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FLESH RESIDUE CONCENTRATIONS OF ORGANOCHLORINE PESTICIDES IN FARMED AND WILD SALMON FROM BRITISH COLUMBIA, CANADA

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Abstract—The present study reports measured levels of organochlorine pesticides (OCPs) in commercial salmon feed ($n = 8$) and farmed Atlantic, coho, and chinook salmon ($n = 110$), as well as wild coho, chinook, chum, sockeye, and pink salmon ($n = 91$). Flesh residue concentrations (ng/g wet weight) of dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexanes (HCHs), chlordanes, chlorobenzenes (CBz) and cyclodiene pesticides (e.g., dieldrin, mirex) were 2 to 11 times higher ($p < 0.05$) in farmed salmon compared with wild salmon. Concentrations were positively correlated with flesh lipid levels. Farmed Atlantic salmon (12–15% lipid) typically exhibited the greatest OCP burdens compared with other salmon species. However, when expressed on a lipid weight basis, concentrations of OCPs (ng/g lipid weight) in wild salmon, in many cases, exceeded those levels in farmed salmon. Observed interspecies and site-specific variations of OCP concentrations in farmed and wild salmon may be attributed to divergent life history, prey/feed characteristics and composition, bioenergetics, or ambient environmental concentrations. Calculated biomagnification factors ($BMF = C_F/C_D$, lipid wt) of OCPs in farmed salmon typically ranged between two and five. Biomagnification of chemicals such as DDTs, chlordanes, and mirex was anticipated, because those compounds tend to exhibit high dietary uptake and slow depuration rates in fish because of relatively high octanol–water partition coefficients (K_{OW}s $> 10^5$). Surprisingly, less hydrophobic pesticides such as hexachlorocyclohexanes and endosulfans (K_{OW}s $< 10^5$) consistently exhibited a high degree of biomagnification in farmed salmon species (BMFs > 5). This is contrary to previous laboratory and field observations demonstrating fish BMFs less than 1 for low K_{OW} chemicals, because of efficient respiratory elimination of those compounds via gills. The results suggest that ambient seawater concentrations and bioconcentration-driven accumulation may play a key role in the bioaccumulation of these relatively more water-soluble contaminants in farmed salmon. Finally, OCP exposure through consumption of British Columbian salmon is found to be low relative to United States national average per capita total exposure levels and provisional tolerable daily intakes. Environ. Toxicol. Chem. 2011;30:2456–2464. © 2011 SETAC

Keywords—Organochlorine Pesticides Salmon Bioaccumulation Human exposure

INTRODUCTION

Recent studies have documented the occurrence of persistent organic pollutants such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzo furans (PCDFs), and organochlorine pesticides (OCPs) in flesh of farmed and wild salmon [1–4]. A major focus has involved risk–benefit analyses, comparing contaminant-related human health risks, relative to health benefits associated with omega-3 highly unsaturated fatty acid (*n*-3 HUFAs) such as eicosapentaenoic acid and docosahexaenoic acid, which are abundant in oily fish such as salmon [4,5].

Although accumulation of persistent organic pollutants in wild Pacific salmon is largely the result of ambient environmental levels and food web biomagnification processes, chemical concentrations in farmed salmon are influenced mainly by concentrations in the supplied feeds [3,4]. Aquaculture operations that use commercial aquafeeds containing relatively contaminated marine fish oils can result in elevated flesh concentrations in farmed fish products [3,4,6–9]. Recent studies show that replacement of marine fish oils (MFOs) with terrestrial-based lipids such as canola oil and poultry fat can effectively reduce contaminant levels in feed, thereby lowering flesh contaminant concentrations in market-size farmed salmon products [7,8,10].

We recently reported results of a comprehensive field survey, involving trace analysis of several environmental contaminants, including PCBs, PCDDs, PCDFs, as well as Hg and other trace elements in numerous samples of farmed and wild salmon ($n = 201$) from coastal British Columbia (BC), Canada [4,11]. The survey included five species of wild Pacific salmon, including chinook (*Oncorhynchus tshawytscha*), coho (*O. kisutch*), sockeye (*O. nerka*), chum (*O. keta*), and pink (*O. gorbuscha*); three species of farmed salmon, including Atlantic (*Salmo salar*), chinook, and coho salmon; and several commercial aquafeeds.

In the present study, we report on the occurrence and levels of 31 OCPs, including those that were monitored in the aforementioned samples. The primary objective of the study was to provide a comparative analysis of OCP residue concentrations in farmed and wild salmon, similar to our previous analyses for PCBs, PCDD/Fs, and trace elements [4,11]. The bioaccumulation behavior of pesticides in farmed salmon is assessed and evaluated. Also, human dietary intake (μg/kg body wt/d) of pesticides through consumption of farmed and wild BC salmon is assessed and compared relative to available provisional tolerable daily intakes (PTDIs).

All Supplemental Data may be found in the online version of this article.

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MATERIALS AND METHODS

Samples

Figure 1 is a map of the study area (coastal BC) showing the various sampling locations. Market-size farmed salmon (Atlantic, coho, and chinook), ($n = 110$) and commercial salmon feed ($n = 8$) were collected from eight BC salmon farm sites during 2003. Wild salmon (coho, chinook, pink, chum, and sockeye; $n = 91$) were sampled across a range of sampling dates and geographical locations from coastal BC waters. All collected samples were analyzed for a suite of 31 OCPs. Details regarding sample collection, storage, and handling are reported elsewhere [4,11].

The survey was a blind study in which the identities of all farmed and wild salmon samples obtained were unknown to the analysts during the analyses. Farmed Atlantic salmon were collected from four different farms located at Broughton, Quatsino Sound, Sechelt Inlet, and Quadra Island, respectively (B, QS, SI1, QI1). Farmed chinook salmon were collected at two farm sites at Quadra Island (QI2, QI3). Farmed coho salmon were collected from two farms located at Jervis Inlet and Sechelt Inlet, respectively (JI, SI2). A total of eight samples of aquafeed were collected ($n = 8$); one representative sample from each of the farm sites fed prior to harvest. Fish were collected both from the farm site (F) and from the corresponding processing plant (PP) at time of arrival. For one source of farmed Atlantic salmon (QI1), we also collected and analyzed those fish at the end of the commercial production line (end-of-line), which represented the market-ready fillet product. Careful removal of bones is conducted in the commercial processing plants. Thus, the final market-ready salmon flesh samples from QI1 were fully processed boneless fillets. A summary of sampling dates, salmon species, and number of fish sampled and their respective mean sizes is provided in the Supplemental Data (Tables S1 and S2).

