

Persistent Organohalogens in Paired Fish Fillet and Eggs: Implications for Fish Consumption Advisories

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S Supporting Information

ABSTRACT: Fish consumption is associated with both health benefits from high-quality proteins, minerals, vitamins, and fatty acids and risks from contaminants in fish. Fish consumption advisories are issued by many government agencies to keep exposure to contaminants at a safe level. Such advisories are typically based on fillets and neglect consumption of other fish parts such as eggs by certain subpopulations. To evaluate potential for dietary exposure to toxic organic chemicals via fish eggs, we analyzed polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCNs), dioxin-like polychlorinated biphenyls (dlPCBs), and polychlorinated dibenzodioxins/furans (PCDD/Fs) in paired fillet and eggs of fish from a tributary to Lake Ontario, one of the North American Great Lakes. All wet weight based concentrations in fish eggs were statistically higher than in the paired fillet samples. In fish eggs, concentrations of Σ_{14} PBDEs, Σ_{14} PCNs, and Σ_{12} dlPCBs were 41–118, 0.3–1.7, and 30–128 ng/g wet weight (ww), respectively; Σ_3 PCDD/Fs and total (dlPCB+ PCDD/Fs) toxic equivalents (TEQs) were 4–22 and 9–54 pg/g ww, respectively. In fillet samples, Σ_{14} PBDEs, Σ_{14} PCNs, and Σ_{12} dlPCBs were 4–116, 0.05–0.66, and 6–85 ng/g, respectively; Σ_3 PCDD/Fs and TEQs were 2–10 and 3.4–31 pg/g ww, respectively. In contrast, the fillets had higher lipid normalized concentrations than the paired egg samples, suggesting that these chemicals did not reach equilibrium between the fillets and eggs. Accordingly, measured concentrations in eggs or empirical relationship with fillet rather than prediction from equilibrium partitioning model should be used to evaluate contaminant exposure via consumption of fish eggs. For fatty fish from the lower Great Lakes area, we suggest one fillet meal be reduced from the advised fish consumption frequency for consumptions of 207 ± 37 , 39 ± 2 , 105 ± 51 , and 119 ± 9 g fish eggs of brown trout, Chinook salmon, Coho salmon, and rainbow trout, respectively.

KEYWORDS: fish consumption advisory, roe eggs, contaminant exposure, POPs, toxics, human health

INTRODUCTION

Fish consumption is regarded as beneficial in the development of the brain and nervous system and in the prevention of coronary heart disease, arrhythmias, and thrombosis^{1,2} due to abundant essential nutrients such as omega-3 polyunsaturated fatty acids, high-quality protein, minerals, and vitamins.^{3,4} Because of such health benefits, fish is commonly considered as a nutritional and healthy food, and thus many dietary guidelines recommend at least two servings of fish per week for adults.^{5–7} Many essential fatty acids cannot be synthesized by the human body and, hence, dietary intake is important.⁸

On the other hand, bioaccumulation results in higher concentrations of many contaminants in fish than in the surrounding environment. Elevated levels of contaminants in fish can cause increased human exposure and health risk from fish consumption. Despite a small fraction of fish in diet, uptake from fish consumption can be a dominant exposure pathway for mercury and many persistent organic pollutants (POPs).^{9,10} Therefore, many environmental or public health agencies^{11,12} have established fish consumption advisories to protect people from being exposed to contaminants more than the safe dosages (tolerable daily intake). On the basis of tolerable daily intake, fish consumption patterns of different age groups, and measured concentrations of contaminants in fish, a recommended fish consumption frequency (number of fish servings per month) is determined and suggested to be followed by the

public. Fish consumption advisories also target specific populations such as women that are pregnant or may become pregnant, whose exposure to toxic contaminants in fish could affect the prenatal exposure and early development of the children. Such practice has been proved effective in reducing the mothers' body concentrations and thus children's prenatal exposures to the most persistent and heavily chlorinated polychlorinated biphenyls by >50%.¹³

It is widely known that contaminant concentrations in fish fillet can significantly vary with fish origin, species, age, size, and sex.^{14–18} Fish consumption advisories usually consider such variations.¹⁹ Additional factors that can affect the risk associated with fish consumption include methods of fish processing used by consumers and a fish portion other than fillet consumed. For example, some consumers eat a fish fillet with the skin intact, whereas some remove skin before eating a fillet. Skin removal has been found to be effective in reducing the intake of many persistent organic contaminants.²⁰ Similarly, various cooking methods can also affect contaminant exposure differently.^{21,22} Some consumers eat other parts of fish such as head, tail, and eggs. Fish eggs, also widely known as roe or caviar, have been

Received: January 7, 2016

Revised: March 14, 2016

Accepted: March 24, 2016

Published: March 24, 2016

considered a delicacy in several traditional cultures, possibly because of observed health benefits associated with omega-3 polyunsaturated fatty acids and proteins.^{23,24}

Although “roe” is generally used for salmon eggs that are commonly used in sushi²⁴ and “caviar” is generally used to refer to unfertilized eggs of sturgeon and paddlefish,²⁵ fish eggs can come from nearly any fish. A survey conducted among Columbia River Basin Indian tribes suggested 43% of fish consumers consumed eggs of salmon and 14% consumed eggs of trout.²⁶ According to a survey on sport fish consumption habits of Ontario anglers, 2.4% consume eggs of sport fish.²⁷ For certain population groups such as Asian Canadians living around the Great Lakes, the percent of populations eating eggs of Great Lakes fish can be >35%.¹³

A number of studies have reported differences in contaminant concentrations in various parts of a fish body. For example, higher concentrations of POPs were observed for fish muscle at the head end than at the tail.²¹ Also, fish eggs have higher average lipid content than the fillet.²⁸ According to bioaccumulation models based on equilibrium phase partitioning,^{29,30} concentrations of organic contaminants in fish eggs can be predicted from the corresponding fillet concentrations using the lipid contents in eggs and fillet. However, maternal transfer of organic contaminants from a fish body to eggs is more complicated than can be explained by the variations in total lipid content using a simple partitioning model.^{31,32} As such, field data would provide useful information for evaluating and improving models that associate contaminant concentrations in fish fillet and eggs.

In this study, we examined the wet weight-based and lipid-normalized concentrations of polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCNs), dioxin-like PCBs (dlPCBs), and polychlorinated dibenzodioxins/furans (PCDD/Fs) in paired fillet and eggs of four species, namely, Chinook salmon (*Oncorhynchus tshawytscha*), Coho salmon (*Oncorhynchus kisutch*), brown trout (*Salmo trutta*), and rainbow trout (*Oncorhynchus mykiss*), sampled from a tributary of Lake Ontario, a North American Great Lake. Using the measured contaminant concentrations, we evaluated the effectiveness of a simple partitioning model in predicting the concentrations in fish eggs from fillet. We quantified the amount of fish eggs with contaminant concentrations equivalent to one meal of fish fillet. This knowledge would be useful to public health officials and fish consumers in managing the risk associated with the consumption of fish eggs.

MATERIALS AND METHODS

Sample Collection. As a part of Ontario's Fish Contaminant Monitoring Program, 11 pairs of skinless fillets and eggs of Chinook salmon, Coho salmon, brown trout, and rainbow trout ranging in size (Table S2) were collected in the fall of 2007 (October 22–November 1) from the Credit River and analyzed for PBDEs, PCNs, PCDD/F, and dlPCBs. The river provides spawning areas to fish and is a tributary of Lake Ontario, a North American Great Lake. Numerous studies have described elevated levels of legacy contaminants as well as contaminants of emerging concern in Lake Ontario fish.^{14,33–39} Fish were filleted, and egg samples were retrieved in the field. The samples were transported on ice to the Ontario Ministry of the Environment and Climate Change laboratory in Toronto, where skin-removed fillets and egg samples were homogenized and kept frozen at −20 °C before the contaminant analyses.

Contaminant Analyses. For each fish, eggs and a skinless, boneless fillet of the dorsal muscle were quantified for lipid content, 17 tri- to deca-PBDEs, 25 PCN congeners (4 coeluted), 12 dioxin-like

PCBs, and the 17 most toxic 2,3,7,8-substituted congeners of PCDD/Fs. Lipid in samples was extracted with methanol and chloroform (2:1) and measured gravimetrically. Concentrations of 17 PBDEs were measured using gas chromatography with high-resolution mass spectrometry (GC-HRMS) based on OMOE method BDE-E3430.⁴¹ All samples were fortified with at least one ¹³C₁₂ isotopically labeled congener for each homologue group (quantification standard). The PBDEs were quantified against these corresponding labeled internal standards. The PCNs were measured using GC-HRMS based on OMOE method PCN-E3431.⁴² PCN-27, -42, -52, -67, -73, and -75 were quantified using isotope dilution, whereas the other PCN congeners were quantified by internal standard methods. The PCDD/Fs and dlPCBs were measured using isotope dilution with GC-HRMS (OMOE method DFPCB-E3418⁴⁰) as described in detail by Bhavsar et al.¹⁴ This method is similar to U.S. EPA 1613 for PCDD/Fs and to U.S. EPA 1668 for dlPCBs. Detailed analytical methods are provided in the Supporting Information.

QA/QC. Instrument precision (within-run precision) was measured by injecting 10 replicates of the calibration solutions (see OMOE method BDE-E3430⁴¹ for concentrations of each chemical). Instrument accuracy (between-run precision) was verified at least twice daily by calculating percent deviation for relative response factor of all parameters in the calibration standard solution, as compared to the most recent calibration curve. These continuing calibration checks were run at the beginning and end of each set of samples analyzed. An acceptance range of ±20% deviation for native congeners (±30% for labeled congeners) was used. A blank sample was fortified with known amounts of native analyte and processed with each sample set. Recoveries and limit of detection of the analytes are listed in Table S1 of the Supporting Information.

Toxic Equivalents (TEQs) Derivation. TEQs were calculated for 7 PCDDs, 10 PCDFs, and 12 dlPCBs, which have toxic equivalent factors (TEFs) set by the World Health Organization (WHO) in 2005.⁴³ Such a TEQ estimation approach relies on the premise that toxicological effects of dlPCBs and PCDD/Fs occur through the same pathway and their dose–response curves are parallel and additive.^{44,45} For a contaminant measured below the method limit of detection (LOD), half of the LOD was used for the TEQ calculations.

RESULTS AND DISCUSSION

Physical Characteristic and Lipid Content. Physical characteristics of the fish samples and lipid content of the paired egg/fillet samples are listed in Table S2. Female fish from which the paired eggs and fillet were collected ranged from 0.495 to 0.913 m in length and from 0.90 to 6.51 kg in weight. The egg samples had a higher lipid fraction ($g_{\text{lipid}}/g_{\text{sample}}$, wet weight) than the corresponding fillet samples (5.2–12.0% and 0.8–6.4%, respectively). Variation in lipid content of the egg samples of different species (coefficient of variation (CV) = 25%) was smaller than the fillet samples (CV = 66%). The ratios of lipid content in eggs to fillet were 1.3–12.5, and no significant correlation was found between the two parameters (Spearman correlation $p = 0.75$). The egg to fillet lipid ratios for Chinook salmon (12.0 and 12.5) were higher than for the other fish species (1.3–4.9) due to both the higher lipid contents in the eggs and the lower lipid contents in the fillet. Such a distinctive characteristic of Chinook salmon can be attributed to the higher depletion of body lipid during the upstream migration for spawning.⁴⁶

Concentrations and Congener Profiles of PBDEs. Wet weight (ww)-based concentrations (C_{ww}) of the 17 PBDE congeners analyzed in the paired egg and fillet samples are presented in Table S3. BDE-85, -138, and -209 were detected in only about 30, 10, and 10% of the samples, and thus we focused on the other 14 PBDE congeners. Total concentrations of the 14 PBDE congeners ($\Sigma_{14}\text{PBDEs}$) in the egg samples ranged

from 41 to 118 ng/g ww (arithmetic mean/median = 70/63 ng/g ww; Figure 1a). Σ_{14} PBDEs in the egg samples were higher

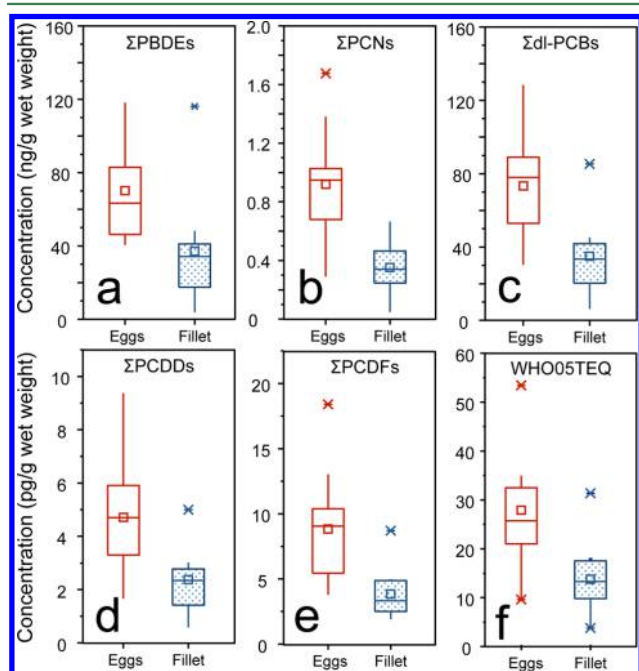


Figure 1. Wet weight-based concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and (f) World Health Organization (WHO) 2005 toxic equivalents (TEQs) in paired samples of fish eggs and skinless and boneless fillet analyzed in this study.

than in the paired fillet samples (paired *t* test one-tailed *p* = 0.002), which had Σ_{14} PBDEs ranging from <8 to 116 ng/g ww (mean/median = 37/34 ng/g ww).

