

CONCENTRATIONS OF SELECTED PERSISTENT ORGANOCHLORINE CONTAMINANTS IN STORE-BOUGHT FOODS FROM NORTHERN ALASKA

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ABSTRACT

Objectives. We address marine and terrestrial mammal blubber, liver, muscle, kidney, heart, tongue, *maktak* and *maktaaq* (epidermis and blubber from bowhead, beluga whales, respectively), and fish muscle and livers, as commonly consumed tissues in subsistence communities across northern Alaska in the context of organochlorine (OC) contamination of store-bought foods. Human exposure to contaminants from biota, as part of a subsistence diet, has been superficially evaluated in numerous studies (focused on liver and blubber), but are limited in the type of tissues analyzed, and rarely consider the contaminants in the alternatives (i.e., store-bought foods).

Study Design. Concentrations from published literature on selected persistent organochlorine contaminants (OCs) in eight tissues of the bowhead whale and other biota (1) were compared to store-bought foods evaluated in this study.

Results. As expected, store-bought foods had lower concentrations of OCs than some tissues of the marine mammals (especially blubber, *maktak*, and *maktaaq*). However, blubber is rarely eaten alone and should not be used to give consumption advice unless considered as a portion of the food item (i.e., *maktak*). This study indicates that the store-bought food alternatives have detectable OC concentrations (e.g., < 0.01 to 22.5 ng/g *w.w.* for hexachlorobenzene) and, in many cases, have greater OC concentrations than some subsistence food items. Many wildlife tissues had OC concentrations similar to those quantified in local store-bought food.

Conclusions. Switching from the traditional diet to western store-bought foods will not always reduce exposure to OCs. However, raw blubber-based products are clearly more contaminated with OCs due to lipid content. A detailed profile of traditional/country foods and western foods consumed by subsistence communities of northern Alaska is required to address chronic exposure in more detail for the diverse sources of foods (subsistence use and commercially available) and the widely varying concentrations of contaminants reported therein. This should be combined

with biomonitoring people dependent upon subsistence foods. Further assessment of essential and non-essential elements, emerging contaminants (e.g. brominated flame retardants), etc. should be conducted in order to improve our understanding of the differences and similarities between wildlife and store-bought foods.

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Keywords: commercial foods, country/traditional foods, organochlorines, pollutants, subsistence

INTRODUCTION

The presence of persistent organochlorine contaminants (OCs) in the Arctic is well known. First detected in the region in the late 1960s (2), OCs have been found throughout the Arctic due to their environmental recalcitrance and long-range transport in air, water and biota (3). The persistence and bioaccumulation potential of OCs is a concern in the arctic environment, where many species feed on prey with greater lipid content compared to phylogenetically similar organisms, or agricultural species inhabiting lower latitude ecosystems. Despite the long distances from sources OCs accumulate to relatively high concentrations in top predatory marine species, such as polar bears (*Ursus maritimus*), ringed seals (*Phoca hispida*), seabirds and toothed whales (3).

The accumulation of OCs in arctic biota remains relevant to indigenous peoples of this region, who may be exposed to OCs at greater concentrations than populations in southern Canada or the USA, due to the consumption of lipid-rich traditional foods (4). However, sources of food from more southerly latitudes, such as commercially available foodstuffs, are not devoid of the OCs (e.g., 5-8). Thus, advice on food consumption, based mostly on

the determination of OCs in subsistence use species and rarely the alternatives available in the communities as “store-bought” foods, is incomplete. To make a thorough assessment of the potential risks to human health from dietary exposure to OCs, the alternatives to traditional country foods that are served as the basis for subsistence-based diets should be evaluated and compared.

The subsistence diet of indigenous peoples of the Arctic varies by region due to prey availability, local hunting practices (9) and the availability of resources from “Outside” (the store). For northern Alaska, we focus the comparison on the bowhead whale and a few other locally relevant marine mammals and fish. Several exposure and risk assessments have recognized the socio-cultural and nutritional benefit of traditional foods to arctic peoples (e.g. 10). However, the accumulation of OCs in Native populations from subsistence arctic communities has raised some questions concerning the suitability for human consumption of terrestrial and marine wildlife and fish from this region (4, 10-13). Concentrations of OCs in marine mammal blubber and liver, along with blubber only from other marine mammals from northern Alaska, have been reported (14-18) and, more recently, human exposure was assessed for

the bowhead whale (eight tissue types) and other subsistence food items (1). However, the exposure to OCs from local store-bought alternatives has not been investigated. In this paper we discuss the potential human exposure to OCs and the implications for human consumption of arctic mammalian tissues compared to select foods purchased locally as “alternatives” to more traditional items.

MATERIAL AND METHODS

Sample collection (store-bought foods)

Various commercially available foods [Cornish game hen, boneless pork loin chop, milkfish (*Chanos chanos*), smoked salmon strips (*Oncorhynchus sp.*), imitation crab flakes, beef shank and tongue, honeycombed tripe (rumen), reindeer steak and steak marrow (*Rangifer tarandus*), lobster tail (possibly *Panulirus argus*), chicken egg yolk, sardines (Western Family Sardines, likely *Sardinella auritas*, lightly smoked in soybean oil with salt), and canned salmon (*O. gorbuscha*; salt)] were obtained from a local market in Barrow, AK, USA, by local residents in 2002. Samples were selected based on their availability and potential dietary importance as a substitute for subsistence foods (i.e. country-based diet) (Fig. 1). Samples were transported to the National Water Research Institute (Environment Canada, Burlington, Ontario, Canada). We expected that the concentrations of OCs in the store-bought foods would be low and this presumption led to pooling of samples to increase the available mass for chemical analyses. All samples were homogenized and stored at -20 °C in pre-cleaned glass containers.

