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Concentrations of persistent organochlorine contaminants in bowhead whale tissues and other biota from northern Alaska: Implications for human exposure from a subsistence diet ☆

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Abstract

Bowhead whale (*Balaena mysticetus*; $n = 5$) blubber, liver, muscle, kidney, heart, diaphragm, tongue, and uncooked maktak (bowhead whale epidermis and blubber) were collected during subsistence hunts at Barrow, AK, USA (1997–1999) to measure concentrations of persistent organochlorine contaminants (OCs). The exposure of humans to OCs via bowhead whales and other biota [fish, ringed (*Phoca hispida*) and bearded seals (*Erignathus barbatus*), and beluga whale (*Delphinapterus leucas*)] as part of a subsistence diet was evaluated. Concentrations of OCs in bowhead whale tissues were correlated with lipid content ($P<0.001$) and were less than levels in other marine mammals reported herein, reflecting the lower trophic status of this cetacean. The relative proportions of hexachlorobenzene (HCB) and sum (Σ) concentrations of chlordane components (ΣCHL), DDT-related compounds (ΣDDT), and polychlorinated biphenyls (ΣPCB) were not statistically different among the tissues analyzed ($P<0.05$). However, relatively higher proportions of hexachlorocyclohexane isomers (ΣHCH), particularly β-HCH, were observed in bowhead whale heart and diaphragm ($P<0.03$). Based on Canadian and World Health Organization daily intake guidelines, "safe" human consumption rates of bowhead whale tissue and other marine biota were calculated. The most restrictive limits (mean value) for daily consumption for bowhead and beluga whale were 302 and 78 g for maktak and maktaaq (beluga whale epidermis and blubber), respectively. The tolerable daily intake limits of dioxin-like compounds from the consumption of bowhead whale blubber and liver were calculated to be 199 g (approximately 600 g for maktak) and 2222 g, respectively. A detailed profile of traditional/country foods consumed by subsistence communities of northern Alaska is required to address chronic exposure in more detail. Overall, bowhead whale tissues and other biota from northern Alaska are safe to consume at, or below, the levels calculated.

Keywords: Arctic; Cetacean; Marine mammal; Subsistence; Traditional food; Tissue distribution

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1. Introduction

The presence of persistent organochlorine contaminants (OCs), a structurally diverse group of agricultural and industrial compounds (or byproducts), in the Arctic is well known. These chemicals were first detected in the region in the late 1960s (Jensen, 1972) and have been found in virtually every compartment in the Arctic due to their environmental recalcitrance and long-range transport from industrial and agricultural activities via atmospheric and oceanic currents (de Wit et al., 2004). The persistence, toxicity, and bioaccumulation potential of OCs is particularly significant in the arctic marine environment, where many species feed on prey with greater lipid content compared to phylogenetically similar species inhabiting lower latitude ecosystems, resulting in the accumulation of OCs to relatively high concentrations in top predatory marine species such as polar bears (*Ursus maritimus*) and ringed seals (*Phoca hispida*) (de Wit et al., 2004). The accumulation of OCs in arctic biota is of concern 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 (Sandau et al., 2000).

The subsistence diet of indigenous peoples of the Arctic varies by region due to prey availability and local hunting practices. In northern Alaska, Inuit have hunted and consumed arctic wildlife and marine mammals, especially the bowhead whale (*Balaena mysticetus*), for many generations (Stoker and Krupnik, 1993). While the bowhead whale stock found in the Bering–Chukchi–Beaufort Seas region was greatly reduced by commercial whaling (Shelden and Rugh, 1996), this population is increasing at a rate of $\approx 3\%$ per year (Raftery and Zeh, 1998) and remains a sociocultural and nutritionally important species to coastal Alaskan subsistence communities (Kassam, 2001).

Several studies have recognized the sociocultural and nutritional benefit of traditional foods to arctic peoples (e.g., Van Oostdam et al., 2003). However, the accumulation of OCs in Native populations from subsistence arctic communities has raised some questions concerning the suitability of terrestrial and marine wildlife from this region for human consumption (Kuhnlein and Chan, 2000; Rubin et al., 2001; Sandau et al., 2000; Van Oostdam et al., 2003). Concentrations of OCs in bowhead whale blubber and liver, along with blubber from other marine mammals from northern Alaska, have been reported (Hoekstra et al., 2002b, 2003b; Krahn et al., 2000; Kucklick et al., 2002; O'Hara et al., 1999). However, the exposure to OCs from consumption of other bowhead whale tissues as part of a subsistence diet has not been investigated.

In this paper, we discuss the potential human exposure to OCs and implications for human consumption of bowhead whale tissues and other biota as part of a subsistence diet. We investigated the presence of several persistent OC groups in bowhead whale tissues that had not been examined previously (muscle, kidney, heart, diaphragm, and tongue) and uncooked bowhead whale maktak, a traditional food item comprising bowhead whale epidermis and blubber. The concentrations and relative abundance of OCs in these bowhead whale tissues are compared to foods in general in this work and will be compared for select foods purchased locally (Barrow, AK, USA) in a separate report.

2. Methodology

2.1. Sample collection

Samples were collected from 1997 to 1999 at Barrow (71°17′N, 156°45′W), Nuiqsut, AK (70°21′N, 151°02′W), and Pt. Lay, AK (69°43′N, 163°00′W) through the North Slope Borough Department of Wildlife Management (Fig. 1) as part of a larger study to investigate the trophic ecology and transfer of persistent OCs in the arctic marine environment of northern Alaska (Hoekstra et al., 2003b). Blubber samples from ringed seals (*P. hispida;* $n = 20$ seals), bearded seals (*Erignathus barbatus;* $n = 20$ seals), and beluga whale (*Delphinapterus leucas*; $n = 20$ whales; $n = 5$ whales with epidermis) were collected from Inuit subsistence harvests. Bowhead whale tissues ($n = 5$ whales) were obtained from the Inuit's subsistence hunt with the permission of the Alaska Eskimo Whaling Commission and Barrow Whaling Captains Association (Barrow). Several species of fish, including arctic char (*Salvelinus alpinus;* $n = 5$ fish), pink salmon (*Oncorhynchus gorbuscha;* $n = 7$ fish), broad whitefish (*Coregonus nasus;* $n = 19$ fish), arctic grayling (*Thymallus arcticus;* $n = 2$ fish), and burbot (*Lota lota;* $n = 15$), were provided by Native (Inuit) subsistence fishers.

