BIOACCUMULATION POTENTIAL OF ORGANIC CONTAMINANTS IN AN ARCTIC MARINE FOOD WEB

by

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MRM. Simon Fraser University, 1999
BSc. Trent University, 1993

THESIS SUBMITTED IN PARTIAL FULFILLMENT OF
THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

In the School of
Resource and Environmental Management

SIMON FRASER UNIVERSITY

Spring 2006

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Degree: **Doctor of Philosophy**

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ABSTRACT

A chemical's octanol-water partition coefficient (K_{OW}) and octanol-air partition coefficient (K_{OA}) are important factors affecting environmental fate and bioaccumulation of persistent organic pollutants (POPs). This thesis involved an investigation of various organic chemicals (ranging in K_{OW} and K_{OA}) in a Canadian Arctic marine food web (53° 59' N, 76° 32' W) aimed to (i) determine levels of PCBs, organochlorine pesticides (OCPs), dialkyl phthalate esters (DPEs) and polybrominated diphenyl ethers (PBDEs) in organisms by high resolution gas-chromatography/high resolution mass-spectrometry (HRGC/HRMS), (ii) evaluate the extent of chemical biomagnification and (iii) identify relationships between biomagnification and influential physical-chemical properties such as K_{OW} and K_{OA}. The results show that recalcitrant Cl_5-Cl_7 PCBs (e.g., PCB 153 and 180) typically exhibited the greatest biomagnification potential and continue to be present at parts per million levels in Arctic biota, some 30 years post-regulatory action. Predator-prey biomagnification factors (BMFs) of PCB 180 ranged from approximately 11.5 in male ringed seals, 45.7 in male beluga whales and 106.3 in common eider ducks. Relatively polar chemicals such as β-HCH (log K_{OW} =3.8) tetrachlorobenzenes (log K_{OW} = 4.5) and β-endosulfan (log K_{OW} = 3.4) in some cases exhibited substantial biomagnification in seaducks and marine mammals. BMFs of β-HCH ranged from 5.2 in common eider ducks, 26.2 in male ringed seals and 50.5 in male beluga whales. No significant biomagnification of β-HCH was observed in invertebrates and fish, likely due to efficient respiratory elimination via gills to water. Extensive biomagnification of β-HCH in air-breathing animals (birds and marine mammals) is likely due the chemical's high resistance to metabolic transformation and slow respiratory elimination through air-exhalation because of its high K_{OA}, (i.e.,log K_{OA} = 8.9). While DPEs and PBDEs were detected at appreciable levels, they appeared to be biotransformed by organisms, demonstrated by very low BMFs and FWMFs compared to recalcitrant PCBs. Further evidence of biotransformation was supported by the detection of primary metabolites, monoalkyl phthalate esters (MPEs) and hydroxylated and methoxylated brominated diphenyl ethers (OH-BDEs / MeO-BDEs). Future regulatory initiatives should include chemical K_{OA} and the formation of potentially toxic metabolites as criteria for assessing the bioaccumulation potential of POPs.

Keywords: biomagnification, contaminants, POPs, K_{OA}, polybrominated diphenyl ethers, phthalate esters, metabolites, Arctic, marine, food web, beluga whales

DEDICATION

I dedicate this work to my wife, Laura and daughter, Erin, who help me keep things in perspective and keep me smiling through it all and to my parents Ken and Carol Kelly who have given great support along this long academic journey.

ACKNOWLEDGEMENTS

I wish to acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada (NSERC), Environment Canada's Northern Ecosystems Initiative (NEI), the Association of Canadian Universities for Northern Studies (ACUNS) and the Indian and Northern Affairs Northern Science Training Program (NSTP). Also, I wish to thank Makivik Coroporation, and Dr. Bill Doidge and Dr. Michael Kwan from the Nunavik Research Centre (NvRC) in Kuujjuaq Quebec for coordinating field sampling in E. Hudson Bay. I would also like to thank members of the REM Environmental Toxicology Research Group and staff at the Institute of Ocean Sciences (IOS) Regional Dioxin Laboratory (RDL). Special thanks in particular go to Joel Blair, Michael Gibbs and Anne Morin for there extensive work on this project. Lastly, I would like to thank Dr. Frank Gobas and members of my examining committee, including Dr. Michael Ikonomou, Dr. Peter Ross, Dr. Margo Moore, Dr. Leah Bendell-Young and Dr. Derek Muir for reviewing this thesis and their helpful comments and guidance to strengthen my work.

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GLOSSARY

Term	Definition
Bioaccumulation	The process where the chemical concentration in an aquatic organism achieves a level that exceeds that in the water as a result of chemical uptake through all routes of chemical exposure (e.g. dietary absorption, transport across the respiratory surface, dermal absorption, inhalation). Bioaccumulation takes place under field conditions. It is a combination of chemical bioconcentration and biomagnification.
Bioaccumulation Factor	The extent of chemical bioaccumulation in an aquatic organism is typically expressed by a chemical bioaccumulation factor (BAF), which is the ratio of the chemical concentrations in the organism (C_B) and the freely dissolved concentration in water (C_W): $BAF = C_B/C_{WD}$ The BAF for air-breathing animals (i.e., reptiles, birds and mammals) is defined as the ratio between chemical concentrations in the organism (C_B) and the surrounding ambient air (C_A): $BAF = C_B/C_A$ As chemical sorption to aerosols and particles can also occur in the atmosphere, the BAF for these air-breathing animals is best expressed in terms of the gas-phase chemical concentration in the air (C_{AG}): $BAF = C_B/C_{AG}$
Bioavailability	In general, bioavailability of a chemical substance in particular environmental media such as water, sediment and the organism's food can be viewed as the fraction of the chemical in the medium that is in a form, shape or condition which can be absorbed by the organism.
Bioconcentration	The process where the chemical concentration in an aquatic organism achieves a level that exceeds that in the water as a result of exposure of the organism to a chemical concentration in the water via the respiratory surface.
Bioconcentration Factor	The extent of chemical bioconcentration is usually expressed in the form of a bioconcentration factor (BCF) which is the ratio of the chemical concentration in the organism (C_B) and the freely dissolved concentration in water (C_{WD}): $BCF = C_B/ C_{WD}$
Biomagnification	The process where the chemical concentration in an organism achieves a level that exceeds that in the organism's diet due to dietary absorption. The extent of chemical biomagnification in an organism is best determined under laboratory conditions where organisms are administered diets containing a certain concentration of the chemical substance while there is no chemical uptake through other routes of exposure (e.g. uptake from water in fish) Biomagnification can also be determined under field conditions based on chemical concentrations in the organism and its diet.

Term	Definition
Biomagnification Factor	The extent of chemical biomagnification is usually expressed in the form of a biomagnification factor (BMF) which is the ratio of the chemical concentration in the organism and the concentration in the organism's diet: $BMF = C_B/C_D$ The chemical concentration in the organism (C_B) and the diet of the organism (C_D) are usually expressed in units of respectively gram of the chemical per kg of the organism and gram chemical per kg of food (lipid wt). Fugacity-based BMFs, the ratio of a chemical's fugacity in an organism (biota) to those fugacities observed in its diet (f_{BIOTA}/f_{DIET} or f_B/f_D), are also commonly used to express the extent of biomagnification.
Food web magnification factor (FWMF)	The extent of amplification of chemical concentrations over the entire food web can be expressed by a food web magnification factor (FWMF), derived from the slopes of the log-linear regressed concentrations in organisms (log C_B) versus trophic position (TP), i.e., $y= mx + b$. Specifically, $FWMFs = 10^m$.
Biotransformation	Biotransformation is the process where chemical substances undergo chemical reactions (chemical or biochemical) in biological organisms. The rate of transformation is usually expressed in terms of a rate constant.
Chemical half-life	This is the time (in hours, days or years) required to reduce the original concentration of a chemical substance in an organism to half the value of the original concentration. If the elimination rate involves transport and transformation processes that follow first order kinetics, the half-life time is related to the combined elimination rate constant k as the half-life time equals 0.693/k.
Dietary Uptake Efficiency or Chemical Assimilation Efficiency (E_D):	This is the fraction of the ingested chemical dose that is actually absorbed by the organism via the gastro-intestinal tract. It is usually expressed in terms of unitless fraction: E_D = absorbed dose /administered dose
Equilibrium	A chemical equilibrium can be viewed as a situation where the chemical substance is distributed among several environmental media (including organisms) according the chemical's physical-chemical partitioning behaviour where concentration ratios between media reflect the chemical's relative solubility in the media. Thermodynamically, an equilibrium is defined as a condition where the chemical's potential (also chemical activity and chemical fugacity) in the environmental media involved are the same. At equilibrium, there is no change in the chemical concentrations in the environmental media over time.
Trophic Level (TL)	A discrete value representing an organism's position in a food web. Typically determined using stable nitrogen isotope analyses, which essentially measures the energy transfer between different organisms.
Trophic position (TP)	The general position an organism occupies in the food web, which can be variable due to omnivorous feeding and seasonality effects on prey selection.

Term	Definition
Non Lipid Organic Matter (NLOM)	In general, hydrophobic organic contaminants such as PCBs and DDT are preferentially stored in lipids. However, when various environmental media exhibit very low lipid levels (e.g., vascular plants), other more prominent non-lipid components such as organic carbon,carbohydrates and proteins may provide this sorptive capacity
Hydrophobicity	It is commonly known that organic contaminants such as PCBs and DDT are "water-hating" chemicals (called hydrophobic chemicals) but rather tend to accumulate or are readily soluble in fat or biological lipids (which is equivalent to octanol). The degree of a chemical's hydrophobicity is directly related to the chemical's octanol-water partition coefficient (K_{OW}).
Octanol-water Partition Coefficient (K_{OW})	This is the ratio of the chemical concentrations in 1-octanol (C_O) and in water (C_W) in an octanol-water system that has reached a chemical equilibrium: $K_{OW}= C_O/C_W$. Since 1-octanol is a good surrogate phase for lipids in biological organisms, the octanol-water partition coefficient represents how a chemical would thermodynamically distribute between the lipids of biological organisms and water. It further represents the lipophilicity and the hydrophobicity of the chemical substance. It is usually referred to as K_{OW} or P, or in its 10-based logarithmic form log K_{OW} or log P, and it is unitless.
Octanol-air partition coefficient (K_{OA})	A quantitative property of organic substances which represents a chemical's tendency to partition into either air and/or octanol (assumed to be a perfect surrogate for biological lipids). K_{OA}'s of most priority pollutants typically range between 10^5 to 10^{12}(or log K_{OW}'s between 5 to 12). Essentially, a chemical's K_{OA} is the ratio of chemical solubility in lipids (i.e., octanol) and air. Organic chemical's with relatively low air solubility are thereby relatively non-volatile, exhibit high K_{OA}'s and will preferentially be retained in biological lipids. Chemicals with high K_{OA}'s indicate more fat-soluble chemicals, while chemical's with low K_{OA}'s will tend to volatilize into the surrounding air. A chemical's K_{OA} is therefore an important factor determining the accumulation of organic contaminants the tissues' of air-breathing animals.
Rate constant	Rate constants are quantities that describe the fraction of the total chemical mass or concentration in a particular medium or organism that is transported from and/or transformed in that medium or organisms per unit of time (e.g., 1/day).
Rate of Uptake & Elimination	The rate of uptake (or elimination) is the amount of chemical (in grams or moles) that is absorbed (or eliminated) by the organism per unit of time. It can be referred to as a flux and it has units of gram chemical per day or moles per day.
Steady-state	This is a situation where the flux of chemical into a medium or organism equals the flux out of that medium or organisms. At steady-state, there will be no net change in mass or concentration of the chemical in the medium or organism. A steady-state is different from an equilibrium in that it is achieved as a result of a balance of transport and transformation processes acting upon the chemical, whereas an equilibrium is the end result of a physical-chemical partitioning process.

Term	Definition
Fugacity	The term fugacity is translated from its Latin origin as "escaping tendency". Chemical fugacity is equivalent to chemical potential and can be measured as the partial pressure (in units of Pascals, Pa) that a chemical exerts within a given matrix. Chemical fugacity is commonly used as an effective surrogate to chemical concentration. The chemical concentration (C in units of mol/m^3) and the chemical fugacity (f in units of Pa) for a given media are related because C equals $f.Z$, where the fugacity capacity (Z in mol/m^3.Pa) indicates the ability of that media to retain chemical within its matrix. Z values are chemical specific, and are related to the phase in which the chemical is sorbed and the temperature of that phase. Fugacity can be derived from known values of C and Z using the equation $f = C/Z$. Chemical concentrations in different environmental media are measured directly (laboratory analysis), while Z values of a given chemical in different media are generally estimated, but can also be measured directly using equilibration experiments. 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 (i.e., a higher fugacity is required to provide movement of chemical from one phase to another). Environmental media are in a state of equilibrium when their respective fugacities are observed to be equal. In fugacity terms, biomagnification is defined as a situation in which chemical fugacity increases with increasing trophic level.
LRTAP	Long range transboundary air pollution protocol.
CEPA	Canadian Environmental Protection Act
UNEP	United Nations Environment Program
AMAP	Arctic Monitoring and Assessment Program
POPs	Persistent Organic Pollutants
PCB	Polychlorinated biphenyls
DDT	Dichlorodiphenyltrichloroethanes
PBDE	Polybrominated diphenyl ethers
PFOS	Perfluoro ocatane sulfonate
OH-BDEs	Hydroxylated brominated diphenyl ethers
MeO-BDEs	Methoxylated brominated diphenyl ethers
HCBz	Hexachlorobenzene
HCH	Hexachlorocyclohexanes
DPE	Diester phthalate
MPE	Monoester phthalate
GIT	Gastro intestinal tract
MMD model	Micelle mediated diffusion model
FFD model	Fat flush diffusion model
GIM model	Gastro intestinal magnification model
Poikilotherm	Cold-blooded animals (invertebrates, fish, amphibians, reptiles)
Homeotherm	Warm blooded animals (birds and mammals)
Water-ventilating ectotherm	Cold-blooded aquatic animals that respire water via gills (i.e., aquatic invertebrates, fish, amphibians etc.)

Term	Definition
Air-breathing endotherms	Warm blooded animals (birds and mammals)
BMF	Biomagnification factor
BAF	Biaccumulation factor
FWMF	Food web magnification factor
HRGC/HRMS	High resolution gas chromatography mass spectrometry
LRGC/LRMS	Low resolution gas chromatography mass spectrometry
MDL	Method Detection Limit
LOQ	Limit of Quantitifcation
SIM	Selective Ion Monitoring
Organism	Scientific Name (Genus spp.)
Primary producers	
Reindeer lichen	*Cladina Rangiferina*
Macro-algae	*Fucus gardneri*
Invertebrates & Fish	
Blue Mussels	*Mytilis edulis*
Arctic cod	*Boreogadus saida*
Four horned sculpin	*Myoxocephalus scorpioides*
Capelin	*Mallotus villosus*
Seabirds	
Common Eider Duck	*Somateria mollissima sedentaria*
White-winged Scoter	*Melanitta fusca*
Marine Mammals	
Ringed seal	*Pusa hispida*
Beluga whales	*Delphinapterus leucas*
Walrus	*Odobenus rosmarus*
Polar Bear	*Ursus maritimus*

CHAPTER 1

INTRODUCTION

1.1 Background

Persistent organic pollutants (POPs) are long-lived and potentially toxic organic chemicals that are resistant to chemical and biological degradation processes. The Arctic is a particularly vulnerable to POPcontamination as a consequence of the global distillation effect that involves long-range transport and condensation at the lower polar temperatures (*1*). POPs can biomagnify in food chains, resulting in chemical concentrations in high trophic level predators that can greatly exceed those concentrations in lower trophic prey species and the surrounding ambient environment (*2-7*). Although first-generation or legacy POPs such as PCBs and DDT were restricted from use in North America and Europe in the 1970's, these compounds are still present in food chains, worldwide. For example, recent studies have shown that biomagnification of PCBs, dioxins and furans in harbour seals (*Phoca vitulina*) and killer whales (*Orcinus orca)* of coastal British Columbia and Washington State has resulted in tissue concentrations in these animals that have in some cases surpassed the adverse affect levels of these compounds that are associated with immunosuppression and endocrine disruption in seals (*8,9*).

In recent years, chemicals of emerging concern, including polybrominated diphenyl ethers (PBDEs) used as flame retardants, phthalate esters (PEs) used as plasticizers and commercial pesticides such as endosulphan have been detected at appreciable levels in fish and wildlife (*10-14*). Also, fluorinated organic compounds such as pefluorooctanoic acid (PFOA) and perfluoro-octane-sulfonate (PFOS) have been recently detected in the tissues of birds and marine mammals from the Canadian Arctic and many other locations in North America, Europe and Asia, suggesting it to be a fairly ubiquitous contaminants (*15-18*). Further evidence suggests PFOS to be more acutely toxic to birds and mammals compared to legacy POPs such as PCBs or DDT (*19-27*). The available data suggest that many of these new chemicals of concern are persistent in the environment, potentially bioaccumulative and are toxic to organisms (i.e., PBT chemicals) and are hence generally regarded as "candidate" POPs. However, convincing evidence regarding

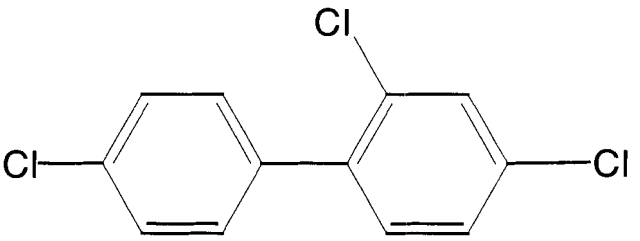
the environmental fate and bioaccumulation potential of these substances is still lacking. The latter is required by regulatory agencies to execute appropriate policy actions.

1.2 Different Classes of Environmental Contaminants.

Legacy POPs such as polychlorinated biphenyls (PCBs) and several organochlorine pesticides (OCPs) such as hexachlorobenzene (HCBz), mirex and dieldrin continue to be dominant environmental pollutants in Arctic ecosystems. Other chemicals of emerging concern include polybrominated diphenyl ethers (PBDEs), phthalate esters (PEs), synthetic musks, perfluorinated acids (PFAs) such as perfluorooctanoic acid (PFOA) and perfluoro-octane-sulfonate (PFOS), endosulphan and metabolic transformation products such as monoalkyl phthalate esters (MPEs) and hydroxylated and methoxylated brominated diphenyl ethers (OH- and MeO-BDEs). The molecular structures of various legacy POPs and other current-use chemicals of emerging concern are shown below in Figures 1.1 to 1.10.

Figure 1.1 Molecular structures of polychlorinated biphenyls

Cl_3 CB-28



Cl_4 CB-52

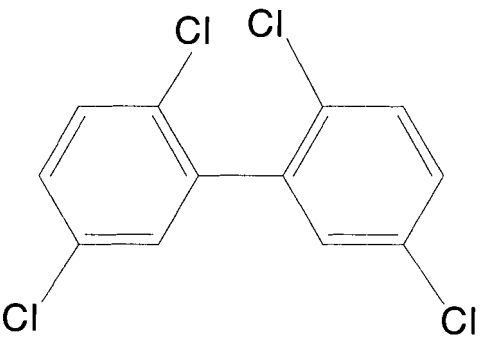
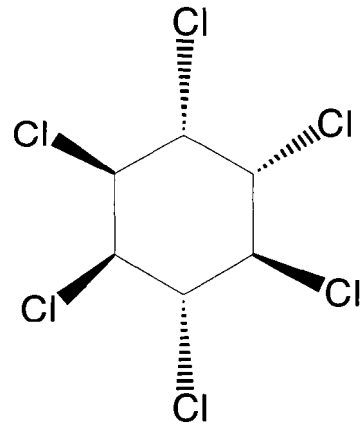
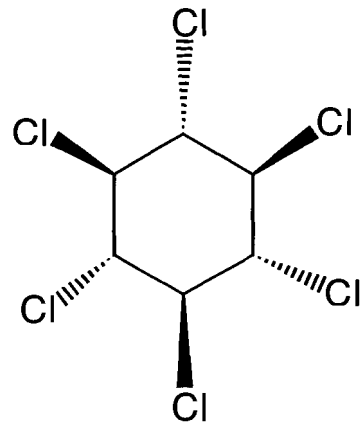


Figure 1.2 Molecular structure of hexachlorocyclohexanes

α-HCH



β-HCH



γ-HCH

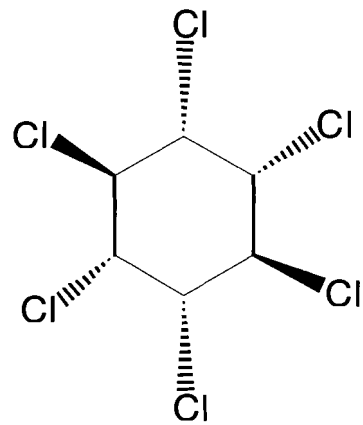
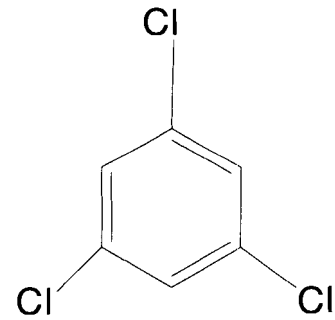
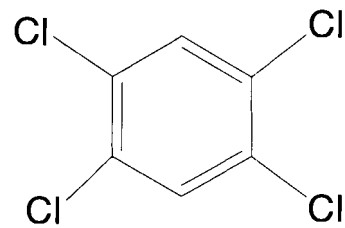


Figure 1.3 Molecular structure of chlorobenzenes

1,3,5 TriCBz



1,2,4,5 TeCBz



HxCBz

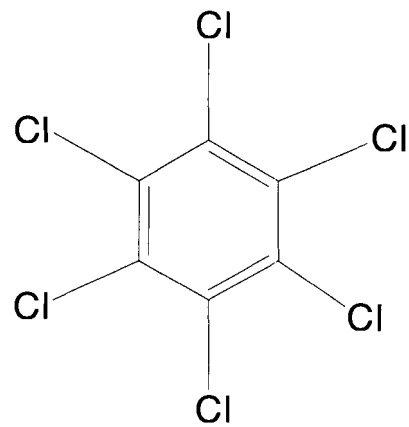
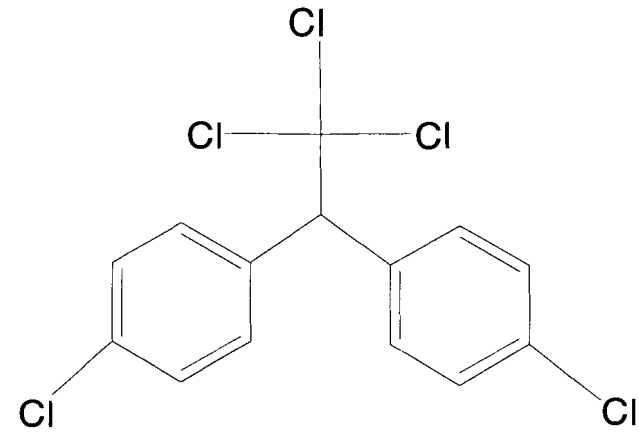
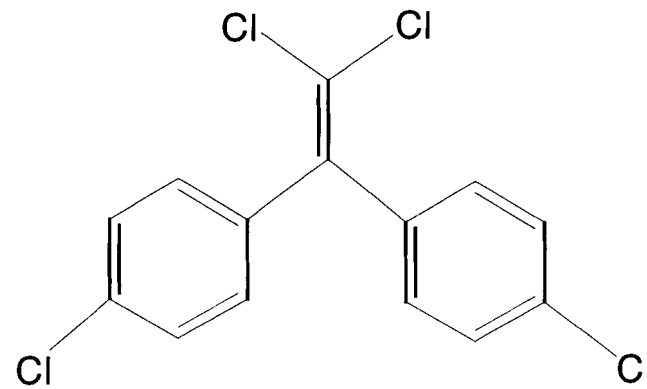


Figure 1.4 Molecular structure of DDTs

p'p' DDT



p'p' DDE



p'p' DDD

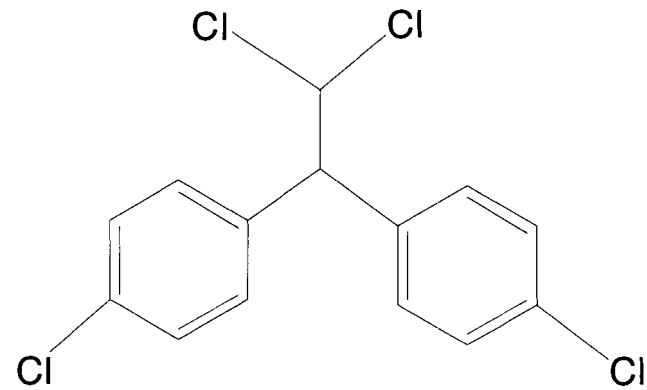
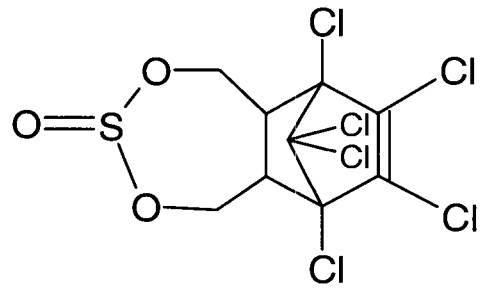


Figure 1.5 Molecular structure of cyclodiene pesticides

α- endosulfan



dieldrin

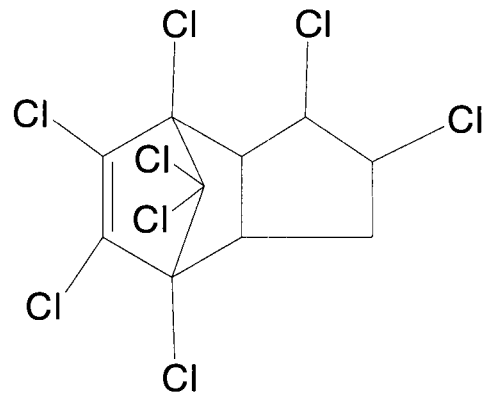
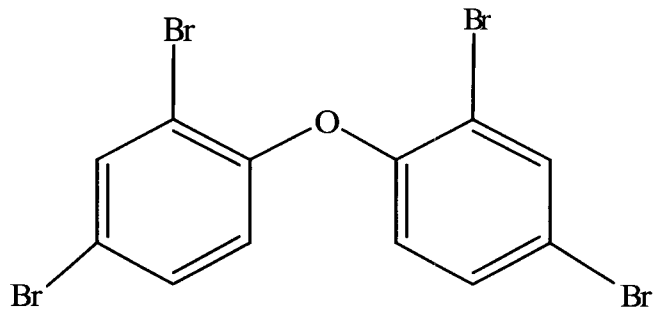
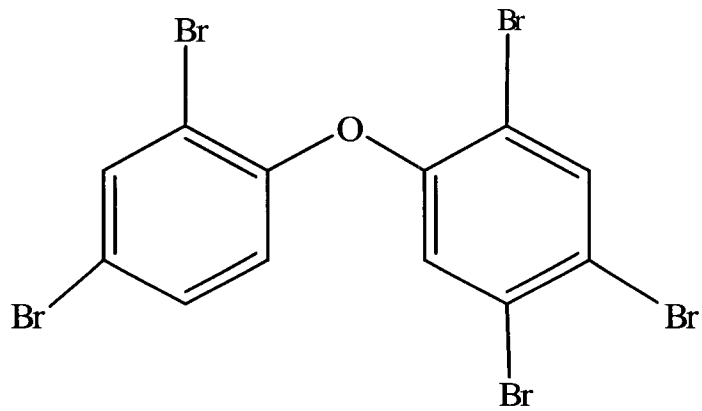


Figure 1.6 Molecular structure of polybrominated diphenyl ethers (PBDEs) and other brominated flame retardants (BFRs)

Br_4BDE 47



Br_5BDE 99



Tetrabromobisphenol A (TBBPA)

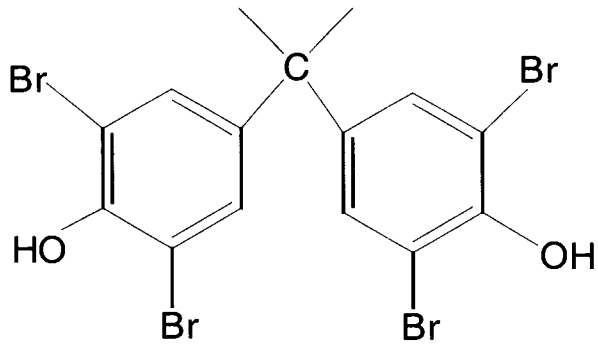
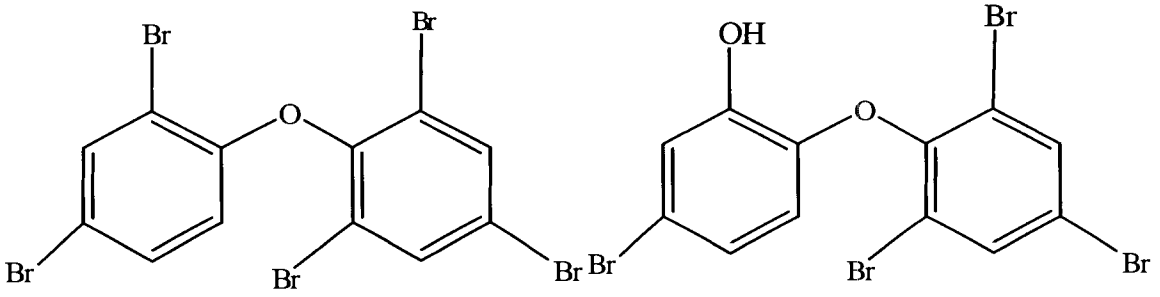


Figure 1.7 Molecular structure of hydroxylated (OH) and methoxylated (MeO) brominated diphenyl ethers

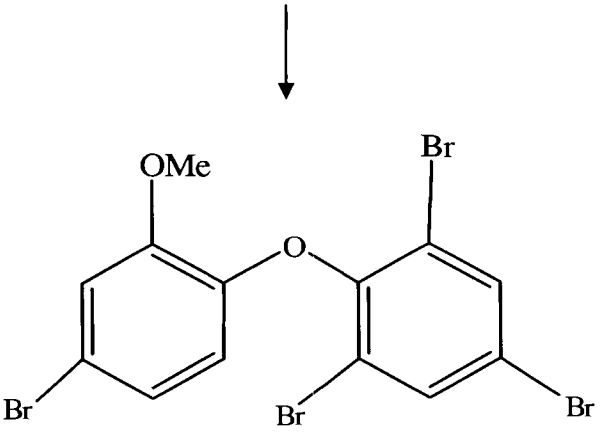
Parent BDE (Precursors)

Hydroxylated (OH) and Methoxylated (MeO) BDE metabolites



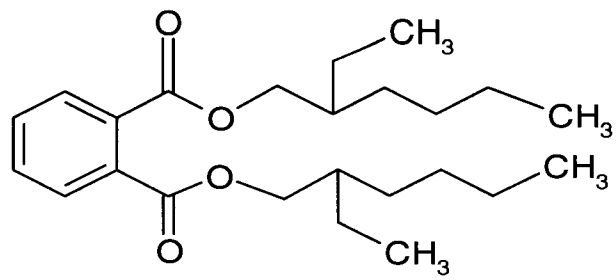
2,2',4,4',6 PBDE (BDE100)

2'OH-2,4,4',6 BDE 75

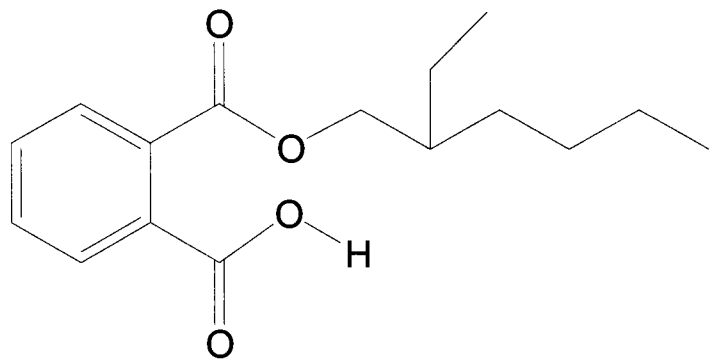


2'MeO-2,4,4',6 BDE 75

Figure 1.8 Molecular structure of Phthalate esters

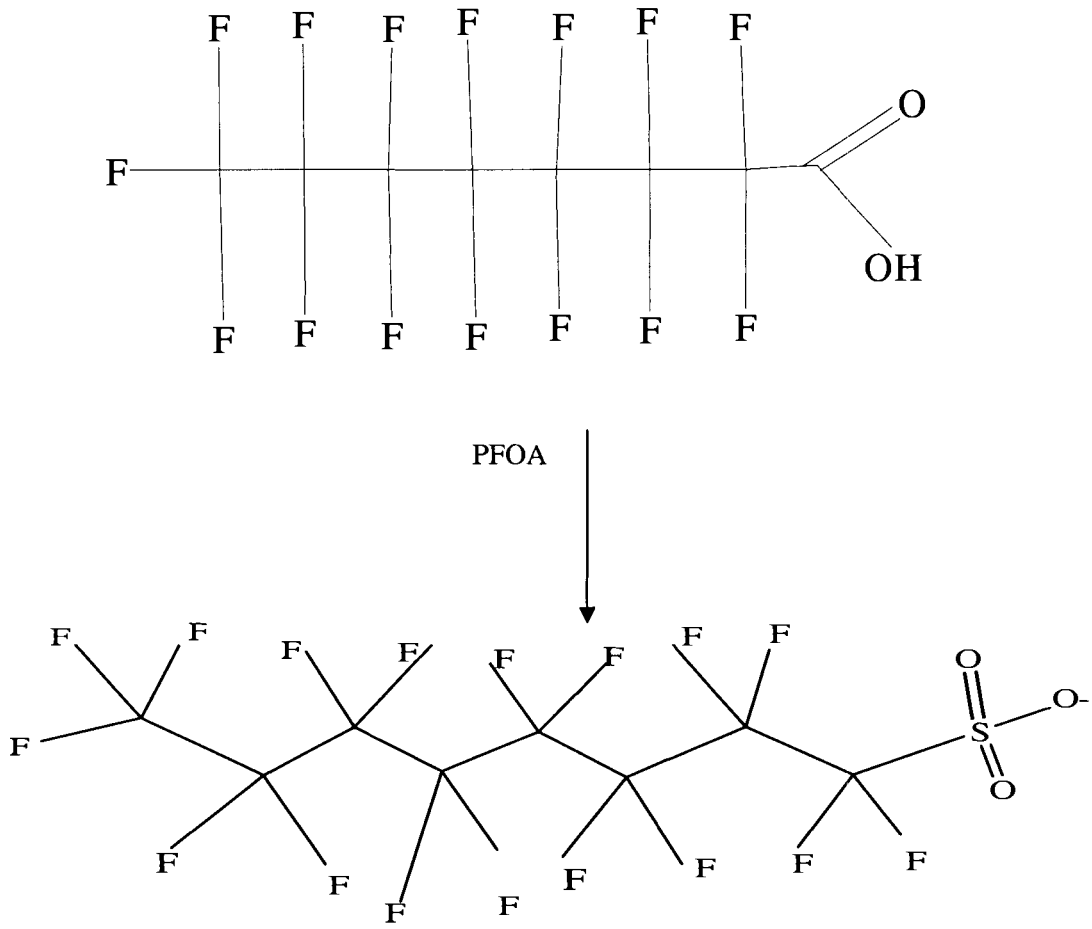


Di (2-ethylhexyl) phthalate (DEHP)



Mono (2-ethylhexyl) phthalate (MEHP)

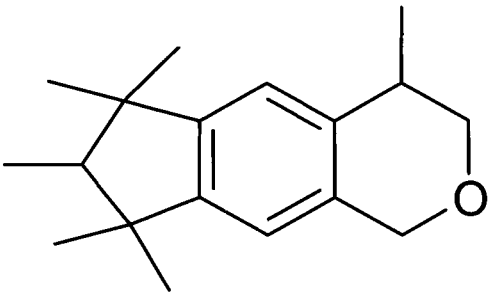
Figure 1.9 Molecular structure of perfluorinated acids



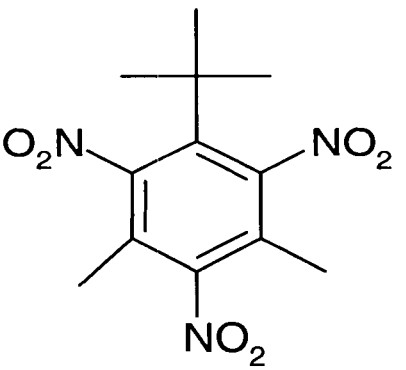
PFOS anion

Figure 1.10 Molecular structure of synthetic musks

Polycyclic musks
(e.g. Galaxolide, HHCB)



Nitro musks
(e.g. musk xylene)



1.3 Temporal Trends of Persistent Organic Pollutants in the Canadian Arctic.

In a recent paper we evaluated the temporal trends of various legacy POPs (e.g., PCBs and dioxins) and new chemicals of concern (e.g., PBDEs) in Canadian Arctic biota (*28*). Levels of past-use persistent organic pollutants (POPs) including PCBs, DDTs and toxaphene in Arctic biota have stabilized or are in decline since regulatory actions imposed during the 1970s and 80s (Figure 1.11). Conversely, "current-use" typically high production volume (HPV) chemicals such as PBDEs have exhibited exponential increases in Canadian Arctic biota. Increasing exposure to PBDEs in wildlife and humans is of concern due to documented adverse effects in laboratory studies, including impacts on neurobehavioural development, thyroid hormone levels and fetal toxicity/teratogenicity at doses in the low mg/kg body weight (*29*). Figure 1.12 illustrates time-series concentrations ($pg \cdot g^{-1}$ lipid wt.) of $\sum$PBDEs, and BDE congeners 47, 99 and 100 in two age-classes of male ringed seal blubber (0-15 and 16-35 years) during the period 1981-2003, along with a temporal trend of worldwide "Penta" BDE production (tonnes/year). Previous studies have demonstrated significant exponential increases of $\sum$PBDE, BDE47 and BDE-99 concentrations in ringed seals aged 0-15 years from Holman Island, NWT during the period 1981-2000, with 4-5 year doubling times (*30*). The most recent 2002 and 2003 samples for the 0-15 year old male ringed seals indicate that mean PBDE have not significantly changed since 2000 (confirmed by one-way Analysis of Variance (ANOVA) $\alpha = 0.05$) confirms no significant differences between mean PBDE concentrations of those animals during 2000, 2002 and 2003 samples. The determination of a statistically significant change in the 2003 time point for 0-15 y males was complicated by a low sample size (n=2) for those animals. Similarly, PBDE concentrations in the older 16-35 age-class of male ringed seals during 2002 and 2003 were comparable and not significantly different from those concentrations observed in 2000 samples. Thus, PBDE levels appear to be stabilizing. However, temporal trends of contaminant levels in marine mammals such as the Holman Island ringed seals is difficult to assess due to natural variability and is greatly affected by sample size and variance.

Figure 1.11 Temporal trends PCBs and PCDDs/PCDFs in ringed seals from Holman Island, NWT, Canada. Data are plotted on a log scale as $ng.g^{-1}$ wet weight for PCBs and $pg.g^{-1}$ wet weight for dioxins and furans. Bars represent arithmetic means and error bars standard deviations. Lines represent linear regression

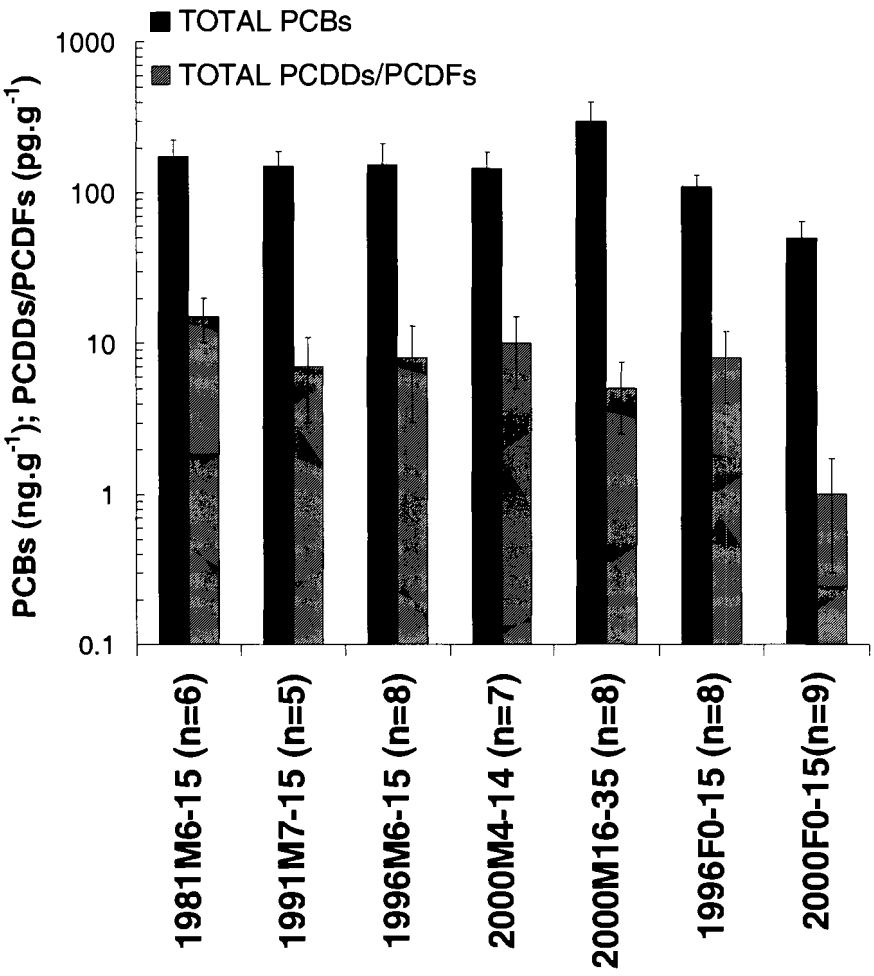
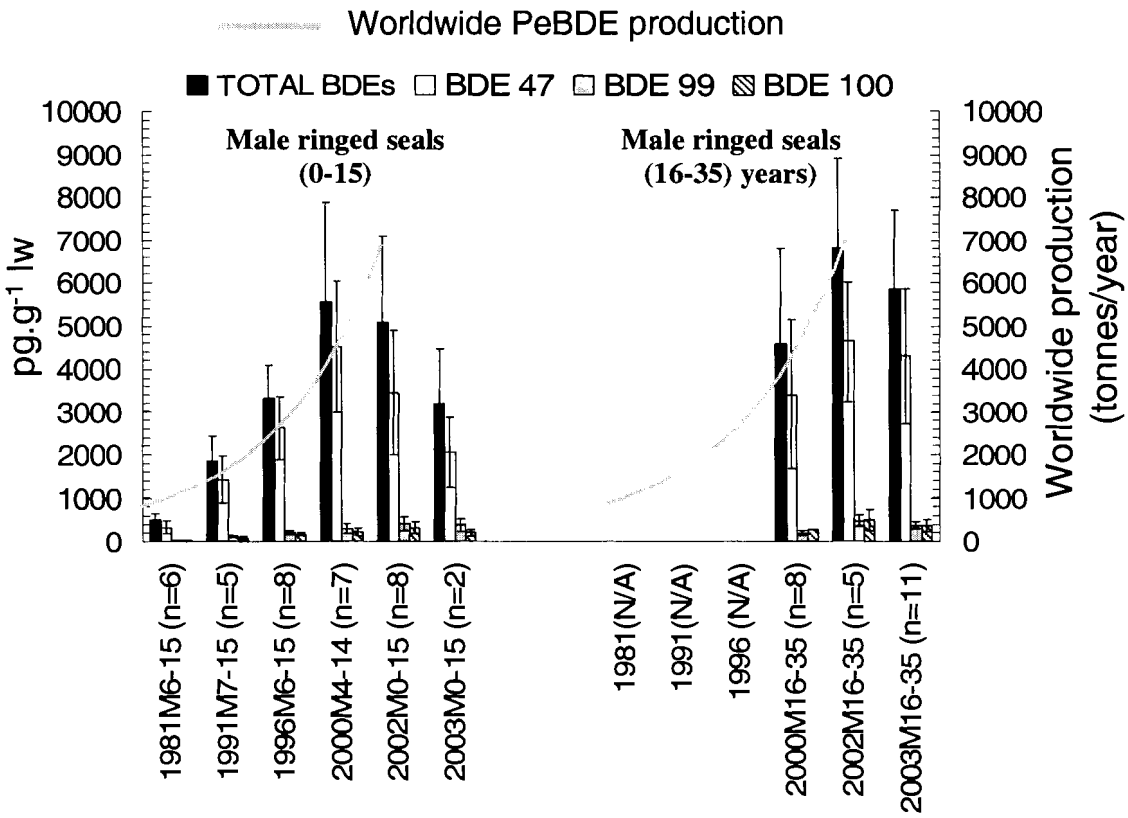


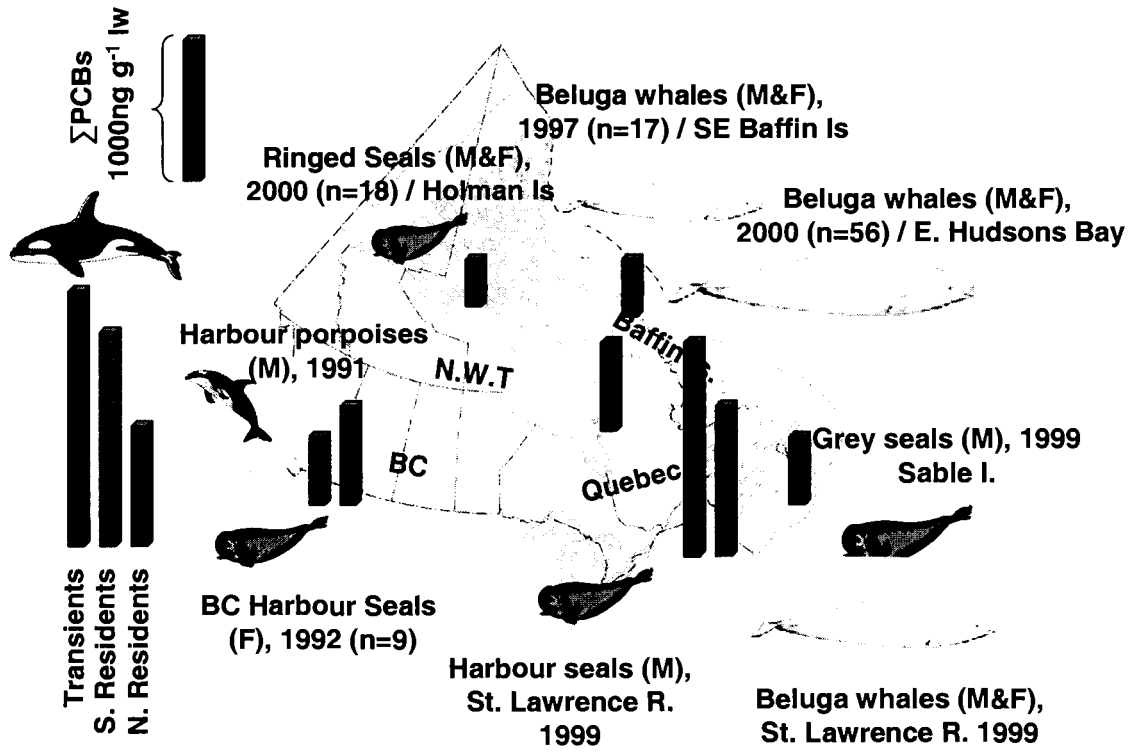
Figure 1.12 Temporal trends for PBDEs in male ringed seals along with worldwide PeBDE production trend (Line). Bars represent arithmetic means and error bars standard deviations.



1.4 Spatial Trends in Marine Mammals from Canadian coastal waters.

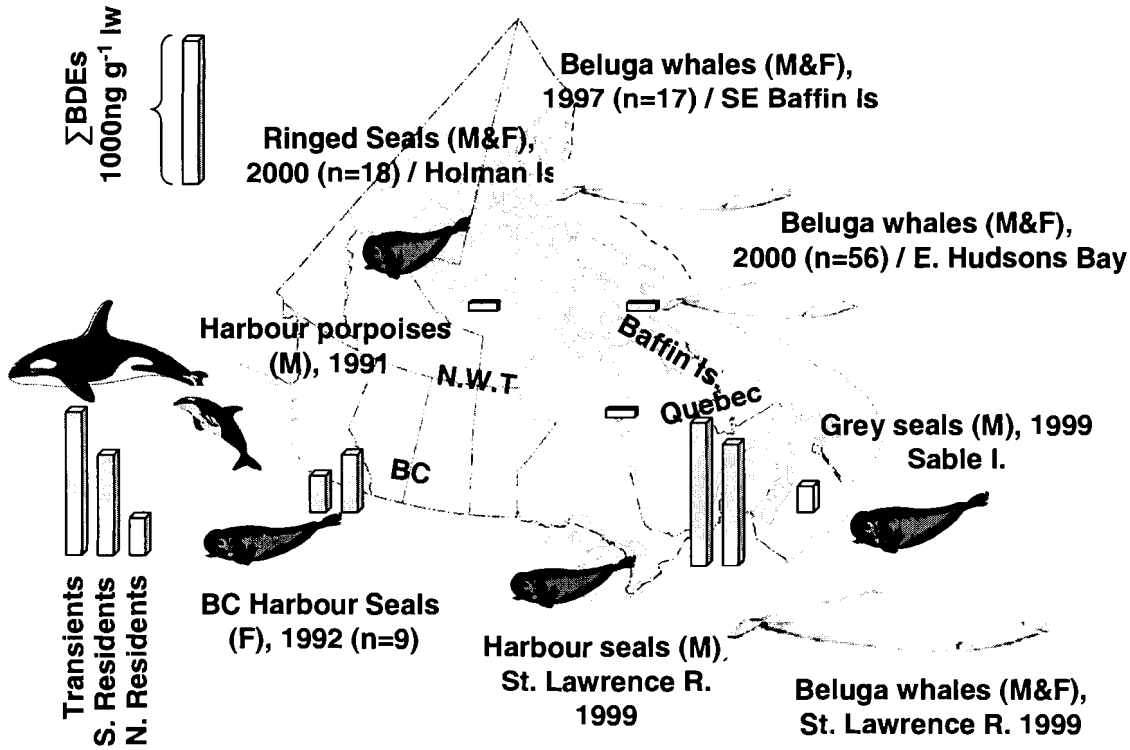
We recently presented data on spatial trends of legacy POPs and PBDEs in biota from Canadian coastal waters (*28,31*). Figure 1.13 and 1.14 summarizes the spatial variation in ∑PCB and ∑PBDE levels among various marine mammals collected off Canada's coasts from 1991-2000, respectively. The most contaminated animals were found in heavily urbanized / industrialized regions, namely, St. Lawrence Estuary (eastern Canada), and Georgia Strait (Lower Mainland of BC). For example, ∑PBDE levels in blubber of St. Lawrence beluga whales and southern resident (Georgia Strait) killer whales were approximately 700 and 650 $ng \cdot g^{-1}$ lipid wt., respectively (*32,33*). ∑PBDE concentrations in Canadian Arctic marine mammals, typically in the low $ng \cdot g^{-1}$ lipid wt. range, are about 100 times lower than ∑PBDE concentrations observed in marine mammals inhabiting more southern/urbanized Canadian waters. For example, SE Baffin Bay whales, E. Hudson's Bay beluga whales and Holman Island Ringed seals are all range between 5-10 $ng \cdot g^{-1}$ lipid wt. (*34*).

Figure 1.13 Average PCB concentrations (ng.g^{-1} lipid) in various marine mammals in Canadian waters.



Note: Map acquired with permission from woldatlas.com at <http://worldatlas.com/aatlas/world.htm>

Figure 1.14 Average PBDE concentrations (ng.g^{-1} lipid) in various marine mammals in Canadian waters.



Note: Map acquired with permission from woldatlas.com at <http://worldatlas.com/aatlas/world.htm>

1.5 Rationale: Towards a science-based approach to toxic substance management.

While legacy POPs such as PCBs and DDT were restricted from use in North America and Europe in the 1970's, many of these compounds are still present at ecologically sensitive levels in the Arctic and worldwide. Studies investigating the distribution of organic contaminants in the environment have consistently shown that non-polar/hydrophobic compounds such as PCBs, DDTs, toxaphene and mirex tend to biomagnify in the food chain, resulting in chemical concentrations in higher trophic level organisms that exceed those concentrations in the organism's prey. Assessing the potential of new and existing commercial chemicals to bioaccumulate in the food chain is an important component of persistent organic pollutant (POP) screening initiatives to identify substances that are persistent (P), bioaccumulative (B), toxic (T), and may undergo long-range transport (LRT). Environment Canada's *Toxic Substance Management Policy* (TSMP, 1995) and the recently revised *Canadian Environmental Protection Act* (CEPA, 1999) constitute the primary federal regulatory instruments for toxic substance management in Canada. Internationally, Canada has been extensively involved with instituting the United Nations Environmental Program (UNEP) long-range transboundary air pollution protocol (LRTAP) on POPs, which is designed to ban worldwide production of priority POPs. It has been well documented that a chemical's hydrophobicity (i.e., its octanol-water partition coefficient or K_{OW}) is a very important factor affecting the extent of bioaccumulation in aquatic organisms and food chains (*3,35,36,37,38*). Toxicokinetic studies in fish have shown that neutral organic substances with log K_{OW}'s < 5 do not biomagnify in aquatic organisms' due to elimination of these less hydrophobic compounds to water *via* the gills (*37, 39,40,41,42*). Based on this science, regulatory agencies in Canada, US and Europe have adopted management policies for POPs (e.g., Canada's Toxic Substance Management Policy, TSMP) that target only those chemicals exhibiting log K_{OW}'s > 5 as being "bioaccumulative". Thus, for the purpose of screening chemicals for PBT and LRT, Canada's TSMP and the UNEP POPs protocol both use a hydrophobicity threshold criterion (octanol-water partition coefficient or $K_{OW} > 10^5$) to identify bioaccumulative substances.

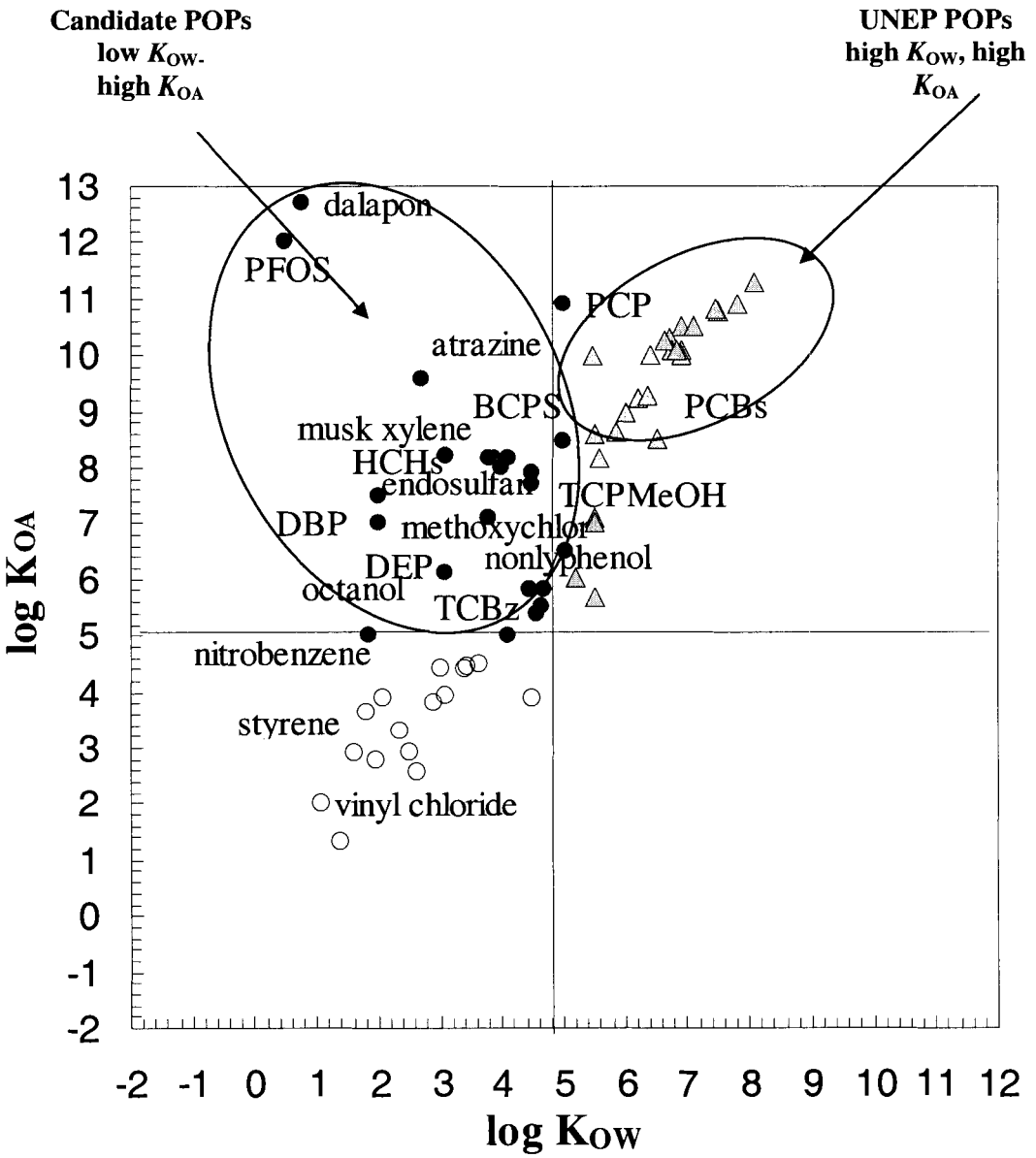
However, our recent studies of POPs bioaccumulation in lichen-caribou-wolf food chains of the Canadian Arctic (*43,44*) indicate that relatively polar chemicals which exhibit log K_{OW}'s < 5 such as β-hexachlorocyclohexane (β-HCH), chlorobenzenes (CBz) and endosulphan, that also exhibit a high octanol-air partition coefficient, K_{OA} (log K_{OA}'s > 6) can biomagnify in air-breathing

animals. The data demonstrate that regardless of the chemical's K_{OW}, air breathing animals apparently cannot eliminate non-metabolizable, "high" K_{OA} compounds because those substances are relatively non-volatile and tend to partition to lipids rather than exhaled to air. As a result, the current K_{OW} threshold criterion (log $K_{OW} > 5$) used for assessing chemical bioaccumulation potential does not adequately reflect the bioaccumulation potential of polar-non-volatile compounds (PNVs) in air-breathing animals because lipid-to-air elimination (rather than lipid-to-water elimination) counteracts gastro-intestinal uptake and biomagnification of non-metabolizable substances in those organisms (*43,44*). A chemical's K_{OA} may therefore better assess chemical bioaccumulation potential than K_{OW} for non-metabolizable organic substances in air-breathing animals. Currently, a considerable number of unregulated commercial chemicals (low K_{OW} - high K_{OA} compounds) may effectively biomagnify in sensitive top-predator birds and mammals, including humans.

As demonstrated in Figure 1.15, many new chemicals of concern such as dialkyl phthalate esters (DPEs), Bisphenol A, Tetrabromobisphenol A (TBBA), Hexabromocyclododecane (HBCD), synthetic musks, dicofol, endosulphan and perfluorinated acids are "low" K_{OW} – "high" K_{OA} substances and hence may, in the absence of metabolic transformation, preferentially biomagnify and accumulate significantly in the tissues of air-breathing animals such as terrestrial reptiles, birds and mammals, including humans. These polar non-volatile (PNV) chemicals, exhibiting log K_{OW}'s < 5 and log K_{OA}'s > 6 are not considered "bioaccumulative" and have therefore been excluded from the Stockholm Convention on POPs. However, in some cases these PNV compounds can exhibit persistence and bioaccumulation comparable to that of hydrophobic POPs such as PCBs, DDTs and mirex (e.g., β-HCH). Also, many of these new chemicals of concern are suspected endocrine disrupting chemicals (EDCs) and have demonstrated immunotoxic, teratogenic or carcinogenic effects in organisms. Some of these commercial chemicals are high production volume chemicals (HPVs). For example, dialkyl phthalate esters (commercial plasticizers) have current annual production volumes in excess of 5 million tonnes globally (*13*). Many of these compounds also appear to be relatively persistent and exhibit physical-chemical properties that permit long-range transport to remote alpine and polar regions. Legacy POPs such as PCBs and DDT are taking decades to purge from the Canadian environment. To avoid similar environmental hazards, current toxic substance management initiatives must effectively identify those substances that pose a risk to ecosystem health due to food chain contamination. This is particularly important in the Canadian Arctic, where local communities rely on "traditional foods" such as caribou, fish, seals, walrus and beluga whales for subsistence throughout the year.

The objective of this study is to investigate the ability of various polar and non-polar organic contaminants (ranging in K_{OW} and K_{OA}) to accumulate in an Arctic marine food web, with focus on bioaccumulation behaviour in air-breathing endotherms such as seaducks and marine mammals. The study's aim is to provide information regarding the bioaccumulation behaviour of new chemicals of concern in the environment, which will help to assess ecosystem health and also aid future decisions regarding the continued production and use of those substances.

Figure 1.15 LogK_{OW} versus LogK_{OA} for various organic chemicals. Open circles represent low K_{OW} – low K_{OA} (polar/volatile) chemicals. Solid circles represent polar non-volatiles (PNVs), low K_{OW} – high K_{OA} chemicals. Solid triangles represent hydrophobic nonpolar/non-volatile chemicals, high K_{OW} – high K_{OA}.



1.6 Hypothesis and Objectives

The purpose of the proposed study is to investigate the extent of food chain bioaccumulation of POPs of emerging concern, which due to their polarity (i.e., log $K_{OW} < 5$) are not recognized as being bioaccumulative by current CEPA 1999 regulations but which because of their low volatility (i.e., log$K_{OA} > 4$) are expected to biomagnify in marine mammals. Legacy POPs such as PCBs continue to be an ecological stressor to the health of marine ecosystems, some 30 years after imposing regulatory measures restricting their manufacturing and/or usage. To prevent the production and use of chemical substances that, through their persistence and bioaccumulation in the food chain, pose threats to environmental and human health, it is important to understand the bioaccumulation mechanisms and to identify the role that physical-chemical properties play in the chemical's ability to biomagnify in the food chain. Knowledge of the relationship between physical-chemical properties and their bioaccumulation behaviour will assist in the development of more effective regulation of chemical usage in Canada and elsewhere in the world, which ultimately plays a key role in safeguarding animal and ecosystem health.

The hypothesis is that relatively polar substances (i.e., chemicals with low K_{OW}'s) that are relatively non-volatile (i.e., high K_{OA}'s) and non-metabolizable can substantially biomagnify in Arctic marine food chains. The objectives of the current study are:

1.7 Short Term Objectives

(1) to measure concentrations of a number of legacy and "candidate" POPs in selected organisms of a marine web from eastern Hudson's Bay, including tissue samples of water ventilating ectotherms (bivalves, fish) and air-breathing endotherms (i.e., eider ducks, beluga whales, ringed seals).

(2) to document the extent of biomagnification for those substances in this food web by comparing chemical concentrations at each trophic level.

(3) to identify the physical-chemical properties of substances that can biomagnify and attain elevated levels in top-predator air-breathing animals of an Arctic marine food web.

1.8 Long Term Objectives

By studying the bioaccumulation of various polar and non-polar organic chemicals (ranging in K_{OW} and K_{OA}) in an Arctic marine food web we aim to (i) demonstrate the degree of chemical biomagnification in a marine food web that includes marine seabirds and mammals and (ii) identify the physical-chemical properties that influence chemical biomagnification in marine food chains. Identifying the effects of critical chemical properties (e.g., K_{OW} and K_{OA}) on chemical biomagnification that will aid regulatory agencies in assessing the bioaccumulation potential of manufactured chemicals. The results are expected to identify the food chain transfer pathways that may be implicated in health effects of POPs in susceptible species such as beluga whales. This work aims to provide information regarding the bioaccumulation behaviour of new chemicals of concern in the Arctic environment, which may help to assess the health status of the eastern Arctic marine ecosystem and the related human health impacts to northern Aboriginal populations. These findings may therefore aid future international initiatives/controls regarding those substances.

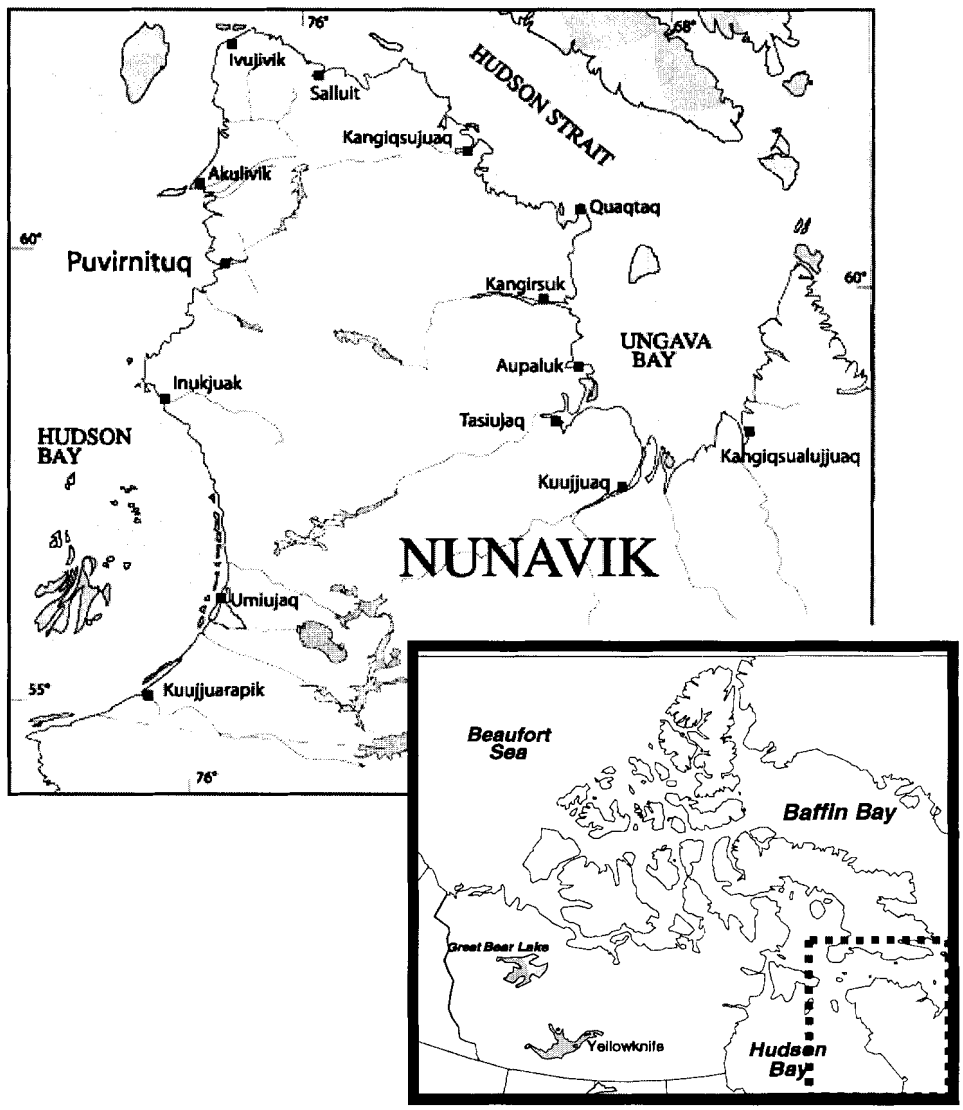
1.9 Project Design and Methodologies

The project involves a bioaccumulation field-study of organic chemicals in selected organisms of the eastern Hudson Bay (EHB) coastal marine food web to identify contaminant sources and quantify biomagnification factors (BMFs) of these compounds in free-ranging seabirds and marine mammals. The focus of the study is targeted at assessing the bioaccumulation potential of various new chemicals of emerging concern. These include dialkyl phthalate esters (DPEs), monoalkyl phthalate esters (MPEs) polybrominated diphenyl ethers (PBDEs) and their hydroxylated (OH-BDEs) and methoxylated derivatives (MeO-BDEs), chlorobenzenes (CBz), hexachlorocyclohexanes (HCHs) and endosulphan.

1.9.1 Sample collections.

During the months of May to August between 1999 and 2003 various biological samples were collected along the eastern Hudson Bay coastline in close proximity to the Inuit village Umiujaq (64° 15′N 113° 07′ W), (Figure 1.16). Biota samples included lichens (*Cladina rangiferina*), inter-tidal macro-algae (*Fucus gardneri*), blue mussels (*Mytilis edulis*), fish: Arctic cod (*Boreogadus saida*), capelin (*Mallotus villosus*) and sculpin (*Myoxocephalus scorpioides*), and tissues and organs of harvested common eider ducks (*Somateria mollissima sedentaria*) and marine mammals including beluga whales (*Delphinapterus lecuas*) and ringed seals (*Pusa hispida*). Samples of bottom sediments were collected using a petit ponar grab at between 25-80 meter depths. Tissue samples (stomach contents, liver, muscle and blubber) of harvested seaducks and marine mammals were collected as part of northern Quebec Inuit subsistence hunts. Beluga whale samples were mainly collected from the E. Hudson Bay beluga stock summering habitat, in close proximity to the Nastapoka River estuary and the Inuit village of Umiujaq (64° 15′N 113° 07′ W) during the summer subsistence hunts. Samples of ringed seal samples were obtained from various locations across northern Quebec (Nunavik) and Labrador (Makovik). Extraordinary care was employed during field collections to avoid sample contamination by dialkyl phthalate esters (DPEs) and polybrominated diphenyl ethers, which are abundant in various consumer products (e.g., plastics, polyurethane foam). Firstly, samples of E. Hudson Bay sediments, macro-algae, lichens, fish and beluga whales were collected in individual 50 mL solvent-rinsed glass jars with aluminum foil lined caps (no plastics were used) and stored at –30 °C prior to chemical analysis. Secondly, laboratory gloves were not used (due to potential DPE contamination). Thirdly, tissue samples from fish and beluga whale tissues were excised using solvent-rinsed disposable scalpel blades. However, ringed seal (blubber) and seaduck (liver and adipose tissue) samples, graciously provided by D.C.G. Muir (Environment Canada's, National Water Research Institute, NWRI, Burlington, ON) and M. Kwan (Nunavik Research Centre, NvRC, Kuujuaq, Quebec), respectively, were collected using plastic sampling bags and hence were not analyzed for plasticizers (i.e., phthalate esters). Appendix 1 summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex and age.

Figure 1.16 Map of study area.



Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

1.9.2 Food web characterization and designation of organism trophic levels.

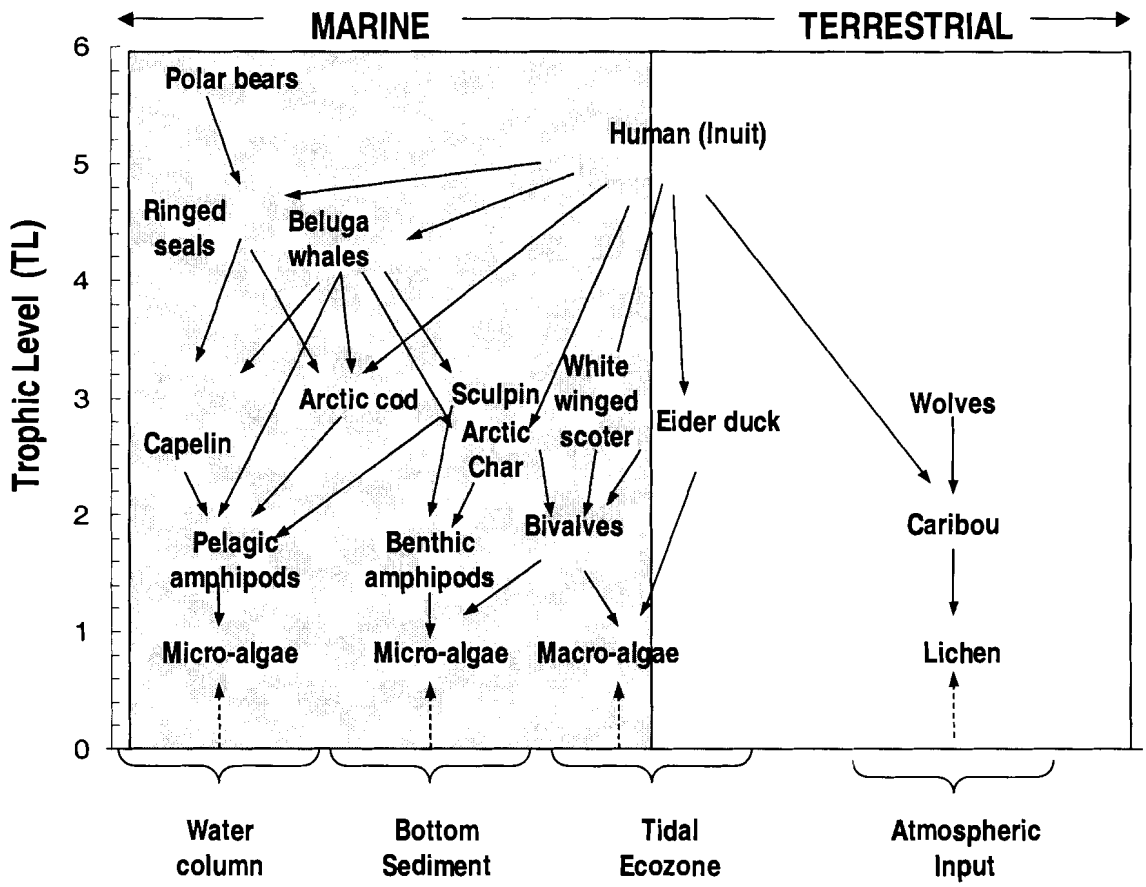
Figure 1.17 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been described by extensive ^{15}N and ^{13}C isotope enrichment analyses involving numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of $TL = 1 + (\delta^{15}N - 5.4)/3.8$. More recent studies using $\delta^{15}N$ measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 summarizes these previous $\delta^{15}N$ measurements and TL ranges for the various organisms within these Arctic marine food webs. While $\delta^{15}N$ values for the various marine organisms undoubtedly vary geographically (even between Arctic systems), the trophic level estimates based on $\delta^{15}N$ measurements are quite comparable between these Arctic marine food webs. For example, Arctic cod and sculpin are shown occupy trophic level range between 3.3 to 3.6. Similarly, fish-eating Arctic resident species such as ringed seals and beluga whales from across the Canadian Arctic have been determined to occupy a TL range of approximately 4.1 to 4.6. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific $\delta^{15}N$ measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

Table 1.1 Compilation of previous studies involving δ15N measurements and trophic level estimates of Arctic biota.

