Concentrations of Polychlorinated Biphenyls (PCB's), Chlorinated Pesticides, and Heavy Metals and Other Elements in Tissues of Belugas, *Delphinapterus leucas*, from Cook Inlet, Alaska

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Introduction

The beluga, *Delphinapterus leucas*, is circumpolar Arctic in distribution and occurs in individual stocks throughout its range. Many of these stocks are distinct, genetically separate populations (e.g. St. Lawrence River estuary population in eastern Canada; Cook Inlet population in northwestern Gulf of Alaska), while for many other groups the degree of population separation is questionable. Based on the previous suggestions by Frost and Lowry (1990) and recent genetic evidence by O'Corry-Crowe et al. (1997), there appear to be two populations in Alaska (Cook Inlet and Bering Sea) containing five beluga whale stocks.

The Cook Inlet stock represents a geographically isolated, subarctic population of this species. The remaining four Alaska stocks, comprising the Bering Sea population, are the Bristol Bay, Norton Sound, eastern Chukchi Sea, and eastern Beaufort Sea stocks. The majority of the Bering Sea population appear to spend the winter in ice-free areas of the Bering Sea, while some may overwinter in polynyas of the Bering Strait and Chukchi Sea, and in the Gulf of Anadyr and Bristol Bay (Hazard, 1988; Frost and Lowry, 1990). Although Cook Inlet belugas may be found there during all seasons, they occasionally appear in the Gulf of Alaska as far east as Yakutat and as far west as the Shelikof Strait (Calkins and Pitcher, 1977; Hubbard et al., 1999). The move-

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ABSTRACT—Tissues from Cook Inlet beluga whales, Delphinapterus leucas*, that were collected as part of the Alaska Marine Mammal Tissue Archival Project were analyzed for polychlorinated biphenyls (PCB's), chlorinated pesticides, and heavy metals and other elements. Concentrations of total PCB's (∑PCB's), total DDT (∑DDT), chlordane compounds, hexachlorobenzene (HCB), dieldrin, mirex, toxaphene, and hexachlorocyclohexane (HCH) measured in Cook Inlet beluga blubber were compared with those reported for belugas from two Arctic Alaska locations (Point Hope and Point Lay), Greenland, Arctic Canada, and the highly contaminated stock from the St. Lawrence estuary in eastern Canada. The Arctic and Cook Inlet belugas had much lower concentrations (∑PCB's and ∑DDT were an order of magnitude lower) than those found in animals from the St. Lawrence estuary. The Cook Inlet belugas had the lowest concentrations of all (∑PCB's averaged 1.49 ± 0.70 and 0.79 ± 0.56 mg/kg wet mass, and ∑DDT averaged 1.35 ± 0.73 and 0.59 ± 0.45 mg/kg in males and females, respectively). Concentrations in the blubber of the Cook Inlet males were significantly lower than those found in the males of the Arctic Alaska belugas (∑PCB's and ∑DDT were about half). The lower levels in the Cook Inlet animals might be due to differences in contaminant sources, food web differences, or different age distributions among the animals sampled. Cook Inlet males had higher mean and median concentrations than did females, a result attributable to the transfer of these compounds from mother to calf during pregnancy and during lactation. Liver concentrations of cadmium and mercury were lower in the Cook Inlet belugas (most cadmium values were <1 mg/kg and mercury values were 0.704–11.42 mg/kg wet mass), but copper levels were significantly higher in the Cook Inlet animals (3.97–123.8 mg/kg wet mass) than in Arctic Alaska animals and similar to those reported for belugas from Hudson Bay. Although total mercury levels were the lowest in the Cook Inlet population, methylmercury concentrations were similar among all three groups of the Alaska animals examined (0.34–2.11 mg/kg wet mass). As has been reported for the Point Hope and Point Lay belugas, hepatic concentrations of silver were relatively high in the Cook Inlet animals and positively correlated with mercury and selenium concentrations in the liver.*

ment of these animals out of Cook Inlet and into the relatively ice-free Gulf of Alaska may occur during conditions of heavy ice in Cook Inlet (Calkins, 1983; Hansen and Hubbard, 1999).

The beluga whale is an important subsistence food resource across the North American Arctic. In Alaska, about 30 coastal villages of the Arctic Ocean and Bering Sea and communities located in Cook Inlet regularly take this species for food. The beluga is hunted primarily for its meat and maktaaq (i.e. skin plus blubber). These products are both consumed locally and are frequently distributed to villages that do not have access to this resource. As a result of the recent decline in the numbers of Cook Inlet belugas and in response to petitions from the State of Alaska and several organizations, NOAA's National Marine Fisheries Service (NMFS) has proposed that this population of belugas be listed as depleted under the Marine Mammal Protection Act (Fed. Regist., 19 Oct. 1999).

Belugas have the potential to accumulate relatively high concentrations of persistent contaminants in their tissues. This species feeds near the top of the marine food web and is therefore exposed to chemicals that accumulate and are biomagnified in the food chain (e.g. mercury, polychlorinated biphenyls (PCB's), etc.). The diets of these toothed cetaceans (odontocetes) vary depending on the summering areas of the individual stocks and the annual and seasonal variability in the abundance of suitable prey. Prey species include various fishes, crangonid shrimp, squid, and octopus (Calkins, 1983; Seaman et al., 1986).

The beluga whale is relatively long-lived (>35 yr) and maintains a thick layer of lipid-rich blubber, thus providing a storage depot for the accumulation of lipophilic contaminants (e.g. PCB's and chlorinated pesticides). For all animals, persistent organic contaminants are lipophilic (tending to associate with lipids) and accumulate in body fat. Mammals have various physiological mechanisms for metabolizing and excreting these compounds, the efficacy of which varies among species and depending on the chemical characteristics of the individual compounds. Although mechanisms for ridding the body of these compounds are available, the processes are usually slow, and the tendency is for the materials to accumulate in body fat over time. The beluga whale is common in nearshore waters and can be commonly found several kilometers up major river systems. Due to this behavior, this species has the potential to be exposed to coastal anthropogenic contaminants.

An important source of exposure to consider for all higher latitude biota, including belugas, is the apparently pervasive release of persistent contaminants in lower latitudes and their transport to remote areas. Many studies have documented global contamination by persistent organic contaminants and the role that atmospheric transport plays in the movement of such compounds from lower to higher latitudes (Barrie et al., 1992; Wania and Mackay, 1993; Iwata et al., 1994; Tanabe et al., 1994). Ocean currents probably also play a role, particularly for transport in ice-covered seas and for those compounds that are relatively hydrophilic (e.g. HCH and toxaphene). It is generally accepted that, except for localized "hot spots" of anthropogenic contamination, the anthropogenic contaminant concentrations reported for tissues of North American Arctic marine mammals probably originate from lower latitudes.

Since 1992, blubber, liver, kidney, and muscle specimens have been collected as part of the Alaska Marine Mammal Tissue Archival Project (AMMTAP) from beluga whales taken by Alaska Native subsistence hunters in Cook Inlet and archived under cryogenic conditions at the National Biomonitoring Specimen Bank at the National Institute of Standards and Technology, Gaithersburg, Md. The AMMTAP, which began in 1987 as part of the Outer Continental Shelf Environmental Assessment Program (OCSEAP), is described in Becker et al. (1988). In addition to Cook Inlet beluga whales, AMMTAP, in collaboration with the North Slope Borough Department of Wildlife Management, has obtained tissue samples from belugas taken in subsistence hunts from Point Hope (eastern Beaufort Sea stock) and Point Lay, Alaska (eastern Chukchi Sea stock). The eastern Beaufort Sea stock is thought to include the animals that migrate with bowhead whales, *Balaena mysticetus*, during the latter's spring migration and that spend the summer at the mouth of the Mackenzie River and Amundsen Gulf in the eastern Beaufort Sea (O'Corry-Crowe et al., 1997). The eastern Chukchi Sea stock includes animals of Kotzebue Sound as well as those that occur at Kasegaluk Lagoon (Point Lay) during the summer.

Aliquots of the Cook Inlet specimens, collected by the AMMTAP and archived in the NBSB, and additional samples collected from the same animals were analyzed for persistent organic contaminants (i.e. PCB's, and chlorinated pesticides) and inorganic constituents (elements), including heavy metals, and methylmercury. The results of the analyses for persistent organic contaminants for the Cook Inlet animals as well as the eastern Beaufort Sea and eastern Chukchi Sea belugas have been recently published elsewhere as part of an overall report on the concentration of these compounds in Alaskan beluga whales (Krahn et al., 1999). In addition, data have been published on belugas from Canada and Greenland that can be used for comparisons with the Cook Inlet animals (Deitz et al., 1990; Hansen et al., 1990; Muir et al., 1990b; Wagemann et al., 1990; Stern et al., 1994; Wagemann et al., 1996; Wade et al., 1997; Muir et al., 1999). This paper compares the tissue concentrations of PCB's, chlorinated pesticides, and elements (including heavy metals) in the Cook Inlet belugas with concentrations reported for the eastern Chukchi Sea and eastern Beaufort Sea animals, and Canadian and Greenland belugas.

Methods

Specimen Collections

Cook Inlet beluga blubber, liver, and kidney were selected for analyses from specimens stored under cryogenic conditions (–150°C) in the National Biomonitoring Specimen Bank at the National Institute of Standards and Technology (NIST) (Wise and Koster, 1995). These specimens were collected from

1992 to 1996 by the NMFS Western Alaska Field Office in collaboration with local Alaska Native hunters and with the support of the Alaska Beluga Whale Committee (Table 1). Collection procedures followed those standard AMMTAP protocols designed to preserve sample integrity and minimize sample contamination from handling (Becker et al., 1991). Blubber specimens consisted of whole thickness sections of blubber from the skin to the muscle, while kidney specimens consisted of whole kidney tissue (cortex and medulla). The gender of each animal and standard body measurements were recorded. Standard body measurements included: standard length (rostrum to fluke notch), axillary girth, fluke width, and blubber thickness. Teeth were collected, and the age of each animal was determined based on number of growth layer groups counted in a thin longitudinal section taken from the middle of the mandibular tooth (Burns and Seaman, 1986). Ages were determined by Robert Suydam, North Slope Borough Department of Wildlife Management, Barrow, Alaska, and Barbara Mahoney, NMFS, Anchorage, Alaska.

Table 1.—Cook Inlet belugas sampled for persistent organic contaminant and element analyses.