Sample extraction and clean-up

Analysis of OCP residues in salmon and aquafeed samples was conducted using gas chromatography/high-resolution mass spectrometry (GC/HRMS) at the Institute of Ocean Sciences (Sidney, BC, Canada). Target analytes were chlorobenzenes (CBz), including 1,3,5 TriCBz, 1,2,4 TriCBz, 1,2,3 TriCBz, 1,2,3,5/1,2,4,5 TeCBz, 1,2,3,4 TeCBz, PeCBz, HxCBz; hexachlorocyclohexanes (HCHs), including α-HCH and β-HCH, γ-HCH; dichlorodiphenyltrichloroethanes (DDTs), including *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD *p,p′*-DDD *o,p′*-DDT, *p,p′*-DDT; chlordanes (*trans*-chlordane, *cis*-chlordane *trans*-nonachlor, *cis*-nonachlor, oxychlordane, heptachlor, heptachlor epoxide); cyclodienes (aldrin, deildrin, mirex, endosulphan, endosulfan sulfate, endrin, mirex); and octacholorostyrene. Organochlorine pesticides were co-extracted along with PCBs and PCDD/Fs in these samples, which have been reported elsewhere [4]. Sample extraction, cleanup, and analysis procedures, described briefly below, are more thoroughly detailed by Ikonomou et al. [4,12–14].

Frozen tissue samples (~10 g wet weight) were thawed and spiked with a mixture of surrogate internal standards for analysis of PCDD/Fs, PCBs (17 $^{13}C_{12}$-labeled PCDDs and PCDFs except OCDF), and 15 $^{13}C_{12}$-labeled PCBs. Tissue samples were then mixed with Na_2SO_4 in a mortar, transferred quantitatively into an extraction column, and extracted with approximately 250 ml DCM/hexane (1:1 v/v). Thirty percent of the extract solution by weight was transferred into a 250-ml round bottom flask. This 30% fraction was reduced to 5 to 10 ml by rotary evaporation and transferred to a screw cap vial. This pesticide split was stored at −20°C until pesticide sample cleanup and analyses. The 70% portion of the sample was used for analyses of PCDD/Fs, PCBs, and PBDEs.

The pesticide split from the original extraction was spiked with surrogate spiking standards for pesticide analysis (d_3-1,3,5

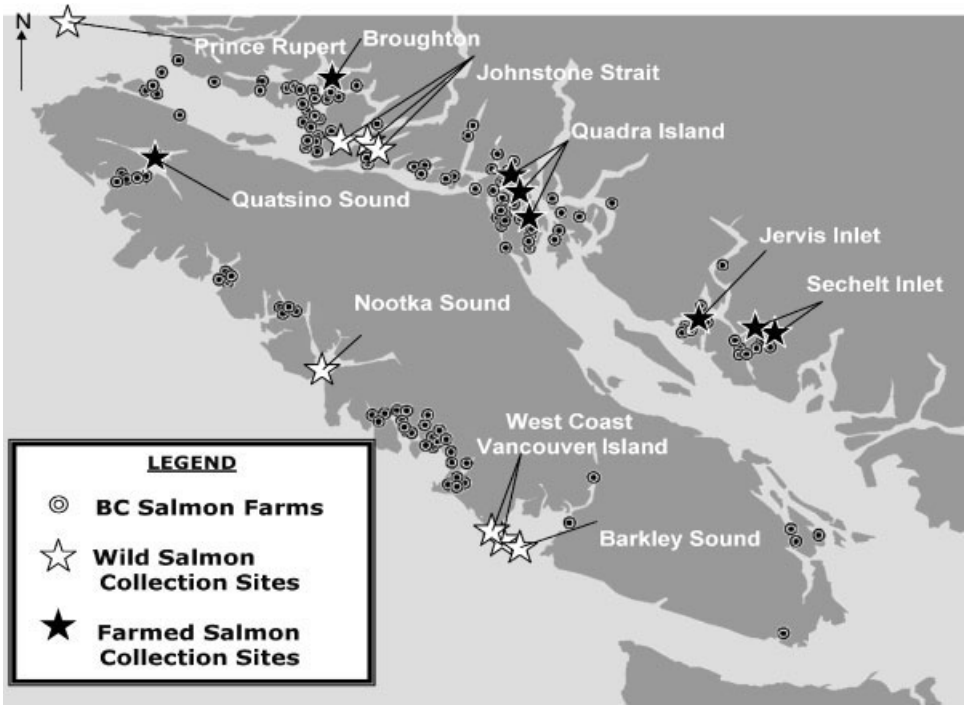

Fig. 1. Map of study area showing salmon farm locations in coastal British Columbia, Canada (bullseye), salmon farm locations sampled (solid stars), and wild salmon sampling locations (open stars).

TriCBz $^{13}C_{12}$-1,2,3,4 TeCBz, $^{13}C_{12}$ HxCBz, $^{13}C_{12}$-β-HCH, $^{13}C_{12}$-γ-HCH,^{13}C mirex, $^{13}C_{12}$-oxychlordane, $^{13}C_{12}$-dieldrin, $^{13}C_{12}$-*p,p′,* DDT, $^{13}C_{12}$- *o,p′* DDT, $^{13}C_{12}$-*p,p′,* DDE, $^{13}C_{12}$-heptachlor epoxide, and $^{13}C_{12}$-*trans*-nonachlor) and cleaned-up using Florisil chromatography (8 g, 1.2% water deactivated). The Florisil column was eluted with 60 ml 1:1 dichloromethane (DCM):hexane. The eluant was collected, reduced to less than 100 µl, spiked with surrogate recovery standard ($^{13}C_{12}$-PCB 47), and submitted for GC/HRMS for pesticide analysis.