	Lancaster Sound (Canada) Hobson and Welch (*45*) $\delta^{15}N$ ‰ ± SD/ (TL)	**Northwater Polyna (Canada), Hobson and colleagues** (*47,48*) $\delta^{15}N$ ‰ ± SD/ (TL)	**Barents Sea (Norway) Hop et al.,** (*46*) $\delta^{15}N$ ‰ ± SD/ (TL)	**Beaufort-Chukchi Seas, (Alaska, USA) Hoekstra et al,** (*49*) $\delta^{15}N$ ‰ ± SD/ (TL)
Primary Producers				
Ice algae	7.5 ± 0.1 (1)	5.1 ± 0.3 (1)	-	-
L. solidungula (Kelp)	7.1 ± 1.3 (1)	-		-
L. longicruris (Kelp)	7.6 ± 0.9 (1)	-	-	-
Invertebrates				
Hiatella arctica (Bivalve)	9.8 ± 0.5 (2.2)	9.1 ± 0.7 (2.3)	-	-
Calanus sp. (copepod)	9.2 ± 0.5 (2.0)	7.7 ± 0.1 (2.0)	8.1 ± 0.1 (2.0)	9.8 ± 0.9 (2.0)
Parathemisto libellula (Pelagic amphipod)	11.7 ± 0.7 (2.7)	9.7 ± 0.1 (2.5)	7.1 (1.7)	-
Gammarus wilkitzkii (Ice amphipod)	11.5 ± 0.3 (2.6)	-	7.6± 0.3 (1.9)	-
Mysids	10.3± 0.3 (2.1)			-
Krill (*Thyanoessa sp.*)	-	-	8.5± 0.1 (2.1)	-
Fish				
Arctic Char (*Salvelinus sp.*)	-	-	-	13.8± 0.3 (3.1)
Arctic cod (*Boreogadus saida*)	15.2 ± 0.7 (3.6)	14.0 ± 0.2 (3.6)	13.0 ± 0.3 (3.3)	14.5 ± 0.4 (3.3)
Sculpin (*Myoxocephalus sp*)	15.2 (3.6)	-	-	15.4 ± 0.9 (TL=3.5)
Birds				
Brunnich's guillemot (*Uria lomvia*)	15.8 ± 0.7 (4.1)	14.1 ± 0.1 (4.0)	11.8 ± 0.2 (3.3)	-
Black guillemot (*Cepphus grylle*)	15.4 ± 0.7 (3.9)	13.7 ± 0.2 (3.9)	13.4 ± 0.2 (3.8)	
Kittiwake (*Rissa tridactyla*)	15.4 ± 0.9 (4.0)	13.7 ± 0.2 (3.9)	12.7 ± 0.2 (3.6)	-
Glaucous gull (*Larus hyperboreus*)	17.0 ± 0.9 (4.4)	16.2 ± 0.3 (4.6)	15.2 ± 0.1 (4.3)	-
Marine Mammals				
Walrus (*Odobenus rosmarus*)	12.5 ± 0.6 (2.9)	-	-	-
Bowhead Whale (*Balaena mysticetus*)	-	-	-	13.5 ± 0.1 (2.8)

	Lancaster Sound (Canada) Hobson and Welch (*45*)	Northwater Polyna (Canada), Hobson and colleagues (*47,48*)	Barents Sea (Norway) Hop et al., (*46*)	Beaufort-Chukchi Seas, (Alaska, USA) Hoekstra et al, (*49*)
Beluga whale (*Delphinapterus leucas*)	16.6 ± 0.6 (3.9)	16.0 ± 0.2 (4.1)	-	16.4 ± 0.1 (3.8)
Beareded Seal (*Erignathus barbatus*)	16.8 ± 0.2 (4.0)	-	-	16.6 ± 0.1 (3.8)
Ringed Seal (*Phoca hispida*)	17.3 ± 1.1 (4.1)	17.5 ± 0.2 (4.6)	14.5 ± 0.4 (3.8)	16.6 ± 0.1 (4.1)
Harp Seal (*Phoca groenlandica*)	-	-	13.6 ± 0.2 (3.4)	-
Polar Bear (*Ursus maritimus*)	21.1 ± 0.6 (5.1)	-	-	-

Figure 1.17 Schematic illustration of organisms (including humans) comprising the Canadian Arctic marine and terrestrial food webs and associated trophic level (TL) and feeding interactions.



1.9.3 Ultra-Trace Chemical analyses

Four separate cleanup and quantification methodologies for environmental and biological samples have been developed for the analysis of (i) PCBs and organochlorine pesticides (***Chapter 3*** and ***Chapter 4***), PBDEs (***Chapter 6***) by high resolution gas-chromatography –mass spectrometry (HRGC/HRMS), (ii) Dialkyl phthalate esters by low resolution gas chromatography mass spectrometry (LRGC/LRMS), (***Chapter 5***), (iii) monoalkyl phthalate esters (MPEs, i.e., metabolites of DPE parent compounds) by LC/ESI-MS (***Chapter 5***) and (iv) Hydroxylated and Methoxylated PBDEs by HRGC/HRMS (***Chapter 7***).

1.9.4 Data analyses

Selected physical-chemical properties including molecular weights (MW, g mol^{-1}), log octanol water partition coefficient log K_{OW}, log octanol-air partition coefficient log K_{OA}, Henry's Law Constants (H, Pa m3 mol^{-1}) and water solubility (C_WSoL, ng·L^{-1}) were compiled for PCBs and OC pesticides using references *50-56* and are summarized in Appendix 2. To enable direct comparisons of chemical concentrations between various environmental media and organisms it is important to correct chemical concentration data to a common unit expression. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~ 5 - 98%) , wet weight chemical concentrations (C, ng·g^{-1} ww) were expressed solely on a lipid weight basis by the equation: C_L = C ww. ÷ ϕL in units of ng·g^{-1} lipid. For matrix with very low lipid fractions (ϕL < 1%), such as sediments, vegetation and algae tend to solubilize organic contaminants in non-lipid biomolecules such as organic carbon (OC) or non-lipid organic matter (NLOM) rather than in extractable lipids (*13,57,58,59*). Thus, sediments, macro-algae and lichens were normalized to a lipid equivalent fraction (ϕLeq) using the equation CLeq = Cww ÷ ϕLeq. Lipid equivalent fractions (ϕLeq) for sediments were determined following reference (*35*) such that ϕ Leq = ϕL + 0.35ϕ_{OC}, where the constant 0.35 represents findings that organic carbon has approximately 35% sorptive capacity of octanol. For macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and NLOM (ϕ_{NL}) fractions following the equation: ϕLeq = ϕL + 0.035ϕ_{NL}, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42, 44*). Because chemical concentrations exhibited log-normal distributions the data were transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for individual compounds and

compound class summations for the various samples collected and analyzed as part of the present study (i.e., sediments, lichens, macro-algae, bivalves, fish, beluga whales and ringed seals). In addition, we also compiled literature reported concentration data for PCBs and OC pesticides in Canadian Arctic biota, including invertebrates (*4*), walrus (*Odobenus rosmarus*) (*60*) polar bears (*Ursus maritimus*) (*61*), barren-ground caribou (*Rangifer tarrandus*) (*43,62,63*), wolves (*Canis lupus*) (*43,63*) and northern Quebec Inuit women (i.e., breast milk samples from references *63,64*) to compare contaminant concentrations, profiles and BMFs in various wildlife species and humans that generally subsist within the same food web.

PCB congeners were categorized by planarity and Cl-substitution patterns, following classifications presented by Boon and colleagues (*65*): i.e., Group I CBs, congeners without vicinal hydrogen atoms are generally non-metabolizable CBs; Group II, congeners with vicinal *ortho-meta* H atoms and 2 *ortho* Cls have a limited metabolism potential in some organisms; Group III, same as II but with 1 *ortho* Cl can be metabolized by induction of methylcholanthrene (MC) type isozymes of the cytochromeP450 monooxygenase enzyme family (i.e., CYP 1A enzymes); Group IV, congeners with vicinal *meta-para* H atoms and ≤ 2 *ortho* Cls can be metabolized by induction of phenobarbitol (PB) type isozymes (i.e., CYP 2B enzymes); Group V, same as IV but with 3 *ortho* Cls may also induce CYP 2B type metabolism. A total of 169 di-ortho and mono-ortho substituted PCB congeners were analyzed (see Appendix 2). Due to several coeluting di-ortho (DO) and mono-ortho (MO) PCBs we have summarized a total of 148 PCB congeners. When environmentally dominant CB congeners coeluted with environmentally irrelevant congeners, we have for the purposes of this study, assumed the coeluting concentration as the single dominant compound. For example, CB153/132 concentrations (coeluting congeners in HRGC/HRMS method) are expressed solely as a CB153 concentration because of that congeners dominant contribution in environmental and biological samples. Specifically, this assumption was used for CBs 52, 101, 118 and 138. One-Way Analyses of Variance (ANOVA) and Tukey's HSD comparison tests were performed on calculated log-transformed concentrations to evaluate differences between mean chemical concentrations observed in sediments, invertebrates, fish and beluga whales.

1.9.5 Evaluative parameters for assessing chemical bioaccumulation potential.

Using lipid corrected chemical concentrations in biota calculated several evaluative parameters commonly used to assess the bioaccumulation potential of organic contaminants. The first

parameter is the food web magnification factors (FWMFs), a marker of cumulative bioaccumulation across the entire food-web, was determined from the log-linear regression between $\log_{10}$ analyte concentrations in biota (C_B) and trophic level (TL):

$$\text{Log } C_B = (m \times TL) + b \quad (1)$$

where m and b are the empirical slope and y-intercept, respectively (*13*). Following reference *13*, FWMFs are calculated as the antilog of the slope (m), (i.e., FWMF $=10^m$). We determined separate FWMFs for (i) water-ventilating ectotherms (invertebrates and fish), (ii) air-breathing endotherms (birds and marine mammals) and (iii) the overall food web. FWMFs > 1 indicate trophic transfer and step-wise amplification in the food web, while FWMFs near or less than unity represent trophic dilution.

The second and third parameters were predator/prey biomagnification factor (BMFs) and bioaccumulation factors (BAFs), respectively. BMFs are the ratio of the lipid corrected concentrations in a given predator (C_B, lipid) and its prey (C_D, lipid), i.e., BMF = C_B/C_D. Species-specific BAFs are the ratio of the chemical concentration in the organism (C_B) and the organism's surrounding ambient environment, which is generally expressed as freely dissolved seawater concentrations for water-ventilating organisms (i.e., BAF = C_B/C_{WD} mol·m^{-3}) and as gas-phase air concentrations for air-breathing animals (BAF = C_B/C_{AG} mol·m^{-3}). Thus, BAFs for E. Hudson Bay water-ventilating ectotherms (e.g., sculpin, cod) were calculated using measured freely dissolved Arctic seawater concentrations (C_{WD} mol m^{-3}) from references *63,66,67,68,* while BAFs for air-breathing endotherms (e.g., seaducks, belugas and ringed seals) were determined using measured vapor phase Arctic air concentrations (C_{AG} mol m^{-3}) from references *63,69,70,71.* If there were no observed air or water concentrations for a given chemical (as was the case for phthalate esters) concentrations in Arctic seawater do not exist),we needed to first estimate freely dissolved ambient concentrations of those compounds in air and seawater from our observed concentrations measured in lichens and macro-algae, respectively. Specifically, freely dissolved concentrations in air (C_{AG} mol m^{-3}) and seawater (C_{WD} mol m^{-3}) were estimated using the following equations involving observed chemical concentrations in lichens (C_{LICHEN}, mol·m^{-3} lipid equivalent)) and macro-algae (C_{ALGAE}, mol·m^{-3} lipid equivalent)), in accordance with predicted BAFs of the chemical in those media using logBAF-logK_{OW} and logBAF-logK_{OA} relationships for POPs documented in reference *72*,

$$C_{AG} = C_{LICHEN} / BAF_{LICHEN} \quad (2)$$

$$C_{WD} = C_{ALGAE} / BAF_{ALGAE} \quad (3)$$

where the logarithm of the BAF in lichens (log BAF_{LICHEN}) is determined from the chemical's logK_{OA}, while the BAF of the chemical in macro-algae (log BAF_{ALGAE}) is determined from the chemical's logK_{OW} using the following second order log-log quadratic relationships observed for POPs in reference *72*,

$$\log BAF_{LICHEN} = -0.102 \cdot \log K_{OA}^2 + 2.246 \cdot \log K_{OA} - 3.521 \quad (4)$$

$$\log BAF_{ALGAE} = -0.115 \cdot \log K_{OW}^2 + 2.016 \cdot \log K_{OW} - 1.946 \quad (5)$$

The fourth evaluative parameter involved the determination a chemical elimination index (EI), which is calculated using the observed BMF of the compound of interest (BMF_i) and the BMF_{MAX} (assumed to equal BMF_{CB180}) by the following equation

$$EI = \log (BMF_{MAX}) - \log(BMF_i) \quad (6)$$

The above EI calculation is essentially equivalent to the PCB metabolic index (MI) previously used by Tanabe and colleagues (*73,74*). We have chosen to represent the "metabolic index" as an "elimination index" because many of the compounds in our analyses are relatively hydrophilic organic chemicals (log K_{OW}'s < 5) and/or volatile (log K_{OA} < 5) and hence may undergo substantial urinary/respiratory elimination (in addition to *in vivo* metabolic degradation). For those compounds, the MI therefore more accurately represents an organism's overall ability to excrete a given contaminant (i.e., chemical elimination via metabolism and/or urine, feces, respiration etc.) and thus should be presented as an elimination index (EI). For hydrophobic non-volatile compounds such as PBDEs, exhibiting logK_{OW}'s > 5 and log K_{OA}'s > 7, the terms EI and MI are synonymous sand can thus be used interchangeably. The EI is a useful indicator of apparent deviations from the maximum biomagnification potential (i.e., BMF_{MAX}) as a result of the various chemical elimination processes in organisms. Near zero EI values suggest highly persistent compounds (i.e., comparable to Cl_7-CB180), while elevated EI values (> 1) suggest the presence of metabolic and/or other elimination processes. A fifth parameter, namely a biodilution

factor (BDF), can be used to represent this deviation from the BMF_{MAX} and is calculated by the equation:

$$\text{BDF} = \text{antilog EI} = 10^{\log(BMF_{MAX}) - \log(BMF_i)} \quad (7)$$

1.10 Thesis Scope and Organization of Chapters

The general scope of this thesis involves investigation into the bioaccumulation behaviour and biomagnification potential of organic contaminants in fish, wildlife and humans. This work included completion of a review paper (***Chapter 2***), results from a field study involving the analysis of several organic contaminants and/or metabolites in a Canadian Arctic coastal marine food web (***Chapters 3 – 7***) and a summary chapter involving a general discussion on the determinants of biomagnification potential of organic contaminants in marine food webs (***Chapter 8***). Field collected samples of sediments, lichens, macro-algae, fish, marine mammals and seaduck tissues' were analyzed for several target analytes, including 169 Polychlorinated biphenyl congeners (PCBs), 31 Polybrominated Diphenyl Ethers (PBDEs), 30 Organochlorine Pesticides (OCPs), 8 Dialkyl Phthalate Esters (DPEs), 9 Monoalkyl Phthalate Esters (MPEs), 30 hydroxylated brominated diphenyl ethers (OH-BDEs) and 30 methoxylated brominated diphenyl ethers (MeO-BDEs). Determination of concentrations and accumulation patterns of PCBs, OC pesticides and dialkyl phthalate esters in E. Hudson Bay sediment and biota samples are presented in ***Chapter 3***. This paper evaluates the observed accumulation patterns in the E. Hudson Bay marine food web and the associated dietary exposure to aboriginal Inuit communities that utilize marine biota for subsistence. ***Chapter 3*** also investigates relationships between age, sex, tissue specific accumulation patterns and maternal transfer for E. Hudson Bay beluga whales (*Delphinapterus leucas*). Inter-tissue differences and maternal transfer estimates were determined using chemical concentrations determined in different tissues/media (blubber, liver, muscle, whole blood, milk). ***Chapter 4*** is a trophodynamic analysis of persistent organic pollutants (POPs) in the E. Hudson Bay marine food web. This paper looks at the bioaccumulation behviour of the various target compounds in terms of trophic level and investigates relationships between physical chemical properties such as chemical K_{OW} and K_{OA} and chemical biomagnification. Data presented includes food web magnification factors (FWMFs), species-specific biomagnification factors (BMFs) and bioaccumulation factors (BAFs)

for water-ventilating ectotherms (fish) and air-breathing endotherms (seaducks and marine mammals). The primary focus of this paper is to investigate the biomagnification potential of low K_{OW} – high K_{OA} compounds such as HCHs and CBz. ***Chapter 5*** is an investigation of the distribution of dialkyl phthalate esters (DPEs) and their mono-alkyl phthalate ester (MPE) metabolites in the E. Hudson Bay marine food web. FWMFs, BMFs, BAFs, Relative patterns (i.e., % relative contributions) and metabolic index (MI) values were determined for the various DPEs and compared to organochlorine data. The paper documents evidence of metabolism (i.e., detection of MPEs) and trophic dilution of DPEs in the Arctic marine food web (i.e., FWMFs < 1). ***Chapter 6*** is a study of PBDE congeners in E. Hudson Bay marine food web. This paper documents levels of BDE congeners and ∑PBDEs in various organisms of the food web. The extent of BDE bioaccumulation in the food web is investigated by comparison of concentrations in biota versus organism trophic level (TL) and estimation of FWMFs. ***Chapter 7*** presents measured concentrations of several hydroxylated and methoxylated BDEs, which are potential PBDE metabolites (i.e., biotransformation to OH-BDEs through primary hydroxylation and subsequent transformation to MeO-BDEs through secondary methylation). OH- and MeO-BDEs levels are compared to observed levels of parent PBDE congeners. The potential sources and toxicological significance of these compounds are discussed. ***Chapter 8*** is a summary and of the findings presented in the preceding chapters and involves a general discussion regarding the chemical and biological determinants of bioaccumulation potential of organic contaminants in marine food webs.

CHAPTER 2

INTESTINAL ABSORPTION AND BIOMAGNIFICATION OF ORGANIC CONTAMINANTS IN FISH, WILDLIFE AND HUMANS

2.1 Introduction

It is widely recognized that emissions of persistent organic chemicals can result in ubiquitous dispersal in local and global environments and bioaccumulation in organisms. Equilibrium partitioning of dispersed chemical causes bioconcentration into organism lipids via passive molecular diffusion. Bioconcentration factors (BCFs), the ratio of a chemical's equilibrium concentration in an organism (C_B, wet wt. basis) and the organism's respired media (C_R), (i.e., $BCF = C_B/C_R$), are largely dependent on an organism's lipid content. However, equilibrium concentrations of hydrophobic chemicals (expressed on a lipid wt. basis) will be equivalent among different organisms, regardless of lipid content. In addition to bioconcentration, some organic contaminants such as polychlorinated biphenyls (PCBs) and DDT are also known to biomagnify, resulting in chemical concentrations (on a lipid wt. basis) in an organism (C_B) that exceed concentrations in consumed prey (C_D), (*2 3,5,6,43,75,76*). Concentration-based biomagnification factors are typically reported on a lipid wt. basis ($BMF_C = C_B/C_{D,}$ lipid wt). Bioaccumulation factors (i.e., $BAFs = C_B/C_R$), represent bioconcentration + biomagnification. Food chain biomagnification occurs when lipid wt. concentrations increase with increasing trophic position (TP), (i.e., $C_{TP,4} > C_{TP,3} > C_{TP,2} > C_{TP,1}$). These bioaccumulative substances are of great concern due to their potential to attain toxicologically significant tissue and organ residue concentrations in high trophic level species such as predatory fish, birds and mammals (including humans), (*8,77,78*). DDT induced egg-shell thinning in birds of prey such as the peregrine falcon (*Falco peregrinus*) during the 1960s is perhaps the most notorious example of the potential deleterious effects of bioaccumulative substances (*78,79*).

To avoid the perils of bioaccumulative substances such as DDT and PCBs, governments in Canada, the United States and Europe have launched proactive and preventative measures to reduce or eliminate similar future risks from "current-use" and "proposal-stage" chemicals of

commerce. For example, Canada has adopted a Toxic Substances Management Policy under the Canadian Environmental Protection Act in accordance with the recent Stockholm Convention on Persistent Organic Pollutants (POPs), (*80*). This policy considers virtual elimination of chemicals that meet criteria for persistence (P), bioaccumulation (B) and inherent toxicity (T). Current bioaccumulation criteria (i.e., B criteria) identify "bioaccumulative" substances as those compounds that exhibit bioaccumulation or bioconcentration factors (BAFs or BCFs) greater than 5,000 in aquatic organisms, or (in the absence of BAF or BCF data) chemicals with octanol-water partition coefficients (K_{OW}) greater than 10^5. The K_{OW} threshold criterion is a marker of bioaccumulation potential in aquatic organisms because of the mechanistic understanding that chemicals with K_{OW}'s < 10^5 may bioconcentrate in aquatic organisms (i. e., accumulation from water *via* the organism's respiratory surface such as gills) but do not biomagnify due to efficient clearance of chemical to water *via* gill ventilation (*36,37,39, 81*).

This simple K_{OW} based structure activity relationship for bioaccumulation of commercial chemicals was derived from observations and biomagnification models for aquatic organisms (*3,36,42,81*). Its adoption in policies implies that it is also considered to be appropriate for assessing bioaccumulation in numerous other organisms including birds, reptiles, mammals and humans. In addition, current mechanistic models may not be appropriate for all organisms nor for all classes of compounds. For example, as is illustrated in Figure 2.1, the magnitude of the biomagnification factor (BMF_C lipid wt.) of stable non-metabolizable compounds (e.g., PCB 153) varies by orders of magnitude among different classes of organisms. Reported BMFs of PCB 153 typically range between 5-10 for invertebrates and fish (*6,36,37,81,82*), 5-10 for caribou, dairy cows and shrews (*43,83,84,85*), 20-60 for birds (*86-88*), 30-40 for mustelids (*89*) and approaching 100 or greater for marine mammals (*8,63, 90-94*), wolves (*43*) and humans (*95*). Investigations of POPs in the Great Lakes by Norstrom and colleagues highlighted that BMFs of organochlorines in Lake Ontario herring gulls were approximately 10 times higher than those BMFs in Lake Ontario Coho salmon (*88*). Numerous other studies have documented this interspecies variability in BMFs between aquatic poikilotherms (invertebrates, fish) and homeotherms (bids and mammals), (*2,5,46*). Consequently, utilization of observed BMFs in aquatic organisms as a surrogate for bioaccumulation potential can substantially underestimate the extent of chemical biomagnification in birds and mammals. Also, some relatively hydrophilic compounds such as β-hexachlorocyclohexane (b-HCH), α-endosulfan and chlorobenzenes (log K_{OW}'s range between 3.8- 4.5) have shown considerable biomagnification in air-breathing organisms (i.e., birds and mammals) while they are not known to biomagnify in aquatic

organisms (*6,14,43*). Another example is Perfluoro-octane-sulfonate (PFOS), which is a water-soluble, non-volatile anionic compound. This substance does not meet the current K_{OW} criterion for bioaccumulative substances and does not biomagnify in fish (*96*). However, it is efficiently absorbed *via* dietary exposures, biomagnifies and persists in the liver and blood of air-breathing animals (*15-17, 97)* and is inherently toxic (*98,99*). Modeling studies of POPs bioaccumulation in arctic caribou and wolves (*44*) have illustrated that relatively hydrophilic (i.e., polar, K_{OW}'s $<10^5$) but non-volatile compounds (i.e., octanol-air partition coefficients or K_{OA}'s $> 10^5$) that are resistant to metabolism (half-life > 60 days) biomagnify due to efficient gastro-intestinal absorption and very slow lipid-to-air elimination *via* respired air. This emerging evidence indicates that the current K_{OW} based classification of chemicals is not an adequate model to identify substances with a bioaccumulative potential in food webs that include mammals, birds and humans. The significance of this issue is emphasized by the fact that approximately two-thirds of the chemical substances used commercially in Canada have been indicated as having possible bioaccumulative potential in birds and mammals (including humans). Of these substances, about half are polar non-volatile compounds (PNVs) with a low K_{OW} (i.e., log K_{OW}'s < 5), whose bioaccumulation potential may have been miscategorized (*100*). These chemicals include many chemicals of concern such as flame retardants, surfactants, pesticides, plasticizers, fluorinated and alkylphenol ethoxylates, pharmaceuticals, sunscreen agents and synthetic fragrances (*101*).

To develop better and more proactive policies for identifying "bioaccumulative" substances, it is important to better characterize the dominant underlying processes and mechanisms driving the biomagnification phenomenon. These processes include dietary absorption, various elimination processes, metabolic transformation and growth dilution. In this paper, we will review the current thinking on dietary absorption and biomagnification of organic substances. Several research groups have recently made significant contributions in this area. The objectives of our paper are first to review the current state of knowledge of mechanisms and models of intestinal absorption and bioaccumulation of organic chemicals in wildlife and humans and secondly to discuss the implications of these models for assessing the bioaccumulative potential of organic substances. Specifically, we outline and evaluate four different intestinal absorption and biomagnification mechanisms and models. It is our hope that this discussion will lead to the formulation of better models to assess the biomagnification behavior of organic chemicals in food webs.

2.2 Theory

The fugacity approach. In this study we will use the fugacity approach to formulate models for dietary absorption and biomagnification. Mackay and colleagues have previously illustrated the benefits of using chemical fugacity to describe and quantify chemical transport in environmental systems and food webs (*102,103*). The fugacity of a chemical (*f*, in units of Pascal) for a given phase is related linearly to its molar concentration (C in $mol \cdot m^{-3}$) by the fugacity capacity (Z, $mol \cdot m^{-3} \cdot Pa$) of the phase in which the chemical is solubilized:

$$f = C/Z \quad (1)$$

The fugacity capacity is compound and phase specific and represents the ability of that phase to sorb and retain a given chemical within its matrix. In essence, the fugacity is a measure of the chemical's concentration normalized to the chemical's solubility in the medium it resides in. The ratio of the fugacity capacities (Z) of two adjacent media or compartments *i* and *j* (i.e., *Zi/Zj*), can be viewed as a partition coefficient *Kij,* which is equivalent to *Ci/Cj* at equilibrium. Thus, $K_{OW} = Z_O/Z_W = C_O/C_W$, while $K_{OA} = Z_O/Z_A = C_O/C_A$.

In the fugacity approach, chemical uptake and clearance processes occur via advection, diffusion or reaction (e.g., metabolism). In fugacity format, transport of chemical for these processes are described in terms of transport parameters (or D values in units of $mol \cdot Pa \cdot d^{-1}$), which are related to concentration-based rate constants (k, d^{-1}). Relatively large values represent fast processes, while small D-values indicate slower processes. If transport of chemicals between different media is diffusive in nature, then D can be determined from the chemical's molecular diffusivity (B, m^2/d), the surface area of diffusion (A, m^2), the fugacity capacity (Z) of the phase, and the length of the diffusion path (d, m).

$$D_{DIFFUSION} = (BAZ)/d \quad (2)$$

Diffusive processes between phases or compartments are reversible and the D value for diffusion from medium *i* to *j* is equal to that from medium to *j* to *i*. The diffusive flux (denoted as *N*, $mol \cdot d^{-1}$) between media or compartments can be described by:

$$N = D(f_i - f_j) \quad (3)$$

where the term $(f_i - f_j)$ represents the departure from equilibrium or driving force between two phases (i.e., media) or compartments *i* and *j*. If chemical fugacities are not equal, the direction of chemical flow occurs from the high fugacity compartment to the low fugacity compartment. If chemical fugacities become equal, the two compartments have attained a chemical equilibrium and the chemical flux (N) is zero. If the transport process is advective in nature, the transport parameter D is the product of a flow rate (G, $m^3 \cdot d^{-1}$) of a given medium and the fugacity capacity (Z in $mol \cdot m^{-3} \cdot Pa$) of the chemical in the advective medium.

$$D_{ADVECTION} = GZ \quad (4)$$

Advective transport processes between phases or compartments are unidirectional and in cases where a distribution is the result of advective inflows and outflows, the net flux (denoted as *N*, $mol \cdot d^{-1}$) between media or compartments can be described by:

$$N = D_i \cdot f_i - D_j \cdot f_j \quad (5)$$

Equation 5 illustrates that transport will take place until $D_i \cdot f_i$ equals $D_j \cdot f_j$ at which N=0 (i.e. steady-state) and the fugacities f_i/f_j equal D_i/D_j. If chemical is generated or depurated by a chemical reaction (e.g. metabolism), the D-value is related to the first order rate constant, the compartment volume (V, m^3) and the Z of the phase:

$$D_{METABOLISM} = VkZ \quad (6)$$

D-values are equivalent to conductivities for chemical mass. Hence their reciprocal (1/D) can be viewed as a resistance to the mass transport a chemical encounters in a given phase. If transport and reaction processes occur in series the reciprocal D values add to give the total resistance to chemical transport:

$$1/D_{TOTAL} = 1/D_1 + 1/D_2 + 1D_3, \text{ etc.} \quad (7)$$

If chemical transport and transformation occur in parallel, D-values are additive:

$$D_{TOTAL} = D_1 + D_2 + D_3, \text{ etc.} \quad (8)$$

The ability to add serial resistances or parallel transport parameters is convenient for quantifying chemical transfer in biological systems due to the presence of multiple, simultaneously occurring processes and/or resistance pathways.

In fugacity terms, biomagnification in the food chain is defined as a state where fugacities increase with increasing trophic level (e.g., $f_{VEGETATION} < f_{HERBIVORE} < f_{CARNIVORE}$). The purpose of this paper is to explore the underlying mechanism of this phenomenon and explore how this mechanism can be formalized in a fugacity based model that is useful for bioaccumulation assessment.

2.2.1 Intestinal absorption of xenobiotic molecules.

Numerous reviews of lipid absorption and digestive physiology have been conducted (*104-108*). Figure 2.2 depicts the transcellular migration path of dietary fats and environmental contaminants (i.e., xenobiotics) from lumen across the epithelium of the gut wall. The formation of mixed micelles from bile salt molecules in the gastro-intestinal tract (GIT) and their function with respect to lipid digestion is well documented (*104,105,107,108*). The purpose of bile salt micelles in the GIT is twofold: one is to remove monoglycerides and free fatty acids (FFA) from the vicinity of digesting fat globules so the digestion process can proceed unabated and secondly to act as a transport medium by "ferrying" monoglycerides, FFA's, fat soluble vitamins A, D and K and thus consequently hydrophobic organic contaminants across an unstirred water layer (UWL) to the luminal membrane. This stagnant water layer is estimated to be approximately 0.05 to 2 mm in thickness in humans and is relatively more acidic (pH between 5.1 and 6.3) than the aqueous phase of the intestinal contents. The greater acidity aids the diffusion of fatty acids across the membrane by increasing the fraction of the unionized free fatty acid in the aqueous phase. It is believed that a pH drop within the UWL adjacent to the brush border membrane (BBM) causes dissociation of the micelles, which then results in fatty acids and xenobiotics (e.g., PCBs) separating from the micelles and permeating through the water phase into the brush border membrane (BBM) of the epithelial cells as monomers (*107*). However, there is some evidence that suggests collisional contact of mixed micelles and intestinal cell membranes causes release of lipids and chemical directly into the BBM (*105*). The significance of the latter process is that it would essentially eliminate the resistance to membrane transfer posed by the aqueous diffusion

barrier adjacent to the BBM and bypass diffusion altogether. Molecules in the intestine can be transported across the epithelium of the gut by either a transcellular route (across the plasma membrane of the epithelial cells) or by a paracellular route (across tight junctions between epithelial cells). While water and polar organic compounds may be transported by both routes, the tight junctions are generally impermeable to large non-polar organic molecules (e.g. fats and PCBs with minimal internal cross sections > 1Å). Hence, those molecules are transported exclusively by the transcellular route (*106*). Beyond the brush border membrane in the cytosol, the absorbed digestion products (i.e., fatty acids, monoglycerides) are resynthesized into triglycerides, which are subsequently "packaged" with phospholipids and apoproteins to form lipid vesicles (referred to as lipoproteins or chylomicrons). The lipid vesicles then migrate to the basolateral membranes where they are released by exocytosis into lymph and/or venous blood. Compounds absorbed directly in the cytosol (individual molecules) can then either diffuse directly across the basolateral membrane into portal blood or become solubilized in lipoproteins and subsequently released via exocytosis. Dulfer et al. (*109*) have recently formalized the above transcellular migration path of organic chemicals in terms of chemical fugacities and transport parameters (D-values) for the various intestinal components (see Appendix 3).

2.3 Mechanisms of Biomagnification

2.3.1 Biomass Conversion Model

The first mechanistic explanation of the biomagnification phenomenon was documented by Woodwell (*76*) based on the observed increase in concentrations of PCBs and DDT in biota with trophic level in a marine aquatic food web. Woodwell reasoned that PCBs and DDTs were efficiently ingested and absorbed in association with food but depurated at a rate slower than the consumption of biomass needed for energy requirements. In fugacity terms, this process can be formulated as:

$$N_B = V_B Z_B df_B/dt = D_D \cdot f_D - D_E \cdot f_B \quad (9)$$

Where N_B is the net absorption of chemical by the organism (i.e. $V_B \cdot Z_B \cdot df_B/dt$); $D_D \cdot f_D$ the rate of chemical absorption (in units of $mol \cdot d^{-1}$) via dietary ingestion and $D_E \cdot f_B$ the rate of chemical depuration (in units of $mol \cdot d^{-1}$) *via* all possible routes. D_D the transport parameter of chemical absorption *via* dietary ingestion ($mol \cdot d^{-1} \cdot P1$), f_D is the chemical fugacity in the diet, D_E is the

transport parameter for chemical depuration (mol·d^{-1}·P1) and, f_B is the chemical fugacity in the organism. At steady state ($N_B = 0$), equation 9 becomes $f_B/f_D = D_D/D_E$, which illustrates that biomagnification can occur for chemicals for which $D_E < D_D$.

One of the characteristics of this mechanism is that chemical is moved from a low fugacity in the prey to a high fugacity in the predator. This constitutes a mass transport against the thermodynamic gradient, which indicates ingested chemical is predominantly absorbed via a non-diffusive active transport process. A second feature of this mechanism is that the magnification of the chemical concentration occurs as a result of energy consumption in the tissues of the organism. The latter has led to the application of bioenergetic models to estimate the degree of chemical magnification (*110*).

2.3.2 Digestion Model

Hydrophobic organic chemicals as well as monoglycerides and fatty acids are ideally suited to diffuse through and pass biological membranes due to their lipophilicity (*111*) and hence do not require an active transport mechanism for absorption. To explain the apparent transport of hydrophobic organic chemicals against the thermodynamic gradient, it was hypothesized that organic chemicals are magnified in the gastro-intestinal tract (GIT) of organisms as a result of food digestion (*35,39,112*). In this mechanism the chemical concentration and fugacity in the GIT of an organism is raised as food is absorbed. In essence, digestion of consumed food in the GIT concentrates ingested chemical residues in a reduced and compositionally altered digesta matrix, which causes a fugacity "pump" or gastro-intestinal magnification (i.e., f_G exceeds f_D). This creates a positive thermodynamic gradient between the GIT and the organism (i.e., $f_G > f_B$). This gradient is required for net passive absorption of chemical. If elimination by excretion or metabolism is negligible, the chemical concentration and fugacity in the organism will increase to match that in the GIT. The latter will cause the concentration in the organism to exceed that in its food.

This mechanism can be formalized in fugacity terms in a two-compartment model, consisting of a gastrointestinal tract with a volume V_G (in m^3) and an organism compartment with volume V_B (in m^3) representing the animal's overall contaminant storage in various tissues and viscera (Figure 2.3). To maintain simplicity, the GIT is viewed as a single well-mixed homogenous compartment. While these simplifications have adequately represented POPs bioaccumulation in several

organisms (*36,44,83,102*), more complex multi-compartment bioaccumulation models can be utilized if tissue/organ specific resolution is desired (*113,114*). Chemical enters the GIT at a rate of $D_D \cdot f_D$, which is equal to the product of the volumetric feeding rate G_D ($m^3 \cdot d^{-1}$), the fugacity capacity of the food Z_D ($mol \cdot Pl \cdot m^{-3}$) and the fugacity in the diet f_D, i.e. $G_D \cdot Z_D \cdot f_D$ or simply $G_D \cdot C_D$. Digested food leaves the GIT in fecal matter at a rate of $D_F \cdot f_G$, which is equal to the product the fecal egestion rate G_F ($m^3 \cdot d^{-1}$), the fugacity capacity of the feces Z_G and the fugacity in the fecal matter f_G, i.e., $G_F \cdot Z_G \cdot f_G$ or simply $G_F \cdot C_G$. Chemical moves from the GIT to the organism at a rate of $D_{GB} \cdot f_G$. The reverse transport takes place at a rate of $D_{BG} \cdot f_B$. The net flux N_G into the GIT can therefore be expressed as:

$$N_G = V_G \cdot Z_G \cdot df_G / dt = D_D \cdot f_D + D_{BG} \cdot f_B - (D_F + D_{GB}) \cdot f_G \quad (10)$$

The organism compartment, which for reasons of simplicity is also represented as a single well mixed compartment receives chemical from contaminant flux between the GIT and organism ($D_{GB}f_G$), and uptake via the respiratory route ($D_R f_R$), i.e. uptake from water or air. Chemical depuration ($mol \cdot d^{-1}$) can occur through excretion into the digesta ($D_{BG} \cdot f_B$) and subsequent fecal excretion, respiratory elimination ($D_R f_R$), urinary excretion ($D_U f_U$), reproductive transfer and lactation ($D_{REPRO} f_B$), metabolic transformation D_M (equivalent to $k_M C_B V_B$) where k_M (d^{-1}) is the metabolic transformation rate constant of the chemical in the organism. Growth dilution ($D_B \cdot f_B$) caused by an animal's increase in body storage volume (V_B) and lipid content (v_{LB}) over time, essentially dilutes internal chemical concentrations (C_B) while increasing storage capacity (i.e., Z_B). Conversely, depletion of an animal's fat reserves concentrates chemical residues while decreasing storage capacity (i.e., Z_B). Growth can be particularly important for nursing newborns, organisms that periodically undergo significant seasonal body condition changes (e.g., hibernating mammals) and physiologically stressed organisms (e.g., diseased animals). The net flux N_B into the organism can therefore be expressed as:

$$N_B = V_B \cdot Z_B \cdot df_B / dt = D_R f_R + D_{GB} f_G - (D_{BG} + D_R + D_M + D_B + D_U + D_{REPRO}) \cdot f_B \quad (11)$$

If it can be assumed that the GIT is at steady-state, i.e. $N_G = 0$ and f_G equals $(D_D \cdot f_D + D_{BG} \cdot f_B)/(D_{GB} + D_F)$, which after substitution into equation 11 yields an interesting expression for the fugacity based biomagnification factor (BMF_f) at steady state:

$$BMF_f = f_B/f_D = (D_R + (D_D/D_F))\cdot(D_{GB}/(D_{BG} + D_R + D_M + D_B + D_U + D_{REPRO})) \quad (12)$$

This equation illustrates the role of some of the key factors in controlling the biomagnification factor of organic chemicals. It illustrates that food digestion is a key factor in the magnification of the chemical as (D_D/D_F) which equals $(G_D \cdot Z_D/G_F \cdot Z_G)$ or $(G_D/G_F)\cdot(Z_D/Z_G)$ will exceed 1 in proportion to the extent to which the fecal excretion rate G_F drops below the dietary intake rate G_D and to the degree to which the fugacity capacity of the diet Z_D is lowered to Z_G as a result absorption of lipids, proteins and other dietary components. Following a 3-phase partitioning model employed in reference (*44*), Z_D/Z_G can be derived as:

$$Z_D/Z_G = C_B/C_G = K_{DG} =$$

$$(\nu_{LD} + 0.035\cdot\nu_{NLD} + \nu_{WD}/K_{OW})/(\nu_{LG} + 0.035\cdot\nu_{NLG} + \nu_{WG}/K_{OW}) \quad (13)$$

where ν_{LD}, ν_{NLD} and ν_{WD} are the lipid , non-lipid organic matter and water contents of ingested food (kg /kg wet wt. food) and ν_{LG}, ν_{NLG} and ν_{WG} are the lipid, non-lipid organic matter and water contents of the gut contents (kg /kg wet wt. digesta). Equation 12 also illustrates the role of metabolic transformation and other mechanisms of elimination. If the exchange of chemical between the GIT and the organism is dominated by passive diffusion, D_{GB} and D_{BG} are equal and the depuration processes combine to drive $(D_{GB}/(D_{BG} + D_R + D_M + D_B + D_U + D_{REPRO}))$ below 1. In the hypothetical case of a complete absence of depuration routes (i.e., D_R , D_M, D_B, D_U and D_{REPRO} are all zero) the maximum attainable BMF_f approaches:

$$BMF_f = f_B/f_D = (D_D/D_F) = (G_D/G_F)\cdot(Z_D/Z_G) \quad (14)$$

This result is the same as for the biomass conversion model. The essence of the digestion model is that food digestion is the key process responsible for actual magnification of the chemical concentration in the predator. A key assumption of this model is that the exchange of chemical between the GIT and the organism is dominated by passive diffusion (i.e., molecular diffusion is the rate limiting step in GIT-organism chemical transport). This means that D_{GB} and D_{BG} are equal. It views micellar transport (D_{MIC}) and subsequent diffusion of the chemical through unstirred water layers (D_W) and the phospholipid bilayers (D_L) as processes applying in series, i.e.

$$1/D_{GB} = 1/D_{MIC} + 1/D_W + 1/D_L \quad (15)$$

with the slowest step in the chain of events controlling the overall rate of gastro-intestinal uptake. As discussed in reference (*40*), this implies that dietary absorption rates (e.g. quantified by the gross dietary absorption efficiency E_D) can be expected to fall with increasing K_{OW}, as the low aqueous concentrations of highly hydrophobic compounds in the unstirred water layers control the rate of intestinal uptake, i.e.,

$$1/E_D = \alpha \cdot K_{OW} + \beta \quad (16)$$

where α and β are organism specific constants.

Figure 2.4 is an illustrative example of GI magnification model predicted fugacities (nPa) at steady-state of a non-metabolizable, non-volatile and hydrophobic compound such as PCB 153 in an aquatic poikilotherm (e.g., fish) and a homeotherm (e.g., mammal). Figure 2.4 shows a fugacity increase from 1 nPa in consumed food (f_D = 1 nPa) to approximately 8 nPa in the GIT for fish. The 8 times fugacity increase in the GIT, due to a G_D/G_F ratio of approximately 2 (i.e., 50% food absorption) and a Z_D/Z_G ratio of approximately 4 (based on 95% lipid, 60% non-lipid organic matter and 95% water extraction efficiency), results in a gastro-intestinal magnification factor (GIMF) of 2 x 4 or 8 in fish. Thus, the steady state fugacities for food (f_D), intestinal tissues (f_I), body tissues (f_B), and fecal matter (f_F) in fish are (f_D:$f_I$$f_B$:$f_F$ = 1:8:8:8) under these hypothetical conditions. For homeotherms, figure 2.4 shows a fugacity increase from 1 nPa in consumed food (f_D = 1 nPa) to approximately 80 nPa in the GIT. The digestion model explains the comparatively high BMFs observed in homeotherms (e.g., BMF = $f_B/f_D = G_D/G_F \cdot Z_D/Z_G$ = 80) compared to fish (e.g., BMF = $f_B/f_D = G_D/G_F \cdot Z_D/Z_G$ = 8) as a result of a greater efficiency of the digestive system. A more efficient digestive system means that organisms exhibit larger G_D/G_F (e.g. 20 based on a 95% food absorption) and Z_D/Z_G (e.g. between 4 to 10, based on a $\geq$ 98 lipid, $\geq$ 60% non-lipid organic matter and 95% water extraction efficiency). The magnitude of the Z drop is also sensitive to the lipid content of the prey species (i.e., quantity of dietary lipids ingested),(*42*). The combined effect of food absorption (G_D/G_F) and extraction of dietary constituents (Z_D/Z_G) will produce greater gastro-intestinal magnification factors and BMFs (~ 80 or greater, i.e. $\geq$ 10 times greater than the BMF in fish) and a fugacity distribution of f_D:$f_I$$f_B$:$f_F$ = 1:80:80:80. Table 1, showing a comparison of dietary absorption parameters and BMF_{MAX} values for fish, birds, terrestrial and marine mammals (*42,115-121*), illustrates that homeothermic carnivores (typically

exhibiting a BMF_{MAX} 10-20 times higher than fish) tend to consume larger amounts of lipid-rich prey and have a higher degree of lipid and food absorption compared to fish.

There is considerable evidence for the role of food digestion on dietary absorption and biomagnification. Initial evidence supporting the digestion model comes from experiments (*40*), in which three batches of fish were fed low fat (LF), medium fat (MF) and high fat (HF) diets. The three diets contained a series of hydrophobic organic chemicals at the same concentration but at varying fugacities (i.e., $f_{LF} > f_{MF} > f_{HF}$, due to $Z_{LF} < Z_{MF} < Z_{HF}$). The authors hypothesized that if molecular diffusion were rate limiting then GIT-organism chemical uptake rates (N_D, mol d^{-1}) and absorption efficiencies (E_D) would increase with decreasing lipid content in the food (due to elevated fugacity). Conversely, if micelle facilitated diffusion were dominating then increased lipid content should result in higher uptake N_D, and absorption efficiencies (E_D). The results showed that dietary uptake rates increased with reduced lipids in consumed food due to increased chemical fugacity in the diet. This suggested that the hydrophobic organic chemicals tested (PCBs and chlorobenzenes) were absorbed via passive diffusion and that a positive fugacity gradient between the intestines and the organism is a key determinant for dietary absorption. The authors concluded that lipid vesicle transport (i.e., micelle-facilitated diffusion) was not rate limiting in the GIT-organism flux in the exposed fish because increased lipid ingestion did not result in increased chemical uptake (N_D) or absorption (E_D).

Further evidence to illustrate that the chemical fugacity in the intestinal tract can be raised over that in the diet, came from three sets of laboratory studies with guppies, goldfish and adult rainbow trout and a comparative field study (*40,41,42*). Direct and indirect measurements of the fugacity of a series of hydrophobic organic chemicals in the intestinal content of these fish species showed that fugacities in the diet are raised in the intestines as a result food digestion. The studies showed that the occurrence of a fugacity pump in the GIT is mainly due the lipid absorption efficiency (approximately 92% in fish) to be greater than the chemical absorption efficiency (i.e. approximately 75% or less). The fact that lipids are absorbed from the gut lumen at a faster rate than the chemical produces an increase in fugacity in the gut lumen over that in the diet consumed. The magnitude of the fugacity increase observed in fish (i.e., 8 times increase from f_D to f_G) corresponded to a 4 times decrease from Z_D to Z_G and a 2 times drop in digesta volume ($G_D/G_F = 2$).

The relationship between dietary absorption efficiency and chemical K_{OW} has been previously investigated in fish (*37,39,40*), birds (*115*) dairy cows (*83,119*) and humans (*95,120*). Data from these comparable studies (plotted in Figure 2.5) illustrates that absorption of ingested chemical in both homeotherms (ring doves, dairy cows and humans) and an aquatic poikilotherm (fish) show a tendency to be relatively constant for low K_{OW} substances, but drops with increasing K_{OW} (E_D drops significantly when log $K_{OW} > 7$). Gobas and colleagues (*40,41,42*) have suggested the declining trend in E_D in fish is consistent with a diffusion controlled dietary absorption mechanism, where micellar transport and diffusion through unstirred water layers and diffusion through phospholipid bilayers apply in series. For low K_{OW} substances, micellar transport and/or phospholipid bilayer diffusion are the rate determining step while diffusion through unstirred water layers is rate limiting for very high K_{OW} substances with very low solubilities in the water layers. The authors further suggest that if gastro-intestinal absorption processes would apply in parallel, then micellar transport should control gastro-intestinal uptake of the higher K_{OW} chemicals and should be similar for all compounds.

2.3.3 Micelle Mediated Diffusion Model

To explain the higher BMFs in homeotherms compared to aquatic poikilotherms, Drouillard and Norstrom (*115*) proposed that micelle mediated diffusion can produce a magnification effect in addition to or in place of food digestion. This process involves micelle facilitated chemical transport from the bulk lumen to the organism (i.e., GIT-to-organism) through unidirectional advection of mixed micelles across the aqueous resistance of the unstirred water layer (UWL), while the reverse flux (i.e., organism-to-GIT) is somewhat reduced because micelles become dissociated within an acidic pH microclimate present at the vicinity of the intestinal wall. In essence, the MMD model assumes intestinal absorption of chemical (enhanced by mixed micelle facilitation) occurs in the upper GIT in association with dietary lipid absorption, while chemical elimination (decoupled in time and space) occurs at a much slower rate in the lower digestive tract. Thus, the mixed micelle transport in the upper intestine causes the rate of chemical uptake across the UWL into gut tissue to be substantially faster than the rate of reverse diffusion back to the intestine. In fugacity terms, the transport parameter D_{GB} in equations 11 and 12 is greater than D_{BG}. This results in a sustained fugacity increase in the organism's tissues over that in the intestines and the original diet consumed. The authors propose that the higher energetic demands of homeothermic animals (birds and mammals) compared to fish results in higher feeding rates in homeothermic animals. The higher feeding rates produce greater mixed micelle concentrations in

the GIT and hence greater chemical uptake rates through direct transfer of the chemical containing micelles to intestinal tissue. This ultimately causes a high fugacity build up in the animal's tissues due to very slow diffusive elimination rate back to the GIT.

This mechanism was formalized in fugacity format by Cahill et al. (*113*) using a physiologically based pharmacokinetic (PBPK) model that incorporates a micelle mediated uptake mechanism. In their generalized PBPK model designed for evaluating multi-residue toxicokinetics, gastrointestinal uptake is described as a parallel aqueous and micelle mediated diffusion. The micelle mediated diffusion model assumes gastro-intestinal uptake (D_{GB}) is described as the sum of simultaneous parallel processes including micellar transport (D_{MIC}) direct aqueous diffusion (D_W) and diffusion across the cell membrane (D_{CELL}), as described by Dulfer et al. (*109*):

$$D_{GB} = D_{MIC} + D_W + D_{CELL} \quad (17)$$

This model of intestinal absorption assumes unidirectional micelle facilitated diffusion across the UWL followed by molecular diffusion through the cell membrane, while aqueous molecular diffusion of contaminant into gut tissue is bi-directional. The primary difference of this model to that of digestion hypothesis is the assumption of unequal chemical uptake (D_{GB}) and elimination (D_{BG}), specifically that $D_{GB} > D_{BG}$. A D_{GB}/D_{BG} ratio greater than 1 inherently suggests that chemical is more efficiently absorbed from the intestine than they are lost to the intestine. This ratio represents an additional magnification factor to any magnification that may occur as a result of food digestion. The latter is illustrated by equation 12, which in the hypothetical case of a complete absence of depuration routes (i.e., D_R, D_M, D_B, D_U and D_{REPRO} are zero) simplifies to:

$$BMF_f = f_B/f_D = (D_D/D_F)\cdot(D_{BG}/D_{BG}) = (G_D/G_F)\cdot(Z_D/Z_G)\cdot(D_{GB}/D_{BG}) \quad (18)$$

The MMD model therefore suggests the larger BMFs exhibited by homeotherms compared to aquatic poikilotherms can be explained by a larger D_{GB}/D_{BG} ratio in homeotherms.

Figure 2.6 illustrates theoretical MMD model predicted fugacities (nPa) of typical hydrophobic POPs at steady-state in a fish and a homeotherm (e.g., mammal). The disparity in D-values across the gut wall is the central distinction of the MMD hypotheses (i.e., $D_{GB} > D_{BG}$). The steady state fugacities for food (f_D), intestinal tissues (f_I), body tissues (f_B), and fecal matter (f_F), are f_D:f_I:f_B:f_F = 1:8:8:7 for fish and f_D:f_I:f_B:f_F = 1:80:80:40 for a mammal.

There have been numerous studies providing evidence for the co-transport of organic chemicals with lipid vesicles in the GIT and/or lymphatic flow (*108,122-125*). Vetter et al. (*126*) used light microscopy to examine intestinal contents and tissues' following the absorption of administered benzo(a)pyrene (BaP) in killifish. Their results indicated co-assimilation of dietary fats and chemical *via* lipid vesicles into the BBM and into fat droplets within the enterocytes and that separation of chemical from dietary lipids occurs primarily after lipid absorption and reassembling in the enterocyte. However, other studies of intestinal absorption of BaP suggest separation of chemical from dietary lipids occurs in the lumen, followed by passive diffusion of single monomers into the enterocyte (*127*).

The semi-empirical fugacity based cell-line model developed by Dulfer et al. (*109*), (see Appendix 3) provides some empirical evidence of the presence of a micelle-facilitated transport mechanism for hydrophobic contaminants across human intestinal membranes. Specifically, in-vitro studies of PCB absorption in human colorectal carcinoma derived cells (Caco-2 cells), (*109*) show that the presence of mixed micelles can increase chemical flux into intestinal cells more than 1000 fold compared to cell lines without mixed micelles due to the relatively high affinity of hydrophobic organic chemicals for mixed micelles (Z_{MIC}). The authors suggest that micelle mediated transport of chemical from the upper intestine (gut lumen to intestinal tissue) is likely a substantially faster process than the reverse transport back across the UWL (intestinal-tissue to gut lumen) because the latter process is assumed to occur by diffusion alone.

Drouillard and Norstrom (*115*) in their study of dietary uptake of PCB congeners in ring doves (*Streptotpelia risoria*) found dietary absorption efficiencies of PCBs (93 to 83% over a log K_{OW} range of 5 to 7.5) were comparable to the lipid absorption efficiency (90%). Furthermore, the authors found PCB congeners entered blood plasma at similar rates to dietary lipids. These findings suggest that lipids and ingested contaminant are absorbed in association, which indicates that a fugacity pump in the GIT may not occur because the onset of gastro-intestinal magnification is primarily caused by a higher rate of lipid removal from the GIT compared to the rate of chemical absorption. The authors observed a small (~10%) decline in absorption efficiencies of PCB in ring doves with increasing K_{OW} (see figure 2.5), which was attributed to solubility limitations of those high K_{OW} compounds in the nucleus of the mixed micelles interior, rather than kinetic limitations across the stagnant aqueous UWL (as is suggested in the digestion model). This argument is supported by measurements of membrane/water partition coefficients (K_{MW}) for chemicals of varying K_{OW} (*128,129*), which illustrate that the solubility of hydrophobic

organic chemicals in membrane vesicles increases with increasing K_{OW} up to a maximum K_{OW} value of approximately 7 and then drops with further increasing K_{OW} (see Appendix 4). In a complementary depuration study of gavaged PCBs in ring doves, Drouillard and Norstrom (*116*), reported an approximate 30% decline in Excreta/carcass partition coefficients (K_{ExC}) (over the log K_{OW} range of 5 to 7.5). The authors suggest the more pronounced 30% drop in K_{ExC} during the depuration experiment, compared to the slight 10% drop in absorption efficiencies during the uptake experiments (*115*), indicates that for the more hydrophobic PCBs gut-to-organism uptake exceeds organism-to-gut transfer (i.e., $D_{GB} > D_{BG}$).

2.3.4 "Fat Flush" diffusion hypothesis

Schlummer et al. (*95*) recently presented a fat-flush diffusion (FFD) model for intestinal uptake in humans. This model is based on the premise that the lipid influx into intestinal cells acts to enhance GI magnification and diffusive uptake from intestinal contents. The authors postulate that the fugacity capacity of intestinal cells (Z_I) increases during periods of active food digestion, when dietary lipids are hydrolyzed to monoglycerides and fatty acids and subsequently resynthesized into triglycerides in the enterocyte. The Z of the re-formed triglycerides in intestinal tissues (Z_{TRI}) is greater than the monoglycerides or fatty acids originating from the mixed micelles in the lumen (Z_{MIC}), (*109,123*), which thereby enhances the diffusion gradient from the lumen into the intestine. The resulting lipid swelling in the enterocyte causes a downward pressure on the fugacity in the intestinal cells as f_I is inversely proportional to Z_I. At the same time the fugacity in the gut lumen (i.e., GIT) is raised as Z_G drops as a result of lipid transfer from the lumen to the intestinal cells. This effect produces a positive fugacity gradient between the gut lumen and the intestinal cells resulting in net absorption. While the fat-flush effect has only been formalized to assess human dietary exposures, it is likely to also occur in other organisms (especially those which digest large quantities of lipids such as top-predator carnivores). Since previous fugacity measurements in dietary accumulation studies with fish by Gobas and colleagues indicate the fat-flush does not occur in fish (*40,41,42*), we will confine the following evaluation of the FFD model to humans.

Figure 2.7 illustrates the fat-flush effect over the course of a digestion event. At the time of ingestion f_D (1 nPa) is lower than f_I (80 nPa). As food digestion and absorption ensues, f_G increases and f_I drops simultaneously, allowing for net passive diffusion of chemical into the intestinal tissue (subscript I). During the rapid period of efficient lipid absorption and

chylomicron transport of resynthesized triglycerides there is a subsequent removal of chemical from the digesta in the GIT (causing f_G to drop as digestion proceeds). Equilibrium partitioning between the digesta and intestinal tissue continues over the 2-4 hour period of digesta transit in the upper gut until a partitioning equilibrium between the intestinal tissues and digesta is achieved (e.g., f_G is shown to equilibrate with f_I at 16 nPa). Absorbed lipids and chemical are transported from intestinal tissue to the liver, and eventually an equilibrium within the body is restored (f_I increases to 80 nPa). Simultaneously, the digesta (f_G = 16 nPa) is advectively transported into the lower digestive tract during the later stages of the digestion event and diffusive elimination back to the feces becomes possible because f_I (80 nPa) > f_F (16 nPa). In essence, the absorption of dietary lipids simultaneously causes a fugacity increase in the digesta moving through the upper intestine and simultaneously an influx of lipid pools in the brush border membrane, which in effect, increases the Z of the intestinal tissue (resulting in a temporary fugacity drop in intestinal tissue). The combined effect of the high fugacity in the gut contents (f_G) and the reduced fugacity in the intestinal tissue (f_I) facilitates efficient chemical absorption due to the temporary thermodynamic gradient. The fugacity in the lower digestive tract (i.e., feces) is only indirectly influenced by the fat flush, as the fat flush has subsided by the time the digesta arrive there.

Figure 2.8a further examines the FFD model predictions of a human subject from the general population, exhibiting POPs tissue residue levels approximately 80 times that of consumed food (i.e., f_B/f_D = BMF_f = 80). Specifically, the fugacity in the diet at 1 nPa may attain approximately a 16 fold fugacity increase in the upper gut (i.e. f_G =16 nPa) due to food absorption and digestion (i.e., GI magnification), and a simultaneous $\geq$ 5 times increase in the fugacity capacity of intestinal tissue Z_I, that causes the fugacity in the intestinal tissues to drop. The diffusive gradient from the digesta into the intestinal tissue results and chemical is taken up (i.e., net uptake occurs) until a partitioning equilibrium between the digesta and the intestinal tissue is achieved (e.g., f_D:f_G:f_I:f_B = 1:16:16:80). Consequently, in the lower digestive tract the fugacity in the digesta is considerably lower than in the intestinal tissue (i.e., f_B:f_I:f_G = 80:80:16). Based on the findings of Rozman et al. (*130*) that indicated contaminant elimination in rats occurs mainly in the large intestine, Schlummer and colleagues initially presumed that once the fat-flush subsides this fugacity gradient would result in diffusive elimination of chemical to feces in the lower digestive tract (similar to the MMD model). However, in later work Moser and McLachlan noted that the experimental evidence in the animal literature is inconsistent on this issue. For example, similar depuration studies in rats by Yoshimura and Yamamota (*131*) and Richter and Schafer (*132*) have indicated organism-to-intestine transfer of tetrachlorobiphenyl and hexachlorobenzene,

respectively, occurs by passive diffusion in the upper intestine. In their assessment of a non-absorbable fat substitute's effects on human digestive elimination of POPs, Moser and McLachlan concluded that chemical concentrations observed in the feces must be the result of an equilibration process within the intestinal tract, since increasing the fugacity capacity of the feces increased the rate of chemical elimination (*133*). Furthermore, laboratory measurements indicated that the fugacity in human feces is considerably lower than the fugacity in the body (*80*). This led them to conclude that the fugacity gradient in the lower digestive tract does not result in significant chemical elimination, likely due to the absence of micelles in the lower digestive tract and hence slower mass transfer from the brush border into the lumen of the intestine (*134,135*). Hence, they concluded the equilibration occurs in the upper gut.

McLachlan and colleagues have conducted several investigations on intestinal absorption of various POPs in humans and agricultural food chains, with a focus on the bioavailability and intestinal absorption/desorption kinetics of polychlorinated dioxins and furans (*83,84,95,119,120,136,137*). Studies involving human infants have shown net dietary absorption efficiencies of most POPs are typically greater than 90% (*136,137*), while similar studies in adult human subjects show more variable results, ranging from high absorption efficiencies of 87% to instances of net excretion (*95,120*). Studies by Schlummer et al. (*95*) and Rohde et al. (*136*) have shown that the net dietary absorption efficiency of a given compound in adult human subjects is highly dependent on the chemical concentration in blood lipids. If concentrations in blood lipids were comparable to levels in the background population, net contaminant absorption was generally observed. When observed concentrations in blood lipids were high compared to the background population (e.g., due to occupational exposure), net contaminant excretion was observed. Furthermore, the chemical elimination rate was linearly correlated with the concentration in the blood lipids. In fugacity terms, this implies that subjects excrete chemical if the blood-to-food fugacity ratio f_B/f_D is high, while net absorption typically occurs when fugacities in blood are low compared to food (i.e., low f_B/f_D ratios). These findings suggest that when tissue residue levels of a compound are high relative to the diet, efficient elimination occurs (likely due to a diffusive equilibration between the digesta and the wall of the upper digestive tract). This scenario is illustrated in figure 2.8b, showing a human subject who has approximately 15 times higher tissue residue levels compared to the general population (e.g., f_B = 1,200 nPa, perhaps due to occupational exposure) but consumes the same diet at 1 nPa (i.e., f_B/f_D = 1,200). In this case, the depression of the fugacity in the intestinal tissue (from 1,200 nPa to 240 nPa) is not sufficient to bring it below the fugacity in the digesta (which rises to 80 nPa due to food

digestion). Consequently, chemical moves along the diffusive gradient (from high fugacity in the body to low fugacity in the digesta) and net chemical excretion occurs until a partitioning equilibrium between the digesta and the intestinal tissue is achieved (i.e., f_D:f_G:f_I:f_B = 1:240:240:1,200).

The above evaluations of the fat flush effect indicate that the intestine/digesta partition coefficient (K_{IG}) at the point of chemical absorption and desorption in the upper GIT is a critical parameter. K_{IG} will largely be dependent on the respective volumes and fugacity capacities of those compartments (i.e., V_IZ_I versus V_GZ_G). Currently, the relative partitioning capacities and contaminant kinetics at the intestinal tissue-digesta interface are not fully understood. The question remains whether the dissociation of micelles at the gut wall (as described by the MMD model) precludes equilibrium of very hydrophobic POPs during chemical depuration back into the gut lumen. Specifically, further work is needed to resolve the issue of the disparity between D_{GB} and D_{BG}.

2.4 Discussion

The review of the various proposed models on dietary absorption and biomagnification illustrates that while there are some key differences, the models show a tendency to converge and build on each other and are not mutually exclusive. The digestion model illustrates the role of food digestion on dietary absorption and magnification on the organism level. The fat-flush and micelle mediated diffusion models describes how lipid digestion absorption and chemical intestinal absorption/desorption are linked at the tissue level and alerts us to the possible existence of a magnification mechanism in addition to food digestion. The models combined provide a good theoretical framework for exploring the role of physical-chemical properties on biomagnification that may be useful for chemical hazard assessment.

The key difference between the digestion and the micelle mediated diffusion model concerns the role of micelles in transporting chemicals across gastro-intestinal membranes. This particular part of the larger process of lipid absorption still remains unresolved. However, its use in models may have a significant impact on the selection of molecular descriptors for biomagnification. The MMD model predicts that BMF_{MAX} of a given compound is dependent on the chemical's K_{OW} primarily due to the fact that $D_{GB} > D_{BG}$. Specifically, the MMD model predicts the BMF_{MAX} of non-metabolizable compounds will be positively correlated with the chemical's K_{OW} because

unidirectional micellar transport (and hence dietary uptake) enhances the rate if absorption for high K_{OW} chemicals, while the reverse transport process from the organism to the gut lumen is reduced by an increase in K_{OW} (due to the absence of micelles in the lower GIT). Alternatively, the digestion model assumes passive diffusion across the gut wall encounters similar diffusive restrictions during absorption and desorption (i.e., $D_{GB} = D_{BG}$), and indicates that the maximum bioaccumulation potential of compounds (i.e. in absence of depuration via metabolism, urine excretion and other mechanisms) is relatively universal and equivalent to $(G_D/G_F)\cdot(Z_D/Z_G)$ which is largely independent of K_{OW}. The fat-flush model can effectively complement both the digestion or micelle mediated diffusion model, depending on how intestinal desorption (organism-to-gut elimination) is envisioned. Currently, the influence of K_{OW} on the BMF_{MAX} in the fat-flush diffusion model is not fully understood and is essentially dependent on whether contaminant absorption and desorption are assumed to be decoupled processes or an equilibration in the upper GIT (i.e., $D_{GB} = D_{BG}$ versus $D_{GB} > D_{BG}$).

Regardless of the intestinal absorption/desorption mechanism, the BMF is ultimately the result of competing rates of chemical uptake from the gastro-intestinal tract (GIT) and other potential chemical elimination routes (i.e., respiration, urinary excretion and metabolism), which are determined by a combination of organism physiology and the physical-chemical properties of the compound. For aquatic poikilotherms, respiratory elimination makes a key contribution to the overall elimination of hydrophobic organic chemicals. Elimination to water (i.e., gill ventilation) has been repeatedly demonstrated to be inversely related to the chemical's K_{OW}. Hence, an increase in K_{OW} causes a slower rate of chemical elimination from the organism, allowing the fugacity in the organism to achieve levels that are closer to that in the gastro-intestinal tract. For non-metabolizable chemicals with K_{OW}'s greater than 10^5, respiratory elimination is small compared to dietary elimination and biomagnification occurs. For air-breathing homeotherms, respiratory elimination is not to water but to the air. Respiratory elimination via lipid-air exchange declines with increasing octanol-air partition coefficient (K_{OA}), causing chemicals to approach a maximum biomagnification potential with increasing K_{OA}. It has been suggested that if K_{OA} exceeds 10^5, respiratory elimination is too small to effectively reduce the biomagnification effect in the GIT of many mammals hence biomagnification can occur (*44*). Only if the substance is rapidly eliminated to urine (e.g., log K_{OW} is less than approximately 2) or rapidly metabolized, can biomagnification be prevented. Diminished BMFs due to metabolic transformation are more common in birds and mammals compared to fish, since those organisms generally have a greater capacity to metabolize organic contaminants (*2,138,139*). The bioaccumulation potential of

organic chemicals in aquatic organisms is best assessed by K_{OW}, while bioaccumulation potential in air-breathing organisms is best described by K_{OA} and K_{OW} *(44,100)*. If K_{OW} and K_{OA} were to follow a simple single universal relationship among chemical classes, it would be possible to use K_{OW} alone as a predictor of biomagnification, but this is not the case *(44,100,140)*. Based on their K_{OW} and K_{OA}, chemicals can be categorized in four groups: polar non-volatiles (PNVs), non-polar non-volatile compounds (NPNVs), non-polar volatile (NPV) or polar volatile (PVs). Figure 2.9 illustrates this categorization using a limited number of chemicals for which bioaccumulation properties are relatively well known. Polar volatiles (bottom left quadrant) include compounds such as styrene and vinyl chloride and have no inherent bioaccumulative properties in either air-breathing or aquatic organisms. NPNVs (top-right quadrant) represent the majority of POPs such as PCBs and several organochlorine pesticides (e.g., mirex) and are inherently bioaccumulative in both aquatic and air-breathing organisms. Polar non-volatiles (PNVs, top-left quadrant) do not biomagnify in aquatic organisms (due to low K_{OW}), but may substantially biomagnify in air-breathing organisms (due to a high K_{OA}) unless they are efficiently metabolized at a significantly high rate or depurated by urinary excretion. Examples of these relatively hydrophilic compounds exhibiting some degree of bioaccumulation potential include hexachlorocyclohexanes (HCHs), endosulfan, atrazine, bis-4-chlorophenyl sulfone (BCPS), tris-chlorophenyl methanol (TCPMeOH) and PFOS. Figure 2.9 shows no existing compounds with non-polar volatile characteristics (bottom right). This group of chemicals may be quite rare, but theoretically involve chemicals with an inherent potential to biomagnify in water-respiring organisms but not in air-breathing organisms.

We feel that current regulatory initiatives aimed to identify bioaccumulative substances do not fully recognize some of the fundamental processes controlling biomagnification in air-breathing homeotherms and aquatic poikilotherms. This is a serious short-coming of current regulatory initiatives as the bioaccumulative properties of many commercial chemicals may be mis-assessed or underestimated. This review illustrates that there is significant evidence from theory (highlighted in this paper) and practice to indicate that BMFs in homeotherms can not only be higher than those in aquatic organisms but also follow different relationships with the physical-chemical properties of chemicals. Considering that the PBT regulations are primarily geared to protect human health, this is a matter of some priority. Further investigations into the mechanism of bioaccumulation in homeotherms is important, as is research on the parameterization of bioaccumulation models. Measurements of feeding rates, chemical and lipid absorption efficiencies, the disparity between gastro-intestinal transport parameters D_{GB} and D_{BG}, intestine-

digesta partition coefficients (K_{IG}), fugacity capacities of food, digesta and fecal matter, digesta transit times and steady state fugacity ratios between food:digesta:feces:organism (i.e., f_D:f_G:f_F:f_B) are likely to be of crucial importance in this endeavor.

2.5 Acknowledgements

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies.

2.6 Tables

Table 2.1 Summary of physiological parameters, feeding rates, digestion efficiencies and observed biomagnification factors (BMF_C lipid wt.) for fish (poikilotherms) and birds and mammals (homeotherms).

	Rainbow [a] trout	Dairy [b] cow	Caribou [c]	Ring [d] Dove	Harp [e] seal	Humans [f]	Stellar [g] Sea Lion	Wolves [h]
Body weight (BW), (kg)	0.439	250	140	0.16	57	80	600	90
Whole body lipid content (L_B) (%BW)	3%	20%	4%	17%	42%	20%	40%	21%
Energetic requirements (% BW· d^{-1})	1.2%	2%	2.1%	7.7%	3.3%	1.1%	5%	2.8%
Feeding rate, kg· d^{-1}	0.004	4	3	0.01179	1.881	0.9	30	2.5
Lipid content food (%)	18%	1 %	0.05%	9.10%	8%	20%	9%	12%
Fecal egestion rate, kg· d^{-1}	0.002	1.6	1.2	0.0039	0.38	0.032	1.80	0.13
Lipid assimilation efficiency (%)	92%	60%	60%	98%	94%	95%	97%	98%
Food assimilation efficiency (%)	50%	60%	60%	70%	80%	92%	94%	95%
G_D/G_F	2	2	2.5	3.4	5	13	17	20
Z_D/Z_G [i]	4	2	0.8	5	4	6	6	7
E_{MAX}	60%	80%	-	97%	-	100%	-	-
Observed BMF_{MAX} (C_B/C_D lipid wt.)	8	4	3	52	23	≥ 80	≥ 100	≥ 100

[a] Rainbow trout data from (*42*) [b] Dairy cow data from (*83,119*); [c] Caribou data from (*44*); [d] Ring dove data from (*115,116*); Harp seal data from (*117*); [f] Human data from (*95,120*); [g] Stellar sea lion data from (*118,121*); [h] Wolf data from (*44*); [i] Z_D/Z_G ratios were estimated using 3-phase partitioning model in *44*.

2.7 Figures

Figure 2.1 Reported biomagnification factors (BMF_{MAX}) of PCB153 in organisms from various freshwater, terrestrial and marine ecosystems. BMF_{MAX} values represent lipid equivalent or lipid normalized concentration-based BMFs (C_B/C_D lipid). References for BMF data are as follows: Amphipods (*6*) mysids, smelt and alewife (*82*), crayfish, zebra mussels and Lake trout (*81*), Atlantic cod and ringed seals (*46*), Arctic char, beluga whale and polar bears (*63*), caribou and wolves (*43*), Dairy cows (*84*), shrews (*85*), mustelids and otters (*89*), humans (*95*), gray whales (*93*), North Atlantic Right whale (*94*), bottlenose dolphin (*91*), Baikal seals (*92*), herons (*86*), herring gulls (*87,88*), Minke whales (*90*), and killer whales (*8*). White bars represent invertebrates, light gray bars represent fish, dark gray bars represent terrestrial mammals and black bars represent marine mammals.

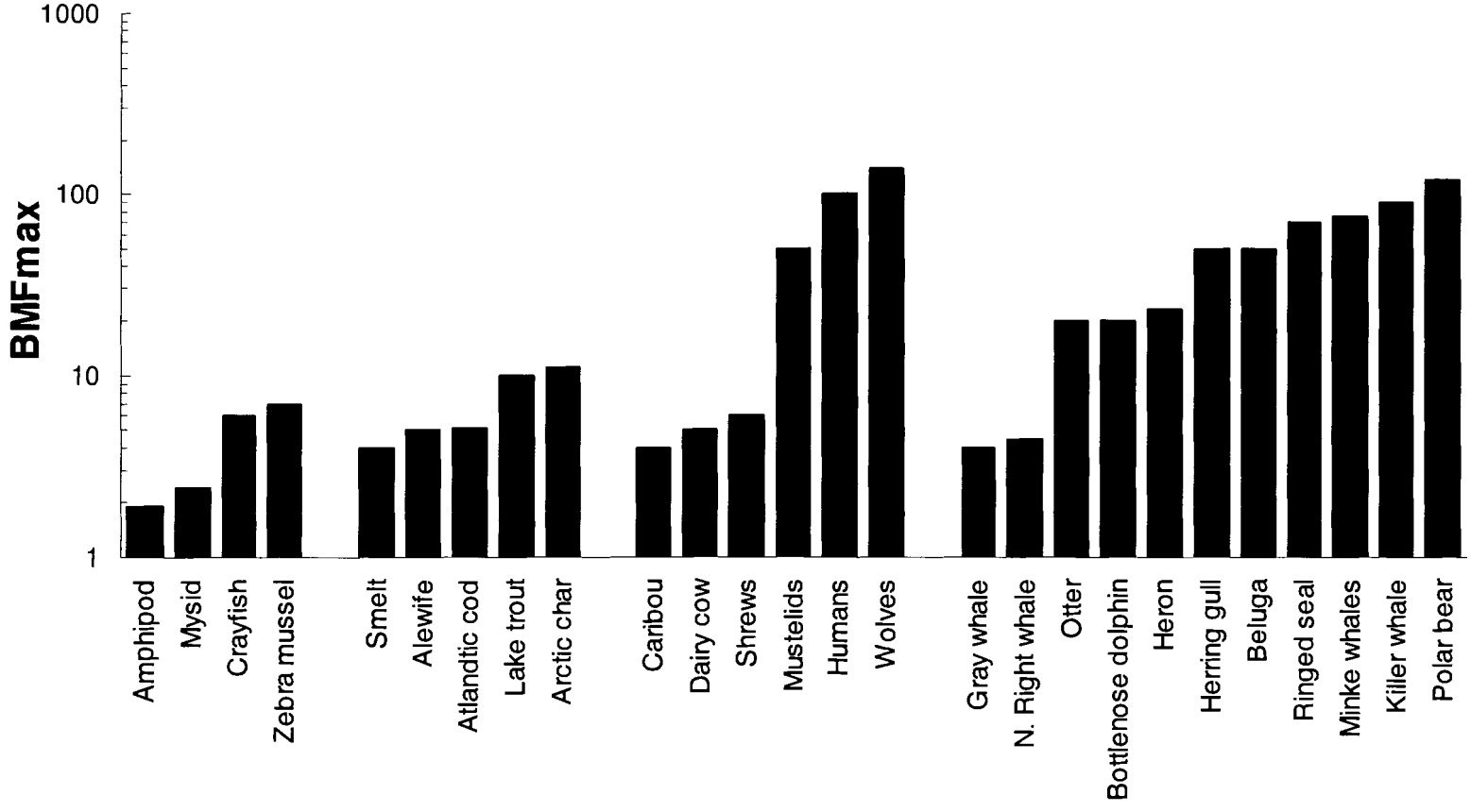


Figure 2.2 Schematic illustration of gastro-intestinal uptake of dietary fats and xenobiotics. Illustration, modified from reference (*106*).