Animal ID no.	Gender	Age (yr)	Length (cm)	Tissues	Date sampled	Comment
692-BLKA-015	M	8.5	374	Blubber, liver	6 Oct. 1992	
692-BLKA-016	M	Adult	472	Blubber	23 July 1994	
692-BLKA-017	F	Unknown	305	Blubber	22 July 1994	
692-BLKA-018	M	Unknown	305	Blubber	23 July 1994	
692-BLKA-020	F	2	240	Blubber, liver	20 May 1995	
692-BLKA-021	M	10.5	409	Blubber, liver	3 June 1995	
692-BLKA-022	F	10	360	Blubber, liver	9 May 1995	
692-BLKA-023	F	12.5	353	Blubber, liver	1 June 1995	
692-BLKA-024	F	15	368	Blubber, liver	5 June 1995	With fetus
692-BLKA-025	F	Fetus	142.5	Blubber, liver	5 June 1995	Near term
692-BLKA-026	M	Adult	422	Blubber, liver	19 June 1995	
692-BLKA-027	M	9	377	Blubber, liver	27 June 1995	
692-BLKA-028	M	9	391	Blubber, liver	28 June 1995	
692-BLKA-029	M	Adult	413	Liver	11 Aug. 1995	
692-BLKA-031	F	Adult	367	Blubber	18 June 1995	Lactating
692-BLKA-032	F	Adult	256	Blubber	15 July 1996	
692-BLKA-033	F	Adult	359	Blubber	30 July 1996	Lactating
692-BLKA-034	F	Adult	377	Blubber	29 Aug 1997	
692-BLKA-035	M	Adult	415	Blubber	7 Oct. 1996	
692-BLKA-036	M	Adult	429	Blubber	7 Oct. 1996	
692-BLKA-037	M	Adult	367	Blubber	7 Oct. 1996	

Sample Preparation

Each tissue specimen to be analyzed (approximately 150 g) was homogenized using a cryogenic procedure designed to reduce the likelihood of changes in sample composition due to thawing and refreezing (Zeisler et al., 1983). Samples of this tissue homogenate, a frozen (nonfreeze-dried) powder, were aliquoted into Teflon[1] jars (10 mL) for storage (at – 80°C) until analyses were performed.

PCB and Chlorinated Pesticide Analyses

Analyses for PCB congeners and chlorinated pesticides were performed at the NIST Gaithersburg, Md., Laboratory, NMFS Northwest Fisheries Science Center (NWFSC), Seattle, Wash., and the Department of Fisheries and Oceans (DFO), Winnipeg, Man., Can. NIST methodology has been described in detail by Schantz et al. (1996). Samples (2–3 g) were mixed with sodium sulfate (approximately 100 g), internal standards were added, and the mixture Soxhlet extracted (18 h) using methylene chloride. The majority of lipids and biogenic material were removed by size exclusion chromatography (SEC) and then polychlorinated biphenyl (PCB) and pesticides fractions were isolated by normal-phase liquid chromatography (LC) on a semi-preparative-scale aminopropylsilane column. The fractions were analyzed by gas chromatography with electron capture detection (GC-ECD). NIST Standard Reference Materials (SRM's) 1588 (Organics in Cod Liver Oil) and 1945 (Organics in Whale Blubber) were analyzed with each set of samples for quality control.

Analytical procedures used by the NWFSC have been described by Krahn et al. (1988) and Sloan et al. (1993). Samples (1–3 g) were extracted following addition of internal standards, by maceration with sodium sulfate and methylene chloride. The methylene chloride extract was then filtered through a column of silica gel and alumina and concentrated for additional cleanup. High-performance liquid chromatography (HPLC) with a size-exclusion column was used to separate lipids and other biogenic material from a fraction containing the PCB's and chlorinated pesticides. This fraction was analyzed by GC-ECD and identification of individual compounds was performed in selected samples by GC-mass spectrometry. NIST SRM 1945 was analyzed with each set of samples.

DFO analytical procedures have been described by Muir et al. (1988 a,b), Muir et al. (1990a), and Stern et al. (1994). Samples (2–5 g) were mixed with anhydrous sodium sulfate and extracted by ball-milling (30 min) with hexane following addition of internal standards. The extract was allowed to stand for 4 h and then centrifuged (1,000 rpm). Lipids were removed using size exclusion chromatography (SEC), and three fractions were obtained using Florisil. The fractions (PCB congeners/4,4'-DDE/mirex, toxaphene/chlordane/mirex, heptachlor epoxide, and dieldrin) were then analyzed by GC-ECD using the same type of column as employed by NIST. SRM's 1588 and 1945 were analyzed with every 20 blubber samples for quality control. Toxaphene was quantified with a modification of previously published procedures (Muir et al., 1988b; Stern et al., 1994). Response factors for individual toxaphene peaks were calculated from weight percent of each peak in the total ion chromatogram of a toxaphene standard as determined by electron impact mass spectrometry on a GC-mass selective detector. Total toxaphene ($\sum$Toxaphene) was the sum of the concentrations of 19 peaks (Muir et al., 1990b).

[1]Mention of trade names or commercial firms does not imply endorsement by the National Marine Fisheries Service, NOAA.

The three laboratories that contributed data to this paper participate in an annual interlaboratory comparison exercise as part of the marine mammal quality assurance component of the NMFS Marine Mammal Health and Stranding Response Program. The results of these analyses were in good agreement (Schantz et al., 1996; Becker et al., 1999). Also, any differences among the laboratories for specific analytes were much less than the final differences between belugas from different locations using the databases from any of the three laboratories. Therefore, combining databases and using the means for samples for which there were results from more than one laboratory was justifiable.

Element Analysis

Instrumental neutron activation analysis (INAA) was used by NIST to analyze liver and kidney samples. INAA is a multielement analytical technique that provides data for a large number of trace elements using only a limited amount of sample. The INAA method is used routinely to measure 37 elements in the NBSB specimens: sodium (Na), magnesium (Mg), aluminum (Al), chlorine (Cl), potassium (K), calcium (Ca), scandium (Sc), vanadium (V), manganese (Mn), iron (Fe), cobalt (Co), copper (Cu), zinc (Zn), arsenic (As), selenium (Se), bromine (Br), rubidium (Rb), strontium (Sr), molybdenum (Mo), silver (Ag), cadmium (Cd), tin (Sn), antimony (Sb), iodine (I), cesium (Cs), barium (Ba), lanthanum (La), cerium (Ce), samarium (Sm), europium (Eu), terbium (Tb), hafnium (Hf), tantalum (Ta), gold (Au), mercury (Hg), thorium (Th), and uranium (U).

The INAA method, previously detailed by Mackey et al. (1995, 1996), consists of exposing samples and standards to a neutron field to produce radioactivity and measuring the energy and amount of resulting radiation. In preparation for INAA, subsamples of the frozen powder were lyphilized at a pressure of 1 Pa for 5 days during which the temperature was gradually increased from –20°C to 5°C. The dried powder was weighed into two 200 mg aliquots and each aliquot formed into a disk using a commercial stainless steel die and hydraulic press. The disks were packaged individually in acid-washed linear polyethylene (LPE) film. Since Hg analysis cannot be performed on these packaged disks due to permeation of volatile Hg into the film during irradiation, two 100 mg aliquots of the powder were placed in acid-washed quartz vials. The vials were flash frozen in liquid nitrogen prior to sealing to avoid any evaporative losses of elemental Hg or Hg compounds.

For each analysis, aliquots of powdered SRM's were packaged in the same way and were included in the analysis scheme for the purpose of quality control. Standards consisting of known amounts of each element were deposited onto filter papers, and formed into disks to insure consistent counting geometry between samples, controls, and standards. Analyses of SRM 1577a Bovine Liver and, beginning in 1991, a QA pilot whale liver tissue homogenate (Wise et al., 1993) were included with all multielement INAA measurements. Analyses of SRM 2710 Montana Soil and SRM 1571 Orchard Leaves were included with all Hg measurements.The irradiation and counting times for INAA were chosen to optimize the number of elements that can be determined and the detection limits for each. All irradiations were done at the NIST Reactor at a reactor power of 20 MW, which corresponds to a neutron fluence rate of approximately 2.0×10^{13} $cm^{-2}s^{-1}$.

In addition to INAA, total mercury analysis was performed at NIST using cold vapor atomic absorption spectrometry (CVAAS) following microwave digestion. For this procedure, 0.5 g of frozen homogenate was placed in a 100 mL Teflon PFA microwave container and the sample dissolved by microwave digestion using an acid mixture of 4 mL of HNO_3, 2 mL of $HClO_4$, and 0.5 mL of HF. After digestion and cooling, the sample was transferred to a 30 mL polypropylene bottle and diluted to a final volume of 25 mL with high-purity water containing 1% $K_2Cr_2O_7$. The weight and density of the final solutions were used in the calculation of the analyte concentration. The sample was then analyzed by cold vapor generation using the FIAS-200 flow injection system and Perkin-Elmer 5000 AAS. Standards were prepared from a 1,000 mg/mL stock standard. Two or three separate analysis runs (with a new set of calibration standards prepared for each run) were made for each sample preparation. Two or three separate sample preparations were made for each sample.

Methylmercury Analysis

Methylmercury analysis was performed at NIST using gas chromatography with atomic emission detection (GC-AED) as described by Donais et al. (1996). For this procedure, four methylmercury chloride calibration solutions and one ethyl mercury chloride internal standard solution were prepared gravimetrically in toluene. Subsamples of 0.2–0.5 g of frozen homogenates of the liver and kidney samples were extracted in 1 mL $CuSO_4$ solution (16 g of $CuSO_4$ to 100 mL of water), 4 mL acidic KBr solution (18 g of KBr and 5 mL concentrated H_2SO_4 diluted to 100 mL with chromatography grade water), and 2 mL toluene. Approximately 5 g of activated copper powder was added to each subsample prior to extraction. Following 1 h of agitation, each sample was centrifuged and the separated toluene layer was removed. The sample was extracted a second time with 2 mL of toluene (agitation for 10 min), the two toluene extractions combined, and the extracts for each sample/standard were spiked with a known mass of internal standard solution. The spiked extract was then concentrated to 0.5 mL and injected onto a preparative size exclusion chromatography column to remove lipids and biogenic material. One aliquot of each standard solution and one subsample of each tissue were extracted and analyzed by GC-AED in triplicate. NIST Pilot Whale Liver Control Material was analyzed with each set of samples for quality control.

Results and Discussion

PCB's and Chlorinated Pesticides

Results from analyses of blubber from 20 Cook Inlet beluga whales (10 males and 10 females) for PCB's and chlorinated pesticides are shown in Tables

Table 2.—Concentrations of ∑PCB, ∑DDT, ∑toxaphene, and ∑chlordane in blubber of Cook Inlet belugas compared with that reported for belugas from other North American locations. Values (mg/kg wet mass) are given as mean ±1 standard deviation.