Analysis of OCPs by GC/HRMS

Pesticide quantification was conducted at the Institute of Ocean Sciences using GC/HRMS. Analytes were separated using a DB-5 fused silica capillary column (60 m × 0.25 mm inner diameter, 0.1-µm film thickness, J&W Scientific). One µl analyte solution was injected in splitless mode into the split/splitless injector that was maintained at 300°C. The temperature program used under constant pressure (80 kPa for either column) was as follows: hold at 80°C for 3 min, 15°C/min to 160°C, 5°C/min to 300°C, hold 5 min. The GC/mass spectrometry interface temperature was 270°C and the ion source, 310°C. For all analyses, the HRMS was operated at 10,000 resolution under positive electron impact conditions, and data were acquired in the single ion resolving mode. Quantification ions (*m/z*) and isotope ratio control limits are shown in the Supplemental Data (Table S3).

Quality assurance/quality control measures. All samples were analyzed in batches of 12, each consisting of one procedural blank, one in-house reference material, and nine samples, of which one was analyzed in duplicate. Analytes were identified only when the GC/HRMS data satisfied all of the following criteria: two isotopes of the analyte were detected by their exact masses with the HRMS operating at 10,000 resolution during the entire chromatographic run; the retention time of the analyte peak was within 3 s of the predicted time obtained from analysis of authentic compounds in the calibration standards; the maxima for both characteristic isotopic peaks of an analyte coincided within 2 s; the observed isotope ratio of the two ions monitored per analyte were within 15% of the theoretical isotopic ratio; and the signal-to-noise ratio resulting from the peak response of the two corresponding ions was $\geq$3 for proper quantification of the analyte. Chemical concentrations were calculated by the internal standard isotope-dilution method, using mean relative response factors determined from calibration-standard runs made before and after each batch of samples was analyzed. Recoveries of individual internal standards typically ranged from 40 to 110%. Only the most volatile compounds (HCHs and chlorobenzenes) had recoveries between 40 and 70%, for the rest of the analytes the recoveries were between 70 and 110%. Concentrations of all analytes were corrected for percent recoveries of the internal standards.

Method detection limits (MDLs) were determined as three standard deviations (SD) above the mean blank levels. When analyte concentrations were nondetectable in blanks, the MDL was set equal to the instrument detection limit, determined from the analyte peak response with a signal-to-noise ratio of three. The instrument detection limit for individual pesticides varied between 0.0001 and 1.73 ng/g. Some OCPs (tri- and tetra-chlorobenzenes and γ-HCH) were routinely observed in procedural blanks. In many cases, concentrations of those compounds were below MDL (mean + 3 SD of blank levels) and were not included in mean calculations or statistical analyses. For most of the OCP analyses, blank levels were relatively low compared with extracted sample matrix (<10% of sample contribution), and hence concentrations were not blank corrected.

Body measurements and lipid content determination

Fish weight (kg) and fork length (cm) were determined for individual fish during field sampling. Lipid contents of all samples were determined gravimetrically from a parallel extraction of a 5-g subsample (wet wt). Samples were homogenized in $NaSO_4$ and extracted with 100 ml of 1:1 DCM:hexane as discussed previously, reduced by turbo-evaporation to a few milliliters, transferred into a preweighed aluminum dish, dried at 40°C overnight, and subsequently weighed. The measured weight of remaining lipid was used to calculate percent lipid of original wet sample weight. Moisture content was determined by oven-drying the sample at 40°C for 48 h and weighing the sample before and after drying.

Data analysis

The OCP concentrations were compiled on a wet weight basis (ng/g wet weight), as well as lipid-corrected values (ng/g lipid weight). Geometric means ± one standard deviation SD or corresponding 95% confidence intervals (CI_{95}) were determined if frequency of detection was greater than 50%. Samples exhibiting nondetectable concentrations were excluded from mean concentration calculations. Statistical analyses were conducted using JMP 9.02 (SAS Institute). One-way analyses of variance and Tukey's honestly significant difference comparison tests were performed to evaluate differences between mean chemical concentrations observed in aquafeeds or fish species. Bivariate regression was used to assess the influence of lipid content or fish size on observed OCP concentrations.

Biomagnification factors (BMFs) were determined as the ratio of observed lipid-corrected pesticide concentration in fish muscle (C_F) and feed sample (C_D), ($BMF = C_F/C_D$, lipid weight). Biomagnification factors were determined for Atlantic salmon at four farm sites (B, QS, SI, QI), for farmed chinook salmon at two sites (QI2, QI3), and for farmed coho salmon at two sites (JI, SI2), using observed concentrations in those fish and corresponding feed sample collected at each site. Mean BMFs were then determined for the three species of farmed salmon (Atlantic, chinook, and coho). Note the uncertainty involved in these BMF estimates, primarily because the data for one representative feed sample are used to represent the dietary concentration. Nevertheless, we believe that the provided BMF calculations do provide a measure of chemical biomagnification potential in these farmed salmon.

To assess human dietary exposure of selected OCPs, observed flesh residue concentrations in farmed and wild BC salmon (present study) were used to calculate contaminant intakes: daily intake (µg/d) = pesticide concentration (µg/g wet weight) × consumption rate (g/d). Daily contaminant exposure estimates through consumption of BC salmon were based on a food consumption rate of two 140-g servings/week for a 70-kg person. Theoretical minimum and maximum daily intakes (µg/kg body weight/d) were determined using lower and upper bound 95% confidence limits. Exposure estimates were compared with the United States average per capita exposure estimates [15] and PTDI, µg/kg body weight/d, as set by Health Canada [16] and the Joint Meeting on Pesticides Residues, under the auspices of the Food and Agriculture Organization and the World Health Organization [17,18].

