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MERCURY AND OTHER TRACE ELEMENTS IN FARMED AND WILD SALMON FROM BRITISH COLUMBIA, CANADA

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Abstract—The present study reports measured levels of Hg and other trace elements in commercial salmon feed; farmed Atlantic, coho, and chinook salmon (n = 110); and wild coho, chinook, chum, sockeye, and pink salmon (n = 91). Metal concentrations in farmed and wild salmon from British Columbia, Canada, were relatively low and below human health consumption guidelines. Methylmercury in all salmon samples (range, 0.03–0.1 μg/g wet wt) were below the 0.5 μg/g guideline set by Health Canada. Negligible differences in metal concentrations were observed between the various species of farmed and wild salmon. Metal concentrations generally were higher in commercial salmon feed compared to farmed salmon. Mercury showed slight bioaccumulation potential in farmed salmon, with biomagnification factors (BMFs) ranging between 0.8 and 1.9. Other metals, such as Cd, Pb, and Ni, exhibited biodilution, with BMFs of much less than one. The relatively low degree of biomagnification of metals observed in farmed salmon likely resulted from the combination of low gastrointestinal absorption efficiency, negligible transfer to muscle tissue relative to other compartments, and a high degree of growth dilution in these fish. Human dietary exposure calculations indicate intakes of Hg, Cd, Pb, Cu, As, and Ni via farmed and wild British Columbia salmon are a relatively small percentage of total intakes (0.05–32%) compared to other Canadian foodstuffs, such as fruits, vegetables, chicken, and beef (68–99%). Although total dietary exposure of Cd, Pb, and Cu approached provisional tolerable daily intake levels, the contribution from British Columbia salmon was less than 2%. Our findings indicate farmed and wild British Columbia salmon remain a safe source of omega-3 highly unsaturated fatty acid intake for cardioprotective and, possibly, other health benefits.

Keywords—Mercury Metals Salmon Bioaccumulation Human exposure

INTRODUCTION

Important human health benefits relate to an adequate intake of omega-3 highly unsaturated fatty acids (n-3 HUFAs), such as eicosapentaenoic acid and docosahexaenoic acid, which can be obtained through weekly consumption of oily fish, such as salmon. In particular, n-3 HUFAs are believed to reduce the likelihood of cardiovascular disease, inflammatory responses and conditions, and certain cancers as well as to enhance brain, cognitive, and ocular development and function [1–4]. Consequently, global demand for wild and farmed salmon from Europe and North America has been rising in recent years. British Columbia, Canada, is an important producer of both farmed and wild salmon. Much of the farmed salmon sold in the United States is imported from British Columbia. Internationally, British Columbia salmon and trout production represented approximately 5% of global production for these products in 2002 [5,6] (http://www.statcan.ca/english/freepub/23-222-XIE/23-222-XIE2003000.pdf, http://www.fao.org/docrep/009/a0699e/A0699E00.htm). Positive health benefits of an oily fish diet may be diminished, however, by negative effects associated with bioaccumulation and elevated levels of environmental contaminants, such as polychlorinated biphenyls (PCBs), dioxins, pesticides, and heavy metals.

In some cases, a high degree of chemical bioaccumulation can lead to elevated tissue residue levels, which may exceed human consumption guidelines (http://www.cfsan.fda.gov/~frf/sea-mehg.html). Elevated human dietary exposure to some chemicals can potentially increase the risk of cancer, immune and cognitive dysfunction, and birth and developmental effects. For example, a recent global assessment of organochlorine contaminants in farmed and wild salmon indicated that frequent consumption of certain farmed Atlantic salmon products poses a greater cancer risk compared to frequent consumption of wild salmon [7]. Our recent analyses of PCBs, dioxins and furans, and Hg in farmed and wild salmon from coastal British Columbia showed that both wild and farmed British Columbia salmon exhibit levels well below human health consumption guidelines for those chemicals [8] and are sufficiently low to allow weekly consumption of salmon as a safe dietary source of n-3 HUFAs. This previous study [8], which investigated those contaminants in five species of wild Pacific salmon, including chinook (*Oncorhynchus tshawytscha*), coho (*Oncorhynchus kisutch*), sockeye (*Oncorhynchus nerka*), chum (*Oncorhynchus keta*), and pink (*Oncorhynchus gorbuscha*), as well as three species of farmed salmon, including Atlantic (*Salmo salar*), chinook, and coho salmon, also demonstrated a large variation in contaminant levels between species. The present study reports on trace element concentrations that were monitored in the aforementioned samples of commercial feed and British Columbia salmon.

Unlike synthetic organic pollutants, such as pesticides and PCBs, trace elements in the environment can originate from both anthropogenic sources, such as mining and combustion of fossil fuels, as well as from natural sources, such as bedrock, volcanoes, and forest fires [9]. Elements generally are clas-

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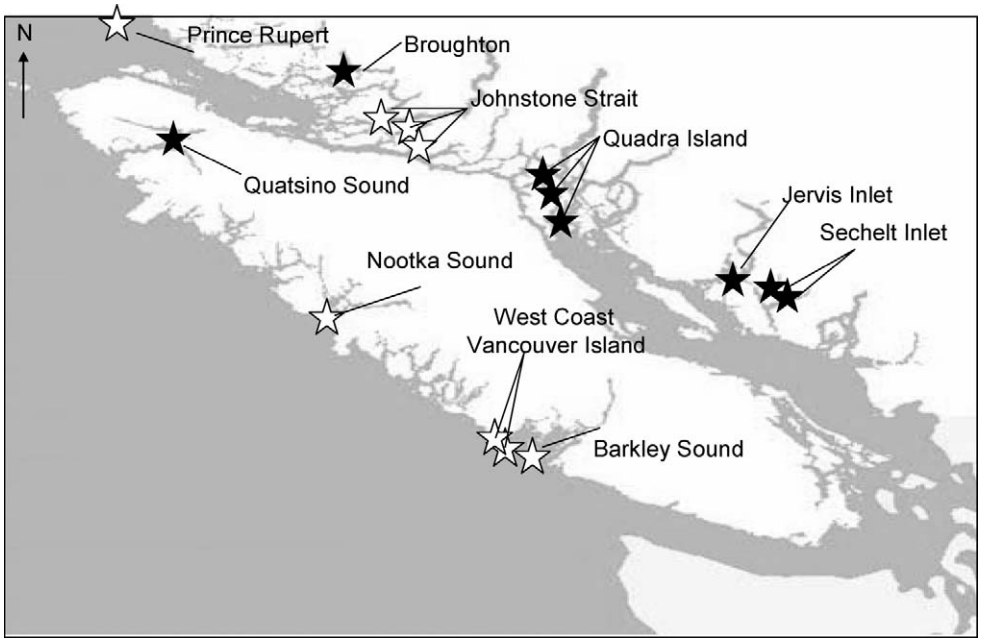

