



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, octachlorostyrene, polychlorinated biphenyls (PCB), dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyldichloroethane (DDD), and dichlorodiphenyldichloroethylene (DDE). Four fish species namely brown trout (*Salmo trutta*), Chinook salmon (*Oncorhynchus tshawytscha*), 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.

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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 tshawytscha*), 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 $-20\text{ }^{\circ}\text{C}$ 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 $\mu\text{g/g}$ 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, Ni⁶³ ECD, MDL = 20 ng/g wet weight (ww)), HCB, OCS and mirex (HP 5890 GC, Ni⁶³ ECD, MDL = 1, 1 and 5 ng/g ww, respectively), and α -chlordane, DDT and metabolites (HP 6890 Series Plus GC, dual column micro Ni⁶³ 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:

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

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

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