Location (Date)	Gender	*n*	Age (yr)	∑PCB's	∑DDT	∑Toxaphene	∑Chlordane	Ref.[2]
West Greenland (1989–90)	M	71		5.38 ± 2.27	4.06 ± 2.50	3.69 ± 1.46	2.41 ± 1.08	1
Nuussuaq/ Disko Bugt	F	67		3.74 ± 2.31	2.60 ± 1.94	3.01 ± 1.62	1.79 ± 1.11	1
Cumberland Sound (1983)	M	6	7.3 ± 6.5	4.91 ± 0.25	6.83 ± 1.89	5.78 ± 5.39	2.38 ± 0.40	2
Pangnirtung	F	6	8.1 ± 7.3	1.15 ± 0.41	0.93 ± 0.55	1.77 ± 1.76	0.62 ± 0.15	2
St. Lawrence (1986–87)	M	4	17.5 ± 9.1	75.8 ± 15.3	101 ± 32.6	14.7 ± 2.46	7.43 ± 0.63	2
estuary	F	5	15.6 ± 10.4	37.3 ± 22.0	23.0 ± 17.3	6.34 ± 3.51	3.55 ± 1.99	2
E. Hudson Bay (1984–85)	M	6	15.6 ± 10.4	2.77 ± 0.51	2.27 ± 0.68	4.13 ± 0.82	1.86 ± 0.35	2
Nastapoka	F	6	17.0 ± 6.3	1.23 ± 0.84	0.98 ± 0.73	1.99 ± 1.10	0.87 ± 0.58	2
W. Hudson Bay (1986)	M	4	13.0 ± 4.8	3.12 ± 0.34	3.13 ± 0.20	5.10 ± 0.42	2.33 ± 0.26	2
Eskimo Point	F	4	10.3 ± 4.1	0.96 ± 1.00	0.85 ± 0.96	1.77 ± 1.41	0.85 ± 0.80	2
Jones Sound (1986)	M	8	4.4± 2.2	2.53 ± 0.57	1.96 ± 0.32	4.25 ± 1.02	1.87 ± 0.44	2
Grise Fjord	F	7	4.6 ± 2.9	2.46 ± 1.98	2.19 ± 1.69	3.74 ± 2.12	1.84 ± 1.13	2
Beaufort Sea (1983, 1987, 1989)	M	10	17.0[3]	3.33 ± 0.85	2.20 ± 0.83	3.83 ± 1.16	1.75 ± 0.41	2
Mackenzie R. & Point Hope	F	4	10.2 ± 7.0[4]	1.80 ± 0.77	0.95 ± 0.38	2.22 ± 1.05	0.99 ± 0.46	2, 3, 4
E. Chukchi Sea (1990,1996)	M	11	12.2 ± 4.5[5]	5.20 ± 0.90	3.63 ± 0.90	3.93 ± 1.16	2.42 ± 0.46	3, 4
Point Lay, AK	F	8	16.4 ± 7.5	1.50 ± 1.12	0.93 ± 0.85	2.62 ± 2.07	0.79 ± 0.61	3, 4
Cook Inlet, AK (1992–97)	M	10	9.2 ± 0.9[6]	1.49 ± 0.70	1.35 ± 0.73	2.40 ± 1.06	0.56 ± 0.25	3, 4
	F	10	9.9 ± 5.6[6]	0.79 ± 0.56	0.59 ± 0.45	2.02 ± 0.46	0.30 ± 0.22	3, 4

[1] ∑PCB's for the Beaufort Sea, eastern Chukchi Sea, and Cook Inlet animals are from Krahn et al. (1999). For the other beluga whale stocks, ∑PCB's are from Muir et al. (1990b). ∑Chlordane is the sum of the concentrations of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, and nonachlor III. ∑DDT is the sum of the concentrations of 2,4′-DDT, 4,4′-DDT, 2,4′-DDD, 4,4′-DDD, 2,4′-DDE, and 4,4′-DDE.

[2] 1 = Stern et al. (1994); 2 = Muir et al. (1990b); 3 = Becker et al. (1995b); 4 = Krahn et al. (1999).

[3] n = 2.

[4] n = 3.

[5] n = 10.

[6] n = 4.

Table 3.—Concentrations of dieldrin, HCB, ∑HCH, and mirex in blubber of Cook Inlet belugas compared with that reported for belugas from other North American locations. Values (mg/kg wet mass) are given as mean ± 1 standard deviation.

Location (Date)	Gender	*n*	Age (yr)	Dieldrin	HCB	∑HCH[1]	Mirex	Ref.[2]
Cumberland Sound (1983)	M	6	7.3 ± 6.5	0.91 ± 0.26	0.96 ± 0.18	0.39 ± 0.11	0.01 ± 0.01	2
Pangnirtung	F	6	8.1 ± 7.3	0.20 ± 0.33	0.18 ± 0.04	0.24 ± 0.06	0.01 ± 0.00	2
St. Lawrence (1986–87)	M	4	17.5 ± 9.1	0.93 ± 0.12	1.34 ± 0.44	0.37 ± 0.11	1.00 ± 0.64	2
estuary	F	5	15.6 ± 10.4	0.56 ± 0.31	0.60 ± 0.43	0.24 ± 0.10	1.11 ± 0.99	2
E. Hudson Bay (1984–85)	M	6	15.6 ± 3.0	0.28 ± 0.09	0.30 ± 0.19	0.21 ± 0.06	0.02 ± 0.01	2
Nastapoka	F	6	17.0 ± 6.3	0.14 ± 0.10	0.14 ± 0.12	0.15 ± 0.04	0.01 ± 0.01	2
W. Hudson Bay (1986)	M	4	13.0 ± 4.8	0.36 ± 0.07	0.61 ± 0.05	0.24 ± 0.07	0.36 ± 0.07	2
Eskimo Point	F	4	10.3 ± 4.1	0.14 ± 0.12	0.19 ± 0.18	0.15 ± 0.04	0.14 ± 0.12	2
Jones Sound (1986)	M	8	4.4 ± 2.2	0.34 ± 0.11	0.50 ± 0.21	0.19 ± 0.09	0.01 ± 0.00	2
Grise Fjord	F	7	4.6 ± 2.9	0.33 ± 0.23	0.39 ± 0.21	0.16 ± 0.08	0.01 ± 0.01	2
Beaufort Sea (1983, 1987, 1989)	M	10	17.0[3]	0.23 ± 0.05	0.59 ± 0.13	0.23 ± 0.06	0.04 ± 0.01	2
Mackenzie R. & Point Hope	F	4	10.2 ± 7.0[4]	0.16 ± 0.08	0.42 ± 0.29	0.27 ± 0.12	0.02 ± 0.01	2, 3, 4
E. Chukchi Sea (1990,1996)	M	11	12.2 ± 4.5[5]	0.39 ± 0.09	0.81 ± 0.12	0.33 ± 0.76	0.06 ± 0.02	3, 4
Point Lay, AK	F	8	16.4 ± 7.5	0.12 ± 0.10	0.23 ± 0.28	0.25 ± 0.12	0.02 ± 0.01	3, 4
Cook Inlet, AK (1992–97)	M	10	9.2 ± 0.9[6]	0.09 ± 0.05	0.22 ± 0.09	0.21 ± 0.07	0.01 ± 0.01	3, 4
	F	10	9.9 ± 5.6[6]	0.06 ± 0.05	0.15 ± 0.13	0.17 ± 0.05	0.01 ± 0.00	3, 4

[1] The sum of α-, β-, and γ-HCH.

[2] 1 = Stern et al. (1994); 2 = Muir et al. (1990b); 3 = Becker et al. (1995b); 4 = Krahn et al. (1999).

[3] n = 2.

[4] n = 3.

[5] n = 10.

[6] n = 4.

2 and 3 together with data for animals from Point Lay (eastern Chukchi Sea), Point Hope and Mackenzie River (eastern Beaufort Sea), five additional locations in Canada, and one location in Greenland. The sampling locations are shown in Figure 1. Data for the Alaska animals were previously reported by Becker et al. (1995b), Becker et al. (1997), and Krahn et al. (1999). Data for the Canadian and Greenland animals were taken from Muir et al. (1990b) and Stern et al. (1994). As these data were generated over a period of about 10 years, the question of temporal comparability arises. Recent results from across the Canadian Arctic indicate no significant change in the concentrations of PCB's, chlordane compounds, and toxaphene in beluga whale blubber over a 10-yr period (1983–94) and no change in DDT over a 20-yr period (1972–94) (Muir et al., 1999). These findings provide some confidence for comparing concentrations of persistent organic contaminants among the belugas from different locations used in this paper.

In Table 2, ∑PCB's as reported by Krahn et al. (1999) for the eastern Beaufort Sea, eastern Chukchi Sea, and Cook Inlet animals were calculated based on the procedure of Lauenstein et al. (1993) by multiplying the sum of the concen-

Figure 1.—Sampling sites for North American belugas: St. Lawrence River estuary (off map), western Greenland (Disko Bugt), Cumberland Sound (Pangnirtung), eastern Hudson Bay (Nastapoka River), western Hudson Bay (Eskimo Point and Southeast Baffin Island), Jones Sound (Grise Fjord), eastern Beaufort Sea (Mackenzie River and Point Hope), eastern Chukchi Sea (Point Lay), and Cook Inlet.

trations of the following congeners: 18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, and 209 by 2. For the other beluga locations, ∑PCB's was the sum of all congeners measured (65). Muir et al. (1990b) stated that 12 congeners accounted for >50% of the ∑PCB's in all of their samples: 52, 66, 95, 101, 99, 118, 149, 151, 153, 138, 187/182, and 180. It therefore appears that the ∑PCB's shown in Table 2 are roughly comparable for belugas for all locations. ∑Chlordanes is the sum of the concentrations of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, and nonachlor III. ∑DDT is the sum of the concentrations of 2,4′-DDT, 4,4′-DDT, 2,4′-DDD, 4,4′-DDD, 2,4′-DDE, and 4,4′DDE. Data for dieldrin, hexachlorobenzene (HCB), hexachlorocylohexane (the sum of α-, β-, and γ-HCH), and mirex are presented in Table 3.

For all Alaska belugas, PCB and chlorinated pesticide concentrations were higher in males than in females. This pattern is reported routinely for marine mammals, since females are able to transfer some of their body loads to the fetus during gestation and to their young through lactation. These avenues for decreasing body loads are not avail-

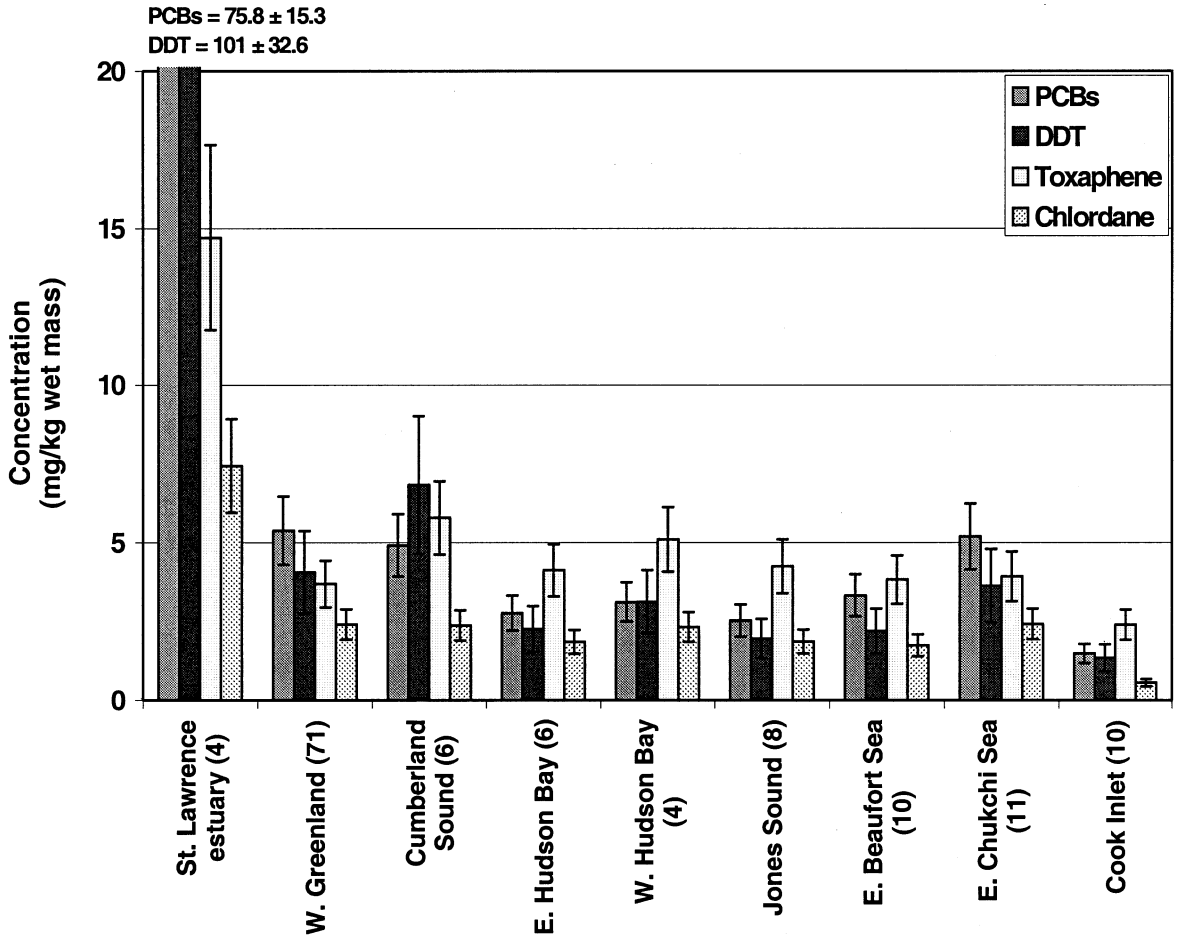

Figure 2.—Concentration (mean ±1 standard deviation) of PCB's, DDT, toxaphene, and chlordane in the blubber of male North American belugas. PCB's is ∑PCB's as described by Krahn et al. (1999) and Muir et al. (1990b). DDT is ∑DDT (sum of 2,4′-DDT, 4,4′-DDT, 2,4′-DDD, 4,4′- DDD, 2,4′-DDE, and 4,4′-DDE). Chlordane is ∑chlordane (sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, and nonachlor III). Numbers of animals are in parentheses.

