

**TROPHIC TRANSFER OF PERSISTENT ORGANIC POLLUTANTS
IN AN ARCTIC TUNDRA ECOSYSTEM**

by

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ABSTRACT

The purpose of this study was to investigate the trophic transfer of Persistent Organic Pollutants (POPs) in lichen-caribou-wolf food-chains of the Canadian Arctic. Chemical fugacities of various organic contaminants, including hexachlorocyclohexane (HCH), hexachlorobenzene (HCB) and polychlorinated biphenyl (PCB) congeners, increased with increasing trophic level in lichen-caribou-wolf food-chains from Canada's central and western Arctic. A fugacity-based bioaccumulation model that explains this biomagnification due to gastrointestinal magnification was developed and applied to lichen-caribou-wolf food-chains from Cambridge Bay, Bathurst Inlet and Inuvik. A dynamic head-space methodology was developed to measure fugacity capacities of lichens (Z_D) and caribou fecal pellets (Z_G). The values of Z_D and Z_G were also estimated by using theoretical calculations. The ratio Z_D/Z_G represents the extent of food digestion in an organism and is an important parameter in this fugacity-based bioaccumulation model. The fugacity capacities of lichens and caribou fecal pellets measured in the laboratory experiments were greater than those values estimated by theoretical calculations. The higher fugacity capacities of lichens and caribou fecal pellets measured in laboratory experiments were attributed to methodological errors. The model is time-dependent and simulates chemical bioaccumulation over the life-time of caribou and wolves from observed chemical concentrations in two common tundra lichens (*Cladina rangiferina* and *Cetraria nivalis*). The model slightly under-predicted chemical concentrations in female

caribou in July (model bias = 0.92) and over-predicted chemical concentrations in male caribou in September (model bias = 2.71). The model predicted biomagnification factors (BMFs) for male caribou in July was approximately 20. The predicted BMFs for wolves relative to lichens was approximately 190. The predicted BMFs were equal for various chemicals, with octanol-water partition coefficients (K_{ow}) ranging from 10^4 to 10^8 .

Current management policies under Environment Canada's Toxic Substance Management Plan (*TSMF*) target chemicals that exhibit K_{ow} 's greater than 10^5 because their biomagnification in aquatic food-chains has not been documented. The observed BMF of beta-HCH ($K_{ow} = 10^{4.5}$) in caribou and wolves at Bathurst Inlet were 14 and 170, respectively. These results suggest that chemicals that exhibit K_{ow} 's less than 10^5 can biomagnify in terrestrial food-chains, and hence question the ability of current management strategies to protect terrestrial ecosystems from the accumulation of bioaccumulative and potentially toxic substances.

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CHAPTER 1

INTRODUCTION

International concern regarding global environmental degradation has increasingly emerged in the past few decades. Scientific studies have provided governments, industry and the general public with information surrounding these global issues. Environmental problems such as acid rain, ozone depletion, biodiversity, nuclear fallout and global warming have previously been debated on the international stage (McCormick, 1985; Leggett, 1990). More recently, the issue of environmental contaminants has become the focus of much international attention. In particular, persistent organic pollutants (POPs) such as PCBs and DDT have been extensively studied since the late 1960's.

Recent evidence suggests that emission of these chemicals throughout industrial and developing countries can result in environmental impacts several thousands of kilometers away (Bidleman *et al.*, 1989; Welch *et al.*, 1991; Wania and Mackay, 1996). Scientists have detected high levels of industrial chemicals in pristine environments far removed from source emission locations. Further scientific evidence has shown that cold environments may act as a sink for globally circulating organic chemicals. Previous studies have documented elevated levels of organic contaminants in environmental and biological samples from the Arctic

(Muir *et al.*, 1988; Norstrom *et al.*, 1988; Bidleman *et al.*, 1995; Zhu *et al.*, 1995; Stern *et al.*, 1997; Addisson *et al.*, 1998), Antarctic (Oehme *et al.*, 1995; Lenihan *et al.*, 1995; Bacci *et al.*, 1986), and more recently in alpine ecosystems (Blais *et al.*, 1998). Concentrations of organic chemicals in these colder ecosystems can be several times greater than those found in warmer climates. A more detailed discussion of the sources, sinks and pathways of POPs in Arctic ecosystems can be found in Appendix V. This scientific evidence suggests the occurrence of an increased loading of particular environmental contaminants to polar and alpine ecosystems. Once a chemical enters an ecological system the fate and potential ecological impacts of that substance depends on the chemical's environmental persistence and bioaccumulative potential in local food-chains.

Bioaccumulation of organic chemicals in ecological food-chains is a complex and not well understood process. Many persistent organic pollutants (POPs), such as DDT and PCBs have been shown to bioaccumulate in organisms and biomagnify through trophic levels (Oliver and Niimi, 1988; Gobas, 1993a; Morrison *et al.*, 1997). However, the mechanisms influencing food-chain bioaccumulation are not completely understood. In absence of this scientific understanding, policy makers tend to regulate chemicals by taking action on substances that meet certain criteria. Environment Canada's Toxic Substances Management Plan (*TSMP*) and the Long-Range Transboundary Air Pollution (*LRTAP*) *POPs Protocol* initiated by the United Nations Environment Program have adopted similar assessment

criteria for regulating environmental contaminants (Appendix IV). To assess the bioaccumulation potential of a candidate substance, these policies target chemicals that exhibit bioaccumulation or bioconcentration factors (BAFs/BCFs) greater than 5000, or chemicals with a $\log K_{ow}$ greater than 5. Therefore, chemicals that have octanol-water partition coefficients (K_{ow} 's) less than 10^5 are considered non-bioaccumulative under the *TSMP and LRTAP POPs Protocol*. These measures of chemical bioaccumulation potential are usually determined experimentally. BAFs and BCFs are most commonly determined through short-term chemical exposure experiments involving small laboratory organisms such as guppies, goldfish (McConnel *et al.*, 1978; Gobas *et al.*, 1988; Fisk *et al.*, 1998).

In the assessment criteria outlined in the *TSMP and LRTAP POPs Protocol*, the bioaccumulation of a substance observed in chemical exposure experiments using laboratory organisms is assumed to be representative of bioaccumulation in all species and for all life-stages. Ecological food-chains are dynamic systems consisting of many different organisms, each having specific life-history's and evolutionary strategies. Also, the current approach for developing bioaccumulation criteria is based on observed bioaccumulation in aquatic organisms but does not consider mechanistic differences in terrestrial animals. This simplified approach adopted by management agencies for assessing bioaccumulation of chemical substances may not completely denote the potential of a candidate substance to bioaccumulate in food-chains. Scientific understanding of species and ecosystem specific mechanisms affecting

bioaccumulation may warrant a more accurate assessment of a chemical's ability to bioaccumulate in food-chains.

The purpose of the present study is to investigate trophic transfer of persistent organic pollutants in lichen-caribou-wolf food-chains of the Canadian Arctic. The objectives of this study are to: (i) evaluate food-chain bioaccumulation of organic chemicals in an Arctic tundra ecosystem; (ii) develop a mechanistic bioaccumulation model representing bioaccumulation of organic contaminants in the lichen-caribou-wolf food-chains of the Canadian Arctic; (iii) parameterize the model based on laboratory experiments and a literature reviews of organic contaminants in tundra ecosystems; and (iv) validate the bioaccumulation model by comparing model predicted chemical concentrations in caribou and wolves with observed chemical concentrations in caribou and wolf tissues measured during an independent study conducted by the Government of the Northwest Territories. Chapter 2 involves a trophodynamic analysis of chemical transfer in Arctic tundra ecosystems. Specifically, Chapter 2 involves calculating chemical fugacities from observed chemical concentrations in lichen-caribou-wolf food-chains from the central and western Arctic to assess food-chain bioaccumulation of persistent organic pollutants (POPs). Chapter 3 reports on a study that investigates the gastro-intestinal magnification of organic chemicals in barren-ground caribou (*Rangifer tarandus*). Chapter 4 is a modelling study which includes development and field-validation of a mechanistic model representing bioaccumulation of

organic contaminants in lichen-caribou-wolf food-chains of Canada's central and western Arctic.

CHAPTER 2

Trophodynamics and food-chain bioaccumulation of persistent organic pollutants (POPs) in Arctic tundra ecosystems

INTRODUCTION

Atmospheric input of POPs has been recognized as the major pathway by which these chemicals enter the terrestrial environment of the Arctic (Carlberg *et al.*, 1983; Thomas *et al.*, 1992; Landers *et al.*, 1995; Elkin and Bethke, 1996).

Chemical input to the tundra ecosystem may involve gaseous partitioning between air and vegetation, particulate deposition, and wet deposition in the form of rain or snow. Snow is of particular interest in the tundra ecosystem due to the annual longevity of snowfall and snowpack accumulation in this region. It has been previously shown that snow deposition may scavenge significant quantities of airborne organic contaminants (Franz *et al.*, 1998; Wania *et al.*, 1999). Seasonal distribution and depositional processes of organic substances in the Arctic atmosphere may therefore influence chemical loadings to a particular region on the tundra.

Tundra vegetation in the Arctic consists of many species of lichens, vascular plants, and small shrubs. Chemical accumulation and storage in vegetation can

be viewed as the primary source of chemical exposure to foraging herbivores, which may subsequently affect bioaccumulation in top-predator carnivores. Thus, bioconcentration of chemical substances from the air and snow into tundra vegetation may play a crucial role in the bioaccumulation of organic contaminants in tundra food-chains of the Arctic.

Dense lichen mats make up a large proportion of ground cover vegetation in tundra ecosystems of the Arctic (Miller, 1976). Lichens are comprised of two separate organisms that have developed symbiotic relationships to sustain life. One organism is a fungus, while the other consists of either green alga cells or several blue-green cyanobacteria, depending on the species. The green algae or blue-green cyanobacteria contain chlorophyll and thus are capable of photosynthesis. Because lichens lack a root system they depend entirely on the atmosphere for moisture and nutrient uptake. The photosynthesizing cells of the lichen are encased by a fungus which provide moisture and nutrients to the algae or cyanobacteria. In return, the algae or cyanobacteria photosynthesize food for the fungus which lacks chlorophyll and thus is incapable of photosynthesis. Lichens do not possess a waxy cuticle or internal transport mechanisms, both of which can inhibit bioconcentration depending on a chemical's hydrophobicity. Lichens have been extensively used in chemical biomonitoring studies because of their non-selective chemical accumulation mechanisms and their widespread global distribution (Bacci *et al.*, 1986; Villeneuve *et al.*, 1988; Muir *et al.*, 1993). The bioconcentration process in

plants has previously been explained by an equilibrium partitioning, resulting in a gaseous air-leaf exchange (Patterson *et al.*, 1991; Polder *et al.*, 1997; Thomas *et al.*, 1998; Wagrowski and Hites, 1998). However, lichens in the Arctic may experience long periods covered by deep snowpacks. The snowpack contains contaminants that have been scavenged from the previous winter's snowfall events. During a snowmelt period, typically from late May to early June, a significant amount of stored chemical in the snowpack may be discharged and hence accumulate in the underlying lichens.

An important tundra food-chain is that of lichens, barren-ground caribou (*Rangifer tarandus*) and tundra wolves (*Canis lupus*). The state of this food-chain is of particular interest because lichen-caribou-human food-chains are prominent throughout the Canadian Arctic (Hall, 1989). Caribou are migratory herbivores, feeding primarily on lichens but may also forage on other vegetation such as cotton grass (*Eriophorum latifolium*) and willows (*Salix spp.*). The approximate geographic ranges of caribou herds in the central and western Arctic of Canada are shown in Figure 2.1. Caribou from these herds are a major source of the nutritional requirement for wolves in the Arctic (Bergerud and Ballard, 1988; Dale *et al.*, 1994). In some regions, wolf pack migration and den site selection have shown strong correlation with seasonal movements of caribou herds (Heard and

Williams, 1992). Figure 2.1 illustrates the range of timber, tundra and true arctic wolves in the Canadian Arctic.

Caribou-dependent relationships have been observed in wolves on the range of the Bathurst caribou herd. Wolf dens on the Bathurst range have been located near tree-line in close proximity to the migration path of the Bathurst caribou herd (Figure 2.1). Relationships between lichen, caribou, and wolves in tundra ecosystems constitute a linear food-web structure in which caribou forage on lichens and wolves predate mainly on local caribou. Analysis of the chemical dynamics within this food-chain may elicit a better understanding of the transport and accumulation of organic chemicals in terrestrial ecosystems. The objective of this chapter is to investigate the extent of chemical biomagnification in lichen-caribou-wolf food-chains of Canada's central and western Arctic. For this purpose, chemical concentrations of various organic contaminants have been compiled from measurements in air, snow, lichens, caribou, and wolves from the central and western Arctic, and expressed in their corresponding fugacities.



Figure 2.1: Approximate geographic ranges of barren-ground caribou (*Rangifer tarandus*) and North American Timber, Tundra and Arctic wolves (*Canis lupus*). Also shown are the range and calving grounds for Bluenose (Inuvik), Victoria Island (Cambridge Bay), Beverly, and Bathurst (Bathurst Inlet) caribou herds.

THEORY

Chemical dynamics within a tundra ecosystem involves input from atmospheric sources (air, rain, snow), environmental transport processes (volatilization, biological degradation), and food-chain bioaccumulation. Because chemical transport between environmental and biological media is driven by fugacity differences, rather than differences in chemical concentrations, it is useful to convert concentrations (C) into chemical fugacities (f). Fugacity can be viewed as an “escaping tendency” of a chemical between different environmental media, such as transport from the atmosphere to vegetation (bioconcentration) and conversely from vegetation back to the atmosphere (volatilization). Fugacity is equivalent to chemical potential and can be measured as the partial pressure (in Pascals) that a chemical exerts within a given matrix. The chemical concentration (C in mol/m^3) and the chemical fugacity (f in Pa) in a given media are related because C equals $f \cdot Z$, where the fugacity capacity (Z in $\text{mol}/\text{m}^3 \cdot \text{Pa}$) indicates the ability of that media to retain chemical within its matrix. Passive chemical transport between different environmental media can occur when thermodynamic gradients between the media exist, resulting in net chemical transport from moving from media of high fugacity to low fugacity. Environmental media are in a state of equilibrium when their respective fugacities are observed to be equal. In fugacity terms, biomagnification of organic contaminants in a food-chain occurs when fugacities in organisms increase with increasing trophic level. Fugacity-based biomagnification factors (BMFs) are then the ratio of chemical fugacities in an

organism to those fugacities observed in its prey (f_B/f_D). An analysis of chemical fugacities observed in environmental media (e. g., air-snow) and biological compartments (e. g., lichen-caribou-wolves) may better assess chemical transport and biomagnification in ecological systems.

The fugacity capacity of Arctic air (Z_A) at a mean annual summer temperature of 10°C is 0.00043 mol/m³·Pa, following the ideal gas law (i. e., $Z = 1/RT$). Storage of hydrophobic organic chemicals within the matrix of environmental media can be associated with lipids and organic carbon. The fugacity capacity of lipid (Z_L) is assumed to be equal to the fugacity capacity of octanol (Z_O) and is calculated as $Z_L = Z_O = K_{OW} \cdot Z_W$. K_{OW} is the octanol-water partition coefficient of the chemical and Z_W is the fugacity capacity of water which is the reciprocal of the chemical's Henry's law constant (H , in units of Pa·m³/mol) of a given chemical. For media with very low lipid fractions such as vegetation, the chemical may be associated with organic carbon to a greater extent rather than with lipids within its matrix. The fugacity capacity of organic carbon has been shown to be approximately 41% of that of octanol (Karickhoff, 1981). Thus, the organic carbon-water partition coefficient (K_{OC}) is calculated as $K_{OC} = 0.41 \cdot K_{OW}$. Morrison *et al.* (1996) presented a calculation for the fugacity capacity of low-lipid media using K_{OC} , the density and organic carbon content of the media. Following this method, the fugacity capacity of lichens can be calculated as $Z_{LICHEN} = Z_W \delta_D \phi_D K_{OC}$, where δ_D is the density (in kg/L), and ϕ_D is the organic carbon content of the media.

The chemical fugacities (f , in units of Pascals) can be calculated from observed concentrations (C , mol/m³) and corresponding fugacity capacities (Z , mol/m³·Pa) of the media as f is inversely proportional to C (i. e., $f = C / Z$). Fugacities in lichens are calculated as the chemical concentration (mol/m³ dry tissue) divided by the fugacity capacity of lichen (Z_{LICHEN}). To calculate fugacities in biota (f_{B} , caribou and wolves), the lipid normalized chemical concentrations in biological tissues (mol/m³ lipid) are divided by the fugacity capacity of lipid (Z_{L}). The chemical fugacities in air (f_{A}) and water (f_{W}) can be calculated as $f_{\text{A}} = C_{\text{A}} / Z_{\text{A}}$ and $f_{\text{W}} = C_{\text{W}} / Z_{\text{W}}$. Also, the chemical fugacities in spring meltwater can be calculated from the chemical concentration in meltwater and the fugacity capacity of water by the equation, $f_{\text{MELT WATER}} = C_{\text{MELT WATER}} / Z_{\text{W}}$.

METHODS

Field collection and data compilation

Figure 2.2 shows sampling locations for the collection of vegetation and caribou fecal pellets (present study), caribou and wolf tissues (Government of Northwest Territories, GNWT supplied data), and air-snowfall-snowpack (Jensen *et al.*, 1998). Vegetation including two common lichen species (*Cladina rangiferina* and *Cetraria nivalis*) and leaves of tundra willows (*Salix glauca*) and caribou fecal pellets representing the diet and gastro-intestinal contents of barren-ground caribou (*Rangifer tarandus*), respectively were collected at different locations

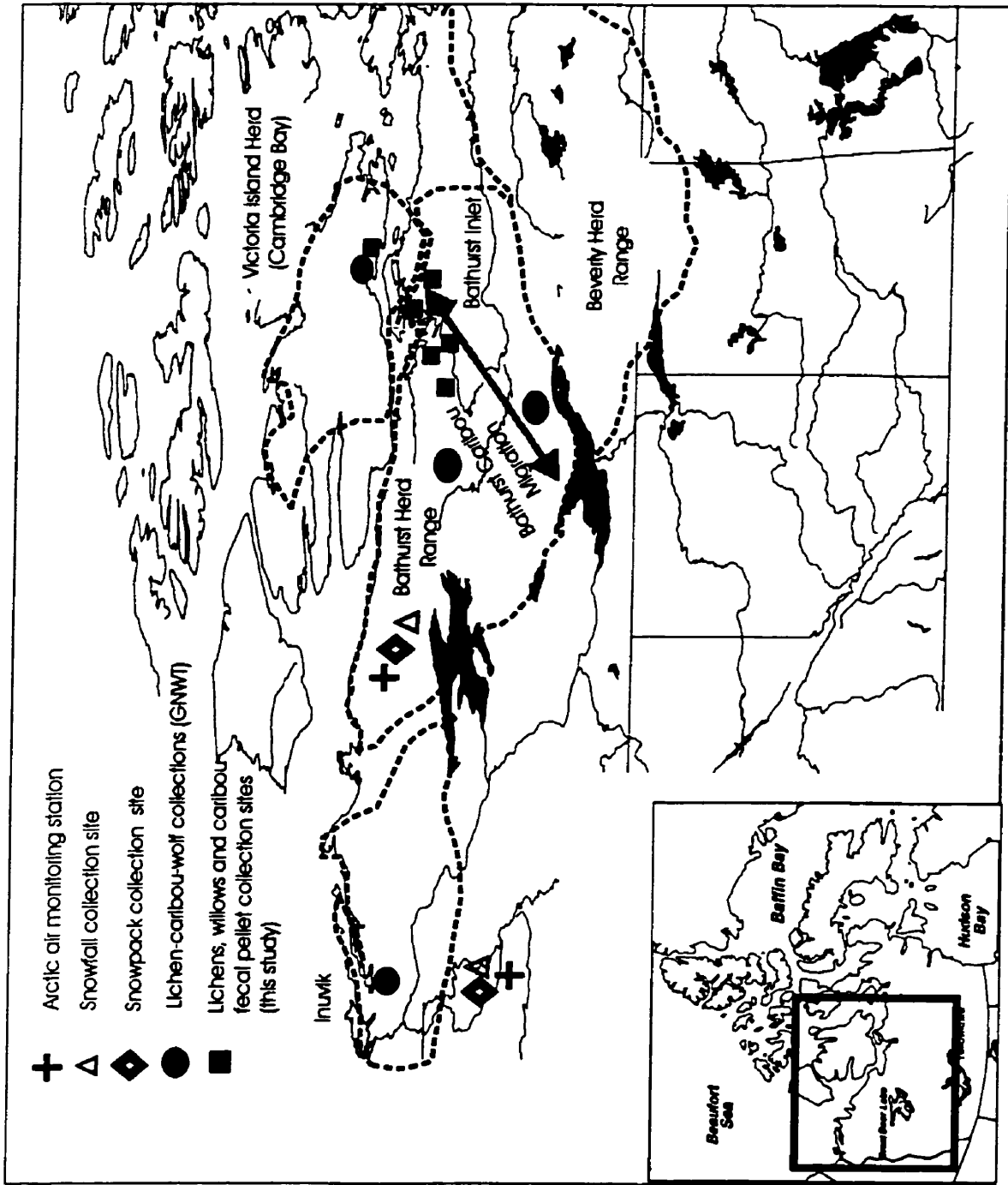


Figure 2.2: Collection sites for sampling of air, snowfall, snowpacks, lichens, caribou, wolves, and caribou fecal pellets.

surrounding Bathurst Inlet during May-June 1997 and July 1998. Vegetation and caribou scat samples were identified by comparison with Pielou (1994). Caribou fecal pellets were collected from individual scat piles in order to represent the GIT content of individual animals. Scat samples were collected fresh from animals observed grazing on lichens near Bathurst Inlet. At each sampling location, 3 to 6 independent samples of caribou fecal pellets, lichens and tundra willows were collected for chemical analysis. Samples were stored at -10°C in 5 mL glass vials.

In 1992, the Canadian Atmospheric Environment Service (AES) collected and analyzed air, snowfall and snowpacks for organochlorine contaminants at different locations in the central and western Arctic. Field sampling and chemical analysis methodologies for air and snow samples are outlined in the 1998 Canadian Arctic Contaminants Assessment Report (CACAR) (Jensen *et al.*, 1998).

From 1992 to 1995, the government of the Northwest Territories, Department of Renewable Resources recently completed a biomonitoring study involving chemical analyses of lichens, liver, muscle and fat tissues of caribou and wolves collected from various locations in the Canadian Arctic. At three locations, Inuvik (Bluenose herd), Cambridge Bay (Victoria Island herd), and Bathurst Inlet (Bathurst herd), organochlorine concentrations have been analyzed in lichens, caribou, and wolf tissues. These data, generated by the Government of the Northwest Territories, constitute a compilation of current levels of organochlorine

contaminants in lichen-caribou-wolf food-chains from Canada's central and western Arctic.

Chemical analysis

Preparation and cleanup of vegetation and caribou fecal samples were performed by the Great Lakes Institute for Environmental Research (GLIER) according to the methods of Lazar *et al.* (1992). Samples (approximately 10 g wet wt.) were homogenized with 20 g Na₂SO₄ using a glass mortar and pestle. The homogenate powder was then extracted using a Na₂SO₄ column by eluting with 50 mL of hexane. The extract was collected and evaporated to 2 mL. 2 mL of DCM was then added to this extract and then transferred to a Gel Permeation Column (GPC) filled with 50 g of BioBeads, S-X33 (BioRad) in 50% DCM/hexane solution (V/V).

The lipid fraction from the GPC was collected and discarded. The remaining 150 mL of eluent from the GPC was collected and transferred to a 1x 35 cm glass column prepared with 6 g of Florisil (60 -100 μm mesh) topped with 2 cm of anhydrous Na₂SO₄. Three fractions were eluted using hexane (fraction 1), 15% DCM/hexane (fraction 2), and 50% DCM/hexane (fraction 3). Each fraction was evaporated to 2 mL and analyzed using a Hewlett-Packard model 5890 gas-chromatograph with electron capture detection (GC-ECD). Chemical identification and quantification was performed by comparing the sample peak against the respective peak areas from calibration standards for each of the three fractions

supplied by the Canadian Wildlife Service Laboratory, Hull, Quebec, Canada. The detection level was 0.05 µg/kg, and recovery efficiencies were all greater than 90%. The lipid content was determined on sub-samples of the extracts and measured gravimetrically using one-tenth of the extract. The lipid content was reported as a percentage of the samples wet weight. Moisture content was determined by comparing the sample's wet and dry weights after oven-drying a 1 g sample at 125 ° C for 24 hr. Organic carbon content of lichen and fecal samples were estimated from loss on ignition by heating samples at 600 °C for 24 hr.

Data analysis and statistics

Chemical concentrations in vegetation and caribou fecal pellets collected from Bathurst Inlet, Cambridge Bay and Inuvik were compiled and separated by species and sampling location. Because lichens from the Bathurst region were collected during spring and summer field seasons, chemical concentrations in lichens were also separated by season. Chemical concentrations on a dry weight basis (ng/g dry wt.) were determined by dividing the wet weight chemical concentrations by the percent dry matter (%DM) of each sample. The arithmetic means and the corresponding standard deviations of the dry weight concentrations were calculated and reported for each sampling location. Total PCBs were reported as the sum of the 43 PCB congeners analyzed. For non-

detectable PCB congeners the sample detection limit was used in the calculation of total PCBs.

Chemical concentrations in liver, muscle and fat tissues of caribou and wolves collected near Bathurst Inlet, Cambridge Bay and Inuvik by the Government of the Northwest Territories were compiled and separated by tissue and sex. Fresh weight chemical concentrations were lipid normalized by dividing fresh weight chemical concentrations by the lipid content of each sample. The arithmetic means and standard deviations of chemical concentrations (ng/g lipid) in liver, muscle and fat were calculated for caribou and wolves at the three sampling locations. Because caribou from Bathurst Inlet were collected during summer (July) and fall (September), the chemical concentrations observed in tissues of Bathurst caribou were further separated by season. Total PCBs were reported as the sum of the 43 PCB congeners analyzed. For non-detectable PCB congeners in caribou and wolf samples, the sample detection limit was used in the calculation of total PCBs.

Chemical fugacities (f , in units of Pa) in lichens, caribou and wolves were calculated using the equation ($f = C/Z$). Fugacities in lichens were calculated by dividing the molar chemical concentrations of dry lichen tissue (mol/m^3 dry tissue) by the fugacity capacity of lichen (Z_{Lichen}). Similarly, fugacities in caribou and wolves were calculated by dividing molar concentrations (mol/m^3 lipid) in fat tissue samples by the fugacity capacity of lipids (Z_L). The chemical fugacities calculated

from the observed chemical concentrations in lichens, caribou and wolves from Bathurst Inlet, Cambridge Bay and Inuvik were then transformed logarithmically to determine the geometric mean (GM) and standard deviations of the GM. The geometric mean was calculated as the antilog of the logarithmic averages. The upper and lower standard deviations were determined by multiplying and dividing the GM by the antilog of the logarithmic standard deviation, respectively.

One-way Analyses of Variance (ANOVA) tests were performed to determine statistically significant differences between the mean logarithms of chemical fugacities in lichens, caribou and wolves. Data representing chemical concentrations in lichen-caribou-wolf food-chains is most comprehensive for species on the Bathurst range. Lichens on the Bathurst range were collected from several locations during summer and spring, while Bathurst caribou were collected during fall and summer field seasons. Also, accurate information pertaining to the sex and age of caribou and wolves sampled from the Bathurst range are available. For these reasons, ANOVA's using chemical fugacities in the lichen-caribou-wolf food-chain at Bathurst Inlet were conducted to investigate chemical bioaccumulation relationships with age, sex and season. Specifically, ANOVA's using a significance level of $\alpha = 0.05$ were conducted to determine statistically significant differences between the means of concentrations of various substances in (i) lichens collected during spring and summer, (ii) caribou and wolves of different age and sex, and (iii) caribou collected in summer and fall. The sample

size (n), F-value , F_{critical} and p values were reported for each ANOVA. Statistical significance was indicated when F-values exceeded F_{critical} .

RESULTS AND DISCUSSION

Geographic variation of POPs in lichen-caribou-wolf food-chains

Spatial bioaccumulation patterns of total PCBs in lichen-caribou-wolf food chains of the central and western Arctic are illustrated in Figure 2.3. Fugacities of total PCBs in summer collected lichens ranged from approximately 6.4×10^{-12} Pa at Inuvik to 1.3×10^{-11} Pa and 2.7×10^{-11} Pa observed at Bathurst Inlet and Cambridge Bay, respectively. Analysis of caribou fat samples showed that fugacity of total PCBs in caribou ranged from approximately 5.5×10^{-12} Pa at Inuvik to 2.4×10^{-11} Pa and 3.0×10^{-11} Pa in caribou at Bathurst and Cambridge Bay were, respectively. The fugacities of Total PCBs in caribou demonstrate a spatial trend where chemical fugacities are lower in the western Arctic compared to the fugacities observed in animals from the central Arctic. Chemical fugacities of Total PCBs in wolves from these three locations demonstrated a different spatial bioaccumulation pattern than lichens and caribou. Bathurst wolves had the highest levels of PCBs, exhibiting an average chemical fugacity of approximately 1.4×10^{-9} Pa. Cambridge Bay wolves, despite higher fugacities of PCBs in caribou from this region, had lower fugacities (1.26×10^{-10} Pa) than wolves sampled at Inuvik (2.1×10^{-10} Pa). The lower levels

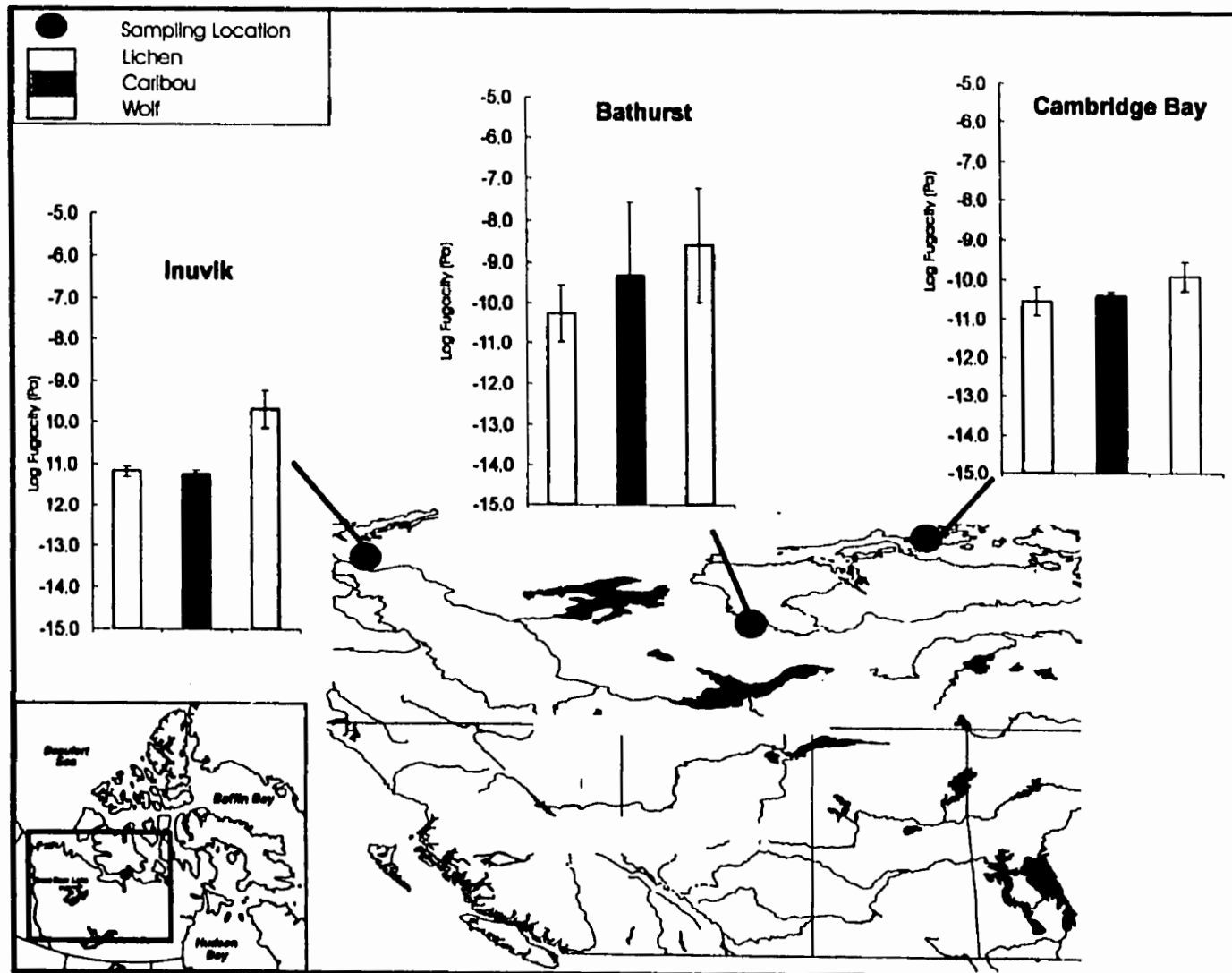


Figure 2.3: Fugacity (Pascals) of total PCBs in the lichen-caribou-wolf food-chains of Canada's central and western Arctic. Bar charts represent the logarithms of geometric means for total PCB fugacities (Pa) in lichens, caribou and wolves. Error bars represent standard deviations of the geometric means.

observed in Cambridge Bay wolves may occur because these wolves rely on other prey species besides caribou for their nutritional requirements. Wolves have been shown to predate on multiple prey species depending on prey availability (Kuyt, 1972; Dale *et al.*, 1994; and Messier, 1994). Alternative prey such as muskox (*Ovibos moschatus*) and Arctic hare (*Lepus arcticus*) may be important dietary components to wolves on Victoria Island. Lower chemical fugacities in these alternative prey species on Victoria Island would explain the reduced chemical fugacities observed in wolves sampled near Cambridge Bay.

In general, fugacity of total PCBs in the lichen-caribou-wolf food chains were greater in samples from the central Arctic compared to samples collected from Inuvik in the western Arctic. Male caribou sampled at Cambridge Bay exhibited significantly higher fugacities of total PCBs than male caribou sampled at Bathurst Inlet ($n=4$, $p= 0.003$, F -value of $20.9 > F_{\text{CRITICAL}}$ of 5.98). The chemical fugacities of total PCBs in the Bathurst male caribou were significantly greater than those in male caribou sampled at Inuvik ($n=4$, $p=0.02$, F -value of $8.5 > F_{\text{CRITICAL}}$ of 5.98). No statistically significant differences in total PCB fugacities were detected between wolves sampled at the three sampling locations ($n=10$, $p=0.32$, F -value of $1.02 < F_{\text{CRITICAL}}$ of 4.4). The chemical fugacities of total PCBs in caribou in Canada's central and western Arctic exhibit a trend where the fugacity in caribou at Inuvik is lowest, while the fugacities in caribou from Cambridge Bay are the highest. Elkin and Bethke (1996) observed decreasing chemical concentrations of organochlorine contaminants in barren-ground caribou from the eastern Arctic

herds (Baffin Island) to those sampled in the western Arctic (Inuvik). This decreasing concentration gradient of organochlorines from east to west has been demonstrated in other terrestrial and marine biota in the Canadian Arctic (Hebert *et al.*, 1996; Weis and Muir 1997). The increased levels of organic contaminants in the eastern Arctic have been attributed to its proximity to industrial regions of eastern North America and the predominate easterly winds that exist over the tundra (Landers *et al.*, 1995).

Bioconcentration in tundra lichens

Table 2.1 summarizes chemical concentrations of organochlorine contaminants observed in lichens, willows and caribou fecal pellets collected near Bathurst Inlet and Cambridge Bay. Also shown in this table are the chemical concentrations of organochlorine contaminants measured in lichens during the Government of Northwest Territories biomonitoring program. The lipid content (% of dry weight) of lichens and caribou fecal pellets collected during the present study were found to be $0.45 \pm 0.14\%$ and $0.97 \pm 0.30\%$, respectively. The organic carbon contents (% dry weight) of lichens and caribou fecal pellets were determined to be $96.1 \pm 0.06\%$ and $84.7 \pm 0.003\%$. The density of lichens and caribou fecal matter were found to be 0.54 ± 0.09 kg/L and 0.86 ± 0.15 kg/L, respectively. The principal organochlorine contaminants in lichen samples were α -HCH, HCB, PCB153,

Table 2.1: Arithmetic means and standard deviations of chemical concentrations (ng/g dry wt.) of organochlorines in three lichen species (*Cladina rangiferina*, *Cladina mitis*, and *Cetraria nivalis*), tundra willow (*Salix glauca*), and caribou fecal pellets collected at sampling locations from the central and western Arctic of Canada.

Bathurst West

Chemical	Brown Sound		S. glauca		C. rangiferina		Bathurst Calving Grounds (Hood River)		n=6
	n=6	n=6	n=6	n=6	n=3	n=3	n=3	n=3	
% Lipids	0.30	1.30							
% Moisture	45.51	47.88							
	C. nivalis	S. glauca	C. rangiferina	C. rangiferina	Caribou	Caribou	Caribou	Caribou	SD
	(summer)	(summer)	(spring)	(spring)	(spring)	(spring)	(summer)	(summer)	SD
1,2,4,5 TCB	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	< 0.03	< 0.03	< 0.03	-
1,2,3,4TCB	< 0.05	< 0.05	0.75	0.75	0.63	0.09	0.63	< 0.03	-
QCB	0.08	< 0.05	0.49	0.49	0.10	0.04	0.10	< 0.03	-
alpha-HCH	2.30	0.75	4.46	4.46	1.04	0.23	1.04	0.66	0.26
beta-HCH	< 0.05	0.09	0.01	0.01	0.09	0.00	0.09	0.12	0.08
gamma-HCH	0.43	0.12	0.76	0.76	0.12	0.04	0.12	0.17	0.09
HCB	0.38	0.08	1.70	1.70	3.93	0.03	3.93	1.64	0.33
OCS	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
Oxychloridane	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
Transchlordanane	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	0.04	0.03
Cischlordanane	0.06	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	0.10	0.02
Transnonachlor	< 0.05	< 0.05	0.11	0.11	0.17	0.03	0.17	0.36	0.57
Cisnonachlor	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	0.06	0.01
p,p' DDE	0.06	< 0.05	2.10	2.10	< 0.08	0.12	< 0.08	0.05	0.03
p,p' DDD	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
p,p' DDT	0.17	< 0.05	0.44	0.44	< 0.08	0.01	< 0.08	< 0.03	-
Photomirex	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
Mirex	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
Heptachlor epoxide	< 0.05	< 0.05	NA	NA	NA	NA	NA	0.04	0.01
Dieldrin	0.12	0.29	NA	NA	NA	NA	NA	0.20	0.05
PCB31	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
PCB28	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	< 0.03	-
PCB52	< 0.05	< 0.05	0.93	0.93	< 0.08	0.07	< 0.08	0.09	0.02
PCB49	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	0.27	0.19
PCB44	< 0.05	< 0.05	< 0.07	< 0.07	< 0.08	-	< 0.08	0.05	0.02

Table 2.1 continued

Chemical	Brown Sound			Bathurst Calving Grounds (Hood River)			
	%Lipids % Moisture	0.30 45.51 n=6 <i>C.nivalis</i> (summer)	1.30 47.88 n=6 <i>S.glauca</i> (summer)	<i>C. rangiferina</i> (spring)	0.89 67.90 (spring) Caribou fecal	1.01 10.46 (summer) Caribou fecal	n=6 SD
PCB42	< 0.05	< 0.05	< 0.05	< 0.07	< 0.08	< 0.03	-
PCB64	< 0.05	< 0.05	< 0.05	< 0.07	< 0.08	< 0.03	-
PCB74	< 0.05	< 0.05	< 0.05	< 0.07	< 0.08	< 0.03	-
PCB70	< 0.05	< 0.05	< 0.05	1.21	< 0.08	< 0.03	-
PCB66/95	< 0.05	< 0.05	< 0.05	1.34	< 0.08	< 0.03	-
PCB60	< 0.05	< 0.05	< 0.05	0.64	< 0.08	< 0.03	-
PCB101	< 0.05	< 0.05	< 0.05	2.52	< 0.08	< 0.03	-
PCB99	< 0.05	< 0.05	< 0.05	1.14	< 0.08	< 0.03	-
PCB97	< 0.05	< 0.05	< 0.05	0.80	< 0.08	< 0.03	-
PCB87	< 0.05	< 0.05	< 0.05	< 0.07	< 0.08	< 0.03	-
PCB110	< 0.05	< 0.05	< 0.05	2.66	< 0.08	< 0.03	-
PCB151	< 0.05	< 0.05	< 0.05	0.57	< 0.08	< 0.03	-
PCB149	0.10	< 0.05	< 0.05	2.30	0.12	< 0.03	0.02
PCB118	< 0.05	< 0.05	< 0.05	3.78	< 0.08	0.09	0.03
PCB146	< 0.05	< 0.05	< 0.05	0.33	< 0.08	< 0.03	-
PCB153	< 0.05	< 0.05	< 0.05	3.33	0.10	< 0.03	0.03
PCB105	< 0.05	< 0.05	< 0.05	1.36	< 0.08	< 0.03	-
PCB141	< 0.05	< 0.05	< 0.05	1.28	< 0.08	< 0.03	-
PCB138	< 0.05	< 0.05	< 0.05	4.97	< 0.08	0.04	0.01
PCB129	< 0.05	< 0.05	< 0.05	0.33	< 0.08	< 0.03	-
PCB162/167	< 0.05	< 0.05	< 0.05	0.40	< 0.08	< 0.03	-
PCB183	< 0.05	< 0.05	< 0.05	0.25	< 0.08	< 0.03	-
PCB185	< 0.05	< 0.05	< 0.05	< 0.07	< 0.08	< 0.03	-
PCB174	< 0.05	< 0.05	< 0.05	0.43	< 0.08	< 0.03	-
PCB171	< 0.05	< 0.05	< 0.05	0.91	< 0.08	< 0.03	-

Table 2.1 continued

Chemical	Brown Sound				Bathurst Calving Grounds (Hood River)					
	0.30 45.51 n=6 <i>C.nivalis</i> (summer)	SD	1.30 47.88 n=6 <i>S.glaucæ</i> (summer)	SD	<i>C. rangiferina</i> (spring)	n=3 SD	0.89 67.90 (spring) Caribou fecal	n=3 SD	1.01 10.46 (summer) Caribou fecal	n=6 SD
PCB200	< 0.05	-	< 0.05	-	0.16	0.01	< 0.08	-	< 0.03	-
PCB172	< 0.05	-	< 0.05	-	< 0.07	-	< 0.08	-	< 0.03	-
PCB180	< 0.05	-	< 0.05	-	1.01	0.04	< 0.08	-	< 0.03	-
PCB170/190	< 0.05	-	< 0.05	-	0.83	0.03	< 0.08	-	< 0.03	-
PCB201	< 0.05	-	< 0.05	-	0.13	0.03	< 0.08	-	< 0.03	-
PCB203	< 0.05	-	< 0.05	-	0.08	0.01	< 0.08	-	< 0.03	-
PCB195	< 0.05	-	< 0.05	-	< 0.07	-	< 0.08	-	< 0.03	-
PCB194	< 0.05	-	< 0.05	-	< 0.07	-	< 0.08	-	< 0.03	-
PCB206	< 0.05	-	< 0.05	-	< 0.07	-	< 0.08	-	< 0.03	-
PCB189	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB77	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB126	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB169	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Total PCBs	2.01	0.04	1.94	-	34.60	1.21	0.44	0.10	0.74	0.21
Arochlor12:54:1260	< 0.05	-	0.51	0.32	67.19	2.76	0.79	0.06	0.54	0.05
Arochlor1250	< 0.05	-	< 0.05	-	8.82	0.39	0.17	0.15	0.09	0.06

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Table 2.1 continued

Bathurst East												
Omingmaktok												
	<i>C. nivalis</i>	n=6	<i>C. rangiferina</i>	n=6	<i>S. glauca</i>	n=6	<i>C. nivalis</i>	n=3	<i>C. rangiferina</i>	n=3	Caribou	(spring) n=3
	(summer)	SD	(summer)	SD	(summer)	SD	(spring)	SD	(spring)	SD	fecal pellets	SD
%Lipids	0.48		0.24		0.78		0.35		0.57		0.89	
% Moisture	8.76		9.21		9.30		81.20		75.56		72.49	
1,2,4,5 TCB	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
1,2,3,4TCB	< 0.05	-	< 0.06	-	< 0.05	-	0.35	0.05	< 0.10	-	0.19	0.01
QCB	0.08	0.07	0.11	0.07	< 0.05	-	0.37	0.06	0.58	0.07	0.12	0.03
alpha-HCH	2.02	1.25	1.19	0.88	0.53	0.07	3.39	0.18	2.20	0.13	1.71	0.02
beta-HCH	< 0.05	-	0.08	0.02	0.11	0.11	0.01	0.00	0.10	0.00	0.23	0.01
gamma-HCH	0.29	0.13	0.24	0.16	0.20	0.03	0.91	0.11	0.66	0.03	0.18	0.00
HCB	0.21	0.05	0.53	0.46	0.10	0.03	2.66	1.44	9.94	0.28	4.06	0.09
OCS	< 0.05	-	< 0.06	-	< 0.05	-	0.10	0.00	< 0.10	-	< 0.08	-
Oxychlordanes	< 0.05	-	< 0.06	-	0.08	0.03	< 0.10	-	< 0.10	-	< 0.08	-
Transchlordanes	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
Cischlordanes	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
Transnonachlor	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	0.12	0.02
Cisnonachlor	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
p,p' DDE	< 0.05	-	0.08	0.03	0.13	0.02	2.43	0.12	1.01	0.04	< 0.08	-
p,p' DDD	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
p,p' DDT	0.10	0.07	< 0.06	-	< 0.05	-	< 0.10	-	0.25	0.04	< 0.08	-
Photomirex	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
Mirex	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
Heptachlor epoxide	< 0.05	-	< 0.06	-	< 0.05	-	NA	NA	NA	NA	NA	NA
Dieldrin	< 0.05	-	0.07	0.03	< 0.05	-	NA	NA	NA	NA	NA	NA
PCB31	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB28	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB52	< 0.05	-	< 0.06	-	< 0.05	-	0.22	0.22	0.89	0.06	< 0.08	-
PCB49	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB44	< 0.05	-	< 0.06	-	< 0.05	-	0.22	0.11	< 0.10	-	< 0.08	-

Table 2.1 continued

	C. nivalis (summer)		C. rangiferina (summer)		S. glauca (summer)		C. nivalis (spring)		Huikitak River .rangiferina n=3 (spring)		Caribou fecal pellets (spring)	
	n=6	SD	n=6	SD	n=6	SD	n=3	SD	n=3	SD	n=3	SD
%Lipids	0.48		0.24		0.78		0.35		0.57		0.89	
% Moisture	8.76		9.21		9.30		81.20		75.56		72.49	
PCB42	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB64	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB74	< 0.05	-	< 0.06	-	0.06	0.02	< 0.10	-	< 0.10	-	< 0.08	-
PCB70	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	0.15	0.09	< 0.08	-
PCB66/95	< 0.05	-	< 0.06	-	< 0.05	-	0.56	0.01	0.62	0.08	< 0.08	-
PCB101	< 0.05	-	< 0.06	-	< 0.05	-	1.62	0.24	1.44	0.08	< 0.08	-
PCB99	< 0.05	-	< 0.06	-	< 0.05	-	0.65	0.14	0.68	0.09	< 0.08	-
PCB97	< 0.05	-	< 0.06	-	< 0.05	-	0.36	0.05	0.56	0.06	< 0.08	-
PCB87	< 0.05	-	< 0.06	-	< 0.05	-	1.32	0.06	< 0.10	-	< 0.08	-
PCB110	< 0.05	-	< 0.06	-	< 0.05	-	1.43	0.10	1.62	0.07	< 0.08	-
PCB151	< 0.05	-	< 0.06	-	< 0.05	-	0.27	0.01	0.39	0.03	< 0.08	-
PCB149	< 0.05	-	< 0.06	-	< 0.05	-	1.14	0.08	1.40	0.15	0.10	0.01
PCB118	< 0.05	-	< 0.06	-	< 0.05	-	1.94	0.25	2.07	0.09	< 0.08	-
PCB146	< 0.05	-	< 0.06	-	< 0.05	-	0.20	0.09	0.14	0.04	0.09	0.02
PCB153	< 0.05	-	< 0.06	-	< 0.05	-	1.95	0.16	1.92	0.21	0.11	0.06
PCB105	< 0.05	-	< 0.06	-	< 0.05	-	0.62	0.06	0.76	0.08	< 0.08	-
PCB141	< 0.05	-	< 0.06	-	< 0.05	-	0.68	0.02	0.72	0.08	< 0.08	-
PCB138	< 0.05	-	< 0.06	-	0.06	0.02	2.66	0.20	2.80	0.30	< 0.08	-
PCB129	< 0.05	-	< 0.06	-	< 0.05	-	0.10	0.00	0.17	0.03	< 0.08	-
PCB182/187	< 0.05	-	< 0.06	-	< 0.05	-	0.29	0.01	0.33	0.02	< 0.08	-
PCB183	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	0.13	0.01	< 0.08	-
PCB185	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB174	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	0.24	0.02	< 0.08	-
PCB171	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	0.46	0.02	< 0.08	-
PCB200	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB172	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-

Table 2.1 continued

	Omingmaktok						Huikitak River				Caribou (spring) n=3	
	<i>C. nivalis</i> (summer)	n=6 SD	<i>C. rangiferina</i> (summer)	n=6 SD	<i>S. glauca</i> (summer)	n=6 SD	<i>C. nivalis</i> (spring)	n=3 SD	<i>C. rangiferina</i> (spring)	n=3 SD	fecal pellets	(spring) n=3 SD
% Lipids	0.48		0.24		0.78		0.35		0.57		0.89	
% Moisture	8.76		9.21		9.30		81.20		75.56		72.49	
PCB180	< 0.05	-	< 0.06	-	< 0.05	-	0.57	0.11	0.57	0.07	< 0.08	-
PCB170/190	< 0.05	-	< 0.06	-	< 0.05	-	0.38	0.03	0.46	0.04	< 0.08	-
PCB201	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	0.24	0.04	< 0.08	-
PCB203	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB195	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB194	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB206	< 0.05	-	< 0.06	-	< 0.05	-	< 0.10	-	< 0.10	-	< 0.08	-
PCB189	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB77	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB126	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB169	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Total PCBs	2.06	0.21	2.32	0.03	1.98	0.02	18.56	1.78	20.68	1.54	0.44	0.21
Arochlor12:54:1260	< 0.05	-	< 0.06	-	0.48	0.53	35.91	2.66	37.88	4.01	0.89	0.22
Arochlor1250	< 0.05	-	< 0.06	-	< 0.05	-	4.96	0.99	4.98	0.61	< 0.08	-

Table 2.1 continued

	Victoria Island		(Cambridge Bay)		
	%Lipids % Moisture	0.25 <i>C. nivalis</i> (Summer) n=6 9.32 (Present Study) SD	(Summer) n=3 <i>C. rangiferina</i> * (Elkin, 1995)	(Summer) n=3 <i>C. mitis</i> * (Elkin, 1995)	(Summer) n=3 <i>C. nivalis</i> * (Elkin, 1995)
1,2,4,5 TCB	< 0.10	-	< 0.01	< 0.01	< 0.01
1,2,3,4TCB	0.09	0.05	0.21	0.09	0.09
QCB	0.15	0.08	0.50	0.34	0.48
alpha-HCH	5.74	3.64	1.10	0.84	3.43
beta-HCH	0.13	0.05	< 0.01	< 0.01	0.01
gamma-HCH	0.80	0.45	0.51	0.44	0.87
HCB	1.42	0.65	1.14	1.95	1.06
OCS	< 0.10	-	0.05	< 0.01	0.05
Oxychlorane	< 0.10	-	< 0.01	< 0.01	0.08
Transchlorane	< 0.10	-	< 0.01	< 0.01	< 0.01
Cischlorane	< 0.10	-	0.09	< 0.01	0.09
Transnonachlor	0.17	0.11	< 0.01	< 0.01	< 0.01
Cisnonachlor	< 0.10	-	0.06	< 0.01	< 0.01
p,p' DDE	< 0.10	-	< 0.01	< 0.01	< 0.01
p,p' DDD	< 0.10	-	0.11	< 0.01	< 0.01
p,p' DDT	0.12	0.06	0.30	< 0.01	0.38
Photomirex	< 0.10	-	< 0.01	< 0.01	< 0.01
Mirex	< 0.10	-	< 0.01	< 0.01	< 0.01
Heptachlor epoxide	0.14	0.02	0.11	< 0.01	0.19
Dieldrin	0.14	0.08	< 0.01	< 0.01	< 0.01
PCB31	< 0.10	-	< 0.01	< 0.01	< 0.01
PCB28	< 0.10	-	< 0.01	< 0.01	< 0.01
PCB52	< 0.10	-	0.15	< 0.01	0.13
PCB49	< 0.10	-	< 0.01	< 0.01	< 0.01
PCB44	0.11	0.03	< 0.01	< 0.01	< 0.01
PCB42	< 0.10	-	< 0.01	< 0.01	< 0.01

Table 2.1 continued

	%Lipids	0.25 <i>C. nivalis</i> (Summer) n=6		SD	(Summer) n=3 <i>C. rangiferina</i> * (Elkin, 1995)	(Summer) n=3 <i>C. mitis</i> * (Elkin, 1995)	(Summer) n=3 <i>C. nivalis</i> * (Elkin, 1995)
		% Moisture	9.32 (Present Study)				
PCB64		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB74		< 0.10	-		0.07	< 0.01	< 0.01
PCB70		< 0.10	-		0.09	< 0.01	0.09
PCB66/95		< 0.10	-		0.10	< 0.01	0.10
PCB60		< 0.10	-		0.01	< 0.01	< 0.01
PCB101		< 0.10	-		< 0.01	< 0.01	0.10
PCB99		< 0.10	-		0.07	< 0.01	0.09
PCB97		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB87		0.11	0.05		< 0.01	< 0.01	< 0.01
PCB110		< 0.10	-		0.08	< 0.01	0.08
PCB151		0.08	0.03		< 0.01	< 0.01	< 0.01
PCB149		0.31	0.18		< 0.01	< 0.01	< 0.01
PCB118		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB146		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB153		< 0.10	-		0.09	< 0.01	0.08
PCB105		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB141		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB138		< 0.10	-		0.10	< 0.01	0.08
PCB129		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB182/187		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB183		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB185		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB174		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB171		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB200		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB172		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB180		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB170/190		< 0.10	-		< 0.01	< 0.01	< 0.01
PCB201		< 0.10	-		< 0.01	< 0.01	< 0.01

Table 2.1 continued

	0.25	C. nivalis (Summer) n=6	(Summer) n=3	(Summer) n=3	(Summer) n=3	(Summer) n=3
%Lipids	9.32 (Present Study)	SD	C. rangiferina* (Elkin, 1995)	C. mitis* (Elkin, 1995)	C. nivalis* (Elkin, 1995)	
PCB203	< 0.10	-	< 0.01	< 0.01	< 0.01	< 0.01
PCB195	< 0.10	-	< 0.01	< 0.01	< 0.01	< 0.01
PCB194	< 0.10	-	< 0.01	< 0.01	< 0.01	< 0.01
PCB206	< 0.10	-	< 0.01	< 0.01	< 0.01	< 0.01
PCB189	NA	NA	< 0.01	< 0.01	< 0.01	< 0.01
PCB77	NA	NA	< 0.01	< 0.01	< 0.01	< 0.01
PCB126	NA	NA	< 0.01	< 0.01	< 0.01	< 0.01
PCB169	NA	NA	< 0.01	< 0.01	< 0.01	< 0.01
Total PCBs	4.04	-	1.19	0.43	1.10	1.10
Arochlor12:54:1260	< 0.10	-	NA	NA	NA	NA
Arochlor1250	< 0.10	-	NA	NA	NA	NA

Table 2.1 continued

Bluenose Range (Inuvik)

	<i>C. mitis</i> (Elkin, 1995) (summer)	SD	<i>C. nivalis</i> (Elkin, 1995) (summer)	SD	<i>C. rangiferina</i> (Elkin, 1995) (summer)	SD
1,2,4,5 TCB	< 0.01	-	< 0.01	-	< 0.01	-
1,2,3,4TCB	0.02	0.02	0.07	0.02	0.08	0.02
QCB	0.21	0.05	0.20	0.03	0.32	0.07
alpha-HCH	1.62	0.20	2.63	0.14	0.56	0.17
beta-HCH	< 0.01	-	< 0.01	-	< 0.01	-
gamma-HCH	0.57	0.02	0.73	0.04	0.19	0.04
HCB	0.43	0.02	0.50	0.02	1.05	0.26
OCS	< 0.01	-	< 0.01	-	< 0.01	-
Oxychlorane	< 0.01	-	0.02	0.02	< 0.01	-
Transchlorane	< 0.01	-	< 0.01	-	< 0.01	-
Cischlorane	0.04	0.03	0.08	0.02	< 0.01	-
Transnonachlor	< 0.01	-	0.08	0.02	< 0.01	-
Cisnonachlor	< 0.01	-	< 0.01	-	< 0.01	-
p,p' DDE	0.08	0.06	0.06	0.01	< 0.01	-
p,p' DDD	< 0.01	-	0.05	0.04	< 0.01	-
p,p' DDT	< 0.01	-	0.18	0.01	< 0.01	-
Photomirex	< 0.01	-	< 0.01	-	< 0.01	-
Mirex	< 0.01	-	< 0.01	-	< 0.01	-
Heptachlor epoxide	< 0.01	-	0.05	0.04	< 0.01	-
Dieldrin	0.04	0.02	0.09	0.03	< 0.01	-
PCB31	< 0.01	-	< 0.01	-	< 0.01	-
PCB28	< 0.01	-	< 0.01	-	< 0.01	-
PCB52	0.07	0.01	0.09	0.01	< 0.01	-
PCB49	< 0.01	-	< 0.01	-	< 0.01	-
PCB44	< 0.01	-	< 0.01	-	< 0.01	-
PCB42	< 0.01	-	< 0.01	-	< 0.01	-

Table 2.1 continued

Inuvik	<i>C. milvis</i> (Elkin, 1995) (summer)	SD	<i>C. nivalis</i> (Elkin, 1999 (summer)	SD	<i>C. rangiferina</i> (Elkin, 1995 (summer)	SD
PCB64	< 0.01	-	< 0.01	-	< 0.01	-
PCB74	< 0.01	-	< 0.01	-	< 0.01	-
PCB70	< 0.01	-	0.03	0.03	< 0.01	-
PCB66/95	0.04	0.02	0.06	0.02	< 0.01	-
PCB60	< 0.01	-	< 0.01	-	< 0.01	-
PCB101	0.02	0.02	0.04	0.03	< 0.01	-
PCB99	< 0.01	-	0.03	0.03	< 0.01	-
PCB97	< 0.01	-	< 0.01	-	< 0.01	-
PCB87	< 0.01	-	< 0.01	-	< 0.01	-
PCB110	< 0.01	-	< 0.01	-	< 0.01	-
PCB151	< 0.01	-	< 0.01	-	< 0.01	-
PCB149	< 0.01	-	0.04	0.03	< 0.01	-
PCB118	< 0.01	-	< 0.01	-	< 0.01	-
PCB146	< 0.01	-	< 0.01	-	< 0.01	-
PCB153	0.03	0.03	0.03	0.03	< 0.01	-
PCB105	< 0.01	-	< 0.01	-	< 0.01	-
PCB141	< 0.01	-	< 0.01	-	< 0.01	-
PCB138	0.04	0.02	0.03	0.03	< 0.01	-
PCB129	< 0.01	-	< 0.01	-	< 0.01	-
PCB 182/187	< 0.01	-	< 0.01	-	< 0.01	-
PCB183	< 0.01	-	< 0.01	-	< 0.01	-
PCB185	< 0.01	-	< 0.01	-	< 0.01	-
PCB174	< 0.01	-	< 0.01	-	< 0.01	-
PCB171	< 0.01	-	< 0.01	-	< 0.01	-
PCB200	< 0.01	-	< 0.01	-	< 0.01	-
PCB172	< 0.01	-	< 0.01	-	< 0.01	-
PCB180	< 0.01	-	< 0.01	-	< 0.01	-
PCB 170/190	< 0.01	-	< 0.01	-	< 0.01	-
PCB201	< 0.01	-	< 0.01	-	< 0.01	-

Table 2.1 continued

Inuvik	<i>C. mitis</i> (Elkin, 1995) (summer)	SD	<i>C. nivalis</i> (Elkin, 199 (summer)	SD	<i>C. rangiferina</i> (Elkin, 1995 (summer)	SD
PCB203	< 0.01	-	< 0.01	-	< 0.01	-
PCB195	< 0.01	-	< 0.01	-	< 0.01	-
PCB194	< 0.01	-	< 0.01	-	< 0.01	-
PCB206	< 0.01	-	< 0.01	-	< 0.01	-
PCB189	< 0.01	-	< 0.01	-	< 0.01	-
PCB77	< 0.01	-	< 0.01	-	< 0.01	-
PCB126	< 0.01	-	< 0.01	-	< 0.01	-
PCB169	< 0.01	-	< 0.01	-	< 0.01	-
Total PCBs	0.57	0.07	0.69	0.19	0.43	-
Arochlor1254:1260	NA	NA	NA	NA	NA	NA
Arochlor1250	NA	NA	NA	NA	NA	NA

Table 2.1 continued

Bathurst Range July 1992 (Elkin and Bethke)

	<i>C. mitis</i> (Elkin, 1995)	SD	<i>C. nivalis</i> (Elkin, 1995)	SD	<i>C. rangiferin</i> (Elkin, 1995)	SD
1,2,4,5 TCB	< 0.01	-	0.03	0.02	0.02	0.02
1,2,3,4TCB	0.08	0.09	0.03	0.03	0.11	0.02
QCB	0.32	0.09	0.16	0.13	0.31	0.17
alpha-HCH	1.51	0.15	2.52	1.46	0.55	0.19
beta-HCH	< 0.01	-	< 0.01	-	< 0.01	-
gamma-HCH	0.65	0.06	1.28	1.19	0.24	0.10
HCB	0.36	0.01	0.79	0.89	0.82	0.28
OCS	< 0.01	-	< 0.01	-	< 0.01	-
Oxychlorane	< 0.01	-	< 0.01	-	< 0.01	-
Transchlorane	< 0.01	-	0.04	0.04	< 0.01	-
Cischlorane	< 0.01	-	0.05	0.06	< 0.01	-
Transnonachlor	< 0.01	-	0.04	0.05	0.02	0.01
Cisnonachlor	0.03	0.03	0.04	0.04	0.01	0.00
p,p' DDE	0.06	0.07	0.04	0.06	0.02	0.01
p,p' DDD	0.07	0.08	0.14	0.12	0.02	0.01
p,p' DDT	0.26	0.35	0.50	0.41	0.05	0.06
Photomirex	< 0.01	-	< 0.01	-	0.18	0.22
Mirex	< 0.01	-	< 0.01	-	< 0.01	-
Heptachlor epoxide	< 0.01	-	0.06	0.04	< 0.01	0.01
Dieldrin	< 0.01	-	0.02	0.02	< 0.01	-
PCB31	< 0.01	-	< 0.01	-	< 0.01	-
PCB28	< 0.01	-	< 0.01	-	< 0.01	-
PCB52	0.07	0.08	0.03	0.03	0.04	0.04
PCB49	< 0.01	-	< 0.01	-	< 0.01	-
PCB44	< 0.01	-	< 0.01	-	< 0.01	-
PCB42	< 0.01	-	< 0.01	-	< 0.01	-

Table 2.1 continued

Bathurst	<i>C. milis</i> (Elkin, 1995)	SD	<i>C. nivalis</i> (Elkin, 1995)	SD	<i>C. rangiferin</i> (Elkin, 1995)	SD
PCB64	< 0.01	-	< 0.01	-	< 0.01	-
PCB74	< 0.01	-	0.02	0.02	0.02	0.03
PCB70	0.04	0.04	0.05	0.04	0.06	0.05
PCB66/95	0.04	0.04	0.06	0.04	0.06	0.04
PCB60	< 0.01	-	< 0.01	-	< 0.01	-
PCB101	0.04	0.04	0.04	0.05	0.05	0.05
PCB99	< 0.01	-	0.03	0.02	0.02	0.02
PCB97	< 0.01	-	< 0.01	-	0.02	0.02
PCB87	< 0.01	-	< 0.01	-	0.03	0.03
PCB110	< 0.01	-	0.02	0.02	0.03	0.05
PCB151	< 0.01	-	0.04	0.03	< 0.01	-
PCB149	< 0.01	-	0.04	0.03	< 0.01	-
PCB118	0.03	0.03	0.02	0.01	< 0.01	-
PCB146	< 0.01	-	< 0.01	-	< 0.01	-
PCB153	0.04	0.04	0.05	0.03	0.02	0.01
PCB105	< 0.01	-	< 0.01	-	< 0.01	-
PCB141	< 0.01	-	< 0.01	-	< 0.01	-
PCB138	0.07	0.01	0.04	0.03	0.02	0.02
PCB129	< 0.01	-	< 0.01	-	< 0.01	-
PCB182/187	< 0.01	-	0.02	0.01	< 0.01	-
PCB183	< 0.01	-	< 0.01	-	< 0.01	-
PCB185	< 0.01	-	< 0.01	-	< 0.01	-
PCB174	< 0.01	-	< 0.01	-	< 0.01	-
PCB171	< 0.01	-	< 0.01	-	< 0.01	-
PCB200	< 0.01	-	< 0.01	-	< 0.01	-
PCB172	< 0.01	-	< 0.01	-	< 0.01	-
PCB180	0.03	0.03	0.02	0.01	< 0.01	-
PCB170/190	< 0.01	-	< 0.01	-	< 0.01	-
PCB201	< 0.01	-	< 0.01	-	< 0.01	-

Table 2.1 continued

Bathurst	<i>C. mitis</i> (Elkin, 1995)	SD	<i>C. nivalis</i> (Elkin, 1995)	SD	<i>C. rangiferin</i> (Elkin, 1995)	SD
PCB203	< 0.01	-	< 0.01	-	< 0.01	-
PCB195	< 0.01	-	< 0.01	-	< 0.01	-
PCB194	< 0.01	-	< 0.01	-	< 0.01	-
PCB206	< 0.01	-	< 0.01	-	< 0.01	-
PCB189	< 0.01	-	< 0.01	-	< 0.01	-
PCB77	< 0.01	-	< 0.01	-	< 0.01	-
PCB126	< 0.01	-	< 0.01	-	< 0.01	-
PCB169	< 0.01	-	< 0.01	-	< 0.01	-
Total PCBs	0.70	0.18	0.76	0.26	0.69	0.30
Arochlor12:54:1260	NA	NA	NA	NA	NA	NA
Arochlor1250	NA	NA	NA	NA	NA	NA

PCB138, PCB110, PCB101, PCB118, PCB170/190 and PCB66/95. Figure 2.4 illustrates the logarithms of fugacities of α -HCH, HCB and total PCBs in two common tundra lichens (*Cladina rangiferina* and *Cetraria nivalis*) collected at different locations within the study area.