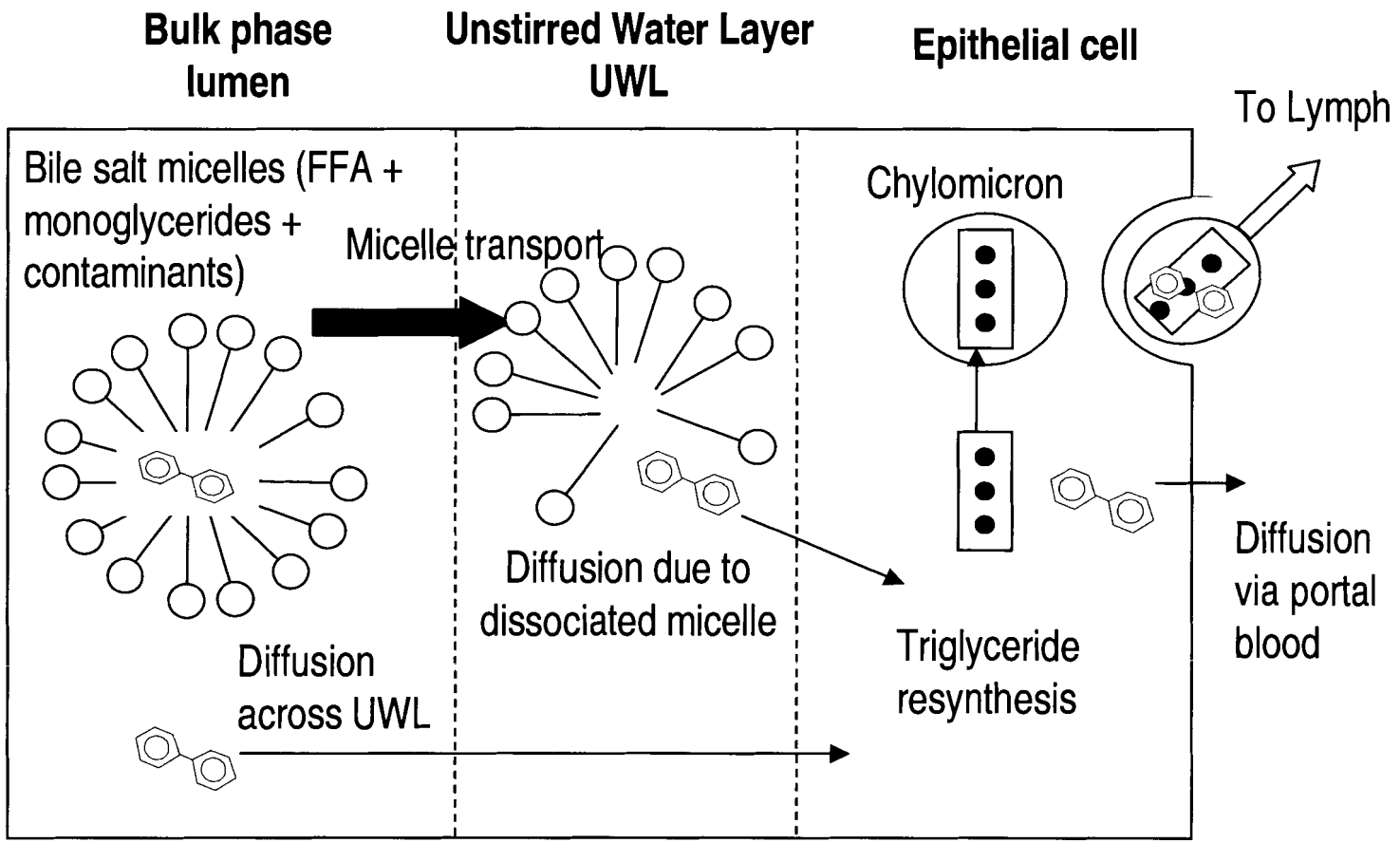


Figure 2.3 Conceptual illustration of a two-compartment model of uptake and elimination of organic chemicals in a generic water-ventilating or air-respiring organism. The gastro-intestinal tract compartment (denoted as subscript G) is shown to include the intestinal wall (denoted as subscript I), separated by an unstirred water layer (UWL).

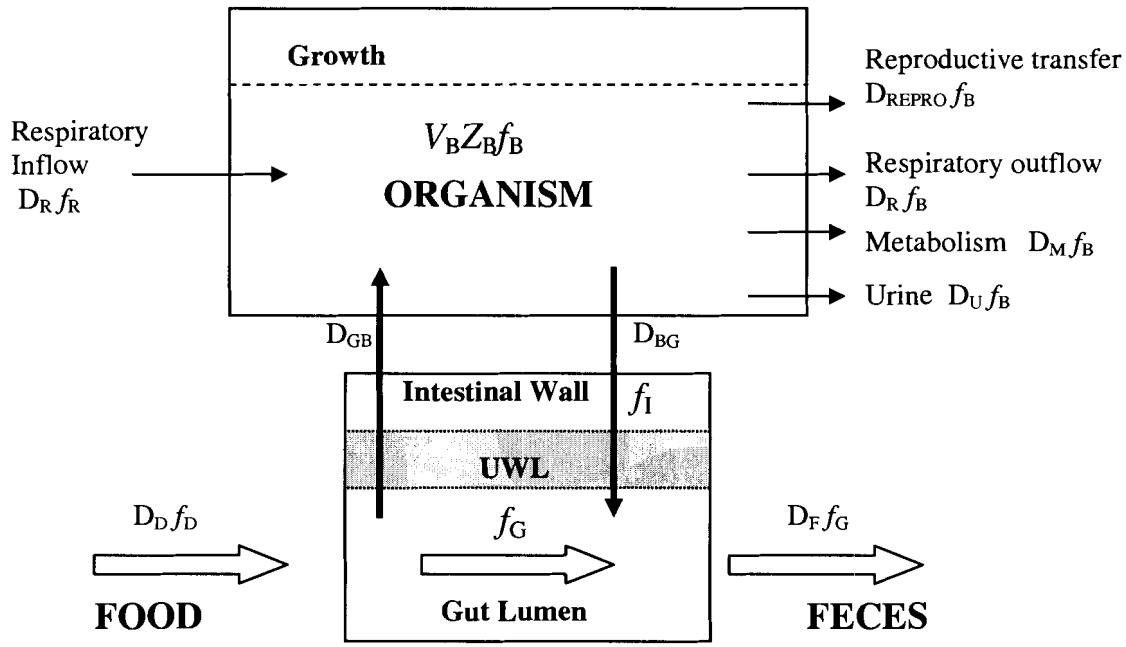


Figure 2.4 Steady state conditions of chemical fugacities in the gastro-intestinal tract (GIT) and organism for fish and homeothermic organisms such as birds and mammals following gastro-intestinal magnification model. The gastro-intestinal tract compartment (denoted as subscript G) is shown to include the intestinal wall (denoted as subscript I), separated by an unstirred water layer (UWL).

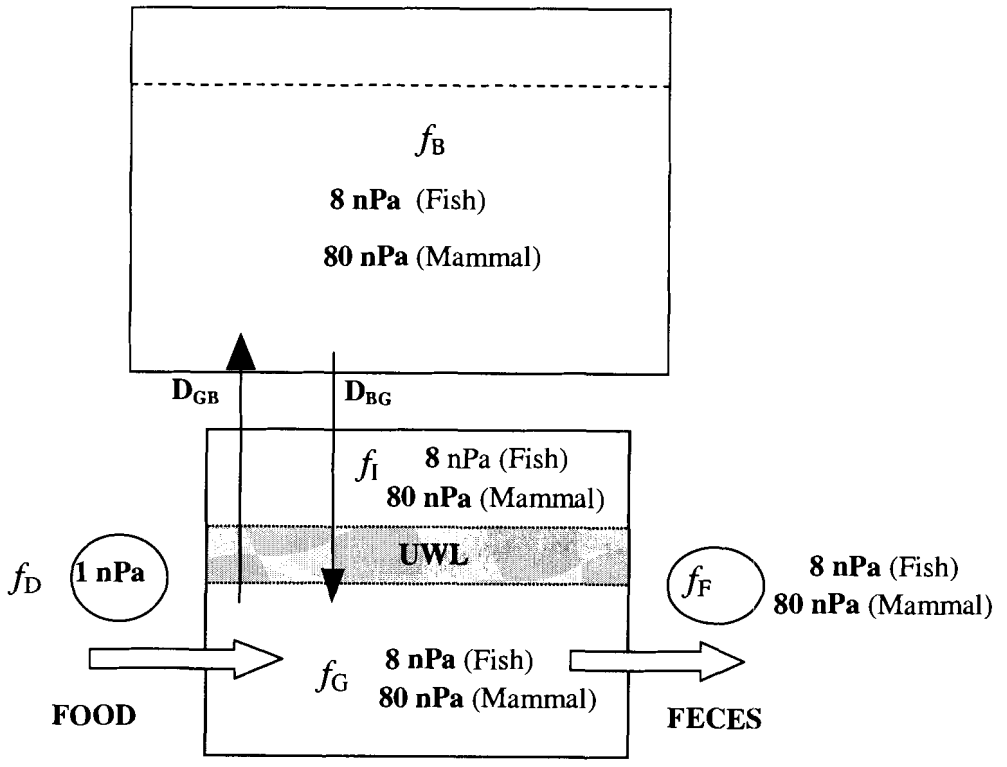


Figure 2.5 Dietary absorption efficiencies of various POPs reported in the literature for several organisms, including fish (*39*), dairy cows (*119*), humans (*120*) and ring doves (*115*) versus chemical log K_{OW}. Trend lines represent non-linear regressions: $1/E_D = 5.3 \times 10^{-8} K_{OW} + 2.3$ for fish data; $1/E_D = 2.9 \times 10^{-8} K_{OW} + 1.2$ for dairy cows; $1/E_D = 2.4 \times 10^{-9} K_{OW} + 1.04$ for ring doves; and $1/E_D = 1.55 \times 10^{-9} K_{OW} + 1.01$ for human data.

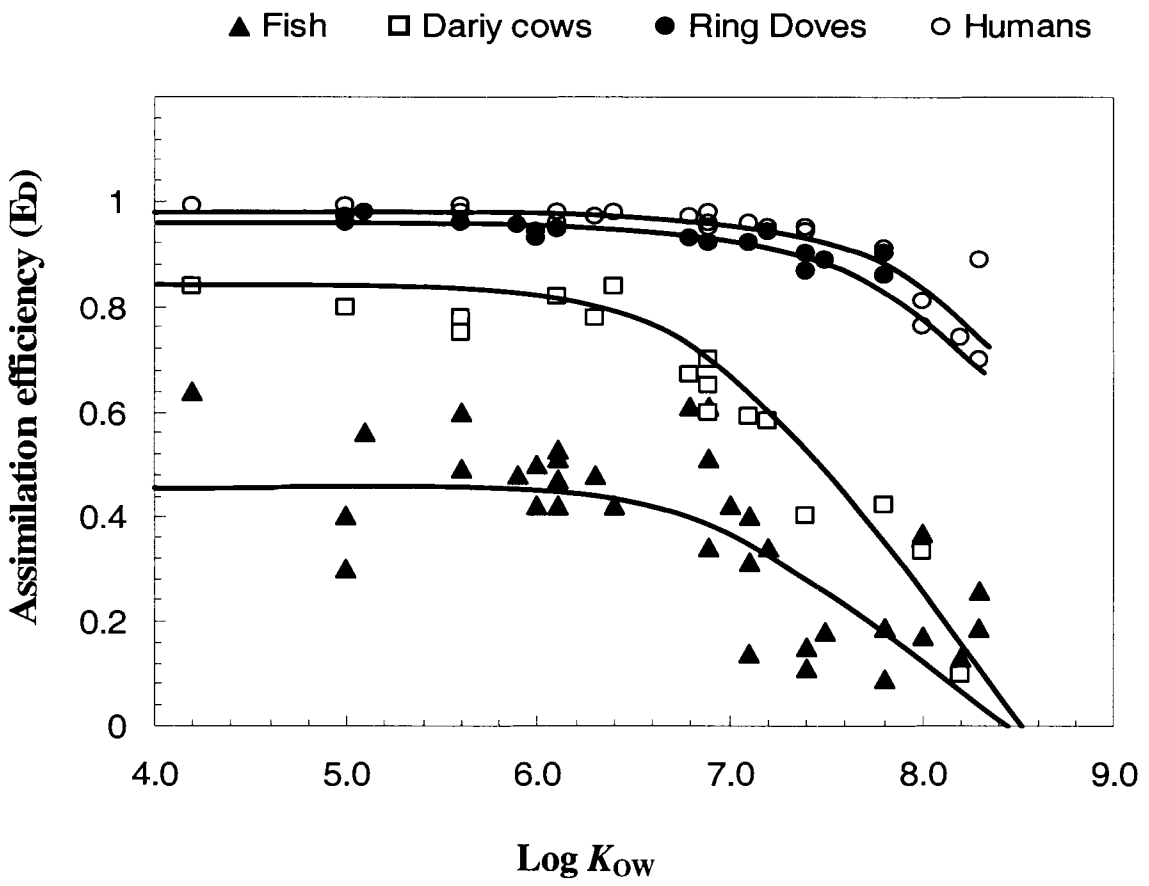


Figure 2.6 Steady state conditions of chemical fugacities in the GIT and organism for fish and homeothermic organisms such as birds and mammals following the unidirectional micelle mediated diffusion model. The gastro-intestinal tract compartment (denoted as subscript G) is shown to include the intestinal wall (denoted as subscript I), separated by an unstirred water layer (UWL). Model predictions based on mixed micelle enhancement factor, i.e., D_{GB}/D_{BG} ratio of approximately 1.2 for fish and 2.0 for bids and mammals.

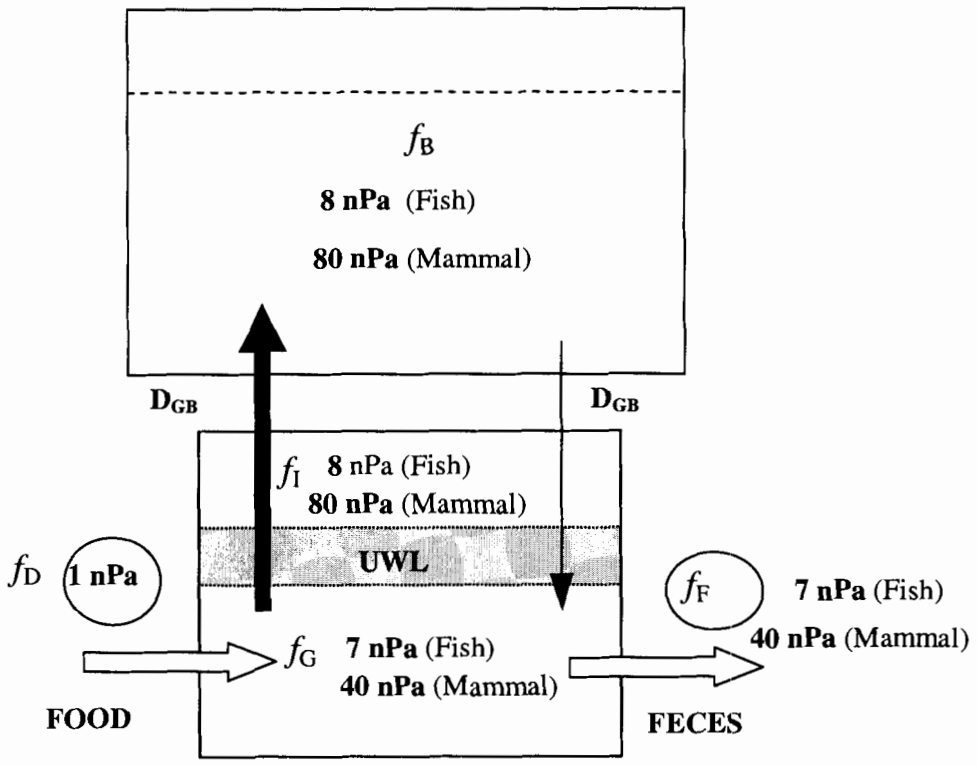


Figure 2.7 Schematic illustration of the fat-flush hypothesis, modified from Schlummer et al. (95)

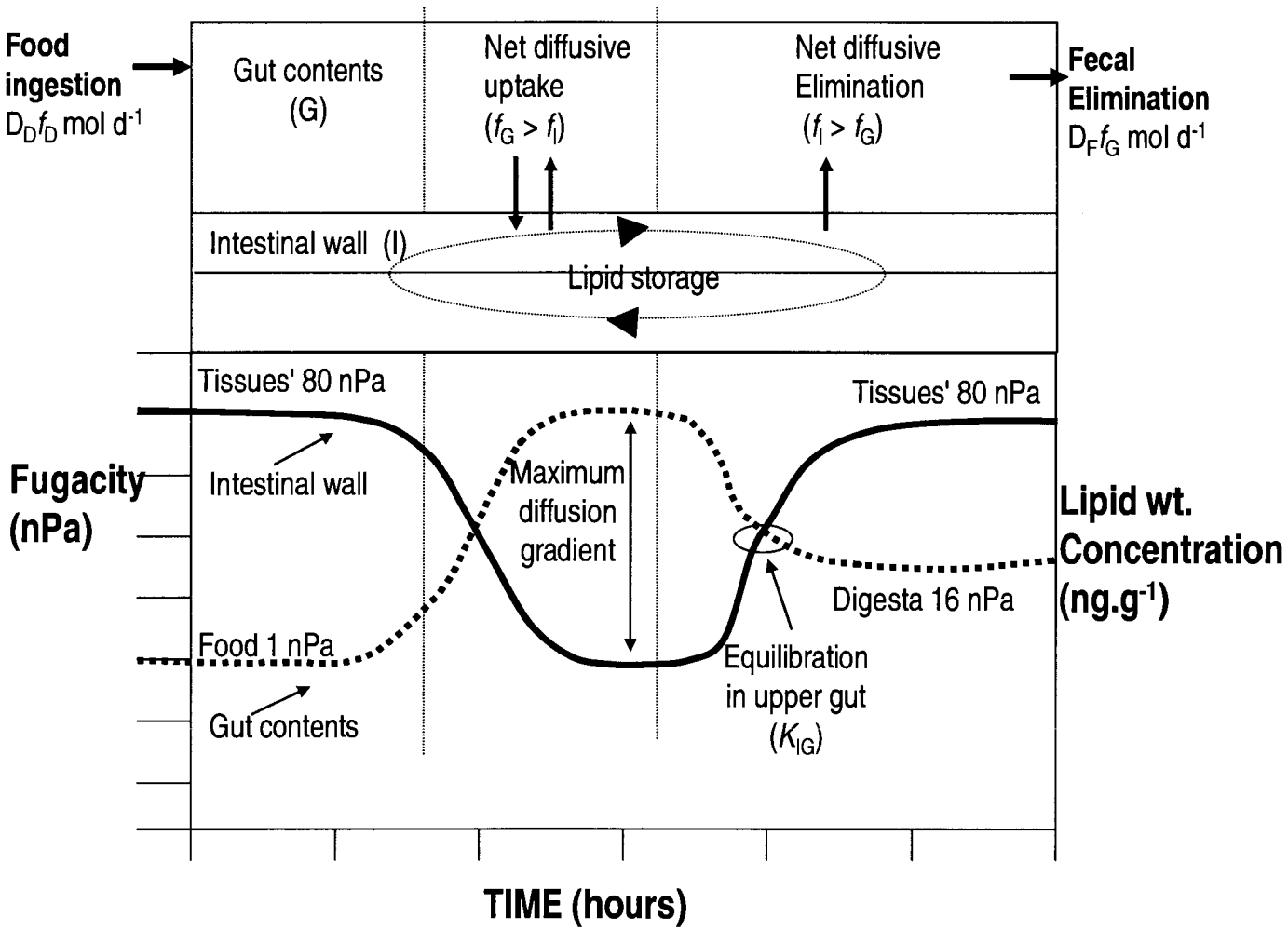


Figure 2.8 Steady state conditions of chemical fugacities in the GIT and organism following the fat-flush diffusion model for (a) human subjects from the general population (b) occupationally exposed persons. The gastro-intestinal tract compartment (denoted as subscript G) is shown to include the intestinal wall (denoted as subscript I), separated by an unstirred water layer (UWL).

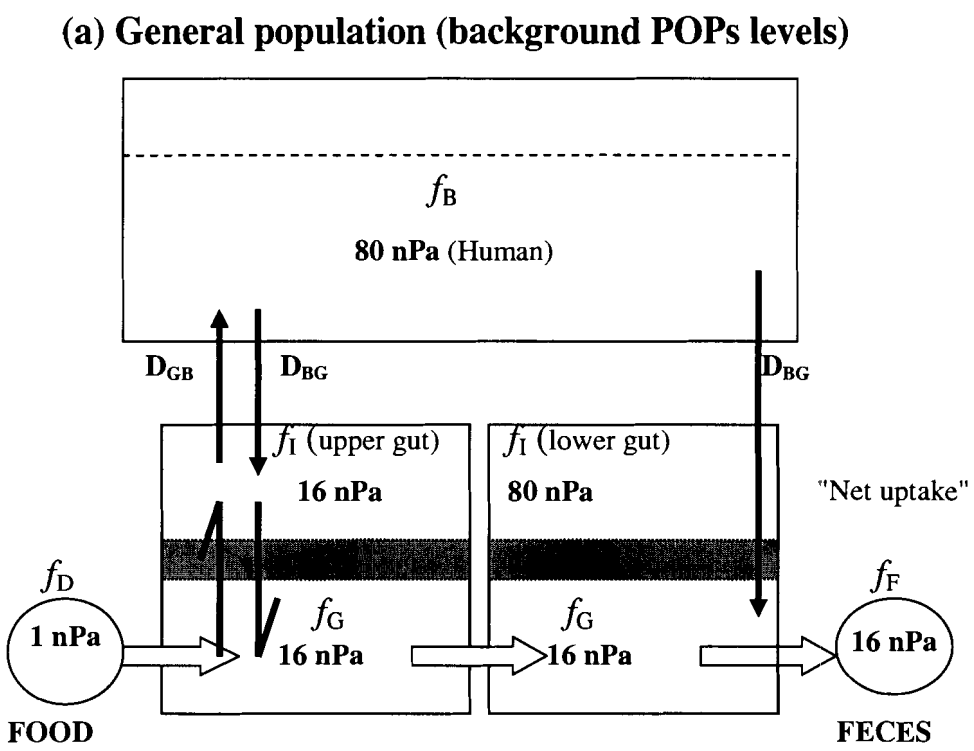


Figure 2.8 continued

(b) Occupational exposure (elevated POPs tissue residue levels)

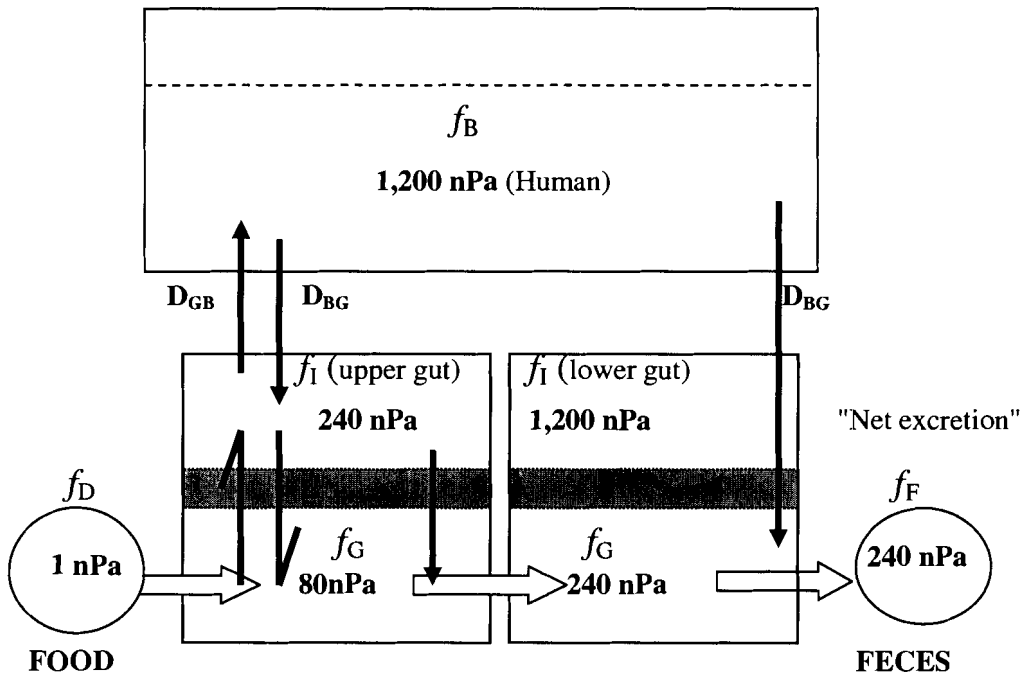
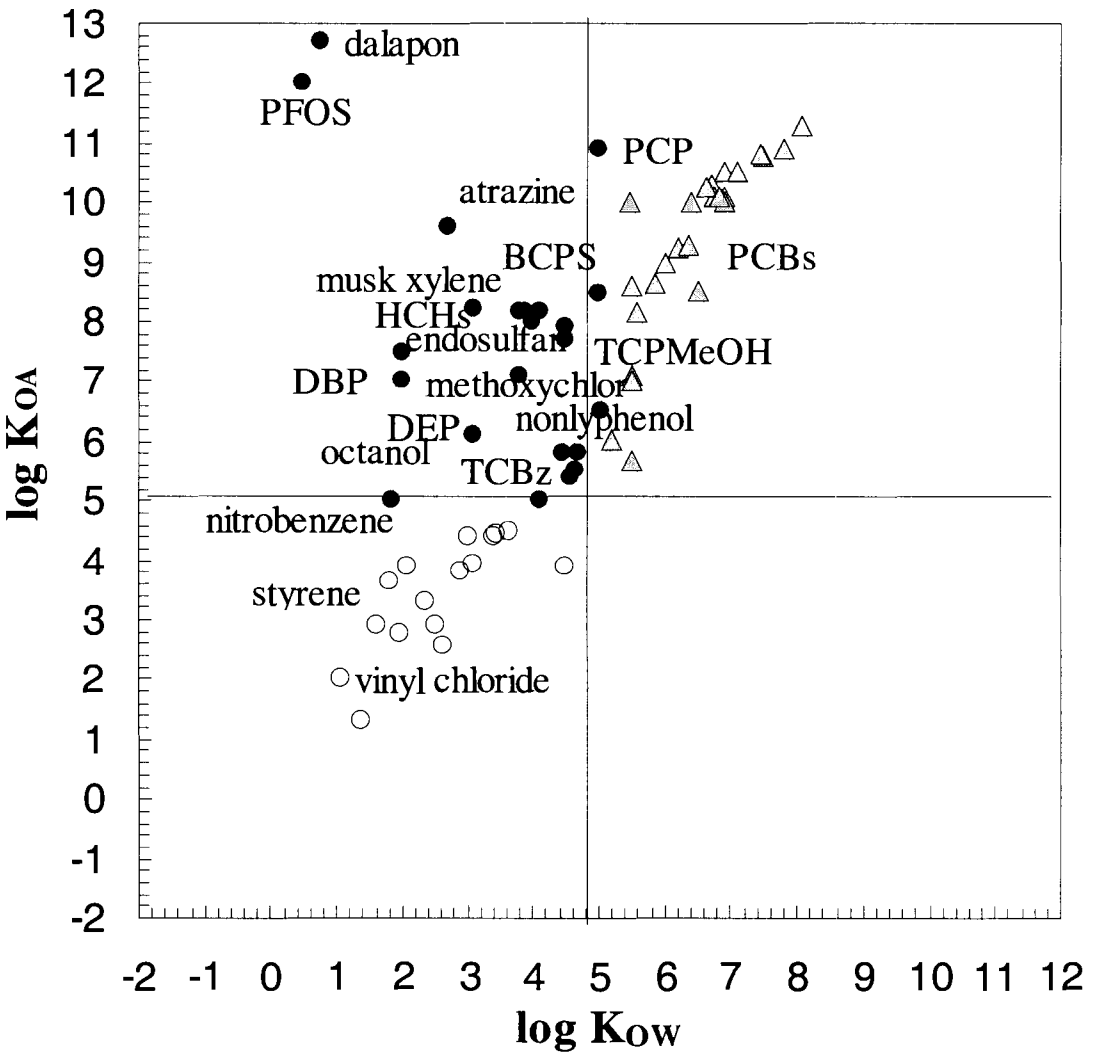


Figure 2.9 Plot of log K_{OW} versus log K_{OA} for various organic chemicals, characterized into four quadrants: polar non-volatile (PNVs), non-polar non volatile (NPNVs), polar volatile (PVs) and non-polar volatile (NPV). Open circles represent polar/volatile, low K_{OW} – low K_{OA} chemicals. Solid circles represent PNVs, low K_{OW} – high K_{OA} chemicals. Solid triangles represent NPNVs, high K_{OW} – high K_{OA}. PCP = pentachlorophenol, PFOS= perfluoro-octane sulfonate, BCPS = bis (4-chlorophenyl) sulfone, HCH = hexachlorocyclohexanes, TCPMeOH = tris (4-chlorophenyl) methanol, DEP = diethyl phthalate, DBP = di-n-butyl phthalate, TCBz = trichlorobenzenes



CHAPTER 3

BIOACCUMULATION OF POPS IN A CANADIAN ARCTIC MARINE FOOD WEB AND RELATED HUMAN DIETARY EXPOSURE OF AN ABORIGINAL, INUIT POPULATION

3.1 Introduction

Numerous studies involving the accumulation of industrial and agricultural chemicals such as PCBs and DDTs, hexachlorobenzene (HCBz), toxaphene and chlordane in Arctic ecosystems have been conducted over the past several decades (*2,75,141,142,143,144, 145*). These persistent organic pollutants (POPs) are thereby classified as they generally exhibit a high degree of environmental persistence (P), bioaccumulation potential in food chains (B) and are inherently toxic to organisms (T). In essence, the physical chemical (physical-chemical) properties of these compounds (i.e. non-labile, hydrophobic and semi-volatile) allows for the potential of (i) long-range transport (LRT) and subsequent deposition to the Arctic environment due to a cold condensation effect at low circumpolar temperatures (*1*), (ii) prolonged environmental persistence and (iii) biomagnification in the food chain (*2,4,75,142,143*). Elevated levels of PCBs and OC pesticides such as DDT, dieldrin, mirex and hexachlorobenzene in aboriginal peoples from the Canadian Arctic have been reported in recent years. For example, Dewailly and colleagues (*64,146,147*) showed PCB and OC pesticide levels in Inuit women from northern Québec were approximately 8 times higher than those levels in breast milk samples from women of European descent living in southern regions of Canada. Similar observations have been observed in Inuit from Greenland (*148,149*). Because of socio-economic and cultural reasons fish and seabirds and marine mammals represent a significant portion (15 - 45%) of the annual Inuit diet and hence exposure to environmental contaminants is of particular interest for public health authorities in the Arctic regions. Numerous studies of contaminant accumulation in the Canadian Arctic physical and biotic environments (including human exposure) have been conducted over the several decades. Aboriginal peoples in the Canadian north, including numerous Inuit communities, are involved in contaminant related research programs and are apprised of findings of toxicological significance (e.g., contaminated food sources). For example, the Canadian Arctic

Indigenous Peoples against POPs (CAIPAP) was actively involved in the United Nations Environment Program (UNEP) POPs protocol negotiations (*150*).

The majority of the twelve notorious (i.e., legacy POPs, known as the dirty dozen), recently been targeted for virtual elimination worldwide following international agreement on the 2004 Stockholm Convention on POPs (*80*), have already been banned or restricted from use since the 1970s (e.g., PCBs). While, this multilateral environmental agreement (MEA) is an important initial action towards better global environmental quality protection, there remain numerous other current-use commercial chemicals with similar physical-chemical properties not yet fully assessed for their PBT and LRT potential. For example, brominated flame retardants such as polybrominated diphenyl ethers (PBDEs), which are structurally similar to PCBs, have been detected at appreciable and exponentially increasing levels in Arctic biota and suggests these relatively high-production volume (HPV) substances also undergo long-range transport and substantially accumulate in Arctic food chains (*30,151*). Other HPV compounds of emerging concern include dialkyl phthalate esters (DPEs) and synthetic musks and current use pesticides such as endosulfan, are potential PBT LRT chemicals and are tentatively classified as "candidate" POPs. Thus, further work towards understanding the physical-chemical and biological factors affecting POPs bioaccumulation behaviour is important for assessing the environmental hazards of novel classes of compounds.

Accumulation of organic contaminants in marine ecosystems is controlled by a combination of chemical bioconcentration and biomagnification. Bioconcentration of exogenous chemical into environmental and biological media occurs mainly through passive molecular diffusion into lipid or other biomolecular substrates such as organic carbon. Organic carbon (OC) is a particularly important "solvent" for the sorption of water-borne contaminants to particulate matter and bottom sediments (*57,152*). Bioconcentration in biota is significantly influenced by the hydrophobicity of the chemical, denoted by the octanol-water partition coefficient (K_{OW}), and the organism's whole-body lipid fraction (ϕL). The bioconcentration factor (BCF) in water-ventilating organisms such as invertebrates and fish is the ratio of a chemical's equilibrium concentration in an organism (C_B, wet wt. basis) and freely dissolved water (C_{WD}), i.e., $BCF = C_B/C_{WD}$. BCFs in aquatic organisms tend to be greatest for highly lipophilic chemicals and in high trophic organisms (e.g., salmonids, > 10% lipid). Chemical biomagnification *via* dietary exposure and absorption can occur, resulting in chemical concentrations (on a lipid wt. basis) in an organism (C_B) that exceed concentrations in consumed prey (C_D), (*2,3,5,42,43,46*). The extent of

biomagnification is represented by a biomagnification factor (BMF), which is the ratio of the lipid corrected concentrations in a given predator (C_B, lipid) and its prey (C_D, lipid), i.e., BMF = C_B/C_D. Following chemical uptake *via* respiration (i.e., bioconcentration) and dietary absorption (biomagnification), the overall resulting bioaccumulation in the organism is dependent on the relative rates of chemical depuration *via* fecal egestion, respiration, urinary excretion, metabolic transformation and lactation (for female mammals). Rates of chemical uptake and depuration can vary substantially among compounds and organisms due to differences in (i) physical-chemical properties and (ii) organism physiology and toxicokinetics (e.g. water ventilating fish versus air-breathing mammals). It is well understood that hydrophobic organic contaminants such as PCBs and OC pesticides substantially biomagnify due to efficient absorption *via* dietary exposure and negligible depuration kinetics and oxidative biotransformation by cytochrome P450 enzymes. Cl_6-CB153 and Cl_7-CB180 tend to exhibit the maximum biomagnification factors in organisms (i.e., BMF_{MAX}) compared to other POPs (*5,37,39,43*) These recalcitrant compounds therefore serve as a standard for which to compare the bioaccumulation behaviour of other less understood organic contaminants.

The two main objectives of this study are to (i) determine concentrations and evaluate bioaccumulation patterns of PCB congeners and several OC pesticides in a Canadian Arctic marine food web and (ii) evaluate the relationship between POP concentrations observed in aboriginal, Inuit peoples and important traditional/country foods from within this food web. Specifically, this paper presents measured chemical concentrations of PCBs and OC pesticides in marine sediments and biota from a Canadian Arctic coastal marine food web, including samples of lichens, macro-algae, invertebrates, fish, seaducks and marine mammals. Levels, trends and evaluative parameters such as predator-prey biomagnification factors (BMFs) and biotransformation capacity index values and related human exposure of POPs *via* consumption of fish and wildlife are summarized and discussed.

3.2 Materials and Methods

3.2.1 Sample collections

During the months of May to August between 1999 and 2003 various environmental and biological samples were collected along the eastern Hudson Bay coastline. Beluga whale samples were mainly collected in close proximity to the Nastapoka River estuary near the Inuit village of

Umiujaq (64° 15′N 113° 07′ W) during the summer months (July-August), while ringed seal samples were obtained from various locations across northern Quebec and Labrador (Makovik), (Figure 3.1). For details see *Chapter 1, Section 1.9.1* and Appendix 1, which summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex, age and condition.

3.2.2 Food web characterization and designation of organism trophic positions.

Figure 3.2 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been established by extensive ^{15}N and ^{13}C isotope enrichment analyses involving numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of $TL = 1 + (\delta^{15}N - 5.4)/3.8$. More recent studies using $\delta^{15}N$ measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 (see *Chapter 1*) summarizes these previous $\delta^{15}N$ measurements and TL ranges for the various organisms within these Arctic marine food webs. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 4.5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific $\delta^{15}N$ measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson

Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

3.2.3 Extraction, cleanup and analysis of PCBs and OC Pesticides.

Select samples (approximately 10 g wet wt for lichens, macro-algae and sediment, 5-15 g for fish, 2 g for beluga whale liver and 0.5 g for beluga whale blubber) were homogenized with approximately 20 g Na_2SO_4 with mortar and pestle. Extracted fish tissue sub-samples consisted of excised muscle tissue (i.e., no skin), with the exception of capelin (which consisted of pooled whole fish). Sub-samples of other tissue samples (e.g., seaduck and marine mammal tissue samples) were excised from the interior of frozen samples to reduce potential contact contamination during collection and/or storage. The homogenate powder was transferred to a glass extraction jar, spiked with surrogate spiking standards for PCBs (^{13}C PCB congeners 28, 52, 101, 128, 156, 180, 194, 206, 209), Pesticides (d_3 1,3,5 Trichlorobenzene, ^{13}C 1,2,3,4 Tetrachlorobenzene, ^{13}C Hexachlorobenzene, ^{13}C beta HCH, ^{13}C lindane (gamma-HCH), ^{13}C mirex, ^{13}C oxychlordane, ^{13}C dieldrin, ^{13}C *p,p'* DDT, ^{13}C o,*p'*DDT, ^{13}C *p,p'* DDE, ^{13}C heptachlor epoxide, and ^{13}C *trans*-nonachlor). The spiked samples were then extracted with 30 mL of 1:1 (v/v) DCM/Hexane in a Branson 5210 ultrasonic water-bath (Branson Ultrasonics Co., CT) for 20 min. Once the suspended particles settled, the supernatant was removed, then extraction was repeated two more times with fresh solvent. The combined extracts were concentrated to ca. 2 mL with a gentle stream of high-purity nitrogen. Relatively low lipid samples (< 5% lipid w/w) such as cod and sculpin tissue were quantitatively transferred onto a 350 mm x 10 mm i.d. glass column packed with 8 g 100% activated florisil (60 -100 μm mesh, activated at 400 C overnight). High lipid samples (>5% lipid w/w) such as marine mammal blubber were first passed through a Gel Permeation Column (GPC) filled with 70 g of BioBeads, S-X33 (BioRad) in 50% DCM/hexane solution (V/V). The lipid fraction from the GPC (180 mL) was collected and discarded, while the remaining 300 mL of eluent from the GPC was collected evaporated to near dryness and solvent exchanged into hexane for further cleanup by Florisil. Three fractions were then eluted using 60 mL hexane (fraction 1), 60 mL 15% DCM/hexane (fraction 2), and 120 mL 50% DCM/hexane (fraction 3). The four fractions were combined in a single 500 mL boiling flask and evaporated to a final volume of 100 uL. The extracts were then spiked with recovery standards (^{13}C PCB 111 for PCB, and ^{13}C PCB 47 for pesticide quantifications) and analyzed by HRGC/HRMS using two separate instrument conditions (*153*). Method blanks, consisting of Na_2SO_4, were extracted according to the same procedure as

environmental samples and analyzed with every batch of twelve samples to check for contamination of the extracts.

3.2.4 Data analysis.

To enable direct comparisons of chemical concentrations between various environmental media and organisms it is important to correct chemical concentration data to a common unit expression. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~ 5 - 98%) , wet weight chemical concentrations (C, $ng \cdot g^{-1}$ ww) were expressed solely on a lipid weight basis by the equation: $C_L = C$ ww. $\div \phi L$ in units of $ng \cdot g^{-1}$ lipid. For matrix with very low lipid fractions ($\phi L < 1\%$), such as sediments, vegetation and algae tend to solubilize organic contaminants in non-lipid biomolecules such as organic carbon (OC) or non-lipid organic matter (NLOM) rather than in extractable lipids (*13,57,58,59*). Thus, sediments, macro-algae and lichens were normalized to a lipid equivalent fraction (ϕLeq) using the equation CLeq = Cww $\div$ ϕLeq. Lipid equivalent fractions (ϕLeq) for sediments were determined following reference (*35*) such that ϕ Leq = $\phi L + 0.35\phi_{OC}$, where the constant 0.35 represents findings that organic carbon has approximately 35% sorptive capacity of octanol. For macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and NLOM (ϕ_{NL}) fractions following the equation: ϕLeq = $\phi L + 0.035\phi_{NL}$, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42, 44*). Because chemical concentrations exhibited log-normal distributions the data were transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for individual compounds and compound class summations for the various samples collected and analyzed as part of the present study (i.e., sediments, lichens, macro-algae, bivalves, fish, beluga whales and ringed seals). In addition, we also compiled literature reported concentration data for PCBs and OC pesticides in Canadian Arctic biota , including invertebrates (*4*), walrus (*Odobenus rosmarus*) (*60*) polar bears (*Ursus maritimus*) (*61*), barren-ground caribou (*Rangifer tarrandus*) (*43,62,63*), wolves (*Canis lupus*) (*43,63*) and northern Quebec Inuit women (i.e., breast milk samples from references *64,63*) to compare contaminant concentrations, profiles and BMFs in various wildlife species and humans that generally subsist within the same food web.

PCB congeners were categorized by planarity and Cl-substitution patterns, following classifications presented by Boon and colleagues (*65*): i.e., Group I CBs, congeners without

vicinal hydrogen atoms are generally non-metabolizable CBs; Group II, congeners with vicinal *ortho-meta* H atoms and 2 *ortho* Cls have a limited metabolism potential in some organisms; Group III, same as II but with 1 *ortho* Cl can be metabolized by induction of methylcholanthrene (MC) type isozymes of the cytochromeP450 monooxygenase enzyme family (i.e., CYP 1A enzymes); Group IV, congeners with vicinal *meta-para* H atoms and ≤ 2 *ortho* Cls can be metabolized by induction of phenobarbitol (PB) type isozymes (i.e., CYP 2B enzymes); Group V, same as IV but with 3 *ortho* Cls may also induce CYP 2B type metabolism. A total of 169 di-ortho and mono-ortho substituted PCB congeners were analyzed (see Appendix 2). Due to several coeluting di-ortho (DO) and mono-ortho (MO) PCBs we have summarized a total of 148 PCB congeners. When environmentally dominant CB congeners coeluted with environmentally irrelevant congeners, we have for the purposes of this study, assumed the coeluting concentration as the single dominant compound. For example, CB153/132 concentrations (coeluting congeners in HRGC/MS method) are expressed solely as a CB153 concentration because of that congeners dominant contribution in environmental and biological samples. Specifically, this assumption was used for CBs 52, 101, 118 and 138. One-Way Analyses of Variance (ANOVA) and Tukey's HSD comparison tests were performed on calculated log-transformed concentrations to evaluate differences between mean chemical concentrations observed in sediments, invertebrates, fish and beluga whales.

3.2.5 Calculation of Biomagnification Factors (BMFs) and Elimination Index (EI).

See *Chapter 1, Section 1.9.5*

3.3 Results and Discussion

Levels of PCBs and organochlorine pesticides (OCPs) measured in E. Hudson Bay marine sediments and biota, including bivalves, fish, seaducks and marine mammals are show in Appendices 5 and 6, respectively. The data are not blank subtracted as procedural blanks for PCBs and organochlorine pesticides were generally low or non-detectable. Method detection limits (MDLs) were determined as the instrument limit of quantification (LOQ) on the HRMS. Levels and accumulation patterns of various PCBs and OCPs are described in the following sections.

3.3.1 PCB levels in E. Hudson Bay marine sediments and biota

Organic carbon normalized chemical concentrations in sediments were approximately 2 to 3 orders of magnitude higher than those dry wt. concentrations, primarily due to the very low OC content of these sandy sediments (0.179 ± 0.099%). Organic carbon normalized ∑CB concentrations in sediments were approximately 63.2 $ng \cdot g^{-1}$ OC wt. Cl_2 and Cl_3 congeners generally exhibited greater concentrations than higher chlorinated Cl_4-Cl_7 congeners in sediments. For example, concentrations of Cl_2-CB8/5 Cl_3-CB28 and Cl_6-CB153 were 3.25, 2.28 and 1.45 $ng \cdot g^{-1}$ OC wt., respectively. ∑PCB concentrations ($ng \cdot g^{-1}$ lipid equivalent wt.) varied widely among E. Hudson Bay biota species, including 1.30 $ng \cdot g^{-1}$ in macro-algae, 4.22 $ng \cdot g^{-1}$ in lichens, 60.7 $ng \cdot g^{-1}$ in cod, 602 $ng \cdot g^{-1}$ in male ringed seals, 734 $ng \cdot g^{-1}$ in eider ducks, 2,950 $ng \cdot g^{-1}$ in white-winged scoters and 3,690 $ng \cdot g^{-1}$ in male beluga whales. Dewailly and colleagues (*146,154,155*) reported relatively high concentrations ($ng \cdot g^{-1}$ lipid wt.) of PCBs and OC pesticides in human breast milk from northern Quebec Inuit women during the late 1980s, including a mean ∑PCB concentration of approximately 1,050 ± 141 $ng \cdot g^{-1}$ lipid wt. ,which is approximately 10 times lower than ∑PCB concentrations of approximately 10,200 $ng \cdot g^{-1}$ lipid wt. observed in E. Hudson Bay polar bears, another apex predator species (*61*). A more recent survey of breast milk from northern Quebec Inuit women between 1996 and 2001 (*156*) showed ∑PCB concentrations in Inuit breast milk (386 $ng \cdot g^{-1}$ lipid wt) have declined slightly since the 1980s. The predominant congeners we measured in marine biota samples included Cl_2 -CB8/5, Cl_3-CB28, Cl_3-CB31, Cl_4-CB52, Cl_5-CB95, Cl_5-CB101, Cl_5-CB110, Cl_5-CB118, Cl_6-CB153, Cl_6-CB138, Cl_7-CB187/182, Cl_7-CB180, with Cl_6 congeners CB153 and CB138 generally exhibiting the highest levels in organisms. The highest mean concentrations of the dominant CB153 hexachloro congener were observed in white-winged scoters (841 $ng \cdot g^{-1}$ lipid) and 16-35 year old male beluga whales (518 $ng \cdot g^{-1}$ lipid).

3.3.2 Organochlorine pesticide (OCP) levels in E. Hudson Bay marine sediments and biota

For OCPs, 1,2,4 TriCBz , PeCBz, HCBz, HCH isomers, *trans*-nonachlor, *p'p'* DDT and *p'p'* DDE were the dominant contaminants detected in E. Hudson bay sediment and biota. Primary metabolites of heptachlor (heptachlor epoxide), technical chlordane (oxychlordane) and endosulfan (endosulfan sulfate) were also present in fish, seaducks and marine mammals. Lipid equivalent OC pesticide concentrations in lichens and macro-algae were low in the 0.1 to 1 $ng \cdot g^{-1}$ range with a rank order of ∑HCHs > ∑CBz > ∑DDTs > ∑Chlordanes. For fish, seaducks and

marine mammals, OC pesticide levels were in the 10 to 1,000 ng·g^{-1} lipid wt. range, with a rank order of ∑DDTs > ∑Chlordanes > ∑CBz > ∑HCHs, and are comparable to other studies of POPs in Arctic fish, seabirds and marine mammals (*2,6,139*). Other cyclodiene pesticides including β-endosulfan, aldrin, dieldrin and mirex were present in biota samples in the 1 -50 ng·g^{-1} lipid wt. range. The highest lipid normalized pesticide concentrations in E. Hudson Bay biota were observed in male beluga whale blubber, HCBz (346 ng·g^{-1}), *p'p'* DDE (1,700 ng·g^{-1}) and *trans*-nonachlor (872 ng·g^{-1}). Dewailly and colleagues (*155,156*) reported OC pesticides in human breast milk from northern Quebec Inuit women during the late 1980s, including dieldrin (37 ng·g^{-1}), HCBz (136 ng·g^{-1}) and *p'p'* DDE (1,212 ng·g^{-1}), which in some cases were eight to ten times higher than pesticide residue levels in Caucasian Canadian women from southern Quebec. Similar to PCBs, concentrations of OC pesticides in from the 1996-2001 Inuit breast milk survey (*156*) were approximately two to five times lower than those concentrations measured in the late 1980s, including HCBz (50.2 ng·g^{-1}) and *p'p'* DDE (420 ng·g^{-1}). For beluga whales, primary metabolite concentrations were equal to or greater than concentrations of the parent compound. For example, levels of oxychlordane (732 ng·g^{-1}) and heptachlor epoxide (201 ng·g^{-1}) were ≥ than technical chlordane constituents (*cis* and *trans* chlordane and heptachlor, at approx. 1 - 170 ng·g^{-1}). Concentrations of *p'p'* DDE (1,700 ng·g^{-1}) were approximately four times greater than *p'p'* DDE (428 ng·g^{-1}), (i.e., DDE/DDT ratio ~ 4). In contrast, concentrations of endosulfan sulfate (0.86 ng·g^{-1} lipid wt.), the primary metabolite of endosulfan was relatively low (i.e., near detection limits) compared to β-endosulfan (12.6 ng·g^{-1} lipid wt.). While β-endosulfan was commonly detected in fish, marine mammals and seaduck tissues, α-endosulfan was only consistently detected at very low levels in male ringed seal blubber (0.3 ng·g^{-1} lipid wt.). There currently exists a paucity of endosulfan concentration data for Arctic biota. Our results for E. Hudson Bay marine biota suggest (i) β-endosulfan is the dominant isomer (with α-endosulfan only being detected in male ringed seals) and (ii) marine mammals and seaducks may efficiently metabolize/eliminate both parent endosulfans (i.e., α and β isomers) and the primary metabolite. Phase I biotransformation products (i.e., primary metabolites such as endosulfan sulfate) can undergo additional Phase II metabolic reactions involving conjugation of endogenous hydrophilic biomolecules (e.g., Glucuronic acid) thereby enhancing water solubility and further elimination through urine and/or bile (*113,117,157,158,159,160*).

3.3.3 Comparison to other Arctic POPs bioaccumulation studies.

An important distinction between our study and previous studies of Arctic marine ecosystem contamination is the use of HRGC/HRMS for chemical concentration quantification (as compared to GC ^{63}Ni electron capture detection (ECD) typically used in past analyses). For example, much of the POPs concentration data for Canadian Arctic biota, generated by the Northern Contaminants Program (NCP) over the past decades for invertebrates and fish (*4,150,161,162,163*), marine mammals (*150,163,164*) and seabirds (*150,163*) are the result of GC-ECD quantification. While direct comparison is difficult due to spatial and temporal differences, these previous reports of POPs in Arctic in biota are in general agreement with our measured POPs concentrations by HRGC/HRMS. For example, previous reports of concentrations ($ng \cdot g^{-1}$ lipid wt.) in Arctic cod for $\sum$PCBs (range = 66-95), $\sum$DDTs (range = 66-120) and $\sum$HCHs (range =39-49), which are comparable to our measured concentrations of 60.7, 50.1 and 9.8 $ng.g^{-1}$ lipid wt. for $\sum$PCBs, $\sum$DDTs and $\sum$HCHs, respectively. Similarly, previous concentration measurements of POPs in E. Hudson Bay beluga whales (between 1995 and 2000) by GC-ECD (*164*) are generally comparable to those concentrations we determined for E. Hudson Bay belugas in this study (1999-2002). For example, previous concentrations ($ng \cdot g^{-1}$ lipid wt.) in male beluga whales (blubber sampled near Pangnirtung, during 1996 and 1997) for PCB-153 (range = 366-556), *p'p'* DDE (range = 1,700 - 3,670) and β-HCH (range =25-53), which are comparable to our measured concentrations of 518, 1,700 and 42.3 $ng.g^{-1}$ lipid wt. for PCB-153, *p'p'* DDE and β-HCH, respectively.

We did however observe a substantial difference between our measurements and previously reported concentrations of endosulfan sulphate (primary metabolite of endosulfan). For example, concentrations reported in male beluga whales from Sanikiluaq (1996-1998) ranged between 33.7-60.9 $ng \cdot g^{-1}$ lipid wt (*164*), while our concentrations were low (approx. 0.9 $ng \cdot g^{-1}$ lipid wt) and generally near detection limits. Reasons for the approximately 50 times higher endosulfan sulfate concentrations in the previous studies of Arctic beluga whales compared to the E. Hudson Bay animals in the present study are not apparent. However, it is conceivable that previous measurements of endosulfans in Arctic beluga whales could be artifically high due to mis-identification/coelution of analytes as those samples were analyzed using GC-ECD, rather than HRGC/HRMS. Clearly, some degree of cross-checking of GC-ECD with HRGC/HRMS results would be prove beneficial for future chemical analyses of Arctic field samples.

3.3.4 Contaminant accumulation patterns of PCBs and organochlorine pesticides.

Figure 3.3 illustrates the relative congener contribution (i.e., % composition) for total organochlorines (PCBs+ OC pesticides) observed in E. Hudson Bay sediment and biota. In these plots, contaminant burden profiles shown for lichens (collected on land in close proximity to marine sampling locations) can be viewed as an atmospheric "signal" resulting from air-borne contaminant exposure processes. Similarly, contaminant profiles shown for sediments and macro-algae represent an aquatic "signal" of water-borne chemical in the marine system, while those profiles for biota are indicative of food web bioaccumulation processes and subsequent chemical residue distributions in organism tissues'. Additional plots for the various organochlorine compound classes are presented in Figure 3.4, including (a) PCBs, (b) chlorobenzenes, (c) HCHs, (d) DDTs, (e) cyclodienes and (f) chlordanes and are described in the following sub-sections.

Figure 3.3 shows that the ∑PCB atmospheric and aquatic "signal" is relatively small compared to other organochlorine contaminants such as chlorobenzenes and HCHs, but those relative ∑PCB burdens tend to be elevated in organisms of the food web and humans. Cl_5-Cl_8 CB congeners are the dominant CBs in marine biota and humans. Figure 3.4a illustrates the CB congener profiles for different components of the food web, showing a general trend of lower chlorinated CBs in sediments, lichens and macro-algae and increasing in chlorination with increasing trophic level. A more detailed examination of CB congener patterns was conducted by calculating CB_X/CB153 ratios (R^{153}), i.e., congener specific R^{153} values (Appendix 7). Cl_2 to Cl_8-CB homologues exhibited relatively equal contributions to total PCBs, with lower chlorinated Cl_2 and Cl_3 congeners (e.g., CB8/5, CB28, CB 31 and CB16/32) contributing substantially to ∑PCBs in lichens, sediments and macro-algae. For example, lipid equivalent concentrations of CB8/5, CB28, CB31 and 16/32 in macro-algae were approximately 1.0, 0.7, 0.9 and 1.5 $ng \cdot g^{-1}$ lipid equivalent, respectively, and were greater than those concentrations of CB153 (~0.12 $ng \cdot g^{-1}$lipid equivalent). Corresponding R^{153} values for CB8/5, CB28, CB31 and CB16/32 in macro-algae were 1.7, 1.4, 1.6 and 2.7, respectively. Similar patterns and R^{153} values were observed in lichens. The relatively high degree of accumulation of Cl_2 and Cl_3 CB congeners observed in lichens and macro-algae (i.e., R^{153} values >1) is likely due (i) the relatively higher vapor pressures and water solubilities of those congeners and consequently (ii) increased exposure of those lower chlorinated CBs through ambient air and seawater, respectively. The R^{153} values and hence CB patterns in lichens, sediments and macro-algae were quite different from R^{153} values observed in

biota: i.e., bivalves, fish, seaducks and marine mammals. Not surprisingly, recalcitrant (Group I and Group II) Cl_6-Cl_8 CB congeners demonstrated a high degree of persistence in organisms of the food web with a concentration rank order of: CB153 > CB138 > CB99 > CB180 > CB 187/182 > CB170/190.

Bivalves and fish generally showed similar CB patterns, where Cl_3 to Cl_8 -CB homologues were dominant, with CB153 having the highest concentrations. For example, levels of CBs 28, 52, 99, 138 and 153 in Arctic cod were approximately 0.81, 1.23, 3.44, 5.81 and 10.9 $ng \cdot g^{-1}$ lipid, respectively. R^{153} for CB138 in cod was approx. 0.54, while CBs 101, 99, 118 and 180 were all slightly greater than 0.2. Capelin showed a unique CB congener pattern compared to other fish species. The comparatively high R^{153} values of CB-101 (0.68), CB-99 (0.5) and CB-95 (0.46) in capelin correspond to higher tissue burdens equal to approximately 12.3, 9.0 and 8.2, $ng \cdot g^{-1}$ lipid respectively. In general, seaducks and marine mammals exhibited similar CB patterns, with $Cl_4$to Cl_8 -CB homologues being dominant. For example, concentrations of CBs 99, 138,153 180 in eider duck (liver samples) were approximately 17.5, 63.8, 103 and 24.8 $ng \cdot g^{-1}$lipid wt., respectively. Concentrations of CB99, 138,153 180 in beluga whales (blubber) were 192, 384, 518 and 104 $ng \cdot g^{-1}$ lipid, respectively. The CB pattern for ringed seals and white-winged scoters differed somewhat from other animals. In particular, white-winged scoters appear to exhibit a very different CB pattern than eider ducks, which are also a molluscivorous species and hence occupy equivalent trophic positions. In contrast to eider ducks however, the scoters exhibited relatively low R^{153} values for several Cl_3-Cl_5 congeners, including CB28 (0.01), 52 (0.01), 95 (0.01), 99 (0.14) and 101 (0.03). This distinct CB profile (with negligible accumulation of those Cl_3-Cl_5 congeners) in scoters may the result of higher cytochrome P450-1A and 2B enzyme activity in those organisms (i.e., greater metabolic transformation) and/or accumulation of CBs from an alternative food source with a distinct CB signature. Also, ringed seals exhibited relatively low R^{153} values for CB52 (0.07), 95 (0.01) and 149 (0.05) as compared with R^{153} values equal to 0.32, 0.33 and 0.42 for those congeners in beluga whales. The comparatively low values of R^{153} for CB52, 95 and 149 in ringed seals thereby suggests those animals (i.e., pinnepeds) have a greater ability to efficiently metabolize those congeners *via* CYP450 1A and 2B enzyme activity compared to belugas (i.e., cetaceans). Cetaceans as an organism class have previously been identified as having reduced CYP-1A and 2B capacity to metabolize PCB congeners, compared with other marine mammals and seabirds (*65*). R^{153} values reported here for ringed seals are comparable to R^{153} values reported in Arctic ringed seals and walrus (*2,60*). R^{153} values shown for E. Hudson Bay polar bears are consistent with previous observations of Canadian

Arctic polar bears (*2,61,75*) and highlights the fact those high trophic predators only substantially retain CB congeners 99, 153, 138, 180,170/190 and 194. In general, the observed CB congener levels and trends in E. Hudson Bay biota are consistent with other studies of Arctic marine mammalian food webs, where a general progression of higher burdens of higher chlorinated CBs moving up the food chain has been observed (*2,75,139*). Also, the above evaluation of CB congener R^{153} values in E. Hudson Bay organisms also highlights the effect of compound selective metabolism *via* CYP1A and 2B enzymes in different species on chemical bioaccumulation profiles.

Figure 3.3 shows ∑HCH and ∑Chlorobenzenes exhibit substantial atmospheric and aquatic "signals" in relation to other OCPs and PCBs. However, burdens of these compounds in organisms of the food web and humans are relatively small compared to PCBs, DDTs and the cyclodienes. Similar OC pesticides accumulation patterns have been observed in other Arctic marine food web studies (*2,6,139*). Figure 3.4b shows that the chlorobenzene composition in lichens (the atmospheric signal) is mainly comprised of HCBz, while the Tri and Tetra CBz tend to be dominant in sediments and macro-algae (the aquatic signal) and is likely related to the relatively high vapor pressure and water solubility of those compounds. α-HCH is the dominant hexachlorocyclohexane isomer representing the atmospheric and aquatic signal (Figure 3.4c). However, β-HCH is shown to accumulate extensively in various organisms of the food web and humans, indicating the high degree of recalcitrance of the β isomer compared to the apparently more labile α- and γ- isomers, which are likely biotransformed by birds and mammals. Figure 3.4d illustrates *p',p'* DDE (the primary metabolite of technical DDT) is the most dominant DDT component in the E. Hudson Bay food web and humans. For cyclodienes (Figure 3.4e), the dominant compounds are mirex, dieldrin and heptachlor epoxide. While *cis* and *trans* chlordane are relatively abundant in lichens, sediments and macro-algae, *cis* nonachlor and *trans*-nonachlor and oxychlordane (the primary metabolite of technical chlordane) is shown to be the dominant chlordane constituent in the food web and humans (Figure 3.4f). This is consistent with other studies indicating that major components of technical chlordane (*cis* and *trans* chlordane) are biotransformed to oxychlordane and that minor components (*cis* nonachlor and *trans* nonachlor) are relatively non-metabolizable and bioaccumulative in food webs (*4,5,6,139*)

3.3.5 Age and Sex related Trends and Maternal Transfer.

We observed significant age and sex related differences in POP concentrations in E. Hudson Bay beluga whales. Figure 3.5 illustrates the age relationship of (a) ∑PCBs and (b) β-HCH concentrations in E. Hudson Bay beluga whales (categorized as calves and females and males between the ages of 3 and 35). ∑PCBs concentrations in male beluga whales are shown to increase linearly, i.e., ∑PCBs =117 (MALE AGE) + 1,290, r^2 =0.436. Several recalcitrant Group I and II CB congeners (e.g., CB153, 180, 138) and pesticide components *p',p'*DDE, *trans*-nonachlor and mirex exhibited similar significant increasing concentration trends with age for male beluga whales. β-HCH (shown in figure 3.5b) exhibits a slight decreasing trend with age for male beluga whales. Hexachlorobenzene (HCBz), a prominent OCP in Arctic marine mammals, also showed no significant relationship with male or female beluga age. Numerous studies of organochlorines in marine mammals (*114,139,165,166,167*) have shown similar age and sex dependent bioaccumulation behaviour (i.e., rank order of chemical concentration are typically males > calves ≥ females). Figure 3.5b shows relatively high β-HCH levels in two beluga calves (i.e., approx. 130 and 170 $ng \cdot g^{-1}$ lipid), which were elevated about 10 to 15 times above blubber and milk concentrations of β-HCH in adult female beluga whales (see Appendix 6). The relatively high levels observed in some calves may be due to elevated contaminant exposure and/or the first birth for the mother (both of which cause higher chemical concentrations in milk) or slow growth rate and reduced fat deposition in the calf (causing an internal concentration amplification). We observed no significant differences in POP concentrations between male and female ringed seals. The effect of age and maternal transfer (i.e., chemical depuration by mother and the corresponding accumulation by calf) is complex and involves temporal changes in ambient contaminant levels and patterns (over decade life spans of these animals) and substantial physiological condition (e.g., animal growth rates) and may require the use of a life-time/generational simulation models (*114*) to fully evaluate these observed temporal trends.

3.3.6 BMF and Elimination Index estimations.

BMFs and EI estimates of PCBs and several OC pesticides in fish, seaducks and marine mammals from E. Hudson Bay are summarized in Appendix 8. Group I CB congeners such as Cl_6-CB153 and Cl_7-CB180 typically exhibited the highest BMFs in organisms of the E. Hudson Bay food web. BMFs of recalcitrant PCBs differed substantially between taxa. For example,

BMFs of CB180 in air-breathing endotherms such as eider ducks (95.9), male beluga (41.8) and male ringed seals (11.5) were approximately 10-30 times higher than those BMFs in water-ventilating ectotherms such as Arctic cod (3.5) and sculpin (3.7). BMF values for CB180 in beluga whales were approximately 42, 11 and 5 for males (aged 3-32 years), females (aged 4-35 years) and calves (< 1 year), respectively. Also, relatively large differences in BMFs of CB congeners were observed between different species of seaducks and marine mammals. For example, calculated BMFs of CB180 for white-winged scoters/mussels, walrus/mussels and polar bear/ringed seal, were approximately 635, 288 and 94, respectively. While a CB180 BMF equivalent to approximately 100 for polar bears is quite feasible and comparable to previously reported seal-to-bear BMFs (*2,139*), the relatively high BMF_{MAX} (i.e., BMF of CB180) estimated for E. Hudson Bay white-winged scoters (~600) and walrus (~300) are likely overestimated because of using incorrect prey species and hence dietary exposure concentrations. For example, a previous study by Muir and colleagues (*60*) indicated elevated organochlorine contaminant levels in E. Hudson Bay walrus sampled during the 1990's were likely the result of those animals utilizing ringed seals as a portion of their typical molluscivorous diet. If we assume walrus even consume a small quantity of ringed seals to supplement overall energy requirements (e.g., 10% ringed seal component in walrus' mixed diet), the walrus/mixed diet CB-180 BMF is approximately 25.6, which seems a more realistic BMF for these animals. White-winged scoters also may feed at a higher trophic level in the E. Hudson Bay food web (e.g., scavenging marine mammal carcasses). However, another plausible explanation for the relatively high levels and BMFs in these molluscivorous seaducks is enhanced accumulation of PCBs and other contaminants *via* bivalves from relatively more contaminated habitats during winter months. Specifically, white-winged scoters from the eastern Canadian Arctic tend to migrate south to utilize eastern United States coastal waters during the months of November to March (*168*). The overall rank order of POP BMFs in air-breathing endotherms was generally: BMF-polar bear ~ BMF eider ducks > BMF-beluga > BMF-seals.

BMFs of organochlorines in beluga whales, ringed seals and eider ducks were in some cases unreasonably high (i.e., BMF between 100-300) and exceeded BMF_{MAX} values (see Appendix 8). Potential overestimations of predator/prey BMFs (i.e., 250 - 675) have previously been indicated in Arctic seabirds such as Glaucous gulls (*Larus hyperboreus*), Black legged kittiwakes (*Rissa tridactyla*) and Northern fulmars (*Fulmaris glacialis*) and has been attributed to occasional scavenging higher trophic animal carcasses (e.g., ringed seals) and/or from higher contaminant exposure from more polluted environments along southern migration routes and over-wintering

habitats (*5,169*). The seemingly inflated BMFs for some organochlorines (i.e., BMF between 100-300) in resident belugas, ringed seals and eider ducks in the present study (see Appendix 8) may be the result of an underestimation of the organism's dietary exposure (i.e., prey concentration, C_D). For example, the BMF of CB-149 for male beluga/ cod was ~ 144, approximately 4 times greater than the BMF for CB-180, (i.e., BMF_{MAX}). However, Arctic cod (an important prey species for beluga whales) exhibit relatively low levels of CB-149 (1.4 $ng \cdot g^{-1}$), compared to other fish such as capelin (6.9 $ng \cdot g^{-1}$). A different CB congener pattern for CB149 and other CB congeners is illustrated by unique R^{153} values for capelin compared with other E. Hudson Bay fish species (see Appendix 7). Thus, a BMF for CB-149 in male belugas relative to capelin as a prey species (BMF beluga/capelin) is approximately 28.4. This somewhat more plausible CB149 BMF value for male beluga indicates that CB149 burdens in these animals may primarily be from dietary exposure *via* consumption of capelin or other prey species (e.g., krill or benthic invertebrates) that exhibit relatively high levels of CB-149. Inter-species variation of contaminant levels in prey organisms becomes very important when estimating BMFs for opportunistic feeders such as beluga whales that can feed on several species of pelagic and/or benthic invertebrates and fish (*63,170,171*). Also, BMF calculations based on Arctic cod diet (e.g., beluga/cod BMFs) were calculated using muscle tissue concentrations in cod. It should be noted that different BMF values may be observed when using chemical concentrations from other tissues (e.g., liver) or whole body concentrations of a given prey species (if there are significant contaminant burden differences between the organism's tissues'). The above examples highlight the natural variability and potential errors associated with field-surveyed predator-prey BMFs and shows the importance of understanding predator-prey relationships, inter-tissue chemical toxicokinetics, organism migration chronology and life-history.

While recalcitrant compounds such as Group I PCBs tend to exhibit efficient accumulation and very slow elimination, other chemicals may undergo enzymatic metabolism (i.e., *via* cytochrome P-450 isozymes), urinary excretion and respiratory elimination, which can act alone or together to reduce an organism's contaminant burden and ultimately lower the chemical's BMFs. Near zero EI values suggest highly persistent compounds (i.e., comparable to CB-180), while elevated EI values (> 1) suggest the presence of metabolic and/or other elimination processes. Appendix 9 illustrates EI value for several Group I-V PCB congeners. Some Group III and IV PCB congeners exhibited relatively high elimination index (EI) values in the various organisms of the E. Hudson Bay food web, indicating induction of both CYPIA and CYP2B isozymes. Low MW congeners such as CB-6 within Group III and CB-4/10 within Group IV exhibited the highest

degree of metabolism, with EI values ranging between 1.5 and 2.2. While high EI values for these less chlorinated and hence less hydrophobic CBs (i.e., log K_{OW}'s ~ 5 for Cl_2-CBs) in gill breathing water-ventilating ectotherms (e.g., cod, sculpin) may be the result of respiratory elimination to water *via* equilibrium partitioning, the EI values > 1 in air-breathing endotherms such as seaducks and marine mammals indicates efficient internal biotransformation of these compounds because substantial respiratory elimination *via* air is likely negligible due high K_{OA}'s of Cl_2 to Cl_{10} PCBs (i.e., log K_{OA}'s > 7 for Cl_2-Cl_{10} CBs). Metabolic transformation rates (k_M, d^{-1}) can vary substantially between species, gender and age-class, and contributes significantly to chemical BMFs and observed contaminant profiles in marine food webs (*2,65,89,167*). For example, male ringed seals appear to readily metabolize Cl_4CB45 (EI = 1.9 and BMF = 0.13), while male beluga whales exhibit relatively limited capacity to transform this compound (EI = 0.1 and a BMF = 27.2). The EI and BMF data suggest the rank order for CB metabolism capacity in Arctic marine biota is ringed seals > seaducks > beluga > cod.

While originally described as a metabolic index (MI) to evaluate biotransformation of PCB congeners, the elimination index (EI) can also be used to evaluate the relative persistence or biodilution of OC pesticides or any other organic compound class of interest. Negligible biotransformation was observed for the semi-volatile hydrophobic pesticides such as the cyclodienes (e.g., chlordanes, mirex and dieldrin) in seaducks and marine mammals (i.e., EIs < 0.5, with BMFs ranging between ~5 and 100). Moderate EIs and BMFs of β-endosulfan were observed in beluga whales and ringed seals (BMFs between 5 to 10), indicating some metabolism. This is supported by the detection of endosulfan sulfate in beluga and ringed seal tissues (see Appendix 6). Relatively high EI values of α and γ-HCHs observed in seaducks and marine mammals indicate very efficient biotransformation of those isomers. In contrast, β-HCH exhibited EI values near zero in seaducks and marine mammals, with relatively high BMFs in beluga whales (~50) and ringed seals (~20), indicating high resistance to metabolic transformation. The highest EI values and hence lowest BMFs were exhibited by the moderately polar/ volatile compounds such as Cl_3 and Cl_4 chlorobenzenes. Relatively low BMFs of Cl_3 to Cl_4 CBz in E. Hudson Bay seaducks and marine mammals (BMFs ~ 0 to 5) indicates these relatively polar/volatile chemicals are efficiently eliminated *via* metabolism, respiration and/or urinary excretion.

3.3.7 Compound and Species-specific Biotransformation Capacity.

Because the majority of POPs are relatively non-polar (K_{OW}'s > 5) and non-volatile (K_{OA}'s > 7) compounds, metabolic transformation tends to be the rate determining process for determining their biomagnification potential in organisms. Biotransformation of xenobiotic organic contaminants is a bio-reactive process including phase I reactions (e.g., oxidation, reductive or hydrolytic) and subsequent phase II reactions (glutathione or sulfate conjugation). However, physical-chemical properties also play a role in the determination of metabolic transformation rate constants. Specifically, previous studies have demonstrated signficant trends in metabolic transformation efficiency with the number of chlorine atoms and chemical K_{OW} (*73,74*), which are both strongly correlated with molecular weight. Appendix 10 shows the strong positive relationships between molecular weight (for PCBs and OCPs) and their chemical K_{OW} and K_{OA}, respectively. Figure 3.6 illustrates EI values and hence the biotransformation efficiency for Group I-V PCBs in (a) beluga whales, (b) ringed seals and (c) eider ducks. In general, low MW PCBs with 3-4 increase with decreasing molecular weight. These EI data appear to indicate an increasing trend with decreasing MW only for eider ducks. It is difficult to observe a clear EI-MW relationship for beluga whales and ringed seals, mainly due to large variability in their EI values. For air-breathing animals such as eider ducks, beluga whales and ringed seals, molecular weight, chemical K_{OW} and molecular structure of a compound undoubtedly influence metabolic transformation rates of hydrophobic non-volatile contaminants (e.g., PCBs). Species related differences in CYP1A and CYP2B type biotransformation efficiency is a crucial determinant of bioaccumulation potential and resulting accumulation patterns of hydrophobic non-volatile compounds in the food web. Figure 3.7 shows elimination index (EI) values for 2,3',4,4' CB66 (CYP1A) and 2,2,'5,5' CB52 (CYP2B) reported in various organisms (including E. Hudson Bay seaducks and marine mammals) and illustrates the relatively high degree of interspecies variation of CYP1A and CYP2B activity among species. The relatively high EI values for CB66 and CB52 in E. Hudson Bay polar bears (this study) are comparable to those reported in sea otters, dogs and humans (*74*) and indicates the relatively high CYP1A and CYP2B activities of those animals. E. Hudson Bay eider ducks and white-winged scoters exhibited similar biotransformation capacity as other seabirds (black tailed gull and tufted puffin), while E. Hudson Bay belugas and ringed seals exhibit a lower metabolic capacity compared to other marine mammals (e.g., Largha seals and Dall's porpoise), (*74*). Figure 3.7 also reveals that marine mammals generally exhibit a higher capacity for CYP1A transformation compared to CYP2B type transformation, while bird species tend to exhibit the reverse (i.e., higher CYP2B activity).

A high CYP2B induction in E. Hudson Bay seaducks is supported by our observations of very low BMFs for Group IV and V PCB congeners such as in those animals (see Appendix 8).

3.3.8 Biomagnification and dietary exposure of POPs in the Aboriginal Inuit population.

Using reported human breast milk POP concentrations in northern Quebec Inuit women between 1996 and 2001 (*156*), we estimated a BMF of CB180 in humans (Inuit women/Traditional Diet) of 4.6 (see Appendix 8), which is relatively low compared to CB180 BMFs observed in seaducks and marine mammals (BMFs between 20 and 100) and is substantially lower than previously reported BMFs of approximately 50 - 100 for recalcitrant PCBs (e.g., CB180) in other human populations (*95,172*). An Inuit women/mixed fish BMF for CB180 was approximately 19.0, which is comparable to previously reported CB180 BMFs in Quebec Inuit women during the late 1980s in reference *155*, (i.e., Inuit women/Arctic char BMF = 32). While the aboriginal Inuit population in the Canadian Arctic has been shown to greatly utilize traditional/country foods (fish, caribou, seals, beluga whales and eider ducks), on average about 15 to 40% of daily energy requirements (*150*), a significant portion (> 50%) of today's Inuit diet consists of market foods (e.g., beef, pork, poultry) from the agricultural food chain. Thus, our BMF estimates for Inuit (relative only to traditional/country foods) may substantially underestimated because organochlorine concentrations in agricultural food chains tend to be much lower than in marine biota (i.e., fish, seaducks and marine mammals).