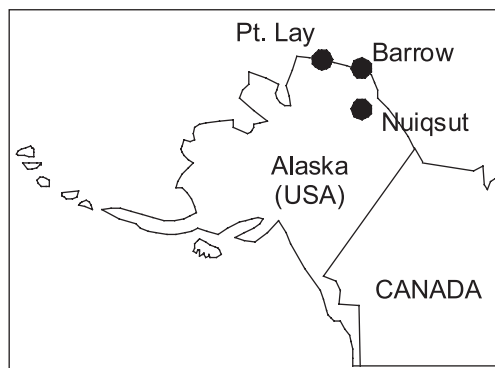


Figure 1. Sampling sites (*) of biota and anadromous fish at Pt. Lay (beluga only), Nuiqsut (arctic grayling, burbot and broad whitefish only) and Barrow, Alaska, USA (1997 – 1999), and store-bought foods at Barrow, Alaska.

Chemicals and Standards

All solvents (pesticide grade) were obtained from Caledon Laboratories. (Georgetown, ON, Canada). American Chemical Society-grade granular sodium sulfate (Na_2SO_4) was obtained from EM Science (Gibbstown, NJ, USA). Pesticide-grade dry silica (60 – 200 mesh) was obtained from ACP (Montréal, PQ, Canada). SX-3 Biobeads (200 – 400 mesh) used in gel permeation chromatography columns were purchased from Bio-Rad Laboratories (Hercules, CA, USA).

Extraction and clean-up of store-bought food samples

Samples of store-bought foods (approximately 10 – 20 grams, wet weight; *w.w.*) were extracted using previously described techniques with minor modification (Hoekstra *et al.*, (14)). Samples were homogenized with sodium sulphate (Na_2SO_4) and spiked with a mixture of internal standards (two polychlorinated biphenyl (PCB) congeners, CB-30 and CB-204; δ -substituted hexachlorocyclohexane (δ -HCH), 1,3,5-tribromobenzene, and 1,2,4,5-tetrabromobenzene) to monitor the efficiency

of the extraction protocol. Samples were extracted with dichloromethane (DCM) using Soxhlet extraction for 16 hours and subsequently passed through an Allihn funnel containing sodium sulfate, and concentrated. Lipids and other bioorganic materials in each sample were removed using gel permeation chromatography (GPC) and lipid percent was determined gravimetrically. The analyte sample was concentrated and separated on 8 grams of 100% activated silica gel into two fractions: 65 mL of 100% hexane (F1) and 90 mL of 50% hexane: 50% dichloromethane (F2). Endrin ketone and 1,3 dibromobenzene were added as laboratory spiking surrogates to determine fractionation performance. Samples were rotary evaporated, transferred to 2,2,4-trimethylpentane (iso-octane) and concentrated to 1000 μ L.

PCB/OC pesticide analysis

PCB and OC pesticide analysis on both sample fractions was performed using a Hewlett Packard (Wilmington, DE, USA) 6890 gas chromatograph (GC) with a ^{63}Ni electron capture detector (ECD). Pulsed splitless injections of 1 μ L volumes were performed by a HP 7683 autosampler and a HP 7683 Series Inject with a splitless time of 1.0 min (injector temperature set at 250 $^{\circ}\text{C}$). Compound separation was completed using a 30 m \times 0.25 mm (i.d.) HP-5MS column (internal film thickness 0.25 μm ; Wilmington, DE, USA) with H_2 carrier gas (at a constant flow rate of 1.1 mL/min). Nitrogen was used as the makeup gas for the ECD (detector temperature: 350 $^{\circ}\text{C}$). The oven temperature program was initiated at 80 $^{\circ}\text{C}$ (held 2.0 min), ramped to 90 $^{\circ}\text{C}$ at 10 $^{\circ}\text{C}/\text{min}$, then ramped at 2.5 $^{\circ}\text{C}/\text{min}$ to 285 $^{\circ}\text{C}$ (5 min hold time) and maintained till

the completion of the 86-minute run. Sample quantification was performed using multiple external standards obtained from the National Laboratory for Environmental Testing (Environment Canada, Burlington, ON, Canada) that were analyzed after every 10 samples. Sum (Σ) of chlordane (ΣCHL) includes technical products *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; the sum of *o,p'*- and *p,p'*-substituted DDT, DDE, and DDD are designated as ΣDDT ; sum of α -, β - and γ -HCH isomers (ΣHCH); sum of PCB congeners (ΣPCB) CB-4/10, 5/8, 6, 7/9, 12/13, 15/17, 16, 18, 19, 21/33/53, 22, 24/27, 25, 26, 28, 29/54, 31, 32, 40, 41/71, 42, 43, 44, 45, 46, 47/48, 49, 50, 51, 52, 55, 56/60, 59, 63, 64, 66/95, 70, 74, 76/98, 82, 83, 84/92, 81, 85, 87, 91, 97, 99, 100, 101, 105, 107/147, 110, 114, 118, 119, 128, 129/178, 130/176, 132, 133/149, 135, 136, 137, 138, 141/179, 143, 144, 145, 151, 153, 156, 158, 163, 167, 170/190, 172, 173/202, 174, 171, 175, 177, 182, 183, 185, 187, 197, 180, 189, 193, 191, 194, 195, 196/203, 198, 199, 201, 205, 206, 207, 208 and CB-209 (IUPAC designation) and the ΣPCB_{10} is the sum of PCB congeners CB-28, 31, 52, 101, 105, 118, 138, 153, 156 and CB-180 (IUPAC designation)