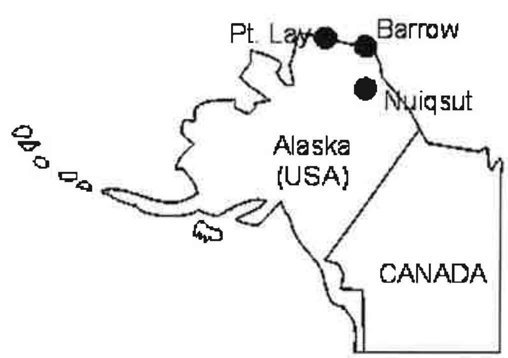

Fig. 1. Sampling sites (●) of biota and fish at Pt. Lay (beluga only), Nuiqsut (arctic grayling, burbot, and broad whitefish only), and Barrow, AK, USA (1997–1999).

Field sampling techniques of mammalian tissues have been previously described (Becker et al., 1991; Hoekstra et al., 2002b). Representative samples of uncooked bowhead and beluga whale *maktak* and *maktaaq*, respectively, were prepared using an epidermis-to-blubber size ratio of 1:2 (i.e., the typically consumed dimension). Samples were initially collected in the field (from each animal) in Whirl-Pak bags and then were transported to the National Water Research Institute (Environment Canada, Burlington, Ont., Canada) under US Export and Canadian Import permits in accordance with the Convention on International Trade in Endangered Species (US694250 and CA-CW-IM-0053-00, respectively) and the US Marine Mammal Protection Act (Permit No. 782-1399). External surfaces of all samples were trimmed and removed (to minimize possible contamination from sample bags), homogenized, and stored at −20 °C in precleaned glass containers.

2.2. Extraction and analysis of non-planar polychlorinated biphenyls and OC pesticides

Biota samples were extracted and analyzed using previously described techniques with only minor modification (Hoekstra et al., 2002b). In brief, fish samples and full-thickness blubber cores from marine mammals were homogenized prior to OC extraction. Tissue subsamples were mixed with sodium sulfate (Na_2SO_4) and spiked with a mixture of internal standards [two polychlorinated biphenyl (PCB) congeners, CB-30 and CB-204, δ-HCH (hexachlorocyclohexane), 1,3,5-tribromobenzene, and 1,2,4,5-tetrabromobenzene] to monitor analyte recovery. Fish and bowhead whale tissues (except blubber and maktak) were weighed (10–15 g) and extracted with dichloromethane (DCM) via Soxhlet, whereas marine mammal blubber and maktak/maktaaq samples (1–3 g) were extracted with DCM using a Polytron homogenizer (Brinkmann, Westbury, NY, USA). Lipids and other bioorganic materials in each sample were removed using gel-permeation chromatography. The lipid percentage of each was determined gravimetrically on a subsample of the extract.

The analytes for each sample were concentrated and separated on 100%-activated silica gel into two fraction, hexane (Fraction 1) and hexane:DCM (1:1 by volume; Fraction 2), transferred to 2,2,4-trimethylpentane (isooctane), and concentrated. Analysis of OCs in both fractions was performed using a Hewlett-Packard 5890 gas chromatograph (Wilmington, DE, USA) with an ^{63}Ni-electron capture detector (Hoekstra et al., 2002c). Sample quantification of OCs was performed using multiple external standards provided by the National Laboratory for Environmental Testing (NLET; Environment Canada, Burlington, Ont., Canada), which were analyzed after every 10 samples. The concentrations of hexachlorobenzene (HCB) and sum (Σ) group concentrations of chlordane-related products (ΣCHL), DDT-compounds (ΣDDT), hexachlorocyclohexane isomers (ΣHCH), and ΣPCB are provided in Tables 1–3.

2.3. Coplanar PCBs and polychlorinated dibenzo-p-dioxin and dibenzofuran s in bowhead whale blubber and liver

A limited number of bowhead whale blubber and liver tissues ($n = 5$; tissues selected from the same specimen) were randomly selected for analysis of coplanar PCBs and polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/F) congeners. Other tissues were omitted from this analysis because coplanar PCB and PCDD/F concentrations in arctic wildlife are generally found at a fraction of ΣPCB levels (de Wit et al., 2004) and were likely to be below method detection limits.

For coplanar PCBs, bowhead whale blubber (1–2 g) and liver (5–10 g) samples were accurately weighed and homogenized with Na_2SO_4. Samples were spiked with ^{13}C-labelled CB-77, CB-126, and CB-169 to monitor extraction efficiency. Samples were extracted overnight in *n*-hexane:DCM (50:50, by volume) using a Soxhlet apparatus and concentrated. Lipid percentage was determined gravimetrically using an aliquot of the extract. The remaining portion was eluted through an acidified silica gel column (22% H_2SO_4 by weight, 6 g) to remove lipids and concentrated.

Coplanar PCBs were separated from non-planar OCs by high-pressure liquid chromatography (HPLC) (Bagheri et al., 1993; Lundgren et al., 2002). In brief, samples were manually injected into an Agilent 1100 HPLC system equipped with a diode-array detector (Agilent Technologies, Wilmington, DE, USA). The HPLC separation of planar from non-planar PCBs was performed on two 15 cm × 4.6 mm (internal diameter, i.d.) Cosmosil PYE-5 columns with a particle size of 5 μm (Nacalai Tesque, Japan). Columns were connected in series and temperature was maintained at 20 °C. Hexane was used for the elution of PCBs at a flow rate of 1 mL min^{-1}. The fraction containing planar PCBs was collected in a quantity of between 11 and 20 mL.