Fig. 1. Map of study area showing sampling locations of farmed salmon (★) and wild salmon (☆) from coastal British Columbia, Canada.

sified as being nonessential (e.g., Pb, Hg, Cd, Ni, and As) or essential/beneficial (e.g., Mg, Mn, Cu, Zn, Se, and Co). Accumulation of metals in the tissues of fish depends primarily on ambient water concentrations, levels in prey or commercial feed, and chemical uptake and elimination kinetics [10]. Other factors, such as chemical speciation/bioavailability as well as fish growth cycle, age, and trophic position, also can influence the extent of metal accumulation in fish. For example, large, long-lived, predatory fish, such as tuna, tend to exhibit high methylmercury (MeHg) levels because of their relatively long life span and high trophic position [11].

The majority of studies of metals in fish have focused on Hg, Cd, and Pb. In particular, investigations of MeHg (along with total Hg [THg]) have been reported frequently because of the relatively high toxicological risks from this highly ubiquitous and toxic organometallic substance to ecological and human health [12–14]. Foran et al. [15] recently reported measured concentrations of several trace elements in the flesh of farmed and wild salmon purchased from commercial suppliers in Canada, the United States, Europe, and South America. Their study concluded that metal concentrations in farmed and wild salmon were relatively low and, hence, posed no apparent risks to human health. Also in that study, no substantial differences in metal concentrations were observed between farmed and wild salmon, which is contrary to previous findings for organochlorine contaminants [7,8,16].

In the present study, we report the findings of a comprehensive investigation that involved measurement of Hg (THg and MeHg) and 18 other trace elements in commercial salmon feed as well as in the flesh of three farmed and five wild salmon species from British Columbia. To evaluate the bioaccumulation behavior of Hg and other trace elements in farmed salmon, we compare measured levels and patterns of those contaminants in commercial salmon feed and farmed salmon flesh. To assess potential human health risks, we provide a human exposure assessment of selected trace metals based on estimated metal intake from consumption of farmed and wild British Columbia salmon as well as other common Canadian foodstuffs.

MATERIALS AND METHODS

Samples

Figure 1 is a map of the study area (coastal BC, Canada) showing sampling locations of the various farmed and wild salmon products. Market-size farmed salmon (i.e., Atlantic, coho, and chinook; $n = 110$) and commercial salmon feed ($n = 8$) were collected from eight British Columbia salmon farm sites. Wild salmon (i.e., coho, chinook, pink, chum, and sockeye; $n = 91$) were sampled across a range of sampling dates and geographical locations from coastal British Columbia waters. All samples were analyzed for THg and various trace elements. Methylmercury was measured in a subset of salmon ($n = 22$) and all eight farmed salmon diet samples. Previous studies have relied mainly on fish samples from retail food outlets, but the present study design controlled for sample origins, handling procedures, and storage conditions. The survey was a blinded study, in which the identities of all farmed and wild salmon samples obtained were unknown to the analysts during the analyses. For farmed salmon, we collected fish from both the farm site and the corresponding processing plant at the time of arrival. For one source of farmed Atlantic salmon (Venture Point Farm, Sonora Island, BC, Canada), we also collected and analyzed those salmon at the end of the commercial production line, which represented the market-ready product. It should be noted that bones are carefully removed in the commercial processing plants. Thus, the final market-ready salmon flesh samples from Venture Point Farm were fully processed boneless fillets. Full details regarding sample collection, storage, and handling are available in the Supplemental Data (http://dx.doi.org/10.1897/07-527.S1). A summary of sampling dates, salmon species, and number of fish sampled as well as their respective mean sizes also is provided in the Supplemental Data (Tables S1 and S2; http://dx.doi.org/10.1897/07-527.S1).

Chemical analysis

All samples of salmon flesh (fillet) and commercial feed were analyzed for THg and 18 trace elements (Ag, Al, As, Ba, Cd, Co, Cr, Cu, Mg, Mn, Ni, Pb, Rb, Se, Sr, Tl, V, and Zn). Methylmercury was determined in feed samples as well as in selected salmon samples. Trace element analyses were conducted at the University of Manitoba (Winnipeg, MB, Canada) and Norwest Labs (Surrey, BC, Canada). Measurements of Hg (THg and MeHg) were conducted at the Fisheries and Oceans Canada's Freshwater Institute (Winnipeg, MB). Trace elements were measured using inductive coupled plasma–mass spectrometry following standard methods (U.S. Environmental Protection Agency method 6020 [17]). Briefly, approximately 1 g of tissue was digested in 5 ml of HNO_3 and 0.5 ml of H_2SO_4 at 125°C overnight. Hydrogen peroxide was then added dropwise to clear the digest, which was then reheated, with more H_2O_2 being added until clear. The digested sample was diluted to 25 ml with water. Analysis was then performed by inductive coupled plasma–mass spectrometry using an ELAN Dynamic Reaction Cell II (PerkinElmer, Wellesley, MA, USA) at a plasma power of 1,100 W and a sample flow rate of approximately 1 ml/min. The instrument was optimized daily for nebulizer gas flow, lens voltage, and autolens to obtain optimum sensitivity. Elements were run in the standard mode except for Se and Fe, which were run in the dynamic reaction cell mode with methane (0.6 L/min) and ammonia (0.5 L/min) as their respective reaction gases to eliminate polyatomic interferences.

Total Hg and MeHg analyses were conducted using cold-vapor atomic absorption spectroscopy and gas chromatography–electron-capture detection, respectively [18]. For THg, approximately 0.2 g of tissue was digested in 1 ml of HNO_3 and 3 ml of HCl at 90°C for 2 h. After digestion, the mixture was diluted to a volume of 25 ml with water. Analysis of THg was performed by cold-vapor atomic absorption spectroscopy with 2% stannous chloride reductant in 15% HCl. For MeHg, an approximately 0.2-g sample was homogenized with a solution of acidic sodium bromide and copper sulfate and extracted into a toluene phase. Methylmercury was then partitioned into an aqueous thiosulfate solution and, subsequently, by addition of potassium iodide, was back-extracted into toluene. Methylmercury in this final extract was measured on a model 3400 gas chromatograph (Varian, Palo Alto, CA, USA) equipped with a 5-m, SPB-5 megabore column and ^{63}Ni electron-capture detector.