Analytical quality assurance (store-bought foods)

Average (± 1 SD) sample recovery of PCB and OC surrogate standards was 103% \pm 18% and concentrations were adjusted accordingly. Detection limits ranged from 0.01 to 0.1 ng g^{-1} for individual PCB congeners and OC pesticides and related compounds. The quality assurance protocol included extraction and analysis of laboratory blanks with every batch of 10 samples, the use of a standard

or certified reference materials (SRM1974a Organics in Muscle Tissue (*Mytilus edulis*) from the National Institute of Standards and Technology, Gaithersburg, MD, USA and CRM DF-2525 Lake Ontario Lake trout muscle (*Salvelinus namaycush*) from Environment Canada, National Laboratory for Environmental Testing, Burlington, ON, Canada), and participation in an international inter-laboratory comparison program on PCB analysis (QUASIMEME, Aberdeen, UK). Results are blank subtracted.

RESULTS AND DISCUSSION

It is no surprise that, in some cases, OC levels in store-bought food are not lower than in some traditional wildlife foods, except for blubber and blubber containing products. The OC concentrations in selected tissues from subsistence species are presented in Tables I-II (adapted from Hoekstra *et al.*, (1)). Store-bought food items and OC concentrations are provided in Table III, along with a list of the OCs analyzed in this study. Store-bought terrestrial

Table I. Mean (\pm 1 SD) and median of lipid content (%) and sum (Σ) concentrations of persistent organochlorine contaminant classes and HCB (ng g^{-1} , wet weight) in bowhead whale (*Balaena mysticetus*) tissues and uncooked maktak from Alaska (1997 – 1999), adapted from Hoekstra *et al.* (2005).

Tissue	Statistic	% Lipid	HCB	Σ CHL	Σ DDT	Σ HCH	Σ PCB
Blubber (n=5)	Mean \pm SD	83.7 \pm 11.0	100 \pm 24	255 \pm 122	377 \pm 343	297 \pm 126	354 \pm 211
	Median	83.0	112	239	258	273	313
Maktak (n=5)	Mean \pm SD	56.1 \pm 5.0	30.5 \pm 5.24	95.3 \pm 27.1	57.3 \pm 11.6	65.7 \pm 16.9	105 \pm 28.2
	Median	59.5	28.1	90.6	54.9	59.6	93.1
Kidney (n=5)	Mean \pm SD	8.00 \pm 5.76	5.50 \pm 3.06	7.82 \pm 4.72	6.37 \pm 4.20	7.20 \pm 5.37	12.0 \pm 8.91
	Median	7.11	5.70	7.90	5.98	6.21	10.6
Liver (n=5)	Mean \pm SD	6.58 \pm 1.70	3.17 \pm 1.77	5.48 \pm 2.22	3.72 \pm 1.54	9.45 \pm 2.22	9.10 \pm 4.13
	Median	6.89	3.66	5.70	3.69	9.14	9.04
Muscle (n=5)	Mean \pm SD	2.39 \pm 1.85	1.60 \pm 0.96	2.32 \pm 1.17	1.71 \pm 1.01	2.74 \pm 1.68	1.87 \pm 0.90
	Median	1.96	1.16	1.63	2.15	1.86	1.30
Heart (n=5)	Mean \pm SD	1.65 \pm 0.25	1.31 \pm 0.51	1.53 \pm 0.92	0.25 \pm 0.18	3.50 \pm 0.99	1.95 \pm 0.85
	Median	1.52	1.14	1.02	0.26	3.14	1.66
Diaphragm (n=5)	Mean \pm SD	2.39 \pm 2.19	1.96 \pm 1.26	2.69 \pm 2.06	1.59 \pm 1.04	4.47 \pm 2.51	3.43 \pm 1.94
	Median	0.87	1.33	1.56	0.96	3.06	2.62
Tongue (n=5)	Mean \pm SD	10.2 \pm 2.3	8.58 \pm 1.54	18.4 \pm 3.76	15.8 \pm 4.67	22.0 \pm 3.76	15.7 \pm 1.57
	Median	10.7	7.93	19.7	17.2	23.3	15.8

Σ CHL=sum of chlordane technical products *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; Σ DDT = sum of *o,p'*- and *p,p'*-substituted DDT, DDE, and DDD; Σ HCH = sum of α -, β - and γ -HCH isomers; Σ PCB = sum of PCB congeners CB-4/10, 5/8, 6, 7/9, 12/13, 15/17, 16, 18, 19, 21/33/53, 22, 24/27, 25, 26, 28, 29/54, 31, 32, 40, 41/71, 42, 43, 44, 45, 46, 47/48, 49, 50, 51, 52, 55, 56/60, 59, 63, 64, 66/95, 70, 74, 76/98, 82, 83, 84/92, 81, 85, 87, 91, 97, 99, 100, 101, 105, 107/147, 110, 114, 118, 119, 128, 129/178, 130/176, 132, 133/149, 135, 136, 137, 138, 141/179, 143, 144, 145, 151, 153, 156, 158, 163, 167, 170/190, 172, 173/202, 174, 171, 175, 177, 182, 183, 185, 187, 197, 180, 189, 193, 191, 194, 195, 196/203, 198, 199, 201, 205, 206, 207, 208 and CP-209 (IUPAC designation).