Figure 3.8 illustrates chemical concentrations ($ng \cdot g^{-1}$ lipid equivalent) of the various organisms of the E. Hudson Bay marine food web versus organism trophic level (TL) for (a) $\sum$PCBs, (b) $\sum$DDTs, (c) $\sum$Chlordanes and (e) $\sum$HCHs. Previously reported POPs concentrations in Canadian Arctic biota, such as polar bear, walrus, caribou, wolves and Inuit women breast milk are added for comparison. It should be noted that some of these previous concentration data were determined 10-15 years prior to the current study. For example, chemical concentrations in barren-ground caribou and wolves (*62,150*) were determined during 1995-1996 sampling of the eastern Canadian Arctic terrestrial food chain (i.e., lichen→ caribou→-wolf trophic transfers). However, temporal trend studies of POPs in Canadian Arctic biota (especially high trophic animals) have not observed significant concentration changes over the past 15 years (*68*). In general, the data in figure 3.8 illustrates the overall food web magnification potential of selected POPs along a traditional Inuit food chain from the Canadian Arctic. PCBs and OC pesticide concentrations increased significantly ($p < 0.05$) with increasing trophic level (i.e., biomagnify),

with the exception of ∑DDTs and ∑HCHs in the lichen-caribou-wolf food chain. The amplification of POP concentrations in the E. Hudson Bay marine food web was greatest for ∑PCBs, ranging from approximately 6 $ng \cdot g^{-1}$ lipid equivalent in macro-algae to 10,200 $ng \cdot g^{-1}$ lipid in adult male polar bears. Conversely, the least food web magnification was demonstrated by ∑HCHs, ranging from approximately 30 $ng \cdot g^{-1}$ lipid equivalent in macro-algae to approx. 430 $ng \cdot g^{-1}$ lipid in adult male polar bears. Also, from our pattern analysis (see Figure 3.4 a-f), concentrations of ∑PCBs, ∑DDTs, ∑Chlordanes and ∑HCHs in higher trophic animals and humans are dominated by Cl_6-Cl_7 CB congeners, *p',p'* DDE, oxychlordane/*trans*-nonachlor and β-HCH, respectively. The data shown in figure 3.8 reveal: (i) organochlorine levels are generally 10-100 higher in marine biota compared to terrestrial species, (ii) POP concentrations in humans (i.e. Inuit women) that are known to consume significant amounts of fish and wildlife for subsistence are generally consistent with estimates based on trophic position within this food web (i.e., concentrations are ordered polar bears > humans > traditional foods) and (iii) the overall extent of food web magnification is greatest in the marine food web (due, in part, to relatively more trophic transfer sequences). Thus, human dietary exposure of POPs *via* traditional/country foods is lowest for consumption of fish and caribou and highest for consumption of marine mammals (ringed seals, beluga, walrus).

Another significant observation of the data in figure 3.8 is the two to five times decrease in Inuit breast milk POP concentrations between 1989 and 2001. The decline in chemical concentrations in northern Quebec Inuit during this period is likely the result of changes in dietary habits towards non-traditional market food items as POP concentrations in the Arctic environment and biota have not been declined signficantly between 1989 and 2001 (*69,150,173,174,175*). The declining trends of POP levels in Inuit women (e.g., breast milk, cord blood) undoubtedly will result in reduced prenatal exposure (placental transfer) and post-partum (*via* nursing) maternal transfer of potentially carcinogenic, teratogenic, neuro and immmunotoxic chemicals to susceptible infants. Although organochlorine levels in Inuit women are somewhat elevated above levels observed in the general Canadian female population, previous benefit-risk characterization and assessments have advised Inuit populations to continue current consumption levels of traditional foods and breast-feeding newborns because of the significant nutritional benefits (*150*).

In summary, the determinants of chemical concentrations and accumulation patterns in fish, seabirds and mammals in Arctic marine food webs involve a complexity of temporal-spatial, biological and physical-chemical factors, organism migration chronology/life history, predatory-

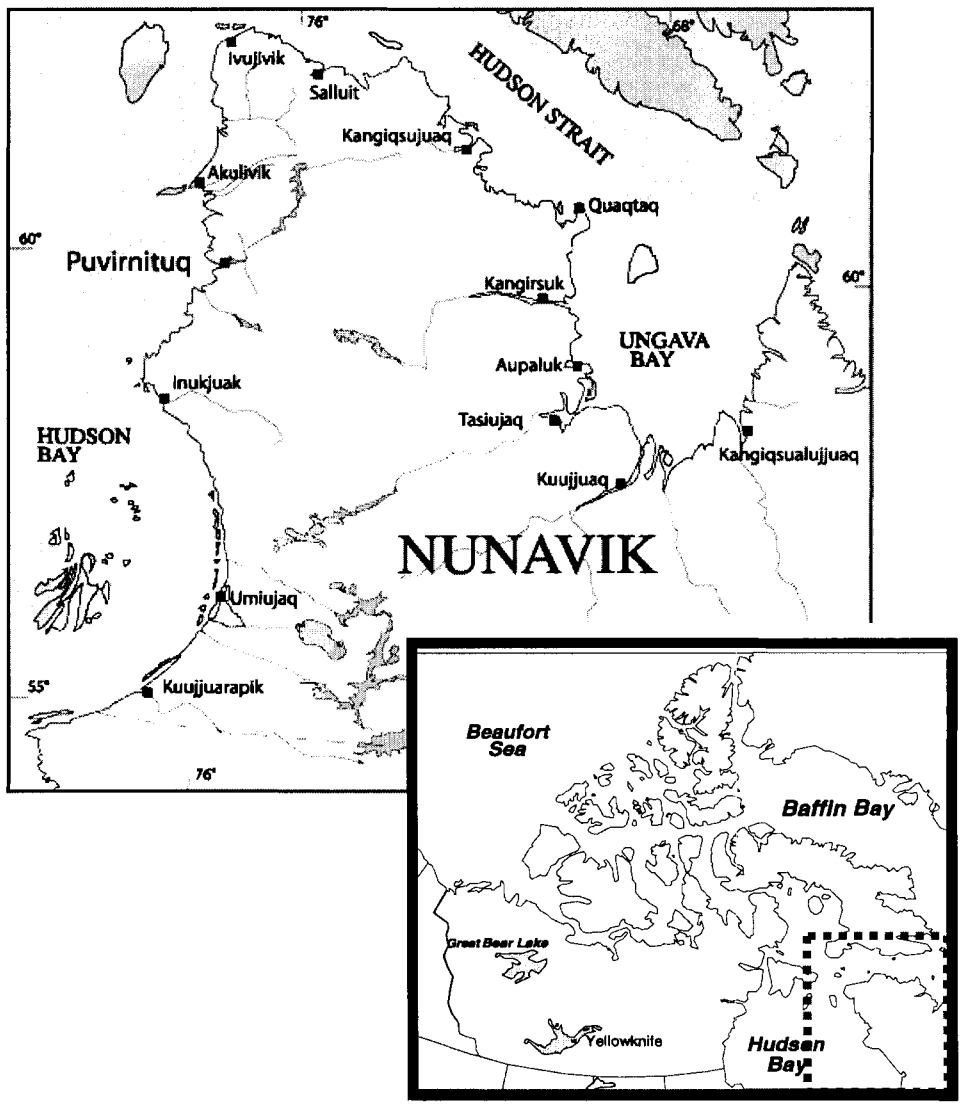
prey relationships, organism physiology and metabolic capacity. Simultaneous processes involving such factors ultimately determine contaminant levels and patterns in various organisms of this Arctic food web, which subsequently delineates the extent of human dietary exposure. The highly dynamic and variable nature of chemical fate and bioaccumulation processes in marine food webs renders it difficult to discern observed contaminant accumulation patterns, especially for new chemicals with which environmental exposure data is minimal or non-existent. Evaluation of biomagnification factors (BMFs) and elimination indexes (EIs), relative to well-established recalcitrant POPs (e.g., Group I PCBs, PCB153 and 180 = BMF_{MAX}) is a useful approach for assessing the chemical fate and bioaccumulation behaviour of novel environmental contaminants. For example, our observations of POPs biomagnification in this Arctic food web may be used as a marker of bioaccumulation and persistence for which to assess parallel accumulation of current-use high production volume (HPV) chemicals of concern such as dialkyl phthalate esters (DPEs), polybrominated diphenyl ethers (PBDEs) and perfluorinated acids (PFAs) such pefluorooctanoate (PFOA, $C_8F_{15}COO^-$) and perfluorooctane sulfonate (PFOS, $C_8F_{17}SO_3^-$). The aim of such studies should be to investigate important physical-chemical and biological determinants of chemical biomagnification. This may further benefit our predictive capability regarding the trophic transfer and persistence of organic contaminants in ecological food chains.

3.4 Acknowledgements.

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies and wish to thank Dr's Bill Doidge, Michael Kwan and Susan Sang, Derek Muir and Northern Quebec Inuit communities of Umiujaq and Inukjuaq for coordinating/aiding collection of field samples.

3.5 Figures

Figure 3.1 Map showing general study area of E. Hudson Bay and various Nunavik Inuit communities of northern Quebec, Canada.



Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

Figure 3.2 Schematic illustration of organisms (including humans) comprising the Canadian Arctic marine and terrestrial food webs and associated trophic level (TL) and feeding interactions.

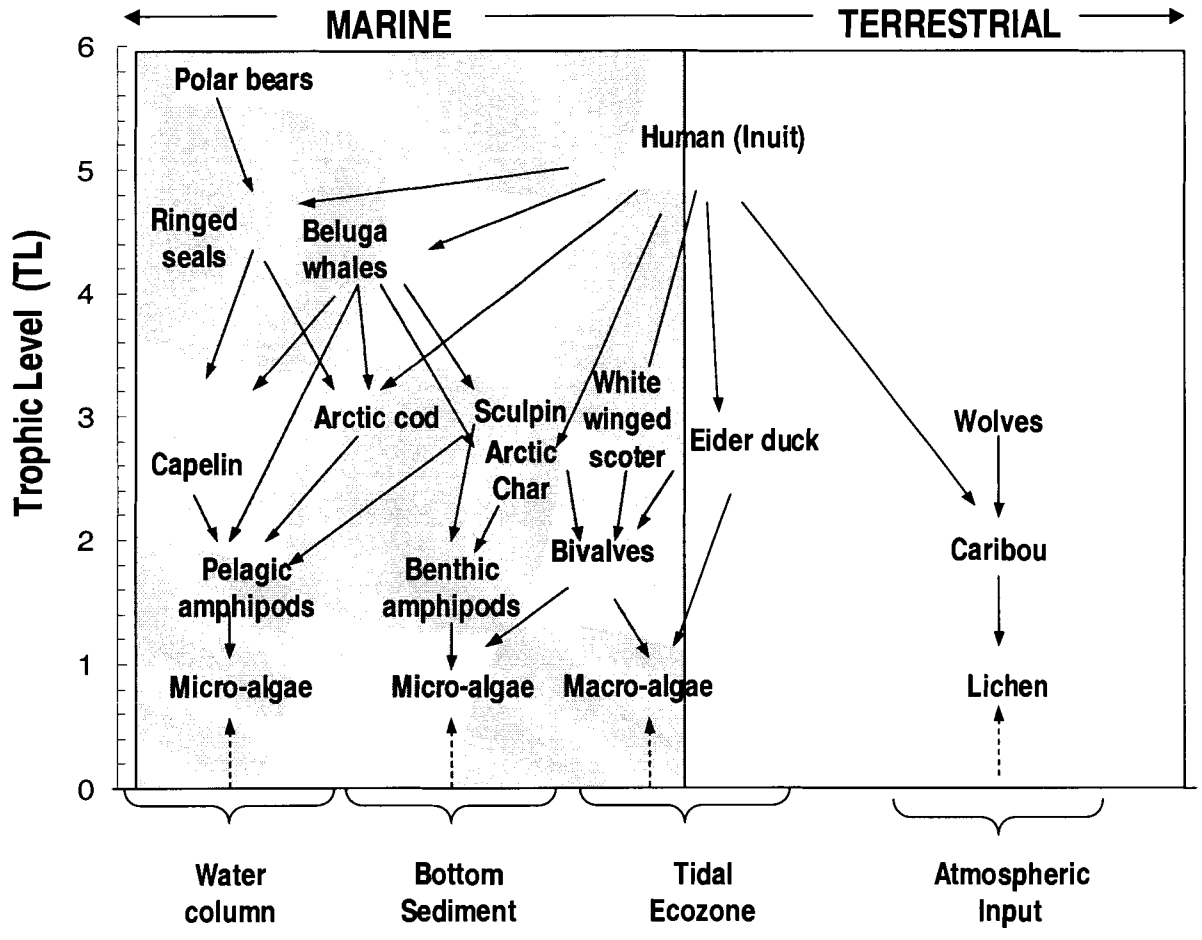


Figure 3.3 % composition of organochlorines (pesticides + PCBs) in environmental and biological samples from E. Hudson Bay.

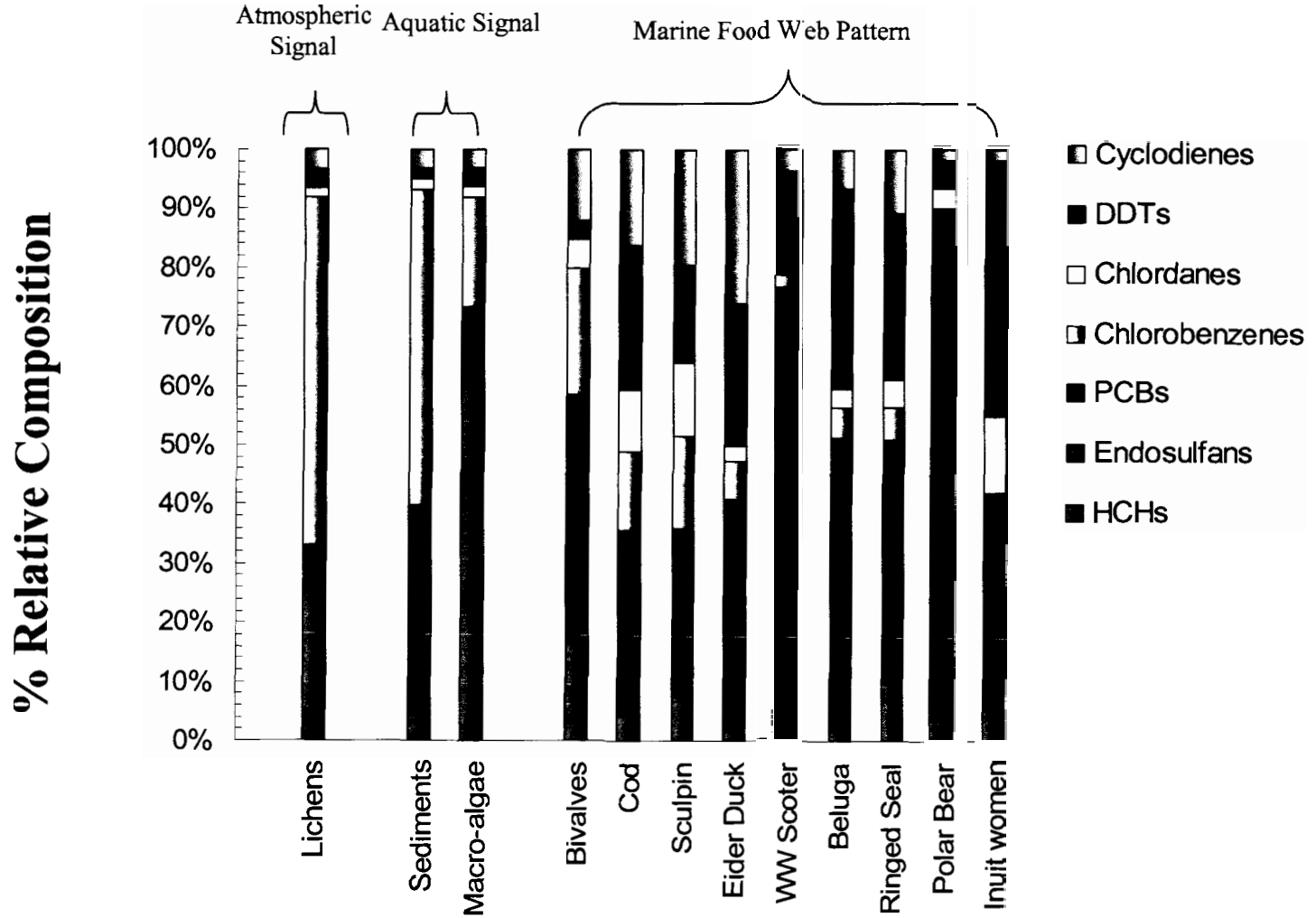


Figure 3.4 % Composition plots for (a) PCBs, (b) Chlorobenzenes, (c) HCHs, (d) DDTs, (e) Cyclodienes and (f) Chlordanes in environmental and biological samples from E. Hudson Bay.

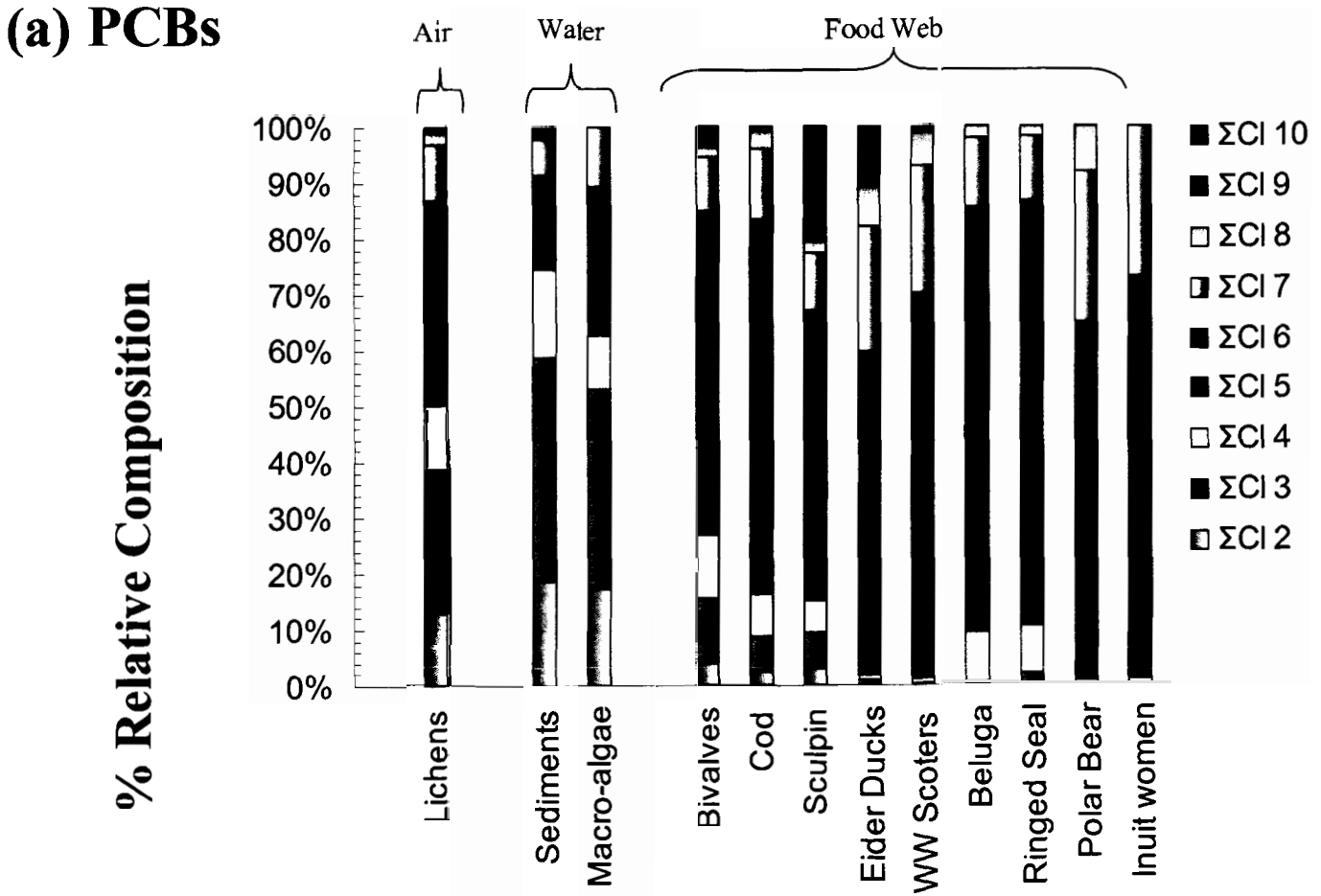


Figure 3.4 continued.

(b) Chlorobenzenes

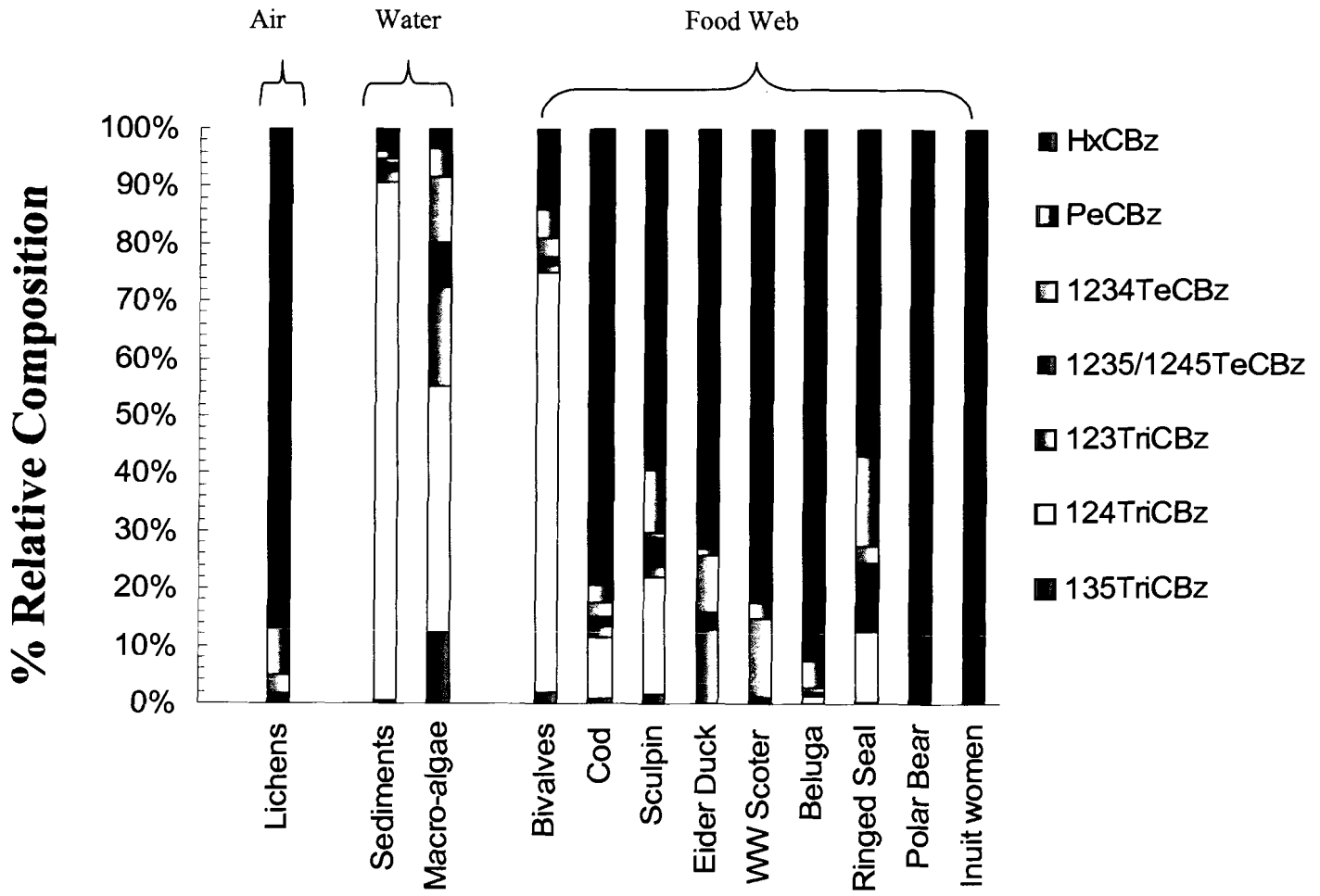


Figure 3.4 continued.

(c) HCHs

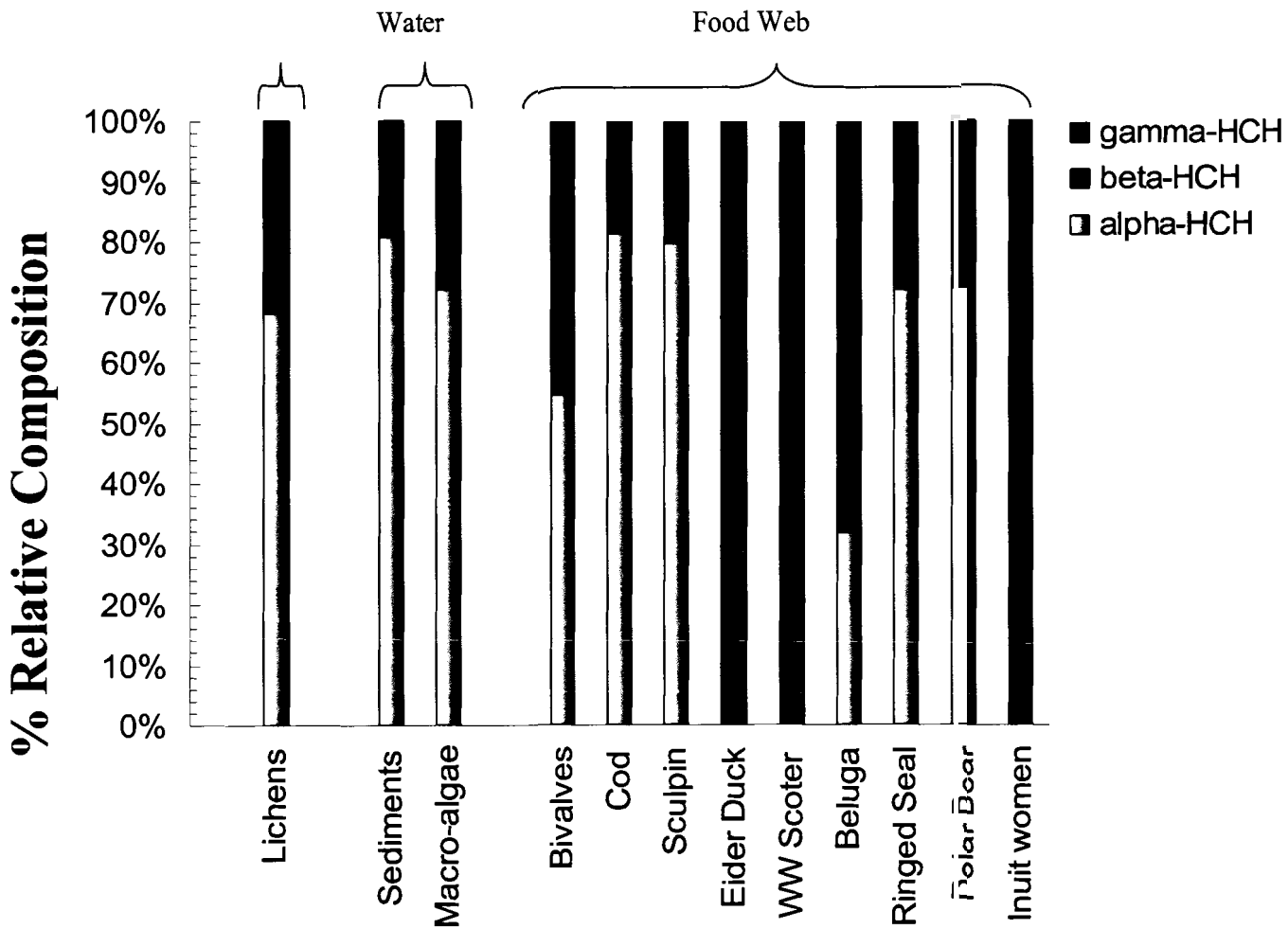
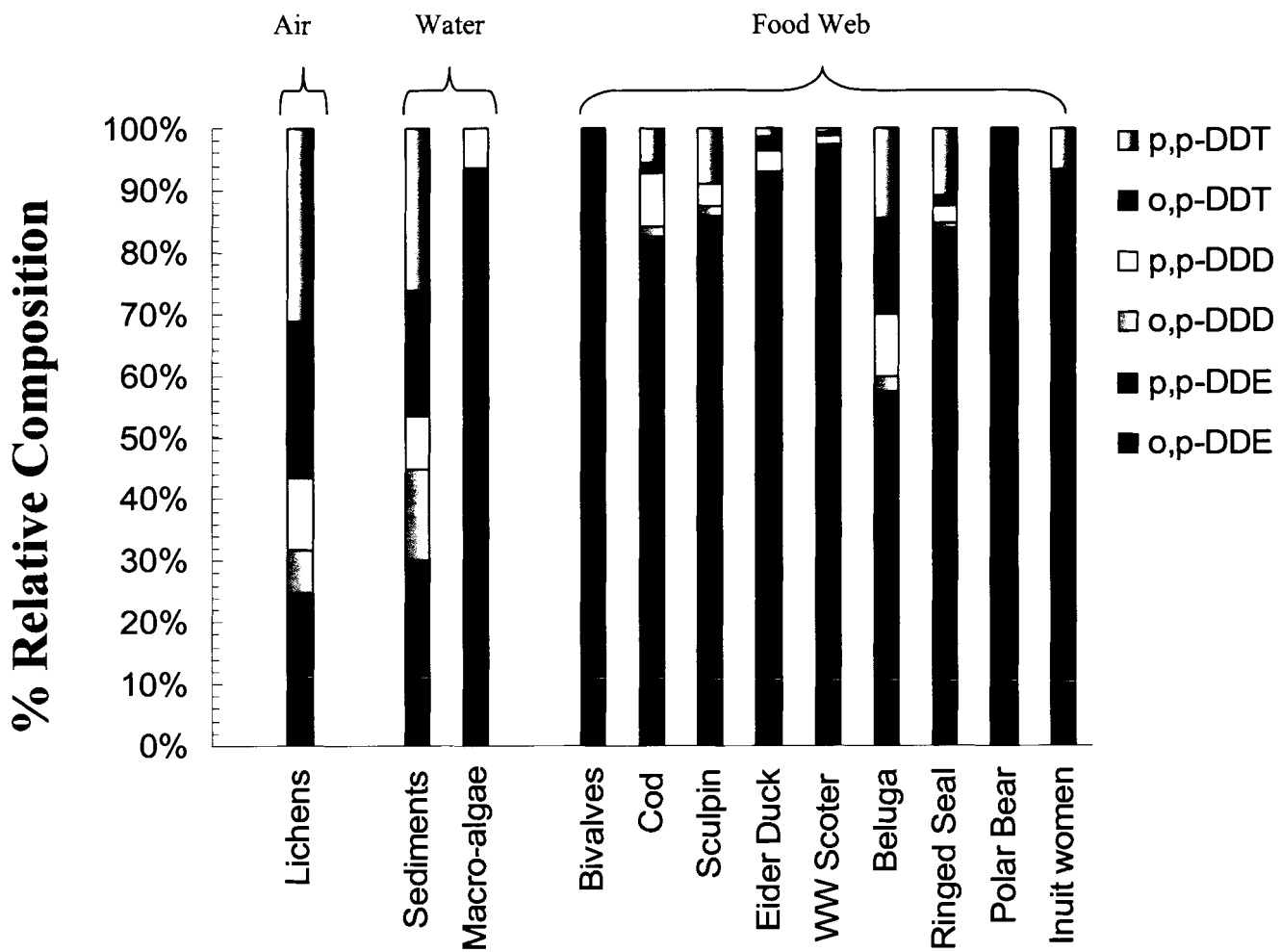


Figure 3.4 continued.

(d) DDTs



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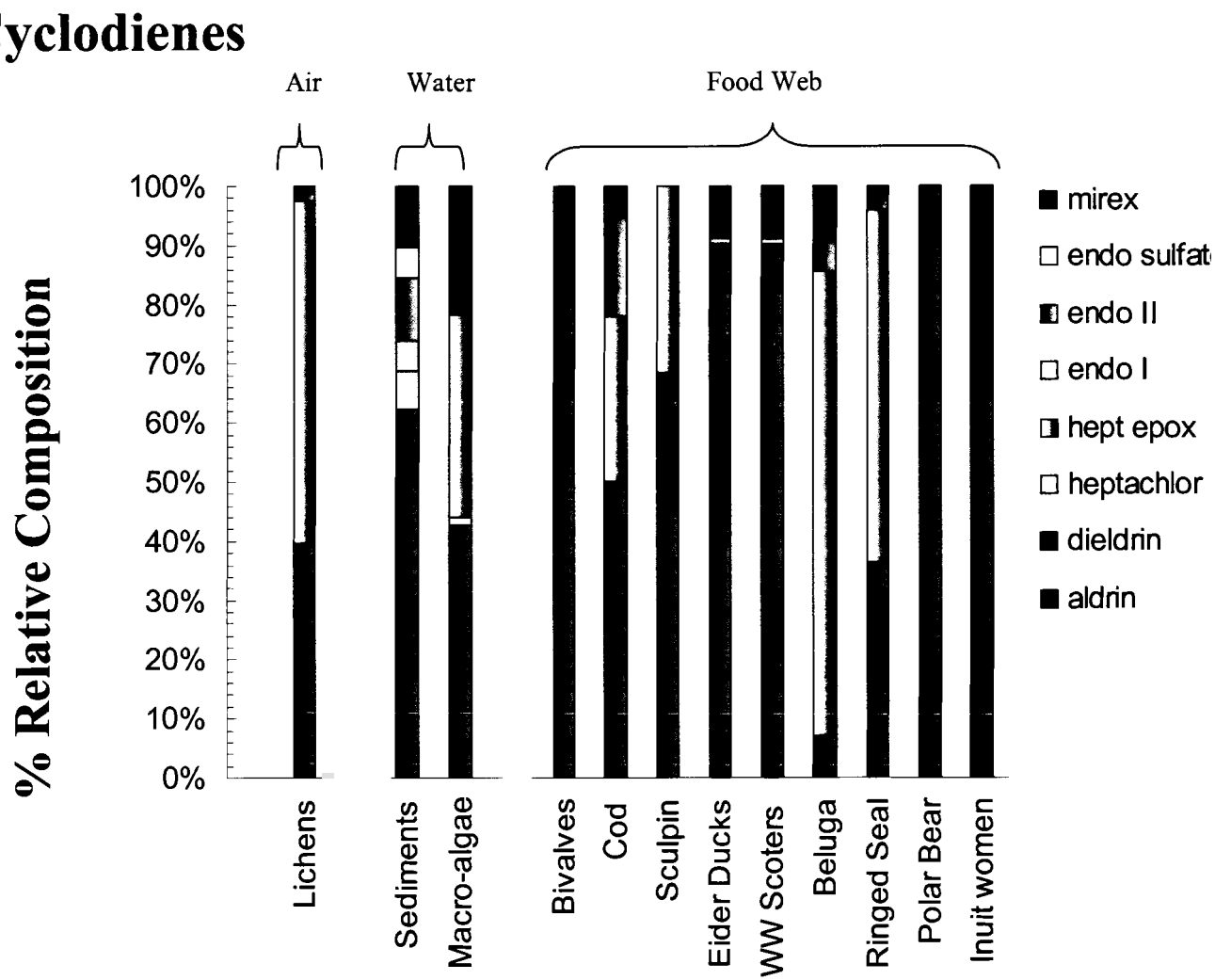


Figure 3.4 continued.

(f) Chlordanes

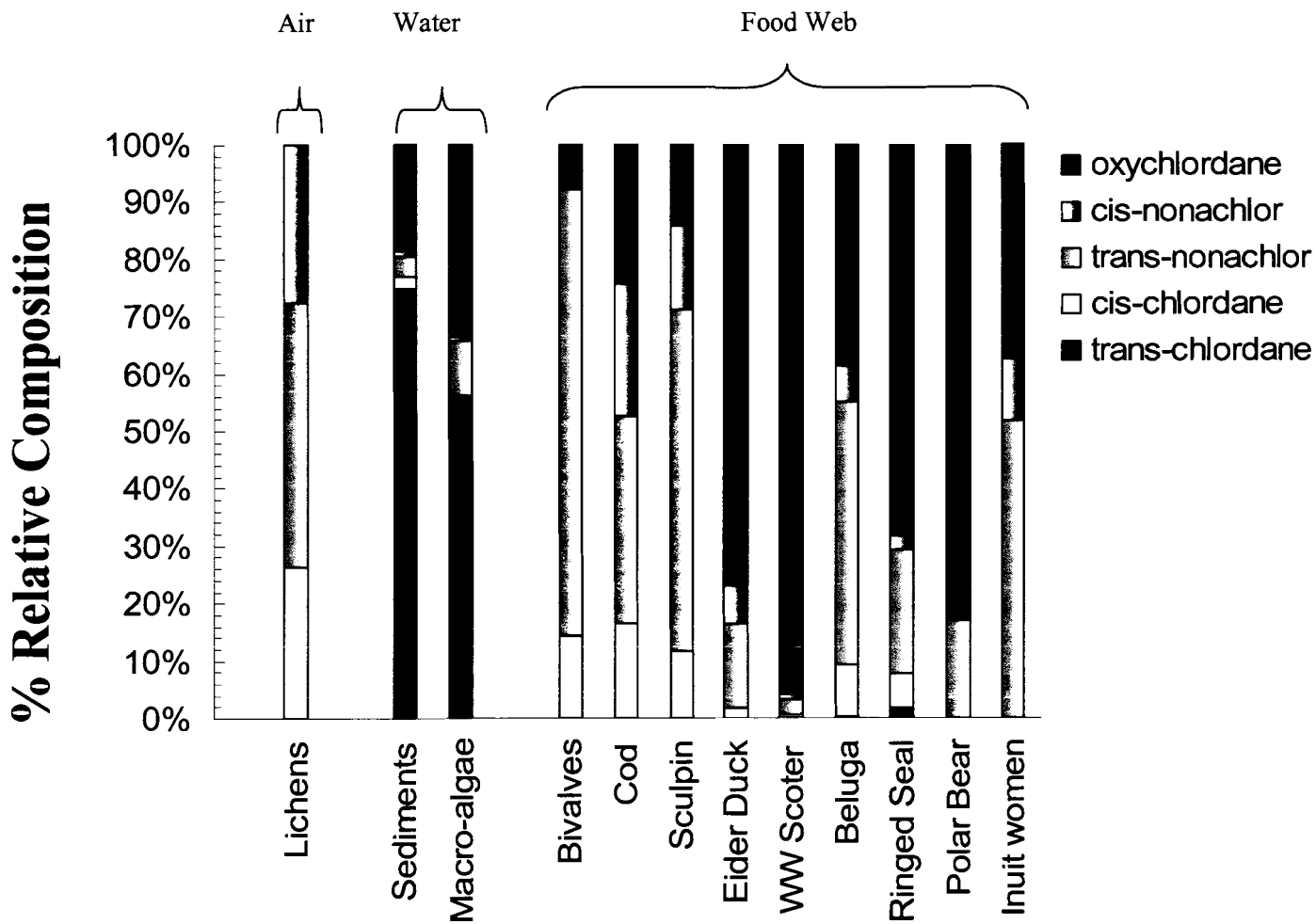


Figure 3.5 Relationship between animal age (years) and chemical concentration (ng·g^{-1} lipid) in beluga whales for (a) ∑PCBs and (b) β -HCH. Data for males are shown as dark circles, females as white circles and calves as gray circles.

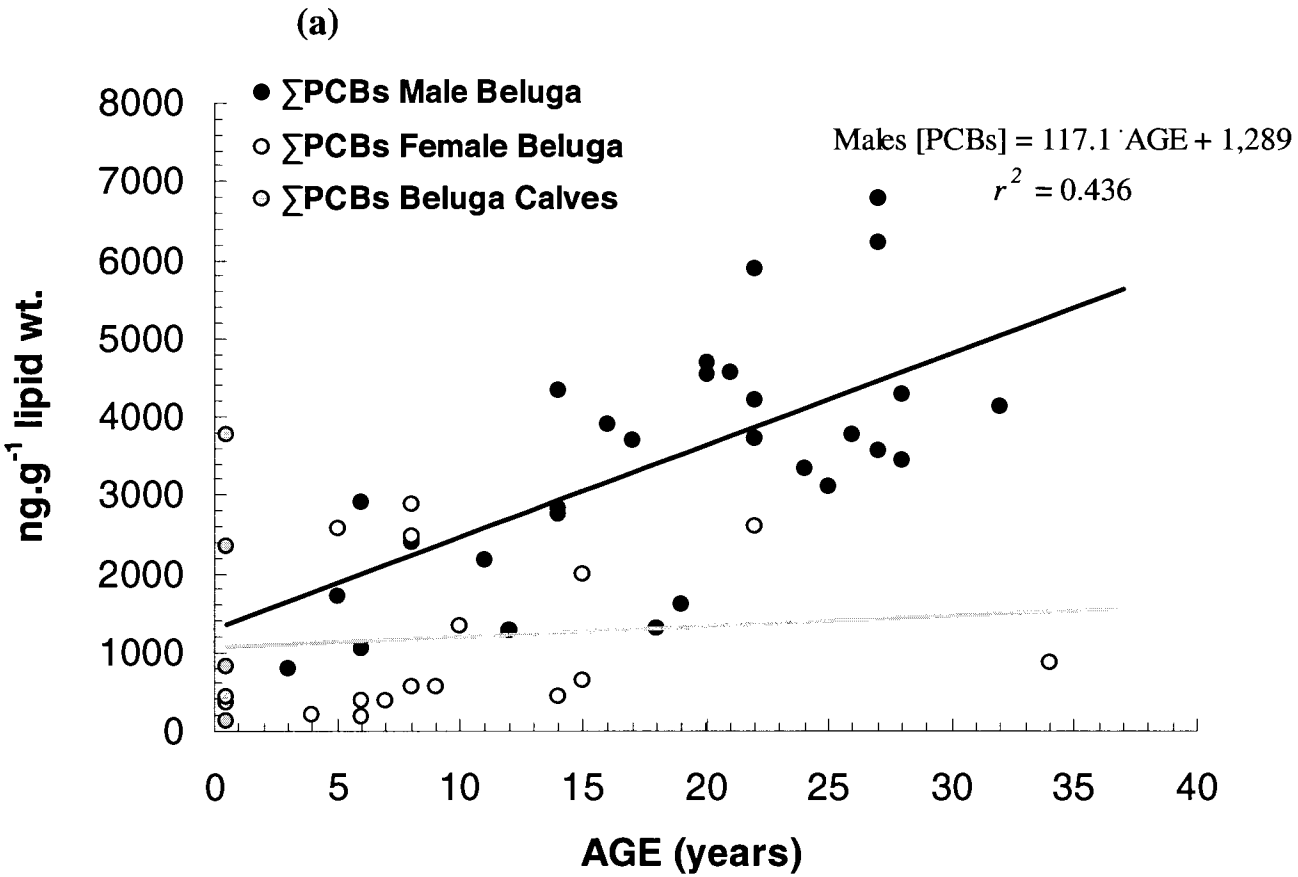


Figure 3.5 continued.

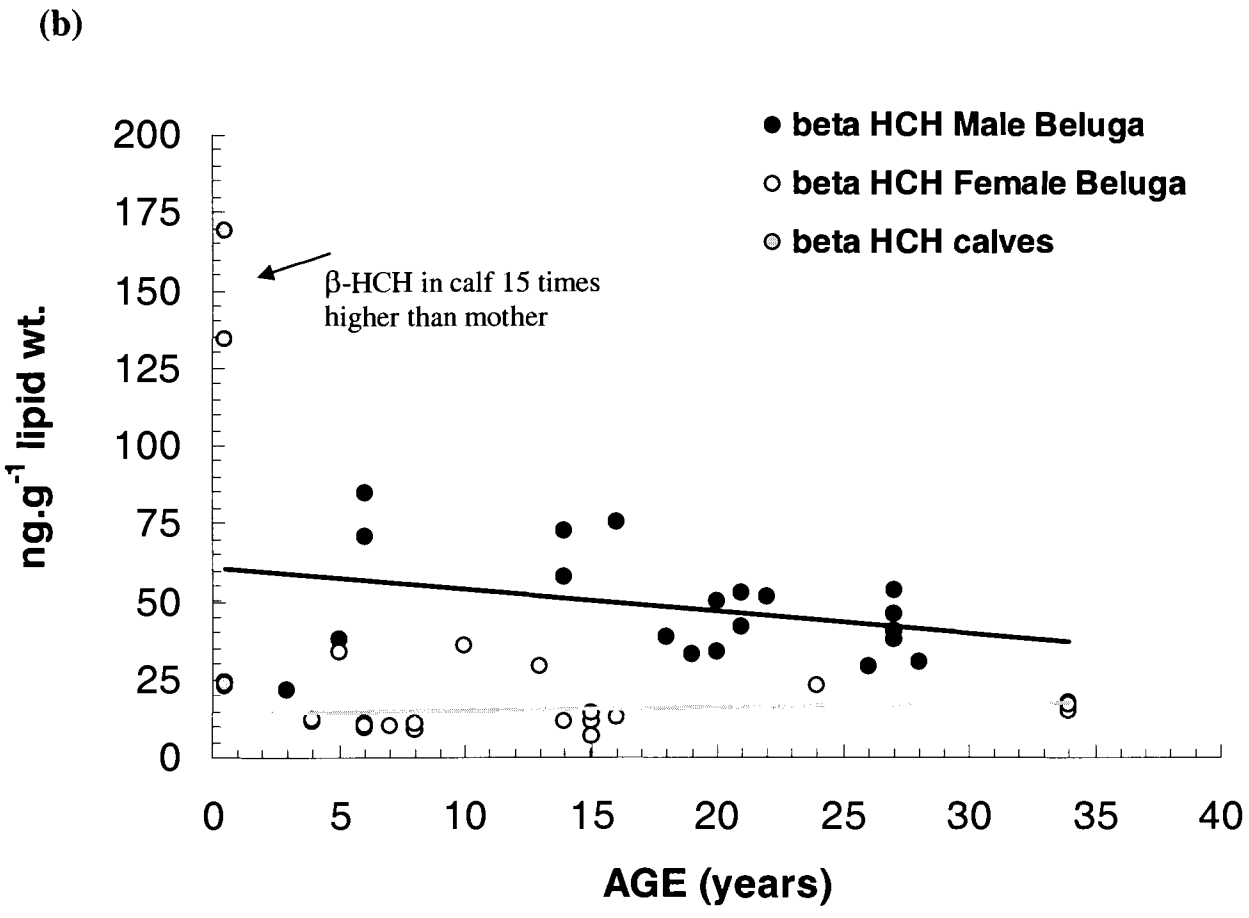
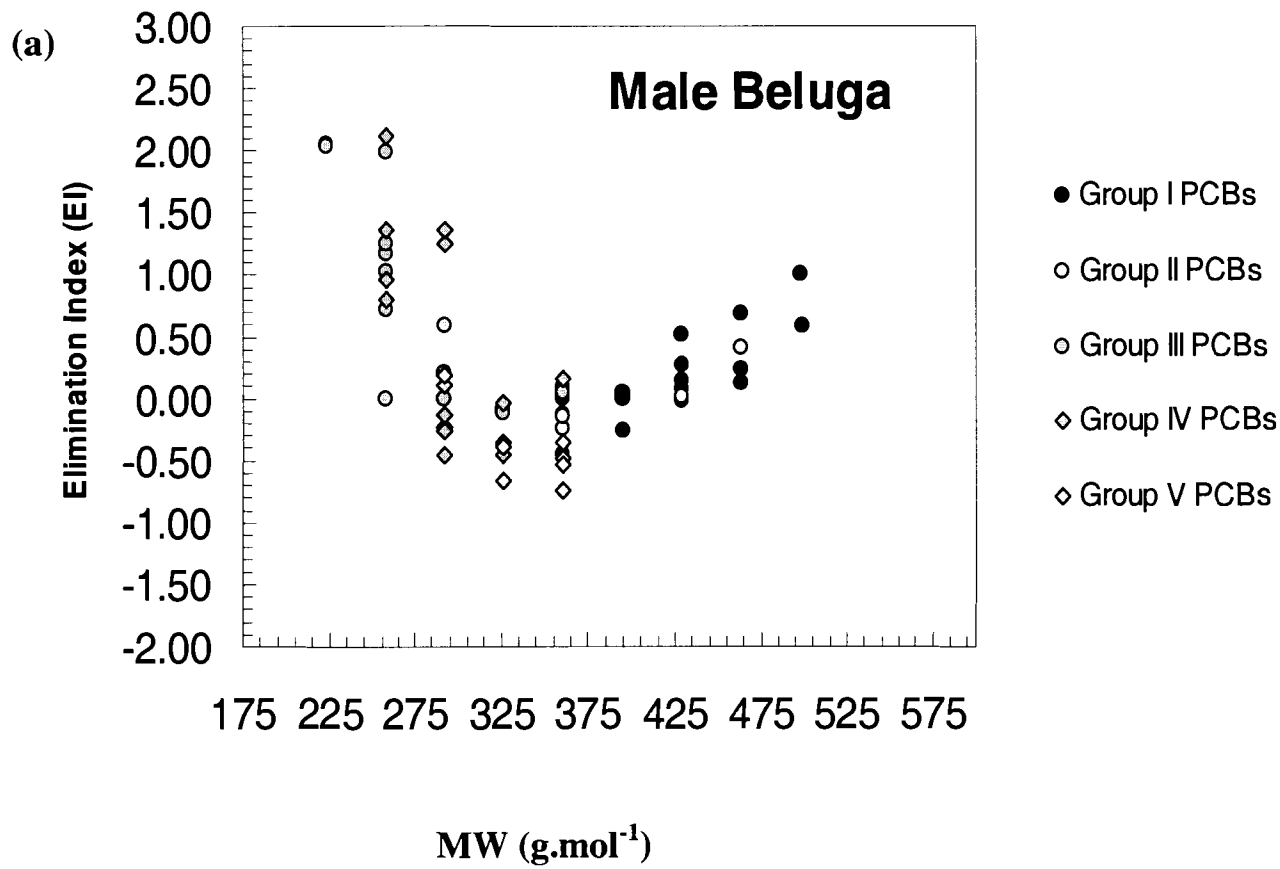


Figure 3.6 Relationship between chemical elimination index (EI) and molecular weight (MW) for Group I – V PCB congeners in (a) male beluga whales, (b) male ringed seals and (c) eider ducks.



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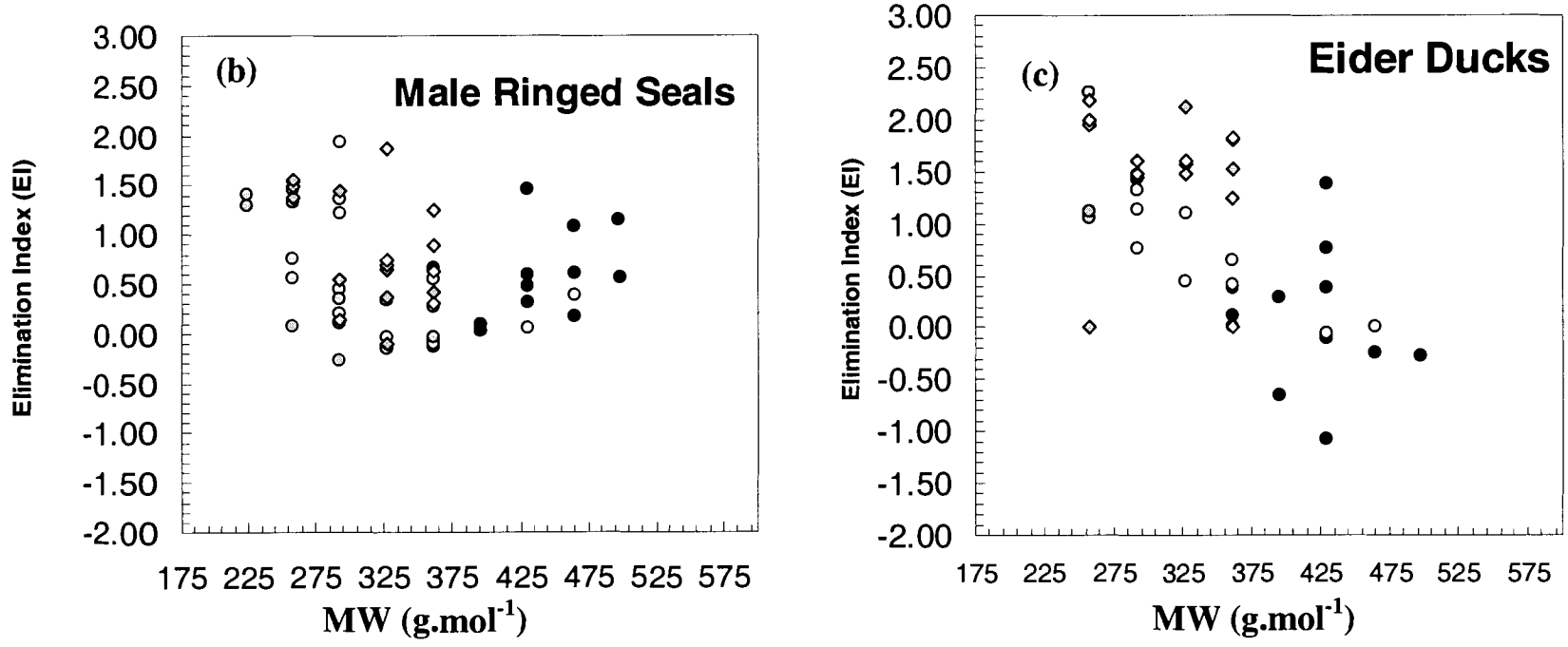


Figure 3.7 Elimination Index (EI) values for 2,3´,4,4´ CB66 (Group III congener representing CYP1A-type biotransformation) and 2,2´,5,5´ CB52 (Group IV congener representing CYP2B-type biotransformation) in various organisms of the E. Hudson Bay marine food web in comparison to EI values previously reported in other organisms. Data for ringed seals, beluga whales, white winged scoters and eider ducks represent EI values in E. Hudson Bay animals (this study), while data shown for other animals are from Kannan et al. (*74*).

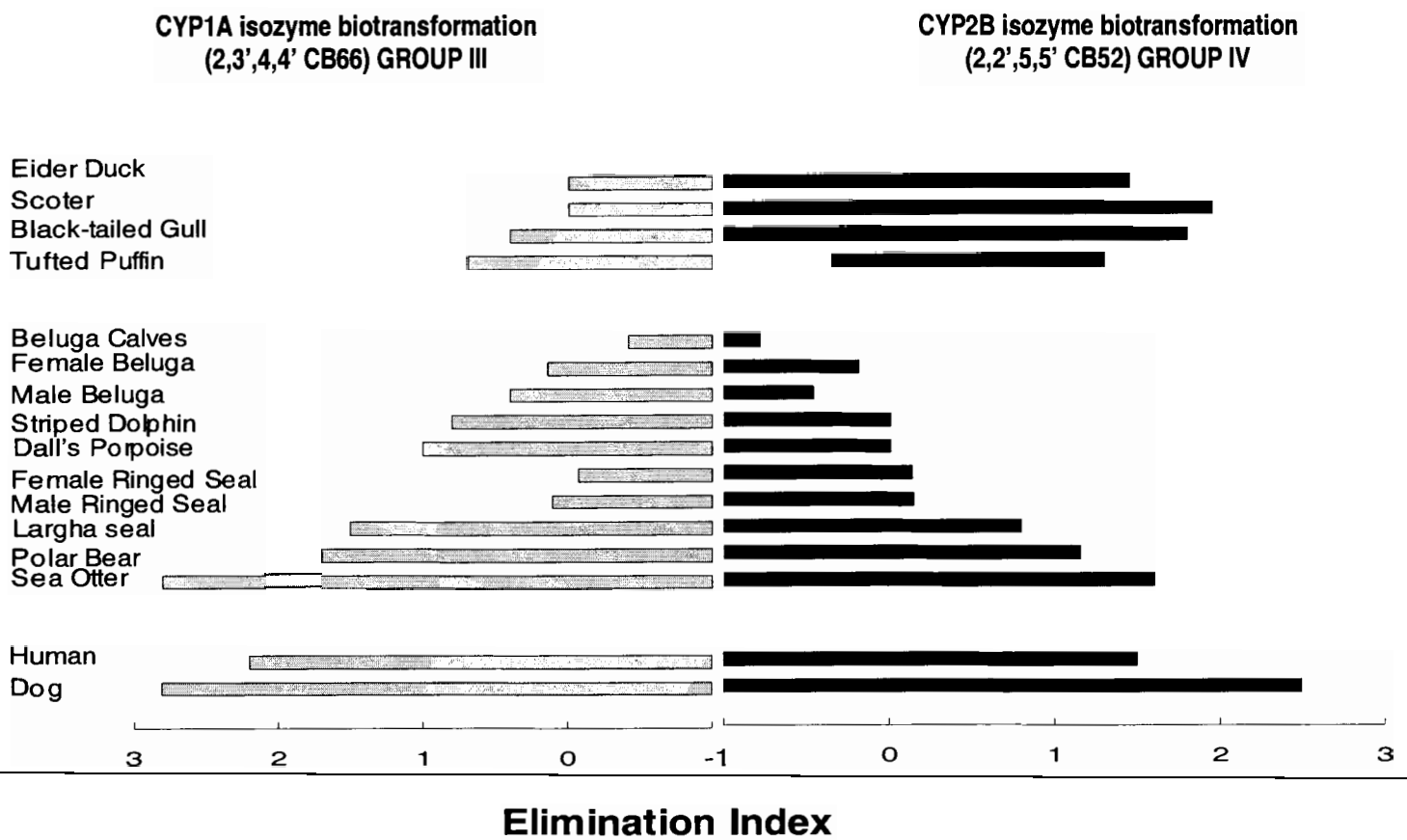
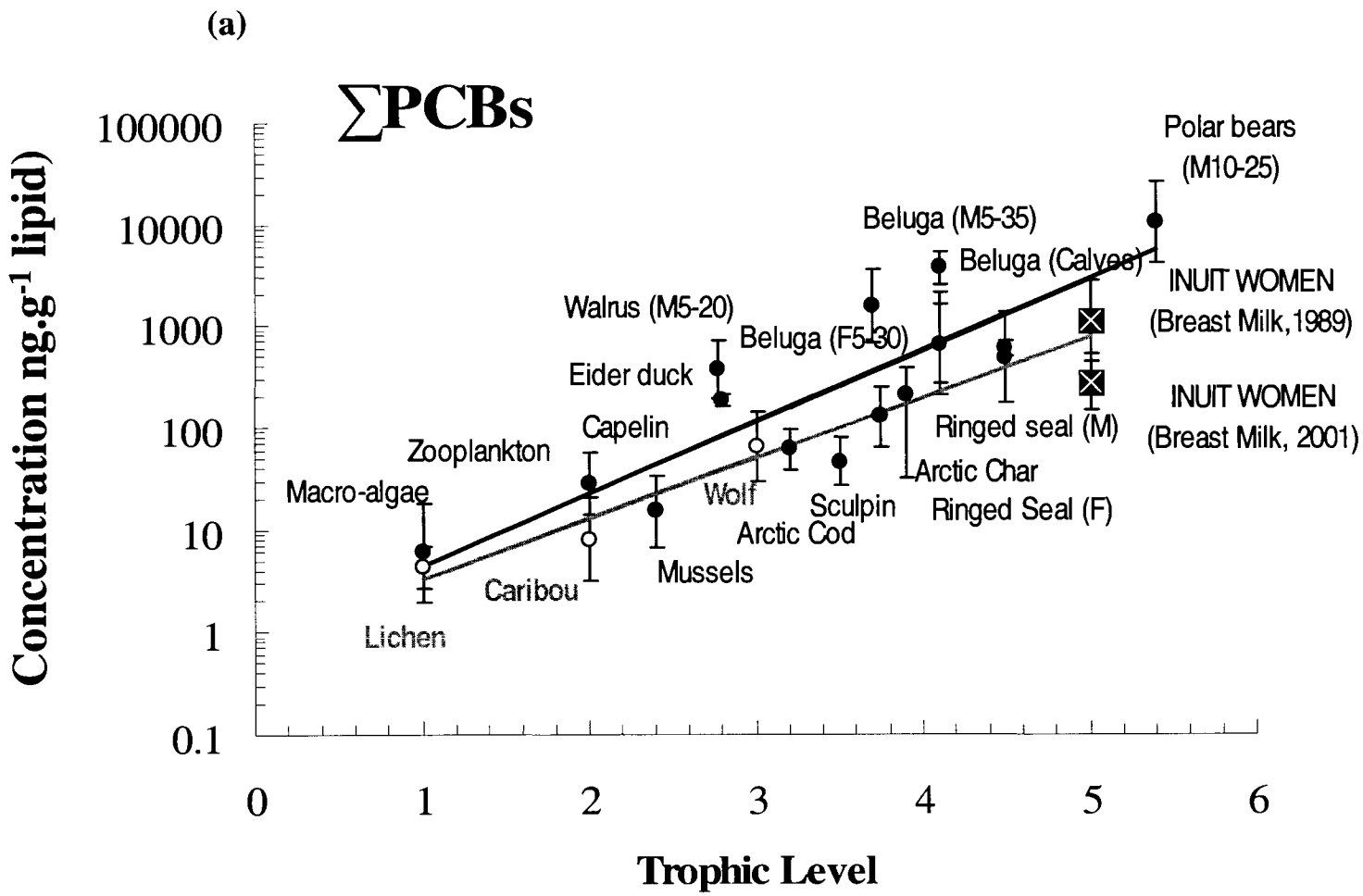


Figure 3.8 Chemical concentrations in (i) organisms of the E. Hudson Bay marine food web (ng·g^{-1} lipid), (ii) organisms of E. Canadian Arcticterrestrial food chain (lichen-caribou-wolves) and breast milk from northern Quebec Inuit women 1989 and 2001 data) all plotted against trophic level (TL) for (a) PCBs, (b) DDTs, (c) Chlordane and (d) HCHs. Black line represents data for marine food web, thin and gray line represents data for terrestrial food chain.



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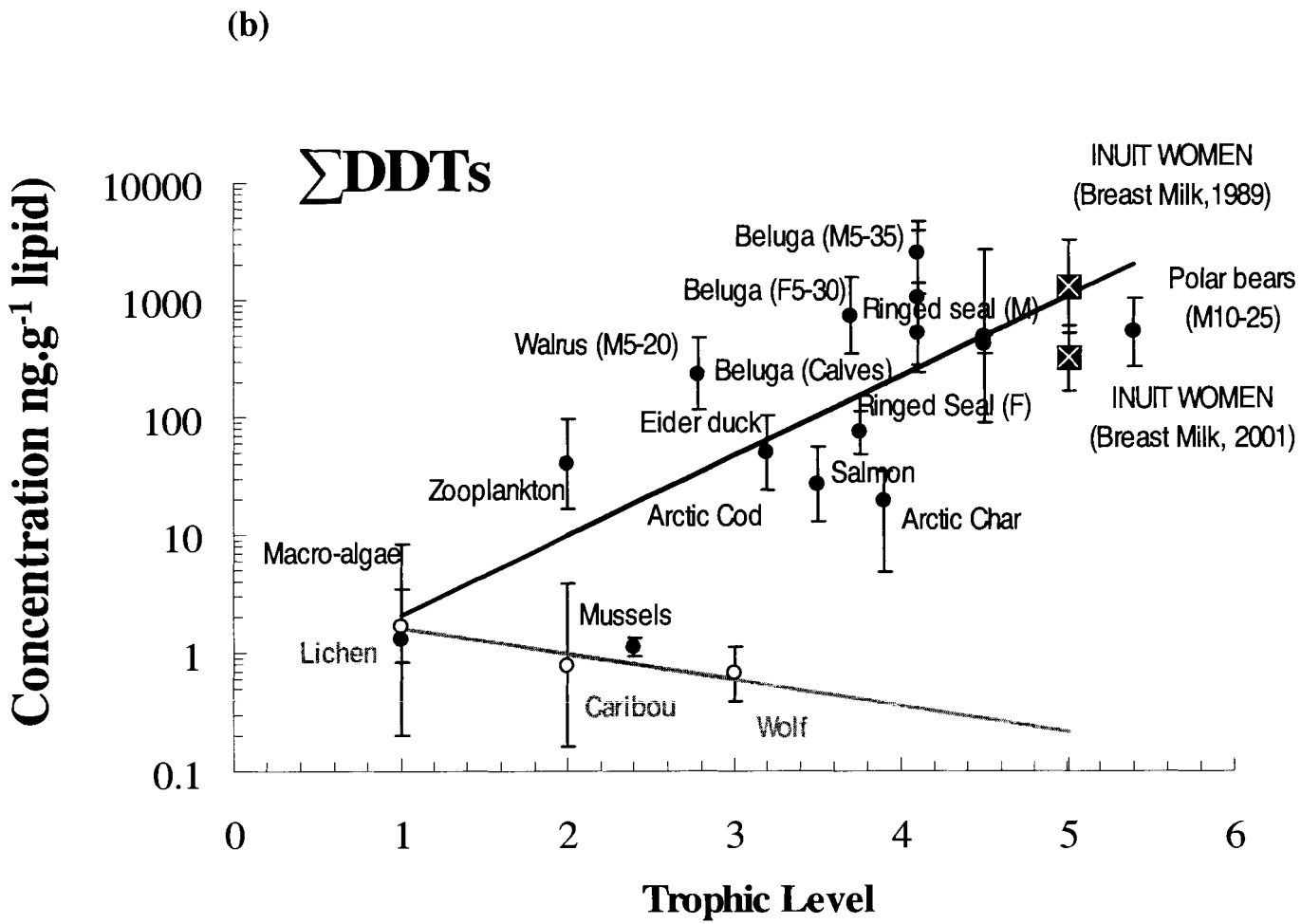

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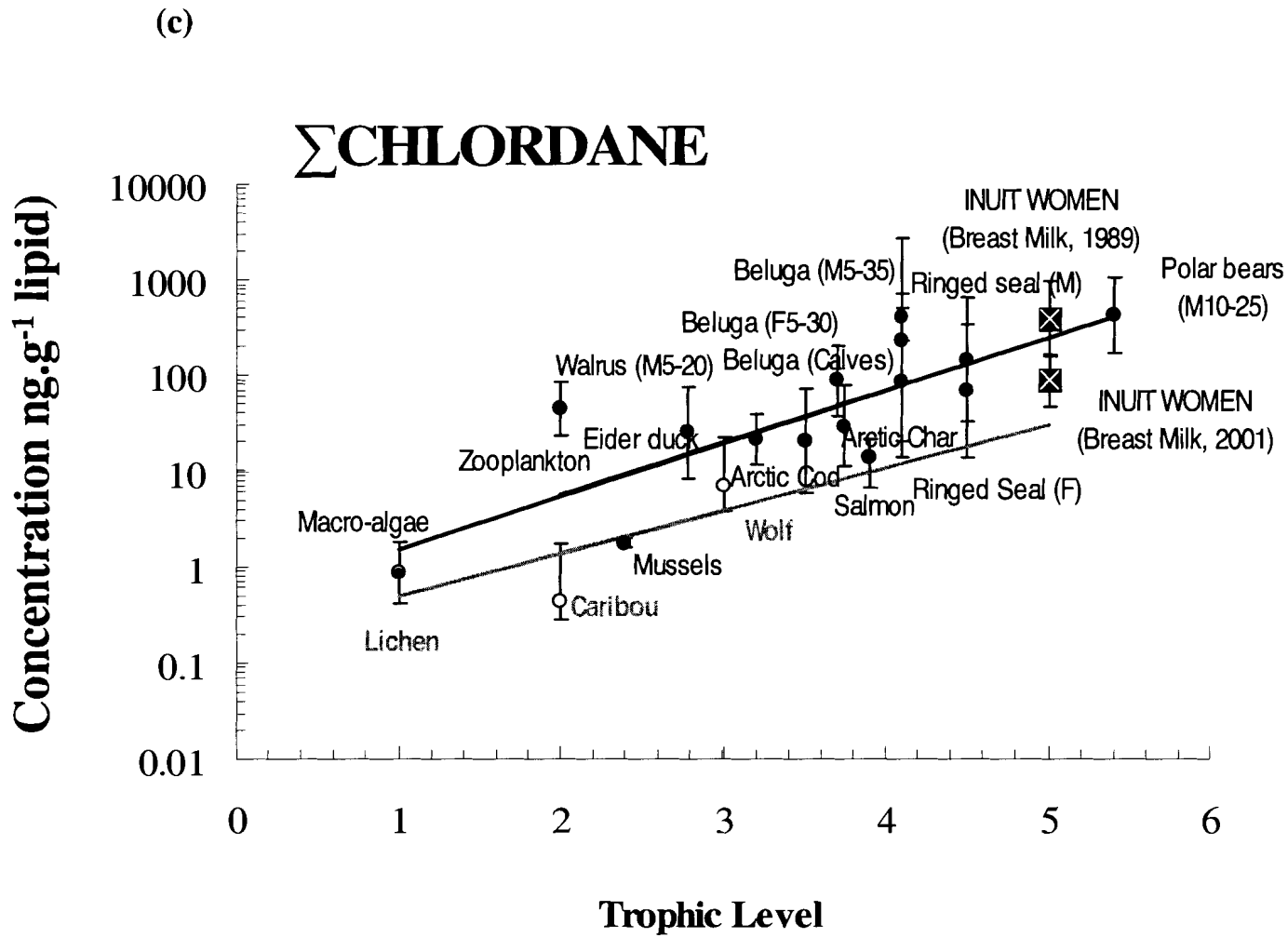

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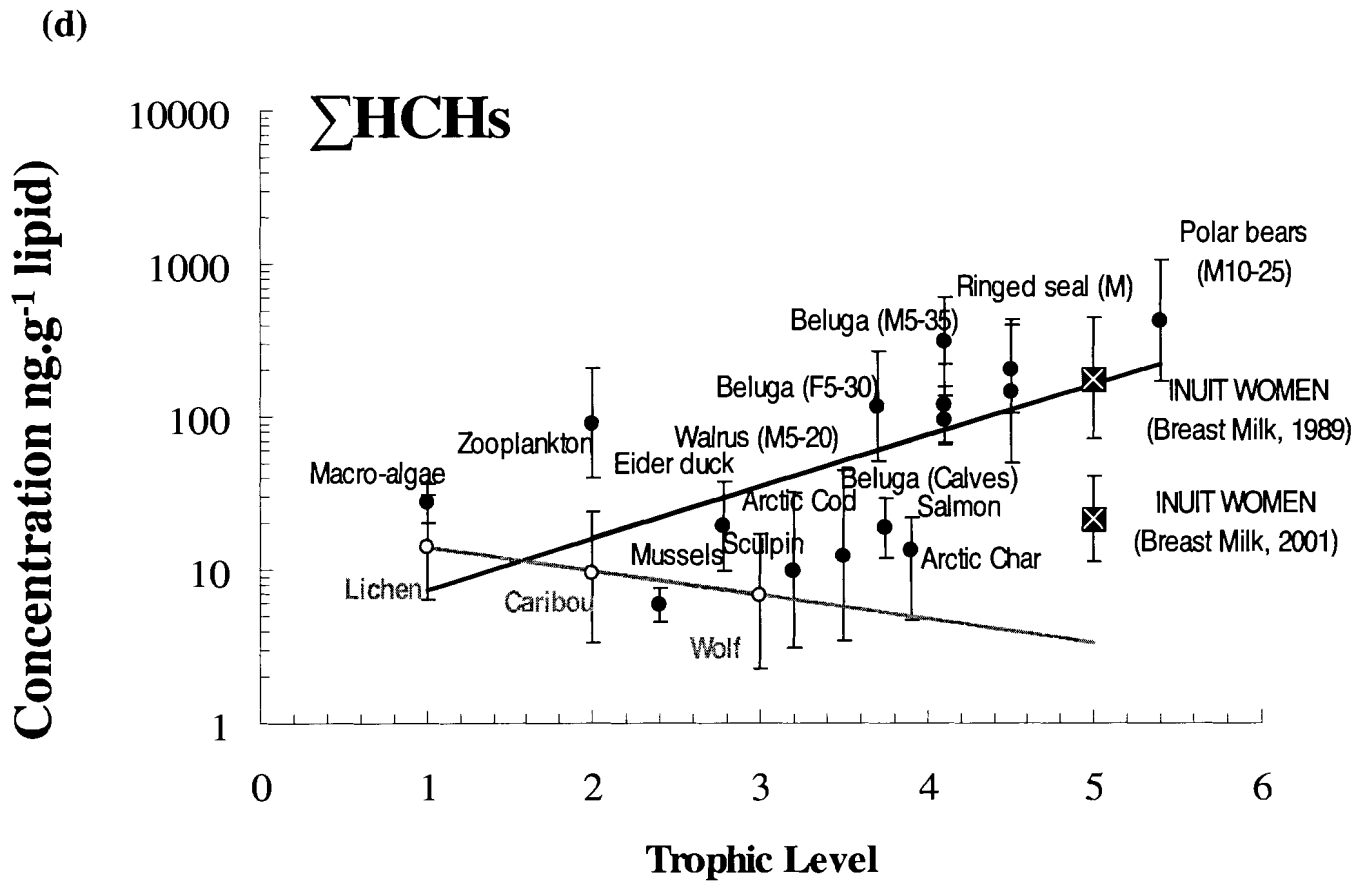


CHAPTER 4

PHYSICAL-CHEMICAL AND BIOLOGICAL DETERMINANTS OF BIOMAGNIFICATION POTENTIAL OF ORGANIC CONTAMINANTS IN AN ARCTIC MARINE FOOD WEB

4.1 Introduction

Persistent organic pollutants (POPs) are classified as long-lived and potentially toxic organic chemicals that are resistant to chemical and biological degradation processes. Cold environments such as alpine and Arctic ecosystems can be particularly sensitive to long-range transport, accumulation and persistence of globally circulating POPs because of enhanced chemical inputs caused by a "cold condensation" effect and slower degradation rates at lower temperatures (*1,176*). Numerous studies of the distribution and bioaccumulation behaviour of legacy POPs such as PCBs and DDT have been conducted on aquatic freshwater and marine ecosystems (*2,3,5,6,46,75*) and to a lesser extent terrestrial systems (*43,44,84*). These compounds also can biomagnify in food chains, resulting in elevated tissue residue concentrations in high trophic level predators that can greatly exceed those concentrations in prey species and the surrounding ambient environment. The significance of the biomagnification phenomenon is that it can potentially raise tissue concentrations in sensitive high-trophic animals that surpass adverse affect levels for toxicological endpoints such as neurobehavioural development, thyroid hormone disruption and fetal toxicity/teratogenicity. For example, biomagnification was essentially the initial biochemical catalyst responsible for organochlorine contaminant induced egg-shell thinning in North American birds of prey in the 1970s (*78*).

A primary objective of the 2004 Stockholm Convention on POPs (*80*) is to implement measures towards the virtual elimination of twelve legacy POPs, namely PCBs, DDT, dioxins/furans, chlordanes, hexachlorobenzene, dieldrin, aldrin, endrin, mirex, heptachlor and chloroboranes (i.e.,toxaphene). However, many of these compounds have long since been prohibited for commercial applications. Thus, current toxic substance management initiatives (both domestic and under UN auspices) aim to assess the persistence (P), bioaccumulation (B), toxicity (T), and

long-range transport (LRT) potential of new and existing commercial chemicals. It is widely recognized that a chemical's hydrophobicity (delineated by the octanol-water partition coefficient or K_{OW}) is a very important factor affecting environmental fate and bioaccumulation in organisms and food chains (*3,4,7,35,37,177*). The relationship between chemical K_{OW} and toxic effects has also been well established. Overton and Meyer during the early 20th Century first demonstrated that increasing chemical lipophilicity (i.e., chemical K_{OW},) thereby enhances chemical toxicity (*178-181*). Common toxicological endpoints such as LC_{50} and LD_{50}'s for neutral organic chemicals are strongly related to their K_{OW} as enhanced partitioning into lipid-rich central nervous system membranes occurs with increasing chemical hydrophobicity (*182-185*). Controlled toxicokinetic studies in laboratory fish have demonstrated that substances with log K_{OW}'s < 5 do not biomagnify in aquatic organisms' due to elimination of these less hydrophobic compounds to water *via* the gills (*37,41,42*). Based on this science, regulatory agencies in Canada, US and Europe have adopted management policies for POPs (e.g., Canada's Toxic Substance Management Policy, TSMP) that identity chemicals exhibiting log K_{OW}'s > 5 as "bioaccumulative". However, a number of studies have shown that aquatic gill-ventilating invertebrates and fish (i.e., water-ventilating ectotherms) exhibit very different POPs bioaccumulation behaviour and potentials compared to higher trophic air-breathing endotherms such as birds and mammals (*5, 46,172*).