The coplanar PCB fraction was fortified in isooctane and concentrated to 100 μL. Analysis of coplanar PCBs was performed using an Agilent 6890N gas chromatograph equipped with a 5973N mass selective detector using negative chemical ionization (Bagheri et al., 1993). The gas chromatograph capillary fused-silica column used was an Agilent HP-5MS (30 m × 0.25 mm i.d. × 0.25-μm film thickness). Helium was selected as the carrier and methane was used as the chemical reagent gas.

Extraction and analysis of PCDD/Fs in bowhead whale blubber and liver samples by Axys Analytics (Sidney, BC, Canada) used previously

Table 1
Mean (±1 SD), median, and range of lipid content (%) and sum (Σ) concentrations of persistent organochlorine contaminant classes and HCB ($ng\,g^{-1}$, wet wt) in tissue and uncooked maktak from five ($n=5$) bowhead whales (*Balaena mysticetus*) collected in 1997–1999

Tissue	% Lipid	HCB	ΣCHL	ΣDDT	ΣHCH	ΣPCB
Blubber	83.7±11.0	100±24	255±122	377±343	297±126	354±211
	83.0	112	239	258	273	313
	62.3–99.3	73.0–273	94.5–701	118–1925	92.2–763	153–1305
Maktak	56.1±5.0	30.5±5.24	95.3±27.1	57.3±11.6	65.7±16.9	105±28.2
	59.5	28.1	90.6	54.9	59.6	93.1
	49.5–60.0	26.0–39.1	69.4–136	48.1–77.1	50.1–87.4	81.8–153
Kidney	8.00±5.76	5.50±3.06	7.82±4.72	6.37±4.20	7.20±5.37	12.0±8.91
	7.11	5.70	7.90	5.98	6.21	10.6
	2.75–15.0	2.49–8.13	3.10–12.4	2.24–11.4	2.05–14.3	3.97–22.7
Liver	6.58±1.70	3.17±1.77	5.48±2.22	3.72±1.54	9.45±2.22	9.10±4.13
	6.89	3.66	5.70	3.69	9.14	9.04
	3.36–10.4	1.73–9.35	2.30–10.6	1.35–6.72	6.25–13.82	1.66–16.5
Muscle	2.39±1.85	1.60±0.96	2.32±1.17	1.71±1.01	2.74±1.68	1.87±0.90
	1.96	1.16	1.63	2.15	1.86	1.30
	0.80–5.34	0.69–3.02	1.20–3.69	0.59–2.81	1.41–5.48	1.18–3.19
Heart	1.65±0.25	1.31±0.51	1.53±0.92	0.25±0.18	3.50±0.99	1.95±0.85
	1.52	1.14	1.02	0.26	3.14	1.66
	1.45–2.05	0.98–2.21	0.99–3.13	0.03–0.46	2.39–4.88	1.26–3.38
Diaphragm	2.39±2.19	1.96±1.26	2.69±2.06	1.59±1.04	4.47±2.51	3.43±1.94
	0.87	1.33	1.56	0.96	3.06	2.62
	0.66–4.98	0.90–3.82	1.03–5.46	0.74–2.93	2.38–7.26	1.57–6.28
Tongue	10.2±2.3	8.58±1.54	18.4±3.76	15.8±4.67	22.0±3.76	15.7±1.57
	10.7	7.93	19.7	17.2	23.3	15.8
	7.11–12.4	7.60–10.9	12.9–21.4	9.13–19.8	16.5–24.9	14.1–17.3

Values provided for each parameter are arithmetic mean±1 SD, median, and data range. HCB, hexachlorobenzene; ΣCHL, sum concentration of *cis*- and *trans*-chlordane, *cis*- and *trans*-non-achlor, and oxychlordane; ΣDDT = sum of *o,p′*- and *p,p′*-DDT, DDE, and DDD; ΣHCH, sum of α-, β-, and γ-HCH; ΣPCB, sum of 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, 81, 82, 83, 85, 87, 91, 92/84, 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, 151, 153, 156, 158, 163, 167, 170/190, 171, 172, 173/202, 174, 175, 177, 180, 182, 183, 185, 187, 189, 191, 193, 194, 195, 196/203, 197, 198, 199, 201, 205, 206, 207, 208, and CB-209 (IUPAC designations).

described techniques with only minor modification (US EPA, 1994). Bowhead blubber and liver samples were homogenized with Na_2SO_4 and spiked with a mixture of ^{13}C-labelled PCDD and PCDF isomer (as listed in Table 3) surrogate standards to monitor extraction efficiency. Samples were extracted in toluene via Soxhlet under clean-room laboratory conditions (positive pressure, carbon and HEPA filters) and concentrated. Extracts were cleaned up and PCDD/Fs were isolated through a series of chromatographic columns (US EPA, 1994). Analysis was performed using a high-resolution gas chromatograph coupled to a high-resolution mass spectrometer. Compound separation was accomplished using a DB-5 capillary chromatography column (60 m × 0.25 mm i.d. × 0.1-μm film thickness; J&W Scientific, Folsom, CA, USA). A second column, DB-225 (30 m × 0.25 mm i.d. × 0.15-μm film thickness), was used for confirmation of 2,3,7,8-tetraCDF.

2.4. Analytical quality assurance

Recovery of non-planar PCB and OC surrogate standards ranged from 70% to 98% and concentrations were adjusted accordingly. Detection limits ranged from 0.02 to 0.1 $ng\,g^{-1}$ for individual PCB congeners (including non-ortho Cl-ubstituted PCBs) and OC pesticides and 0.2–5.0 $pg\,g^{-1}$ for PCDD/F analysis. Quality assurance protocol included extraction and analysis of laboratory blanks with every batch of 10 samples and the use of two standard reference materials (SRM1588 Organics in Cod Liver Oil and SRM1945 Organics in Whale Blubber Homogenate) from the National Institute of Standards and Technology (Gaithersburg, MD, USA). Results of SRM analysis were within 20% of certified values. The laboratory that conducted coplanar PCB analyses (NLET) had successfully participated in an international interlaboratory comparison program on PCB analysis (QUASIMEME, Aberdeen, UK).

