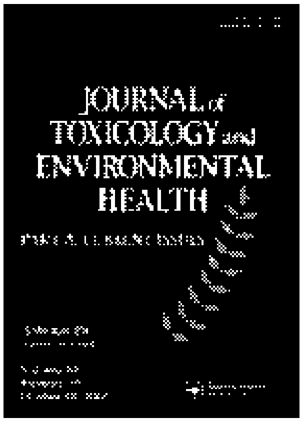

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ORGANOCHLORINE AND METAL CONTAMINANTS IN TRADITIONAL FOODS FROM ST. LAWRENCE ISLAND, ALASKA

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Marine mammals (bowhead whale, walrus, and various seals) constitute the major component of the diet of the Yupik people of St. Lawrence Island, Alaska. St. Lawrence Island residents have higher serum concentrations of polychlorinated biphenyls (PCB) than in the general U.S. population. In order to determine potential sources, traditional food samples were collected from 2004 to 2009 and analyzed for PCBs, three chlorinated pesticides, and seven heavy metals (mercury, copper, zinc, arsenic, selenium, cadmium, and lead). Concentrations of PCB in rendered oils (193–421 ppb) and blubber (73–317 ppb) from all marine mammal samples were at levels that trigger advisories for severely restricted consumption, using U.S. Environmental Protection Agency (EPA) fish consumption advisories. Concentrations of pesticides were lower, but were still elevated. The highest PCB concentrations were found in polar bear (445 ppb) and the lowest in reindeer adipose tissue (2 ppb). Marine mammal and polar bear meat in general have PCB concentrations that were 1–5% of those in rendered oils or adipose tissue. PCB concentrations in organs were higher than meat. Concentrations of metals in oils and meats from all species were relatively low, but increased levels of mercury, cadmium, copper, and zinc were present in some liver and kidney samples. Mercury and arsenic were found in lipid-rich samples, indicating organometals. These results show that the source of the elevated concentrations of these contaminants in the Yupik population is primarily from consumption of marine mammal blubber and rendered oils.

St. Lawrence Island, Alaska, is located in the northern Bering Sea, closer (61 km) to the Chukotkan Peninsula of northern Russia than to the Alaska mainland at Nome, which lies 322 km to the east of the community of Gambell. The island is home to approximately 1600 Yupik people who reside in the villages of Gambell and Savoonga. Situated at 63°10′ N latitude and 172°12′ W longitude and 240 miles south of the Arctic Circle, St. Lawrence Island is subject to long-range transport of semivolatile pollutants through what has become known as the "grasshopper" effect. This reflects the fact that semivolatile compounds, such as polychlorinated biphenyls (PCB) and chlorinated pesticides, are carried

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via atmospheric transport to the cold Arctic, where they condense from the vapor phase and deposit (Wania and Mackay 1995; Gouin et al. 2004). Because PCB are persistent and lipophilic, these compounds bioaccumulate in the food supply. There also are two formerly used defense sites on the island that constitute local sources of exposure to contaminants, including metals, which are also transported via particulates coming from Asia. Carpenter et al. (2005) previously reported that serum concentrations of PCB are higher in the Yupik than the general U.S. population. The most important route of exposure to the St. Lawrence Island Yupik people is almost certainly dietary intake. Therefore, the aim of the present study was to determine levels of PCB, three chlorinated pesticides, and seven heavy metals in a variety of traditional foods.

The Yupik people of St. Lawrence Island are heavily reliant on subsistence foods, particularly marine mammals. Marine mammals (walrus, seal, and bowhead whale) make up 94 and 88% of the total subsistence harvest at Gambell and Savoonga, respectively, while fish and other aquatic organisms make up only 4 and 5%, respectively (Amasuk and Trigg 2007). Traditional diets high in marine mammals and fish are major sources of persistent organic pollutants and heavy metals for many Arctic Indigenous Peoples (Kuhnlein and Chan 2000; Odland et al. 2003), and also for other indigenous populations (Stewart et al. 2011). Even plants have been documented to accumulate PCB (Hermanson and Hites 1990; Dayan and Koch 2002; Lovett et al. 1997), and therefore were also included in this study as plant consumption is also part of the subsistence diet on St. Lawrence Island.

Subsistence hunting of traditional food animals is necessary in most rural Arctic Indigenous communities, and contributes to the overall health of these communities in numerous ways. Villages rely on harvesting available foods to ensure care for all community members in need. Cash economies are often tenuous or nonexistent, and opportunities for local employment are scarce. Even when available, imported foods are often not an economically feasible option. Communal harvesting and sharing practices are central to the spiritual and cultural vitality of a village. These practices reinforce interdependence of community members and overall community self-reliance (AMAP 2002). Traditional foods provide beneficial nutrients such as omega-3 fatty acids and fat-soluble vitamins (Van Oostdam et al. 1999). When the traditional diet is replaced by imported foods, numerous adverse health effects are observed, including increases in obesity, diabetes, anemia, and dental problems, as well as decreases in physical activity and resistance to disease (Suk et al. 2004).

There is evidence that Alaska Natives and specifically the Yupik people of St. Lawrence Island are disproportionately exposed to recalcitrant pollutants. Previous studies demonstrated that the St. Lawrence Island Yupik people have elevated serum concentrations of PCB compared to the average U.S. population (Carpenter et al. 2005). Similar studies of Yupik women from the Yukon-Kuskokwim River Delta reported elevated serum concentrations of hexachlorobenzene (HCB), DDE, and PCB, as well as polybrominated diphenyl ethers (PBDE) (AMAP 2009). An earlier study (1980–1989) of Aleut women reported serum PCB concentrations even higher than those in St. Lawrence Island Yupik people (Rubin et al. 2001).

In comparison to other Arctic regions, there is a paucity of data on concentrations of recalcitrant pollutants in traditional foods from Alaska. Unlike the situation with food consumed by the Inuit in Eastern Canada and Greenland, where contaminants originate primarily from North America, marine mammals in the northern Pacific are likely to be affected more by contaminants coming from Asia. In many regions of the Arctic, recalcitrant pollutants reach concentrations that produce adverse health effects in food animals (Letcher et al. 2010), and by implication are likely to also produce adverse health effects in humans who consume these animals. Furthermore, there is reason for concern that the situation will become worse in the era of global warming of the Arctic (Jenssen 2006; Ford and Pearce 2010). Increasing temperatures associated with climate change may affect contaminant fluxes into Arctic ecosystems, and release previously sequestered contaminants into the environment, compounding this effect (Noyes et al. 2009). The multiple beneficial properties of traditional foods and their integral role in the vitality of northern Indigenous communities make it essential to understand the current extent of contamination in these food sources and to follow changes in the future.

There has been a great deal of past documentation indicating the food supply from subsistence lifestyles to be a source of contamination to Native American groups (Harris and Harper 1997; Hoekstra et al. 2005; Burger et al. 2007). At the same time, all too often the exposed populations are unaware of their exposure and/or of ways to reduce their exposure to contaminants. This is true not only for native populations, but for the U.S. population as a whole (Burger and Gochfeld 2009), making it that much more important for the results of this type of research to be made available for public health officials.

In this study a diverse set of traditional foods harvested by the inhabitants of St. Lawrence Island was analyzed at their request for four environmentally persistent organochlorines (OC)—PCB, the pesticides HCB, mirex, and *p,p′*-DDE, a breakdown product of the insecticide DDT—and seven heavy metals: arsenic (As), cadmium (Cd), copper (Cu), lead (Pb), mercury (Hg), selenium (Se), and zinc (Zn).

MATERIALS AND METHODS

Sampling

Samples were collected from 2005 to 2009 by community field researchers from the villages of Gambell and Savoonga as well as the Northeast Cape, one of the former defense sites. The Northeast Cape is near spring hunting camps used by multiple Yupik families. It is also an important location used throughout the year as a drinking-water source for travelers and as a refuge during inclement weather. Traditional food samples were collected at the time of butchering, while prepared food and rendered oil samples were collected from households as available. Plant samples were collected seasonally. In total, 327 samples were collected (at least 10 g free of fur and bone) for PCB and pesticide analysis, and 216 samples were collected for metal analysis, using a Teflon-coated knife. All liquids/oils were collected using a clean glass Pasteur pipette or eye dropper. Knives, pipettes, and eyedroppers were washed thoroughly with soap and water and rinsed with distilled water prior to sampling and between samples. Samples for PCB and pesticide analysis were placed in clean glass jars with lined lids and labeled with a marker. Samples for metals analysis were placed in separate plastic bags, labeled, and then placed into second plastic bags. Field blanks were collected during each sampling event. All samples were frozen as soon as possible. Samples were shipped on icepacks to the University at Albany for analysis.