Our recent works on POPs in Arctic caribou and wolves indicates that chemicals such as chlorobenzenes (CBz) hexachlorocyclohexanes (HCHs) can biomagnify in air-breathing endotherms because those compounds are moderately polar (i.e. log K_{OW}'s range between 2 - 5) but relatively non-volatile due to high octanol-air partition coefficients (K_{OA}'s > 10^5), (*7,8*). These studies suggest that regardless of the chemical's K_{OW}, air-breathing endotherms may not efficiently eliminate relatively non-metabolizable "high" K_{OA} compounds due to a combination of (i) efficient gastro-intestinal uptake and (ii) negligible lipid to air respiratory elimination rates of those compounds. We estimate that about 30% of the ~ 23,000 commercial substances currently on Canada's Domestic Substances List (DSL) exhibit this moderately polar and non-volatile criteria and hence may potentially biomagnify in various air-breathing taxa including reptiles, amphibians, birds, marine and terrestrial mammals (*100,140*). We have recently proposed the development of a novel QSAR involving chemical K_{OA} for future POPs screening initiatives (*172*). We feel that to ensure future POPs initiatives and regulations effectively apply to all organisms and ecosystems it is important to further investigate chemical bioaccumulation behaviour as a function of the chemical's K_{OA}.

In this paper, we investigate the effect of biological factors such as organism metabolic capacity and physiology (i.e., water-ventilating ectotherms vs. air-breathing endotherms) and also physical-chemical properties (e.g., K_{OW} and K_{OA}) on the biomagnification potential of various organic contaminants, including PCBs, DDTs, chlorobenzenes (CBz), hexachlorocylohexanes (HCHs), and cyclodiene pesticides. Residue concentrations of these compounds in species of algae, invertebrates, fish, seaducks and marine mammals sampled from Eastern Hudson Bay (EHB) in Canada's eastern low-Arctic region were determined by high-resolution gas chromatography-mass spectrometry (HRGC/HRMS). Chemical concentrations in organisms of varying trophic level are presented and evaluative bioaccumulation parameters such as elimination index (EI), chemical biomagnification factors (BMFs), bioaccumulation factors (BAFs) and food web magnification factors (FWMFs) are calculated and discussed.

4.2 Materials and Methods

4.2.1 Sample collections.

During the months of May to August between 1999 and 2003 various biological samples were collected along the eastern Hudson Bay coastline in close proximity to the Inuit village Umiujaq (64° 15'N 113° 07' W), (Figure 4.1). For details see *Chapter 1, Section 1.9.1* and Appendix 1, which summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex, age and condition.

4.2.2 Food web characterization and designation of organism trophic levels.

Figure 4.2 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been established by extensive ^{15}N and ^{13}C isotope enrichment analyses involving numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of $TL = 1 + (\delta^{15}N - 5.4)/3.8$. More recent studies using $\delta^{15}N$ measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 (see *Chapter 1*) summarizes these

previous $\delta^{15}N$ measurements and TL ranges for the various organisms within these Arctic marine food webs. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 4.5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific $\delta^{15}N$ measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

4.2.3 Extraction, cleanup and analysis of POPs in tissue samples.

Tissue samples (approximately 10 g wet wt for lichens and macro-algae, 5-15 g for fish, 2 g for beluga whale liver and 0.5 g for blubber (beluga whales and ringed seals) were homogenized with approximately 20 g Na_2SO_4 with mortar and pestle. Sub-samples of other tissue samples (e.g., seaduck and marine mammal tissue samples) were excised from the interior of frozen samples to reduce potential contact contamination during collection and/or storage. The homogenate powder was transferred to a glass extraction jar, spiked with ^{13}C-labeled procedural internal standards (Cambridge Isotope Laboratories, Andover, MA), including PCB surrogate solution (approx. 2000 pg of each ^{13}C-CBs 28, 52, 101, 128, 156, 180, 194, 206, 209), and OC pesticide surrogate solution (approx. 5000 pg of each d_3 1,3,5 Trichlorobenzene, ^{13}C 1,2,3,4 Tetrachlorobenzene, ^{13}C Hexachlorobenzene, ^{13}C beta HCH, ^{13}C lindane (gamma-HCH), ^{13}C mirex, ^{13}C oxychlordane, ^{13}C dieldrin, ^{13}C *p,p'* DDT, ^{13}C o,p' DDT, ^{13}C *p,p'* DDE, ^{13}C heptachlor epoxide, and ^{13}C trans nonachlor). The spiked samples were then extracted with 30 mL of 1:1 (v/v) DCM/Hexane in a Branson 5210 ultrasonic water-bath (Branson Ultrasonics Co., CT) for 20 min. Once the suspended particles settled, the supernatant was removed, then extraction was repeated two more

times with fresh solvent. The combined extracts were concentrated to ca. 2 mL with a gentle stream of high-purity nitrogen. Relatively low lipid samples (< 5% lipid w/w) such as cod and sculpin tissue were quantitatively transferred onto a 350 mm x 10 mm i.d. glass column packed with 8 g 100% activated florisil (60 -100 μm mesh, activated at 400 °C overnight). High lipid samples (> 5% lipid w/w) such as salmon, and beluga blubber were first passed through a Gel Permeation Column (GPC) filled with 70 g of BioBeads, S-X33 (BioRad) in 50% DCM/hexane solution (V/V). The lipid fraction from the GPC (180 mL) was collected and discarded, while the remaining 300 mL of eluent from the GPC was collected evaporated to near dryness and solvent exchanged into hexane for further cleanup by Florisil. Three fractions were then eluted using 60 mL hexane (fraction 1), 60 mL 15% DCM/hexane (fraction 2), and 120 mL 50% DCM/hexane (fraction 3). The four fractions were combined in a single 500 mL boiling flask and evaporated to a final volume of 100 uL. The extracts were then spiked with recovery standards (^{13}C-CB111 for PCBs, and ^{13}C-CB47 for pesticide quantifications) and analyzed by HRGC/MS using two separate conditions. Method blanks, consisting of Na_2SO_4, were extracted according to the same procedure as environmental samples and analyzed with every batch of 12 samples to check for contamination of the extracts. The samples were analyzed by HRGC/MS in batches of twelve, each containing one procedural blank and 11 samples. Duplicates and analyte spiked matrices were occasionally extracted and analyzed to evaluate extraction/cleanup efficacy.

4.2.4 Data compilation/treatment and statistics.

Physical-chemical properties including molecular weights (MW, g mol^{-1}), log octanol water partition coefficient log K_{OW}, log octanol-air partition coefficient log K_{OA}, Henry's Law Constants (H, Pa m3 mol^{-1}) and water solubility (C_WSoL, ng·L^{-1}) were compiled for several PCBs and OCPs using references *50-56*,(see Appendix 1). The target analytes in the present study exhibit a wide range in chemical K_{OW} and K_{OA} values which allow for a more robust assessment of the influence of chemical polarity and volatility on chemical bioaccumulation behaviour in the food chain. To enable direct comparisons of POPs between different environmental media and organisms it is important to correct chemical concentration data to a common unit expression such as lipid equivalent concentrations. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~1 - 98%) , wet weight chemical concentrations (C, ng·g^{-1} ww) were expressed solely on a lipid weight basis by the equation: C_L = C ww. ÷ ϕL in units of ng·g^{-1} lipid wt.. For some biological matrices with very low lipid fractions (ϕL < 1%), such as vegetation and algae tend to solubilize organic contaminants in non-lipid biomolecules (i.e., non-

lipid organic matter, NLOM) rather than in extractable lipids (*13,57,58,59*). Thus, for macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and NLOM (ϕ_{NL}) fractions following the equation: $\phi Leq = \phi L + 0.035\phi_{NL}$, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42,44*). Because chemical concentrations exhibited log-normal distributions and were hence transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for POPs in the various organisms collected and analyzed as part of the present study (i.e., lichens, macro-algae, bivalves, fish, beluga whales and ringed seals). In addition, we also compiled literature reported concentration data for PCBs and OC pesticides in Canadian Arctic biota , including invertebrates (*4*), walrus (*Odobenus rosmarus*) (*60*) polar bears (*Ursus maritimus*) (*61*), barren-ground caribou (*Rangifer tarrandus*) (*43,62,63*), wolves (*Canis lupus*) (*43,63*) and northern Quebec Inuit women (i.e., breast milk samples from references *64,63*) to compare contaminant concentrations, profiles and BMFs in various wildlife species and humans that generally subsist within the same food web.

PCB congeners were categorized by planarity and Cl-substitution patterns following Boon and colleagues Group I-V metabolic classification (*65*). Group I and II congeners are generally non-metabolizable in most organisms, Group III CBs with vicinal *ortho-meta* H atoms 1 *ortho* Cl can be metabolized by induction of methylcholanthrene (MC) type isozymes of the cytochrome P450 monooxygenase enzyme family (i.e., CYP 1A enzymes) and Group IV and V congeners, with vicinal *meta-para* H atoms and 1- 3 *ortho* Cls can be metabolized by induction of phenobarbitol (PB) type isozymes (i.e., CYP 2B enzymes). A total of 169 di-ortho and mono-ortho substituted PCB congeners were analyzed (see Appendix 2). Due to several coeluting di-ortho (DO) and mono-ortho (MO) PCBs we have summarized a total of 148 PCB congeners. When environmentally dominant CB congeners coeluted with environmentally irrelevant congeners, we have for the purposes of this study, assumed the coeluting concentration as the single dominant compound. For example, Cl_6-CB153/132 concentrations (coeluting congeners in HRGC/MS method) are expressed solely as a Cl_6-CB153 concentration because of that congeners dominant contribution in environmental and biological samples. Specifically, this assumption was used for CBs 52, 101, 118 and 138. Linear regressions and One-Way Analyses of Variance (ANOVA) tests were performed on calculated log-transformed concentrations to determine statistically significant differences between the geometric means of concentrations in biota.

4.2.5 Evaluative parameters for assessing chemical bioaccumulation potential.

See *Chapter 1, Section 1.9.5*

4.3 Results and Discussion

4.3.1 Chemical concentration relationships with trophic level and FWMFs.

The levels of PCBs and OCPs in E. Hudson Bay marine biota, including bivalves, fish, seaducks and marine mammals are presented in greater detail in a preceding paper (*186*) and are summarized in Appendices 5 and 6, respectively. The data are not blank subtracted as procedural blanks for PCBs and organochlorine pesticides were generally low or non-detectable. Method detection limits (MDLs) were determined as the instrument limit of quantification (LOQ) on the HRMS. Results from log-linear regressions analyses of organism chemical concentrations (C_B) and trophic level (TL) and corresponding food web magnification factors (FWMFs) (i) water-ventilating ectotherms, (ii) air-breathing endotherms and (iii) the overall food are summarized in Appendix 11. Four different types of C_B-TL relationships were identified, including (i) strong positive C_B-TL relationships for both water-ventilating ectotherms and air-breathing endotherms with FWMFs ~ 5-14, (ii) moderate positive relationships for both organism groups, FWMFs ~ 2-6, (iii) negative or no relationship (i.e., FWMFs < 1 and (iv) no food web magnification for water-ventilating ectotherms (FWMF < 1) but positive C_B-TL relationships for air-breathing endotherms with FWMFs in those animals > 3. Strong positive C_B-TL relationships in both organism groups were observed for the highly chlorinated (Cl_6-Cl_9) recalcitrant PCBs (Group I and II congeners) such as Cl_6-CB138, Cl_7-CB180 and Cl_6-CB153. Figure 4.3 is a plot of CB153-TL regression lines for water-ventilating ectotherms, air-breathing endotherms and the overall food web together with observed Cl_6-CB153 concentrations (GM $\pm$ 1 SD ng·g^{-1} lipid), with estimated slopes equivalent to approximately 1.05, 1.04 and 0.84, respectively. This corresponds to FWMFs of CB153 equal to approximately 11.0 for the overall food web, 11.3 for air-breathing endotherms and 6.84 for water-ventilating ectotherms, respectively. CB153 concentration data for eastern Arctic amphipods (*4*), male walrus (*60*), male polar bears (*61*) and breast milk samples of Inuit women (*64*), while not used in our regression analyses, are plotted in figure 4.3 for comparison and appear to generally agree with the CB153 –TL regression models.

Strong positive C_B-TL relationships were also observed for several hydrophobic OC pesticides such as *trans*-nonachlor, *p,p'*DDE, HCBz and dieldrin (shown in Figure 4.4a), demonstrating the high degree of biomagnification potential and persistence of these compounds in the Arctic marine food web. That higher concentrations in air-breathing endotherms (i.e., seaducks and marine mammals) tend to drive up the overall FWMF for recalcitrant POPs is expected due to generally higher dietary absorption efficiencies (E_D) and biomagnification factors (BMFs) in those organisms compared to invertebrates and fish (*5 46,88*). Less chlorinated (Cl_3) Group III-V metabolizable PCBs such as CB28 (Group III) and CB18 (Group IV) tend to show significant yet slightly weaker relationships with trophic level (Figure 4.4b and 4.4c), likely due to more efficient CYP1A and 2B enzymatic biotransformation of those congeners. TriCBz exemplify chemicals that can demonstrate a negative/near zero slope and FWMFs < 1, indicating trophic dilution. 1,3,5 TriCBz (shown in Figure 4.4d) is moderately polar (log K_{OW} ~ 3.5) and volatile (log K_{OA} ~ 5.8), which suggests it should be efficiently eliminated by both aquatic water-ventilating ectotherms *via* lipid-water equilibrium partitioning (*37,39*) and by air-breathing endotherms *via* lipid-air exhalation route (*44*). Conversely, moderately polar (log K_{OW} ~ 3.8 – 4.5) yet less volatile (log K_{OA}'s ~ 8.2 - 10.5) such as TeCBz and HCH isomers (Figure 4.4e-h) exhibit biomagnification in air-breathing endotherms (i.e., FWMFs >>1) but demonstrate trophic dilution for water-ventilating ectotherms (i.e., FWMFs < 1). For example, FWMFs of β-HCH in E. Hudson Bay water-ventilating ectotherms and air-breathing endotherms were approximately 0.9 and 3.0, respectively. Biomagnification of β-HCH has previously been observed in Arctic seabirds and mammals (*5,43,46,60,169,187,188*).

4.3.2 POPs biomagnification potential.

BMFs and Elimination Index (EI) estimations in E. Hudson Bay organisms (summarized in Appendix 8) are presented in more detail in reference *186*. Briefly, BMFs of POPs varied widely between different chemicals and different organisms, ranging from zero to 250. Group I CB congeners such as CB153 and CB180 typically exhibited the highest BMFs in organisms of the E. Hudson Bay food web, which is consistent with other food web bioaccumulation studies (*2,3,37,43*). Those BMFs can therefore be viewed as standard measure for an organism's mechanistic biomagnification potential (i.e., BMF_{MAX}). BMFs of recalcitrant PCBs differed substantially between the two organism groups. For example, BMFs of CB180 in air-breathing endotherms such as eider ducks (95.9), male polar bears (94.0), male beluga (45.7) and male ringed seals (11.5) were approximately 10-30 times higher than those BMFs in water-ventilating

ectotherms such as Arctic cod (3.5) and sculpin (3.7). The rank order of BMFs in air-breathing endotherms was generally: BMF-polar bear ~ BMF eider ducks > BMF-beluga > BMF-seals. These data suggest the maximum biomagnification potential (i.e., BMF_{MAX}) is not necessarily related to the organism's trophic position, otherwise ringed seals (TL ~ 4.5) would have exhibited a higher BMF than eider ducks (TL ~ 2.8) and beluga whales (TL ~ 4.1). Relatively low BMFs of POPs in Arctic seals compared to other Arctic marine mammals has been previously observed (*5,6,49,117,189*). Thus, while chemical concentration increases over the entire food web are strongly related to trophic level, organism-specific biomagnification potential (e.g., BMF_{MAX}) may be more a function of prey properties (e.g., lipid content) and biochemical processes such as intestinal physiology/absorption efficiency (*172*).

Some Group III and IV PCB congeners exhibited relatively high elimination index (EI) in the various organisms of the E. Hudson Bay food web, indicating induction of both CYPIA and CYP2B isozymes (Appendix 8). Low MW congeners such as Cl_2-CB6 within Group III and Cl_2-CB4/10 within Group IV exhibited the highest degree of metabolism, with EI values ranging between 1.5 and 2.2. While high EI values for these less chlorinated and hence less hydrophobic CBs (i.e., log K_{OW}'s ~ 5 for Cl_2-CBs) in gill breathing water-ventilating ectotherms (e.g., cod, sculpin) may be the result of respiratory elimination to water *via* equilibrium partitioning, the EI values > 1 in air-breathing endotherms such as seaducks and marine mammals indicates efficient internal biotransformation of these compounds because substantial respiratory elimination *via* air is likely negligible due high K_{OA}'s of Cl_2 to Cl_{10} PCBs (i.e., log K_{OA}'s > 7 for Cl_2-Cl_{10} CBs. The EI and BMF data suggest the rank order for CB metabolism capability in Arctic marine biota is polar bear > walrus > ringed seals > seaducks > beluga > cod.

In general, biotransformation of non-volatile hydrophobic pesticides such as DDTs and cyclodienes (e.g., chlordanes and mirex) in seaducks and marine mammals was negligible (i.e., EIs < 0.5, with BMFs ranging between 5-100). Relatively high EI values were observed for all HCH isomers in water-ventilating ectotherms (EI > 1.5 and BMFs < 1 for cod and sculpin). EI values of α and γ-HCHs observed in air-breathing endotherms indicate efficient biotransformation of those isomers in seaducks and marine mammals. HCHs are moderately polar but relatively non-volatile pesticides (i.e., log K_{OW}'s ~ 3.8 and log K_{OA}'s > 8), which suggests eliminated kinetics *via* gill ventilation is likely dominant in aquatic water-ventilating ectotherms but metabolic transformation is likely dominant route of elimination in due to negligible respiratory elimination. β-HCH exhibited EI values near zero in air-breathing

endotherms, with relatively high BMFs in beluga whales (~50) and ringed seals (~20), which are comparable to CB180 and CB153 BMFs in those organisms. The substantial biomagnification of β-HCH in these higher trophic air-breathing endotherms is likely due to a combination of (i) efficient dietary uptake (ii) relatively high resistance to metabolic transformation and (iii) very slow elimination rates through respiration (due to high K_{OA}) and urinary excretion (log K_{OW} >2). The highest EI values and hence lowest BMFs were exhibited by relatively low molecular weight, polar and volatile compounds such as Cl_3 and Cl_4 chlorobenzenes. For the water-ventilating ectotherms, high EI values and hence low BMFs of Cl_3 to Cl_4 CBz (BMFs < 1) is likely due to efficient elimination through the gills, which has been well documented laboratory and field studies involving aquatic invertebrates and fish (*37,39*). Unlike the HCHs, the relatively high volatility of these compounds (log K_{OA}'s ~5) may result in elevated chemical elimination through respiration in air-breathing endotherms. Thus, relatively low BMFs of moderately polar/volatile chemicals such as the chlorobenzenes (i.e., BMFs between 1-5 for air-breathing endotherms) may be the combined effect of metabolism and respiratory elimination. It is difficult to discern the relative importance of respiratory elimination versus metabolism of these "low" K_{OA} compounds using field surveyed concentration data and will likely require studies in laboratory animals under controlled conditions. However, investigations of occupational exposure of volatile industrial chemicals such as N-nitrosamines and styrene with log K_{OA}'s ~2 -4 (*190,191,192*) and medical studies of inhalation anaesthetic agents such as isoflurane and halothane (log K_{OA} between 1 -2), (see references *182,193 194,195,196*) highlight the importance of lung-air equilibrium partitioning as a driving mechanism for elimination of moderately polar and volatile (i.e., "low" K_{OA}) chemicals in air-breathing animals.

4.3.3 Effect of K_{OW} and K_{OA} on chemical bioaccumulation potential.

Lipid corrected chemical bioaccumulation factors (log BAFs) for selected PCBs and OC pesticides are summarized in Appendix 12, along with corresponding K_{OW} and K_{OA} values. Figure 4.5 shows two log-log plots of recalcitrant CB congeners and OC pesticide BAFs for the various organisms for various species of the E. Hudson Bay food web, including (a) BAFs (C_B/C_{WD}) for water-ventilating ectotherms plotted versus chemical K_{OW} and (b) BAFs for air-breathing endotherms (C_B/C_{AG}) plotted against chemical K_{OA}. Second order quadratic regression models best fit these BAF-K_{OW} and BAF-K_{OA} relationships. The dashed lines shown in figure 4.5 are 1:1 log-log relationships for plots of BAFs-K_{OW} (4.5a) and BAF-K_{OA} (4.5b) and hence represents a chemical equilibrium between organism lipids or lipid equivalent media (e.g.,

organic matter) and the ambient environment. BAFs of moderately polar compounds such as the CBz and HCHs in E. Hudson Bay water-ventilating ectotherms (i.e., bivalves, sculpin and cod) along with aquatic macro-algae are equivalent or less than the predicted lipid-water equilibrium concentrations (1:1 K_{OW}:BAF line). In general, BAFs show an initial linear increase (~10^4 and 10^8) for chemicals with logK_{OW}'s between 2 and 6, then plateau for chemicals with logK_{OW}'s > 6, with maximum BAFs approaching~ 10^{10} for Arctic cod. BAFs for macro-algae are in close agreement with the predicted lipid-water equilibrium concentrations, which is expected for primary producers where the primary route of chemical uptake and elimination is by passive diffusion *via* seawater. The slight drop in BAFs in macro-algae for chemicals with logK_{OW} > 6 may be attributed to kinetically limited uptake and/or errors associated with freely dissolved water concentration measurements of these highly hydrophobic compounds (e.g., Cl_7 – Cl_{10} CBs), both of which have been cited as causal factors of reduced BAFs of high K_{OW} substances in aquatic organisms (*3,7,37,42*). BAFs for cod and sculpin are comparable and both are higher than BAFs observed in bivalves. The higher degree of chemical accumulation in fish compared to bivalves may be due to a combination of higher trophic level feeding and more efficient digestive tracts of fish species compared to bivalves. It is evident from figure 4.5a that the relatively low K_{OW} compounds are efficiently eliminated to ambient water through gill ventilation/passive diffusive partitioning, while concentrations of more hydrophobic chemicals are magnified above equilibrium concentrations because of an increased importance of dietary exposure of those compounds.

Figure 4.5b illustrates similar quadratic relationships for BAFs of air-breathing endotherms versus chemical K_{OA}. However, chemical BAFs in those animals (i.e., seaducks and marine mammals) were three to four orders of magnitude greater than those BAFs in the water-ventilating ectotherms, with maximum BAF values of approximately 10^{12} to 10^{13} for Cl_6-Cl_7 CB congeners. BAFs for terrestrial lichens from E. Hudson Bay (also plotted in figure 4.5b), exhibit a linear increase between K_{OA}'s of 10^5 to 10^7 and are comparable to predicted lipid-air equilibrium concentrations. The observed decrease of BAFs in lichens for high K_{OA} chemicals (K_{OA}'s > 10^8) is likely due to kinetically limited gaseous deposition and hence insufficient air-vegetation exchange to achieve equilibrium, which has been demonstrated in previous studies of POPs accumulation in lichens and vascular plants (*44,197,198*). BAFs in ringed seals, beluga and eider ducks were comparable and showed linear increases between K_{OA} 10^5 to 10^8. BAFs of chemicals with relatively low K_{OA} (i.e., 10^5 to 10^6), mainly the Tri to Penta chlorobenzenes, are near the predicted lipid-air equilibria and suggests these compounds are not biomagnified but

rather achieve a chemical equilibrium between chemical concentrations in organism lipids and the animal's surrounding ambient air. The linear increase of BAFs for male ringed seals plateau around 10^{12} for chemicals with $K_{OA} > 10^8$, while male beluga BAFs exhibit a decline for chemicals exceeding $K_{OA} \sim 10^{10}$ (possibly the result of decreased absorption efficiency of the very hydrophobic Cl_8-Cl_{10} CB congeners). Eider duck BAFs are relatively linear over the K_{OA} range of the target compounds (10^5 to 10^{12}) and do not show declines for the highly hydrophobic CB congeners. It appears this avian species may more efficiently absorb very hydrophobic high MW compounds compared to other marine mammals, i.e., ringed seals (pinnipeds) and beluga whales (cetaceans). This is plausible given that birds generally have very high bioenergetic demands and tend to exhibit very rapid and efficient food assimilation to utilize maximize quantities of required dietary constituents (*199*) and may be particularly true for Arctic resident species like the common eider duck that subsist in subzero temperatures much of the year (*63,170*). In general, the BAFs for air-breathing endotherms shown in figure 4.5b indicate that maximum bioaccumulation potential is observed for non-metabolizable chemicals with K_{OA}'s between 10^8 and 10^{10} such as Cl_5-Cl_7 CB congeners (e.g., Cl_6-CB153), which is due to efficient dietary uptake and very slow respiratory and urinary elimination of those hydrophobic non-volatile compounds. Also, decreased bioaccumulation potential can occur when K_{OA} becomes low ($< 10^6$) as chemical volatility increases and those compounds may be efficiently eliminated through the respiratory route. Relatively high K_{OA} chemicals $>10^{10}$, which typically also exhibit very high K_{OW}'s ($> 10^8$) also show decreased bioaccumulation potential because insufficient assimilation in the digestive tract and hence increased advective elimination *via* passage of digesta and excretion of fecal matter.

Appendix 8 shows predator/prey biomagnification factors (BMFs) of the various POPs in E. Hudson Bay air-breathing endotherms, including ringed seals, beluga whales and eider ducks. For the purpose of BMF-K_{OA} regression analyses in this study, presumed "metabolizable" compounds were those chemicals with log $K_{OA} > 7$ and elimination index > 1.5. Calculated BMFs for readily metabolizable PCBs and OC pesticides were not included in those regressions. More volatile compounds such as the chlorobenzenes (i.e., log K_{OA}'s < 7) were included in the BMF-K_{OA} regressions because elimination of these compounds may also be influenced by passive respiratory elimination kinetics in air-breathing endotherms (i.e., *via* lipid-air partitioning) rather than solely due to metabolic transformation.

The log-linear BMF-K_{OA} relationships shown for male ringed seals and male belugas were best fit using a second order quadratic regression model, while a linear model best fit the eider duck BMF-K_{OA} relationship. BMF-K_{OA} relationship was strongest for male beluga BMF = -3.57 $\log K_{OA}^2$ + 66.1$\log K_{OW}$ - 260 (r^2 = 0.434), compared to male ringed seals BMF = -0.922 $\log K_{OA}^2$ + 15.9$\log K_{OA}$ - 58.3 (r^2 = 0.276) and eider ducks BMF = 18.4 $\log K_{OA}$ – 113 (r^2 = 0.357). Regressions of BMFs and K_{OW} were also performed and generally exhibited weaker correlations than the BMF-K_{OA} relationships. With the exception of trichlorobenzenes, all POPs are shown to biomagnify, exhibiting BMFs greater than unity. Increasing BMFs of the Tri to Penta chlorobenzenes tend to drive an initial increasing BMF trend between $\log K_{OA}$ 5-7. For ringed seals and beluga whales, the slight decreasing trend for very high K_{OA} ($\log K_{OA}$ > 11) is due to lower BMFs of Cl_8 and Cl_9 PCBs. The decreasing BMFs for very these relatively high K_{OA} compounds may be due to reduced gastro-intestinal assimilation of those compounds in ringed seals and beluga whales. Low dietary absorption efficiencies (E_D) of relatively large hydrophobic molecules such as the octa-deca chlorobiphenyls observed in laboratory animals (*40,115,119, 200*) is a potential explanatory factor for the relatively low BMFs of those compounds observed in fish, wildlife and humans (*172*).

A key observation in the BMF data is the fact that relatively polar compounds (i.e., $\log K_{OW}$'s < 5) such as HCHs (β and γ isomers) and chlorobenzenes (Tetra and Penta) exhibit quite high biomagnification potential in ringed seals and beluga whales, with BMFs equal to ~3-5 for TeCBz, ~10-20 for PeCBz and γ-HCH and ~30-50 for β-HCH. In fact, the relatively high BMFs of β-HCH in ringed seals (20.3) and beluga whales (50.1) exceed the BMFs of Cl_7-CB180 in those animals. This high degree of β-HCH biomagnification has previously been reported in other air-breathing endotherms, including various species of seabirds and marine mammals (*5,6,46*) and terrestrial mammals (*43*). Recent investigations of technical lindane components (i.e., α, β ,γ, δ -HCH isomers) indicate the β isomer exhibits different physical chemical properties and environmental partitioning/transport behaviour. For example, β-HCH has been shown to have an unusually low Henry's Law Constant (~ 0.045 Pa m^{-3} mol^{-1}) and correspondingly high chemical K_{OA} (i.e., $\log K_{OA}$ = 8.9 at 25 °C) compared to the other HCH isomers (*201*). This deviation of physical chemical properties for β-HCH has been suggested as the cause of seemingly high sorption rates to aerosols and subsequent atmospheric "washout" of this isomer to ocean surface waters *via* rain and snow scavenging events (*66*). Our data from the present study indicate that the relatively low HLC and high K_{OA} of β-HCH may be a key factor

causing the high degree of biomagnification of this compound in air-breathing endotherms in this study (i.e., ringed seals and beluga whales) and may explain similar observations of β-HCH biomagnification in other air-breathing species (*5,43,46,169*). For air-breathing endotherms, the extent of chemical biomagnification of lipid soluble organic contaminants is determined largely by the competing rates of chemical uptake and loss through (i) intestinal absorption, (ii) respiration and (iii) metabolism. Dietary exposure studies of air-breathing endotherms (birds and mammals) show that intestinal absorption of ingested POPs is very efficient (> 90%) for moderately polar substances (log K_{OW} between 3 and 7), but tends to drop slightly when log K_{OW} exceeds ~ 7 (*83*). Thus, if the organism lacks the capacity to metabolize a given compound, respiratory elimination *via* lipid-air partitioning (a K_{OA} controlled process) becomes the key process controlling the elimination kinetics of the absorbed chemical. Our model simulation of POPs accumulation in terrestrial mammals indicate that non-metabolizable "low" K_{OW} compounds (i.e., moderately polar) that also exhibit logK_{OA} > 5 can effectively biomagnify because of insufficient lipid-to-air volatilization in the animal's lungs (*44*). The moderately polar compounds observed to biomagnify in E. Hudson Bay ringed seals and beluga whales (e.g., CBz and HCHs) in this study all exhibit log K_{OA}'s > 5. While some metabolism and/or respiratory elimination of these "low" K_{OW} compounds may likely occur, the BMF data suggest the two processes combined do not adequately void chemical biomagnification for Tetra and Penta CBz and γ-HCH and β-HCH in these animals. Moreover, the rank order of BMFs for these "low" K_{OW} compounds, i.e., BMFs of β-HCH > γ-HCH~ PeCBz > TeCBz > TriCBz, correspond to increasing K_{OA}'s of those compounds, indicating the potential importance of respiratory elimination kinetics.

4.3.4 Regulatory implications of bioaccumulative "low" K_{OW} – "high" K_{OA} chemicals.

Results from our field survey of POPs concentrations in the E. Hudson Bay marine food web indicate many of these compounds can efficiently accumulate, biomagnify, persist in organisms and exhibit sequentially increasing concentrations with increasing trophic level (i.e., algae → invertebrates → fish → seabirds ~ marine mammals). Recalcitrant PCBs such as Cl_6-CB153 and Cl_7-CB180 typically exhibit the greatest prey to predator biomagnification potential (i.e., highest FWMFs and BMFs) and undergo a trophic amplification equivalent to a factor of ~ 3-5 for water-ventilating ectotherms and ~ 20-75 for air-breathing endotherms. The higher degree of biomagnification in air-breathing endotherms compared to water-ventilating ectotherms is likely the effect of more efficient digestive systems and intestinal absorption of organic chemicals in

those animals. Thus, the cause of elevated contaminant burdens observed in high trophic air-breathing endotherms such as beluga whales, ringed seals, polar bears and humans is essentially a twofold effect involving (i) elevated dietary exposure concentrations in mid-trophic prey species due to multiple energy transfers and chemical biomagnification steps in the lower food web and (ii) a large concentration amplification (i.e., high BMF) following prey consumption. These inherent differences in mechanistic biomagnification potential between taxa are implicit in the overall food web magnification factors (FWMFs).

A recent review of FWMFs of POPs in Arctic marine food webs by Borga et al. (*189*) highlights the various biological and chemical factors influencing bioaccumulation of persistent organochlorine contaminants in several Arctic marine food webs. The FWMFs determined for the E. Hudson Bay marine food web determined in the current study are generally comparble to previous FWMF values reported for European and Canadian Arctic marine food webs, including the Barents Sea (*46*), the Northwater Polyna (*5*), Beaufort-Chukchi Seas (*49*), which are all substantially higher than those reported for the White Sea food web in the Russian Arctic (*188*). For example, the FWMF for PCB-153 in E. Hudson Bay (this study) was approximately 11.02 compared to 18.8, 9.7 and 6.7 for the Barents Sea, Northwater Polyna and Beaufort-Chukchi Seas food webs, respectively. In contrast, the observed FWMF for PCB-153 in the White Sea food web was substantially lower (approx. 2.9). The lower FWMF in the White Sea study is likely the result of only using one high trophic level species in the regression analyses (i.e., relationship of invertebrates→fish →seals), while the other studies generally included chemical concentration data for several high trophic animals (e.g., beluga whales, seabirds). Thus, the class of the selected organisms (and the numbers of species within each class) for a particular experimental design can therefore substantially affect a chemical's FWMF for a given food web. Thus, compilation and comparison of literature reported FWMFs for the purpose of chemical risk assessment should be conduced with some caution.

In addition to the observed differences in mechanistic differences (e.g., BMF_{MAX}) between water-ventilating ectotherms and air-breathing endotherms (attributable to organism digestion efficiencies), our results indicate that differences in physical chemical properties (K_{OW}, K_{OA}) and organism-specific respiratory elimination kinetics (lipid-to-water versus lipid-to-air partitioning) are key factors affecting bioaccumulation potential in aquatic and air-breathing animals. Specifically, "low" K_{OW} – "high" K_{OA} compounds such as HCHs and chlorobenzenes were observed to biomagnify in air-breathing endotherms such as ringed seals, beluga whales and eider

ducks, but not water-ventilating ectotherms (i.e., fish). The available data suggest that both water-ventilating ectotherms and air-breathing endotherms efficiently absorb these compounds through the diet *via* gastro-intestinal uptake, but only the aquatic organisms (i.e., water-ventilating ectotherms) can effectively eliminate them through passive lipid-water respiration. For air-breathers (i.e., air-breathing endotherms), respiratory elimination of these compounds through alveolar air may simply be insufficient (due to a high K_{OA}) to counter act dietary uptake rates. This is consistent with our previous findings showing substantial biomagnification of "low" K_{OW} – "high" K_{OA} compounds in barren-ground caribou and wolves from the Canadian Arctic (*43*). It is clear that in order for these "low" K_{OW} – "high" K_{OA} chemicals to exhibit biomagnification and hence attain relatively high tissue concentrations in air-breathing animals, metabolic transformation must be negligible. For example, model simulations of terrestrial mammals indicates that internal chemical half lives ($T_{1/2}$) of less than approximately 30 days are required to negate biomagnification of these "low" K_{OW} – "high" K_{OA} (*44*). In essence, all compounds exhibiting moderate to high hydrophobicity (K_{OW}'s between 2 to 8) and volatility (K_{OA}'s between ~ 6 and 12), in absence of any biotransformation, have the potential to biomagnify in air-breathing organisms. For compounds with those properties, dietary uptake and absorption is high, respiratory elimination is low and consequently chemical elimination *via* metabolic transformation is the primary mechanism which to reduce extensive accumulation. It is well documented that large differences in metabolic transformation rates (k_M) between species and between individuals of a species is a paramount parameter influencing inter- and intra-species variations of bioaccumulation potential and contaminant accumulation patterns. Thus, assessing the extent of compound specific metabolism and also the fate and elimination behaviour of formed metabolites is an important challenge for regulatory agencies conducting risk assessments of commercial chemicals.

Following the recent endorsement of the Stockholm Convention on POPs, government agencies from Canada, the United States and the European Union are now faced with the challenge of categorizing tens of thousands of commercial chemicals in terms of their PBT LRT status. Environment Canada for example has commenced categorization of substances included on Canada's Domestic Substance List (DSL), which involves approximately 23,000 registered chemicals of commerce. For the vast majority of those chemicals, there is a paucity of data regarding environmental fate, food chain bioaccumulation and human exposure potential. Thus, regulators must rely to a great extent on Quantitative Structure Activity Relationships (QSARs) and generic environmental fate and bioaccumulation models to characterize the relative

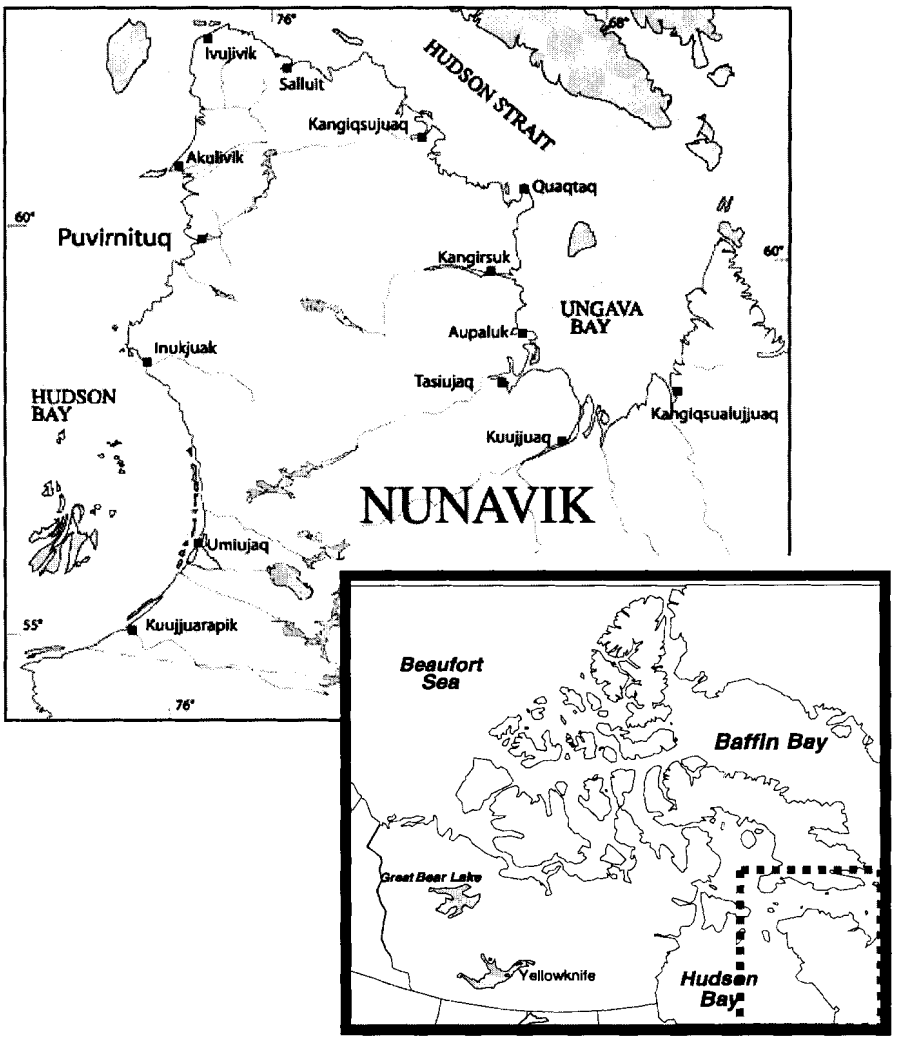
environmental and human health hazards posed by these substances. QSARs and mechanistic models used for assessing POPs bioaccumulation behaviour were initially developed using data generated from numerous field and laboratory studies of PCBs, and OC pesticides, primarily in aquatic organisms and food chains. For example, the most commonly used QSAR for identifying "bioaccumulative" substances is the K_{OW} threshold criterion (log K_{OW}'s > 5), is based on past observations that water-ventilating organisms efficiently eliminated those relatively more hydrophilic compounds to ambient water *via* the gills. Our results showing the ability of "low" K_{OW} – "high" K_{OA} compounds to biomagnify in air-breathing animals highlights the need to develop future QSARs that also include chemical volatility, i.e., octanol-air partition coefficient's (K_{OA}) and metabolic transformation rates (k_M). Specifically, we suggest further development of mechanistic simulation models based on key chemical and biological input parameters such as K_{OW}, K_{OA} and k_M, and which also incorporate a lung-to-air respiratory elimination mechanism for better representation of organic contaminant bioaccumulation in air-breathing animals. Future B criterion and associated QSARs for assessing bioaccumulation potential of commercial chemicals should include K_{OW} and K_{OA} criteria and also k_M's of targeted compounds in different taxa. This task will require future estimation or preferably direct measurement of physical-chemical properties such as K_{OW} and K_{OA} along with animal exposure studies to document the degree of metabolism (k_M) and the occurrence of any significant biomagnification (BMFs >>1). Also, because some metabolites can exhibit substantial persistence and toxicity in organisms, future B criteria should also include assessment of the fate and bioaccumulation potential of both parent compounds and recalcitrant metabolic transformation products.

4.4 Acknowledgements

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies and wish to thank Dr's Bill Doidge, Michael Kwan and Susan Sang, Derek Muir and Northern Quebec Inuit communities of Umiujaq and Inukjuaq for coordinating/aiding collection of field samples.

4.5 Figures

Figure 4.1 Map showing general study area of E. Hudson Bay and various Nunavik Inuit communities of northern Quebec, Canada.



Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

Figure 4.2 Conceptual illustration of E. Hudson Bay marine food web organisms and assigned trophic levels (TL).

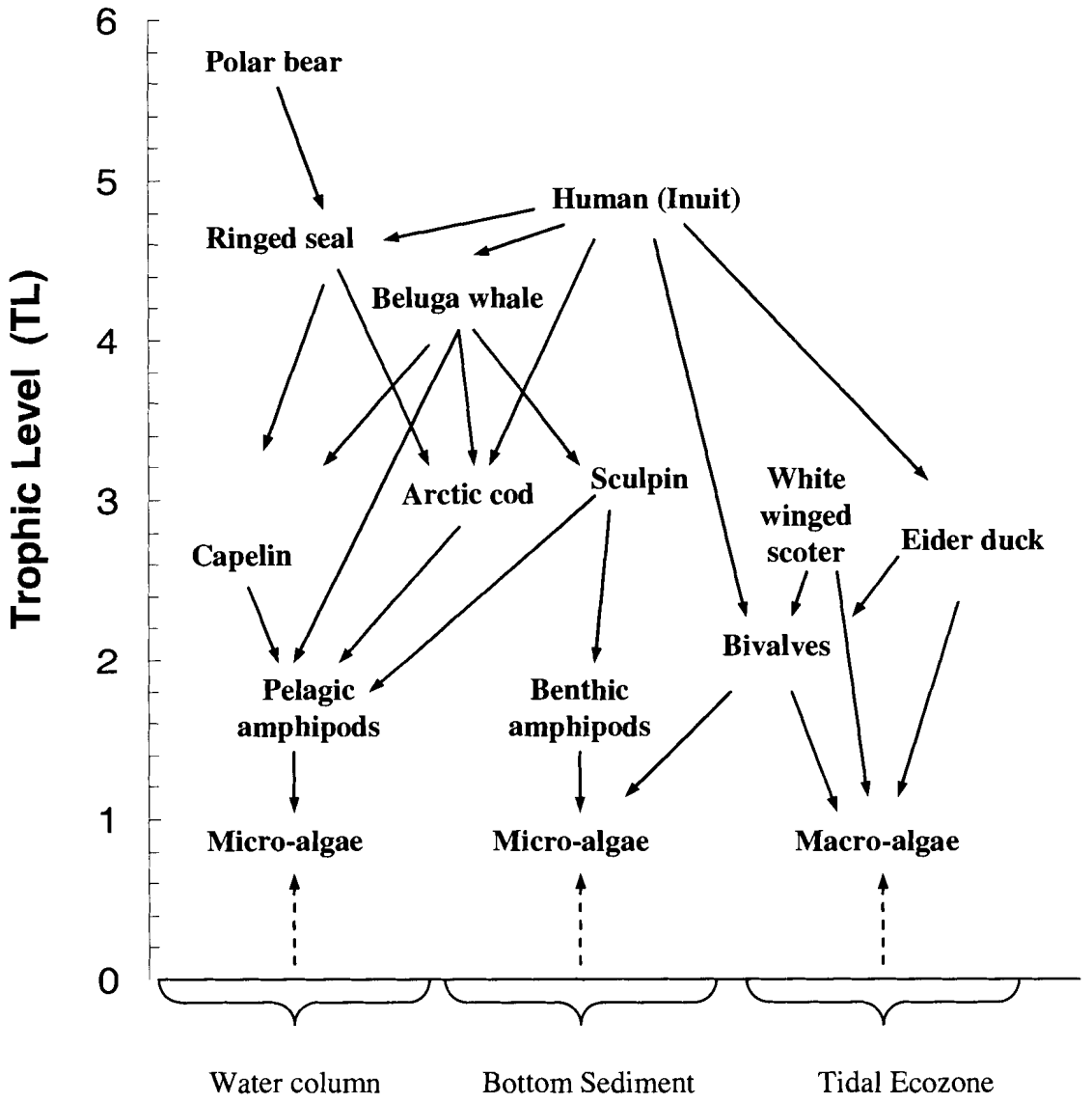


Figure 4.3 Relationship between Cl_6-CB153 concentrations ($ng \cdot g^{-1}$ lipid) and trophic level (TL) for the various water-ventilating ectotherms and air-breathing endotherms of the E. Hudson Bay marine food web.

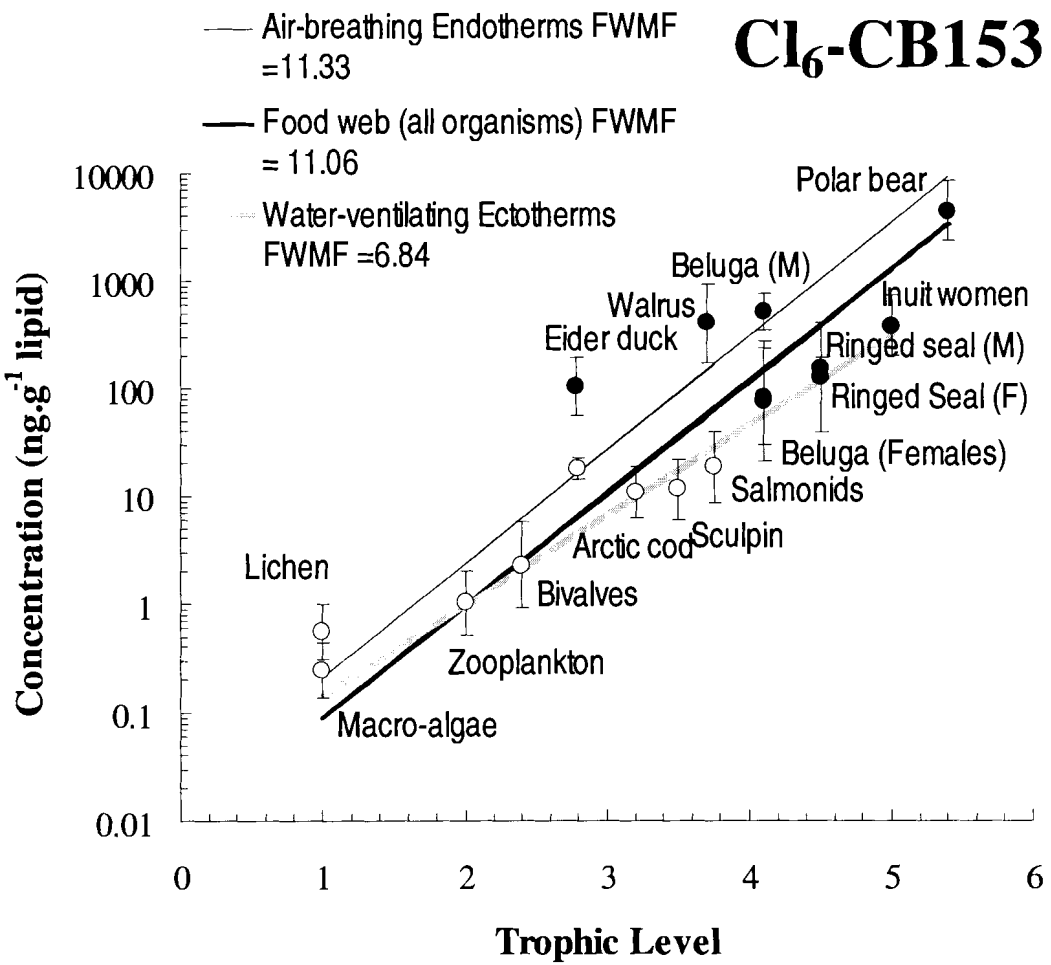
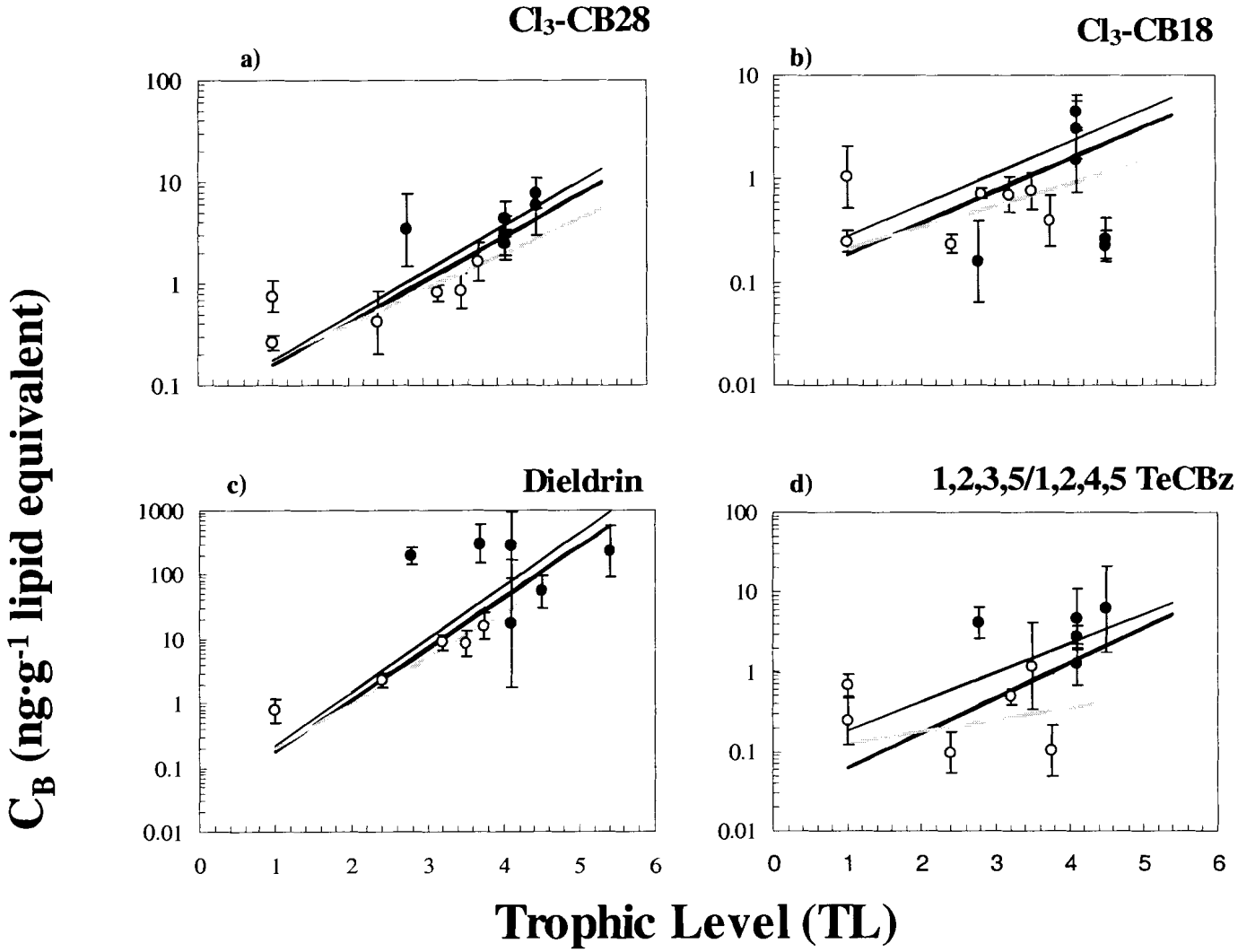


Figure 4.4 Relationship between chemical concentration (ng·g^{-1} lipid) in various organisms in the E. Hudson Bay marine food web versus organism trophic level (TL) for (a) CB28 (b) CB18 (c) dieldrin (d)1,2,3,5/1,2,4,5 TeCBz, (e) α-HCH, (f) 1,3,5 TriCBz, (g) γ-HCH and (h) β-HCH. Thick black line represents data for whole food web, thin black line represents air-breathing endotherms, and gray line represents water-ventilating ectotherms



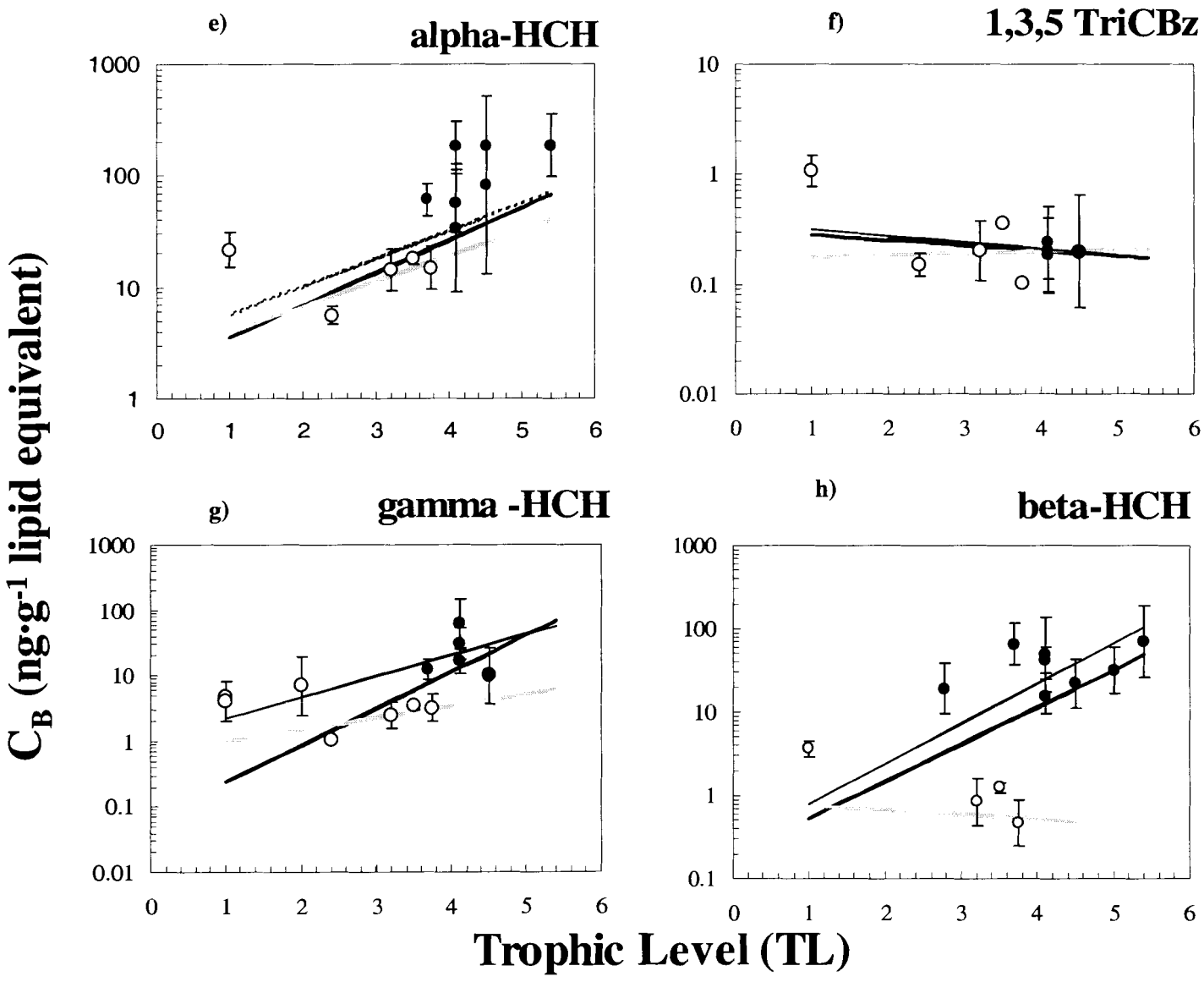
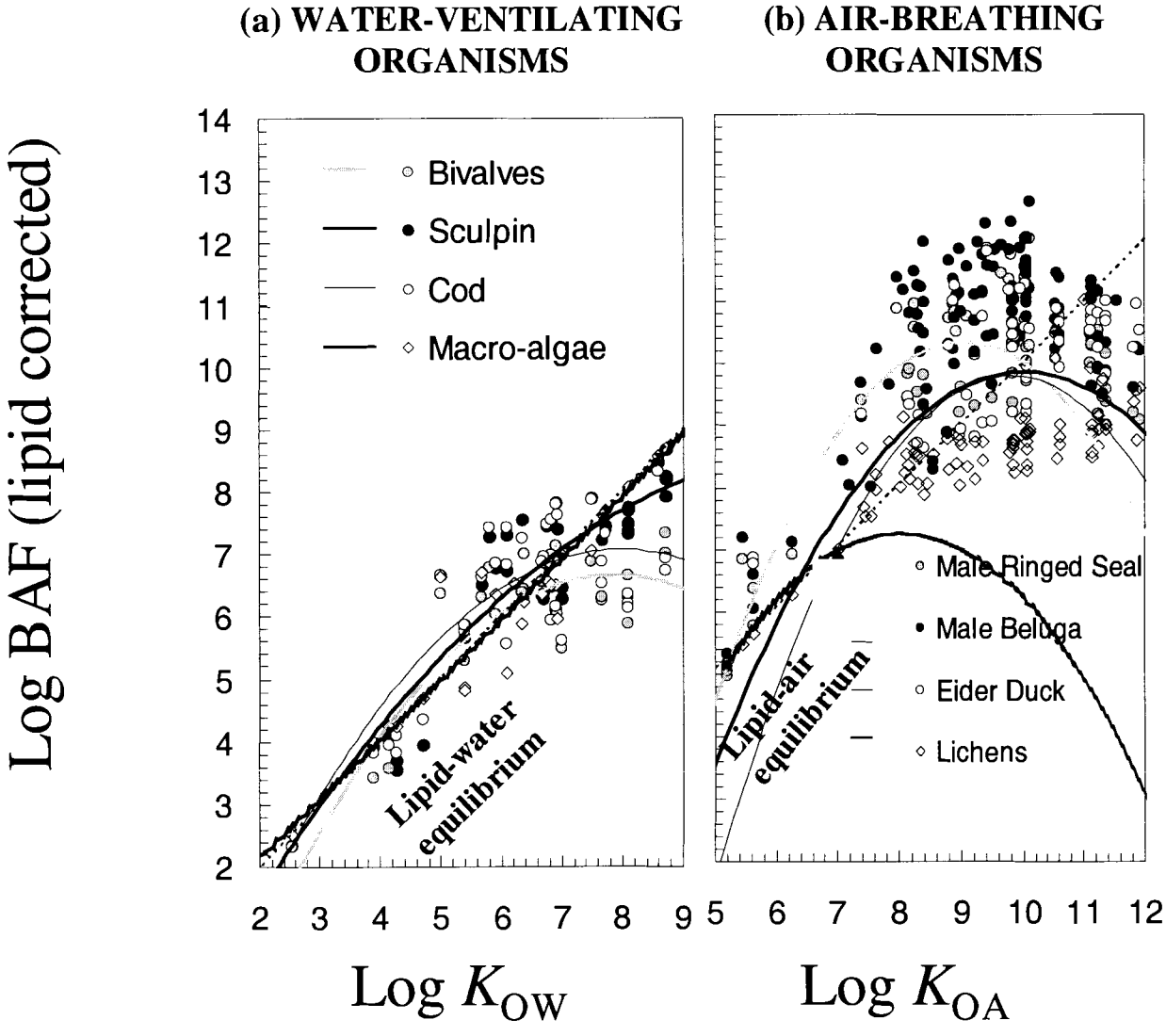

e)
alpha-HCH
f)
1,3,5 TriCBz
g)
gamma -HCH
h)
beta-HCH
C_B (ng·g^{-1} lipid equivalent)
Trophic Level (TL)

Figure 4.5 Relationships between chemical bioaccumulation factors (plotted as log BAFs) in (A) water-ventilating ectotherms (C_B/C_{WD} mol m^{-3} lipid) such as bivalves, Arctic cod and sculpin and (B) for air-breathing endotherms determined as (C_B/C_{AG} mol m^{-3} lipid) in eider ducks, male ringed seals and beluga whales. Second order quadratic regressions were as follows: Lichens: $\log BAF = -0.102 \cdot \log K_{OA}^2 + 2.25 \cdot \log K_{OA} - 3.52$; Macro-algae: $-0.115 \cdot \log K_{OW}^2 + 2.02 \cdot \log K_{OW} - 1.95$; Bivalves: $\log BAF = -0.102 \log K_{OW}^2 + 2.19 \log K_{OW} - 1.53$ ($r^2 = 0.777$); Sculpin: $\log BAF = -0.266 \log K_{OW}^2 + 3.93 \log K_{OW} - 5.33$ ($r^2 = 0.774$); Arctic cod: $\log BAF = -0.203 \log K_{OW}^2 + 3.45 \log K_{OW} - 4.65$ ($r^2 = 0.834$); Male beluga: $\log BAF = -0.286 \log K_{OA}^2 + 5.66 \log K_{OA} - 16.6$ ($r^2 = 0.818$); Male ringed seals: $\log BAF = -0.142 \log K_{OA}^2 + 3.54 \log K_{OA} - 9.55$ ($r^2 = 0.918$) and eider ducks: $\log BAF = -0.13 \log K_{OA}^2 + 3.77 \log K_{OA} - 13.0$ ($r^2 = 0.854$).


(a) WATER-VENTILATING ORGANISMS
(b) AIR-BREATHING ORGANISMS
Log BAF (lipid corrected)
Bivalves
Sculpin
Cod
Macro-algae
Lipid-water equilibrium
Male Ringed Seal
Male Beluga
Eider Duck
Lichens
Lipid-air equilibrium
Log K_{OW}
Log K_{OA}

CHAPTER 5

BIOTRANSFORMATION AND TROPHIC DILUTION OF DIALKYL PHTHALATE ESTERS IN A CANADIAN ARCTIC MARINE FOOD WEB

5.1 Introduction

Dialkyl phthalate esters (DPEs) are branched alkyl esters produced from the esterification of phthalic acid (1,2-Benzenecarboxylic Acid) and are interchangeably referred to as phthalate esters or phthalates. Phthalates have been manufactured since the early 1900s and used extensively as plasticizers in various industrial and consumer products. An important commercial use of phthalates has been as plasticizers in flexible polyvinyl chloride (PVC) such as medical tubing and vinyl floor tiles, however these compounds are also used in lubricating oils, paints, photographic film and cosmetics (*202*). These compounds are high production volume (HPV) chemicals with current global total phthalate production levels at approximately 4.3 million tonnes/year (*203*). Phthalate levels in indoor air of household and office buildings can be relatively high (50 – 4800 $ng \cdot m^3$) due to the high frequency of phthalate containing consumer products (*204*). Previous works in North America and Europe and Asia have reported phthalate concentrations of around 500-1,000 $pg \cdot m^{-3}$ in the atmosphere (*205,206*) and part per million (ppm) levels in environmental samples such as sediments and surface waters (*12,207,208,209*), suggesting these HPV substances are ubiquitous and relatively stable in the global environment. There is evidence suggesting some phthalates such as di-ethylhexyl phthalate (DEHP) and di-n-butyl phthalate (DBP) may be endocrine disrupting chemicals (EDCs), which can cause reproductive and developmental effects at high exposure levels (50-300 mg/kg BW/day) (*210,211*). Consequently, public concern over the environmental and human impacts of these compounds has increased significantly in recent years. Regulatory authorities and industry representatives have recently initiated evaluative reviews of phthalate esters for Persistence (P), Bioaccumulation potential (B) and Toxicity (T) under the United Nations Long Range Transboundary Air Pollution Protocol (LRTAP) for identifying Persistent Organic Pollutants (POPs).

Phthalates vary in alkyl chain length and branching and span a wide range of physical chemical properties. The majority of commercially marketed phthalates are produced as individual compounds (e.g., dimethyl phthalate or DMP). However, phthalates with alkyl chain lengths greater than 6 carbon atoms are also manufactured as complex isomeric mixtures (e.g., C6, C7, C8, C9 and C10 are available as phthalate isomeric mixtures). DPEs are commonly subdivided into three molecular weight (MW) categories. Group I: Low MW phthalates, esterified with alcohols having straight-chain carbon backbones of $\leq$ C3 include di-methyl phthalate (DMP) and diethyl phthalate (DEP). Group II: Transitional MW phthalates esterified with alcohols having straight-chain carbon backbones of C4-C6 including di n butyl phthalate (DBP), butyl benzyl phthalate (BBP) di-ethylhexyl phthalate (DEHP), di-iso-hexyl (C6) and di-iso-heptyl (C7) isomeric mixtures. Group III: High MW phthalates esterified with alcohols having straight-chain carbon backbones of > C7 including di-*n* octyl phthalate (DnOP) di-*n*-onyl phthalate (DnNP) and also di-iso-octyl (C8), di-iso-nonyl (C9) and di-iso-decyl (C10) isomeric mixtures. Molecular weights range from 194 g mol^{-1} for di-methyl phthalate (DMP) to 530 g mol^{-1} for Di-tridecyl phthalate (DTDP). Phthalates range widely in polarity (i.e., K_{OW}'s range from $10^{1.6}$-$10^{12.1}$for DMP to DTDP) and volatility (i.e., K_{OA}'s range from $10^{7.01}$ – $10^{13.1}$ for DMP to DTDP), indicating large differences in environmental partitioning and bioaccumulation behaviour for this group of compounds. Table 5.1 lists and summarizes several physical-chemical properties including molecular weights (MW, g mol^{-1}), log octanol water partition coefficient log K_{OW}, log octanol-air partition coefficient log K_{OA}, Henry's Law Constants (*H,* Pa m3 mol^{-1}) and water solubility (C_WSoL, ng·L^{-1}) for DPEs and their de-esterified monoester metabolites, i.e., monoalkyl phthalate esters (MPEs). Many of the group II and III DPEs (i.e., transitional and high MW congeners, > 350 g·mol^{-1}) such as di-ethyl hexyl phthalate (DEHP), di-n-octyl (DnOP) and di-n-nonyl (DnNP) phthalate exhibit similar physical-chemical properties as PCBs and other legacy POPs and hence may be susceptible to long-range transport, biomagnification and accumulation in Arctic ecosystems and are tentatively classified as "candidate" POPs.

However, laboratory investigations using fish show bioconcentration factors (i.e., BCFs = concentration in organism ÷ concentration in water) of several phthalates were lower than expected based on K_{OW} values (*202,212,213*). It has been hypothesized that DPEs undergo enzymatic biotransformation in the intestinal tract and/or tissues of organisms and likely do not biomagnify in food chains (*202*). A recent field-survey of DPE concentrations in organisms from Canada's west coast reported phthalate ester concentrations ranging from approximately 2.17 ng·g^{-1} lipid for di-*n*-nonly phthalate (DnNP) in dogfish to 28,700 ng·g^{-1} lipid for C8 isomers in

plankton (*13*). The potentially bioaccumulative "high" K_{OW} DPEs (i.e., Group II and III DPEs: di-(2-ethylhexyl), di-n-octyl, di-n-nonyl, C8, C9, and C10), did not biomagnify as lipid normalized concentrations significantly declined with increasing trophic position and stable isotope ratios ($\delta^{15}N$). Food -web magnification factors (FWMFs) of DPEs in this Pacific coastal marine food web were low, ranging between 0.25 and 0.48. In contrast, PCBs measured in the same food web exhibited significant biomagnification potential with FWMFs ranging between 2 to 9 (*13*).

Previous studies of DPEs indicate these compounds can be metabolized *in vivo* by hydrolytic de-esterification to monoalkyl phthalate esters (MPEs), (*214,215,216,217*) resulting in trophic dilution in food webs (*12,13*). However, analysis of their bioaccumulation in Arctic biota remains important because of their mega tonne production volumes and similar physical-chemical properties to other POPs. Moreover, there remains increasing concerns regarding the potential toxicological impacts of the primary de-esterified metabolites, i.e., monoalkyl phthalate esters (MPEs) on organism reproduction and neurodevelopment (*218*). Nomenclature and physical chemical properties of several MPEs are shown in Table 5.1 with corresponding diester parent compounds and include mono-methyl (MMP), mono-ethyl (MEP), mono-butyl (MBuP), mono-butyl-benzyl (MBzP), mono-2-ethylhexyl (MEHP), mono-*n*-octyl (MnOP) and monoesters of isomeric diester mixtures (i.e., MoC6, MoC7, MoC9, MoC10). DPE metabolism is viewed as a multi phase process (*219,220,221*). Appendix 13 illustrates this metabolic pathway of a diester (DEHP). Following dietary exposure, Phase I biotransformation or de-esterification of DEHP can occur *via* hydrolysis in the upper gastro-intestinal tract (GIT) by pancreatic lipases where the monoester (MEHP) and corresponding alcohol 2-ethylhexanol (2-EH) are formed. Absorbed parent DEHP can also be metabolized in liver or blood of the organism by enzymatic lipases (*222*). Phase II biotransformation involves the glucuronidation (reaction of free monoester with glucuroic acid) of MEHP resulting in a MEHP-glucuronide conjugate. The glucuronidation of the free monoester is believed to increase the water solubility and hence enhance urinary excretion of the metabolite in animals. Alternatively, absorbed MEHP can be excreted in urine unaltered or further metabolized in the liver to produce even more hydrophilic oxidative products (*via* ω, ω -1, ω -2 oxidation), (*223*). There is some evidence that free mono alkyl phthalates of several diesters may actually be culpable of the observed reproductive and developmental impacts in laboratory animals (*224,225,226,227*). The available data indicate that monoester phthalates are likely bioreactive molecules *in vivo* and may have potential to cause toxicological effects in organisms. However, there is currently a large information gap regarding exposure levels and bioaccumulation behaviour of MPEs in organisms and food webs.

In this paper, we present the findings from a field study, involving the analysis of several dialkyl phthalate esters and monoester metabolites and polychlorinated biphenyls (PCBs) in various organisms of a sub-arctic coastal marine food web. The study involved the analysis of eight individual diesters, DMP, DEP, DiBP, DBP, BBP, DEHP, DnOP, DnNP, five isomeric DPE mixtures (C6, C7, C8, C9 and C10), ten free form monoester phthalates (i.e., non conjugated), (MMP, MEP, MBuP, MBzP, MEHP, MnOP, MoC6, MoC7, MoC9, MoC10) and several PCB congeners in samples of plankton and macro-algae, various fish species and marine mammals. K_{OW}'s of the selected PCB congeners were within the range of the DPE K_{OW}'s and varied from $10^{5.24}$ for PCB-18 to $10^{8.18}$ for PCB-209. The objective of the study are threefold: (i) compare bioaccumulation behaviour of DPEs with that of know POPs such as recalcitrant PCB congeners (e.g., PCB 153, 180 etc.) by assessing lipid equivalent concentrations of those compounds in organisms of various trophic levels, (ii) determine presence and levels of monoester phthalates (MPEs) in livers of beluga whale (i.e., primary DPE metabolites) and (iii) compare DPE, MPE and PCBs concentrations at our relatively remote sub-arctic field site in north eastern Canada to levels reported in more urbanized locations. To our knowledge, these are the first reported measurements of phthalate esters in the Arctic environment.

5.2 Materials and Methods

5.2.1 Sample collections.

During the months of May to August between 1999 and 2003 various biological samples were collected along the eastern Hudson Bay coastline in close proximity to the Inuit village Umiujaq (64° 15′N 113° 07′ W), (Figure 5.1). For details see *Chapter 1, Section 1.9.1* and Appendix 1, which summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex, age and condition.

5.2.2 Food web characterization and designation of organism trophic positions.

Appendix 14 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been established by extensive ^{15}N and ^{13}C isotope enrichment analyses involving

numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of TL = 1 + (δ^{15}N -5.4)/3.8. More recent studies using δ^{15}N measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 (see *Chapter 1*) summarizes these previous δ^{15}N measurements and TL ranges for the various organisms within these Arctic marine food webs. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 4.5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific δ^{15}N measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

5.2.3 Extraction, cleanup and analysis of DPEs.

The method used for co-extraction and cleanup of DPEs and PCBs in sediments and biota samples have been recently published elsewhere (*12*). Briefly, approximately 2 g of sediment or 5 g of biota sample was weighed, spiked with the suite of deuterated and^{13}C-labeled surrogate internal standards, including approx. 50-100 ng of each d_4-DMP, d_4-DBP and d_4-DnOP for diester phthalates and approx. 2,000-5,000 pg of each of ^{13}C PCB congeners 28, 52, 101, 128, 156, 180, 194, 206, 209 for PCBs, then blended with 15 to 20 g of pre-baked Na_2SO_4, and ground with mortar and pestle to a free-flowing powder. Sub-samples of other tissue samples (e.g., seaduck and marine mammal tissue samples) were excised from the interior of frozen samples to reduce potential contact contamination during collection and/or storage., then extracted by ultrasonic

solvent extraction with 50 mL of 1:1 (v/v) DCM/Hex using a Branson 5210 ultrasonic water-bath (Branson Ultrasonics Co., CT) for 10 min, and shaken on a shaker table (Eberbach Co., MI) also for 10 min. Once the suspended particles settled, the supernatant was removed. The extraction was repeated two more times with fresh solvent. The combined extracts were concentrated to ~ 5 mL with a gentle stream of high-purity nitrogen. The concentrate was quantitatively transferred onto a 350 mm x 10 mm i.d. glass column packed with 15 g deactivated alumina (15% HPLC water, w/w) and capped with 1-2 cm of anhydrous Na_2SO_4. To prepare samples for GC/MS analysis, the alumina column was eluted with three 30 ml fractions of (1) hexane; (2) 1:9 DCM/Hex; (3) 1:1 DCM/Hex. The third fraction (1:1 DCM/Hex fraction) was evaporated to approximately 100 µL and spiked with isotope-labelled surrogate performance standards (d_4-DEP and d_4-BBP) before GC/MS analysis.

For DPEs, low resolution gas chromatography LRGC/MS analyses were carried out on a Finnigan Voyager GC/MS system (Manchester, UK) which consisted of a Finnigan 8000 Series gas chromatograph, a Finnigan Voyager quadrupole mass spectrometer (1000 amu mass range) and a CTC A200S autosampler. Instrument control, data acquisition and data processing were performed using the Finnigan Masslab software. The mass spectrometer was operated in the positive EI mode with an electron energy of 70 eV. Data were acquired in the selective ion monitoring mode (SIM, m/z 149 for all phthalates except 163 for DMP) with a dwell time of 100 ms and a delay time of 10 ms. A J&W DB-5 fused silica capillary column (30 m x 0.25 mm, 0.25 µm film thickness) was used for separation. Splitless injections of 1 µL sample extract and 0.5 µL of air were made and the carrier gas used was Helium at a flow rate of 1 mL/min. The GC temperature program was: 70 °C (hold 1 min) to 180 at 12 °C/min, to 240 °C at 5 °C/min and to 300 °C at 5 °C/ min (hold 10 min). The injection port was at 260 °C, the GC/MS interface at 250 °C and the ion source at 200 °C. Criteria for identification and quantification by the isotope dilution method, along with quality control measures undertaken during GC/MS analysis, are detailed elsewhere (*12*). Two procedural blanks, consisting of Na_2SO_4, were extracted according to the same procedure as samples and analyzed with every batch of 10 samples to check for contamination of the extracts.