2.5. Statistical analysis and calculations

Statistical analyses were conducted using the SYSTAT statistical package, Version 8.0 (SPSS, Chicago, IL, USA). All statistical tests were two-tailed and maximum probability of a Type-I error (α) was

Table 2
Mean (±1 SD), median, and range of lipid content (%) and sum (Σ) concentrations of persistent organochlorine contaminant classes and HCB (ng g^{-1}, wet wt) in various marine mammals and fish from Alaska (1997–1999)

Species	Tissue	% Lipid	HCB	ΣCHL	ΣDDT	ΣHCH	ΣPCB
Beluga whale	Blubber	84.8±8.15	239±91.4	816±84.0	1979±954	212±161	2730±1024
(*Delphinapterus leucas*)	n=20	84.0	233	792	2120	175	2680
		71.4–100	55.8–487	135–1936	254–4210	86.7–865	620–5248
	Maktaaq	44.2±9.60	76.3±21.5	520±226	324±95.3	66.7±23.1	962±264
	n=5	48.3	77.9	453	305	64.9	898
		28.3–53.3	39.2-106	258-922	194–483	43.3–97.0	619–1390
Ringed seal	Blubber	82.2±13.6	17.3±11.7	488±400	269±198	203±127	691±526
(*Phoca hispida*)	n=20	86.1	12.3	400	230	141	495
		54.7–100	4.95–49.5	68.8–1484	28.6–818	37.2–1068	124–2114
Bearded seal	Blubber	79.5±3.96	6.65±0.55	188±19.4	156±21.2	89.3±36.4	293±30.9
(*Erignathus barbatus*)	n=7	84.4	7.52	217	138	64.8	270
		60.7–90.0	3.92–8.32	123–245	111–267	25.1–300	196–411
Pink salmon	Fillet	6.34±0.26	1.45±0.52	1.32±0.27	1.83±0.27	1.36±0.58	2.63±3.50
(*Oncorhynchus gorbuscha*)	n=7	6.46	2.10	1.18	1.63	1.58	2.89
		5.21–7.10	1.10–3.25	0.54–2.05	0.98–2.11	0.92–2.15	0.57–5.86
Arctic char	Fillet	4.54±1.79	1.44±0.53	1.35±1.10	1.62±0.38	1.81±0.88	2.34±0.95
(*Salvelinus alpinus*)	n=5	3.64	1.23	1.29	1.48	1.65	2.54
		2.36–7.32	0.63–1.81	0.18–2.17	1.18–2.15	1.06–2.25	0.89–3.15
Broad whitefish	Fillet	4.19±1.04	0.47±0.18	0.49±0.46	0.14±0.28	0.22±0.09	2.89±2.84
(*Coregonus nasus*)	n=19	3.98	0.42	0.39	0.06	0.22	1.64
		2.60–6.01	0.25–0.91	0.10–2.21	0.02–1.20	0.06–0.40	0.16–10.5
Arctic grayling	Fillet	4.45±2.70	4.83±2.59	1.28±0.90	1.32±0.72	0.26±0.29	2.13±1.71
(*Thymallus arcticus*)	n=2	—	—	—	—	—	—
		2.54,6.36	3.00,6.67	0.64,1.91	0.81,1.83	0.06,0.47	0.92,3.34
Burbot	Liver	40.6±8.93	9.74±2.70	25.2±14.7	16.2±5.89	4.41±1.58	47.3±17.7
(*Lota lota*)	n=15	38.9	9.96	22.7	15.8	4.55	47.2
		31.4–62.8	3.31–13.8	3.74–63.4	3.88–27.4	1.60–6.79	15.7–81.9

Values provided for each parameter are arithmetic mean±1 SD, median (if applicable), and data range; *n*, number of specimens analyzed.

Table 3
The mean (±1 SD) tolerable daily intake limits (Eq. (2)) for various subsistence dietary items based on dietary exposure guidelines[a,b] and the wet weight concentrations of persistent organochlorine contaminants listed in Tables 1 and 2

		Tolerable daily intake limits[c] (g)				
Species	Tissue	HCB	ΣCHL	ΣDDT	ΣHCH	ΣPCB
Bowhead whale	Blubber	184±80	149±43	4680±1940	67±15	218±72
	Maktak	687±99	389±100	25,000±4,270	302±73	697±152
	Tongue	2390±352	1974±494	96,800±38,100	980±197	4,480±448
	Liver	2930±1563	7594±2823	>500,000	2185±411	7200±1877
	Kidney	5130±3050	6270±4,078	>325,000	4800±3890	9510±6950
	Muscle	16,200±8600	18,453±8476	>500,000	9730±4470	43,800±17,100
	Diaphragm	21,600±14,400	20,663±12,650	>500,000	5960±2680	26,100±13,900
	Heart	25,300±8050	27,800±11,935	>500,000	6400±1310	40500±15,600
Beluga whale	Blubber	97±63	43±40	1316±1,196	122±44	32±21
	Maktaaq	276±106	80±33	4727±1,435	358±127	78±22
Ringed seal	Blubber	1924±1010	182±176	13,300±11,700	212±147	203±144
Bearded seal	Blubber	3000±328	200±22	9,788±1060	436±113	254±25
Fish[c]	Fillet	31,500±20,800	90,646±86,740	>500,000	89,300±87,500	64,700±88,000
Burbot	Liver	2250±468	1795±712	80,330±19,100	4940±2250	1450±390
TDI (μg kg^{-1} day^{-1})		0.3[a]	0.5[b]	20[a]	0.3[a]	1.0[a]

[a]Health Canada (1996).
[b]US EPA (1999) and WHO (1993).
[c]TDIL values calculated from pooled fish data (except burbot).