RESULTS AND DISCUSSION

Observed pesticide concentrations

Measured flesh residue concentrations of OCPs (ng/g wet weight) in farmed and wild BC salmon, as well as commercial aquafeed, are provided in Table S4 of the Supplemental Data. In terms of frequency of detection, most OCPs were routinely detected in sample extracts. Aldrin, heptachlor, heptachlor epoxide, 1,2,3 TriCBz, 1,3,5 TriCBz, and 1,2,3,5/1,2,4,5 TeCBz were routinely observed in samples of aquafeed and farmed salmon, but only occasionally (frequency of detection $< 50\%$) in wild Pacific salmon samples. The 1,2,4 TriCBz and γ-HCH concentrations were below MDLs in all samples. Mean wet weight concentrations of individual OCPs (ng/g wet weight) in all sources of farmed and wild salmon varied widely, ranging from less than 0.001 to 1.3 (Fig. 2). The rank order of OCP concentrations in salmon was generally DDTs > CHLs ∼ CBz ∼ dieldrin > endosulfans ∼ endrin ∼ octacholorostyrene ∼ mirex ∼ aldrin. The *p,p′*-DDE consistently exhibited the highest concentrations in feed and salmon samples. Among OCP compounds, *p,p′*-DDE was found in the highest concentrations, including detection of 62.7 ng/g in a sample of commercial feed and 43.7 ng/g in a farmed Atlantic salmon from Broughton. The observed concentrations are generally comparable to those previously reported by Hites et al. [3]. Similar to those previous reports, we observed significantly higher concentrations of some OCPs in farmed salmon compared with wild salmon. Specifically, concentrations of individual DDTs, HCHs (α and β isomers), chlordanes, and various cyclodiene pesticides (dieldrin, Mirex) in farmed salmon were 2 to 11 times higher ($p < 0.0001$) compared with wild salmon sources. Chemical analyses also revealed the presence of various metabolites. In addition to *p,p′*-DDE (primary DDT metabolite), we observed detectable concentrations of endosulfan sulfate and oxychlordane and heptachlor epoxide in both farmed and wild salmon samples (Fig. 2).

The profiles of the various OCP classes were comparable among aquafeed and different salmon species studied (Supplemental Data, Fig. S1). For example, *p,p′*-DDE was the dominant DDT component, constituting 60 to 80% of ΣDDTs in feed, as well as farmed and wild salmon; HxCBz was consistently the dominant chlorobenzene (>80% of ΣCBz); dieldrin was the dominant cyclodiene pesticide (60 to 80% of Σcyclodienes); α-HCH and β-HCH generally made up 60% and 40% of total HCH burdens, respectively. *trans*-Nonachlor was the dominant chlordane component, followed by *cis*-chlordane and *cis*-nonachlor.

Interspecies and site-specific variation

Interspecies and site-specific comparisons reveal that flesh residue concentrations of OCPs (ng/g wet wt) varied substantially between the different groups of farmed salmon sampled (Fig. 3). One-way analysis of variance and Tukey's honestly significant difference comparison confirm that farmed Atlantic salmon exhibited significantly higher ($p < 0.05$) OCP concentrations (ng/g wet weight) compared with other farmed species. Among wild salmon, wild chinook typically exhibited the highest OCP flesh residue concentrations. However, concentrations of some OCPs (chlorobenzenes, chlordanes, and other cyclodienes) in wild sockeye from Johnstone Strait were comparable to those in wild chinook.

Much of the observed variation in flesh residue concentrations of OCPs (ng/g wet weight) can be explained by lipid content and fish size. Not surprisingly, bivariate regression analyses showed flesh residue concentrations of OCPs, lipophilic substances, were positively correlated with fish lipid content. Measured lipid contents in farmed salmon ($11.5 \pm 3.5\%$) were significantly higher ($p < 0.05$) compared with wild salmon (3.0 ± 2.4). Farmed Atlantic salmon, which have significantly higher lipid contents ($13.5 \pm 2.9\%$) compared with other farmed species, exhibit the highest flesh concentrations of OCP (Supplemental Data, Fig. S2). This is consistent with observations of PCBs, which demonstrated that farmed Atlantic salmon exhibit three to eight times higher concentrations compared with all other salmon sources [4]. The effect of lipid content is not as apparent for wild salmon species, because lipid contents were not significantly different in these fish ($p > 0.05$), (Supplemental Data, Fig. S2).

Regression analyses also showed that wet weight OCP concentrations were positively correlated with fish size. For example, relatively strong correlations were observed between ΣDDTs (ng/g wet wt) and fish weight (kg) for wild chinook salmon ($r^2 = 0.49$) and farmed Atlantic salmon ($r^2 = 0.55$). (Supplemental Data, Fig. S3). For farmed Atlantic salmon, fish size was significantly correlated with lipid content ($r^2 = 0.41$), which is consistent with previous observations of captive salmonids fed unrestricted diets [19,20]. Conversely, no

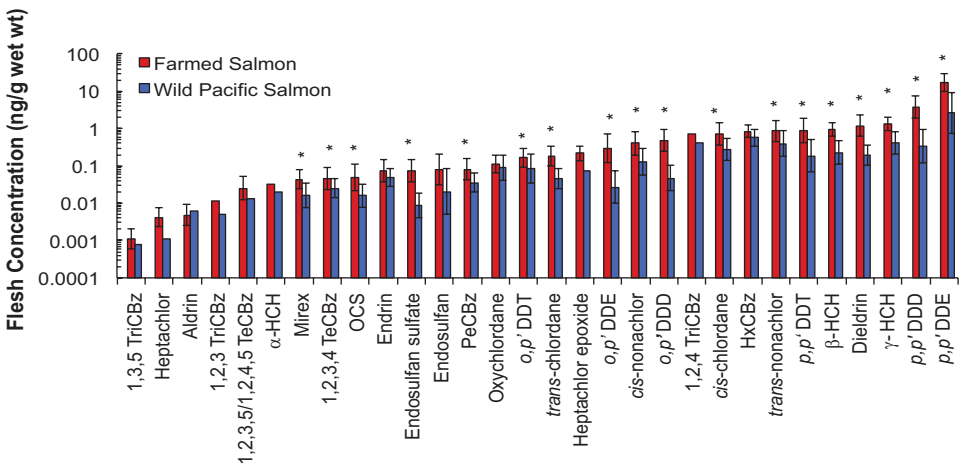

Fig. 2. Concentrations (ng/g wet wt) of organochlorine pesticides measured in farmed salmon (left column) and wild salmon (right column) from British Columbia. Data represent geometric means ± 1 standard deviation. * = significant deference ($p < 0.05$) between farmed and wild salmon. See text for definitions of acronyms. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com]