Results from a one-way Analysis of Variance (ANOVA) confirmed a significant increase in chemical fugacities of alpha-HCH, HCB and total PCBs in lichen samples (*C. rangiferina* and *C. nivalis*) collected during the 1997 spring snowmelt period (See Figure 2.4 for details). The increased chemical fugacities observed in lichens collected during spring over those collected in the summer may be representative of increased contaminant loading from snowpack meltwater. The environmental fate of contaminants associated with a snowpack is determined by the chemical release back into the atmosphere (volatilization) and uptake by underlying vegetation (bioconcentration). For less volatile chemicals such as PCBs, chemical loss to the atmosphere by volatilization may be small. Thus, snowpacks may accumulate significant quantities of non-volatile chemicals. Snowfall deposition and snowpack accumulation can vary significantly between regions and even closely situated locations. Regional weather patterns (precipitation, wind) and topography (eskers, lee side slopes) may play important roles in the fate of environmental contaminants associated with snow. Geographic distributions of airborne chemicals may then be affected by differences in regional loadings due to variability of snowfall patterns. Regional variations of snow

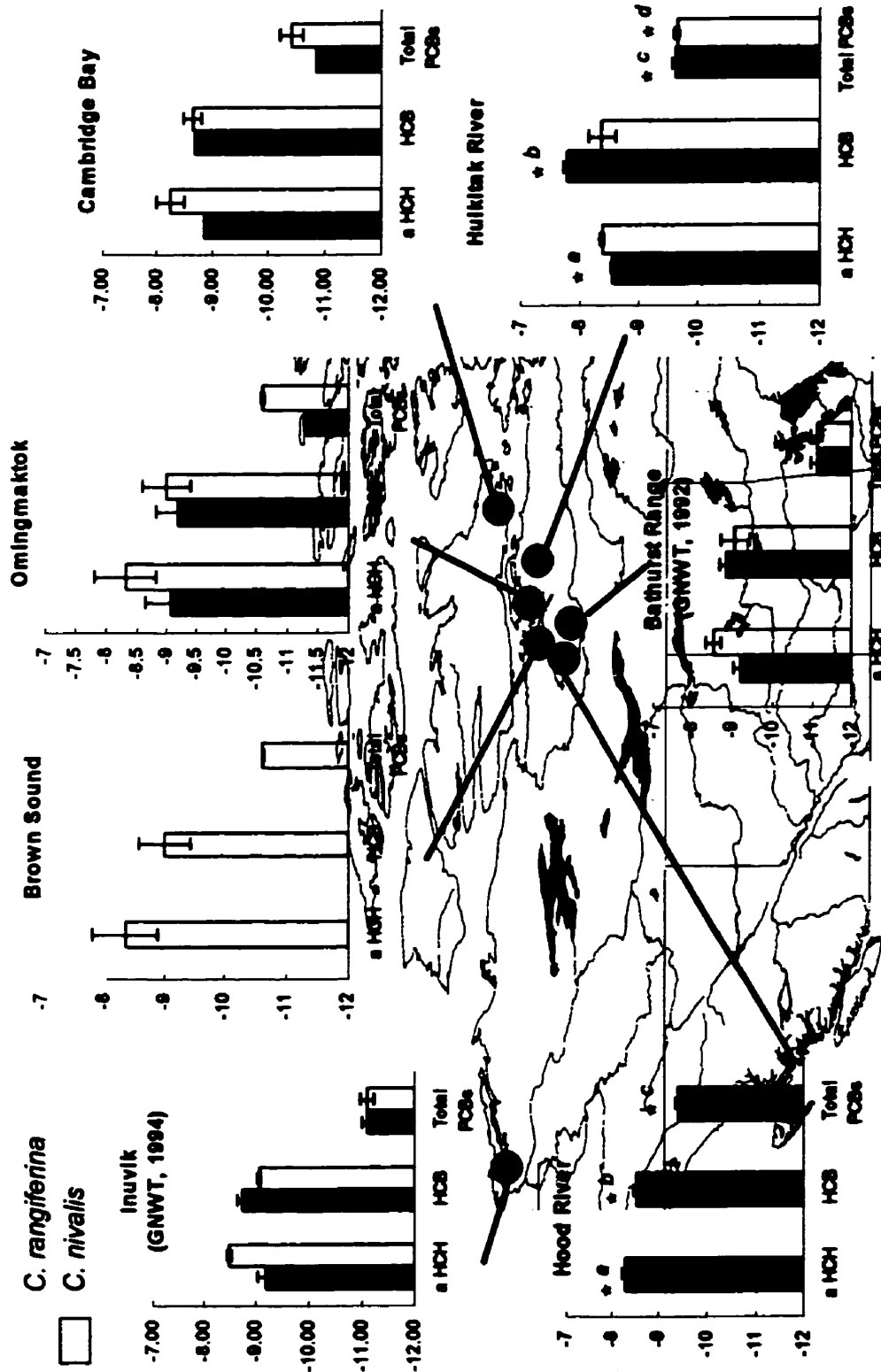


Figure 2.4: Logarithms of fugacities (Pa) of HCB, total PCBs and alpha-HCH in two common lichen species (*Cleidea rangiferina* and *Cetraria nivalis*) at sampling locations in the central and western Arctic. Data are reported as geometric means. Error bars represent the standard deviations of the geometric means. * indicates statistical significance at $\alpha = 0.05$ between mean chemical fugacities in lichens collected in spring compared to lichens collected in summer. ^a alpha-HCH in *C. rangiferina* significant for $n=6$, F-value (23.9) > Fcritical(5.12), $P=0.008$. ^b HCB in *C. rangiferina* significant for $n=6$, F-value (10.12) > Fcritical (5.32), $p = 0.01$. ^c Total PCBs in *C. rangiferina* significant for ($n=6$, F-value (59.1) > Fcritical (5.11), $P = 0.00003$. ^d statistically significant for Total PCBs in *C. nivalis* for $n=3$, F-value(237.9) > Fcritical (7.71) at $P = 0.0001$.

accumulation across the Arctic tundra could result in different exposure levels to lichens and hence influence lichen bioconcentration patterns. Geographic variability of chemical concentrations in tundra lichens would suggest that caribou may be exposed to various levels of contaminants during migratory grazing periods. Spatial and temporal changes in dietary exposure levels should be considered to fully assess chemical bioaccumulation in migratory species such as barren-ground caribou.

Table 2.2 summarizes the observed chemical concentrations of Σ HCHs, HCB, Σ DDTs, Σ Chlordanes and total PCBs measured in air, snow and lichens sampled from Canada's central Arctic region. The octanol-air partition coefficients (K_{OA}) for these chemicals were calculated by dividing the chemical's octanol-water partition coefficient (K_{OW}) by the dimensionless air-water partition coefficient (K_{AW}). Mean annual atmospheric concentrations of Σ HCHs (91.3 pg/m^3) and HCB (52.7 pg/m^3) in Arctic air were higher than atmospheric concentrations of Σ DDTs (1.4 pg/m^3), Σ Chlordanes (5.6 pg/m^3) and total PCBs (18.9 pg/m^3). Despite lower concentrations in Arctic air, total PCBs measured in snowfall and snowpacks from the central Arctic were greater than concentrations of Σ HCHs, HCB, Σ DDTs and Σ Chlordanes in these media. The chemical concentration of total PCBs in meltwater ($4.1 \pm 2.9 \text{ ng/L}$), exceed concentrations of the other compounds by at least a factor of 5. The chemical concentrations in the spring meltwater were calculated from observed chemical concentrations measured in snowpacks from

Table 2.2: Observed chemical concentrations in air, snowfall, snowpack, lichens in the Central Arctic. Values in brackets represent the standard deviations of the observed data. Chemical concentrations in meltwater were calculated from observed concentrations in the snowpack by using a snowpack density of 0.9 kg/L.

Chemicals *	Log K _{ow}	Air (pg/m ³)	Snowfall (ng/L)	Snowpack (ng/L)	Meltwater (ng/L)	Lichen (May) (ng/g dry wt.)	Lichen(July) (ng/g dry wt.)
ΣHCH K _{ow} = 4 H = 2	7.6	Mean 91.30 Min 27.60 Max 178.0	0.95 (0.30)	0.76 (0.93)	0.84 (1.03)	5.22 (1.94)	1.51 (1.29)
HCB K _{ow} = 5.5 H = 65	7.1	Mean 52.7 Min 12.6 Max 196.0	0.1 (0.07)	0.16 (0.1)	0.18 (0.11)	5.5 (3.34)	0.53 (0.46)
ΣDDTs K _{ow} = 6.1 H = 15	8.7	Mean 1.4 Min 0.3 Max 3.2	0.24 (0.20)	0.09 (0.11)	0.04 (0.12)	2.61 (0.67)	0.28 (0.24)
ΣChlordanes K _{ow} = 6.3 H = 18	8.8	Mean 5.6 Min 1.9 Max 10.4	0.31 (0.32)	0.04 (0.05)	0.10 (0.11)	0.5 (0.12)	0.27 (0.10)
Total PCBs K _{ow} = 6.9 H = 10	9.1	Mean 18.9 Min 7.6 Max 41.2	2.1 (0.66)	4.1 (2.9)	4.56 (3.22)	34.6 (1.21)	2.32 (0.98)

* HCHs are comprised of alpha-HCH, beta-HCH and gamma-HCH. ΣDDTs consist of p' p' DDT, DDE and DDD. ΣChlordanes is the sum of the oxychlordane, transchlordane, cischlordane, transnonachlor and cisnonachlor. Total PCBs represent the sum of 43 PCB congeners analyzed in these media.

the central Arctic. The density of a snowpack (approximately 0.9 kg/L) was used to convert the chemical concentration in the snowpack to a meltwater equivalent concentration (ng/L water). With the exception of Σ Chlordanes, chemical concentrations in lichens collected during a spring snowmelt period (May-June, 1997) exceeded the chemical concentrations observed in summer collected lichens by at least a factor of 3. The increased chemical concentrations observed in lichens collected during spring may be the result of increased bioconcentration of chemical associated with meltwater during the spring snowmelt period. As temperatures rises in spring, water associated with snowpacks slowly evaporates, resulting in a smaller volume of water (i. e., meltwater). This transformation can cause non-volatile chemicals previously stored in the snowpack to be discharged with the spring meltwater.

The volumetric bioconcentration factors (BCF_V) (i.e., C_{LICHEN}/C_{AIR}), calculated for Σ HCH, total PCBs, HCB, Σ DDT and Σ Chlordanes in lichens increase with increasing octanol-air partition coefficient (K_{OA}) (Figure 2.5). The BCF_V is calculated as ratio of the chemical concentration in lichens (mol/m^3) to the concentration of chemical in the atmosphere (mol/m^3). BCF_V 's for lichens in spring were calculated from chemical concentrations observed in lichens collected from Bathurst Inlet in this study. To calculate BCF_V 's for summer lichens, chemical concentrations in lichens collected near Bathurst Inlet from this study were combined with concentrations in lichens observed by the Government of the

Atmospheric Partitioning of Organic Contaminants in Lichens from the Central Arctic

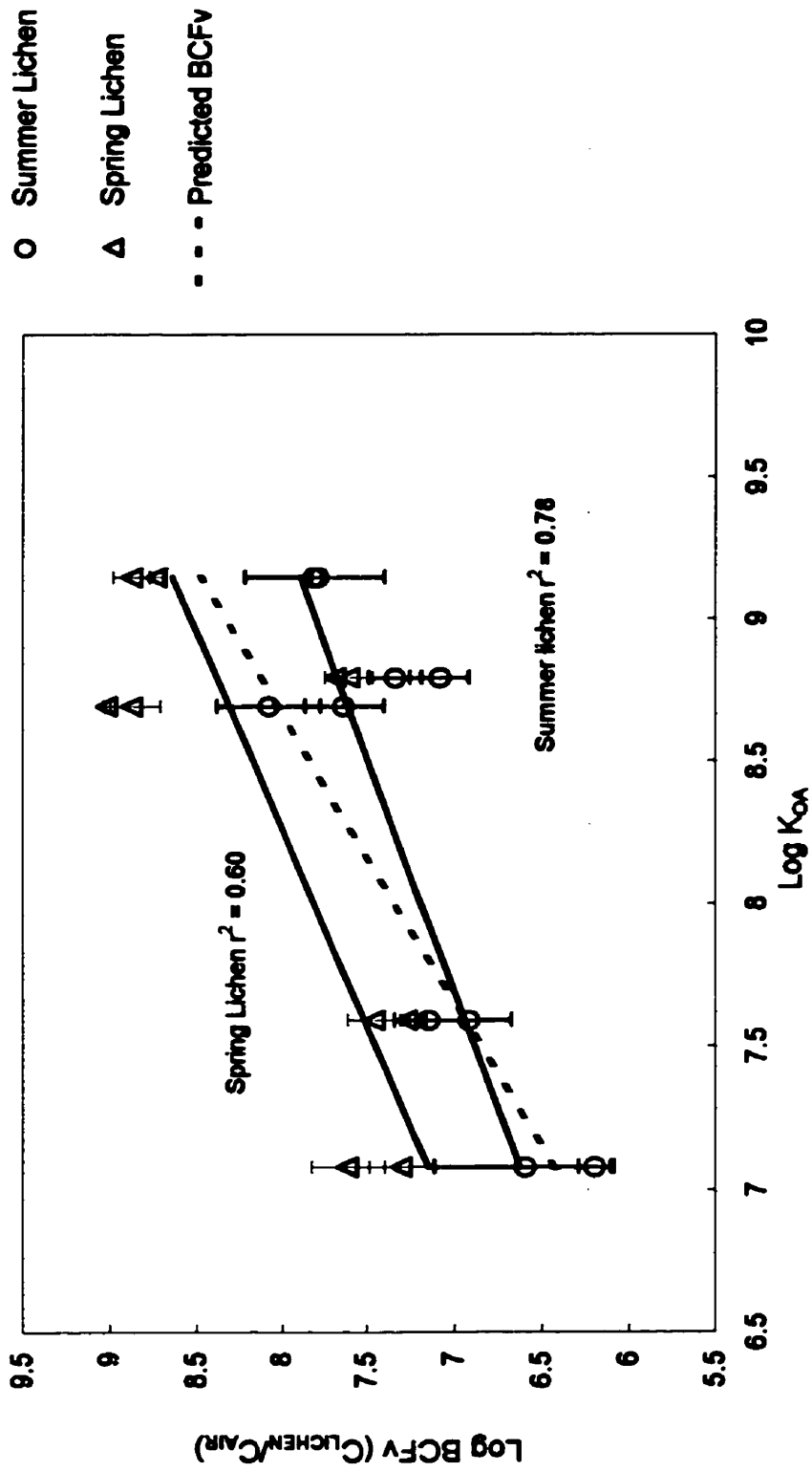


Figure 2.5: Volumetric bioconcentration factors (BCFv) in lichens versus the chemical's octanol-air partition coefficient (K_{OA}). BCFv's are calculated as the ratio of the chemical concentrations in lichens (mol/m^3) to chemical concentrations in air (mol/m^3). Error bars represent the standard deviations of chemical concentrations in lichens.

Northwest Territories. The observed BCF_V 's in lichens collected during summer near Bathurst Inlet are similar to the predicted BCF_V 's under the assumption of equilibrium (or equifugacity) between lichens and air. At equilibrium, the chemical fugacities in lichens are equal to those fugacities in the air, and the chemical concentration in lichens is equal to the product of the fugacity in air and the fugacity capacity of lichens (i. e., $C_{LICHEN} = f_A \cdot Z_{LICHEN}$). Thus, the predicted BCF_V 's in lichens at equilibrium with air are calculated by this concentration in lichens at equilibrium (C_{LICHEN} , mol/m³) divided by the chemical concentration in air (C_A , mol/m³). It is reasonable to expect that lichens exposed to Arctic air during summer months have reached a chemical equilibrium with ambient atmospheric concentrations. Observed BCF_V 's in lichens collected during spring snowmelt were higher than the BCF_V 's in summer lichens. The elevated BCF_V 's in spring lichens are supported by the statistically significant differences ($p < 0.05$) between the mean chemical fugacities in lichens collected in spring and lichens collected in summer. The increase of BCF_V 's in spring collected lichens above the predicted BCF_V 's suggests a chemical dis-equilibrium between lichens and the atmosphere may occur during spring runoff events. It is conceivable that lichens may bioconcentrate organic contaminants associated with surficial spring runoff.

Snowpack accumulation on the Arctic tundra can be the result of several months of snowfall events. Airborne contaminants scavenged by snowfall may accumulate in snowpacks, increasing in quantity throughout the winter months. As summer approaches and temperatures increase, the snowpack begins to slowly evaporate. During evaporation of a snowpack, organic chemicals may be retained in the snowpack until it is discharged as meltwater. Once associated with the snowpack, the volatility of the chemical substance will influence the degree of volatilization to the air, and hence its quantity in the snowpack.

The octanol-air partition coefficient (K_{OA}) of a chemical is representative of its atmospheric partitioning behaviour. The chemical volatility decreases with increasing K_{OA} . Thus, chemicals that exhibit greater K_{OA} 's are less volatile and may accumulate more in snowpacks than volatile chemicals. It is hypothesized that the quantity of chemical retained in a snowpack and the compositional change of snow to meltwater during evaporation may increase chemical fugacities in spring runoff. The elevated fugacities in the meltwater may result in net chemical transport into underlying lichens. To investigate this hypothesis it is important to compare chemical fugacities in air, spring meltwater and lichens during summer and spring snowmelt periods. This analysis may elicit a better understanding of seasonal bioconcentration of organic contaminants in lichens.

Figure 2.6 illustrates the logarithms of chemical fugacities in air, spring meltwater and lichens during summer and spring near Bathurst Inlet for Σ HCHs, HCB, Σ DDTs, Σ Chlordanes and total PCBs. Chemical fugacities of these compounds in Arctic air are represented by mean annual fugacities (\pm min/max fugacities, Pa) in the central Arctic region. Chemical fugacities observed in Arctic air are comparable to those fugacities observed in lichens collected near Bathurst Inlet during summer. The chemical fugacities of Σ HCHs, HCB, Σ DDTs, and total PCBs in lichens collected in spring are shown to be greater than fugacities in lichens collected in summer and Arctic air. The increased chemical fugacities observed in lichens collected in spring coincide with fugacity increases in meltwater that exceed fugacities in Arctic air and lichens collected in summer. The elevated chemical fugacities in spring meltwater above the fugacities in snow-covered lichens would provide fugacity gradients by which chemical transport from meltwater to lichens could occur during a spring snowmelt event. The fugacity of total PCBs in meltwater were 2 orders of magnitude higher than the fugacity of total PCBs observed in lichens collected during the spring snowmelt period.

The assumption that all chemical stored in a snowpack will be dissolved during a spring runoff may somewhat over-predict the chemical fugacity in meltwater.

PCBs are hydrophobic organic substances that exhibit low volatility and a high degree of environmental persistence once associated with environmental media. It has previously been recognized that PCBs and other hydrophobic contaminants

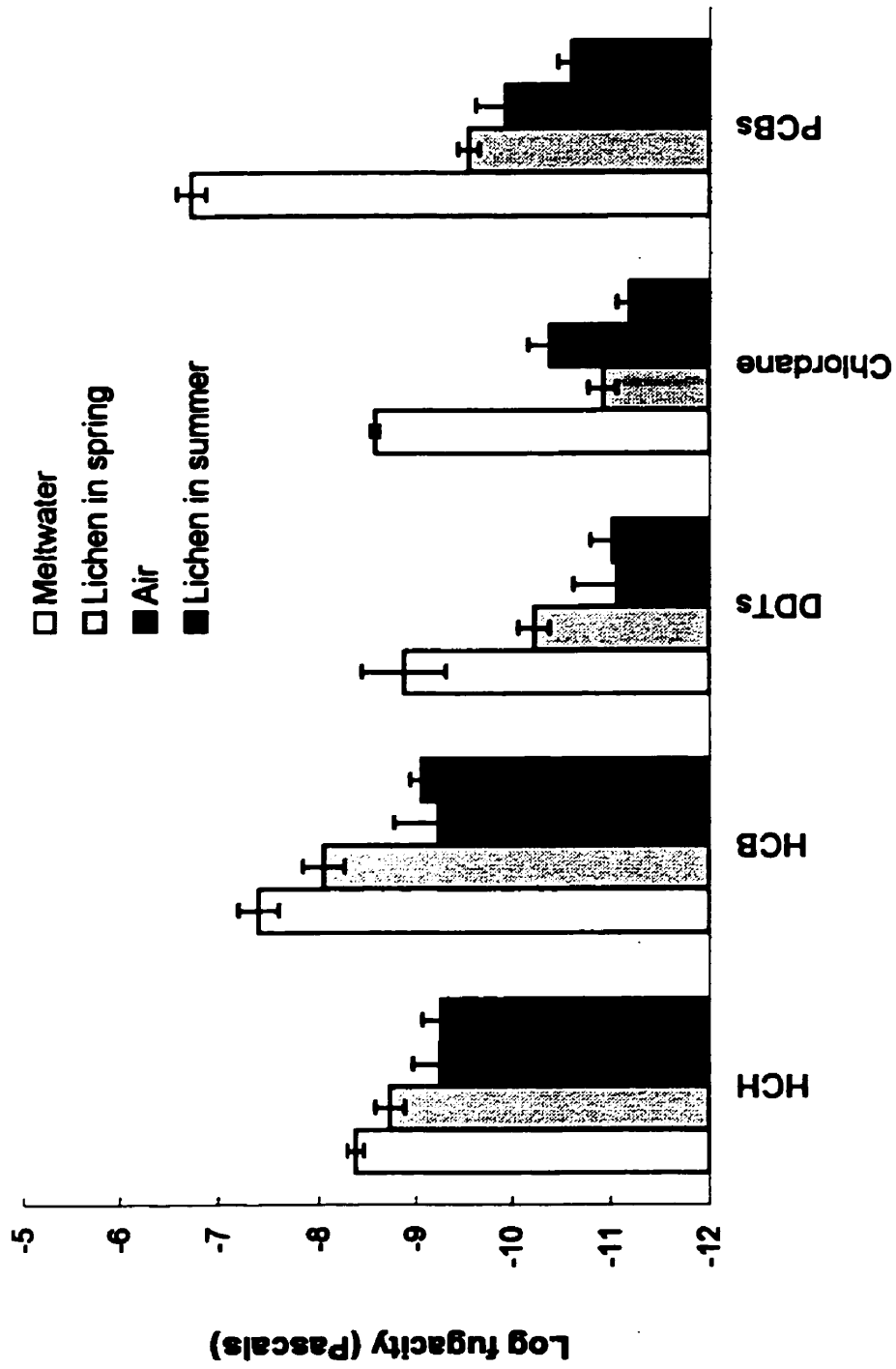


Figure 2.6: Logarithms of chemical fugacities (in units of Pa) of HCH, HCB, DDTs, Chlordanes and PCBs in air (f_{AIR}), meltwater ($f_{MELT\ WATER}$) and lichens (f_{LICHEN}). *C. rangiferina* and *C. nivalis* in summer and spring. Error bars represent standard deviations of the logarithmic data. Chemical fugacities in lichens were calculated from observed concentrations in lichens from all sampling locations during summer and spring collections.

can be associated with particulate matter during meltwater discharge periods (Barrie *et al.*, 1992). An increased fraction of chemical associated with particulate matter within a snowpack would decrease the amount of chemical dissolved in meltwater, and hence result in a lower chemical fugacity in meltwater. Chemical bound to particulate matter may be transported to plant surfaces by particulate adsorption. In this situation, the increased chemical concentration in lichens would be the result of increased surficial adsorption of chemical rather than an increased fugacity.

Bioaccumulation in terrestrial wildlife

The chemical concentrations (ng/g lipid) of organic contaminants in liver, muscle and fat tissues of caribou and wolves from Bathurst Inlet, Cambridge Bay and Inuvik are summarized in Appendix I. The lipid normalized concentrations for most of the organochlorine chemicals analyzed in different tissues of caribou and wolves were comparable, generally within a factor of 2. This suggests that the chemical fugacities in liver, muscle and fat tissues of caribou and wolves are equal. Since the chemical fugacities are the same for these tissues, we can assume that the chemical fugacity in caribou and wolves is uniform. The corresponding chemical fugacities (Pa) in the lichen-caribou-wolf food-chains from Bathurst Inlet, Cambridge Bay and Inuvik are shown in Appendix II.

Fugacities of total PCBs for individual caribou and wolves collected from Cambridge Bay and Bathurst Inlet are shown in Figure 2.7 to illustrate relationships of age, sex and season on chemical bioaccumulation. The fugacity of total PCBs in female caribou of different age-classes at Cambridge Bay demonstrated a trend in which animals < 3 years attained higher fugacities than animals over the age of 5. No statistical inference regarding the effect of age on bioaccumulation in these animals was possible due to the limited number of samples within each age class. However, the lower fugacities of total PCBs observed in older female caribou, compared to animals less than 3 years may represent differences in lactational excretion between these age-classes. Conversely, the higher fugacities of total PCBs in younger animals may be the result of increased chemical exposure from ingesting milk while nursing. Chemical transfer via milk ingestion by nursing newborns is an age-specific mechanism that has been shown to influence chemical bioaccumulation in mammals (Borrell *et al.*, 1995).

The chemical fugacities of total PCBs in male caribou collected near Bathurst Inlet in July and September were 4.7×10^{-10} Pa (range of 1 SD = 1.78×10^{-12} to 2.3×10^{-8} Pa) and 1.7×10^{-11} Pa (range of 1SD = 7.8×10^{-12} to 2.6×10^{-11} Pa), respectively. Results of a one-way ANOVA showed fugacities of total PCBs in male caribou were significantly greater ($n = 5$, F-value (13.9) > F_{critical} (5.4) and p -value of 0.009 at a significance level of $\alpha = 0.05$) in animals collected in July

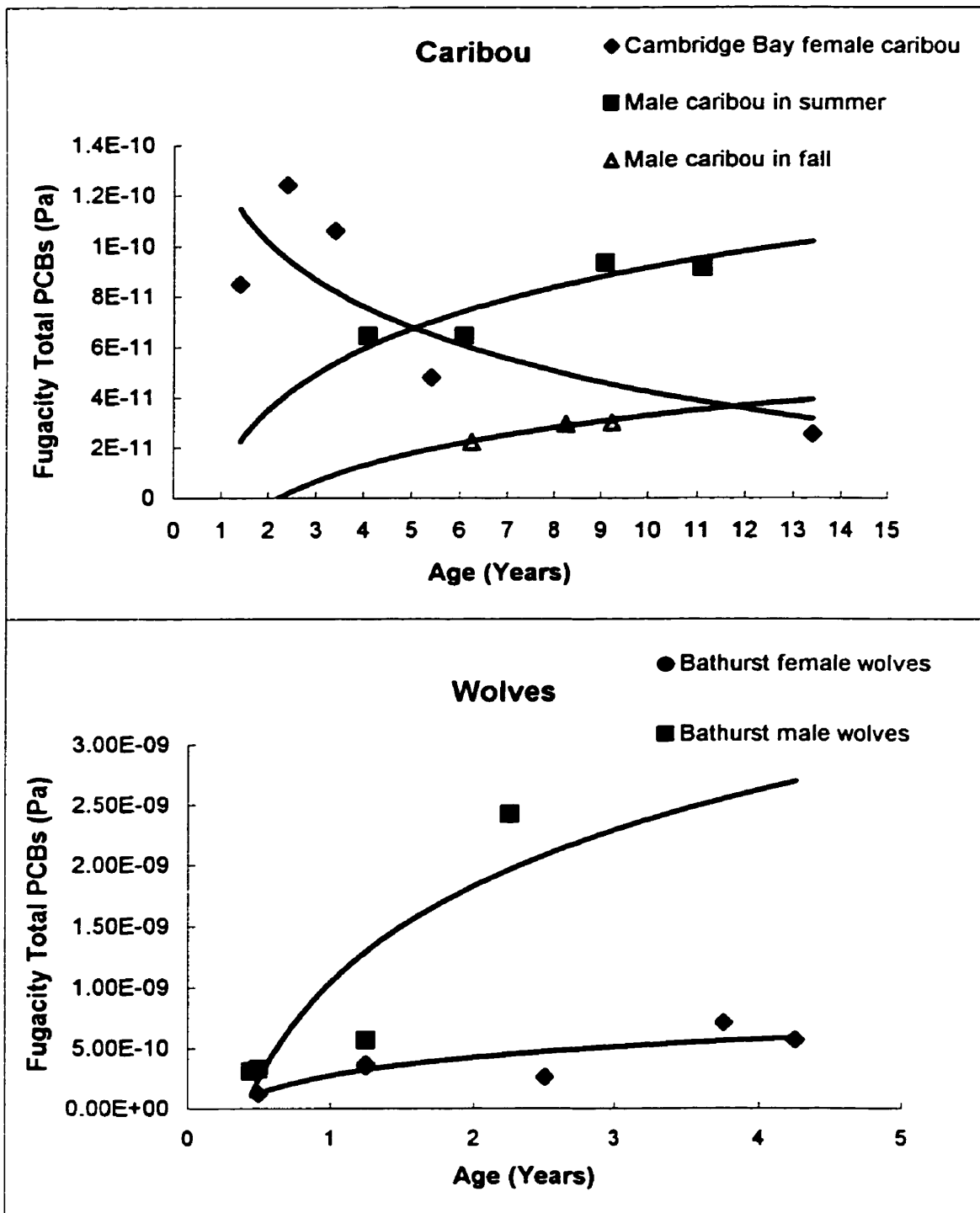


Figure 2.7: Relationships between age, sex and season on chemical fugacities (Pa) of total PCBs in caribou and wolves. Fugacities of total PCBs in male caribou collected on the Bathurst range in July (n=5) were significantly greater ($P < 0.05$) than male animals collected September.

compared to fugacities in male sampled in September. Caribou experience seasonal fluctuations in body fat, exhibiting high fat content in late fall and low fat content in early summer. It seems that male caribou can exhibit significantly greater chemical fugacities in summer, during periods when fat reserves are depleted after the previous winter. The reduced fat content during summer months would concentrate the chemical stored within the animal's body fat. This increased chemical concentration (ng/g lipid) corresponds to an increased chemical fugacity in animals during summer months. The lower fugacities observed in female caribou in July compared to fugacities in males collected during July may be due to chemical elimination by lactating females during the calving season (i. e., early June). An important distinction between male and female mammals is the lactational elimination of chemical by females. This additional elimination mechanism specific to lactating females has been used to explain lower levels of POPs in female mammals (Norstrom *et al.*, 1988; Borrell *et al.*, 1995). No significant differences ($n = 5$, F-value (2.57) < F_{Critical} (5.3) and p -value of 0.14 at a significance level of $\alpha = 0.05$) between fugacities of total PCBs in male and female caribou sampled in September were detected.

For Bathurst wolves, no statistical differences ($n = 4$, F-value (1.19) < F_{Critical} (5.98) and p -value of 0.31 at a significance level of $\alpha = 0.05$) between total PCB fugacities in male and females were detected. Due to small sample sizes, no statistical analyses testing for differences in chemical bioaccumulation between wolves of different ages were conducted. However, the trend in these data

suggest that older wolves seem to attain higher fugacities of total PCB compared to fugacities observed in younger animals from the same region.

The age and sex of an animal within a dynamic ecological population are two important factors affecting the bioaccumulation of organic contaminants. Because age and sex specific mechanisms such as fat production and depletion, nursing and lactation can influence life-time chemical bioaccumulation in terrestrial mammals they should be considered when assessing bioaccumulation in wildlife populations. Age-specific prey selection can also affect chemical bioaccumulation in both males and females of a population. Other seasonal effects, such as fasting and hibernation may affect internal pharmacokinetics, and hence may influence chemical bioaccumulation. Fasting and hibernation involve the utilization of fat reserves by the organism during times of food shortages. As fat reserves in the organism are depleted, the chemical concentrations can increase, thus elevating the chemical fugacity. This elevated fugacity may result in net passive diffusion of chemical into other tissues of the organism (i. e., muscle, liver, heart, etc).

In both caribou and wolves, oxychlordanes contributed a higher percentage to total chlordanes components than *trans*-nonachlor. The increased proportion of oxychlordanes to *trans*-nonachlor in tissues of caribou and wolves suggest metabolic transformation of chlordanes. Some organisms have the capacity to metabolize xenobiotic substances such organochlorine contaminants. This metabolic capacity is related to the presence and activity of cytochrome enzymes. In particular,

cytochrome CPY450 2B and CPY450 1A enzyme activity are responsible for the metabolic transformation of most POPs in terrestrial mammals. Because oxychlordan is a persistent metabolite of chlordan and nonachlor compounds, an increased amount of oxychlordan relative to *trans*-nonachlor indicates the presence of metabolic activity of cytochrome CPY 450 2B.

Figure 2.8 shows the proportion of specific PCB congeners to Total PCBs measured in lichens, caribou and wolf tissues from the Bathurst Inlet. PCB congeners 31 to 206 on the x-axis are ordered by PCB IUPAC number, which increases with increasing chlorination (i. e., homologue sequences: trichlorinated biphenyls -3 chlorines to nonachlorinatedbiphenyls-9 chlorines). An increasing number of chlorine molecules on PCB compounds is associated with increased hydrophobicity and molecular weight. These PCB congener profiles show that lichens, caribou and wolves exhibit congener specific bioaccumulation. Metabolism of specific PCB congeners with vicinal hydrogen atoms at the *meta-para* positions by CPY450 2B and CPY450 1A enzymes has been reported in various fish and mammalian species (Muir *et al.*, 1988; Norstrom and Muir, 1994). As a result, species-specific metabolic transformation of certain substances can alter chemical profiles in tissues of organisms at different trophic levels. PCB congener profiles in wolves demonstrate fewer PCB compounds compared to the number of congeners in

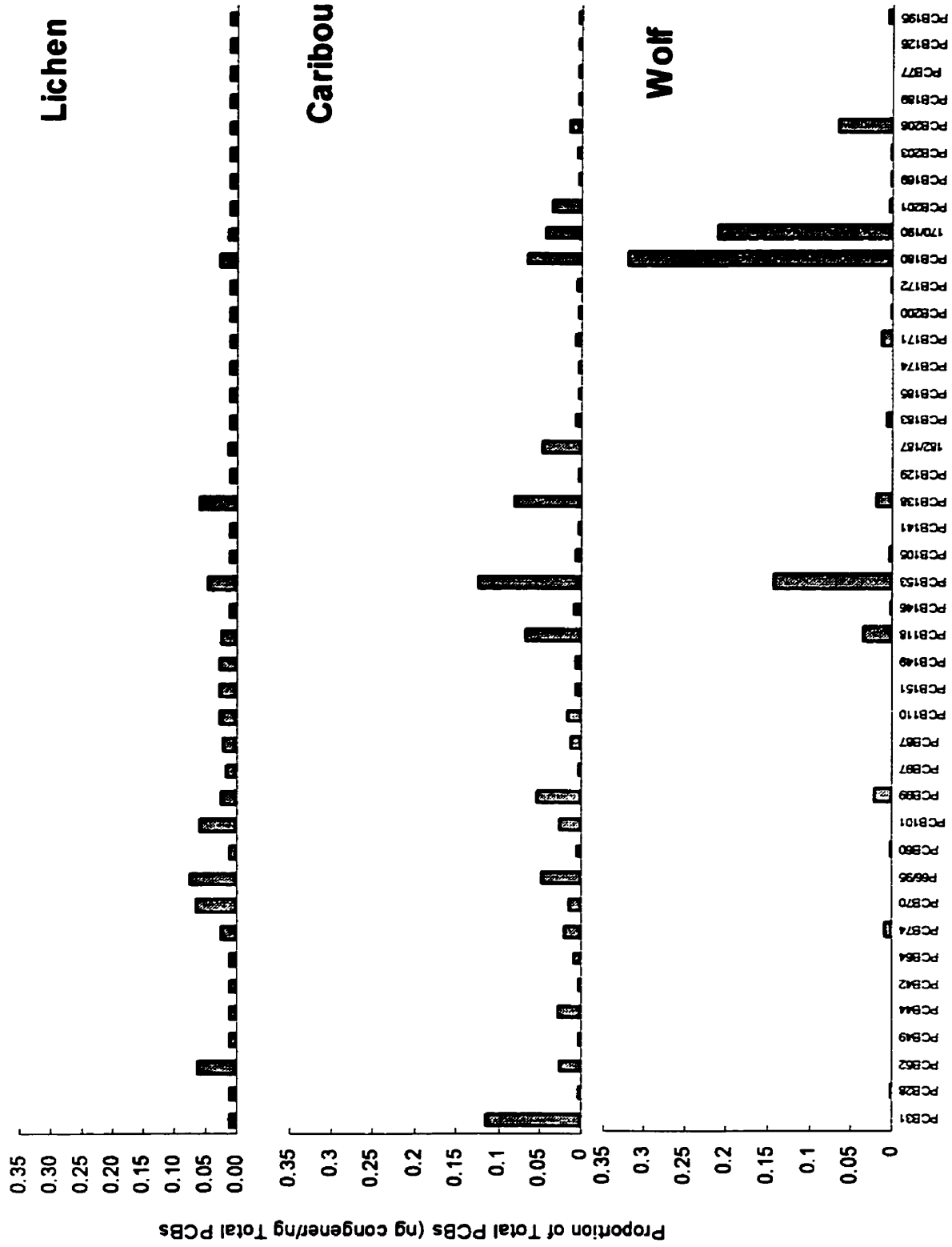


Figure 2.8: Composition of the mixture of polychlorinated biphenyl congeners in the lichen-caribou-wolf food chain at Bathurst Inlet. Bar charts represent the proportion of specific PCB congeners to Total PCBs (ng congener/ng Total PCBs).

lichens and caribou (Figure 2.8). The reduction in PCB congeners in top-predators occur in other mammalian food-chains (Muir *et al.*, 1988; Norstrom and Muir, 1994). Tundra wolves seem to have the ability to metabolize certain PCB congeners but not others. One hexachloro congener (PCB 153), two pentachloro congeners (PCB 99 and 118), two heptachloro congeners (PCB 180, 170/190), one octachloro congener (PCB 194) and one nonachloro congener (PCB 206) are the only congeners that substantially bioaccumulate in wolves.

Food-chain bioaccumulation

Food-chain bioaccumulation is defined as a stepwise increase of chemical fugacities in organisms with increasing trophic level. Figure 2.9 shows chemical fugacities of hexachlorocyclohexane (HCH) components, hexachlorobenzene (HCB) and total PCBs in the lichen-caribou-wolf food-chain on the Bathurst range. Total PCBs and HCB seem to bioaccumulate in this food-chain, demonstrating a step-wise fugacity increase with increasing trophic level. Results from a one-way ANOVA showed no statistically significant differences ($n = 10$, F-value (2.23) < F_{Critical} (5.98) and p -value of 0.19 at significance level of $\alpha = 0.05$) between fugacities of alpha-HCH in caribou and wolves. However, results from a one-way ANOVA (for $n = 10$, F-value (76.1) > F_{Critical} (5.31) and p -value of 2.3×10^{-5} at significance level of $\alpha = 0.05$) showed chemical fugacities of beta-HCH in wolves (9.6×10^{-9} Pa, range of 1 SD = 3.2×10^{-9} to 2.8×10^{-9}) to be significantly greater

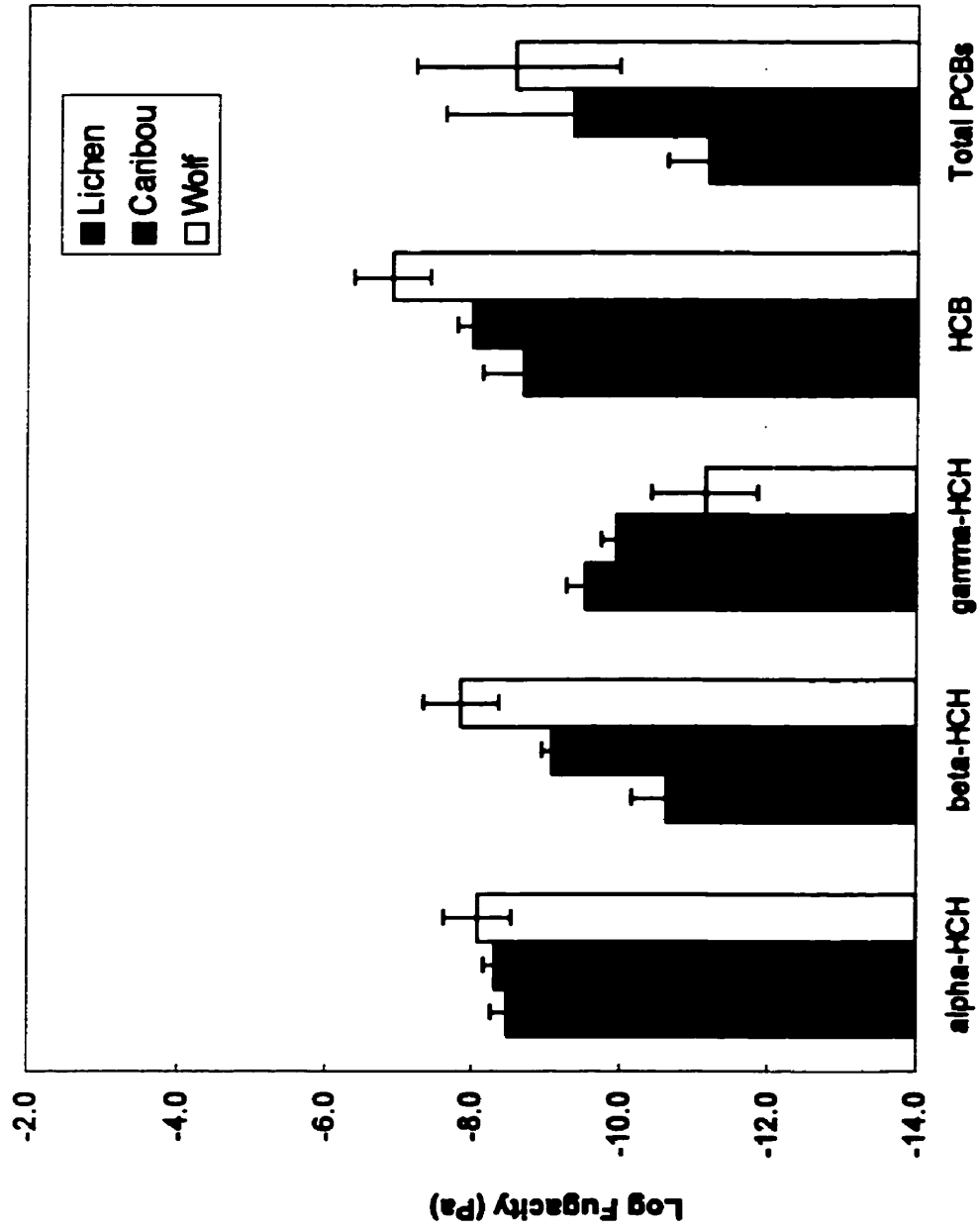


Figure 2.9: Logarithms of chemical fugacities (Pa) of alpha-, beta-, and gamma-HCH, HCB and Total PCBs in lichen-caribou-wolf food-chain at Bathurst Inlet. Bar charts represent the logarithms of geometric means and error bars represent the logarithmic standard deviations.

than fugacities of beta-HCH observed in caribou (1.9×10^{-10} Pa, range of 1 SD = 1.4×10^{-10} to 2.4×10^{-10}). Conversely, chemical fugacities of gamma-HCH in wolves (5.6×10^{-12} Pa, range of 1 SD = 1.0×10^{-12} to 3.1×10^{-11}) were shown to be significantly lower ($n = 10$, F-value (11.1) > F_{Critical} (4.41) and p -value of 0.003 at significance level of $\alpha = 0.05$) than fugacities of gamma-HCH in caribou (3.74×10^{-11} Pa, range of 1 SD = 2.8×10^{-11} to 4.86×10^{-11}). The decreased chemical fugacities of gamma-HCH with increasing trophic level are indicative of trophic dilution via metabolic transformation.

Logarithms of the observed fugacity-based BMFs (f_B/f_D) for various organic chemicals in caribou and wolves in relation to the chemical's $\log K_{ow}$ are illustrated in Figure 2.10. BMFs of alpha-HCH in caribou ranged from 1.0 to 2.2 for caribou collected in fall and summer, respectively. The observed BMFs for beta-HCH in caribou ranged from 3.9 for the animals collected in fall to 16.2 for animals in summer. Also, 1,2,4,5 Tetrachlorobenzene (TCB) demonstrated BMF values of 2.2 and 5.6 for caribou collected in September and July, respectively. In general, BMFs in caribou are shown to increase for chemicals with increasing K_{ow} between 10^4 and 10^7 . PCB 153 ($K_{ow} = 10^{6.9}$) exhibited the largest BMF values in caribou from Bathurst Inlet. The BMF for PCB153 in male caribou collected in July (28.6) was shown to be greater than BMFs observed in caribou in September (4.2) by a factor of approximately 7. For chemicals with K_{ow} 's greater than 10^7 , the BMFs in caribou are shown to decline with increasing K_{ow} . The smaller BMFs for high K_{ow}

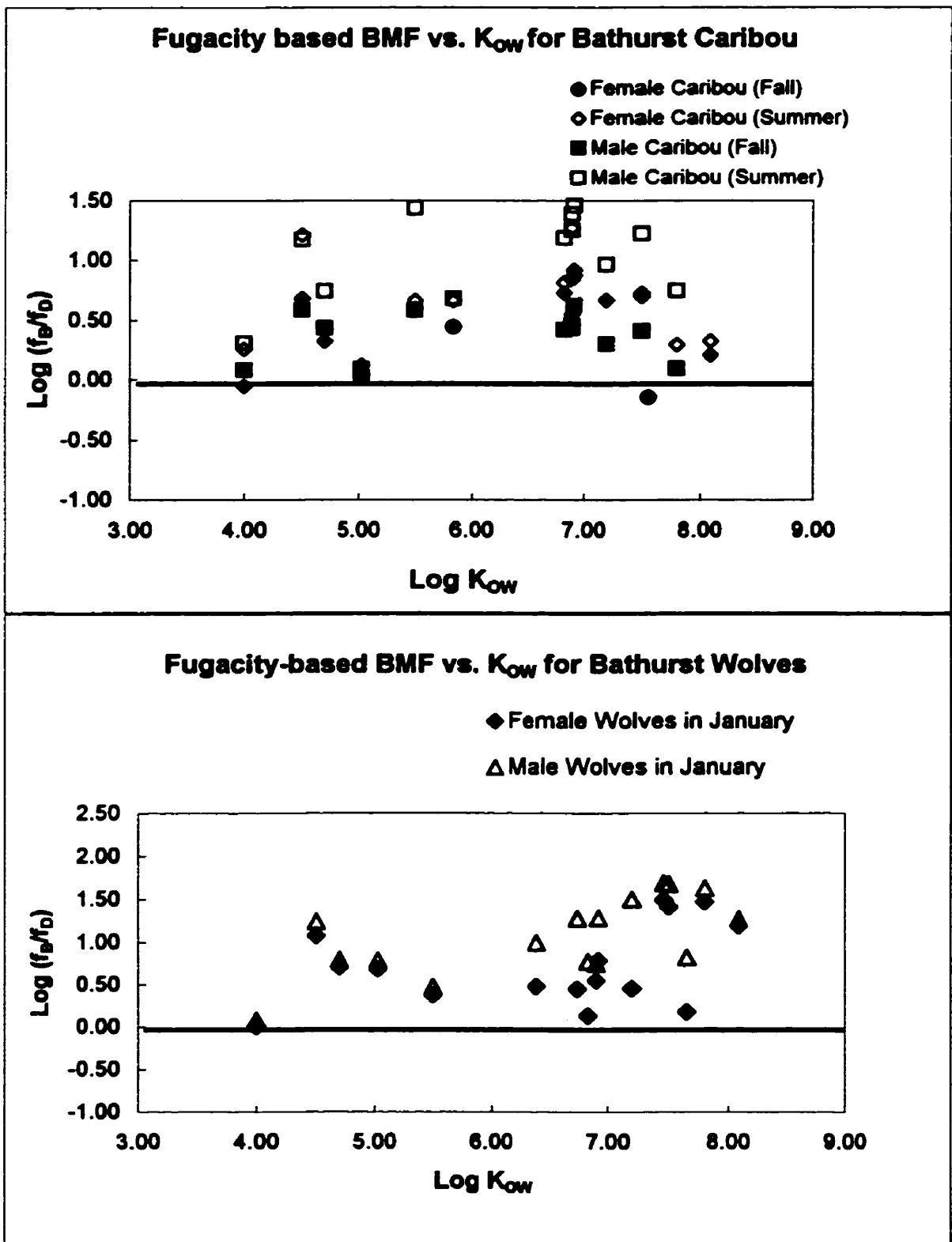


Figure 2.10: Logarithms of the fugacity-based biomagnification factors (f_B/f_D) versus chemical K_{ow} observed in caribou and wolves collected at Bathurst Inlet. Bioaccumulation occurs when BMF values are above solid line (i. e., $f_B/f_D > 1$).

chemicals in caribou may be due to a decreased absorption efficiency of these chemicals. The decline in BMFs for very hydrophobic chemicals in caribou may also be the result of non-steady state conditions in the field. The kinetics of very hydrophobic organic chemicals in organisms can be slower than chemicals with lower K_{ow} 's. BMFs of alpha-HCH, beta-HCH and 1,2,4,5-TCB in male wolves (i. e., $f_{WOLF}/f_{CARIBOU}$) from Bathurst Inlet were found to be 1.0, 17.87 and 6.19, respectively. In general, the BMFs observed in wolves increase with increasing $\log K_{ow}$. The BMFs in wolves were greater than BMFs in caribou for chemicals with K_{ow} 's greater than 10^7 . The BMF of PCB 180 in male wolves (47.2) was found to be 15 times greater than the BMF of PCB 180 in caribou in the fall (2.55), and 3 times greater than BMF of PCB 180 in caribou collected in the summer (16.19). Wolves seem to have the ability to absorb and bioaccumulate very hydrophobic chemicals following dietary exposure.

In this study, fugacities of various organic chemicals in lichen-caribou-wolf food-chains from the central and western Arctic were compared to assess food-chain bioaccumulation. These findings suggests that beta-HCH ($K_{ow} = 10^{4.5}$) and 1,2,4,5 TCB ($K_{ow} = 10^{4.7}$) may have the potential to bioaccumulate in these food chains. The hydrophobicity criteria that denotes chemicals with K_{ow} 's greater than 10^5 as bioaccumulative is commonly used to assess the bioaccumulation potential of new and existing chemical substances. The current approach targets chemicals with K_{ow} 's greater than 10^5 for possible management actions such as chemical bans or regulation. These results indicate that the current approach for assessing

bioaccumulative substances may underestimate the potential of chemicals exhibiting K_{ow} 's less than 10^5 to bioaccumulate in terrestrial food-chains.

CHAPTER 3

Gastro-intestinal magnification in barren-ground caribou (*Rangifer tarandus*): Mechanism of biomagnification and food-chain bioaccumulation

INTRODUCTION

Many persistent organic pollutants such as PCBs, DDT and toxaphene biomagnify, resulting in chemical concentrations on a lipid weight basis in a consumer organism that exceed the concentrations in the organism's prey (Connolly and Pederson, 1988; Norstrom and Muir, 1994). Traditionally, biomagnification has been described by concentration-based biomagnification factors (BMFs), which are represented by the ratio of chemical concentrations on a lipid weight basis in the consumer to the concentrations in the organism's diet (C_B/C_D). The concentration-based BMFs assume that lipids solely constitute the chemical storage capacity within the consumer and prey organisms. For vegetation species that can have very low lipid contents (< 1%), normalizing chemical concentrations on a lipid weight basis may not accurately denote the chemical storage capacity within this media. Organic carbon is a major component of vegetation, and hence may contribute more to the chemical storage capacity within vegetation. Chemical transport between biological media is driven by fugacity differences, rather than differences in chemical concentrations. Biomagnification occurs when chemical fugacities in a consumer organism (f_B)

exceed the fugacities observed in the organism's diet (f_D). For these reasons, fugacity-based BMFs (f_B/f_D) may better assess biomagnification while providing more insight into the mechanisms driving this process.

The biomagnification phenomenon has previously been explained by the loss of biomass in food-chains as a result of energy conversion at each trophic level (Woodwell, 1967). However, recent investigations on the dietary uptake and biomagnification of organic chemicals in fish have generated information pertaining to the mechanisms driving food-chain bioaccumulation (Gobas *et al.*, 1993b; 1999). Although these findings were presented for fish, similar mechanisms are expected to occur in mammals. Results from these thermodynamic studies show that food digestion and food absorption can raise the chemical fugacity in the gastro-intestinal tract of fish above the fugacity in the ingested food. Fugacity can be viewed as an "escaping tendency" of a chemical between different environmental media, such as transport from water to air (volatilization), water to biota (bioconcentration) or gastro-intestinal tract (GIT) to biota via food digestion and absorption (biomagnification). Fugacity is equivalent to chemical potential and can be measured as the partial pressure (in Pascals) that a chemical exerts within a given matrix. The chemical concentration (C in mol/m^3) and the chemical fugacity (f in Pa) in a given media are related because C equals $f \cdot Z$, where the fugacity capacity (Z in $\text{mol/m}^3 \cdot \text{Pa}$) indicates the ability of that media to retain chemical within its matrix. Passive transport between different environmental media occurs only when there is a fugacity gradient, causing net

chemical transport from media with high fugacity to low fugacity. Thus, if food digestion and food absorption elevates the chemical fugacity in the GIT of a consumer organism above the fugacity in the ingested prey organism, net-passive diffusion of chemical from the GIT to the organism's tissues can occur. The chemical fugacity in the consumer organism can then achieve a value that exceeds the fugacity in its prey, thus demonstrating the biomagnification phenomenon.

This fugacity-based explanation of biomagnification differs from conventional theory that has previously been used by bioenergetic-based models to predict chemical transport in food-chains. While the fugacity-based biomagnification mechanism has been confirmed for fish, it has not been tested for other classes of organisms, such as mammalian herbivores. In order to develop models that can predict the degree of bioaccumulation in terrestrial mammals, it is important that the model provide a reasonable description of the actual mechanism driving biomagnification. Hence, the purpose of the present study is to investigate the importance of gastro-intestinal magnification of organic contaminants in barren-ground caribou (*Rangifer tarandus*).

