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Polychlorinated dibenzodioxins, dibenzofurans and non-ortho substituted polychlorinated biphenyls in caribou (*Rangifer tarandus*) from the Canadian Arctic

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Abstract

The presence of contaminants in the Arctic environment has raised concerns regarding levels in wildlife and possible effects on the health of wildlife populations. In addition, contaminants in wild foods are of particular concern to those people who rely on these foodstuffs for a significant portion of their diet. Among the most toxic contaminants found in the environment are the polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and non-ortho substituted polychlorinated biphenyls (NOPCBs). Few data exist documenting the levels of these compounds in Arctic terrestrial wildlife. In 1993, caribou samples were obtained from three herds in the Yukon Territory (Finlayson, Tay and Bonnet Plume) and from four herds in the Northwest Territories (Bathurst, Southampton Island, Cape Dorset and Lake Harbour). High resolution gas-chromatography/mass spectrometry was used to measure contaminant concentrations. Wet weight concentrations of PCDDs, PCDFs and NOPCBs were greater in fat tissue than in muscle and liver, however, concentrations in all tissues were extremely low. Lipid normalized concentrations were greater in muscle and liver than in fat, indicating that equilibrium partitioning is not the only process regulating tissue concentrations of these contaminants. There were no significant relationships between concentrations of individual congeners and caribou age. Concentrations of the non-ortho substituted PCBs #126 and #169 were greater in caribou from the eastern Arctic, although levels in all herds were low. 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) toxic equivalents were also low in tissues from all herds. The presence of these compounds in the Arctic can likely be attributed to long-range atmospheric transport. The levels documented in this study are some of the lowest ever reported in wildlife and are unlikely to pose a threat to caribou or their human consumers.

Keywords: Caribou (*Rangifer tarandus*); Dibenzodioxins; Dibenzofurans; Polychlorinated biphenyls; Canadian Arctic

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1. Introduction

Previous studies have documented the broad distribution of persistent contaminants in the Arctic (Barrie et al., 1992; Muir et al., 1992; Thomas et al., 1992). The presence of these compounds in the environment is a concern because of their potential for adversely affecting the health of wildlife and human populations. Elevated levels of the heavy metal cadmium have been found in caribou kidney and liver from Québec, the Yukon and Northwest Territories (Crête et al., 1989; Gamberg and Scheuhammer, 1994). Organochlorine contaminants are also a concern because they accumulate in fat and are transferred through food webs. Therefore, organisms at the top of the food web, such as humans, are often exposed to the highest concentrations of these contaminants. Existing data indicate that organochlorine concentrations in caribou from the Canadian Arctic are low (Thomas et al., 1992; Gamberg, 1993; Elkin and Bethke, 1995). However, there has been no systematic attempt to quantify concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs) and non-ortho substituted polychlorinated biphenyls (NOPCBs) in caribou from across the Canadian North. These compounds are of particular interest because they are some of the most toxic chemicals found in the environment. The mechanism by which they exert their toxicity is similar, hence, they may act in an additive fashion to exacerbate their impact on wildlife and humans (Safe, 1990). Therefore, to fully evaluate the potential toxicity to wildlife, all three groups should be examined. This study investigated the concentrations of a wide variety of PCDD, PCDF and NOPCB congeners in female caribou from the Yukon and Northwest Territories (NWT). Geographic differences in contaminant concentrations were assessed, in addition to the influence of age on contaminant accumulation.

2. Materials and methods

2.1. Sample collection

In March 1993, 20 female caribou (*Rangifer tarandus*) were shot from each of the Finlayson, Tay and Bonnet Plume herds (Fig. 1). Liver, muscle and subcutaneous back or kidney fat samples were taken from each animal within 1 h of death. Kidney fat was collected only when the animals did not have sufficient subcutaneous fat reserves for a sample to be obtained. Approximately 500 g of liver, gastrocnemius muscle and fat were stored in individual hexane and acetone-rinsed glass jars sealed with teflon caps. All samples were stored at −20°C. Age determinations were made for each animal by examining an incisor using the tooth cementum technique.