established at 0.05. The relationship between PCB and OC concentrations (wet weight; wet wt) with the percentage of extractable lipids in various bowhead whale tissues was investigated using Model-II linear regression. All other statistical comparisons of OC concentrations were performed using lipid-adjusted concentrations to reduce the variability of lipid content on analyte concentrations (Hebert and Keenleyside, 1995). The influence of tissue type and lipid content on the relative proportion of ΣOCs among bowhead whale tissues was investigated using a general linear model (GLM):

$$\Sigma\text{OCs proportion} = \mu + \text{tissue} + \text{lipid} + (\text{tissue} \times \text{lipid}) + \varepsilon, \qquad (1)$$

where μ is a constant and ε is the error term. Preliminary analysis indicated that the first-order interaction term (tissue × lipid) was not a significant factor according to Type-III sums-of-squares and was subsequently removed from the GLM. The Scheffé's method was selected to assess all a posteriori pairwise comparisons of group means deemed significant by the GLM.

ΣPCB concentrations (wet wt) in all biota were compared (via the Z test) to threshold concentrations (wet wt) in food destined for human consumption that range from 0.2 $\mu g\,g^{-1}$ (meat), to 0.5 $\mu g\,g^{-1}$ (poultry), and to 2.0 $\mu g\,g^{-1}$ in fish as recommended by Health Canada (Hing, 1998). Calculations of tolerable daily consumption (or intake) limits (TDIL) of bowhead whale tissues and other marine biota as food were calculated using the following equation:

$$\text{TDIL (g)} = \frac{\text{TDI}\,(\mu g\,kg^{-1}\,day^{-1}) \times \text{BW (kg)}}{\text{TC}\,(\mu g\,g^{-1}, \text{wet})}, \qquad (2)$$

where TC is the mean tissue concentration of ΣOC expressed in $\mu g\,g^{-1}$ (wet wt) and BW is the assumed body weight of an average adult human consumer (70 kg). The established tolerable daily intake (TDI) guidelines indicate that daily intake should not exceed 20 $\mu g\,kg^{-1}$ BW of ΣDDT, 0.3 for ΣHCH, 1.0 for ΣPCB, and 0.27 for HCB (Health Canada, 1996). For ΣCHL, the provisional tolerable daily intake (PTDI) value of 0.5 $\mu g\,kg^{-1}$ of body wt was used (US EPA, 1999; WHO, 1993). Estimating exposure to neonates and children was not attempted in this manuscript, as separate studies addressing concentrations of OCs in cord blood, breast milk, etc. are underway.

The potential human exposure to PCDDs and other dioxin-like contaminants in bowhead whale blubber and liver was investigated by normalizing to concentrations to 2,3,7,8-tetraCDD equivalency (TEQ) using toxic equivalency factors provided in Van den Berg et al. (1998). Additionally, TCDD-equivalent TDIL values were calculated using the Canadian TDI value of 10 $pg\,kg^{-1}$ BW day^{-1} (assuming a 70-kg adult consumer) from Health Canada (1996).

3. Results and discussion

3.1. OC concentrations in bowhead whale tissue

Concentrations of sum OC groups in bowhead whale tissues and other biota, including fish, are presented in Tables 1 and 2. The trophic transfer of OCs in this near-shore marine food web has been previously discussed (Hoekstra et al., 2003b). In summary, the varying concentrations and relative bioaccumulation of OCs in marine mammals and fish species mostly results from differences in the physical–chemical properties of OCs and the feeding strategy and biotransformation capacity of the species studied in this region (Hoekstra et al., 2003b).

Log_{10}-transformed, wet wt ΣOC and HCB concentrations in the bowhead whale were significantly correlated with the lipid content of each tissue ($P<0.01, r^2 = 0.81$). The influence of lipid content on OC concentrations is particularly evident when comparing contaminant levels in maktak and blubber. The lipid content of maktak was approximately <30% of the lipid percentage in corresponding blubber samples. However, the geometric mean concentrations of ΣOC in maktak were approximately 15–30% of mean blubber values (Table 1). While the decrease in the extractable portion of non-polar lipids from blubber to maktak is an important variable affecting OC concentrations, the likely heterogeneous stratification of OCs in blubber is also a significant factor (Aguilar et al., 1999). However, lipid-adjusted OC concentrations in biopsy samples of beluga whales from the Cook Inlet, Alaska were similar to full-core blubber samples obtained by necropsy (Krahn et al., 2003). Owing to the discrepancy between these studies, direct comparisons of OC data from biopsy (or maktak) and complete blubber-core samples should be conducted with caution.

In marine mammals, blubber is the major repository for many of the recalcitrant, lipophilic OCs (O'Shea, 1999). Subsequently, most investigations emphasize the determination of residue concentrations in this tissue to address various parameters related to organism health. Contaminant concentrations in cetaceans have been reported in other compartments, such as whole blood (or erythrocytes), liver, kidney, and muscle (e.g., Bruhn et al., 1995; Hoekstra et al., 2002b, 2003a; O'Shea, 1999). However, the detailed analysis of OCs in various cetacean tissues from the same specimen is limited compared to the amount of existing literature on contaminants in blubber.

The overall percentage of HCB, ΣDDT, ΣCHL, and ΣPCBs to the total OC concentration in bowhead whale was generally consistent among all tissues analyzed. The relative proportions of HCB (GLM, $F_{7,31} = 0.61$; $P = 0.74$), ΣDDT (GLM, $F_{7,31} = 0.45$; $P = 0.84$), ΣCHL (GLM, $F_{7,31} = 2.20$; $P = 0.08$), and ΣPCB (GLM, $F_{7,31} = 0.14$; $P = 0.91$) were not statistically different among bowhead whale tissues. While the limited number of tissue samples available in this study decreased the power of the statistical analysis, our findings are consistent with previous observations on the homogeneous distribution of lipid-normalized OC levels in cetacean tissues (Marsili and Focardi, 1997; Muir et al., 1999).