The ratios of $C_{WW}(\Sigma_{14}\text{PBDEs})$ in the eggs to fillet samples (presented as distribution coefficients between paired egg and fillet or D_{EF} hereafter) were 0.9–10.8. Except for Chinook salmon, D_{EF} values of individual PBDE congeners were negatively correlated with the number of bromines of the congeners (*p* < 0.05, Spearman correlation coefficient between −0.58 and −0.96, Figure S1). Chinook salmon and Coho salmon had significantly different D_{EF} values from all other fish species studied (*p* < 0.002, ANOVA with Tukey hoc multiple comparisons), but no significant difference was observed between brown trout and rainbow trout (*p* = 0.26).

Congeners (BDE-47, -99, and -100) associated with the PentaBDE commercial mixture contributed ~85% of the Σ_{14} PBDEs (Figure 2a). BDE-47 was the predominant congener and had similar contributions to Σ_{14} PBDEs in the eggs and fillets (57 and 55%, respectively). Good correlations (*r* > 0.99) were observed between $C_{WW}(\text{BDE-47})$ and $C_{WW}(\Sigma_{14}\text{PBDEs})$. BDE-47 can thus serve as a good indicator to assess Σ_{14} PBDEs in eggs, fillet, and the corresponding dietary exposure. On the basis of the data from this study, we found the relationships of $C_{WW}(\Sigma_{14}\text{PBDEs}) = (1.67 \pm 0.07) \times C_{WW}(\text{BDE-47}) + (2.77 \pm 2.90)$, $R^2 = 0.985$ for eggs, and $C_{WW}(\Sigma_{14}\text{PBDEs}) = (2.04 \pm 0.06) \times C_{WW}(\text{BDE-47}) + (3.41 \pm 1.38)$, $R^2 = 0.993$ for fillet (Figure S2).

Lipid (mass fraction) normalized concentrations (C_{LW}) of Σ_{14} PBDEs in the eggs ranged from 410 to 1380 ng/g_{lipid}

(mean/median = 930/990 ng/g_{lipid}). $C_{LW}(\Sigma_{14}\text{PBDEs})$ in the fillet ranged from 470 to 2500 ng/g_{lipid} (mean/median = 1400/1460 ng/g_{lipid}; Figure 3a). In contrast to $C_{WW}(\Sigma_{14}\text{PBDEs})$, $C_{LW}(\Sigma_{14}\text{PBDEs})$ values were lower in eggs than in the corresponding fillet (paired *t* test one-tailed *p* = 0.0015); only one pair of rainbow trout samples had $C_{LW}(\Sigma_{14}\text{PBDEs})$ higher in the eggs than the corresponding fillet. The $C_{LW}(\Sigma_{14}\text{PBDEs})$ ratios of eggs to fillet ranged from 0.4 to 1.2, and no significant differences were observed among the fish species (ANOVA *p* = 0.094).

PBDE congener profiles in the fish egg and fillet samples were dominated by BDE-47, a major congener of the penta-BDE technical mixture (Figure 2). BDE-209 was measured at low concentrations when infrequently detected in the fish fillet and egg samples. This is in contrast to high concentrations measured in abiotic environments such as sediment and wastewater effluent.^{47,48} The lower concentrations of BDE-209 in fish fillet and eggs than in abiotic environments can be attributed to (1) lower bioavailability of BDE-209 than other PBDE congeners due to higher particle bound or dissolved organic carbon bound fractions, which limit accumulation through skin and gill;^{49,50} (2) degradation of BDE-209 to lighter PBDE congeners in the fish body;^{51,52} and (3) higher affinity of BDE-209 for liver than for muscle and other fish tissues.^{53,54}

We also noted that for most paired egg and fillet samples, heavier PBDE congeners have lower distribution coefficients between egg and fillet, suggesting that the equilibrium partition into the lipid of egg and fillet cannot fully explain the chemical distribution between eggs and fillet. Transfer of heavier PBDE congeners from the body to eggs can be kinetically limited and may not reach equilibrium between body tissues and eggs.⁴⁶ For Chinook salmon, we did not observe more preferential distribution of lighter PBDE congeners to the eggs. The unique characteristic for Chinook salmon is the depletion of lipid storage during spawning.⁴⁶

Concentrations and Congener Profiles of PCNs.

Concentrations of 25 PCN congeners (4 coeluted) in the paired egg and fillet samples analyzed in this study are presented in Table S4. PCN-31, -46, -48, -70, and -75 were detected in <30% of either the eggs or fillet samples. Although PCN-13, -27, -28, -36, -63, and -74 were detected in >50% of the egg samples, they were detected in <40% of the fillet samples, and the total concentrations of these 6 PCNs accounted for only 0.3–2.2% of $\Sigma_{20}\text{PCNs}$. Our further data analysis thus focused on the other 14 PCN congeners. Total concentrations of the 14 PCN congeners in the egg samples ($\Sigma_{14}\text{PCNs}$) ranged from 0.28 to 1.67 ng/g ww (mean/median = 0.92/0.95 ng/g ww). $\Sigma_{14}\text{PCNs}$ in the fillet samples were lower and ranged from 0.05 to 0.66 ng/g ww (mean/median = 0.34/0.34 ng/g ww; Figure 1b).

On a wet weight basis, each of the PCN congeners had higher concentrations in egg than fillet samples. D_{EF} of the PCN congeners ranged from 1.2 to 10.4. Chinook salmon had significantly higher D_{EF} (4.5–10.4) than Coho salmon (1.4–8.4), brown trout (1.4–2.2), and rainbow trout (1.2–2.8) (*p* < 0.006; two-factor (congeners and species) ANOVA and Tukey post hoc multiple comparison). D_{EF} of brown trout and rainbow trout showed no significant differences (*p* > 0.05). PCN-52/60, PCN-42, and -66/67 accounted for >80% of $C_{WW}(\Sigma_{14}\text{PCNs})$ for both egg and fillet samples (Figure 2b). PCN-52 and PCN-60 were more abundant than the other PCN

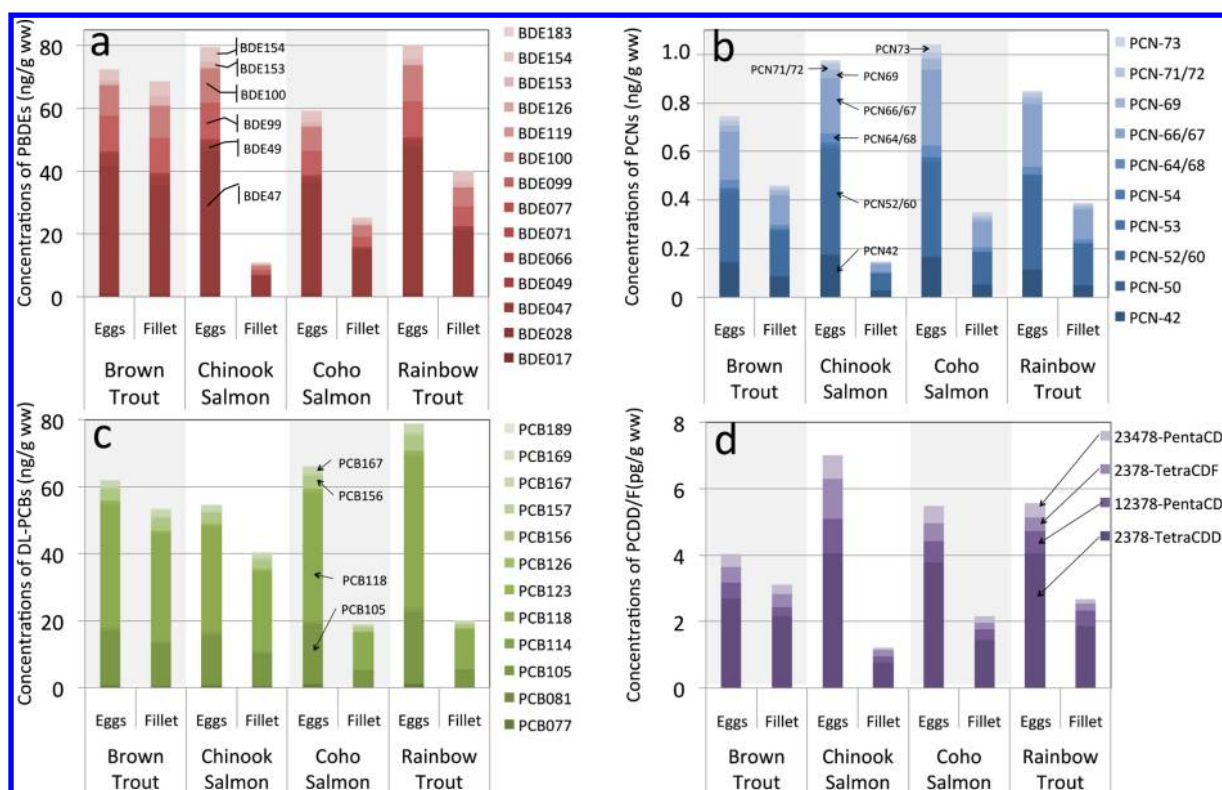


Figure 2. Wet weight-based concentrations and congener profiles of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), and (d) polychlorinated dibenzyl dioxins/furans (PCDD/Fs) in paired samples of fish eggs and skinless and boneless fillets of brown trout, Chinook salmon, Coho salmon, and rainbow trout.

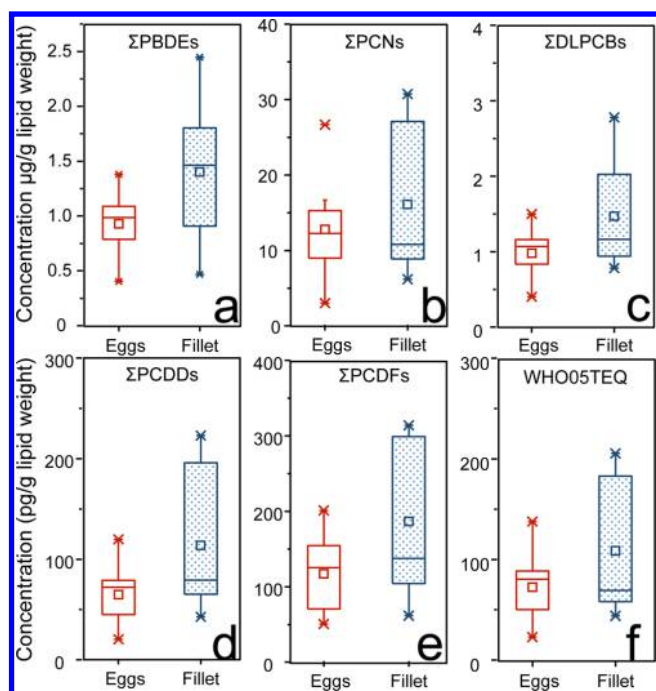


Figure 3. Lipid weight normalized concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and (f) World Health Organization 2005 toxic equivalents (WHO05TEQ) in paired fish eggs and skinless and boneless fillet samples analyzed in this study.

congeners in both egg and fillet samples and contributed 35–55% of $C_{WW}(\Sigma_{14}PCNs)$.