THEORY

For terrestrial mammals, bioaccumulation of organic contaminants is primarily the result of chemical exposure via food ingestion. In fugacity terms, dietary accumulation of chemical (biomagnification) can be explained by net passive diffusion along thermodynamic gradients within the GIT of an organism. A fugacity gradient between an organism's GIT contents and biotic tissues may occur as a result of food digestion and absorption in the GIT. Food digestion causes a change in food composition, and hence a change in the fugacity capacity (Z) of food as it passes through the GIT. The process by which digestible products in the organism's food are extracted by the organism is expected to decrease the fugacity capacity in the GIT (Z_G) below that in its food (Z_D). The reduced fugacity capacity in the gastro-intestinal content would correspond to an increased fugacity in the GIT (i. e., $f_G = C_G/Z_G$). The reduction of GIT content as food is absorbed may act to increase the chemical concentration (i. e., the chemical mass per unit volume of the GIT content), resulting in a fugacity increase. The combination of food absorption and digestion may elevate the fugacity in the GIT above the fugacity in the organism's food. It is hypothesized that this increase in chemical fugacity within the GIT may allow for net passive diffusion of chemical to the organism.

Following the explanation given for biomagnification in fish by Gobas *et al.* (1998), it is hypothesized that biomagnification of organic chemicals in caribou follows

similar mechanisms (Figure 3.1). According to this mechanism caribou ingest lichens at an ingestion rate of G_D (m^3/day) and excrete fecal matter at a rate of G_F (m^3/day). The ruminant digestive system is treated as a single GIT compartment in which chemical fugacity is assumed to be uniform. The flux of chemical into the GIT (N_G) and out of the GIT via fecal excretion (N_F) in units of mol/day can be calculated as $G_D \cdot C_D$ and $G_F \cdot C_G$, respectively. Because fugacity is linearly related to concentration, N_G can also be expressed as $G_D \cdot Z_D \cdot f_D$ (mol/day) and N_F as $G_F \cdot f_G \cdot Z_G$ (mol/day).

Figure 3.1a provides an illustrative example of how caribou with an internal fugacity of 1 Pa ingests lichens with a fugacity of 1 Pa, at an ingestion rate of 3 m^3/day and excretes fecal matter at a rate of 1 m^3/day . For net uptake of chemical to occur from the GIT to the organism, a fugacity gradient between the GIT and organism must be attained unless the chemical is taken up actively. However, active transport mechanisms for highly hydrophobic xenobiotic chemicals are unlikely to exist and have never been reported. If no food absorption or digestion occurs, the fugacity in the GIT remains the same as the fugacity observed in the food (i. e., 1 Pa). In this situation, no net chemical transport across the intestinal walls of the GIT could occur since there is no fugacity gradient (Figure 3.1a). However, if food absorption occurs, the contents

(A) Food Ingestion ($G_b = 3$)

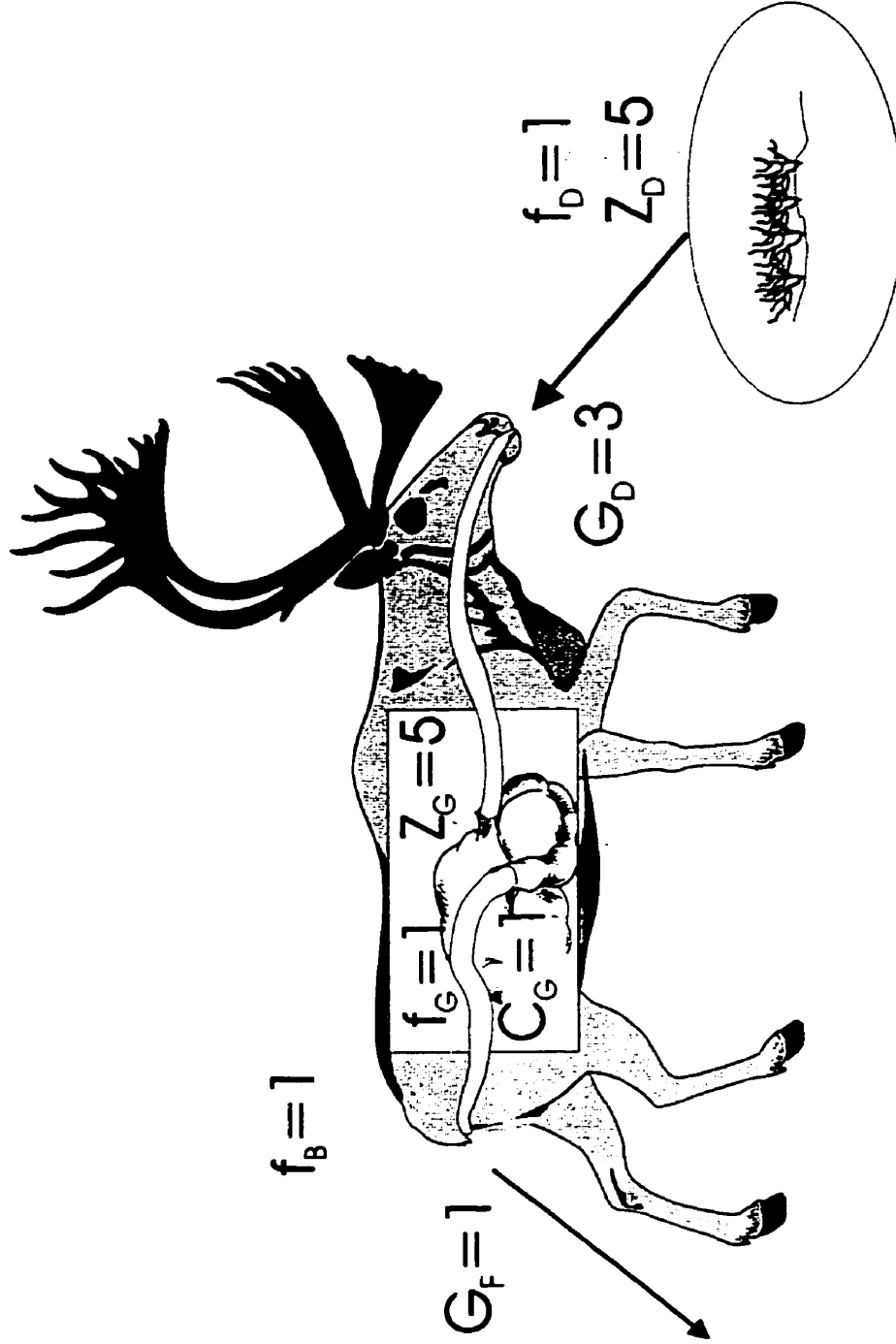


Figure 3.1: Illustrative conceptual diagram of gastrointestinal magnification of organic contaminants in caribou. Food digestion and food absorption can raise the fugacity in the GIT thus allowing for the animal to achieve a fugacity above that in its food.

(B) Food Absorption $G_D/G_F = 3$

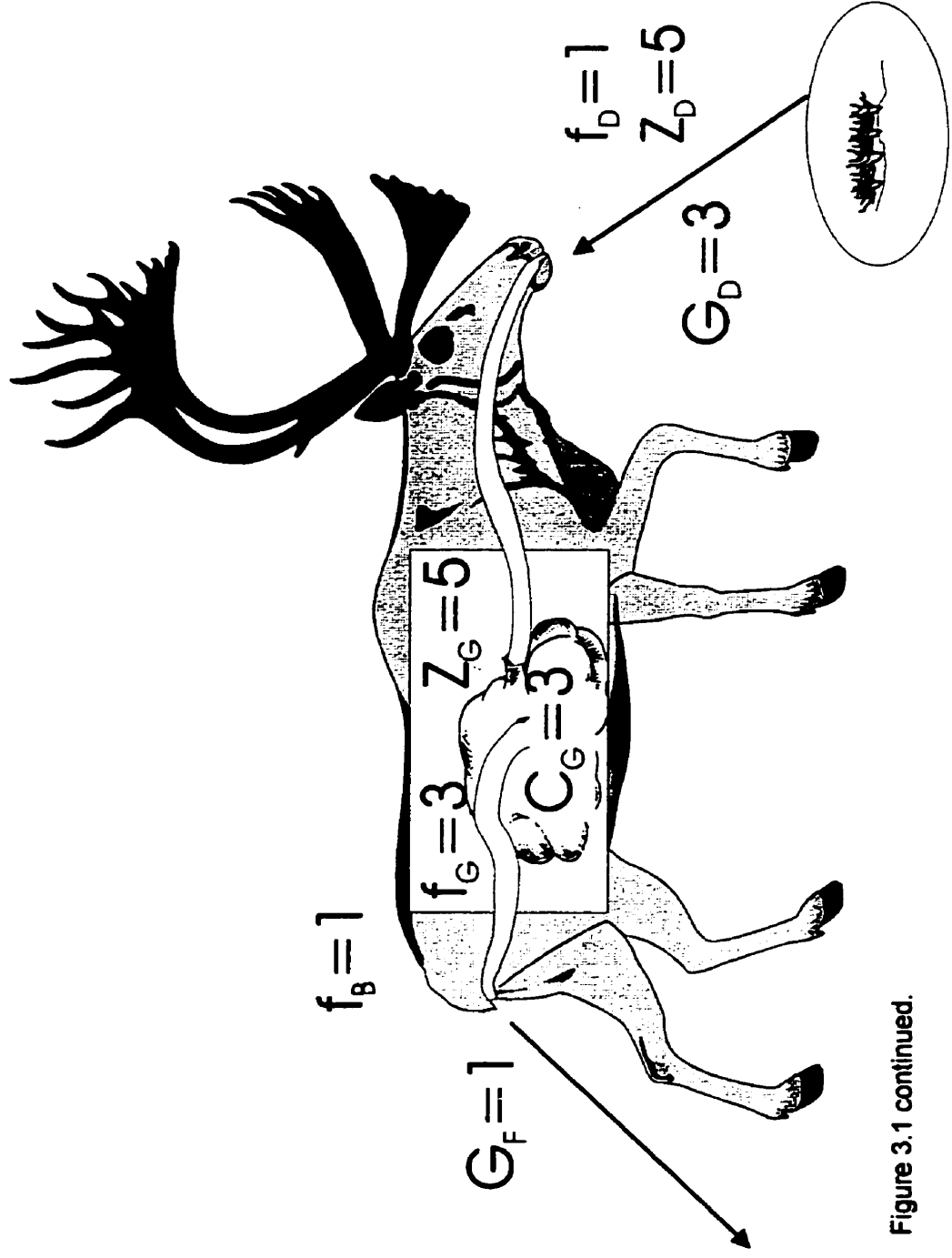


Figure 3.1 continued.

(C) Food Digestion $Z_D/Z_G = 5$

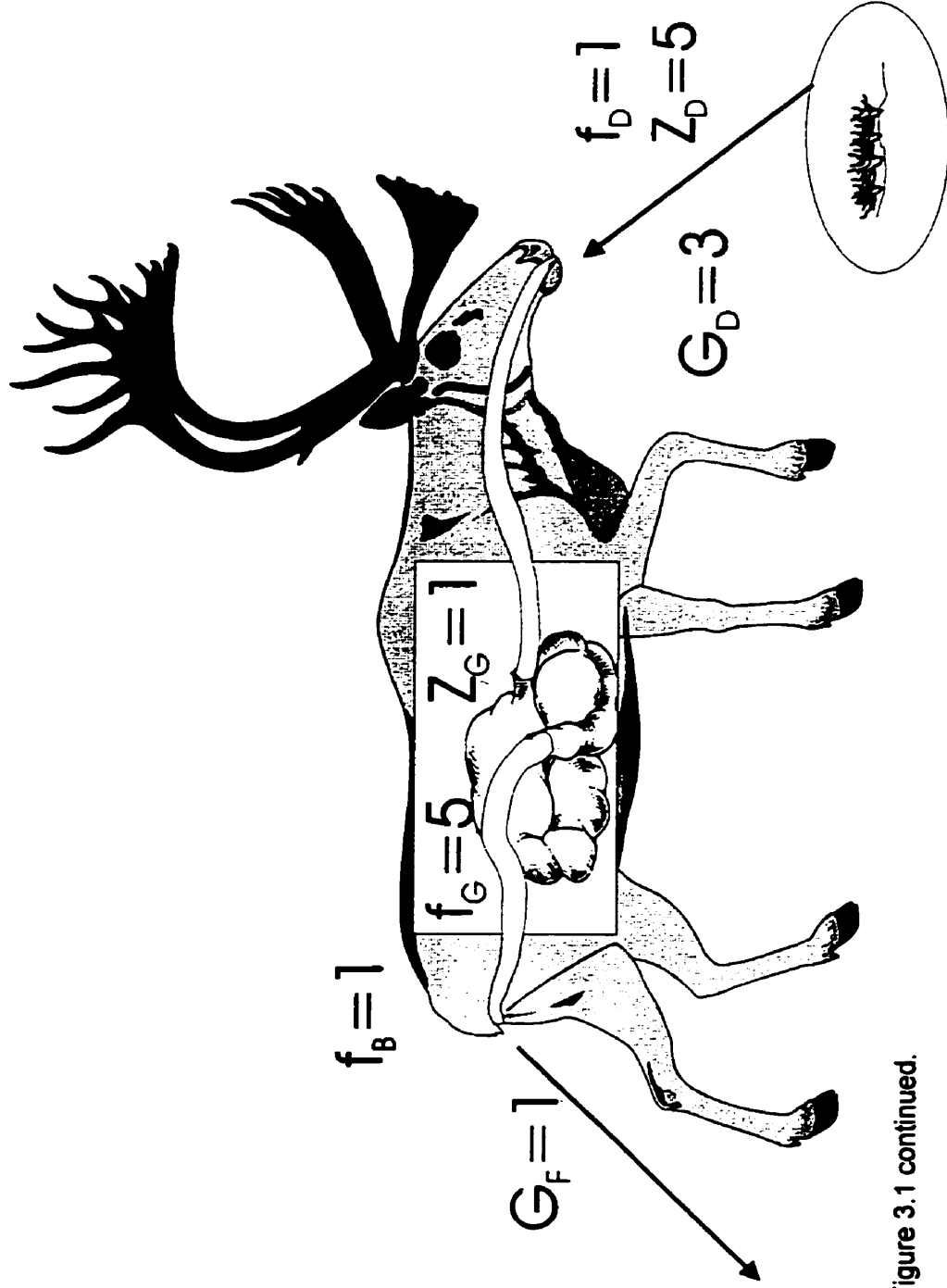


Figure 3.1 continued.

(D) Effects of Food Digestion and Food Absorption

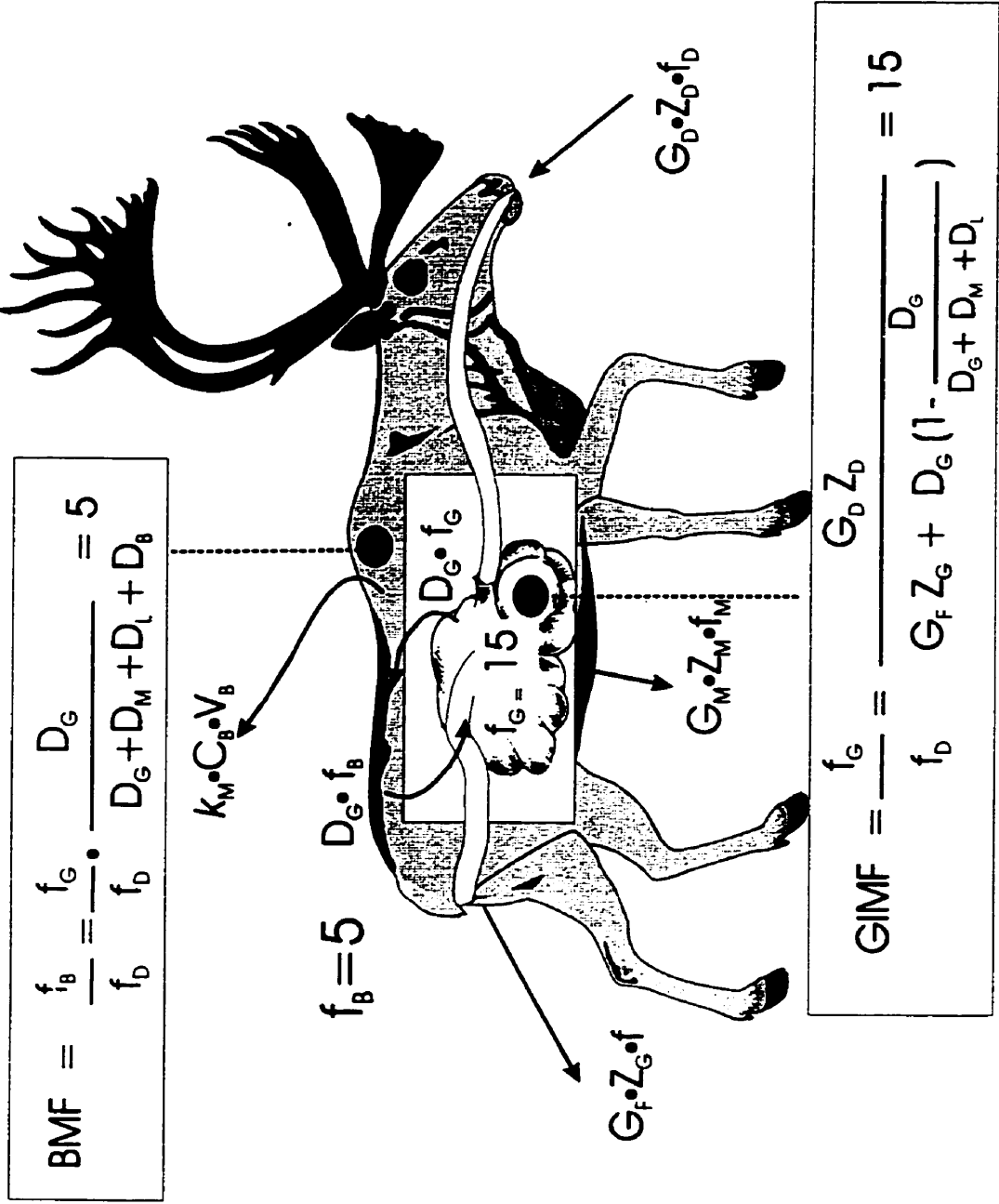


Figure 3.1 continued.

within the GIT is reduced as digestible matter is absorbed by the organism. If we assume the volume of the GIT (V_G) remains constant, the replacement of food during a grazing period would gradually concentrate ingested chemical within the GIT. Figure 3.1b shows how an increase of chemical in the GIT caused by food absorption would result in an increased fugacity in the GIT (i. e., $f_G=3$). The extent of food absorption, represented by (G_D/G_F) is shown to be a factor of 3.

The role of food digestion in elevating the fugacity in the GIT above that of the ingested food can be viewed as the ratio of the fugacity capacities of food and the GIT (Z_D/Z_G). For carnivores, food digestion in the GIT involves extraction of lipids associated with a prey organism. Chemical storage within organisms having moderate to high lipid content is assumed to be associated with lipids. The extraction of lipids from food alters the composition of the GIT content, hence reducing the fugacity capacity of the GIT contents (Z_G) below that of the ingested food (Z_D). A similar process is believed to occur in ruminant caribou foraging on lichens. Lichens are comprised mainly of cellulose. A large portion of the cellulose fibers are in the form of hemicellulose (> 80%). The lipid content of lichens and other vegetation is usually about 1%. The chemical storage capacity within lichens may be more closely associated with cellulose fibers rather than lipids.

The processes involved with ruminant digestion of cellulose have been well documented (Ferguson, 1985; Hans and White, 1991; Aagnes *et al.*, 1995). Ruminants possess a large stomach compartment, the rumen, that provides an environment for cultures of bacteria and protozoa to digest vast amounts of ingested cellulose. The ruminant digestive process begins with the mastication of vegetative material associated with secretion of saliva. Ruminant animals then re-chew regurgitate portions of the ingested food to increase the surface area of the ingesta for more effective breakdown of cellulose to glucose by microorganisms. The microorganisms within the rumen breakdown the transformed glucose to volatile fatty acids, mainly comprised of acetic acid. The fatty acids are the major carbon source available for ruminant metabolism. The microorganisms also provide the ruminant with vital amino acids through conversion of proteins and nitrogen entering the rumen. The absorption of the digested products within the ruminant GIT occur mainly in the small intestine, however absorption of volatile fatty acids may also occur in the rumen and reticulum. Absorption of the various digestive products can occur as a result of active transport of macromolecules or passive diffusion across the inner lining of the stomach and intestinal walls.

It is believed that organic chemicals associated with the digestive products in the GIT are absorbed across the intestinal walls via passive diffusion because of their hydrophobicity and lack of internal function within an organism. Figure 3.1c exemplifies how the fugacity capacity of the GIT ($Z_G=1$) is reduced from the original fugacity capacity of the ingested lichen ($Z_D=5$) by cellulose extraction and

digestion in the GIT. The combined effects of food digestion and food absorption can result in a chemical fugacity in the GIT ($f_G=15$) that exceeds the fugacity in the ingested lichens ($f_D=1$) (Figure 3.1d). This elevated fugacity in the GIT provides a thermodynamic gradient by which net passive diffusion of chemical from the GIT to the organism's tissues can occur. This is illustrated in Figure 3.1d and shows that following food digestion and food absorption, the fugacity in the caribou ($f_B=5$) can be elevated above that in the lichens ($f_G=1$).

So far, this mechanistic explanation of biomagnification has not considered chemical loss by metabolic transformation, lactational excretion and urinary excretion, all of which can affect bioaccumulation of organic chemicals in terrestrial mammals. Transport parameters (or D values) representing chemical loss mechanisms are expressed in units of mol/Pa • day. D values substitute the products of the flow rate of a given medium ($G \text{ m}^3/\text{day}$) and the fugacity capacity (Z in mol/ $\text{m}^3 \cdot \text{Pa}$) of that medium. The transport of chemical via food intake (D_D) is then the product of the dietary intake rate (G_D) and the fugacity capacity of the food (Z_D). Chemical transport from the GIT to the organism (D_G), occurs via blood perfusion across the stomach and intestinal walls and is calculated as the product of the food absorption rate (G_A) and the fugacity capacity of the GIT (Z_G). Transport of chemical via fecal excretion (D_F) is calculated as $G_F Z_G$. Transport parameters for lactational elimination (D_L) and urinary excretion (D_U) are equivalent to $G_L Z_M$ and $G_U Z_U$, respectively. Chemical transport by metabolic transformation (in units of mol/Pa•day) is represented as the transport parameter

(D_M) and is calculated as $k_M Z_B V_B$. The rate constant k_M (d^{-1}) is the metabolic transformation rate of chemical in the organism's tissues, Z_B is the fugacity capacity of the organism ($mol/m^3 \cdot Pa$) in the organism, and V_B (m^3) is the volume of the organism. In addition, growth dilution (D_B) can occur when an animal's increase in body weight over time decreases chemical concentrations in the animal, although no chemical is actually excreted. Growth dilution depends on the organism's rate of growth (k_G), in relation to chemical uptake and elimination.

Chemical fluxes (mol/day) for uptake and elimination process are calculated as the product of the transport parameters (D , $mol/Pa \cdot day$) and the chemical fugacities (f , Pa) in a given medium. If the chemical loss by metabolism ($D_M f_B$), lactational excretion ($D_L f_M$) and growth dilution (D_B) are small compared to gastrointestinal uptake ($D_G f_G$), the extent of food digestion and food absorption can be viewed as the primary factors controlling bioaccumulation in the organism. However, if these additional loss mechanisms are significant compared to chemical loss via fecal excretion, these factors can be included to formulate a GIT magnification factor $GIMF$ (f_G/f_D) and biomagnification factor BMF (f_B/f_D) denoted as:

$$(GIMF) = \frac{f_G}{f_D} = \frac{G_D Z_D}{G_F Z_F + D_G (1 - D_G / (D_G + D_M + D_L + D_B))} \quad (\text{Equation 3.1})$$

$$(BMF) = \frac{f_a}{f_o} = \frac{f_a}{f_o} \times \frac{D_o}{(D_o + D_m + D_L + D_B)} \quad (\text{Equation 3.2})$$

The GIMF, determined by the extent of food digestion and food absorption will determine the chemical fugacity in the GIT. The fugacity achieved in the GIT will then determine the resulting fugacity in the animal. For hydrophobic, non-metabolizable substances (K_{ow} 's greater than 10^6) the rate of chemical elimination by metabolic transformation and urine excretion are negligible and unable to reduce the chemical fugacity achieved in the animal. In this case, biomagnification may occur, resulting in a chemical fugacity and hence chemical concentration in the animal that is elevated above the fugacity in the animal's food. However, chemicals that are efficiently metabolized in the organism may subsequently reduce the higher fugacity achieved in the organism. In this case, the chemical fugacity and concentration in the animal would be approximately equal or lower than that in its food, thus eliminating the biomagnification phenomenon. Lactational excretion, specific to female mammals, is an important loss mechanism that can also reduce the chemical fugacity achieved in an organism. The extent of these chemical uptake and elimination processes need to be quantified to properly assess the degree to which chemical bioaccumulation in these organisms.

For the purpose of modelling biomagnification and food-chain bioaccumulation it is important to incorporate the extent of food digestion and food absorption in an organism. Parameterization of dietary intake rates (G_D), fecal excretion rates (G_F), and the fugacity capacities of food (Z_D) and GIT content (Z_G) is important for accurate representation of GIT magnification in terrestrial mammals. Dietary intake and fecal excretion rates have been documented in previous studies for different wildlife species. Field studies can also be conducted to determine these parameters in cases where there are no literature values. The fugacity capacity of food and GIT contents (Z_G), are not well documented and cannot be easily determined by observation.

Gobas *et al.*, 1993b used a static head-space methodology to measure Z_D and Z_G for different fish species. In the present study, a dynamic head-space methodology is developed and applied to derive fugacity capacities of lichens (Z_D) and caribou fecal pellets (Z_G). Chemical concentrations in lichens (C_D) and in caribou fecal pellets (C_G) were determined from chemical analysis of lichens and caribou fecal pellets collected near Bathurst Inlet. From the observed chemical concentrations (C) and fugacity capacities (Z) of lichens and caribou fecal samples, the fugacities (f) can be calculated because fugacity is inversely proportional to concentration by the fugacity capacity of each media ($f = C / Z$). Chemical fugacities in lichens, caribou fecal pellets and caribou fat samples are determined to investigate gastro-intestinal magnification (f_G/f_D) and biomagnification (f_B/f_D) in barren-ground caribou.

MATERIALS AND METHODS

Field study and chemical analysis

Lichens and caribou fecal pellets were collected on the range of the Bathurst caribou herd and analyzed for organochlorine chemical concentrations. The methodology section in Chapter 2 summarizes collection procedures for samples and protocols for chemical analysis.

Materials

1,2,4,5-Tetra-, penta-, and hexachlorobenzene (purity > 99%) were obtained from Aldrich. 2,2',5,5'-Tetra- and 2,2',4,4',6,6'-hexachlorobiphenyl were obtained from Analabs. Analytical grade hexane and toluene were obtained from BDH Inc. (Vancouver, Canada). Silica gel 100/200 μm mesh were obtained from Supelco Canada Ltd. Anhydrous sodium sulfate, obtained from J.T. Baker Chemical Co., was heated at 550 °C before use.

Dynamic head-space analysis method to determine Z_o and Z_g

Gobas *et al.* (1993b and 1999) measured fugacities of various organic chemicals in the diet and fecal matter of fish. These experiments involved the addition of chemical to food and fecal samples by direct injection of a chemical solution made up with petroleum ether. The samples were then tightly sealed in 2mL vials and allowed to reach an equilibrium state with the gas-phase within the sample vessel. Chemical concentrations in the gas-phase were then determined and related to the chemical's fugacity according to the Ideal Gas Law. The chemical concentrations in food and fecal matter were determined by solvent extractions. The fugacity capacity of the fish food and fecal matter were then derived as the ratio of the chemical concentrations (mol/m^3) and fugacities (Pa) in these media (i. e., $Z = C / f$). For environmental media such as plants that have low volumetric lipid fractions, direct injection of chemical solution is not an appropriate contamination method because the solvents may extract vital components contributing to the fugacity capacity of the media. Therefore, a dynamic head-space methodology that contaminates samples by gas exposure was developed and used to determine the fugacity capacities of lichens and caribou GIT content.

Reindeer lichen (*Cladina rangiferina*), a common tundra lichen, was used to represent the diet of barren-ground caribou. Caribou fecal pellets collected from calving grounds of the Bathurst caribou herd were used to represent the gastrointestinal contents of caribou. The dynamic head-space method involves

exposure of lichen and caribou fecal samples to a contaminated gas-mixture. The apparatus developed for these experiments includes a glass column as a thermal desorption chamber and a rotary evaporator as a deposition vessel (Figure 3.2). Chemicals including 1,2,4,5-Tetrachlorobenzene (TCB), Pentachlorobenzene (QCB), Hexachlorobenzene (HCB), and 2,2',4,4',6,6' Hexachlorobiphenyl (HPCB) were added to a glass column containing glass beads. To increase the chemical vapour pressure, hence its chemical concentration in the gas-phase, the temperature inside the glass column was increased to approximately 40 °C. The temperature increase was achieved by applying insulated 20 gauge nichrome wire to the outside of the column. A voltage regulator (approximately 10 Volts) was used to calibrate the heat output of the wire such that a temperature within the glass column was 40 °C. The glass column was attached at one end to an ultra-high-pure grade Nitrogen cylinder. To retain the moisture content of lichens and fecal samples during exposure, the nitrogen gas was passed over a volumetric flask containing water. The volumetric flask of water was situated before the column containing chemical and was kept cool using crushed ice. The other end of the column was attached to a Rotary evaporator. Samples were placed into a 500 mL round bottom flask and attached to the rotary evaporator. The contaminated gas followed a temperature gradient within the apparatus, flowing from the heated glass column (40 °C) to the sample flask at room temperature (approximately 20 °C). To reduce chemical condensation, a water bath calibrated to 25 °C was applied to the sample flask. The flow rate (F, mL/min) of nitrogen

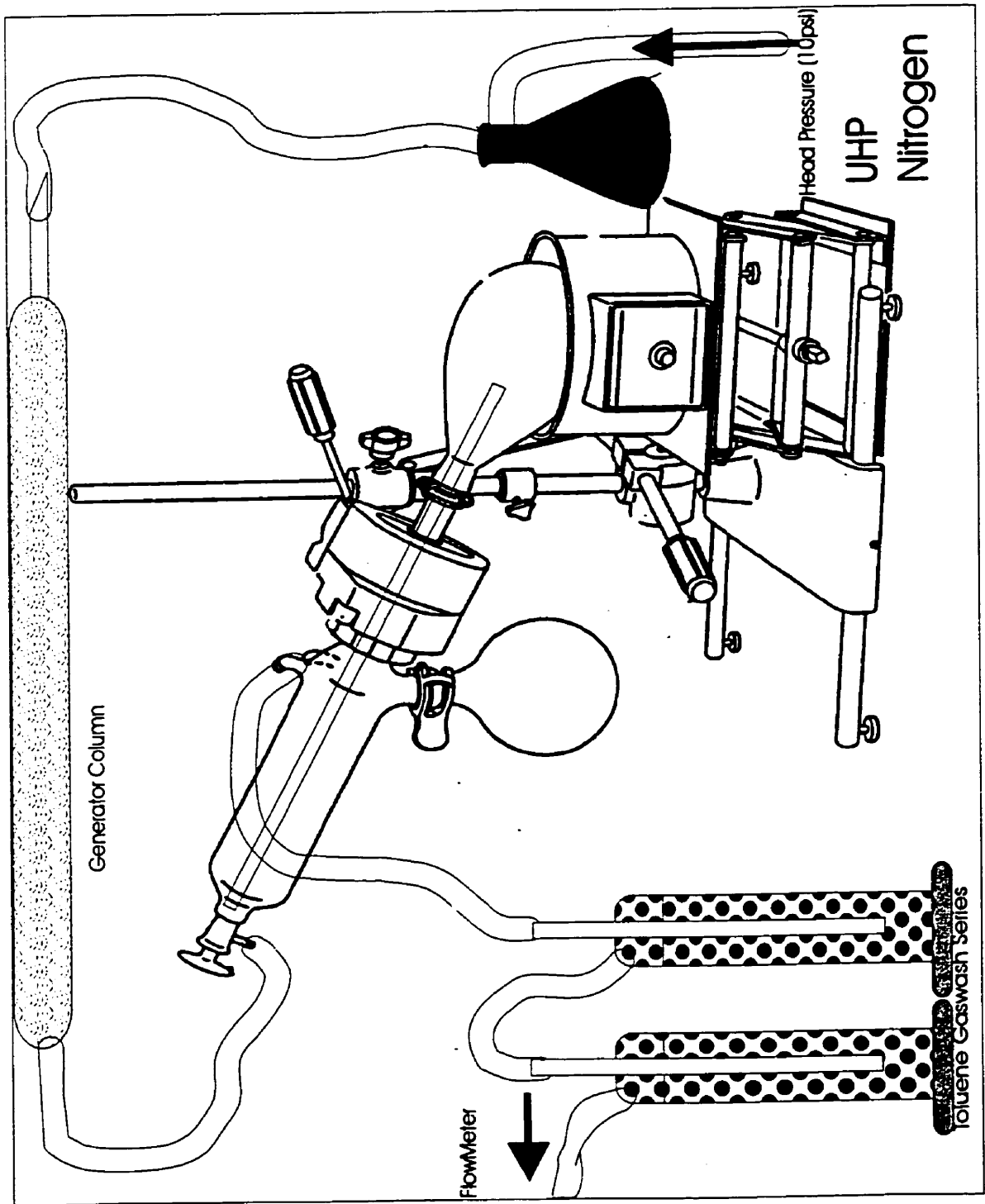


Figure 3.2: Apparatus used in dynamic head-space exposure experiments involving lichens and caribou fecal pellets.

passing through the system was measured with a bubble meter situated at the end of the apparatus. Contaminated gas was passed over the sample (approximately 20 g lichen or fecal matter) at a flow rate of approximately 40 mL/min. The outflow of gas from the rotary evaporator was attached to two gas-wash bottles in series containing 250 mL of toluene each.

After 3, 5, 9, 13, 20, 25, 32, and 45 days, measurements of chemical concentrations of analytes in the sample (C_B) and the nitrogen above the sample (C_N) were performed. To determine the chemical concentration in the sample approximately 0.5 g of contaminated sample was removed from the exposure vessel. Samples were weighed and then ground with sodium sulfate using a glass mortar and pestle. The sample was then added to a glass column containing from bottom to top, glass wool, 4 g sodium sulfate, and 4 g acidified silica gel. The columns were eluted with 250 mL of hexane. The eluent was concentrated to 2 mL and analyzed by gas chromatography. Also on each sampling day, the contents of each gas-wash bottle were concentrated using a rotary evaporator equipped with a 65 °C water. Samples were concentrated to 0.5 mL and analyzed for chemical concentrations by gas chromatography. The amount of chemical detected in the two gas-wash bottles ($X_1 + X_2$, in mg), the measured flow rate (F , in mL/min.) and exposure time (T , in min.) were used to determine the average concentration of chemical in the head-space (C_N , in mg/L) for each sampling interval ($C_N = (X_1 + X_2) \cdot FT$). Chemical losses (ϕ) associated with the efficiency of the gas-wash bottles was calculated as ($\phi = 1 - X_2/X_1$). This value represents an efficiency of the gas-

wash bottles to trap chemical, and was used to correct for chemical loss in the gas-wash bottles during the experiments. The loss of chemical during sample preparation and extraction were determined by conducting 3 recovery trials, in which 10 μ L of a 1 mg/L solution containing the above analytes was added to 0.5 g of lichens. The sample preparation and extraction methods outlined above were performed for the contaminated samples and analyzed by gas chromatography. Chemical loss attributed to sample preparation and extraction was then reported as the ratio of the recovered chemical mass to the mass of chemical in the original spiked sample.

Gas chromatographic analyses were performed on a Hewlett-Packard HP5890, equipped with a 30m DB-5 capillary column (J&W Scientific), a 63Ni Electron Capture Detector (ECD), a cool-on column injector, and an integrator. The ECD was set to 300°C, while the column temperature program ranged from 35°C to 250°C. The carrier gas was ultra-high-pure (UHP) grade helium, and the make-up gas was UHP grade 5% methane-95% argon. 1 μ L of sample extracts were injected using a 10 μ L Hamilton syringe attached to a 7673 Hewlett Packard automatic sampler. A series of external standards ranging from 0.001 mg/L to 1 mg/L were prepared from the pure chemicals. These standards were used to identify and quantify sample peaks during chromatographic analysis. The limits of quantitation (LOQ) for analytes used in the experiment were determined by multiplying the lowest observed concentration of these chemicals by a factor of 2. The LOQ's for TCB, QCB, HCB and HPCB were found to be 0.003 mg/L, 0.0006

mg/L, 0.0004 mg/L and 0.0008 mg/L, respectively. The LOQ's for analyte concentrations in lichen and caribou fecal samples were determined to be 3.3×10^{-6} mg/g, 5.8×10^{-7} mg/g, 4.1×10^{-7} mg/g and 8.6×10^{-7} mg/g for TCB, QCB, HCB and HPCB, respectively. The LOQ's for analytes in the gas-phase were determined to be 1.7×10^{-8} mg/L, 2.9×10^{-9} mg/L, 2.1×10^{-9} mg/L and 4.3×10^{-9} mg/L for TCB, QCB, HCB and HPCB, respectively.

Once the chemical concentration in the sample establishes a constant concentration with time, a chemical equilibrium state between the sample and the gas-phase has been achieved. At equilibrium, the chemical fugacity in the gas-phase (f_N) can be determined from the concentration in the air (C_N), since $f_N = C_N/Z_N$, where Z_N is $(1/RT)$ and is equal to $0.00041 \text{ mol/m}^3 \cdot \text{Pa}$ at 25°C . After a chemical equilibrium is attained, the fugacity in the food (f_D) or fecal (f_G) samples is equal to the fugacity in the air (f_N) above the sample. The fugacity capacities of the food and fecal matter for the chemicals can then be derived from the measured concentrations in these media and their corresponding fugacities:

$$Z_D = C_D / f_D$$

$$Z_G = C_G / f_G$$

where; C_D, C_G are measured concentrations (mol/m^3) food and feces, respectively;

f_D, f_G are the chemical fugacities (Pa) in food and feces, respectively.

Alternative methods used to calculate Z_D and Z_G

The fugacity capacity of lipids (Z_L) can be approximated by the fugacity capacity of octanol (Z_O). The fugacity capacity of water (Z_W) is the reciprocal of the chemical's Henry's law constant ($1/H$). Thus, the octanol-water partition coefficient (K_{OW}) can be expressed as $Z_O \div Z_W$. The fugacity capacity of environmental media containing high lipid fractions can then be calculated as the product of the chemical's K_{OW} and Z_W . For media having very low lipid fractions, organic carbon may contribute more to the fugacity capacity of that media than lipids. The fugacity capacity of organic carbon is thought to be approximately 41% of pure octanol (Karickhoff, 1981). Therefore, a chemical partition coefficient between water and organic carbon (K_{OC}) is substituted by $0.41 \cdot K_{OW}$. Based on K_{OC} , the organic carbon content (ϕ) and the density (δ_D) of lichens and caribou fecal pellets, a fugacity capacity of lichen (Z_D) and caribou GIT content (Z_G) can be estimated by $Z = Z_W \delta_D \phi K_{OC}$. The fugacity capacity of lichens (Z_D) was also calculated from chemical concentrations observed in Arctic air and lichens collected from the central Arctic. Observed concentrations of hexachlorobenzene (HCB) and HPCB

measured in Arctic air (C_A), their corresponding fugacities in air (f_A), and their concentrations in lichens (C_D) collected near Bathurst Inlet were used to estimate the fugacity capacity of lichens (Z_D), (i. e., $Z_D=C_D/f_D$). In this calculation of Z_D it is assumed that lichens collected from the study area are in equilibrium with the air in the central Arctic, hence the fugacity in lichen (f_D) was presumed to be equal to the fugacity in the air (f_A). The chemical fugacity in air was derived from the observed chemical concentrations measured in Arctic air (in units of mol/m^3), which are reported in Jensen *et al.* (1998) and a fugacity capacity in air (Z_A) equal to $0.00043 \text{ mol/m}^3 \cdot \text{Pa}$ at a mean summer temperature of 10°C (i. e., $f_A = C_A/Z_A$).

The fugacity capacity in caribou GIT content was also calculated by the equation $Z_G = (1 - \alpha) \cdot Z_D$, where α is the cellulose extraction efficiency of caribou. The organic carbon content of caribou fecal pellets was shown to be 12 % less than the organic carbon content measured in lichens. The organic carbon contents of lichens and caribou fecal pellets were determined by loss on ignition (LOI), which is an assessment of total organic matter in a sample. If we assume the storage capacity of lichens and caribou GIT content to be associated with cellulose, the cellulose content of these media may better represent their respective fugacity capacities. The digestibility and removal of cellulose (α) by caribou during digestion of lichens has previously been shown to be approximately 80% (Boertje, 1990). Therefore, the fugacity capacity of caribou GIT may be estimated as $Z_G = (1 - \alpha) \cdot Z_D$, where α is the cellulose extraction efficiency of caribou.

RESULTS AND DISCUSSION

Fugacity capacity measurements under laboratory conditions

The lipid content of lichens and caribou fecal pellets collected during the present study were found to be $0.45 \pm 0.14\%$ and $0.97 \pm 0.30\%$, respectively. The organic carbon contents of lichens and caribou fecal pellets were determined to be $96.1 \pm 0.06\%$ and $84.7 \pm 0.003\%$. The density of lichens and caribou fecal matter were 0.54 ± 0.09 kg/L and 0.86 ± 0.15 kg/L, respectively. Chemical loss by sample extraction and clean-up procedures used on lichen and caribou fecal pellets was negligible, exhibiting recoveries of 98.6 ± 3.1 %. Chemical loss during evaporation of toluene gas-wash bottles ranged from 68.8 ± 3.1 % for TCB to 78.6 ± 4.8 % for HPCB.

The concentration of analytes in the gas-phase during the lichen exposure experiment increased from day 3 to day 5, then declined until reaching a constant concentration after day 13 (Figure 3.3a). Lichens exposed to the contaminated gas flow seemed to reach a state of equilibrium after approximately day 20 of the experiment (Figure 3.3b). The concentration of analytes in the gas-phase during

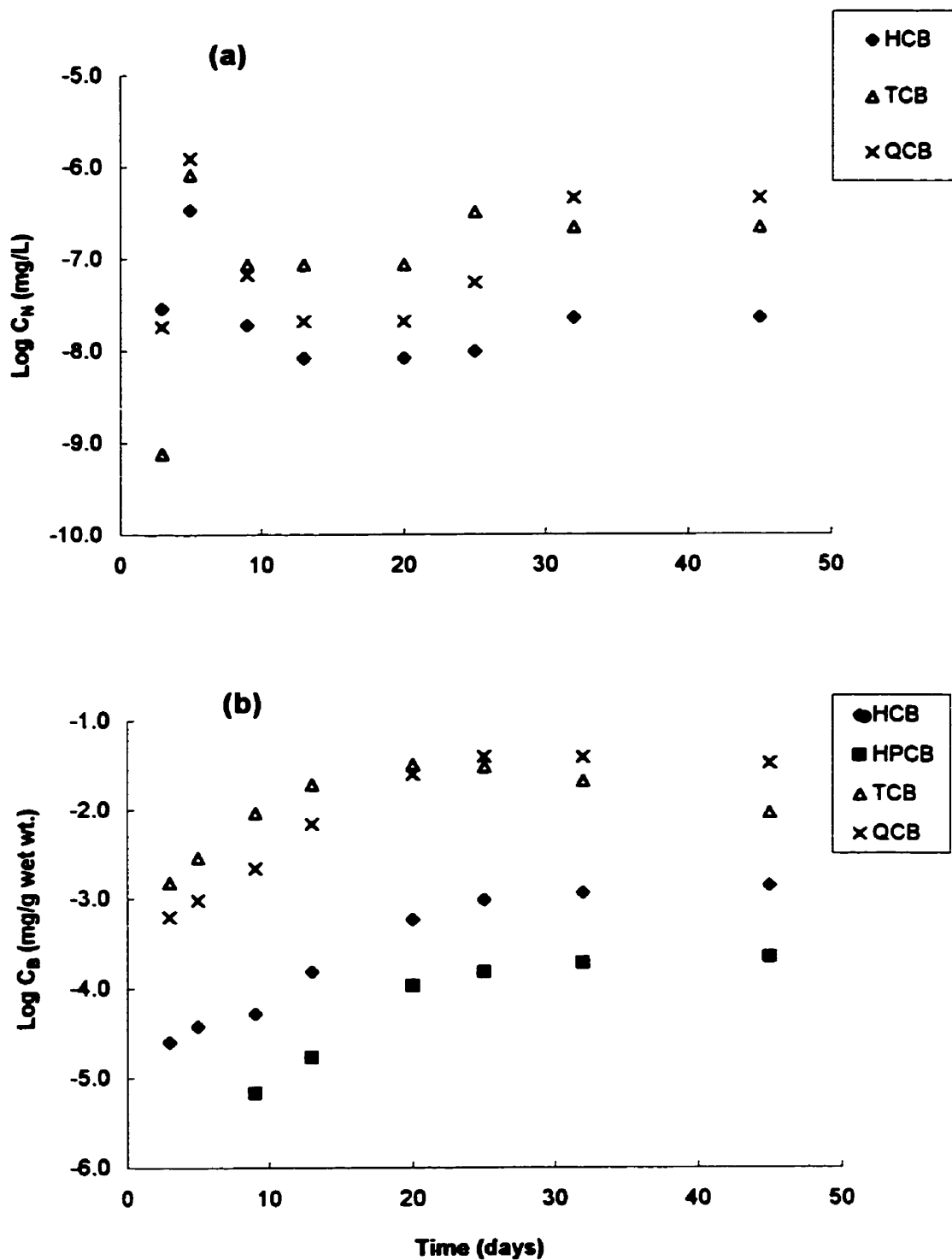


Figure 3.3: Uptake Curves showing logarithms of chemical concentrations in (a) gas-phase (mg/L) (b) lichens (mg/g wet wt.) over the 45-day exposure experiment.

the caribou fecal exposure experiment are shown in Figure 3.4a. The concentrations of TCB and HCB in the head-space during caribou fecal exposures declined from day 3 to day 5 and then remained relatively constant for the duration of the experiment. The concentrations of TCB in the gas-phase during fecal exposures declined slightly by day 13, and remained constant until day 32 but afterwards increased. The concentrations of HPCB during caribou fecal pellet exposures generally remained constant throughout the experiment. Caribou fecal matter seemed to reach an initial chemical equilibrium with the gas-phase after day 9. On day 20, the chemical concentrations in the fecal pellets increased and then remained constant for the duration of the experiment (Figure 3.4b). The ratio of the chemical concentrations (mol/m^3) in lichens and caribou fecal samples to the chemical fugacities (Pa) in the gas-phase during the exposure experiments represent the fugacity capacities (Z , in units of $\text{mol}/\text{m}^3 \cdot \text{Pa}$) of these media. The logarithms of the fugacity capacities of lichens (Z_D) and caribou fecal pellets (Z_G) as a function of exposure time are shown in Figure 3.5a and Figure 3.5b, respectively. Z_D values for TCB declined from $4.8 \times 10^5 \text{ mol}/\text{m}^3 \cdot \text{Pa}$ on day 3 to achieve a value of approximately $1.1 \times 10^5 \text{ mol}/\text{m}^3 \cdot \text{Pa}$ on day 45. The values of Z_D for HCB increased initially and then remained constant from day 25 to day 45. For QCB, Z_D initially increased from day 3 to day 13, remained constant from day 13 to day 25, and then declined to $1.5 \times 10^4 \text{ mol}/\text{m}^3 \cdot \text{Pa}$ on day 45. In the caribou fecal exposure experiment the temporal trends for Z_G were similar for all the analytes. The Z_G values generally remained constant from day 5 to day 9 and then

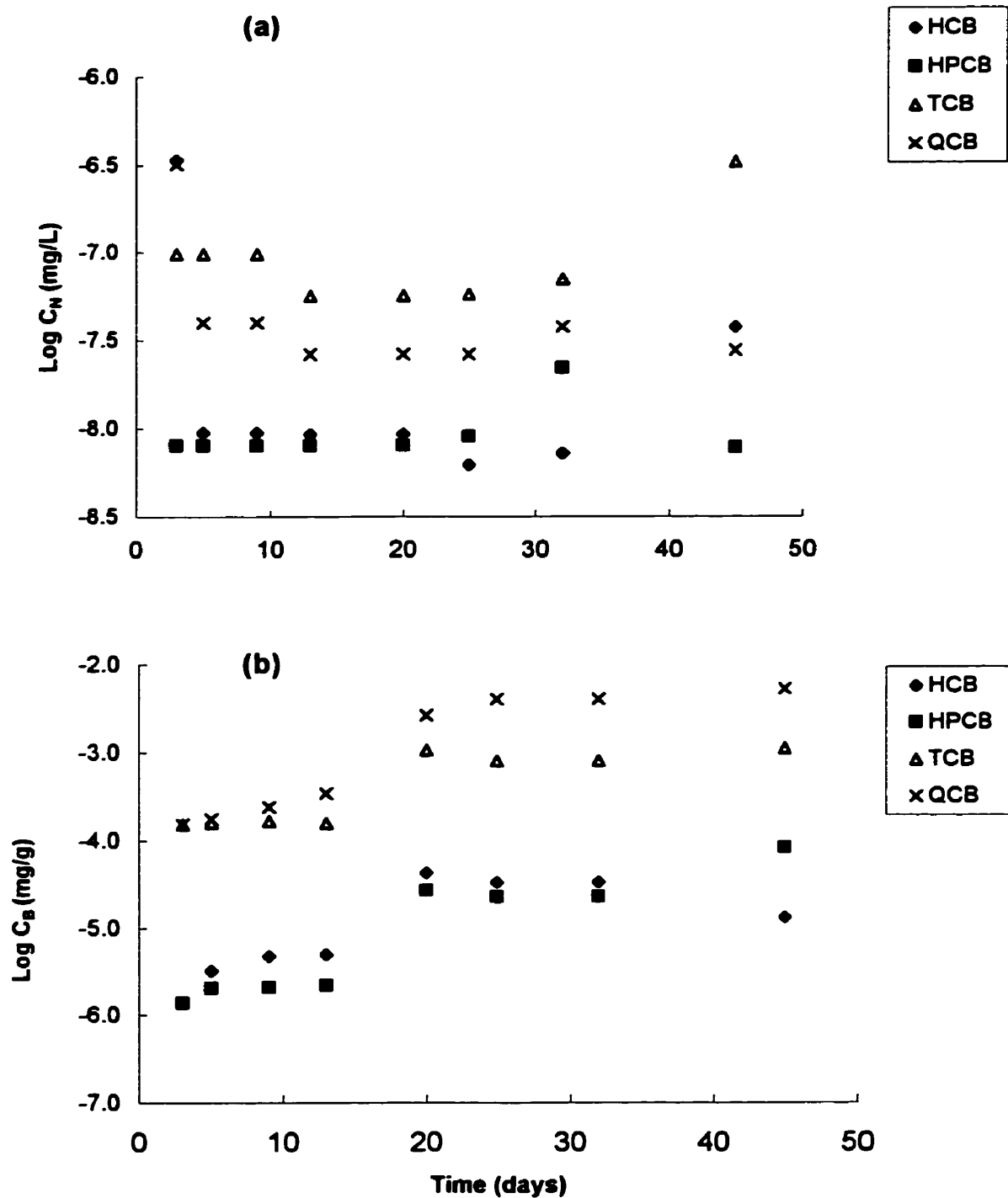


Figure 3.4: Uptake Curves showing logarithms of chemical concentrations in (a) gas-phase (mg/L) (b) caribou fecal pellets (mg/g wet wt.) over the 45-day exposure experiment.

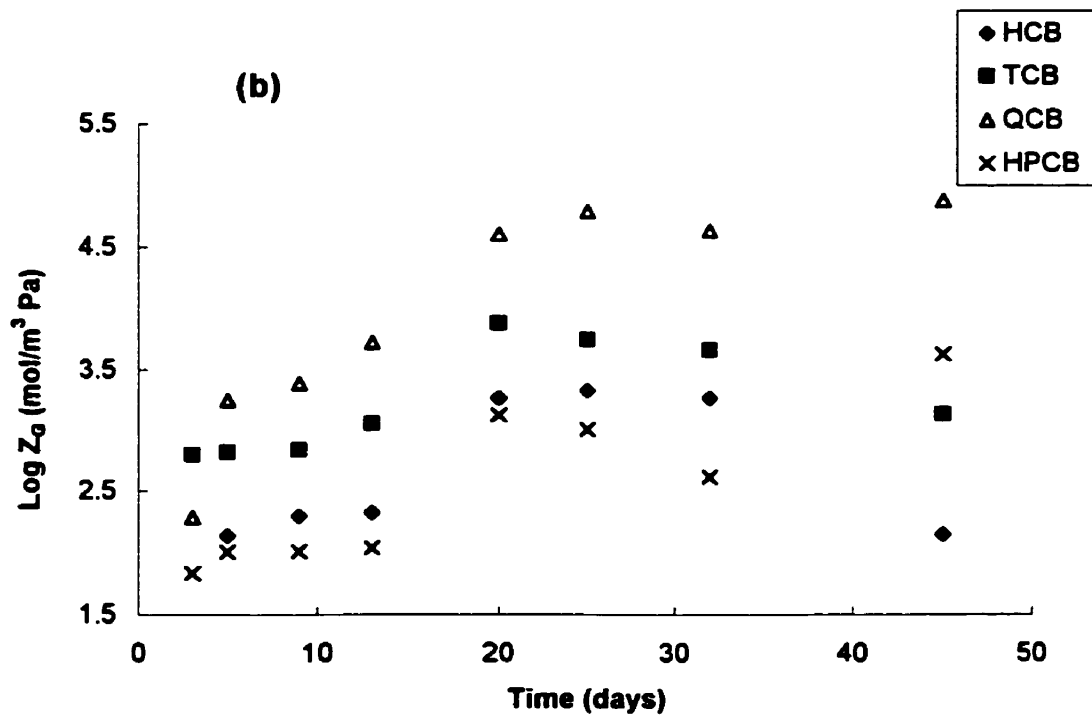
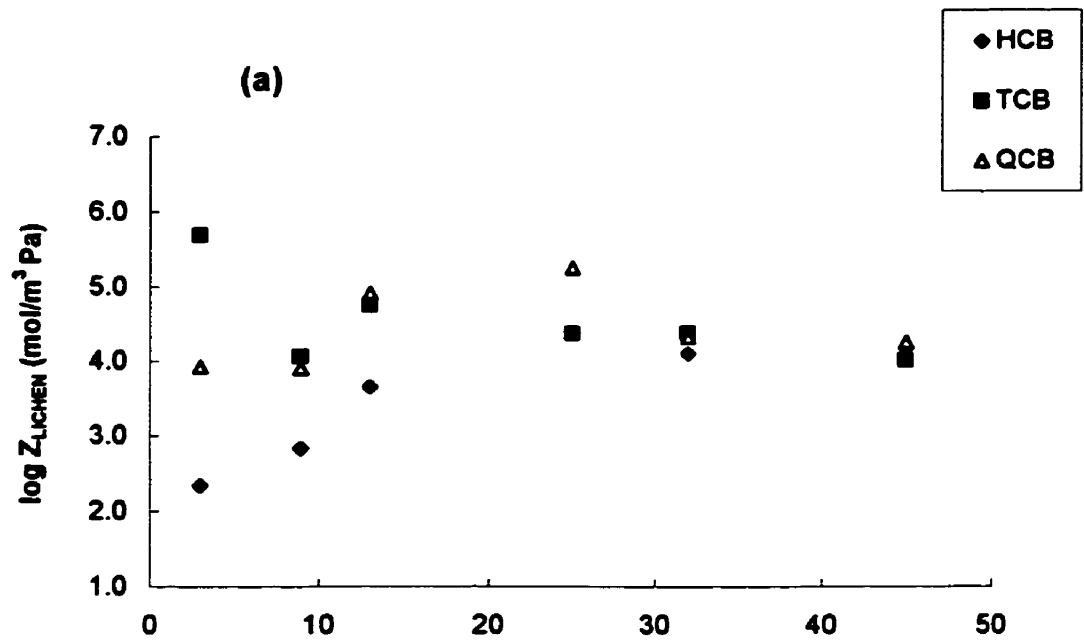


Figure 3.5: Logarithms of the fugacity capacity ($\text{mol}/\text{m}^3 \text{ Pa}$) of (a) lichens (Z_0) and (b) caribou fecal pellets (Z_G) for analytes used in 45 day exposure experiments.

increased between day 13 and day 20. For TCB, QCB and HCB, Z_G values remained constant from day 20 to day 32. The Z_G values for HPCB remained constant from day 20 to day 25, declined until day 32 and then increased again on day 45.

The experimentally derived values of Z_D and Z_G shown in Table 3.1 are based on the equilibrium conditions observed on day 45 for lichens and day 32 for caribou fecal pellets. The Z_D values derived from the experiments ranged from 1.1×10^4 mol/m³.Pa for TCB to 1.5×10^4 mol/m³.Pa for HCB. Z_G values ranged from 4540 mol/m³.Pa for TCB to 4.3×10^4 mol/m³.Pa for QCB. The ratio of Z_D/Z_G for lichens and caribou GIT content represents the extent of digestion for barren-ground caribou. The Z_D/Z_G ratio observed in these experiments was 2.3 for TCB, 0.24 for QCB and 6.9 for HCB. For TCB and QCB the fugacity capacity values of lichen and caribou GIT content exceeded the fugacity capacity of Z_o . The perception that lichens, containing very low lipid (<1%), have a greater chemical storage capacity than pure octanol is unlikely. The higher values of Z_D and Z_G determined from the experimental data may be the result of experimental error. In this methodology, the two factors controlling the magnitude of Z_D and Z_G values are the chemical concentration measured in the gas-phase and the chemical concentration measured in the exposed samples of lichens or fecal matter. Sources of error that would explain elevated Z value results are: (i) error in the measurement of gaseous concentrations due to an inefficiency of the gas-wash series to trap

Table 3.1: Fugacity capacities of lichen (Z_D) and caribou GIT (Z_G). Range of standard deviations are represented as the sum of the relative errors from chemical concentration, organic carbon content and density measurements of lichens.

Chemical	Log K_{ow}	Z_w^a calculated mol/m ³ Pa	Z_o^a calculated mol/m ³ Pa	Z_D^b calculated (field data) mol/m ³ Pa	Z_D^c calculated mol/m ³ Pa	Z_G^d calculated mol/m ³ Pa	Z_G^e calculated mol/m ³ Pa	Z_D/Z_G calculated mol/m ³ Pa	Z_D Experiment mol/m ³ Pa	Z_G Experiment mol/m ³ Pa	Z_D/Z_G Experiment mol/m ³ Pa
1,2,4,5 Tetrachlorobenzene	4.5	0.010	316	-	71 ± 16	153 ± 35	14 ± 3	5.1 ± 2.2	1.1 × 10 ⁴	4540	2.3
Pentachlorobenzene	5.0	0.013	1388	-	313 ± 73	413 ± 95	62 ± 11	5.0 ± 2.1	1.7 × 10 ⁴	4.3 × 10 ⁴	0.24
Hexachlorbenzene	5.5	0.015	4865	1617 ± 1028	1098 ± 258	1507 ± 347	219 ± 38	5.0 ± 2.1	1.5 × 10 ⁴	1834	6.9
2,2,4,4,6,6 Hexachlorobiphenyl	6.8	0.063	3.2 × 10 ³	2.7 × 10 ⁴ ± 1.7 × 10 ⁴	7.1 × 10 ⁴ ± 1.6 × 10 ⁴	1.6 × 10 ⁵ ± 3.6 × 10 ⁴	1.4 × 10 ⁴ ± 0.0014	5.1 ± 2.2	ND	4230	-

^a Z_o and Z_w were calculated using Kow and Henry's Law Constants from (Mackay *et al.*, 1992; Hawker and Connell, 1988; Dunnivant and Elzerman, 1992).

^b Air concentrations obtained from the 1999 CACAR report (Jensen *et al.*, 1998) were used to derive the fugacity capacities of field collected lichens.

^c Fugacity capacities of lichens (Z_D) were calculated using the equation $Z_D = Z_w \delta_{D/D} K_{OC}$ (Morrison *et al.*, 1996).