able to the males. Because males tend to have higher concentrations of these compounds in their blubber, their values may be of more concern for human consumption health issues. However, levels in females and their transfer to offspring may be of more concern to the health of the animal population.

In the case of the beluga whales shown in Table 2, the variability in blubber concentrations of PCB's and chlorinated pesticides in males was less than in females for all belugas. Thus, males were selected for comparison of these compounds among the different belugas from different locations (Fig. 2, 3). The beluga whales from the St. Lawrence estuary had the highest concentrations of these compounds, with both ∑PCB's and ∑DDT being an order of magnitude greater than the other animals and mirex being on average more than twice as great as levels reported for belugas from other locations. The higher level of mirex in the St. Lawrence estuary animals (Fig. 3) is not surprising, since the St. Lawrence River drains an area having industrial sources for this compound. In St. Lawrence estuary belugas, ∑PCB's and ∑DDT contributed much more to the total persistent organic pollutant concentrations than in either the North American Arctic animals or Cook Inlet animals (Fig. 4). On average, these two groups of compounds made up 85–90% of the total blubber concentration in the St. Lawrence belugas, while contributing 40–50% in belugas from the other locations. On the other hand, toxaphene contributed a much larger fraction to the Arctic and Cook Inlet animals (20–50% on average) than was reported for the St. Lawrence estuary animals (10%).

Among the North American belugas, the Cook Inlet animals had the lowest blubber concentrations of PCB's and chlorinated pesticides (Fig. 2, 3). Krahn et al. (1999) compared both males and females from the Cook Inlet population with those from the eastern Chukchi and eastern Beaufort Sea belugas and found that the concentrations of these compounds were significantly lower in the Cook Inlet males. Although the concentrations in the Cook Inlet females appeared to be lower than those in the

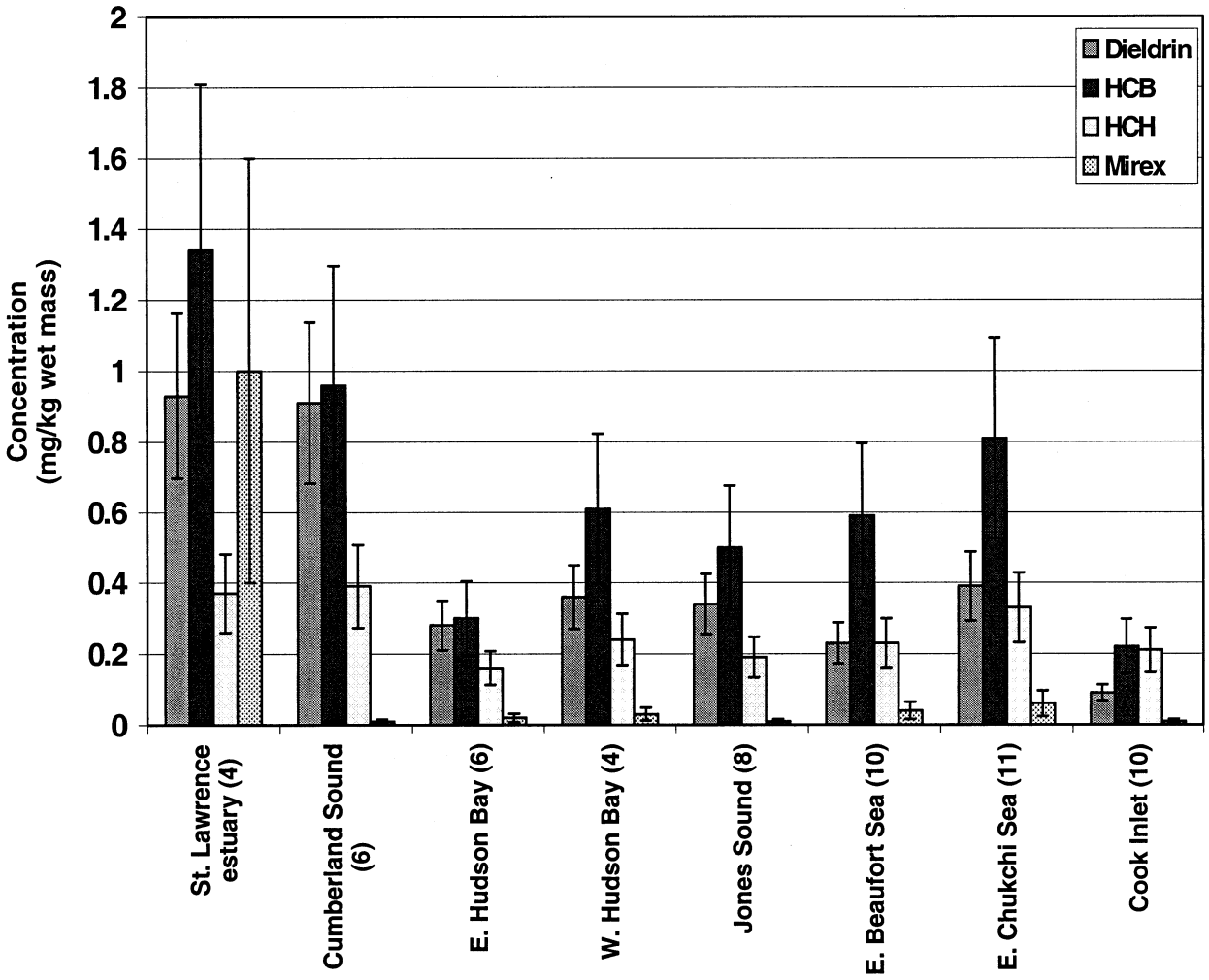

Figure 3.—Concentration (mean ±1 standard deviation) of dieldrin, HCB, HCH, and mirex in the blubber of male North American belugas. HCH is ∑HCH (sum of α-, β-, and γ-HCH). Numbers of animals are in parentheses.

eastern Chukchi Sea and eastern Beaufort Sea belugas, the differences were not statistically significant. The ratios of 4,4′-DDE to ∑DDT in the Cook Inlet animals were basically the same as those for other beluga whales in Alaska and Canada (0.47–0.57), suggesting uniform temporal exposure across the higher latitudes of North America for DDT (Krahn et al., 1999). However, concentrations of ∑chlordane were substantially lower in the Cook Inlet animals than in belugas from other locations, indicating a much lower contribution from this class of compounds to the total persistent organic contaminant loads in the Cook Inlet belugas than in the North American Arctic animals (Fig. 3, 4). Using the ratio of ∑chlordanes/∑PCB's, Krahn et al. (1999) found that the contribution of chlordane compounds to the total loads was substantially less in the Cook Inlet animals (~0.35) than in the eastern Beaufort Sea and eastern Chukchi Sea animals (~0.55), more than twice as high as for the St. Lawrence beluga whales (~0.1), and about half that for belugas from Hudson Bay and Jones Sound (~0.8). The differences in these ratios suggest different sources of these compounds, which may be related to geographic and latitudinal differences in atmospheric transport patterns and processes or different feeding habits or prey availability among these groups of belugas.

Elements

For most marine mammal species, little is known about normal concentration ranges of elements in specific tissues. Concentrations of essential elements such as zinc, copper, and selenium, and potentially toxic elements, such as cadmium and mercury, are commonly measured in liver and kidney. One must use caution comparing concentrations of specific elements in the tissues of marine mammals with those in terrestrial or freshwater species. Unlike persistent organic anthropogenic contaminants, all elements (including the potentially toxic ones) occur naturally in the earth's crust, and in many cases elevat-