Σ PCB₁₀ = sum of PCB congeners CB-28, 31, 52, 101, 105, 118, 138, 153, 156 and CB-180 (IUPAC designation)

food items indicated a range of lipid content from 0.7% (pork chop, Cornish game hen) to 58.8% (reindeer steak marrow; Table III). As expected, the marine mammal tissues had a much larger range of values for lipid content (1.65 to 84.8%) (Tables I and II). As would be expected, the OCs were generally higher in items with higher lipid content. The reindeer steak marrow (58.8% lipid) contained the greatest concentration of hexachlorobenzene (HCB), sum concentrations of hexachlorocyclohexane (Σ HCH), and Σ PCB and Σ PCB₁₀ of the store-bought terrestrial foods. The sum chlordane (Σ CHL) concentration is greatest in chicken egg yolk (23.8% lipid), while Σ DDT is highest in beef tongue (12.1% lipid) for the store-bought terrestrial foods.

The lipid content of store-bought aquatic food ranged from 0.5 to 19.9%, much less than for the marine mammals. The highest lipid content item from the aquatic system (sardines) contained the highest concentration of PCBs (Σ PCB and Σ PCB₁₀) relative to all other store-bought aquatic foods. Canned salmon (4% lipid) contained the highest concentrations of HCB, Σ DDT and Σ HCH; and smoked salmon contained the second highest levels in HCB, Σ DDT and Σ HCH, but the highest concentrations of Σ CHL, of all store-bought aquatic foods (Table III). Overall, the two processed salmon products were found to have the highest levels of HCB, Σ DDT and Σ HCHs. Compared to other regions and species these concentrations are relatively low. The effects of processing

Table II. Mean (\pm 1 SD) and median of lipid content (%) and sum (Σ) concentrations of persistent organochlorine contaminant classes and HCB (ng g⁻¹, wet weight) in various marine mammals and fish from Alaska (1997 – 1999), adapted from Hoekstra et al. (2005).

Species	Tissue	Statistic	% Lipid	HCB	Σ CHL	Σ DDT	Σ HCH	Σ PCB
Beluga whale (<i>Delphinapterus leucas</i>)	Blubber	Mean \pm SD	84.8 \pm 8.15	239 \pm 91.4	816 \pm 84.0	1979 \pm 954	212 \pm 161	2730 \pm 1024
	n=20	Median	84.0	233	792	2120	175	2680
	Maktaaq	Mean \pm SD	44.2 \pm 9.60	76.3 \pm 21.5	520 \pm 226	324 \pm 95.3	66.7 \pm 23.1	962 \pm 264
Ringed seal (<i>Phoca hispida</i>)	n=5	Median	48.3	77.9	453	305	64.9	898
	Blubber	Mean \pm SD	82.2 \pm 13.6	17.3 \pm 11.7	488 \pm 400	269 \pm 198	203 \pm 127	691 \pm 526
Bearded seal (<i>Erignathus barbatus</i>)	n=20	Median	86.1	12.3	400	230	141	495
	Blubber	Mean \pm SD	79.5 \pm 3.96	6.65 \pm 0.55	188 \pm 19.4	156 \pm 21.2	89.3 \pm 36.4	293 \pm 30.9
Pink salmon (<i>Oncorhynchus gorbuscha</i>)	n=7	Median	84.4	7.52	217	138	64.8	270
	Fillet	Mean \pm SD	6.34 \pm 0.26	1.45 \pm 0.52	1.32 \pm 0.27	1.83 \pm 0.27	1.36 \pm 0.58	2.63 \pm 3.50
Arctic char (<i>Salvelinus alpinus</i>)	n=7	Median	6.46	2.10	1.18	1.63	1.58	2.89
	Fillet	Mean \pm SD	4.54 \pm 1.79	1.44 \pm 0.53	1.35 \pm 1.10	1.62 \pm 0.38	1.81 \pm 0.88	2.34 \pm 0.95
Broad whitefish (<i>Coregonus nasus</i>)	n=5	Median	3.64	1.23	1.29	1.48	1.65	2.54
	Fillet	Mean \pm SD	4.19 \pm 1.04	0.47 \pm 0.18	0.49 \pm 0.46	0.14 \pm 0.28	0.22 \pm 0.09	2.89 \pm 2.84
Arctic grayling (<i>Thymallus arcticus</i>)	n=19	Median	3.98	0.42	0.39	0.06	0.22	1.64
	Fillet	Mean \pm SD	4.45 \pm 2.70	4.83 \pm 2.59	1.28 \pm 0.90	1.32 \pm 0.72	0.26 \pm 0.29	2.13 \pm 1.71
Burbot (<i>Lota lota</i>)	n=2	Median	–	–	–	–	–	–
	Liver	Mean \pm SD	40.6 \pm 8.93	9.74 \pm 2.70	25.2 \pm 14.7	16.2 \pm 5.89	4.41 \pm 1.58	47.3 \pm 17.7
	n=15	Median	38.9	9.96	22.7	15.8	4.55	47.2

Descriptions for Σ CHL, Σ DDT, Σ HCH, Σ PCB are the same as in Table I.

of the salmon tissue are difficult to determine, but could be a significant factor (i.e., desiccation and changes in wet weight concentrations, contamination from processing, etc.).

