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Effects of skin removal on contaminant levels in salmon and trout filets

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HIGHLIGHTS

- The effects of skin removal on concentrations of mercury and persistent organochlorines in four fish species were assessed.
- Concentrations of the lipophilic organochlorines declined after skin removal, which reduced the lipid contents of the filets.
- Mercury concentrations increase after skin removal, indicating mercury is mainly associated with fish muscles.
- Trimming skin from salmon and trout filets before consumption is helpful in reducing exposure to toxic contaminants.

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ABSTRACT

Skin removal is a generally accepted method to reduce exposure to contaminants through fish consumption. However, inconsistent results from studies on the effectiveness of this method suggest influence of other factors such as characteristics of contaminants and fish species. This study investigated the effects of skin removal on the lipid contents and concentrations of total mercury, α-chlordane, hexachlorobenzene, mirex, octachlorosytrene, polychlorinated biphenyls (PCB), dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyldichloroethane (DDD), and dichlorodiphenyldichloroethylene (DDE). Four fish species namely brown trout (*Salmo trutta*), Chinook salmon (*Oncorhynchus tschawytscha*), coho salmon (*Oncorhynchus kisutch*) and rainbow trout (*Oncorhynchus mykiss*) sampled from the Credit River, Ontario, Canada were considered. Concentrations of all the lipophilic organic contaminants decreased significantly (median 17–37%) after removing skins from filets of brown trout, Chinook salmon and coho salmon, but not of rainbow trout. In contrast, the concentrations of mercury tended to be either similar or marginally higher after removing skins from filets of all four species; however, the amount of mercury would have likely declined or remained unchanged. Overall, removal of skin before consuming a fish filet is recommended to reduce exposure to contaminants widely found in Ontario fish.

1. Introduction

Fish consumption contributes to numerous nutritional and health benefits to human health because fish generally contain higher amounts of essential nutrients such as high-quality protein, minerals, vitamins and omega-3 (n−3) polyunsaturated fatty acids (PUFAs) (Domingo, 2007). n−3 PUFAs such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) are beneficial in the prevention of coronary heart disease, arrhythmias, and thrombosis (Kinsella et al., 1990). Therefore, the dietary guidelines by several health organizations including the World Health Organization (WHO, 2002), Health Canada (Health Canada, 2007), the United States Department of Agriculture (USDA, 2010) and the American Heart Association (AHA) (Domingo, 2007) recommend adults to have at least two servings of fish per week.

On the other hand, concerns exist on the exposure to the elevated concentrations of contaminants accumulated in fish and the health risk involved in fish consumption (Alcock et al., 1998; Mozaffarian and Rimm, 2006). Although fish consumption comprises only a small portion of human diet, it is the major pathway of human exposure to various contaminants such as persistent organic pollutants (POPs) and mercury (Alcock et al., 1998; Clarkson, 1993). As potential endocrine disruptors, POPs such as dioxin, polychlorinated-biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) may affect human health by mimicking natural hormones and altering the normal regulatory function of the immune, nervous, and endocrine systems (Crisp et al., 1998). Mercury has been linked to neurological deficits and developmental delay in children with prenatal exposure (Counter and Buchanan, 2004). The extent of exposure and the adverse health effect largely depend on the contaminant concentrations in the fish consumed.

Concentrations of contaminants in fish from the same habitat area can vary depending on fish age, gender, species etc. (Gewurtz et al.,

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2011b; Monod and Keck, 1982; Rypel et al., 2007). Such variations could be caused by different physiological characteristics such as diet, metabolism and lipid content for different fish species (Clark et al., 1990; Gewurtz et al., 2011a). Concentrations of contaminants also vary within fish body. For example, higher concentrations of PCBs and DDT were found at the head end compared to the tail end with a peak in the central section of Atlantic salmon (*Salmo salar*) (Bayen et al., 2005). Such variation in contaminant concentration within a fish is largely due to differential lipid distribution within the fish body (Bayen et al., 2005). The wet weight based lipid contents for ten different tissue types of Atlantic Salmon varied widely (2–38%) and the skin had a lipid content twice as high as the white muscle (Aursand et al., 1994). Fatty tissues such as the skin generally contain a higher concentration of lipophilic organic contaminants within the fish body (Davis et al., 2002; Hora, 1981). Therefore, skin removal before eating a fish filet is recommended by many agencies that issue fish consumption advisories (OMOE, 2009; Virginia Department of Health, 2008).

Despite studies indicating that the skin removal from fish filet decreases concentrations of organic contaminants (Aursand et al., 1994; Hora, 1981), the amount of reduction is highly variable among contaminants and fish species (Domingo, 2007; Foran et al., 2005). In some cases, even increased wet-weight based contaminant concentrations after skin removal have been reported (Dellinger et al., 1995; Shaw et al., 2006). Considering the variable effects of skin removal on contaminant concentrations in fish, more fish species- and contaminant-specific information on the effects of skin removal is needed to accurately advise on how fish consumers can minimize exposure to toxic contaminants.