^d Fugacity capacities of Z_G were calculated based on organic carbon content using the equation $Z_G = Z_w \delta_{G/G} K_{OC}$ (Morrison *et al.*, 1996).

^e Fugacity capacity of caribou GIT content (Z_G) was calculated using the equation $Z_G = (1 - \alpha) \cdot Z_D$. The extraction of cellulose (α) by caribou was assumed to be 80% (Boertje, 1990).

chemical in the gas-phase; and (ii) an increased concentration in the exposed sample due to condensation of chemical from the gas-phase to the surface of the sample.

Estimated fugacity capacities of lichens (Z_D) and caribou GIT (Z_G)

The estimated values of Z_D and Z_G for lichens and caribou GIT content are shown in Table 3.1. The range of standard deviations were calculated by adding the relative errors associated with the organic carbon content and density of lichens and caribou fecal pellets. Estimated Z_D values ranged from $71 \pm 16 \text{ mol/m}^3 \cdot \text{Pa}$ for TCB to $7.1 \times 10^4 \pm 1.6 \times 10^4 \text{ mol/m}^3 \cdot \text{Pa}$ for HPCB. Calculations of Z_D for HCB and HPCB using chemical concentrations observed in Arctic air and field collected lichens elicited values similar to the estimated Z_D values based on organic carbon. The estimates for Z_G ranged from 14 for TCB to $1.4 \times 10^4 \text{ mol/m}^3 \cdot \text{Pa}$ for HPCB. The experimentally derived Z_D and Z_G values were 1 to 2 orders of magnitude higher than the calculated estimates. The ratio of Z_D/Z_G using the estimated fugacity capacities of lichens and caribou GIT content was 5.1 ± 2.1 .

Gastro-intestinal magnification in barren-ground caribou

The chemical fugacities of α -HCH, HCB and PCB congeners 52, 118, 153, and 180 varied among lichens (f_D), caribou GIT content (f_G) and caribou fat (f_B) collected from the range of the Bathurst caribou herd (Figure 3.6). The standard deviations

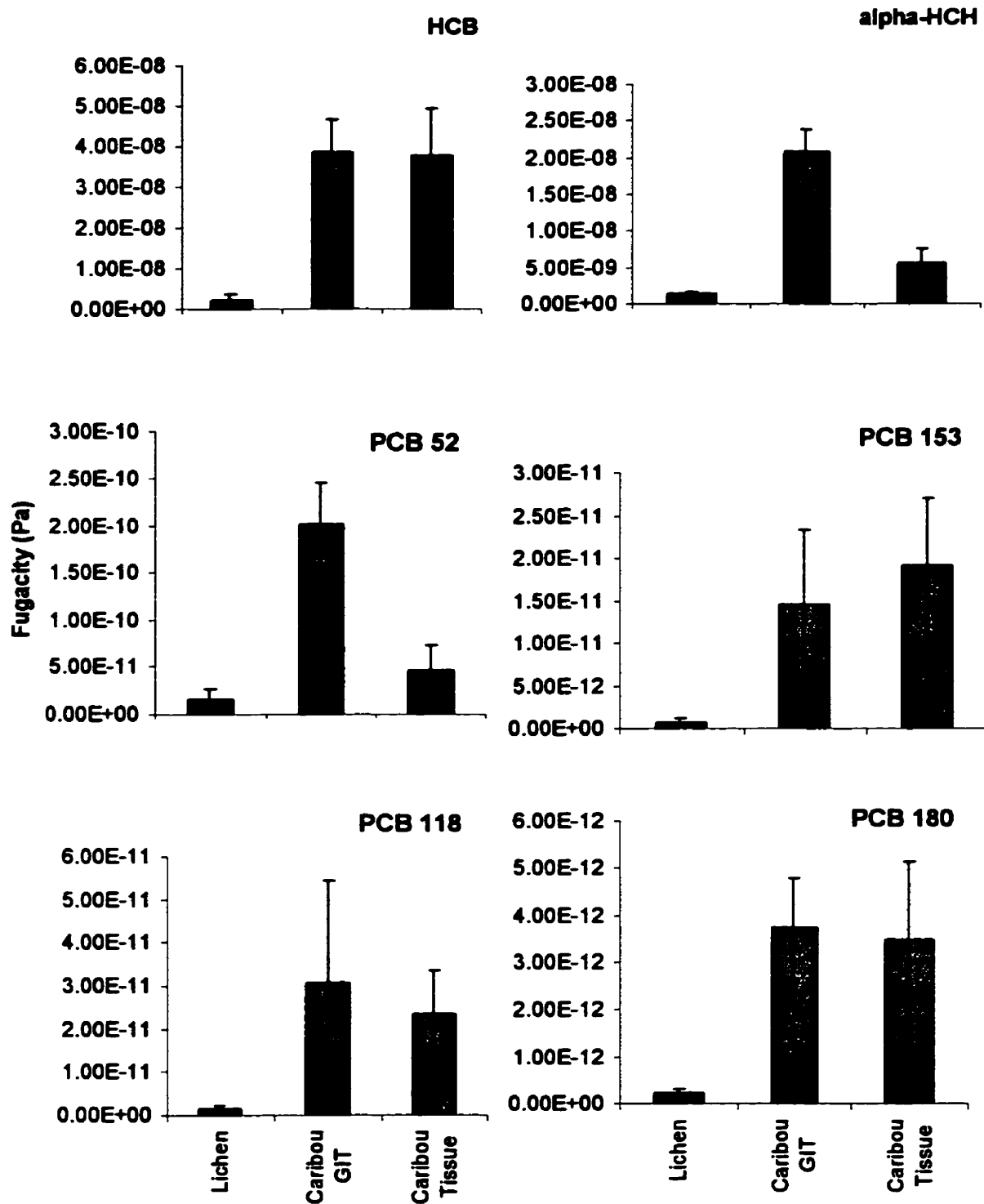


Figure 3.6: Observed fugacities (Pa) of HCB, alpha-HCH, PCB congeners 52, 138,153,180 in the lichens (diet), caribou GIT content, and caribou fat tissue. Error bars for lichens and caribou GIT content represent sum of the relative errors associated with the measured chemical concentrations, organic carbon content and densities of lichens and caribou fecal pellets. Error bars for caribou fat tissue are the standard deviations of the observed chemical concentrations in caribou fat samples.

for chemical fugacities in lichens represent the sum of the relative errors associated with the measured chemical concentrations, organic carbon content and density of lichens. The standard deviations for chemical fugacities in caribou represent the relative error associated with the measured chemical concentrations in caribou fat samples. The chemical fugacities in these media are derived from their observed chemical concentrations (mol/m^3) and estimated fugacity capacities ($\text{mol}/\text{m}^3\cdot\text{Pa}$) of lichen (Z_D), caribou fecal pellets (Z_G) and caribou lipids (Z_o) (Table 3.1). The estimated ratio of Z_D/Z_G (5 ± 2) was used in these fugacity calculations. For each of these chemicals the fugacities in the GIT contents were elevated above the fugacities in lichens. The chemical fugacities in fat tissue samples of caribou were also all elevated above the fugacity in lichens. However, the fugacity increase of PCB 52 and α -HCH in caribou fat was less pronounced than fugacities of other chemicals. This suggests that caribou may eliminate PCB 52 and α -HCH following gastro-intestinal uptake. Possible routes of elimination of these chemicals in caribou are by metabolic transformation and milk excretion by lactating females. Also, the animals growth over time can also cause chemical concentrations, and hence fugacities to decrease in the organism.

The extent of gastro-intestinal magnification of different substances is demonstrated by a fugacity-based gastro-intestinal magnification factor (GIMF). The GIMF is calculated as the ratio of the fugacity in the organism's GIT to the fugacity in its food (f_o/f_D). Similarly, the extent of biomagnification is determined by a biomagnification factor (BMF), expressed as the fugacity ratio between an

organism and its food (f_B/f_D). The chemical fugacities in lichens (f_D), caribou GIT (f_G) and caribou (f_B) used to calculate the fugacity-based GIMFs and BMFs were calculated from the observed chemical concentrations in these media and their corresponding fugacity capacities (i. e., $f = C/Z$). GIMFs and BMFs for the trophic transfer of lichen-caribou are illustrated for different organic contaminants of various K_{ow} in Figure 3.7. GIMFs for these chemicals were all around 15. The fugacity-based BMFs for lichen-caribou ranged from approximately 5 (α -HCH) to 25 (PCB 153). The reason that BMFs for α -HCH and PCB 52 are substantially lower than their corresponding GIMFs may be due to metabolic transformation of these chemicals in caribou. For these chemicals, the fugacity achieved in the caribou as a result of gastro-intestinal magnification may be reduced by chemical elimination via metabolic transformation in the organism's tissues. The BMFs of PCB 153 (25.7) and PCB 118 (24.1), were greater than the GIMFs of these chemicals in caribou. There is substantial error in the calculation of these BMFs due to variability in the observed concentrations (i. e., C_D , G_G , and C_B), and the calculations of fugacity capacities (Z_D , Z_G and Z_B). The error associated with these fugacities and hence the fugacity-based BMFs could explain the apparently higher but not statistically significant increase in BMFs of PCB 153 and PCB 118 observed in caribou.

GIT and Biomagnification Factors of Organic Chemicals in Barren-ground Caribou

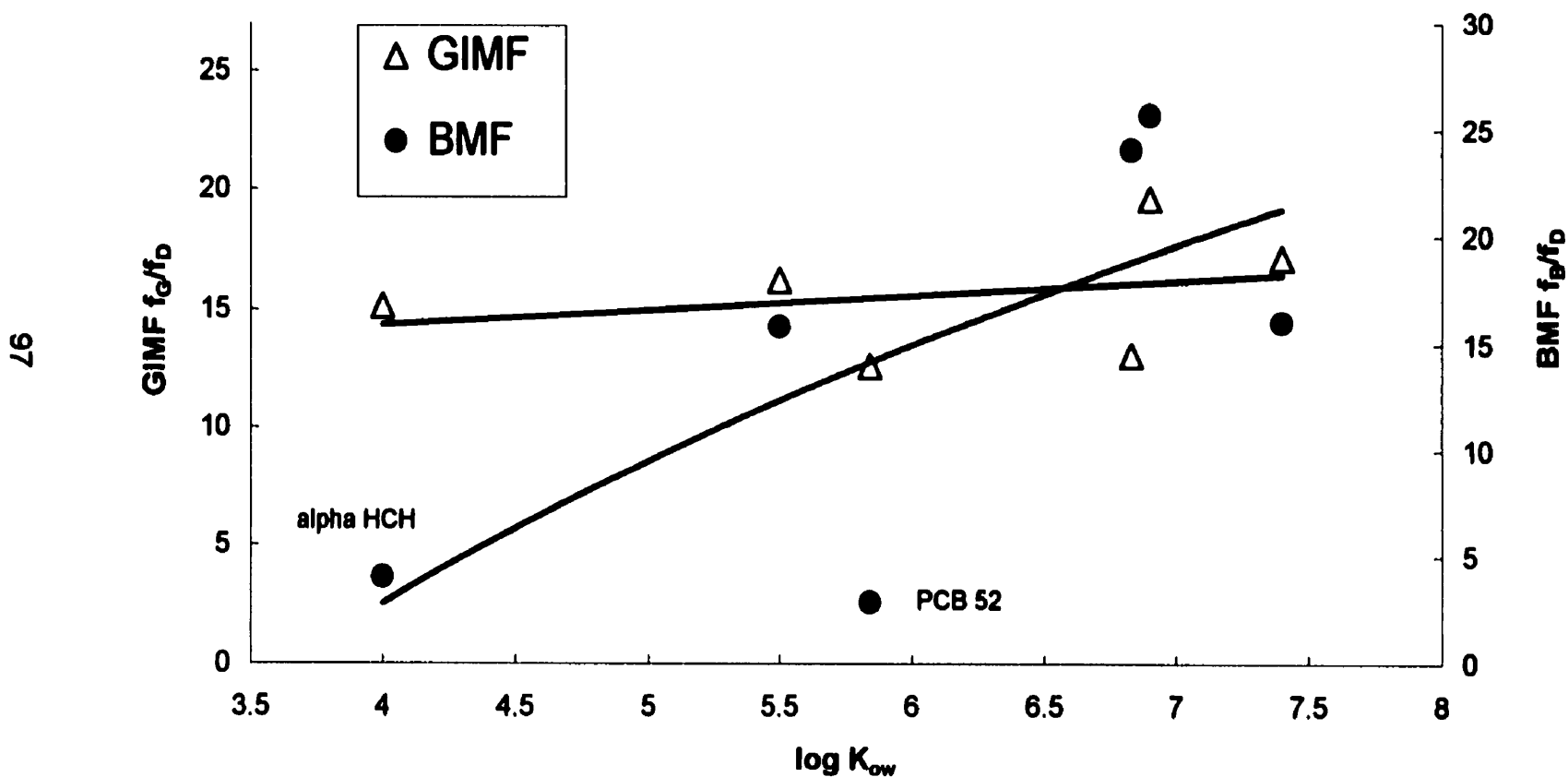


Figure 3.7: Gastro-intestinal magnification factors (GIMF) (f_G/f_D) and Biomagnification factors BMF (f_B/f_D) calculated from fugacities (Pa) in lichens, caribou fecal pellets and caribou tissue collected on the Bathurst range.

CHAPTER 4

Development and field-validation of a fugacity-based bioaccumulation model for organic contaminants in mammals: Application to the lichen-caribou-wolf food-chains of Canada's central and western Arctic

INTRODUCTION

Environmental studies of persistent organic pollutants (POPs), such as DDT and PCBs have been conducted for many species and ecosystems (Elliot *et al.*, 1988; Muir *et al.*, 1988; Oliver and Niimi, 1988; Elkin and Bethke, 1996). Recent policy initiatives involving management of these and other classes of organic chemicals have been developed by different agencies worldwide. In 1995, Environment Canada introduced the *Toxic Substances Management Policy (TSMP)*. More recently in 1998, Canada and other industrial nations began negotiations on a *POPs protocol* under the United Nations Environment Program's (UNEP) Convention for Long-Range Transboundary Air Pollution (LRTAP). A major focus of these policies is to characterize the environmental hazard of chemicals based on specific assessment criteria. Under this screening approach, chemicals that are considered bioaccumulative (BCF or BAFs > 5000), or exhibit an octanol-water partition coefficient (K_{ow}) greater than 10^5 , are targeted for virtual elimination from the environment. The bioaccumulation criteria are largely the result of bioaccumulation experiments or biomonitoring studies involving aquatic species, generally small forage fish (Gobas *et al.*, 1988; Fisk *et al.*, 1998).

Chemical bioaccumulation is a complex process and can be influenced by many dynamic variables. The mechanisms driving these processes are not completely understood for all organisms. Many factors such as diet, life-stage, and physiological processes can affect bioaccumulation of organic contaminants. Ecological and physiological variability between ecosystems and species can differ substantially. It is reasonable to suggest that chemical bioaccumulation could also differ among species and ecosystems.

Hydrophobic organic contaminants associate with lipids and to a lesser extent with organic carbon within organisms. Ruminant herbivores foraging on vegetation consisting mainly of cellulose may accumulate organic contaminants very differently than top-predator carnivores feeding on high lipid muscle and fat tissues. McLachlan (1994) presented a mechanistic model representing chemical bioaccumulation of organic contaminants in a feeding cow. In his study, McLachlan also investigated the mechanisms associated with milk production and excretion in a lactating cow. A significant physiological difference that exists between mammals and other classes of animals is the ability of the females to produce and excrete milk for newborns growth and development. Lactational excretion of hydrophobic chemicals has been suggested as a limiting factor for lower contaminant levels observed in females from marine mammal populations (Muir *et al.*, 1988; Borrell *et al.*, 1995). The natural variability of ecological and physiological factors affecting chemical bioaccumulation is not fully considered in

current policies regarding environmental contaminant management. Establishing bioaccumulation criteria based on experimentally derived BAF or BCF values may not properly assess individual species bioaccumulation and food-chain bioaccumulation. Ecosystem-specific bioaccumulation models may aid this process by incorporating life-time exposure, ecosystem properties, and physiological characteristics.

In this study, a bioaccumulation model for assessing biomagnification and food chain bioaccumulation of organic contaminants in terrestrial ecosystems is developed. The model is applied to an Arctic terrestrial food-chain involving lichens (*Cladina rangiferina* and *Cetraria nivalis*), barren-ground caribou (*Rangifer tarandus*) and wolves (*Canis lupus*). The fugacity-based bioaccumulation model is used to predict chemical concentrations in caribou and wolves of different sex and age classes. This ecosystem is a good candidate for a food-chain modelling study because it illustrates a simple, linear food web structure (Figure 4.1). A previous study investigating organic contaminants in a lichen-caribou-wolf food-chain from northern Sweden was conducted by Villeneuve *et al.* (1985). More recently, a biomonitoring program by the Government of the Northwest Territories (GNWT) was conducted to determine current levels of organochlorine contaminants in

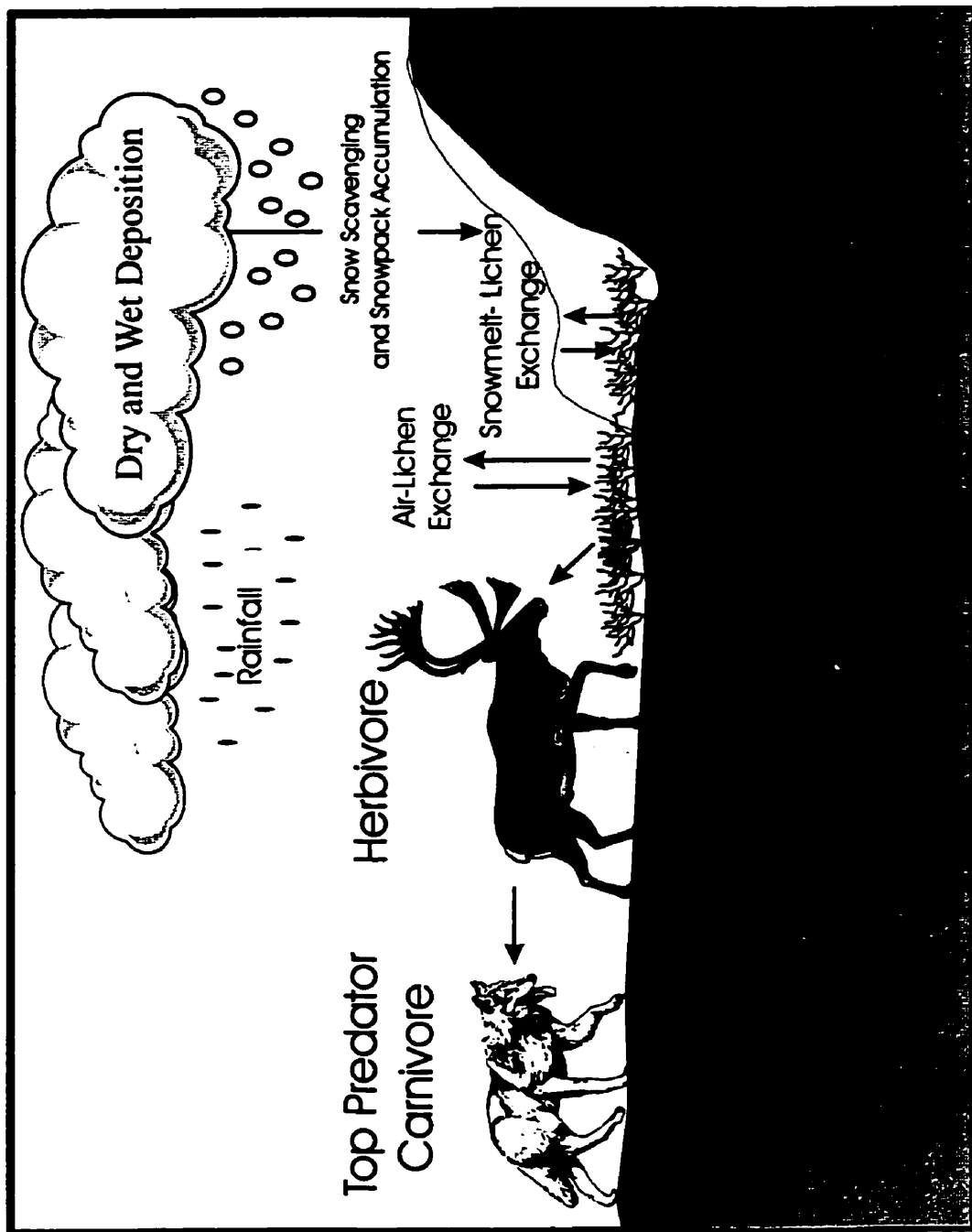


Figure 4.1: Conceptual diagram of the chemical dynamics within an Arctic tundra food-chain involving lichen, caribou, and wolves.

lichen-caribou-wolf food-chains of Canada's central and western Arctic. Chemical concentrations measured in caribou and wolves from Cambridge Bay, Bathurst Inlet and Inuvik (Appendix III) are used to "validate" the model predicted concentrations of organochlorine contaminants in the lichen-caribou-wolf food-chains from these regions.

THEORY

Bioaccumulation of hydrophobic organic contaminants in the tissues of terrestrial mammals involves two routes of exposure; (i) absorption of chemical from air via respiration in the lungs (bioconcentration), and (ii) absorption of chemical from digestion of food (biomagnification) ¹. For mammals, chemical exposure from air is through passive diffusion of airborne contaminants to the organism *via* inhalation and to a lesser extent dermal absorption through the skin surface. However, the predominant route of exposure for chemical accumulation in tissues of terrestrial mammals is through dietary intake and biomagnification. The degree of bioaccumulation in mammals depends on the chemical's properties (i. e., K_{ow}) and biological characteristics such as diet composition, life history, and physiological processes in the gastro-intestinal tract (GIT) of the organism. An important bioaccumulation mechanism specific to newborn mammals, is chemical uptake and elimination *via* milk ingestion by nursing newborns and milk excretion by lactating females. The biomagnification process between consumer and prey organisms at different trophic levels in ecological food-webs can result in food-chain bioaccumulation², causing chemical concentrations on a lipid weight basis (i. e., ng of chemical/g lipid) in organisms with higher trophic status to be greater than those in organisms of lower trophic status. In some cases, metabolic

¹ Biomagnification results when chemical concentrations on a lipid weight basis in a consumer organism exceed the concentration of chemical in the prey species.

²Food-chain bioaccumulation results when chemical concentrations on a lipid weight basis in organisms of a known food-web structure increase with increasing trophic level.

transformation, lactational excretion and the animals growth over time can cause trophic dilution³.

Terrestrial mammals, including humans can accumulate organic contaminants through inhalation, digestion and absorption through skin surfaces. Chemical elimination routes include loss by urinary excretion, metabolic transformation, lactation and fecal excretion. In addition, growth dilution can occur when an animal's body weight increases over time, causing decreased chemical concentrations in the animal, although the chemical is not excreted.

Bioaccumulation in terrestrial mammals can be described by a two compartment model. If exposure of chemical from inhalation of airborne contaminants is insignificant, the model consists of a gastrointestinal tract (GIT), treated as a separate compartment from the organism, and an organism compartment. The GIT and organism are viewed as a single compartments, in which the fugacity of chemical is uniform. There are diffusive fluxes between the GIT and organism compartments via blood perfusion. Advective fluxes of chemical occurs as intake through food and milk ingestion, fecal excretion out of the GIT, and milk excretion out of female mammals. This model is based on the premise that dietary uptake of chemical is the dominant route of chemical uptake bioaccumulation in terrestrial mammals. Although chemical uptake from food consumption is assumed to be the predominant route of chemical exposure to terrestrial mammals, chemical uptake

³ Trophic dilution results when chemical concentrations on a lipid weight basis decline with increasing trophic level due to efficient elimination mechanisms such as metabolic transformation.

from air and skin can be significant for volatile chemicals that can attain high fugacities in the atmosphere relative to those in the organism's food.

The thermodynamic principle of chemical fugacity is useful in explaining chemical transport between environmental and biological media because net passive diffusion of a chemical between different media (i. e., GIT to the organism) occurs in response to fugacity gradients, rather than differences in chemical concentration. Passive transport of chemical between different environmental media occurs only when there is a fugacity gradient, causing net chemical transport from media with high fugacity to low fugacity. The fugacity-based modelling approach, originally described by Mackay (1995), has been subsequently used to describe environmental fate and bioaccumulation processes in aquatic (Gobas *et al.*, 1993; 1998) and agricultural food chains (McLachlan, 1994). To express the bioaccumulation process in fugacity terms, advective fluxes and diffusive flows are expressed as transport parameters (or D values in units of mol/Pa.day) and concentrations are expressed as fugacities (in units of Pascals). The transport parameters (or D values) for advective and diffusive fluxes can be described as the product of the flow rate (G in m^3/day) of a given medium and the fugacity capacity (Z in $\text{mol}/\text{m}^3\cdot\text{Pa}$) of that medium. For example, the transport parameter representing dietary intake (D_D) is the product of the dietary intake rate (G_D) and the fugacity capacity of the ingested food (Z_D). The chemical flux, representing uptake and elimination of chemical (in units of mol/day) are calculated as the product of the transport parameter of a given medium and the

chemical fugacity of that medium. Thus, chemical uptake by ingestion of food is expressed as $D_D f_D$ (mol/day).

Figure 4.2 is a conceptual illustration of the chemical uptake and elimination processes in terrestrial mammals. In this model, net uptake of chemical by caribou and wolves is assumed to be the combined effect of chemical uptake from food ($D_D f_D$), air ($D_A f_A$), and dermal absorption ($D_S f_A$). Chemical loss from the animals may occur by exhalation ($D_E f_A$), urine excretion ($D_U f_U$), fecal excretion ($D_F f_G$), lactation ($D_L f_M$). Chemical elimination by metabolic transformation (in units of mol/day) is represented as $k_M C_B V_B$. The rate constant k_M (d^{-1}) is the metabolic transformation rate of chemical in the organism and is calculated as $0.693/T_{1/2}$, where $T_{1/2}$ (days) is the chemical's half-life in biological tissue. C_B is the chemical concentration (mol/m^3) in the organism, and V_B (m^3) is the volume of the organism.

In essence, the model is characterized by a GIT compartment which receives a flow rate (m^3/day) of food (G_D) and excretes fecal matter at a flow rate of G_F (in m^3/day). The model treats the GIT as well-mixed compartment, thus the digested products in the GIT and fecal matter have the same composition, hence the same fugacity capacity ($Z_G = Z_F$). The organism compartment receives chemical from passive diffusion across the stomach and intestinal walls of the GIT at a rate of

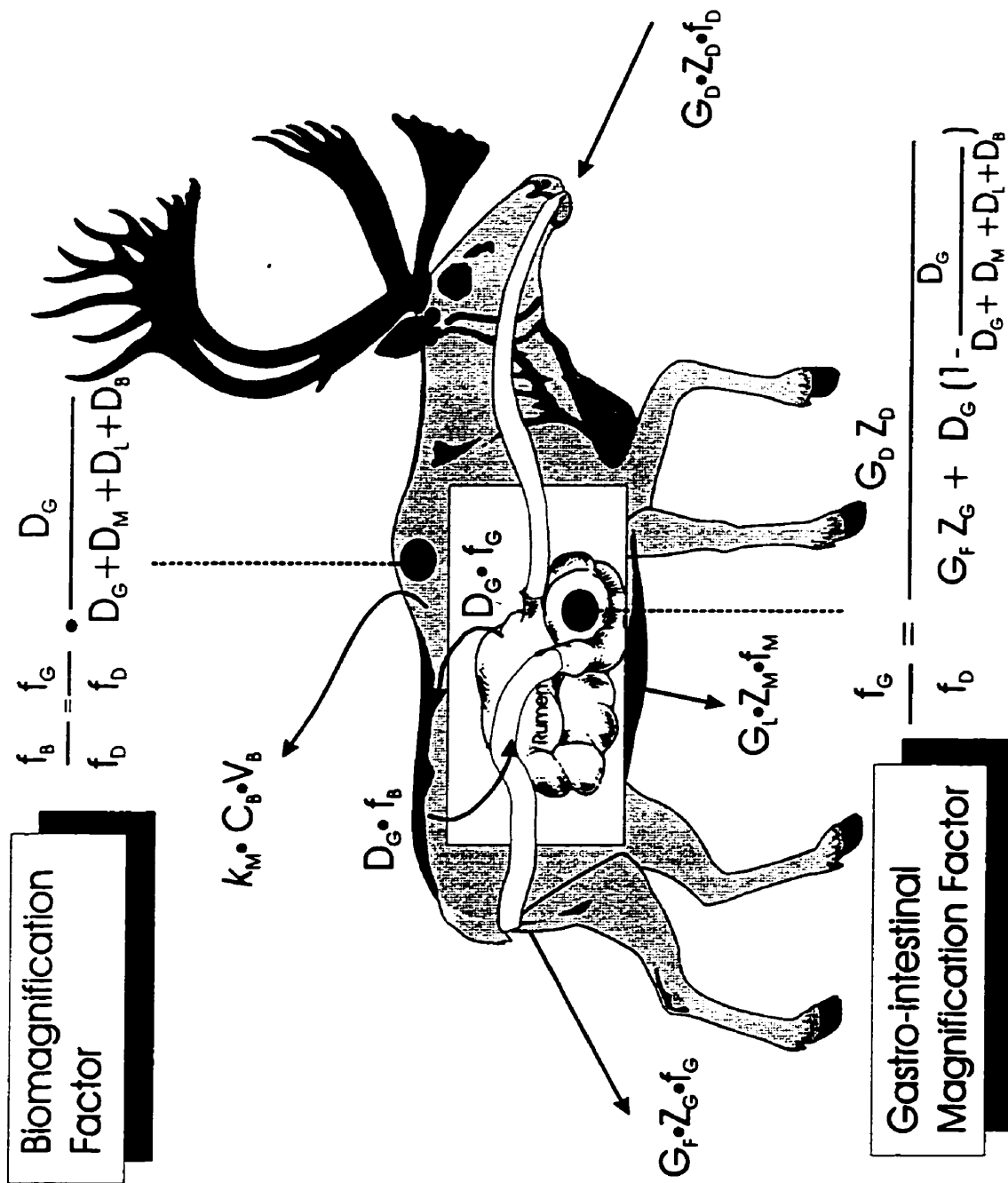


Figure 4.2: Conceptual model of chemical uptake and elimination in terrestrial mammals.

(G_G in m^3/day). The storage capacity of the organism compartment is determined by the volumetric lipid fraction relative to the total volume of the organism. Thus, the chemical storage capacity of the organism is represented by the product of the organism's volume (V_B) and its fugacity capacity (Z_B).

In this two compartment model, a chemical flux into the GIT (N_G , mol/day) occurs during periods of food ingestion. The time-dependent expression for the chemical flux to the GIT is then:

$$N_G = d(V_G Z_G f_G)/dt = D_D f_D + D_G f_G - (D_F + D_G) \cdot f_G \quad (\text{Equation 4.1})$$

Thermodynamic experiments have shown that food digestion and food absorption in the GIT can elevate the chemical fugacity in GIT (f_G) above that in food (f_D). As food passes through the GIT, lipids or organic carbon associated with the food are removed, resulting in a decrease in the fugacity capacity of the GIT contents (Z_G) below the fugacity capacity of the ingested food (Z_D). Food digestion, characterized by ratio of Z_D/Z_G , can raise the chemical fugacity in the GIT of the organism. The fugacity capacity of food (Z_D) can be estimated by the expression $Z_D = Z_w \delta_D \phi_D K_{OW}$ for lipid-rich foods, such as prey in a carnivore's diet. When food is very low in lipids, such as plants, the chemical is assumed to be associated mainly with the organic carbon within the vegetative structure. Thus, the fugacity capacity of vegetation in herbivorous diets can be estimated by $Z_D = Z_w \delta_D \phi_D K_{OC}$.

Z_w is the fugacity capacity of water and is the reciprocal of the Henry's Law Constant (H in units of $\text{Pa}\cdot\text{m}^3/\text{mol}$) of a given chemical, δ_D is the density of the food item (in kg/L), ϕ_D is the lipid or organic carbon content of the food, K_{ow} is the octanol-water partition coefficient and K_{oc} is the organic carbon-water partition coefficient. The fugacity capacity of the excreted feces (Z_F) following digestion can be estimated by the expression $Z_F = (1-\alpha) \cdot Z_D$, where α is the extraction efficiency of lipids or organic carbon associated with the organism's diet.

Food absorption, characterized by the ratio of dietary intake to excretion (G_D/G_F), results in a reduced substrate volume within the GIT. The reduction of substrate can cause chemical concentrations in the GIT to increase. The increased chemical concentration due to food absorption in the GIT also causes an increased chemical fugacity in the GIT. The combined effect of food digestion and food absorption result in a chemical fugacity in the GIT (f_G) that is elevated above the fugacity in its food (f_D). The ratio of f_G/f_D can be viewed as a gastro-intestinal magnification factor and is calculated as:

$$f_G/f_D = D_D / \{D_F + D_G(1 - D_D / (D_G + D_W + D_B + D_U))\} \quad (\text{Equation 4.2})$$

The extent to which f_G is elevated above f_D (i. e., GIT magnification) depends on degree of food digestion and food absorption in the GIT. The increase in chemical fugacity in the GIT allows for net passive diffusion of chemical from the GIT to the organism. Passive diffusion of chemical via blood perfusion across the stomach and intestinal walls of the GIT can occur as a diffusive flow in both directions

between the GIT and biotic tissues of the organism. Transport of chemical from the GIT to the organism via passive diffusion can then elevate the fugacity in the organism (f_B) above that fugacity in its food (f_D) (i. e., biomagnification).

Following gastro-intestinal uptake of chemical ($D_G f_G$), the magnitude of biomagnification (f_B/f_D) depends on the rate of chemical elimination from the organism through urinary excretion (D_U), exhalation (D_A), lactation (D_L), metabolic transformation (D_M) as well as growth dilution (D_B), determined by the organism's rate of growth. The ratio of f_B/f_D is the fugacity-based biomagnification factor and is calculated as,

$$f_B/f_D = f_D/f_D \cdot (D_G/(D_G+D_A+D_U+D_M+D_L+D_B)) \quad (\text{Equation 4.3})$$

The mass balance equation representing the chemical flux (mol/day) from the GIT into the organism (N_B) is represented by the equation,

$$N_B = d(V_B Z_B f_B)/dt = D_A f_A + (D_G f_G) - (D_G + D_L + D_A + D_M + D_B) \cdot f_B \quad (\text{Equation 4.4})$$

At steady state in the GIT (i. e., $N_G = 0$), the chemical fugacity is then,

$$f_G = (D_D f_D + D_G f_B)/(D_G + D_F) \quad (\text{Equation 4.5})$$

If we assume this steady state condition in the GIT Equation 4.5 can be substituted into equation 4.4, resulting in an overall chemical flux (N_B , in mol/day) to the organism of

$$N_B = d(V_B Z_B f_B)/dt = D_A f_A + E_D D_G f_D - E_D D_F f_B - (D_L + D_A + D_U + D_M + D_R) \cdot f_B \quad (\text{Equation 4.6})$$

where, E_D represents an organism's dietary uptake efficiency and can be calculated as $D_G/(D_F + D_G)$. The dietary uptake efficiency (E_D) is a representation of an organism's ability to transfer chemical between the GIT and its biotic tissues. The value of E_D depends on an organism's digestion strategy and the composition of its diet. The definitions and symbols used to represent parameters in the bioaccumulation model are summarized in Table 4.1.

If $C = f \cdot Z$, then the chemical concentration (C , in mol /m³) of a given media can be calculated as product of the fugacity (f , in Pa) and fugacity the capacity (Z , in units of mol/m³ .Pa) of that media. The time-dependent calculation of chemical fugacity using equation 4.6, ($dt = 1$ day), represents the chemical fugacity in the organism for a given day within the organism's lifetime. The chemical concentration in the organism's tissues (C_B , mol/m³ lipid) can then be derived using the fugacity capacity of lipid (Z_L) and the calculated fugacity in the organism (f_B), with the equation $C_B = f_B \cdot Z_L$

Table 4.1: Definition, symbols and units of parameters used in the model representing chemical bioaccumulation of organic chemicals in terrestrial mammals.

Parameter	Units	Definition
f_B, f_D, f_G, f_F, f_M	Pa	Chemical fugacity in biota(_B), diet (_D), GIT(_G), feces (_F), and milk (_M)
N_B, N_G	$\text{mol}\cdot\text{d}^{-1}$	Chemical flux in biota, and GIT
V_B, V_G	m^3	Volume of biota, and GIT
G_D, G_F	$\text{m}^3 \cdot \text{d}^{-1}$	Dietary intake and fecal excretion rates, respectively
$D_D, D_A, D_F, D_G, D_B, D_{MI}, D_L$	$\text{mol}/\text{Pa}\cdot\text{day}$	Transport parameters of chemical intake from consumption of prey, air, chemical egestion through feces, chemical diffusion between the GIT and the organism, growth, chemical intake from milk ingestion and chemical loss through lactation
Z_B, Z_G, Z_D	$\text{mol}/\text{m}^3\cdot\text{Pa}$	Fugacity capacity of biota, GIT and feces, and prey
E_D	unitless	Dietary absorption efficiency

MODEL SIMULATIONS

Chemical fugacities in lichens (f_D) are the base input to this food-chain bioaccumulation model. Chemical fugacities in lichens, representing the diet of caribou (f_D) from Bathurst Inlet, Cambridge Bay and Inuvik were calculated from the observed chemical concentrations in lichens collected from these regions and are reported in Appendix II. Vegetation samples, including two common tundra lichens (*Cladina rangiferina* and *Cetraria nivalis*) were collected on the range of the Bathurst caribou herd in May-June 1997 and July 1998. From 1993 to 1995, the Department of Renewable Resources of the Government of the Northwest Territories (GNWT) conducted a biomonitoring program which provided chemical concentrations of various organochlorine contaminants lichens in caribou and wolves from Inuvik, Cambridge Bay and Bathurst Inlet. The age, sex and weight of caribou and wolves sampled from these regions were determined at the time of collection. The chemical fugacities calculated for adult female caribou and are used to characterize the fugacity of a newborn (f_B) and also the fugacity in the newborn's milk diet (f_M). To test this food-chain bioaccumulation model, simulations of 22 organic chemicals including PCBs, chlorobenzenes and various organochlorine pesticides over a 15 year period were conducted to generate predicted chemical concentrations in caribou and wolves from Cambridge Bay (Victoria Island herd), Bathurst Inlet (Bathurst herd) and Inuvik (Bluenose herd). The time-step (dt) used in model simulations was 1 day. The initial chemical fugacity (f_0 , in Pa) in caribou and wolves (i. e., $dt = 0$) was determined from the

observed chemical concentrations in adult female caribou and wolves (C_B , mol/m³ lipid) and the fugacity capacity of lipid (Z_L , mol/m³·Pa) using the equation $f_o = C_B/Z_L$.

Natural variability within ecological systems is a well known phenomenon, and variability of chemical dynamics within ecological food-chains occur (Villeneuve *et al.*, 1985; Muir *et al.*, 1988; Norstrom *et al.*, 1988; Elkin and Bethke, 1996).

Predator-prey interactions of migratory mammals such as the relationship that exists between barren-ground caribou wolves in the Arctic are dynamic. Because ecosystems are dynamic and variable in nature, contaminant exposure to animals can vary between seasons and among animals of different age-classes or sex.

For example, the maternal fugacity, hence the internal concentration of a pregnant female can influence the chemical concentrations of a newborn later in life.

Migratory herbivores and co-migratory predators, such as caribou and wolves may experience various exposure levels of contaminants through food ingestion.

Previous modelling studies involving trophic transfer of contaminants have included natural variability in model forecasts (Gobas, 1993a). In this study, Monte-Carlo simulations (MCS) with sample sizes of 10,000 were conducted using Crystal Ball © (Decisioneering) to include the sources of variability associated with the observed maternal and dietary concentrations. The natural variability associated with dietary and maternal chemical concentrations for caribou and wolves differs among chemicals. In addition to the natural variability associated

with dietary and maternal chemical concentrations, inherent model parameters such as feeding rates, fecal excretion rates, lactation rates, lipid contents and the animal's body weight can also vary among animals in a population.

A sensitivity analysis was performed to test the sensitivity of model outputs to changes in model input parameters. The sensitivity analysis was conducted for PCB 180 in a newborn calf (125 days old), adult female caribou (6.25 years old) and an adult male caribou (6.25 years old). The base-line concentrations of PCB 180 for the sensitivity analysis included 0.03 ng/g dry wt. In lichens, 0.5 ng/g lipid in pre-natal female (i. e., maternal concentrations). The value of several model parameters were independently reduced by 10% to determine the change in PCB 180 concentrations (ng/g lipid) predicted for a newborn calf , adult female caribou and an adult male caribou.

MODEL PARAMETERIZATION

Parameters values for chemical properties and biological characteristics used in model simulations are summarized in Tables 4.2 and 4.3, respectively. The model parameters used in the MCS were assumed to exhibit normal distributions.

Therefore, the mean and standard deviations of the parameters were used in the MCS to calculate the standard deviations of the predicted concentrations. To determine the natural variability inherent to the model, MCS were conducted using only the variability associated with inherent model parameters. Inherent model parameters include feeding rates (G_D), fecal excretion rates (G_F), lactation rates (G_L), lipid contents (ϕ_B), and the animal's size (V_B). The variability associated with these inherent model parameters is the same for all chemicals. Thus, the standard deviations calculated from the MCS using only inherent model parameters are used to represent model confidence for predictions from all simulations. Variability in physical-chemical properties was not considered because of the difficulties of assessing variability in these values.

Chemical properties

The chemicals and their physicochemical properties were obtained from Mackay *et al.*, 1992; Hawker and Connell, 1988; and Dunnivant and Elzerman, 1992.

Logarithms of water partition coefficients ($\log K_{OW}$), Henry's law constants

Table 4.2: Physicochemical properties of organic chemicals used in model simulations.

CHEMICAL	Log K _{ow} ^{a,b}	Henry's Law Constant (H) ^{c,d}	Molecular Weight ^e	Half-life in Tissue T _{1/2} (days)
Alpha HCH	4.0	2	400	7.14
Beta HCH	4.5	1	400	NA
Dieldrin	5.4	2	374	NA
1,2,4,5 TCB	4.7	101	215	33
QCB	5.01	75	245	27
Hexachlorobenzene	5.5	65	285	210
PCB 31	5.67	24	292	196
PCB52	5.84	20	292	500
PCB 66/95	6.2	20	292	670
PCB 99	6.39	88	326	NA
PCB 101	6.38	9	326	1000
PCB 118	6.74	23	326	NA
PCB 149	6.67	9	326	NA
PCB 138	6.83	8	361	NA
PCB 153	6.9	10	361	>1000
PCB 182/187	7.2	8	395	NA
PCB 180	7.4	25	358	NA
PCB 201	7.62	38	430	NA
PCB 206	8.09	23	462	>1000
PCB 170/190	7.3	12	395	NA
PCB 194	7.8	16	395	>1000
Heptachlor epoxide	4.98	5	395	NA
Mirex	6.89	15	545	NA
Octachlorostyrene	6.29	15	379	NA

^a Octanol-water partition coefficients (logK_{ow}) for PCBs were obtained from Hawker and Connell, 1988.

^b Octanol-water partition coefficients (logK_{ow}) for other organochlorine compounds were obtained from Mackay *et al.*, 1995.

^c Henry's Law constants (Pa•m³/mol) for PCBs were obtained from Dunnivant and Elzerman, 1992.

^d Henry's law constants (Pa•m³/mol) for other organochlorine compounds were obtained from Mackay *et al.*, 1995.

^e Molecular weights (g/mol) for all compounds were obtained from Mackay *et al.*, 1995.

^f Half-lives in biotic tissues were obtained from Mackay *et al.*, 1995, using results observed in rainbow trout.

Table 4.3: Model Parameterization.

Barren-ground caribou (*Rangifer tarandus*) model parameters

Diet composition =	50% <i>Cladina rangiferina</i> , 50% <i>Cetraria nivalis</i>
Feeding rate (G_D) =	2.1 ± 1.5 kg/day
Fecal excretion rate =	0.66 ± 0.4 kg/day
Urinary excretion rate =	0.45 L/day
Dietary uptake efficiency (E_D) =	50%
Lactation rate (G_L) =	1.2 L/day ^a
Suckling rate ($G_{MI} = G_L$) =	1.2 L/day ^a
Lipid content in caribou milk =	15 ± 5 %
Weaning begins =	130 days after birth
Mean annual lipid content =	8% ^b

Tundra wolf (*Canis lupus*) model parameters

Diet composition =	100% caribou
Feeding rate (G_D) =	3.5 ± 1.5 kg caribou /day ^c
Fecal excretion rate =	1.1 ± 0.5 kg/day
Urinary excretion rate =	0.25 L/day
Dietary uptake efficiency (E_D) =	50%
Litter size (L) =	3 ± 3 pups/female ^d
Lactation rate (G_L)•litter size =	(1 L/day)•L ^d
Suckling rate (G_{MI}) =	1 L/day ^d
Lipid content of wolf milk =	15 ± 5 %
Weaning begins =	68 days after birth
Mean annual lipid content =	12 ± 8 % ^e

^a Suckling rates, weaning and calf growth rates of caribou were obtained from Lavigueur and Barrette (1992). In the MCS calculation, adult female caribou are assumed to exhibit an equal probability of lactating (i. e., $G_L = 0 - 1.2$ L/day).

^b Seasonal lipid contents for male and female caribou were obtained from Miller and Broughton (1974), and Dauphine (1979) are shown in Figure 4.3.

^c See text for a detailed description of demographic distribution calculation of caribou in the diet of tundra wolves.

^d Suckling rates, weaning and pup growth for wolves were obtained from Kuyt (1972). In the MCS calculation, adult female wolves are assumed to exhibit an equal probability of lactating (i. e., litter size (L) = 0 - 6 pups at a lactation rate of $G_L = 1$ L/day).

^e Seasonal lipid changes for male and female wolves were obtained from Kuyt (1972) and are shown in Figure 4.4.

(Pa·m³/mol) and molecular weights (g/mol) compiled from these sources are listed in Table 4.2.

Maternal transfer and dietary intake

Caribou: Barren-ground caribou (*Rangifer tarandus*) are migratory ungulates that can travel across vast distances over the course of a year. A pregnant female caribou gives birth to a calf in spring (late May- early June) following the northern migration to the herd's calving grounds. The age at first conception for female caribou is 2 years, although yearlings can conceive (Dauphine, 1976). Suckling rates, weaning and calf growth rates have been documented in captive woodland caribou (*Rangifer tarandus caribou*) by Lavigueur and Barrette (1992). A suckling rate of approximately 1.2 L/day was observed in captive adult female caribou. Therefore, a milk ingestion rate (G_{MI}) for caribou calves of 0.0012 m³/day is used to represent milk intake of caribou in the model. The fat content of caribou milk is approximately 20%. Caribou calves can suckle milk from multiple females within the herd. For the purpose of this study, the lactation rate of the post-birthing female is assumed to be equal to the calf's suckling rate ($G_L = 0.0012$ m³/day). Weaning of caribou calves from a milk diet to lichens begins approximately 130 days after birth.

In winter, caribou forage mainly on lichens buried beneath the snow (Miller, 1976; Holleman *et al.*, 1979). In summer, lichens comprise much of the caribou diet, but other vegetation such as *Eriophorum* flowers and tundra willows (*Salix glauca*) are

consumed by caribou grazing on their summer range (Miller, 1976). For the purpose of this study, caribou are assumed to feed mainly on two common tundra lichens (*Cladina rangiferina* and *Cetraria nivalis*). The daily intake of lichen by foraging caribou (G_D) on the tundra was found to be approximately 2.1 ± 1.5 kg/day (Holleman *et al.*, 1979). This corresponds to a dietary intake of lichens by caribou (G_D) of 0.0021 ± 0.0015 m³/day. Chemical fugacities in lichens (f_D) were calculated from observed chemical concentrations (C_D , mol/m³) and the fugacity capacity of lichens (Z_D) (i. e., $f_D = C_D \div Z_D$). The fugacity capacity of lichen was estimated using the equation $Z_D = \rho \varnothing (0.41Kow \cdot Zw)$, where; ρ is the density of lichen (0.54 ± 0.09 kg/L) and \varnothing is the organic carbon content of lichen ($96.1 \pm 0.06\%$).

Wolves: Wolves in the Canadian Arctic mate in late march. Following a 60 to 65 day gestation period, adult females (> 2 years) may give birth to litters of two to seven pups. Kuyt (1972) observed dietary intake of captive adult wolves and newborn wolf pups. Observations from these dietary and growth studies of captive wolves are used to represent milk and caribou ingestion rates for wolves in the model. Wolf pups suckle milk (18 % fat) at a rate (G_{MI}) of approximately 0.001 m³/day. The pups are weaned from milk to a meat diet (i. e., caribou) after 68 days.

In many regions of the Arctic, wolves follow caribou migration patterns to fulfill dietary requirements throughout the year (Heard and Williams, 1992). In other regions, wolf packs may switch to alternative prey at different times of the year. Bathurst wolves migrate in close approximation to the Bathurst caribou herd, and

thus are assumed to feed close to 100% on caribou. In this study, wolf packs from Inuvik and Cambridge Bay are assumed to prey solely on caribou on their geographic ranges. Kuyt (1972) estimated wolves on the barren-ground caribou range on average need approximately 23 caribou per year to fulfill their annual nutritional requirements. The daily intake of caribou, which includes muscle, fat, and viscera, estimated by Kuyt was 3.5 ± 1.5 kg/day. Kuyt further calculated that these 23 caribou would be comprised of 5 calves, 2 yearlings and 16 adult caribou 2 years or older. Model predicted chemical fugacities in calves, yearlings and adult caribou were used to represent the chemical intake via food ingestion by wolves. Due to the seasonal fluctuation of chemical fugacity expected in caribou (i. e., f_{CARIBOU} in summer $> f_{\text{CARIBOU}}$ in fall) the chemical fugacity in the diet of wolves is seasonally dependent. Therefore, a dietary intake rate (G_D) of 0.0035 ± 0.0015 m³/day and the average of chemical fugacities in 5 calves, 2 yearlings and 16 mature caribou (f_D) at a given time-step in the model simulation was used to represent dietary exposure of chemical to wolves.

Ecology and physiology

Caribou: Barren-ground caribou can exhibit large fluctuations in body fat content throughout the year (Dauphine, 1979; Adamczewski, *et al.*, 1986; Huot, 1988). Various estimation techniques to measure fat content of barren-ground caribou have been reported in the literature (Davis *et al.*, 1987; Gerhart *et al.*, 1996). Lipid contents reported in caribou using these various methods ranged

from 2% in calves to over 20% in adult males. Growth rates, body condition and composition of barren-ground caribou calves and adults have previously been reported by Chan-McLeod *et al.* (1994), Lavigne and Barrette (1992) and Miller and Broughton (1974). Observed body weights of barren-ground caribou from these studies ranged from 5 kg for newborn calves to approximately 150 kg in adult bulls. The fugacity capacity of caribou is determined by its lipid content using the expression $Z_B = \varphi \cdot Z_L$, where φ is the lipid content (%) and Z_L is the fugacity capacity of lipid. The body weight of the animal is expressed as a volume (V_B , m^3). The product $V_B \cdot Z_B$ represents the chemical storage capacity of the organism. The time-dependent changes in lipid content (% lipid) and body size expressed in units of volume (m^3) for male and female caribou used in the model simulations are shown in Figure 4.3.

The extent of food digestion and food absorption in caribou is a function of the extraction of digestible vegetation matter (e. g., cellulose) and the excretion of indigestible vegetation material (e. g., lignins). During grazing periods, the fugacity capacity of the caribou GIT content (Z_G) of caribou is assumed to be 5 times less than the fugacity capacity of the ingested lichens (Z_D). The value of Z_G in the GIT of grazing caribou is based on the assumption that the extraction

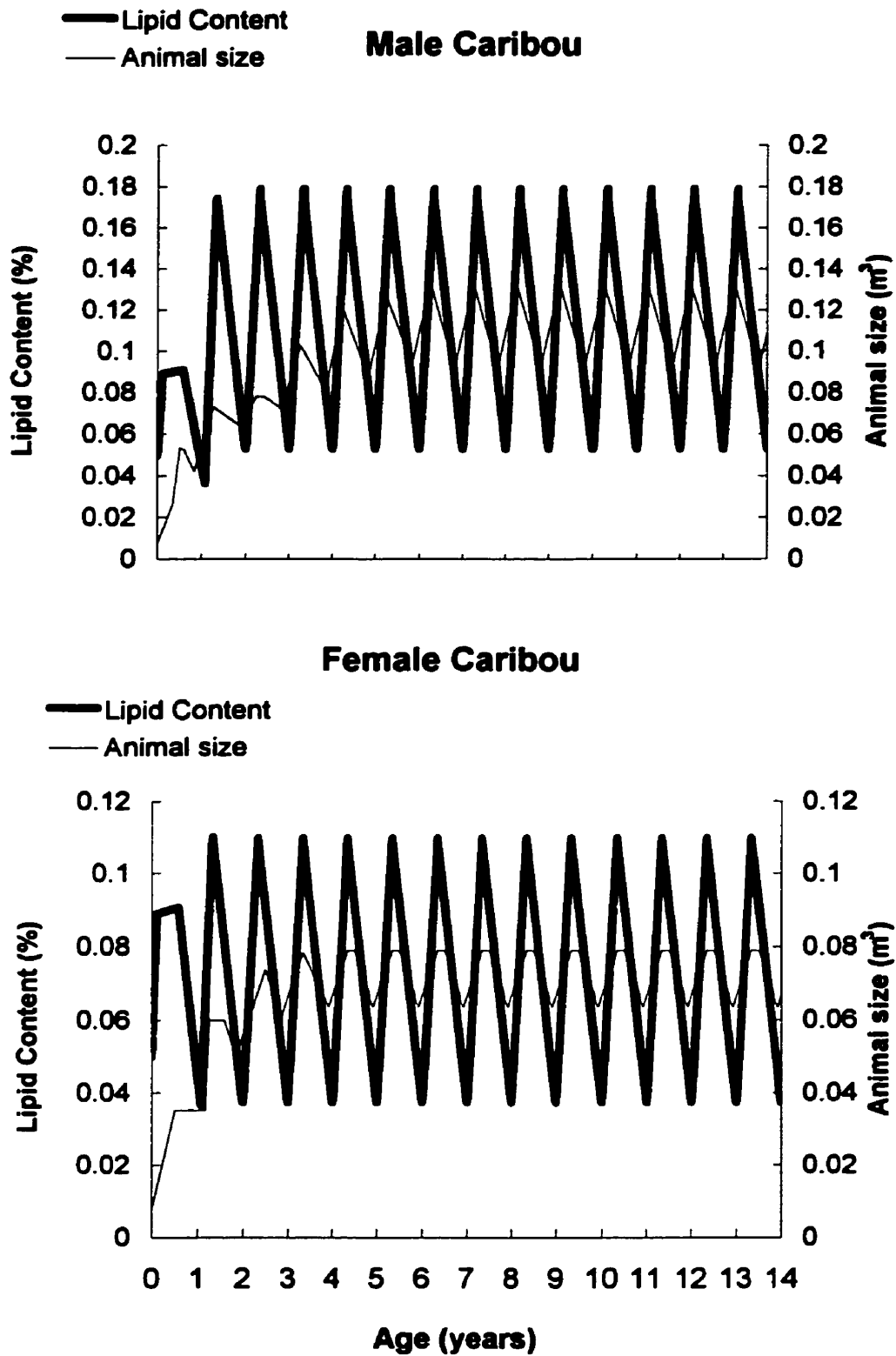
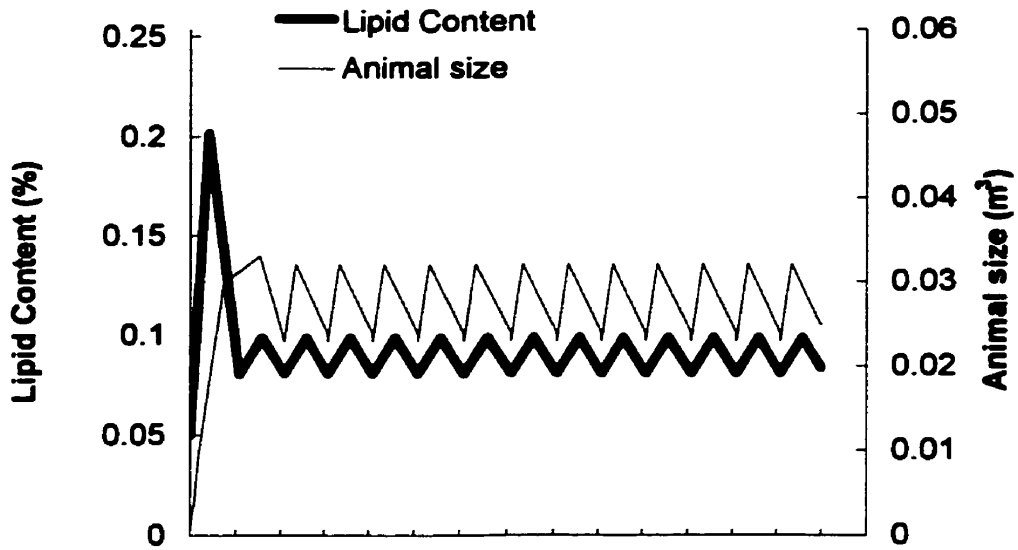


Figure 4.3: Time-dependent changes in animal size, as expressed by volume (m^3) and lipid contents (% lipid) of male and female caribou used in model simulations.

efficiency of digestible cellulose (α) in caribou is approximately 80% (Boertje, 1990). The fugacity capacity of the GIT contents (Z_G) in newborn caribou calves digesting milk is related to the extraction of lipids in the milk. If a lipid extraction efficiency (α) of 90% is assumed, then a Z_G in a nursing calf is estimated to be 10 times less than the fugacity capacity of the ingested milk (Z_M). During a captive feeding study, excretion of fresh feces in goats was shown to be approximately 60% of the ingested feed (Rozman *et al.*, 1984). If we assume that 50% of the excreted fecal matter is comprised of body water, then the excretion of dry fecal matter is approximately 30% of the ingested feed. A fecal excretion rate (G_F) of 0.00066 m³/day, a value approximately 30% of the ingestion rate (G_D), is used to represent the rate of chemical elimination by fecal excretion in caribou. For newborn caribou calves, a fecal excretion rate (G_F) of 0.0004 m³/day was determined from the ingestion rate of milk ($G_{MI} = 0.0012$ m³/day).

Wolves: Newborn pups are approximately 0.5 kg at birth. Kuyt (1972) observed rapid growth of wolf pups in the first month of life, at which time the pups continues to grow but at a slower rate. The average weight of adult male wolves is approximately 30 - 40 kg, while mature females are smaller exhibiting body weights from 30 - 35 kg. Like caribou, the fat content and body weights of wolves can also fluctuate throughout seasons. Following these observations, the time-dependent volume of wolves (V_B , m³) and lipid contents of male and female wolves were calculated and are shown in Figure 4.4.

Female Wolf



Male Wolf

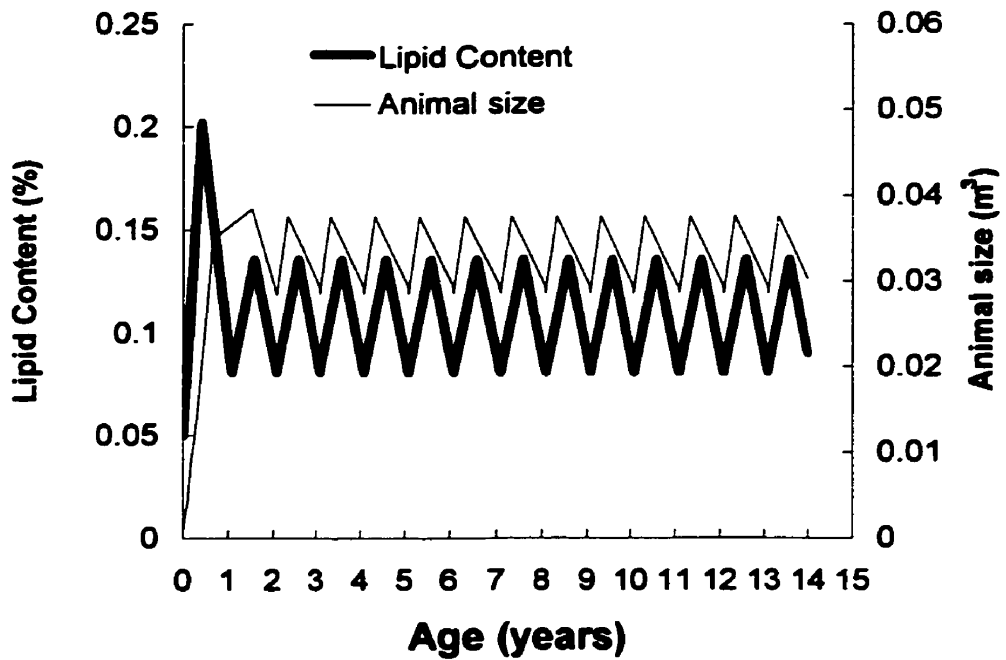


Figure 4.4: Time-dependent changes in animal size, expressed as volume (m³) and lipid contents (% lipid) for male and female wolves used in the model simulations

The extent of food digestion (Z_D/Z_G) in wolves is related to the extraction of lipids associated with its food. A lipid extraction efficiency for wolves is assumed to be approximately 90%. This results in a fugacity capacity of GIT contents of wolves (Z_G) that is 10 times less than the fugacity capacity of caribou (Z_D) and ingested milk (Z_M). The fecal excretion rates (G_F) for wolves is assumed to be approximately 30% of the ingested food.