$C_{LW}(\Sigma_{14}PCNs)$ in the eggs ranged from 2.9 to 26.5 ng/g_{lipid} (mean/median = 12.7/12.1 ng/g_{lipid}). $C_{LW}(\Sigma_{14}PCNs)$ in the fillet ranged from 5.9 to 30.4 ng/g_{lipid} (mean/median = 15.9/10.6 ng/g_{lipid}; Figure 3b). There was no significant difference in lipid-normalized $\Sigma_{14}PCN$ concentrations between the eggs and corresponding fillet ($p = 0.051$; paired t test one-tailed). The ratios of $C_{LW}(\Sigma_{14}PCNs)$ in eggs to fillet ranged from 0.5 to 1.3, and no significant differences were observed among the species ($p = 0.16$; ANOVA).

Concentrations and Congener Profiles of dlPCBs. Wet weight-based concentrations of 12 individual dlPCB congeners ($C_{WW}(\Sigma_{12}dlPCBs)$) are presented in Table S5. All dlPCBs were detected in all egg and fillet samples. $C_{WW}(\Sigma_{12}dlPCBs)$ in the eggs ranged from 30 to 128 ng/g ww (mean/median = 75/78 ng/g ww) and in the fillets ranged from 6 to 85 ng/g ww (mean/median = 34/33 ng/g ww; Figure 1c). All eggs had higher $C_{WW}(\Sigma_{12}dlPCBs)$ than the corresponding fillets. D_{EF} of $\Sigma_{12}dlPCBs$ (ww) for the paired eggs and fillets ranged from 1.0 to 6.4, and significant differences existed among all of the species (p range <0.001–0.01; two-factor (congeners and species) ANOVA and Tukey post hoc multiple comparisons). Chinook salmon had the largest D_{EF} (5.6–7.1), followed by Coho salmon (0.8–5.8), rainbow trout (1.0–2.5), and brown trout (0.7–1.6). D_{EF} values of individual dlPCB congeners in brown trout, Coho salmon, and rainbow trout were negatively correlated with the number of chlorines in congeners ($p < 0.01$, Spearman correlation coefficients between -0.80 and -0.94). No significant correlations were observed for Chinook salmon ($p > 0.06$).

Two mono-ortho congeners, PCB-118 and -105, were the most abundant dlPCBs, and they contributed 54–62% and 24–30% of Σ_{12} dlPCBs, respectively, in both eggs and fillets (Figure 2c). Contributions of all the other individual dlPCBs in the eggs and fillets were <15%. The non-ortho congeners (i.e., PCB-77, -81, -126, and -169) accounted for only 0.8–2% of Σ_{12} dlPCBs in the eggs and fillets. The two most toxic dlPCBs, PCB-126 and -169, contributed 0.2–0.4% and 0.01–0.04% of Σ_{12} dlPCBs, respectively, in the eggs and fillets. These observations are similar to those reported for fillets of a wide variety of fish.⁵⁵

The lipid weight-based concentrations C_{LW} (Σ_{12} dlPCBs) were lower in the eggs compared to the fillets (ranging from 400 to 1500 and from 780 to 2800 ng/g_{lipid} for the eggs and fillets, respectively; $p = 0.008$, paired t test one-tailed; Figure 3c), except for one egg–fillet pair of Coho salmon as well as of rainbow trout. The ratios of C_{LW} (Σ_{12} dlPCBs) between paired eggs and fillets ranged from 0.4 to 1.3, and no significant differences were observed among the species ($p = 0.13$, ANOVA).

Concentrations and Congener Profiles of PCDD/Fs.

Wet weight-based concentrations of 7 PCDD and 10 PCDF congeners measured in this study are presented in Tables S6 and S7, respectively. Hexa-, hepta-, and octa-PCDD/Fs were below the detection limits in all egg and fillet samples and were not considered in the data analysis. 2,3,7,8-TetraCDD and 1,2,3,7,8-pentaCDD were detected in all egg samples and >70% of the fillet samples. 2,3,7,8-TetraCDD had the highest concentrations among all PCDD/Fs (Figure 2d). C_{ww} of 2,3,7,8-tetraCDD in the eggs ranged from 0.7 to 7.4 pg/g ww (mean/median = 3.6/3.5 pg/g ww) and were higher than the corresponding fillets, which ranged from <0.2 to 4 pg/g ww (mean/median = 1.6/1.5 pg/g ww). Concentrations of 1,2,3,7,8-pentaCDD were lower than those of 2,3,7,8-tetraCDD in both egg and fillet samples (0.3–1.4 and <0.2–0.53 pg/g ww, respectively). 2,3,7,8-TetraCDF was the most abundant PCDF and detected in all egg and fillet samples. C_{ww} values of 2,3,7,8-tetraCDF in the eggs were 1.4–13.0 pg/g ww (mean/median = 6.1/6.2 pg/g ww) and in the fillets were 0.7–5.6 pg/g ww (mean/median = 2.4/2.4 pg/g ww). 2,3,4,7,8-PentaCDF was detected in all egg samples (0.7–3.6 pg/g ww), but quantifiable levels in the fillets were found for only one brown trout and one Coho salmon (2.0 and 1.4 pg/g ww, respectively). 1,2,3,7,8-PentaCDF was measured in the eggs at <0.15–0.7 pg/g ww, but was below the detection limit in all of the fillets.

D_{EF} of 2,3,7,8-tetraCDD, 1,2,3,7,8-pentaCDD, and 2,3,7,8-tetraCDF, three PCDD/Fs frequently detected in both egg and fillet samples, ranged from 0.9 to 7.3, and there were species-specific differences ($p < 0.001$, two-factor ANOVA). Tukey post hoc multiple comparisons indicated D_{EF} of the three PCDD/Fs in Chinook salmon (4.8–7.3) were higher than in the other fish species ($p < 0.001$). Coho salmon had slightly higher ($p = 0.03$) D_{EF} (1.1–5.2) than brown trout (0.9–2.6) but had no difference ($p = 0.23$) when compared with D_{EF} of rainbow trout (1.3–2.4).

Lipid-normalized concentrations of Σ PCDD/Fs (Figure 3d,e) in the eggs ranged from 70 to 300 pg/g_{lipid} (mean/median = 160/170 pg/g_{lipid}) and in the fillets ranged from 80 to 380 pg/g_{lipid} (mean/median = 200/160 pg/g_{lipid}). No significant differences in C_{LW} (Σ PCDD/Fs) were found between the paired eggs and fillets ($p = 0.06$, paired t test one-tailed). The ratios of C_{LW} (Σ PCDD/Fs) in eggs to fillets

ranged from 0.6 to 1.4, and no significant differences were observed among the species ($p = 0.073$, ANOVA).

Toxic Equivalent (TEQ). TEQs based on WHO 2005 toxic equivalent factors (TEF)⁴³ and the measured concentrations of

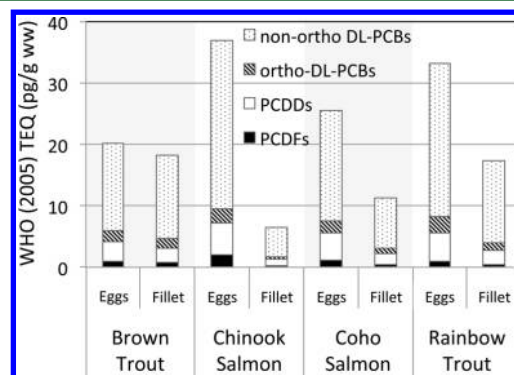


Figure 4. Contributions of dioxin-like polychlorinated biphenyls (dlPCBs) and polychlorinated dibenzyl dioxins/furans (PCDD/Fs) to WHO-2005 toxic equivalents (TEQ) in paired samples of fish eggs and skinless and boneless fillet of brown trout, Chinook salmon, Coho salmon, and rainbow trout.

PCDD/Fs and dlPCBs in the paired eggs and fillets are presented in Table S8. The TEQs for the eggs ranged from 9.3 to 53.0 pg/g ww (mean/median = 27.5/25.4 pg/g ww) and for the fillets ranged from 3.4 to 31.0 pg/g ww (mean/median = 13.4/13.0 pg/g ww). The contributions of dlPCBs to the TEQs for the eggs and fillets were greater (>75%) than those of PCDDs and PCDFs, which contributed 6–20% and 2–7%, respectively (Figure 4). Contributions of dlPCBs to the TEQs were dominated by PCB-126, a non-ortho dlPCB with the highest TEF among all dlPCB congeners. Although PCB-126 accounted for only <0.5% of Σ_{12} dlPCBs in concentration, its contribution to the TEQ represented >80% of the total contribution by dlPCBs due to the higher TEF (0.1) value than for the other dlPCBs. After PCB-126, 2,3,7,8-TCDD was the second largest contributor to TEQ and accounted for 4–17% and 3–15% of TEQ for the eggs and fillets, respectively. These results are in agreement with the observations reported in previous studies.^{55–57}

D_{EF} of TEQ ranged from 1.0 to 6.0. Species-specific differences were found for D_{EF} of TEQ ($p = 0.002$, one-factor ANOVA). Tukey post hoc multiple comparisons indicated D_{EF} values of TEQ for Chinook salmon (5.6–6.0) were higher than those for the other fish species ($p < 0.002$). No significant differences ($p > 0.05$) in D_{EF} of TEQ were found among Coho salmon (1.3–3.0), brown trout (1.8–2.0), and rainbow trout (1.0–1.4).

Implication for Dietary Exposures. The part of fish consumed may vary from skin-removed fillet to organs to eggs.^{13,27} As indicated by chemical analysis of paired fillet and egg samples, eggs have higher wet weight-based concentrations of the persistent organohalogenes. To keep exposure to these contaminants under a safe level, advised maximum meals of the corresponding fillet need to be reduced when one consumes fish eggs. As determined by the toxicologically relevant doses and concentrations of the persistent organohalogenes in the fillet, maximum meals of fillet within a given period (e.g., per month) were limited by TEQs. On the basis of the WHO 2005 TEQs and additional contributions of TEQs from PCNs as calculated using the TEFs suggested by previous studies (PCN-

54, 1.7×10^{-4} ; PCN-64/68, 1.5×10^{-4} ; PCN-66/67, 2.3×10^{-3} ; PCN-69, 2.0×10^{-3} ; PCN-71/72, 7.0×10^{-6} ; PCN-73, 1.0×10^{-3}),^{58,59} the amount of eggs with contaminant levels equivalent to one meal of fillet was determined (Figure 5).

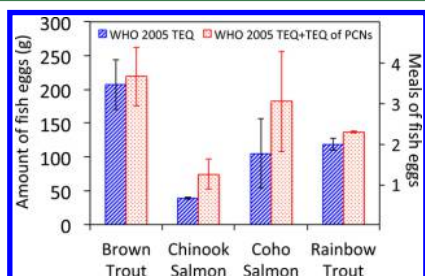


Figure 5. Amount (g, left axis) and meals (60 g/meal) of fish eggs that are equivalent to one typical meal of fish (227 g or 8 oz or half a pound fillet per meal used in Ontario's Fish Consumption Advisories¹¹). Calculations were based on WHO 2005 TEQ and WHO 2005 TEQ plus TEQ contributed by polychlorinated naphthalenes (PCNs).