PCB and Pesticide Analysis

Methods for PCB and pesticide analysis were previously published (DeCaprio et al. 2000). In brief, OC were extracted and analyzed using a dual-column gas chromatograph with electron capture detection, with analysis of 101 PCB congeners (83 individual congeners and 18 congeners as pairs or triplets) and 3 pesticides (DDE, HCB, and mirex). The method detection limit (MDL) for individual congeners ranged from 0.01 to 0.15 ppb (median 0.02 ppb, where 1 ppb = 1 ng/g). All results were calculated with values below the MDL set to zero. Calibration standards and QA/QC methods used were previously published (DeCaprio et al. 2000).

Tissue Sample Extraction Extraction methods for meat and skin tissue samples have been published (Hardell et al. 2010). Adipose tissue and blubber samples were extracted using this method, but with a sample of approximately 1 g, while plant samples were approximately 10 g.

Rendered Oil Extraction Rendered oil (0.2 g) was added to a clean 10-ml glass test tube. The method blank was an empty clean glass test tube, carried through the entire extraction process. Method controls were prepared using 1 ml of the standard solution (200 ng/ml) carried through the extraction process. Hexane (2 ml) and 10 μl surrogate standard were added with 1 ml H_2SO_4. Samples were vortexed for 1 min, and 1 ml was transferred to a Florisil-packed column for sample cleanup.

Sample Cleanup Sample extracts were cleaned up according to methods previously published (Hardell et al. 2010).

Metal Analysis

Sample Preparation Thawed samples were homogenized with a stainless-steel immersion blender. The blender was cleaned after each sample by wiping with a paper towel, washing with Citranox in deionized water, and rinsing with 18.3-MΩ deionized water. For each sample, a 0.25 ± 0.01g (wet weight) aliquot was digested according to a modified U.S. Environmental Protection Agency (EPA) method 3052 in a CEM MARS-5 microwave oven. For this method, a weighed sample was placed in a Teflon liner; 1 ml of laboratory-grade 30% hydrogen peroxide (FMC Corporation, ultrapure) and 9 ml of high-purity nitric acid (Alfa-Aesar Environmental Grade) were added, and the solution was heated gradually to a temperature of 180°C and held there for 10 min. A reagent blank and NIST Certified Reference Material was digested with each batch of 10 samples. After digestion, the solution was quantitatively transferred to a 50-ml Digitube; 0.1 ml of inductively coupled plasma–mass spectroscopy (ICP-MS) internal standard containing 50 ppm of Rh, In, and Bi in 5% nitric acid was added, and the solution was diluted to 50 ml with 18.3-MΩ deionized water. The digested solutions were stored for a period of days to several months. For Hg analysis, samples were measured directly from 50-ml tubes. For ICP-MS analysis, a 1-ml aliquot was transferred to a 14-ml polystyrene test tube and diluted to 10 ml total volume with 18.3 MΩ deionized water a day or two prior to analysis.

Calibration Standard and Quality Assurance/Quality Control Method blanks, duplicates, and matrix spikes were included in each sample batch for quality assurance. Standard reference materials, NIST 1566b oyster tissue, DOLT 3 dogfish liver, and DORM 3 fish protein, were measured with each sample batch to assess method precision and accuracy. For Hg, the coefficients of variation, expressed as relative standard deviation (%RSD) for nanograms per gram (ppb) analysis of NIST1566b, DOLT 3, and DORM 3, were 8, 3, and 13%, respectively. The average measured concentrations of the NIST 1566b, DOLT 3, and DORM 3 standards were not significantly different from the certified values within a 95% confidence interval. NIST 1566b was used for quality control with ICP-MS.

Mercury Analysis Mercury analysis was carried out by cold vapor atomic absorption spectrometry with a Leeman Labs Hydra AA automated Hg analyzer. HCl rinse solutions (Fisher, trace metal grade), $SnCl_2$ reductant (Fisher, Hg analysis grade), and calibration standards were prepared from high-purity reagents immediately prior to analysis. Calibration curves were generated using four or five aqueous HCl calibration standards (0.01 to 20 μg/L Hg) for each analytical batch. Method blanks, sample duplicates, and certified reference materials (NIST 1566b oyster tissue [0.037 μg/g Hg], DORM 3 fish protein [0.409 μg/g Hg], or DOLT 3 dogfish liver [3.37 μg/g Hg]) were included in each analysis batch for quality assurance. The limit of detection and limit of quantitation reported were 3 and 10 standard deviations of replicate analyses of the blank, respectively.

Other Trace Metal Analysis Trace metal analysis (other than Hg) was conducted by inductively coupled plasma–mass spectrometry (ICP-MS) on a PerkinElmer/Sciex Elan 6100 DRC. Isotopes analyzed were ^{63}Cu, ^{64}Zn, ^{75}As, ^{82}Se, ^{114}Cd, ^{121}Sb, ^{206}Pb, ^{207}Pb, and ^{208}Pb, with ^{103}Rh, ^{115}In, and ^{209}Bi as internal standards. Due to the natural isotopic variation of Pb, the sum of three Pb isotopes was used to determine total Pb concentration. Blank and standard solutions were prepared at the time of analysis from high-purity reagents. The limits of detection and limits of quantification were calculated as already described for Hg.

Data Analysis

Summary statistics were calculated using Microsoft Excel (7.0). Samples for PCB and pesticides with no detectable levels were treated as zero. Generally, samples that are non-detects have levels between zero and the limit of detection; therefore, by treating the non-detects as zero likely result in reported concentrations being lower than actual concentrations. We chose to report conservatively due to the low sample sizes for many sample sets. For statistical calculations, metals concentrations below the method detection limit were assigned a value of the detection limit divided by the square root of two.

Risk-based consumption limits were calculated for PCB and for PCB, *p,p'*-DDE, and HCB combined using U.S.EPA-derived methodology for fish (U.S.EPA 2000).

The equation used for cancer health endpoints is based on the cancer slope factors for each compound (Table 2), and calculated to prevent 1 excess cancer in 100,000 over a 70-yr exposure:

$$CR_{lim} = ARL \times BW/CSF \times C_m \qquad (1)$$

where CR_{lim} is the maximum allowable fish consumption rate (kg/d), ARL is the maximum acceptable individual lifetime risk level (10^{-5}, unitless), BW is the consumer body weight (70 kg, used here for this purpose), C_m is the measured concentration of a chemical contaminant m in a given species of fish (mg/kg), and CSF is the cancer slope factor [2.0, 1.6, and 0.34 $(mg/kg\text{-}d)^{-1}$ for PCB, HCB, and DDE (using Total DDT CSF), respectively].

To calculate the allowable number of meals of a specified meal size that may be consumed over a given time period, Eq. (2) was used:

$$CR_{mm} = CR_{lim} \times T_{ap}/MS \qquad (2)$$

where CR_{mm} is the maximum allowable fish consumption rate (meals per year), CR_{lim} is the maximum allowable fish consumption rate (kg/d), T_{ap} the time averaging period (365.25 days per year), and MS is the meal size (0.227 kg fish/meal size used).