Fig. 1. Location of caribou herds: 1, Finlayson; 2, Tay; 3, Bonnet Plume; 4, Bathurst; 5, Southampton; 6, Cape Dorset; 7, Lake Harbour.

From the four Northwest Territories herds (Fig. 1), subcutaneous back fat samples had been collected previously as part of a study examining levels of other contaminants in caribou (Elkin and Bethke, 1995). These collections had been made on the following dates: Southampton Island (November 1991), Cape Dorset and Lake Harbour (April 1992), Bathurst (July and September, 1992). These samples had been stored at −20°C after collection. Only females were analyzed to facilitate comparison with the Yukon caribou.

2.2. Sample processing

Subcutaneous fat samples were analyzed in this study so that comparisons could be made among the Yukon and NWT herds. Fat samples were analyzed first because hydrophobic compounds, such as PCDDs, PCDFs and NOPCBs are stored in lipid. If these contaminants are not found in fat

tissue then their wet weight concentrations in liver or muscle would be lower because these tissues contain a much lower proportion of lipid. Fat samples from the Finlayson herd were analyzed first to determine if there was a relationship between age and PCDD/PCDF levels. Individual samples were pooled according to age and five separate age pools resulted: age 2 ($n = 3$), age 3 ($n = 2$), age 4 ($n = 6$), age 5 ($n = 6$) and age 6 ($n = 2$). One caribou, aged 10, was not included in the pools and was, therefore, not analyzed. As levels of all compounds were extremely low in the Finlayson fat samples, only one liver and one muscle sample were analyzed. The sample analyzed (age 3 pool) was selected at random. Contaminant concentrations were compared in fat, liver and muscle of the age 3 pool to assess the tissue distribution of the contaminants in the Finlayson caribou.

2.3. Chemical analysis

Sample extraction. After grinding the liver and muscle samples with anhydrous sodium sulphate, neutral extraction of lipids was achieved using 1:1 dichloromethane/hexane. For the fat samples, lipid extraction was achieved using the following method. Fat samples (10–15 g) were rendered in a homogenizer (Sorvall Omni-mixer 17105) in three consecutive steps with 50% dichloromethane/hexane (once with 100 ml for 20 min, and twice with 50 ml for 10 min). After each rendering, the extract was decanted through anhydrous sodium sulphate retaining most of the connective tissue in the homogenizer. The sodium sulphate column was rinsed with 100 ml of 50% dichloromethane/hexane. The volume of the combined lipid extracts was reduced in preparation for gel permeation chromatography (GPC) clean-up. Lipids and biogenic compounds were removed by GPC and alumina column clean-up (Norstrom et al., 1986). Separation of PCDDs, PCDFs and non-ortho PCBs from other contaminants was accomplished using a carbon/fibre column (Norstrom and Simon, 1991; Ford et al., 1993). Separation of the PCDDs/PCDFs from the NOPCBs was completed using Florisil column chromatography.

Analysis. Quantitative analysis for PCDDs/PCDFs and non-ortho PCBs was performed using a VG Autospec double-focusing high resolution mass spectrometer linked to a Hewlett Packard 5890 Series II high resolution gas chromatograph (equipped with a Carlo Erba CTC-A200S autosampler). The mass spectrometer was operated in the selected ion monitoring (VOLTAGE SIR) mode using 10 000 resolution for PCDDs/PCDFs and 7000 resolution for NOPCBs.

Quantitation. Each sample was spiked with $^{13}C_{12}$-labelled PCDD, PCDF (T_4CDD/T_4CDF to H_7CDD/H_7CDF and OCDD) and NOPCB (PCB-77, -126 and -169) internal standards (Wellington Lab.). This was carried out prior to lipid extraction in the case of liver and muscle samples and prior to the GPC clean-up for the fat samples. These internal standards were used to perform internal standard quantitation and to calculate internal standard recoveries. Prior to analysis, two other isotopically-labelled standards (1,2,3,4-

Table 1
PCDD, PCDF and PCB congeners included in the calculation of 2,3,7,8-TCDD equivalents (TEQs)