The range of Σ PCBs concentrations for store-bought foods was 0.4 to 9.40 ng/g *w.w.* (Tables III and IV). It should be noted that 9 of 17 mean concentrations of OCs in wildlife tissues, sampled in Hoekstra *et al.* (1), also fell into this range (Table IV). The ranges of concentrations for HCB (< 0.01 to 22.5 ng/g *w.w.*) for store-bought foods (Tables III and IV) indicate that HCB concentrations for 11 of 16 wildlife tissues sampled (Tables I and II) occur within this range. Other comparisons show similar patterns, suggesting that store-bought alternatives may not provide a “less contami-

nated” alternative to many traditional country foods.

Table IV lists the various matrices assessed in rank order for lipid percentage and OC concentration for the 31 matrices (14 store-bought and 17 subsistence use items) studied. Not surprisingly, the first four items are blubber, maktak and maktaaq of the whales and seals. However, it was surprising that reindeer marrow (store-bought) was fifth highest in lipid content and HCB (Table IV). Items ranked 6 through 10 included expected items, such as blubber and maktaaq, except for the sardines, which ranked tenth for sum PCBs (9.40 ng g⁻¹ *w.w.*). Items ranked 11th through 15th included organs and muscles of various species, with chicken egg yolk Σ CHL and Σ PCB (2.74 and 7.77 ng g⁻¹, *w.w.*),

Table III. Sum (Σ) concentrations (ng/g; wet weight) of persistent organochlorine (OC) contaminant classes and HCB (ng g⁻¹, wet weight) in selected store-bought foods from Barrow, Alaska.

Food Item	No. of Pools	% Lipid	HCB	Σ CHL	Σ DDT	Σ HCH	Σ PCB	Σ PCB ₁₀
Terrestrial Biota								
Boneless Pork								
Loin Chop	1	0.7	<0.01	0.01	0.03	<0.01	2.48	0.28
Beef Shank	1	2.6	0.10	0.22	0.98	<0.01	0.96	0.21
Beef Tongue	1	12.1	0.61	0.46	2.09	0.07	2.14	0.81
Reindeer Steak	1	2.5	0.93	0.03	0.03	0.26	2.03	0.25
Reindeer								
Steak Marrow	1	58.8	22.5	0.32	0.15	1.22	7.85	2.86
Cornish								
Game Hen	2	0.7, 0.7	<0.01, <0.01	0.08, 0.01	0.12, 0.01	0.03, 0.06	2.77, 3.81	0.51, 0.28
Chicken Egg Yolk	1	23.8	0.01	2.74	0.15	<0.01	7.77	0.21
Honeycombed								
Tripe	1	1.9	0.02	0.04	0.43	0.02	1.15	0.32
Aquatic Biota								
Milkfish	1	5.6	0.03	0.07	0.40	0.19	2.64	0.72
Smoke Salmon								
Strips	1	2.1	0.35	1.01	0.92	0.53	2.72	0.66
Sardines	1	19.9	0.02	0.27	0.86	0.03	9.40	2.22
Canned Salmon	1	4.0	0.37	0.56	1.19	0.69	4.77	0.96
Imitation								
Crab Flakes	1	0.8	0.01	0.04	0.12	0.02	0.40	0.06
Lobster Tail	1	0.5	<0.01	0.03	0.01	<0.01	0.83	0.09

Descriptions for Σ CHL, Σ DDT, Σ HCH, Σ PCB are the same as in Table I.

beef tongue Σ DDT (2.09 ng g⁻¹, w.w.); and reindeer marrow (7.85 ng g⁻¹, w.w.), canned salmon (4.77 ng g⁻¹, w.w.), and Cornish game hen (2.77 and 3.81 ng g⁻¹, w.w.) for Σ PCBs mixed in with various subsistence use items.

There is no clear demarcation in OC contamination of store-bought foods versus subsistence foods. However, due to blubber and blubber-based products, a trend for higher

concentrations of OCs is evident in marine mammal-based foods. Continuing this theme, food items ranked from 15 through 28 are a mixture of store-bought foods and subsistence items. Store-bought foods dominated the final 28 through 31 items for lowest in lipid content and lowest OC concentrations. Based on these data, it is not obvious that switching to similar store-bought alternatives would have a

Table IV. Rank order of store-bought foods OC concentrations sampled in Barrow Alaska (bold italics) compared to mean OCs concentrations of select tissues from subsistence use species of northern Alaska, including lipid content (%) and sum (Σ) concentrations of persistent organochlorine contaminant classes and HCB (ng g⁻¹, wet weight).