Equation (3) was used to calculate CR_{lim} for carcinogenic health endpoints for the mixture of all the analyzed contaminants in each species, assuming additive carcinogenic effects of the contaminants. Equation (2) was then applied to calculate the meal consumption limits for the mixtures of carcinogens.

$$CR_{lim} = (ARL)BW)/\sum_{m=1}^{x} C_m(CSF_m) \qquad (3)$$

RESULTS

PCB and Pesticides

Seal, Whale, and Walrus Blubber and Polar Bear and Reindeer Adipose Tissue Table 1 shows levels of PCB, DDE, HCB, and mirex in blubber and adipose tissue samples obtained from bearded, ringed, and spotted seals, bowhead whale, walrus, polar bear, and reindeer. All samples contained detectible PCB, and most contained detectible DDE and HCB, although at considerably lower levels than were found for PCB. Mirex was not found at high levels in most samples. The highest PCB concentration was observed in polar bear adipose tissue, which was expected since polar bear feed primarily on seals and constitute the apex predator of the Arctic ecosystems. Notable levels were present in all three species of seal (bearded, ringed, and spotted). Bowhead whale, which feeds low on the food chain, had a surprisingly elevated amount of PCB in blubber. Walrus blubber samples contained intermediate but significant levels of PCB. Reindeer, which feed on plants and lichens, had low concentrations of PCB but did show concentrations of DDE and HCB in adipose tissue samples that were at concentrations higher than those for PCB, although lower than that in the marine mammal adipose tissue.

Rendered Oil Samples Rendered oil samples (Table 2) contained the greatest

TABLE 1. PCB and Pesticides Results (ppb, w/w) in Blubber/Adipose Tissue

Species and sample type	PCBs	*p,p'*-DDE–85	HCB	Mirex
Bearded seal blubber (*n* = 21)	116.37 ± [illegible] (17.10–354.65)	38.28 ± 47.11 (0.07–193.50)	3.35 ± 1.77 (0–6.33) ND = 1	1.43 ± 1.83 (0–6.76) ND = 7
Ringed seal blubber (*n* = 2)	73.33 ± 10.24 (66.09–80.57)	9.27 ± 10.03 (2.17–16.36)	7.66 ± 8.97 (1.31–14.00)	0.35 ± 0.49 (0–0.69) ND = 1
Spotted seal blubber (*n* = 6)	190.44 ± [illegible] (38.26–545.25)	59.06 ± 143.35 (0–349.36) ND = 1	38.78 ± 76.07 (0.70–193.50)	4.75 ± 4.85 (0–13.22) ND = 1
Bowhead blubber (*n* = 3)	317.67 ± 87.43 (224.43–397.86)	6.29 ± 1.16 (5.36–7.59)	23.82 ± 2.14 (21.60–25.87)	0.26 ± 0.01 (0.25–0.27)
Bowhead mungtak[a] (*n* = 7)	143.61 ± 188.71 (0.09–537.90)	4.72 ± 3.38 (1.09–11.04)	11.59 ± 14.77 (1.49–34.81)	0.13 ± 0.13 (0–0.31) ND = 2
Walrus blubber (*n* = 29)	33.89 ± 34.20 (3.07–187.97)	1.03 ± 0.89 (0–3.54) ND = 2	0.36 ± 1.14 (0–6.18) ND = 2	0.79 ± 0.88 (0–3.17) ND = 10
Polar bear blubber (*n* = 2)	445.95 ± 193.37 (309.22–582.68)	25.82 ± 0.19 (25.68–25.95)	8.22 ± 1.75 (6.98–9.46)	0.82 ± 1.16 (0–1.64) ND = 1
Reindeer alunga[b] (*n* = 2)	0.16 ± 0.05 (0.12–0.19)	2.24 ± 1.38 (1.26–3.21)	0.09 ± 0.12 (0–0.17) ND = 1	0 ND = 2
Reindeer adipose tissue (*n* = 6)	2.35 ± 1.50 (0.22–3.55)	11.39 ± 13.59 (0.54–37.67)	2.45 ± 5.57 (0–13.8) ND = 4	0 ND = 6

Note. Results are presented as the average, standard deviation, range, and the number of non-detects (ND).

[a] Mungtak is the Yupik term for blubber and skin of the bowhead whale.

[b] Alunga is the Yupik term for the lining of the reindeer's stomach.

TABLE 2. PCB and Pesticides (ppb, w/w) in Rendered Oils

Species and sample type	PCBs	*p,p'*-DDE	HCB	Mirex
Bearded seal rendered oil (*n* = 4)	241.93 ± 124.03 (129.67–402.27)	44.79 ± 21.39 (23.58–71.69)	10.30 ± 11.53 (4.34–27.53)	2.75 ± 1.46 (1.59–4.89)
Ringed seal rendered oil (*n* = 2)	421.11 ± 64.38 (375.59–466.63)	64.85 ± 60.63 (21.98–107.72)	12.90 ± 7.00 (7.95–17.85)	2.09 ± 2.96 (0–4.18) ND = 1
Spotted seal rendered oil (*n* = 2)	251.43 ± 147.95 (146.79–356.06)	6.53 ± 2.23 (4.95–8.10)	15.84 ± 11.13 (7.97–23.71)	3.49 ± 0.98 (2.79–4.18)
Bowhead whale rendered oil (*n* = 3)	354.06 ± 201.22 (158.92–560.83)	26.43 ± 14.06 (10.27–35.14)	16.97 ± 0.76 (16.46–17.79)	2.90 ± 2.64 (0–5.17) ND = 1
Walrus oil (*n* = 8)	193.61 ± 131.81 (36.04–440.44)	6.14 ± 10.80 (0–30.53) ND = 1	4.33 ± 6.40 (0–17.78) ND = 1	4.22 ± 2.02 (0.79–6.75)

Note. Results are presented as the average, standard deviation, range, and number of non-detects (ND).

concentrations of PCB, ranging from a sample average of 193.6 ppb in walrus (*n* = 8) to 421.1 ppb in ringed seal (*n* = 2). The highest individual rendered oil sample was 560.8 ppb in bowhead whale (*n* = 3, range = 158.9–560.8 ppb). The rendered oils uniformly had higher concentrations of PCB than did blubber and adipose tissue samples. This was also the case for most, but not all, of the pesticides. The ringed seal oil had both the largest individual sample and average concentrations of DDE at 107.2 and 64.9 ppb respectively. Only one rendered oil (walrus) sample was a non-detect (ND) for DDE. The highest individual sample concentration of HCB was in a bearded seal sample, 27.5 ppb, and the highest averaged concentration was measured in the bowhead whale, 16.9 ppb. The only HCB ND was in a walrus oil sample. Averaged sample mirex concentrations ranged from ND to 4.2 ppb, with 2 NDs, one bowhead oil and one ringed seal oil. The ringed seal samples contained much larger concentrations of DDE than the other oils, with an average concentration of 64.9 ppb compared to the spotted seal which showed the lowest concentrations with an average of 6.5 ppb. Mirex concentrations were markedly lower.

Meat Samples Table 3 shows PCB and pesticide concentrations in 90 meat samples. Total PCB averages ranged from 0.1 ppb in reindeer rump meat to 38.7 ppb in hair seal (Yupik term for young ringed seal) meat. Only one meat sample (walrus) was a non-detect for PCB. Walrus meat samples ranged from ND to15.1 ppb, $n = 28$. A bowhead meat sample had the highest individual total PCB concentration of 102.8 ppb, with bowhead samples ranging from 1.0–102.8 ppb, $n = 4$.