PCDDs	TEF	PCDFs	TEF	PCBs	TEF
2378-T4D	1.000	2378-T4F	0.100	IUPAC #77	0.0005
12378-P5D	0.500	12378-P5F	0.050	IUPAC #126	0.1000
123478-H6D	0.100	23478-P5F	0.500	IUPAC #169	0.0100
123678-H6D	0.100	123478-H6F	0.100	IUPAC #105	0.0001
123789-H6D	0.100	123678-H6F	0.100	IUPAC #118	0.0001
1234678-H7D	0.010	123789-H6F	0.100		
12346789-OD	0.001	234678-H6F	0.100		
		1234678-H7F	0.010		
		1234789-H7F	0.010		
		12346789-OF	0.001		

Toxic equivalency factors (TEFs) are shown beside each congener and are taken from NATO/CCMS (1988) and Ahlborg et al. (1994).

Table 2
Concentrations ($ng \cdot kg^{-1}$, lipid weight) of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and non-ortho substituted PCBs (NOPCBs) in caribou fat, liver and muscle tissue collected from three herds in the Yukon during 1993

Location	Finlayson		Finlayson		Finlayson		Finlayson		Finlayson		Finlayson		Finlayson		Tay		Bonnet Plume	
Pool	Age 2		Age 3		Age 3		Age 3		Age 4		Age 5		Age 6		Age 5.3		Age 6.6	
Sample type	Fat		Fat		Liver		Muscle		Fat		Fat		Fat		Fat		Fat	
No. in pool	3		2		2		2		6		3		1		13		14	
% Lipid	88.7		85.5		4.1		1.8		87.2		71.3		88.1		84.2		83.8	
	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL
PCDDs																		
2378-T4D	ND	0.34	ND	0.16	ND	8.05	ND	18.33	ND	0.03	ND	0.25	ND	1.48	ND	0.05	ND	0.11
Total T4D	ND		ND		ND		ND		ND		ND		ND		ND		ND	
12378-P5D	ND	0.19	ND	0.05	8.05	2.68	ND	7.22	ND	0.09	ND	0.22	ND	1.04	0.12	0.02	ND	0.13
Total P5D	ND		ND		8.05		ND		ND		ND		ND		0.12		ND	
123679-H6D	ND	0.11	ND	0.02	ND	3.41	ND	9.44	ND	0.16	ND	0.07	ND	0.61	ND	0.02	ND	0.06
123478-H6D	ND	0.11	ND	0.02	ND	3.41	ND	9.44	ND	0.16	ND	0.07	ND	0.61	0.06	0.02	ND	0.06
123678-H6D	ND	0.08	ND	0.02	ND	3.41	ND	9.44	ND	0.16	ND	0.06	ND	0.48	ND	0.02	ND	0.07
123789-H6D	ND	0.10	ND	0.02	ND	3.41	ND	9.44	ND	0.16	ND	0.07	ND	0.61	0.02	0.02	ND	0.07
Total H6D	ND		ND		ND		ND		ND		ND			0.08			ND	
1234679-H7D	ND	0.16	ND	0.13	ND	2.20	ND	12.22	ND	0.13	ND	0.35	ND	1.67	ND	0.02	ND	0.10
1234678-H7D	ND	0.16	ND	0.13	ND	2.20	ND	12.22	ND	0.13	ND	0.35	ND	1.67	0.25	0.02	ND	0.10
Total H7D	ND		ND		ND		ND		ND		ND		ND	0.25			ND	
12346789-OD	0.38	0.14	2.36	0.05	10.73	1.71	ND	8.33	1.08	0.07	ND	0.20	ND	0.49	1.09	0.04	0.66	0.01
PCDFs																		
2368-T4F															0.06	0.04	ND	0.16
2378-T4F	ND	0.21	ND	0.06	ND	10.24	ND	15.00	ND	0.03	ND	0.15	ND	1.03	0.14	0.02	ND	0.16
2367-T4F															ND	0.02	ND	0.16
Total T4F	ND		ND		ND		ND		ND		ND		ND		0.20		ND	
12468-P5F															0.05	0.04	ND	0.11
12478-P5F	ND	0.19	ND	0.12	ND	2.44	ND	6.67	ND	0.13	ND	0.06	ND	0.20	0.24	0.04	0.25	0.11
12378-P5F	ND	0.19	ND	0.12	ND	2.44	ND	6.67	ND	0.13	ND	0.06	ND	0.20	0.06	0.04	ND	0.11
23478-P5F	ND	0.19	ND	0.12	ND	2.44	ND	6.67	ND	0.13	ND	0.07	ND	0.24	0.17	0.04	ND	0.11
23467-P5F	ND	0.19	ND	0.12	ND	2.44	ND	6.67	ND	0.13	ND	0.07	ND	0.24	ND	0.04	ND	0.11
Total-P5F	ND		ND		ND		ND		ND		ND		ND		0.52		0.25	