This study examines the effect of fish skin removal on the filet concentrations of various contaminants including total mercury, α-chlordane, hexachlorobenzene, mirex, octachlorosytrene, total-PCB and DDT (including its metabolites) in four fish species from Credit River (Ontario, Canada). The species considered are brown trout (*Salmo trutta*), Chinook salmon (*Oncorhynchus tschawytscha*), coho salmon (*Oncorhynchus kisutch*) and rainbow trout (*Oncorhynchus mykiss*). These fish from the Credit River spend a substantial amount of their life time in Lake Ontario, Canada.

2. Method

2.1. Sample collection and preparation

The Sport Fish Contaminant Monitoring Program of the Ontario Ministry of the Environment (OMOE) monitors various contaminants in sport and forage fish samples collected from >2000 locations across Ontario, Canada, and advises on safe consumption of fish (Bhavsar et al., 2011). As a part of the monitoring program, 18 samples of brown trout, 58 samples of Chinook salmon, 23 samples of coho salmon and 13 samples of rainbow trout were collected from a fish ladder location (Streetsville) in the Credit River (Ontario, Canada). The river is home to the brown trout population and provides spawning areas for Chinook salmon, coho salmon and rainbow trout. The number of fish samples collected in a sampling season dictated the sample sizes. Once collected, the fish were measured for their size (length and weight), sexed, and filleted. Two filets, one with skin-on and one with skin removed, were collected from each fish. The samples were ground and stored in glass vials at −20 °C until chemical analysis.

2.2. Sample analysis

All 112 pairs of skin-on and -off fish samples were analyzed for α-chlordane, hexachlorobenzene (HCB), mercury, mirex, octachlorosytrene (OCS), PCB, 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (*p,p′*-DDD), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethene (*p,p′*-DDE) and 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (*p,p′*-DDT), and 40 (10 per species; skin-on and -off; randomly selected) samples were analyzed for dioxins/furans/dioxin-like PCB at the OMOE laboratories in Toronto (Canada). The OMOE laboratories are accredited by the Canadian Association for Laboratory Accreditation (CALA) and monitor the performance of the methods through laboratory intercalibration studies (e.g., Northern Contaminants Program — NCP, and Quality Assurance of Information for Marine Environmental Monitoring in Europe — QUASIMEME). The methods are described in detail by Gewurtz et al. (2010) and summarized below.

2.2.1. Mercury

The OMOE method HGBIO-E3057 (OMOE, 2006) was used for mercury analysis. Homogenized fish tissue sample (0.2–0.4 g) was digested with 4:1 concentrated sulfuric to nitric acid (v/v). The diluted digestates were analyzed for total mercury by cold vapor-flameless atomic absorption spectroscopy (gold-film Jerome Model 511 Hg Analyzer, method detection limit MDL = 0.01 μg/g ww). Calibration curves were based on five concentrations encompassing the range of tissue concentrations and were accepted if correlation coefficients were ≥0.990.

2.2.2. Percent lipid, total-PCB, and other organochlorines

Total-PCB, α-chlordane, HCB, mirex, OCS, *p,p′*-DDD, *p,p′*-DDE and *p,p′*-DDT were analyzed using OMOE method E3136 (OMOE, 2007). 5 g homogenized sample of fish skin-on or skin-off filet was spiked with decachlorobiphenyl and 1,3,5-tribromobenzene, digested with hydrochloric acid, and extracted with hexane/dichloromethane. Lipid content was determined gravimetrically. Gas chromatography (GC) with electron capture detection (ECD) was used for the analysis of PCBs (HP 6890 GC, Ni^{63} ECD, MDL = 20 ng/g wet weight (ww)), HCB, OCS and mirex (HP 5890 GC, Ni^{63} ECD, MDL = 1, 1 and 5 ng/g ww, respectively), and α-chlordane, DDT and metabolites (HP 6890 Series Plus GC, dual column micro Ni^{63} ECD, MDL = 2 ng/g ww). Method blanks and matrix spikes were processed with each set of 20 to 30 samples. Calibration curves were based on six concentrations encompassing the range of tissue concentrations and were accepted if correlation coefficients were ≥0.985.

2.3. Data analysis

One-tailed Wilcoxon signed-rank test was performed (SPSS 12.0) at the significant level of 0.05 to compare concentrations of the contaminants in the fish filet with skin and after skin removal. As a non-parametric statistical test, the Wilcoxon signed-rank test does not require the population to follow a normal distribution and is used as an alternative to the paired Student's t-test (Wilcoxon, 1945). Even when the normality assumption holds, the testing power from the Wilcoxon signed-rank test does not decline more than 5% compared to power of the t-test (Lehmann, 1999; Sawilowsky, 2005).