Pesticide concentrations were low overall, with sample averages ranging from ND to 6.8 ppb, ND to 1.4, and ND to 0.4 ppb, respectively, for DDE, HCB, and mirex. There were 23 ND for DDE, with the highest individual sample

TABLE 3. PCB and Pesticides in Meat (ppb, w/w)

Species and sample type	PCBs	*p,p′*-DDE+85	HCB	Mirex
Bearded seal dried meat ($n = 2$)	5.05 ± 0.11 (4.97–5.12)	0.93 ± 1.05 (0.19–1.67)	0.04 ± 0.04 (0.01–0.07)	0.11 ± 0 (0.11–0.11)
Bearded seal meat ($n = 23$)	2.60 ± 2.64 (0.07–8.99)	0.55 ± 0.85 (0–4.01) ND = 3	0.13 ± 0.26 (0–1.16) ND = 3	0.04 ± 0.08 (0–0.38) ND = 13
Ringed seal meat ($n = 4$)	5.06 ± 4.07 (2.43–11.12)	1.37 ± 1.30 (0.85–3.79)	0.11 ± 0.06 (0.04–0.79)	0.03 ± 0.03 (0–0.07) ND = 1
Spotted seal meat ($n = 7$)	6.08 ± 5.46 (0.10–13.31)	1.57 ± 2.06 (0.03–4.79)	0.39 ± 0.63 (0–1.78) ND = 1	0.04 ± 0.05 (0–0.11) ND = 3
Bowhead meat ($n = 4$)	27.20 ± 50.42 (0.99–102.83)	0.27 ± 0.28 (0–0.64) ND = 1	0.58 ± 0.28 (0.26–0.85)	0.08 ± 0.11 (0–0.22) ND = 2
Walrus breast meat ($n = 2$)	14.26 ± 4.68 (10.95–17.57)	2.46 ± 1.76 (1.21–3.70)	0.14 ± 0.16 (0.03–0.25)	0.10 ± 0.04 (0.07–0.13)
Walrus meat ($n = 28$)	1.72 ± 3.33 (0–15.07) ND = 1	0.01 ± 0.04 (0–0.11) ND = 15	0.07 ± 0.13 (0–0.51) ND = 19	0.08 ± 0.21 (0–1.03) ND = 20
Polar bear meat ($n = 3$)	13.23 ± 12.27 (4.03–27.16)	0.76 ± 0.72 (0.12–1.54)	0.29 ± 0.17 (0.17–0.48)	0.02 ± 0.03 (0–0.05) ND = 1
Reindeer meat ($n = 8$)	2.37 ± 2.28 (0.08–6.94)	1.81 ± 2.88 (0–8.13) ND = 1	0.29 ± 0.50 (0–1.41) ND = 4	0.00 ND = 8
Arctic cod meat ($n = 3$)	6.78 ± 7.18 (0.74–14.71)	0.40 ± 0.39 (0.06–0.83)	0.31 ± 0.37 (0.06–0.74)	0.01 ± 0.02 (0–0.03) ND = 2
Dolly Varden meat ($n = 2$)	2.55 ± 1.99 (1.14–3.96)	0.25 ± 0.07 (0.20–0.30)	0.29 ± 0.08 (0.23–0.34)	0.00 ND = 2
Sculpin meat ($n = 3$)	0.93 ± 0.78 (0.37–1.48)	0.29 ± 0.34 (0.05–0.53)	0.12 ± 0.03 (0.10–0.14)	0.01 ± 0.01 (0–0.02) ND = 1
Halibut meat ($n = 1$)	24.17	0.38	0.16	0.02
Pink salmon meat ($n = 1$)	31.71	6.21	1.3	0.1

Note. Results are presented as the average, standard deviation, range, and number of non-detects (ND).

concentration in reindeer meat (8.4 ppb). HCB analysis had 31 NDs, with average concentrations ranging from ND ppb in snow goose ($n = 2$) to 8.3 ppb in hair seal meat ($n = 1$). The highest individual concentration was also the hair seal meat sample. Several samples had no detectable mirex, with average mirex results ranging from ND to 0.2 ppb. The highest individual sample result was in walrus meat (1 ppb). Bowhead whale meat total PCB concentrations were among the highest, averaging 27.2 ppb. HCB concentrations averaged 0.6 ppb. Eider meat samples ranged from 1.1 to 2.4 ppb.

Organ Samples Table 4 provides PCB and pesticides results obtained from the major organs of seal, whale, walrus, reindeer, and cormorant brain. Skin and liver samples had some of the highest concentrations of total PCB and pesticides among the organ samples. Liver not only had higher PCB levels than other organs, but also had some of the higher concentrations of HCB and DDE. PCB concentrations measured in ringed seal liver ranged from 2.2–13.4 ppb. Intestines also had higher concentrations of total PCB as well as HCB and DDE. Bowhead whale skin had markedly elevated concentrations of PCB, DDE, and HCB. As with the meat samples, the reindeer organs showed the lowest concentrations of PCB and the three pesticides.

Figure 1 illustrates the relative distribution of PCB in the various tissues from ringed seals.

TABLE 4. PCB and Pesticides (ppb, w/wt) in Various Mammalian Organs

Species and sample type	PCBs	p,p'-DDE–85	HCB	Mirex
Bearded seal liver ($n = 3$)	2.25 ± 2.21 (0.32–4.66)	0.97 ± 0.55 (0.27–1.23)	0.04 ± 0.02 (0.02–0.06)	0 ND = 3
Ringed seal liver ($n = 3$)	8.08 ± 5.77 (1.94–13.38)	0.70 ± 0.17 (0.56–0.89)	0.10 ± 0.04 (0.05–0.13)	0.04 ± 0.04 (0–0.08) ND = 1
Ringed seal kidney ($n = 2$)	3.39 ± 2.00 (1.97–4.80)	0.65 ± 0.74 (0.13–1.17)	0.11 ± 0.05 (0.07–0.14)	0.02 ± 0.02 (0–0.03) ND = 1
Ringed seal heart ($n = 2$)	8.24 ± 1.29 (7.33–9.15)	1.59 ± 1.00 (0.88–2.29)	0.08 ± 0.11 (0–0.15) ND = 1	0.05 ± 0.01 (0.04–0.05)
Ringed seal intestine ($n = 2$)	9.62 ± 3.45 (7.18–12.06)	1.40 ± 1.20 (0.55–2.24)	0.31 ± 0.21 (0.16–0.46)	0.02 ± 0.02 (0–0.03) ND = 1
Bowhead skin[a] ($n = 2$)	57.52 ± 40.16 (29.13–85.91)	4.36 ± 2.48 (0.60–4.36)	1.62 ± 0.98 (0.93–2.31)	0.03 ± 0.04 (0–0.06) ND = 1
Walrus liver ($n = 15$)	1.97 ± 3.04 (0.02–10.90)	0.01 ± 0.03 (0–0.09) ND = 7	0.05 ± 0.09 (0–0.29) ND = 7	0.06 ± 0.11 (0–0.34) ND = 6
Walrus heart ($n = 5$)	2.55 ± 4.32 (0.03–10.18)	0.07 ± 0.14 (0–0.32) ND = 1	0.08 ± 0.16 (0–0.36) ND = 2	0.03 ± 0.05 (0–0.11) ND = 2
Walrus intestine ($n = 4$)	7.35 ± 5.66 (0.92–13.76)	0.30 ± 0.37 (0.04–0.28)	0.15 ± 0.24 (0–0.53) ND = 1	0.35 ± 0.53 (0.02–1.14)
Reindeer liver ($n = 4$)	0.18 ± 0.21 (0–0.48) ND = 1	0.96 ± 0.28 (0.55–1.21)	0 ND = 4	0 ND = 4
Reindeer kidney ($n = 4$)	0.03 ± 0.04 (0–0.08) ND = 2	0.63 ± 0.17 (0.48–0.86)	0 ND = 4	0 ND = 4
Reindeer heart ($n = 2$)	0.06 ± 0.04 (0.03–0.08)	0.56 ± 0.20 (0.42–0.70)	0 ND = 2	0 ND = 2

Note. Results are presented as the average, standard deviation, range, and number of non-detects (ND).

[a] Bowhead was the only species with skin samples analyzed.

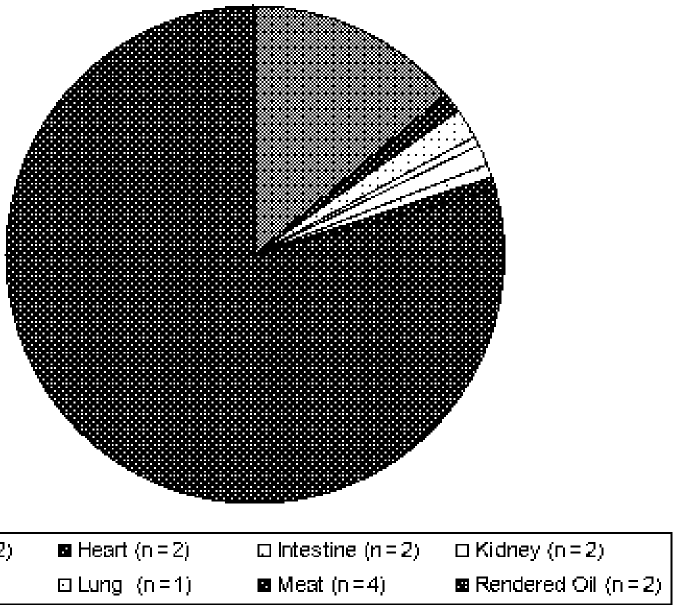

FIGURE 1. Distribution of percentage of total PCB in different types of ringed seal samples (color figure available online).