5.2.4 Extraction, cleanup and analysis of MPEs.

Approximately 5 g tissue samples were dried with approximately 20g of Na_2SO_4 using mortal and pestle, spiked with 50uL of MBP-d_4 (12ppm) and MEHP-d_4 (12ppm) internal surrogate standards

and extracted in 20 mL 1:1 DCM: Acetone by ultrasonic solvent extraction, 3 times 15 minutes each. The extract was evaporated to dryness, resuspended in 1 mL of CH_3CN with 5-6 ml of the acidic buffer prior to solid phase microextraction (SPE). The SPE Oasis cartridge (6cc, 500mg) was prepared by washing with CH_3CN (5.0 mL) followed by water (5.0 mL) and acidic buffer (10.0 mL). The sample was loaded onto the cartridge, then wash the cartridge with an additional 10ml of acidic buffer, then 20 mL of water (discard). The analytes were then eluted from the washed cartridge with CH_3CN (5mL) followed by EtOAc (5 mL). Any remaining water was removed with Na_2SO_4 and the extract was transferred to a centrifuge tube with CH_3CN. The eluate was evaporated to dryness under a stream of dry nitrogen and reconstituted in approximately 5mL of 1:1 DCM:Hexane prior to GPC. GPC column was loaded and eluted with 140mL of 1:1 DCM:Hexane (discarded) and then further eluted with 300mL of 1:1 DCM:Hexane which is collected in a 500mL flat bottom flask. Samples are rotary evaporated to approx. 1mL and transferred to centrifuge tubes and evaporated to dryness and resuspended in about 100 uL of CH_3OH and spiked with MPE performance standard 50uL MiNP-d_4 (12ppm) prior to LC/MS. LC/MS for MPEs was conducted using a Luna-column 5u Phenyl-hexyl 250x1 mm at a flow rate of 0.05 ml/min, Gradient from 100%A (A =5% CH3CN , 95%H2O, 1 mM NH4OAc) to 100%B (B= 90% CH3CN, 10% H2O, 1 mM NH4OH)within 5 minute, 18 minutes at 100%B, then back to 100%A within 5 minutes. The MS conditions were electrospray ionisation, negative ionisation mode, 120 degrees C - source temperature, Capillary voltage 3.99kV, Cone voltage 22V. The MS was operated in the SIM mode monitoring m/z 193 for MEP, m/z 249 for MoC6, m/z 255 for MBzP, m/z 263 for MoC7, m/z 277 for MEHP and MnOP, m/z 291 for MoC9, m/z 305 for MoC10.

5.2.5 Data treatment/compilation and statistics.

To enable direct comparisons of contaminant burdens between different environmental media and organisms it is important to correct chemical concentration data to a common unit expression such as lipid equivalent concentrations. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~1 - 98%), wet weight chemical concentrations (C, $ng \cdot g^{-1}$ ww) were expressed on a lipid weight basis by the equation: $C_L = C$ ww. $\div \phi L$ in units of $ng \cdot g^{-1}$ lipid. For some biological matrices with very low lipid fractions ($\phi L < 1\%$), such as vegetation and algae tend to solubilize organic contaminants in non-lipid biomolecules (i.e., non-lipid organic matter, NLOM) rather than in extractable lipids (*13 57,58,59*). Thus, for macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and

NLOM (ϕ_{NL}) fractions following the equation: $\phi Leq = \phi L + 0.035\phi_{NL}$, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42,44*). Because chemical concentrations exhibited log-normal distributions and were hence transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for POPs in the various organisms collected and analyzed as part of the present study (i.e., lichens, macro-algae, bivalves, fish, beluga whales and ringed seals).

5.2.6 Evaluative parameters for assessing chemical bioaccumulation potential.

See *Chapter 1, Section 1.9.5*

5.3 Results and Discussion

5.3.1 Data qualification.

Concentrations of dialkyl phthalate esters (DPEs) and mono alkyl phthalates (MPEs) in E. Hudson Bay sediments and biota are summarized in Appendix 15, and shows geometric means ± 95% confidence limits, along with corresponding lipid, lipid equivalent, moisture and organic carbon contents. Procedural blanks for PCBs and OC pesticides were generally low or non-detectable and method detection limits (MDLs) were then determined as the instrument limit of quantification (LOQ) on the HRMS. Low-resolution mass spectrometry (LRMS) was used for analysis of DPEs because of relatively strong peak response on the MS following sample cleanup and purification. However, because background contamination during extraction and cleanup can be extensive (even in laboratories used for ultra-trace residue analysis), procedural blanks play an essential role in accurate DPE analysis. Extensive glassware cleaning protocol, solvent distillation and micro-scale solvent extractions and elutions were employed to minimize phthalate MDLs (calculated as the mean + 3 standard deviations of simultaneously extracted procedural blank levels). DPE concentrations (shown in Appendix 15) were corrected for procedural blank contamination (i.e., blank subtracted), however the relative contribution of analyte originating background contamination versus sample matrix was typically low (~0-25% of sample amount). Overall, a vast majority (> 95%) of samples analyzed for DPEs were above analyte MDLs. While great care was taken to avoid phthalate contamination in this current study (e.g., no use of plastics), it is conceiveable that DPE levels observed in these Arctic samples may be the result during transport, storage or sample preparation because of the extremely high levels of DPEs

present in indoor air and dust (*204*). This may be particularly important for DEHP, the most extensively used plasticizer in commercial products.

5.3.2 Levels and congener profiles of dialkyl phthalate esters in E. Hudson Bay food web.

In general, DPE levels in E. Hudson Bay sediment and biota (Appendix 15) were orders of magnitude higher than PCBs (Appendix 5) and OC pesticides (Appendix 6). Figure 5.2 illustrates the relative congener contribution (i.e., % composition) for (a) dialkyl phthalate ester congeners and (b) total DPEs versus other organochlorines (i.e., PCBs and OC pesticides) observed in E. Hudson Bay sediment and biota. In these plots, contaminant burden profiles shown for lichens (collected on land in close proximity to marine sampling locations) can be viewed as an atmospheric "signal" resulting from air-borne contaminant exposure processes. Similarly, contaminant profiles shown for sediments and macro-algae represent an aquatic "signal" of water-borne chemical in the marine system, while those profiles for biota are indicative of food web bioaccumulation processes and subsequent chemical residue distributions in organism tissues'. Figure 5.2a illustrates the relative congener contribution (i.e., % composition) for dialkyl phthalate ester congeners in E. Hudson Bay lichens ("atmospheric signal"), sediments and macro-algae ("aquatic signal") and biota (food web profile). The predominant DPEs in biota were diethyl phthalate (DEP), di-n-butyl phthalate (DBP) and di (2-ethylhexyl) phthalate (DEHP). For macro-algae and fish species (e.g., cod and sculpin), relative DPE tissue contributions were ordered DEHP (88%) > DBP (6%) > DEP (2%). DEHP concentrations were approximately 8,400 $ng \cdot g^{-1}$ lipid in Arctic cod and 12,200 $ng \cdot g^{-1}$ lipid in sculpin. DBP concentrations were slightly lower at 1,810 $ng \cdot g^{-1}$ and 1,240 $ng \cdot g^{-1}$ lipid in cod and sculpin, respectively. For beluga whales, the composition rank order was DBP (45%) > DEHP (20%) > DEP (15%). Lipid normalized concentrations of DBP and DEHP in male beluga livers were approximately 2,100 $ng \cdot g^{-1}$ lipid for DBP and 914 $ng \cdot g^{-1}$ lipid for DEHP. $\sum$DPEs exhibit nearly 100% of the atmospheric and aquatic "signals" in relation to other OCPs and PCBs. Similarly, high DPE levels observed in E. Hudson Bay fish (cod and sculpin) result in tissue burdens > 95% of the total DPE + organochlorine accumulation levels. For beluga whales, $\sum$DPE burdens are ~28% of the total DPE + organochlorine levels, which is comparable to beluga whale tissue burdens for $\sum$PCBs (40.8%) and $\sum$DDTs (21.3%).

While there are substantially more individual compounds comprising $\sum$organochlorines (i.e., 209 PCBs congeners + HCHs, + CBz + cyclodienes etc) compared to only eight DPE congeners that

constitute ∑dialkyl phthalate esters, tissue burdens of ∑DPEs in organisms of the E. Hudson Bay food web are generally greater than ∑organochlorines (Figure 5.2b). Specifically, ∑DPEs exhibit nearly 100% of the atmospheric and aquatic "signals" in relation to other OCPs and PCBs. Figure 5.2b also shows that relatively high DPE levels observed in E. Hudson Bay fish (cod and sculpin) result in tissue burdens > 95% of the total DPE + organochlorine accumulation levels. For beluga whales, ∑DPE burdens are ~28% of the total DPE + organochlorine levels, which is comparable to beluga whale tissue burdens for ∑PCBs (40.8%) and ∑DDTs (21.3%).

5.3.3 Levels of monoalkyl phthalate esters (MPEs).

Concentrations ($ng \cdot g^{-1}$ wet weight) of monoalkyl phthalate esters (MPEs) along with DPEs in stomach contents and liver tissue of Beluga whales are shown in Table 5.2. Wet weight concentrations are used here to compare tissue burdens of DPEs and MPEs in biota samples because of the highly polar nature of monoester phthalates and hence their likely sorption to aqueous rather than lipid fractions of organism tissues'. The monoesters metabolites of DBP (i.e., monobutyl phthalate, MBP), and DEHP (i.e., mono-ethylhexyl phthalate, MEHP) were the only MPEs detected in biota samples. Concentrations of MBP in stomach contents and liver were approximately 13.7 and 22.5 $ng \cdot g^{-1}$ wet wt., respectively. Concentrations of MEHP in stomach and liver were approximately 10.8 and 33.5 $ng \cdot g^{-1}$ wet wt., respectively. In general, diester phthalate concentrations were slightly higher than the corresponding monoester metabolite. There were no significant differences ($p < 0.05$) between individual compound concentrations in stomach and liver samples. In beluga livers, DBP concentrations were significantly higher ($p < 0.05$) than MBP concentrations, while DEHP concentrations in liver were approximately equal to MEHP concentrations in liver. Specifically, the DBP/MBP ratio of 3.77 and DEHP/MEHP ratio of 0.96 were observed in beluga liver. The data indicate that diester phthalates are transformed to monoesters in beluga whales. Also, the site of diester metabolism appears to be in the intestinal tract because the monoesters were detected at appreciable levels in the stomach contents. Relatively high concentrations of MBP and MEHP in beluga whale liver suggest these metabolic transformation products of the parent DPEs (DBP and DEHP) may accumulate and persist in the tissues' of these animals.

5.3.4 Spatial Trends of DPE levels in biota.

While numerous studies of phthalate ester toxicokinetics in laboratory animals have been conducted (*212,215,219,228,229*), few data regarding levels and bioaccumulation behaviour of DPEs in fish and wildlife exist. The DPE concentrations reported in the E. Hudson Bay food web in the present study are comparable to DPE levels we have recently reported in a marine biota from the west coast of Canada (*12,13*). For example, DEP, DBP and DEHP concentrations in Pacific staghorn sculpin (*Leptocottus armatus*) sampled in 1998 near Vancouver, Canada were approximately 490, 2,450 and 3,720 $ng \cdot g^{-1}$ lipid. The fact that we observed comparable levels and accumulation profiles of DPEs in a Pacific urbanized coastal food web and eastern Arctic marine food webs suggests atmospheric levels and patterns of DPEs may be relatively uniform across large geospatial ranges. Figure 5.3, showing reported concentrations of DEHP in fish, seabirds and mammals from North America and Europe during the 1980s and 1990s (see references *230,231,232,233*), indicates DEHP levels measured in biota from the United States, Canada, and western Europe are comparable (~1,500- 10,000 $ng \cdot g^{-1}$). Thus, accumulation of DPEs in organisms and food webs may be more influenced by long-range transport and global circulation of DPEs rather than local point sources of DPE contamination.

5.3.5 Concentration relationships with trophic level and FWMFs.

Results from log-linear regressions analyses of organism chemical concentrations (C_B) and trophic level (TL) and corresponding food web magnification factors (FWMFs) for (i) water-ventilating ectotherms, (ii) air-breathing endotherms and (iii) the overall food are summarized in Appendix 16. Strong positive C_B-TL relationships in both organism groups were observed for the highly chlorinated (Cl_6-Cl_9) recalcitrant PCBs (Group I and II congeners) such as Cl_6-CB138, Cl_7-CB180 and Cl_6-CB153. Figure 5.4 shows C_B-TL regression lines for water-ventilating ectotherms, air-breathing endotherms and the overall food web together with observed concentrations of PCB153 (4a), DMP (4b), BBP (4c), DEHP (4d) and DiBP (4e). For CB153, estimated slopes were approximately 1.05, 1.04 and 0.84 for water-ventilating ectotherms, air-breathing endotherms and the overall food web, respectively. This equates to FWMFs of CB153 of approximately 6.84 for water-ventilating ectotherms, 11.33 for air-breathing endotherms and 11.02 for the overall food web respectively. In contrast to PCB-TL relationships, lipid equivalent concentrations of phthalate esters DMP, BBP, DEHP and DiBP, did not change significantly with increasing trophic level (Figure 5.4b-e). For example, FWMFs for DMP, BBP, DEHP and DiBP

over the entire food web were 0.84. 0.87, 0.80 and 0.85, respectively. Using concentration data only for aquatic organisms, those FWMFs were slightly higher at 1.95, 1.32, 1.12 and 1.25 for DMP, BBP, DEHP and DiBP, respectively. This is due to slightly higher DPE levels in fish compared macro-algae. Higher DPE levels in fish compared to macro-algae (assumed to be in equilibrium with surface seawater concentrations) indicate fish species may undergo a small DPE concentration amplification due to inefficient metabolic transformation of diesters within those species. In general however, the data indicate that dialkyl phthalate esters do not biomagnify in this marine food web (i.e., FWMFs ~ unity), compared to extensive biomagnification observed for PCBs (e.g., CB153 FWMF > 11).

5.3.6 Estimated Air (C_{AG}) and Seawater (C_{WD}) Concentrations and BAFs.

Table 5.3 shows (i) physical chemical properties, log K_{OW}, log K_{OA}, water solubility, vapour pressures (ii) estimated air and seawater concentrations of using an equilibrium partitioning model and calculated chemical concentrations in lichens and macro-algae, respectively and (iii) calculated bioaccumulation factors (log BAFs) in various species of the E. Hudson Bay food web for the eight dialkyl phthalate esters and several PCB congeners. In general, estimated air and seawater concentrations of DPEs and PCBs are shown to decline for the more hydrophobic and less volatile compounds (i.e., low water solubility/low volatility). Estimated air concentrations for DPEs ranged from ~0.5 -2,000 $pg \cdot m^{-3}$, while estimated seawater concentrations of DPEs ranged from~ 3.4×10^{-4} to 1,000 $ng \cdot L^{-1}$. Among the eight diesters, DBP (with a log K_{OW} = 4.27) exhibited the highest estimated concentrations in both air (~ 2,190 $pg \cdot m^{-3}$) and seawater (875 $ng \cdot L^{-1}$). DEHP, which exhibits similar hydrophobicity to Cl_7-PCBs (e.g., logK_{OW} DEHP = 7.7 and log K_{OW} Cl_7-CB180 = 7.5), was estimated to have concentrations of approximately 88.0 $pg \cdot m^{-3}$and 0.11 $ng \cdot L^{-1}$ in air and seawater, respectively. Atmospheric and surface water concentrations of DEHP near the Great Lakes during the 1980s ranged between ~500 and 5,000 $pg \cdot m^{-3}$ in air and ~30-300 $ng \cdot L^{-1}$ in freshwater (*205*). A recent study of phthalates in indoor air (*204*) has reported mean gas-phase DEHP levels in homes ranged from approximately 59 to 1,000,000 $pg \cdot m^{-3}$ (median value of 77,000 $pg \cdot m^{-3}$). This relatively high concentration of DEHP in indoor air (i.e., ~80,000 $pg \cdot m^{-3}$) is approximately 1,000 times higher than our estimated air concentrations from a remote marine location in the Canadian Arctic (~88 $pg \cdot m^{-3}$). However, our estimated concentrations of phthalates in Arctic air and seawater appear to be orders of magnitude higher than other common organochlorine contaminants that accumulate in the Arctic environment. For example, DEHP concentrations in air and seawater were approximately 4,000 – 9,000 times

greater than concentrations of Cl_7-CB180 in air (0.02 $pg \cdot m^{-3}$) and seawater (1.17×10^{-5} $ng \cdot L^{-1}$), respectively. High ambient environmental levels of dialkyl phthalate esters (compared to PCBs) is not unexpected because these substances are current-use megatonne commercial chemicals (*12,13*), whereas PCBs are globally discontinued substances and correspondingly have exhibited stabilized levels in the Arctic in recent years (*71*).

BAFs of dialkyl phthalate esters and PCB congeners are shown to increase with increasing chemical hydrophobicity/decreasing volatility. For example, log BAFs of phthalate esters (log K_{OW}'s ~1.61 to 8.6) relative to seawater in macro-algae, fish and male beluga whales ranged from approximately 1for DMP (log K_{OW}'s = 1.61) to 8 for DnNP (log K_{OW}'s = 8.6). Similarly, log BAFs for male beluga whales relative to air concentrations ranged from ~ 7 for DMP (log K_{OA} = 7.01) and ~11 for DnOP (log K_{OA} = 10.53). These data show that dialkyl phthalate ester BAFs relative to water (BAF = C_B/C_{WD}) and air (BAF = C_B/C_{AG}) are equivalent to those chemicals K_{OW} and K_{OA} and indicate DPEs in biota exhibit equilibrium concentrations as predicted by physical chemical properties. This equilibrium observation is also shown to occur for PCBs in macro-algae in seawater (i.e., macro-algae BAFs ~ K_{OW}'s). However, BAFs for PCB congeners in fish and male beluga whales are much higher than their K_{OW} and K_{OA}'s, indicating concentrations of those compounds in biota are elevated above equilibrium conditions with the surrounding environment. This is a common observation for PCBs in food webs, due to their extensive biomagnification potential and high metabolic resistances. Overall, the BAF data in Table 5.3 suggest that PCBs biomagnify in E. Hudson Bay biota, while dialkyl phthalate esters appear to bioaccumulate into organism tissues but are efficiently eliminated and/or transformed (e.g., hydrolysis) to a point where DPE concentrations in organisms attain a chemical equilibrium with ambient DPE levels in the environment. It should be noted that the presented BAFs reported for dialkyl phthalate esters should be utilized with caution due to the fact they were calculated using estimated air and seawater concentrations (based on equilibrium assumptions with measured lichens and macro-algae concentrations). In particular, the accuracy of the BAFs for the more hydrophobic and non-volatile DPEs (log K_{OW} > 8 and log K_{OA} > 9) may be substantially over-estimated because those compounds may exhibit kinetically limited uptake, rather than equilibrium conditions. This violation of the equilibrium assumption for these non-polar/non-volatile compounds (e.g., DEHP, DnOP, DnNP) would result in an underestimate of chemical air and seawater concentrations based on equilibrium partitioning (e.g., derived from K_{OA} and K_{OW}) and a subsequent error (overestimate) in the BAFs.

5.3.7 Biomagnification Potential of Dialkyl Phthalate Esters.

In a recent preceding study (*186*) we reported and evaluated biomagnification factors (BMFs) and elimination index (EIs) and biodilution factors (BDFs) of PCBs and OC pesticides in E. Hudson Bay organisms. Figure 5.5 illustrates EI values for selected Group I-V PCB congeners compared to dialkyl phthalate esters. EI values near zero indicate negligible metabolism, while EI values > 1 suggest substantial metabolism and low biomagnification potential. Recalcitrant PCBs (Group I and II congeners) tend to exhibit efficient accumulation, very slow kinetic elimination and are generally resistant to metabolic transformation. Consequently, those compounds typically exhibit the greatest BMFs in organisms (i.e., BMF_{MAX}). For example, the relatively low EI values of CB180 (i.e., EI ~ 0) correspond to a BMF of CB180 in male beluga whales (beluga/Arctic cod) of approximately 41.8. In comparison, all eight dialkyl phthalate ester congeners exhibited relatively high EI values (> 1.5) in male beluga whales, indicating substantial DPE biotransformation in those animals (Figure 5.5). These relatively high EI values of DPEs correspond to BMFs of those compounds in beluga whales (beluga/Arctic cod) generally equal to or less than unity, indicating no biomagnification. Thus, in comparison to recalcitrant PCBs (e.g., BMF CB180), dialkyl phthalate esters exhibit biodilution factors (BDFs) of approx. 40 (i.e., 40 times reduced from BMF_{MAX}). The reduced biomagnification potential and accumulation of DPEs in beluga tissues (i.e., "biodilution") is likely due to *in vivo* hydrolytic de-esterification of parent DPEs into their primary mono-alkyl phthalate ester metabolites, which has been previously observed in laboratory animals (*222,234,235,236*).

Figure 5.6 shows chemical biomagnification factors (BMFs) for PCBs, organochlorine pesticides (OCPs) and dialkyl phthalate esters (DPEs) in E. Hudson Bay beluga whales versus the chemical's octanol-air partition coefficient (K_{OA}). This plot shows substantially lower BMFs for the dialkyl phthalate esters in beluga whales (BMFs $\leq$ 1) compared to PCBs and OC pesticides (i.e., BMFs >50) that exhibit similar physical-chemical properties. The high degree of chemical biomagnification of these compounds such as Cl_5-to Cl_7-PCB congeners in air-breathing animals such as beluga whales is essentially the result of (i) very efficient assimilation through dietary exposure and (ii) very low metabolic transformation rates (k_M) *in vivo*. For example, dietary absorption efficiencies (E_D) are typically between 90 - 100% in birds and mammals (*172*) and chemical half-lives ($T_{1/2}$) of Group I PCBs such as CB153 in tissues' of organisms can exceed 1,000 days (*52*). For the dialkyl phthalate esters, dietary absorption (E_D) is also very efficient, but can result in metabolic transformation, with $T_{1/2}$ estimates on the order of hours (*237*).

Substantial biotransformation was observed for all eight dialkyl phthalate ester congeners in male beluga (i.e., EI > 1.5). BMFs of DPEs in beluga whales (beluga/cod) from eastern Hudson Bay were are all less than 1.0, indicating dialkyl phthalate esters do not biomagnify in the food web. The low biomagnification potential of DPEs in beluga tissues is likely the result of *in vivo* hydrolytic de-esterification of DPE congeners to their primary mono-alkyl phthalate ester metabolites. Although rapid metabolism of DPEs ultimately diminishes the bioaccumulation potential and hence concentrations in biota, the relatively high levels of these HPV commercial chemicals observed in the ambient environment (i.e., atmospheric and aquatic signals) nonetheless result in relatively high DPE tissue residue burdens in organisms of the E. Hudson Bay food web. We estimate the dietary exposure of DEHP to beluga whales via Arctic cod (8,400 $ng \cdot g^{-1}$ lipid) is approximately 0.0025 mg/kg/day, based on a 30 kg cod diet for beluga whales (see reference *114*). This is far lower than reported no-observable adverse effect levels for DEHP ($NOAEL_{DEHP}$, ~ 100 mg/kg/day). However, our observations regarding the formation of equal amounts of monoesters in tissues of beluga whales may be of toxicologically significant, especially as previous studies indicate these relatively more bio-active metabolites may be the key derivative associated with observed endocrine disruption and teratogenic effects of phthalate ester exposure in laboratory animals (*218*). Further investigation into the fate, toxicokinetics and toxicological significance of observed levels of monoester phthalates in marine organisms is required to fully evaluate the ecological risk posed by the extensive commercial use of dialkyl phthalate esters.

5.4 Acknowledgements.

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies and wish to thank Dr's Bill Doidge, Michael Kwan and Susan Sang, Derek Muir and Northern Quebec Inuit communities of Umiujaq and Inukjuaq for coordinating/aiding collection of field samples.

5.5 Tables

Table 5.1 Physical-chemical properties of dialkyl and monoalkyl phthalate esters.

Chemical Name	Formula	CAS #	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3 \cdot P1$)	MW ($g \cdot mol^{-1}$)	Le Bas Molar Volume ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure VPsL (Pa)
Dialkyl Phthalate Esters									
Di-methyl (DMP)	$C_{10}H_{10}O_4$	131-11-3	1.61	7.01	0.01	220	206.4	5.22×10^{9}	0.263
Di-ethyl (DEP)	$C_{12}H_{14}O_4$	84-66-2	2.54	7.55	0.03	266	254.0	5.91×10^{8}	0.065
Di-iso-butyl (DIBP)	$C_{16}H_{22}O_4$	84-69-5	4.27	8.54	0.13	278	342.8	9.90×10^{6}	0.001
Di-n-butyl (DBP)	$C_{16}H_{22}O_4$	84-74-2	4.27	8.54	0.13	287	342.8	9.90×10^{6}	0.001
Benzyl-butyl (BBP)	$C_{19}H_{20}O_4$	85-68-7	4.70	8.78	0.21	295	364.8	3.80×10^{6}	0.0025
Di-(2-ethyl hexyl) (DEHP)	$C_{24}H_{38}O_4$	117-81-7	7.73	10.53	3.95	358	520.4	2.49×10^{3}	0.00003
Di-n-octyl (DnOP)	$C_{24}H_{38}O_4$	11-78-4	7.73	10.53	3.95	395	520.4	2.49×10^{3}	0.00003
Di-n-nonyl (DnNP)	$C_{26}H_{42}O_4$	84-76-4	8.60	11.03	9.26	450	564.8	3.08×10^{2}	0.00001
Monoalkyl Phthalate Esters									
Mono-methyl (MMP)	$C_9O_4H_8$	4376-18-5	1.17	-	-	180	-	1.54×10^{10}	-
Mono-ethyl (MEP)	$C_{10}O_4H_{10}$	2306-33-4	1.67	-	-	194	-	4.86×10^{9}	-
Mono-butyl (MBuP)	$C_{12}O_4H_{14}$	131-70-4	2.53	-	-	222	-	6.50×10^{8}	-
Mono-Benzyl-butyl (MBzP)	$C_{15}O_4H_{12}$	2528-16-7	2.95	-	-	256	-	2.59×10^{8}	-
Mono-(2-ethyl hexyl) (MEHP)	$C_{16}O_4H_{22}$	4376-20-9	4.25	-	-	278	-	1.11×10^{7}	-
Mono-n-nonyl (MnOP)	$C_{16}O_4H_{22}$		4.25	-	-	278	-	1.11×10^{7}	-

Chemical Name	Formula	CAS #	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3 \cdot P1$)	MW ($g \cdot mol^{-1}$)	Le Bas Molar Volume ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure VPsL (Pa)
Mono-C9-iso-mix	$C_{17}O_4H_{24}$	-	4.68	-	-	292	-	3.99×10^6	-
Mono-C10-iso-mix	$C_{18}O_4H_{26}$	-	5.11	-	-	306	-	1.43×10^6	-

Table 5.2 Concentrations of DPEs and MPEs ($ng \cdot g^{-1}$ lipid) in beluga whale stomach contents and liver tissue and corresponding DPE/MPE ratios.

	Beluga Stomach Contents			Beluga Liver		
	GM	*(95% CL)*	DPE/ MPE	*GM*	*(95% CL)*	DPE/ MPE
Dialkyl Phthalates						
DMP	0.36	*0.069-1.85*	-	2.53	*0.79-8.14*	-
DEP	2.10	*0.60-7.32*	-	28.9	*8.21-102*	-
DIBP	0.82	*0.19-3.44*	-	6.47	*1.44-29.2*	-
DBP	5.90	*1.16-30.1*	**0.43**	84.9	*25.7-281*	**3.8**
BBP	1.56	*0.48-5.0*	-	14.9	*3.91-57.5*	-
DEHP	25.2	*7.21-88.2*	**2.3**	32.3	*3.92-265*	**0.96**
DnOP	0.39	*0.063-2.46*	-	2.71	*0.44-16.8*	-
DnNP	0.79	*0.12-5.16*	-	3.42	*0.44-26.6*	-
∑DPEs	34.8	*8.9-135*	**2.5**	207	*60.4-710*	**4.4**
Monoalkyl Phthalates						
MBP	13.7	*1.69-110*	-	22.5	*7.20-70.3*	-
MEHP	10.8	*2.12-55.4*	-	33.5	*5.24-215*	-
∑MPEs	13.8	*1.99-95.5*	-	47.1	*10.4-214*	-

Table 5.3 Logarithms of Bioaccumulation Factors (log BAFs) of DPEs and PCBs in macro-algae, fish and beluga whales from E. Hudson Bay, along with estimated gas-phase air concentrations (C_{AG}, pg m^{-3}) and freely dissolved surface seawater concentrations (C_{WD} ng·L^{-1}) and the chemical's K_{OW} and K_{OA}.

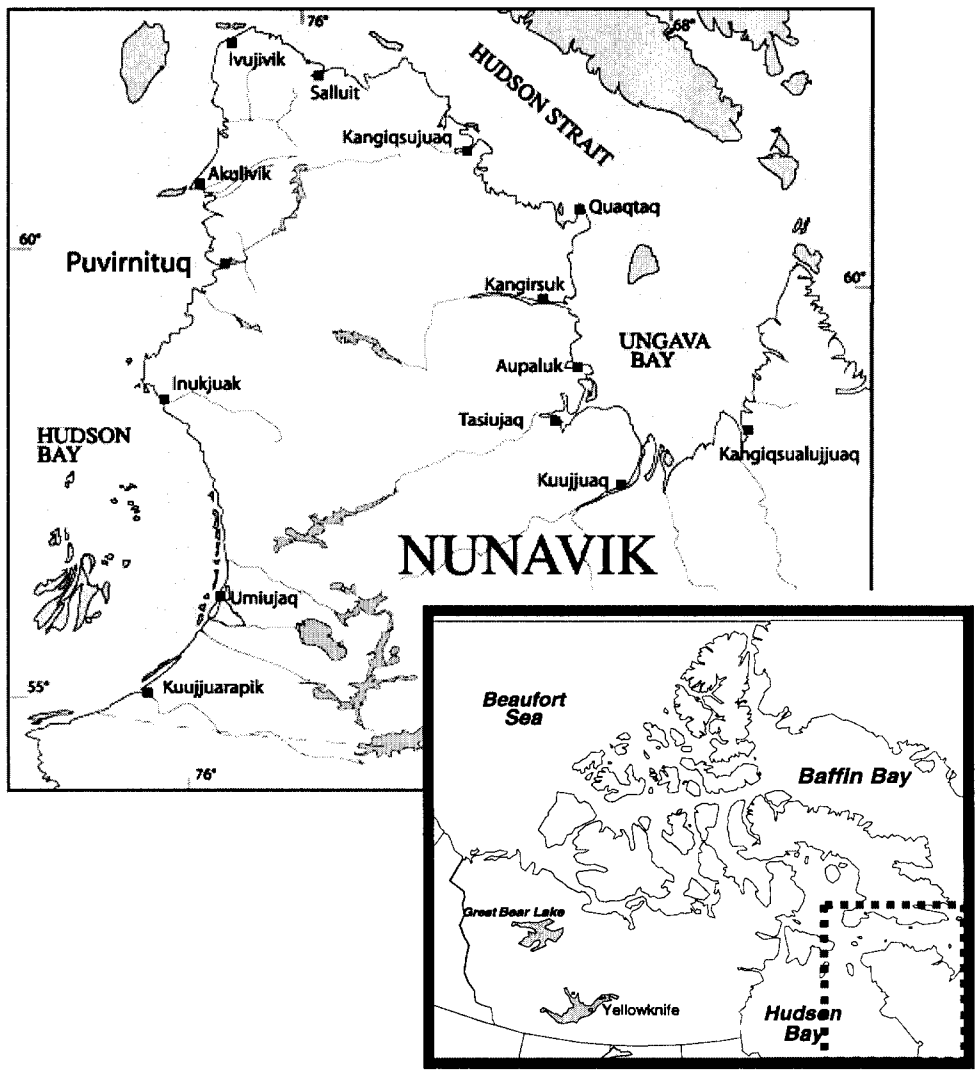
	Log K_{OW}	Log K_{OA}	C_{AG} (pg m^{-3})	C_{WD} (ng L^{-1})	Log BAF Macro-algae/ Water	Log BAF Cod/ Water	Log BAF Sculpin/ Water	Log BAF Male Beluga/ Water	Log BAF Male Beluga/ Air
DMP	1.61	7.01	475.65	2.99E+01	2.5	3.0	3.0	2.05	6.8
DEP	2.54	7.55	2192.33	2.52E+01	3.4	3.9	3.9	3.94	8.0
DIBP	4.27	8.54	102.70	1.74E-01	5.1	5.6	5.2	5.19	8.4
DBP	4.27	8.54	567.50	2.45E+00	5.1	5.3	5.1	4.66	8.3
BBP	4.70	8.78	56.05	1.92E-01	5.6	5.9	5.5	5.37	8.9
DEHP	7.73	10.53	87.95	3.15E-03	8.6	8.9	9.0	8.69	10.2
DnOP	7.73	10.53	0.96	4.83E-05	8.6	8.9	8.9	9.25	11.0
DnNP	8.60	11.03	0.41	1.01E-05	9.5	9.9	10.0	-	-

Table 5.3 continued.

	Log K_{OW}	Log K_{OA}	C_{AG} ($pg \cdot m^{-3}$)	C_{WD} ($ng \cdot L^{-1}$)	Log BAF Macro-algae/ Water	Log BAF Cod/ Water	Log BAF Sculpin/ Water	Log BAF Male Beluga/ Water	Log BAF Male Beluga/ Air
CB28	5.0	6.88	1.16	0.00018	6.6	6.7	6.7	7.3	9.4
CB118	6.74	8.24	0.53	0.00010	6.6	7.5	7.4	9.2	11.4
CB153	6.9	9.79	0.68	0.00017	5.9	7.8	7.8	9.4	11.8
CB138	6.83	10.02	0.34	0.00016	6.5	7.6	7.5	9.3	12.0
CB180	7.5	9.83	0.58	0.00003	6.4	7.9	7.9	9.5	11.2
CB187/182	7.2	10.57	0.40	-	-	-	-	12.1	11.4
CB194	7.8	11.24	0.07	-	-	-	-	12.0	11.2
CB206	8.1	11.36	0.05	-	-	-	-	11.9	10.7
CB209	8.4	11.88	0.06	-	-	-	-	12.0	10.3

5.6 Figures

Figure 5.1 Map showing general study area of E. Hudson Bay and various Nunavik Inuit communities of northern Quebec, Canada.



Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

Figure 5.2 % composition of DPEs in commercial phthalate usage and in environmental and biological samples from E. Hudson Bay.

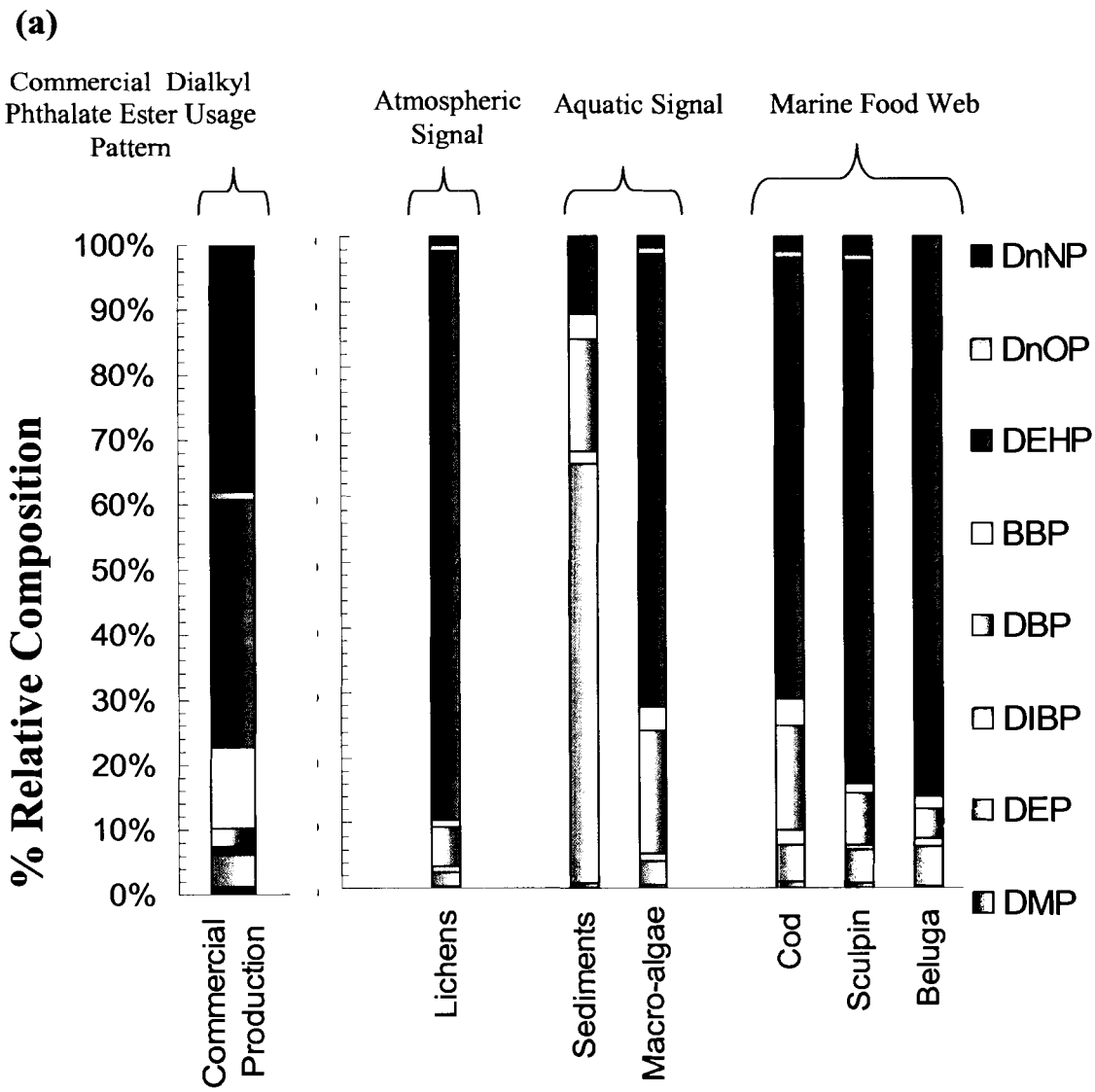


Figure 5.2 continued.

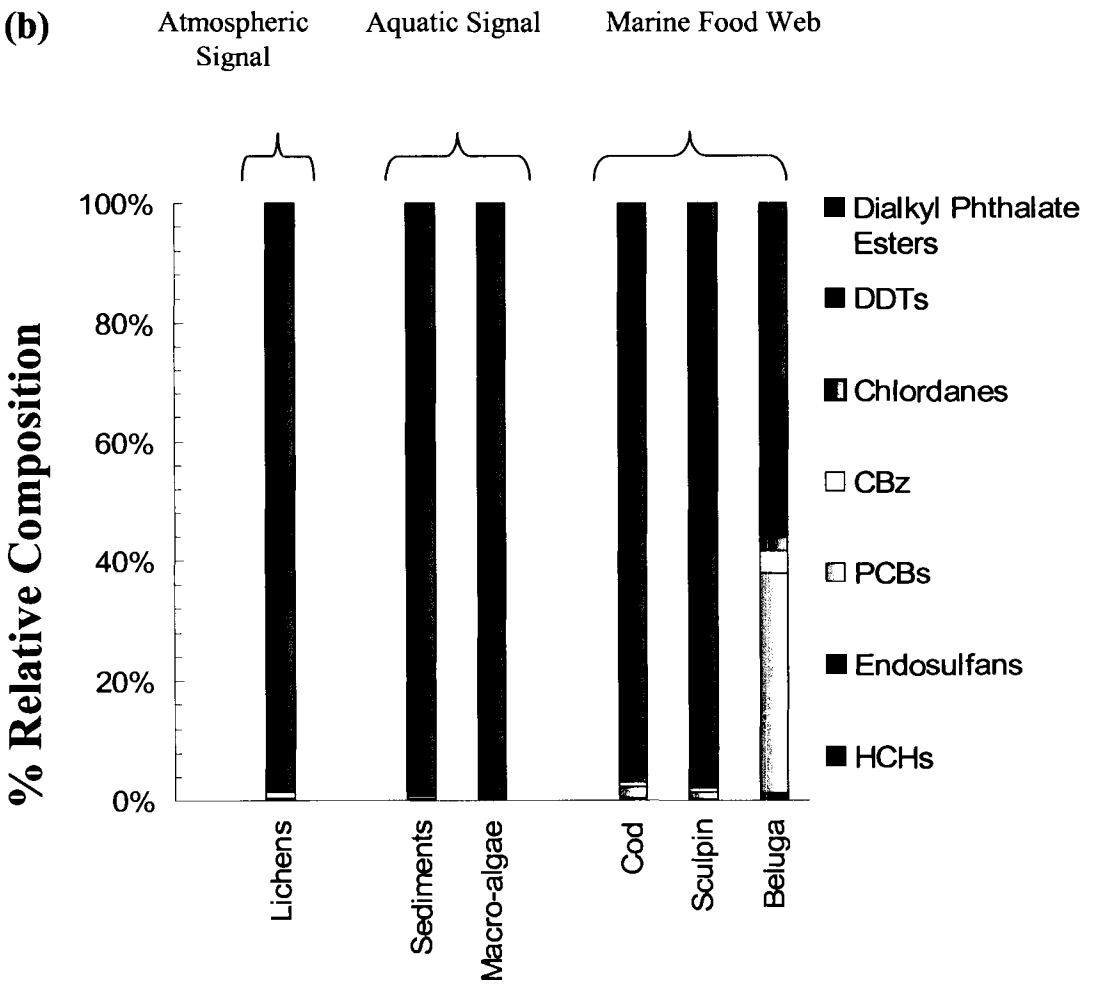
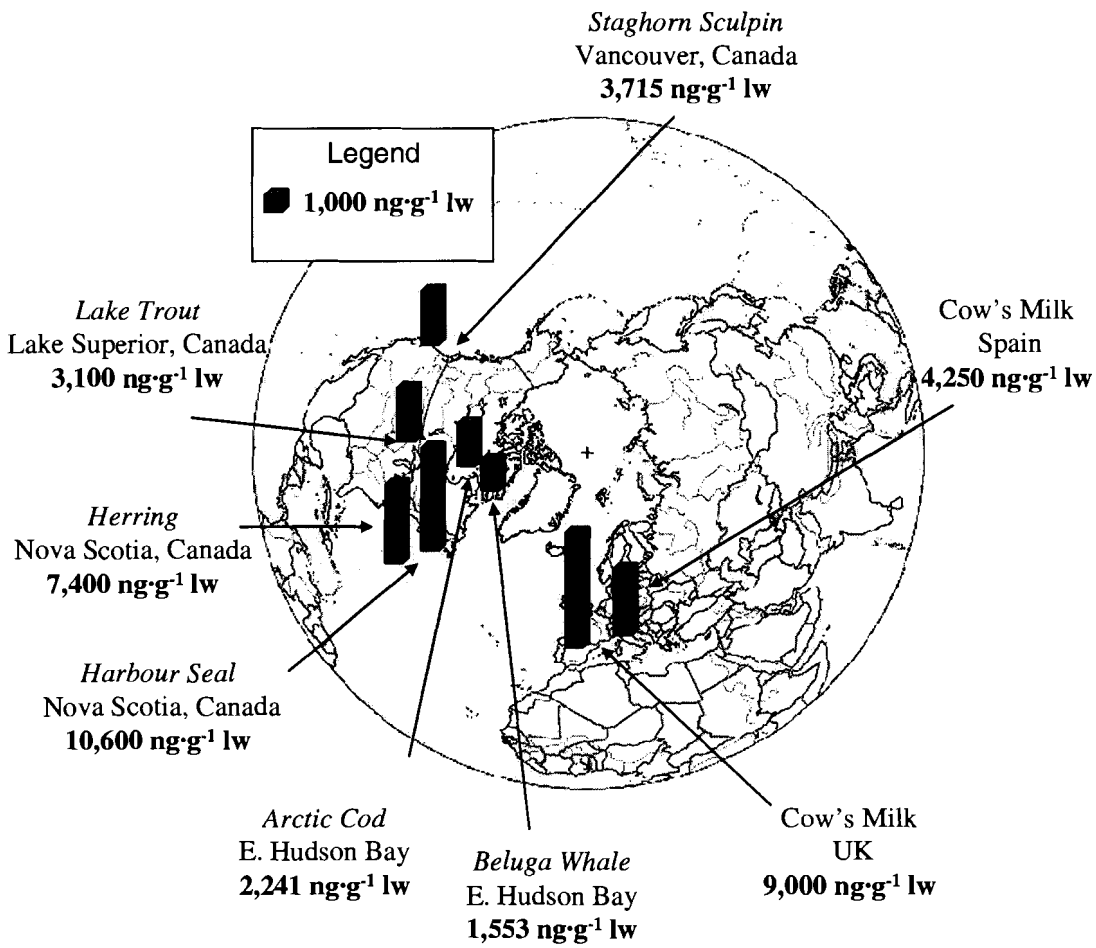


Figure 5.3 Geospatial variation of DEHP concentrations reported in biota from North America and Europe.



Note: Map acquired with permission from woldatlas.com at <http://worldatlas.com/aatlas/world.htm>

Figure 5.4 Relationship between chemical concentration in various water-ventilating ectotherms and air-breathing endotherms of the E. Hudson Bay marine food web (ng.g^{-1} lipid) and trophic level (TL) for (a) CB153 , (b) DMP, (c) BBP, (d) DEHP and (e) DIBP. Thick black line represents data for whole food web, thin black line represents air-breathing endotherms, and gray line represents water-ventilating ectotherms.

(a)

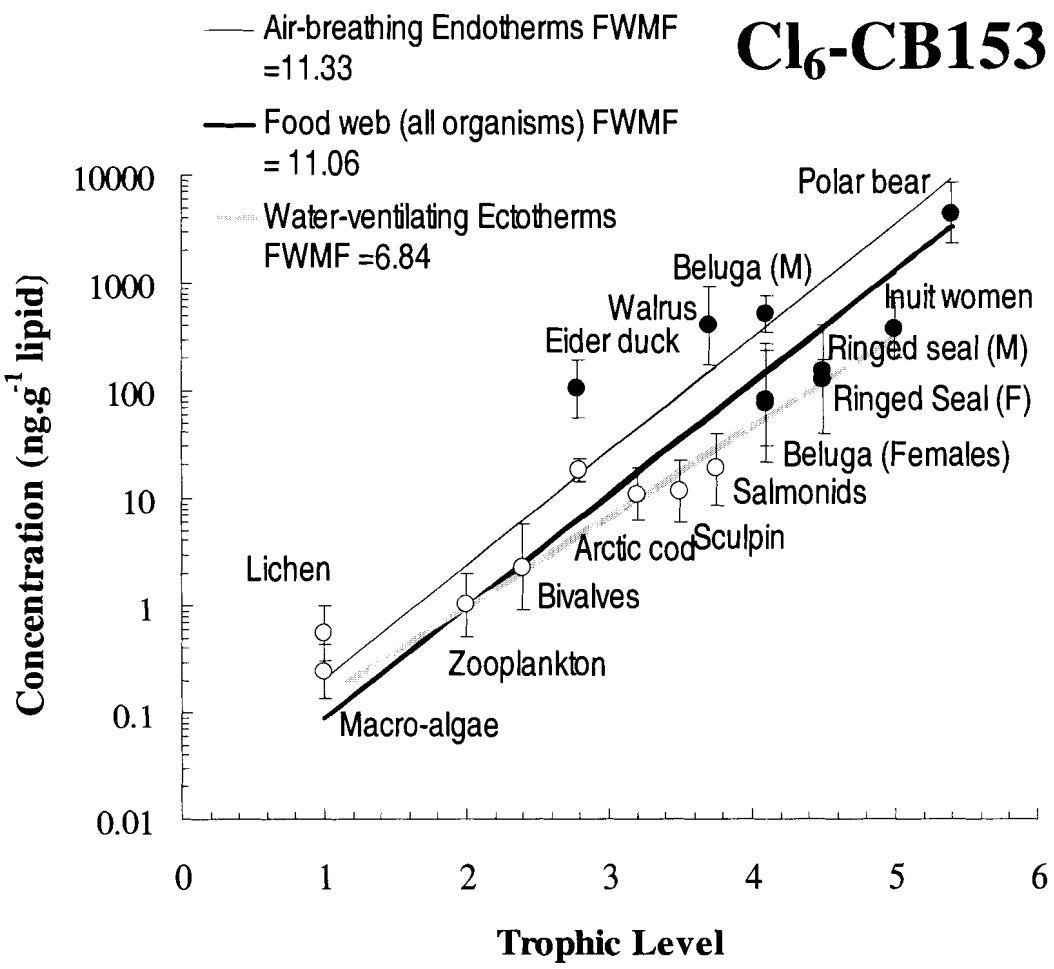


Figure 5.4 continued.

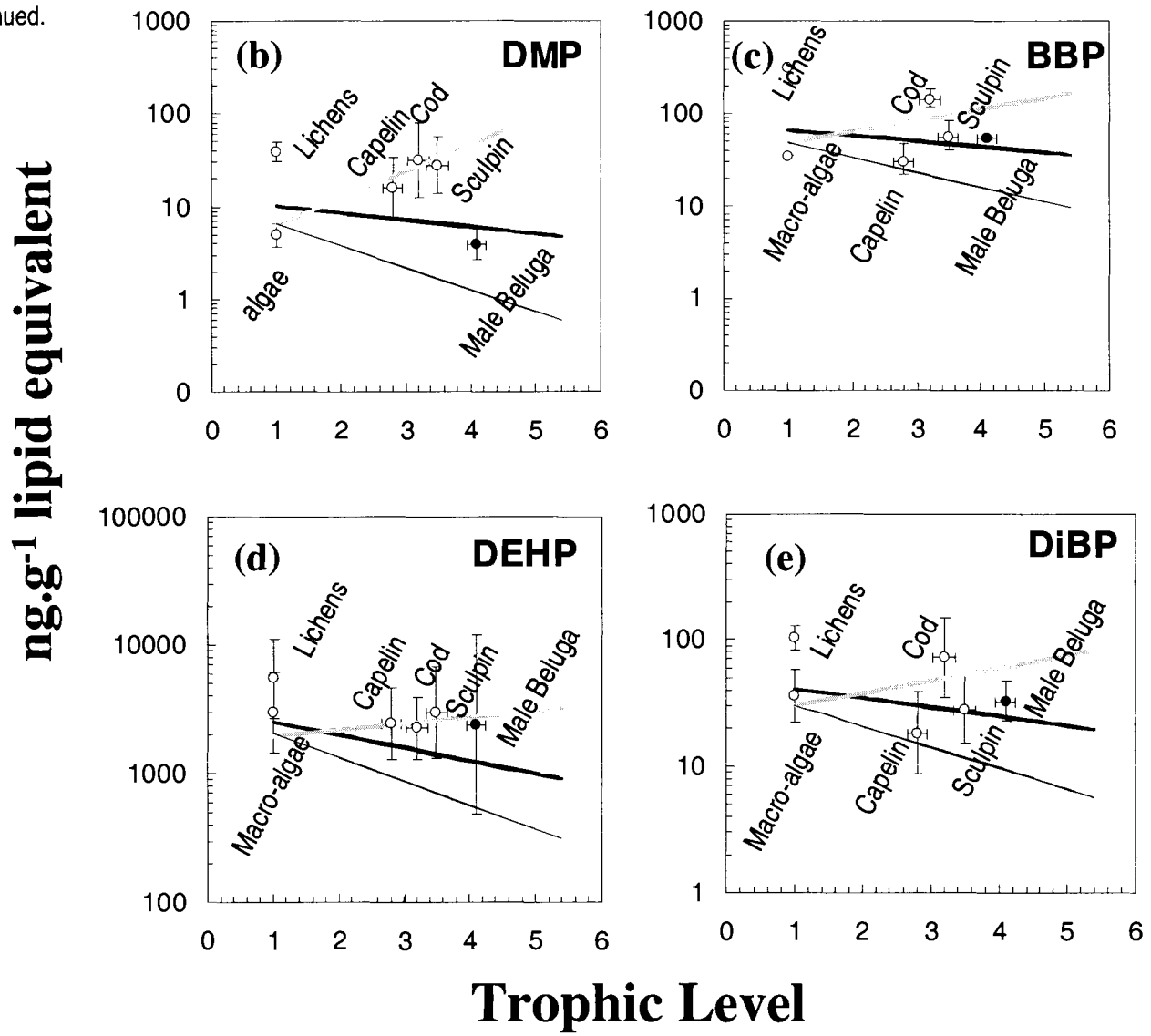


Figure 5.5 Elimination Index (EI) values for Group I-V PCB congeners and DPEs in male beluga whales from E. Hudson Bay.

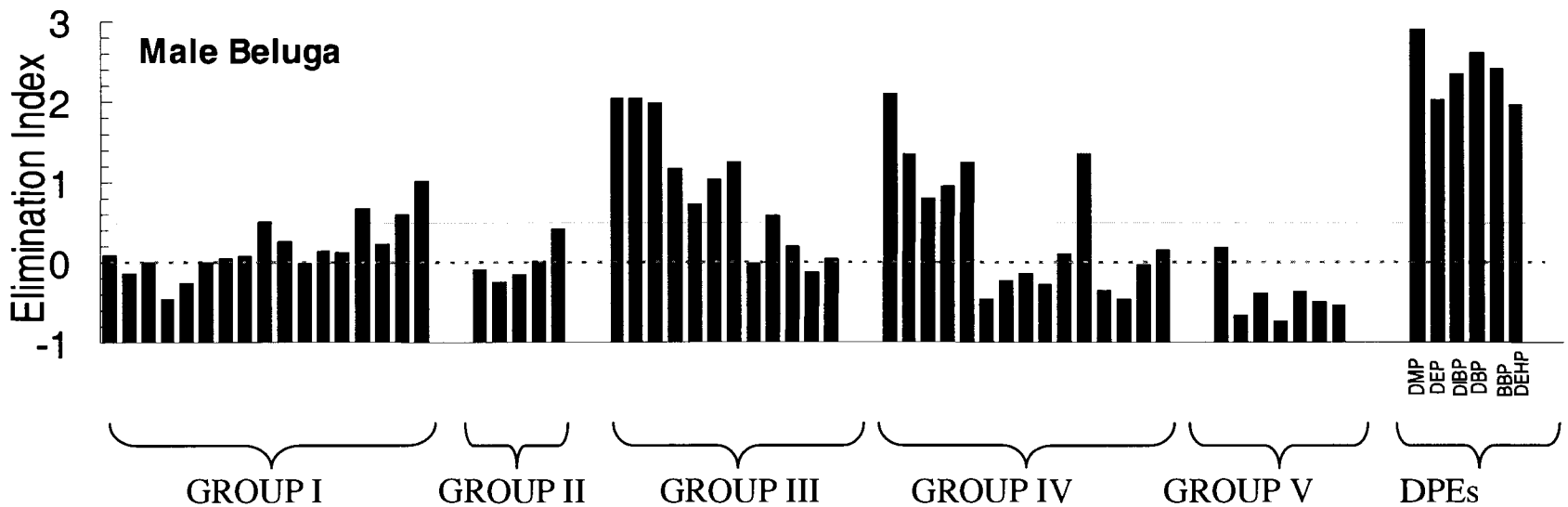
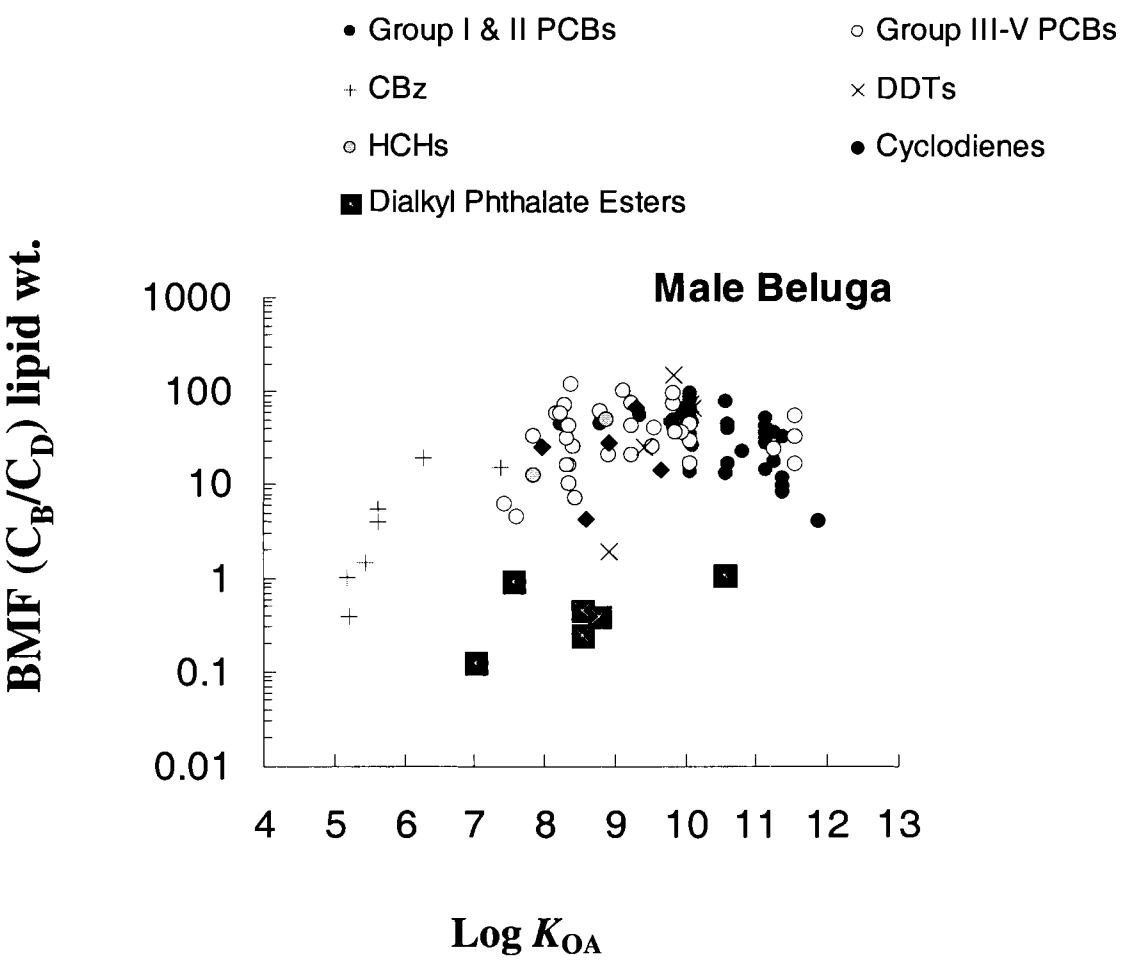


Figure 5.6 Relationships between observed BMFs (C_B/C_D) in male beluga and log K_{OA} for recalcitrant POPs such as PCBs and OC pesticides, with DPE BMFs plotted for comparison.



CHAPTER 6

BIOACCUMULATION POTENTIAL OF POLYBROMINATED DIPHENYL ETHERS IN A CANADIAN ARCTIC MARINE FOOD WEB

6.1 Introduction

Polybrominated diphenyl ethers (PBDEs) are an important class of brominated flame retardants (BFRs) used extensively in industrial and commercial products, including polyurethane foams, textiles, furniture, appliances and computers. The large market demand for PBDE application in commercial products has resulted in high production volumes (HPVs) of these compounds in North America and Europe. Bioaccumulation potential is an important aspect for assessing the overall risk posed by organic chemicals. While persistent organic pollutants (POPs) such as PCBs, DDTs and toxaphene have consistently shown a high degree of biomagnification in food chains, "current-use" high production volume (HPV) chemicals such as brominated diphenyl ethers (BDEs) have not been fully evaluated for their bioaccumulation potential. Field surveys of BDEs in biota have indicated BDE biomagnification comparable to PCBs (*238*), while others have found minimal or no biomagnification potential of BDEs (*151*). Because BDEs exhibit similar physical-chemical properties as legacy POPs such as PCBs, e.g., octanol-water partition coefficients (K_{OW}'s) ranging between 10^6 and 10^{11}, these compounds, in the absence of environmental and/or biological degradation mechanisms such as microbial degradation, photolysis and metabolic transformation, have potential to bioaccumulate and biomagnify in food chains.

Recent studies of BDE uptake and depuration in laboratory animals such as rats and fish have provided much insight into the complexities regarding toxicokinetics and persistence of major BDE congeners, including deca BDE209 (*239,240,241,242,243*). Specifically, these studies indicate that (i) BDEs exhibit very high dietary absorption efficiencies (>80%), with the exception of the superhydrophobic (i.e., log K_{OW} = 10.5) deca BDE209 (0.02-1%), (ii) substantial debromination of BDEs can occur in organisms, resulting in the bioformation of lower

brominated congeners and (iii) oxidative biotransformation of BDEs *via* cytochrome P450 mediated metabolism can generate several hydroxylated BDEs (OH-BDEs). *In vivo* debromination through cleavage of the relatively weak C-Br bond can result in the formation of major BDE congeners (i.e., present in commercial mixtures) such as BDE-47, -66, -99, -100, -153 and -154 or unknown BDEs, including several BDEs of the penta-to-octa homologues. For example, Kierkegaard et al. (*239*) and Stapleton et al. (*241*) showed the major debromination products of deca BDE209 in pike (*Esox lucius*) and carp (*Cyprinus carpio*), respectively were BDE 153, 154 and several unknown hexa to octa BDEs. Stapleton et al. (*240*) showed significant debromination of BDE99→BDE47 and BDE183→BDE154 within the intestinal tract of common carp (*Cyprinus carpio*), indicating cleavage of the C-Br bond can occur *via* microbial enzyme activities in the gut of organisms. Also, reductive debromination of BDE congeners can occur in the environment through anaerobic microbial and/or photochemical degradation (*244*). The overall degradation pathways (i.e., in the environment and within organisms) for major BDE congeners arising from the commercial penta mixtures are as follows: BDE-153 → BDE-99 → BDE-47 and BDE-183 → BDE-154 → BDE-100. Thus, successive debromination steps with increasing trophic level may result in higher tissue residue burdens of lower brominated congeners (e.g., Br_4-BDE47). Consequently, observed biomagnification factors (BMFs) of these lower brominated congeners in organisms may be inflated due to enhanced formation of those compounds from higher brominated homolougues, assuming no further debromiantion or oxidative metabolism occurs. However, these lower brominated congeners may also be further debrominated and hence not accumulated to any great extent. For example, Br_3-BDE28 was evidently formed as a debromination product of BDE47 following BDE99→BDE47 debromination in dietary uptake studies with carp (*240*). Furthermore, Stapleton et al. (*240*) concluded from a mass balance during BDE debromination study in carp that the extensive degradation of the parent BDEs administered could not fully be explained by debromination, indicating some metabolic transformation to other products such as OH-BDEs *via* hydroxylation. This assertion is supported by observations of six tetrabromo OH-BDEs in tissues of fish exposed to BDE47 (*239*). Thus, the rate limiting steps for determining the extent of congener-specific biomagnification of BDEs in a given organism is likely threefold, involving (i) dietary uptake rate (ii) the rate of debromination and (iii) metabolic transformation to OH-BDEs or other metabolites.

The degree of accumulation and recalcitrance of BDEs and their metabolites (debromination products + oxidative metabolites) in organisms and food chains is of particular concern due to

observed adverse effects in laboratory animals at doses in the low mg/kg body weight (*245*). To better aid PBDE risk evaluations, determination of congener specific bioaccumulation behaviour parameters such as predator/prey biomagnification factors (BMFs) and food web magnification factors (FWMFs) and identification of debromination products and metabolic transformation products is needed. In this paper we present measured concentrations of PBDEs in various organisms of a Canadian Arctic marine food web. A comparison of lipid equivalent concentrations of BDEs among organisms is conducted to calculate evaluative parameters such as BMFs and FWMFs. The relative bioaccumulation potential of these high production volume (HPV) "current-use" flame-retardants is compared to legacy POPs such as PCBs and DDTs. Debromination, biotransformation and subsequent trophic dilution of BDEs in the Arctic marine food webis discussed.

6.2 Materials and Methods

6.2.1 Sample collections.

During the months of May to August between 1999 and 2003 various biological samples were collected along the eastern Hudson Bay coastline in close proximity to the Inuit village Umiujaq (64° 15′N 113° 07′ W), (Figure 6.1). For details see *Chapter 1, Section 1.9.1* and Appendix 1, which summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex, age and condition.

6.2.2 Food web characterization and designation of organism trophic levels.

Appendix 14 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been established by extensive ^{15}N and ^{13}C isotope enrichment analyses involving numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of $TL = 1 + (\delta^{15}N - 5.4)/3.8$. More recent studies using $\delta^{15}N$ measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 (see *Chapter 1*) summarizes these

previous $\delta^{15}N$ measurements and TL ranges for the various organisms within these Arctic marine food webs. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 4.5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific $\delta^{15}N$ measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

6.2.3 Extraction, cleanup and analysis of BDEs.

Details of our methods for BDE analysis of environmental and biological samples and QA/QC procedures are detailed fully in reference *246*. Briefly, tissue samples (approximately 10 g wet wt for lichens, macro-algae and sediment, 5-15 g for fish, 2 g for beluga whale liver and 0.5 g for blubber (beluga whales and ringed seals) were homogenized with approximately 20 g Na_2SO_4 with mortar and pestle. Sub-samples of other tissue samples (e.g., seaduck and marine mammal tissue samples) were excised from the interior of frozen samples to reduce potential contact contamination during collection and/or storage. The homogenate powder was transferred to a glass extraction jar, spiked with ^{13}C-labeled procedural internal standards (Cambridge Isotope Laboratories, Andover, MA), approx. 2000-5000 pg of each ^{13}C BDEs (^{13}C BDEs 3,15, 28 47, 77, 118, 99, 100,153 and 183). The spiked samples were then extracted with 30 mL of 1:1 (v/v) DCM/Hexane in a Branson 5210 ultrasonic water-bath (Branson Ultrasonics Co., CT) for 20 min. Once the suspended particles settled, the supernatant was removed and then extraction was repeated two more times with fresh solvent. The combined extracts were concentrated to ca. 2 mL with a gentle stream of high-purity nitrogen. Relatively low lipid samples (< 5% lipid w/w)

such as cod and sculpin tissue were quantitatively transferred onto a 350 mm x 10 mm i.d. glass column packed with 8 g 100% activated florisil (60 -100 μm mesh, activated at 400 C overnight). High lipid samples (>5% lipid w/w) such salmon, and beluga blubber were first passed through a Gel Permeation Column (GPC) filled with 70 g of BioBeads, S-X33 (BioRad) in 50% DCM/hexane solution (V/V). The lipid fraction from the GPC (180 mL) was collected and discarded, while the remaining 300 mL of eluent from the GPC was collected evaporated to near dryness and solvent exchanged into hexane for further cleanup by Florisil. Three fractions were then eluted using 60 mL hexane (fraction 1), 60 mL 15% DCM/hexane (fraction 2), and 120 mL 50% DCM/hexane (fraction 3). The four fractions were combined in a single 500 mL boiling flask and evaporated to a final volume of 100 μL. Quantification of BDEs was determined by high resolution gas-chromatography (GC-HRMS) using a Micromass Ultima HR-mass spectrometer coupled with an HP 5890 Series II GC and a CTC A200S autosampler (CTC Analytics, Zurich, Switzerland). The GC column used was a 15 m high temperature DB-5-HT (0.225 mm i.d. × 0.1 μm film thickness). The HRGC was operated in splitless mode was used with the purge valve being activated 2 min following sample injection. Ultra high purity helium at 80 kPa was used as the carrier gas using the following temperature program: hold at 100 °C for 1 min, 2 °C min^{-1} to 140 °C, 4 °C min^{-1} to 220 °C, 8 °C min^{-1} to 330 °C and hold for 1.2 min. For all analyses, the MS was operated at 10,000 resolution in the positive ion mode at 39 eV energy and data were acquired in the single ion resolving mode (SIR). Analytes were identified by retention time (RT) comparison relative to authentic calibration standards. For Br_1 and Br_2 homologues and Br_4-BDE77, the two most abundant isotopes representing the parent ion $[M^+]$ were monitored. For all other homologues (Br_3-Br_7 congeners) the two dominant isotopes representing the $[M\text{-}2Br]^+$ fragment were monitored. Quantification ions were *m/z* 323.8785 for Br_4-BDEs, 403.7870 for Br_5-BDEs, 481.6975 for Br_6-BDEs and 561.6060 for Br_7-BDEs Concentrations were calculated by the internal standard isotope dilution method using mean relative response factors (RRFs) determined from a calibration standards, run prior to and following sample analyses. A total of 31 individual mono- to hepta- BDE congener peaks and three co-eluting bands (each composed of two congeners) were identified and quantified, establishing the initial data set of 37 congeners overall: BDE-1, -2, -3, -7, -8/11, -10, -12, -13, -15, -17, -25, -28/33, -30, -32, -35, -37, -47, -49, -66, -71, -75, -77, -85, -99, -100, -116, -119, -138/166, -140, -153, -154, -155, -181, -190. Method blanks, consisting of Na_2SO_4, were extracted according to the same procedure as environmental samples and analyzed with every batch of 12 samples to check for contamination of the extracts.

6.2.4 Data treatment/compilation and statistics.

To enable direct comparisons of POPs between different environmental media and organisms it is important to correct chemical concentration data to a common unit expression such as lipid equivalent concentrations. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~1 - 98%), wet weight chemical concentrations (C, $ng \cdot g^{-1}$ ww) were expressed solely on a lipid weight basis by the equation: C_L = C ww. ÷ ϕL in units of $ng \cdot g^{-1}$ lipid. For some biological matrices with very low lipid fractions (ϕL < 1%), such as vegetation and algae tend to solubilize organic contaminants in non-lipid biomolecules (i.e., non-lipid organic matter, NLOM) rather than in extractable lipids (*13,57,58,59*). Thus, for macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and NLOM (ϕ_{NL}) fractions following the equation: $\phi Leq = \phi L + 0.035\phi_{NL}$, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42,44*). Because chemical concentrations exhibited log-normal distributions and were hence transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for POPs in the various organisms collected and analyzed as part of the present study (i.e., lichens, macro-algae, bivalves, fish, beluga whales and ringed seals). In addition, we also compiled literature reported concentration data for PCBs and OC pesticides in Canadian Arctic biota , including invertebrates (*4*), walrus (*Odobenus rosmarus*) (*60*) polar bears (*Ursus maritimus*) (*61*), barren-ground caribou (*Rangifer tarrandus*) (*43,62,63*), wolves (*Canis lupus*) (*43,63*) and northern Quebec Inuit women (i.e., breast milk samples from references *64,63*) to compare contaminant concentrations, profiles and BMFs in various wildlife species and humans that generally subsist within the same food web.

Evaluative parameters for assessing chemical bioaccumulation potential.

See *Chapter 1, Section 1.9.5*

6.3 Results and Discussion

6.3.1 Levels and congener profiles of BDEs in marine sediments and biota.

Levels of brominated diphenyl ethers (BDEs) in E. Hudson Bay marine sediment and biota samples are summarized in Appendix 17. The data are not blank subtracted as procedural blanks for BDEs were generally low or non-detectable. Method detection limits (MDLs) were determined as the instrument limit of quantification (LOQ) on the HRMS. The data are presented as geometric means ± 95% confidence limits, along with corresponding lipid, lipid equivalent, moisture and organic carbon contents. In general, BDE levels in E. Hudson Bay sediment were equal to or greater than levels of PCBs that we recently reported in those samples (*186*), (See Appendix 5 and 6). Fourteen major BDE congeners were regularly detected in sediments and biota, including BDE-15, -30, -17, -28/33, -49, -47, -66, -77, -100, -99, -118, -153, -154 and -183, with BDE congeners 47, 99, 100 and 154 being the most dominant compounds. In addition, we detected several unidentified (UI) tri, penta and hexa bromodiphenyl ethers, primarily in marine mammal and seaduck samples. These unknown BDEs included Br_3 (UI TriBDE #2), Br_5 (UI PeBDE #1), Br_5 (UI PeBDE #2), Br_5 (UI PeBDE #3), Br_5 (UI PeBDE #4), Br_5 (UI PeBDE #5), Br_5 (UI PeBDE #6), Br_5 (UI PeBDE #7), Br_5 (UI PeBDE #8), Br_6 (UI HxBDE #1) and Br_6 (UI HxBDE #2), (see Appendix 17). The most dominant unidentified BDEs observed in biota samples were Br_5 (UI PeBDE #4), with a relative retention time to BDE 47 (RRT_{BDE47}) equal to 1.061, followed by Br_6 (UI HxBDE #2), with a RRT_{BDE47} equal to 1.221 on the employed 15m DB-5-HT column.

Organic carbon corrected ∑BDE (Br_1-Br_7) concentrations in sediments were ~ 81 ng·g^{-1} OC wt., comparable to ∑PCBs (62 ng·g^{-1} OC wt.). The dominant BDE in sediment (Br_5-BDE99) exhibited significantly higher concentrations (67.7 ng·g^{-1} OC wt) than dominant PCB congeners such as Cl_6-CB153 (3.2 ng·g^{-1} OC wt). Because of the very low organic carbon content of the highly mineralized sediments of E. Hudson Bay (i.e., TOC was ~ 0.1%), the presented OC corrected chemical concentrations in sediments are 2 to 3 orders of magnitude higher than dry wt. concentrations. Observed BDE concentrations in biota samples were generally lower than PCB concentrations. ∑BDE concentrations did not vary substantially among biota (between ~1 and 325 lipid ng·g^{-1} lipid equivalent), including approximately 324 ng·g^{-1} lipid equivalent in macro-algae, 9.3 ng·g^{-1} lipid equivalent in lichens, 9.8 ng·g^{-1} lipid in cod, 72.8 ng·g^{-1} lipid in sculpin, 13.6 ng·g^{-1} lipid in male ringed seals, 19.7 ng·g^{-1} lipid in eider ducks, 71.3 ng·g^{-1} lipid in white-

winged scoters and 27.0 $ng \cdot g^{-1}$ lipid in male beluga whales. Conversely, $\sum$PCB concentrations were observed to vary widely (orders of magnitude) among biota, ranging from approximately 1.30 $ng \cdot g^{-1}$ lipid equivalent in macro-algae, 4.22 $ng \cdot g^{-1}$ lipid equivalent in lichens, 60.7 $ng \cdot g^{-1}$ lipid in cod, 602 $ng \cdot g^{-1}$ lipid in male ringed seals, 734 $ng \cdot g^{-1}$ lipid in eider ducks, 2,950 $ng \cdot g^{-1}$ lipid in white-winged scoters and 3,410 $ng \cdot g^{-1}$ lipid in male beluga whales. The highest concentrations of BDE47 were observed in white-winged scoter liver (14.7 $ng \cdot g^{-1}$ lipid) and male beluga whales blubber (15.4 $ng \cdot g^{-1}$ lipid), which were significantly lower ($p < 0.05$) than CB153 concentrations of in scoters (841 $ng \cdot g^{-1}$ lipid) and male beluga (448 $ng \cdot g^{-1}$ lipid). The $\sum$BDEs higher levels (~ 4 times) in molluscivorous white winged scoters (71.3 $ng \cdot g^{-1}$ lipid) compared to eider ducks (19.7 $ng \cdot g^{-1}$ lipid), which also feed on bivalves and hence occupy comparable trophic positions, may be the result of enhanced contaminant accumulation *via* bivalves from relatively more contaminated habitats in the eastern United States coastal waters during the months of November to March, (i.e., over-wintering habitat) (*168*). We have previously suggested this as a possible explanation for an observed 10 times greater PCB burden in this migratory seaduck compared to the Arctic resident common eider ducks. The relatively high BDE levels observed in ambient environmental samples such as sediments, lichens and macro-algae suggest (compared to levels of PCBs) is not surprisingly considering those compounds are a high production volume (HPV) current-use brominated flame retardants (BFRs) that has experienced exponential global production rates during the 1980s and 1990s (*30,247*), while PCBs have been restricted internationally since the 1970s. While PCBs levels in the Arctic have generally stabilized over the past decade (*30,69,71,173*) those compounds continue to be a dominant organohalogen contaminant in Canadian Arctic marine biota, especially for higher trophic and long-lived species such as seabirds and marine mammals.