Based on the measured wet-weight contaminant concentrations in skin-on and skin-off fish filet, one thousand bootstrapping case resamplings were conducted and applied in constructing the distribution of the relative change of contaminant concentrations in fish after skin removal. Bootstrapping is a technique to estimate the population features via resampling the observed data (Chernick, 2008). By performing bootstrapping resampling, the bias caused by the outliers from a limited sampling size can be eliminated.

The concentration of the organic contaminants is hypothesized to be related to the lipid content in the fish sample. To investigate the effect of lipid reduction on the contaminants' concentrations in the filet after skin removal, a regression model was developed:

$$C_{SBF} = \beta_0 + \beta_1 C_{FSO} + \beta_2 (L_{SBF} - L_{FSO}) \quad (1)$$

where C_{SBF} is the wet-weight based contaminant concentration in the skinless and boneless filet (SBF); C_{FSO} and $L_{SBF} - L_{FSO}$ are the contaminant concentration in the filet with skin on (FSO) and the change of

lipid content after skin removal, respectively. β_0, β_1 and β_2 are the regression coefficients. For each contaminant and each fish species, 1000 cases from the bootstrap resampling were analyzed using the model constructed in SPSS v12.0.

3. Results and discussion

3.1. Fish characteristics

Based on multiple comparison of Kruskal–Wallis one-way analysis of variance (Kruskal and Wallis, 1952), Chinook salmon was the longest (median [interquartile range] = 90.5 [76, 95.9] cm) among the four fish species considered in this study. The length of coho salmon (68.0 [63.7, 70.5] cm) was similar to that of rainbow trout (64.0 [61.8, 66.5] cm) and brown trout (59.1 [55.1, 64.0] cm). Chinook salmon (7.9 [4.5, 9.5] kg) was also heavier than Coho salmon (4.8 [3.9, 5.3] kg), rainbow trout (3.0 [2.8, 3.5] kg) and brown trout (2.5 [2.1, 3.3] kg). The lipid content in filets with skin on (FSO) was in the order: coho salmon (7.4 [5.6, 8.2] %) ≈ brown trout (5.0 [3.8, 7.0] %) > rainbow trout (3.8 [1.7, 5.3] %) ≈ Chinook salmon (4.0 [2.3, 5.5] %). The lipid content in filet with skin off was lower than that with skin on but followed the same order: coho salmon (5.6 [4.2, 7.5] %) > Chinook salmon (3.3 [2.0, 4.6] %) ≈ brown trout (4.4 [3.2, 5.5] %) ≈ rainbow trout (3.0 [2.1, 6.8] %). The detailed descriptive statistics on the fish characterizations are given in Table S1 of the supplementary material.

3.2. Contaminant levels

The median and interquartile range of lipid contents and concentrations of each contaminant in the four fish species are presented in Table 1; other descriptive statistics are provided in Table S2 of the supplementary material. PCB and *p,p′*-DDE are predominant among the nine contaminants we investigated. Since the relative abundance of various chemicals would largely depend on the environment where the fish lived and vary from one location to another in the Great Lakes (Bhavsar et al., 2008, 2007, 2010), no further effort was spent comparing concentrations between different chemicals.

When comparing the wet-weight based contaminant concentrations in the four fish species, we note that mirex, octachlorostyrene, PCB, *p,p′*-DDD and *p,p′*-DDE in FSO as well as mirex and octachlorostyrene in SBF had the same ranks among the four species (Chinook salmon > brown trout > coho salmon > rainbow trout). The next predominant rank pattern was Chinook salmon > brown trout > rainbow trout > coho salmon, which was observed for mercury in FSO and SBF and for PCB and *p,p′*-DDT in SBF. Different from the two predominant rank patterns, α-chlordane (FSO and SBF) was in the order of coho salmon > Chinook salmon > rainbow trout > brown trout; hexachlorobenzene and *p,p′*-DDT in FSO were in the order of brown trout > Chinook salmon > coho salmon > rainbow trout; hexachlorobenzene and *p,p′*-DDD in SBF were in the order of brown trout > Chinook salmon > rainbow trout > coho salmon.

Table 2
p-Values from one-tailed Wilcoxon Signed Rank test on skin-on and -off filet lipid content and wet-weight based contaminant concentrations.

	Brown trout	Chinook	Coho	Rainbow trout
	N = 19	N = 58	N = 23	N = 13
Lipid content	**0.047**	<**0.001**	**0.007**	0.500
α-Chlordane	**0.008**	<**0.001**	<**0.001**	0.378
Hexachlorobenzene	**0.007**	<**0.001**	<**0.001**	0.237
Mercury [a]	<**0.001**	<**0.001**	0.454	0.078
Mirex	**0.004**	<**0.001**	<**0.001**	0.238
Octachlorostyrene	**0.003**	<**0.001**	<**0.001**	0.264
PCB	**0.005**	<**0.001**	<**0.001**	0.319
p,p′-DDD	**0.032**	<**0.001**	<**0.001**	0.361
p,p′-DDE	**0.032**	<**0.001**	**0.002**	0.240
p,p′-DDT	<**0.001**	<**0.001**	<**0.001**	0.216

[a] The alternative hypothesis for mercury is different from others. When the test result is significant, it means that the concentration of mercury increased after skin removal. The *p*-values at 0.05 significance have been highlighted in bold.