Quality-assurance/quality-control measures included the extraction and analysis of procedural blanks, duplicate samples, and certified reference materials (CRMs), including CRM 2976A and CRM 2976-B (National Institute of Standards and Technology, Gaithersburg, MD, USA) as well as DORM-2A, DOLT-3, TORT-1A, TORT-1B, and TORT-1C (National Research Council Canada, Ottawa, ON). Method detection limits were determined as threefold the standard deviation of the procedural blanks and ranged from 0.0001 to 0.005 μg/g wet weight.

Data analysis

Chemical concentration data (μg/g wet wt) were expressed as the geometric mean ± standard deviation or corresponding 95% confidence interval. Mercury and Se data were further translated into molar concentrations to assess molar Se to Hg ratios as described in previous studies [19–21]. Specifically, wet-weight Se and Hg concentrations were expressed as molar values using the respective atomic weights of Se (78.96 g/mol) and Hg (200.59 g/mol). Analyses of variance and Tukey's post hoc tests were performed to evaluate chemical concentration differences between various sites and/or species. Biomagnification factors (BMFs) were determined for the various elements and MeHg in farmed salmon, calculated as the ratio of mean wet-weight concentrations in salmon flesh (C_F) to commercial salmon diet (C_D; i.e., BMF = C_F/C_D, wet wt). We assessed human dietary exposure of Hg, Cd, Pb, Cu, As, and Ni via consumption of British Columbia salmon and other Canadian foodstuffs, such as fruits, vegetables, poultry, beef, pork, milk, and eggs, using observed metal concentrations in British Columbia salmon (present study) and previously reported metal concentrations in Canadian domestic foodstuffs by the Canadian Food Inspection Agency (http://www.inspection.gc.ca/english/fssa/microchem/resid/residfse.shtml). Daily intakes (μg/d) of metals from these various dietary sources were determined from observed metal concentrations and estimated food consumption rates (Table S3 [http://dx.doi.org/10.1897/07-527.S1]). Mean exposure estimates (μg/kg body wt/d) for the various food sources were then calculated for a 70-kg body weight. Theoretical maximum daily intakes (μg/kg body wt/d) were determined using maximum reported metal concentrations. Mean exposure estimates and theoretical maximum daily intakes were then compared to provisional tolerable daily intakes (PTDIs; μg/kg body wt/d) as proposed by the Joint Food and Agricultural Organization of the United Nations/World Health Organization Expert Committee on Food Additives [22] (http://jecfa.ilsi.org).

RESULTS AND DISCUSSION

Concentrations in farmed and wild salmon

Concentrations (μg/g wet wt) of THg, MeHg, and 18 other trace elements in the various species/locations of farmed and wild British Columbia salmon are provided in Table S4 (http://dx.doi.org/10.1897/07-527.S1). The percentage of samples with detectable concentrations for each analysis is given in Table S5 (http://dx.doi.org/10.1897/07-527.S1). With the exception of Ag, Ba, Cd, and Se, metals were detected in all samples analyzed. Figure 2, which illustrates mean concentrations observed in the flesh of all sources of farmed and wild salmon, demonstrates a wide range of concentrations depending on the metal under consideration (0.001–250 μg/g). Concentrations of nonessential elements ranged from 0.001 μg/g for Tl, Cd, and Ag to 1 μg/g for As, Rb, and Al. Concentrations of Pb, Ni, and THg were between 0.01 and 0.05 μg/g. Essential/beneficial element concentrations ranged between 0.01 to 0.1 μg/g for Co, Se, and Mn; 1 to 5 μg/g for Cu and Zn; and more than 250 μg/g for Mg. Observed concentrations of Mg, Zn, and Se were comparable to previous reports from nutritional studies of British Columbia salmon products [23]. In general, element concentrations observed in these British Columbia salmon are comparable to those recently reported by Foran et al. [15]. Their survey of metals in salmon documented significant differences between farmed and wild fish for a number of trace elements (Co, Cu, Cd, and As). In contrast, in the present study, we did not detect significantly higher concentrations of As in farmed compared to wild salmon flesh. Also, we did not observe significantly higher concentrations of Co, Cu, and Cd in the flesh of wild compared to farmed fish; however, we did observe flesh Hg concentrations in wild British Columbia salmon that were significantly (threefold) higher than those concentrations in farmed salmon. In addition, we

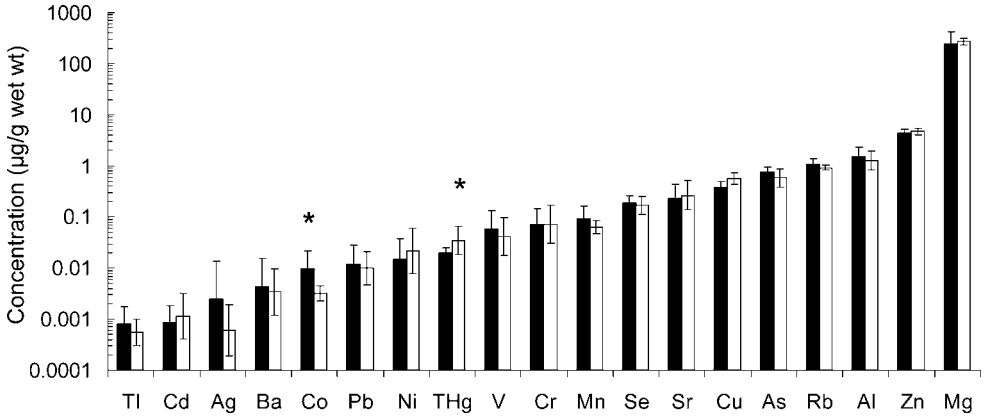

Fig. 2. Concentrations (µg/g wet wt) of 19 trace elements measured in farmed salmon (■) and wild salmon (□) from British Columbia, Canada. Error bars represent ± one standard deviation. An asterisk indicates a significant difference ($p < 0.05$) between farmed and wild salmon concentrations. THg = total Mercury.

observed flesh concentrations of Co in wild British Columbia salmon that were significantly (threefold) lower than those concentrations in farmed fish.