The relative abundance of ΣHCH in the bowhead whale was significantly influenced by tissue type (GLM, $F_{7,31} = 10.9$; $P < 0.001$). The proportions of ΣHCH of total OC concentrations in heart and diaphragm were significantly greater compared to other tissues (Fig. 2, top; $P < 0.03$). The greater percentage of ΣHCH in heart and diaphragm is due to the accumulation of β-HCH in these compartments (Fig. 2, bottom; $P < 0.01$). These results are consistent with the tissue-specific accumulation of HCH in rats, in which the accumulation of β-HCH in the heart was significantly greater than that of α-HCH; the predominant isomer in all other tissues (Siddiqui et al., 2003). To our knowledge, this is the first report of isomer-specific accumulation of HCH in any mysticete. Preferential accumulation of isomers could have toxicological implications (Willett et al., 1998) and may therefore be useful for future hazard assessments.

3.2. Human dietary exposure to ΣOCs and HCB

The ΣPCB concentrations in bowhead whale blubber exceeded the guideline for ΣPCB in meat (i.e., livestock) by approximately 80% ($P < 0.001$), but did not exceed those suggested for fish and poultry as recommended by Health Canada (Table 1, $P > 0.25$; Hing, 1998). All other bowhead whale tissues, along with all fish samples, did not exceed any of the suggested ΣPCB guidelines (Table 1, $P < 0.01$). Concentrations of ΣPCBs in the bearded seal blubber were statistically greater than the Health Canada ΣPCB threshold level for meat ($P < 0.01$), whereas ΣPCBs in ringed seals exceeded both meat and poultry guidelines ($P < 0.02$; both comparisons). As expected, ΣPCBs in beluga whale blubber exceeded all threshold levels, including those for fish (Table 2, $P < 0.004$), whereas beluga maktaaq ΣPCB concentrations only exceeded meat and poultry guidelines ($P < 0.001$). It should be noted that the maximum ΣPCB threshold levels were established for known or estimated consumption rates for the general Canadian population (Hing, 1998) and not those of indigenous peoples using traditional foods. Unfortunately, this information is not available for the subsistence communities in northern Alaska.

Tolerable daily consumption (or intake) limits for various marine mammal tissues and fish varied substantially among tissues and species (Table 3) and are likely better guidelines than those discussed above using

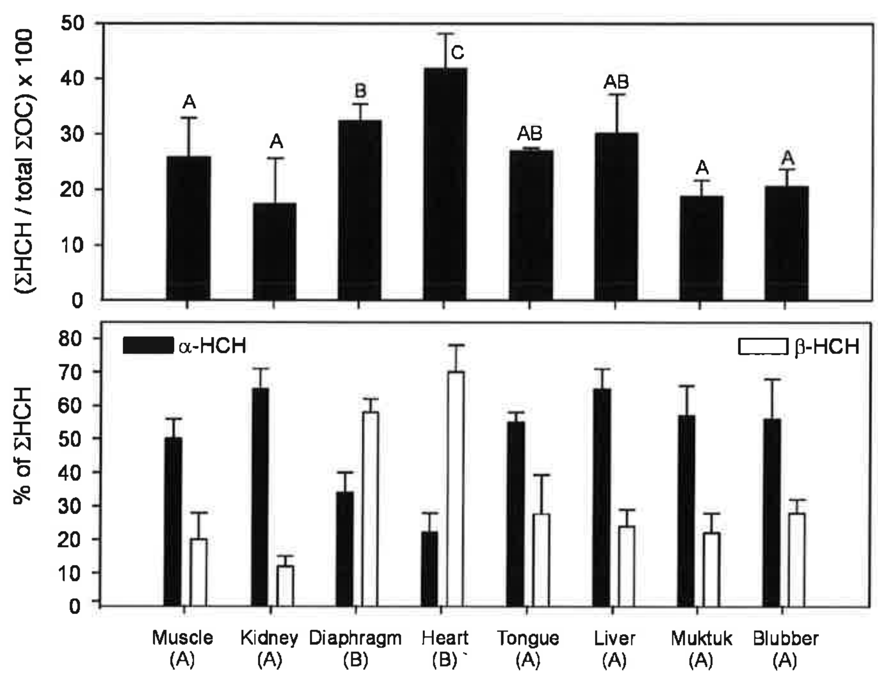

Fig. 2. The mean (± 1 SD) proportion of ΣHCH to the total OC concentrations (top) and relative percentage of α- and β-HCH isomers (to ΣHCH) among various bowhead whale tissues and maktak ($n = 5$, all groups). Graph bars designated with the same letters were not statistically different (GLM, Scheffé test, $P < 0.05$).

tissue residue limits (i.e., threshold levels). The maximum allowable daily intake for bowhead whale blubber would be 67 g (or 469 g per week) over an entire human lifespan as established by ΣHCH concentrations. However, cetacean blubber is typically consumed with epidermis (collectively called maktak and maktaaq for bowhead and beluga whales, respectively) as part of a subsistence diet in northern Alaska. Thus, TDILs for bowhead whale maktak provides a more realistic exposure scenario to OCs than those values calculated from the consumption of blubber alone. The TDIL for maktak would be 687 g for HCB, 389 g for ΣCHL, 302 g for ΣHCH, 697 g for ΣPCB, and approximately 25,000 g for ΣDDT. The most restrictive TDIL for maktak (302 g per day, or 2114 g per week) indicates that a consumer can intake more than 2 kg (approximately 4.4 lb) of maktak every week for his or her entire life and be below the critical level of exposure for these OCs. The mean TDILs calculated for other bowhead whale tissues were significantly greater than those determined for blubber due to the relatively lower concentrations of ΣOCs in other compartments and thus can be safely consumed in larger amounts.