EVALUATION OF MODEL BEHAVIOUR

To assess the predictability of the model, model predicted chemical concentrations in caribou and wolf tissues (ng/g lipid) are compared to the observed chemical concentrations in animals sampled from Cambridge Bay, Bathurst Inlet and Inuvik. The chemical concentrations in individual caribou and wolves of known age and sex used for model validation are shown in Appendix IV. Model predictions corresponding to the specific age and sex of animals sampled from Cambridge Bay, Bathurst Inlet and Inuvik were compared to the chemical concentrations measured in fat samples of individual animals.

Systematic error in the model was determined by comparing the model predicted concentrations (C_P) with the observed chemical concentrations (C_O). Specifically,

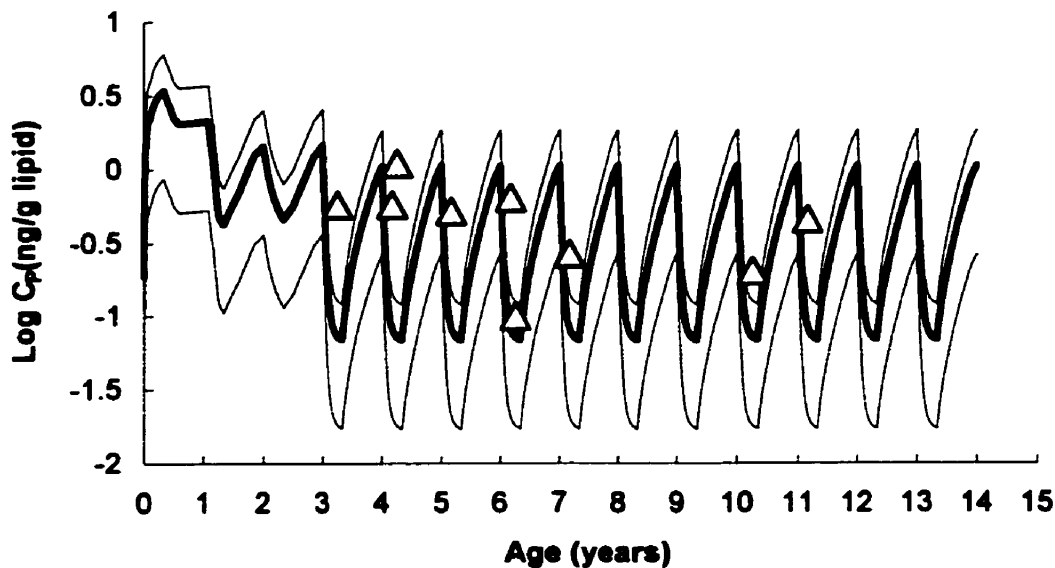
logarithms of the C_P/C_O ratios were calculated for each prediction. The $\log C_P/C_O$ values for multiple predictions (i. e., for all chemicals) were combined to generate a population of predicted versus observed values for a given simulation (e. g. Bathurst male caribou in September). The mean of the population of $\log C_P/C_O$ ratios were used to evaluate the model bias, while the confidence interval (i. e., $1.96 \times SD$) of the $\log C_P/C_O$ population was used to express the uncertainty of the model predictions.

RESULTS AND DISCUSSION

Life-time bioaccumulation profiles in caribou and wolves

The predicted life-time bioaccumulation profiles of PCB180 in caribou and wolves from Bathurst Inlet are illustrated in Figures 4.5 and 4.6, respectively. The model predicted chemical concentrations of PCB 180 in Bathurst female caribou during July and September were 0.32 ± 0.31 ng/g lipid and 0.24 ± 0.23 ng/g lipid, respectively. Chemical concentrations and standard deviations observed in adult female caribou (ages ranging from 3 to 11) in July and September at Bathurst Inlet were 0.49 ± 0.14 ng/g lipid and 0.52 ± 0.39 ng/g lipid, respectively. The model predicted chemical concentration of PCB 180 was 2.2 times lower than the observed concentrations in female caribou sampled at Bathurst Inlet during September. The lower concentrations of PCB 180 predicted in female caribou following the calving

Life-time Bioaccumulation of PCB180 in Bathurst Female Caribou



Life-time Bioaccumulation of PCB180 in Bathurst Male Caribou

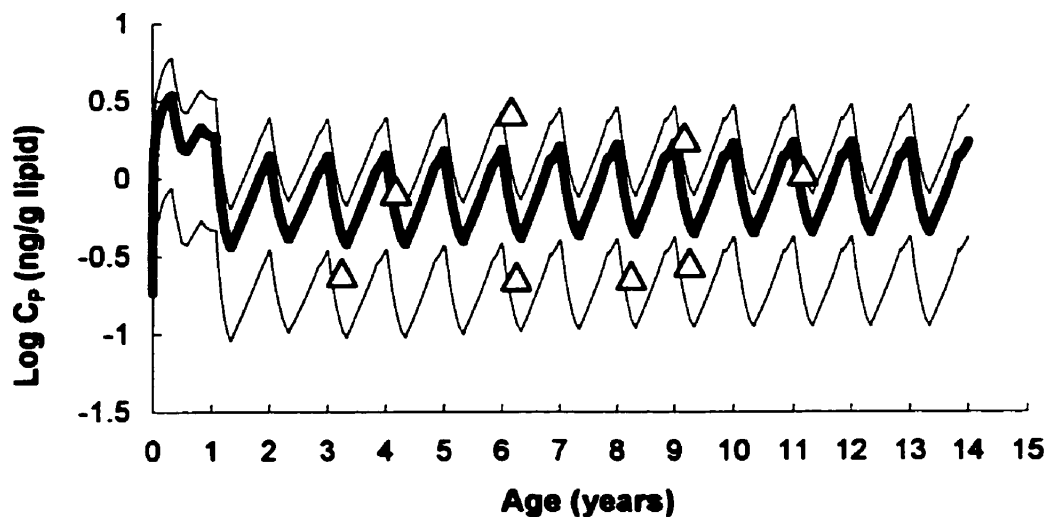
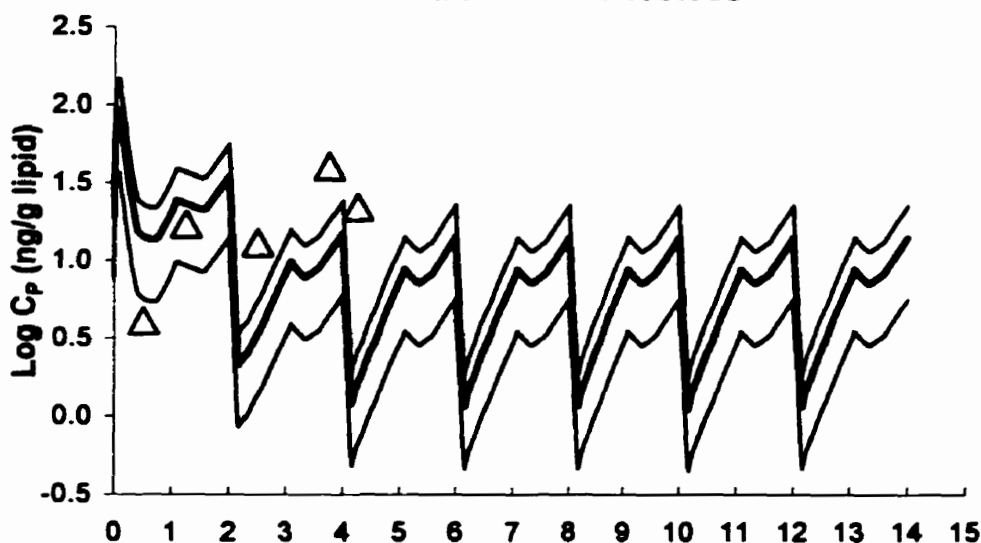


Figure 4.5: Predicted chemical concentrations (log concentration, ng/g lipid) of PCB 180 over the life-time of caribou from Bathurst Inlet. Solid line represents the model predicted chemical concentrations. Lines above and below the predicted values are the standard deviations of the mean calculated from Monte Carlo Simulation. Δ represents the observed chemical concentrations in fat samples of individual caribou from Bathurst Inlet.

Life-time Bioaccumulation of PCB180 in Bathurst Female Wolves



Life-time Bioaccumulation of PCB 180 in Bathurst Male Wolves

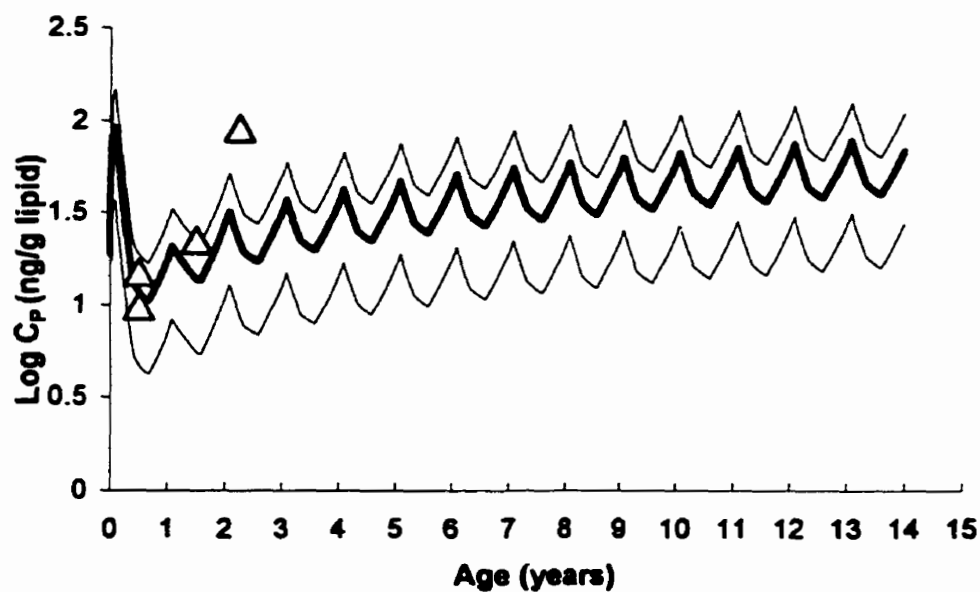


Figure 4.6: Logarithms of predicted chemical concentrations (ng/g lipid) of PCB 180 over the lifetime of wolves from Bathurst Inlet. Solid line represents the model predict chemical concentrations. Lines above and below the predicted values are the standard deviations calculated by the MCS. Δ represents the observed chemical concentrations individual wolves sampled at Bathurst Inlet.

season (i. e., fall) may be due to the presence of lactating and non-lactating females in the caribou herd. Also, the model may under-predict the chemical concentrations in post-natal females by over-estimating their whole-body lipid content. After calving in June and nursing through September, the post-natal female may have reduced fat deposits, and hence lower whole-body lipid contents. The model predicted chemical concentrations of PCB 180 in male caribou during July (1.8 ± 1.45 ng/g lipid) were similar to the observed concentrations in male caribou (ages ranging from 3 to 11) sampled in July at Bathurst Inlet (1.73 ± 0.77 ng/g lipid). However, the model predictions for Bathurst male caribou in September (0.48 ± 0.36 ng/g lipid) over-estimated the concentrations of PCB 180 observed in male caribou sampled during September (0.25 ± 0.03 ng/g lipid).

The predicted chemical concentration of PCB 180 in a 6-month-old female wolf at Bathurst Inlet (19.4 ± 10.9 ng/g) was greater than the observed concentration measured in a 6-month-old female wolf from Bathurst Inlet (4.1 ng/g lipid). The chemical concentration of PCB 180 predicted in a 3-year-old female wolf (38.5 ± 23.3 ng/g lipid) was similar to the observed concentrations measured in a 3-year-old female (40.2 ng/g lipid) sampled at Bathurst Inlet. Chemical concentrations (ng/g lipid) of PCB 180 predicted in male wolves aged 6 months, 1 year and 2 years from Bathurst Inlet were 11.7 ± 7.01 , 15.5 ± 9.27 and 46.5 ± 27.9 , respectively. Model predictions for 6-month and 1-year old male wolves were similar to the concentrations observed in a 6-month and 1-year old male at Bathurst Inlet.

However, the model prediction for a 2-year old male wolf was less than the observed chemical concentration of PCB 180 in a 2-year old male wolf (90.61 ng/g lipid) sampled at Bathurst Inlet.

At the age of maturity, approximately 2 years of age, chemical concentrations in female caribou decline due to elimination *via* lactational excretion. If pregnancies then occur on an annual basis, a cyclical pattern of chemical elimination and accumulation occurs. After the initial increase from birth and subsequent suckling, the chemical concentrations in adult male caribou also exhibits a cyclical bioaccumulation behaviour. The fugacity fluctuations observed in male caribou result from seasonal changes in lipid content and body size (Figure 4.5). The observed chemical concentrations in male caribou sampled during July were approximately 7 times greater than those concentrations found in males sampled in September. In the model, the chemical fugacity in caribou and wolves is hypothesized to fluctuate throughout periods of fat deposition in the fall and fat depletion during spring and early summer. As the animal stores fat for an oncoming winter, increased lipid content results in dilution of chemical, thus causing a reduction in chemical fugacity. Conversely, fat depletion in the early summer would concentrate chemical within the reduced lipid reserves, causing the fugacity in the animal to increase. Based on seasonal changes in lipid content and body size, the model prediction of chemical dynamics during fat mobilization in fall and depletion in summer is consistent with the observed data.

Figure 4.6 illustrates the chemical bioaccumulation profiles of PCB 180 in male and female wolves that are assumed to prey solely on Bathurst caribou. The predicted chemical concentrations in male and female wolves increases initially until the wolves are approximately 80 days old, at which time the pups are weaned. Following weaning, the chemical fugacity and hence chemical concentrations in wolves declines as a result of body growth and lipid production. At this life-stage, the flux of chemical transported to the animal via food ingestion is offset by the increasing chemical storage capacity within the animal. As growth and lipid production in wolves stabilize, further ingestion of chemical associated with food increases the chemical fugacity and hence concentrations in body tissues. For females, the chemical fugacity declines after the age of 2, the time when the first pregnancy and pup rearing occurs. Social hierarchies inherent to wolf packs can influence the age at which a female wolves will mate (Banfield, 1974). The dominant male and his mate are at the top of the pack hierarchy. Although juvenile female wolves become sexually mature at 2 years old, social dynamics within a wolf pack will determine when a female breeds. The chemical fugacity in male wolves exhibits a cyclical exponential increase. Fluctuation of chemical fugacities in male wolves are the result of seasonal changes in lipid contents, body size and chemical fugacities in prey.

Maternal transfer of contaminants during nursing

The logarithms of the model predicted fugacities (in units of Pa) of PCB 180 in a newborn caribou calf and a lactating female following a calving event are shown in Figure 4.7. The newborn calf and the female caribou at the time of birth exhibit equal fugacities (4.0×10^{-13} Pa). The newborns are suckling milk that is comprised of a high lipid fraction (>15%), at an ingestion rate (G_{MI}) of 1.2 L/day. The chemical fugacity in the milk is equal to the fugacity in the female during the time of lactation. The fugacity capacity of milk is related to its lipid content (ϕ_M), by $Z_M = K_{OW}Z_W\phi_M$. The post-natal female caribou is excreting milk at a rate of 1.2 L/day. During the nursing process, chemical losses *via* milk excretion can reduce the chemical fugacity in a lactating female. When the female stops lactating (after 130 days), the elimination of chemical *via* milk excretion ceases, causing the chemical fugacity in the female to increase. The fugacity in the female caribou after 365 days is equal to fugacity it exhibited at the time of birth (4.0×10^{-13} Pa).

The early exposure of contaminant to newborns through milk ingestion results in an initial increase in chemical fugacity in caribou calves. After approximately 125 days, the calves are weaned and begin to graze primarily on lichens. The chemical fugacity in milk (f_M) (i. e., newborn's diet) is 6 times greater than the fugacity in lichens (f_D) due to chemical biomagnification in the adult female caribou.

Model Predicted Fugacity of PCB180 in a Newborn Calf and Lactating Adult Female Caribou During Nursing

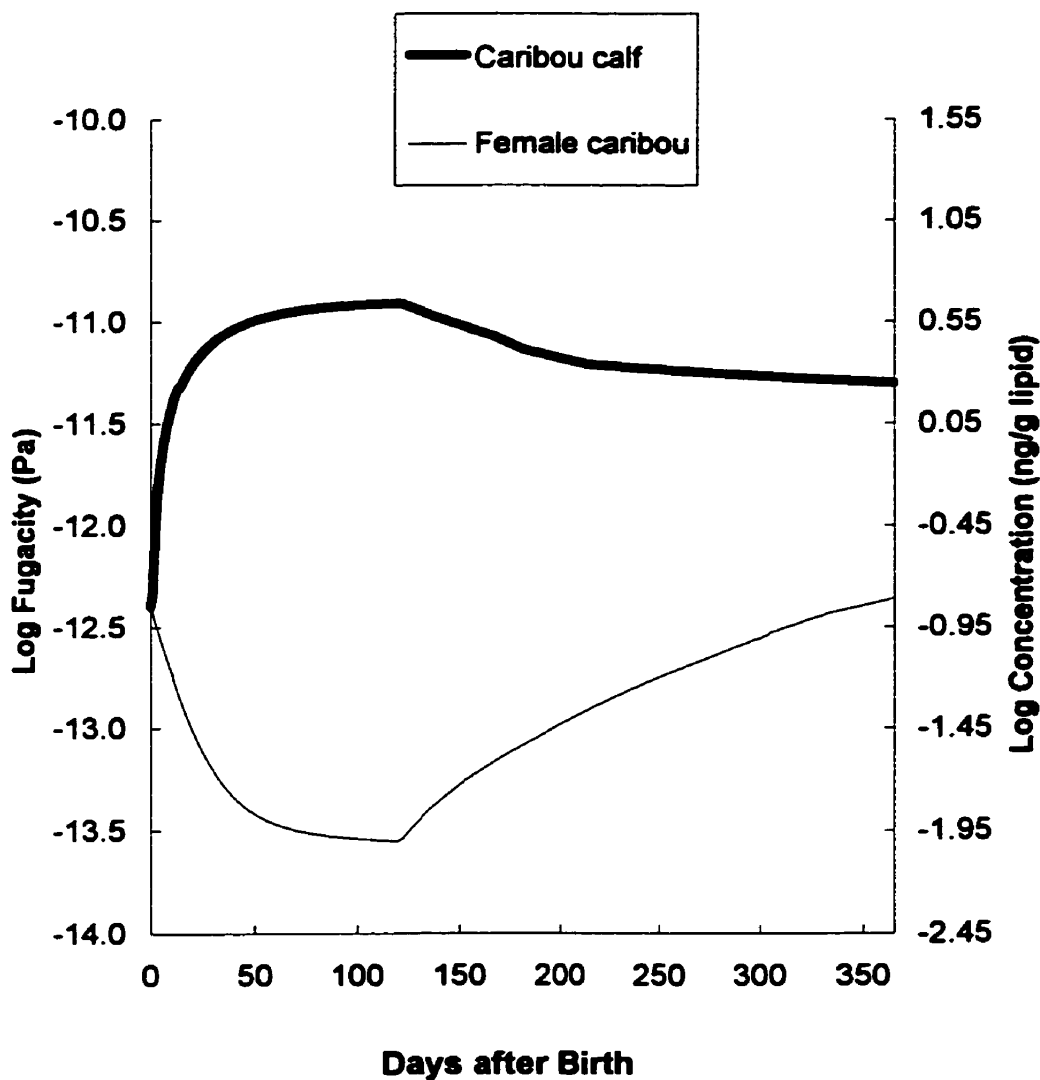


Figure 4.7: Logarithms of model predicted fugacities (Pa) and chemical concentration (ng/g lipid) in a lactating female caribou and nursing calf. Caribou calves are weaned a approximately 160 days after birth.

Another important factor affecting chemical bioaccumulation in nursing newborns is the extent of digestion and absorption of the high-lipid milk diet. If the newborn is able to efficiently absorb and digest lipids from the milk diet, a large GIMF is expected to occur, causing a larger degree of chemical uptake from the GIT to the calf. In this model, the fugacity capacity of the GIT content (Z_G) for a nursing newborn caribou calf is calculated by the equation $Z_G = (1 - \alpha) \cdot Z_M$, where α is the extraction efficiency of lipids in the milk and Z_M is the fugacity capacity of milk.

Digestion and absorption of milk during nursing can raise the chemical fugacity in the GIT of a caribou calf (f_G) above the fugacity in the ingested milk. The ratio of Z_M/Z_G for a nursing caribou calf is 10, while the Z_D/Z_G ratio for caribou grazing on lichens is 5. The larger diet/GIT ratio of fugacity capacities in nursing calves can cause increased gastro-intestinal magnification of chemical and hence increase biomagnification at this life-stage. During the dietary transition from milk to lichen, the calf continues to grow while ingesting less chemical associated with its food. The smaller Z_D/Z_G ratio exhibited in caribou grazing on lichens compared to calves ingesting milk results in a reduced GIT magnification of chemical and hence a reduced biomagnification in grazing calves. The chemical fugacity of PCB 180 achieved in a yearling 365 days after birth (4.9×10^{-12} Pa), is 12.5 times greater than the fugacity in the post-natal female (4.0×10^{-13} Pa).

The rate of growth and lipid content during the nursing stage may also influence chemical bioaccumulation in nursing newborns. As chemical associated with milk is ingested and absorbed, the calf continues to grow and produce body fat. A newborn that exhibits a fast growth rate or high lipid content compared to the rate of chemical flux into the animal (N_B , $\mu\text{mol}\cdot\text{d}^{-1}$) tends to “dilute” their internal chemical concentrations, resulting in lower chemical fugacities and concentrations than at birth. Conversely, animals that exhibit slow rates of growth and low lipid contents may “concentrate” chemical in their tissues, causing increased chemical fugacities and concentrations in the animal.

Sensitivity of model parameters

Results from the sensitivity analysis are shown in (Figure 4.8). The value of several model parameters were independently reduced by 10% to determine the change in PCB 180 concentrations (ng/g lipid) predicted for a newborn caribou calf, adult female caribou and adult male caribou relative to the baseline case. For a newborn caribou calf, internal chemical concentrations were sensitive to the parameters associated with digestion of milk. The rate of milk ingestion (G_M), concentration in milk (C_M), dietary uptake efficiency (E_D), lipid extraction efficiency (α) and the

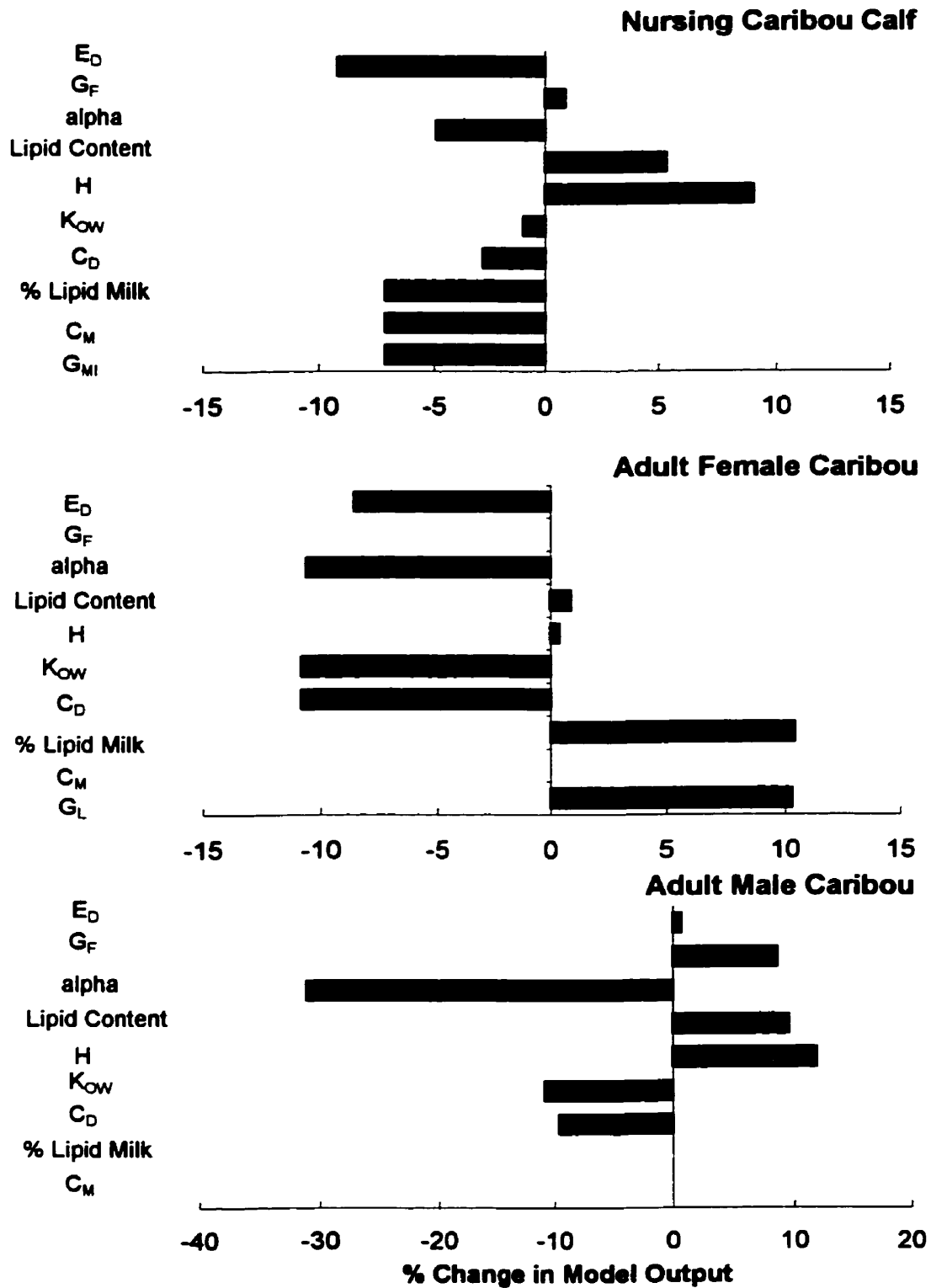


Figure 4.8: Results of sensitivity analyses on model input parameters, showing percent change (%) of model predicted chemical concentrations in a yearling, adult female and adult male caribou.

lipid content in milk each caused 5 to 10% reductions in predicted chemical concentrations in a nursing newborn calf. The chemical's Henry's Law constant was also shown to be an important input parameter, causing a 9% increase in the predicted concentration of PCB 180 in a newborn calf. For an adult female caribou, internal chemical concentrations were sensitive to changes in parameters associated with the digestion of lichens. In addition to E_D and α , the chemical concentration in lichens (C_D) cause a 10.8% reduction in the predicted chemical concentration in an adult female caribou. A 10% reduction in the lactation rate (G_L) and the % lipid in caribou milk each caused a 10.3% and 10.4% increase in chemical concentration predicted in an adult female. A 10% change in the octanol-water partition coefficient (K_{ow}) of the chemical caused a 10.8% reduction in the predicted chemical concentration for an adult female. C_D and α each caused a 9.6% and 31.1% reduction in the predicted chemical concentration in an adult male caribou.

Because model predicted chemical concentrations in caribou were sensitive to input parameters associated with dietary intake and digestion, the magnitude of gastro-intestinal magnification in caribou was further investigated. The gastro-intestinal magnification factor (GIMF) is associated with parameterization of food absorption and digestion and is expressed as the ratio $G_D Z_D / G_F Z_G$. Results from three simulations using GIMFs of PCB 180 in male caribou of 5, 15 and 50 were compared to the observed chemical concentration of PCB 180 in male caribou in July (1.65 ng/g lipid). Using a GIMF value of 15, the predicted chemical concentration of PCB 180 was 1.37 ng/g. A GIMF scenario of 50 resulted in an

internal chemical concentration of 4.5 ng/g lipid. A chemical concentration of 0.37 ng/g lipid was predicted with a GIMF value of 5. The intermediate GIMF value of 15 was shown to best represent the observed bioaccumulation in caribou. The GIMF of 15 in caribou is based on the assumption that food absorption (G_D/G_F) is approximately 3 and the extent of food digestion (Z_D/Z_G), based on a cellulose extraction efficiency of 80% ($\alpha = 0.8$), is approximately 5 (See Chapter 3). Based on previous research in ruminant dietary absorption and digestion physiology, a GIMF of 15 in barren-ground caribou is a realistic estimate.

The model calculates chemical loss *via* metabolic transformation (mol/day) by the expression, $k_M \cdot C_B \cdot V_B$, where the metabolic transformation rate (k_M , d^{-1}) is derived from the chemical's half-life ($T_{1/2}$) in tissue by the equation $k_M = 0.693/T_{1/2}$. Because the model incorporates metabolic transformation of chemical in the organism, the significance of this loss mechanism can be explored for chemicals that exhibit different half-lives. For example, the model predicted concentration of alpha-HCH in a 1-year-old male wolf at Bathurst Inlet, assuming no metabolic transformation, was 105.4 ng/g lipid. However, if metabolic transformation of alpha-HCH is considered by the input of a k_M value equal to $0.098 d^{-1}$, derived from a half-life of 7.14 days for alpha-HCH in rainbow trout (See Table 4.2), the model predicted chemical concentration of this chemical was 6.83 ng/g lipid. The observed chemical concentration in a 1-year-old male wolf sampled at Bathurst Inlet was 10.07 ng/g lipid (Appendix III). These results suggest that the extent of chemical loss *via*

metabolic transformation of chemical in the animal can be explained by the chemical's half-life in biota.

Food-Chain Bioaccumulation

Model predicted chemical concentrations of various organic contaminants in lichen-caribou-wolf food-chains at Bathurst Inlet, Cambridge Bay and Inuvik are consistent with the observed concentrations. Chemical concentrations of PCBs measured in caribou were lower than concentrations of HCB and alpha HCH. These concentration differences are revealed by clustering of these chemical compounds on the observed versus predicted plots (Figure 4.9).

Chemical concentrations of PCBs, chlorobenzenes and organochlorine pesticides in these plots are shown to be variable among wolves. The model over-estimated the chemical concentrations reported in male wolves from Cambridge Bay by a factor of 10.4. The lower chemical concentrations observed in Cambridge Bay male wolves may be the result of predation on prey species other than caribou within their geographic range. The Bathurst and Bluenose caribou herds are comprised of several thousands of animals, migrating bi-annually on their respective ranges. The frequency of caribou on Victoria Island is less than the frequency of caribou observed on the Bathurst or Bluenose ranges. It is then conceivable that resident

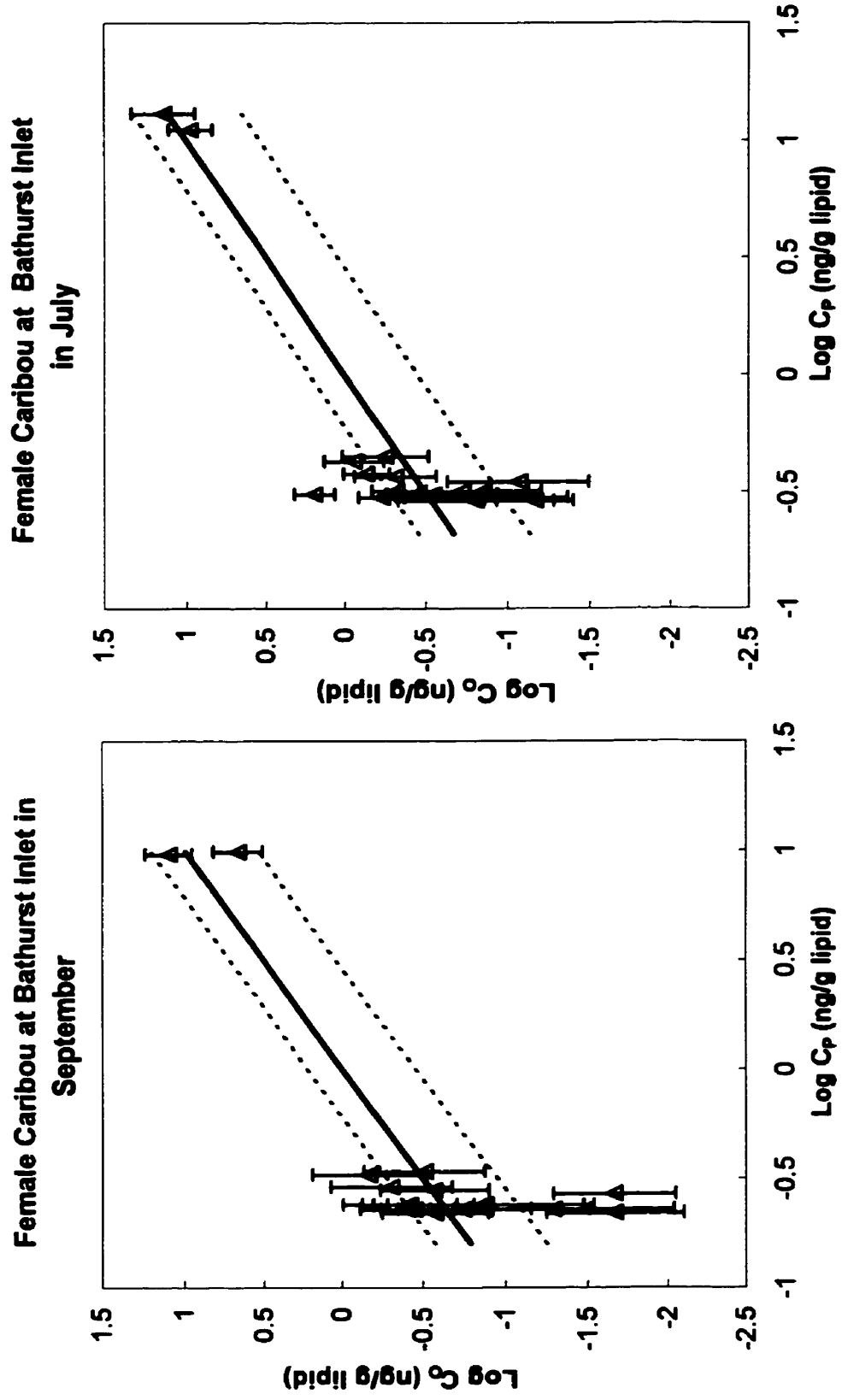


Figure 4.9: Observed versus predicted chemical concentrations for caribou and wolves from Bathurst Inlet, Cambridge Bay and Inuvik. Data points and error bars represent the logarithms of the geometric mean (GM) and standard deviations of the GM, respectively. Solid line represents a perfect model fit. Dashed lines indicate the inherent model confidence, represented as the standard deviations calculated by MCS using inherent model parameters (See Table 4.3).

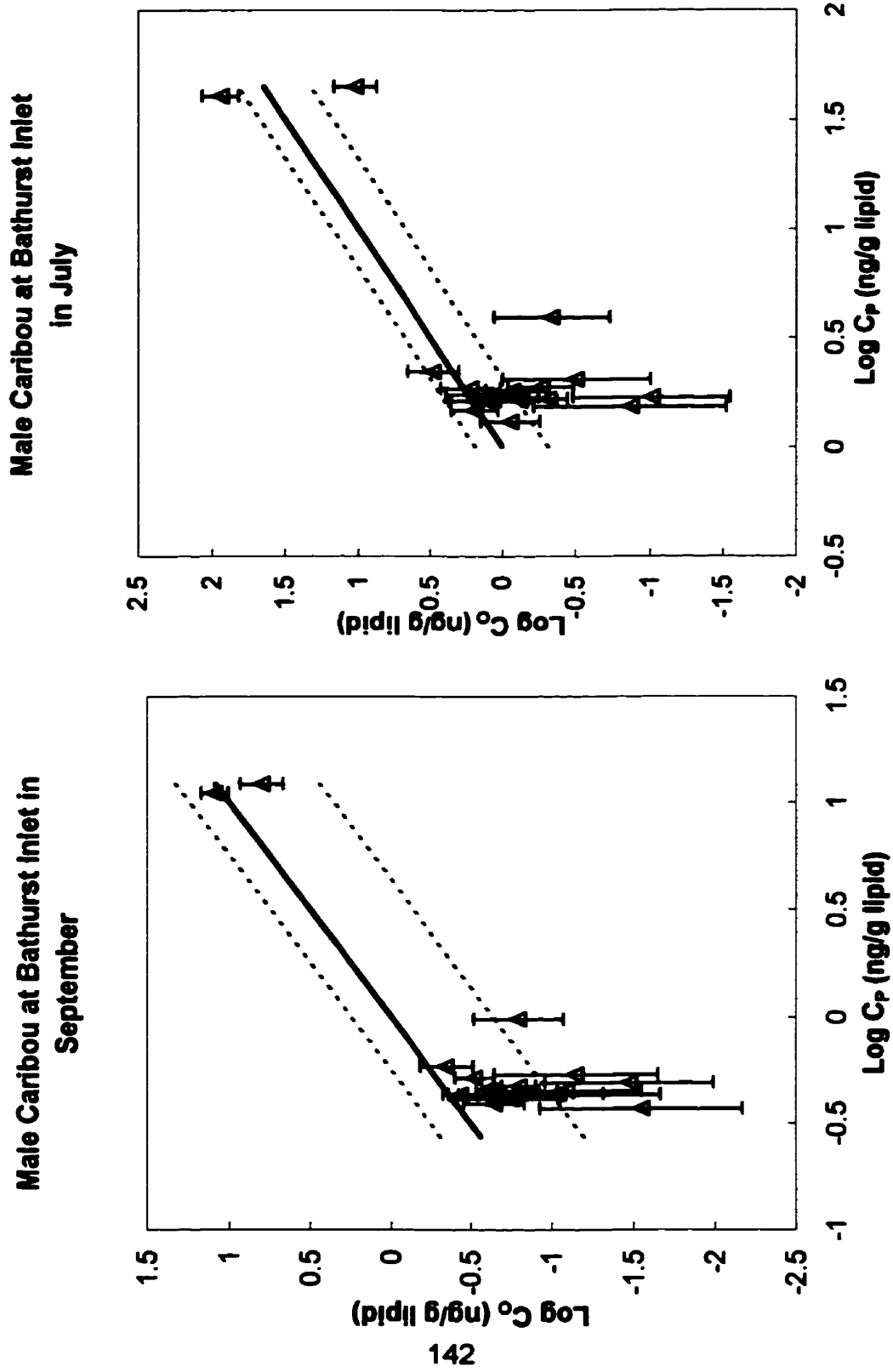


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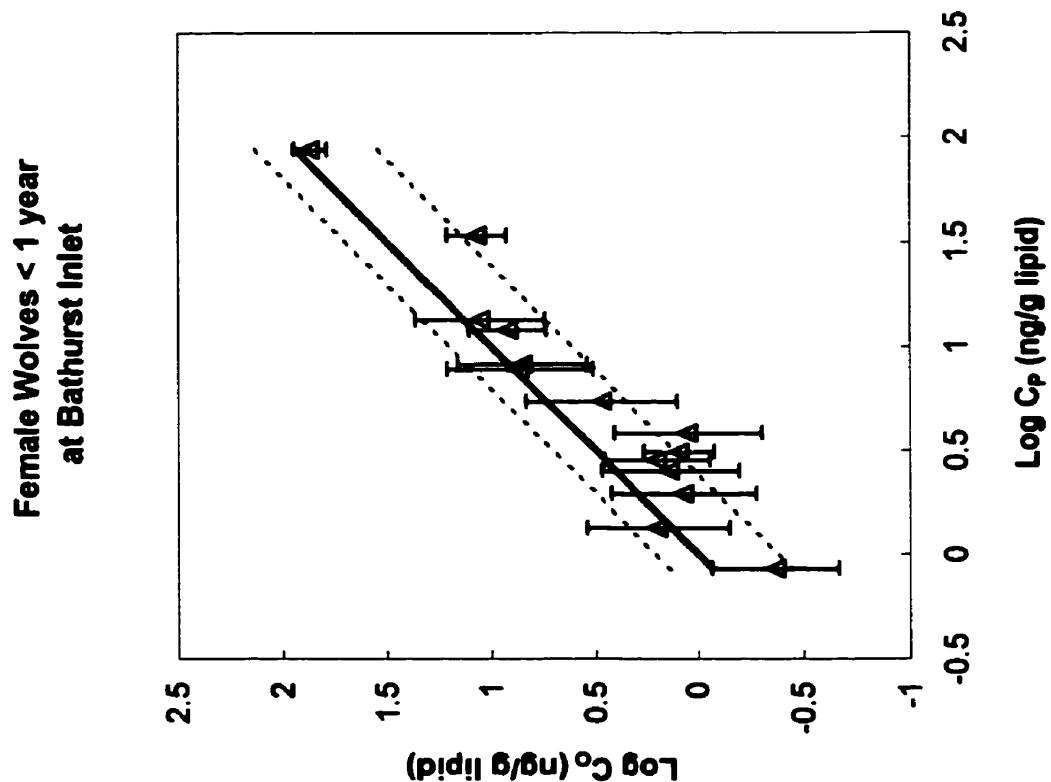
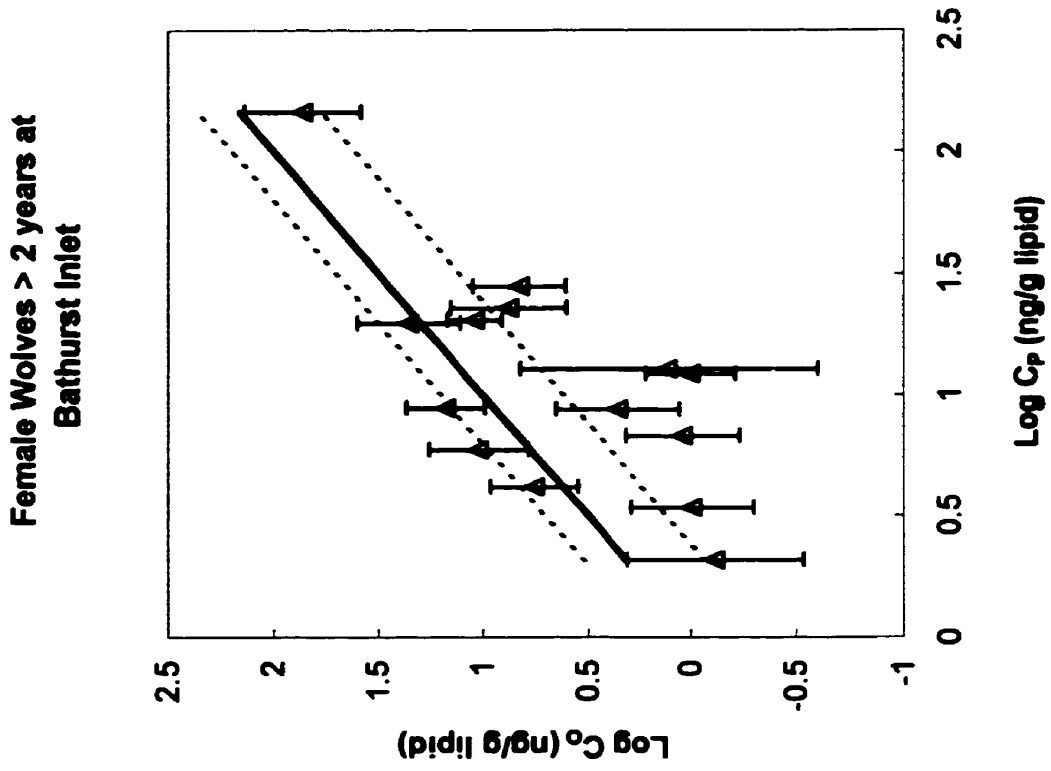


Figure 4.9 Continued

Male Wolves at Bathurst Inlet

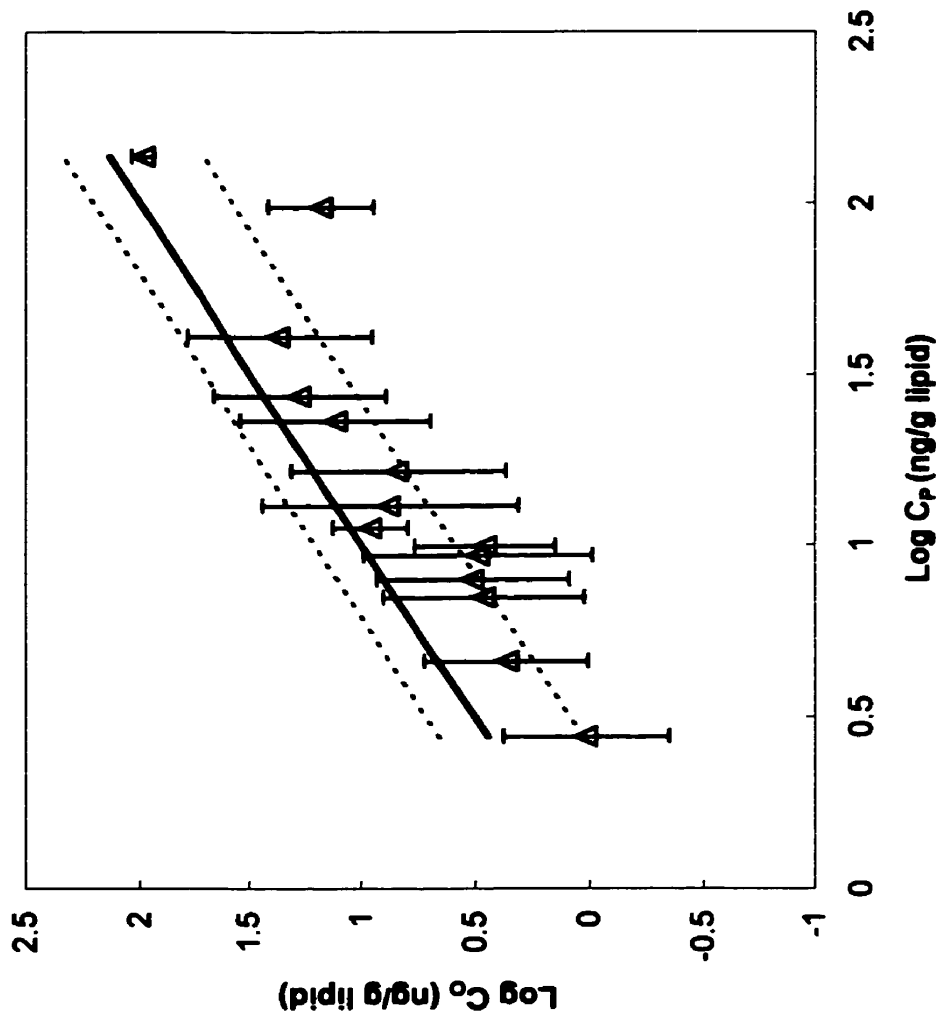
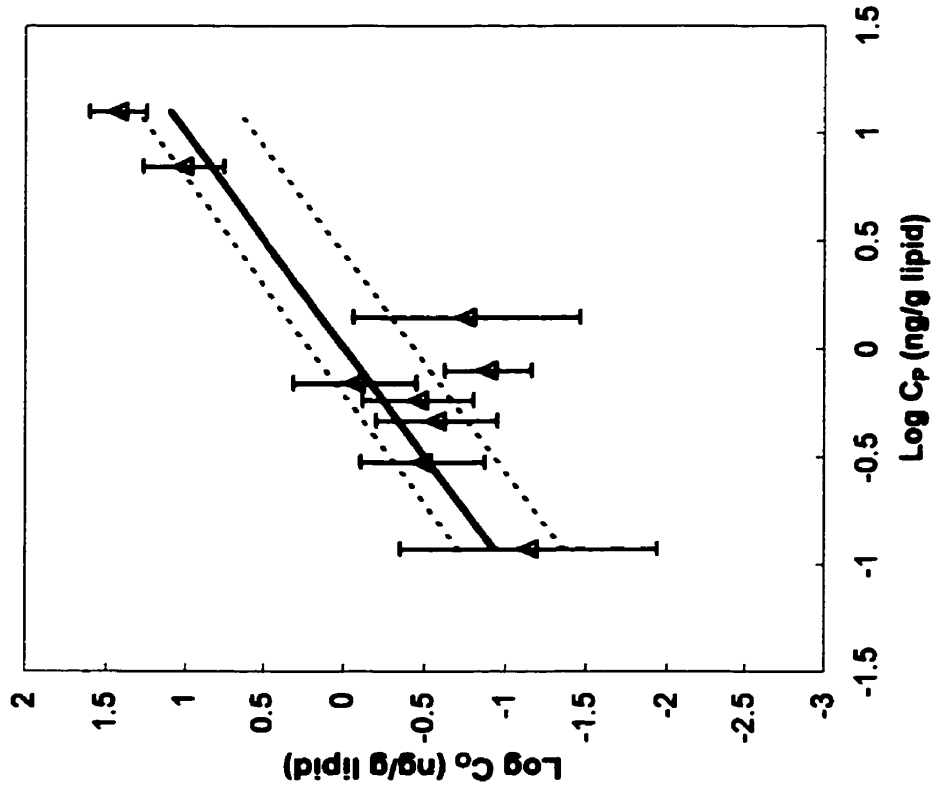
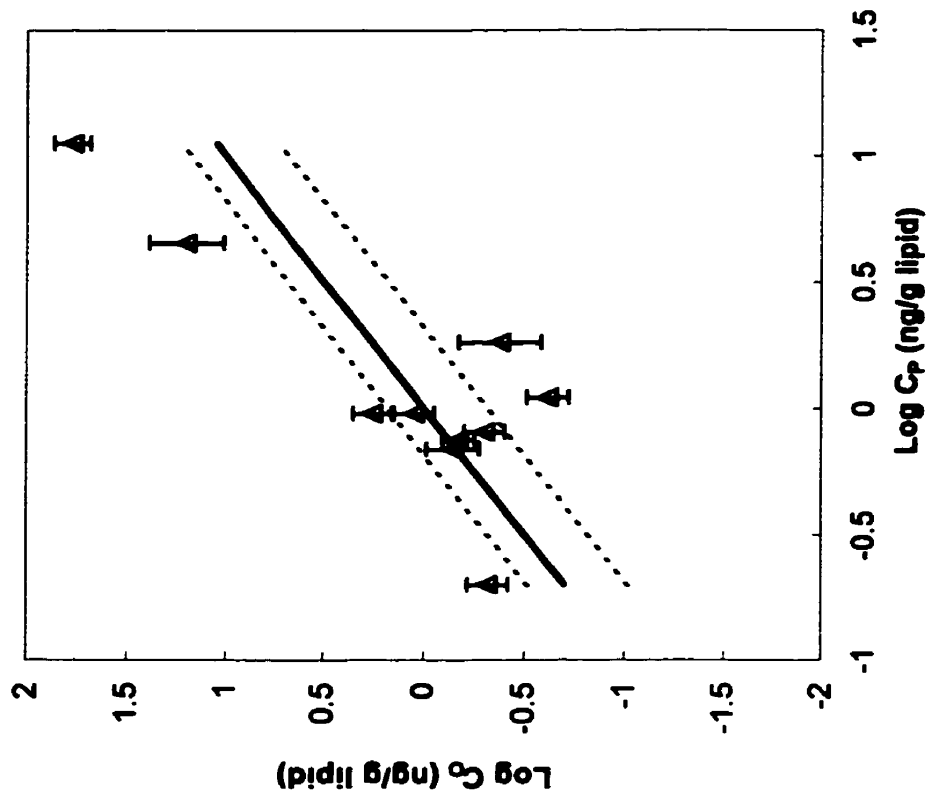


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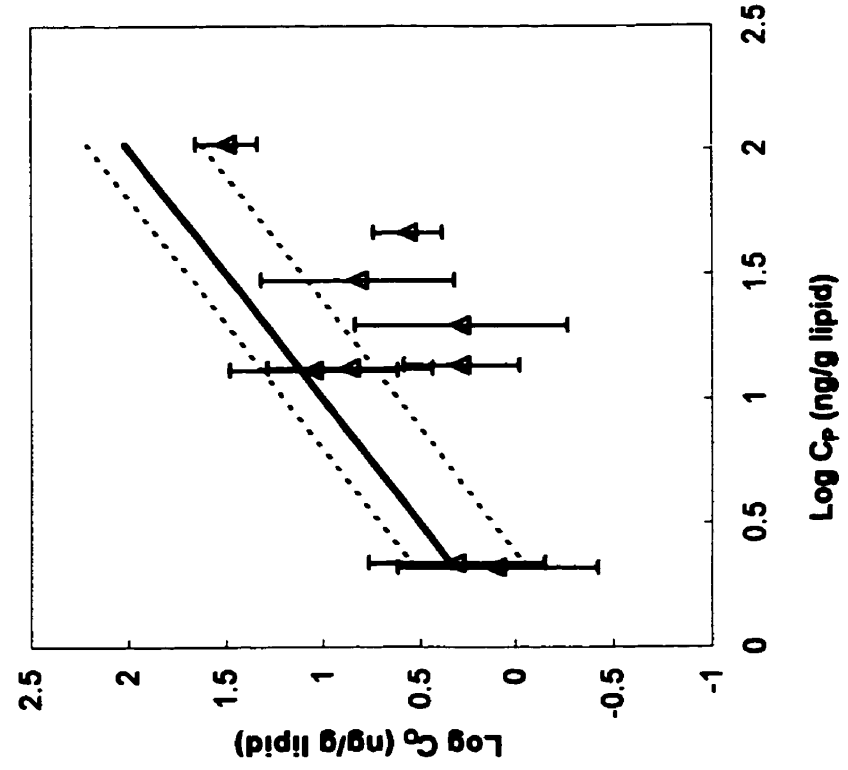
Female Caribou at Cambridge Bay



Male Caribou at Cambridge Bay



Female Wolves at Cambridge Bay



Male Wolves at Cambridge Bay

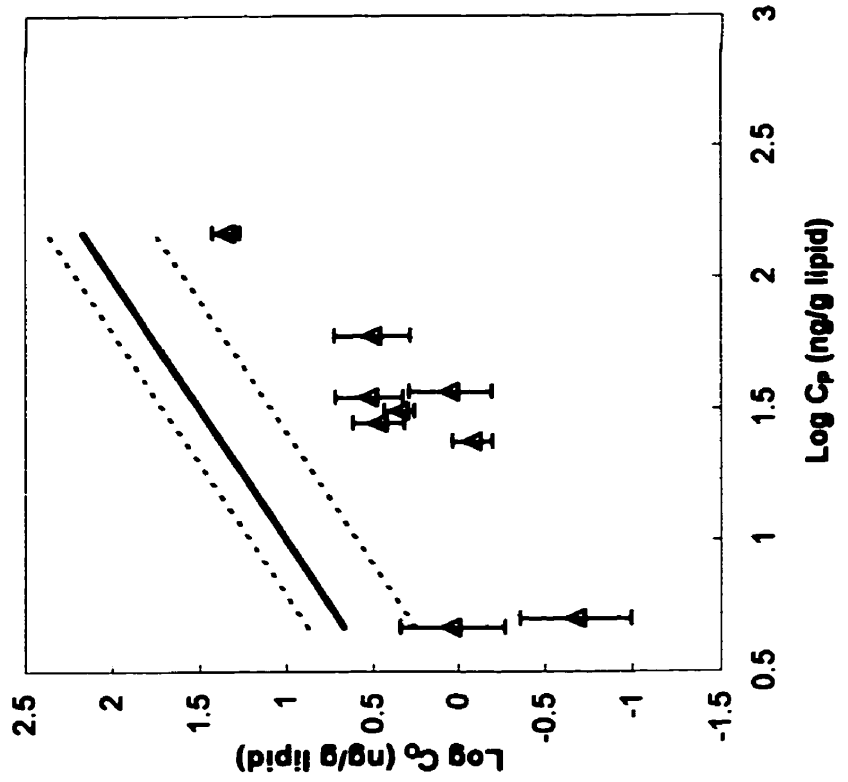
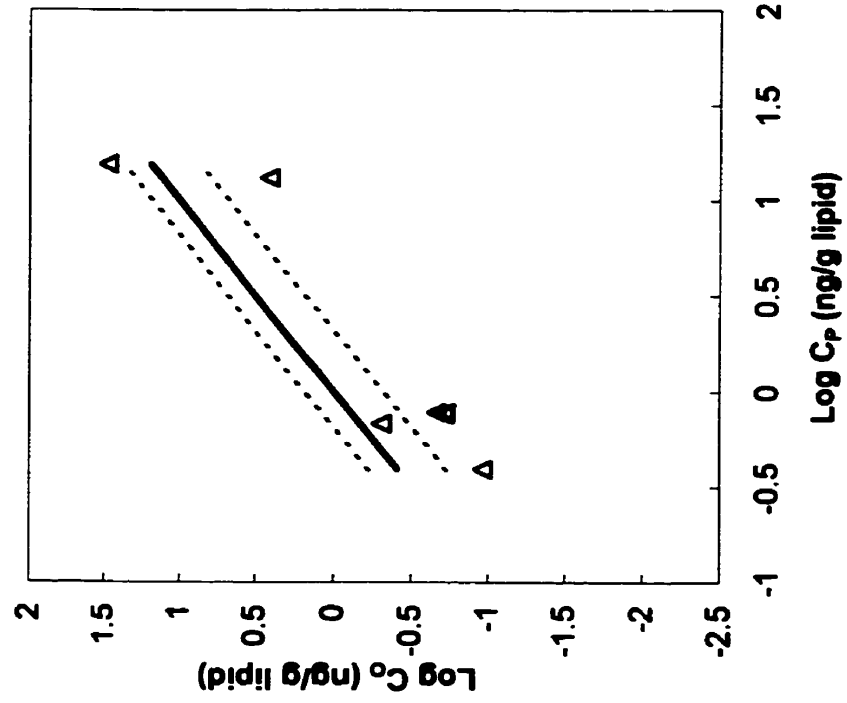


Figure 4.9 Continued

Female Caribou at Inuvik



Male Caribou at Inuvik

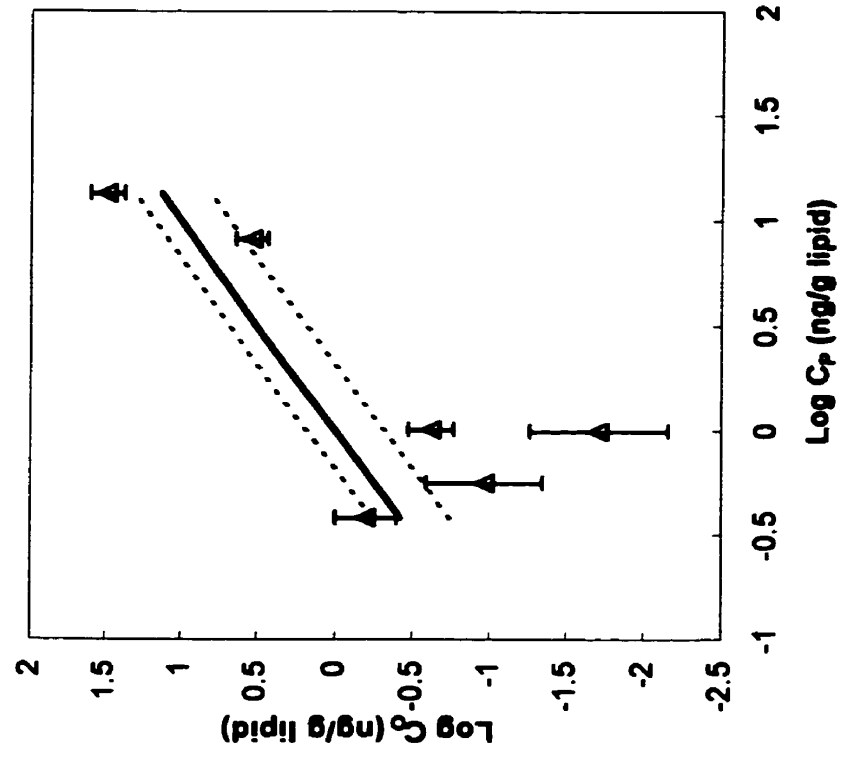


Figure 4.9 Continued

Male and Female Wolves at Inuvik

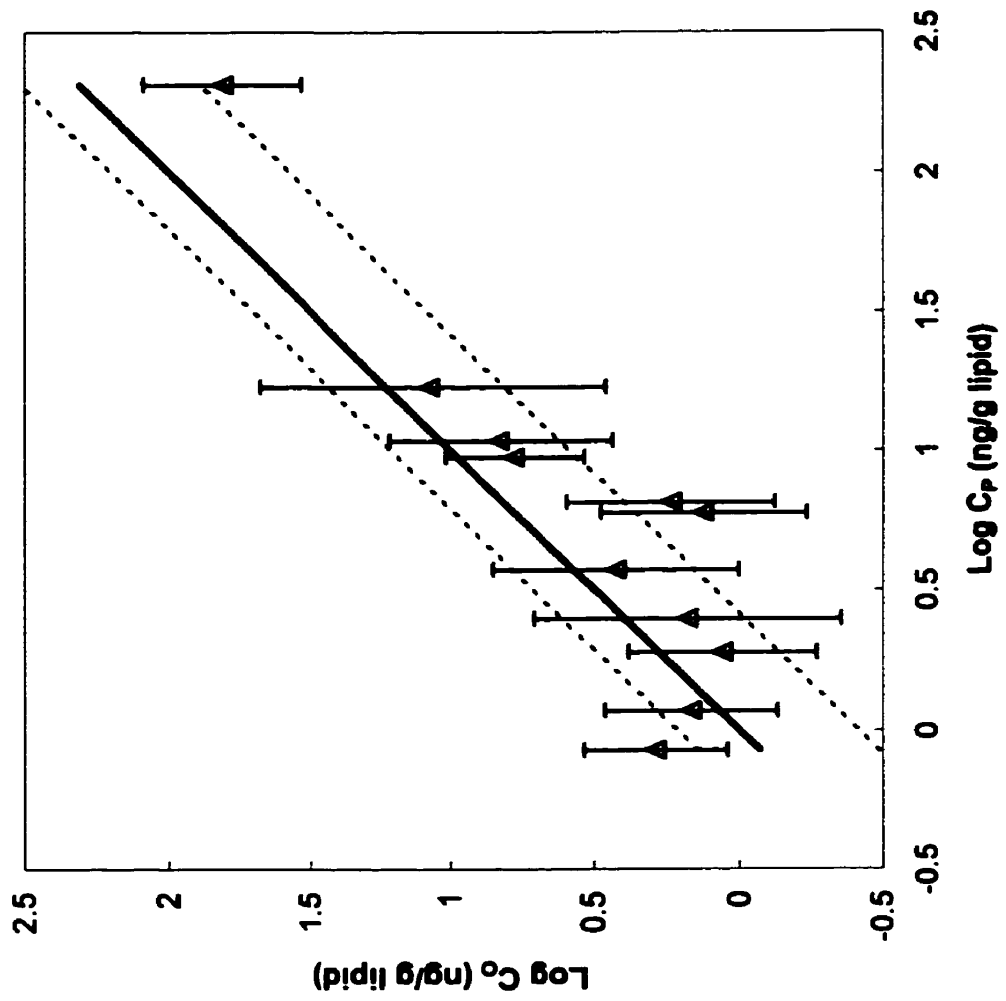


Figure 4.9 Continued

wolves on Victoria Island may rely on prey species other than caribou for sustenance. Wolves on barren-ground caribou ranges can prey on muskox, Arctic hare, Arctic ground-squirrels, geese and other species (Kuyt, 1972). These alternate prey may have substantially lower chemical concentrations than those exhibited by caribou on Victoria Island. If wolves on Victoria Island are feeding on prey species that exhibit lower chemical concentrations than caribou in that region, contrary to the assumption of a 100% caribou diet, observed chemical concentrations in these wolves would be less than expected.