Our analysis of a subset of salmon samples ($n = 22$) that were analyzed for MeHg showed that on average, 97.1% of the THg was in the organometallic MeHg form. This is consistent with previous studies that have reported more than 90% of THg in the flesh of salmonids as the relatively more toxic MeHg compared to inorganic Hg [12]. Conversely, in the case of As, the inorganic form tends to pose the greatest toxicological risk to organisms. In the present study, we only measured total As for British Columbia salmon. Recent studies of farmed and wild salmon [15], however, indicate that the organic form represents 100% of total As, because no inorganic As has been detected. Thus, concentrations of As in these farmed and wild British Columbia salmon (range, 0.3–1.9 µg/g) likely represent levels of the relatively less toxic organic As.

We observed no significant differences in trace element concentrations between farmed Atlantic salmon sampled before commercial processing and those market-ready products obtained postprocessing (Fig. S1 [http://dx.doi.org/10.1897/07-527.S1]). Concentrations of some metals in the market-ready boneless fillets may potentially be lower following processing because of extensive removal of bones, which can be a major storage compartment for trace elements [10]. Alternatively, metal contamination also can occur during handling and processing, which can thereby enhance human exposure via consumption [24]. The present study, however, indicates that negligible losses or gains (i.e., contamination) of trace elements occur during processing of these farmed salmon products.

Interspecies and site-specific variation

Figure 3 illustrates measured concentrations of THg, Cd, Pb, Ni, As, and Cu in the various species of farmed and wild British Columbia salmon, along with those same concentrations in commercial salmon feed. With the exception of THg, concentrations of metals in commercial salmon feed were significantly higher ($p < 0.01$) than concentrations in farmed and wild salmon. In general, concentrations in feed samples were comparable to levels reported in previous studies of physical and chemical characterization of commercial feed pellets used in British Columbia salmon farms [25]. Interspecies comparisons revealed that individual metal concentrations did not vary substantially between species of farmed salmon; however, wild piscivorous chinook salmon (the longest-lived of the salmon species studied) exhibited significantly higher (two- to eightfold, $p < 0.05$) concentrations of THg compared to those levels observed in the smaller, shorter-lived, planktivorous species, such as pink and chum (Fig. 3). For other metals, such as Cd, Pb, and Ni, concentrations differed slightly between wild salmon species, but no species or site-specific accumulation pattern was apparent. In contrast to our previous findings of organochlorines in these salmon [8], farmed Atlantic salmon do not appear to exhibit elevated metal concentrations compared to other species of British Columbia salmon (farmed or wild). It was shown previously that farmed Atlantic salmon exhibited three- to eightfold higher PCB concentrations compared to all other salmon sources in British Columbia [8]. Comparatively higher levels of lipophilic organic contaminants, such as PCBs, in farmed Atlantic salmon are attributed to the significantly higher lipid contents in those fish (>15%).

For metals, which generally are non–fat soluble compounds, lipid contents have less influence on flesh residue concentrations compared to their concentrations in prey and their uptake and elimination kinetics. Metals are well known to accumulate in organisms, but they often exhibit biodilution in organisms and food chains rather than biomagnification [21,26,27]. Biodilution is defined here as a situation in which element concentrations on a wet-weight basis are lower in fish compared to those levels in consumed food. A notable exception is MeHg, which is a relatively hydrophobic compound and tends to biomagnify (i.e., to increase in concentration with increasing trophic level) [13,21]. Thus, higher observed Hg and As concentrations in chinook likely result from the combination of higher Hg and As levels in forage fish (chinook diet) compared to those in lower-trophic-level zooplankton (pink and chum diet) and greater longevity in chinook. Observed interspecies variation of other metals among wild salmon (Fig. 3) may reflect geographic variation of those metals in British Columbia coastal waters as well as species-specific differences in life history and bioenergetics.

Bioaccumulation behavior of metals in farmed and wild salmon

The fact that metal concentrations in commercial feed samples generally were higher than concentrations in farm-raised