123468-H6F	ND	0.15	ND	0.09	ND	4.15	ND	5.00	ND	0.09	ND	0.11	ND	0.84	ND	0.04	ND	0.10
124678-H6F	ND	0.15	ND	0.09	ND	4.15	ND	5.00	ND	0.09	ND	0.11	ND	0.84	ND	0.04	ND	0.10
124689-H6F	ND	0.15	ND	0.09	ND	4.15	ND	5.00	ND	0.09	ND	0.11	ND	0.84	ND	0.04	ND	0.10
123478-H6F	ND	0.15	ND	0.09	ND	4.15	ND	5.00	ND	0.09	ND	0.11	ND	0.84	ND	0.04	ND	0.10
123678-H6F	ND	0.12	ND	0.08	ND	4.15	ND	5.00	ND	0.09	ND	0.10	ND	0.48	0.06	0.02	ND	0.10
123789-H6F	ND	0.10	ND	0.11	ND	4.15	ND	5.00	ND	0.09	ND	0.15	ND	0.61	0.11	0.04	ND	0.10
234678-H6F	ND	0.10	ND	0.12	ND	4.15	ND	5.00	ND	0.09	ND	0.18	ND	0.61	0.19	0.04	ND	0.10
Total H6F	ND		ND		ND		ND		ND		ND		ND		0.36		ND	
1234678-H7F	ND	0.15	ND	0.13	ND	2.20	ND	1.67	ND	0.16	ND	0.15	ND	0.75	0.15	0.01	ND	0.04
1234689-H7F	ND	0.15	ND	0.13	ND	2.20	ND	1.67	ND	0.16	ND	0.15	ND	0.75	ND	0.01	ND	0.04
1234789-H7F	ND	0.15	ND	0.13	ND	2.20	ND	2.22	ND	0.16	ND	0.15	ND	0.75	0.08	0.01	ND	0.05
Total H7F	ND		ND		ND		ND		ND		ND		ND		0.23		ND	
12346789-OF	ND	0.10	ND	0.18	ND	1.71	ND	12.22	ND	0.07	ND	0.24	ND	0.41	0.25	0.04	ND	0.06
NOPCBs																		
IUPAC #37	1.41	0.76	12.29	1.24	176.34	17.07	212.78	21.11	10.44	0.26	1.46	0.32	4.68	2.94	3.98	0.70	3.93	1.58
IUPAC #77	2.12	0.37	6.84	0.30	43.90	5.12	80.56	9.44	3.70	0.09	1.43	0.25	2.97	1.29	3.04	0.59	2.92	0.36
IUPAC #81	0.28	0.28	0.73	0.30	ND	5.12	ND	9.44	0.54	0.09	0.28	0.25	ND	1.29	ND	0.59	0.36	0.36
IUPAC #126	6.34	0.26	7.08	0.57	50.00	7.80	ND	14.44	7.11	0.07	6.12	0.21	5.30	1.11	5.74	0.18	9.64	0.25
IUPAC #169	0.83	0.37	1.02	0.23	ND	6.83	ND	17.78	0.79	0.06	0.76	0.27	0.90	0.90	0.58	0.26	0.75	0.43
Recoveries	% Recoveries for 13-C-12																	
2378-T4D	113		87		91		91		82		90		113		92		100	
12378-P5D	120		92		83		85		77		76		111		87		83	
123678-H6D	93		83		87		83		81		79		86		87		91	
1234678-H7D	91		91		90		89		97		63		101		89		93	
12346789-OD	59		68		69		70		93		36		72		84		76	
2378-T4F	94		91		80		82		79		72		95		89		80	
12378-P5F	120		88		80		83		81		72		104		86		72	
123478-H6F	87		71		82		93		75		75		87		84		81	
1234678-H7F	79		82		83		80		96		66		84		80		78	
PCB #77	69		61		52		49		109		76		73		66		38	
PCB #126	91		65		65		64		111		100		88		77		44	
PCB #169	94		66		67		67		108		104		86		75		43	

ND, not detected.