The highest concentration was found in the rendered oil, which is significantly higher than that in blubber. The other organs have relatively low concentrations, consistent with the fact that their lipid content is much lower.

Bird, Plants, Berries, and Marine Invertebrate Samples Table 5 presents PCB and pesticide levels in other samples, although for most only a few were analyzed. The glaucous gull meat (n = 1) had one of the highest total PCB concentrations, 78.1 ppb. Arctic loon and pelagic cormorant adipose tissue and brain were also high.

Fourteen plant samples of several species were analyzed. Results ranged from 0.01 to 1.3 ppb for PCB, ND to 0.06 ppb for DDE, 0.01 to 0.27 ppb for HCB, and 0 to 0.06 ppb for mirex. The prepared roseroot had more than twofold greater PCB concentration compared to the second highest plant sample, salmon berries, and contained an almost 10-fold higher concentration of DDE than the other samples. The higher concentrations in the prepared plant samples are likely a result of contaminant contributions from the cooking oils used to prepare them.

While there were relatively few samples of invertebrates and seaweed analyzed, PCB concentrations were low, ranging from 0.4 to 3.7 ppb. Uupa (sea peaches, a tunicate species) were the only invertebrate samples found to contain mirex, with an average concentration of 0.03 ppb, and had the highest concentration of PCB (2.62 ppb) and HCB (0.03 ppb) of all the invertebrate and seaweed samples. Seaweed had the second highest concentration of PCB among the invertebrate and seaweed samples (2.4 ppb), yet the second lowest concentration of HCB (0.03 ppb), equal with the concentration in the shelled clam.

Metals

Blubber, Adipose Tissue, Rendered Oil, and Alumguk Results for blubber, adipose tissue, and alumguk and rendered oil samples are given in Table 6. Blubber, adipose tissue, and rendered oil were uniformly low in Cu, Zn, Se, Ca, and Pb, with the vast majority of measurements for these elements below the limit of detection. In contrast, As was found in all blubber and rendered oil samples with mean concentrations of 2.3 (n = 39) and 2.7 μg/g (n = 14), respectively. Arsenic was also present in about half of the adipose tissue samples with a mean of 0.43 μg/g (n = 8).

TABLE 5. PCB and Pesticides in Birds, Plants, Berries, and Other Seafood

Species and sample type	PCBs	p,p'-DDE+85	HCB	Mirex
Birds				
Arctic loon adipose tissue ($n = 1$)	87.3	9.81	10.78	1.09
Pelagic cormorant/loon liver/brain ($n = 3$	10.10 ± 13.23 (0.29–5.19)	0.95 ± 0.25 (0.66–1.12)	0.31 ± 0.28 (0.03–0.58	0.02 ± 0.03 (0–0.05) ND = 1
Arctic ($n = 1$)	5.53	0.72	0.59	0.06
Common eider skin ($n = 1$)	0.78	0.17	0.13	0.01
Glaucous gull meat ($n = 1$)	78.05	11.29	34.21	1.9
Northern pintail skin ($n = 1$)	12.13	4.24	0.83	3.14
Arctic loon meat ($n = 3$)	4.70 ± 4.04 (0.89–8.93)	0.77 ± 0.46 (0.39–1.31)	0.63 ± 0.34 (0.32–1.00)	0.08 ± 0.08 (0–0.16) ND = 1
Common eider meat ($n = 2$)	1.78 ± 0.90 (1.14–2.41)	0.26 ± 0.12 (0.17–0.34)	0.43 ± 0.21 (0.28–0.57)	0.07 ± 0.08 (0.01–0.13)
Snow goose meat ($n = 2$)	0.06 ± 0.07 (0.01–0.11)	0.04 ± 0.06 (0–0.08) ND = 1	0.00 ND = 2	0.00 ND = 2
Pelagic cormorant adipose tissue ($n = 1$)	61.45	10.57	10.26	0 ND = 1
Pelagic cormorant liver ($n = 1$)	1.4	0.91	0.91	0 ND = 1
Plants and berries				
Roseroot greens ($n = 2$)	0.68 (0.21–1.15)	0.05 (0.05–0.05)	0.02 (0.01–0.02)	0.06 (0–0.13) ND = 1
Salmon berries ($n = 1$)	1.33 ± 1.60 (0.04–3.65)	0.01 ± 0.01 (0–0.02) ND = 1	0.03 ± 0.07 (0–0.13) ND = 3 = 1	0.03 ± 0.03 (0.01–0.07)
Prepared plant mix ($n = 1$)	0.73	0.02	0.04	0
Prepared roseroot ($n = 1$)	3.73	0.06	0.27	0 ND = 1
Greens ($n = 1$)	0.01	0 ND = 1	0.03	0 ND = 1
Fireweed greens ($n = 3$)	0.39 ± 0.40 (0.11–0.84)	0.01 (0–0.03) ND = 2	0.01 (0.01–0.01)	0.03 ± 0.03 (0–0.06) ND = 1
Crowberries ($n = 1$)	0.29	0 ND = 1	0.01	0 ND = 1
Other seafood				
Blue king crab meat ($n = 3$)	1.67 ± 2.23 (0.18–4.18)	0.14 ± 0.15 (0.03–0.31)	0.02 ± 0.03 (0–0.05) ND = 1	0.00 ND = 3
Sea peaches/tupa ($n = 3$)	2.62 ± 2.17 (0.41–4.74)	0.05 ± 0.06 (0–0.12) ND = 1	0.16 ± 0.11 (0.08–0.28)	0.03 ± 0.03 (0–0.06) ND = 1
Seaweed (kelp) ($n = 1$)	2.38	0 ND = 1	0.03	0 ND = 1
Shelled clam ($n = 1$)	0.89	0.03	0.03	0 ND = 1
Razor clam (imaamaq) ($n = 2$)	0.75 (0.35–1.14)	0.10 (0–0.2) ND = 1	0.01 (0–0.01) ND = 1	0 ND = 2
Sea cucumber (tukugnak) $n = 1$	0.52	0.02	0.14	0 ND = 1
Blue king crab eggs ($n = 6$)	1.11 ± 0.43 (0.93–1.98)	0.29 ± 0.13 (0.13–0.49)	0.38 ± 0.13 (0.19–0.59)	0.01 ± 0.01 (0–0.03) ND = 3
Blue king crab meat ($n = 3$)	1.67 ± 2.23 (0.18–4.18)	0.14 ± 0.15 (0.03–0.31)	0.02 ± 0.03 (0–0.05) ND = 1	0 ND = 3

TABLE 6. Trace Metals Results, µg/g (w/w), in Blubber, Adipose Tissue, Alumguk,[a] and Rendered Oils