The ages of wolves sampled at Cambridge Bay were not determined at the time of collection. In absence of this information, the model predictions for Cambridge Bay wolves were based on an age range of wolves that was observed at Bathurst Inlet (i. e., 6 months to 4 years). Specifically, an average of predicted chemical concentrations in wolves aged 6 months to 4 years was used to compare to the observed concentrations in wolves from Cambridge Bay. Male wolves sampled at Cambridge Bay may consist of young animals. Because, chemical concentrations in wolves are expected to increase with age, comparing predicted chemical concentrations in adult wolves with observed concentrations in animals less than 1 year may also be a source of error in this prediction.

Quantification of model bias and uncertainty for simulation of chemical bioaccumulation in the lichen-caribou-wolf food-chain at Bathurst Inlet was conducted by calculating the mean of $\log(C_F/C_O)$ and its 95% confidence

intervals, respectively (Table 4.4). Perfect model agreement is represented by a model bias value (i. e., $\text{antilog}\{\sum_{i=1}^n \log(C_{P,i}/C_{O,i})\}$) equal to 1. The model slightly under-predicted chemical concentrations in female caribou in July (model bias = 0.92) and over-predicted chemical concentrations in male caribou in September (model bias = 2.71). The simulations of chemical bioaccumulation in female caribou and female wolves exhibited the greatest uncertainty, represented by 95% confidence intervals that were 11.18 and 11.41 times the predicted values. Model predictions for Bathurst male caribou in September were 2.71 times greater than the observed concentrations. The over-estimation of chemical concentration in males during the fall may be the result of under-estimating the lipid content of these animals. In late summer adult male caribou begin to store fat deposits, mainly in the form of subcutaneous or "back" fat around the saddle and rump of the animal. Fat deposition continues into September and October until the rutting period is reached. The model assumes male caribou in September to have a whole-body lipid content of approximately 20%. Male caribou at Bathurst Inlet exhibiting whole-body lipid contents greater than 20% could explain the model's over-estimation of chemical concentrations in male caribou in the fall.

The model predicted chemical concentrations in adult female wolves was 1.94 times greater than the concentrations observed in adult female wolves at Bathurst Inlet. This over-estimation of chemical bioaccumulation in adult female wolves may be due to the presence of lactating and non-lactating females in the population. The model assigns an equal probability for pregnancy and lactation in a female every year

Table 4.4: Model bias, expressed as the mean $\log C_P/C_O$, and model uncertainty, as expressed by its 95% confidence interval.

SIMULATION	MODEL BIAS	UNCERTAINTY
	$\text{antilog}\{\sum_{i=1}^n \log(C_{P,i}/C_{O,i})\}$	$\text{antilog}\{95\% \text{ CI } \sum_{i=1}^n \log(C_{P,i}/C_{O,i})\}$
Male Caribou (July)	1.87	7.57
Male Caribou (September)	2.71	6.56
Female Caribou (July)	0.92	6.17
Female Caribou (September)	1.17	11.18
Male Wolf 6 months to 2 years of age	1.52	3.79
Female Wolf < 1 year of age	1.56	5.02
Female Wolf 2 years of age	1.94	11.41

following its respective age of maturity. This simplification of lactational excretion would over-estimate chemical loss *via* lactation in female animals that have not recently had reared offspring.

The fugacity-based biomagnification factors, BMFs (f_b/f_D), of PCB 180 predicted by the model are comparable to the observed BMFs in caribou and wolves at Bathurst Inlet (Table 4.5). For each model simulation, chemical fugacities (Pa) in lichens are calculated from the observed chemical concentration (C , mol/m³ dry tissue) and the fugacity capacity of lichen (Z_{LICHEN} , mol/m³·Pa): $f = C/Z$. Predicted chemical fugacities (Pa) in caribou and wolves were calculated during model simulations using Equation 4.6. The observed chemical fugacities (Pa) in caribou and wolves were calculated from the observed chemical concentrations (C , mol/m³ lipid) and the fugacity capacity of lipid (Z_L , mol/m³·Pa) using the equation $f = C \div Z$. Male caribou sampled in July exhibited BMFs that were 4 times greater than males of the same age sampled in September at Bathurst Inlet. The model seemed to over-predict slightly the BMF in male caribou in September and under-predict the BMF expected in females during summer and fall.

A food-chain multiplier for Bathurst wolves (i. e., $\text{BMF}_{\text{WOLF}} \times \text{BMF}_{\text{CARIBOU}}$) represents chemical bioaccumulation in wolves relative to lichens. The model predicted food-chain multiplier for PCB 180 in Bathurst wolves were comparable to the observed values (Table 4.5). The model predicted food-chain multiplier for male wolves in

Table 4.5: Observed and predicted concentrations, BMFs and food-chain multipliers of PCB 180 in caribou and wolves from Bathurst Inlet. Values in brackets represent the standard deviations of either the observed data or model predictions.

Species	Predicted Concentration	Observed Concentration	Predicted BMF (f_B/f_D)	Observed BMF (f_B/f_D)
Lichen (ng/g dry wt.) <i>C. rangiferina</i> , <i>C. nivalis</i>	-	0.04 (0.02)	-	-
Caribou (ng/g lipid)				
Male (Fall)	0.5 (0.3)	0.3 (0.03)	5.6	3.1
Male (Summer)	1.8 (1.3)	1.7 (0.8)	22.5	20.7
Female (Fall)	0.2 (0.2)	0.5 (0.4)	3.0	6.5
Female (Summer)	0.3 (0.29)	0.5 (0.1)	4.1	6.1
Wolf (ng/g lipid)				
Male (Fall)	40.8 (32.0)	34.4 (37.8)	49.1	41.7
Female (Fall)	16.7 (11.5)	19.0 (11.8)	20.5	23.3
Food-Chain-Multiplier ($BMF_{WOLF} \times BMF_{CARIBO}$)				
Male Wolf (Fall)	-	-	211.3	238.5
Male Wolf (Summer)	-	-	653.7	657.6
Female Wolf(Fall)	-	-	88.3	99.7
Female Wolf (Summer)	-	-	273.3	274.9

summer (653.7) and fall (211.3) were greater than values predicted in females during summer (273.3) and fall (88.3). The food-chain multiplier for wolves can be viewed as a fugacity-based biomagnification factor (BMF) that is calculated as the ratio of chemical fugacities in wolves to the fugacity in lichens (f_{WOLF}/f_{LICHEN}). Similarly, the fugacity-based BMF for caribou is calculated as the chemical fugacity in caribou divided by the chemical fugacity in lichens ($f_{CARIBOU}/f_{LICHEN}$). The logarithms of model predicted and observed fugacity-based BMFs of chemicals with log K_{OW} 's ranging from 4.0 to 8.0 in male caribou ($f_{CARIBOU}/f_{LICHEN}$) and male wolves (f_{WOLF}/f_{LICHEN}) from Bathurst Inlet are illustrated in Figure 4.10. The model predicted BMF for male wolves in the fall (194.9) is shown to be 9 times greater than the predicted fugacity-based BMF in male caribou during summer (22.5). The model predicted fugacity-based BMFs for caribou and wolves are based on the assumption that no metabolic transformation of chemical occurs in these animals. Chemicals that exhibit fugacity-based BMFs in caribou and wolves that exceed a value of 1 indicate chemical bioaccumulation (i. e., f_{WOLF} and $f_{CARIBOU} > f_{LICHEN}$), and hence can be considered bioaccumulative substances in this food-chain. The fugacity of beta-HCH observed in male caribou during summer and male wolves sampled in fall at Bathurst Inlet were respectively 14 times and 170 times greater than the fugacity of beta-HCH in lichens on the Bathurst range.

The bioaccumulation criteria outlined in the *TSMP and LRTAP POPs Protocol* identify chemical's having K_{OW} 's less than 10^5 or exhibit BCFs or BAFs in aquatic

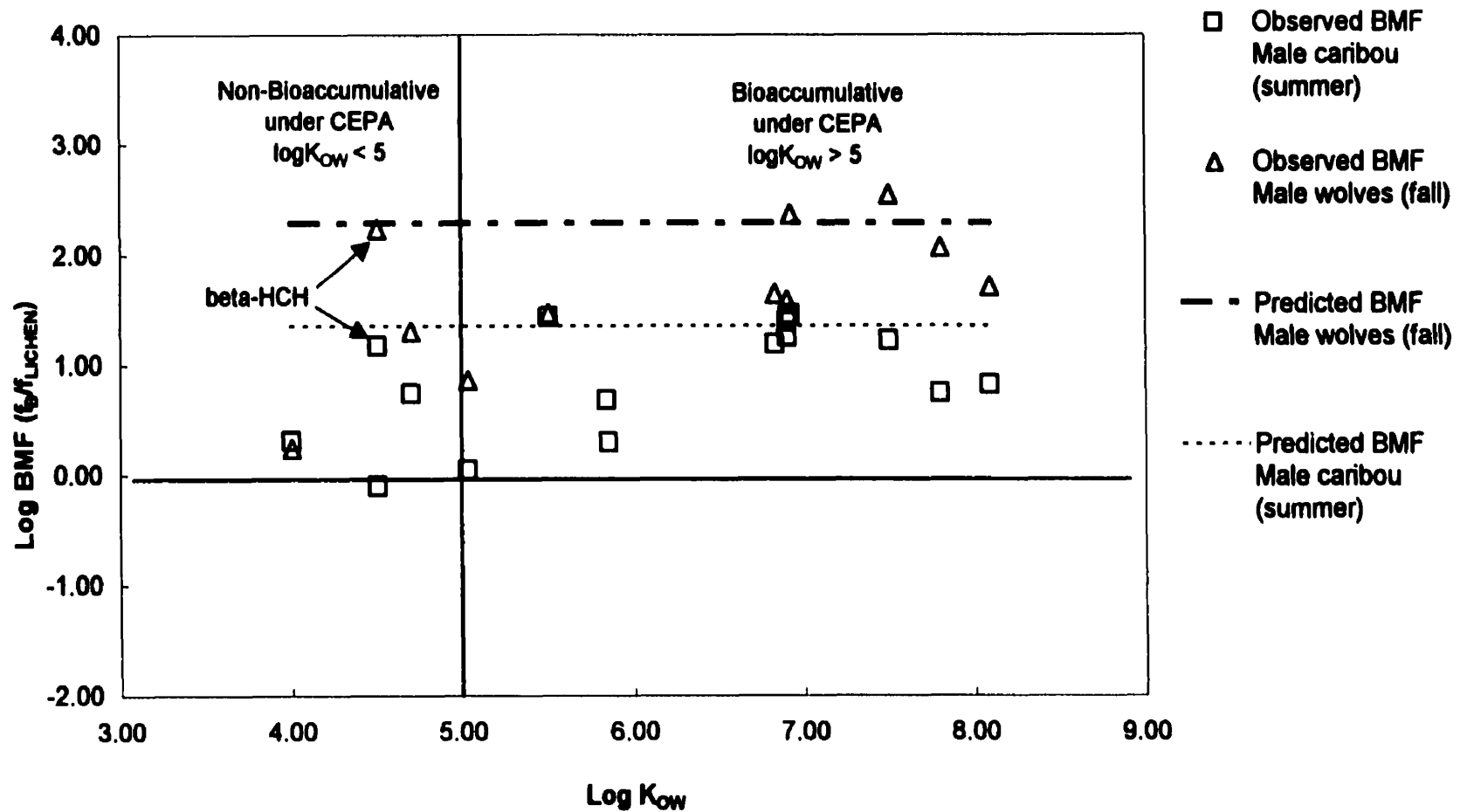


Figure 4.10: Logarithms of observed and predicted fugacity-based BMFs (f_B/f_{LICHEN}). Chemical magnification is defined as a situation where BMF is greater than 1.

organisms less than 5000 as non-bioaccumulative substances. This bioaccumulation criteria is not applicable to terrestrial ecosystems, due to the fact that chemical's having K_{ow} 's less than 10^5 are shown to bioaccumulate and biomagnify in this lichen-caribou-wolf food chain. It seems that caribou and wolves are unable to metabolize beta-HCH, resulting in biomagnification and food-chain bioaccumulation of this compound. The bioaccumulation criteria currently used for screening and management initiatives of POPs does not consider biomagnification of non-metabolizable chemicals that exhibit K_{ow} 's less than 10^5 . Thus, the usage and subsequent emission of non-metabolizable, low K_{ow} chemicals such as beta-HCH into the environment may result in food-chain bioaccumulation of these substances in terrestrial food-chains.

Bioaccumulation potential of a substance is related to its ability to accumulate and biomagnify in food-chains. Recent policy initiatives associated with Canada's *TSMP* and the *LRTAP POPs Protocol* assume a substance to be bioaccumulative if it exhibits a K_{ow} greater than 10^5 . Results from this study suggest that chemical bioaccumulation in food-chains is not solely dependent on a chemical's hydrophobicity. Many factors associated with an organism's taxonomic class, life-history, age, sex and physiology can play important roles in chemical bioaccumulation. Targeting chemicals that simply demonstrate a $K_{ow} > 10^5$, or are shown to bioaccumulate in aquatic organisms fails to address issues such as maternal-newborn chemical transfer, metabolism and metabolites, and depletion of fat reserves inherent to terrestrial mammals.

The pesticide Lindane (Hexachlorocyclohexane or HCH), is used extensively throughout equatorial regions around world. This extensive emission of HCH into the atmosphere at equatorial release locations has resulted in substantial chemical input to Arctic ecosystems. Chemical concentrations of alpha-HCH ($K_{ow} = 10^4$) in lichens collected along the migration route of the Bathurst caribou are approximately 3 mg/kg dry wt. Chemical concentrations of alpha-HCH in male caribou at Bathurst Inlet in September (6.58 mg/kg lipid) and July (11.12 mg/kg lipid) are comparable to chemical concentrations in male wolves (9.64 mg/kg lipid) sampled in fall near Bathurst Inlet. The concentration of beta-HCH ($K_{ow} = 10^{4.5}$) in male wolves (17.10 mg/kg lipid) is greater than beta-HCH concentrations observed in male caribou (1.47 mg/kg lipid) and lichens (0.08 mg/kg dry wt.) sampled at Bathurst Inlet. In Chapter 2, the chemical fugacity of beta-HCH in wolves sampled at Bathurst Inlet were significantly greater ($p < 0.05$) than the fugacity of beta-HCH in caribou and lichens collected on the Bathurst range, indicating the ability of this compound to biomagnify in the lichen-caribou-wolf food-chain. These results elude to preferential bioaccumulation of Lindane components in this food-chain, regardless of the chemical's low K_{ow} . Chemical concentrations of a substance in an organism's tissues are related to: (i) the physicochemical properties of the substance, (ii) the organism's physiological bioaccumulation capabilities, and (iii) the concentration of chemical in its diet (mg/kg), which is related the organism's dose in mg/kg/day. For extensively used chemicals such as Lindane, ambient environmental concentrations may attain high levels, resulting in an increased dose to terrestrial mammals. A

high dose of non-metabolizable chemicals exhibiting K_{ow} 's less than 10^5 may result in substantial accumulation in terrestrial mammals. In this scenario, the current bioaccumulation criteria which targets substances having a $K_{ow} > 10^5$ is not applicable to terrestrial food-chains. The goal of initiating political action for chemical management should be to target chemicals that exhibit the ability to accumulate in the environment, so as to fully protect all organisms at all life-stages from potential toxic effects. Current management policies associated with the management of environmental contaminants may not accomplish this goal for terrestrial mammals.

In many regions of the Canadian Arctic, lichen-caribou-wolf food-chains represent a linear food-web structure, where caribou forage primarily on lichens and wolves rely entirely on caribou for food. Analyses of chemical dynamics within these food-chains of Canada's central and western Arctic were conducted to investigate mechanistic explanations of the observed chemical bioaccumulation patterns. The fugacity-based bioaccumulation model outlined in this chapter provides a mechanistic explanation for observed bioaccumulation patterns in lichen-caribou-wolf food-chains in the Canadian Arctic. The model predictions of internal chemical concentrations in caribou and wolves in this study were comparable to observed chemical concentrations in caribou and wolves sampled at Bathurst Inlet, Cambridge Bay and Inuvik. The bioaccumulation model presented in this chapter simulates chemical bioaccumulation of organic chemicals in terrestrial food-chains based on the inputs of chemical properties (e. g., K_{ow}), various ecological parameters and the ambient

environmental chemical concentrations (i. e., chemical concentrations in vegetation).

Therefore, this model can be applied to other terrestrial ecosystems to investigate bioaccumulation mechanisms and bioaccumulation potential of various organic chemicals. Further analysis of bioaccumulation mechanisms and their effects on food-chain bioaccumulation for both new and existing organic chemicals should accompany current and future policy action regarding the usage, manufacturing and disposal of organic chemicals.

CHAPTER 5

Summary and Conclusions

In May of 1997 and July of 1998, vegetation and caribou fecal pellets were collected from various locations on the range of the Bathurst Caribou herd and analyzed for organochlorine pesticide and polychlorinated biphenyl (PCB) concentrations. Alpha-hexachlorocyclohexane (α -HCH), hexachlorobenzene (HCB), and PCB congeners 101, 118, 149, 153, 138, and 180 were detected in all samples. Lichens had higher concentrations of pesticides and PCBs than tundra willow (*Salix glauca*). The chemical fugacity of PCBs, DDTs, chlordanes, HCH (Lindane) and HCB in lichens were similar to the chemical fugacities observed in Arctic air. These results suggest that a chemical equilibrium between lichens and air exists during times of atmospheric exposure.

Chemical concentrations of less volatile hydrophobic compounds in lichens were significantly greater in samples collected during spring snowmelt. Increased chemical concentrations in lichens during spring snowmelt suggest chemical bioconcentration from contaminants stored in the previous winter's snowpack. The higher chemical concentrations found in lichens during spring may represent internal bioconcentration or exterior accumulation via surficial deposits of chemical associated with particulate matter. These results further demonstrate the importance

of snowfall and snowpack dynamics on bioconcentration of organic chemicals in tundra lichens.

The biomagnification model presented in this study depends on the characterization of food absorption (G_D/G_F) and food digestion (Z_D/Z_G). The extent of dietary bioaccumulation of organic chemicals depends on the relationship between the dietary intake rate G_D , the fecal excretion rate G_F , as well as the fugacity capacities of the ingested food (Z_D) and the fecal matter (Z_F). Dietary intake and absorption data are readily available from the literature for many mammals. However, documentation of fugacity capacities of various media are not well documented. A dynamic head-space methodology using chlorobenzenes and polychlorinated biphenyls (PCBs), was then developed to characterize the fugacity capacities of food and fecal samples of barren-ground caribou. Experimental derivation of Z_D and Z_G values in lichens and caribou fecal pellets from head-space analyses were shown to be inaccurate. Estimated values of Z_D and Z_G were used for further calculation of chemical fugacities in food and GIT contents of barren-ground caribou. If the extent of food digestion (Z_D/Z_G) was estimated to be a factor of 5, and food absorption (G_D/G_F) a factor of 3, the chemical fugacities in caribou GIT were shown to be elevated above lichens for various organic chemicals. Based on food digestion and food absorption estimates, the gastro-intestinal magnification factor (GIMF) in caribou was approximately 15.

In Chapter 4, a mechanistic model was presented to assess the exposure and bioaccumulation of organic contaminants in terrestrial mammals. This time-dependent model simulates the exposure of chemical to newborns through milk ingestion during their nursing life-stage. The model also accounts for temporal and seasonal variation of parameters such as feeding rates, dietary preference, fat content, lactation and body mass. The model was applied to an Arctic terrestrial ecosystem to predict internal concentrations of organic chemicals in barren ground caribou (*Rangifer tarandus*), and wolves (*Canis lupus*) from a range of observed concentrations in two common lichen species (*Cladina rangiferina* and *Cetraria nivalis*).

Sensitivity analyses on model parameters suggest the importance for accurate representation of contaminant levels in the an organism's diet, chemical partitioning properties (K_{ow} , Henry's Law constants), and the extent of gastro-intestinal magnification of chemical based on GIMF values in caribou and wolves. These investigations have further shown the complexities surrounding chemical dynamics within ecological food-chains and indicates the need for further research into the driving mechanisms effecting food-chain bioaccumulation. The results from this study provide a mechanistic explanation for chemical bioaccumulation patterns of organic contaminants in lichen-caribou-wolf food-chains across the central and western Arctic of Canada.

Chemical bioaccumulation in organisms is a complex process, involving many independent factors. The current approach used to screen new and existing chemicals for bioaccumulation potential involve an assessment of the chemical's hydrophobicity (K_{ow}) and observed bioaccumulation in laboratory experiments. This approach may not accurately quantify chemical bioaccumulation in ecological food-chains. An alternative approach may be to utilize verifiable models that represent the ecological and physiological mechanisms associated with chemical bioaccumulation. Application of mechanistic models that incorporate ecosystem and species specific parameters may better assess the potential of a substance to bioaccumulate in ecological food-chains. Consequently, these models may aid policy-makers in the derivation of chemical screening and assessment strategies.

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APPENDIX I

Chemical concentrations (ng/g lipid) of organic contaminants in the liver, muscle and fat tissues of caribou and wolves from Cambridge Bay, Bathurst Inlet, and Inuvik.

Appendix I: Chemical concentrations (ng/g lipid) of organic contaminants in liver, muscle and fat tissues of caribou and wolves sampled at Cambridge Bay, Bathurst Inlet and Inuvik. Logarithms of the octanol-water partition coefficient ($\log K_{ow}$) of the chemical is also shown.

Cambridge Bay Caribou

CHEMICAL	logK _{ow}	Female Caribou n=5						Male caribou n=5					
		Liver		Muscle		Fat		Liver		Muscle		Fat	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1,2,4,5 TCB	4.70	< 0.23	-	< 0.62	-	0.43	0.19	< 0.23	-	< 0.52	-	0.73	0.21
1,2,3,4TCB	4.46	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
QCB	5.03	0.68	1.11	< 0.62	-	0.66	0.22	2.18	0.48	< 0.52	-	0.98	0.20
alpha-HCH	4.00	37.30	2.01	19.56	3.07	12.54	6.27	45.77	6.78	21.20	2.53	17.01	7.59
beta-HCH	4.00	8.57	2.09	10.69	5.86	0.59	0.15	7.83	2.15	8.75	3.24	0.83	0.20
gamma-HCH	4.50	0.23	0.02	3.93	8.23	0.47	0.16	< 0.23	-	-	0.09	0.55	0.16
HCB	5.50	36.77	5.90	34.22	7.00	28.53	9.65	66.46	15.32	54.80	12.89	60.43	12.82
OCS	6.90	3.02	1.51	0.62	0.16	0.35	0.25	8.15	3.13	< 0.52	-	0.76	0.23
Oxychloridane	6.90	55.56	15.98	2.12	3.58	0.22	0.11	112.44	75.68	< 0.52	-	0.43	0.22
Tranachlordane	6.90	< 0.23	-	< 0.62	-	< 0.01	-	0.91	1.34	< 0.52	-	< 0.01	-
Ciechloridane	6.90	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
Transnonachlor	6.90	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	0.13	0.07
Cianonachlor	6.90	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
P,p' DDE	6.90	< 0.23	-	< 0.62	-	< 0.01	-	1.19	1.37	< 0.52	-	0.53	0.46
P,p' DDD	6.90	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
P,p' DDT	6.00	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
Photomirex	6.00	3.25	1.03	< 0.62	-	0.05	0.05	4.78	1.01	< 0.52	-	0.12	0.05
Mirex	6.00	4.43	1.47	< 0.62	-	0.07	0.07	6.26	1.28	< 0.52	-	0.16	0.13
Heptachlor epoxide	6.00	12.06	3.51	< 0.62	-	0.28	0.17	14.34	2.39	< 0.52	-	0.46	0.19
Dieldrin	6.20	4.30	5.42	< 0.62	-	< 0.01	-	0.63	0.79	< 0.52	-	< 0.01	-
PCB31	5.60	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB28	5.60	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB52	5.84	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	0.17	0.05
PCB49	5.85	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB44	5.75	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB42	5.76	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB64	5.95	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB74	6.20	< 0.23	-	< 0.62	-	0.12	0.10	< 0.23	-	< 0.52	-	0.31	0.05
PCB70	6.20	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	< 0.01	-
PCB66/95	6.20	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	0.25	0.06
PCB60	6.11	< 0.23	-	< 0.62	-	< 0.01	-	< 0.23	-	< 0.52	-	0.16	0.13

Appendix I continued
Cambridge Bay Wolves

CHEMICAL	Female Wolf n=5						Male Wolf n=5						
	Liver		Muscle		Fat		Liver		Muscle		Fat		
logK _{ow}	mean	SD	Mean	SD	Mean	SD	mean	SD	Mean	SD	Mean	SD	
1,2,4,5 TCB	4.70	3.42	3.93	1.42	2.24	2.22	1.54	< 0.23	-	< 0.32	-	0.87	0.36
1,2,3,4TCB	4.46	< 0.17	-	< 0.28	-	0.02	0.02	< 0.23	-	< 0.32	-	0.05	0.06
QCB	5.03	4.68	3.91	3.60	2.89	2.27	1.38	0.75	1.28	0.76	0.65	0.85	0.23
alpha-HCH	4.00	6.43	4.37	6.07	2.86	3.87	1.55	4.93	2.94	4.52	1.34	3.08	1.05
beta-HCH	4.00	14.32	9.73	13.97	9.12	9.94	8.10	3.41	2.53	4.53	0.92	2.27	0.47
gamma-HCH	4.50	< 0.17	-	< 0.28	-	< 0.01	-	0.23	0.01	< 0.32	-	0.06	0.08
HCB	5.50	73.84	51.72	54.94	34.47	33.27	13.21	30.55	16.62	33.92	3.11	22.65	3.83
OCS	6.90	27.69	9.87	5.30	3.09	1.71	0.73	8.58	4.23	1.45	0.60	0.69	0.40
Oxychlorane	6.90	483.21	576.14	67.89	55.41	21.54	24.92	69.45	38.08	13.18	3.99	2.79	2.18
Transchlorane	6.90	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
Cischlorane	6.90	< 0.17	-	< 0.28	-	0.04	0.06	< 0.23	-	< 0.32	-	< 0.01	-
Transnonachlor	6.90	7.59	14.84	3.64	6.86	1.98	3.54	0.48	0.61	0.61	0.72	0.41	0.50
Cisnonachlor	6.90	< 0.17	-	< 0.28	-	0.17	0.29	< 0.23	-	< 0.32	-	0.04	0.04
p,p' DDE	6.90	< 0.17	-	< 0.28	-	0.81	1.61	< 0.23	-	< 0.32	-	0.14	0.22
p,p' DDD	6.90	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
p,p' DDT	6.00	< 0.17	-	< 0.28	-	< 0.01	-	5.45	8.15	< 0.32	-	< 0.01	-
Photomirex	6.00	10.88	10.66	2.72	2.25	1.20	1.01	2.30	1.27	< 0.32	-	0.34	0.27
Mirex	6.00	14.75	15.35	2.36	1.77	0.71	0.42	4.72	3.51	< 0.32	-	0.31	0.32
Heptachlor epoxide	6.00	57.93	91.01	5.67	7.64	3.23	3.84	21.86	19.00	2.52	1.95	1.42	1.39
Dieldrin	6.20	77.67	122.62	4.26	8.11	3.91	5.70	36.21	37.80	2.76	3.02	2.40	2.97
PCB31	5.60	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB28	5.60	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB52	5.84	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB49	5.85	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB44	5.75	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB42	5.76	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB64	5.95	1.04	1.73	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB74	6.20	1.91	3.48	< 0.28	-	1.27	1.93	< 0.23	-	< 0.32	-	0.23	0.10
PCB70	6.20	< 0.17	-	< 0.28	-	0.08	0.13	< 0.23	-	< 0.32	-	< 0.01	-
PCB66/95	6.20	< 0.17	-	< 0.28	-	0.26	0.49	< 0.23	-	< 0.32	-	< 0.01	-
PCB60	6.11	< 0.17	-	< 0.28	-	1.20	2.37	< 0.23	-	< 0.32	-	< 0.01	-

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Appendix I continued

Cambridge Bay Wolves

CHEMICAL	logKow	Liver		Muscle		Fat		Liver		Muscle		Fat	
		mean	SD	Mean	SD	Mean	SD	mean	SD	Mean	SD	Mean	SD
PCB101	6.38	< 0.17	-	< 0.28	-	0.08	0.13	0.23	0.01	< 0.32	-	< 0.01	-
PCB99	6.39	91.21	143.94	19.27	26.09	6.34	9.20	11.85	7.20	3.70	2.20	1.20	0.89
PCB97	6.29	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB87	6.29	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB110	6.48	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB151	6.64	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB149	6.67	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	0.02	0.03
PCB118	6.74	11.08	20.80	3.39	6.36	3.86	6.52	< 0.23	-	< 0.32	-	0.64	0.39
PCB146	6.89	1.00	1.66	< 0.28	-	0.38	0.69	< 0.23	-	< 0.32	-	0.10	0.09
PCB153	6.92	49.10	72.55	20.73	21.21	10.94	12.94	5.94	3.55	6.45	2.02	3.61	2.17
PCB105	6.65	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB141	6.82	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB138	6.83	13.85	23.27	5.51	7.10	3.78	5.30	0.95	1.76	0.57	0.60	1.30	0.88
PCB128	6.73	< 0.17	-	< 0.28	-	0.08	0.12	< 0.23	-	< 0.32	-	0.02	0.02
PCB182/187	7.20	< 0.17	-	< 0.28	-	0.16	0.31	< 0.23	-	< 0.32	-	0.03	0.04
PCB183	7.00	5.01	9.69	< 0.28	-	0.60	0.68	< 0.23	-	< 0.32	-	0.18	0.15
PCB185	7.11	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB174	7.11	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB171	7.11	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB200	7.20	< 0.17	-	< 0.28	-	0.21	0.39	< 0.23	-	< 0.32	-	0.03	0.05
PCB172	7.33	< 0.17	-	< 0.28	-	0.08	0.12	< 0.23	-	< 0.32	-	< 0.01	-
PCB180	7.50	92.98	123.77	16.61	13.75	15.04	11.15	7.33	4.75	5.10	1.88	3.66	1.90
PCB170/190	7.46	32.99	59.04	9.98	9.46	9.89	7.91	2.09	2.19	1.36	1.68	2.30	1.05
PCB201	7.62	0.77	1.20	< 0.28	-	0.14	0.19	< 0.23	-	< 0.32	-	0.02	0.03
PCB203	7.65	1.04	1.73	< 0.28	-	0.08	0.11	< 0.23	-	< 0.32	-	0.04	0.06
PCB195	7.56	0.51	0.67	< 0.28	-	0.12	0.09	< 0.23	-	< 0.32	-	0.03	0.04
PCB194	7.80	18.11	23.98	11.56	10.11	4.52	2.80	< 0.23	-	0.61	0.72	0.84	0.47
PCB206	8.09	18.04	25.80	6.48	4.67	1.82	1.34	< 0.23	-	< 0.32	-	0.27	0.23
PCB189	7.71	0.81	1.28	< 0.28	-	0.44	0.30	< 0.23	-	< 0.32	-	0.05	0.06
PCB77	6.36	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB126	6.89	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
PCB169	7.42	< 0.17	-	< 0.28	-	< 0.01	-	< 0.23	-	< 0.32	-	< 0.01	-
Total PCBs	6.92	339.45	494.59	93.53	98.75	61.38	65.22	28.39	19.47	17.79	9.11	14.58	8.65
Arochlor1254:1260	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Appendix I continued
 Bathurst Female Caribou

CHEMICAL	logKow	Female Caribou(September) Fat Mobilization n=5						Female Caribou (July - Depleted Lipid Reserves) n=4					
		Liver		Muscle		Fat		Liver		Muscle		Fat	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1,2,4,5 TCB	4.70	< 0.26	-	< 0.54	-	0.22	0.08	0.19	0.01	< 0.98	-	0.27	0.10
1,2,3,4TCB	4.46	< 0.26	-	< 0.54	-	0.03	0.02	0.19	0.01	< 0.98	-	< 0.06	-
QCB	5.03	< 0.26	-	< 0.54	-	0.42	0.20	0.19	0.01	< 0.98	-	0.50	0.17
alpha-HCH	4.00	11.80	3.70	8.52	1.21	4.85	1.41	20.81	9.43	12.11	4.16	9.87	3.63
beta-HCH	4.00	1.82	3.46	2.39	4.25	0.47	0.34	8.21	5.21	5.03	9.13	1.61	0.43
gamma-HCH	4.50	0.26	0.17	< 0.54	-	0.22	0.11	0.19	0.01	0.98	0.19	0.78	0.36
HCB	5.50	9.47	1.79	7.54	1.52	13.00	4.04	16.39	11.33	16.25	12.79	15.18	8.52
OCS	6.90	1.36	2.40	< 0.54	-	0.33	0.20	0.52	0.45	< 0.98	-	0.27	0.34
Oxychlorane	6.90	18.38	13.20	< 0.54	-	0.13	0.07	20.68	8.60	< 0.98	-	0.07	0.04
Transchlorane	6.90	< 0.26	-	< 0.54	-	0.22	0.31	0.19	0.01	< 0.98	-	0.22	0.23
Cischlorane	6.90	< 0.26	-	< 0.54	-	0.21	0.32	0.19	0.01	< 0.98	-	0.12	0.13
176 Transnonachlor	6.90	< 0.26	-	< 0.54	-	0.18	0.25	0.68	1.07	< 0.98	-	0.10	0.10
Cisnonachlor	6.90	< 0.26	-	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	< 0.06	-
p,p' DDE	6.90	1.88	1.56	2.26	2.61	0.95	0.87	1.33	1.64	2.10	2.41	2.57	2.66
p,p' DDD	6.90	< 0.26	-	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	< 0.06	-
p,p' DDT	6.00	< 0.26	-	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	< 0.06	-
Photomirex	6.00	1.11	0.53	< 0.54	-	< 0.02	-	1.32	0.37	< 0.98	-	< 0.06	-
Mirex	6.00	0.71	0.56	< 0.54	-	0.04	0.04	1.98	0.51	< 0.98	-	0.08	0.05
Heptachlor epoxide	6.00	0.67	0.90	< 0.54	-	0.03	0.03	1.37	0.93	< 0.98	-	0.14	0.18
Dieldrin	6.20	< 0.26	-	< 0.54	-	< 0.02	-	1.41	1.66	< 0.98	-	< 0.06	-
PCB31	5.60	< 0.26	-	< 0.54	-	0.51	0.48	0.19	0.01	< 0.98	-	0.92	1.19
PCB28	5.60	< 0.26	-	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	0.06	0.02
PCB52	5.84	2.29	2.78	2.25	3.93	0.34	0.23	0.19	0.01	1.95	2.17	0.55	0.30
PCB49	5.85	< 0.26	-	< 0.54	-	0.08	0.09	0.19	0.01	< 0.98	-	0.17	0.16
PCB44	5.75	< 0.26	-	< 0.54	-	0.26	0.22	0.19	0.01	< 0.98	-	0.22	0.23
PCB42	5.76	< 0.26	-	< 0.54	-	0.05	0.07	0.19	0.01	< 0.98	-	< 0.06	-
PCB64	5.95	0.40	0.14	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	< 0.06	-
PCB74	6.20	1.95	2.26	1.25	1.70	0.17	0.14	0.19	0.01	1.76	1.74	0.20	0.16
PCB70	6.20	1.46	2.63	1.82	2.98	0.24	0.30	0.19	0.01	1.37	0.88	0.37	0.32
PCB66/95	6.20	1.52	2.77	2.54	4.57	0.42	0.38	0.19	0.01	2.15	2.81	0.65	0.38
PCB60	6.11	0.93	1.44	< 0.54	-	< 0.02	-	0.19	0.01	< 0.98	-	0.25	0.42
PCB101	6.38	1.86	3.52	3.39	6.49	0.34	0.27	0.19	0.01	2.15	2.81	0.46	0.31
PCB99	6.39	1.36	2.40	1.54	2.34	0.23	0.14	0.19	0.01	< 0.98	-	0.37	0.10
PCB97	6.29	< 0.26	-	< 0.54	-	0.11	0.13	0.19	0.01	< 0.98	-	0.12	0.13
PCB87	6.29	1.06	1.74	1.68	2.66	0.21	0.18	0.19	0.01	2.34	3.04	0.26	0.24

Appendix I continued

Bathurst Female Caribou

CHEMICAL	logK _{ow}	Female Caribou(September) Fat Mobilization				Female Caribou(July - Depleted Lipid Reserves)					
		Liver	Muscle	Fat		Liver	Muscle	Fat			
		Mean	SD	Mean	SD	Mean	SD	Mean	SD		
PCB110	6.48	1.52	2.77	1.97	3.29	0.19	0.17	< 0.19	1.31	0.45	0.34
PCB151	6.64	0.93	0.87	1.39	2.02	0.16	0.12	< 0.19	1.74	0.12	0.13
PCB149	6.67	0.79	1.15	1.68	2.66	0.25	0.19	< 0.19	1.74	0.17	0.17
PCB118	6.74	0.89	1.37	1.68	2.66	0.47	0.29	< 0.19	1.31	0.62	0.19
PCB146	6.89	< 0.26	-	< 0.54	-	0.12	0.08	< 0.19	-	0.09	0.03
PCB153	6.92	0.76	1.07	2.11	3.61	0.85	0.50	1.55	1.59	1.37	0.88
PCB105	6.65	< 0.26	-	< 0.54	-	0.08	0.10	< 0.19	-	0.10	0.05
PCB141	6.82	< 0.26	-	< 0.54	-	0.08	0.05	< 0.19	-	< 0.06	-
PCB138	6.83	0.86	1.29	2.25	3.93	0.63	0.42	1.13	1.29	1.76	1.74
PCB129	6.73	< 0.26	-	< 0.54	-	0.03	0.02	< 0.19	-	< 0.06	-
PCB182/187	7.20	< 0.26	-	< 0.54	-	0.43	0.24	0.67	1.07	< 0.98	0.29
PCB183	7.00	< 0.26	-	< 0.54	-	0.14	0.09	< 0.19	-	0.11	0.11
PCB185	7.11	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.06	-
PCB174	7.11	< 0.26	-	< 0.54	-	0.04	0.04	< 0.19	-	< 0.07	0.03
PCB171	7.11	0.47	0.46	< 0.54	-	0.07	0.05	< 0.19	-	< 0.06	-
PCB200	7.20	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.06	-
PCB172	7.33	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.06	-
PCB180	7.50	0.69	0.93	< 0.54	-	0.52	0.40	0.43	0.54	< 0.98	0.49
PCB170/190	7.46	< 0.26	-	< 0.54	-	0.24	0.21	< 0.19	-	< 0.98	0.25
PCB201	7.62	1.23	1.32	< 0.54	-	0.24	0.23	< 0.19	-	< 0.98	0.33
PCB203	7.65	< 0.26	-	< 0.54	-	0.08	0.08	< 0.19	-	< 0.98	0.07
PCB195	7.56	< 0.26	-	< 0.54	-	0.07	-	< 0.19	-	< 0.98	< 0.06
PCB194	7.80	< 0.26	-	< 0.54	-	0.22	0.18	< 0.19	-	< 0.98	0.18
PCB206	8.09	< 0.26	-	< 0.54	-	0.15	0.19	< 0.19	-	< 0.98	0.19
PCB189	7.71	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.98	< 0.06
PCB77	6.36	< 0.26	-	< 0.54	-	0.06	0.10	< 0.19	-	< 0.98	0.10
PCB126	6.89	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.98	< 0.06
PCB169	7.42	< 0.26	-	< 0.54	-	< 0.02	-	< 0.19	-	< 0.98	< 0.06
Total PCBs	6.92	20.97	30.93	25.55	42.83	8.13	6.39	6.69	4.68	21.50	21.77
Arochlor12:54:1260		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Appendix I continued
 Bathurst Male Caribou

CHEMICAL	logK _{ow}	Male Caribou (September Fat Mobilization) n=5						Male Caribou (July - Depletion of Lipid Reserves) n=5											
		Liver	Mean	SD	Muscle	Mean	SD	Fat	Mean	SD	Liver	Mean	SD	Muscle	Mean	SD	Fat	Mean	SD
1,2,4,5 TCB	4.70	< 0.18	-	< 0.64	-	0.28	0.02	< 0.18	-	< 0.87	-	< 0.87	-	0.57	0.18	-	< 0.03	-	-
1,2,3,4TCB	4.46	< 0.18	-	< 0.64	-	0.04	0.06	< 0.18	-	< 0.87	-	< 0.87	-	0.42	0.15	-	11.12	4.33	-
QCB	5.03	< 0.18	-	< 0.64	-	0.47	0.10	0.25	0.14	< 0.87	-	< 0.87	-	1.47	0.88	-	16.20	11.12	4.33
alpha-HCH	4.00	15.96	4.52	11.54	4.28	6.58	1.88	20.12	7.08	22.26	16.20	11.12	4.33	1.47	0.88	-	16.20	11.12	4.33
beta-HCH	4.00	1.45	2.84	8.60	8.07	0.38	0.10	8.50	5.15	< 0.87	-	< 0.87	-	1.47	0.88	-	16.20	11.12	4.33
gamma-HCH	4.50	< 0.18	-	< 0.64	-	0.28	0.06	< 0.18	-	< 0.87	-	< 0.87	-	0.97	0.62	-	16.20	11.12	4.33
HCB	5.50	9.43	0.90	10.16	3.09	12.54	2.47	82.77	58.09	97.11	91.63	90.99	27.08	0.97	0.62	-	16.20	11.12	4.33
OCS	6.90	2.90	4.13	< 0.64	-	0.25	0.11	3.49	4.88	4.95	6.48	1.66	0.63	0.97	0.62	-	16.20	11.12	4.33
Oxychloridane	6.90	21.52	4.05	< 0.64	-	0.27	0.32	44.47	26.69	1.79	2.03	0.60	0.32	0.97	0.62	-	16.20	11.12	4.33
Tranethylordane	6.90	< 0.18	-	< 0.64	-	0.05	0.06	0.25	0.14	< 0.87	-	< 0.87	-	0.21	0.38	-	16.20	11.12	4.33
Ciechloridane	6.90	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.13	0.22	-	16.20	11.12	4.33
Tranonachlor	6.90	< 0.18	-	< 0.64	-	< 0.02	-	1.41	1.78	< 0.87	-	< 0.87	-	0.05	0.03	-	16.20	11.12	4.33
Cienonachlor	6.90	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	< 0.03	-	-	16.20	11.12	4.33
P,p' DDE	6.90	1.18	1.44	3.48	2.70	0.39	0.46	1.45	1.49	2.29	2.03	3.32	3.68	< 0.03	-	-	16.20	11.12	4.33
P,p' DDD	6.90	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.16	0.22	-	16.20	11.12	4.33
P,p' DDT	6.00	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.20	0.39	-	16.20	11.12	4.33
Photomirex	6.00	1.13	0.78	< 0.64	-	< 0.02	-	2.13	0.83	< 0.87	-	< 0.87	-	0.09	0.11	-	16.20	11.12	4.33
Mirex	6.00	1.46	1.77	< 0.64	-	0.06	0.07	3.02	1.11	< 0.87	-	< 0.87	-	0.30	0.37	-	16.20	11.12	4.33
Heptachlor epoxide	6.00	0.47	0.65	< 0.64	-	0.13	0.14	2.46	1.50	< 0.87	-	< 0.87	-	0.23	0.15	-	16.20	11.12	4.33
Dieldrin	6.20	< 0.18	-	< 0.64	-	< 0.02	-	1.50	2.97	< 0.87	-	< 0.87	-	0.17	0.22	-	16.20	11.12	4.33
PCB31	5.60	< 0.18	-	< 0.64	-	0.44	0.44	< 0.18	-	< 0.87	-	< 0.87	-	< 0.03	-	-	16.20	11.12	4.33
PCB28	5.60	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.27	0.43	-	16.20	11.12	4.33
PCB52	5.84	0.72	1.21	< 0.64	-	0.11	0.07	< 0.18	-	< 0.87	-	< 0.87	-	0.58	0.76	-	16.20	11.12	4.33
PCB49	5.85	< 0.18	-	< 0.64	-	0.02	0.00	< 0.18	-	< 0.87	-	< 0.87	-	0.18	0.33	-	16.20	11.12	4.33
PCB44	5.75	< 0.18	-	< 0.64	-	0.11	0.10	< 0.18	-	< 0.87	-	< 0.87	-	0.31	0.52	-	16.20	11.12	4.33
PCB42	5.76	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.10	0.17	-	16.20	11.12	4.33
PCB64	5.95	< 0.18	-	< 0.64	-	0.04	0.06	< 0.18	-	< 0.87	-	< 0.87	-	0.07	0.10	-	16.20	11.12	4.33
PCB74	6.20	< 0.18	-	< 0.64	-	0.08	0.04	< 0.18	-	< 0.87	-	< 0.87	-	0.55	0.34	-	16.20	11.12	4.33
PCB70	6.20	< 0.18	-	< 0.64	-	0.06	0.05	< 0.18	-	< 0.87	-	< 0.87	-	0.33	0.50	-	16.20	11.12	4.33
PCB66/95	6.20	< 0.18	-	< 0.64	-	0.19	0.10	< 0.18	-	< 0.87	-	< 0.87	-	0.69	0.75	-	16.20	11.12	4.33
PCB60	6.11	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.87	-	0.21	0.15	-	16.20	11.12	4.33
PCB101	6.38	< 0.18	-	< 0.64	-	0.11	0.06	< 0.18	-	< 0.87	-	< 0.87	-	0.52	0.63	-	16.20	11.12	4.33
PCB99	6.39	< 0.18	-	< 0.64	-	0.21	0.11	1.46	1.59	< 0.87	-	< 0.87	-	0.97	0.47	-	16.20	11.12	4.33
PCB97	6.29	< 0.18	-	< 0.64	-	< 0.02	-	0.39	0.49	< 0.87	-	< 0.87	-	0.11	0.18	-	16.20	11.12	4.33
PCB87	6.28	< 0.18	-	< 0.64	-	0.05	0.07	< 0.18	-	< 0.87	-	< 0.87	-	0.21	0.33	-	16.20	11.12	4.33

Appendix I continued

Bathurst Male Caribou

CHEMICAL	logK _{ow}	Male Caribou (September Fat Mobilization) n=5						Male Caribou (July - Depletion of Lipid Reserves)=5						
		Liver		Muscle		Fat		Liver		Muscle		Fat		
		mean	SD	Mean	SD	Mean	SD	mean	SD	Mean	SD	Mean	SD	
PCB110	6.48	< 0.18	-	< 0.64	-	0.07	0.05	< 0.18	-	< 0.87	-	0.30	0.40	
PCB151	6.64	< 0.18	-	< 0.64	-	0.03	0.03	< 0.18	-	< 0.87	-	0.08	0.09	
PCB149	6.67	< 0.18	-	< 0.64	-	0.03	0.03	< 0.18	-	< 0.87	-	0.16	0.29	
PCB118	6.74	< 0.18	-	< 0.64	-	0.26	0.10	0.21	0.07	< 0.87	-	1.68	0.72	
PCB146	6.89	< 0.18	-	< 0.64	-	0.03	0.01	0.18	0.02	< 0.87	-	0.22	0.16	
PCB153	6.92	< 0.18	-	< 0.64	-	0.47	0.17	6.51	5.12	< 0.87	-	3.25	1.32	
PCB105	6.65	< 0.18	-	< 0.64	-	0.03	0.02	< 0.18	-	< 0.87	-	0.33	0.12	
PCB141	6.82	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.04	0.03	
PCB138	6.83	< 0.18	-	< 0.64	-	0.31	0.09	4.88	4.09	< 0.87	-	1.81	0.84	
PCB129	6.73	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.33	0.20	
PCB182/187	7.20	< 0.18	-	< 0.64	-	0.19	0.17	1.02	1.89	< 0.87	-	0.87	0.44	
PCB183	7.00	< 0.18	-	< 0.64	-	0.03	0.01	< 0.18	-	< 0.87	-	0.17	0.11	
PCB185	7.11	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.03	-	
PCB174	7.11	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.06	0.07	
PCB171	7.11	< 0.18	-	< 0.64	-	0.03	0.02	< 0.18	-	< 0.87	-	0.18	0.16	
PCB200	7.20	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.06	0.04	
PCB172	7.33	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.14	0.12	
PCB180	7.50	< 0.18	-	< 0.64	-	0.25	0.04	0.78	1.08	< 0.87	-	1.65	0.76	
PCB170/190	7.46	< 0.18	-	< 0.64	-	0.17	0.05	< 0.18	-	< 0.87	-	0.92	0.39	
PCB201	7.62	< 0.18	-	< 0.64	-	0.14	0.05	0.49	0.69	< 0.87	-	0.89	0.35	
PCB203	7.65	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.14	0.10	
PCB195	7.56	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.24	0.11	
PCB194	7.80	< 0.18	-	< 0.64	-	0.11	0.08	< 0.18	-	< 0.87	-	0.51	0.17	
PCB206	8.09	< 0.18	-	< 0.64	-	0.06	0.06	< 0.18	-	< 0.87	-	0.61	0.33	
PCB189	7.71	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.03	-	
PCB77	6.36	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	0.08	0.11	
PCB126	6.89	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.03	-	
PCB169	7.42	< 0.18	-	< 0.64	-	< 0.02	-	< 0.18	-	< 0.87	-	< 0.03	-	
Total PCBs	6.92		0.72	1.21	0.00	0.00	3.61	2.09	15.94	15.06	< 0.87	-	19.83	13.10
Arochlor12:54:1260	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

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Appendix I continued

Bathurst Wolves

CHEMICAL	kow	Female Wolf		n=6		Male Wolf		n=5						
		Liver	mean	SD	Muscle	Mean	SD	Liver	mean	SD	Muscle	Mean	SD	Fat
1,2,4,5 TCB	4.70	1.06	0.62	1.15	1.03	1.71	0.71	1.38	0.44	< 0.51	-	2.07	0.67	
1,2,3,4TCB	4.46	< 0.21	-	< 0.43	-	0.13	0.06	< 0.21	-	< 0.51	-	0.10	0.10	
QCB	5.03	1.71	0.56	3.09	0.94	2.20	0.81	1.75	0.81	3.58	1.04	2.77	0.94	
alpha-HCH	4.00	2.96	0.60	8.78	3.89	8.19	3.57	3.53	1.55	8.65	3.02	9.64	3.91	
beta-HCH	4.00	6.36	5.12	24.46	17.73	11.72	3.26	7.94	5.63	39.14	23.63	17.10	10.66	
gamma-HCH	4.50	< 0.21	-	0.63	0.43	0.05	0.06	< 0.21	-	< 0.51	-	0.03	0.04	
HCB	5.50	38.03	17.07	82.42	35.48	78.39	30.09	44.96	11.70	93.24	6.44	97.33	11.26	
OCS	6.90	10.83	6.60	3.03	1.19	2.21	1.40	5.60	1.50	3.93	2.72	3.53	2.96	
Oxychlorodane	6.90	44.61	14.71	17.30	7.00	7.53	5.25	48.96	27.04	28.54	16.30	13.52	7.26	
Tranochlorodane	6.90	0.49	0.53	1.26	1.40	0.07	0.08	0.36	0.30	0.83	0.62	0.13	0.11	
Cischlorodane	6.90	0.26	0.16	0.59	0.34	0.02	0.02	< 0.21	-	< 0.51	-	0.07	0.05	
Transnonachlor	6.90	0.37	0.27	0.51	0.19	0.16	0.13	0.46	0.51	< 0.51	-	0.81	0.30	
Tranonachlor	6.90	0.21	0.05	0.54	0.41	0.02	0.01	< 0.21	-	< 0.51	-	0.06	0.02	
p,p' DDE	6.90	2.84	3.22	< 0.43	-	0.02	0.02	2.10	2.08	0.72	0.40	< 0.01	-	
p,p' DDD	6.90	0.65	0.56	< 0.43	-	< 0.1	-	1.08	0.62	< 0.51	-	0.02	0.01	
p,p' DDT	6.00	< 0.21	-	< 0.43	-	0.01	0.00	< 0.21	-	< 0.51	-	0.21	0.39	
Photomirex	6.00	0.98	0.55	0.71	0.62	0.51	0.43	0.66	0.20	2.50	3.59	1.07	1.06	
Mirex	6.00	1.68	0.76	1.09	0.96	0.78	0.76	1.24	0.31	0.84	0.77	1.36	1.06	
Heptachlor epoxid	6.00	7.01	5.60	2.98	0.82	1.51	0.91	8.22	2.80	5.07	3.53	2.93	2.24	
Dieldrin	6.20	3.60	6.29	0.93	0.73	< 0.1	-	0.62	0.80	< 0.51	-	0.12	0.23	
PCB31	5.60	< 0.21	-	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	< 0.01	-	
PCB26	5.60	< 0.21	-	< 0.43	-	0.06	0.05	< 0.21	-	< 0.51	-	0.12	0.01	
PCB52	5.84	1.33	1.15	< 0.43	-	< 0.1	-	0.70	0.66	< 0.51	-	0.17	0.19	
PCB49	5.85	0.26	0.16	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	0.06	0.10	
PCB44	5.75	< 0.21	-	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	< 0.01	-	
PCB42	5.76	< 0.21	-	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	< 0.01	-	
PCB64	5.95	< 0.21	-	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	< 0.01	-	
PCB74	6.20	0.49	0.45	< 0.43	-	0.58	0.25	0.91	0.93	1.66	2.42	2.39	3.63	
PCB70	6.20	0.62	0.59	0.69	0.17	0.02	0.02	0.46	0.51	1.59	0.92	0.12	0.21	
P66/95	6.20	0.69	0.84	0.59	0.34	0.04	0.07	0.51	0.61	< 0.51	-	0.22	0.19	
PCB60	6.11	0.47	0.59	< 0.43	-	0.09	0.09	< 0.21	-	< 0.51	-	0.80	1.14	
PCB101	6.38	0.80	0.66	< 0.43	-	0.01	0.00	0.56	0.71	< 0.51	-	0.14	0.16	
PCB99	6.39	2.58	1.04	2.35	1.10	1.33	0.72	16.90	25.70	2.35	1.51	4.38	4.86	
PCB97	6.29	< 0.21	-	< 0.43	-	< 0.1	-	< 0.21	-	< 0.51	-	< 0.01	-	
PCB87	6.29	0.47	0.40	< 0.43	-	< 0.1	-	0.31	0.20	< 0.51	-	< 0.01	-	

Appendix I continued
Inuvik Caribou

CHEMICAL	logK _{ow}	Female Caribou n=2						Male caribou n=8					
		Liver		Muscle		Fat		Liver		Muscle		Fat	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1,2,4,5 TCB	4.70	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
1,2,3,4TCB	4.46	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
QCB	5.03	< 0.20	-	< 0.43	-	0.50	0.07	< 0.24	-	< 0.55	-	0.70	0.38
alpha-HCH	4.00	7.37	2.03	6.71	2.60	2.60	0.60	23.41	36.81	3.42	1.88	3.48	0.78
beta-HCH	4.00	2.90	0.95	2.83	0.38	0.35	0.10	3.58	1.12	3.54	1.33	0.28	0.07
gamma-HCH	4.50	< 0.20	-	< 0.43	-	0.07	0.08	< 0.24	-	< 0.55	-	0.05	0.07
HCB	5.50	41.00	8.13	39.36	1.28	31.16	12.30	36.15	9.87	33.87	9.87	31.91	9.71
OCS	6.90	1.14	1.35	< 0.43	-	0.14	0.05	1.24	0.91	< 0.55	-	0.21	0.07
Oxychlorane	6.90	18.25	16.89	< 0.43	-	< 0.01	-	17.97	7.93	< 0.55	-	0.07	0.08
Transchlorane	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
Cischlorane	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
182 Transnonachlor	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
Cisnonachlor	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
p,p' DDE	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
p,p' DDD	6.90	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
p,p' DDT	6.00	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
Photomirex	6.00	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
Mirex	6.00	0.75	0.77	< 0.43	-	< 0.01	-	0.91	0.92	< 0.55	-	< 0.01	-
Heptachlor epoxide	6.00	1.98	0.17	< 0.43	-	< 0.01	-	1.94	0.34	< 0.55	-	< 0.01	-
Dieldrin	6.20	1.35	1.65	< 0.43	-	< 0.01	-	2.20	0.91	< 0.55	-	< 0.01	-
PCB31	5.60	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB28	5.60	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB52	5.84	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB49	5.85	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB44	5.75	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB42	5.76	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB64	5.95	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB74	6.20	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB70	6.20	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB66/95	6.20	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB60	6.11	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB101	6.38	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB99	6.39	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB97	6.29	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB87	6.29	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-

Appendix I continued
Inuvik Caribou

CHEMICAL	logK _{ow}	Female Caribou n=2						Male caribou n=8					
		Liver		Muscle		Fat		Liver		Muscle		Fat	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
PCB110	6.48	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB151	6.64	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB149	6.67	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB118	6.74	< 0.20	-	< 0.43	-	0.14	0.18	< 0.24	-	< 0.55	-	0.04	0.08
PCB146	6.89	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB153	6.92	< 0.20	-	< 0.43	-	0.22	0.02	< 0.24	-	< 0.55	-	0.25	0.09
PCB105	6.65	< 0.20	-	< 0.43	-	0.05	0.05	< 0.24	-	< 0.55	-	0.03	0.04
PCB141	6.82	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB138	6.83	< 0.20	-	< 0.43	-	0.19	0.02	< 0.24	-	< 0.55	-	0.23	0.05
PCB129	6.73	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB182/187	7.20	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB183	7.00	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB185	7.11	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB174	7.11	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB171	7.11	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB200	7.20	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB172	7.33	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB180	7.50	< 0.20	-	< 0.43	-	0.11	0.01	< 0.24	-	< 0.55	-	0.13	0.06
PCB170/190	7.46	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB201	7.62	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB203	7.65	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB195	7.56	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB194	7.80	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB206	8.09	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB189	7.71	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB77	6.36	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB126	6.89	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
PCB169	7.42	< 0.20	-	< 0.43	-	< 0.01	-	< 0.24	-	< 0.55	-	< 0.01	-
Total PCBs	6.92	< 0.20	-	< 0.43	-	0.71	0.28	< 0.24	-	< 0.55	-	0.69	0.32
Arochlor12:54:1260		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250		NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

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Appendix I continued
Inuvik Wolves

CHEMICAL	logKow	Inuvik WOLVESn=10					
		Liver		Muscle		Fat	
		mean	SD	Mean	SD	Mean	SD
1,2,4,5 TCB	4.70	1.06	1.83	< 0.45	-	1.75	0.99
1,2,3,4TCB	4.46	< 0.18	-	< 0.45	-	0.10	0.07
QCB	5.03	4.42	1.86	2.72	1.65	2.21	1.13
alpha-HCH	4.00	6.93	3.30	8.12	3.17	6.76	3.40
beta-HCH	4.00	9.33	6.37	13.40	8.99	9.78	5.40
gamma-HCH	4.50	0.28	0.26	1.26	1.66	0.08	0.10
HCB	5.50	91.79	55.60	76.05	39.58	78.20	52.84
OCS	6.90	9.92	3.74	2.88	2.03	1.70	1.05
Oxychlorane	6.90	59.35	20.72	16.22	8.44	5.57	3.07
Transchlorane	6.90	< 0.18	-	0.54	0.30	0.10	0.14
Cischlorane	6.90	< 0.18	-	< 0.45	-	0.01	0.00
Transnonachlor	6.90	0.31	0.38	< 0.45	-	0.37	0.55
Cisnonachlor	6.90	< 0.18	-	< 0.45	-	0.04	0.09
p,p' DDE	6.90	3.04	9.06	4.64	12.19	2.53	6.92
p,p' DDD	6.90	< 0.18	-	< 0.45	-	0.03	0.05
p,p' DDT	6.00	< 0.18	-	< 0.45	-	0.03	0.04
Photomirex	6.00	1.07	0.98	0.51	0.16	0.51	0.33
Mirex	6.00	3.91	4.33	0.82	0.80	0.63	0.40
Heptachlor epoxide	6.00	16.11	10.05	1.64	1.61	1.72	0.82
Dieldrin	6.20	10.48	10.53	1.24	1.81	0.15	0.28
PCB31	5.60	< 0.18	-	< 0.45	-	< 0.01	-
PCB28	5.60	< 0.18	-	< 0.45	-	< 0.01	-
PCB52	5.84	< 0.18	-	< 0.45	-	0.13	0.14
PCB49	5.85	< 0.18	-	< 0.45	-	< 0.01	-
PCB44	5.75	< 0.18	-	< 0.45	-	0.04	0.08
PCB42	5.76	< 0.18	-	< 0.45	-	0.04	0.08
PCB64	5.95	< 0.18	-	< 0.45	-	< 0.01	-
PCB74	6.20	0.29	0.30	< 0.45	-	0.69	0.35
PCB70	6.20	< 0.18	-	< 0.45	-	0.03	0.03
PCB66/95	6.20	< 0.18	-	< 0.45	-	0.12	0.13
PCB60	6.11	< 0.18	-	< 0.45	-	< 0.01	-
PCB101	6.38	< 0.18	-	< 0.45	-	0.10	0.11
PCB99	6.39	4.92	2.64	1.60	1.52	1.42	0.90
PCB97	6.29	< 0.18	-	< 0.45	-	< 0.01	-
PCB87	6.29	< 0.18	-	< 0.45	-	0.04	0.07

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Appendix I continued
Inuvik Wolves

CHEMICAL	logKow	Inuvik WOLVESn=10					
		Liver		Muscle		Fat	
		mean	SD	Mean	SD	Mean	SD
PCB110	6.48	< 0.18	-	< 0.45	-	0.03	0.06
PCB151	6.64	< 0.18	-	< 0.45	-	< 0.01	-
PCB149	6.67	< 0.18	-	< 0.45	-	< 0.01	-
PCB118	6.74	5.42	5.33	3.38	3.31	3.78	2.93
PCB148	6.89	0.63	1.47	0.84	1.26	0.18	0.26
PCB153	6.92	22.80	31.34	14.76	18.79	9.03	6.78
PCB105	6.65	0.30	0.34	< 0.45	-	0.57	0.60
PCB141	6.82	< 0.18	-	< 0.45	-	< 0.01	-
PCB138	6.83	7.38	15.67	3.28	7.84	2.31	2.03
PCB129	6.73	< 0.18	-	< 0.45	-	0.04	0.03
PCB182/187	7.20	1.60	4.50	1.06	1.96	0.14	0.31
PCB183	7.00	2.45	4.69	1.01	1.30	0.48	0.47
PCB185	7.11	< 0.18	-	< 0.45	-	< 0.01	-
PCB174	7.11	< 0.18	-	< 0.45	-	< 0.01	-
PCB171	7.11	2.01	2.23	0.71	0.77	1.17	1.03
PCB200	7.20	< 0.18	-	< 0.45	-	0.48	0.53
PCB172	7.33	< 0.18	-	< 0.45	-	0.07	0.05
PCB180	7.50	35.59	32.49	16.10	13.95	27.61	42.89
PCB170/190	7.46	13.82	11.48	10.16	6.78	9.52	8.43
PCB201	7.62	1.86	4.69	0.78	1.09	0.16	0.10
PCB203	7.65	1.90	5.45	0.73	0.92	0.11	0.10
PCB195	7.56	0.45	0.90	4.88	5.13	0.22	0.21
PCB194	7.80	6.86	8.38	6.75	8.08	6.70	8.36
PCB206	8.09	7.21	10.61	6.17	8.25	2.76	3.60
PCB189	7.71	< 0.18	-	< 0.45	-	0.47	0.52
PCB77	6.36	< 0.18	-	< 0.45	-	< 0.01	-
PCB126	6.89	< 0.18	-	< 0.45	-	< 0.01	-
PCB169	7.42	< 0.18	-	< 0.45	-	< 0.01	-
Total PCBs	6.92	115.28	142.51	72.23	80.93	68.43	81.16
Arochlor12:54:1260	NA	NA	NA	NA	NA	NA	NA
Arochlor1250	NA	NA	NA	NA	NA	NA	NA

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APPENDIX II

Chemical fugacities (Pa) of organic contaminants in lichens, caribou and wolves from Cambridge Bay, Bathurst Inlet, and Inuvik.