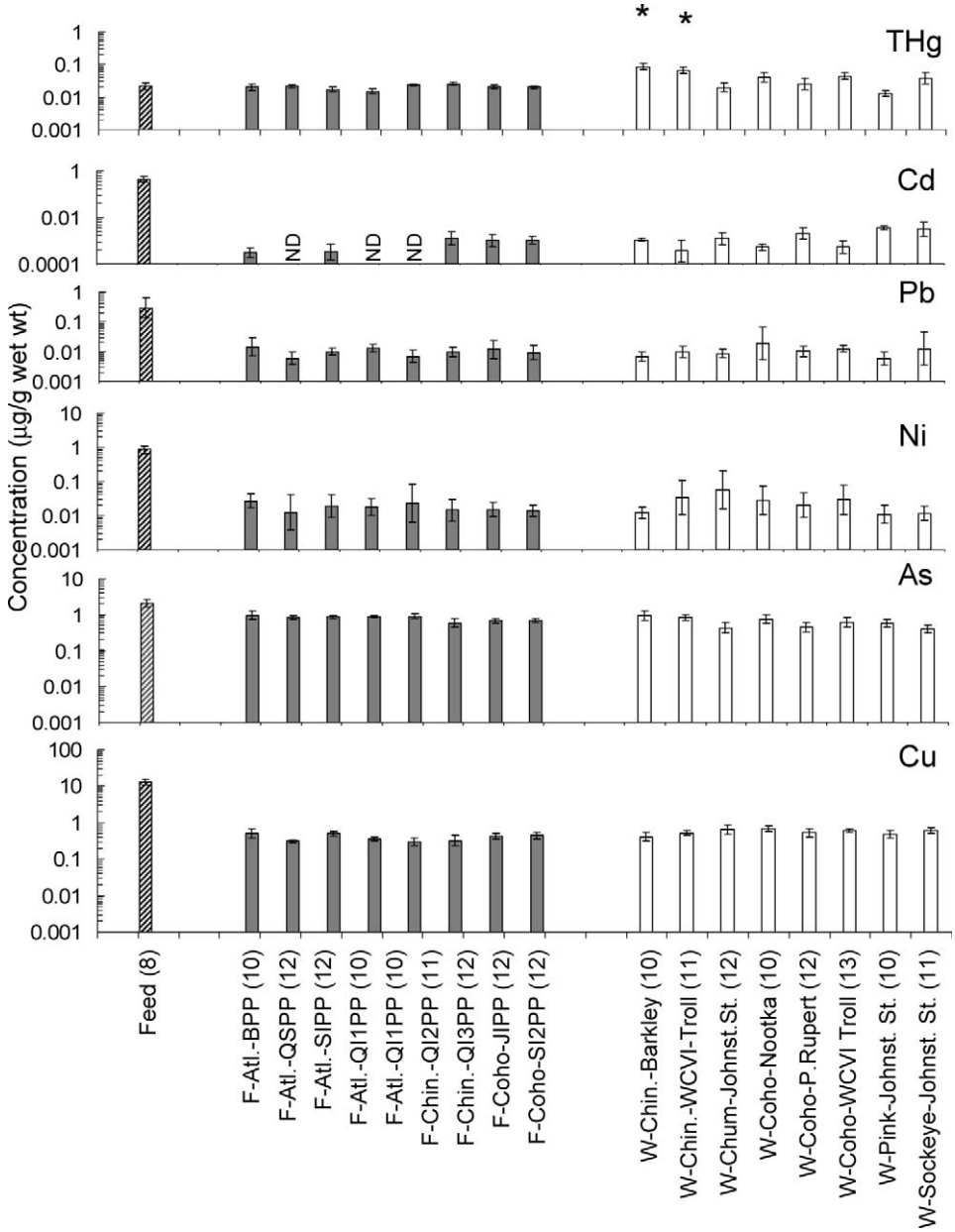

Fig. 3. Concentrations (µg/g wet wt) of total mercury (THg), Cd, Pb, Ni, As, and Cu measured in commercial salmon feed (▨), farmed salmon (i.e., farmed Atlantic, chinook, and coho [■]) and wild salmon (i.e., wild chinook, chum, coho, pink, and sockeye [□]) from British Columbia, Canada. Error bars represent ± one standard deviation. An asterisk indicates significantly higher ($p < 0.05$) concentrations compared to other wild salmon species. Farmed salmon samples were obtained on arrival at the processing plant (PP) and include farmed Atlantic salmon (F-Atl.) from Broughton (BPP), Quatisino Sound (QSPP), Sechelt Inlet (SI1PP), and Quadra Island (Ql1PP and QI2PP); farmed Chinook (F-Chin) from Quadra Island (QI3PP and QI4PP); and farmed coho (F-Coho) from Jervis Inlet (JIPP) and Sechelt Inlet (SI2PP). Wild salmon sources include wild Chinook (W-Chin.) from Barkley Sound (Barkley) and West Coast Vancouver Island (WCVI-Troll); wild chum (W-Chum) from Johnstone Strait (Johnst. St.); wild coho (W-Coho) from Nootka Sound (Nootka), Prince Rupert (P. Rupert), and West Coast Vancouver Island (WCVI-Troll); wild pink salmon (W-Pink) from Johnstone Strait (Johnst. St.); and wild sockeye (W-sockeye) from Johnstone Strait (Johnst. St). Numbers in parentheses represent sample size. ND = not detected.

salmon (Fig. 2 and Table S4 [http://dx.doi.org/10.1897/07-527.S1]) indicates low biomagnification potential. Calculated BMFs of THg, MeHg, and other trace elements for three species of farmed salmon (Atlantic, coho, and chinook) are shown in Table 1. Trace elements generally exhibited BMFs of less than one. Biomagnification factors of Cd were very low, ranging between 0.002, 0.002, and 0.005 in farmed Atlantic, coho, and chinook, respectively. Biomagnification factors of Pb, Cu, and Ni ranged between 0.01 and 0.07 in those farmed salmon. In our previous study of organohalogens, such as PCBs in British Columbia salmon [8], we observed moderate biomagnification of those compounds in farmed salmon (i.e., BMF range, 1.5–3). Biodilution of trace metals has been documented previously in aquatic invertebrates and fish [26]. Biomagnification factors of Hg (THg and MeHg) were slightly higher compared with other metals and ranged between 0.8 and 2. Biomagnification factors in these farmed salmon generally are comparable with BMFs of metals in wild fish [21,26,28]. For example, in a pelagic Arctic marine food web, THg and MeHg BMFs in Arctic cod were between 1.5 and 3, whereas metals, such as Cd, Ni, and Pb, exhibited significant biodilution with BMFs of less than one in those fish [21].

Table 1. Biomagnification factors (BMFs) of total mercury (THg), methylmercury (MeHg), and other trace elements in farmed Atlantic, coho, and chinook salmon from British Columbia, Canada

	BMF		
	Farmed Atlantic	Farmed coho	Farmed chinook
Mercury			
THg	0.94	0.78	1.3
MeHg[a]	1.3	0.86	1.9
Other elements			
Ag	0.038	0.012	0.003
Al	0.052	0.1	0.1
As	0.43	0.37	0.33
Ba	0.005	0.002	0.001
Cd	0.002	0.002	0.005
Co	0.004	0.004	0.025
Cr	0.18	0.1	0.24
Cu	0.03	0.036	0.03
Mg	0.16	0.19	0.2
Mn	0.004	0.003	0.003
Ni	0.037	0.016	0.048
Pb	0.056	0.072	0.044
Rb	0.33	0.35	0.38
Se	0.19	0.21	0.25
Sr	0.009	0.007	0.007
Tl	0.045	0.18	0.061
V	0.15	0.12	0.12
Zn	0.025	0.029	0.034

[a] The BMFs of MeHg in farmed salmon were determined using measured MeHg concentrations in salmon feed at the corresponding farm but estimated MeHg concentrations in farmed salmon flesh. For this purpose, we assumed that farmed salmon flesh exhibit similar MeHg content as wild salmon flesh, which was approximately 97% of THg concentrations.