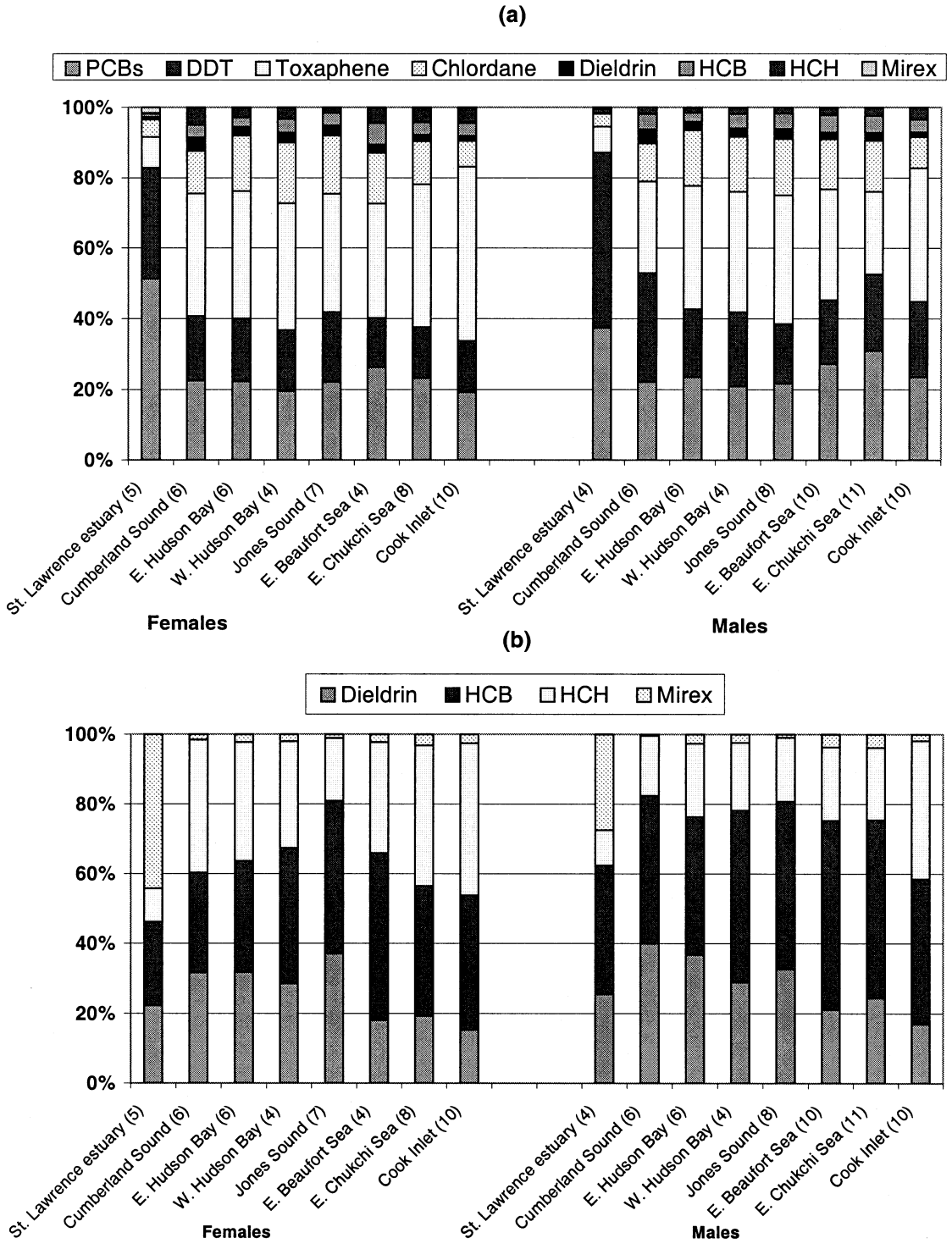

Figure 4.—Relative contributions to the persistent organic pollutant concentrations in blubber from North American belugas. PCB's is ∑PCB's as described by Krahn et al. (1999) and Muir et al. (1990b). DDT is ∑DDT (sum of 2,4′-DDT, 4,4′-DDT, 2,4′-DDD, 4,4′-DDD, 2,4′-DDE, and 4,4′-DDE). Chlordane is ∑chlordane (sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, and nonachlor III). HCH is ∑HCH (sum of α-, β-, and γ-HCH). Numbers of animals are in parentheses.

Table 4.—Comparison of means and ranges of concentration values of selected elements measured in the livers of Alaska belugas. Values (mg/kg, wet mass) are given as mean ± 1 standard deviation. Data for the Point Hope and Point Lay animals are from Becker et al. (1995b).

	Eastern Beaufort Sea (1989) Point Hope, Alaska		Eastern Chukchi Sea Point Lay, Alaska		Cook Inlet Cook Inlet, Alaska	
Element	Males (1)	Females (3)	Males (7)	Females (3)	Males (6)	Females (4)
Na	960.5	1198 ± 145.6 1049–1340	1397 ± 150.8 1136–1576	1321 ± 176.8 1141–1494	1331 ± 191 1101–1567	1204 ± 200 983–1449
Mg	111.8	151.4 ± 70.4 78.5–219	116.9 ± 33.18 58.5–164	157.6 ± 16.10 140.5–172.5	149.8 ± 16.9 131.2–177.4	134.5 ± 26 111–162.2
Cl	1384	1481 ± 83.0 1392–1556	1814 ± 87.98 1638–1880	1684 ± 171.6 1486–1790	1610 ± 269 1312–1971	1312 ± 198 1251–1561
K	2272	2524 ± 196.2 2315–2704	2222 ± 486.9 1523–2700	1754 ± 601.9 1326–2442	2898 ± 310.8 2552–3306	2849 ± 322 2516–3021
Ca	30.82	26.3 ± 0.878 25.30–26.92	31.64 ± 10.55 20.5–44	32.7 ± 7.145 28.5–40.95	41.6 ± 6.46 34.7–50.3	26.7 ± 2.91 24–30.95
V	0.034	0.079 ± 0.026 0.049–0.095	0.1644 ± 0.063 0.037–0.205	0.1529 ± 0.1092 0.0898–0.279	0.041 ± 0.012 0.021–0.054	0.034 ± 0.022 0.015–0.065
Mn	2.506	3.211 ± 0.134 3.093–3.357	1.939 ± 0.313 1.62–2.4	1.918 ± 0.2812 1.656–2.215	2.17 ± 0.33 1.70–2.52	2.651 ± 0.72 1.617–3.254
Fe	587.5	457.3 ± 143.6 362.5–622.5	599 ± 92.95 474.5–726	558.2 ± 224.3 332–780.5	316.9 ± 116.7 228.0–494.5	235.0 ± 149.0 100.0–443.0
Co	0.011	0.1 ± 0.06 0.01–0.18	0.012 ± 0.002 0.008–0.015	0.015 ± 0.006 0.008–0.021	0.009 ± 0.002 0.006–0.012	0.0281 ± 0.041 0.005–0.0895
Cu	12.36	12.86 ± 1.336 12–14.40	12.66 ± 6.449 6.85–26.4	21.18 ± 17.43 7.1–40.68	48.93 ± 39.79 15.62–123.8	29.26 ± 20.09 3.97–48.27
Zn	22.62	33.67 ± 4.324 30.24–38.53	23.86 ± 2.758 21.1–28.65	23.68 ± 2.62 20.90–26.1	27.26 ± 2.265 24.56–30.64	24.38 ± 1.591 22.66–26.07
As	0.177	0.215 ± 0.046 0.163–0.252	0.160 ± 0.203 0.065–0.616	0.180 ± 0.020 0.162–0.201	0.078 ± 0.023 <0.05–0.120	0.356 ± 0.329 <0.07–0.815
Se	6.243	8.472 ± 5.261 3.961–14.25	18.55 ± 8.770 7.01–29.36	37.33 ± 33.07 18.20–75.51	4.347 ± 1.561 2.907–6.088	2.620 ± 1.547 1.078–4.215
Br	25.62	24.36 (n = 1)	20.45 ± 1.704 18.36–23.06	25.31 ± 9.373 17.22–35.58	17.83 ± 4.26 13.17–22.48	17.28 ± 7.321 10.22–25.00
Rb		1.346 (n = 2) 1.174–1.518	1.050 ± 0.463 0.09–1.493	1.717 (n = 1)	1.765 ± 0.267 1.372–2.084	1.387 ± 0.174 1.222–1.602
Ag	14.58	20.83 ± 8.538 14.36–30.51	24.32 ± 10.65 14.38–40.69	46.62 ± 52.98 10.05–107.4	6.778 ± 4.169 1.513–11.61	4.383 ± 4.463 0.637–9.787
Cd	0.75	1.307 ± 0.745 0.455–1.840	1.884 ± 0.438 1.14–2.41	3.333 ± 0.501 2.755–3.645	<1 <0.44–<1	0.63 ± 0.155 <0.5–0.74
Cs	0.021	0.038 ± 0.007 0.031–0.038	0.028 ± 0.003 0.024–0.033	0.033 ± 0.003 0.030–0.035	0.051 ± 0.024 0.009–0.075	0.0644 ± 0.0094 0.0556–0.0746
Hg	3.52	5.462 ± 4.428 1.397–10.18	36.53 ± 16.81[1] 17.73–50.14	52.64 ± 22.86 27.85–72.9	5.454 ± 3.471 2.98–11.42	2.568 ± 1.816 0.704–5.03
Me-Hg	0.49	0.513 ± 0.231 0.37–0.78	1.517 ± 0.592[1] 0.86–2.01	1.153 ± 0.395 0.85–1.6	1.47 ± 0.66 0.800–2.11	0.52 ± 0.25 0.34–0.70

[1] n = 3

ed tissue levels in marine animals may reflect natural geochemical and biological food web processes rather than anthropogenic sources. This is not to say that anthropogenic contamination of the marine environment by heavy metals and metalloids does not occur. They do occur, but the identification of contamination and the source of this contamination is much more difficult with heavy metals than with the persistent organic contaminants, since all of the latter are anthropogenic in origin.

The concentrations of 19 elements and methylmercury measured in the livers of 10 Cook Inlet belugas are presented and compared in Table 4 with data published on belugas sampled by AMMTAP from Point Hope and Point Lay, Alaska (Becker et al. 1995b). In addition, liver concentrations of copper, cadmium, mercury, selenium, and zinc in these animals are compared in Table 5 with published data from west Greenland (Dietz et al., 1990; Hansen et al., 1990) and six locations in Canada (Wagemann et al., 1990; Wagemann et al., 1996). The values reported from Canada (the largest database) are based on dry mass concentrations. Therefore, for direct comparison with these published values, the wet mass concentrations of these five elements in the Alaska animals were converted to dry mass values using percent moisture data for each individual sample. The conversion factors of wet mass to dry mass for the Cook Inlet and

Table 5.—Concentrations of Cu, Cd, Hg, Se, and Zn in livers of Cook Inlet belugas compared with that reported for belugas from other North American locations. Values (mg/kg dry mass) are given as mean ± 1 standard deviation.

Location (Date)	n	Age (yr)	Cu	Cd	Hg	Se	Zn	Ref.[1]
West Greenland (1980-87)	40		not analyzed	8.84 (median)	10.5 ± 13.0[2]	14.8 (median)	114 (median)	1, 2
Cumberland S., Pangnirtung (1984)	11	11.2 ± 3.7	60.7 ± 36.0	23.5 ± 23.9	18.7 ± 16.8	10.31 ± 5.57	101 ± 25.9	3
St. Lawrence estuary (1982–86)	30	17.4 ± 8.5	37.3 ± 34.5	0.58 ± 0.41	126 ±161	79.2 ± 110	98.4 ±41.8	3
E. Hudson Bay, Nastapoka R. (1984)	15	13.2 ± 7.7	150 ± 200	18.9 ± 9.88	38.4 ± 48	16.7 ± 7.97	93.2 ± 9.99	3
W. Hudson Bay, Eskimo Pt (1984)	23	11.2 ± 6.7	117 ± 250	25.0 ± 22.9	24.9 ± 25.2	15.7 ± 8.78	90.4 ± 31.4	3
W. Hudson Bay, S.E. Baffin I. (1984–94)	139	11.9 ± 6.0	76.8 ± 131	26.0 ± 19.5	33.6 ± 33.0	21.4 ± 12.7	115 ± 32.2	4
Jones Sound, Grise Fjord (1984)	17	5.6 ± 4.8	39.1 ± 21.1	12.2 ± 14.1	8.27 ± 7.71	9.21 ± 4.74	93.6 ± 18.8	3
E. Beaufort, Mackenzie R. (1981, 84)	43	13.9 ± 5.5	50.3 ± 49.0	8.52 ± 5.42	44.1 ± 45.5[3]	23.3 ±19.7	92.0 ± 15.5	3
E. Beaufort, Mackenzie R. (1993–94)	77	19.3 ± 6.6	45.2 ± 28.4	9.08 ± 4.16	108 ± 98.8	75.2 ± 55.6	112 ± 20.0	4
E. Beaufort, Point Hope, AK (1989)	4	7.0 ± 2.5	48.0 ± 6.08	4.47 ± 2.50	18.8 ± 13.0	30.0 ± 15.0	120 ± 29.6	3
E. Chukchi Sea, Point Lay, AK (1990)	10	14.1 ± 6.8 [4]	61.6 ± 42.3	9.38 ± 3.39	179 ± 78.6[5]	97.2 ± 76.7	96.0 ± 11.5	5
Cook Inlet, AK (1992–95)	10	9.6 ± 3.7[6]	162 ± 130	2.39[7]	16.3 ± 13.0	14.3 ± 7.0	102 ±10.7	6

[1] 1 = Dietz et al. (1990); 2 = Hansen et al. (1990); 3 = Wagemann et al. (1990); 4 = Wagemann et al. (1996); 5 = Becker et al. (1995b); 6 = NIST (this paper).
[2] n = 6.
[3] n = 42.
[4] n = 2.
[5] n = 6.
[6] n = 8.
[7] Value based on n = 2; all other measures were below detection, ranging from <0.44 to <1mg/kg.

