductive status, and individual capacity for metabolism and elimination of POPs) may be driving contaminant uptake and accumulation in farmed salmon.

Interestingly, when skin was removed, stronger correlations were observed between lipid content and concentrations of most of the POPs including the dioxin-like PCBs (as WHO PCB TEQs) in our farmed and wild salmon samples (Figure S3). In skin-off samples, there were positive relationships for total PCBs ($R^2 = 0.63$), WHO PCB TEQs ($R^2 = 0.72$), DDTs ($R^2 = 0.61$), and HCHs ($R^2 = 0.43$), but not for CHLs and HCB. Although internal transport and deposition of contaminants in fish is not well understood, these findings, along with the lack of a consistent reduction in POPs in skin-off samples, suggest that body tissues besides subdermal depots harbor lipophilic contaminants in salmon.

Differences between Farmed and Organically Farmed Salmon. Although the Norwegian organically farmed salmon were fed certified organic feed lower in fish oil content (≥28% of the total diet) and were reportedly "virtually free of dioxins and PCBs resulting in lower ICES7 concentrations" (sum of ICES 7 set of PCBs: IUPAC numbers 28, 52, 101, 118, 138, 153, 180) compared with conventional farm-raised salmon, these organic samples were highest in lipid content (18.4%) and their total PCB burdens (mean 27 ng/g, ww) were significantly higher ($p < 0.01$) than those in farmed salmon from the Northeast region. Total $PCBs_{soc45}$ in our organic samples were also more than 2-fold higher than the ICES7 PCB levels reported by the most recent monitoring data for Norwegian conventionally farmed salmon (*33*). Compared with our results, the ICES7 PCB concentrations reported by the Norwegians were similar (mean ~12 ng/g, ww), but concentrations of DDT, HCB, and dioxin-like PCBs (as WHO PCB TEQs) in our organic samples were more than 2-fold higher. This suggests that measuring only the ICES7 PCBs may significantly underestimate the total PCB content (here, by a factor of 2.25), as other PCB congeners present in salmon are not measured.

Global Comparison of Concentrations. Total PCBs and WHO PCB TEQs in our farmed and wild salmon samples were compared with those reported in salmon from different regions (Figure 5). Such comparisons are limited because of the numerous sources of variability across the studies; therefore, only a few specific comparisons are discussed here to place the results in perspective with those of others. Total PCB concentrations in our farmed salmon samples from Maine (15.7 ng/g, ww) and eastern Canada (14 ng/g, ww) are 2- to 3-fold lower than those previously reported by Hites et al. (*5*) for farmed salmon from these areas. For the Maine farmed salmon, the differences disappear when the data are lipid-adjusted, while PCB concentrations in salmon from eastern Canada are still 2.5 times lower. Higher mean PCB concentrations (~ 40−50 ng/g, ww) have been reported in farmed salmon from western Canada and northern Europe (*5*−*8*). A recent analysis of farmed salmon purchased in retail outlets in Spain, Italy, Portugal, and Belgium found slightly lower PCB concentrations (33.9 ng/g, ww) in European farmed salmon, although the region of origin was unknown (*34*). On a lipid weight basis, the highest PCB concentrations were reported in farmed salmon from British Columbia (371.5 ng/g, lw; 48.3 ng/g, ww) (*6*), followed by Scotland (308 ng/g, lw; 51.1 ng/g, ww) (*7*); these levels are 2-fold higher than those in our organically farmed Norwegian samples (147.3 ng/g, lw; 27 ng/g, ww). PCB levels in our wild Alaskan Chinook samples (72.3 ng/g, lw; 5.5 ng/g, ww) were similar to those reported for Alaskan Chinook in other studies (*5, 6*).