% Lipid	HCB	Σ CHL	Σ DDT	Σ HCH	Σ PCB
1 Beluga* Blubber 84.8	Beluga Blubber 239	Beluga Blubber 816	Beluga Blubber 1979	Bowhead blubber 297.0	Beluga blubber 2730
2 Bowhead blubber 83.7	Bowhead blubber 100.0	Beluga maktaaq 520	Bowhead blubber 377	Beluga Blubber 212	Beluga maktaaq 962
3 Ringed blubber 82.2	Beluga maktaaq 76.3	Ringed blubber 488	Beluga maktaaq 324	Ringed blubber 203	Ringed blubber 691
4 Bearded blubber 79.5	Bowhead maktak 30.5	Bowhead blubber 255.0	Ringed blubber 269	Bearded blubber 89.3	Bowhead blubber 354.0
5 RSMARROW** 58.8	RSMARROW 22.5	Bearded blubber 188	Bearded blubber 156	Beluga maktaaq 66.7	Bearded blubber 293
6 Bowhead maktak 56.1	Ringed blubber 17.3	Bowhead maktak 95.3	Bowhead maktak 57.3	Bowhead maktak 65.7	Bowhead maktak 105
7 Beluga maktaaq 44.2	Burbot liver 9.74	Burbot liver 25.2	Burbot liver 16.2	Bowhead tongue 22.0	Burbot liver 47.3
8 Burbot liver 40.6	Bowhead tongue 8.58	Bowhead tongue 18.4	Bowhead tongue 15.8	Bowhead liver 9.45	Bowhead tongue 15.7
9 YOKE 23.8	Bearded blubber 6.65	Bowhead kidney 7.82	Bowhead kidney 6.37	Bowhead kidney 7.20	Bowhead kidney 12.0
10 SARDINES 19.9	Bowhead kidney 5.50	Bowhead liver 5.48	Bowhead liver 3.72	Bowhead diaphragm 4.47	SARDINES 9.40
11 BEEF TONGUE 12.1	Ta fillet 4.83	YOKE 2.74	BEEF TONGUE 2.09	Burbot liver 4.41	Bowhead liver 9.10
12 Bowhead tongue 10.2	Bowhead liver 3.17	Bowhead diaphragm 2.69	Pink fillet 1.83	Bowhead muscle 2.74	RSMARROW 7.85
13 Bowhead kidney 8.00	Bowhead diaphragm 1.96	Bowhead muscle 2.32	Bowhead muscle 1.71	Bowhead heart 3.50	YOKE 7.77
14 Bowhead liver 6.58	Bowhead muscle 1.60	Bowhead heart 1.53	Char fillet 1.62	Char fillet 1.81	CAN SALMON 4.77
15 Pink fillet 6.34	Pink fillet 1.45	Char fillet 1.35	Bowhead diaphragm 1.59	Pink fillet 1.36	HEN 2.77, 3.81
16 MILKFISH 5.6	Char fillet 1.44	Pink fillet 1.32	Grayling fillet 1.32	RSMARROW 1.22	Bowhead diaphragm 3.43
17 Char fillet 4.54	Bowhead heart 1.31	Grayling fillet 1.28	MILKFISH 1.19	CAN SALMON 0.69	Whitefish 2.89
18 Grayling fillet 4.45	REINDEER 0.93	SMOKE SALMON 1.01	SHANK 0.98	SMOKE SALMON 0.53	SMOKE SALMON 2.72
19 Whitefish fillet 4.19	Whitefish fillet 0.47	CAN SALMON 0.56	SMOKE SALMON 0.92	REINDEER 0.26	MILKFISH 2.64
20 CAN SALMON 4.0	BEEF TONGUE 0.61	Whitefish fillet 0.49	SARDINES 0.86	Grayling fillet 0.26	Pink 2.63
21 Bowhead muscle 2.39	CAN SALMON 0.37	BEEF TONGUE 0.46	TRIPE 0.43	Whitefish fillet 0.22	PORK 2.48
22 SHANK 2.6	SMOKE SALMON 0.35	RSMARROW 0.32	MILKFISH 0.40	MILKFISH 0.19	Char 2.34
23 REINDEER 2.5	SHANK 0.10	SARDINES 0.27	Bowhead heart 0.25	BEEF TONGUE 0.07	BEEF TONGUE 2.14
24 Bowhead diaphragm 2.39	MILKFISH 0.03	SHANK 0.22	YOKE 0.15	HEN 0.03, 0.06	Grayling 2.13
25 SMOKE SALMON 2.1	SARDINES 0.02	MILKFISH 0.07	RSMARROW 0.15	SARDINES 0.03	REINDEER 2.03
26 TRIPE 1.9	TRIPE 0.02	HEN 0.08, 0.01	Whitefish fillet 0.14	IMITATION CRAB 0.02	Bowhead heart 1.95
27 Bowhead heart 1.65	YOKE 0.01	TRIPE 0.04	IMITATION CRAB 0.12	TRIPE 0.02	Bowhead muscle 1.87
28 IMITATION CRAB 0.8	IMITATION CRAB 0.01	IMITATION CRAB 0.04	HEN 0.12, 0.01	HEN <0.01	TRIPE 1.15
29 PORK 0.7	HEN <0.01	LOBSTER 0.03	REINDEER 0.03	PORK <0.01	SHANK 0.96
30 HEN 0.7, 0.7	PORK <0.01	REINDEER 0.03	PORK 0.03	SHANK <0.01	LOBSTER 0.83
31 LOBSTER 0.5	LOBSTER <0.01	PORK 0.01	LOBSTER 0.01	LOBSTER <0.01	IMITATION CRAB 0.40

* Subsistence wildlife species: Beluga = Beluga whale (*Delphinapterus leucas*), Bowhead = Bowhead whale (*Balaena mysticetus*), Ringed = Ringed seal (*Phoca hispida*), Bearded = Bearded seal (*Erignathus barbatus*), Pink = Pink salmon (*Oncorhynchus gorbuscha*), Char = Arctic char (*Salvelinus alpinus*), Whitefish = Broad whitefish (*Coregonus nasus*), Grayling = Arctic grayling (*Thymallus arcticus*), Burbot (*Lota lota*).