There were fish species-specific differences in the amount of fish eggs with contaminant levels equivalent to one meal of fillet. On the basis of WHO 2005 TEQ for dioxins/furans/dlPCBs, one can eat only 39 ± 2 g Chinook salmon eggs instead of one meal of the fillet without exceeding a safe dosage. In contrast, as much as 207 ± 37 , 105 ± 51 , and 119 ± 9 g eggs of brown trout, Coho salmon, and rainbow trout, respectively, would equal one meal of the corresponding fillet. Although advised maximum meals of fillets would decrease when PCNs are also considered in the TEQ scheme, the amount of eggs equivalent to one fish meals would increase (Figure 5) due to

the lower distribution coefficient (0.5–1.3) between eggs and fillet for the TEQ contributed by PCNs than that (1.0–6.0) for WHO 2005 TEQ. In this case, amounts of eggs one can consume by replacing one meal of brown trout, Chinook salmon, Coho salmon, and rainbow trout fillet were calculated at 219 ± 43 , 75 ± 22 , 182 ± 74 , and 137 ± 2 g, respectively.

To our knowledge, there is no widely applicable generic information on the amount of fish eggs consumed in a meal; however, up to 60 g of fish eggs per meal has been estimated considering cholesterol in fish eggs and percent daily values recommended for cholesterol by the U.S. Department of Agriculture.⁶⁰ Accordingly, for every fish egg meal (60 g), reductions of 0.3 ± 0.1 , 1.5 ± 0.1 , 0.6 ± 0.1 , and 0.5 ± 0.0 in advised fillet meal(s) of brown trout, Chinook salmon, Coho salmon, and rainbow trout are recommended, respectively, based on the WHO 2005 TEQs.

Note that for fatty fish in Lake Ontario in general, total PCBs are the major contaminant restricting consumption frequency recommended by the advisories.¹¹ Although total PCBs were not considered in this study, dlPCBs (a subset of among the most toxic PCBs) were included in the TEQ values. Because there is a linear relationship between total PCBs and dlPCB-TEQ⁵⁶ and because dlPCB-TEQ are typically the major contributors to total TEQs for the Great Lakes fish,⁵⁷ the one fillet meal equivalent values derived for fish eggs in this study should be appropriate for the species considered.

We recognize the limited sample sizes considered in this study, and as such, present the results as a preliminary chemical assessment and consumption advisory of fish eggs. The results highlight the importance of reducing consumption of fillet if fish eggs are also consumed. There were large variations in contaminant levels and distributions between fillet and eggs

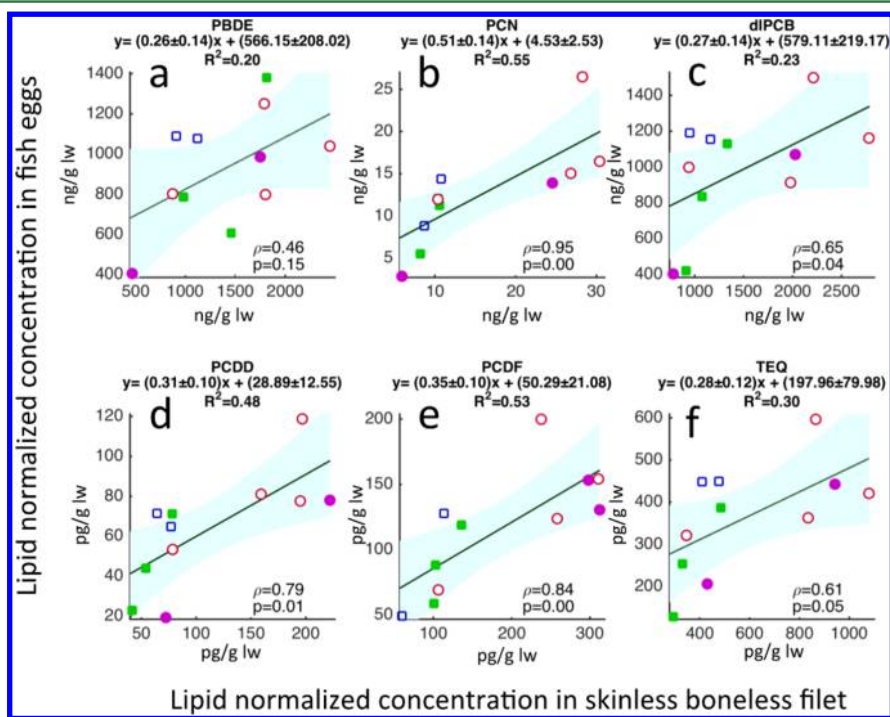


Figure 6. Relationship between lipid-normalized concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and World Health Organization (WHO) 2005 toxic equivalents (TEQs) in paired fish fillet (x-axis) and eggs (y-axis): green filled squares, brown trout; blue hollow squares, rainbow trout; red filled circles, Coho salmon; purple hollow circles, Chinook salmon. ρ and p are the coefficient of Spearman correlation and its significance, respectively.

even within a fish species. As such, a more comprehensive follow-up study is recommended to strengthen the findings of this study.

Influence of Biological Parameters. Concentrations of PBDE, PCN, dlPCB, and TEQ in the fillet samples had significant positive correlations ($p < 0.05$) with the lipid contents in fillet (Figure S3a–c,f). Lipid content can explain 75% variability in the PBDE and dlPCB concentrations, 66% variability in TEQs, and 46% variability in the PCN concentrations. PCDD/F concentrations in fillet appeared to increase with lipid contents, yet such correlations were insignificant at the 95% confidence interval (Figure S3d,e). Neither weight nor length of fish had significant correlations with chemical concentrations in fillet (Figures S4 and S5). In contrast to fillet, lipid content showed no statistically significant correlation with concentrations of the chemicals in eggs (Figure S6).

Significant correlations existed between lipid-normalized concentrations in fillet and eggs for all contaminants except PBDE (Figure 6). However, only 23–55% of the variability in the contaminant distributions between fish fillet and eggs can be explained by the differences in lipid content between them (Figure 6). Furthermore, lipid-normalized fillet concentrations were greater than the corresponding egg concentrations (Figure 3), which is in contrast to a previous suggestion that lipid-normalized concentrations of POPs in eggs and adult fish are generally equal.³¹ The results from this study suggest that the lipid content in fillet and eggs and/or lipophilicity of a contaminant are not primary determinants for contaminant concentrations in fish eggs. This also implies that equilibrium partitioning determined by lipid contents of biological tissues may not be appropriate to predict concentrations of POPs in one tissue from another. As such, to evaluate dietary exposure to contaminants in fish eggs and to propose consumption advisory, measured data rather than predictions based on equilibrium partitioning should be used.

Some PBDEs can be debrominated by fish,⁵² which may lead to its redistribution between eggs and fillet independent of the lipid content. In addition to chemical degradation, other factors that can potentially affect chemical distributions between fish fillet and eggs include (1) nonequilibrium or unsteady state between fish body and eggs;⁴⁶ (2) different types of lipid in fish eggs and fillet;^{61,62} (3) influence of other nonlipid organic matter;⁶³ and/or (4) depletion of body lipid due to the long-range migration of fish during spawning seasons.⁴⁶

In summary, we examined the wet weight-based and lipid-normalized concentrations of PCDD/Fs, dlPCBs, PBDEs, and PCNs in paired fish fillet and eggs of four fish species from a tributary to Lake Ontario, Canada. On a lipid-normalized basis, concentrations of all contaminants in all fish species considered in the study were about 50–75% lower in eggs than in the fillets. However, on a wet weight basis, concentrations were higher in the eggs compared to the corresponding fillets. These higher levels result in lower comparable amounts of fish eggs than fillets that could be safely consumed without exceeding the tolerable daily intakes. Most importantly, the results indicated that a simple equilibrium and lipid-based partitioning model may be an ineffective tool to predict concentrations of the studied POPs in fish eggs from the corresponding fillet concentrations.

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jafc.6b00089.

Additional seven tables and six figures (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We acknowledge the help of Steve Petro, Chris Mahon, Emily Awad, Eric Reiner, Terry Kolic, and Karen MacPherson of the Ontario Ministry of Environment and Climate Change.

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Supporting Information

Persistent organohalogens in paired fish fillet and eggs: Implications for fish consumption advisories

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Number of Pages: 17

Number of Figures: 6

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Details on chemical extractions and analyses

Concentrations of 17 PBDEs were measured using the gas chromatography with high resolution mass spectrometric (GC-HRMS, resolution >10 000) based on OMOE method BDE-E343041 and Kolic et al (2009). ~5 g of samples were mixed with diatomaceous earth and extracted and cleaned with an automated Fluid Management System (FMS, Waltham, MA). Pressurized liquid extraction was conducted in a 40 mL stainless steel cells filled with the sample and diatomaceous earth mix using hexane and DCM. The sample extracts were then concentrated to ~ 1 mL using a rotary evaporator, transferred to 40-mL vials and diluted to ~35 mL with hexane. Samples were loaded on columns pre-packed with Teflon silica (FMS, PCB-HCDS-ABN) followed by carbon/celite (FMS, PCBC-CCE) for the fractionation of the target compounds. PBDEs were separated using HP 7890A GC with HRMS. Samplers were injected using splitless mode onto a DB5-HT column (15 m × 0.25 mm × 0.10 µm film thickness, J&W Scientific, Folsom, CA). The GC conditions were: 110°C hold for 1 min, ramp to 200°C at 40.0°C/min, ramp to 330°C at 10°C/min hold for 5.5 min. The carrier gas was He with a constant flow rate of 1.0 mL/min constant. Mass spectral conditions and criteria for positive identification are summarized in Table II. The non-BDE halogenated flame retardants were analyzed on Agilent Technologies 6890 Plus GC interfaced to a Waters Autospec-Premier HRMS (Waters) in EI positive with an electron energy of 40 eV. Split/splitless injection was used with a direct injection sleeve (1.5 mm i.d.; Supelco). The chromatographic separation for the non-BDE HFRs was carried out on a DB-5HT 15 m × 0.25 mm × 0.10 µm (J&W Scientific). The GC condition was: 100°C for 1 min, increase to 210°C at 10.0°C/min, to 310°C at 20°C/min and then hold for 6 min. Helium (1.0 mL/min) was used as the carrier gas.

PCNs, PCDD/Fs and dlPCBs were measured using GC-HRMS based on OMOE method PCN-E343142 and DFPCB-E341840. Homogenized fish tissue samples (~5 g ww) were fortified with ¹³C-PCNs (¹³C-PCNs 42, 27, 52, 64, and 75; Cambridge Isotope Laboratories), ¹³C-PCDD/Fs, and ¹³C-DL-PCBs (Wellington Laboratories). Samples were digested in hydrochloric acid and extracted with hexane. The extracts were passed through an anhydrous sodium sulfate/sulfuric acid-silica column, followed column cleanup. The extract was loaded first to a column containing 1.5 g of 10% [w/w] silver nitrate, 1.0 g of activated silica, 2.0 g of 33% [w/w] sodium hydroxide/silica, 1.0 g of activated silica, 4.0 g of 44% [w/w] sulfuric acid/silica, 2.0 g of activated silica, and 2.0 g of anhydrous sodium sulfate and then eluted with hexane. The eluted sample was further cleaned up with a second column containing 0.35 g of 5% (w/w) carbon-activated silica, which was eluted with 40 ml of 25% (v/v) dichloromethane/hexane (fraction 1; mono-ortho-PCBs). The column was then inverted to reverse the flow and eluted with 160 ml of toluene to isolate the PCDD/Fs, coplanar PCBs, and PCNs (fraction 2). The two fractions of extracts were reduced in volume by rotary and nitrogen evaporators.

The samples were analyzed by gas chromatography-high-resolution mass spectrometry using a Micromass Autospec Ultima mass spectrometer coupled to a Hewlett-Packard HP6890 gas chromatograph (Agilent Technologies). PCDD/Fs and DL-PCBs were separated with a DB-5 column (length, 40 m; inner diameter, 0.18 mm; film thickness, 0.18 µm; J&W Scientific) and PCNs were separated with a RTX-Dioxin-2 column (length, 40 m; inner diameter, 0.20 mm; film thickness, 0.18 µm; Restek).

Samples were injected in splitless mode with He carrier gas (1.5 cm/s PCDD/Fs, 0.8 cm/s for PCNs). The injector- and transfer-line temperatures were maintained at 280°C (250°C for PCNs) and 300°C, respectively. The GC temperature program for PCDD/Fs and DL-PCBs was 100°C for 1 min, from 100 to 200°C at 40°C/min, from 200 to 235°C at 3°C/min, hold at 235°C for 10 min, from 235 to 300°C at 6°C/min, and hold at 300°C for 12 min; for PCNs: 100°C for 1 min, from 100 to 200°C at 25°C/min, hold at 200°C for 5 min, from 200 to 235°C at 2.5°C/min, hold at 235°C for 3 min, from 235 to 267°C at 3°C/min, from 267 to 300°C at 10°C/min, and hold at 300°C for 3 min.