Total WHO PCB TEQ values in our Maine and eastern Canadian samples (0.7 and 0.6 ng/g, ww, respectively) are 3−4 times lower than total TEQs of PCBs and PCDD/Fs reported by Hites et al. (*5*) in farmed salmon from northern Europe, 1.6 and 2.5 times lower than values in salmon from Maine and eastern Canada, respectively, and slightly lower than those of Chilean farmed salmon. On a lipid weight basis, the differences are no longer evident for Maine farmed salmon while TEQs in our eastern Canadian samples are still 2.3 times lower than those reported by the Hites study. However, it should be noted that the WHO TEQ values in the present study were calculated from 7 (of 12) non-*ortho* and mono-*ortho* PCBs (IUPAC numbers 77, 81, 126, 169, 105, 118, and 189) and thus underestimate the total TEQ as other isomers were present that were not measured.

The WHO PCB TEQ values of 2.85 pg/g ww in our organically farmed Norwegian samples are in the higher range of the values of 0.7−3 pg WHO TEQ, ww, reported in farmed salmon from around the world. In contrast with Karl et al. (*24*) who found higher levels of WHO TEQs in conventionally farmed salmon versus organically farmed salmon, total WHO PCB TEQ values in our Norwegian organic samples were higher than the total TEQs of PCBs and PCDD/Fs reported in conventionally farmed salmon from Norway (*5*) and 4- to 5-fold higher than the WHO PCB TEQs in our conventionally farmed salmon from North America.

Implications for the Consumer. The importance of labeling salmon as farmed and identifying the region of origin has been emphasized as a means to helping the consumer avoid unnecessary exposure to highly contaminated fish. However, given the significant intra-regional variations in concentrations in our samples, it appears that the consumer's ability to accurately predict and minimize exposure to contaminated farmed salmon through such labeling may be limited. The lower PCB and WHO TEQ concentrations in our Canadian samples compared with results of the Hites study (*5*) could reflect efforts by industry to reduce contaminant loads in salmon feed (*15*, *16*). Alternatively, it is possible that the salmon were collected from different farms using different feeding practices at different times of the year. Variability in lipid content and composition, as well as batch-to-batch variation in feed could also account for differences in levels. Removal of skin has been reported to reduce contaminant levels in fish. In our farmed salmon, skin removal resulted in highly variable reductions of lipids and POPs, and in some cases, contaminant concentrations were higher in skin-off samples. This observation is in conflict with the current paradigm that removal of skin and adipose tissue reduces