** Store-bought food codes (words in all-caps in Table): Boneless Pork Loin Chop = PORK, Beef Shank = SHANK, Beef Tongue = BT, Reindeer Steak = REINDEER, Reindeer Steak Marrow = RSMARROW, Cornish Game Hen = HEN, Chicken Egg Yolk = YOKE, Honeycombed Tripe = TRIPE, Milkfish = MILKFISH, Smoke Salmon Strips = SMOKE SALMON, Sardines = SARDINES, Canned Salmon = CAN SALMON, Imitation Crab Flakes = IMITATION CRAB, Lobster Tail = LOBSTER.

dramatic impact on reducing OC exposure, if one considers the same tissue types and basic lipid contents. Clearly, blubber and foods composed of blubber are unique, as there are no suitable, commercially available, “replacement” items with relatively high lipid contents that are predisposed to accumulating the OCs.

Careful interpretation of the food items reported in this study is required. For example, the effect of food preparation on OC concentrations was not studied. Previous investigations have noted a reduction of Σ PCB and other OC concentrations in fish due to cooking (19, 20). While the effect of rendering, fermentation, and other food preparation practices used by Native Alaskans (21), on OC concentrations is unknown, the estimated daily intake values calculated from raw tissues in (1) likely overestimate OC exposure from these subsistence dietary items when cooked. Some of the store-bought items are ready to eat (smoked salmon, sardines), while others require processing and would need to be assessed after preparation for consumption to truly reflect the OC exposure. As indicated in this study, the store-bought foods are not always less contaminated than similar food items (i.e., fish products). Dougherty *et al.* (7) showed that average food ingestion exposures for the whole US population exceeded benchmark concentrations for chlordane, DDT, dieldrin, dioxins and polychlorinated biphenyls, when non-detects were assumed to be equal to zero. For each of these pollutants, exposure through fish consumption accounts for a large percentage of food exposures. Interestingly, Focant *et*

al. (8) reported that background concentrations of PCDDs, PCDFs, nonortho-PCBs, and mono-ortho-PCBs in fast food samples collected in different countries appeared to be very low.

The socio-cultural and nutritional benefits of a subsistence diet to the residents of northern Alaska must be considered when discussing switches to store-bought foods. Preliminary information on the nutritional quality of bowhead whale tissues and other wildlife suggest that these subsistence dietary items are excellent sources of nutritionally essential minerals, fatty acids and fat-soluble vitamins, such as vitamins A, D and E (e.g. 10, 22-26). The increasing prevalence of “Western”, or modern, diets over traditional foods is thought to have negative health consequences, in part due to the increased consumption of total fat, saturated fats and sucrose above recommended levels (25, 10). Upon considering the risks associated with exposure to contaminants relative to the known benefits of traditional foods, as well as the risks associated with eating poor replacement foods (e.g. 27, 12), the consumption of bowhead whale tissues and other marine biota by Inuit of northern Alaska should be maintained and encouraged, as suggested for traditional foods in general by Verbrugge and Middaugh (28) and Arnold and Middaugh (29). By using the balanced approach nutrients and contaminants, this information will allow individuals to decide for themselves which dietary habits best suits their lifestyle. In order to do this, there must be a general sense of how traditional foods compare to the store-bought alternatives with respect to contamination by OCs.

Conclusions

In conclusion, persistent OCs were quantified in numerous tissues of Alaska marine mammals and fish (marine and freshwater) and compared to store-bought food “alternatives”. As expected, lipid content significantly influenced the concentrations of Σ OCs in store-bought foods and subsistence use foods. Thus, blubber and blubber-based products have the highest concentrations of OCs for both wildlife and store-bought foods. There was no clear indication that store-bought foods were significantly less contaminated than comparable traditional (wildlife) foods, except for blubber and blubber-containing products (as there was no blubber analog in the store). We conclude that switching from non-blubber tissues to local store-bought alternatives will not eliminate OC exposure and may only provide a slight reduction, if any, that may be at the expense of much less nutrient-rich products and the elimination of important, healthy, socio-cultural practices. We did not assess a blubber analog from the store and, thus, cannot provide information comparing a “substitute”, but we recognize that blubber is the most contaminated tissue. Due to this contamination, we would encourage the assessment of the impact of food processing on resultant nutrients and contaminants in blubber (raw, boiled, fermented, pickled, etc.). The data presented here do not consider essential and non-essential elements, and emerging contaminants (e.g. brominated flame retardants). However, we recognize them as important components and, as such, will be reported upon elsewhere.

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REFERENCES

1. Hoekstra PF, O'Hara TM, Backus SM, Hanns C, Muir DCG. Concentrations of persistent organochlorine contaminants in bowhead whale tissues and other biota from northern Alaska: Implications to human exposure from a subsistence diet. *Environ Res* 2005; **98** (3): 329-340.
2. Jensen S. The PCB story. *Ambio* 1972; **1**: 123-131.
3. de Wit CA, Fisk AT, Hobbs KE, et al. Persistent organochlorine pollutants. In: de Wit CA, Fisk AT, Hobbs KE, Muir DCG (Eds.) *Arctic Monitoring and Assessment Program (AMAP) II. 2004. Arctic Monitoring and Assessment Program*, Oslo, Norway.
4. Sandau CD, Ayotte P, Dewailly E, Duffe J, Norstrom RJ. Analysis of hydroxylated metabolites of PCBs (OH-PCBs) and other chlorinated phenolic compounds in whole blood from Canadian Inuit. *Environ Health Perspect* 2000; **108**: 611-616.
5. Huwe JK. Dioxins in food: a modern agricultural perspective. *J Agric Food Chem* 2002; **50**: 1739-1750.
6. Himberg KK. Coplanar polychlorinated biphenyls in some Finnish food commodities. *Chemosphere* 1993; **27**: 1235-1243.
7. Dougherty CP, Henricks Holtz S, Reinert JC, Panyacosit L, Axelrad DA, Woodruff TJ. Dietary Exposures to Food Contaminants across the United States. *Environ Res* 2000; **84**(2): 170-185.