PCDD/Fs and DL-PCBs were quantified using isotope dilution (¹³C-PCDD/Fs and ¹³C-DL-PCBs). PCN-27, 42, 52, 67, 73 and 75 were quantified using isotope dilution, while the other PCN congeners were quantified by internal standard methods.

FIGURES

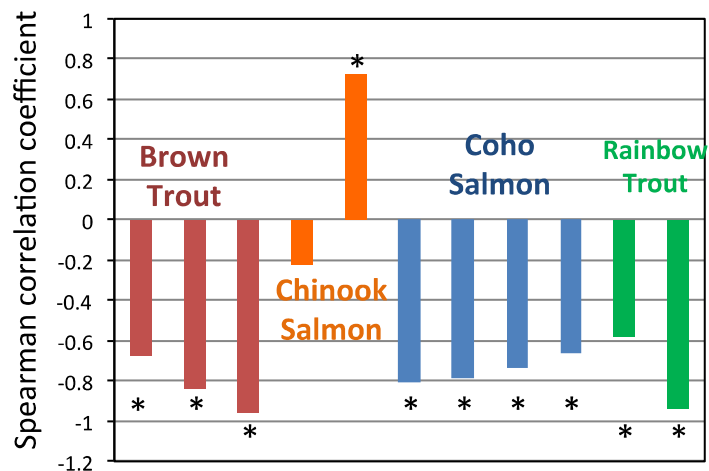


Figure S1. Correlations between fish egg/fillet distribution coefficients (D_{EF}) and the number of bromines in the PBDE congeners. The stars indicate significant spearman correlations ($p < 0.05$).

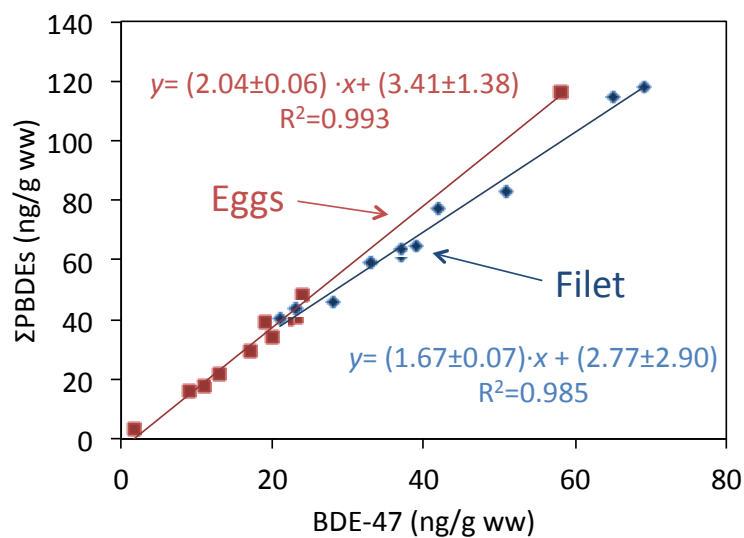


Figure S2. Relationships between BDE-47 with total PBDE concentrations in fish fillet and egg samples

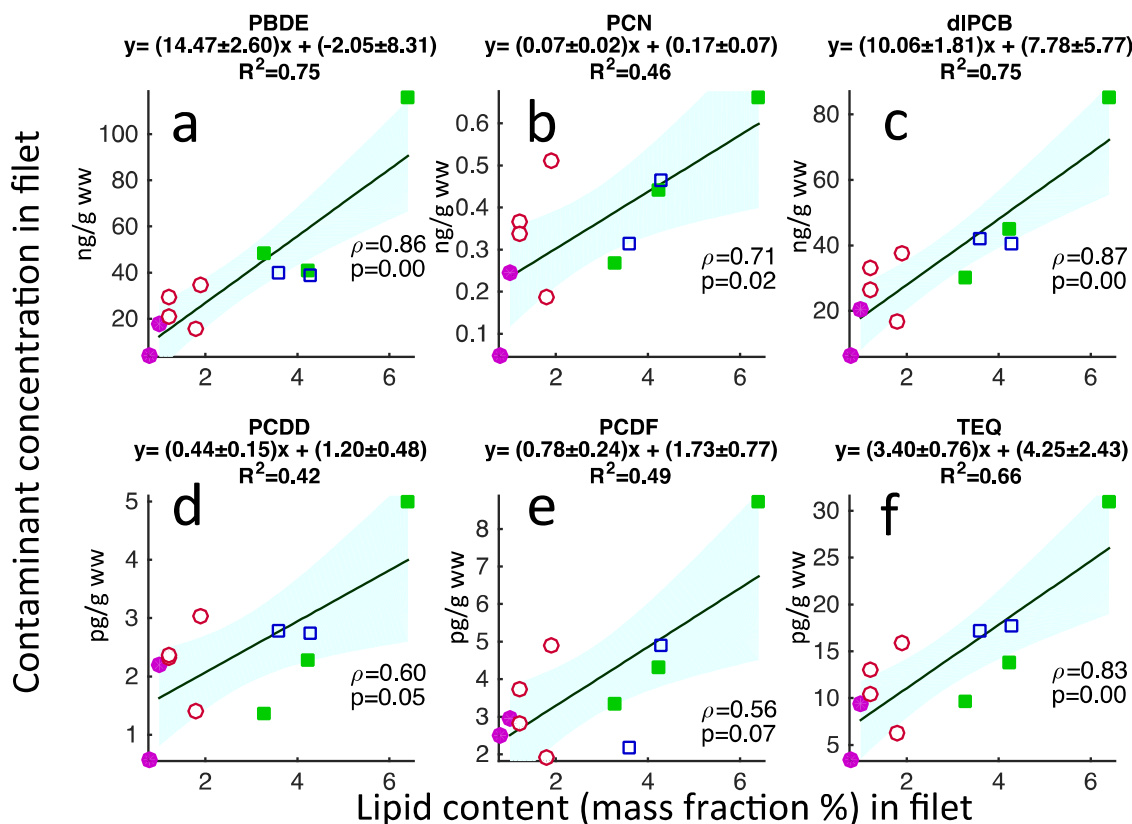


Figure S3. Relationship between lipid content and wet weight based concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dIPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and World Health Organization (WHO) 2005 toxic equivalents (TEQs) in the skinless and boneless fillet samples. Green filled square: Brown Trout; blue hollow square: Rainbow Trout; red filled circle: Coho Salmon; purple hollow circle: Chinook Salmon. ρ and p are coefficient of Spearman correlation and its significance.

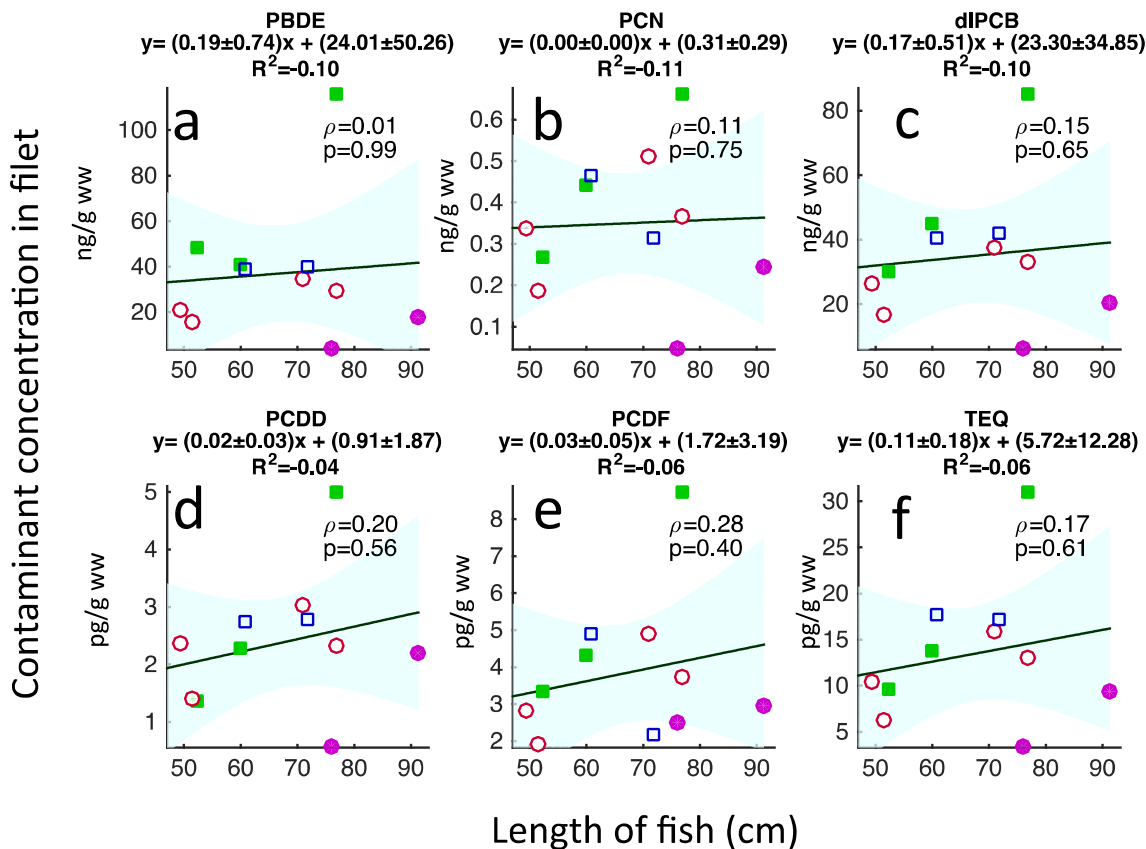


Figure S4. Relationship between fish length (cm) and wet weight based concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and World Health Organization (WHO) 2005 toxic equivalents (TEQs) in the skinless and boneless fillet samples. Green filled square: Brown Trout; blue hollow square: Rainbow Trout; red filled circle: Coho Salmon; purple hollow circle: Chinook Salmon. ρ and p are coefficient of Spearman correlation and its significance.

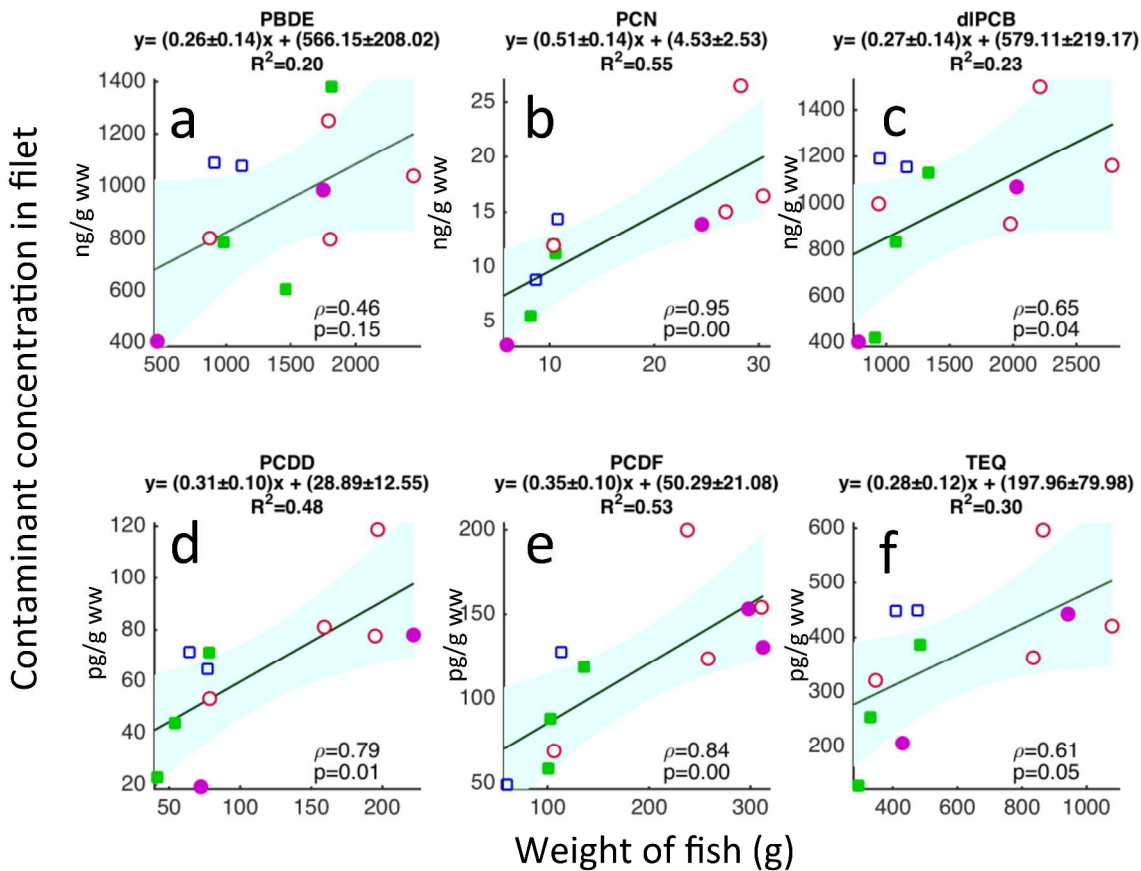


Figure S5. Relationship between fish weight (cm) and wet weight based concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dIPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and World Health Organization (WHO) 2005 toxic equivalents (TEQs) in paired skinless and boneless fillet samples. Green filled square: Brown Trout; blue hollow square: Rainbow Trout; red filled circle: Coho Salmon; purple hollow circle: Chinook Salmon. ρ and p are coefficient of Spearman correlation and its significance.