3.3. Changes in contaminants after skin removal

Wilcoxon signed-rank tests were applied in comparing lipid content and wet-weight based contaminant concentrations (C_{WW}, ng/g_{ww}) between FSO and SBF. Based on pre-screening of the data, a trend of decreased levels was observed for lipid content and C_{WW} after skin removal. Since statistical inference should rely on the rejection of the

Table 1
Lipid contents (%) and concentrations (ng/g ww) of contaminants in filets with skin-on (FSO)[a] and skin-off (SBF)[b] of four fish species. The results are presented as medians and interquartile ranges.

		Brown trout	Chinook	Coho	Rainbow trout
Lipid content (%)					
FSO[a]		5.0 [3.8, 7.0]	4.0 [2.3, 5.5]	7.4 [5.6, 8.2]	3.8 [1.7, 5.3]
SBF[b]		4.4 [3.2, 5.5]	3.3 [2.0, 4.6]	5.6 [4.2, 7.5]	3.0 [2.1, 6.8]
Concentration of contaminants (ng/g ww)					
a-Chlordane	FSO	14 [10, 18]	21 [15, 29]	25 [20, 37]	17 [14, 20]
	SBF	12 [10, 13]	17 [14, 23]	19 [14, 24]	17 [13, 20]
Hexachlorobenzene	FSO	10 [8, 15]	10 [7, 12]	8 [7, 10]	7 [6, 10]
	SBF	9 [5, 12]	8 [6, 11]	6 [3, 8]	6 [5, 8]
Mercury	FSO	180 [160, 190]	265 [220, 310]	150 [140, 170]	165 [95, 210]
	SBF	190 [170, 210]	285 [220, 330]	150 [140, 170]	160 [130, 200]
Mirex	FSO	185 [170, 230]	215 [180, 330]	195 [165, 240]	125 [98, 160]
	SBF	145 [135, 173]	190 [155, 235]	150 [115, 190]	140 [90, 165]
Octachlorostyrene	FSO	21 [18, 31]	22 [18, 32]	19 [15, 28]	12 [11, 20]
	SBF	16 [12, 20]	19 [15, 23]	15 [12, 20]	16 [10, 17]
PCB	FSO	2480 [2000, 3040]	2090 [1700, 3280]	1940 [1460, 2320]	1650 [1310, 2275]
	SBF	1910 [1610, 2440]	1825 [1380, 2270]	1610 [1100, 1900]	1710 [1240, 2150]
p,p′-DDD	FSO	47 [37, 61]	44 [35, 59]	33 [26, 51]	30 [23, 40]
	SBF	37 [30, 49]	35 [27, 46]	25 [20, 32]	28 [21, 33]
p,p′-DDE	FSO	644 [279, 848]	674 [528, 1123]	547 [310, 692]	482 [369, 618]
	SBF	447 [311, 643]	578 [397, 808]	384 [288, 531]	366 [300, 725]
p,p′-DDT	FSO	58 [41, 92]	51 [35, 76]	34 [25, 60]	40 [29, 45]
	SBF	40 [27, 57]	42 [25, 55]	25 [16, 33]	37 [33, 49]

[a] FSO: boneless filet with skin on.
[b] SBF: skinless and boneless filet.

null hypothesis and accepting the alternative hypothesis to avoid the Type II error, the alternative hypothesis set up for lipid content and C_{WW} of organic contaminants was $L_{FSO} > L_{SBF}$ and $C_{FSO} > C_{SBF}$. For mercury, however, a trend of increased concentration was observed after skin removal. Therefore, the alternative hypothesis for mercury was $C_{FSO} < C_{SBF}$.

As indicated by the *p*-values from the Wilcoxon signed-rank tests (Table 2), the lipid content for all the fish species except rainbow trout had a statistically significant decrease after skin removal. Accordingly, C_{WW} of the lipophilic organic contaminants had a statistically significant decrease after skin removal for all the fish species except rainbow trout. For mercury, statistically significant increase in concentration was observed in brown trout and Chinook salmon, but not in coho salmon and rainbow trout.

Since lipid content is generally a principal factor in controlling the lipophilic contaminant levels in fish, Wilcoxon signed-rank tests were also conducted on the lipid normalized contaminant concentrations (C_{LW}, ng/g_{lipid}). Comparing the test results for C_{LW} (Table S3) with that for C_{WW} (Table 2), no statistically significant reduction of C_{LW} after skin removal was observed in brown trout for all chemicals except octachlorostyrene and *p,p′*-DDT. However, similar to C_{WW}, statistically significant reduction of C_{LW} was found for the organic contaminants in Chinook salmon (except hexachlorobenzene and octachlorostyrene) and in coho salmon (except PCB and *p,p′*-DDE).