BDE concentrations observed in E. Hudson Bay sediments and biota (this study) are generally comparable to BDE levels reported elsewhere in Canadian Arctic biota (*30,34,248*). For example, $\sum$BDE levels in marine sediments from this study (0.15 $ng \cdot g^{-1}$ dry wt.) are comparable to recent $\sum$BDE measurements (*174,248*) of marine sediments from the high Canadian Arctic (0.107 - 0.297 $ng \cdot g^{-1}$ dry wt.). Also, $\sum$BDEs of 34 $ng \cdot g^{-1}$ lipid in E. Hudson Bay male belugas (this study) were similar to $\sum$BDE levels of ~ 16 $ng \cdot g^{-1}$ lipid reported in male beluga whales sampled during the same period (i.e., 1997-98) from relatively nearby S.E. Baffin Island in the eastern Canadian Arctic (*34*). A recent study of the polar cod-beluga and polar-cod-ringed seal-polar bear trophic transfer of BDEs in the Norwegian Arctic marine food web (eastern Svalbard), (*151*) reported relatively higher BDE levels compared to the Canadian Arctic biota. For example,

BDE47 in blubber biopsies of live-captured male beluga whales from Svalbard (90 ng·g^{-1} lipid) were approximately 6 times higher than BDE47 in E. Hudson Bay male beluga blubber (15 ng·g^{-1} lipid). The presence of any significant circumpolar variation in BDE levels is difficult to currently assess because of very low number of Svalbard belugas blubber samples analyzed for BDEs (n=4). However, a series of recent papers comparing POPs levels and patterns in Canadian Arctic and central Barents Sea biota indicates distinct differences between these two parallel Arctic food webs, with a general trend towards elevated concentrations in Barents Sea biota (*249,250*).

BDE levels in E. Hudson Bay biota (this study) are substantially lower than BDE concentrations documented in marine biota proximate to more urbanized/industrialized marine systems (*32,33,251,252,253*). For example, average concentrations of BDE47 in Columbia River whitefish in western Canada (190 ng·g^{-1} lipid) (*252*)are approximately 40 times higher than our observed BDE47 levels in E. Hudson Bay Arctic cod (5 ng·g^{-1} lipid). BDE47 concentrations in St. Lawrence male beluga whales (210 ng·g^{-1} lipid) (*32*), southern resident male killer whales from British Columbia (450 ng·g^{-1} lipid) (*33*) and male harbour seals from San Francisco Bay (2,040 ng·g^{-1} lipid) (*251*) are approximately 15-140 times higher than BDE47 levels in E. Hudson Bay male belugas (15 ng·g^{-1} lipid). For seabirds, BDE47 levels reported in eggs of double crested cormorants (*Phalacrocorax auritus*), great blue herons (*Ardea herodias*) from the Georgia Basin-Puget Sound system near Vancouver, Canada have been measured at approximately 250 and 1,365 ng·g^{-1} lipid, respectively (*253*) and are approximately 15-300 times higher than our measurements of BDE47 in eider ducks (4 ng·g^{-1} lipid) and white winged scoters (15 ng·g^{-1} lipid) from E. Hudson Bay.

Figure 6.2 illustrates the BDE congener compositions (i.e., % contributions for Br_3-Br_7 congeners) in the commercial pentabromodiphenyl ether formulation Bromkal® (a) versus the observed composition pattern in E. Hudson Bay sediments and biota (b), along with corresponding observed BDE concentrations (ng·g^{-1} lipid equivalent) in those media. Commercial Penta BDE formulations (e.g., Bromkal 70-5DE) traditionally consist of 50-62% Br_5-BDEs and 24-38% tetra-BDEs (e.g., Br_4-BDE47), with the major penta contribution being from BDE congeners 99 and 100 at a ratio of approximately 85:15 w/w (*254*). In figure 6.2, the BDE profiles shown for lichens (collected on land in close proximity to marine sampling locations) can be viewed as an atmospheric "signal" resulting from air-borne contaminant exposure processes. Similarly, contaminant profiles shown for sediments and macro-algae

represent an aquatic "signal" of water-borne chemical in the marine system, while those profiles for biota are indicative of food web bioaccumulation processes and subsequent chemical residue distributions in organism tissues'. The observed BDE composition profiles in lichens (representing the ambient atmospheric signal) and sediments and macro-algae (representing the ambient aquatic signal) are dominated by BDE99 and are comparable to the commercial Bromkal® formulation profile. For example, the BDE99:BDE100 ratio for lichens, macro-algae and sediment were ~ 83:17, 84:16, and 80:20, respectively. However, for bivalves, fish, seaducks and marine mammals (i.e. the food web pattern), the BDE congener pattern changes and generally predominates in the order Br_4-BDE47 > Br_5-BDE99 > Br_5-BDE100, which is consistent with congener specific BDE bioaccumulation patterns observed in other food webs (*151,255*). While the BDE99:BDE100 ratio for ambient environmental samples (i.e., lichens, macro-algae and sediments) was generally equivalent to commercial mixture (~80:20), that ratio for organisms of the food web including bivalves (~74:26), cod (~62:38), eider ducks (51:49), white winged scoters (~30:70), ringed seals (66:34) and beluga whales (~ 40:60) were generally lower, indicating an increased elimination of BDE99 and an increased accumulation of BDE100 in biota. This decline in BDE99:100 ratios from ambient media (e.g., air, sediments) to biota has previously been observed in other studies of BDEs bioaccumulation in marine food webs (*11,256*). The observed BDE congener pattern trend towards lower brominated congeners in higher trophic level organisms follows an expected trend as BDE debromination mediated *via* metabolic, photolytic, or other abiotic degradation processes may lead to the following major BDE congener degradation pathways: BDE-153 → BDE-99 → BDE-47 and BDE-183 → BDE-154 → BDE-100. The latter conversion pathway (i.e., octabrom BDE) and also debromination of deca BDE-209 is likely negligable in the Arctic because of substantially lower atmospheric flux of those higher brominated congeners to the Arctic environment compared to more urbanized locations (*257,258*). In general however, bioconversion/biodegredation of BDE congeners with increasing trophic level, birds and marine mammals may have a high BDE burden predominantly of lower brominated congeners (i.e., mainly Br_4-BDE47), which are though generally to have lower toxicological thresholds than higher brominated (Br_5-Br_7) congeners (*245*).

6.3.2 Chemical concentration relationships with trophic level and FWMFs.

Results from log-linear regression analyses of organism chemical concentrations (C_B) and trophic level (TL) and corresponding food web magnification factors (FWMFs) for (i) water-ventilating ectotherms, (ii) air-breathing endotherms and (iii) the overall food are summarized in Appendix

18. The strongest positive C_B-TL relationships were observed for the highly chlorinated (Cl_6-Cl_9) recalcitrant PCBs (Group I and II congeners) such as Cl_6-CB138, Cl_7-CB180 and Cl_6-CB153. Figure 6.3 shows C_B-TL regression lines for water-ventilating ectotherms, air-breathing endotherms and the overall food web together with observed concentrations of PCB153 (6.3a), BDE47 (6.3b), BDE99 (6.3c), BDE100 (6.3d). For CB153, estimated slopes were approximately 1.05, 1.04 and 0.84 for water-ventilating ectotherms, air-breathing endotherms and the overall food web, respectively. This equates to FWMFs of CB153 of approximately 6.8 for water-ventilating ectotherms, 11.3 for air-breathing endotherms and 11.0 for the overall food web respectively. In contrast, lipid equivalent concentrations of BDEs did not change significantly with increasing trophic level (Figure 6.3b-d). FWMFs of BDE47, 99 and 100 over the entire food web were 1.21, 0.55 and 0.83, respectively. In general, the data indicate that BDEs exhibit far lower biomagnification potential in this marine food web (i.e., FWMFs ≤ unity), compared to hydrophobic POPs such as Cl_6-Cl_8 PCBs (e.g., CB153 FWMF > 11).

6.3.3 Chemical BAFs and Concentration Estimates in Air (C_{AG}) and Seawater (C_{WD}).

Table 6.1 shows (i) physical chemical properties, log K_{OW}, log K_{OA}, (ii) measured air concentrations, (iii) estimated air and seawater concentrations of using an equilibrium partitioning model and calculated chemical concentrations in lichens and macro-algae, respectively and (iv) lipid corrected bioaccumulation factors (log BAFs) for several BDE congeners and organochlorines (PCBs and OC pesticides) in various species of the E. Hudson Bay food web. Weekly measurements of BDEs in Arctic air have recently been conducted under the Canadian Northern Contaminants Program (*174*), with high volume samplers stationed in the high Canadian Arctic at Alert (82.30° N 62.20° W), Tagish Yukon (60.20° N 134.12° W) and Dunai, Eastern Siberia (74.60° N 124.30° W). Additional studies of BDEs in the Norwegian Arctic air have also been recently conducted (*259,260,261*). Average ∑BDE (Br_2-Br_7) concentrations in air at Alert (240 pg·m^{-3}) and Tagish (424 pg·m^{-3}) were significantly higher than samples analyzed at Dunai (14 pg·m^{-3}) and northern Norway (~1.5 pg·m^{-3}). The elevated levels at the Canadian Arctic sites are suspected to be the result of volatilization of BDEs from local incineration of discarded BDE containing household and/or commercial products and hence the actual BDE air concentrations in the Canadian Arctic are likely closer to those levels observed in the Siberian and Norwegian Arctic (i.e., ~ 1-10 pg·m^{-3}). Estimated vapour phase air concentrations (C_{AG}) for BDEs ranged from 0.81 to 6.59 pg·m^{-3}, while estimated freely dissolved seawater concentrations (C_{WD}) of BDEs ranged from 0.0008 ng·L^{-1} to 0.024 ng·L^{-1}. BDE47, 99 and 100 exhibited the

highest estimated concentrations in air and seawater. BDE47, which exhibits similar hydrophobicity to Cl_7-PCBs (e.g., logK_{OW} BDE47 = 7.66 and log K_{OW} Cl_7-CB180 = 7.5), was estimated to have concentrations of approximately 5.92 pg·m^{-3}and 0.0125 ng·L^{-1} in air and seawater, respectively. Estimated BDE47 concentrations in E. Hudson Bay air and seawater were approximately 300-1,000 times greater than concentrations of Cl_7-CB180 in air (0.02 pg·m^{-3}) and seawater (1.17×10^{-5} ng·L^{-1}), respectively.

Bioaccumulation factors (BAFs) of Br_3 to Br_7-BDEs in macro-algae and fish, relative to estimated seawater concentrations are shown to increase slightly over a log K_{OW} range of 6.84 to 8.71 from ~ 6.45 for Br_3-BDE28/33 (log K_{OW} = 6.84) to 6.87 for Br_6-BDE154 (log K_{OW} = 8.10). For lichens, estimated log BAFs of Br_3 to Br_7-BDEs (log K_{OA}'s ~ 9.5 to 11.96), relative to previously measured gas-phase air concentrations of BDEs, unexpectedly did not increase with increasing K_{OA}. Specifically, log BAFs in lichens included values of ~ 9.14 for Br_4-BDE47 (log K_{OA}= 10.53), ~ 9.88 for Br_5-BDE99 (log K_{OA} = 11.31), and ~ 8.62 for Br_7-BDE154 (log K_{OA}= 11.92). BAFs for male beluga whales relative to air concentrations ranged from ~ 7 for Br_3-BDE28/33 (log K_{OA} = 9.50) and ~11 for Br_6-BDE153 (log K_{OA} = 11.82). These data show that BAFs of BDE congeners in E. Hudson Bay organisms relative to water (BAF = C_B/C_{WD}) and air (BAF = C_B/C_{AG}) are generally equal to or less than the compounds corresponding K_{OW} and K_{OA} and indicate BDEs in biota exhibit equilibrium concentrations as predicted by physical chemical properties. The BAF data for E. Hudson Bay biota shown in Table 6.1 suggest that PCBs biomagnify in organisms, while BDEs (with comparable physical chemical properties) conversely achieve a chemical equilibrium with ambient BDE levels in the environment.

6.3.4 Biomagnification potential of BDEs.

Figure 6.4 illustrates concentrations (ng.g^{-1} lipid) of several major BDE congeners and two unknown BDE congeners (one unknown penta and one unknown Hexa BDE) in E. Hudson Bay Arctic cod and beluga whales (including data for females, milk, calves and males). There were small significant increases ($p < 0.05$) of BDE lipid corrected BDE levels from Arctic cod to male beluga for BDE 47 and 100, 153, and 154, while no significant differences were observed between cod and beluga for BDE 99. The predator/prey BDE biomagnification factors (BMFs) for male beluga relative to Arctic cod (i.e., C_{BELUGA}/C_{COD} lipid) were ~ 1 for BDE99, 2.8 for BDE47, 2.4 for BDE100, 3.4 for BDE153 and 4.1 for BDE154. The observed BMFs of major BDE congeners (e.g., BDE-47, -99, -100) in E. Hudson Bay male beluga are very low compared

to other hydrophobic organohalogens with comparable physical chemical properties such as PCB153 and 180, which exhibited male beluga/cod BMFs of approximately 40. Also, shown in figure 6.4 are the concentrations of the two most predominant unidentified BDEs (Br_5-UI-PeBDE#4 and Br_6-UI-HxBDE#2) observed in tissues of Arctic cod and beluga whales. No biomagnification of the unknown hexa BDE Br_6-UI-HxBDE#2 was observed. However, a substantial increase in the unknown penta Br_5-UI-PeBDE#4 was observed between cod and beluga whales. The BMF of Br_5-UI-PeBDE#4 in male beluga/cod of approximately 29.6. The concentration of the unknown Br_5-UI-PeBDE#4 in male beluga whale blubber (5.63 $ng \cdot g^{-1}$ lipid) exceeded concentrations of major congeners BDE99 (2.34 $ng \cdot g^{-1}$ lipid) and BDE100 (3.07 $ng \cdot g^{-1}$ lipid), but was lower than BDE47 (15.2 $ng \cdot g^{-1}$ lipid). Because this unknown penta BDE was not observed in the ambient environment (sediments) and organisms of the lower food web (macro-algae, bivalves), this compound may be a recalcitrant debromination product originating from exposure to higher brominated hexa to deca BDEs (e.g., 153, 154, 183, or 209).

Beluga calves exhibited comparable BDE concentrations to those observed in male beluga whales, which may be attributed to maternal transfer of BDEs during the nursing period (~ 2 year duration for beluga whales). Levels of BDE47 and BDE100 in beluga calves were significantly higher ($p < 0.05$) than those concentrations in female beluga milk, indicating that a small amplification (i.e., biomagnification) of BDEs may occur in newborns during this early life-stage, i.e., BDE BMFs for beluga calves relative to mother's milk (i.e., C_{BELUGA}/C_{MILK} lipid) were 2.9 for BDE47 and 2.4 for BDE100. Figure 6.5 illustrates chemical concentrations in male, female and calves (blubber) as a function of animal age for (a) PCB153, (b) BDE99 and (c) BDE47. While significant concentration-age relationships were observed for recalcitrant PCBs such as CB153, no significant age trends were observed for BDEs. The absence of an increasing BDE concentration trend is likely due to the debromination/metabolic transformation of those compounds in beluga whales, thus decreasing the bioaccumulation potential over the lifetime of the animal.

Species-specific biomagnification factors (BMFs), elimination Index (EI) and biodilution factors (BDFs) of Br_3-Br_7 BDEs and selected PCB congeners and OC pesticides in E. Hudson Bay organisms are summarized in Appendix 19. EI values, BMFs and BDFs of PCBs and OC pesticides are presented in more detail in a preceding study of POPs bioaccumulation in E. Hudson Bay organisms (*186*). Figure 6.6 illustrates EI values for selected Group I-V PCB congeners compared to several BDE congeners in E. Hudson Bay male beluga whales. The data

show suggest that compared to recalcitrant PCBs (e.g., CB153 and CB180), BDE congeners exhibit relatively high EI values in male beluga whales (EIs > 1) and are more comparable to metabolizable PCB congeners, i.e., Group III-V congeners), indicating the presence of relatively high metabolic transformation rates of BDEs in beluga whales. The relatively high EI values for BDEs in beluga whales (EIs > 1) corresponds to correspondingly low BDE biomagnification factors (BMFs) in those animals (male beluga/Arctic cod), ranging between ~ 1 for BDE99 and 4.1 for BDE154. Specifically, compared to recalcitrant PCBs (e.g., BMF CB180), BDEs exhibit biodilution factors (BDFs) between 15 and 35 in beluga whales (i.e., 15-35 times reduced from BMF_{MAX}). Similar biomagnification parameters estimates (i.e., EIs, BMFs, BDFs) were observed for BDEs in ringed seals, eider ducks and white winged scoters (Appendix 19). The reduced biomagnification potential and accumulation of major BDEs in seaducks and marine mammals (i.e., "biodilution") is likely due to a combination of debromination and metabolic transformation *in vivo*. In particular, the absence of any significant biomagnification of BDE47 (an expected debromination product) indicates this congener may undergo further biotransformation in Arctic seducks and marine mammals, perhaps involving the oxidative formation of OH-BDEs as observed in laboratory studies (*239,243*). Several OH-BDEs have recently been reported in two Norwegian Arctic top-predators, glaucous gulls (*Larus hyperboreus*) and polar bears (*Ursus maritimus*) (*262*).

Figure 6.7 shows chemical biomagnification factors (BMFs) for PCBs, organochlorine pesticides (OCPs) and brominated diphenyl ethers (BDEs) in (a) ringed seals and (b) beluga whales versus the chemical's octanol-air partition coefficient (K_{OA}). This plot shows substantially lower BMFs of BDEs (i.e., BMFs equal to 2-4) compared to PCBs and OC pesticides (i.e., BMFs ~50) that exhibit similar physical-chemical properties. The high degree of chemical biomagnification of legacy POPs such as Cl_5-to Cl_7-PCB congeners in air-breathing animals such as beluga whales is essentially the result of (i) very efficient assimilation through dietary exposure and (ii) very low metabolic transformation rates (k_M) *in vivo*. For example, dietary absorption efficiencies (E_D) are typically between 90 - 100% in birds and mammals (*172*) and chemical half-lives ($T_{1/2}$) of Group I PCBs such as CB153 in tissues' of organisms can exceed 1,000 days (*52*). However, other chemicals may undergo enzymatic metabolism (i.e., *via* cytochrome P-450 isozymes), urinary excretion and respiratory elimination, or in the case of BDEs (debromination) which can act alone or together to reduce an organism's contaminant burden and ultimately lower the chemical's BMFs. For relatively hydrophobic (log K_{OW} > 5) and non-volatile chemicals (log K_{OA}'s > 6) such as PCBs and BDEs, respiratory elimination *via* alveolar air in air-breathing endotherms is

negligible and the primary route of chemical elimination is *via* metabolic transformation. Because dietary absorption of BDEs is also very efficient, >80% (*242,263*), the apparent reduced biomagnification of these compounds in E. Hudson Bay seaducks and marine mammals is likely due to relatively rapid metabolic transformation in those animals (*in vivo* $T_{1/2}$ values on the order of days to weeks).

While our study of BDEs in the E. Hudson Bay food web suggests substantial debromination/biotransformation and hence low biomagnification potential of BDEs in Canadian Arctic biota, other studies have observed a relatively high degree of BDE biomagnification in seabirds and marine mammals. For example, a recent study of BDEs in Norwegian Arctic marine mammals (beluga, ringed seals and polar bears) presented BMFs of BDE47, 99 and 100 in beluga whales ranging between ~20 and 60 (*151*). BDE47 was the only BDE congener detected in Svalbard Polar bears from the same study and exhibited very low BMFs (bear/seal BMF ~ 1.6), indicating efficient metabolic clearance of BDEs in polar bears. It should be noted that BMFs in beluga whales from Svalbard (*151*) were based on data from only four adult male individuals (n =4), two juvenile animals (n=2) and three polar cod samples. Other studies have reported varying degrees of BDE BMFs in biota from the North Sea (*264*) and Baltic food webs (*10,259*), including BDE47 BMFs of ~ 7 in grey seals (seal/herring lipid) to over 17 for osprey and guillemot eggs (egg/sea salmon lipid). The seemingly large disparity between metabolic capacity of BDEs in E. Hudson Bay and Svlabard beluga whales is not clear. It is possible that CYP enzyme induction rates and/or BDE debromination rates differ among the two populations of beluga whales. Also, seasonal fluctuations in BDE levels in blubber layer during the summer sampling period (i.e., depletion of fat reserves) may have caused relatively high BDE concentrations in Svalbard belugas (hence high BMFs). Another more plausible reason for these large differences is the use of inaccurate dietary concentrations in the BMF calculation. For example, beluga whales from these studies may utilize different prey species which exhibit different contaminant levels and patterns than cod. It is also important to note that chemical concentrations in cod muscle tissue were used in the present study and may differ from whole body concentrations (even on a lipid weight basis), which could ultimately alter BMF values. In summary, further studies regarding congener-specific debromination, biotransformation and metabolite (i.e., OH-and possibly MeO-BDE) formation rates of BDEs *in vivo* will aid future initiative to assess the relative persistence (P), bioaccumulation potential (B) and toxicity of these high-production volume (HPV) commercial chemicals.

6.4 Acknowledgements

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies and wish to thank Dr's Bill Doidge, Michael Kwan and Susan Sang, Derek Muir and Northern Quebec Inuit communities of Umiujaq and Inukjuaq for coordinating/aiding collection of field samples.

6.5 Tables

Table 6.1 Logarithms of Bioaccumulation Factors (log BAFs) of BDEs and PCBs in macro-algae, fish and beluga whales from E. Hudson Bay, along with estimated gas-phase air concentrations (C_{AG}, pg·m^{-3}) and freely dissolved surface seawater concentrations (C_{WD} ng·L^{-1}) and the chemical's K_{OW}, K_{OA}.

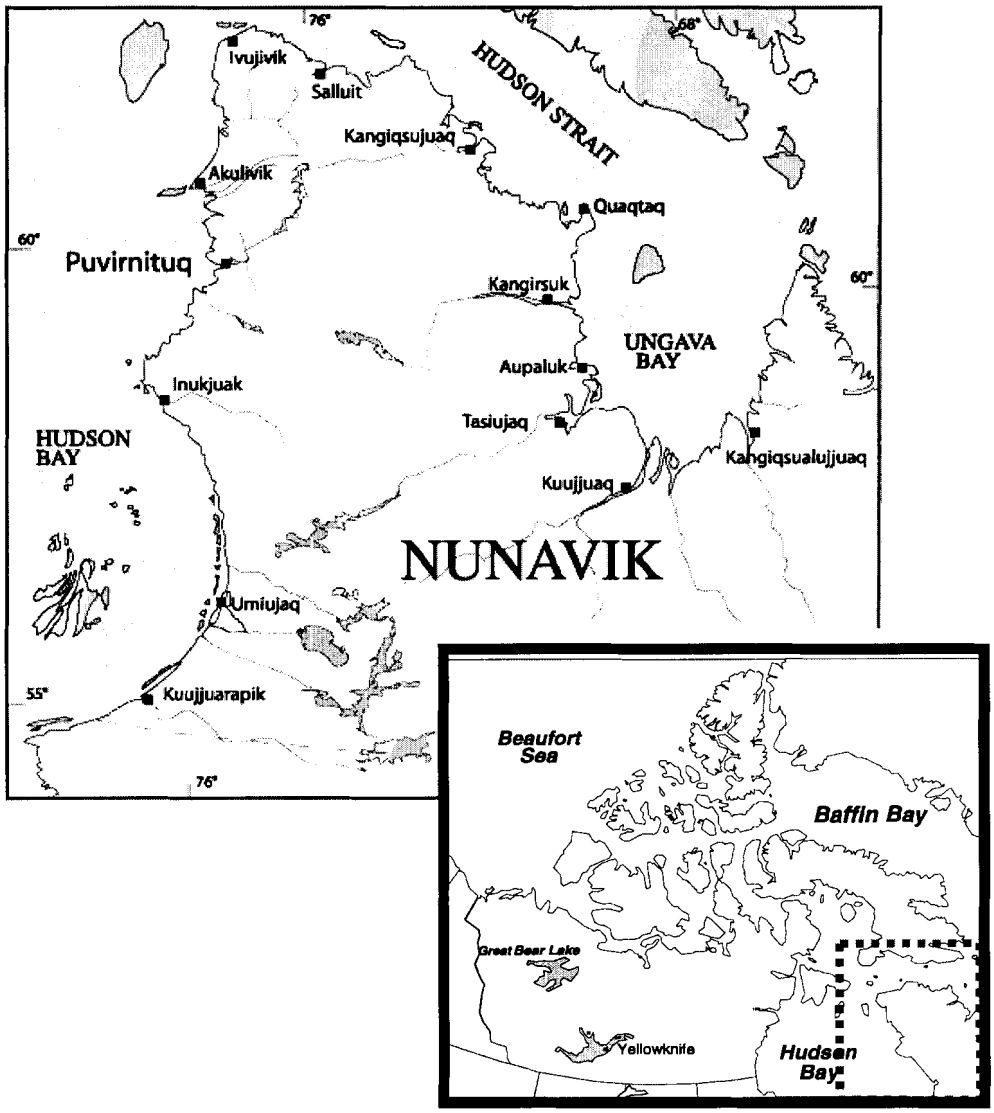
	Log K_{OW}	Log K_{OA}	C_{AG} observed (pg·m^{-3})	C_{WD} observed (ng·L^{-1})	C_{AG} estimated (pg·m^{-3})	C_{WD} estimated (ng·L^{-1})
BDEs						
BDE28/33	6.84	9.50	0.1	-		0.0008
BDE47	7.66	10.53	0.3	-	5.92	0.0125
BDE100	7.00	11.13	0.1	-	1.32	0.0090
BDE99	8.10	11.31	0.7	-	6.59	0.0239
BDE153	8.71	11.82	0.2	-	0.91	0.0029
BDE154	8.10	11.92	0.1	-	0.81	0.0050
BDE183	8.71	11.96	-	-	-	-
OCPs						
1,2,3,5/1,2,4,5 TeCBz	4.50	5.63	-	-	574	156
1,2,3,4 TeCBz	4.50	5.64	-	-	2,310	230
PeCBz	5.03	6.27	-	-	1,390	91.9
HCB	5.50	7.38	63	1.4	1,140	555
α-HCH	3.89	7.61	61	2.2	19.5	5,860
β-HCH	3.81	8.88	0.19	0.03	0.02	1,010
γ-HCH	4.14	7.85	7.6	0.28	8.19	1,210
CB28	5.0	6.88	1.16	0.0002	1.88	7.9×10^{-3}
CB118	6.74	8.231	0.53	0.0001	1.09	7.46×10^{-5}
CB153	6.9	9.79	0.68	0.00017	0.040	7.42×10^{-5}
CB138	6.83	10.02	0.34	0.00016	0.023	8.0×10^{-5}
CB180	7.5	9.83	0.58	0.00003	0.020	1.17×10^{-5}
CB187/182	7.2	10.57	0.40	-	0.0031	2.49×10^{-5}
CB194	7.8	11.24	0.07	-	0.00057	8.74×10^{-7}
CB206	8.1	11.36	0.05	-	0.00020	4.18×10^{-7}
CB209	8.4	11.88	0.06	-	0.000047	2.19×10^{-7}

Table 6.1 continued.

	Log BAF Lichen/ Air	Log BAF Macro-algae/ Water	Log BAF Cod/ Water	Log BAF Sculpin/ Water	Log BAF Eider Duck/ Air	Log BAF Male Ringed Seal/ Air	Log BAF Male Beluga/ Air
BDEs							
BDE28/33	-	6.5	5.6	6.1	9.1	9.4	9.7
BDE47	9.1	6.7	5.6	6.3	10.1	10.5	10.7
BDE100	8.9	6.5	5.2	6.0	10.5	10.0	10.5
BDE99	9.9	6.8	4.9	6.1	9.7	9.5	9.5
BDE153	9.1	6.9	4.9	6.4	10.3	9.2	9.6
BDE154	8.6	6.8	4.9	6.1	10.5	9.1	10.2
BDE183	-	6.5	5.6	6.1	-	-	-
OCPs							
1,2,3,5/1,2,4,5 TeCBz	5.6	-	-	-	6.9	6.4	6.6
1,2,3,4 TeCBz	5.6	-	-	-	6.8	5.8	6.0
PeCBz	6.3	-	-	-	-	6.9	7.1
HCB	8.6	-	-	-	9.2	9.4	9.7
α-HCH	8.2	4.0	3.8	3.9	-	8.4	8.6
β-HCH	-	-	-	-	11.0	11.0	11.4
γ-HCH	8.7	4.2	3.9	4.1	-	9.3	9.7
CB28	8.4	6.6	6.7	6.7	9.5	9.8	9.4
CB118	8.6	6.6	7.5	7.4	-	10.9	11.4
CB153	8.6	5.9	7.8	7.8	11.2	11.4	11.8
CB138	8.9	6.5	7.6	7.5	11.3	11.4	12.0
CB180	8.4	6.4	7.9	7.9	10.7	10.7	11.2
CB187/182	8.5	-	-	-	10.9	10.5	11.4
CB194	9.2	-	-	-	10.8	10.5	11.2
CB206	9.0	-	-	-	11.0	10.1	10.7
CB209	8.8	-	-	-	11.0	9.6	10.3

6.6 Figures

Figure 6.1 Map showing general study area of E. Hudson Bay and various Nunavik Inuit communities of northern Quebec, Canada.



Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

Figure 6.2 % composition of BDEs in commercial penta-formulation (Bromkal 70-5DE mixture) and in environmental and biological samples from E. Hudson Bay.

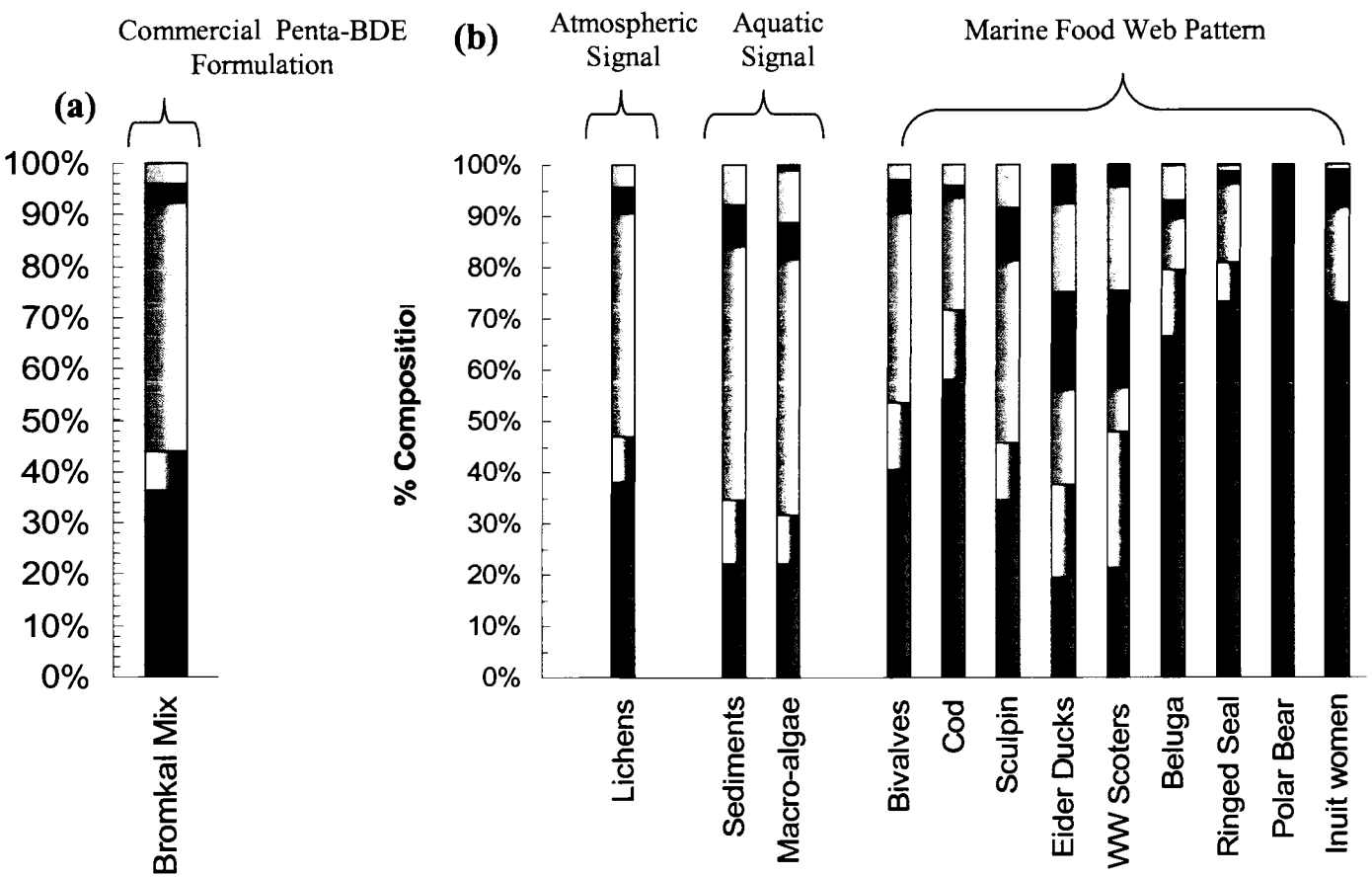


Figure 6.2 continued.

(c)

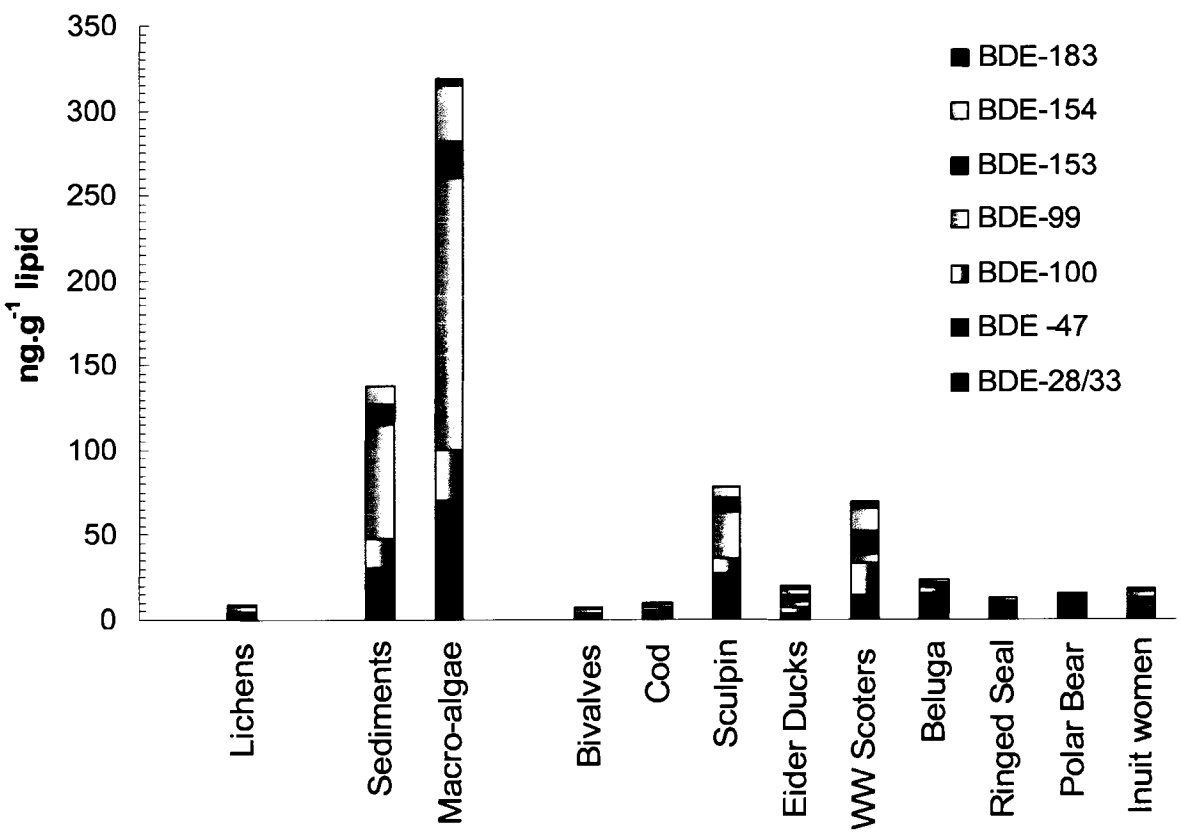


Figure 6.3 Relationship between chemical concentration in various water-ventilating ectotherms and air-breathing endotherms of the E. Hudson Bay marine food web ($ng \cdot g^{-1}$ lipid) and trophic level (TL) for (a) CB153 , (b) BDE47, (c) BDE99 and (d) BDE100. Thick black line represents data for whole food web, thin black line represents air-breathing endotherms, and gray line represents water-ventilating ectotherms.

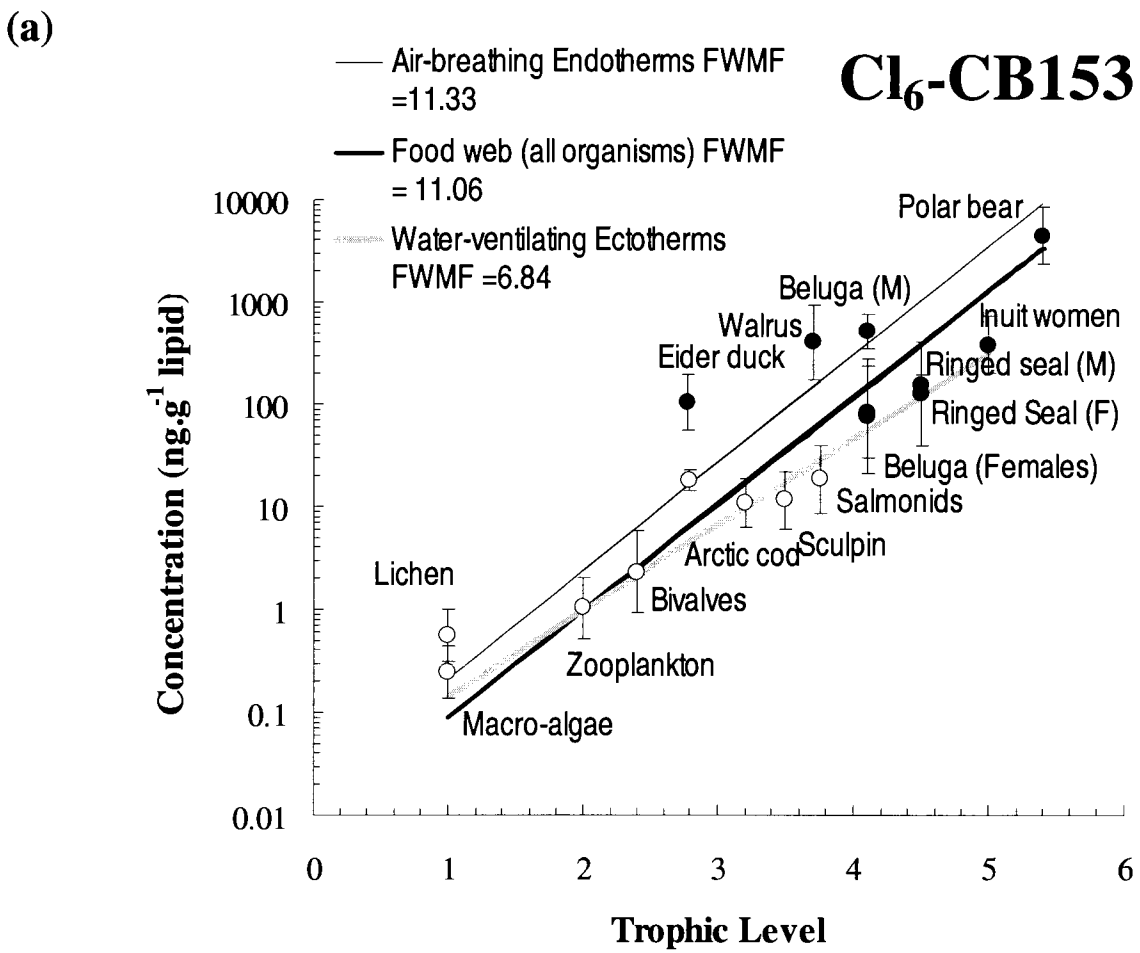


Figure 6.3 continued.

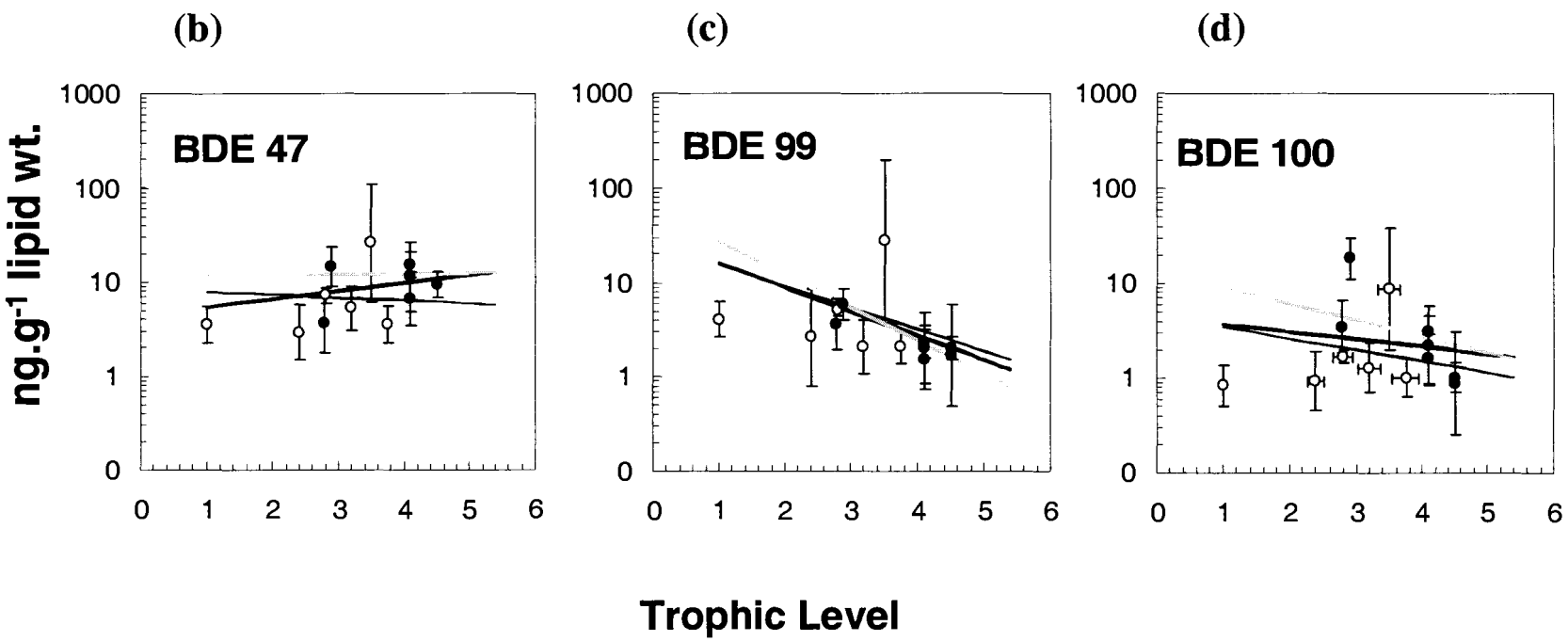


Figure 6.4 Concentrations (ng·g^{-1} lipid) of several BDE congeners in Arctic cod and tissue samples of beluga whales, including those concentrations in female blubber, female milk, blubber from calves < 1year old and male beluga blubber (aged 6-35 years)

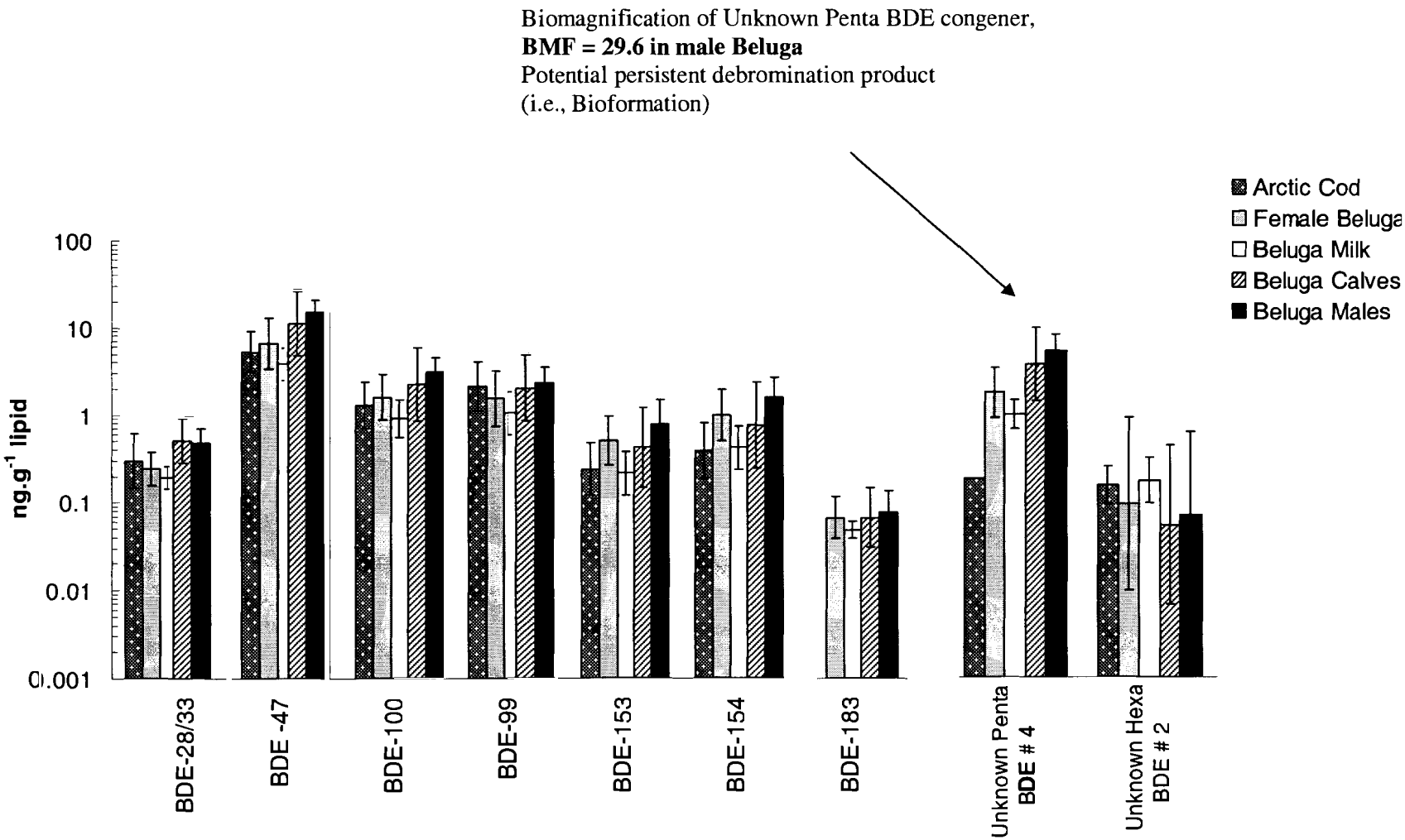


Figure 6.5 Chemical concentrations ($ng \cdot g^{-1}$ lipid wt.) versus animal age (years) beluga whales (blubber) from E. Hudson Bayfor (a) CB153 and (b) BDE47 and (c) BDE99. Data for males are shown as dark circles, females as white circles and calves as gray circles.

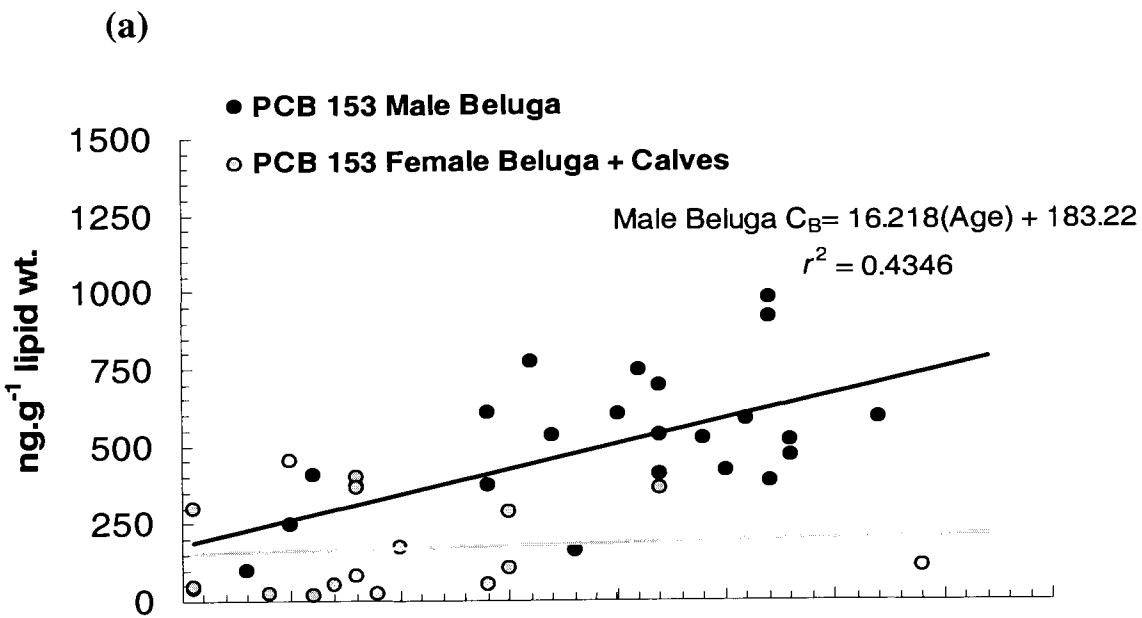


Figure 6.5 continued.

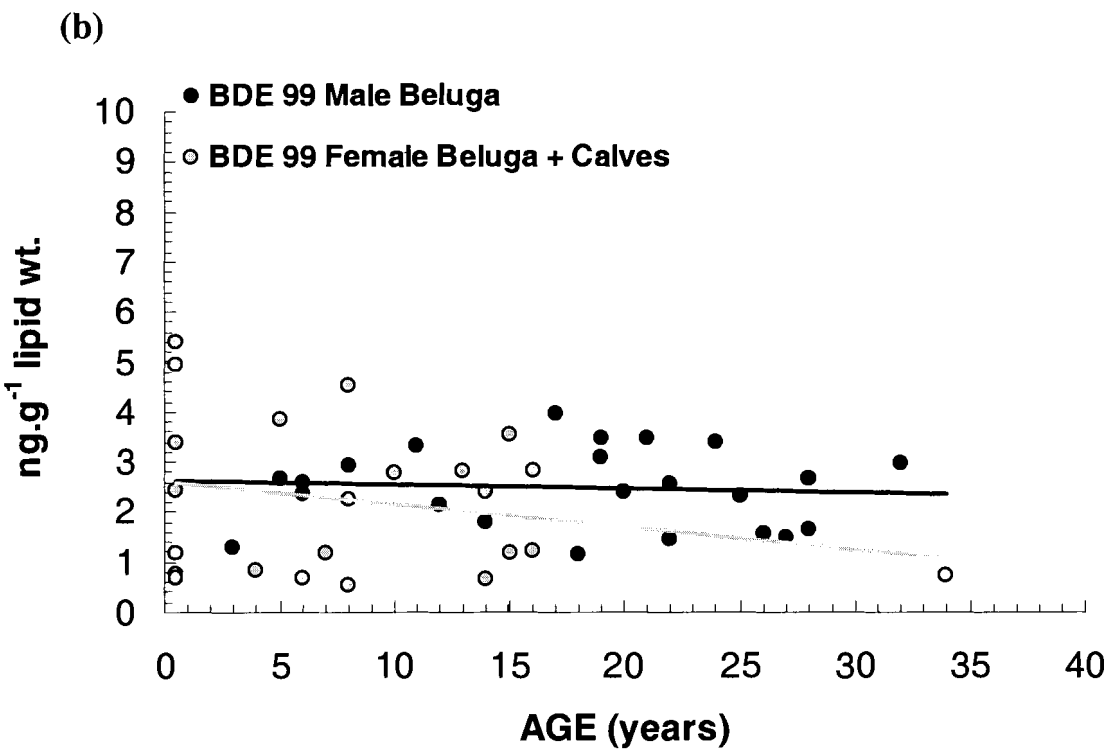


Figure 6.5 continued.

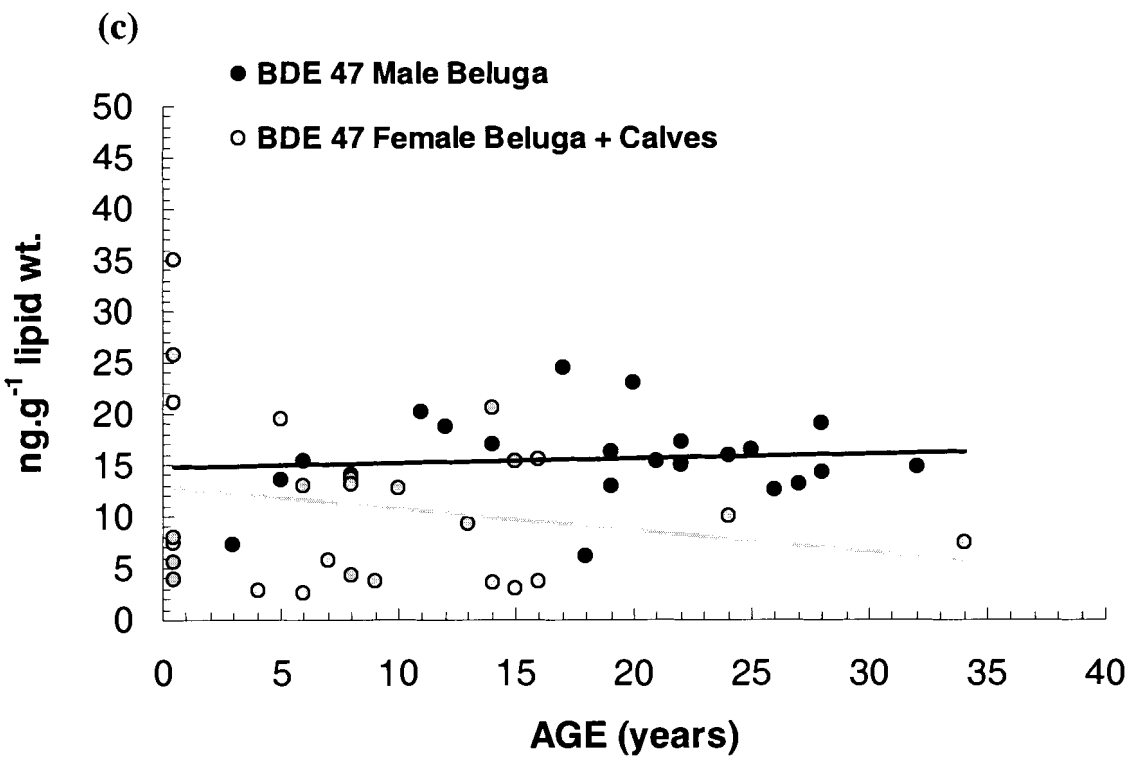


Figure 6.6 Elimination Index (EI) values for Group I-V PCB congeners and BDEs in male beluga whales from E. Hudson Bay.

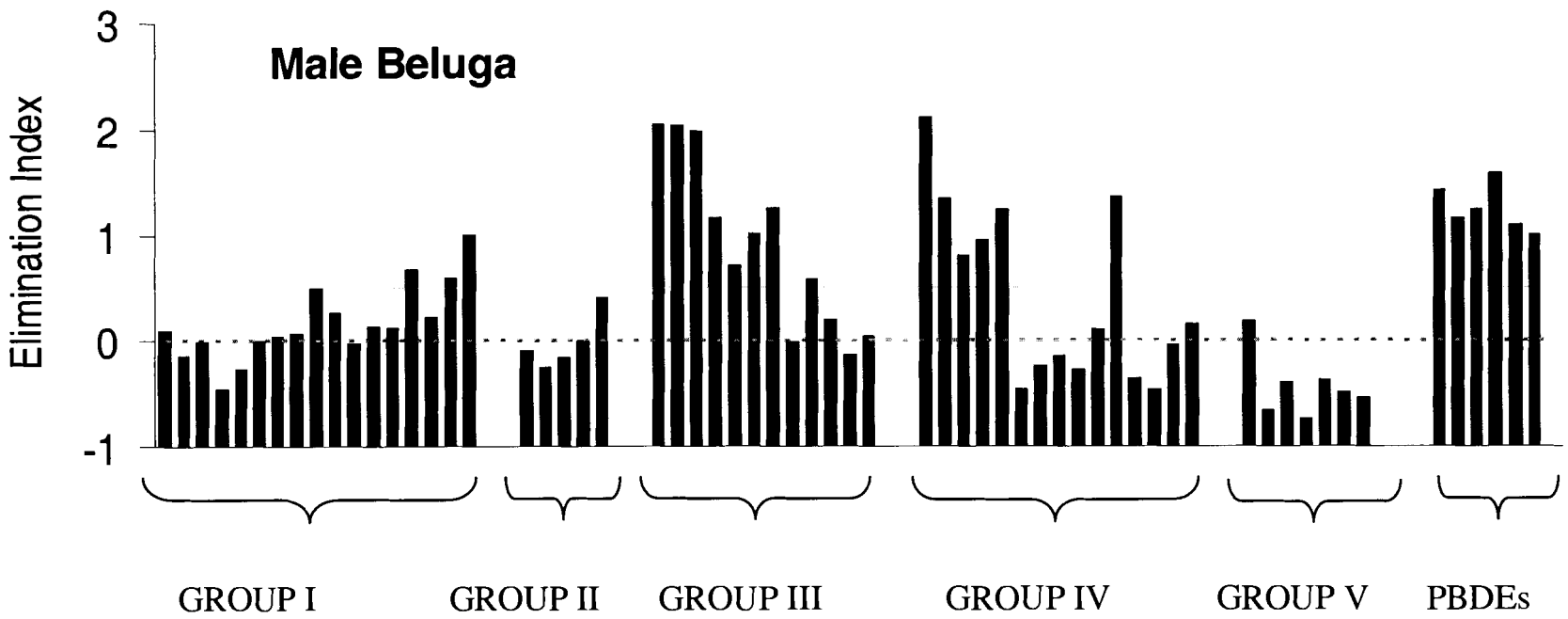
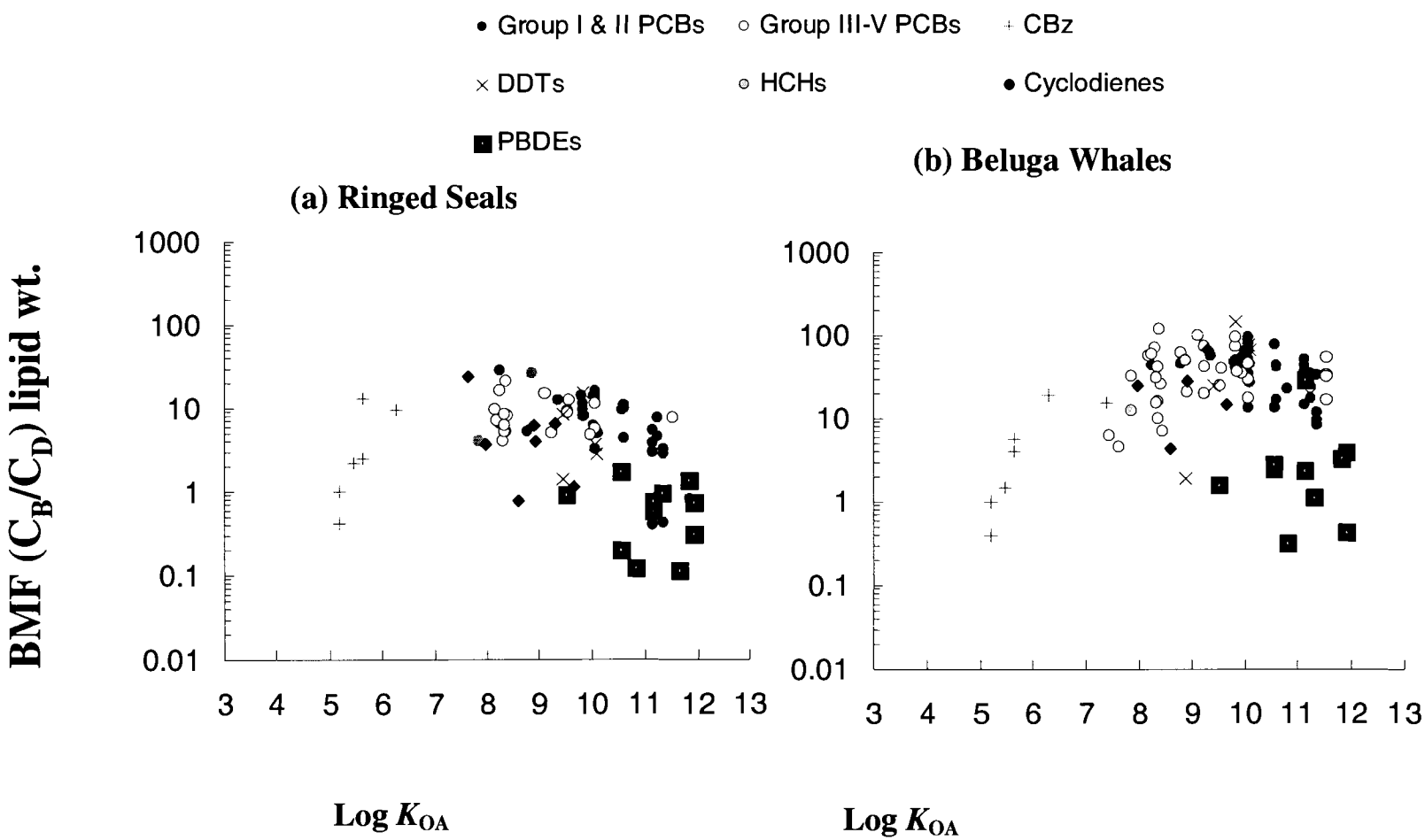


Figure 6.7 Relationships between observed BMFs (C_B/C_D) in male beluga and log K_{OA} for recalcitrant POPs such as PCBs and OC pesticides, with BDE BMFs plotted for comparison.



CHAPTER 7

HYDROXYLATED AND METHOXYLATED POLYBROMINATED DIPHENYL ETHERS IN A CANADIAN ARCTIC MARINE FOOD WEB: PBDE METABOLITES OR NATURAL BIOGENIC ORGANOHALOGENS?

7.1 Introduction

The extensive use of polybrominated diphenyl ethers (PBDEs) in commercial applications and products since the early 1980s has resulted in extremely high production volumes (HPVs) of these compounds in North America and Europe. Temporal and spatial trend studies of brominated diphenyl ethers (BDE) indicate concentrations of these compounds are exponentially increasing in the environment (*30,252*). BDEs can also undergo long range transport (LRT) and accumulate in relatively pristine regions such as alpine (*265*) and Arctic ecosystems (*30*), thousands of kilometres from point sources. BDEs do not exhibit dioxin-like toxicity but have been reported to negatively affect thyroid function (*266,267*) and are characterized as potential endocrine disrupting compounds (EDCs). Critical effects of penta-BDE technical mixtures on thyroid hormone levels and neurobehavioral development are established from 0.6 mg/kg body weight in rats and mice (*29*).

Recent studies have focused on *in vivo* debromination and/or oxidative metabolism and the subsequent formation of hydroxy (OH-) substituted PBDEs in laboratory animals (*239,268*). Hydroxy (OH-) and methoxy (MeO-) substituted BDEs have been identified in wild fish (*269*), birds (*270*), marine mammals (*10*) and humans (*271*). To date the most comprehensive field surveys of OH and MeO-BDEs compounds include a study of the Norwegian Arctic glaucous gulls (*Larus hyperboreus*) and polar bears (*Ursus maritimus*), (*262*) and a study of pelagic and benthic freshwater fish from the Detroit River (*272*). OH-BDEs are structurally very similar to the thyroid hormone thyroxine (T4), and hence may have negative impacts on thyroid function (*273,274*). Specifically, OH-BDEs can compete with the binding of T4 to plasma thyroid

hormone- transporter transthyretin (TTR) (*275*). 6-OH-BDE 47 has been identified as a primary metabolite of BDE47 *via* cytochrome P450 mediated metabolism and has recently been detected in human plasma (*271*) and has a high binding affinity to TTR (*276*). Thus, toxicokinetics of OH-BDEs may be an important factor in observed BDE congener toxicity.

While certain OH-BDEs have been confirmed as metabolites of various major BDE congeners (e.g., BDE47 → 6 OH-BDE-47), other hydroxylated and methoxylated bromodiphenyl ethers have been identified as naturally produced compounds (i.e., biogenic formation of organohalogens), originating from production *via* species of marine sponges and/or algae. Indeed, the diversity and numbers of naturally occurring organohalogens in the marine environment (including bromodiphenyl ethers) is vast (*277,278*). 2'-MeO-BDE68 (frequently detected in marine fish, mammals and seabirds) is most likely the result of biogenic formation due to the lack of a major BDE precursor (*269,279,280*). For example, 2'-MeO-BDE68 and 6-MeO-BDE 47 were detected in pike from Swedish waters and no correlation was found between PBDEs and methoxylated derivatives temporal trends (*279*), indicating the source of the MeO-Tetra-BDEs was of natural origin and from within a freshwater system. Radiocarbon ($\Delta^{14}C$) of isolated 2-(2',4'-dibromophenoxy)-3,5-dibromoanisole (6 MeO-BDE-47) and 2-(2',4'-dibromophenoxy)-4,6-dibromoanisole (2' MeO-BDE 68) of 10 kg of blubber sampled from a stranded True's beaked whale positively revealed that those compounds are naturally produced organohalogens and indicates that naturally produced methoxy BDEs can accumulate and persist in the food chain (*281*). However, numerous known naturally occurring OH- and MeO-BDEs may originate both from biogenic formation and anthropogenic sources (i.e., PBDE metabolism). Thus, the environmental presence of naturally occurring OH- and MeO-BDEs adds an additional layer of complexity in assessing the biotransformation of BDE congeners.

Our recent analyses of PBDE (Br_2-Br_7 congeners) concentrations in marine sediments and biota from the E. Hudson Bay in the Canadian eastern Arctic region showed BDEs exhibited very low biomagnification potential compared to recalcitrant PCB congeners (e.g., CB153, 180), which indicates substantial BDE debromination and/or biotransformation (*282*). Metabolic transformation of BDEs in E. Hudson Bay seaducks and marine mammals was supported by the observations that tetra and penta BDE congeners such as Br_4-BDE47, Br_5-BDE99 and Br_5-BDE100 exhibited metabolic index values (MIs > 1) comparable to known metabolizable PCB congeners (i.e., Group III-V CBs). In the present study we identify and quantify levels of OH- and MeO-BDEs in marine sediments and biota from E. Hudson Bay, Canada. Accumulation

patterns and potential origins (anthropogenic versus biogenic) of OH- and MeO-BDEs are discussed.

7.2 Materials and Methods

7.2.1 Sample collections.

During the months of May to August between 1999 and 2003 various biological samples were collected along the eastern Hudson Bay coastline in close proximity to the Inuit village Umiujaq (64° 15′N 113° 07′ W), (Figure 7.1). For details see *Chapter 1, Section 1.9.1* and Appendix 1, which summarizes information for individual seaducks and marine mammals sampled, including species, tissue/viscera type, collection date, sampling location, length, girth, sex, age and condition.

7.2.2 Food web characterization and designation of organism trophic levels.

Appendix 14 is a schematic illustration of common organisms and approximate trophic positions within the Arctic marine food web, including primary producers (i.e., lichens and macro algae), bivalves (blue mussels), fish (e.g., arctic cod) and marine mammals such as beluga whales, ringed seals, walrus polar bears and humans. Trophic levels (TL) of Canadian arctic marine biota have previously been established by extensive ^{15}N and ^{13}C isotope enrichment analyses involving numerous species of invertebrates, fish, seabirds and marine mammals from the eastern Canadian Arctic (*45*), resulting in the general equation of $TL = 1 + (\delta^{15}N - 5.4)/3.8$. More recent studies using $\delta^{15}N$ measurements to establish trophodynamics of several Arctic marine food webs include analyses of biota from marine food webs, including the Barents Sea (*46*), Northwater Polyna (*47,48*) and the Beaufort-Chukchi Seas (*49*). Table 1.1 (see *Chapter 1*) summarizes these previous $\delta^{15}N$ measurements and TL ranges for the various organisms within these Arctic marine food webs. For the purpose of the current study we utilized TL determinations in references *45,47,48* and assigned primary production matrices such as lichens and macro-algae a trophic level (TL) equal to 1.0 and Mollusca (i.e., bivalves) such as blue mussels were assigned at a TL of approx. 2.0. Specifically, fish included arctic cod (TL= 2.9), sculpin (TL = 3.6) and estuarine salmon (TL = 3.9). Seaducks included molluscivorous common eiders (TL= 2.8). Marine mammals include molluscivorous walrus (TL = 3.4), invertebrate/fish eating ringed seals (TL ~ 4.1) and beluga whales (TL = 4.7) and top-predator polar bears (TL = 5.5) that consume ~100% ringed seals. Several Inuit communities such as Umiujaq, Inukjuak and Akulivik substantially

utilize coastal E. Hudson Bay fish, birds and marine mammals for subsistence and hence likely occupy a TL somewhere between ringed seals polar bears in the region (i.e., TL = 4.5). It should be noted that these assigned trophic levels are best estimates in absence of sample-specific $\delta^{15}N$ measurements for the E. Hudson Bay marine biota and hence should be used with caution. However, these assigned trophic levels are supported by strong data from multiple Arctic marine systems and provides a general framework representing the trophodynamics of the E. Hudson Bay marine food web, including the algae → invertebrate → fish → avian/mammal trophic transfers.

7.2.3 Chemicals and reagents.

PBDE Analytical Standard Solution EO-4980 was purchased from Cambridge Isotope Laboratories, Inc. (MA, USA). The components of this solution were: 3 mono BDEs (BDE 1, 2 and 3), 7 diBDEs (BDE 7, 8, 10, 11, 12, 13 and 15), 8 triBDEs (BDE 17, 25, 28, 30, 32, 33, 35 and 37), 6 tetraBDEs (BDE 47, 49, 66, 71, 75 and 77), 7 pentaBDEs (BDE 85, 99, 100, 116, 118, 119 and 126), 5 hexaBDEs (BDE 138, 153, 154, 155 and 166) and 3 heptaBDEs (BDE 181, 183 and 190). The concentrations of each compound ranged from 100 pg/μL for the mono congeners to 250 pg/μL for the hepta congeners. Synthesised OH and MeO-PBDEs were: 2'-MeO-2,4,4'-BDE 28 (abbreviated name 2'MeO-BDE 28), 4'-MeO-2,2',4-BDE 17 (4'MeO-BDE 17), 4'-OH-2,2',4-BDE 17 (4'OH-BDE 17), 2'-MeO-2,4,4',6-BDE 75 (2' MeO-BDE 75), 6-MeO-2,2',4,4-BDE 47 (6MeO- BDE 47), 2'-MeO-2,4,4',5-BDE 74 (2'MeO-BDE 74), 6'-MeO-2,3',4,4'-BDE 66 (6'MeO-BDE 66), 2'-OH-2,4,4,6-BDE 75 (2'OH-BDE 75), 6-OH-2,2',4,4'-BDE 47 (6OH-BDE 47), 2'-OH-2,4,4',5-BDE 74 (2'OH-BDE 74), 6'-OH-2,3',4,4'-BDE 66 (6'OH-BDE 66). Stock solutions of OH and MeO-BDEs were prepared at 890-3110 μg/mL in nonane and working solutions at 500 ng·mL^{-1} in hexane. The surrogate standard solution EO-5100 (^{13}C labelled BDE) containing ^{13}C-DBE 3, ^{13}C-BDE 15 ^{13}C-BDE 28, ^{13}C-BDE 47, ^{13}C-BDE 99, ^{13}C-BDE 100, ^{13}C-BDE 118, ^{13}C-BDE 126, ^{13}C-BDE 153 and ^{13}C-BDE 183 at 100 to 250 ng·mL^{-1} was used to quantify both parent and OH and MeO-BDE. ^{13}C-BDE 77 from Cambridge Isotope Laboratories was used as internal standard. Solvents used and H_2SO_4 were from Merck (Germany).

7.2.4 Extraction and cleanup of BDEs.

Details of our methods for PBDE analysis of environmental and biological samples and QA/QC procedures are detailed fully in reference *246*. Briefly, tissue samples (approximately 10 g wet

wt for lichens, macro-algae and sediment, 5-15 g for fish, 2 g for beluga whale liver and 0.5 g for blubber (beluga whales and ringed seals) were homogenized with approximately 20 g Na_2SO_4 with mortar and pestle. Sub-samples of other tissue samples (e.g., seaduck and marine mammal tissue samples) were excised from the interior of frozen samples to reduce potential contact contamination during collection and/or storage. The homogenate powder was transferred to a glass extraction jar, spiked with ^{13}C-labeled procedural internal standards (Cambridge Isotope Laboratories, Andover, MA), approx. 2000-5000 pg of each ^{13}C BDEs (^{13}C BDEs 3,15, 28 47, 77, 118, 99, 100,153 and 183). The spiked samples were then extracted with 30 mL of 1:1 (v/v) DCM/Hexane in a Branson 5210 ultrasonic water-bath (Branson Ultrasonics Co., CT) for 20 min. Once the suspended particles settled, the supernatant was removed and then extraction was repeated two more times with fresh solvent. The combined extracts were concentrated to ca. 2 mL with a gentle stream of high-purity nitrogen. The lipid content was determined gravimetrically on sub-samples of the extracts and 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 1 g of sample at 125 ° C for 24 hr. Relatively low lipid samples (< 5% lipid w/w) such as cod and sculpin tissue were quantitatively transferred onto a 350 mm x 10 mm i.d. glass column packed with 8 g 100% activated Florisil (60 -100 μm mesh, activated at 400 C overnight). High lipid samples (>5% lipid w/w) such salmon, and beluga blubber were first passed through a Gel Permeation Column (GPC) filled with 70 g of BioBeads, S-X33 (BioRad) in 50% DCM/hexane solution (V/V). The lipid fraction from the GPC (180 mL) was collected and discarded, while the remaining 300 mL of eluent from the GPC was collected evaporated to near dryness and solvent exchanged into hexane for further cleanup by Florisil. Three fractions were then eluted using 60 mL hexane (fraction 1), 60 mL 15% DCM/hexane (fraction 2), and 120 mL 50% DCM/hexane (fraction 3). The four fractions were combined in a single 500 mL boiling flask and evaporated to a final volume of 100 μL.