Minimum detection limits (MDLs) corrected for percent lipid and percent recoveries are provided for each compound in each sample.

T_4CDD and 1,2,3,7,8,9-H_6CDD) were added to the cleaned dioxin/furan extracts and PCB-112 was added to the NOPCB fraction. These served as recovery standards for the quantification of internal standard recoveries.

An external standard mixture (containing native quantitation standards, internal standards and recovery standards) was analyzed along with the samples. Relative response factors (RRF) for each pair of native analyte and $^{13}C_{12}$-labelled compound were calculated daily.

Two characteristic ions (the most abundant ions in the molecular cluster) were monitored for each native and $^{13}C_{12}$-labelled compounds, within established retention time windows. The identity of the peaks was confirmed by monitoring accurate masses (4 decimal places) running the mass spectrometer in high resolution mode.

Residue levels for PCDDs/PCDFs and NOPCBs were determined by using an internal standard quantitation method. Internal standard quantitation was based on the integrated areas (sum of the two ions) measured for native congeners, compared directly with the integrated areas measured for the $^{13}C_{12}$-labelled internal standards in the sample, and using the isomer specific RRF values (determined on the same day).

2.4. Quality assurance

Recoveries for $^{13}C_{12}$-PCDDs/PCDFs and -NOPCBs were calculated by comparing the integrated areas of the labelled internal standards and areas of the recovery standards in samples, with the areas of these compounds measured in the external standard mixture analyzed along with the samples. Internal standard recoveries for PCDDs/PCDFs were >70% for most of the samples. Recoveries for NOPCBs were <70% because of problems occurring during the alumina column clean-up. Because calculations were based on internal standard quantitation, all reported levels should be accurate within ±15%.

Minimum detection limits (MDL, signal/noise = 3) were calculated for every congener examined. PCDD/PCDF and NOPCB analysis of a quality control sample (1989 Herring Gull egg pool from Lake Ontario) was performed along with the samples. Method blanks ($^{13}C_{12}$-PCDDs/PCDFs/NOPCBs) were subjected to the entire clean-up procedure and analyzed along with the caribou samples.

2.5. Calculation of 2,3,7,8-TCDD toxic equivalents

Toxic equivalents (TEQs) were calculated for each congener by multiplying congener concentration (wet weight) by a congener-specific toxic equivalency factor (TEF) (NATO/CCMS 1988; Ahlborg et al., 1994). The resulting TEQs were then summed to provide an indication of the toxic potential of the sample expressed as 2,3,7,8-TCDD equivalents. The compounds which were included in the calculation of TEQs, along with their TEFs, are shown in Table 1. These congeners have been identified as being the primary contributors to TEQ concentrations (Ahlborg et al., 1994). PCB congeners #37 and #81 were not included in the TEQ calculation because TEFs for these congeners were not available. Initially, TEQs were calculated only for the caribou from the Northwest Territories because data for the ortho-substituted PCBs, IUPAC #105 and #118, were available only from these herds (Elkin and Bethke, 1995). Some ortho-substituted congeners have been shown to be important contributors to overall TEQ concentrations in a variety of samples (Ahlborg et al., 1994). For the Yukon caribou, data for the two ortho-substituted PCB congeners were not available. Therefore, TEQ levels in Yukon caribou were calculated using two steps: (1) the TEQs contributed by the congeners analyzed in this study were calculated and (2) the percent contribution of the ortho-substituted congeners to overall TEQ levels in Northwest Territories caribou was calculated and the TEQ levels in the Yukon caribou were increased by that percentage, ~12%.