8. Focant J-F, Pirard C, De Pauw E. Levels of PCDDs, PCDFs and PCBs in Belgian and international fast
9. Fuller AS, George JC. Evaluation of subsistence harvest data from the North Slope Borough 1993 census for eight North Slope villages: for the calendar year 1992. Department of Wildlife Management, Barrow, AK, USA. 1997.
10. Van Oostdam J, Donaldson S, Feeley M, Tremblay N. Human health. In: Canadian Arctic Contaminants Assessment Report II. 2003. Ministry of Public Works and Government Services Canada, Ottawa, ON, Canada, pp -127.
11. Hing MC. A database for environmental contaminants in traditional foods in northern and arctic Canada: developments and applications. *Food Add Contam* 1998; 15: 127-134.
12. Kuhnlein HV, Chan HM. Environment and contaminants in traditional food systems of northern indigenous peoples. *Annu Rev Nutr* 2000; 20: 595-626.
13. Rubin CH, Lanier A, Socha M, Brock JW, Kieszak S, Zahm S. Exposure to persistent organochlorines among Alaska Native women. *Int J Circumpolar Health* 2001; 60: 157-169.
14. Hoekstra PF, O'Hara TM, Pallant SJ, Muir DCG. Bioaccumulation of organochlorine contaminants in bowhead whales (*Balaena mysticetus*) from Barrow, Alaska. *Arch Environ Contam Toxicol* 2002; 42: 497-507.
15. Hoekstra PF, O'Hara TM, Fisk AT, Borgå K, Solomon KR, Muir DCG. Trophic transfer of persistent organochlorine contaminants (OCs) within an arctic marine food web from the southern Beaufort-Chukchi Seas. *Environ Pollut* 2003; 124: 509-522.
16. Krahn MM, Burrows DG, Stein JE, et al. White whales (*Delphinapterus leucas*) from three Alaskan stocks: concentrations and patterns of persistent organochlorine contaminants in blubber. *J Cetacean Res Manage* 2000; 1: 239-249.
17. Kucklick JR, Struntz WDJ, Becker PR, York GW, O'Hara TM, Bohonowich JE. Persistent organochlorine pollutants in ringed seals and polar bears collected from northern Alaska. *Sci Total Environ* 2002; 287: 45-59.
18. O'Hara TM, Krahn MM, Boyd D, Becker PR, Philo LM. Organochlorine contaminant levels in Eskimo harvested bowhead whales of arctic Alaska. *J Wildlife Dis* 1999; 35: 741-752.
19. Kamrin MA, Fischer LJ. Current status of sport fish consumption advisories for PCBs in the Great Lakes. *Regul Toxicol Pharmacol* 1999; 29: 175-181.
20. Zabik ME, Zabik MJ, Booren AIM, et al. Pesticides and total polychlorinated biphenyls in chinook salmon and carp harvested from the Great Lakes: effects of skin-on and skin-off processing and selected cooking methods. *J Agricult Food Chem* 1995; 43: 993-1001.
21. Kassam K-AS. Passing on the knowledge: mapping human ecology in Wainwright, Alaska. 2001. Arctic Institute of North America, University of Calgary, Calgary, AB, Canada.
22. Blanchet C, Dewailly É, Ayotte P, Bruneau S, Receveur O, Holub BJ. Contribution of selected traditional and market foods to the diet of Nunavik Inuit women. *Can J Diet Prac Res* 2000; 61: 50-59.
23. Kenny DE, O'Hara TM, Chen TC, Lu Z, Tian X, Holick MF. Vitamin D content in Alaskan Arctic zooplankton, fishes, and marine mammals. *Zoo Biology* 2004; 23(1): 33-43.
24. Nobmann ED, Byers T, Lanier AP, Hankin JH, Jackson MY. The diet of Alaska Native adults: 1987 – 1988. *Am J Clin Nutr* 1992; 55(5): 1024-1032.
25. Nobmann ED, Lanier AP. Dietary intake among Alaskan native women resident of Anchorage, Alaska. *Int J Circumpolar Health* 201; 60: 123-137.
26. Woshner VM, O'Hara TM, Bratton GR, Suydam RS, Beasley VR. Concentrations and interactions of selected essential and non-essential elements in bowhead and beluga whales of arctic Alaska. *J Wildlife Dis* 2001; 37: 693-710.
27. Egeland GM, Feyk LA, Middaugh JP. The use of traditional foods in a healthy diet in Alaska: risks in perspective. *State of Alaska Epidemiology Bulletin* 1998; Vol. 2 No. 1 (January).
28. Verbrugge LA, Middaugh JP. Use of traditional foods in a healthy diet in Alaska: risks in perspective. Second Edition: Volume 1. Polychlorinated biphenyls (PCBs) and related compounds. *State of Alaska Epidemiology Bulletin* 8:1-62.
29. Arnold SM, Middaugh JP. Use of traditional foods in a healthy diet in Alaska: risks in perspective. Second Edition: Volume 2. Mercury. *State of Alaska Epidemiology Bulletin* 8:1-48.

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