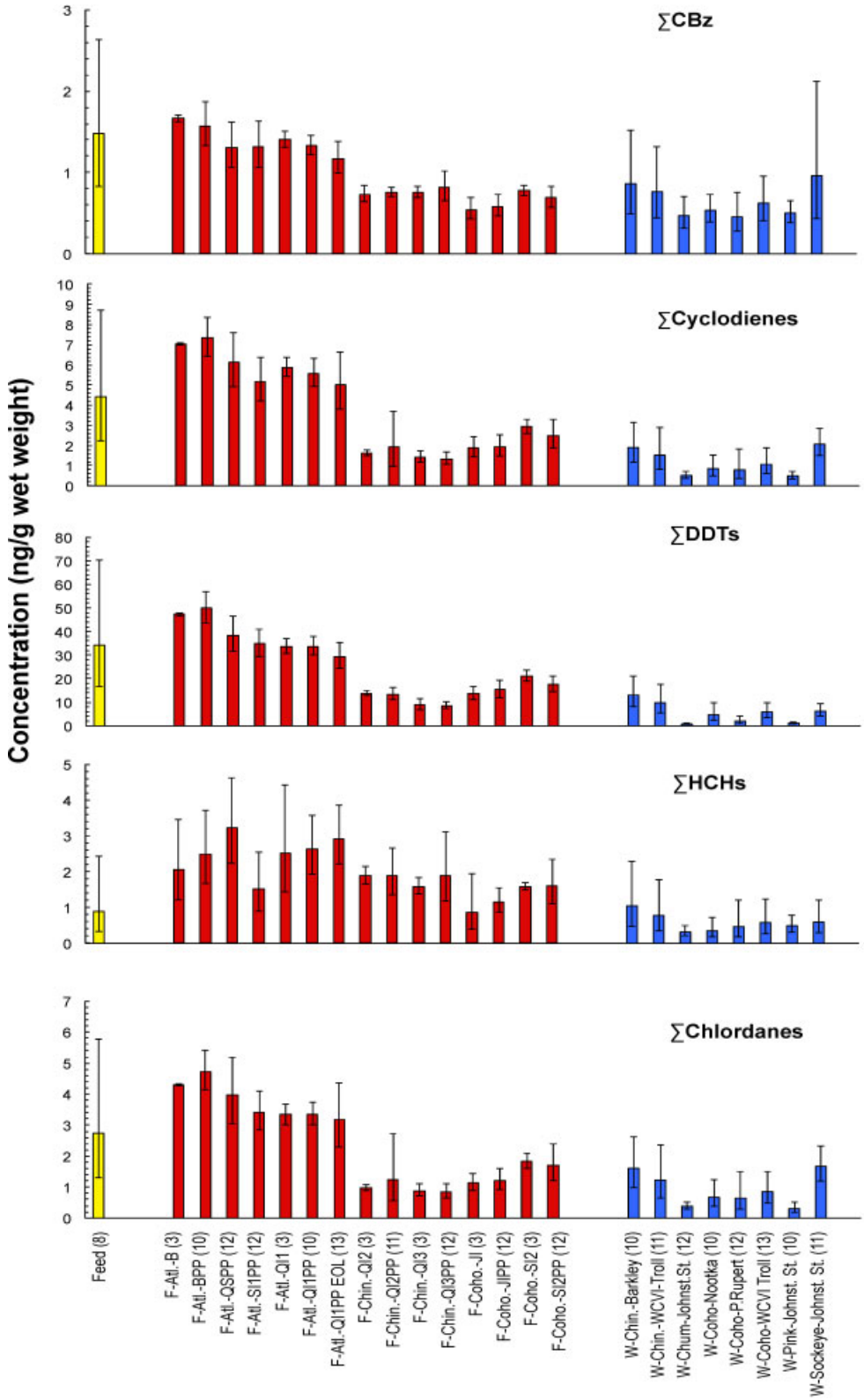

Fig. 3. Concentrations (ng/g wet wt) of chorobenzenes (ΣCBz), cyclodienes (ΣCyclodienes), dichlorodiphenyltrichloroethanes (ΣDDTs), hexachlorocyclohexanes (ΣHCHs), and chlordanes (ΣChlordanes), measured in commercial salmon feed (left column), farmed salmon (center columns), and wild salmon (right columns) from British Columbia, Canada. Error bars represent ±1 standard deviation. Farmed salmon samples, including those collected at farm sites or on arrival at the processing plant (PP), are as follows: farmed Atlantic salmon (F-Atl.) from Broughton (B, BPP), Quatsino Sound (QSPP), Sechelt Inlet (SI1PP), and Quadra Island (QI1, QI1PP, QI1PP-end-of-line); farmed chinook (F-Chin) from Quadra Island (QI2, QI2PP, QI3, QI3PP); farmed coho (F-coho) from Jervis Inlet (JI, JIPP) and Sechelt Inlet (SI2, SI2PP). Wild salmon sources include wild chinook (W-Chin.) from Barkley Sound (Barkley) and West Coast Vancouver Island (WCVI-Troll); wild chum (W-Chum) from Johnstone Strait (Johnst. St.); wild coho (W-coho) from Nootka Sound (Nootka), Prince Rupert (P. Rupert), and West Coast Vancouver Island (WCVI-Troll); wild pink salmon (W-Pink) from Johnstone Strait (Johnst. St.); wild sockeye (W-Sockeye) from Johnstone Strait (Johnst. St). Data represent geometric means ± 1 standard deviation. Numbers in parentheses represent sample size. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com]

relationship between fish size and lipid content was observed for wild salmon in the present study.