Species and sample type	Cu	Zn	As	Se	Cd	Hg	Pb
Bearded seal blubber (n = 11)	1.6 ± 1.1 (<1.5–4.0) ND = 9	14 ± 16 (<13–61) ND = 10	2.1 ± 0.8 (0.5–3.0)	0.7 ± 0.3 (<0.9–1.2) ND = 10	<0.06 ND = 11	0.074 ± 0.073 (<0.002–0.190) ND = 3	<0.15 ND = 11
Spotted seal blubber (n = 3)	<1.5 ND = 3	<13 ND = 3	2.4 ± 1.2 (1.4–3.8)	<0.9 ND = 3	<0.06 ND = 3	0.005 ± 0.003 (<0.002–0.007) ND = 1	<0.15 ND = 3
Walrus blubber (n = 24)	<1.5 ND = 24	<13 ND = 24	2.6 ± 1.3 (0.7–4.9)	<0.9 ND = 24	<0.06 ND = 24	0.089 ± 0.065 (<0.002–0.158) ND = 11	<0.15 ND = 24
Polar bear adipose tissue (n = 3)	<1.5 ND = 3	<13 ND = 3	0.13 ± 0.07 (<0.06–0.17) ND = 1	<0.9 ND = 3	<0.06 ND = 3	0.055 ± 0.093 (<0.002–0.162) ND = 2	<0.15 ND = 3
Reindeer alumguk (n = 2)	<1.5 ND = 2	15 ± 3 (13–17)	<0.06 ND = 2	<0.9 ND = 2	<0.06 ND = 2	0.195 ± 0.003 (0.193–0.197)	0.35 ± 0.11 (0.27–0.43)
Reindeer adipose tissue (n = 2)	<1.5 ND = 2	<13 ND = 2	<0.06 ND = 2	<0.9 ND = 2	<0.06 ND = 2	0.157 ± 0.006 (0.152–0.161)	<0.15 ND = 2
Bearded seal rendered oil (n = 3)	<1.5 ND = 2	<13 ND = 3	3.0 ± 0.1 (2.9–3.1)	<0.9 ND = 2	<0.06 ND = 3	0.131 ± 0.005 (0.127–0.134)	<0.15 ND = 2
Spotted seal rendered oil (n = 2)	<1.5 ND = 2	<13 ND = 2	1.74 ± 0.04 (1.7–1.8)	<0.9 ND = 2	<0.06 ND = −2	0.130 ± 0.013 (0.121–0.139)	<0.15 ND = 2
Bowhead whale rendered oil (n = 2)	<1.5 ND = 2	<13 ND = 2	1.1 ± 0.1 (1.0–1.2)	<0.9 ND = 2	<0.06 ND = 2	0.141 ± 0.004 (0.138–0.143)	<0.15 ND = 2
Walrus oil (n = 7)	<1.5 ND = 7	<13 ND = 7	3.9 ± 1.7 (1.4–5.5)	<0.9 ND = 7	<0.06 ND = 7	0.059 ± 0.072 (<0.002–0.140) ND = 2	<0.15 ND = 7

Note. Data are presented as average, standard deviation, range, and number of non-detects (ND).

[a]Alumguk is Yupik for skin plus adipose tissue.

Mercury was detectable in more than half of the blubber, oil, and adipose tissue samples with a mean of 0.11 µg/g. Alumguk (reindeer skin with adipose tissue; n = 2) showed no detectable As and elevated mean Zn (15 µg/g), Hg (0.2 µg/g), and Pb (0.35 µg/g), relative to the blubber, adipose tissue, and oil samples. A sample of minke whale skin had 64 µg/g Zn, suggesting Zn in Alumguk is in the skin. No mungtak (blubber and skin of the bowhead whale) samples were measured for metals. In general, As and Hg concentrations of seal and walrus rendered oils were equal to or quantitatively elevated with respect to concentrations in corresponding blubber samples.

Meat Results for meat samples are given in Table 7. Copper concentrations in meat were higher in reindeer (4.7 ± 3.1, n = 8) and birds (5.8 ± 1.7, n = 12), than in walrus (1.4 ± 0.4, n = 30) and seals, whales, and polar bears (1.6 ± 1.0, n = 31). Average Zn concentrations were lowest in birds (16.7 ± 5.5 µg/g) and highest in walrus (55 ± 20 µg/g), with considerable overlap among individuals from all species.

In marine mammals, As meat concentrations were considerably lower than in blubber. Mean meat As for walrus, seal, and whale was approximately 0.35 µg/g (n = 58), with wide variation among individuals. Birds had lower mean As levels (0.27 ± 0.25) than marine mammals. All polar bear and reindeer samples were at or below the detection limit for As. Selenium was detected in nearly all walrus samples (2.7 ± 1.9) but was not detected in most other meat samples. Cadmium was below detection in most meat samples, with notable exceptions in king eider (0.82 µg/g) and walrus (0.42 µg/g). For Hg, walrus (n = 30) ranged from below detection to 0.19 µg/g. The highest meat Hg concentration occurred in Arctic loon (0.49 ± 0.2 µg/g; n = 3). Lead was near or below detection in the vast majority of samples.

Trace Metals in Organs, Fish, and Invertebrates Metal results for liver, kidneys,

TABLE 7. Trace Metal Results, μg/g (w/w), in Meat Samples

Species and sample type	Cu	Zn	As	Se	Cd	Hg	Pb
Bowhead whale (n = 4)	3.6 ND = 3	38 ± 23 (<13–56) ND = 2	0.51 ± 0.38 (0.07–0.90)	0.78 ± 0.29 (<0.9–1.27) ND = 3	<0.06 ND = 4	0.032 ± 0.019 (<0.002–0.047) ND = 1	<0.15 ND = 4
Arctic loon (n = 3)	4.0 ± 1.3 (3.2–5.1)	14 ± 1 (13–15)	0.43 ± 0.06 (0.37–0.47)	<0.9 ND = 3	<0.06 ND = 3	0.491 ± 0.023 (0.467–0.513)	<0.15 ND = 3
Sandhill crane (n = 2)	6.3 ± 0.2 (6.2–6.4)	33.9 ± 0.3 (33.7–34.1)	0.43 ± 0.06 (0.37–0.47)	<0.9 ND = 2	<0.06 ND = 2	0.148 ± 0.005 (0.144–0.151)	<0.15 ND = 3
Snow goose (n = 2)	5.9 ± 0.1 (5.9–6.1)	12 ± 5 (<13–16) ND = 1	<0.06 ND = 2	<0.9 ND = 2	0.15 ± 0.15 (<0.04–0.25) ND = 1	0.149 ± 0.001 (0.148–0.149)	<0.15 ND = 2
Polar bear (n = 3)	1.6 ± 0.5 (<1.5–2.1) ND = 1	45 ± 11 (33–53)	0.05 ± 0.02 (<0.06–0.08) ND = 1	<0.9 ND = 2	<0.06 ND = 3	0.032 ± 0.0029 (0.011–0.065)	0.16 ± 0.09 (<0.15–0.26) ND = 2
Reindeer (n = 8)	4.7 ± 3.1 (<1.5–9.6) ND = 1	33 ± 17 (<13–61) ND 1	<0.06 ND = 8	<0.9 ND = 8	<0.06 ND = 8	0.003 ± 0.002 (<0.002–0.006) ND = 5	<0.15 ND = 8
Walrus (n = 30)	1.4 ± 0.4 (<1.5–2.4) ND = 18	55 ± 19 (15–93)	0.32 ± 0.35 (<0.06–1.71) ND = 1	2.7 ± 1.9 (<0.9–9.3) ND = 4	0.08 ± 0.09 (<0.06–0.42) ND = 23	0.004 ± 0.007 (<0.002–0.019) ND = 20	<0.15 ND = 30
Spotted seal (n = 4)	<1.5 ND = 4	27 ± 13 (<13–41) ND = 1	1.1 ± 1.6 (0.19–3.4)	0.8 ± 0.3 (<0.9–1.3) ND = 3	<0.06 ND = 2	0.021 ± 0.017 (<0.002–0.037) ND = 3	<0.15 ND = 2
Bearded seal (n = 17)	1.5 ± 0.7 (<1.5–3.1) ND = 9	52 ± 34 (14–136)	0.25 ± 0.15 (0.11–0.63)	0.90 ± 0.64 (<0.9–3.16) ND = 13	0.07 ± 0.05 (<0.06–0.20) ND = 13	0.052 ± 0.077 (<0.002–0.337) ND = 5	<0.15 ND = 2

Note. Data are presented as average, standard deviation, range, and number of non-detects (ND).