Appendix II: Geometric means (GM) of organochlorine fugacities (Pa) in lichens, caribou and wolves from Cambridge Bay, Bathurst Inlet and Inuvik. Range of standard deviations are represented as lower and upper 1 standard deviation of the geometric means.

Cambridge Bay	logK _{ow}	GM	lichen n=9		female caribou n=5			male caribou n=5		
			lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
1,2,4,5 TCB	4.7	8.93E-10	2.83E-10	2.83E-09	2.88E-09	1.66E-09	4.98E-09	5.45E-09	4.11E-09	7.22E-09
1,2,3,4TCB	4.46	3.03E-09	1.83E-09	5.03E-09	1.69E-10	1.39E-10	2.05E-10	1.69E-10	1.55E-10	1.85E-10
QCB	5.03	1.31E-09	6.57E-10	2.62E-09	1.63E-09	1.13E-09	2.34E-09	2.49E-09	2.05E-09	3.01E-09
alpha-HCH	4	4.08E-09	1.73E-09	9.60E-09	5.19E-09	2.89E-09	9.30E-09	7.91E-09	5.12E-09	1.22E-08
beta-HCH	4	6.79E-11	1.85E-11	2.50E-10	3.00E-10	2.34E-10	3.84E-10	4.06E-10	3.22E-10	5.13E-10
gamma-HCH	4.5	2.62E-10	1.67E-10	4.12E-10	6.68E-11	4.57E-11	9.77E-11	8.53E-11	6.56E-11	1.11E-10
HCB	5.5	2.38E-09	1.67E-09	3.38E-09	1.93E-08	1.28E-08	2.89E-08	4.28E-08	3.47E-08	5.30E-08
OCS	6.9	8.23E-13	3.81E-13	1.78E-12	1.35E-12	5.72E-13	3.17E-12	3.63E-12	2.66E-12	4.96E-12
Oxychlorane	6.9	7.25E-13	2.66E-13	1.98E-12	8.75E-13	4.60E-13	1.67E-12	1.92E-12	1.12E-12	3.29E-12
Transchlorane	6.9	5.76E-13	1.82E-13	1.82E-12	6.25E-14	5.15E-14	7.60E-14	6.27E-14	5.74E-14	6.84E-14
Cischlorane	6.9	6.52E-13	3.05E-13	1.39E-12	4.35E-14	3.58E-14	5.29E-14	4.36E-14	4.00E-14	4.76E-14
Transnonachlor	6.9	5.14E-13	1.26E-13	2.09E-12	2.52E-13	8.44E-14	7.50E-13	4.07E-13	2.33E-13	7.10E-13
Cisnonachlor	6.9	2.28E-13	8.41E-14	6.17E-13	2.03E-14	1.67E-14	2.47E-14	2.04E-14	1.87E-14	2.22E-14
p,p' DDE	6.9	1.86E-13	5.88E-14	5.88E-13	1.96E-13	4.84E-14	7.92E-13	6.51E-13	2.88E-13	1.47E-12
p,p' DDD	6.9	9.71E-13	3.50E-13	2.70E-12	8.08E-14	6.65E-14	9.82E-14	8.10E-14	7.42E-14	8.84E-14
p,p' DDT	6	1.43E-11	4.95E-12	4.11E-11	6.42E-13	5.28E-13	7.80E-13	6.44E-13	5.90E-13	7.03E-13
Photomirex	6	5.91E-12	1.87E-12	1.87E-11	2.10E-12	6.38E-13	6.94E-12	5.60E-12	3.85E-12	8.15E-12
Mirex	6	4.24E-12	1.34E-12	1.34E-11	1.59E-12	4.31E-13	5.90E-12	4.71E-12	2.06E-12	1.08E-11
Heptachlor epoxide	6	1.30E-11	5.30E-12	3.16E-11	9.17E-12	1.92E-12	4.38E-11	2.16E-11	1.35E-11	3.45E-11
Dieldrin	6.2	4.29E-12	1.13E-12	1.63E-11	4.05E-13	3.33E-13	4.92E-13	4.06E-13	3.72E-13	4.43E-13
PCB31	5.6	1.99E-11	6.28E-12	6.28E-11	2.16E-12	1.78E-12	2.62E-12	2.17E-12	1.98E-12	2.36E-12
PCB28	5.6	1.99E-11	6.28E-12	6.28E-11	2.16E-12	1.78E-12	2.62E-12	2.17E-12	1.98E-12	2.36E-12
PCB52	5.84	2.11E-11	9.29E-12	4.77E-11	1.24E-12	1.02E-12	1.51E-12	1.61E-11	1.19E-11	2.17E-11
PCB49	5.85	1.12E-11	3.53E-12	3.53E-11	1.21E-12	1.00E-12	1.48E-12	1.22E-12	1.12E-12	1.33E-12
PCB44	5.75	1.50E-11	4.46E-12	5.05E-11	1.53E-12	1.26E-12	1.86E-12	1.53E-12	1.40E-12	1.67E-12
PCB42	5.76	1.38E-11	4.35E-12	4.35E-11	1.49E-12	1.23E-12	1.82E-12	1.50E-12	1.37E-12	1.63E-12
PCB64	5.95	8.88E-12	2.81E-12	2.81E-11	9.65E-13	7.94E-13	1.17E-12	9.67E-13	8.86E-13	1.06E-12
PCB74	6.2	6.20E-12	2.28E-12	1.68E-11	2.62E-12	5.47E-13	1.26E-11	1.33E-11	1.13E-11	1.55E-11
PCB70	6.2	8.14E-12	3.80E-12	1.74E-11	5.43E-13	4.46E-13	6.59E-13	5.44E-13	4.98E-13	5.94E-13
PCB66/95	6.2	8.33E-12	3.87E-12	1.79E-11	5.55E-12	2.95E-12	1.05E-11	1.06E-11	8.24E-12	1.36E-11
PCB60	6.11	6.14E-12	1.94E-12	1.94E-11	6.67E-13	5.49E-13	8.11E-13	4.98E-12	1.15E-12	2.17E-11
PCB101	6.38	5.50E-12	2.55E-12	1.19E-11	2.42E-12	7.39E-13	7.93E-12	5.15E-12	3.77E-12	7.03E-12

Appendix II continued

Cambridge Bay

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	logK _{ow}	GM	lichen	n=9	female			male		
			lower 1	upper 1	GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
			SD	SD						
PCB99	6.39	4.58E-12	2.14E-12	9.76E-12	8.73E-12	3.98E-12	1.92E-11	1.26E-11	9.93E-12	1.59E-11
PCB97	6.29	3.63E-12	1.15E-12	1.15E-11	3.95E-13	3.25E-13	4.80E-13	3.96E-13	3.63E-13	4.32E-13
PCB87	6.29	3.61E-12	1.08E-12	1.21E-11	3.95E-13	3.25E-13	4.80E-13	3.96E-13	3.63E-13	4.32E-13
PCB110	6.48	3.08E-12	1.28E-12	7.39E-12	3.87E-13	1.55E-13	9.64E-13	4.76E-13	1.40E-13	1.62E-12
PCB151	6.64	1.30E-12	4.59E-13	3.66E-12	1.76E-13	1.45E-13	2.14E-13	1.77E-13	1.62E-13	1.93E-13
PCB149	6.67	2.96E-12	5.37E-13	1.63E-11	1.65E-13	1.36E-13	2.00E-13	1.65E-13	1.51E-13	1.80E-13
PCB118	6.74	1.29E-12	4.08E-13	4.08E-12	7.34E-12	3.46E-12	1.56E-11	1.17E-11	9.39E-12	1.45E-11
PCB146	6.89	7.87E-13	2.66E-13	2.32E-12	4.06E-13	9.74E-14	1.69E-12	1.42E-12	9.97E-13	2.02E-12
PCB153	6.92	1.23E-12	5.84E-13	2.58E-12	6.34E-12	2.66E-12	1.51E-11	1.34E-11	1.07E-11	1.68E-11
PCB105	6.65	1.57E-12	5.00E-13	4.93E-12	6.49E-13	1.63E-13	2.59E-12	3.78E-13	7.69E-14	1.86E-12
PCB141	6.82	1.07E-12	3.39E-13	3.39E-12	1.17E-13	9.59E-14	1.42E-13	1.17E-13	1.07E-13	1.28E-13
PCB138	6.83	1.54E-12	7.19E-13	3.30E-12	6.49E-12	2.94E-12	1.43E-11	9.19E-12	7.27E-12	1.16E-11
PCB129	6.73	1.09E-12	3.44E-13	3.44E-12	2.87E-13	7.51E-14	1.10E-12	1.17E-12	7.97E-13	1.72E-12
PCB182/187	7.2	3.69E-13	1.17E-13	1.17E-12	4.40E-13	1.97E-13	9.83E-13	9.87E-13	7.29E-13	1.34E-12
PCB183	7	5.85E-13	1.85E-13	1.85E-12	1.58E-13	3.94E-14	6.37E-13	1.56E-13	5.75E-14	4.24E-13
PCB185	7.11	4.54E-13	1.44E-13	1.44E-12	4.93E-14	4.06E-14	6.00E-14	4.95E-14	4.53E-14	5.40E-14
PCB174	7.11	4.54E-13	1.44E-13	1.44E-12	4.93E-14	4.06E-14	6.00E-14	4.95E-14	4.53E-14	5.40E-14
PCB171	7.11	4.54E-13	1.44E-13	1.44E-12	7.48E-14	2.44E-14	2.30E-13	5.16E-13	4.40E-13	6.06E-13
PCB200	7.2	3.69E-13	1.17E-13	1.17E-12	4.01E-14	3.30E-14	4.87E-14	4.02E-14	3.68E-14	4.39E-14
PCB172	7.33	2.74E-13	8.65E-14	8.65E-13	2.97E-14	2.45E-14	3.61E-14	1.23E-13	4.63E-14	3.29E-13
PCB180	7.5	1.85E-13	5.85E-14	5.85E-13	5.20E-13	2.11E-13	1.28E-12	1.08E-12	8.99E-13	1.30E-12
PCB170/190	7.46	1.86E-13	5.89E-14	5.89E-13	1.22E-13	2.06E-14	7.26E-13	7.81E-13	6.22E-13	9.81E-13
PCB201	7.62	1.29E-13	4.08E-14	4.08E-13	6.98E-14	1.38E-14	3.53E-13	3.83E-13	2.95E-13	4.97E-13
PCB203	7.65	1.20E-13	3.80E-14	3.80E-13	1.98E-14	7.94E-15	4.94E-14	2.05E-14	8.14E-15	5.17E-14
PCB195	7.56	1.48E-13	4.68E-14	4.68E-13	2.30E-14	8.52E-15	6.21E-14	6.00E-14	1.35E-14	2.66E-13
PCB194	7.8	8.52E-14	2.69E-14	2.69E-13	1.44E-14	4.44E-15	4.65E-14	8.94E-14	7.33E-14	1.09E-13
PCB206	8.09	4.37E-14	1.38E-14	1.38E-13	4.75E-15	3.91E-15	5.77E-15	2.83E-14	8.34E-15	9.59E-14
PCB189	7.71	NA	NA	NA	1.14E-14	9.37E-15	1.38E-14	1.14E-14	1.05E-14	1.25E-14
PCB77	6.36	NA	NA	NA	2.55E-13	2.10E-13	3.10E-13	2.55E-13	2.34E-13	2.79E-13
PCB126	6.89	NA	NA	NA	7.52E-14	6.19E-14	9.14E-14	7.54E-14	6.91E-14	8.23E-14
PCB169	7.42	NA	NA	NA	3.28E-14	1.13E-14	9.48E-14	2.23E-14	2.04E-14	2.43E-14
Total PCBs	6.93	2.77E-11	1.19E-11	6.50E-11	2.03E-11	7.79E-12	4.40E-11	4.05E-11	3.32E-11	4.94E-11

Appendix II continued

Cambridge Bay

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	logK _{ow}	female wolves			male wolves		
		GM	n=6 lower 1 SD	upper 1 SD	GM	n=6 lower 1 SD	upper 1 SD
1,2,4,5 TCB	4.7	1.44E-08	7.24E-09	2.86E-08	6.17E-09	3.84E-09	9.90E-09
1,2,3,4TCB	4.46	2.04E-10	1.03E-10	4.05E-10	3.11E-10	8.79E-11	1.10E-09
QCB	5.03	4.96E-09	2.47E-09	9.96E-09	2.13E-09	1.62E-09	2.80E-09
alpha-HCH	4	1.82E-09	1.20E-09	2.75E-09	1.47E-09	1.05E-09	2.08E-09
beta-HCH	4	3.64E-09	1.36E-09	9.74E-09	1.12E-09	9.14E-10	1.38E-09
gamma-HCH	4.5	1.70E-12	1.57E-12	1.84E-12	4.15E-12	9.69E-13	1.78E-11
HCB	5.5	2.27E-08	1.57E-08	3.30E-08	1.61E-08	1.35E-08	1.92E-08
OCS	6.9	7.79E-12	4.63E-12	1.31E-11	2.98E-12	1.57E-12	5.57E-12
Oxychlorane	6.9	6.43E-11	1.96E-11	2.12E-10	1.13E-11	5.69E-12	2.23E-11
Transchlorane	6.9	5.35E-14	4.94E-14	5.81E-14	5.14E-14	5.08E-14	5.21E-14
Cischlorane	6.9	7.07E-14	2.00E-14	2.50E-13	3.58E-14	3.53E-14	3.62E-14
Transnonachlor	6.9	1.74E-12	2.93E-13	1.04E-11	8.65E-13	3.18E-13	2.38E-12
Cisnonachlor	6.9	6.79E-14	9.96E-15	4.63E-13	3.39E-14	1.13E-14	1.02E-13
p,p' DDE	6.9	7.27E-14	4.22E-15	1.25E-12	5.54E-14	8.50E-15	3.81E-13
p,p' DDD	6.9	6.92E-14	6.38E-14	7.51E-14	6.65E-14	6.56E-14	6.73E-14
p,p' DDT	6	5.50E-13	5.07E-13	5.98E-13	5.28E-13	5.21E-13	5.35E-13
Photomirex	6	4.41E-11	1.58E-11	1.23E-10	1.43E-11	7.83E-12	2.80E-11
Mirex	6	2.06E-11	8.34E-12	5.09E-11	8.58E-12	4.03E-12	1.83E-11
Heptachlor epoxide	6	1.04E-10	3.59E-11	2.99E-10	5.57E-11	2.74E-11	1.13E-10
Dieldrin	6.2	6.11E-11	1.67E-11	2.24E-10	4.87E-11	1.87E-11	1.27E-10
PCB31	5.6	1.85E-12	1.70E-12	2.01E-12	1.78E-12	1.75E-12	1.80E-12
PCB28	5.6	1.85E-12	1.70E-12	2.01E-12	1.78E-12	1.75E-12	1.80E-12
PCB52	5.84	1.06E-12	9.81E-13	1.15E-12	1.02E-12	1.01E-12	1.04E-12
PCB49	5.85	1.04E-12	9.59E-13	1.13E-12	9.99E-13	9.86E-13	1.01E-12
PCB44	5.75	1.31E-12	1.21E-12	1.42E-12	1.26E-12	1.24E-12	1.27E-12
PCB42	5.76	1.28E-12	1.18E-12	1.39E-12	1.23E-12	1.21E-12	1.24E-12
PCB64	5.95	8.26E-13	7.61E-13	8.96E-13	7.93E-13	7.83E-13	8.04E-13
PCB74	6.2	2.34E-11	5.51E-12	9.94E-11	9.19E-12	5.76E-12	1.47E-11
PCB70	6.2	1.02E-12	1.98E-13	5.28E-12	4.46E-13	4.40E-13	4.52E-13
PCB66/95	6.2	1.45E-12	1.52E-13	1.39E-11	4.46E-13	4.40E-13	4.52E-13
PCB60	6.11	2.64E-12	1.26E-13	5.54E-11	5.49E-13	5.42E-13	5.58E-13
PCB101	6.38	6.93E-13	1.39E-13	3.46E-12	2.95E-13	2.91E-13	2.99E-13

Appendix II continued

Cambridge Bay

	logK _{ow}	female wolves			male wolves		
		GM	n=6 lower 1 SD	upper 1 SD	GM	n=6 lower 1 SD	upper 1 SD
PCB99	6.39	7.50E-11	1.95E-11	2.88E-10	2.54E-11	1.43E-11	4.52E-11
PCB97	6.29	3.38E-13	3.12E-13	3.67E-13	3.25E-13	3.21E-13	3.29E-13
PCB87	6.29	3.38E-13	3.12E-13	3.67E-13	3.25E-13	3.21E-13	3.29E-13
PCB110	6.48	2.18E-13	2.01E-13	2.37E-13	2.10E-13	2.07E-13	2.12E-13
PCB151	6.64	1.51E-13	1.39E-13	1.64E-13	1.45E-13	1.43E-13	1.47E-13
PCB149	6.67	1.41E-13	1.30E-13	1.53E-13	1.91E-13	8.19E-14	4.48E-13
PCB118	6.74	1.45E-11	3.00E-12	7.00E-11	6.21E-12	3.47E-12	1.11E-11
PCB146	6.89	5.99E-13	7.22E-14	4.96E-12	5.31E-13	1.76E-13	1.61E-12
PCB153	6.92	4.85E-11	1.52E-11	1.55E-10	2.36E-11	1.42E-11	3.92E-11
PCB105	6.65	1.48E-13	1.36E-13	1.60E-13	1.42E-13	1.40E-13	1.44E-13
PCB141	6.82	9.98E-14	9.20E-14	1.08E-13	9.59E-14	9.46E-14	9.71E-14
190 PCB138	6.83	1.57E-11	4.43E-12	5.60E-11	9.17E-12	5.21E-12	1.61E-11
PCB129	6.73	3.24E-13	6.83E-14	1.53E-12	1.31E-13	6.31E-14	2.73E-13
PCB182/187	7.2	9.56E-14	1.26E-14	7.24E-13	4.92E-14	1.85E-14	1.31E-13
PCB183	7	1.15E-12	1.31E-13	1.01E-11	7.67E-13	4.09E-13	1.44E-12
PCB185	7.11	4.22E-14	3.89E-14	4.58E-14	4.06E-14	4.01E-14	4.11E-14
PCB174	7.11	4.22E-14	3.89E-14	4.58E-14	4.06E-14	4.01E-14	4.11E-14
PCB171	7.11	4.22E-14	3.89E-14	4.58E-14	4.06E-14	4.01E-14	4.11E-14
PCB200	7.2	1.01E-13	1.19E-14	8.64E-13	5.06E-14	1.77E-14	1.44E-13
PCB172	7.33	7.41E-14	1.65E-14	3.34E-13	2.44E-14	2.41E-14	2.48E-14
PCB180	7.5	1.79E-11	6.63E-12	4.86E-11	5.35E-12	3.41E-12	8.39E-12
PCB170/190	7.46	1.18E-11	4.50E-12	3.12E-11	3.43E-12	2.27E-12	5.19E-12
PCB201	7.62	5.22E-14	8.37E-15	3.26E-13	1.66E-14	6.75E-15	4.09E-14
PCB203	7.65	4.56E-14	1.23E-14	1.69E-13	2.33E-14	6.89E-15	7.92E-14
PCB195	7.56	1.00E-13	2.47E-14	4.05E-13	1.97E-14	7.41E-15	5.25E-14
PCB194	7.8	2.52E-12	9.12E-13	6.97E-12	5.50E-13	3.25E-13	9.32E-13
PCB206	8.09	4.74E-13	1.43E-13	1.57E-12	7.92E-14	3.81E-14	1.65E-13
PCB189	7.71	1.90E-13	2.50E-14	1.45E-12	2.20E-14	5.88E-15	8.22E-14
PCB77	6.36	2.18E-13	2.01E-13	2.37E-13	2.10E-13	2.07E-13	2.12E-13
PCB126	6.89	6.44E-14	5.94E-14	6.99E-14	6.19E-14	6.11E-14	6.27E-14
PCB169	7.42	1.90E-14	1.75E-14	2.06E-14	1.83E-14	1.80E-14	1.85E-14
Total PCBs	6.93	1.91E-10	6.36E-11	5.74E-10	6.22E-11	3.82E-11	1.01E-10

Appendix II continued

191	Bathurst	logK _{ow}	lichen(summer)			lichen(spring)		
			n=12			n=6		
			GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
	1,2,4,5 TCB	4.7	4.81E-10	2.17E-10	1.07E-09	1.71E-09	1.43E-09	2.04E-09
	1,2,3,4TCB	4.46	1.43E-09	5.58E-10	3.66E-09	9.86E-09	4.09E-09	2.38E-08
	QCB	5.03	6.44E-10	2.10E-10	1.97E-09	3.02E-09	2.43E-09	3.76E-09
	alpha-HCH	4	1.98E-09	1.20E-09	3.27E-09	4.00E-09	2.92E-09	5.46E-09
	beta-HCH	4	2.31E-11	1.02E-11	5.22E-11	1.69E-11	3.78E-12	7.55E-11
	gamma-HCH	4.5	2.02E-10	9.77E-11	4.18E-10	3.02E-10	2.60E-10	3.52E-10
	HCB	5.5	8.74E-10	4.32E-10	1.77E-09	6.17E-09	2.62E-09	1.45E-08
	OCS	6.9	2.43E-13	1.10E-13	5.37E-13	1.10E-12	9.21E-13	1.32E-12
	Oxychlordan	6.9	2.30E-13	1.02E-13	5.20E-13	1.10E-12	9.21E-13	1.32E-12
	Transchlordan	6.9	2.80E-13	1.21E-13	6.50E-13	1.10E-12	9.21E-13	1.32E-12
	Cischlordan	6.9	2.28E-13	9.10E-14	5.70E-13	7.66E-13	6.41E-13	9.15E-13
	Transnonachlor	6.9	2.11E-13	8.13E-14	5.48E-13	9.22E-13	7.24E-13	1.18E-12
	Cisnonachlor	6.9	1.27E-13	6.14E-14	2.64E-13	3.58E-13	2.99E-13	4.27E-13
	p,p' DDE	6.9	1.32E-13	5.12E-14	3.40E-13	6.91E-12	4.58E-12	1.04E-11
	p,p' DDD	6.9	8.11E-13	2.77E-13	2.37E-12	5.24E-13	1.27E-13	2.16E-12
	p,p' DDT	6	1.58E-11	3.21E-12	7.73E-11	2.85E-11	1.47E-11	5.50E-11
	Photomirex	6	2.36E-12	1.05E-12	5.34E-12	1.13E-11	9.46E-12	1.35E-11
	Mirex	6	1.70E-12	7.51E-13	3.83E-12	8.11E-12	6.78E-12	9.69E-12
	Heptachlor epoxide	6	4.16E-12	1.86E-12	9.29E-12	NA	NA	NA
	Dieldrin	6.2	2.32E-12	6.83E-13	7.86E-12	NA	NA	NA
	PCB31	5.6	7.95E-12	3.52E-12	1.80E-11	3.80E-11	3.18E-11	4.54E-11
	PCB28	5.6	7.95E-12	3.52E-12	1.80E-11	3.80E-11	3.18E-11	4.54E-11
	PCB52	5.84	7.40E-12	2.89E-12	1.89E-11	1.28E-10	4.90E-11	3.33E-10
	PCB49	5.85	4.57E-12	1.96E-12	1.07E-11	2.14E-11	1.79E-11	2.56E-11
	PCB44	5.75	5.63E-12	2.49E-12	1.27E-11	3.38E-11	1.95E-11	5.86E-11
	PCB42	5.76	5.50E-12	2.44E-12	1.24E-11	2.63E-11	2.20E-11	3.14E-11
	PCB64	5.95	3.55E-12	1.57E-12	8.02E-12	1.70E-11	1.42E-11	2.03E-11
	PCB74	6.2	2.61E-12	1.10E-12	6.20E-12	9.55E-12	7.99E-12	1.14E-11
	PCB70	6.2	4.26E-12	1.84E-12	9.89E-12	2.74E-11	8.20E-12	9.17E-11
	PCB66/95	6.2	4.94E-12	2.34E-12	1.04E-11	8.28E-11	5.43E-11	1.26E-10
	PCB60	6.11	2.46E-12	1.09E-12	5.55E-12	4.90E-11	2.71E-11	8.88E-11
	PCB101	6.38	2.41E-12	9.63E-13	6.04E-12	1.28E-10	9.77E-11	1.68E-10
	PCB99	6.39	1.57E-12	7.04E-13	3.51E-12	4.91E-11	3.59E-11	6.70E-11

Appendix II continued

Bathurst

192

	logK _{ow}	lichen(summer)			lichen(spring)		
		GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
PCB97	6.29	1.63E-12	7.14E-13	3.73E-12	4.27E-11	2.99E-11	6.09E-11
PCB87	6.29	1.81E-12	7.64E-13	4.28E-12	1.64E-11	4.10E-12	6.60E-11
PCB110	6.48	1.33E-12	5.55E-13	3.19E-12	9.20E-11	6.93E-11	1.24E-10
PCB151	6.64	9.03E-13	3.96E-13	2.06E-12	1.36E-11	9.72E-12	1.91E-11
PCB149	6.67	1.03E-12	3.30E-13	3.24E-12	5.01E-11	3.64E-11	6.91E-11
PCB118	6.74	6.97E-13	3.37E-13	1.44E-12	6.86E-11	4.94E-11	9.52E-11
PCB146	6.89	3.65E-13	1.62E-13	8.25E-13	4.00E-12	2.48E-12	6.46E-12
PCB153	6.92	5.87E-13	2.59E-13	1.33E-12	4.25E-11	3.20E-11	5.64E-11
PCB105	6.65	6.35E-13	2.81E-13	1.43E-12	2.94E-11	2.06E-11	4.21E-11
PCB141	6.82	4.29E-13	1.90E-13	9.70E-13	1.97E-11	1.45E-11	2.69E-11
PCB138	6.83	8.21E-13	4.26E-13	1.58E-12	6.78E-11	4.98E-11	9.24E-11
PCB129	6.73	4.36E-13	1.93E-13	9.84E-13	4.16E-12	2.45E-12	7.08E-12
PCB182/187	7.2	1.64E-13	7.63E-14	3.54E-13	2.66E-12	2.28E-12	3.09E-12
PCB183	7	2.34E-13	1.04E-13	5.29E-13	1.85E-12	1.23E-12	2.77E-12
PCB185	7.11	1.82E-13	8.04E-14	4.10E-13	8.69E-13	7.27E-13	1.04E-12
PCB174	7.11	1.82E-13	8.04E-14	4.10E-13	2.13E-12	1.13E-12	4.04E-12
PCB171	7.11	1.82E-13	8.04E-14	4.10E-13	3.39E-12	1.27E-12	9.03E-12
PCB200	7.2	1.48E-13	6.54E-14	3.34E-13	9.30E-13	7.34E-13	1.18E-12
PCB172	7.33	1.09E-13	4.85E-14	2.47E-13	5.23E-13	4.38E-13	6.26E-13
PCB180	7.5	9.32E-14	4.10E-14	2.12E-13	2.72E-12	1.99E-12	3.72E-12
PCB170/190	7.46	7.86E-14	3.56E-14	1.74E-13	2.12E-12	1.48E-12	3.03E-12
PCB201	7.62	5.16E-14	2.28E-14	1.16E-13	4.06E-13	2.68E-13	6.16E-13
PCB203	7.65	4.81E-14	2.13E-14	1.09E-13	2.42E-13	2.17E-13	2.70E-13
PCB195	7.56	5.92E-14	2.62E-14	1.34E-13	2.83E-13	2.37E-13	3.38E-13
PCB194	7.8	3.41E-14	1.51E-14	7.70E-14	1.63E-13	1.36E-13	1.95E-13
PCB206	8.09	1.75E-14	7.73E-15	3.95E-14	8.35E-14	6.99E-14	9.99E-14
PCB189	7.71	NA	NA	NA	NA	NA	NA
PCB77	6.36	NA	NA	NA	NA	NA	NA
PCB126	6.89	NA	NA	NA	NA	NA	NA
PCB169	7.42	NA	NA	NA	NA	NA	NA
Total PCBs	6.93	1.24E-11	6.94E-12	2.22E-11	2.76E-10	2.09E-10	3.71E-10

Appendix II continued

	logK _{ow}	female caribou july n=5			male caribou july n=5			female caribou September n=6			male caribou September n=4		
		GM	SD	upper 1	GM	SD	upper 1	GM	SD	lower 1	GM	SD	upper 1
1,2,4,5 TCB	4.7	1.91E-09	1.09E-09	3.38E-09	4.15E-09	3.52E-09	4.90E-09	1.60E-09	1.12E-09	2.28E-09	2.15E-09	2.01E-09	2.30E-09
1,2,3,4TCB	4.46	7.28E-10	5.17E-10	1.03E-09	3.10E-10	2.16E-10	4.44E-10	3.17E-10	1.75E-10	5.74E-10	3.17E-10	1.12E-10	9.02E-10
QCB	5.03	1.22E-09	8.19E-10	1.82E-09	1.34E-09	9.37E-10	1.92E-09	9.76E-10	5.83E-10	1.63E-09	1.18E-09	9.37E-10	1.49E-09
alpha-HCH	4	4.73E-09	3.46E-09	6.46E-09	9.26E-09	2.85E-09	3.01E-08	2.32E-09	1.63E-09	3.31E-09	3.18E-09	2.35E-09	4.30E-09
beta-HCH	4	7.77E-10	5.77E-10	1.05E-09	1.02E-09	2.94E-10	3.56E-09	1.96E-10	1.01E-10	3.80E-10	1.85E-10	1.43E-10	2.40E-10
gamma-HCH	4.5	1.14E-10	7.31E-11	1.78E-10	2.78E-10	4.04E-11	1.91E-09	3.18E-11	2.04E-11	4.98E-11	4.29E-11	3.38E-11	5.43E-11
HCB	5.5	9.96E-09	6.34E-09	1.56E-08	1.11E-07	2.80E-08	4.42E-07	8.99E-09	6.46E-09	1.25E-08	8.91E-09	7.36E-09	1.08E-08
OCS	6.9	7.79E-13	2.60E-13	2.33E-12	4.43E-11	9.46E-13	2.08E-09	1.35E-12	6.36E-13	2.85E-12	1.16E-12	7.71E-13	1.75E-12
Oxychloridane	6.9	3.36E-13	2.06E-13	5.49E-13	1.48E-11	2.73E-13	8.04E-10	5.31E-13	2.64E-13	1.07E-12	5.49E-13	9.72E-14	3.10E-12
Transchloridane	6.9	6.12E-13	1.83E-13	2.05E-12	2.30E-12	1.40E-14	3.76E-10	5.75E-13	1.90E-13	1.74E-12	1.41E-13	5.19E-14	3.81E-13
Cischloridane	6.9	2.91E-13	1.25E-13	6.78E-13	1.06E-12	7.11E-15	1.59E-10	3.48E-13	1.07E-13	1.14E-12	5.27E-14	4.27E-14	6.51E-14
Transnonachlor	6.9	8.77E-14	6.22E-14	1.24E-13	3.21E-13	5.11E-15	2.02E-11	3.32E-14	2.31E-14	4.78E-14	2.46E-14	1.99E-14	3.04E-14
Cisnonachlor	6.9	2.30E-12	6.38E-13	8.31E-12	1.91E-11	3.03E-13	1.20E-09	9.42E-13	2.76E-13	3.21E-12	3.17E-13	6.70E-14	1.49E-12
p,p' DDE	6.9	3.49E-13	2.48E-13	4.92E-13	2.55E-12	1.28E-13	5.07E-11	1.15E-13	8.35E-14	1.59E-13	9.79E-14	7.93E-14	1.21E-13
p,p' DDD	6	2.77E-12	1.97E-12	3.91E-12	9.93E-12	1.65E-13	5.97E-10	9.14E-13	6.64E-13	1.28E-12	7.78E-13	6.30E-13	9.61E-13
p,p' DDT	6	2.77E-12	1.97E-12	3.91E-12	8.81E-12	2.82E-13	2.76E-10	9.14E-13	6.64E-13	1.28E-12	9.69E-13	5.79E-13	1.62E-12
Photomirex	6	2.62E-12	1.56E-12	4.40E-12	1.50E-11	3.54E-13	6.35E-10	9.39E-13	3.94E-13	2.24E-12	1.27E-12	3.61E-13	4.43E-12
Mirex	6	4.55E-12	1.73E-12	1.20E-11	2.10E-11	8.60E-13	5.11E-10	1.26E-12	6.22E-13	2.56E-12	4.22E-12	1.50E-12	1.18E-11
Heptachlor epoxide	6.2	1.75E-12	1.24E-12	2.46E-12	8.52E-12	1.66E-13	4.38E-10	5.77E-13	4.19E-13	7.95E-13	4.91E-13	3.97E-13	6.06E-13
Dieldrin	5.6	4.12E-11	5.28E-12	3.22E-10	1.14E-11	1.60E-12	8.16E-11	3.22E-11	4.26E-12	2.44E-10	2.55E-11	3.10E-12	2.09E-10
PCB31	5.6	9.32E-12	6.61E-12	1.31E-11	5.10E-11	1.68E-12	1.57E-09	3.08E-12	2.23E-12	4.24E-12	2.62E-12	2.12E-12	3.23E-12
PCB28	5.84	4.82E-11	2.68E-11	8.64E-11	9.68E-11	3.57E-12	2.63E-09	2.69E-11	1.25E-11	5.79E-11	7.93E-12	3.03E-12	2.08E-11
PCB52	5.85	1.07E-11	3.69E-12	3.12E-11	2.19E-11	5.53E-13	8.63E-10	4.31E-12	1.25E-12	1.48E-11	1.47E-12	1.19E-12	1.82E-12
PCB49	5.75	1.55E-11	4.82E-12	5.21E-11	4.04E-11	1.01E-12	1.61E-09	1.53E-11	2.92E-12	8.02E-11	7.93E-12	2.04E-12	3.09E-11
PCB44	5.76	6.45E-12	4.58E-12	9.09E-12	1.66E-11	5.38E-13	5.04E-10	3.50E-12	1.23E-12	9.97E-12	1.81E-12	1.47E-12	2.24E-12
PCB42	5.95	4.16E-12	2.95E-12	5.87E-12	1.05E-11	3.52E-13	3.14E-10	1.37E-12	9.97E-13	1.89E-12	1.72E-12	6.02E-13	4.94E-12
PCB64	6.2	7.01E-12	3.25E-12	1.51E-11	7.75E-11	3.64E-12	1.65E-09	4.85E-12	1.38E-12	1.71E-11	2.94E-12	1.31E-12	6.60E-12
PCB74	6.2	9.34E-12	2.53E-12	3.44E-11	2.63E-11	5.43E-13	1.27E-09	5.41E-12	1.28E-12	2.28E-11	1.77E-12	6.73E-13	4.64E-12
PCB70	6.2	2.44E-11	1.32E-11	4.49E-11	8.03E-11	2.61E-12	2.47E-09	1.35E-11	5.66E-12	3.24E-11	7.15E-12	3.90E-12	1.31E-11
PCB66/95	6.11	5.53E-12	1.51E-12	2.03E-11	3.68E-11	1.85E-12	7.30E-10	9.51E-13	6.90E-13	1.31E-12	9.29E-13	5.64E-13	1.53E-12
PCB60	6.38	1.11E-11	5.67E-12	2.16E-11	4.13E-11	9.86E-13	1.73E-09	7.22E-12	2.85E-12	1.03E-11	2.33E-12	8.98E-13	6.04E-12

Appendix II continued

Bathurst		female caribou July n=5				male caribou July n=5			female caribou September n=6			male caribou September n=4			
		logK _{ow}	GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD	
PCB99	6.39	9.00E-12	6.72E-12	1.21E-11	8.50E-11	3.46E-12	2.09E-09	4.57E-12	1.87E-12	1.12E-11	4.52E-12	2.33E-12	8.75E-12		
PCB97	6.29	2.65E-12	1.14E-12	6.15E-12	6.02E-12	1.03E-13	3.51E-10	1.68E-12	3.94E-13	7.18E-12	4.79E-13	3.87E-13	5.91E-13		
PCB87	6.29	5.33E-12	1.71E-12	1.66E-11	1.14E-11	1.88E-13	6.96E-10	3.91E-12	9.48E-13	1.62E-11	9.04E-13	3.15E-13	2.59E-12		
PCB110	6.48	6.22E-12	1.89E-12	2.04E-11	1.65E-11	3.09E-13	8.75E-10	2.36E-12	6.37E-13	8.72E-12	1.03E-12	4.35E-13	2.46E-12		
PCB151	6.64	1.18E-12	5.08E-13	2.75E-12	4.04E-12	7.56E-14	2.16E-10	1.29E-12	3.27E-13	5.09E-12	2.82E-13	1.26E-13	6.30E-13		
PCB149	6.67	1.54E-12	6.06E-13	3.92E-12	3.50E-12	3.09E-14	3.95E-10	1.94E-12	4.68E-13	8.05E-12	2.63E-13	1.18E-13	5.88E-13		
PCB118	6.74	6.62E-12	4.83E-12	9.08E-12	8.28E-11	2.42E-12	2.83E-09	4.44E-12	2.29E-12	8.61E-12	2.68E-12	1.71E-12	4.21E-12		
PCB146	6.89	6.64E-13	4.36E-13	1.01E-12	5.83E-12	9.02E-14	3.78E-10	6.83E-13	2.34E-13	2.00E-12	2.46E-13	1.51E-13	4.00E-13		
PCB153	6.92	6.44E-12	4.20E-12	9.88E-12	1.16E-10	2.81E-12	4.75E-09	5.11E-12	2.27E-12	1.15E-11	3.31E-12	2.26E-12	4.84E-12		
PCB105	6.65	1.22E-12	7.37E-13	2.03E-12	2.03E-11	6.93E-13	5.94E-10	7.19E-13	2.26E-13	2.29E-12	3.64E-13	1.97E-13	6.73E-13		
194 PCB141	6.82	5.78E-13	4.23E-13	7.89E-13	1.64E-12	3.30E-14	8.15E-11	6.29E-13	2.52E-13	1.57E-12	1.41E-13	1.14E-13	1.74E-13		
PCB138	6.83	6.00E-12	4.32E-12	8.35E-12	6.72E-11	1.67E-12	2.70E-09	4.12E-12	1.78E-12	9.53E-12	2.46E-12	1.86E-12	3.24E-12		
PCB129	6.73	5.11E-13	3.62E-13	7.20E-13	1.33E-11	3.38E-13	5.25E-10	2.22E-13	1.10E-13	4.49E-13	1.66E-13	1.15E-13	2.35E-13		
PCB182/187	7.2	4.98E-13	1.43E-13	1.74E-12	1.67E-11	2.91E-13	9.57E-10	1.15E-12	5.35E-13	2.48E-12	3.56E-13	9.72E-14	1.30E-12		
PCB183	7	4.16E-13	1.88E-13	9.21E-13	4.10E-12	7.30E-14	2.31E-10	5.03E-13	1.61E-13	1.57E-12	1.19E-13	7.27E-14	1.96E-13		
PCB185	7.11	2.13E-13	1.51E-13	3.00E-13	6.23E-13	1.55E-14	2.50E-11	7.03E-14	5.10E-14	9.68E-14	5.98E-14	4.84E-14	7.38E-14		
PCB174	7.11	2.65E-13	1.75E-13	4.02E-13	9.75E-13	9.95E-15	9.55E-11	1.33E-13	6.41E-14	2.75E-13	5.98E-14	4.84E-14	7.38E-14		
PCB171	7.11	2.13E-13	1.51E-13	3.00E-13	3.86E-12	5.38E-14	2.77E-10	2.17E-13	8.50E-14	5.53E-13	1.07E-13	5.44E-14	2.09E-13		
PCB200	7.2	1.73E-13	1.23E-13	2.44E-13	9.93E-13	1.33E-14	7.40E-11	5.71E-14	4.14E-14	7.87E-14	4.86E-14	3.93E-14	6.00E-14		
PCB172	7.33	1.28E-13	9.10E-14	1.81E-13	1.07E-12	8.62E-15	1.33E-10	4.23E-14	3.07E-14	5.83E-14	4.97E-14	2.73E-14	9.05E-14		
PCB180	7.5	7.58E-13	5.37E-13	1.07E-12	1.73E-11	2.00E-13	1.50E-09	6.23E-13	2.45E-13	1.58E-12	3.99E-13	3.40E-13	4.68E-13		
PCB170/190	7.46	3.09E-13	1.28E-13	7.43E-13	1.01E-11	1.29E-13	7.97E-10	2.35E-13	5.80E-14	9.55E-13	2.64E-13	2.08E-13	3.35E-13		
PCB201	7.62	3.18E-13	1.61E-13	6.29E-13	7.47E-12	8.35E-14	6.69E-10	1.49E-13	3.46E-14	6.39E-13	1.51E-13	1.06E-13	2.15E-13		
PCB203	7.65	5.64E-14	4.00E-14	7.96E-14	2.25E-13	3.02E-15	1.68E-11	1.86E-14	1.35E-14	2.57E-14	1.58E-14	1.28E-14	1.96E-14		
PCB195	7.56	8.65E-14	5.70E-14	1.31E-13	1.12E-12	9.38E-15	1.35E-10	6.19E-14	1.84E-14	2.08E-13	2.43E-14	1.45E-14	4.06E-14		
PCB194	7.8	1.10E-13	4.72E-14	2.58E-13	3.62E-12	2.75E-14	4.78E-10	3.79E-14	6.83E-15	2.10E-13	2.79E-14	9.03E-15	8.64E-14		
PCB206	8.09	2.05E-14	1.45E-14	2.89E-14	1.05E-13	8.53E-16	1.30E-11	6.76E-15	4.90E-15	9.31E-15	5.75E-15	4.65E-15	7.10E-15		
PCB189	7.71	7.03E-14	3.31E-14	1.49E-13	3.52E-13	1.47E-15	8.41E-11	2.82E-14	8.75E-15	9.11E-14	1.38E-14	1.12E-14	1.70E-14		
PCB77	6.36	1.10E-12	7.81E-13	1.55E-12	2.09E-12	1.23E-13	3.55E-11	3.63E-13	2.63E-13	5.00E-13	3.09E-13	2.50E-13	3.81E-13		
PCB126	6.89	3.25E-13	2.30E-13	4.57E-13	7.02E-12	1.69E-13	2.92E-10	1.82E-13	5.32E-14	6.20E-13	9.11E-14	7.38E-14	1.13E-13		
PCB169	7.42	2.44E-13	1.07E-13	5.59E-13	6.23E-12	8.81E-14	4.41E-10	2.32E-13	5.85E-14	9.17E-13	1.71E-13	9.49E-14	3.08E-13		
Total PCBs	6.93	4.80E-11	2.40E-11	5.91E-11	4.75E-10	7.84E-12	2.32E-08	3.08E-11	1.00E-11	7.31E-11	1.71E-11	7.86E-12	2.61E-11		

Appendix II continued

Bathurst	logK _{ow}	female wolves n=6			male wolves n=4		
		GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
1,2,4,5 TCB	4.7	1.19E-08	6.75E-09	2.10E-08	1.55E-08	1.16E-08	2.07E-08
1,2,3,4TCB	4.46	1.35E-09	4.59E-10	3.98E-09	5.48E-10	1.30E-10	2.30E-09
QCB	5.03	5.35E-09	3.64E-09	7.85E-09	9.06E-09	5.41E-09	1.52E-08
alpha-HCH	4	3.78E-09	2.42E-09	5.89E-09	8.09E-09	2.80E-09	2.34E-08
beta-HCH	4	5.66E-09	4.24E-09	7.57E-09	1.35E-08	3.98E-09	4.55E-08
gamma-HCH	4.5	3.86E-12	1.13E-12	1.32E-11	7.26E-12	1.39E-12	3.78E-11
HCB	5.5	5.28E-08	3.47E-08	8.03E-08	1.23E-07	3.75E-08	4.05E-07
OCS	6.9	9.42E-12	5.14E-12	1.73E-11	8.12E-11	2.79E-12	2.36E-09
Oxychlorane	6.9	2.85E-11	1.19E-11	6.82E-11	3.50E-10	1.42E-11	8.62E-09
Transchlorane	6.9	1.64E-13	4.49E-14	5.98E-13	2.24E-12	4.91E-14	1.02E-10
Cischlorane	6.9	5.23E-14	2.47E-14	1.11E-13	1.04E-12	1.64E-14	6.62E-11
Transnonachlor	6.9	3.39E-13	9.74E-14	1.18E-12	1.08E-11	5.59E-13	2.08E-10
Cisnonachlor	6.9	2.28E-14	1.29E-14	4.02E-14	6.33E-13	1.50E-14	2.68E-11
p,p' DDE	6.9	2.36E-14	1.25E-14	4.44E-14	1.39E-13	2.53E-15	7.67E-12
p,p' DDD	6.9	7.21E-14	6.96E-14	7.47E-14	5.18E-13	2.14E-14	1.26E-11
p,p' DDT	6	5.73E-13	5.53E-13	5.93E-13	5.21E-12	4.00E-13	6.78E-11
Photomirex	6	2.05E-11	9.76E-12	4.32E-11	1.19E-10	1.58E-11	8.97E-10
Mirex	6	2.11E-11	9.34E-12	4.79E-11	1.22E-10	1.40E-11	1.06E-09
Heptachlor epoxide	6	6.38E-11	3.14E-11	1.30E-10	3.72E-10	4.50E-11	3.08E-09
Dieldrin	6.2	3.61E-13	3.49E-13	3.74E-13	3.23E-12	2.40E-13	4.35E-11
PCB31	5.6	1.93E-12	1.86E-12	2.00E-12	4.98E-12	8.07E-13	3.07E-11
PCB28	5.6	7.49E-12	2.55E-12	2.20E-11	5.07E-11	8.32E-12	3.09E-10
PCB52	5.84	1.11E-12	1.07E-12	1.15E-12	1.68E-11	5.06E-13	5.59E-10
PCB49	5.85	1.08E-12	1.05E-12	1.12E-12	6.56E-12	7.59E-13	5.68E-11
PCB44	5.75	1.36E-12	1.32E-12	1.41E-12	3.84E-12	5.24E-13	2.82E-11
PCB42	5.76	1.33E-12	1.29E-12	1.38E-12	3.78E-12	5.09E-13	2.80E-11
PCB64	5.95	8.60E-13	8.30E-13	8.91E-13	2.72E-12	2.95E-13	2.51E-11
PCB74	6.2	2.35E-11	1.59E-11	3.46E-11	1.63E-10	1.38E-11	1.93E-09
PCB70	6.2	6.52E-13	3.21E-13	1.32E-12	4.26E-12	3.20E-13	5.68E-11
PCB66/95	6.2	7.68E-13	2.54E-13	2.33E-12	1.87E-11	7.02E-13	5.00E-10
PCB60	6.11	2.16E-12	4.99E-13	9.32E-12	2.19E-11	1.88E-12	2.55E-10
PCB101	6.38	3.20E-13	3.09E-13	3.31E-13	6.10E-12	1.13E-13	3.28E-10
PCB99	6.39	2.86E-11	1.51E-11	5.42E-11	2.86E-10	2.41E-11	3.39E-09

195

Appendix II continued

Bathurst

female wolves n=6

male wolves n=4

196

	logK _{ow}	female wolves n=6			male wolves n=4		
		GM	lower 1 SD	upper 1 SD	GM	lower 1 SD	upper 1 SD
PCB97	6.29	3.52E-13	3.40E-13	3.65E-13	1.35E-12	9.92E-14	1.85E-11
PCB87	6.29	3.52E-13	3.40E-13	3.65E-13	1.35E-12	9.92E-14	1.85E-11
PCB110	6.48	2.27E-13	2.20E-13	2.36E-13	1.92E-12	2.92E-14	1.26E-10
PCB151	6.64	2.22E-13	9.32E-14	5.31E-13	2.85E-12	6.43E-14	1.26E-10
PCB149	6.67	1.47E-13	1.42E-13	1.52E-13	7.03E-13	3.32E-14	1.49E-11
PCB118	6.74	1.36E-11	4.27E-12	4.34E-11	4.06E-10	1.49E-11	1.10E-08
PCB146	6.89	5.82E-13	1.97E-13	1.72E-12	7.57E-12	1.98E-13	2.89E-10
PCB153	6.92	5.41E-11	2.98E-11	9.82E-11	7.49E-10	3.13E-11	1.79E-08
PCB105	6.65	5.77E-13	7.26E-14	4.59E-12	3.83E-11	4.43E-13	3.31E-09
PCB141	6.82	1.04E-13	1.00E-13	1.08E-13	5.42E-13	2.16E-14	1.36E-11
PCB138	6.83	9.12E-12	6.06E-12	1.37E-11	1.28E-10	5.81E-12	2.83E-09
PCB129	6.73	1.06E-13	1.02E-13	1.09E-13	1.43E-12	9.61E-14	2.13E-11
PCB182/187	7.2	7.13E-14	2.40E-14	2.12E-13	5.19E-13	1.56E-14	1.73E-11
PCB183	7	1.92E-12	8.70E-13	4.25E-12	2.41E-11	1.07E-12	5.39E-10
PCB185	7.11	4.40E-14	4.25E-14	4.56E-14	2.71E-13	7.73E-15	9.51E-12
PCB174	7.11	4.40E-14	4.25E-14	4.56E-14	4.41E-13	1.50E-14	1.30E-11
PCB171	7.11	1.30E-12	2.00E-13	8.43E-12	4.51E-11	1.42E-12	1.43E-09
PCB200	7.2	1.93E-13	4.95E-14	7.52E-13	1.29E-11	5.77E-13	2.87E-10
PCB172	7.33	2.01E-13	1.14E-13	3.54E-13	1.32E-12	2.07E-14	8.41E-11
PCB180	7.5	2.47E-11	1.15E-11	5.27E-11	2.81E-10	8.18E-12	9.64E-09
PCB170/190	7.46	1.70E-11	8.46E-12	3.40E-11	1.54E-10	4.84E-12	4.91E-09
PCB201	7.62	1.62E-13	4.33E-14	6.07E-13	1.68E-12	1.07E-14	2.63E-10
PCB203	7.65	3.76E-14	6.01E-15	2.35E-13	4.05E-13	1.01E-14	1.62E-11
PCB195	7.56	1.00E-13	3.04E-14	3.30E-13	2.88E-12	4.73E-14	1.75E-10
PCB194	7.8	2.08E-12	8.02E-13	5.40E-12	2.13E-11	5.06E-13	8.93E-10
PCB206	8.09	4.23E-15	4.09E-15	4.38E-15	4.59E-14	4.23E-16	4.97E-12
PCB189	7.71	1.02E-14	9.80E-15	1.05E-14	1.82E-13	3.71E-15	8.93E-12
PCB77	6.36	2.27E-13	2.19E-13	2.35E-13	1.53E-12	1.17E-13	2.00E-11
PCB126	6.89	1.86E-12	8.11E-13	4.28E-12	1.14E-11	6.83E-13	1.91E-10
PCB169	7.42	9.85E-12	3.87E-12	2.50E-11	8.72E-11	3.56E-12	2.14E-09
Total PCBs	6.93	2.43E-10	1.29E-10	4.46E-10	2.45E-09	1.06E-10	5.76E-08