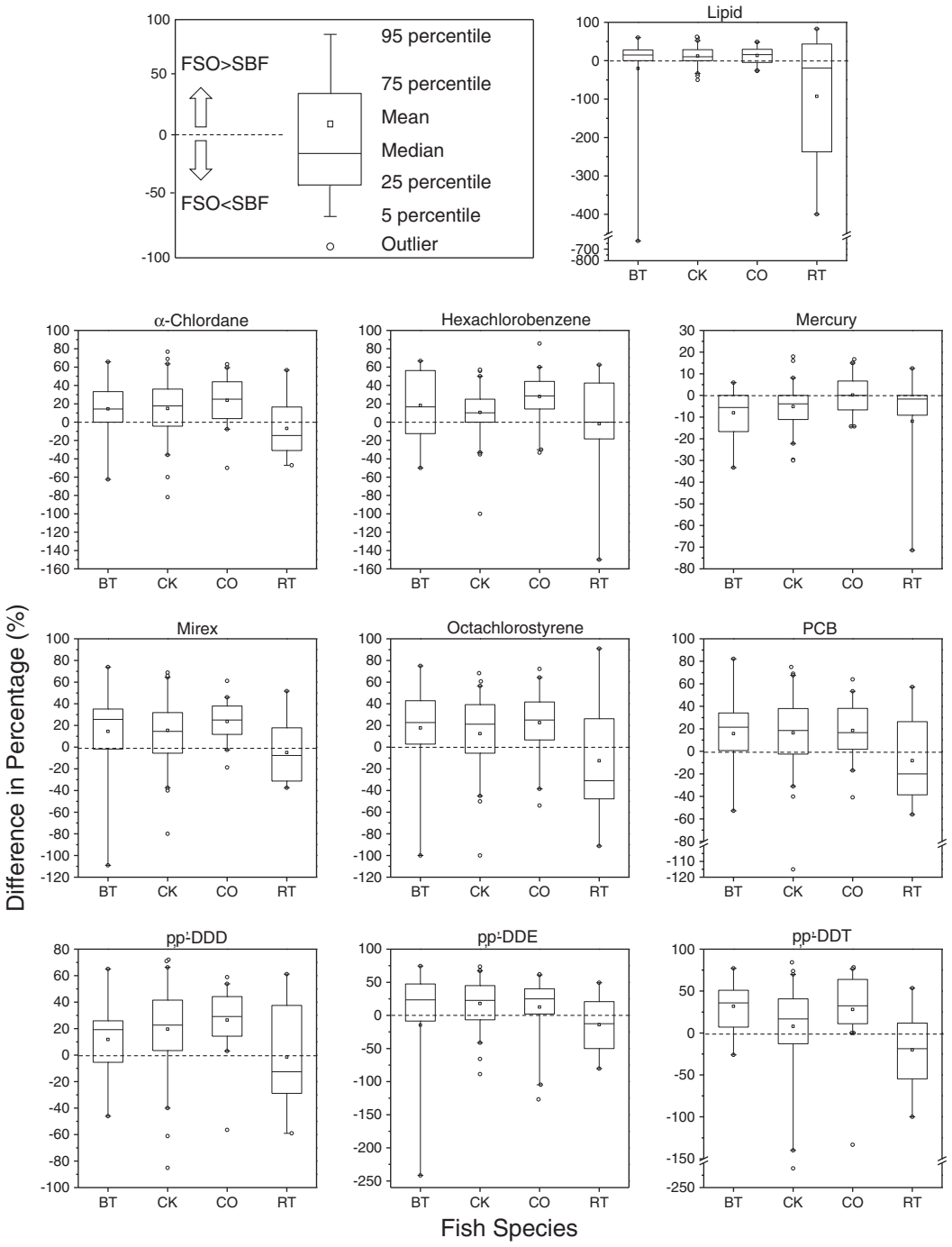

Fig. 1. Differences (in percent or %) in lipid contents and wet-weight based contaminant concentrations between filet with skin-on (FSO) and skinless and boneless filets (SBF) of four fish species (BT: brown trout, CK: Chinook salmon, CO: coho salmon, RT: rainbow trout).

For rainbow trout, there was no statistically significant reduction in C_{LW} after skin removal, which is similar to the findings for C_{WW}.

The relative changes in lipid content and C_{WW} after skin removal varied greatly (Fig. 1). Some outliers (e.g., lipid content of brown trout and rainbow trout) were found to deviate from the medians by as much as 400%; in some cases the direction of the change (i.e., increase or decrease) for the outliers was opposite to the majority of the other samples. In order to eliminate the influence of the outliers on our interpretation, one thousand bootstrap resamplings on the skin-on and -off pair for each chemical and each fish species were performed. The probability distributions on the relative reductions of the contaminants in filets after skin removal constructed from the bootstrap resamplings are shown in Fig. 2. The median and 95% confidence interval (CI) are indicated for each contaminant and fish species. The positive/negative relative change means increased/decreased C_{WW} after the skin removal.