Biomagnification of MeHg has been attributed to the relatively high dietary uptake efficiency and slow elimination kinetics of this relatively more hydrophobic compound [10,26]. Typically, MeHg biomagnification is comparable to that of other hydrophobic organic contaminants, such as PCBs, but other metals, such as Cd, Pb, Ni, Cu, and Ag, generally are absorbed less efficiently and, hence, accumulate only slightly over time (even though their biological half-lives are relatively long). Furthermore, once absorbed, many metals, such as Cd and Pb, in fish are stored preferentially in tissues/compartments other than muscle, such as liver, kidney, and bone; hence, they undergo negligible mass transport to fish flesh [10]. It is important to note that inclusion of such tissues in chemical analyses may result in different BMFs. Because the present study was concerned primarily with the flesh quality of these salmon, we did not include these other constituents in our analyses. Organism growth also can play a key role in the degree of chemical bioaccumulation [29,30]. For example, during periods of rapid or extended growth, absorbed contaminants tend to undergo a dilution effect because of the increasing presence of acquired macronutrients, such as proteins and lipids. Conversely, once growth slows and the organism approaches maximum growth potential (i.e., steady-state condition) or when growth declines during periods of fasting, internal tissue residue concentrations may potentially increase if chemical elimination rates are very slow. Thus, organism-specific bioenergetics and chemical-specific toxicokinetics both influence the bioaccumulation potential of a given contaminant. In biodynamic analyses of metals in aquatic organisms, growth dilution has been cited as a major factor causing much of the variability in metal bioaccumulation [26]. Growth dilution is particularly important for farmed salmon, because those fish exhibit very fast growth rates during their relatively short seawater residency period of 12 to 23 months (depending on the species) compared to two- to five-year life spans for wild salmon. Because of growth dilution in farmed fish, the BMFs of the various metals reported here in farmed salmon may be somewhat lower than those attained in mature wild fish, which likely are near a steady-state growth condition.

We observed a significant relationship between THg flesh concentration and fish size (kg), but only in wild chinook salmon ($r^2 = 0.4$) (Fig. S2 [http://dx.doi.org/10.1897/07-527.S1]). The data show that THg concentrations ranged from approximately 0.01 μg/g for a 2-kg fish to 0.1 μg/g for an 8-kg fish. Previous studies have reported strong positive relationships for THg and fish size [31,32], which is indicative of the relatively high bioaccumulation potential of MeHg. We did not observe a positive THg–fish size relationship in any of the farmed salmon species over the same weight range (2–7 kg) (Fig. S2 [http://dx.doi.org/10.1897/07-527.S1]). The lack of correlation between THg and weight of farmed salmon may result from growth dilution in these fish.

We did not observe significant relationships between molar Hg and molar Se concentrations for any of the farmed or wild salmon species (Fig. S3a [http://dx.doi.org/10.1897/07-527.S1]). Some previous studies have observed strong positive correlations ($r^2 > 0.6$) between Hg and Se levels in aquatic invertebrates, fish, birds, and marine mammals [19–21]. Other studies, however, have found no positive relationships between Hg and Se levels in various marine and freshwater fish species [33,34]. Because Se and Hg concentrations showed no correlation in farmed and wild British Columbia salmon (present study), molar Se to Hg ratios in these fish were quite variable, ranged between 3 to 100, and decreased with increasing THg concentration (Fig. S3b [http://dx.doi.org/10.1897/07-527.S1]). The Se to Hg ratios were similar between farmed salmon (mean ratio, 25) and wild salmon (mean ratio, 18). These Se to Hg ratios in British Columbia salmon are consistent with previously observed Se to Hg ratios in aquatic organisms. For example, Se to Hg ratios of 1 to 100 have been found in muscle samples of Arctic fish from Greenland [20]. The Se to Hg ratio can differ substantially between organisms and between tissues/organs. For example, Se to Hg ratios can range from less than 1 to more than 1,000 between various species of Arctic invertebrates, fish, birds, and mammals [21]. Furthermore, mean Se to Hg ratios observed in those Arctic fish were significantly different for muscle (mean ratio, 38), liver (mean ratio, 198), and kidney (mean ratio, 162). Tracking the degree of Se in a given organism or tissue/organ (relative to Hg) is important for assessing ecotoxicological and human health risks, because several studies have demonstrated that this beneficial element acts to mitigate the toxic effects of Hg in organisms [35,36]. In certain quantities, Se, through various detoxifying mechanisms, may therefore counteract Hg toxicity. These protective benefits, however, may become detrimental if Se exposure becomes too high and surpasses toxicological thresholds [20,37]. The data from the present study demonstrate that the majority of farmed and wild British Columbia salmon have a moderate surplus of Se compared to Hg of approximately 10- to 50-fold in excess on a molar basis, which indicates the presence of potentially beneficial levels of Se in these fish.