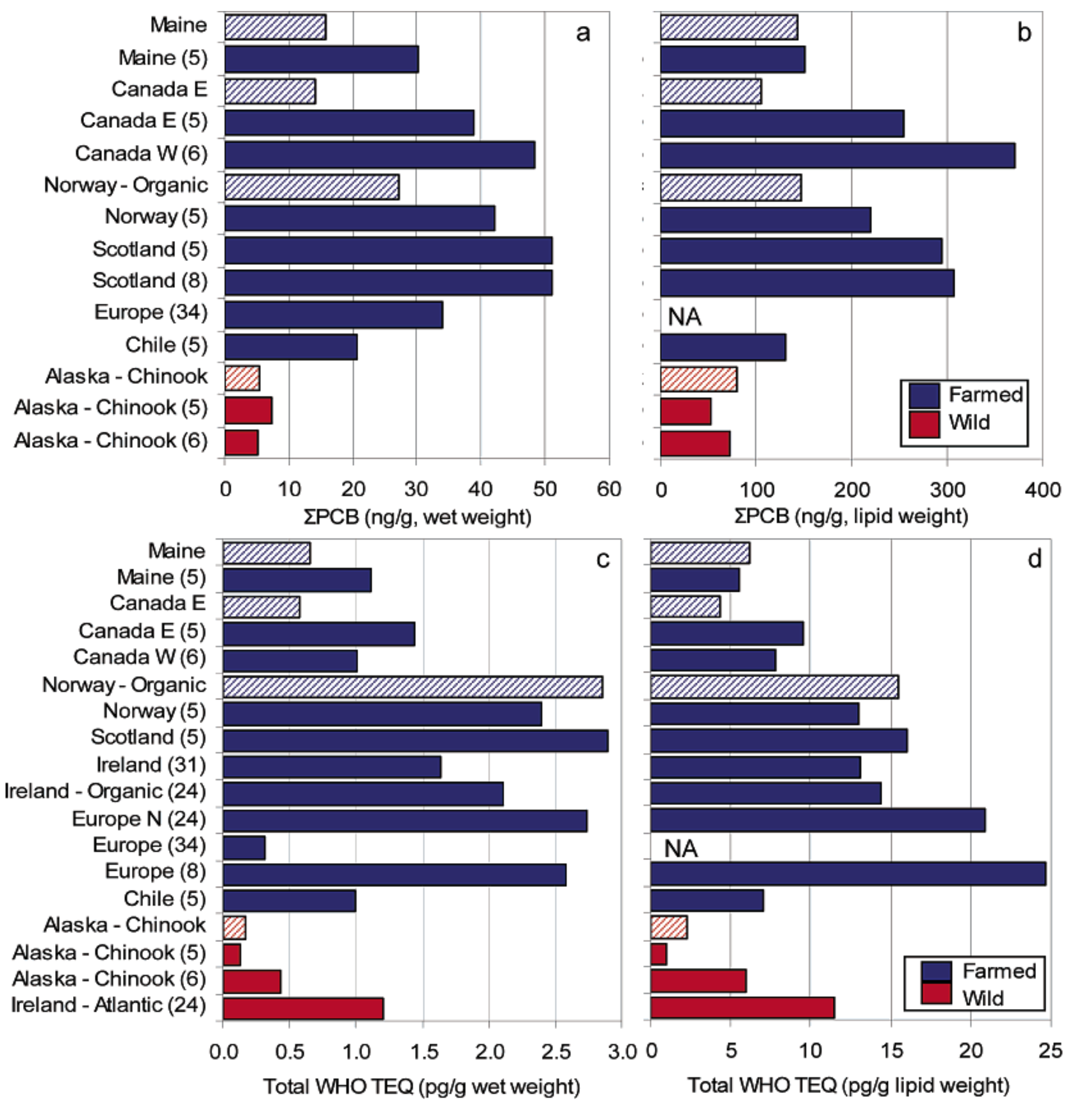

FIGURE 5. Global comparison of total PCB concentrations in wet weight (a) and lipid weight (b) and total WHO TEQ concentrations in wet weight (c) and lipid weight (d). NA = data not available. Results from the current study (in striped bars) represent total WHO TEQs for seven dioxin-like PCBs. Results from studies in refs *6* and *34* represent total WHO TEQs for 12 dioxin-like PCBs. Results of the other studies represent total WHO TEQs for 12 dioxin-like PCBs and 17 PCDD/Fs.

contaminant concentrations by 50% or more. In this study, the highest concentrations of PCBs, dioxin-like PCBs, and DDT were found in organically farmed Norwegian salmon, which poses additional issues for the consumer. Moreover, these samples were found to contain a higher proportion of penta-CB 126, suggesting that the toxicity of fish should be assessed on a congener-specific basis. These observations suggest that purchasing higher priced organically farmed salmon, even when monitoring results are provided, does not necessarily protect the consumer from toxic exposure.

In view of the increasing availability of farmed salmon in the marketplace and rising consumption rates among U.S. residents, the results of this study point to the need for a reassessment of health risks associated with human dietary exposure to farmed salmon, and for the development of consistent consumption advice among agencies for the protection of public health.

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Supporting Information Available

Details of analytical methods, QA/QC, and statistical methods: Table S1, biometric data of samples; figures of PCB homologue profiles (S1), congener profiles for 77, 126, and 99 (S2), plots of lipid content versus POPs (S3). This material is available free of charge via the Internet at http://pubs.acs.org.

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