Medians of 8% and 5% increase in C_{WW} of mercury were observed in brown trout and Chinook salmon. The 95% CIs of mercury in brown trout and Chinook salmon were higher than zero indicating a statistically significant increase in C_{WW}. Since the 95% CIs of mercury in coho salmon and rainbow trout included zero, we cannot infer that there was a significant change in C_{WW} after skin removal. Results of the bootstrap resamplings were consistent with the Wilcoxon signed-rank tests (Table 2). For all the other contaminants in brown trout, Chinook salmon and coho salmon, the median relative reduction after skin removal ranged 17–37%. The upper values of the 95% CIs of the changes for these chemicals were less than zero indicating a statistically significant reduction in C_{WW} after skin removal. The 95% CIs for rainbow trout were wider than those for brown trout, Chinook salmon and coho salmon and included zero; hence, no significant changes in C_{WW} of these organic contaminants after skin removal in rainbow trout could be inferred.

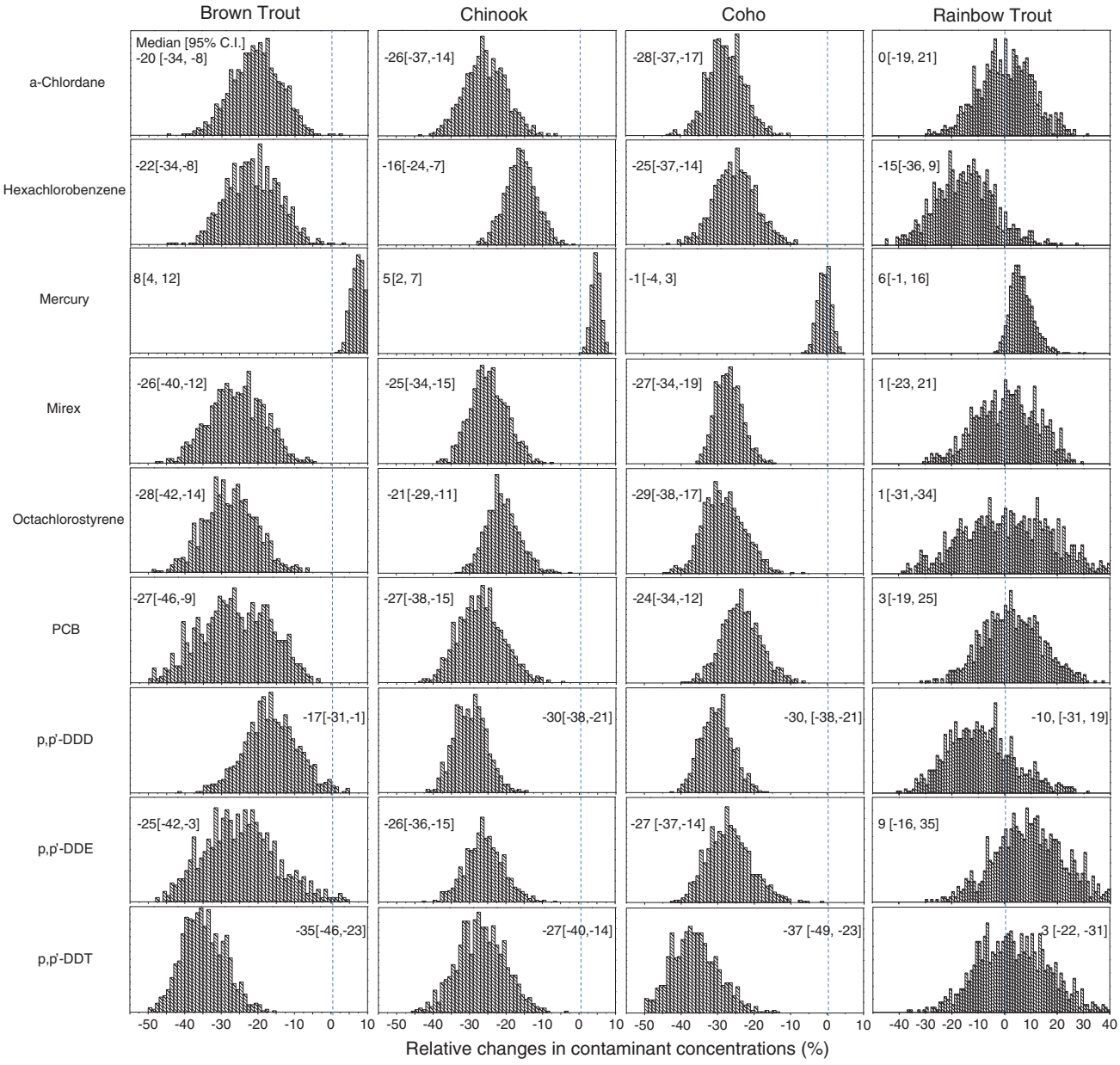

Fig. 2. Distributions of the relative changes in the wet-weight based contaminant concentrations in fish filets after skin removal. The distributions were constructed based on one thousand times bootstrap resampling. The median and 95% confidence interval (CI) are shown for each chemical and fish species.