Point Hope animals were 0.26 ± 0.02 (mean ± 1 SE) and for the Point Lay animals was 0.25 ± 0.01. The data from west Greenland was converted from wet mass to dry mass basis using the conversion factor of 0.25. Since the west Greenland and Canadian papers do not differentiate concentrations in males and females, the data presented in Table 5 represent both genders.

As in the case of the persistent organic contaminants, these regional comparisons use data generated over a decade. Recent studies by Wagemann et al. (1996) indicate that, at least for the eastern Canadian Arctic, there has been little change in the concentrations of cadmium in beluga livers from 1981 to 1994; however, hepatic mercury concentrations were significantly higher in both the eastern Canadian animals and the western (eastern Beaufort Sea) from 1993 to 1994, than from 1981 to 1984. Wagemann et al. (1996) also suggested there was a higher rate of mercury accumulation recently than was the case 15 years before.

Elements that are essential for normal health of animals are well regulated metabolically. One would, therefore, expect little variation in the concentration levels of these elements among individual animals and among populations. For example, the hepatic zinc concentrations among the Alaska belugas vary by 11–25% from the average values. Nonessential elements are not usually regulated metabolically and may bioaccumulate and/or biomagnify. The concentration levels of these elements vary to a greater extent among individual animals and among populations. For example hepatic mercury concentrations of the Alaska belugas span more than an order of magnitude.

Mercury and cadmium are two heavy metals that bioaccumulate, and high concentrations may result in toxic effects to an animal. Cadmium can accumulate to high levels in both kidney and liver of birds and mammals. Cadmium concentrations in the kidneys are usually 3–4 times higher than in the liver of marine mammals. Hammond and Beliles (1980) state that in humans at low doses, kidney concentrations are about ten times higher than liver concentrations, and that with increasing levels of exposure the concentration ratio approaches one. Among Alaska marine mammals, high concentrations of cadmium in livers and kidneys have been reported for both bowhead whale, *Balaena mysticetus* (Krone et al., 1999), and walrus, *Odobenus rosmarus* (Taylor et al., 1989), and probably reflect both biomagnification through the food web and accumulation with increasing age of the animals. As compared to many other marine mammal species, cadmium concentrations are usually relatively low in belugas. In fact, the lowest levels reported for belugas were those found in the St. Lawrence estuary animals (Fig. 5), which are considered to be highly contaminated with anthropogenic pollutants. The concentrations of cadmium in the livers of the Alaska animals are relatively low as compared with those reported for belugas from Hudson Bay and Cumberland Sound in Canada, but similar to those reported for belugas from Greenland and Jones Sound, Canada (Table 5). The lowest hepatic cadmium concentrations were found in the animals from Cook Inlet, where concentrations ranged from the detection limit of <0.44 to 0.74 mg/kg wet mass (Table 4). Kidney concentrations ranged from 2.16 to 8.99 mg/kg wet mass (mean ±1 standard deviation = 3.98 ± 2.08 mg/kg wet mass).

Unlike cadmium, mercury is present at relatively high concentrations in the livers of belugas throughout North America. These levels are quite similar to those reported for Atlantic pilot whales, *Globicephalus melas* (Meador et al., 1993; Becker et al., 1995b; Mackey et al., 1995). Among the Alaska animals, the mean concentration of mercury in livers of the Cook Inlet animals was an order of magnitude lower than that reported recently for the eastern Beaufort Sea and eastern Chukchi Sea animals (significant at the 99% confidence level) (Table 5, Fig. 6a). Compared with most other belugas in North America, hepatic mercury levels in the Cook Inlet animals were at the low end of the range of values and similar to those reported for animals from Greenland and Cumberland Sound, Canada.

The mercury values shown in Tables 4 and 5 are for total mercury, which includes both inorganic and organic forms of this element. The toxic form of this element is methylmercury, which for animals with high total mercury concentrations in livers, usually contributes a relatively small fraction to the total concentration in this organ (Fig. 7a). Although the hepatic total mercury levels were lowest in the Cook Inlet animals, concentrations of methylmercury were similar among all of the Alaska animals. Concentrations in the Cook Inlet belugas ranged from 0.09 mg/kg wet mass in a fetus to 2.11 mg/kg wet mass in a large male (median levels were 0.5 mg/kg wet mass for females and 1.5 mg/kg wet mass for males). Dietz et al. (1990), in their study of belugas and narwhals, *Monodon monoceros*, from western Greenland, found that for total mercury hepatic concentrations of <1 mg/kg wet mass, about half of the mercury was organic and it did not exceed 2.2 mg/kg wet mass even with hepatic total mercury concentrations 50 times greater. The hepatic concentrations in the fetus and the large male from Cook Inlet (0.09–2.11 mg/kg wet mass) represent the largest range of methylmercury values found to date in Alaska belugas.

Marine mammals are exposed to methylmercury through their prey (particularly fish). There is some evidence to support the idea that marine mammals are able to metabolically convert the methylmercury consumed to the relatively nontoxic inorganic mercury that is then stored in the liver. If that is the case, over time the inorganic fraction of the hepatic mercury would be expected to increase, but the methylmercury fraction would not. Methylmercury values for the Alaska belugas generally do not exceed 2 mg/kg wet mass at the highest levels of total mercury (i.e. up to 73 ppm total mercury), and the highest percentage of methylmercury occurs at the low end of the total mercury range. The relationship of percent methylmercury with total hepatic mercury shown in Figure 7b for all of the Alaska belugas supports this hypothesis and suggests that a threshold concentration level may exist, above which methylmercury no longer accumulates in this organ.

The pattern of relative differences in selenium concentrations reported in liver

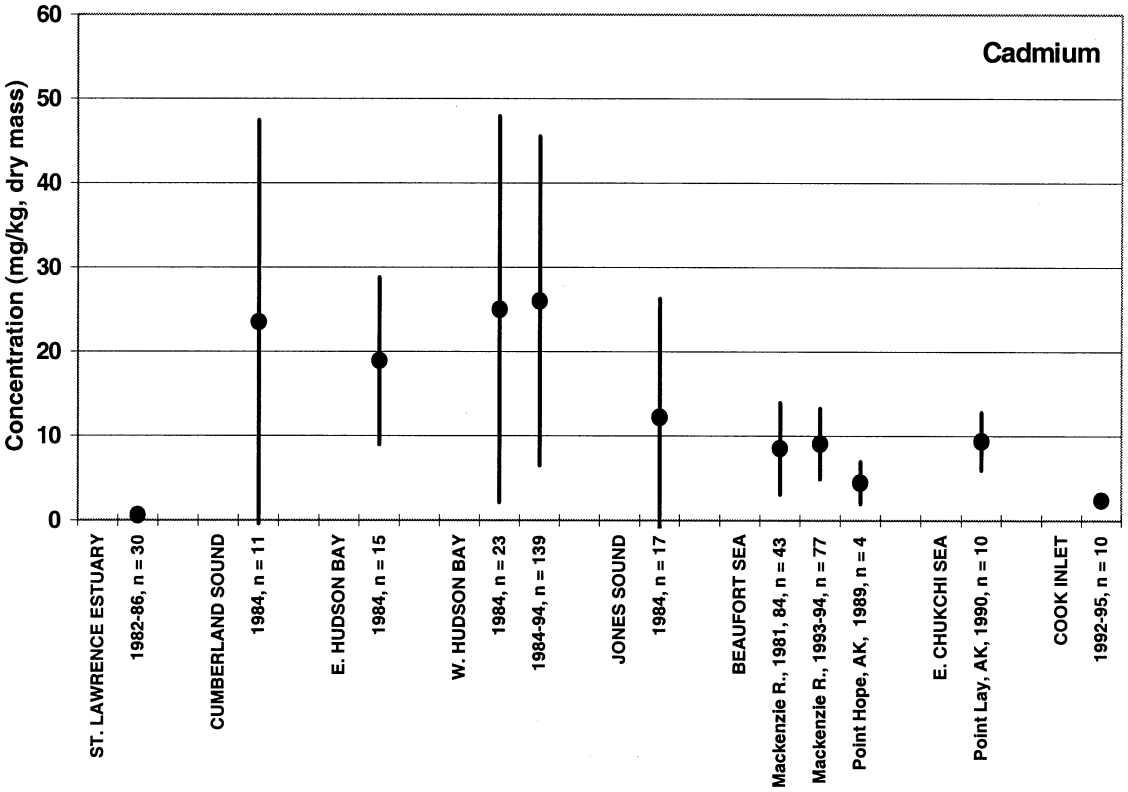

Figure 5.—Concentration of cadmium (mean ±1 standard deviation) in liver tissue of Cook Inlet beluga whales compared to concentrations reported in the livers of other North American populations and stocks of this species. The standard deviation for each mean (●) is indicated by a vertical bar. Geographic locations, dates of sample collections, and numbers of animals are shown on the X axis. Animals from the Mackenzie River and Point Hope are thought to be from the eastern Beaufort Sea stock. Data are from Wagemann et al. (1990), Wagemann et al. (1996), Becker et al. (1995b), and NIST (this paper).

tissue among the North American belugas is similar to that of total mercury (Fig. 6). The positive correlation of concentrations of selenium with mercury in liver tissue has been commonly reported for many species of animals (e.g. Koeman et al., 1973; Wagemann et al., 1983; Julshamn et al., 1987; Muir et al., 1988b; Meador et al., 1993; Mackey et al., 1995). In vertebrates this correlation has been shown for kidney as well as liver. This selenium-mercury relationship is thought to be indicative of both an accumulative and a protective function for the organism by incorporating the mercury within an innocuous complex involving selenium compounds and metal-binding proteins (Hammond and Beliles, 1980).

Selenium was positively correlated with mercury concentrations in liver and kidney tissue of the Cook Inlet belugas as well as the livers of the eastern Beaufort Sea and eastern Chukchi Sea animals. Regression plots of hepatic mercury concentrations versus hepatic selenium concentrations for all of the Alaska belugas yield a linear correlation coefficient (R^2) value of 0.73 (for N = 18).

Although silver is not usually measured in vertebrate tissues, it is one of the 37 elements routinely measured in liver specimens banked by the AMMTAP. Becker et al. (1995a) reported silver concentrations in the livers of eastern Beaufort Sea and eastern Chukchi Sea belugas that are from one to three orders of magnitude higher than what has been reported for other marine mammal species. The silver concentrations were positively correlated with mercury concentrations, selenium concentrations, and with ages of the animals. A linear relationship between hepatic silver and selenium concentrations also has been observed for several other marine mammal species, although hepatic concentrations of silver in other species are much lower (Mackey et al., 1996). This relationship remains when the silver and selenium concentrations for the Cook Inlet animals are added to the Alaska beluga whale database; in fact, the correlation between silver and selenium is much stronger than for mercury and selenium. Regression plots of hepatic silver concentrations versus hepatic selenium concentrations for all of the Alaska belugas yield a linear correlation coefficient (R^2) value of 0.93 (for N = 21).