TABLE 8. Trace Metals (μg/g, w/w) in Organs, Fish, and Shellfish

Species and sample type	Cu	Zn	As	Se	Cd	Hg	Pb
Bearded seal liver (n = 2)	22 ± 8 (16–27)	43 ± 7 (39–48)	0.34 ± 0.16 (0.23–0.45)	3.9 ± 3.5 (1.4–6.4)	4.2 ± 5.0 (0.7–7.8)	3.26 ± 3.67 (0.67–5.86)	<0.15 ND = 2
Walrus liver (n = 14)	16 ± 11 (<1.5–38) ND = 1	34 ± 10 (<13–49) ND = 1	0.27 ± 0.06 (0.19–0.39)	1.6 ± 1.0 (<0.9–4.0) ND = 3	3.8 ± 2.0 (<0.06–7.1) ND = 2	0.511 ± 0.409 (0.79–1.350)	0.13 ± 0.06 (<0.15–0.33) ND = 13
Reindeer liver (n = 4)	16 ± 6 (7–27)	23 ± 2 (20–25)	<0.06 ND 4	0.9 ± 0.2 (0.6–1.2)	0.35 ± 0.06 (0.27–0.40)	0.350 ± 0.255 (<0.002–0.615) ND = 1	0.29 ± 0.08 (0.22–0.38)
Walrus heart (n = 5)	3.6 ± 1.0 (2.5–4.9)	29 ± 13 (18–51)	0.19 ± 0.05 (0.11–0.24)	2.2 ± 0.6 (1.1–2.7)	<0.15 ND = 5	0.045 ± 0.078 (<0.002–0.184) ND = 1	<0.15 ND = 5
Reindeer heart (n = 2)	3.6 ± 0.1 (3.6–3.7)	12 ± 4 (<13–15) ND = 1	<0.06 ND 2	<0.9 ND = 2	<0.15 ND = 2	0.008 ± 0.001 (0.007–0.008)	<0.15 ND = 2
Reindeer kidneys (n = 4)	4.9 ± 0.5 (4.1–5.2)	21 ± 2 (19–23)	<0.06 ND 4	1.4 ± 0.3 (1.1–1.7)	1.7 ± 0.4 (1.4–2.3)	1.43 ± 0.80 (0.67–2.20)	0.17 ± 0.13 (<0.15–0.36) ND = 3
Halibut muscle (n = 2)	<1.5 ND = 2	<13 ND = 2	2.08 ± 0.00 (2.08–2.08)	<0.9 ND = 2	<0.06 ND = 2	0.496 ± 0.017 (0.484–0.508)	<0.15 ND = 2
Crab eggs (n = 7)	11 ± 6 (<1.5–21) ND = 1	35 ± 12 (<13–46) ND = 1	5.70 ± 2.62 (<0.06–7.56) ND = 1	4.8 ± 2.1 (<0.9–7.2) ND = 1	0.29 ± 0.26 (0.14–0.87)	0.024 ± 0.052 (<0.002–0.140) ND = 5	<0.15 ND = 7

Note. Data are presented as average, standard deviation, range, and number of non-detects (ND).

and heart are given in Table 8. Compared to other tissues, liver and kidneys had the highest concentrations of Hg and Cd. Polar bear liver contained the highest Hg level (6.35 μg/g; $n = 1$), followed by seal (2.83 ± 2.70; $n = 3$), walrus (0.58 ± 0.39; $n = 14$), arctic loon (0.55; $n = 1$), and reindeer (0.47 ± 0.13; $n = 4$). Liver Cd was highest in walrus (4.2 ± 2), followed by seal (3.3 ± 3.9), polar bear (2), and pelagic cormorant (0.32; $n = 1$). Reindeer liver also had consistently elevated Pb levels (0.29 ± 0.8), whereas it was not detected in other liver samples. Kidneys, which were only collected from reindeer, showed mean Hg and Cd concentrations of 1.4 ± 0.8 and 1.7 ± 0.4 μg/g, respectively ($n = 4$). Heart tissue of walrus, reindeer, and polar bear displayed similar concentrations to those of average meats for those species. Results from halibut and crab eggs are also shown in Table 8. Halibut muscle contained measurable As (2.08 ± 0.00 μg/g; $n = 2$) and elevated Hg (0.496 ± 0.017), a level that is well above the U.S. EPA guideline of 0.05 μg/g for unlimited consumption. One sample each of salmon berries and of plant greens was analyzed. No metals were detected in either sample.

DISCUSSION

Organochlorines in Yupik Foodstuffs

These results indicate that there are elevated levels of PCB (and to a lesser degree pesticides) in the blubber and rendered oils of all of the marine mammals analyzed, compared to the other sample types. Concentrations of PCB and pesticides were lower (1–10%) in meat samples than in blubber or rendered oils. This is to be expected since all of our results are reported as wet weight values (not lipid adjusted) and these substances concentrate in adipose tissue. Organs have intermediate levels of contaminants, consistent with having a higher lipid content than meat. Because consumption of marine mammal blubber in various forms is a vital component of the diet of the residents of St. Lawrence Island, our observations provide strong evidence that this is the primary source of the elevated blood levels of PCB and pesticides reported in this population (Carpenter et al. 2005). Bjerregaard et al. (2001) also showed a direct correlation between marine diets and OC exposure in various Inuit populations in Greenland.

It was anticipated that animals feeding lower on the food chain would have lesser concentrations of contaminants. The relatively high levels in bowhead whale blubber (317.6 ppb) and rendered oil (354.1 ppb) are remarkable since bowheads are baleen animals that feed low on the food chain. However, these levels are comparable to that reported by Hoekstra et al. (2005) in Barrow bowhead blubber (345 ppb wet weight), although the levels found in bowhead meat (27.2 ppb) are higher than those reported by these investigators (1.8 ppb). O'Hara et al. (1999) noted lipid-adjusted values as the sum of 17 PCB congeners in bowhead blubber from Barrow to be 458.5 ppb. Muir et al. (1999) reported higher concentrations (935 ppb wet weight) in bowhead blubber from the eastern Canadian Arctic. The Arctic beluga whales in general have an order of magnitude higher concentration of PCB. However, they are toothed whales that feed higher on the food chain (Becker 2000; AMAP 2009). Although bowhead whales feed lower on the food chain, the bowhead whale blubber, rendered oil, and skin samples contained some of the highest concentrations of PCB found in our study among all the samples collected. Bowhead whales' primary food source is plankton (Rogachev et al. 2008). Plankton and zooplankton were demonstrated to take up PCB (Ko and Baker 1995; Wang et al. 1998; Harding et al. 1997; Braune et al. 2005). These invertebrates are considered to act as a sink for PCB, by bringing them to the ocean floor as they settle (Berglund et al. 2001). The bowhead mungtak (Yupik for skin plus blubber) was also high in PCB and pesticides, but lower than in bowhead blubber. The average mungtak PCB concentration was 142.61 ppb, comparable to concentrations previously observed in the region (Hoekstra et al. 2005). One factor that may explain why bowheads in our study would have higher concentrations than seals or walrus may be that they live longer and therefore have a longer time to accumulate lipophilic compounds. There was relatively more variation in PCB levels among the bowhead samples than those of the other mammals, probably reflecting the fact that it was not possible to control for age or gender.

Since walrus feed primarily on shellfish it is reasonable that they would have lower levels than seals, who primarily feed on fish. The mean values for walrus blubber in this study (33.9 ppb, $n = 29$) are lower, however, than those reported in a previous study (Seagers and Garlich-Miller 2001) of 27 Bering Sea walrus (450 ppb). The difference may simply reflect the age of the mammals sampled. Gender is also an important variable for which there was no control in our study. Females, as is the case in humans, transfer a significant amount of their body burden to offspring through gestation and lactation, and thus levels in males are in general higher than those in females. Our values are also lower than those reported from the Canadian Arctic from the 1980s by Muir et al. (1999), and from Greenland walrus blubber by Muir et al. (2000).

Bearded seal blubber averaged 106 ppb in total PCB, comparable to other bearded seal PCB concentrations in the area (Hoekstra et al. 2005; Krahn et al. 1997). Ringed seal total PCB concentrations were lower than those reported from other areas of the Arctic (Muir et al. 1999; Fisk et al. 2002; Cleeman et al 2000; Weis and Muir 1997; Mallory et al. 2005). This is to be expected as seals from Alaska were found to generally contain lower OC concentrations than other seals in central and eastern Canada (Braune et al. 2005), and levels are even higher in the European Arctic (Vorkamp et al. 2008; Braune et al. 2005).