In the other biota analyzed, the most restrictive level of daily consumption was calculated to be 32 and 78 g for beluga whale blubber and maktaaq and 182 and 200 g for ringed seal and bearded seal blubber, respectively, based on ΣCHL or ΣPCB concentrations. Consequently, an adult can safely consume 546 g (i.e., 1.20 lb) of maktaaq per week, every week, for an entire lifetime. While beluga blubber is typically consumed as a part of maktaaq, seal blubber is rendered into oil and used as a dip for a variety of frozen and boiled meats and fish by Native Alaskans (Kassam, 2001); therefore, it is more difficult to provide "safe" consumption advice. The maximum TDIL was 31,500 g for HCB in the fish fillets analyzed, whereas the tolerable consumption of burbot liver was more restrictive at approximately 1450 g per day (i.e., 10,150 g or 22.4 lb per week). Given the relatively large amounts of fish and seal oil dip needed to surpass intake guidelines, these species should be considered safe to consume under the guidelines discussed.

It should be noted that the TDIL for bowhead whale tissues and other biota are subject to interpretation due to the discrepancy of ΣCHL TDI values between various regulatory agencies. The WHO provisional TDI value of $0.5\,\mu g\,kg^{-1}\,day^{-1}$ used in this study was calculated by applying a 100-fold "safety factor" (to account for inter- and intraspecies variability) from the chronic "no adverse effect level" (NOAEL) of $50\,\mu g\,kg^{-1}$ using rats exposed to technical chlordane via dietary exposure (WHO, 1993). The same TDI value (or oral reference dose) is provided by the US Environmental Protection Agency (US EPA), which is based on applying a 1/300 uncertainty factor to an NOEAL value of $150\,\mu g\,kg^{-1}\,day^{-1}$ derived from a laboratory experiment in which mice were orally exposed to ΣCHL for 104 weeks (Khasawinah and Grutsch, 1989; US EPA, 1999). However, the Health Canada guideline value of $0.05\,\mu g\,kg^{-1}\,day^{-1}$ for ΣCHL exposure is lower and more conservative (Health Canada, 1996), reflecting a high uncertainty factor (1000) compared to other jurisdictions. However, information on how this Health Canada dietary guideline value was established is not available. Nevertheless, the application of the Health Canada TDI value for ΣCHL would decrease the TDIL for bowhead whale tissues by one order of magnitude (i.e., 14 and 39 g of blubber and maktak, respectively). Based on the data provided by the US EPA and WHO, it is our opinion that the Health Canada guideline is overly protective and would place undue restriction on the consumption of the subsistence foods presented in this manuscript. Therefore, the lifetime exposure guideline of $0.05\,\mu g\,kg^{-1}\,day^{-1}$ for ΣCHL proposed by Health Canada was not used for this analysis.

3.3. PCDDs and dioxin-like OCs in bowhead whale blubber and liver

Relatively low concentrations of coplanar PCBs (i.e., non-*ortho* substituted congeners), mono-*ortho* Cl-substituted PCB congeners, and PCDD and PCDF isomers were detected in bowhead whale blubber and liver samples (Table 4) compared to other OCs. Overall, the profiles of PCDDs and other dioxin-like compounds in bowhead whale blubber are consistent with reports in other cetaceans (e.g., Gauthier et al., 1998; Jarman et al., 1996; Muir et al., 1996). The compound with the largest contribution to total TEQ values is the coplanar PCB congener, 3,3′,4,4′,5-pentaCB (CB-126), which comprised 33 and 72% of the total TEQ value in blubber and liver, respectively. The mono-*ortho* Cl-substituted CB-118 and CB-156 were major contributors to the total TEQ (CB-118 blubber, 22%; liver, 6.0%; CB-156 blubber, 18%; liver, 10%) compared to other congeners. While the mono-*ortho* Cl-substituted PCB congeners CB-123 and CB-157 were not quantified in this study, the abundance of these congeners in arctic biota is minor (de Wit et al., 2004), and their subsequent contribution to the total TEQ is expected to be minimal.

The TCDD-TEQ concentrations were comparable to those reported in various commercially available food commodities with similar lipid content (Himberg, 1993; Dyke and Stratford, 2002; Schecter et al., 1997). For example, wet-weight TEQ concentrations of PCDD/Fs in various food items (e.g., beef, pork, chicken, farm-raised fish, etc.) ranged from 0.03 to $2.90\,pg\,g^{-1}$. However, these TEQ concentrations in the commercial foods did not include coplanar PCBs, which may

Table 4
Mean (±1 SD) concentration (pg g^{-1}, wet weight) and toxic equivalent (TEQ) values for polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyl (PCB) congeners in bowhead whale blubber and liver ($n = 5$)

Compound	Concentration (pg g^{-1} wet wt)		TEF[a]	TEQ (pg g^{-1} wet wt)	
	Blubber	Liver		Blubber	Liver
PCDDs					
1,2,3,4,6,7,8-heptaCDD	ND[b]	0.35±0.18	0.01	—	0.003±0.002
OCDD	ND	1.60±0.42	0.0001	—	0.002±0.001
ΣPCDDs	—	1.95±0.39	—	—	0.004±0.002
PCDFs	ND	ND	—	—	—
Non-ortho Cl-substituted PCBs					
3,3′,4,4′-tetraCB (CB-77)	8.75±4.27	5.50±1.73	0.0001	0.0009±0.0001	0.0006±0.0002
3,4,4′,5-tetraCB (CB-81)	4.95±3.99	ND	0.0001	0.0005±0.0004	—
3,3′,4,4′,5-pentaCB (CB-126)	11.52±4.55	2.28±0.58	0.1	1.152±0.455	0.228±0.058
3,3′,4,4′,5,5′-hexaCB (CB-169)	9.42±4.52	1.49±0.73	0.01	0.094±0.045	0.015±0.007
Σno-PCBs	39.8±16.1	9.27±2.72	—	1.24±0.438	0.243±0.065
Mono-ortho Cl-substituted PCBs					
2,3,3′,4,4′-pentaCB (CB-105)	2418±1175	86.5±40.9	0.0001	0.242±0.118	0.008±0.004
2,3,4,4′,5-pentaCB (CB-114)	1200±763	18.5±15.3	0.0005	0.600±0.381	0.009±0.007
2,3′,4,4′,5-pentaCB (CB-118)	7870±4170	197±106	0.0001	0.787±0.417	0.019±0.011
2,3,3′,4,4′,5-hexaCB (CB-156)	1298±1200	85.2±22.6	0.0005	0.649±0.601	0.031±0.023
2,3′,4,4′,5,5′-hexaCB (CB-167)	479±393	7.0±5.0	0.00001	0.005±0.004	0.001±0.001
2,3,3′,4,4′,5,5′-heptaCB (CB-189)	304±230	ND	0.0001	0.030±0.023	—
Σmo-PCBs	13500±7420	376±200	—	2.31±1.43	0.071±0.010
ΣTEQ				3.52±1.12	0.315±0.031
ΣTEQ range				(2.01–4.95)	(0.293–0.370)