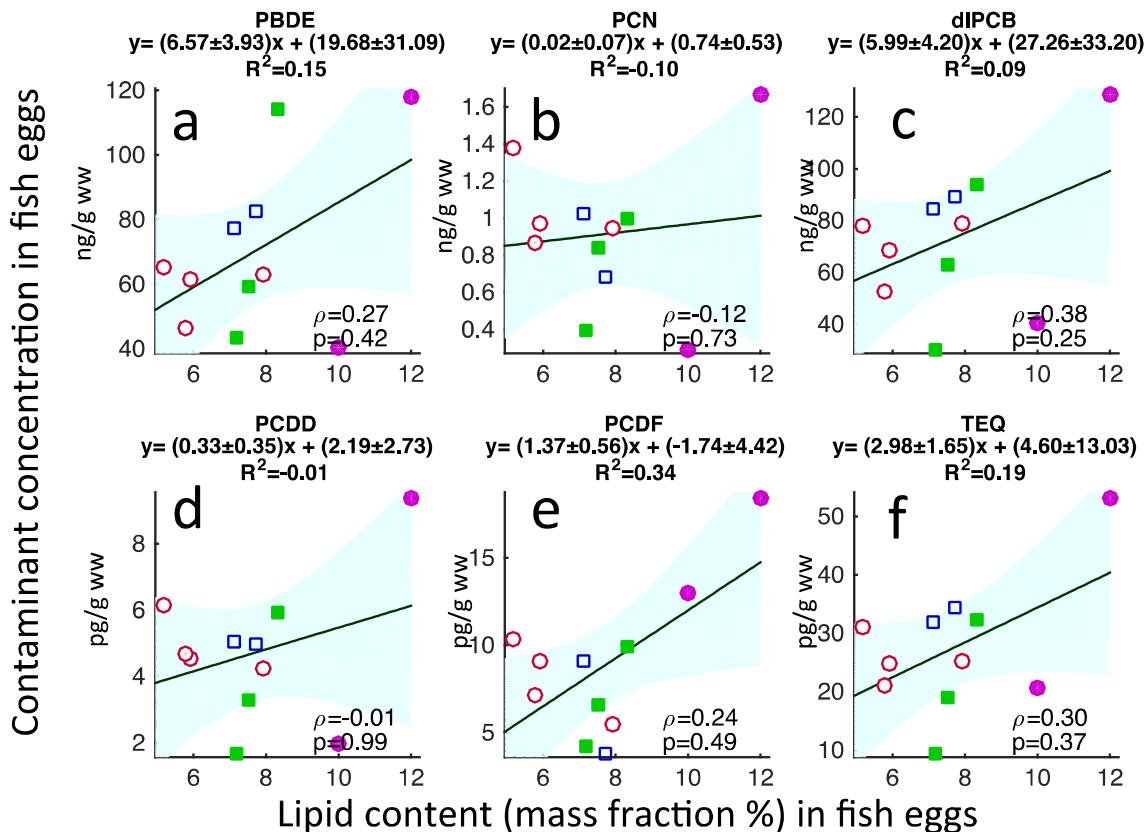


Figure S6. Relationship between lipid content and wet weight based concentrations of (a) polybrominated diphenyl ethers (PBDEs), (b) polychlorinated naphthalenes (PCNs), (c) dioxin-like polychlorinated biphenyls (dlPCBs), (d) polychlorinated dibenzodioxins (PCDDs), (e) polychlorinated dibenzofurans (PCDFs), and World Health Organization (WHO) 2005 toxic equivalents (TEQs) in the fish egg samples. Green filled square: Brown Trout; blue hollow square: Rainbow Trout; red filled circle: Coho Salmon; purple hollow circle: Chinook Salmon. ρ and p are coefficient of Spearman correlation and its significance.

TABLES

Table S1. Recoveries and method detection limits (MDLs) of analytes.

Chemical	Recovery %	MDL (ng/g)	Chemical	Recovery %	MDL (pg/g)	Chemical	Recovery %	MDL (pg/g)	Chemical	Recovery %	MDL (pg/g)
BDE-17	82	0.02	PCN28	99	0.9	2378-TCDF	95	2	PCB077	115	6
BDE-28	95	0.01	PCN36	96	0.6	12378-PCDF	98	8	PCB081	105	12
BDE-47	99	0.02	PCN42	96	1.3	23478-PCDF	101	8	PCB105	142	9
BDE-49	82	0.05	PCN50	100	0.9	123478-HxCDF	105	6	PCB114	100	126
BDE-66	102	0.15	PCN52/60	95	0.4	123678-HxCDF	99	6	PCB118	118	8
BDE-71	84	0.04	PCN53	104	1.8	123789-HxCDF	96	5	PCB123	100	36
BDE-77	104	0.01	PCN54	101	1.8	234678-HxCDF	103	5	PCB126	102	6
BDE-85	99	0.02	PCN64/68	93	0.4	1234678-HpCDF	95	3	PCB156	102	9
BDE-99	98	0.17	PCN66/67	93	0.3	1234789-HpCDF	105	5	PCB157	96	10
BDE-100	89	0.04	PCN69	95	0.6	OCDF	101	29	PCB167	103	7
BDE-119	104	0.02	PCN71/72	88	1.7	2378-TCDD	102	2	PCB169	101	6
BDE-126	102	0.01	PCN73	92	0.5	12378-PCDD	105	9	PCB189	115	12
BDE-138	98	0.07	PCN74	82	1.4	123478-HxCDD	101	6			
BDE-153	90	0.01				123678-HxCDD	88	6			
BDE-154	84	0.02				123789-HxCDD	100	19			
BDE-183	86	0.02				1234678-HpCDD	97	2			
BDE-209	88	2.8				OCDD	102	12			

Table S2. Length and weight of the fish samples, and lipid content (mass fractions) of the paired fish fillet and egg samples.

Sample code		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon				Rainbow Trout	
		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Length (cm)		60	77	52.2	76.1	91.3	76.8	71	51.4	49.5	71.9	60.8
Weight (g)		2960	4870	1560	4660	6510	4950	3910	1530	900	3840	2850
Lipid	Eggs	7.5	8.3	7.2	10	12	5.9	5.8	7.9	5.2	7.7	7.1
Content (%)	Filet	4.2	6.4	3.3	0.8	1	1.2	1.9	1.8	1.2	3.6	4.3

Table S3. Concentrations of polybrominated diphenyl ethers (PBDEs, ng/g wet weight) in the paired fish fillet and egg samples

Sample code		Fish species										
		Brown Trout			Chinook Salmon		Coho Salmon			Rainbow Trout		
		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	Concentrations										
Fish Eggs	BDE-17	0.028	0.025	0.026	0.019	0.022	0.022	0.022	0.021	0.025	0.019	0.019
	BDE-28	1.1	1.6	0.76	0.54	1.8	1.2	0.92	1.1	1	1.1	1.2
	BDE-47	33	65	23	21	69	37	28	37	39	51	42
	BDE-49	3.3	5.7	2.3	0.88	4.9	2	1.4	1.3	1.5	1.6	2.9
	BDE-66	0.51	1.8	0.69	0.57	1.4	0.68	0.51	0.72	1.5	0.96	0.86
	BDE-71	0.028	0.039	0.032	0.028	0.033	0.057	0.052	0.032	0.048	0.04	0.026
	BDE-77	0.013	0.025	<0.02	0.074	0.037	0.023	<0.02	0.019	0.015	<0.02	0.027
	BDE-85	0.28	<0.02	<0.04	<0.03	<0.04	0.034	<0.03	0.073	<0.03	<0.05	<0.04
	BDE-99	11	16	7	7.5	16	7.2	5.4	9.3	8.9	9.8	13
	BDE-100	6.5	16	6.2	6.5	15	7.5	5.9	8.8	8.2	12	11
	BDE-119	0.13	0.33	0.13	0.29	0.27	0.11	0.094	0.16	0.24	0.2	0.19
	BDE-126	0.018	0.042	0.04	0.047	0.099	0.073	0.051	0.025	0.029	0.033	0.036
	BDE-138	0.064	<0.03	<0.07	<0.04	<0.05	<0.07	<0.03	0.025	<0.03	<0.06	<0.06
	BDE-153	1.1	2.4	1.1	0.83	2.9	1.6	1.2	1.3	1.3	1.6	1.7
	BDE-154	2.2	5.5	2.4	2.3	6.7	3.8	2.7	3.6	3.3	4.5	4.3
	BDE-183	0.043	0.035	0.03	0.042	0.07	0.032	0.026	0.031	0.033	0.036	0.051
	BDE-209	<2.00	<2.40	<2.00	<4.50	<1.40	<0.78	<3.70	<1.70	<1.10	<2.40	<3.20
	Σ₁₄PBDEs*	59.0	114	43.7	40.6	118	61.3	46.3	63.4	65.1	82.9	77.3
Fish fillet	BDE-17	0.012	0.022	0.019	<0.00	0.0038	0.0067	0.011	0.0029	0.0037	0.014	0.0077
	BDE-28	0.65	1.2	0.58	<0.08	0.32	0.47	0.49	0.23	0.31	0.44	0.5
	BDE-47	23	58	24	<3.50	11	17	20	9	13	22	19
	BDE-49	1.7	4.2	1.9	<0.16	0.53	0.56	0.69	0.18	0.21	0.83	1.4
	BDE-66	0.47	1.7	0.64	<0.14	0.2	0.32	0.39	0.18	0.24	0.49	0.41
	BDE-71	0.028	0.048	0.034	<0.01	0.0087	0.02	0.031	0.01	0.012	0.018	0.014
	BDE-77	0.01	0.025	0.011	0.012	0.0061	0.0074	0.0073	0.0038	<0.01	0.0088	0.013
	BDE-85	0.013	<0.02	<0.01	<0.04	<0.01	<0.02	0.0087	0.017	0.013	<0.01	<0.01
	BDE-99	6.5	19	8.4	<1.90	2.1	3.7	4.6	2.5	2.7	5.4	6.7
	BDE-100	5.2	18	7	<1.00	2.2	4.3	4.8	2.2	3	6.3	6
	BDE-119	0.093	0.33	0.15	0.044	0.039	0.085	0.07	0.037	0.058	0.12	0.11
	BDE-126	0.038	0.21	0.096	<0.00	0.011	<0.03	0.04	0.008	0.022	0.052	0.051
	BDE-138	<0.03	<0.04	<0.02	<0.01	<0.02	<0.03	<0.02	<0.03	<0.03	<0.01	<0.01
	BDE-153	1.3	4.8	2.2	<0.23	0.37	0.98	1	0.45	0.64	1.8	1.8
	BDE-154	2.1	8.6	3.2	<0.36	0.72	1.9	2.1	0.91	1.2	2.9	3
	BDE-183	0.035	0.061	0.045	<0.01	<0.01	<0.03	0.034	<0.01	0.068	<0.02	0.04
	BDE-209	<1.40	6.4	<1.30	<2.90	<1.00	4.8	<2.10	<2.60	<2.50	<1.30	<1.90
	Σ₁₄PBDEs	41.1	116	48.3	3.8	17.5	29.4	34.3	15.7	21.5	40.4	39.0

*Frequently detected BDE congeners summed up as Σ₁₄PBDEs are highlighted in bold

Table S4. Concentrations of polychlorinated naphthalenes (PCNs, pg/g wet weight) in the paired fish fillet and egg samples.