3. Results and discussion

Concentrations ($ng \cdot kg^{-1}$, lipid weight) of individual PCDD, PCDF and NOPCB congeners are reported for the Yukon caribou in Table 2 and the Northwest Territories caribou in Table 3. Minimum detection limits (MDL) for each compound, in each pooled sample, are also reported. Concentrations are expressed on a lipid weight

Table 3
Concentrations ($ng \cdot kg^{-1}$, lipid weight) of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and non-ortho substituted PCBs (NOPCBs) in caribou fat collected from four herds in the Northwest Territories during 1991/1992

Location	Bathurst		Southampton Is.		Cape Dorset		Lake Harbour	
Pool	Age 4.1		Age 3.6		Age 3.7		Age 4.3	
Sample type	Fat		Fat		Fat		Fat	
No. in pool	7		5		3		4	
% Lipid	43.0		92.3		19.2		80.4	
	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL
PCDDs								
2378-T4D	ND	0.19	ND	0.15	0.73	0.31	0.14	0.02
Total T4D	ND		ND		0.73		0.14	
12378-P5D	ND	0.40	0.30	0.18	1.67	0.78	0.31	0.02
Total P5D	ND		0.30		1.67		0.31	
123679-H6D	ND	0.16	ND	0.09	ND	0.10	ND	0.02
123478-H6D	ND	0.16	0.10	0.09	1.04	0.10	0.16	0.02
123678-H6D	ND	0.16	ND	0.10	0.73	0.10	0.32	0.02
123789-H6D	ND	0.16	ND	0.10	ND	0.10	0.09	0.02
Total H6D	ND		0.10		1.77		0.57	
1234679-H7D	ND	0.09	ND	0.16	ND	0.31	ND	0.02
1234678-H7D	0.33	0.09	ND	0.16	0.73	0.31	0.39	0.02
Total H7D	0.33		ND		0.73		0.39	
12346789-OD	2.14	0.02	1.65	0.04	4.69	0.05	1.13	0.01
PCDFs								
2368-T4F	ND	0.30	ND	0.15	ND	0.99	0.09	0.02
2378-T4F	ND	0.30	0.16	0.15	0.99	0.99	0.21	0.02
2367-T4F	ND	0.30	ND	0.15	ND	0.99	0.05	0.02
Total T4F	ND		0.16		0.99		0.35	
12468-P5F	ND	0.19	ND	0.05	ND	0.10	0.05	0.02
12478-P5F	0.74	0.19	0.40	0.05	0.52	0.10	0.27	0.02
12378-P5F	ND	0.19	ND	0.05	ND	0.10	0.04	0.02
23478-P5F	ND	0.19	0.23	0.09	0.78	0.10	0.41	0.02
23467-P5F	ND	0.19	ND	0.09	0.47	0.10	ND	0.02
Total P5F	0.74		0.63		1.77		0.77	
123468-H6F	ND	0.16	ND	0.04	ND	0.16	ND	0.01
124678-H6F	ND	0.16	ND	0.04	ND	0.16	0.04	0.01
124689-H6F	ND	0.16	ND	0.04	ND	0.16	0.02	0.01
123478-H6F	ND	0.16	ND	0.04	0.42	0.16	0.15	0.01
123678-H6F	ND	0.16	ND	0.04	0.73	0.10	0.11	0.01
123789-H6F	ND	0.16	0.23	0.07	ND	0.16	0.15	0.01
234678-H6F	ND	0.16	ND	0.07	0.83	0.21	0.14	0.01
Total H6F	ND		0.23		1.98		0.61	
1234678-H7F	ND	0.07	ND	0.16	ND	0.31	0.16	0.01
1234689-H7F	ND	0.07	ND	0.16	ND	0.31	ND	0.01
1234789-H7F	ND	0.09	ND	0.16	ND	0.31	0.06	0.01
Total H7F	ND		ND		ND		0.22	
12346789-OF	ND	0.26	ND	0.13	ND	0.21	0.25	0.04
NOPCBs								
IUPAC #37	7.70	1.49	2.60	1.30	15.10	5.47	2.64	0.95
IUPAC #77	5.14	0.23	6.45	0.85	10.31	0.89	5.52	0.75
IUPAC #81	0.58	0.23	0.91	0.85	2.45	0.89	1.74	0.75