[a]Toxic equivalency factors (TEFs) for PCDD/F and non- and mono-*ortho* substituted PCB congeners from Van den Berg et al. (1998).
[b]Not detected at concentrations greater than method detection limits.

represent a significant contribution to the overall ΣTEQ value (Huwe, 2002).

Based on the TDI value of 10 pg kg^{-1} BW day^{-1} (Health Canada, 1996), and assuming a 70-kg adult consumer, the TDIL of dioxin-like compounds from the consumption of bowhead whale blubber and liver was calculated to be 199 g (range, 141–348 g) and 2222 g (range, 1892–2390 g), respectively. As previously mentioned, bowhead whale blubber is typically consumed as maktak, and, therefore, actual exposure levels to OC via consumption will be overestimated only considering blubber concentrations. By comparing differences in ΣOC concentrations between these two food matrices (Table 1), the TCDD–TEQ concentration in maktak is likely three times lower than blubber concentrations, thereby increasing the TDIL to approximately 600 g (i.e., 4.2 kg, or approximately 9.2 lb per week) for this food item. Consequently, the risk to human health associated with exposure to PCDDs and dioxin-like compounds is minimal considering the current toxicological information and contaminant data.

3.4. Limitations and uncertainty

Careful interpretation of the dietary exposure values 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 ΣPCB and other OC concentrations in fish due to cooking (Kamrin and Fischer, 1999; Zabik et al., 1995). While the effect of rendering, fermentation, and other food preparation practices used by Alaska Natives (Kassam, 2001) on OC concentrations is unknown, the estimated daily intake values calculated from raw tissues in this study likely overestimate OC exposure from these subsistence dietary items when cooked. In addition, there are limitations in using TDI or PTDI guidelines to evaluate dietary exposure to anthropogenic compounds. As previously stated, these guidelines assume a life-long exposure and are developed using the NOAEL data from laboratory- or community-based exposure scenarios. These values were subsequently adjusted using conservative safety factors, typically by dividing the NOAEL by 100 or 1000, to compensate for interspecies differences between the test species and humans (Waltner-Toews and McEwen, 1994).

In addition, it should be emphasized that TDI/PTDI values are based on a lifetime of daily exposure. However, most wildlife and fish are hunted and consumed during select months of the year. For example, beluga whales in Wainwright, AK are generally hunted from June to July, whereas bowheads are hunted during two seasonal events in the Fall (September–October) and early Spring (April–May) at Barrow (Kassam, 2001; Fuller and George, 1997). Therefore, the estimated consumption of approximately 300 g of whale tissue (e.g., maktak) per day over an entire lifetime is likely very conservative and overestimates the true degree of risk associated with dietary exposure to OCs from these subsistence foods. However, it should be noted that dietary surveys of these subsistence species are required to properly elucidate the extent of contaminant exposure via dietary intake. The TDIL values determined in this study only considered PCB/OC intake from each individual tissue/food item. Thus, dietary surveys are needed to consider contaminant intakes from all possible dietary sources, along with the seasonal nature of hunting practices, when evaluating which food items significantly contribute to PCB/OC exposure via an actual subsistence diet (e.g., Van Oostdam et al., 2003).

The obvious sociocultural and nutritional benefits of a subsistence diet to the residents of northern Alaska must be considered. Preliminary information on the nutritional quality of bowhead whale tissues and other wildlife suggests 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., Blanchet et al., 2000; Kenny et al., 2004; Nobmann et al., 1992; Nobmann and Lanier, 2001; Van Oostdam et al., 2003; Woshner et al., 2001). The increasing prevalence of "Western" or modern diets, instead of 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 (Nobmann and Lanier, 2001; Van Oostdam et al., 2003). 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., Egeland et al., 1998; Kuhnlein and Chan, 2000), the consumption of bowhead whale tissues and other marine biota by Inuit of northern Alaska should be maintained and encouraged. Using a balanced approach to nutrients and contaminants, this information will allow individuals to decide for themselves which dietary habits best suit their lifestyle.

4. Conclusions

Persistent organochlorine contaminants were quantified in numerous tissues of the bowhead whale and other biota. In general, concentrations were relatively low in bowhead whales compared to other marine mammals and reflect the trophic status of this cetacean (Hoekstra et al., 2002a, 2003b). Lipid content significantly influenced the concentrations of ΣOCs in bowhead whale tissues and accumulation profiles of ΣOC groups were generally homogeneous in bowhead whale tissues. However, cardiac and diaphragm tissues contained a relatively greater percentage of HCH isomers, especially β-HCH, compared to other tissues. ΣOC concentrations were used to evaluate the human consumption rates based on established exposure guidelines. Overall, bowhead whale tissues and other biota from northern Alaska are safe to consume at, or below, the levels calculated in this study. Considering the safety factors applied to establish many of the TDILs, one could exceed the suggested intake levels by 100-fold the TDIL before achieving a "no observed effect" level and thus allow for a rather large tolerable range of intake. Additional information on the dietary profile (e.g., rates of consumption, peak seasons of consumption) of traditional/country foods consumed by the subsistence communities of northern Alaska is required to more definitively address the potential human health risks resulting from chronic exposure to OCs from a subsistence-based diet and the level of nutrient intake.

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