As expected, the highest levels found were in polar bears, which primarily eat seal blubber, while the lowest levels were in reindeer, which are herbivores. The polar bear meat with adipose tissue sample also had a high concentration of PCB (309.2 ppb). Verreault et al. (2005) observed that PCB levels were higher in Alaska polar bears than others from North America. Data showed lipid-adjusted levels of 2980 and 2838 ppb in males and females, respectively, but these values can not be compared with wet weight values.

Levels of PCB in the few fish samples were relatively low, but were higher in salmon and halibut than in cod and Dolly Varden. Both salmon and halibut are predacious and relatively fatty fish (Muir et al. 1999). The levels in pink salmon from St. Lawrence Island were higher than those reported for other wild Alaska pink salmon (Hites et al. 2004), but much lower than those for salmon and halibut obtained from the contaminated Adak Island in the Aleutians of Alaska (Hardell et al. 2010). Crabs, clams, and sea peaches displayed low levels of all contaminants. Plant samples also contained low concentrations.

There was large variation in the levels found in bird samples, as expected based on their diet. Glaucous gull meat was relatively high (78.1 ppb), but levels in snow goose and eider meat were low. These findings are consistent with previous studies (Braune et al. 2005) and reflect the fact that the gulls are scavengers and foragers. The few bird adipose tissue samples contained levels similar to those in the glaucous gull meat, but were much higher than levels in other bird meat samples. Skin, brain, and other bird organ samples were not markedly elevated.

The pesticides in blubber and adipose tissue were found at much lower concentrations than PCB in all samples except for reindeer, but followed the same general pattern of being higher in rendered oils, and higher in polar bear than marine mammals. Concentrations in walrus were less than in seals, and DDE was present at higher concentrations than HCB, while mirex was lowest. The walrus mungnguna (Yupik term for walrus skin with blubber) contained lower concentrations of PCB, HCB, and mirex than the walrus blubber, but double the concentration of DDE. There was a large variation in pesticide concentrations in bowhead samples. Since samples are not separately based on age and gender, these factors may play a role in the large variation.

Metals in Yupik Foodstuffs

Overall, metal concentrations varied less than OC measurements. This may be because the metal concentrations were higher and analyzed with higher detection limits than OC, limiting their detectable range compared to the OC. Mercury was the most detected metal of those analyzed, with concentrations less than 0.52 ppm except for the bearded seal liver and reindeer kidney. This concentration in liver is lower than the average found in bearded seals in Barrow, AK (Dehn et al. 2006). Ringed seal meat Hg concentrations are comparable to those found by Woshner et al. (2001), but the Hg measured in the blubber sample was higher than the average in the Woshner et al. (2001) study.

While the various forms of Hg were not distinguished, it may be assumed that the primary form of Hg in marine mammals is the fat soluble methylmercury (MeHg), since the highest concentrations were observed in the adipose tissue samples. The Joint FAO/WHO Expert Committee on Food Additives established a tolerable intake of 1.6 μg/kg body weight per week for MeHg to protect developing fetuses from neurological effects (JECFA 2004). The concentration in bearded seal liver (3.26 ppm) is sufficiently high to pose health concerns, depending upon level of intake. Zinc and Se were found to affect Hg distribution and protect against its toxicity (Rooney 2007). The Hg in reindeer kidney is likely inorganic Hg, which does not cross the blood–brain barrier.

Arsenic was the second most detected metal, found in 22 of 30 samples analyzed. This analysis only measured total As, but since some of the highest concentrations were observed in the oil and blubber samples it may be assumed that this is primarily organoarsenic. Organoarsenic, especially arsenobetaine, is the form of As most commonly found in fish and seafood. Unlike inorganic As, arsenobetaine is known not to be carcinogenic (Sabbioni et al. 1991). Organoarsenic is also a common component used in military chemicals and in herbicides (Leermakers et al. 2006).

Cadmium concentrations were highest in the organ samples, and were found to increase up the food web. The samples with high levels of Cd correlate with samples that are high in Zn. This may help to reduce Cd-induced toxicity by forming Cd-metallothionein complexes (Ohta and Cherian 1991; 1995) and also via non-metallothionein mechanisms (Mishima et al. 1997). The ringed seal had substantially lower levels than those found in West Greenland ringed seals (Sonne et al. 2009). Bearded seal liver had levels lower than the average noted in Barrow, AK, and ringed seal liver showed levels lower than the average measured in Homan, Canada (Dehn et al. 2006). Polar bear and ringed seal Se concentrations were lower than others observed in Alaska (Woshner et al. 2001).

Human Dietary Implications and Recommendations

The U.S. Environmental Protection Agency (EPA) has published guidelines for sports fish consumption, to protect from cancer and noncancer risk from ingestion of PCB and pesticides (U.S. EPA 2000). These advisories provide recommended frequency of consumption based on contaminant levels in the fish. For PCB the U.S. EPA guidelines indicate a threshold for unlimited fish consumption based on cancer risk at 1.5 ppb and for noncancer risk at 5.9 ppb. Based on these guidelines, even sea plant samples trigger a reduced consumption advisory to reduce cancer risk, with seaweed falling into the 16 meals/mo guideline and sea peaches at 12 meals/mo. Reindeer meat, adipose tissue, and organs were among the lowest in PCB, with meat and organ samples giving an unlimited consumption advisory. Levels of PCB exceeding 100 ppb trigger an advisory recommendation of "no consumption," based on cancer risk. PCB levels in the rendered oil samples of seals (ringed, spotted, and bearded seal), walrus, and bowhead whale greatly exceeded this threshold in our study, as did blubber samples of bearded seal, spotted seal, bowhead whale (blubber and mungtak), and polar bear. While the nutritional and cultural benefits of traditional food are well described (Suk et al. 2004), and omega-3 fatty acids found in marine-based traditional foods may confer certain health benefits, although they are not protective against cancer (MacLean et al. 2006), these results clearly indicate that the diet of residents of St. Lawrence Island would be expected to significantly increase risk of cancer.

Implications for Marine Health

It is important to acknowledge that the health of Arctic marine species is also in jeopardy from these contaminants (Letcher et al. 2010). Seal populations with weakened immune systems resulting from exposure to OC such as PCB have been decimated by diseases that they would normally be able to fight off when their immune systems are not compromised (de Swart et al. 1996) and are linked to increases in the incidences of other diseases (Bredhult et al. 2008). Glaucous gull chicks also exhibited weakened immune systems as a result of exposure to PCB and other persistent organochlorine pollutants (POP) (Sagerup et al. 2009), while polar bears were shown to demonstrate a weakened humoral immunity following exposure to PCB (Lie et al. 2004).

When interpreting these results it is crucial to consider the impacts of climate change and global warming. It has been well documented that as temperatures rise, so does the potential for pollutant mobility from local sources and global migration (Ma et al. 2004), with the Arctic region becoming an increasing sink for pollutants (Macdonald et al. 2005). Therefore, it may be assumed that the concentrations found in the Arctic marine and wildlife will also rise. Climate change also results in severe stress to wildlife, compromising access to food sources and shelter, and also compromising their immune systems, in turn increasing their susceptibility to contaminants (Letchter et al. 2010; Ford and Pearce 2010). This might lead to a decrease in the availability of subsistence foods to the Yupik people, with further adverse effects to their culture and lifestyle. The shrinking sea ice that the marine mammals depend on will decrease the availability of these foods further.

CONCLUSIONS

This study of the dietary sources of the St. Lawrence Island Yupik provides a snapshot not just of the contaminant exposure in their local subsistence diet, but also of the current contaminant exposure of the wildlife in this area. The results demonstrate levels of PCB in traditional foods at concentrations that pose clear adverse effects on human health. Levels of chlorinated pesticides and organic mercury are also sufficiently high to be a reason for concern. Within the context of traditional foods, the lowest contaminants are founds in plants, reindeer meat, and the meat of marine species. Levels of PCB in the blubber/adipose tissue all trigger severe consumption restriction advisories when one applies U.S. EPA guidance for fish, but these guidelines need to be balanced by other considerations including culture and tradition. The primary goal is to preserve the culture and lifestyle associated with traditional foods, but at the same time provide the community members necessary information to make their own informed decisions. However, it is imperative to take measures to reduce exposures where possible and eliminate sources of PCB, chlorinated pesticides, and metals through state, national, and international policy actions.

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