		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon			Rainbow Trout		
Sample code		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	Concentrations										
Fish eggs	PCN-13	0.77	0.25	0.82	1.3	0.75	0.85	0.75	0.71	<0	0.6	<0
	PCN-27	1.2	0.47	1.1	<0	<1	0.9	0.67	<0	0.45	<0	<0
	PCN-28	<0.3	0.38	0.65	0.67	0.72	0.56	0.4	<0.2	<0.3	<0.3	<0.3
	PCN-31	0.27	<0.3	<0.4	<0.2	<0.3	<0.2	<0.3	0.077	<0.4	<0.3	<0.3
	PCN-36	3.1	1.2	2.9	1.3	2.8	2.8	2	1.9	1.7	0.36	0.73
	PCN-42	180	150	97	48	300	170	130	170	190	66	160
	PCN-46	<2.2	3.2	4.1	<0.8	<3.1	<2.5	<2.7	<1.8	<2.4	<1.2	<1.4
	PCN-48	<0.2	<0.3	<0.3	<0.2	<0.3	<0.2	<0.3	<0.3	<0.3	<0.3	<0.3
	PCN-50	5.4	2.5	3.6	2.9	4.4	5.3	2.4	1.8	2.2	<0.5	0.49
	PCN-52/60	310	400	150	160	710	380	310	370	500	290	480
	PCN-53	24	10	15	5.4	24	24	14	16	13	2.8	7.3
	PCN-54	3.5	3.6	2.3	7.4	10	3.9	3.1	2.7	6.2	1.1	4.3
	PCN-63	2.9	2	1.7	1.1	3.7	1.6	1.1	0.4	0.8	0.8	2.5
	PCN-64/68	34	43	13	10	70	43	44	35	69	23	35
	PCN-66/67	210	300	88	34	420	250	260	280	450	250	270
	PCN-69	31	38	12	10	58	41	42	37	65	22	31
	PCN-70	<0.7	<1.0	<0.9	<0.6	<0.9	<0.7	0.74	<0.9	<1.3	<0.8	<1.2
	PCN-71/72	25	27	9	6.4	45	25	24	21	32	16	23
	PCN-73	20	27	4.9	<1.9	25	28	43	12	50	6.6	11
	PCN-74	1.2	2.6	<0.6	<0.8	2.6	0.91	1.1	0.36	<0.8	<0.9	1.5
	PCN-75	<1.2	<1.7	<1.6	<1.1	<0.9	<1.1	<1.0	<1.5	<1.6	<1.6	<1.3
Σ ₁₄ PCNs		843	1001	395	285	1666	970	873	946	1377	678	1022
Fish fillet	PCN-13	<0.4	<0.3	<0.3	<0.5	<0.3	<0.4	<0.4	<0.4	<0.5	<0.3	<0.3
	PCN-27	<0.3	<0.2	<0.2	<0.2	<0.2	<0.3	<0.3	<0.3	<0.3	<0.2	<0.2
	PCN-28	<0.3	<0.3	<0.2	0.096	<0.2	<0.3	<0.4	<0.3	<0.4	<0.2	0.23
	PCN-31	<0.3	0.065	<0.2	<0.3	<0.3	<0.4	0.096	0.12	<0.4	<0.2	<0.3
	PCN-36	1.6	0.97	1.2	<0.3	<0.2	1	<0.9	<0.4	<0.3	<0.3	<0.4
	PCN-42	95	100	64	8	47	65	68	33	44	29	71
	PCN-46	<1.4	<2.3	<2.1	<0.9	<1.2	<1.8	<1.7	<1.1	<1.1	<0.3	<1.1
	PCN-48	<0.3	<0.2	<0.2	<0.3	<0.3	<0.4	<0.3	<0.3	<0.4	0.1	<0.2
	PCN-50	2.7	1.4	1.8	0.39	<0.4	0.63	1.3	<0.5	<0.7	<0.3	<0.6
	PCN-52/60	170	270	110	26	110	150	180	74	120	130	210
	PCN-53	11	6	7	1.2	2.3	6.2	8.5	2.5	<0.8	<0.4	2.8
	PCN-54	1.9	2.5	1.7	1.3	1.6	0.94	1.3	<0.5	0.9	0.46	2.5
	PCN-63	1.3	1.4	0.8	<0.6	<0.5	<1.0	<1.0	<0.6	<1.3	<0.9	0.5
	PCN-64/68	16	26	7.8	1.7	9.1	14	25	6.5	17	11	17
	PCN-66/67	110	200	62	5.8	58	96	160	56	120	120	130
	PCN-69	16	24	7.4	1.5	7.7	13	22	6.8	15	11	14
	PCN-70	<0.9	<0.8	<0.8	<0.8	<0.6	<1.7	0.25	<1.0	<2.2	<1.2	<0.7
	PCN-71/72	12	17	5.6	1	6.1	8.2	13	4.3	8.4	6.6	8.3
	PCN-73	9.1	16	2.8	<1.3	3.5	11	31	3	13	5.4	6.8
	PCN-74	<0.7	1.4	<0.4	<0.4	0.24	<1.1	0.57	<0.7	<1.2	<0.5	<0.5
	PCN-75	<1.6	<1.6	<0.8	<1.2	<0.9	<2.3	<1.0	<1.8	<2.0	<0.9	<1.0
Σ ₁₄ PCNs		444	663	270	48	245	365	510	187	339	314	463

*Frequently detected PCN congeners summed up as Σ₁₄PCNs are highlighted in bold

Table S5. Concentrations of dIPCB (pg/g wet weight) in the paired fish fillet and egg samples.

Sample code			Fish Species										
			Brown Trout			Chinook Salmon		Coho Salmon				Rainbow Trout	
			000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical		Concentrations										
Fish eggs	PCB077	3,3',4,4'-tetraCB	770	810	450	570	1900	840	690	700	790	430	1100
	PCB081	3,4,4',5-tetraCB	48	84	26	43	140	58	47	48	59	63	96
	PCB105	2,3,3'4,4'-pentaCB	16000	25000	7800	12000	34000	18000	14000	20000	20000	23000	22000
	PCB114	2,3,4,4',5-pentaCB	900	1400	430	640	1900	1000	750	1200	1100	1300	1200
	PCB118	2,3'4,4',5-pentaCB	38000	55000	18000	22000	75000	40000	31000	47000	47000	53000	50000
	PCB123	2'3,4,4',5-pentaCB	970	1500	420	470	2200	1100	850	1400	880	1400	1400
	PCB126	3,3'4,4',5-pentaCB	130	220	65	160	370	170	140	180	210	260	230
	PCB156	2,3,3'4,4'5-hexaCB	3200	5200	1800	2400	6700	3800	2900	4400	4300	5000	4400
	PCB157	2,3,3'44'5'-hexaCB	540	1000	270	590	1600	790	630	890	850	1100	960
	PCB167	23',44',55'-hexaCB	1800	3100	850	1200	4100	2400	1700	2600	2400	3000	2700
	PCB169	3,3'4,4'55'-hexaCB	8.3	16	3.7	15	25	13	9.2	11	14	16	11
	PCB189	233'44'55'-heptaCB	220	390	140	210	500	290	210	390	350	390	360
Σ_{12} dIPCBs/ 10^3			62.6	93.7	30.3	40.3	128.4	68.5	52.9	78.8	78.0	89.0	84.5
Fish fillet	PCB077	3,3',4,4'-tetraCB	480	640	370	96	310	340	400	140	170	200	490
	PCB081	3,4,4',5-tetraCB	39	70	21	7.3	25	25	30	8.3	14	25	42
	PCB105	2,3,3'4,4'-pentaCB	10000	21000	7100	1900	5400	7900	9200	4200	6400	10000	9900
	PCB114	2,3,4,4',5-pentaCB	650	1300	430	100	300	470	550	240	380	610	590
	PCB118	2,3'4,4',5-pentaCB	28000	50000	18000	3400	12000	20000	22000	10000	16000	25000	24000
	PCB123	2'3,4,4',5-pentaCB	740	1300	440	74	310	580	640	240	400	640	680
	PCB126	3,3'4,4',5-pentaCB	100	220	71	27	66	94	110	45	72	130	130
	PCB156	2,3,3'4,4'5-hexaCB	2800	5600	2000	340	970	2000	2400	1100	1600	2600	2500
	PCB157	2,3,3'44'5'-hexaCB	520	1200	410	92	230	430	530	230	360	640	580
	PCB167	23',44',55'-hexaCB	1600	3400	1100	180	580	1300	1500	620	970	1700	1600
	PCB169	3,3'4,4'55'-hexaCB	8.3	20	5.5	2.6	4.4	8.6	11	3.7	5.4	12	11
	PCB189	233'44'55'-heptaCB	280	590	210	32	75	220	250	100	160	280	280
Σ_{12} dIPCBs/ 10^3			45.2	85.3	30.2	6.25	20.3	33.4	37.6	16.9	26.5	41.8	40.8

Table S6. Concentrations of polychlorinated dibenzo-p-dioxins (PCDDs, pg/g wet weight) in the paired fish fillet and egg samples.

Sample code		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon				Rainbow Trout	
		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	Concentrations										
Fish eggs	2378-tetraCDD*	2.5	4.7	0.9	0.73	7.4	3.4	3.5	3.3	4.9	4	4.1
	12378-pentaCDD*	0.45	0.7	0.26	0.66	1.4	0.68	0.57	0.52	0.81	0.69	0.65
	123478-hexaCDD	<0.1	<0.1	<0.2	<0.1	<0.2	<0.2	<0.2	<0.1	<0.1	<0.1	<0.1
	123678-hexaCDD	<0.2	<0.3	<0.2	<0.3	<0.3	<0.2	<0.3	<0.1	<0.1	<0.2	<0.1
	123789-hexaCDD	<0.1	<0.2	<0.2	<0.3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1	<0.1
	1234678-heptaCDD	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1	<0.1	<0.2	<0.2	<0.1	<0.2
	OctaCDD	<0.2	<0.3	<0.2	<0.3	<0.2	<0.3	<0.5	<0.2	<0.3	<0.1	<0.1
Fish fillet	2378-tetraCDD*	1.5	4	1	<0.2	1.4	1.5	2.1	0.76	1.4	1.7	2
	12378-pentaCDD*	0.22	0.52	<0.2	<0.2	0.29	0.3	0.5	<0.2	0.43	0.53	0.44
	123478-hexaCDD	<0.2	<0.1	<0.1	<0.1	<0.2	<0.2	<0.1	<0.2	<0.2	<0.2	<0.1
	123678-hexaCDD	<0.2	<0.2	<0.1	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1
	123789-hexaCDD	<0.2	<0.1	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1
	1234678-heptaCDD	<0.2	<0.2	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1
	OctaCDD	<0.3	<0.3	<0.2	<0.2	<0.3	<0.3	<0.2	<0.3	<0.3	<0.3	<0.2

*Frequently detected congeners summed up to Σ_3 PCDD/Fs are highlighted in bold

Table S7. Concentrations of polychlorinated dibenzo-p-furans (PCDFs, pg/g wet weight) in the paired fish fillet and egg samples.