Lipid-corrected OCP data (ng/g lipid wt) are presented in Figure S4 of the Supplemental Data. This data presentation removes the confounding effect of differences in flesh lipid content between the different salmon sources. The observed species and site-specific differences on a lipid weight basis for farmed salmon (Atlantic, chinook, and coho) are similar to those observed for wet weight concentrations (Fig. 3). One-way analysis of variance results demonstrate that lipid corrected OPC concentrations in farmed Atlantic salmon are significantly higher ($p < 0.05$) compared with other farmed species. However, when expressed on lipid weight basis, a somewhat different interspecies and site-specific pattern of OCPs is apparent. First, lipid-corrected concentrations in wild salmon species are comparable to, and in some cases exceed, levels observed in farmed salmon species. For example, DDT concentrations in wild chum salmon from Johnstone Strait (mean = 47.9, $CI_{95} = 18.0$ to127 ng/g lipid) were four times higher ($p < 0.05$) than the highest ΣDDT concentration observed in farmed Atlantic salmon from Broughton (mean = 11.9, $CI_{95} = 5.55$ to 25.4 ng/g lipid). Second, OCP concentrations tend to be highest in wild chinook salmon and lowest in wild pink salmon. However, for the most part, the observed differences in lipid corrected concentrations in these wild salmon are not statistically significant ($p > 0.05$). Some exceptions exist, including observations of significantly higher ΣDDT concentrations in wild chinook ($p < 0.05$), significantly lower chlordane concentrations in wild pink salmon ($p < 0.05$), and significantly higher ($p < 0.05$) concentrations of ΣCBz in wild chum salmon (Supplemental Data, Fig. S4). A plot of lipid corrected ΣDDTs concentrations (ng/g lipid weight) versus fish weight is shown in the Supplemental Data for reference (Fig. S5). The data indicate that OCP concentrations in wild salmon are not correlated with fish size, which did not differ significantly among wild salmon ($p > 0.05$).

The lipid normalized concentration data clearly show that although contaminant burdens in farmed salmon tend to be higher in comparison with wild salmon on a wet weight basis, this is not true when differences in their respective lipid contents are taken into account. The elevated concentrations of OCPs in farmed Atlantic salmon compared with other species is consistent with observations of PCBs and PCDD/Fs, which has been attributed to the fact that farmed salmon are typically supplied more lipid-rich (high-energy) formulated diets [4]. The lack of interspecies differences in OCP concentrations (lipid weight) among wild salmon was somewhat surprising. In particular, OCP concentrations were anticipated to be elevated in chinook salmon. Previous analyses of other persistent organic pollutants (PCBs and PCDD/Fs) showed that chinook salmon exhibited significantly higher ($p < 0.05$) lipid corrected concentrations compared with other wild salmon species [4]. This was attributed to the fact that chinook salmon are piscivorous, longer-lived species that tend to occupy a higher trophic level than the other wild salmon species studied.

In essence, a variety of factors may influence concentrations of hydrophobic organic contaminants in farmed and wild salmon. These include divergent life history, prey/feed characteristics and composition, bioenergetics, and ambient environmental concentrations. For example, detection of relatively high concentrations (lipid normalized) of certain OCPs (chlorobenzenes, chlordanes, cyclodienes) in wild chum and sockeye from Johnstone Strait (Supplemental Data, Fig. S4) compared with wild chinook salmon suggests that the former salmon species may be exposed to elevated ambient concentrations of those OCPs along their migration pathway. Further environmental monitoring of OCPs and other chemicals of concern in BC's coastal waters may provide insight into geographical variation patterns, help identify highly contaminated zones (hot spots), and ultimately serve to strengthen future ecological and human health risk assessments.

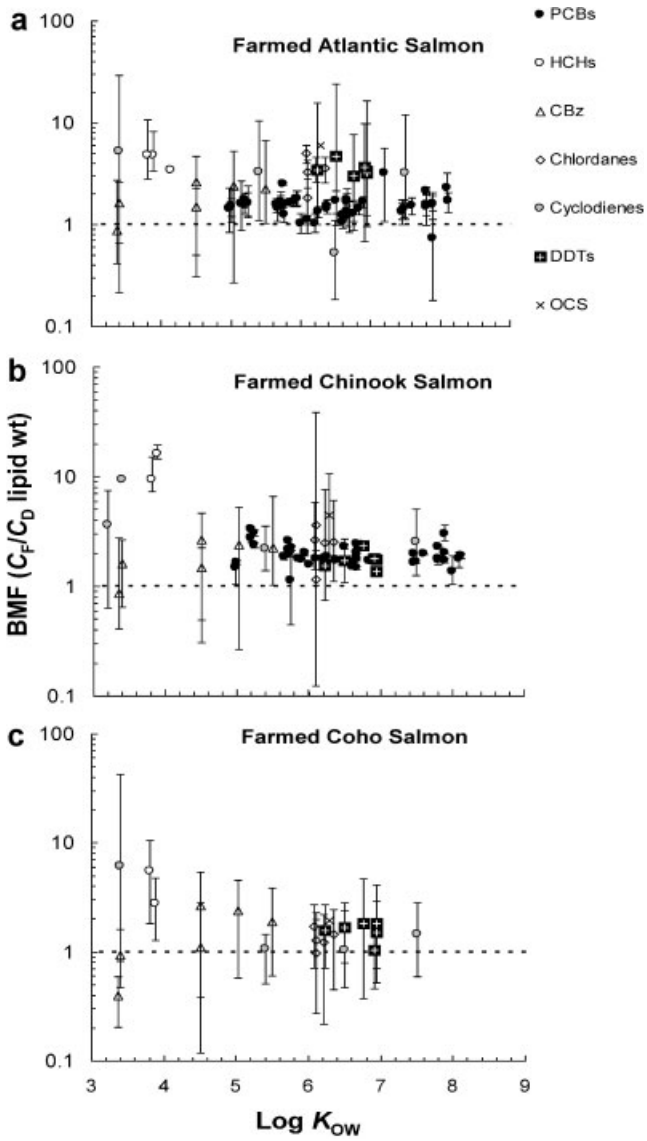

Fig. 4. Relationship between biomagnification factor (BMF = C_F/C_D, lipid wt) of individual polychlorinated biphenyl congeners and organochlorine pesticides versus chemical octanol–water partition coefficient (log K_{OW}) for (**A**) farmed Atlantic salmon, (**B**) farmed chinook salmon, and (**C**) farmed coho salmon.