Table 3 (*Continued*)

Location	Bathurst		Southampton Is.		Cape Dorset		Lake Harbour	
Pool	Age 4.1		Age 3.6		Age 3.7		Age 4.3	
Sample type	Fat		Fat		Fat		Fat	
No. in pool	7		5		3		4	
% Lipid	43.0		92.3		19.2		80.4	
	Conc.	MDL	Conc.	MDL	Conc.	MDL	Conc.	MDL
IUPAC #126	7.16	0.09	26.73	0.53	74.74	0.21	30.73	0.12
IUPAC #169	0.88	0.26	2.28	0.65	11.77	0.68	3.42	0.30
Recoveries			% Recoveries for 13-C-12					
2378-T4D	103		81		90		96	
12378-P5D	88		60		76		93	
123678-H6D	98		83		76		92	
1234678-H7D	101		58		82		92	
12346789-OD	82		54		75		88	
2378-T4F	91		70		79		92	
12378-P5F	79		65		75		91	
123478-H6F	85		59		69		89	
1234678-H7F	83		48		72		84	
PCB #77	70		24		58		52	
PCB #126	75		28		73		68	
PCB #169	75		27		97		68	

ND, not detected.
Minimum detection limits (MDLs) corrected for percent lipid and percent recoveries are provided for each compound in each sample.

basis to facilitate geographic comparisons of levels in fat tissue and to allow examination of the distribution of these compounds in fat, liver and muscle tissue. Levels of PCDDs, PCDFs and NOPCBs were extremely low in animals from all herds.

The relationship between age and lipid-normalized congener concentration was examined using linear regression analysis. Obviously, this could only be completed on congeners which were present at detectable levels in the majority of the Finlayson samples (OCDD and the five NOPCBs). There were no statistically significant relationships between age and OCDD/NOPCB levels in fat tissue from the Finlayson caribou ($P > 0.1$ in all cases), therefore, all female caribou over the age of 1 year were included in the pools for the other six herds. The number of animals and the mean (± 1 S.E.) age of the caribou included in each of the pooled samples was as follows: Bonnet Plume ($n = 14$, 6.6 $\pm$ 0.6 years), Tay ($n = 13$, 5.3 $\pm$ 0.8 years), Bathurst ($n = 7$, 4.1 $\pm$ 0.6 years), Southampton Island ($n = 5$, 3.6 $\pm$ 0.5 years), Cape Dorset ($n = 3$, 3.7 $\pm$ 0.9 years) and Lake Harbour ($n = 4$, 4.3 $\pm$ 0.9 years).

3.1. Geographic trends in fat tissue

In the Finlayson caribou, only octachlorodibenzodioxin (OCDD) was found on a regular basis and only in negligible quantities (maximum 2.36 $ng \cdot kg^{-1}$, lipid wt.) (Table 2). All of the more highly toxic PCDD/PCDF congeners, such as 2,3,7,8-TCDD, were not detected. PCDD, PCDF and NOPCB concentrations in the Tay and Bonnet Plume herds were similar to the Finlayson herd, however, a greater variety of PCDDs and PCDFs were observed in the Tay caribou. This may have been the result of exposure to minor local sources. A variety of PCDDs and PCDFs was also observed in caribou from the Bathurst, Southampton, Cape Dorset and Lake Harbour herds but the levels in these animals were also extremely low (Table 3). The larger number of PCDD/PCDF congeners detected in the Northwest Territories herds may have reflected differences in the long-range atmospheric transport of these compounds to the western and eastern Arctic. This is consistent with the observation that OCDD was the predominant PCDD/PCDF congener found in all samples. Highly chlorinated

dioxins, such as OCDD, are usually indicative of combustion-related sources (Broman et al., 1991), which probably arrived in the Arctic via long-range atmospheric transport (Norstrom et al., 1990).

Non-ortho PCBs were present at low concentrations in all of the caribou from the Yukon and Northwest Territories (Tables 2 and 3). PCB congeners #126 and #169 showed some spatial variability with higher levels in the eastern Arctic. This result corroborates the findings of Elkin and Bethke (1995) who found higher concentrations of ortho-substituted polychlorinated biphenyl congeners in the eastern Arctic. Higher levels of these PCBs in the east are probably also the result of differences in atmospheric circulation patterns, thereby affecting the deposition of these contaminants via long-range atmospheric transport.