Sample code		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon				Rainbow Trout	
		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	Concentrations										
Fish eggs	2378-tetraCDF*	4.6	6.6	2.9	11	13	6.2	4.6	3.6	6.9	1.4	6.8
	12378-pentaCDF	0.31	0.48	<0.16	<0.03	0.7	0.44	0.35	<0.03	0.4	<0.21	0.33
	23478-pentaCDF	1.2	2	0.67	1.2	3.6	1.6	1.6	1.3	2.4	1.6	1.3
	234678-hexaCDF	<0.05	<0.02	<0.11	<0.05	<0.19	<0.12	<0.03	<0.09	<0.04	<0.02	<0.06
	123478-hexaCDF	<0.20	<0.54	<0.20	<0.07	<0.90	<0.49	<0.18	<0.07	<0.79	<0.09	<0.14
	123678-hexaCDF	<0.02	<0.12	<0.08	<0.01	<0.16	<0.03	<0.03	<0.01	<0.01	<0.07	<0.08
	123789-hexaCDF	<0.05	<0.05	<0.01	<0.10	<0.09	<0.05	<0.10	<0.06	<0.06	<0.02	<0.01
	1234678-heptaCDF	<0.05	<0.05	<0.01	<0.08	<0.09	<0.05	<0.06	<0.01	<0.01	<0.01	<0.01
	1234789-heptaCDF	<0.07	<0.04	<0.07	<0.09	<0.13	<0.03	<0.06	<0.03	<0.01	<0.08	<0.05
	OctaCDF	<0.09	<0.01	<0.01	<0.01	<0.18	<0.04	<0.03	<0.09	<0.01	<0.10	<0.09
Fish fillet	2378-tetraCDF*	3.2	5.6	2.5	1.8	2.1	2.4	2.7	0.94	1.5	0.69	3.4
	12378-pentaCDF	<0.01	<0.28	<0.04	<0.19	<0.11	<0.18	<0.13	<0.12	<0.09	<0.12	<0.06
	23478-pentaCDF	<0.94	2	<0.01	<0.10	<0.35	<0.75	1.4	<0.51	<0.33	<0.23	<0.74
	234678-hexaCDF	<0.09	<0.05	<0.05	<0.06	<0.17	<0.16	<0.10	<0.06	<0.11	<0.08	<0.18
	123478-hexaCDF	<0.10	<1.08	<0.26	<0.07	<0.08	<0.35	<0.15	<0.15	<0.29	<0.20	<0.36
	123678-hexaCDF	<0.03	<0.02	<0.01	<0.10	<0.01	<0.11	<0.05	<0.14	<0.14	<0.08	<0.06
	123789-hexaCDF	<0.07	<0.01	<0.03	<0.09	<0.18	<0.12	<0.16	<0.01	<0.09	<0.10	<0.19
	1234678-heptaCDF	<0.03	<0.06	<0.01	<0.06	<0.05	<0.03	<0.01	<0.05	<0.05	<0.09	<0.01
	1234789-heptaCDF	<0.07	<0.04	<0.09	<0.04	<0.01	<0.05	<0.03	<0.07	<0.18	<0.13	<0.19
	OctaCDF	<0.06	<0.01	<0.03	<0.01	<0.05	<0.08	<0.04	<0.20	<0.14	<0.33	<0.13

*Frequently detected congeners summed up to Σ_3 PCDD/Fs are highlighted in bold

Table S8. Concentrations of WHO 2005 Toxic Equivalents (TEQs, pg/g wet weight) in the paired fish fillet and egg samples.

Sample code		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon				Rainbow Trout	
		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	WHO 2005 TEQ										
Fish eggs	2378-tetraCDD	2.5	4.7	0.9	0.73	7.4	3.4	3.5	3.3	4.9	4	4.1
	12378-pentaCDD	0.45	0.7	0.26	0.66	1.4	0.68	0.57	0.52	0.81	0.69	0.65
	123478-hexaCDD	<0.10	<0.10	<0.20	<0.10	<0.20	<0.20	<0.20	<0.10	<0.10	<0.10	<0.10
	123678-hexaCDD	<0.16	<0.25	<0.20	<0.25	<0.34	<0.20	<0.25	<0.10	<0.13	<0.15	<0.14
	123789-hexaCDD	<0.10	<0.20	<0.20	<0.29	<0.20	<0.20	<0.20	<0.20	<0.20	<0.10	<0.10
	1234678-heptaCDD	<0.16	<0.18	<0.19	<0.20	<0.20	<0.12	<0.10	<0.17	<0.16	<0.14	<0.17
	OctaCDD	<0.18	<0.28	<0.20	<0.26	<0.20	<0.27	<0.51	<0.21	<0.34	<0.10	<0.14
	2378-tetraCDF	4.6	6.6	2.9	11	13	6.2	4.6	3.6	6.9	1.4	6.8
	12378-pentaCDF	0.31	0.48	<0.19	<0.57	0.7	0.44	0.35	<0.15	0.4	<0.21	0.33
	23478-pentaCDF	1.2	2	0.67	1.2	3.6	1.6	1.6	1.3	2.4	1.6	1.3
	234678-hexaCDF	<0.10	<0.10	<0.20	<0.10	<0.20	<0.20	<0.12	<0.10	<0.09	<0.10	<0.20
	123478-hexaCDF	<0.45	<0.83	<0.26	<0.22	<1.10	<0.72	<0.58	<0.30	<0.82	<0.80	<0.57
	123678-hexaCDF	<0.10	<0.16	<0.10	<0.10	<0.20	<0.20	<0.10	<0.10	<0.10	<0.10	<0.10
	123789-hexaCDF	<0.10	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.10	<0.10	<0.10	<0.20
	1234678-heptaCDF	<0.07	<0.08	<0.10	<0.10	<0.10	<0.10	<0.07	<0.07	<0.07	<0.07	<0.04
	1234789-heptaCDF	<0.10	<0.10	<0.10	<0.20	<0.20	<0.10	<0.10	<0.10	<0.10	<0.10	<0.06
	OctaCDF	<0.10	<0.10	<0.20	<0.20	<0.20	<0.20	<0.10	<0.20	<0.10	<0.10	<0.10
	3,3',4,4'-tetraCB	0.08	0.08	0.05	0.06	0.19	0.08	0.07	0.07	0.08	0.04	0.11
	3,4,4',5-tetraCB	0.01	0.03	0.01	0.01	0.04	0.02	0.01	0.01	0.02	0.02	0.03
	2,3,3'4,4'-pentaCB	0.48	0.75	0.23	0.36	1.02	0.54	0.42	0.60	0.60	0.69	0.66
	2,3,4,4',5-pentaCB	0.03	0.04	0.01	0.02	0.06	0.03	0.02	0.04	0.03	0.04	0.04
	2,3'4,4',5-pentaCB	1.14	1.65	0.54	0.66	2.25	1.20	0.93	1.41	1.41	1.59	1.50
	2'3,4,4',5-pentaCB	0.03	0.05	0.01	0.01	0.07	0.03	0.03	0.04	0.03	0.04	0.04
	3,3'4,4',5-pentaCB	13.00	22.00	6.50	16.00	37.00	17.00	14.00	18.00	21.00	26.00	23.00
	2,3,3'4,4'5-hexaCB	0.10	0.16	0.05	0.07	0.20	0.11	0.09	0.13	0.13	0.15	0.13
	2,3,3'44'5'-hexaCB	0.02	0.03	0.01	0.02	0.05	0.02	0.02	0.03	0.03	0.03	0.03
	23',44',55'-hexaCB	0.05	0.09	0.03	0.04	0.12	0.07	0.05	0.08	0.07	0.09	0.08
	3,3'4,4'55'-hexaCB	0.25	0.48	0.11	0.45	0.75	0.39	0.28	0.33	0.42	0.48	0.33
	233'44'55'-heptaCB	0.01	0.01	0.00	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01
WHO05TEQs		19.0	32.1	9.3	20.6	53.1	24.8	21.0	25.4	31.0	34.6	31.9

		Fish Species										
		Brown Trout			Chinook Salmon		Coho Salmon			Rainbow Trout		
Sample code		000X9824	000X9827	000X9828	000X9787	000X9807	000X9815	000X9816	000X9818	000X9819	000X9832	000X9833
Portion Type	Chemical	WHO 2005 TEQ										
Fish fillet	2378-tetraCDD	1.5	4	1	<0.20	1.4	1.5	2.1	0.76	1.4	1.7	2
	12378-pentaCDD	0.22	0.52	<0.20	<0.20	0.29	0.3	0.5	<0.20	0.43	0.53	0.44
	123478-hexaCDD	<0.20	<0.10	<0.06	<0.10	<0.20	<0.20	<0.10	<0.20	<0.20	<0.20	<0.08
	123678-hexaCDD	<0.20	<0.21	<0.13	<0.10	<0.20	<0.20	<0.16	<0.20	<0.20	<0.20	<0.14
	123789-hexaCDD	<0.20	<0.10	<0.07	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.10
	1234678-heptaCDD	<0.20	<0.23	<0.12	<0.16	<0.20	<0.20	<0.19	<0.23	<0.15	<0.20	<0.12
	OctaCDD	<0.32	<0.32	<0.18	<0.20	<0.26	<0.28	<0.20	<0.28	<0.31	<0.30	<0.20
	2378-tetraCDF	3.2	5.6	2.5	1.8	2.1	2.4	2.7	0.94	1.5	0.69	3.4
	12378-pentaCDF	<0.20	<0.35	<0.17	<0.21	<0.12	<0.23	<0.22	<0.20	<0.20	<0.18	<0.27
	23478-pentaCDF	<0.98	2	<0.59	<0.29	<0.64	<0.88	1.4	<0.52	<0.95	<0.96	<0.92
	234678-hexaCDF	<0.10	<0.10	<0.10	<0.10	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20
	123478-hexaCDF	<0.45	<1.20	<0.32	<0.10	<0.21	<0.45	<0.57	<0.24	<0.41	<0.59	<0.67
	123678-hexaCDF	<0.10	<0.10	<0.10	<0.10	<0.10	<0.20	<0.10	<0.20	<0.20	<0.10	<0.20
	123789-hexaCDF	<0.10	<0.20	<0.10	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20
	1234678-heptaCDF	<0.09	<0.08	<0.07	<0.10	<0.08	<0.10	<0.10	<0.09	<0.11	<0.10	<0.10
	1234789-heptaCDF	<0.10	<0.10	<0.09	<0.10	<0.10	<0.20	<0.10	<0.10	<0.20	<0.20	<0.20
	OctaCDF	<0.10	<0.10	<0.10	<0.20	<0.11	<0.20	<0.13	<0.20	<0.24	<0.42	<0.20
	3,3',4,4'-tetraCB	0.05	0.06	0.04	0.01	0.03	0.03	0.04	0.01	0.02	0.02	0.05
	3,4,4',5-tetraCB	0.01	0.02	0.01	0.00	0.01	0.01	0.01	0.00	0.00	0.01	0.01
	2,3,3',4,4'-pentaCB	0.30	0.63	0.21	0.06	0.16	0.24	0.28	0.13	0.19	0.30	0.30
	2,3,4,4',5-pentaCB	0.02	0.04	0.01	0.00	0.01	0.01	0.02	0.01	0.01	0.02	0.02
	2,3'4,4',5-pentaCB	0.84	1.50	0.54	0.10	0.36	0.60	0.66	0.30	0.48	0.75	0.72
	2'3,4,4',5-pentaCB	0.02	0.04	0.01	0.00	0.01	0.02	0.02	0.01	0.01	0.02	0.02
	3,3'4,4',5-pentaCB	10.00	22.00	7.10	2.70	6.60	9.40	11.00	4.50	7.20	13.00	13.00
	2,3,3'4,4',5-hexaCB	0.08	0.17	0.06	0.01	0.03	0.06	0.07	0.03	0.05	0.08	0.08
	2,3,3'44'5'-hexaCB	0.02	0.04	0.01	0.00	0.01	0.01	0.02	0.01	0.01	0.02	0.02
	23',44',55'-hexaCB	0.05	0.10	0.03	0.01	0.02	0.04	0.05	0.02	0.03	0.05	0.05
	3,3'4,4'55'-hexaCB	0.25	0.60	0.17	0.08	0.13	0.26	0.33	0.11	0.16	0.36	0.33
	233'44'55'-heptaCB	0.01	0.02	0.01	0.00	0.00	0.01	0.01	0.00	0.00	0.01	0.01
WHO05TEQ		13.9	31.0	9.7	3.4	9.4	12.9	15.9	6.2	10.4	17.2	17.6