Appendix II continued

Inuvik	logK _{ow}	lichen n=9		female caribou n=2			male caribou n=8			wolves (Age and Sex unknown) n=10				
		GM	lower 1	upper 1	GM	lower 1	upper 1	GM	lower 1	upper 1	GM	lower 1	upper 1	
			SD	SD		SD	SD		SD	SD		SD		
1,2,4,5 TCB	4.7	1.92E-10	1.92E-10	1.92E-10	1.05E-10	1.05E-10	1.05E-10	1.04E-10	1.02E-10	1.06E-10	1.13E-08	5.75E-09	2.22E-08	
1,2,3,4TCB	4.46	2.32E-09	1.80E-09	3.00E-09	1.83E-10	1.82E-10	1.83E-10	1.81E-10	1.78E-10	1.84E-10	9.85E-10	3.68E-10	2.63E-09	
QCB	5.03	1.53E-09	1.12E-09	2.09E-09	1.28E-09	1.12E-09	1.46E-09	1.64E-09	1.04E-09	2.58E-09	4.99E-09	2.83E-09	8.81E-09	
alpha-HCH	4	1.57E-09	6.89E-10	3.57E-09	1.28E-09	1.01E-09	1.62E-09	1.70E-09	1.33E-09	2.16E-09	2.97E-09	1.70E-09	5.19E-09	
beta-HCH	4	1.24E-11	1.24E-11	1.24E-11	1.73E-10	1.31E-10	2.29E-10	1.37E-10	1.08E-10	1.74E-10	4.03E-09	1.97E-09	8.25E-09	
gamma-HCH	4.5	1.56E-10	7.73E-11	3.13E-10	6.45E-12	1.36E-12	3.05E-11	3.91E-12	1.26E-12	1.21E-11	6.58E-12	1.83E-12	2.37E-11	
HCB	5.5	1.19E-09	7.66E-10	1.85E-09	2.16E-08	1.44E-08	3.24E-08	2.23E-08	1.71E-08	2.90E-08	4.68E-08	2.48E-08	8.83E-08	
OCS	6.9	1.24E-13	1.24E-13	1.24E-13	6.91E-13	4.91E-13	9.72E-13	1.01E-12	7.08E-13	1.44E-12	6.61E-12	2.91E-12	1.50E-11	
Oxychlorane	6.9	1.56E-13	8.49E-14	2.87E-13	6.77E-14	6.76E-14	6.78E-14	1.65E-13	4.68E-14	5.80E-13	2.18E-11	9.26E-12	5.13E-11	
Transchlorane	6.9	1.24E-13	1.24E-13	1.24E-13	6.77E-14	6.76E-14	6.78E-14	6.70E-14	6.58E-14	6.81E-14	1.68E-13	3.81E-14	7.38E-13	
Cischlorane	6.9	2.64E-13	9.14E-14	7.62E-13	4.71E-14	4.70E-14	4.72E-14	4.66E-14	4.58E-14	4.74E-14	4.71E-14	4.56E-14	4.86E-14	
Transnonachlor	6.9	2.13E-13	6.86E-14	6.61E-13	4.71E-14	4.70E-14	4.72E-14	4.66E-14	4.58E-14	4.74E-14	5.00E-13	1.03E-13	2.43E-12	
Cisnonachlor	6.9	4.03E-14	4.03E-14	4.03E-14	2.20E-14	2.20E-14	2.20E-14	2.17E-14	2.14E-14	2.21E-14	3.01E-14	1.13E-14	8.02E-14	
p,p' DDE	6.9	1.21E-13	4.10E-14	3.56E-13	2.19E-14	2.18E-14	2.19E-14	2.16E-14	2.13E-14	2.20E-14	6.01E-13	9.84E-14	3.67E-12	
p,p' DDD	6.9	2.85E-13	1.07E-13	7.61E-13	8.75E-14	8.74E-14	8.77E-14	8.65E-14	8.51E-14	8.80E-14	1.14E-13	4.99E-14	2.60E-13	
p,p' DDT	6	4.43E-12	9.35E-13	2.10E-11	6.95E-13	6.94E-13	6.96E-13	6.87E-13	6.76E-13	6.99E-13	8.83E-13	4.18E-13	1.87E-12	
Photomirex	6	1.27E-12	1.27E-12	1.27E-12	6.95E-13	6.94E-13	6.96E-13	6.87E-13	6.76E-13	6.99E-13	1.94E-11	7.85E-12	4.77E-11	
Mirex	6	9.13E-13	9.13E-13	9.13E-13	4.99E-13	4.98E-13	5.00E-13	4.93E-13	4.85E-13	5.02E-13	1.84E-11	8.57E-12	3.94E-11	
Heptachlor epoxide	6	2.16E-12	8.68E-13	5.36E-12	6.95E-13	6.94E-13	6.96E-13	6.87E-13	6.76E-13	6.99E-13	7.48E-11	3.77E-11	1.48E-10	
Dieldrin	6.2	2.55E-12	8.35E-13	7.78E-12	4.39E-13	4.38E-13	4.39E-13	4.34E-13	4.26E-13	4.41E-13	1.17E-12	2.28E-13	6.00E-12	
PCB31	5.6	4.28E-12	4.28E-12	4.28E-12	2.34E-12	2.33E-12	2.34E-12	2.31E-12	2.27E-12	2.35E-12	2.34E-12	2.26E-12	2.41E-12	
PCB28	5.6	4.28E-12	4.28E-12	4.28E-12	2.34E-12	2.33E-12	2.34E-12	2.31E-12	2.27E-12	2.35E-12	2.34E-12	2.26E-12	2.41E-12	
PCB52	5.84	8.15E-12	2.63E-12	2.52E-11	1.35E-12	1.34E-12	1.35E-12	1.33E-12	1.31E-12	1.35E-12	6.12E-12	1.50E-12	2.50E-11	
PCB49	5.85	2.41E-12	2.41E-12	2.41E-12	1.31E-12	1.31E-12	1.32E-12	1.30E-12	1.28E-12	1.32E-12	1.31E-12	1.27E-12	1.36E-12	
PCB44	5.75	3.03E-12	3.03E-12	3.03E-12	1.66E-12	1.65E-12	1.66E-12	1.64E-12	1.61E-12	1.66E-12	2.23E-12	8.66E-13	5.75E-12	
PCB42	5.76	2.96E-12	2.96E-12	2.96E-12	1.62E-12	1.61E-12	1.62E-12	1.60E-12	1.57E-12	1.63E-12	2.17E-12	8.56E-13	5.50E-12	
PCB64	5.95	1.91E-12	1.91E-12	1.91E-12	1.04E-12	1.04E-12	1.05E-12	1.03E-12	1.02E-12	1.05E-12	1.04E-12	1.01E-12	1.08E-12	
PCB74	6.2	1.08E-12	1.08E-12	1.08E-12	5.87E-13	5.86E-13	5.88E-13	5.81E-13	5.71E-13	5.91E-13	2.57E-11	1.37E-11	4.80E-11	
PCB70	6.2	1.39E-12	7.06E-13	2.73E-12	5.87E-13	5.86E-13	5.88E-13	5.81E-13	5.71E-13	5.91E-13	8.37E-13	3.97E-13	1.77E-12	
PCB66/95	6.2	2.89E-12	1.13E-12	7.37E-12	5.87E-13	5.86E-13	5.88E-13	5.81E-13	5.71E-13	5.91E-13	2.38E-12	5.39E-13	1.05E-11	
PCB60	6.11	1.32E-12	1.32E-12	1.32E-12	7.23E-13	7.21E-13	7.24E-13	7.14E-13	7.02E-13	7.27E-13	7.22E-13	6.99E-13	7.46E-13	
PCB101	6.38	1.18E-12	4.93E-13	2.83E-12	3.88E-13	3.87E-13	3.89E-13	3.84E-13	3.77E-13	3.90E-13	1.49E-12	4.21E-13	5.28E-12	
PCB99	6.39	8.03E-13	4.08E-13	1.58E-12	3.40E-13	3.39E-13	3.40E-13	5.73E-13	2.08E-13	1.58E-12	2.85E-11	1.35E-11	6.02E-11	

Appendix II continued

Inuvik	logK _{ow}	lichen n=9			female caribou n=2			male caribou n=8			wolves (Age and Sex unknown) n=10		
		GM	lower 1	upper 1	GM	lower 1	upper 1	GM	lower 1	upper 1	GM	lower 1	upper 1
			SD	SD		SD	SD		SD	SD		SD	
PCB97	6.29	7.83E-13	7.83E-13	7.83E-13	4.28E-13	4.27E-13	4.28E-13	4.23E-13	4.16E-13	4.30E-13	4.27E-13	4.14E-13	4.41E-13
PCB87	6.29	7.83E-13	7.83E-13	7.83E-13	4.28E-13	4.27E-13	4.28E-13	4.23E-13	4.16E-13	4.30E-13	5.70E-13	2.31E-13	1.41E-12
PCB110	6.48	5.06E-13	5.06E-13	5.06E-13	2.76E-13	2.76E-13	2.77E-13	2.73E-13	2.68E-13	2.78E-13	3.62E-13	1.55E-13	8.43E-13
PCB151	6.64	3.50E-13	3.50E-13	3.50E-13	1.91E-13	1.91E-13	1.91E-13	1.89E-13	1.86E-13	1.92E-13	1.91E-13	1.85E-13	1.97E-13
PCB149	6.67	5.31E-13	2.31E-13	1.22E-12	1.78E-13	1.78E-13	1.79E-13	1.76E-13	1.73E-13	1.79E-13	1.78E-13	1.72E-13	1.84E-13
PCB118	6.74	2.78E-13	2.78E-13	2.78E-13	6.78E-13	8.17E-14	5.63E-12	2.15E-13	7.66E-14	6.05E-13	2.96E-11	1.12E-11	7.85E-11
PCB146	6.89	1.97E-13	1.97E-13	1.97E-13	1.07E-13	1.07E-13	1.08E-13	1.06E-13	1.04E-13	1.08E-13	6.45E-13	1.57E-13	2.65E-12
PCB153	6.92	3.06E-13	1.28E-13	7.34E-13	1.60E-12	1.47E-12	1.75E-12	1.77E-12	1.26E-12	2.47E-12	4.93E-11	2.02E-11	1.20E-10
PCB105	6.65	3.42E-13	3.42E-13	3.42E-13	4.57E-13	1.29E-13	1.62E-12	2.93E-13	1.24E-13	6.89E-13	5.00E-12	1.84E-12	1.36E-11
PCB141	6.82	2.31E-13	2.31E-13	2.31E-13	1.26E-13	1.26E-13	1.26E-13	1.25E-13	1.23E-13	1.27E-13	1.26E-13	1.22E-13	1.30E-13
PCB138	6.83	3.39E-13	1.42E-13	8.11E-13	1.56E-12	1.56E-12	1.56E-12	1.86E-12	1.50E-12	2.32E-12	1.40E-11	6.16E-12	3.20E-11
PCB129	6.73	2.35E-13	2.35E-13	2.35E-13	1.28E-13	1.28E-13	1.28E-13	1.27E-13	1.25E-13	1.29E-13	2.71E-13	1.18E-13	6.23E-13
198 PCB182/187	7.2	7.95E-14	7.95E-14	7.95E-14	4.34E-14	4.33E-14	4.35E-14	4.29E-14	4.22E-14	4.37E-14	1.22E-13	2.75E-14	5.44E-13
PCB183	7	1.26E-13	1.26E-13	1.26E-13	6.88E-14	6.87E-14	6.89E-14	6.80E-14	6.69E-14	6.92E-14	1.43E-12	3.79E-13	5.38E-12
PCB185	7.11	9.78E-14	9.78E-14	9.78E-14	5.34E-14	5.33E-14	5.35E-14	5.28E-14	5.19E-14	5.37E-14	5.34E-14	5.17E-14	5.51E-14
PCB174	7.11	9.78E-14	9.78E-14	9.78E-14	5.34E-14	5.33E-14	5.35E-14	5.28E-14	5.19E-14	5.37E-14	5.34E-14	5.17E-14	5.51E-14
PCB171	7.11	9.78E-14	9.78E-14	9.78E-14	5.34E-14	5.33E-14	5.35E-14	5.28E-14	5.19E-14	5.37E-14	2.77E-12	8.28E-13	9.28E-12
PCB200	7.2	7.95E-14	7.95E-14	7.95E-14	4.34E-14	4.33E-14	4.35E-14	4.29E-14	4.22E-14	4.37E-14	5.41E-13	8.12E-14	3.60E-12
PCB172	7.33	5.89E-14	5.89E-14	5.89E-14	3.22E-14	3.21E-14	3.22E-14	3.18E-14	3.13E-14	3.24E-14	1.03E-13	3.56E-14	2.99E-13
PCB180	7.5	3.98E-14	3.98E-14	3.98E-14	1.74E-13	1.74E-13	1.74E-13	1.73E-13	7.21E-14	4.16E-13	1.89E-11	4.62E-12	7.70E-11
PCB170/190	7.46	4.01E-14	4.01E-14	4.01E-14	2.19E-14	2.19E-14	2.20E-14	2.17E-14	2.13E-14	2.20E-14	9.93E-12	3.34E-12	2.95E-11
PCB201	7.62	2.78E-14	2.78E-14	2.78E-14	1.52E-14	1.51E-14	1.52E-14	1.50E-14	1.47E-14	1.52E-14	1.20E-13	3.82E-14	3.79E-13
PCB203	7.65	2.59E-14	2.59E-14	2.59E-14	1.42E-14	1.41E-14	1.42E-14	1.40E-14	1.38E-14	1.42E-14	6.66E-14	1.98E-14	2.24E-13
PCB195	7.56	3.19E-14	3.19E-14	3.19E-14	1.74E-14	1.74E-14	1.74E-14	1.72E-14	1.69E-14	1.75E-14	1.64E-13	4.39E-14	6.13E-13
PCB194	7.8	1.83E-14	1.83E-14	1.83E-14	1.00E-14	1.00E-14	1.00E-14	9.91E-15	9.74E-15	1.01E-14	2.52E-12	6.54E-13	9.72E-12
PCB206	8.09	9.41E-15	9.41E-15	9.41E-15	5.14E-15	5.13E-15	5.15E-15	5.08E-15	5.00E-15	5.17E-15	5.67E-13	1.68E-13	1.91E-12
PCB189	7.71	2.26E-14	2.26E-14	2.26E-14	1.23E-14	1.23E-14	1.23E-14	1.22E-14	1.20E-14	1.24E-14	2.24E-13	5.69E-14	8.86E-13
PCB77	6.36	5.05E-13	5.05E-13	5.05E-13	2.76E-13	2.75E-13	2.76E-13	2.73E-13	2.68E-13	2.77E-13	2.76E-13	2.67E-13	2.85E-13
PCB126	6.89	1.49E-13	1.49E-13	1.49E-13	8.14E-14	8.13E-14	8.16E-14	8.05E-14	7.92E-14	8.19E-14	8.14E-14	7.88E-14	8.40E-14
PCB169	7.42	4.40E-14	4.40E-14	4.40E-14	2.40E-14	2.40E-14	2.41E-14	2.38E-14	2.34E-14	2.42E-14	2.40E-14	2.33E-14	2.48E-14
Total PCBs	6.93	6.40E-12	4.84E-12	8.45E-12	5.67E-12	2.10E-12	4.64E-12	5.63E-12	1.93E-12	4.28E-12	2.13E-10	7.34E-11	6.17E-10

APPENDIX III

Chemical concentrations (ng/g lipid) in individual caribou and wolves from Cambridge Bay, Bathurst Inlet, and Inuvik, used for model validation.

Appendix III: Chemical concentrations (ng/g lipid) and ages of individual caribou and wolves sampled from Cambridge Bay, Bathurst Inlet and Inuvik, Cambridge Bay

CHEMICAL	log K _{ow}	ng/g dry wt. Lichen SD	Female caribou (age in years)					Male caribou (age in years)					Female wolf n=5		Male wolf n=5	
			1.40	2.40	3.40	5.40	13.40	5.40	7.40	7.40	7.40	Mean	SD	Mean	SD	
			0.05	0.53	0.23	0.42	0.71	0.20	0.53	0.59	0.98	0.81	2.22	1.54	0.87	0.36
1,2,4,5 TCB	4.70	0.07	0.05	0.53	0.23	0.42	0.71	0.20	0.53	0.59	0.98	0.81	2.22	1.54	0.87	0.36
1,2,3,4TCB	4.46	0.10	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.05	0.06
QCB	5.03	0.25	0.16	1.05	0.62	0.49	0.75	0.41	0.85	0.91	0.86	1.28	2.27	1.38	0.85	0.23
alpha-HCH	4.00	4.42	3.56	22.36	12.75	12.06	4.85	7.21	18.69	11.83	10.46	27.04	3.87	1.55	3.08	1.05
beta-HCH	4.00	0.09	0.07	0.60	0.63	0.55	0.67	0.42	0.66	1.11	0.70	0.85	9.94	8.10	2.27	0.47
gamma-HCH	4.50	0.74	0.39	0.68	0.48	0.48	0.34	0.25	0.52	0.50	0.42	0.78	0.01	0.00	0.06	0.08
HCB	5.50	1.41	0.57	36.67	39.69	28.43	19.69	15.88	51.55	48.00	75.15	67.00	33.27	13.21	22.65	3.83
OCS	6.90	0.08	0.03	0.69	0.60	0.25	0.11	0.12	0.76	0.49	0.73	1.04	1.71	0.73	0.69	0.40
Oxychlorane	6.90	0.08	0.04	0.25	0.37	0.23	0.09	0.09	0.29	0.21	0.68	0.53	21.54	24.92	2.79	2.18
Transchlorane	6.90	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
Chlorane	6.90	0.09	0.03	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.04	0.06	0.01	0.00
Transnonachlor	6.90	0.11	0.12	0.21	0.09	0.11	0.01	0.09	0.07	0.17	0.08	0.21	1.98	3.54	0.41	0.50
Cisnonachlor	6.90	0.08	0.04	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.17	0.29	0.04	0.04
p,p' DDE	6.90	0.07	0.05	0.14	0.39	0.23	0.20	0.01	0.18	1.23	0.28	0.42	0.61	1.61	0.14	0.22
p,p' DDD	6.90	0.08	0.04	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
p,p' DDT	6.00	0.16	0.12	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
Photomirex	6.00	0.07	0.05	0.11	0.11	0.08	0.01	0.01	0.01	0.01	0.12	0.18	1.20	1.01	0.34	0.27
Mirex	6.00	0.07	0.05	0.06	0.19	0.11	0.01	0.01	0.07	0.06	0.20	0.33	0.71	0.42	0.31	0.32
Heptachlor epoxide	6.00	0.12	0.05	0.39	0.44	0.43	0.01	0.22	0.36	0.56	0.23	0.67	3.23	3.84	1.42	1.39
Dieldrin	6.20	0.10	0.09	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	5.70	2.40	2.97
PCB31	5.60	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB28	5.60	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB52	5.84	0.10	0.04	0.01	0.02	0.01	0.01	0.01	0.11	0.22	0.14	0.19	0.01	0.00	0.01	0.00
PCB49	5.85	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB44	5.75	0.08	0.06	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB42	5.76	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB64	5.95	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB74	6.20	0.08	0.04	0.18	0.28	0.13	0.01	0.01	0.28	0.26	0.37	0.33	1.27	1.93	0.23	0.10
PCB70	6.20	0.09	0.03	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB66/65	6.20	0.09	0.03	0.22	0.23	0.16	0.06	0.08	0.18	0.28	0.22	0.32	0.26	0.49	0.01	0.00
PCB60	6.11	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.20	0.31	0.11	1.20	2.37	0.01	0.00

Appendix III continued
Cambridge Bay

CHEMICAL	log K _{ow}	ng/g dry wt. Lichen SD	Female caribou (age in years)			Male caribou (age in years)			Female wolf n=6		Male wolf n=8					
			1.40	2.40	3.40	6.40	13.40	6.40	7.40	7.40	Mean	SD				
PCB101	6.38	0.09	0.03	0.12	0.23	0.14	0.10	0.01	0.15	0.22	0.13	0.25	0.08	0.13	0.01	0.00
PCB99	6.39	0.09	0.03	0.73	0.76	0.38	0.17	0.14	0.58	0.35	0.55	0.57	6.34	9.20	1.20	0.89
PCB97	6.29	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB87	6.29	0.08	0.07	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB110	6.48	0.08	0.04	0.09	0.02	0.01	0.01	0.01	0.01	0.15	0.01	0.01	0.01	0.00	0.01	0.00
PCB151	6.64	0.05	0.04	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB149	6.67	0.21	0.21	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.02	0.03
PCB118	6.74	0.07	0.05	0.88	1.63	0.97	0.29	0.32	0.90	0.87	1.38	1.11	3.86	6.52	0.64	0.39
PCB146	6.89	0.05	0.04	0.13	0.23	0.10	0.01	0.01	0.18	0.11	0.21	0.25	0.38	0.69	0.10	0.09
PCB153	6.92	0.08	0.03	1.90	2.14	0.91	0.39	0.33	1.94	1.32	2.19	1.98	10.94	12.94	3.61	2.17
PCB105	6.65	0.06	0.05	0.01	0.25	0.11	0.07	0.01	0.01	0.01	0.30	0.01	0.01	0.00	0.01	0.00
PCB141	6.82	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB138	6.83	0.09	0.03	1.47	1.75	1.04	0.36	0.33	1.08	0.83	1.45	1.22	3.78	5.30	1.30	0.88
PCB129	6.73	0.07	0.05	0.14	0.12	0.01	0.01	0.01	0.12	0.07	0.14	0.18	0.08	0.12	0.02	0.02
PCB162/187	7.20	0.07	0.05	0.19	0.39	0.16	0.07	0.05	0.22	0.27	0.39	0.40	0.16	0.31	0.03	0.04
PCB183	7.00	0.07	0.05	0.19	0.11	0.01	0.01	0.01	0.07	0.01	0.08	0.01	0.60	0.68	0.16	0.15
PCB185	7.11	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB174	7.11	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB171	7.11	0.07	0.05	0.01	0.14	0.01	0.01	0.01	0.12	0.12	0.12	0.17	0.01	0.00	0.01	0.00
PCB200	7.20	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.10	0.21	0.39	0.03	0.05
PCB172	7.33	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.07	0.01	0.09	0.10	0.08	0.12	0.01	0.00
PCB180	7.50	0.07	0.05	0.68	0.87	0.37	0.13	0.13	0.58	0.57	0.80	0.79	15.04	11.15	3.66	1.90
PCB170/190	7.46	0.07	0.05	0.27	0.44	0.17	0.01	0.01	0.41	0.40	0.65	0.51	9.89	7.91	2.30	1.05
PCB201	7.62	0.07	0.05	0.22	0.32	0.11	0.01	0.01	0.31	0.26	0.47	0.38	0.14	0.19	0.02	0.03
PCB203	7.65	0.07	0.05	0.09	0.11	0.01	0.01	0.01	0.01	0.01	0.08	0.01	0.08	0.11	0.04	0.06
PCB195	7.56	0.07	0.05	0.01	0.02	0.01	0.01	0.01	0.14	0.01	0.21	0.01	0.12	0.09	0.03	0.04
PCB194	7.80	0.07	0.05	0.01	0.12	0.01	0.01	0.01	0.11	0.10	0.16	0.13	4.52	2.60	0.84	0.47
PCB206	8.09	0.07	0.05	0.01	0.16	0.01	0.01	0.01	0.14	0.01	0.17	0.11	1.82	1.34	0.27	0.23
PCB189	7.71	0.01	0.00	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.44	0.30	0.05	0.06
PCB77	6.36	0.01	0.00	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB126	6.89	0.01	0.00	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
PCB169	7.42	0.01	0.00	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.00
Total PCBs	6.92	2.99	1.59	14.06	22.30	21.62	15.95	22.59	13.34	25.63	30.56	35.09	61.60	65.24	14.83	8.66
Arochlor1254:1260		0.10	0.00	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250		0.10	0.00	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Appendix III continued

Bathurst

CHEMICAL	log K _{ow}	ng/g dry wt.		Female caribou (age in years)										
		Lichen	SD	3.3	3.3	4.1	4.3	5.1	6.1	6.3	7.1	10.3	11.1	
1,2,4,5 TCB	4.70	0.04	0.02	0.31	0.15	0.31	0.29	0.33	0.31	0.14	0.09	0.19	0.33	
1,2,3,4TCB	4.46	0.06	0.04	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.06	0.07	
QCB	5.03	0.15	0.13	0.65	0.18	0.26	0.60	0.37	0.58	0.32	0.62	0.34	0.66	
alpha-HCH	4.00	1.72	1.12	4.84	6.32	7.65	5.23	8.07	16.28	2.52	9.26	5.33	8.11	
beta-HCH	4.00	0.04	0.03	1.02	0.34	1.88	0.56	1.31	1.01	0.22	2.05	0.22	1.79	
gamma-HCH	4.50	0.49	0.59	0.19	0.22	0.44	0.18	0.51	0.89	0.12	1.34	0.40	0.73	
HCB	5.50	0.52	0.46	16.01	10.02	30.29	17.68	11.57	12.45	7.96	12.11	13.33	9.50	
OCS	6.90	0.04	0.02	0.50	0.22	0.87	0.56	0.05	0.19	0.09	0.09	0.28	0.13	
Oxychlorane	6.90	0.04	0.02	0.22	0.06	0.13	0.18	0.05	0.04	0.04	0.09	0.13	0.07	
Transchlorane	6.90	0.04	0.02	0.78	0.05	0.04	0.11	0.05	0.39	0.07	0.53	0.07	0.07	
Cischlorane	6.90	0.04	0.03	0.78	0.05	0.04	0.09	0.05	0.35	0.04	0.09	0.07	0.07	
Transnonachlor	6.90	0.04	0.03	0.62	0.08	0.04	0.16	0.05	0.27	0.01	0.09	0.01	0.07	
Cisnonachlor	6.90	0.04	0.02	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.03	0.07	
p,p' DDE	6.90	0.05	0.05	2.05	0.51	0.44	1.71	4.67	1.20	0.10	6.14	0.38	0.40	
p,p' DDD	6.90	0.07	0.06	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
p,p' DDT	6.00	0.20	0.24	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
Photomirex	6.00	0.04	0.02	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
Mirex	6.00	0.04	0.02	0.03	0.02	0.04	0.11	0.05	0.15	0.01	0.09	0.01	0.07	
Heptachlor epoxide	6.00	0.05	0.02	0.03	0.08	0.04	0.02	0.05	0.46	0.01	0.09	0.01	0.07	
Dieldrin	6.20	0.05	0.05	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
PCB31	5.60	0.03	0.02	0.03	0.98	2.05	0.54	0.05	0.04	0.01	0.09	0.97	2.39	
PCB28	5.60	0.04	0.02	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
PCB52	5.84	0.05	0.03	0.37	0.15	0.22	0.38	0.42	0.93	0.10	0.80	0.68	0.40	
PCB49	5.85	0.04	0.02	0.03	0.02	0.04	0.11	0.05	0.35	0.01	0.36	0.24	0.07	
PCB44	5.75	0.04	0.02	0.03	0.34	0.39	0.41	0.05	0.04	0.01	0.09	0.50	0.53	
PCB42	5.76	0.04	0.02	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.18	0.07	
PCB64	5.95	0.04	0.02	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07	
PCB74	6.20	0.04	0.02	0.19	0.09	0.22	0.18	0.19	0.46	0.01	0.09	0.40	0.07	
PCB70	6.20	0.05	0.02	0.19	0.09	0.04	0.16	0.37	0.81	0.01	0.53	0.77	0.07	
PCB66/95	6.20	0.06	0.02	0.37	0.20	0.26	0.38	0.51	1.20	0.10	0.89	1.06	0.40	
PCB60	6.11	0.04	0.02	0.03	0.02	0.04	0.02	0.05	1.01	0.01	0.09	0.01	0.07	

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Appendix III continued

Bathurst

CHEMICAL	log K _{ow}	ng/g dry wt. Lichen SD	Female caribou (age in years)									
			3.3	3.3	4.1	4.3	5.1	6.1	6.3	7.1	10.3	11.1
PCB101	6.38	0.05	0.34	0.14	0.17	0.40	0.33	0.93	0.07	0.62	0.77	0.27
PCB99	6.39	0.04	0.31	0.14	0.44	0.29	0.23	0.50	0.04	0.36	0.38	0.33
PCB97	6.29	0.04	0.03	0.02	0.04	0.29	0.05	0.35	0.01	0.09	0.22	0.07
PCB87	6.29	0.04	0.25	0.06	0.04	0.31	0.19	0.58	0.01	0.45	0.44	0.07
PCB110	6.48	0.04	0.16	0.09	0.04	0.22	0.37	0.70	0.01	0.89	0.47	0.27
PCB151	6.64	0.04	0.22	0.03	0.04	0.27	0.05	0.35	0.01	0.09	0.25	0.07
PCB149	6.67	0.05	0.25	0.11	0.04	0.38	0.19	0.46	0.01	0.09	0.49	0.07
PCB118	6.74	0.04	0.37	0.26	0.74	0.70	0.47	0.85	0.17	0.62	0.84	0.40
PCB146	6.89	0.04	0.19	0.08	0.13	0.20	0.05	0.12	0.01	0.09	0.12	0.07
PCB153	6.92	0.05	1.18	0.57	1.62	1.46	0.65	1.16	0.19	0.62	0.87	0.66
PCB105	6.65	0.04	0.03	0.03	0.17	0.11	0.05	0.12	0.01	0.09	0.27	0.07
PCB141	6.82	0.04	0.12	0.06	0.04	0.13	0.05	0.08	0.01	0.09	0.10	0.07
PCB138	6.83	0.05	0.62	0.51	0.83	1.30	0.56	1.20	0.13	0.71	0.60	0.53
PCB129	6.73	0.04	0.03	0.02	0.04	0.07	0.05	0.04	0.01	0.09	0.01	0.07
PCB162/167	7.20	0.04	0.53	0.35	0.39	0.78	0.05	0.85	0.10	0.09	0.43	0.07
PCB183	7.00	0.04	0.25	0.09	0.04	0.20	0.05	0.31	0.01	0.09	0.15	0.07
PCB185	7.11	0.04	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
PCB174	7.11	0.04	0.03	0.03	0.04	0.11	0.05	0.12	0.01	0.09	0.03	0.07
PCB171	7.11	0.04	0.03	0.08	0.04	0.13	0.05	0.04	0.01	0.09	0.12	0.07
PCB200	7.20	0.04	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
PCB172	7.33	0.04	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
PCB180	7.50	0.04	0.81	0.37	0.57	1.06	0.51	0.66	0.10	0.27	0.28	0.47
PCB170/190	7.46	0.04	0.31	0.20	0.48	0.56	0.23	0.39	0.01	0.09	0.13	0.07
PCB201	7.62	0.04	0.53	0.14	0.52	0.45	0.33	0.31	0.01	0.09	0.09	0.40
PCB203	7.65	0.04	0.19	0.05	0.04	0.14	0.05	0.12	0.01	0.09	0.01	0.07
PCB195	7.56	0.04	0.03	0.02	0.04	0.25	0.05	0.04	0.01	0.09	0.01	0.07
PCB194	7.80	0.04	0.47	0.09	0.39	0.32	0.05	0.23	0.01	0.09	0.18	0.13
PCB206	8.09	0.04	0.43	0.02	0.17	0.25	0.05	0.39	0.01	0.09	0.01	0.27
PCB189	7.71	0.01	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
PCB77	6.36	0.01	0.03	0.02	0.26	0.02	0.05	0.04	0.01	0.09	0.24	0.07
PCB126	6.89	0.01	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
PCB169	7.42	0.01	0.03	0.02	0.04	0.02	0.05	0.04	0.01	0.09	0.01	0.07
Total PCBs	6.92	1.57	12.50	8.84	15.20	16.97	11.96	22.19	7.80	16.98	22.74	20.47
Arochlor1254:1260		0.05	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Arochlor1250		0.05	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Appendix III continued

Bathurst

Male caribou

(age in years)

CHEMICAL	log K _{ow}	ng/g dry wt.		(age in years)										
		Lichen	SD	3.25	4.10	6.10	6.25	8.25	8.25	9.10	9.10	9.25	11.10	
1,2,4,5 TCB	4.70	0.04	0.02	0.29	0.54	0.46	0.28	0.28	0.30	0.46	0.89	0.25	0.50	
i,2,3,4TCB	4.46	0.06	0.04	0.02	0.02	0.03	0.01	0.01	0.15	0.03	0.05	0.01	0.02	
QCB	5.03	0.15	0.13	0.48	0.54	0.28	0.44	0.32	0.49	0.34	0.63	0.60	0.34	
alpha-HCH	4.00	1.72	1.12	4.22	8.29	10.60	8.89	5.15	7.07	8.03	18.60	7.58	10.09	
beta-HCH	4.00	0.04	0.03	0.42	0.99	1.14	0.50	0.28	0.43	1.63	2.92	0.28	0.69	
gamma-HCH	4.50	0.49	0.59	0.23	0.54	0.89	0.32	0.20	0.36	1.03	1.98	0.28	0.42	
HCB	5.50	0.52	0.46	12.21	66.98	124.42	12.55	16.45	9.64	73.42	116.26	11.85	73.88	
OCS	6.90	0.04	0.02	0.19	0.95	2.67	0.21	0.28	0.15	1.63	1.56	0.42	1.47	
Oxychlorane	6.90	0.04	0.02	0.02	0.28	1.11	0.40	0.15	0.02	0.52	0.68	0.78	0.40	
Transchlorane	6.90	0.04	0.02	0.02	0.02	0.89	0.15	0.01	0.02	0.03	0.05	0.03	0.06	
Cischlorane	6.90	0.04	0.03	0.02	0.02	0.52	0.01	0.01	0.02	0.03	0.05	0.01	0.02	
Transnonachlor	6.90	0.04	0.03	0.02	0.02	0.03	0.01	0.01	0.02	0.10	0.05	0.06	0.02	
Cisnonachlor	6.90	0.04	0.02	0.02	0.02	0.03	0.01	0.01	0.02	0.03	0.05	0.01	0.02	
p,p' DDE	6.90	0.05	0.05	0.21	2.13	1.41	1.19	0.24	0.02	3.07	9.64	0.31	0.32	
p,p' DDD	6.90	0.07	0.06	0.02	0.19	0.03	0.01	0.01	0.02	0.03	0.52	0.01	0.02	
p,p' DDT	6.00	0.20	0.24	0.02	0.02	0.89	0.01	0.01	0.02	0.03	0.05	0.01	0.02	
Photomirex	6.00	0.04	0.02	0.02	0.02	0.28	0.01	0.01	0.02	0.10	0.05	0.04	0.02	
Mirex	6.00	0.04	0.02	0.12	0.02	0.92	0.01	0.01	0.02	0.34	0.16	0.15	0.06	
Heptachlor epoxide	6.00	0.05	0.02	0.02	0.02	0.37	0.08	0.07	0.08	0.31	0.31	0.38	0.11	
Dieldrin	6.20	0.05	0.05	0.02	0.02	0.52	0.01	0.01	0.02	0.03	0.28	0.01	0.02	
PCB31	5.60	0.03	0.02	1.06	0.02	0.03	0.01	0.53	0.02	0.03	0.05	0.56	0.02	
PCB28	5.60	0.04	0.02	0.02	0.02	1.01	0.01	0.01	0.02	0.03	0.05	0.01	0.23	
PCB52	5.84	0.05	0.03	0.19	0.11	1.90	0.07	0.11	0.02	0.15	0.52	0.15	0.19	
PCB49	5.85	0.04	0.02	0.02	0.02	0.77	0.01	0.01	0.02	0.03	0.05	0.01	0.06	
PCB44	5.75	0.04	0.02	0.25	0.02	1.23	0.01	0.14	0.02	0.03	0.05	0.15	0.21	
PCB42	5.76	0.04	0.02	0.02	0.02	0.40	0.01	0.01	0.02	0.03	0.05	0.01	0.02	
PCB64	5.95	0.04	0.02	0.15	0.02	0.25	0.01	0.01	0.02	0.03	0.05	0.01	0.02	
PCB74	6.20	0.04	0.02	0.10	0.32	1.11	0.08	0.12	0.02	0.28	0.68	0.09	0.38	
PCB70	6.20	0.05	0.02	0.15	0.04	1.23	0.06	0.06	0.02	0.08	0.21	0.01	0.11	
PCB66/95	6.20	0.06	0.02	0.23	0.28	2.00	0.33	0.14	0.07	0.21	0.63	0.18	0.32	
PCB80	6.11	0.04	0.02	0.04	0.15	0.46	0.01	0.01	0.02	0.15	0.05	0.01	0.25	

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Appendix III continued

Bathurst

Male caribou

(age in years)

CHEMICAL	log Kow	ng/g dry wt.		Male caribou (age in years)											
		Lichen	SD	3.25	4.10	6.10	6.26	8.25	8.26	9.10	9.10	9.10	9.26	11.10	
PCB101	6.38	0.19	0.05	0.17	1.63	0.10	0.08	0.02	0.15	0.42	0.15	0.26	0.23		
PCB99	6.39	0.17	0.04	0.52	1.66	0.32	0.17	0.07	0.83	1.20	0.34	0.63			
PCB97	6.29	0.02	0.04	0.02	0.43	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB87	6.29	0.17	0.04	0.02	0.80	0.01	0.04	0.02	0.03	0.05	0.01	0.13			
PCB110	6.48	0.15	0.04	0.06	1.01	0.03	0.07	0.02	0.08	0.21	0.07	0.15			
PCB151	6.64	0.08	0.04	0.02	0.25	0.01	0.01	0.02	0.03	0.05	0.01	0.08			
PCB149	6.67	0.08	0.05	0.02	0.68	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB118	6.74	0.23	0.04	0.99	2.67	0.28	0.36	0.11	1.39	2.19	0.29	1.16			
PCB146	6.89	0.02	0.04	0.02	0.40	0.04	0.05	0.02	0.15	0.36	0.04	0.17			
PCB153	6.92	0.29	0.05	1.79	4.88	0.50	0.61	0.31	2.99	4.33	0.66	2.27			
PCB105	6.65	0.02	0.04	0.22	0.52	0.06	0.05	0.02	0.26	0.36	0.01	0.31			
PCB141	6.82	0.02	0.04	0.02	0.09	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB138	6.83	0.23	0.05	0.93	3.04	0.29	0.40	0.23	1.60	2.24	0.40	1.24			
PCB129	6.73	0.02	0.04	0.22	0.64	0.01	0.01	0.02	0.21	0.42	0.03	0.17			
PCB182/187	7.20	0.46	0.04	0.47	1.41	0.22	0.06	0.02	0.36	1.15	0.18	0.93			
PCB183	7.00	0.02	0.04	0.02	0.34	0.01	0.04	0.02	0.10	0.21	0.04	0.10			
PCB185	7.11	0.02	0.04	0.02	0.03	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB174	7.11	0.02	0.04	0.02	0.18	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB171	7.11	0.02	0.04	0.15	0.46	0.04	0.07	0.02	0.03	0.16	0.01	0.11			
PCB200	7.20	0.02	0.04	0.02	0.12	0.01	0.01	0.02	0.03	0.05	0.01	0.06			
PCB172	7.33	0.02	0.04	0.02	0.25	0.01	0.06	0.02	0.13	0.26	0.01	0.02			
PCB180	7.50	0.25	0.04	0.82	2.70	0.23	0.28	0.20	1.57	2.08	0.29	1.07			
PCB170/190	7.46	0.15	0.04	0.50	1.50	0.14	0.25	0.15	0.72	1.09	0.16	0.80			
PCB201	7.62	0.08	0.04	0.52	1.35	0.14	0.21	0.11	0.67	1.15	0.16	0.76			
PCB203	7.65	0.02	0.04	0.02	0.31	0.01	0.01	0.02	0.10	0.16	0.04	0.10			
PCB195	7.56	0.02	0.04	0.13	0.40	0.01	0.01	0.02	0.18	0.31	0.01	0.19			
PCB194	7.80	0.25	0.04	0.37	0.80	0.06	0.11	0.07	0.44	0.52	0.09	0.42			
PCB206	8.09	0.02	0.04	0.28	1.17	0.01	0.14	0.02	0.59	0.47	0.12	0.53			
PCB189	7.71	0.02	0.01	0.02	0.03	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB77	6.36	0.02	0.01	0.02	0.28	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB126	6.89	0.02	0.01	0.02	0.03	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
PCB169	7.42	0.02	0.01	0.02	0.03	0.01	0.01	0.02	0.03	0.05	0.01	0.02			
Total PCBs	6.92	8.62	1.57	13.66	46.58	9.56	12.63	10.12	23.01	31.40	13.75	24.72			
Arochlor1254:1260		NA	0.05	NA	NA	NA	NA	NA	NA	NA	NA	NA			
Arochlor1250		NA	0.05	NA	NA	NA	NA	NA	NA	NA	NA	NA			

Appendix III continued

Bathurst Wolves

CHEMICAL	log Kow	Female wolves (age in years)						Male wolves (age in years)							
		0.5	1.3	1.3	2.5	3.8	4.3	0.5	0.5	1.5	1.5	2.3			
1,2,4,5 TCB	4.70	1.46	1.50	2.52	0.53	2.19	2.08	1.65	1.68	1.89	3.06	0.19	0.18	0.01	0.01
1,2,3,4TCB	4.46	0.15	0.17	0.19	0.14	0.14	0.01	0.26	2.24	2.40	4.18	7.27	6.26	10.07	14.99
QCB	5.03	1.88	1.85	3.39	1.15	2.29	2.80	13.93	9.55	12.06	32.85	0.01	0.01	0.01	0.01
alpha-HCH	4.00	5.28	11.91	9.39	4.97	5.02	12.80	104.08	91.42	84.66	109.14	2.18	1.61	2.39	7.94
beta-HCH	4.00	8.04	15.30	13.15	10.61	8.18	15.04	7.87	11.81	10.25	24.14	0.09	0.01	0.27	0.14
gamma-HCH	4.50	0.01	0.01	0.01	0.01	0.14	0.10	0.12	0.01	0.05	0.10	0.25	0.51	0.74	0.98
HCB	5.50	59.42	82.47	81.98	36.35	126.83	83.29	0.03	0.04	0.05	0.09	0.01	0.01	0.01	0.01
OCS	6.90	0.97	1.33	3.06	1.17	4.59	2.15	0.01	0.01	0.01	0.04	0.01	0.01	0.01	0.04
Oxychloridane	6.90	1.88	4.27	13.58	2.41	11.41	11.67	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Transchloridane	6.90	0.01	0.01	0.01	0.04	0.09	0.22	0.01	0.01	0.01	0.10	0.01	0.01	0.01	0.01
Cischloridane	6.90	0.01	0.01	0.01	0.01	0.07	0.01	0.01	0.01	0.01	0.10	0.01	0.01	0.01	0.01
Transnonachlor	6.90	0.09	0.29	0.35	0.01	0.07	0.13	0.01	0.01	0.01	0.04	0.01	0.01	0.01	0.01
Cisnonachlor	6.90	0.01	0.01	0.01	0.01	0.01	0.04	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
p,p' DDE	6.90	0.05	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
p,p' DDD	6.90	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
p,p' DDT	6.00	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Photomirex	6.00	0.16	0.44	0.57	0.23	1.34	0.34	0.36	0.30	1.06	2.57	0.68	0.41	1.58	2.75
Mirex	6.00	0.19	0.64	0.84	0.36	2.28	0.56	1.42	0.98	3.45	5.88	0.01	0.01	0.01	0.47
Heptachlor epoxide	6.00	0.63	2.20	2.80	0.47	1.16	1.76	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Dieldrin	6.20	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB31	5.60	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB28	5.60	0.05	0.01	0.01	0.10	0.08	0.12	0.11	0.13	0.11	0.12	0.25	0.01	0.01	0.40
PCB52	5.84	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.22
PCB49	5.85	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB44	5.75	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB42	5.76	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB64	5.95	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
PCB74	6.20	0.52	0.68	1.03	0.45	0.34	0.46	0.56	0.37	0.80	7.84	0.01	0.01	0.01	0.44
PCB70	6.20	0.06	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.48
PCB66/85	6.20	0.17	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.48
PCB60	6.11	0.01	0.01	0.11	0.01	0.14	0.24	0.01	0.01	0.01	0.75	0.01	0.01	0.01	2.42

Appendix III continued

Bathurst Wolves

CHEMICAL	log K _{ow}	Female wolves (age in years)					Male wolves (age in years)				
		0.6	1.3	1.3	2.6	3.8	4.3	6.6	6.6	1.6	2.3
PCB101	6.38	0.01	0.01	0.01	0.01	0.01	0.21	0.01	0.01	0.01	0.34
PCB99	6.39	0.47	1.96	1.79	0.64	2.15	1.47	1.26	3.26	11.55	
PCB97	6.29	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB87	6.29	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB110	6.48	0.01	0.01	0.01	0.01	0.01	0.17	0.01	0.01	0.01	
PCB151	6.64	0.01	0.01	0.09	0.01	0.01	0.12	0.01	0.04	0.06	
PCB149	6.67	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB118	6.74	2.94	0.67	0.74	0.26	1.25	6.33	1.80	6.71	41.85	
PCB146	6.89	0.04	0.11	0.08	0.20	0.19	0.25	0.06	0.15	0.52	
PCB153	6.92	3.28	12.67	8.69	4.38	15.15	12.06	8.63	18.48	66.52	
PCB105	6.65	0.30	0.01	0.01	0.01	0.01	1.15	0.01	1.21	8.74	
PCB141	6.82	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB138	6.83	0.79	1.60	1.48	0.60	1.59	1.78	1.00	3.39	14.65	
PCB129	6.73	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.04	0.16	
PCB182/187	7.20	0.01	0.01	0.08	0.01	0.01	0.01	0.01	0.01	0.32	
PCB183	7.00	0.13	0.82	0.57	0.21	0.90	0.35	0.26	1.11	5.20	
PCB185	7.11	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB174	7.11	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.09	
PCB171	7.11	0.49	0.24	0.36	0.01	1.01	1.28	0.75	1.69	8.37	
PCB200	7.20	0.20	0.01	0.09	0.01	0.21	0.21	0.32	0.71	3.41	
PCB172	7.33	0.04	0.04	0.09	0.10	0.17	0.09	0.01	0.12	0.37	
PCB180	7.50	4.06	15.67	18.45	12.90	40.23	9.64	15.00	22.38	90.60	
PCB170/190	7.46	2.93	10.66	12.73	9.75	22.96	5.24	8.28	13.12	50.92	
PCB201	7.62	0.13	0.23	0.26	0.01	0.42	0.41	0.01	0.29	0.94	
PCB203	7.65	0.03	0.20	0.09	0.01	0.21	0.22	0.08	0.26	1.54	
PCB195	7.56	0.09	0.66	0.32	0.20	0.65	0.12	0.14	0.48	1.96	
PCB194	7.80	1.16	4.06	5.55	5.62	11.25	2.02	6.63	5.90	28.56	
PCB206	6.09	0.61	1.99	2.70	3.37	6.33	1.14	2.97	2.73	11.92	
PCB189	7.71	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
PCB77	6.36	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.12	
PCB126	6.89	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.10	
PCB169	7.42	0.01	0.01	0.01	0.01	0.47	0.01	0.81	0.01	0.05	
Total PCBs	6.92	18.74	52.45	55.55	39.29	105.96	45.78	48.77	84.18	360.96	
Arochlor1254:1260		NA	NA	NA	NA	NA	NA	NA	NA	NA	
Arochlor1250		NA	NA	NA	NA	NA	NA	NA	NA	NA	

APPENDIX IV

Assessment of Toxic Substances under the Canadian Environmental Protection Act (CEPA) and Environment Canada's Toxic Substance Management Plan (TSMP)

Current POPs Management Policy

Political and legal action toward the management of persistent organic pollutants (POPs) is difficult due to the complexity of present political and socio-economic status among nations that produce and or utilize these chemicals. In 1995, Environment Canada introduced their *Toxic Substances Management Policy (TSMP)*. Embedded in this policy is Part IV of the Canadian Environmental Protection Act, the governing legislation that regulates toxic substances in Canada. More recently in 1998, Canada and other industrial nations began negotiations on a new POPs protocol under the United Nations Environment Program's (UNEP) - *Convention for Long-Range Transboundary Air Pollution (LRTAP)*. Under the *TSMP* and *LRTAP* POPs protocol policy makers have attempted to provide a science-based framework of managing toxic substances in Canada and Internationally.

Scientific assessment of candidate chemicals (new and existing compounds) is an important feature of this policy. The assessment process of a candidate substance under the *TSMP* and Part IV of CEPA is shown in Figure AIV-1. Assessment of chemicals is based on a set of four scientific criteria. The assessment considers if a chemical is (1) toxic as defined within the provisions of CEPA, (2) anthropogenically produced, (3) bioaccumulative, and (4) persistent. If an assessment of a candidate substance is found to satisfy all of the above criteria

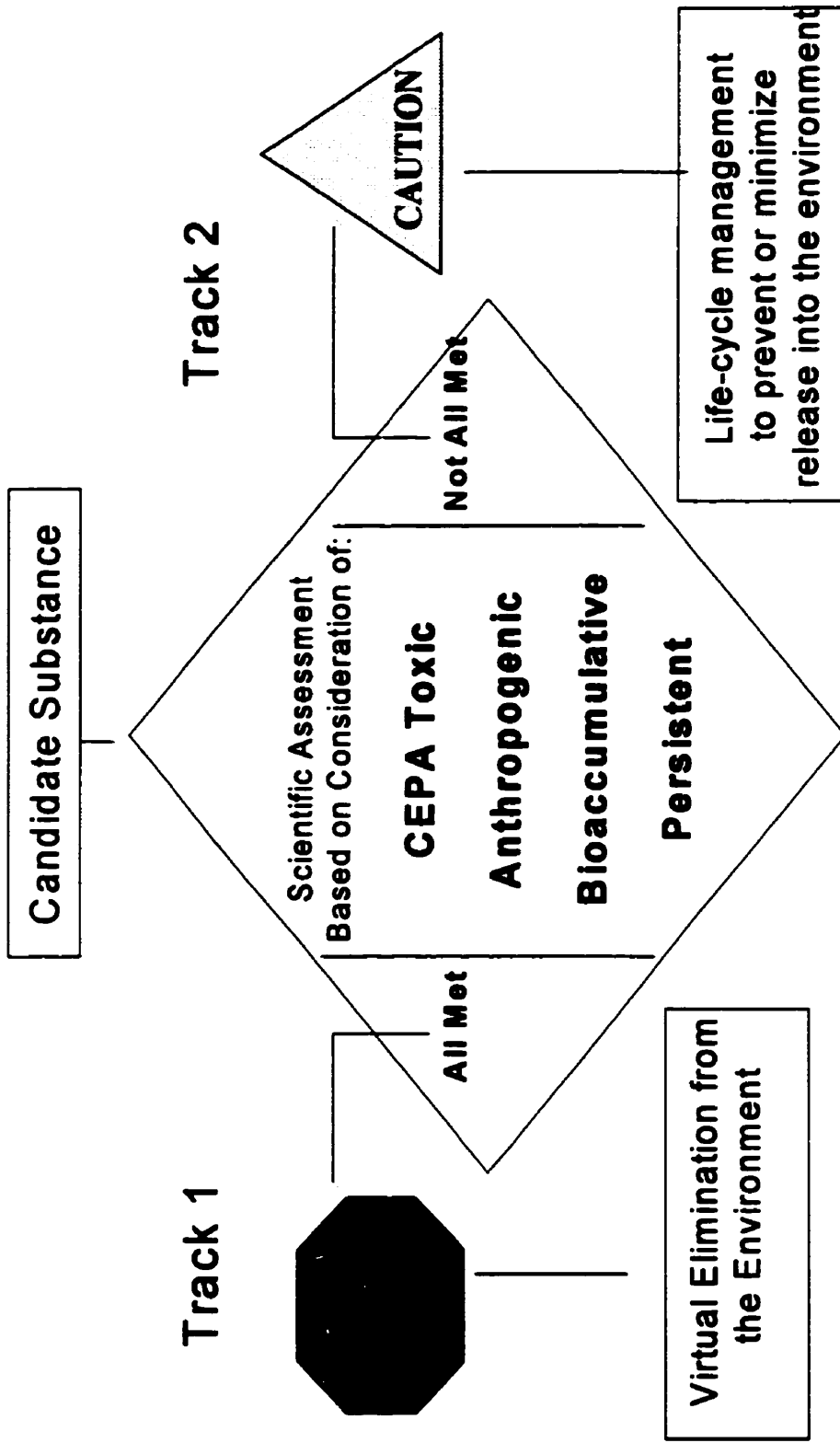


Figure AIV-1: Assessment criteria under CEPA and the TSMP.

are, then a the substance is considered a track 1 substance. The action precipitated by a Track 1 assessment involves virtual elimination from the environment by implementation of production and usage bans. If all of the criteria are not satisfied through the assessment, then the chemical is to go under life-cycle management. This policy action, referred to as "cradle to grave approach", attempts to prevent or minimize the production and release of the substance. Under the screening approach of the TSMP and United Nations LRTAP, chemicals that are considered bioaccumulative are targeted for virtual elimination from the environment. Bioaccumulation criteria are derived from experiments and biomonitoring studies. Generally, the experimentally derived BAFs are the result of dietary exposure experiments with small forage fish such as guppies, rainbow trout and goldfish. Analysis of chemical concentrations in food, water and a test organism's tissues under experimental conditions can elicit bioconcentration (BCF) and bioaccumulation (BAF) factors for a candidate substance. bioaccumulative substances are those chemicals that exhibit BCFs/BAFs greater than 5000 or its octanol-water partition coefficient (K_{ow}) is greater than 10^5 . Do these experimentally derived, single value bioaccumulation criteria completely address the bioaccumulative potential of a chemical in all environments, for all species and life-stages?

Problems with Current Approach

There are inherent problems in the current approach for assessing bioaccumulative capacity of new and existing chemicals. First, this approach sets bioaccumulation criteria based on laboratory analyses of the candidate substance. The observed bioaccumulation in these experiments are intended to be extrapolated to other species and ecosystems. Intraspecies and interspecies variation, ecological conditions, and physiology can lead to different bioaccumulation patterns. Secondly, this approach does not account for sensitive life-stages of some organisms. For example, bioaccumulation of less hydrophobic compounds may be transferred to newborn mammals from lactating females. In addition to physiology, bioaccumulation of organic contaminants is a chemical process that is affected by the dose(mg/kg/day) available for uptake by an organism. The current management policy for screening bioaccumulative substances does not consider the available dose in terms of ambient environmental concentrations. If produced in large enough quantities, chemicals that are considered not to be bioaccumulative substances may pose a hazard to some organisms.

APPENDIX V

Sources, Sinks and Pathways of Persistent Organic Pollutants (POPs)

Persistent Organic Pollutants

Recent evidence suggests that many POPs are volatile enough to evaporate and deposit among air, water and soil at ordinary environmental temperatures (Mackay and Wania, 1996). Warm temperatures favour evaporation from the earth's surface in both tropical and sub-tropical regions of the world. Cool temperatures at higher latitudes favour deposition from the atmosphere. The chemical properties of POPs and the differing ambient temperatures will delineate the loading and exposure of these contaminants to a given region. In addition to the temperature the resulting evaporation or deposition of these chemicals is dependent on their inherent properties such as vapour pressure (P_v), temperature of condensation (T_c). Chemicals with high vapour pressures and low condensation temperatures are more likely to accumulate in polar ecosystems.

Once emitted to the atmosphere, it is theorized that POPs undergo an evaporation and deposition process referred to as a "grasshopper effect". Figure AV-1 illustrates how these chemicals might migrate, rest, and migrate again in tune with seasonal temperature changes at mid-latitudes. If we adopt this hypothesis we would expect to see a concentration gradient, increasing from warm release locations (mid-latitudes) to cold migration points (polar regions). This pattern has been demonstrated in various environmental and biota samples for volatile POPs such as Hexachlorobenzene and Hexachlorocyclohexanes

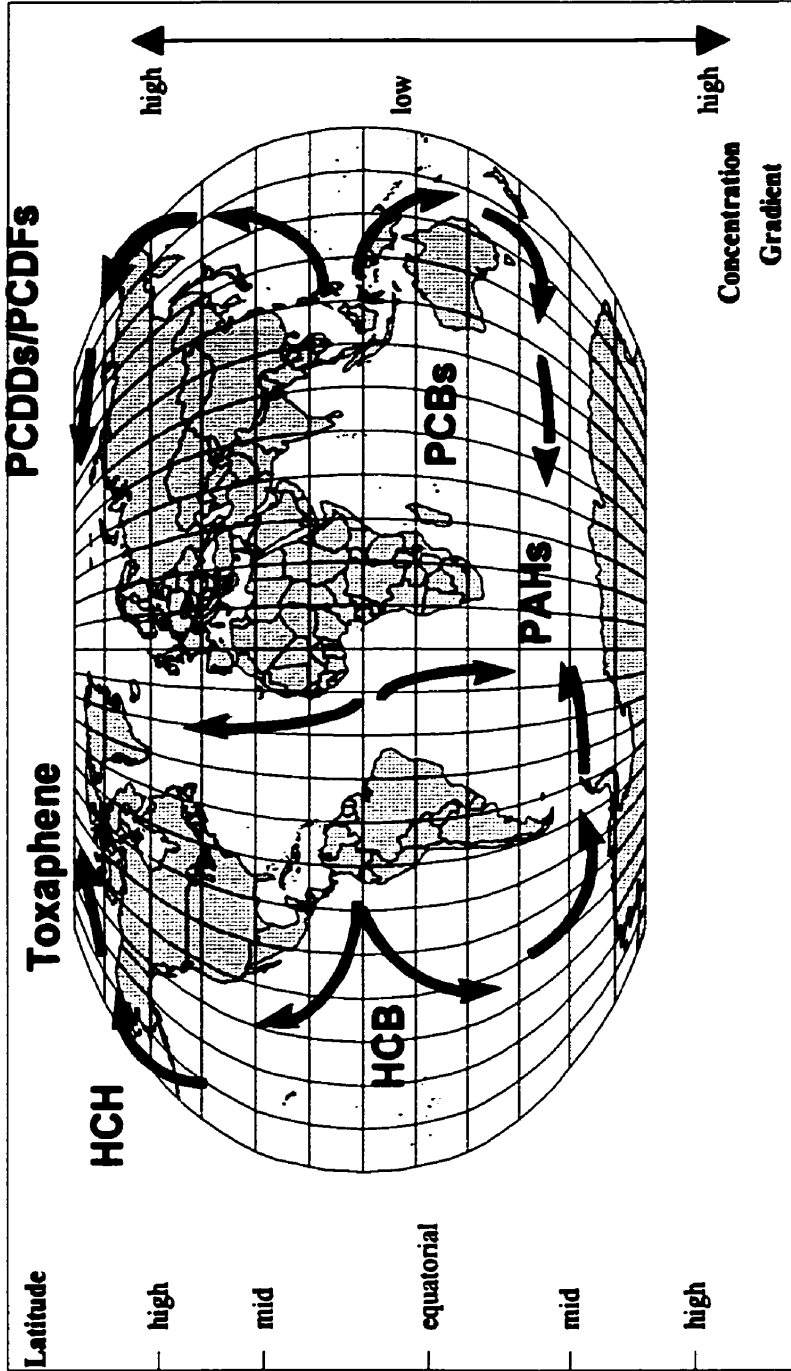


Figure AV-1: Theoretical model of POP migration processes.

(HCHs). Figure AV-2 illustrates an inverted latitudinal concentration profile of Σ HCH in sea water. This concentration profile shows very low concentrations (0.05 – 0.5 ng/L) in the tropics and increasing concentrations with increasing latitude.

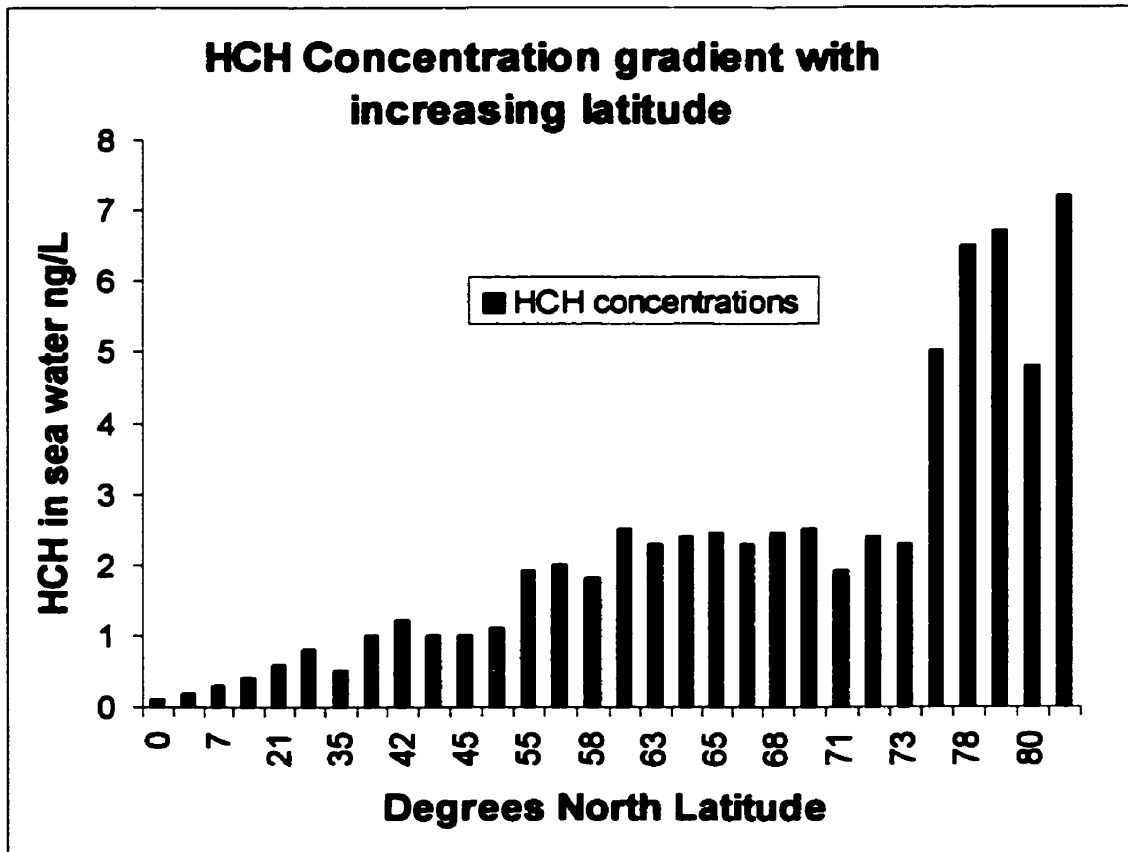


Figure AV-2: Inverted latitudinal concentration profile of Σ HCH in sea water. Source: Mackay and Wania, 1996.

POPs can originate from application of agricultural pesticides, combustion, solvents and by-products from the plastics industry and metal processing.

Political and legal actions to combat the issue of global emission of POPs can be difficult to implement and enforce due to the complexity of the present political

and market-based networks. The increased environmental hazards associated with these chemicals are due to the fact that they bioaccumulate into organisms and may biomagnify through food-chains, they are persistent in the environment and have the potential to elicit severe toxic effects at low concentrations. The scientific evidence surrounding the global distribution mechanisms of POPs further suggests an increased loading to the polar ecosystems. Consequently, this may pose a heightened health risk to arctic ecosystems and polar peoples who utilize fish, wildlife and marine mammals for sustenance.