Physiochemical processes that may be involved in the accumulation and possible interaction of selenium, mercury, and silver in beluga whale livers have been described elsewhere (Becker et al., 1995a; Mackey et al., 1996). Including the silver data for the Cook Inlet animals in the comparison of beluga whales with other marine mammal species supports the pattern of high levels of this element in the liver tissue of beluga

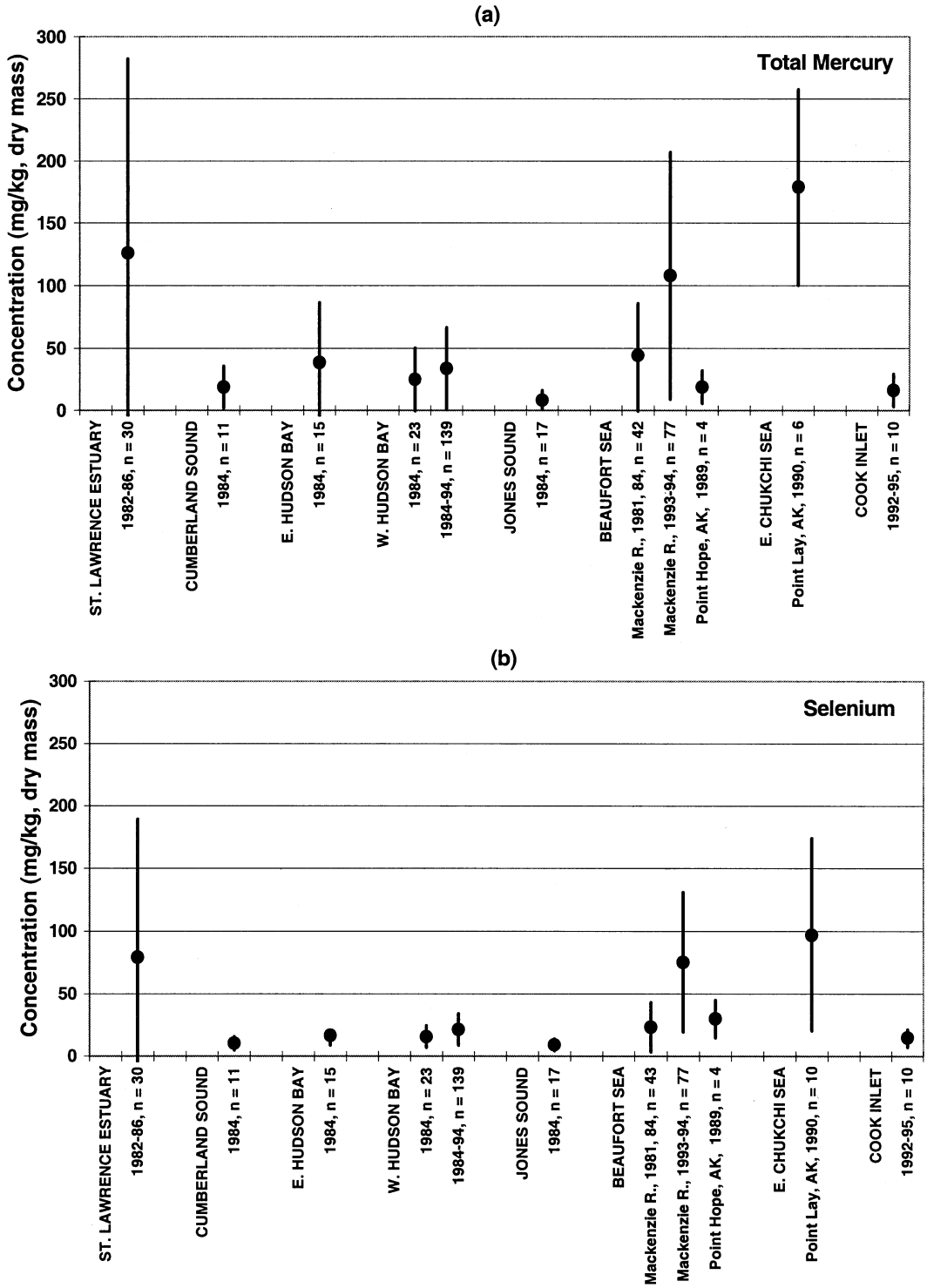

Figure 6.—Concentration of mercury and selenium (mean ±1 standard deviation) in liver tissue of Cook Inlet beluga whales compared to concentrations reported in the livers of other North American populations and stocks of this species. The standard deviation for each mean (●) is indicated by a vertical bar. Geographic locations, dates of sample collections, and numbers of animals are shown on the X axis. Animals from the Mackenzie River and Point Hope are thought to be from the eastern Beaufort Sea stock. Data are from Wagemann et al. (1990), Wagemann et al. (1996), Becker et al. (1995b), and NIST (this paper).

whales. Although the concentrations in the Cook Inlet animals (1.52–11.61 mg/kg wet mass) were significantly less than those reported previously for the eastern Beaufort Sea and eastern Chukchi Sea animals (10.05 to 107.4 mg/kg wet mass), the Cook Inlet beluga whales still had much higher levels than those reported for other marine mammal species (0.01–1.50 mg/kg wet mass) (Becker et al., 1997). These results suggest that the high levels of silver in

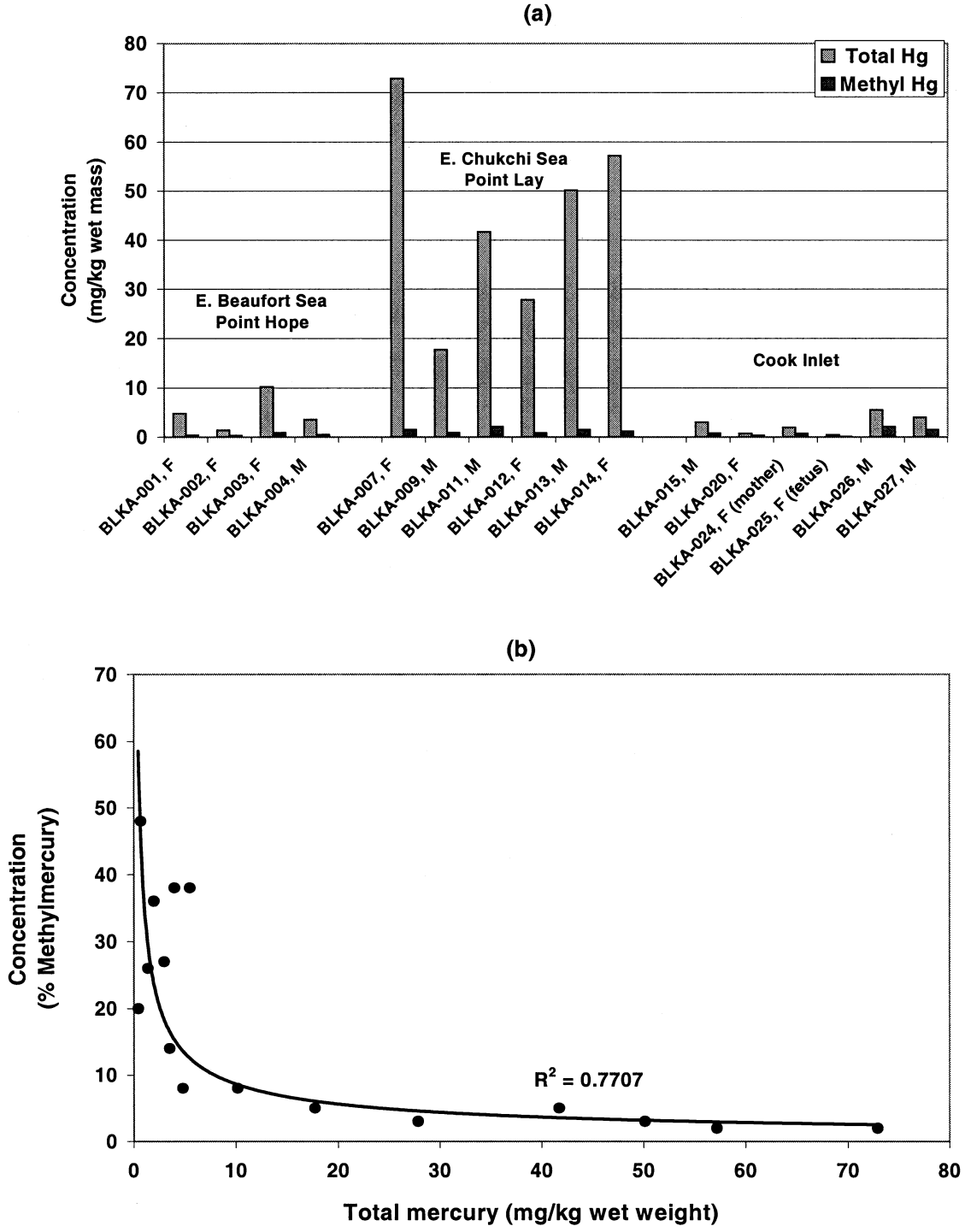

Figure 7.—Mercury in liver tissue of belugas. (a) Concentration of total mercury and methyl mercury in liver tissue of Cook Inlet belugas compared to the Point Hope and Point Lay animals. (b) Relationship between methylmercury and total mercury for beluga liver tissue from all three locations. The data is from Becker et al. (1995b) and NIST (this paper).

livers of beluga whales is a species-specific phenomenon and not a geographic phenomenon; however, data for belugas from other locations, including those from eastern Canada and Greenland, are necessary to determine if this is true.

Copper was present in higher concentrations in the livers of Cook Inlet animals than in the other Alaska animals (Table 4). When compared to the eastern Beaufort Sea and eastern Chukchi Sea belugas, concentrations were on average 2–3 times higher in the Cook Inlet animals (significant at the 95% confidence level). The relatively high average concentration and the high variability of the hepatic copper in the Cook Inlet belugas were similar to what has been reported for belugas from Hudson Bay (Table 5, Fig. 8).

Mackey et al. (1996) reported higher concentrations of vanadium in livers of Alaska marine mammals as compared with marine mammals from U.S. east coast. In the latter case, vanadium concentrations were at or below detection limits (<0.01 mg/kg), while in the livers of Alaska marine mammals, concentrations ranged from 0.02 mg/kg to 3.8 mg/kg wet mass. The highest concentrations were reported by Warburton and Seagars (1993) for walrus (0.25–3.8 mg/kg wet mass), while the lowest were reported by Mackey et al. (1996) for belugas (0.3–1.9 mg/kg wet mass). Vanadium concentrations in livers of the Cook Inlet belugas (Table 4) were the lowest of the Alaska animals (0.021–0.065 mg/kg wet mass), but still substantially higher than what has been reported from the lower latitudes of the United States (Mackey et al., 1996). Mackey et al. (1996), in their discussion of this apparent pattern of higher hepatic vanadium concentrations in marine mammals of the higher latitudes of Alaska, suggest that this may reflect a unique dietary source, a unique geochemical source of this element, or anthropogenic input to the Alaska marine environment. In any case, there is no evidence to indicate that the levels found so far in the Alaska animals pose any health hazard to the animals or to human consumers of these tissues.