3.4. Regression analysis

The regression model (Eq. (1)) was applied to the bootstrap resampled C_{WW} of each contaminant in FSO and SBF of each species. The model's ability to predict (R^2_{adj}) and regression coefficients are reported in Table S4. The R^2_{adj} represents the portion of the model-explained variance in the dependent variable. Of the 36 species/contaminant pairs analyzed, 18 pairs had over 50% of the dependent variance explained by the model (Fig. 3).

The R^2_{adj} highly varied among contaminants and fish species. For example, R^2_{adj} for octachlorostyrene for the four fish species ranged 0.11–0.33 and were generally lower than those of other contaminants. The R^2_{adj} (0.78–0.93) for mercury in all four species were generally higher than the other chemicals. This is because C_{WW} of mercury in SBF and in FSO were highly correlated and other factors such as lipid content have less influence on mercury distribution within the fish body. The R^2_{adj} for mirex for Chinook salmon, coho salmon and rainbow trout ranged 0.41–0.67. However, the model worked poorly for mirex in brown trout and could not explain the majority of the variance in the dataset. The low R^2_{adj} is likely due to other influential factors that were not considered in the model. This limits the use of the model for the purpose of prediction; however, the models provided useful information in data interpretations.

3.5. Effect of lipid on changes in contaminants after skin removal

The normalized regression coefficients (Table S4) were used to assess the effect of lipid content changes on the C_{WW} in the skin-off filets. Since the units of the independents are different, normalized regression coefficients directly reflect the relative contributions of the independents on the dependent. The ratio of B_2 and B_1 was used as lipid contribution index (LCI), where B_1 and B_2 are the normalized regression coefficients for the independents of C_{FSO} and ($L_{SBF} - L_{FSO}$), respectively. Fig. 4 compares the LCIs for the contaminants in the four fish species with an absolute value of LCI reflecting the extent of the contribution. LCI<0 is observed for all the organic contaminants, indicating lipid reduction due to skin removal would contribute to the reduction of C_{WW} in SBF.

Mercury was the only contaminant with LCI>0, which indicates that lipid reduction due to skin removal would contribute to elevated C_{WW} of mercury in SBF. This is consistent with the understanding that mercury mainly accumulates in fish muscles in methylmercury form by binding with sulfur-bearing amino acids (Bloom, 1992; Dellinger et al., 1995). The negative values of LCIs for all the other contaminants indicate that lipid reduction due to skin removal would contribute to lower C_{WW} in SBF. Generally, brown trout and rainbow trout had higher LCIs than Chinook salmon and coho salmon, except for α-chlordane and *p,p′*-DDT, the LCI of which is higher in Chinook salmon than in brown trout. Interestingly, LCI for coho salmon is generally lower although, as noted earlier, the lipid content of coho salmon is the highest among the four fish species considered.

LCI reflects the effect of changes in lipid content on contaminant concentrations, and is expected to relate with the lipophilicity (log K_{OW}, compiled in Table S5) of the contaminants. Significant correlation (Spearman) was observed between log K_{OW} and LCI for brown trout, coho salmon and rainbow trout. Such significant correlations indicate that the variations of the empirically derived LCI among different chemicals in each of fish species can be explained by the variations of K_{OW}. However, no significant such correlation was found for Chinook salmon, indicating K_{OW} may not be a good predictor for the distributions of organic compounds in the lipid content of Chinook salmon as studies (Endo et al., 2011; Geisler et al., 2012) have observed different partitioning properties for different types of lipid and a single predictor may not be able to capture the variability in different types of lipid.

3.6. Factors controlling lipophilic contaminants within fish

Lipid distribution is deemed as the most influential factor in determining the distributions of lipophilic organic contaminants within a fish body. The lipid distribution within a fish body varies not only among fish species but also within each species (Persson et al., 2007). The lipid content in the skin was found two times higher than the red muscle but seven times lower than the white muscle (Aursand et al., 1994). Such differences would affect the reductions in the concentrations of lipophilic contaminants by skin removal. The wide variability in the contaminant reductions due to skin removal of the four species presented in this study (Fig. 2) is similar to a number of other studies (Shaw et al., 2006; Skea et al., 1979; Voiland et al., 1991; Zabik et al., 1995).