3.2. Tissue distribution and human consumption

When expressed on a lipid weight basis, contaminant concentrations, in the age 3 Finlayson pool, are higher in muscle and liver tissue than in fat (Table 2). This indicates that the partitioning of these compounds to caribou tissue is not purely driven by the lipid content of the tissue (i.e. equilibrium partitioning) but that physiological processes may be important in regulating the preferential deposition of these compounds in muscle and liver. From a human consumption perspective, however, it is the wet weight concentrations which are of interest because all tissues are consumed on a wet weight basis. Wet weight concentrations in fat, liver and muscle would be ~75%, 4% and 2% of lipid weight concentrations, respectively. Therefore, we would expect concentrations in fat tissue to represent the maximal concentrations to which the human consumer is exposed. This is emphasized by examining the concentration of 2,3,7,8-TCDD equivalents (TEQs) which were calculated for each pooled sample (Table 4). In 3-year-old caribou from the Finlayson herd, TEQ levels are greater in fat tissue than in liver or muscle. However, the TEQ levels found in all of the caribou tissues are extremely low with a maximum concentration of 3.29 $ng \cdot kg^{-1}$. TCDD equivalents in Arctic marine mammals, such as the beluga (*Delphinapterus leucas*) and narwhal (*Monodon monoceros*), range from ~100–500 $ng \cdot kg^{-1}$ (Norstrom and Muir, 1994). TEQ levels in ringed seal (*Phoca hispida*) are approximately one-tenth those observed in beluga and narwhal but are still one order of magnitude greater than those observed in caribou. The greatest contribution to overall TEQ levels was from the non-ortho substituted PCBs, with PCB #126

Table 4
2,3,7,8-TCDD equivalents (TEQs) ($ng \cdot kg^{-1}$, wet weight) in caribou from the Northwest and Yukon Territories

Herd	Tissue	2,3,7,8-TCDD equivalents ($ng \cdot kg^{-1}$, wet wt.)				
		PCDDs	PCDFs	NOPCBs	PCB 105 + 118	Total TEQs
Lake Harbour	Fat	0.29	0.23	2.50	0.27	3.29
Cape Dorset	Fat	0.34	0.13	0.46	0.30	1.23
Southampton Is.	Fat	0.15	0.14	0.49	0.07	0.85
Bathurst	Fat	<0.01	ND	0.31	0.02	0.33
Finlayson, age 2	Fat	<0.01	ND	0.57	[a]0.08	0.65
Finlayson, age 3	Fat	<0.01	ND	0.62	[a]0.08	0.70
Finlayson, age 3	Liver	0.17	ND	0.21	[a]0.05	0.43
Finlayson, age 3	Muscle	ND	ND	<0.01	[a]<0.01	<0.01
Finlayson, age 4	Fat	<0.01	ND	0.63	[a]0.08	0.71
Finlayson, age 5	Fat	<0.01	ND	0.44	[a]0.06	0.50
Finlayson, age 6	Fat	<0.01	ND	0.48	[a]0.06	0.54
Tay	Fat	0.06	0.12	0.49	[a]0.09	0.76
Bonnet Plume	Fat	<0.01	ND	0.82	[a]0.11	0.93

ND, not detected.
[a]TEQs contributed by PCBs #105 and #118 were estimated for the Yukon caribou. See text for details.

being of particular importance. Their mean contribution to TEQ levels in fat tissue from all of the herds was >70%. These results are similar to those from previous studies which found that PCBs were the major contributors to overall TEQ levels in Arctic marine mammals (Daelemans et al., 1993; Ford et al., 1993).

Our results indicate that PCDDs, PCDFs and NOPCBs are unlikely to pose a threat to either the caribou sampled in this study or to their human consumers. The levels observed can probably be considered to be background concentrations. PCDDs, PCDFs and PCBs are globally distributed and long-range atmospheric transport, with some minor local inputs, can probably account for the PCDD/PCDF and non-ortho PCB levels found in all of the caribou.

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