Comparison of Cook Inlet Belugas with Belugas From Other Locations

Although occurring in what one might consider a region under substantially more anthropogenic influence than belugas from other locations in Alaska, the Cook Inlet animals had lower concentrations of PCB's and chlorinated pesticides than have been reported for other Alaska belugas or for this species from other areas of North America. Although one might suggest that differences be-

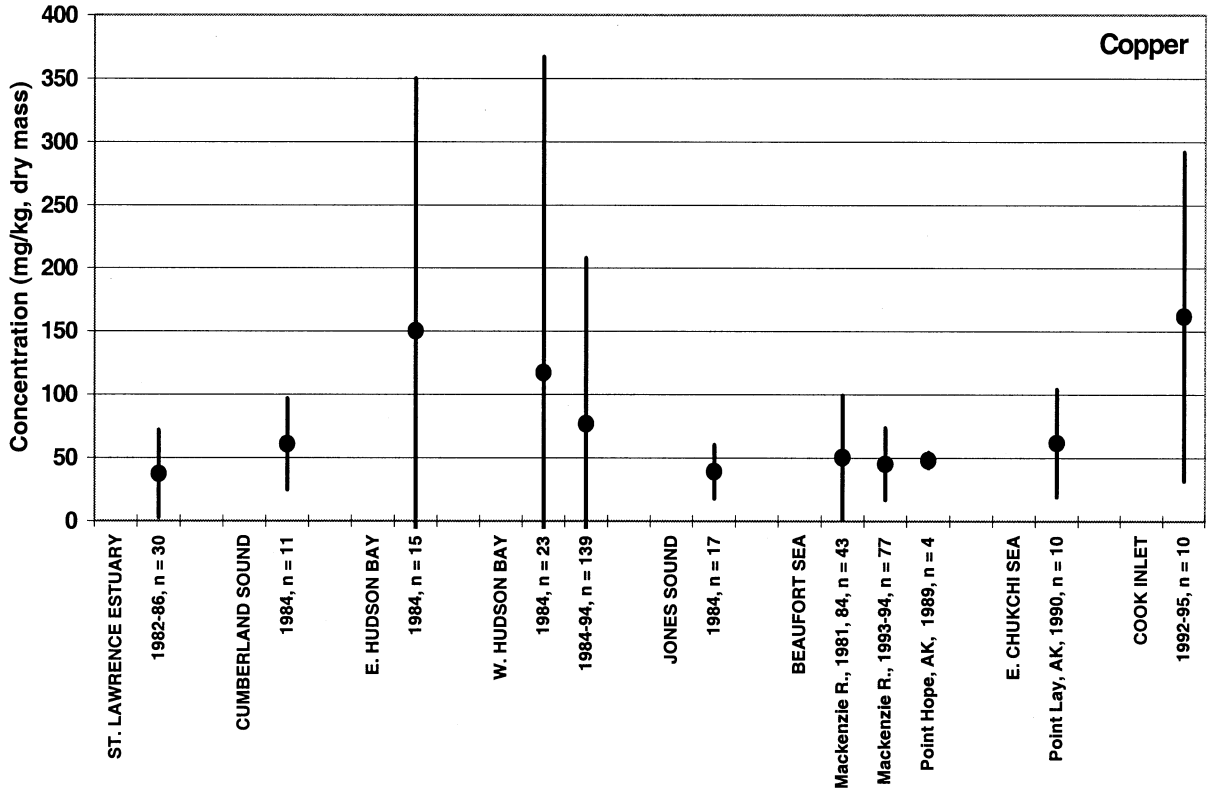

Figure 8.—Concentration of copper (mean ±1 standard deviation) in liver tissue of Cook Inlet beluga whales compared to concentrations reported in the livers of other North American populations and stocks of this species. The standard deviation for each mean (●) is indicated by a vertical bar. Geographic locations, dates of sample collections, and numbers of animals are shown on the X axis. Animals from the Mackenzie River and Point Hope are thought to be from the eastern Beaufort Sea stock. Data are from Wagemann et al. (1990), Wagemann et al. (1996), Becker et al. (1995b), and NIST (this paper).

tween the ages of the Cook Inlet animals and those of the other belugas might be a factor in these concentration differences, there are other differences that suggest that other factors may also be involved. Krahn et al. (1999), using principal components analysis, were able to demonstrate clear pattern differences in individual PCB and chlorinated pesticide analytes between the blubber of the Cook Inlet animals and that of the eastern Chukchi Sea animals. Chlordane compounds also contributed less to the organochlorine contaminant loads of the Cook Inlet animals than for the other North American belugas.

In addition to having relatively low concentrations of persistent organic contaminants in their blubber, the Cook Inlet belugas also had relatively low levels of mercury, selenium, and cadmium in their livers relative to other North American belugas (Table 4) and relatively low levels of silver and vanadium relative to the eastern Beaufort Sea and eastern Chukchi Sea animals (Table 5). Mackey et al. (1996) reported a positive correlation of liver concentrations of all five of these elements with increasing age in belugas. However, only a few of the Cook Inlet animals had known ages, and inclusion of the limited age values for this population to the data for the eastern Chukchi Sea and the eastern Beaufort Sea animals did not result in a significant correlation of any of the elements with age.

In comparison with the other Alaska belugas, the Cook Inlet belugas were also unique in having relatively high hepatic copper concentrations. The toxicological implication of these levels is unknown. Copper, being an essential element, is regulated metabolically in vertebrates and tends to vary among and within species. Diet and age appear to be important factors in accumulation of this element. McClurg (1984) suggested that the relatively high levels of copper in the liver of Ross seals from Antarctica were a reflection of the naturally higher levels of this element in its principal prey, squid. Prey difference might also be a factor in the hepatic copper concentration differences between the Cook Inlet and other Alaska belugas. Copper apparently does not accumulate with age, and, in fact, the highest copper concentrations are usually found in the younger animals. Wagemann et al. (1983) reported that copper concentration was negatively correlated with length for narwhals, and Honda et al. (1983) found the same for striped dolphin, *Stenella coeruleoalba*. However, the limited age data for the Cook Inlet belugas do not allow for an evaluation of age as the determining factor for the relatively high hepatic copper concentration in this population.

Compared with other belugas, the Cook Inlet animals appear to have unique patterns of persistent organic contaminants and elements in their tissues. Such differences for the Cook Inlet animals suggest different sources of these compounds, which may be related to geographic and latitudinal differences in atmospheric transport patterns and processes, geochemical source differences, or different feeding habits or prey availability for these animals.

Human Health Issues

Although lipophilic contaminants such as PCB's and chlorinated pesticides have been found to be generally lower in Arctic Alaska marine mammals than in marine mammals from lower latitudes, the levels in these Arctic animals are of special significance since they provide food for human residents whose diets are relatively high in fat. The beluga may therefore be of particular significance, since it has the potential for concentrating lipophilic compounds in its blubber at relatively high levels. Among the elements measured in the liver and kidney of the Cook Inlet belugas, only mercury would appear to be of any concern to human health. However, liver and kidney of belugas are not usually consumed as subsistence foods. Of more direct concern are the concentrations of the PCB's and chlorinated pesticides in the blubber and their potential health risk to consumers of maktaaq.

Among most of the marine mammals used as a subsistence food in Alaska (i.e. ringed seal, bearded seal, walrus, and bowhead whale), the beluga has relatively high concentrations of persistent organic contaminants in the blubber. Therefore, the consumption of the maktaaq of this species might be of particular concern for long-term human health effects.

Krahn et al. (1999) in their discussion of the persistent organochlorine contaminants found in the Cook Inlet, eastern Beaufort Sea, and eastern Chukchi Sea belugas calculated the allowable dose of contaminants in blubber based on the Allowable Daily Intake (ADI) levels recommended by the Contaminants Toxicology Section, Food Directorate, Health Canada, as described by Jensen et al. (1997). These calculations were based on the ADI of individual compounds (or groups of compounds such as ∑PCB's), the average body weight for an adult human (70 kg), and the mean tissue concentration in mg/kg wet mass. The ADI represents a life-long daily exposure and a significant safety factor has been applied to make these safe and conservative estimates. The ADI also reflects the relative toxicity of the compound. For example, the ADI of ∑PCB's is 1.0, while for ∑chlordane the ADI is 0.05. Their results indicate that the limiting compound for the Cook Inlet beluga whales is toxaphene with blubber consumption rate limit of 7 g/day.

For the eastern Beaufort Sea and eastern Chukchi Sea animals, the limiting compound was found to be chlordane at a recommended daily consumption rate less than half of that for the Cook Inlet animals. Such differences between the Cook Inlet animals and the other belugas reflect both the lower levels of all persistent organic contaminants in the Cook Inlet belugas and the lesser contribution of ∑chlordane to this population.

These calculations should also be viewed with caution. They are useful for comparative purposes among populations of animals; however, they do not provide sufficient information upon which to base dietary advice. Although providing some idea of relative risk from individual compounds and relative differences in these risks among species, the actual health effects from consumption of these compounds intermittently or constantly over a period of time, remain unknown. Potential risks must

also be evaluated relative to the known nutritional benefits of consumption of food derived from marine mammal tissues. It is imperative that other known benefits of subsistence use (cultural, economic, nutritional, and overall health) enter into the risk management equation. Discussions of these issues are provided by Kinloch et al. (1992) and Egeland et al. (1998).

Conclusions

The Cook Inlet belugas had much lower concentrations of PCB's and chlorinated pesticides than those which have been reported for belugas from Point Hope and Mackenzie River (eastern Beaufort Sea animals) and Point Lay (eastern Chukchi Sea animals). As compared to the other persistent organic contaminants, Σchlordane contributed substantially less to the total burden of the compounds in the Cook Inlet belugas than it did to the burden of any of the other belugas. This suggests that there is a different source (geographic or food web) of these compounds for the Cook Inlet belugas. The concentrations of persistent organic contaminants in the Cook Inlet males were higher than in the females from this population; however, the relative difference was much less than for the eastern Beaufort Sea and eastern Chukchi Sea animals.

Also, in the case of heavy metals and other elements, cadmium, mercury, and selenium were much lower in the livers of Cook Inlet animals than all other belugas, and vanadium and silver were lower in the Cook Inlet belugas than in the other Arctic Alaska belugas. Hepatic methylmercury levels were similar to those reported for other beluga whales and did not exceed 2.11 mg/kg wet mass. The silver, selenium, and mercury relationship previously described for other belugas and pilot whales was also found in the Cook Inlet population. The relatively high hepatic concentration of silver, described previously for the eastern Chukchi Sea and eastern Beaufort Sea belugas was also found in the Cook Inlet animals, suggesting a species-specific phenomenon.

An initial estimate of the human health risk associated with the consumption of beluga blubber (based on the Allowable Daily Intake levels recommended by Health Canada) suggested that the principal chemical limiting the recommended consumption rate of Cook Inlet blubber was toxaphene. This was different from the eastern Beaufort Sea and eastern Chukchi Sea belugas, where the limiting compound was chlordane. In the case of all Alaska belugas (including those from Cook Inlet), additional and more thorough evaluation of potential health effects relative to the known health benefits of marine mammal derived food is required to provide adequate and more scientific-based recommendations on human consumption.

Due to the lower concentrations in the Cook Inlet belugas, the effects of PCB's and chlorinated pesticides on animal health may be of less significance for the Cook Inlet animals than for belugas from other locations. However, very little is known about the role that multiple stressors play in the health of individual animals and populations. Biotoxins, bacterial or viral infections, parasitic disease, physical stresses in the environment, periodic limitations in food, or stress of being hunted in combination with the accumulation of toxic chemicals may further compromise animal health. The interaction of such stressors on a declining population, such as the Cook Inlet belugas, and the resulting effects on population recruitment should be a major avenue of future research and evaluation.

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