Biomagnification of OCPs in farmed salmon

The interspecies and site-specific comparison based on lipid corrected OCP concentrations (ng/g lipid weight) also highlight that OCP concentrations in farmed salmon were generally higher than corresponding concentrations observed in commercial feed samples, indicating biomagnification (Supplemental Data, Fig. S4). The concentration of ΣDDTs (ng/g lipid weight) in farmed Atlantic salmon from Broughton (BPP samples: mean = 361, $CI_{95} = 150$–869) was three times higher than lipid corrected ΣDDT concentrations in feed (mean = 109, $CI_{95} =$ 27.8 to 425).

Observed BMFs of individual OCPs in market-size farmed Atlantic, chinook, and coho salmon are provided in the Supplemental Data (Table S5). Biomagnification factors of the various OCPs typically ranged between 2 and 5, indicating biomagnification. Interestingly, less hydrophobic compounds such as HCH isomers and endosulfan ($\log K_{OW} < 5$) relatively high BMFs (between 2.8 and 16), compared with more hydrophobic compounds (e.g., DDTs, chlordanes). For example, mean BMFs of β-HCH in farmed Atlantic, chinook, and coho were 4.8, 9.3, and 5.5, respectively.

A plot of observed BMFs of individual OCPs and PCB congeners for farmed Atlantic, farmed chinook, and farmed coho salmon versus log K_{OW} does not reveal a clear BMF–K_{OW} relationship (Fig. 4). However, the data do indicate a consistent trend of relatively high BMFs for low K_{OW} chemicals (HCHs and endosulfan), compared with more hydrophobic compounds (DDTs, chlordanes, and PCBs). This is contrary to previous observations in laboratory fish [21,22] and field studies [23,24]. The reason for these seemingly high BMFs may be the influence of ambient seawater concentrations of these relatively water-soluble compounds. Freely dissolved water concentrations (C_{WD}) in the range of 0.3 to 1 ng/L have previously been reported for α-HCH in seawater from coastal British Columbia [25]. Using these previously reported water concentrations and measured flesh residue concentrations (the present study), we estimate a log BAF ($\log C_F/C_{WD}$, kg/L) of approximately 3 for α-HCH in BC farmed salmon. This is consistent with previously measured and model-predicted fish BAFs for chemicals in this K_{OW} range [26,27]. Consequently, seawater concentrations and corresponding BAF estimates may be more useful than feed concentrations to estimate flesh residue levels of these relatively more water-soluble contaminants such as HCHs and endosulfan in these farmed salmon.

Human exposure assessment

Human dietary exposure estimates of endosulfan, heptachlor, *p,p′*-DDT, hexachlorobenzene, and dieldrin via consumption of farmed and wild salmon (two 140-g servings per week) ranged between 0.00001 and 0.001 μg/kg body weight/d (Table 1). Organochlorine pesticides exposure levels were two to five times higher for farmed salmon, compared with exposure through consumption of wild Pacific salmon. Chlordane exhibited the highest exposure among the various OCPs investigated. The OCP exposure from consumption of salmon was generally very low compared with the United States national average per capita total exposure levels and PTDIs (Table 1). For example, using CI_{95} daily intake values of dieldrin via weekly BC salmon consumption (farmed or wild), we estimate that salmon contributes only 0.1 to 9% of the total exposure for the general public in the United States (0.03 μg/kg body weight/d) and only 0.06 to 5% of the PTDI (0.05 μg/kg body weight/d; Table 1).

Toxaphene exposure, estimated from previously reported flesh residue levels in BC salmon [3], appears to be higher than other OCPs. For example, toxaphene intakes of 0.03 and 0.004 μg/kg body weight/d were estimated for consumption of farmed and wild salmon, respectively. Consequently, salmon consumption potentially contributes a greater percentage of the total toxaphene daily intake (0.7–40%; Table 1).

Concentrations of organic contaminants in farmed salmon can vary substantially between different regions worldwide [3]. Resulting flesh residue concentrations in farm-raised salmon may be influenced by various factors, including feed composition and duration, bioenergetics, and local ambient environmental concentrations. Of particular importance is the degree of contamination of dietary lipids used in the formulation of commercial aquafeeds [4,6,10,28].

Table 1. Estimated daily intake of organochlorine pesticides (μg/kg body wt/d) for persons consuming BC farmed and wild salmon along with existing provisional tolerable daily intake (PTDI) and the United States per capita exposure levels[a]

	PTDI[b] (μg/kg body wt/d)	Intake from farmed salmon (μg/kg body wt/d)[c]	Intake from wild salmon (μg/kg body wt/d)[c]	United States per capita total exposure (μg/kg body wt/d)[d]	Salmon contribution to total exposure (%)	Salmon contribution to PTDI (%)
Pesticides (μg/kg body wt/d)						
Endosulfan	0.3	0.00005 (*0.00001–0.00023*)	0.00001 (*0.000001–0.0001*)	0.05	*0.003–0.47*	*0.0005–0.08*
Heptachlor	0.1	0.0001 (*0.00004–0.0004*)	0.00004 (*0.00001–0.0001*)	0.03	*0.04–1.34*	*0.02–0.4*
p,p′-DDT	20[e]	0.0005 (*0.0001–0.002*)	0.0001 (*0.00002–0.0006*)	0.1	*0.02–2.1*	*0.002–0.2*[e]
Hexachlorobenzene	0.27	0.0005 (*0.0002–0.001*)	0.0003 (*0.0001–0.001*)	0.04	*0.24–3.6*	*0.04–0.53*
Dieldrin	0.05	0.0007 (*0.0002–0.003*)	0.0001 (*0.00003–0.0004*)	0.03	*0.1–9*	*0.06–5*
Chlordane	0.05	0.0012 (*0.0003–0.005*)	0.0004 (*0.0001–0.002*)	0.07	*0.2–7*	*0.2–9.8*
Toxaphene[e]	0.2	0.03 (*0.0086–0.08*)	0.004 (*0.001–0.012*)	0.2	*0.7–40*	*0.7–40*