In addition to the lipid distribution, differences in chemical properties could also affect contaminant distribution within fish and thereby efficiency of a contaminant reduction due to skin removal. Uptake of lipophilic contaminants by fish occurs mainly via food intake, dermal diffusion and respiration (Mackay and Fraser, 2000); however, the dominant exposure pathway depends on the chemical properties. For chemicals with large molecules and high lipid/water partition coefficients (e.g., log K_{ow} greater than 6), the predominant uptake pathway is via food ingestion (Mackay and Fraser, 2000). After entering the fish body via the gastrointestinal tract, the

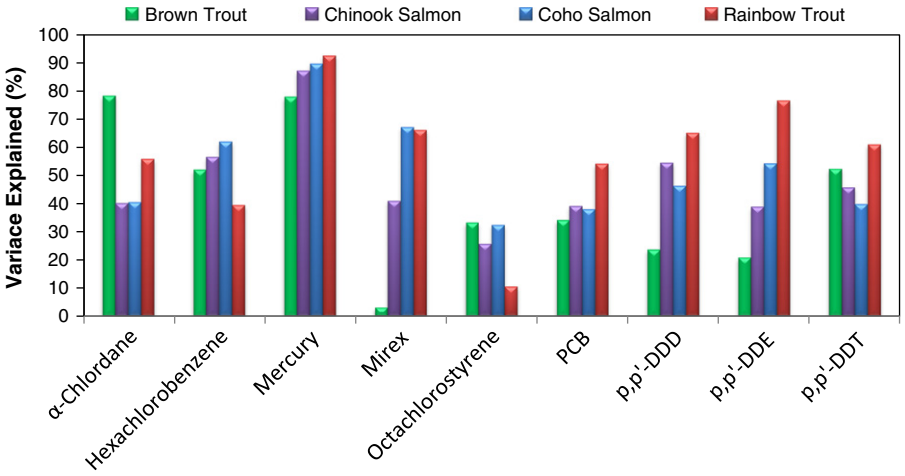

Fig. 3. Variance explained by the regression model for the wet-weight based contaminant concentrations in the skinless and boneless fish filets.

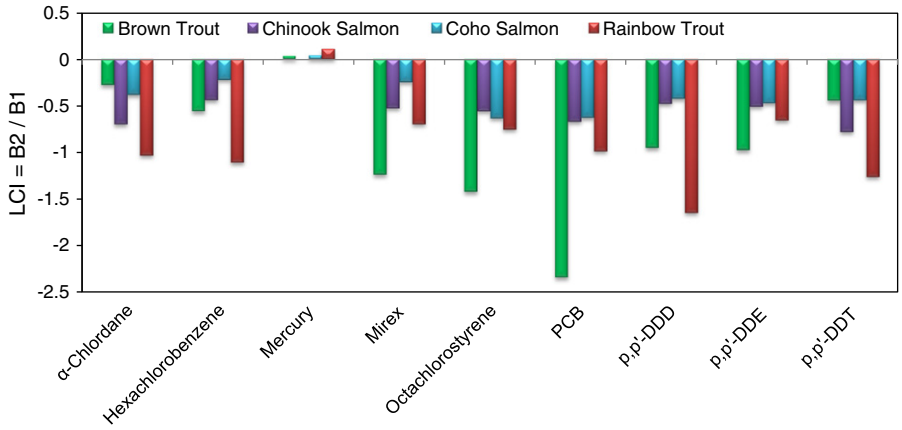

Fig. 4. Lipid contribution index (LCI) as a measure of the effect of lipid reduction on the wet-weight based contaminant concentrations in the skin-off filets.

chemical does not reach equilibrium within the whole fish body instantaneously; the chemical is distributed within the body via the circulatory system and such process is kinetically limited (Krishnan and Peyret, 2009). This can result in a greater accumulation of some POPs in fish muscle lipids than in skin-associated fat, and thereby in differential reductions in lipid content and the POPs via skin removal (Shaw et al., 2008).

4. Implications for fish consumption advisories

Studies have shown inconsistent results on the reduction of organic contaminants by removing fish skin and flesh (Aursand et al., 1994; Foran et al., 2005; Shaw et al., 2006; Skea et al., 1979; Voiland et al., 1991; Zabik et al., 1995). Our study considering brown trout, Chinook salmon, coho salmon and rainbow trout collected from the Credit River, Ontario, Canada found that skin removal significantly reduces (median 17–37%) concentrations of major legacy lipophilic organic contaminant found in Ontario's brown trout, Chinook salmon and coho salmon, but made no statistically significant difference for rainbow trout. Although skin removal tends to increase the mercury concentrations in brown trout, coho salmon and rainbow trout, the total intake of mercury for a given meal size will be lower for the skin-off filet compared to the corresponding skin-on filet. Therefore, we conclude that trimming skin from fish filet before consumption is helpful in reducing exposure to toxic contaminants.

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Appendix A. Supplementary data

Further information on the results can be found in the four tables in the supplementary material. Supplementary materials related to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.10.090.

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