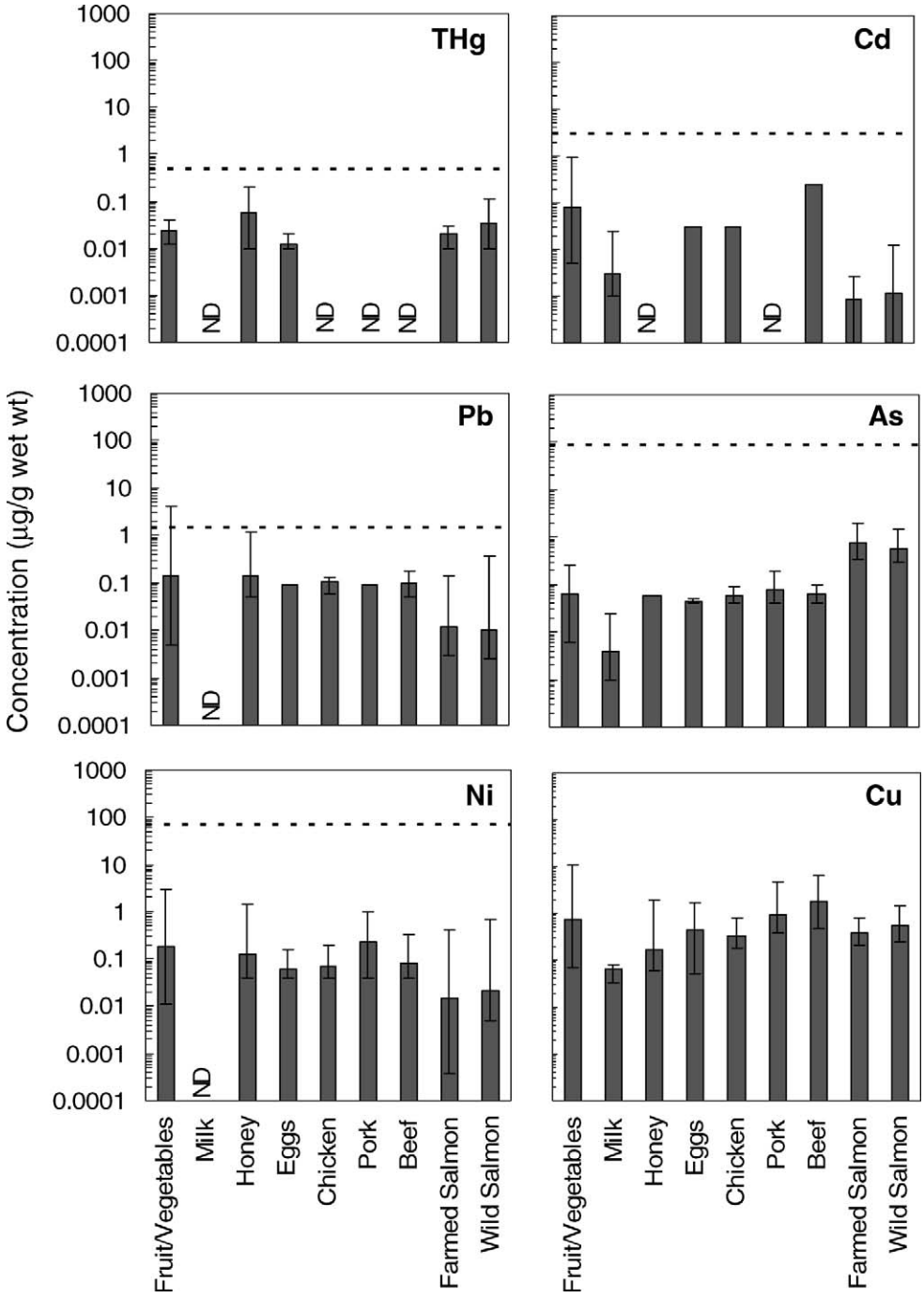

Fig. 4. Concentrations (μg/g wet wt) of total mercury (THg), Cd, Ni, Pb, As, and Cu in the flesh of farmed and wild salmon from British Columbia (BC), Canada, in relation to other foodstuffs (e.g., milk, honey, eggs, chicken, pork, and beef). Error bars represent the range of minimum and maximum levels detected. Threshold concentrations (dashed lines) represent Health Canada standards (maximum level) for retail food products [39] and/or U.S. Food and Drug Administration action levels for metals in crustaceans [38] as follows: THg, 0.5 μg/g (Health Canada); all other levels obtained from the U.S. Food and Drug Administration: Cd, 3.0 μg/g; Ni, 70 μg/g; Pb, 1.5 μg/g; and As, 76 μg/g. ND = nondetectable concentration.

Human exposure assessment

A comparison of measured concentrations of Hg and other trace metals (Cd, Ni, Pb, As, and Cu) in the flesh of farmed and wild British Columbia salmon (present study) to levels reported in other Canadian foodstuffs is shown in Figure 4. Total Hg and As concentrations in British Columbia salmon (both farmed and wild) generally were higher or equivalent to levels reported in other foodstuffs. For example, THg concentrations in British Columbia salmon (farmed + wild mean, 0.03 μg/g) were higher than THg levels reported in chicken, pork, and beef (all nondetectable concentrations, <0.01 μg/g) but were comparable to THg in fruits/vegetables (mean, 0.024 μg/g), honey (mean, 0.056 μg/g), and eggs (mean, 0.012 μg/g). Similarly, concentrations of As in British Columbia salmon (farmed + wild mean, 0.67 μg/g) were higher than those in all other foodstuffs, including milk (mean, 0.004 μg/g), fruits/vegetables (mean, 0.064 μg/g), and meat products (mean, 0.07 μg/g). Conversely, levels of Cd, Ni, Pb, and Cu

Table 2. Estimated human exposure to selected trace metals for persons consuming British Columbia salmon and other domestic Canadian foodstuffs in relation to provisional tolerable daily intake (PTDI) proposed by the Joint Food and Agricultural Organization of the United Nations/World Health Organization Expert Committee on Food Additives[a]

Metal	PTDI (μg/kg body wt/d)		Intake from farmed salmon (μg/kg body wt/d)	Intake from wild salmon (μg/kg body wt/d)	Total intake (μg/kg body wt/d)	% PTDI from salmon	% PTDI from total diet
As	50	Mean	0.2	0.1	1.2	0.76%	2.4%
		Min–max	(*0.1–0.5*)	(*0.09–0.4*)	(*0.5–3.2*)	(*0.4–2%*)	(*1–6%*)
Cd	1	Mean	*0.0002*	*0.0003*	*1.1*[b]	*0.06%*	*100%*[b]
		Min–max	(*0.00001–0.004*)	(*0.00002–0.02*)	(*0.8–5.3*)	(*0.002–0.4%*)	(*79–100%*)
Cu	57	Mean	*0.1*	*0.2*	*12*	*0.5%*	*22%*
		Min–max	(*0.06–0.2*)	(*0.07–0.4*)	(*3.2–83*)[c]	(*0.2–1%*)	(*6–100%*)[c]
Ni	71	Mean	*0.004*	*0.006*	*1.9*	*0.01%*	*2.7%*
		Min–max	(*0.0001–0.1*)	(*0.001–0.2*)	(*0.35–19*)	(*0.002–0.4%*)	(*0.5–26%*)
Pb	4	Mean	*0.003*	*0.003*	*1.4*	*0.2%*	*39%*
		Min–max	(*0.0008–0.04*)	(*0.0007–0.1*)	(*0.5–21*)[c]	(*0.04–4%*)	(*15–100%*)[c]
THg	0.7	Mean	*0.006*	*0.009*	*0.14*	*2.2%*	*20%*
		Min–max	(*0.003–0.009*)	(*0.003–0.03*)	(*0.07–0.26*)	(*1–6%*)	(*9–38%*)

[a] Values in italics represent the range of minimum to maximum estimates. THg = total mercury.
[b] Estimated mean value exceeds PTDI.
[c] Estimated maximum value exceeds PTDI.

in British Columbia salmon generally were lower than those in other foodstuffs. For example, Pb levels in British Columbia salmon (farmed + wild mean, 0.01 μg/g) were relatively low compared to Pb levels in fruits/vegetables (mean, 0.14 μg/g) and meat products (mean, 0.1 μg/g).

Metal concentrations in farmed and wild British Columbia salmon do not exceed U.S. and Canadian human consumption guidelines [38,39] (http://hc-sc.gc.ca/fn-an/pubs/mercur/merc_fish_poisson_e.html). Even the maximum observed concentrations measured in the present study for British Columbia salmon were well below guidelines (Fig. 4). For example, the highest level of As reported in the present study (1.93 μg/g), which was for a farmed Atlantic salmon, was approximately 50-fold below the 86 μg/g guideline set for As in fish. Similarly, the highest level of THg in British Columbia salmon (0.1 μg/g), which was observed in a wild chinook, was below the 0.5 μg/g THg guideline set by Health Canada. Maximum concentrations of Pb in wild chinook (0.35 μg/g) also were well below the 1.5 μg/g guideline for Pb in fishery products. In some cases, however, metal concentrations in other foodstuffs approach or exceed those guidelines. For example, in the present study, maximum reported Pb concentrations in fruits/vegetables (4.2 μg/g) exceed the 1.5 μg/g threshold. The data indicate that metal concentrations in British Columbia salmon exhibit relatively safe levels and generally are equivalent to or lower than levels observed in other common domestic foodstuffs, such as fruits, vegetables, dairy, honey, and meat products.