[a] Values in italics represent 95% confidence interval range (CI_{95}).
[b] PTDIs (μg/kg body wt/d) represent Health Canada [16], or World Health Organization (WHO) [17–19] PTDIs as follows: endosulfan (0.3 μg/kg body wt/d, Health Canada); heptachlor (0.1 μg/kg body wt/d, [17,18]); lindane (0.3 μg/kg body wt/d, Health Canada); DDTs (20 μg/kg body wt/d, Health Canada); hexachlorobenzene (0.27 μg/kg body wt/d, Health Canada); dieldrin (0.05 μg/kg body wt/d, Health Canada); chlordane (0.05 μg/kg body wt/d, [17,18]); toxaphene (0.2 μg/kg body wt/d, Health Canada); dioxins, normalized to 2,3,7,8 tetrachloro-*p*-dibenzodioxin (2,3,7,8 TCDD) toxic equivalent or TEQ (1 pg TEQ/kg body wt/d, Health Canada); dioxin-like polychlorinated biphenyls (DL-PCBs), (1 pg TEQ/kg body wt/d, Health Canada) and THg (0.7 μg/kg body wt/d, Health Canada)
[c] Intake estimates were determined from geometric mean concentrations. Ranges in parentheses represent intakes based on lower and upper 95% confidence interval concentrations.
[d] National per capita average exposure level determined for United States. See Dougherty et al. [15] for details.
[e] PTDI for dichlorodiphenyltrichloroethane (DDT) is for the sum of DDTs, DDDs (dichlorodiphenyldichloroethanes), and DDEs (dichlorodiphenyldichloroethylenes), [18].
[f] Dietary intake estimates were determined using previously reported toxaphene levels in British Columbia farmed and wild salmon [3].
Abbreviations: BC = British Columbia; PTDI = provisional tolerable daily intakes; DDT = dichlorodiphenyltrichloroethane; 2,3,7,8 TCDD = 2,3,7,8 tetrachloro-*p*-dibenzodioxin; TEQ = toxicity equivalent; DL-PCBs = dioxin-like polychlorinated biphenyls; DDD = dichlorodiphenyldichloroethane; DDE = dichlorodiphenyldichloroethylenes.

The aquaculture industry is moving toward greater utilization of terrestrial-based lipid sources (e.g., canola oil and poultry fat) in the formulation of salmon feed, which tend to exhibit substantially lower concentrations of lipophilic contaminants compared with marine fish oils incorporated in traditional salmon feeds. Friesen et al. [10] demonstrated that substitution of these alternative terrestrial-based diets can effectively reduce levels of lipophilic contaminants such as PCBs and PCDD/Fs in market-size salmon by as much as seven times. The farmed salmon from the present study were raised mainly on traditional fish oil–based aquafeeds. Increasing usage of terrestrial-based salmon feeds will likely result in comparable residue level reductions for OCPs such as dieldrin, toxaphene, chlordanes, and DDTs. Human exposure to those contaminants through salmon consumption therefore also may decline in coming years. The same may not be true for less hydrophobic OCPs such as HCHs and endosulfans, because flesh residues of these compounds appear to be more influenced by ambient seawater concentrations rather than dietary exposure.

Despite relatively low OCP exposure associated with salmon consumption, total exposure of some OCPs can in some cases exceed recommended PTDIs (Table 1). For example, the United States national average per capita exposure of toxaphene (0.2 μg/kg body weight/d) and chlordane (0.07 μg/kg body weight/d) are greater than or equal to corresponding PTDIs for those compounds. The hazard ratio, determined as the ratio of estimated exposure to PTDI, is equal to 1.0 and 1.4 for toxaphene and chlordane, respectively (Supplemental Data, Fig. S6). Provisional tolerable daily intakes are inherently conservative estimates, derived from no-observable adverse effect levels and application of safety factors (10–100) to account for interspecies variation and animal size. The fact that estimated United States exposure levels exceed PTDI values is somewhat concerning, especially considering exposure risks may be underestimated, because PTDIs do not consider carcinogenic effects of pesticides (e.g., cancer benchmark concentrations), which would likely result in comparatively higher hazard ratios [15].

The occurrence of residues of persistent environmental contaminants such as PCBs, PCDD/Fs, and pesticides in seafood remains a global public health concern. The data from the present study indicate that human dietary exposure of OCPs via consumption of farmed and wild BC salmon is relatively low compared with total estimated exposure and PTDIs. We have reported similar findings for PCBs, PCDD/Fs, Hg, and other trace elements [4,11]. Together, critical analyses of the available data further suggests that bi-weekly consumption of BC salmon is not anticipated to result in any substantial risks associated with potentially toxic environmental contaminants; hence, salmon remains a safe dietary source of beneficial *n*-3 HUFAs.

SUPPLEMENTAL DATA

Figure S1. Chlorobenzenes vs chlordanes.
Figure S2. DDTs versus lipid content.
Figure S3. DDT levels by fish weight.
Figure S4. Lipid Corrected Concentrations.
Figure S5. DDTs.
Figure S6. Hazard ratios. (317 KB PDF)

Table S1. Farmed salmon samples analyzed for fillet concentrations of organohalogen contaminants.

Table S2. Wild salmon samples analyzed for fillet concentrations of organohalogen contaminants, trace metals, proximate constituents, fatty acids, carotenoid pigments, and gross energy.

Table S3. Pesticide *m/z* (single ion resolving mode) and isotope ratio control limits.

Table S4. Concentrations of organochlorine pesticides.

Table S5. Biomagnification factors (BMFs, C_F/C_D lipid wt) of organochlorine pesticides. (29 KB DOC)

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