Table 2 shows daily intakes (μg/kg body wt/d) of several metals calculated for a 70-kg person consuming a typical North American diet consisting of fruits, vegetables, dairy, eggs, meats, and fish, which includes two 140-g servings of British Columbia salmon per week (one serving farmed + one serving wild). Also shown in Table 2 are the PTDIs (μg/kg body wt/d) of those metals as proposed by the Joint Food and Agricultural Organization of the United Nations/World Health Organization Expert Committee on Food Additives. For the total diet, estimated mean daily intakes ranged from 0.14 μg/kg body weight/d for Hg to 12 μg/kg body weight/d for Cu. For Cd, Pb, Cu, and Ni, the contribution of total intake from salmon generally was small (0.04–2% of total exposure) compared to that from other foodstuffs. Estimated intake of Hg from salmon consumption, however, was slightly higher, at 11% of total exposure. Similarly, As intake from salmon was estimated at 32% of total As intake. The data in Table 2 further highlight the fact that metal exposure from farmed salmon is equivalent to that from wild salmon. For example, mean daily intakes of Hg for farmed and wild British Columbia salmon were 0.006 and 0.009 μg/kg body weight/d, respectively.

Human exposure estimates of metals from consumption of a typical diet generally were below the recommended PTDIs (Table 2). For example, daily intake of Hg from all food sources was only 20% of the PTDI of 0.7 μg/kg body weight/d. Similarly, As and Ni intakes (40-fold below PTDIs) only contributed approximately 2% of PTDIs. In some cases, however, metal exposure approached the PTDI. Mean intake of Cd from all food sources (1.1 μg/kg body wt/d) was equivalent to the PTDI of 1 μg/kg body weight/d set for Cd. Theoretical maximum daily intake for Cd (5.3 μg/kg body wt/d), as determined from maximum observed concentrations of all food sources, indicates that intake may potentially exceed the PTDI by a factor of five. Theoretical maximum daily intakes of Cu and Pb exceed those PTDIs by 1.5- to 5-fold, respectively. Provisional tolerable daily intakes generally are established using empirical no-observable-adverse effect levels and application of safety factors (generally a factor of 10–100) to account for interspecies variation and animal size. Whereas our calculations show that metal exposure related to consumption of farmed and wild British Columbia salmon is very low, the fact that dietary exposure estimates of Cd, Pb, and Cu from other common Canadian foodstuffs (mainly fruits, vegetables, and meat products) can approach and potentially exceed PTDIs is cause for some concern. Metal contamination in human foodstuffs remains an important issue facing nations worldwide. A recent assessment of metal contamination of foodstuffs in the European Union revealed similar results, indicating that metal exposures from common foods exceed PTDIs [40].

CONCLUSION

The accumulation of environmental contaminants, such as metals, pesticides, PCBs, and dioxins, in human food chains is a major public health concern. The present study involved a comprehensive investigation of Hg and other trace elements

in farmed and wild British Columbia salmon. The data indicate that with the exception of Hg, metal concentrations do not differ substantially between farmed and wild British Columbia salmon and exhibit negligible interspecies and size-specific variation. Comparison of metal concentrations in farmed salmon and their feed indicated substantial biodilution of metals in these fish (denoted by BMFs $\ll 1$). Estimates of human dietary exposure indicate that human health risks associated with trace metal exposure via consumption of farmed and wild British Columbia salmon are negligible. Even Hg and As, which are notorious contaminants in seafood worldwide, exhibit relatively low levels in farmed and wild British Columbia salmon—well below human health consumption guidelines. In addition to the fact our previous investigations showed relatively low risks associated with residue flesh levels of PCBs and dioxins and a net health benefit from *n*-3 HUFAs, this further validates the relative safety of farmed and wild British Columbia salmon. The current scientific evidence therefore supports the weekly consumption of oily fish species (including all British Columbia salmon sources) as recommended by the American Heart Association (http://www.americanheart.org/presenter.jhtml?identifier=3013797).

SUPPORTING INFORMATION

Figure S1. Concentrations (μg/g wet wt) of various trace elements in the flesh of one source of farmed Atlantic salmon (i.e., Venture Pt farm Atlantic salmon) prior to entering the processing plant ($n = 13$) and in the end-of-line market-ready boneless fillet ($n = 13$).

Figure S2. Relationship between concentrations (μg/g wet wt) of total mercury (THg) in the flesh and fish weight (kg) for (A) farmed salmon and (B) wild salmon. Regression lines represent THg-weight relationship using data for individual species of wild salmon. Regression results are as follows: wild chinook ($y = 0.0076\ x + 0.0519$, $r^2 = 0.40$), wild coho ($y = 0.0041x + 0.0268$, $r^2 = 0.1$); wild chum ($y = -0.0032\ x + 0.0332$, $r^2 = 0.34$); wild sockeye ($y = 0.0064x + 0.0231$, $r^2 = 0.1$).

Figure S3. Plots showing (A) relationship between concentrations of molar concentrations (mol/m^3) of total mercury (THg) and selenium (Se) in the flesh for all farmed salmon and (B) corresponding Se:Hg ratios (molar basis) in those fish relative to molar THg concentrations.

Table S1. Farmed salmon samples analyzed for fillet concentrations of organohalogen contaminants, trace metals, proximate constituents, fatty acids, carotenoid pigments, and gross energy.

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

Table S3. Food consumption rates (g/d) and corresponding concentrations (μg/g) of THg, Cd, Cu, Ni, Pb, and As used in the human exposure calculations of those metals.

Table S4. Concentrations of mercury and other trace elements (μg/g wet wt) in the commercial feed (for farmed salmon) and fillets of farmed and wild salmon from coastal British Columbia, Canada. Data are reported as geometric means along with 95% confidence intervals. Numbers in brackets represent number of samples analyzed. NA = substance not analyzed in those samples.

Table S5. Percentage (%) of samples detected.

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