7.2.5 Extraction and cleanup of OH-BDEs and MeO-BDEs.

Details of the methods we employed for analysis of OH- and MeO-BDEs are presented in reference *283*. Briefly, tissue samples (approximately 10 g wet wt for lichens, macro-algae and sediment, 5-15 g for fish, 2 g for beluga whale liver and 0.5 g for blubber (beluga whales and ringed seals) were homogenized with approximately 20 g hydromatrix with mortar and pestle. The homogenate powder was spiked with ^{13}C-labeled procedural internal standards (Cambridge Isotope Laboratories, Andover, MA), approx. 2000-5000 pg of each ^{13}C BDEs (^{13}C BDEs 3,15,

28 47, 77, 118, 99, 100,153 and 183), surrogate spiking solution and extracted using pressurized liquid extraction (PLE) with an accelerated solvent extraction (ASE) apparatus, ASE 2000 (DIONEX, USA) using hexane:CH_2Cl_2 (2:1 v/v) at 2000 psi and at a temperature of 100°C (100% flush volume) with a heat-up time of 5 min. Two cycles of extraction were performed during 5 min in static mode. The purge time was of 100 s and the extraction cell volume was of 11 mL. The lipid content was determined gravimetrically from a parallel PLE extraction using the same solvents. To remove bulk lipids, samples were first passed through a large scale manual GPC column-consisting of 70 g BioBeads S-X33 (BioRad) in 1:1 CH_2Cl_2:Hexane (V/V). The lipid fraction from the GPC (180 mL) was collected and discarded, while the remaining 300 mL of eluent was collected evaporated to near dryness and solvent exchanged into hexane for further cleanup by Florisil. A 30 cm column was wet-packed with 8 g of 1.2% deactivated Florisil in hexane. The rotary-evaporated extract was quantitatively placed at the top of the column in 1:1 CH_2Cl_2: Hexane (V/V) and compounds were eluted with 60 ml of 1:1 CH_2Cl_2:Hexane (V/V) and 20 ml of CH_2Cl_2. Since OH-BDE metabolites showed poor response under HRGC/HRMS, sample extacts and also calibration standards for OH-BDE quantification (in parallel) required derivatization. Thus, the resulting extract was evaporated again to approximately 0.5 ml and transferred with toluene, from microvials into centrifuge tubes for the purpose of derivatization (i.e., acetylation) of OH-BDEs. Derivatization was performed as follows: the transferred samples were dissolved in 500 μl of toluene and then 100 μl of pyridine and acetic anhydride were added to the sample. The sample was vortexed for 2 min and heated at 60°C for 30 min. After derivatization, 700 μL of 2x toluene washed water was added to pull out the reaction by-products and left over reagents. The sample was vortexed and back extracted into another centrifuge tube using 3 hexane washes. The extracted sample was passed through a Pasteur pipette filled with hydromatrix to remove any water. The sample was then nitrogen evaporated to 100 μL, transferred to a microvial, nitrogen evaporated to almost dryness and reconstituted in CH_2Cl_2.

The resulting extract was evaporated again to approximately 0.5 ml. OH-BDEs were derivatized (i.e., acetylated) by addition of 100 μl of pyridine and acetic anhydride, 2 min vortex mixing, and 30 min on heating block at 60°C. Following derivatization, 700 μL of 2x toluene washed water was added to pull out the reaction by-products and left over reagents. The sample was vortexed and back extracted into another centrifuge tube using 3 hexane washes. The extracted sample was passed through a Pasteur pipette filled with hydromatrix to remove any water. The sample was then nitrogen evaporated to 100 μL, transferred to a microvial, nitrogen evaporated to almost

dryness and reconstituted in 100 μL of CH_2Cl_2. At this stage, the 100 μl of IS BDE#77 was added giving a total amount of 1200 pg.

7.2.6 Instrumental Analysis.

Analysis of both PBDE and OH and MeO-PBDEs were performed on a HRGC/HRMS using a VG-AutoSpec-S (Micromass, Manchester, UK) equipped with a Hewlett Packard model 5890 series II gas chromatogram (Agilent, Palo Alto, California, USA) and a CTC A200S autosampler (CTC Analytics, Zurich, Switzerland). The GC was operated in the splitless injection mode and the splitless injector purge valve was activated 2 min after sample injection. For PBDEs, a 30 m DB-5 column (0.25 mm i.d. x 0.25 μm film thickness from J&W Scientific (Folsom, CA) was used with UHP He as the carrier gas at a constant head pressure of 25 psi. The temperature program was from 100°C (held for 2 min) to 320°C (held for 2.5 min) at 4°C/min. The injector port, GC-MS interface and the MS ion source were maintained at 300, 270 and 310°C, respectively. For PBDEs, specific analytical conditions are described elsewhere (3). Briefly, quantification of PBDEs was determined by high resolution gas-chromatography (HRGC/HRMS) using a Micromass Ultima HR-mass spectrometer coupled with an HP 5890 Series II GC and a CTC A200S autosampler (CTC Analytics, Zurich, Switzerland). The GC column used was a 15 m high temperature DB-5-HT (0.225 mm i.d. × 0.1 μm film thickness). The HRGC was operated in splitless mode was used with the purge valve being activated 2 min following sample injection. Ultra high purity helium at 80 kPa was used as the carrier gas using the following temperature program: hold at 100 °C for 1 min, 2 °C min^{-1} to 140 °C, 4 °C min^{-1} to 220 °C, 8 °C min^{-1} to 330 °C and hold for 1.2 min. For all analyses, the MS was operated at 10,000 resolution in the positive ion mode at 39 eV energy and data were acquired in the single ion resolving mode (SIR). Analytes were identified by retention time (RT) comparison relative to authentic calibration standards. For Br_1 and Br_2 homologues and Br_4-BDE77, the two most abundant isotopes representing the parent ion [M^+] were monitored. For all other homologues (Br_3-Br_7 congeners) the two dominant isotopes representing the [M-2Br]$^+$ fragment were monitored. Quantification ions were *m/z* 323.8785 for Br_4-BDEs, 403.7870 for Br_5-BDEs, 481.6975 for Br_6-BDEs and 561.6060 for Br_7-BDEs. Concentrations were calculated by the internal standard isotope dilution method using mean relative response factors (RRFs) determined from a calibration standards, run prior to and following sample analyses. A total of 31 individual mono- to hepta- PBDE congener peaks and three co-eluting bands (each composed of two congeners) were identified and quantified, establishing the initial data set of 37 congeners overall: BDE-1, -2, -3, -7, -8/11, -10, -

12, -13, -15, -17, -25, -28/33, -30, -32, -35, -37, -47, -49, -66, -71, -75, -77, -85, -99, -100, -116, -119, -138/166, -140, -153, -154, -155, -181, -190. PBDEs were quantified using ^{13}C BDE 28, 47, 100, 99, 153 and 183 as surrogate standards and ^{13}C BDE 77 as internal recovery standard.

Identification of OH- and MeO-BDEs in biota sample extracts was conducted by relative retention time (RRT) comparisons to authentic synthesized OH and MeO-BDE reference standards by high-resolution gas-chromatography mass spectrometry (HRGC/HRMS). OH- and MeO-BDEs were quantified by the internal standards isotope dilution method using mean relative response factors (RRFs) calculated from derivatized calibration standards. OH and MeO-BDE calibration standards (consisting of native OH and MeO-BDEs + ^{13}C BDE47, ^{13}C BDE100 and ^{13}C BDE77 labelled surrogates) were derivatized in-parallel with extracted samples (see above descrition of acetylation procedure). Specifically, quantification of acetyl OH-BDE derivatives and MeO-BDEs were performed using also the isotope dilution method using ^{13}C BDE47 to quantify -OH and MeO- BDE47 and ^{13}C BDE100 to quantify the rest of the compounds, with ^{13}C BDE 77 as internal recovery standard. Derivatization of OH-BDEs did not affect MeO-BDEs nor the isotope-lablelled surrogate standards (i.e., ^{13}C BDE congeners #47, #100 and #77), as the response in the derivatized and underivatized standard solutions were equivalent. The derivatized standard solution was stable for 7 days, keeping the standards at room temperature (20°C). Thus, new OH- and MeO-BDE calibration solutions were prepared for subsequent acetylation quantification by HRGC/HRMS.

For OH and MeO-BDEs, we used a 30 m DB5 (0.25 mm i.d. x 0.25 μm film thickness from J&W Scientific (Folsom, CA) and a second polar column SP 2331 (30m x 0.25 mm i.d x 0.2 μm film thickness) for peak confirmation and resolution of coeluting compounds. For the DB5 column, conditions used were as follows: 80°C (held for 2 min) to 300°C (held for 10 min) at 10°C/min. The injector, interface and source temperatures were set at 260, 260 and 300°C, respectively. These conditions were used to identify other OH and MeO-PBDEs, for which we did not have standards, through retention time calculation and at the exact mass. In all cases, 1 μl of sample was injected using the splitless injection mode with a splitless time of 1 min. The MS was operated under positive electron ionization conditions with the filament in the trap stabilization mode at 600 μA, an electron energy of 35 eV and perfluorokerosene as calibrant. The instrument was operated at a resolving powder of 10,000 and data were acquired in the selective ion monitoring mode (SIM) mode monitoring the molecular peak [M]+ for mono, di, and BDE 77 and the $[M-2Br]^+$ for or the rest of compounds. Quantification and corresponding confirmation

ions monitored for hydroxylated and methoxylated BDEs included *m/z* 435.8133 and 437.8113 for MeO-Br_3-BDEs; *m/z* 421.7976 and 423.7956 for OH-Br_3-BDEs; *m/z* 515.7217 and 513.7237 for MeO-Br_4-BDEs; *m/z* 501.7061 and 499.7081 for OH-Br_4-BDEs; *m/z* 595.6303 and 593.6323 for MeO-Br_5-BDEs; *m/z* 581.6146 and 579.6166 for OH-Br_5-BDEs.

7.2.7 Quality control.

A total of 23 OH BDEs and 23 corresponding MeO-BDEs were monitored (Table 7.1). Identification and confirmation criteria for PBDE and OH and MeO-PBDE involved: (i) two isotopes of the specific congener were detected by their exact mass at 10,000 resolving power; (ii) the retention time of target compounds was within 3 s to that of a standard; (iii) the peak maxima for both characteristic isotopic ions of a specific congener was within 2 s; (iv) the isotope ratio of the two ions monitored per congener was within 15% of the theoretical isotopic ratio and (v) the signal to noise ratio for both ions of a specific congener was > 3. PBDEs and OH and MeO-PBDEs were quantified by the internal standards isotope dilution method using mean relative response factors (RRFs) calculated from calibration standards. PBDEs were quantified using ^{13}C BDE 28, 47, 100, 99, 153 and 183 as surrogate standards and ^{13}C BDE 77 as internal standard. Quantification of OH and MeO-PBDEs was performed using the isotope dilution method using ^{13}C BDE47 to quantify OH- and Meo-BDE47 and ^{13}C BDE100 to quantify the rest of the OH- and MeO-BDE compounds. Limits of quantitation (LOQ), calculated as a mean of the noise plus three times the standard deviation were approximately 0.005-0.2 $ng \cdot g^{-1}$lipid for OH-BDEs and approximately 0.006-0.05$ng \cdot g^{-1}$lipid for and MeO-PBDEs, respectively (Table 7.1). Method blanks, consisting of Na_2SO_4 (for BDE analysis) or hydromatrix (for OH-and MeO-BDEs), were extracted according to the same procedure as environmental samples and analyzed with every batch of 12 samples to check for contamination of the extracts.

7.2.8 Data treatment/compilation and statistics.

To enable direct comparisons of POPs between different environmental media and organisms it is important to correct chemical concentration data to a common unit expression such as lipid equivalent concentrations. For samples with relatively high lipid fraction (ϕL), e.g., fish, seaduck and marine mammal tissues (ϕL ~1 - 98%) , wet weight chemical concentrations (C, $ng \cdot g^{-1}$ ww) were expressed solely on a lipid weight basis by the equation: C_L = C ww. ÷ ϕL in units of $ng \cdot g^{-1}$ lipid. For some biological matrices with very low lipid fractions (ϕL < 1%), such as vegetation

and algae tend to solubilize organic contaminants in non-lipid biomolecules (i.e., non-lipid organic matter, NLOM) rather than in extractable lipids (*13,57,58,59*). Thus, for macro-algae and lichens, the lipid equivalent fraction was determined as the sum of lipid (ϕL) and NLOM (ϕ_{NL}) fractions following the equation: $\phi Leq = \phi L + 0.035\phi_{NL}$, where the constant 0.035 demonstrates observations that NLOM has approximately 3.5% sorptive capacity of octanol (*42,44*). Because chemical concentrations exhibited log-normal distributions and were hence transformed logarithmically to reduce variance heterogeneity. Geometric means (GM) and the geometric standard deviation (GSD) and 95% confidence limits (CL) were determined for target analytes in the various organisms collected and analyzed as part of the present study (i.e., lichens, macro-algae, bivalves, fish, beluga whales and ringed seals).

7.2.9 Biomagnification Factors (BMF) and Food Web Magnification Factors (FWMF).

See *Chapter 1, Section 1.9.5*

7.3 Results and Discussion

7.3.1 Identification of OH- and MeO-BDEs in environmental and biological samples.

Table 7.1 summarizes the suite of 46 OH- and MeO-BDE compounds we currently monitor by HRGC/HRMS, along with corresponding relative retention times (RRTs), i.e., relative to BDE-47, on the 30 m DB-5 column and limits of quantitation (LOQ) in beluga blubber. RRTs of the eleven compounds for which we have synthesized reference standards (compounds in bold) were determined by direct comparison of RTs to RT of BDE-47, present in our OH- and MeO-BDEs calibration solution. RRTs for the other OH- and MeO-BDEs, which are not currently present in our calibration solution, were determined from previously reported OH- and MeO-BDE RRTs from reference *269*. Positive identification and resolution of coeluting OH- and MeO-BDEs were determined by duplicate analysis using the polar GC column (SP-2331) and subsequent RT shifting behaviour of analytes of interest.

7.3.2 Levels and congener profiles of BDEs, OH-BDEs and MeO-BDEs.

Concentrations ($ng.g^{-1}$ lipid) of major BDE congeners and OH- and MeO-BDEs in E. Hudson Bay samples are summarized in Appendix 20. The data are not blank subtracted as procedural blanks for OH- and MeO-BDEs were generally low or non-detectable. Method detection limits

(MDLs) were determined as the instrument limit of quantification (LOQ) on the HRMS. In general, MeO-BDEs were regularly detected at appreciable quantities in biota samples, while OH-BDEs were detected at very low concentrations and only in marine mammal tissue samples. A wide variety of tri to penta OH- and MeO-BDEs were detected in biota samples, including several *ortho*, *meta* and *para* substituted OH- and MeO-BDE congeners. Figure 7.2 illustrates mean concentrations of several OH-BDEs, MeO-BDEs and parent PBDE compounds detected in E. Hudson Bay male beluga blubber. Relatively high levels of MeO Tetra BDEs were observed in beluga whales from the eastern Canadian Arctic, e.g., 6 MeO-BDE-47 levels were approx. 200 $ng \cdot g^{-1}$ lipid in beluga blubber (Figure 7.2), which is comparable to 6 MeO-BDE47 concentrations reported in gray and ringed seals (95-160 $ng.g^{-1}$ lipid) from the Baltic (*10*), but is substantially less than that measured in the True's Beaked whale (approx. 900 $ng.g^{-1}$ lipid wt.) from the north Atlantic (*281*). In general, MeO-BDE levels were equivalent or greater than parent PBDE congeners. OH-BDEs were detected at low concentrations (i.e., ~ 0.01 $ng \cdot g^{-1}$ lipid) in beluga whale blubber. OH- BDEs were only detected in marine mammals (beluga whales and ringed seals). MeO-BDEs were detected in fish, seaducks and marine mammals, but were not found in ambient samples of sediments, lichens and macro-algae, indicating no local natural sources of MeO-BDEs is apparent, at least in the E. Hudson Bay region.

Figure 7.3 illustrates the BDE congener compositions (i.e., % contributions for Br_3-Br_7 congeners+ 6 MeO-BDE47) in the commercial BDE formulation Bromkal® in comparison to the observed composition pattern in E. Hudson Bay sediments and biota, along with corresponding observed BDE concentrations ($ng \cdot g^{-1}$ lipid equivalent) in those media. The BDE profiles in lichens (collected on land in close proximity to marine sampling locations) can be viewed as an atmospheric "signal" resulting from air-borne contaminant exposure processes. Similarly, contaminant profiles shown for sediments and macro-algae represent an aquatic "signal" of water-borne chemical in the marine system, while those profiles for biota are indicative of food web bioaccumulation processes and subsequent chemical residue distributions in organism tissues. The observed BDE composition profiles in lichens (representing the ambient atmospheric signal) and sediments and macro-algae (representing the ambient aquatic signal) are dominated by BDE99 and have previously been shown to be comparable to the commercial Bromkal® formulation profile (*282*). However, for bivalves, fish, seaducks and marine mammals (i.e. the food web pattern), the BDE congener pattern changes and generally predominates in the order Br_4-BDE47 > Br_5-BDE99 > Br_5-BDE100, indicating debromination towards lower brominated congeners. No MeO-BDEs are present in commercial BDE mixtures

or were detected in the ambient environmental samples from E. Hudson Bay (i.e., lichens, sediments or macro-algae). However, the relative contribution of Br_4-MeO-BDEs (e.g., 6 MeO-BDE47) is shown to increase while BDE-47 amounts diminished with trophic level of the Arctic food web. This is particularly true for beluga whales where 6 MeO-BDE47 represents almost 90% of the total BDE burden, while BDE47 only comprises approximately 8% of the BDE burden.

7.3.3 Trophic Transfer and Food Web Magnification.

Figure 7.4 shows concentrations of (a) PCB153, (b) BDE 47 (c) BDE99, (d) BDE47 and (e) 6 MeO-BDE-47 in selected organisms of the E. Hudson Bay marine food web. 6 MeO- BDE-47 ($ng \cdot g^{-1}$ lipid) increased with each step-wise increase in trophic level, exhibiting similar biomagnification potential as recalcitrant PCBs (e.g., PCB 153). For example, the food web magnification factor (FWMF) of 6 MeO-BDE47 in the E. Hudson Bay food web was approximately was 7.4, which is comparable to the FWMF previously determined for PCB153 (FWMF = 11.0). Similarly, the BMF of 6 MeO-BDE47 in male beluga (beluga/cod) was approx. 47.8 (comparable to the BMF_{MAX}, i.e., CB180 BMF of approx. 45.7). Other MeO- Tetra BDEs (e.g., 2' MeO-BDE68, 6' MeO-BDE-49 and 6 MeO-BDE-99) demonstrated similar high biomagnification potential. Conversely, major BDE congeners -100, -99 and -47 exhibit similar concentrations across trophic levels, indicating debromination / biotransformation and thus trophic dilution.

7.3.4 Potential Sources of OH- and MeO-BDEs in Arctic Biota.

Table 7.2 shows the predominant tri-hexa OH and MeO-BDEs previously reported in (i) tissues of BDE exposed laboratory animals, (ii) field-collected abiotic and biota samples and (iii) isolated as natural products from marine sponges. The only confirmed metabolites determined from controlled exposure studies using BDE-47 include 4'-OH-BDE-49, 4-OH-BDE-42, 6'OH-BDE-49, 3-OH-BDE-47, 6-OH-BDE-47 and 6'-OH-BDE66, indicating the dominant hydroxylated metabolic products of BDEs appear to be *meta* and *para* substituted OH-BDEs (Table 7.2). MeO-TeBDE have been reported in herring, grey and ringed seals, salmon, fish oil at levels between 0.1 to 158 $ng.g^{-1}$ lipid wt. but not in human adipose tissue (*10*). Among various OH- and MeO- substituted BDEs, Marsh *et* al. identified 6-OH/MeO-BDE47 and 6'OH/MeO-BDE49 and 6-OH-BDE-99 in Baltic Sea salmon blood as metabolites of corresponding PBDE congeners

(*269*). Tri and MeO- Tetra-BDEs were identified in salmon, guillemot and Artic cod liver at levels between 0.02 and 16 $ng.g^{-1}$ lipid, although their origin remained unknown (*280*). More recently Ueno *et* al. (*284*) reported detection of several di to pentabromo OH-BDEs in abiotic samples (including surface water, rainfall and snowfall) near urbanized locations in southern Ontario, Canada. The authors suggest the likely source of OH-BDEs in these abiotic samples is from PBDEs entering nearby wastewater treatment plants (WWTPs) either via microbial oxidation or reaction with OH radicals in ozone treated effluents. Interestingly, the majority of the OH-BDEs suspected as point source BDE degradation products in abiotic samples have the hydroxy group substituted in the *ortho* position of the phenyl ring, similar to OH- and MeO-BDEs suspected as natural marine products. These recent findings of an urban point source of OH-BDEs further confounds the distinction of hydroxylated and methoxylated BDEs in the environment and food chains.

2'-MeO-BDE68 and 6-MeO-BDE47 were detected up to 3.6 $ng.g^{-1}$ lipid in pike from Swedish waters and no correlation was found between PBDEs and metabolites (*279*), indicating that the source was other than PBDE metabolism. Verreault et al. (*262*) recently demonstrated very high correlation coefficients between 6 OH-BDE47 and 6 MeO-BDE 47 and also between 6 OH/MeO-BDE47 and the parent BDE47 in glaucous gulls from the Norwegian Arctic, which they indicate as evidence of a natural origin of 6 OH/MeO-BDEs in glaucous gulls. However, regression analyses showing high correlation between parent compound may also be indicative of BDE metabolism as well. Figure 7.5 shows the relationship between (a) BDE-47 and 6 MeO-BDE47 and (b) *p,'p'* DDT and *p',p'* DDE for E. Hudson Bay male beluga whales (this study). Strong correlation was observed between 6 MeO-BDE47 and BDE47 ($r^2 = 0.801$), which is similar to the correlation found between *p,'p'* DDT and its primary metabolite *p',p'* DDE ($r^2 = 0.922$).

Figure 7.6 illustrates levels and patterns (% composition) of BDE47, 100 and 6 MeO-47 in marine animals (seabirds and marine mammals) of the E. Hudson Bay (Canadian Arctic), compared to those animals from the Norwegian Arctic, Baltic, Australia and N. Atlantic. The BDE and MeO-BDE data from these various studies indicate there is a great variability in (i) concentrations of BDE and methoxylated derivatives among marine animals around the world and also (ii) the % contribution of the MeO-BDE relative to the parent BDE. Previous studies of OH- and MeO-BDEs in fish and wildlife have suggested sources of dominant hydroxy and methoxy BDEs (e.g., 6 MeO-BDE-47) are biogenic organohalogens, originating from marine

sponges and/or algae. This appears to be particularly true for OH- and MeO-BDEs with the hydroxylation and/or methylated substitution in the *ortho* position.

The current understanding in this rapidly progressing area of research is that six major OH-BDEs are formed *in vivo* following metabolism of BDE47 (either in the liver or intestinal tract). These OH-Tetra-BDEs have been identified as 4′-OH-BDE-49, 4-OH BDE-42, 6′OH-BDE-49, 3-OH-BDE-47, 6-OH-BDE-47 and 6′-OH-BDE66 (*239*). Of these six confirmed BDE metabolites, 6-OH-BDE-47 has also been shown occur *via* biogenic formation (*285*). To our knowledge, there have been no reports of MeO-BDE formation *via* BDE metabolism and hence these are generally regarded as naturally occurring compounds that can bioaccumulate due to a high estimated octanol-water partition coefficient (K_{OW}). For example, Teuten et al., (*281*) recently estimated the log K_{OW} of MeO-Tetra-BDEs is ~6.85 (comparable to Cl_6-PCBs). However it has been postulated OH-BDEs may be methylated in the intestinal tract of organisms or within marine sediments (i.e., similar to microbial formation of methyl mercury), (*10*). The former mechanism is particularly intriguing due to the likely elimination of relatively polar OH-BDEs to the intestine *via* expelled bile following digestion and absorption. Thus, it is plausible that commonly detected MeO-BDEs such as 6 MeO-BDE47 may be a methylation product of 6 OH-BDE47 (a confirmed BDE metabolite). However, the majority of studies reporting this dominant tetra MeO-BDE in biota samples generally attribute its presence to biogenic formation rather than BDE metabolism. In the present study of PBDEs, OH-BDEs and MeO-BDEs in a Canadian Arctic marine food web suggests that the detected OH- and MeO-BDEs are *in vivo* biotransformation products of PBDEs due to the fact we observed (i) no biomagnification of parent PBDEs and (ii) no measurable quantities of OH- or MeO-BDEs in ambient environmental samples (i.e., sediments and macro-algae). However, further investigation into OH- and MeO- BDEs in Arctic marine food webs is required to fully assess their fate and bioaccumulation behaviour and potential sources of these compounds to the Arctic marine environment.

7.4 Acknowledgements

We acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada, the Association of Canadian Universities for Northern Studies and wish to thank Dr's Bill Doidge, Michael Kwan and Susan Sang, Derek Muir and Northern Quebec Inuit communities of Umiujaq and Inukjuaq for coordinating/aiding collection of field samples.

7.5 Tables

Table 7.1 List of OH- and MeO-BDEs, RRTs (relative to BDE-47) on standard 30 m DB-5 column (relative to BDE-47) and LOQs ($ng \cdot g^{-1}$) for HRGC/HRMS analysis.

OH-BDEs	RRT	LOQ $ng \cdot g^{-1}$ (ww)	MeO-BDEs	RRT	LOQ $ng \cdot g^{-1}$ (ww)
6' OH-BDE-17	0.983	0.017	6' MeO-BDE-17	0.948	0.014
4' OH-BDE-30	0.984	0.017	4' MeO- BDE-30	0.950	0.014
2' OH-BDE-28	0.996	0.017	**2' MeO- BDE-28**	**0.966**	**0.012**
3' OH-BDE-28	1.010	0.017	3' MeO- BDE-28	0.986	0.012
4' OH-BDE-17	**1.010**	**0.017**	**4' MeO- BDE-17**	**0.986**	**0.015**
6' OH-BDE-49	1.039	0.0064	6' MeO- BDE-49	1.024	0.018
2' OH-BDE-68	1.050	0.0064	2' MeO- BDE-68	1.038	0.018
2'-OH-BDE-75	**1.057**	**0.005**	**2'-OMe-BDE-75**	**1.041**	**0.018**
6 OH-BDE-47	**1.061**	**0.008**	**6 MeO -BDE-47**	**1.054**	**0.014**
4' OH-BDE-69	1.064	0.006	4' MeO- BDE-69	1.058	0.016
3 OH-BDE-47	1.074	0.006	**2'-OMe-BDE 74**	**1.063**	**0.049**
2' OH-BDE-66	1.075	0.006	3 MeO-BDE-47	1.072	0.018
5' OH-BDE-47	1.078	0.006	2' MeO-BDE-66	1.073	0.018
4' OH-BDE-49	1.080	0.006	5' MeO-BDE-47	1.077	0.018
2'-OH-BDE 74	**1.081**	**0.007**	**6' MeO-BDE-66**	**1.079**	**0.017**
6' OH-BDE 66	**1.089**	**0.007**	4' MeO-BDE-49	1.079	0.018
4' OH-BDE-121	1.109	0.006	4' MeO-BDE-121	1.118	0.006
4 OH-BDE-42	1.111	0.006	4 MeO-BDE-42	1.121	0.018
6 OH-BDE-90	1.122	0.006	6 MeO-BDE-90	1.136	0.006
6 OH-BDE-99	1.125	0.006	6 MeO-BDE-99	1.140	0.006
4 OH-BDE-90	1.150	0.006	4 MeO-BDE-90	1.173	0.006
2 OH-BDE-123	1.156	0.006	2 MeO-BDE-123	1.181	0.006
6 OH-BDE-85	1.167	0.006	6 MeO-BDE-85	1.197	0.006

Table 7.2 Compilation of major OH- and MeO-BDEs reported in the literature, including samples from laboratory experiments (rats and fish) and field-surveys (i.e. environmental samples: marine sponges, algae, mussels, freshwater and marine fish, marine mammals, birds and humans), including indication of likely sources (i.e., biogenic formation vs. in vivo metabolic formation) and potential BDE precursors.

Compound	Field and Laboratory Observations	Likely Source(s)	Potential Precursor
OH- and MeO-Br_3 Analogues			
6′-OH-BDE-17	**Environment:** red algae/mussels (*285*); Marine Fish (this study), marine mammals (this study), human milk Spain (*283*)	Biogenic/ Metabolite	BDE-28
4′-OH-BDE-17	**Environment**: marine mammals (this study), human milk Spain (*283*)	Metabolite	BDE-28
2′OH-BDE-28	**Environment**:; glaucous gulls (*262*); human milk Spain (*283*)	Metabolite	BDE-28
6′- MeO-BDE-17	**Environment**: human milk Spain (*283*)	Metabolite	BDE-28
2′ MeO-BDE-28	**Environment**: glaucous gulls (*262*)	Metabolite	BDE-28
4′ MeO-BDE-17	**Environment**: human milk Spain (*283*)	Metabolite	BDE-28
4′ MeO-BDE-30	**Environment**: human milk Spain (*283*)	Metabolite	BDE-28
OH- and MeO-Br_4 Analogues			
4-OH BDE-42	**Environment**: Freshwater fish (*272*) polar bears (*262*) **Laboratory**: Rat (*243*), fish (*239*)	Confirmed Metabolite	BDE-47

Compound	Field and Laboratory Observations	Likely Source(s)	Potential Precursor
4′-OH-BDE-49	**Environment**: Surface water (*284*), Rainfall (*284*), Freshwater fish (*272*)*glaucous gulls (262); polar bears (262)* **Laboratory:** Rat (*243*), fish (*239*)	Confirmed Metabolite	BDE-47
6′OH-BDE-49	**Environment**: Surface water (*284*), Rainfall (*284*), glaucous gulls (*262*) **Laboratory:** Rat (*243*), fish (*239*);	Confirmed Metabolite	BDE-47
2′ OH-BDE-68	**Environment**: sponge (*286,287,288,289*); red algae/mussels (*285*); Surface water (*284*), Rainfall (*284*), Freshwater fish (*272*); Marine fish (*269*) glaucous gulls (*262*)	Biogenic	None
3-OH-BDE-47	**Environment:** Freshwater fish (*272*);glaucous gulls (*262*) **Laboratory:** Rat (*243*), fish (*239*);	Confirmed Metabolite	BDE-47
6-OH-BDE-47	**Environment:** sponge (*287,288,289*); Red algae/mussels (*285*); Surface water (*284*), Rainfall (*284*), Freshwater fish (*272*); Marine fish (*269*); glaucous gulls (*262*) human plasma Sweden (*271* **Laboratory:** Rat (*243*), fish (*239*)	Confirmed Metabolite	BDE-47
2′-OH-BDE75	**Environment:** human milk Spain (*283*)	Metabolite	BDE-100
2′-OH-BDE74	**Environment**: human milk Spain (*283*)	Metabolite	BDE-100
6′-OH-BDE66	**Environment:** human milk Spain (*283*) **Laboratory:** Rat (*243*), fish (*239*)	Confirmed Metabolite	BDE-47
6′ MeO -BDE-49	**Environment**: glaucous gulls (*262*)		BDE-47/ BDE-49

Compound	Field and Laboratory Observations	Likely Source(s)	Potential Precursor
2′ MeO -BDE-68	**Environment:** Sponge (*290,291*); Red algae/mussels (*285*); Freshwater fish (*272*); Marine fish (*269*); glaucous gulls (*262*) ; cetaceans (*151,292*))seals (*10*) polar bears (*262*); human milk (*277*)	Biogenic	None
3- MeO -BDE-47	**Environment:** glaucous gulls (*262*); polar bears (*262*)	Metabolite	BDE-47
6- MeO -BDE-47	**Environment:** Sponge (*293*); Red algae/mussels (*285*);Freshwater fish (*272*); Marine fish (*269*); glaucous gulls (*262*), cetaceans (*151,292*), seals (*10*); polar bears (*262*)	Biogenic/ Metabolite	BDE-47
2′-MeO-BDE75	**Environment**: human milk Spain (*283*)	Metabolite	BDE-47
2′- MeO -BDE74	**Environment**: human milk Spain (*283*)	Metabolite	BDE-47
6′- MeO -BDE66	**Environment**: human milk Spain (*283*)	Metabolite	BDE-47
OH- and MeO-Br_5 Analogues			
6-OH-BDE-90	**Environment:** Red algae/mussels (*285*); Surface water (*284*), Rainfall (*284*), Freshwater fish (*272*); Marine fish (*269*)	Biogenic/ Metabolite	BDE-99
6-OH-BDE-99	**Environment:** Sponge (*294*); Red algae/mussels (*285*); Surface water (*284*), Rainfall (*284*), Freshwater fish (*272*); Marine fish (*269*)	Biogenic	BDE-99
2-OH-BDE-123	**Environment:** Sponge (*295*); Red algae/mussels (*285*); Surface water (*284*), Freshwater fish (*272*)	Biogenic	BDE-99
6-MeO-BDE-90	**Environment:** glaucous gulls (*262*); polar bears (*262*)	Biogenic	BDE-99

Compound	Field and Laboratory Observations	Likely Source(s)	Potential Precursor
6-MeO-BDE-99	**Environment**: glaucous gulls (*262*); polar bears (*262*)	Biogenic/ Metabolite	BDE-99
OH- and MeO-Br_6 Analogues			
6-OH-BDE-137	**Environment**: Marine sponge (*286*), Red algae/mussels (*285*)	Biogenic	None

7.6 Figures

Figure 7.1 Map showing general study area of E. Hudson Bay and various Nunavik Inuit communities of northern Quebec, Canada.

Note: Map acquired with permission from Makivik Corporation at <http://www.makivik.org/eng/media_centre/nunavik_maps.htm>

Figure 7.2 Concentrations (ng·g^{-1} lipid) of several OH-BDEs, MeO- BDEs and parent PBDE congeners in E. Hudson Bay beluga whales (blubber). Geometric means (GM) are plotted on a log scale with error bars representing 1 SD of the GM.

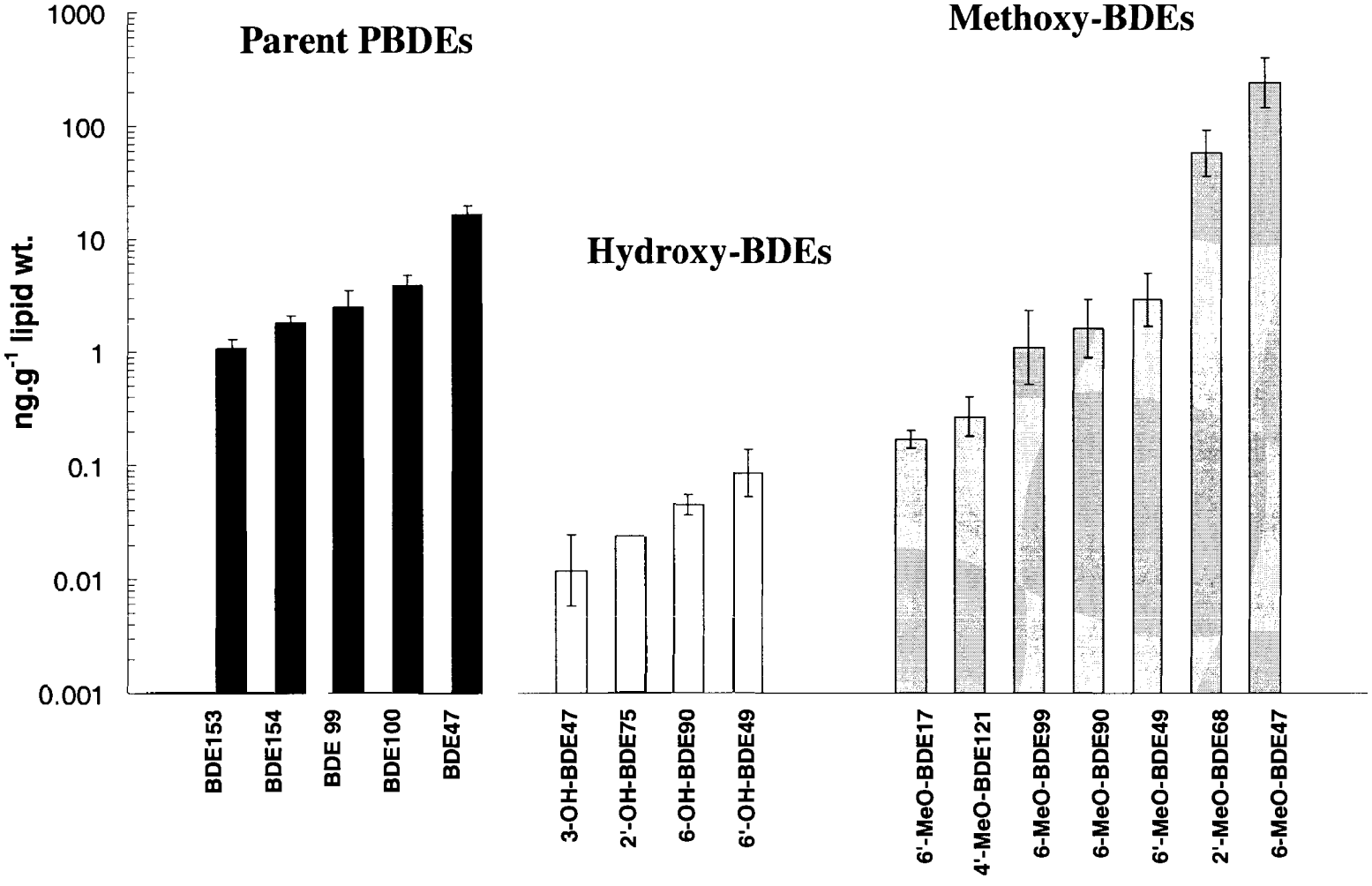


Figure 7.3 % composition plots for several PBDEs and 6 MeO-BDE-47 in E. Hudson Bay marine sediment and biota samples.

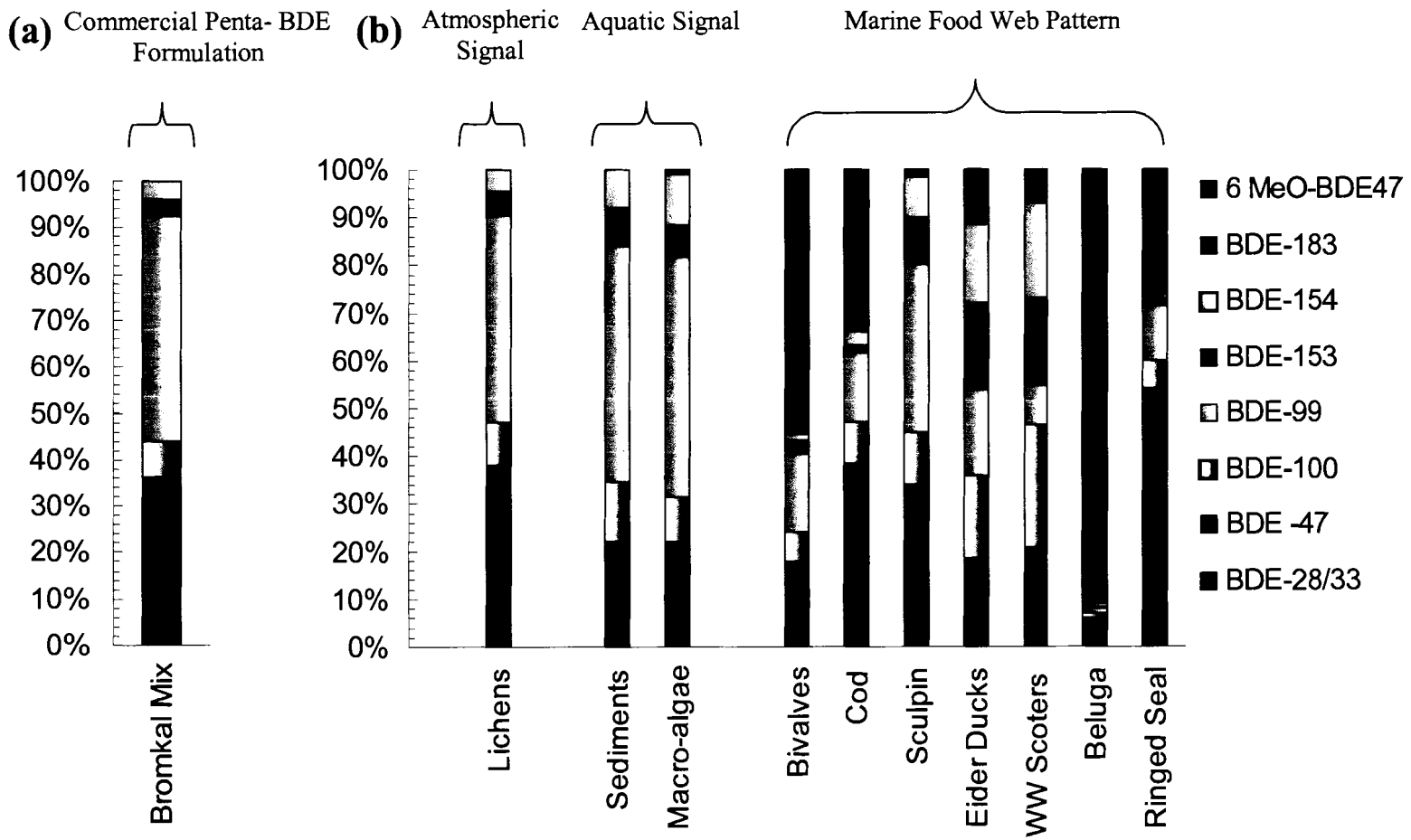


Figure 7.3 continued.

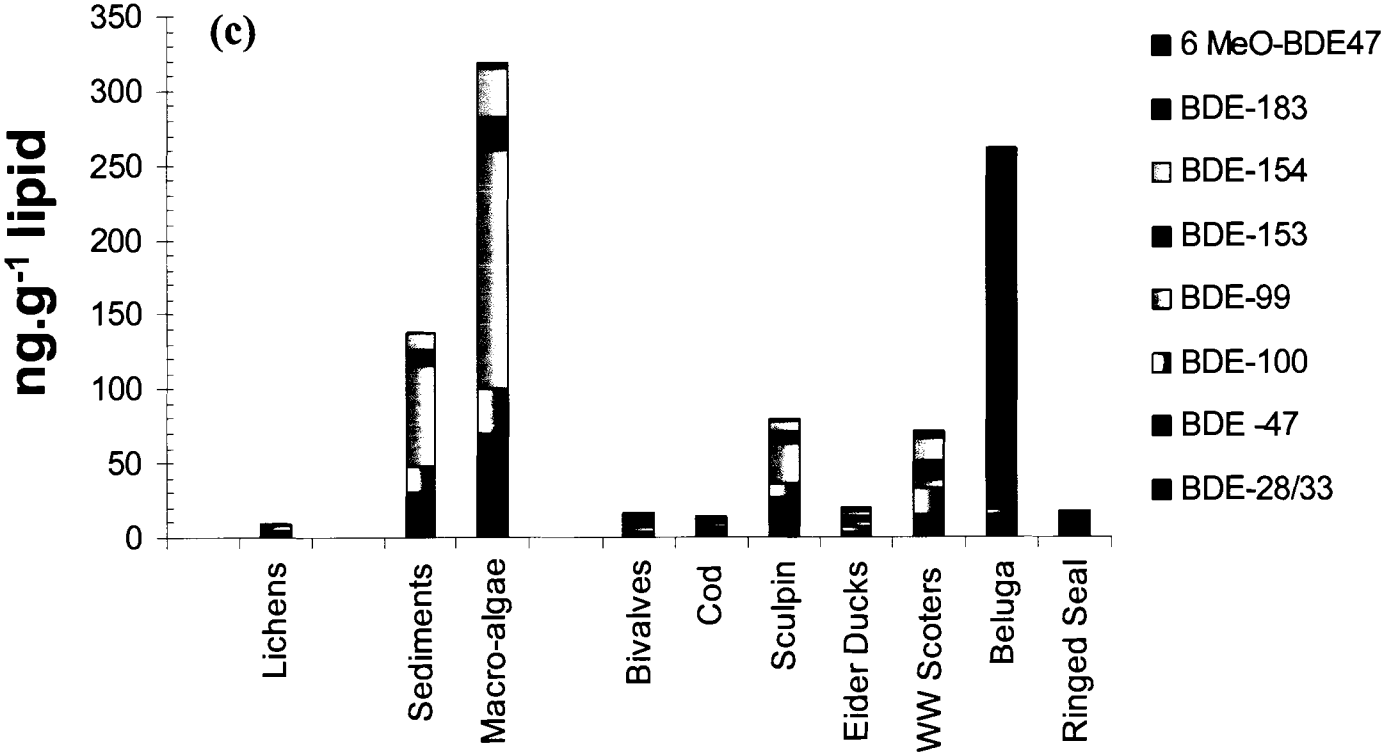


Figure 7.4 Relationship between (a) BDE47 and 6 MeO-BDE-47 and (b) p',p' DDT and p',p'DDE in E. Hudson Bay beluga whale blubber.

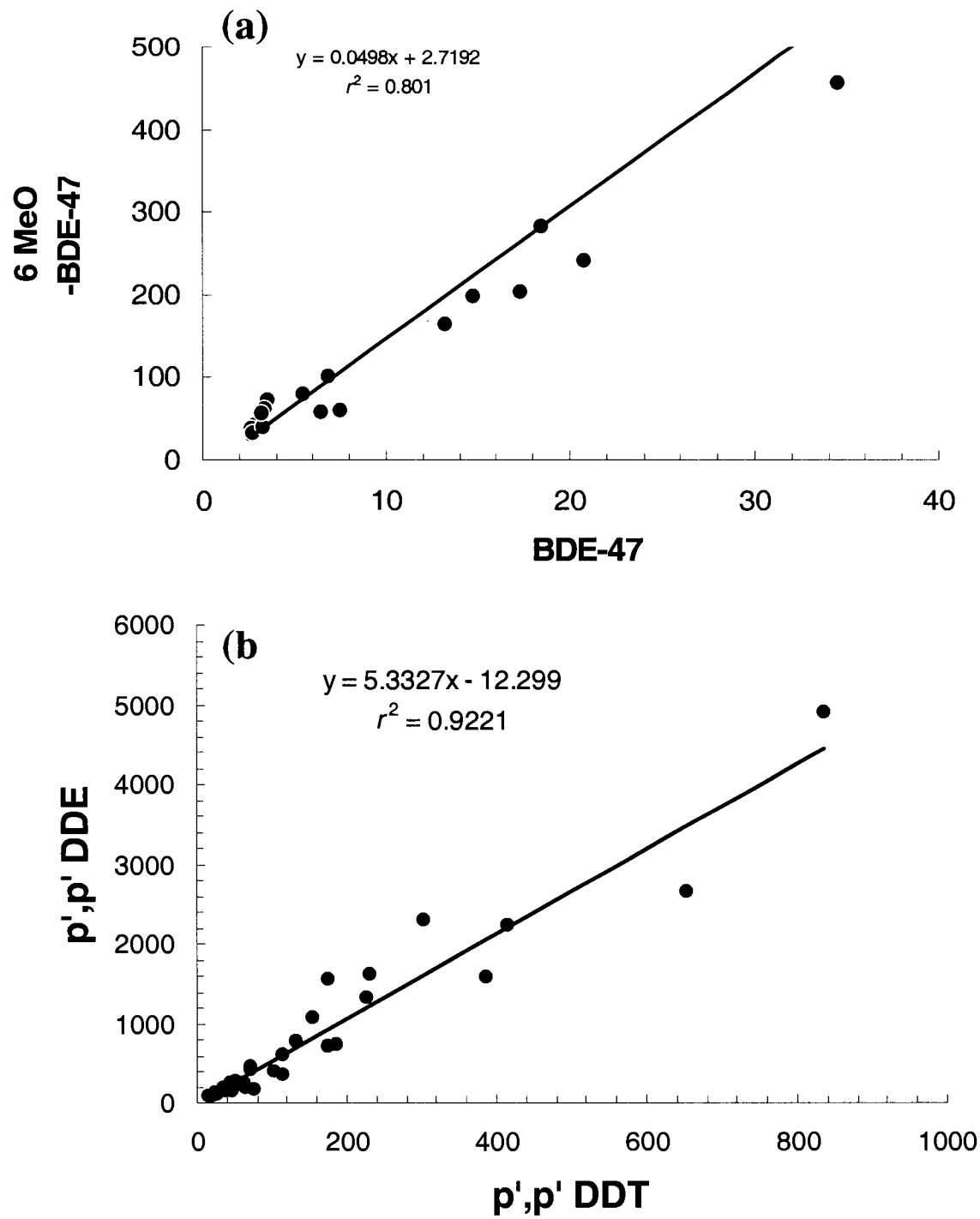


Figure 7.5 Plots of concentrations (ng·g^{-1} lipid) versus trophic level (TL) for (a) CB153 and (b) BDE100 (c) BDE99 (d) BDE47 and (e) 6 MeO-BDE-47 in biota from the E. Hudson Baymarine food web. Thick black line represents data for whole food web, thin black line represents air-breathing endotherms, and gray line represents water-ventilating ectotherms.

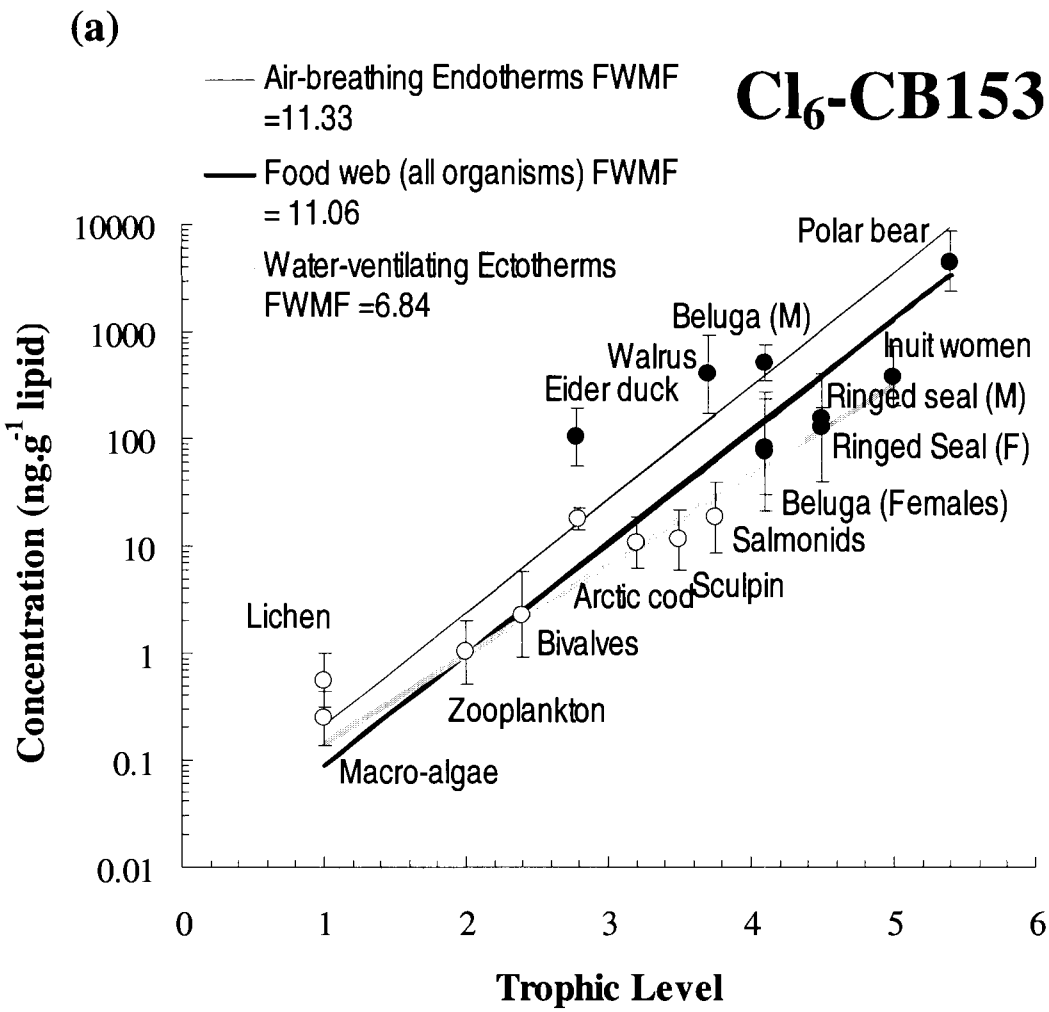


Figure 7.5 continued.

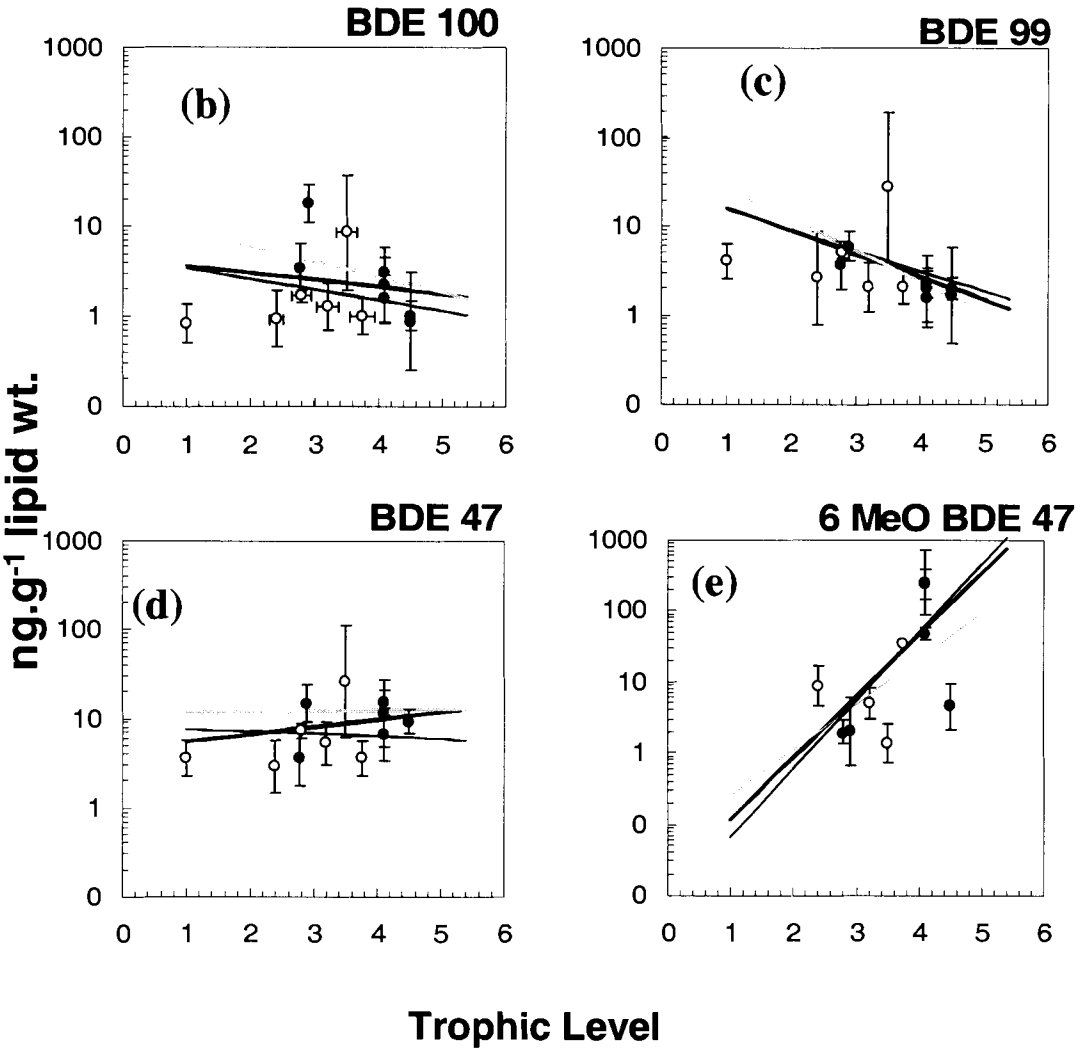


Figure 7.6 Concentrations ($ng \cdot g^{-1}$ lipid) and relative patterns for BDE47, BDE100 and and 6 MeO-BDE-47 in E. Hudson Bay eider ducks, ringed seals and beluga whales in comparison to other species from the Norwegian Arctic, Canadian Arctic (this study), Baltic, Mediterranean and North Atlantic waters. Data for Norwegian beluga whales, ringed seals, glaucous gulls and polar bears were from (*151,262*); data for Baltic grey and ringed seals are from *10*; data for Mediterranean Fin whale and bottlenose dolphin are from *296*; data for North Atlantic True's Beaked whale is from *281*.

Concentration (ng.g-1 lipid)
1000
900
800
700
600
500
400
300
200
100
0
6 MeO-BDE-47
BDE-100
BDE-47
N wegian Arctic
Canadian Arctic (this study)
Baltic
Mediterranean
rth Atlantic
Norwegian Polar Bears
Norwegian Glaucous gulls
Norwegian Ringed Seals
Norwegian Beluga whales
E. Hudson Bay Elder ducks
E. Hudson Bay Ringed Seals
E. Hudson Bay Beluga
Baltic Grey Seals
Baltic Ringed seals
Bottlenose Dolphin
Fin Whale
True's Beaked whale

CHAPTER 8

SUMMARY

8.1 Thesis Summary

The general hypothesis was that relatively polar substances (i.e., chemicals with low K_{OW}'s) that are relatively non-volatile (i.e., high K_{OA}'*s*) and resistant to biotransformation (i.e., metabolism) can substantially biomagnify in Arctic marine food webs. The main objectives of this study were to (i) measure concentrations of several organic contaminants in an Arctic marine food web, (ii) evaluate the biomagnification potential of those compounds and (iii) identify important biological factors and physical-chemical properties that influence food chain bioaccumulation of organic contaminants. To this end, the present thesis has provided novel information regarding current levels and various biological and physical-chemical determinants affecting the bioaccumulation behaviour of organic contaminants in Arctic marine food webs. Much of the chemical concentration data generated from this study are unique. For example, in the present study chemical concentrations of legacy POPs (i.e., PCBs and organochlorine pesticides) in abiotic and biotic Arctic samples were quantified using HRGC/HRMS, which are generally superior to previous POPs concentration data generated by GC-ECD detection. Also, this work has provided chemical concentration data for several new chemicals of emerging concern (e.g., phthalate esters, hydroxylated and methoxylated BDEs), which are currently rare or nonexistent. Findings of particular importance in this thesis include:

- Relatively polar/non-volatile chemicals such as β-HCH, CBz, β-endosulphan (log K_{OW} ~3-4, logK_{OA}> 7) biomagnify in air-breathing animals (marine mammals, seabirds, humans) and hence exhibit substantial food web magnification in the E. Hudson Bay marine food web.

- Other chemicals of emerging concern such as dialklyl phthalate esters (DPEs) and polybrominated diphenyl ethers (PBDEs) exhibit trophic dilution in this food web,

with FWMFs and BMFs < 1. Thus, organism /compound specific metabolic transformation can effectively reduce chemical bioaccumulation potential

- Although some commercial chemicals can be degraded/metabolized, accumulation of metabolites such as monoalkyl phthalate esters (MPEs), OH- and MeO-BDEs, *p',p'* DDE in organisms can occur. Thus, metabolism of parent compound can result in formation of bioaccumulative and potentially toxic compounds.

- These findings highlight the need to modify current bioaccumulation (B) criteria in current toxic substance management policy/legislation (e.g. CEPA, Stockholm Convention on POPs). Specifically, the current K_{OW} threshold criterion ($\log K_{OW} > 5$) does not adequately protect air-breathers animals such as reptiles, amphibians, birds, mammals (including humans). Chemical K_{OA} criteria should be developed to assess biomagnification in air- breathing animals. Also, the issue of metabolism/ degradation of parent compounds and formation of bioaccumulative, potentially toxic metabolites should be addressed. Further development of quantitiative-structure-activity relationships (QSARs), laboratory studies and mechanistic modelling may aid these initiatives.

8.2 Determinants of chemical bioaccumulation potential in marine food webs.

The accumulation and distribution of organic contaminants in food webs is complex and its understanding involves knowledge of several simultaneous processes (*82,177,297*). The prevailing processes in marine systems include (i) inter-media exchange (e.g., sediment-water distribution dynamics), (ii) organism respiration and dietary exposure/elimination kinetics and (iii) organism biotransformation capacity. Distribution of contaminants in media such as bottom sediments, particulate matter in the water column and primary producing algae is mainly controlled by chemical hydrophobicity (K_{OW}) and the sorptive capacity of the matrix (e.g., lipid and/or organic carbon content). For example, waterborne chemicals with K_{OW}'s between 10^6 and 10^8 such as Cl_6 to Cl_8 chlorobiphenyls tend to exhibit the maximum passive accumulation into environmental and biological media *via* passive equilibrium partitioning. However, accumulation of very hydrophobic compounds (K_{OW}'s > 10^8) is kinetically limited. Compounds such as Cl_9 to

Cl_{12} CBs, dioxins/furans are more associated with particulate matter and hence their distribution is more controlled by particle advection than passive sorption kinetics (*58,59,198,298,299,300*). In addition to chemical K_{OW}, the magnitude of lipid and/or organic matter of a given matrix or organism influences equilibrium chemical concentrations. For example, the bottom sediments in the present study of E. Hudson Bay exhibited relatively low dry weight contaminant burdens because those sediments were comprised mainly of sand (i.e., very low organic carbon, ~ 0.001 grams OC per gram dry weight of sediment) and hence have negligible sorptive capacity for hydrophobic organic contaminants.

For invertebrates, fish, seabirds and mammals the extent of overall bioaccumulation is primarily the effect of respiratory uptake and elimination kinetics (**bioconcentration**), gastro-intestinal absorption and exchange kinetics (i.e., **biomagnification**) and metabolic transformation capacity (i.e., **biotransformation**). These processes are also influenced by a combination of physical-chemical properties and various biological factors related to organism physiology and taxa. For example, equilibrium partitioning of chemical between an organism's respiratory medium and respiratory membranes (i.e., bioconcentration *via* gills or lungs) is inherently different for water-ventilating invertebrates and fish compared to air-breathing birds and mammals. Specifically, efficient respiratory elimination of organic chemicals in water-ventilators occurs when $K_{OW} < 10^5$ (*37,41,42*), while respiratory elimination by air-respiring organisms likely occurs only for very volatile compounds (i.e., when chemical $K_{OA} < 6$), (*44*). Chemical biomagnification is the process whereby chemical concentrations in an organism are elevated above the organism's diet due to dietary absorption. Inter-taxa differences in (i) food digestion/absorption efficiencies and (ii) chemical absorption efficiencies (E_D) are the primary reasons for wide ranging observations of biomagnification potential between organisms. For example, BMF_{MAX} values for fish species (~5-10) of birds and mammals (~50-100) correspond to more efficient food and chemical assimilation in avian and mammalian species (*172*).

8.2.1 Kinetics of chemical bioconcentration and biomagnification.

Uptake of contaminants in organisms is from intestinal exposure (consumption of food and/or water) and accumulation across respiratory membranes. Dietary uptake is generally the main route of exposure for the majority of legacy POPs and new chemicals of emerging concern. Chemical BMFs in a given organism are therefore determined primarily by the relative magnitude of competing rates of chemical uptake from the diet (k_{Diet}) *via* the gastro-intestinal tract (GIT) and

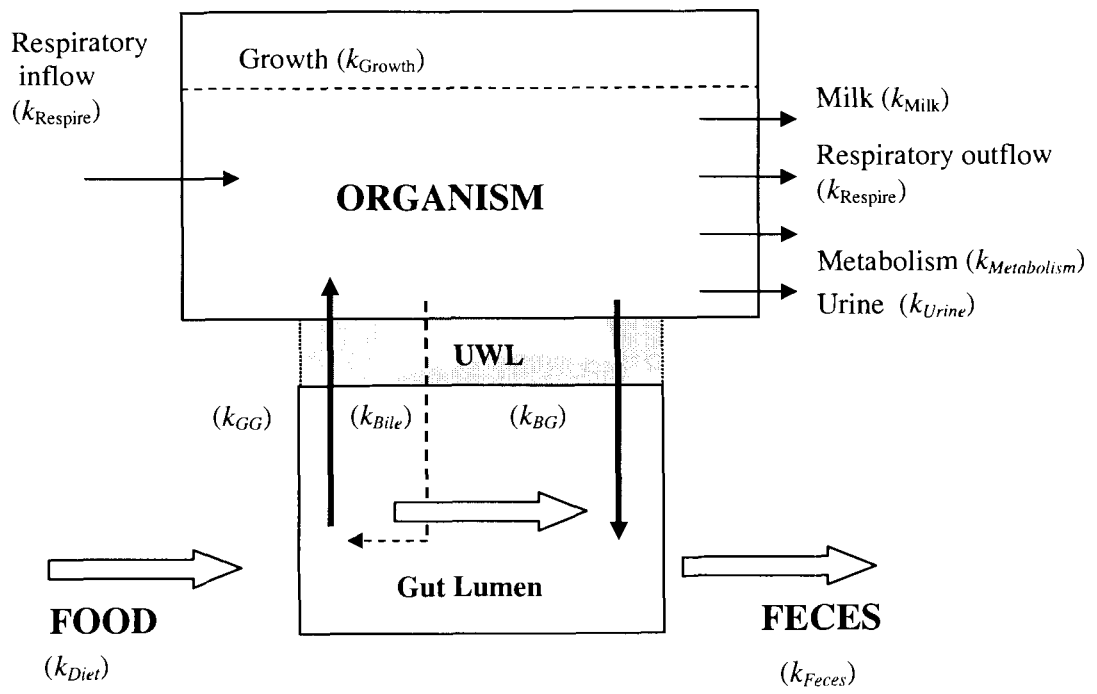
chemical losses by the various elimination routes, including respiration ($k_{Respire}$), urinary excretion (k_{Urine}), fecal egestion (k_{Feces}), bile excretion (k_{Bile}), growth dilution (k_{Growth}), milk excretion *via* lactation (k_{Milk}) and metabolism ($k_{Metabolism}$). Figure 8.1 is a conceptual diagram illustration of a simple two-compartment model (in rate constant format) of the bioaccumulation processes for a generic organism (i.e., either air-breathering endotherms $k_{Respire}$ is for organism-to-air losses or water-ventilating ectotherms where $k_{Respire}$ is for organism-to-water losses). Elimination of contaminants into respired air involves organism-to-air exchange, which is driven mainly by chemical's octanol-air partition coefficient K_{OA}. Elimination into respired water (aquatic organisms only), urine and bile involves exchange between the organism and water and is related to the octanol-water partition coefficient (K_{OW}). Fecal egestion and milk excretion (in female animals) involves chemical exchange between lipids and other non-lipid organic matter (e.g., proteins, carbohydrates etc.) and can be represented by lipid-to-organic matter partition coefficients. The steady-state biomagnification factor (BMF) is represented as:

$$BMF = C_{Biota}/C_{Diet} =$$

$$k_{Diet}/(k_{Respire} + k_{Urine} + k_{Bile} + k_{Feces} + k_{Milk} + k_{Growth} + k_{Metabolism}) \quad (1)$$

where C_{Biota} and C_{Diet} are the steady-state chemical concentrations ($mol.m^{-3}$).

Figure 8.1 Conceptual illustration of a two-compartment bioaccumulation model for a generic organism (air-breathing endotherms such as birds and mammals or aquatic water ventilating ectotherms such as aquatic invetebrates and fish).



The dietary uptake rate constant k_{Diet} can be expressed as:

$$k_{Diet} = E_{Diet}.G_D/V_B \quad (2)$$

where E_{Diet} is the dietary uptake efficiency, G_D is the feeding rate ($m^3 \cdot d^{-1}$) and V_B is the weight of the organism (m^3). The respiratory elimination rate constant, $k_{Respire}$, can be determined as k_{Water} (for aquatic organism) or k_{Air} (for air-breathing animals), expressed as:

$$k_{Respire} = k_{Water} = E_W.G_W/V_B.L_B.K_{OW} \quad (3)$$

and

$$k_{Respire} = k_{Air} = E_{Air}.G_A/V_B.L_B.K_{OA} \quad (4)$$

where G_W is the water ventilation rate over the gills ($m^3 \cdot d^{-1}$), G_A is the air respiration rate ($m^3 \cdot d^{-1}$), V_B is the weight of the organism, L_B is the lipid content of the organism. The urinary excretion rate constant is

$$k_{Urine} = G_U/V_B.L_B.K_{OW} \quad (5)$$

where G_U is the urinary excretion rate ($m^3 \cdot d^{-1}$). The fecal excretion rate constant is

$$k_{Fecal} = G_F/V_B.K_{BF} \quad (6)$$

where G_F is the fecal excretion rate ($m^3 \cdot d^{-1}$) and K_{BF} is the organism-to-feces partition coefficient (kg wet weight organism/kg wet weight feces). The bile excretion rate is

$$k_{Bile} = G_B/V_B.L_B.K_{OB} \quad (7)$$

where G_B is the bile excretion rate ($m^3 \cdot d^{-1}$) and K_{OB} is the octanol-bile partition coefficient which can be expressed as a function of K_{OW}, i.e. $K_{OB} = K_{OW}/\delta$ where δ represents the degree to which bile fluids exceed the solubility of contaminants over that in water. The milk excretion (i.e., lactation) rate constant can be expressed as

$$k_{Milk} = G_M/V_B.L_B.K_{OM} \quad (8)$$

where G_M is the milk excretion rate ($m^3 \cdot d^{-1}$) in female animals and K_{OM} is the octanol-milk partition coefficient. In male animals k_{Milk} is zero.

In the BMF calculation in Equation 1, k_{Growth} (1/d) is the growth dilution rate constant. It does not represent a true elimination pathway. However, an increase in body mass has the effect of "diluting" or reduction in the internal concentration and can be represented as an elimination route.

8.2.2 Non-metabolizable compounds.

For the respiratory elimination route, it is important to distinguish between water-ventilating and air-breathing organisms, as the respired media and hence elimination kinetics are markedly

different. For aquatic water-ventilating organism, elimination to water (i.e., gill ventilation) is well known to be inversely related to the chemical's K_{OW}. Hence, an increase in K_{OW} (i.e., K_{OW}'s greater than 10^5) causes a slower rate of chemical elimination from the organism via respiration. Thus, for non-metabolizable chemicals with K_{OW}'s > 10^5, respiratory elimination is small compared to dietary uptake and biomagnification occurs. For air-breathing homeotherms, respiratory elimination is not to water but to alveolar air *via* lipid-air exchange dynamics in the lungs. Respiratory elimination *via* lipid-air exchange declines with increasing octanol-air partition coefficient (K_{OA}), causing chemicals to approach a maximum biomagnification potential with increasing K_{OA}. It has been suggested that if K_{OA} exceeds 10^5, respiratory elimination is too small to effectively reduce the biomagnification effect in the GIT of many mammals hence biomagnification can occur (*44*). Only if the substance is rapidly eliminated to urine (e.g., logK_{OW} is less than approximately 2) or rapidly metabolized, can biomagnification be prevented. Thus, the bioaccumulation potential of organic chemicals in aquatic organisms is best assessed by K_{OW}, while bioaccumulation potential in air-breathing organisms is best anticipated by K_{OA} and K_{OW} (*44,100*).

8.2.3 Biotransformation.

The metabolic transformation rate constant $k_{Metabolism}$ represents the metabolic transformation rate of the parent compound. Increased $k_{Metabolism}$ (determined from the half life of the parent compound in the organism's intestinal tract or liver) ultimately reduces the biomagnification potential of chemicals. Simultaneously, there are chemical gains *via* formation of the metabolic by-products. Cahill et al. (*113*) recently presented a generalized physiologically based pharmacokinetic (PBPK) model that includes metabolite formation and distribution within organisms. The model represents metabolism as the transformation of parent compound (α), primary metabolites (β), and secondary metabolites (γ). The secondary metabolite (γ) can be either a newly formed chemical species (e.g., via further oxidized species) or a conjugated form of the primary metabolite (e.g., glucuronidated conjugate). Chemical reactions (which can occur in the intestine, liver or blood) include $\alpha \rightarrow \gamma$, $\beta \rightarrow \gamma$ and also γ back into β. In this thesis we observed several primary metabolites of the target compounds analyzed in Arctic biota. Those biotransformations include endosulfan→ endosulfan sulfate, *p',p'* DDT → *p',p'*DDE, technical chlordane → oxychlordane, heptachlor→ heptachlor epoxide, dialkyl phthalate esters→ monoalkyl phthalate esters and PBDEs → OH-BDEs → MeO-BDEs. Metabolism is generally viewed as a sequence of multi-phase reactions, including Phase I biotransformation (hydrolysis,

oxidation reactions) in the upper gastro-intestinal tract, liver or blood, Phase II biotransformation (reaction of primary metabolite with glucuroic acid) resulting in a β-glucuronide conjugate. The glucuronidation of primary metabolite generally increases the water solubility and hence enhance urinary excretion. The non-conjugated (i.e., free form) of the primary metabolite (β) can be absorbed, excreted in urine or further metabolized (e.g., ω, ω -1, ω -2 oxidation).

8.3 Conclusion

In addition to bioaccumulation processes (i.e., bioconcentration, biomagnification and biotransformation), other geospatial and biological factors such as prey selection, migration and habitat usage can also influence chemical bioaccumulation patterns. Migratory seabirds and mammals (e.g., whales) can traverse many degrees of latitude and longitude and hence can experience quite variable dietary exposures. This ecologically driven factor may be the reason for seemingly high levels and unique bioaccumulation patterns of organochlorines observed in E. Hudson Bay white-winged scoters (an Arctic migrant species) compared to common eider ducks (an Arctic resident species), (see *Chapter 3*). The determinants of chemical concentrations and patterns in wild fish, birds and mammals in marine food involve a complexity of temporal-spatial, biological and physical-chemical factors such as organism migration chronology, predatory-prey relationships, chemical K_{OW} and K_{OA}, organism physiology and metabolic capacity. Evaluation of biomagnification factors (BMFs) and elimination indexes (EIs), relative to well-established recalcitrant POPs (e.g., PCB153 and 180 = BMF_{MAX}) is a useful approach for assessing the chemical fate and bioaccumulation behaviour of novel environmental contaminants. The findings in this thesis indicate K_{OA} is an important determinant of chemical biomagnification in air-breathing animals. An organism's metabolic capacity is also a crucial factor affecting chemical bioaccumulation potential and the resulting contaminant profiles observed in different species of the food web. Specifically, primary metabolites, monoalkyl phthalate esters (MPEs) and hydroxylated and methoxylated brominated diphenyl ethers (OH-BDEs / MeO-BDEs) were detected in organisms of the food web. Future regulatory initiatives should include chemical K_{OA} and the formation of potentially toxic metabolites as criteria for assessing POPs bioaccumulation potential.

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APPENDICES

Appendix 1 Description of Marine mammal and Seaduck tissue samples collected in E. Hudson Bay Nunavik region between 1999-2001

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
Female Beluga										
Beluga	F	DL99-01	Nastapoka R	-	F	-	-	-	8	8
Beluga	F	DL99-07	Nastapoka R	-	F	-	-	-	6-10	9
Beluga	F,Mu,L,S, I, B	DL99-25	Nastapoka R	3-Aug-00	F	gray	-	-	15	15
Beluga	F,Mu,L,S, I, B,M	DL2000-01	Nastapoka R.	2-Aug-00	F	adult	330	-	-	21
Beluga	F,Mu,L,S, I, B	DL2000-03	Nastapoka R	3-Aug-00	F	white	-	-	15	15
Beluga	F,Mu,L,S, I, B,M	DL2000-05	Nastapoka R	3-Aug-00	F	white	-	-	34	34
Beluga	F,Mu,L,S, I, B	DL2000-16	Nastapoka R	5-Aug-00	F	white	350	-	-	26
Beluga	F,Mu,L,S, I, B	DL2000-17	Nastapoka R	5-Aug-00	F	white	324	-	-	23
Beluga	F,Mu,L,S, I, B	DL2000-18	Nastapoka R	5-Aug-00	F	gray	-	-	-	7
Beluga	F,Mu,L,S, I, B	DL2000-20	Nastapoka R	5-Aug-00	F	white	360	-	-	19
Beluga	F,Mu,L,S, I, B	DL2000-21	Nastapoka R	5-Aug-00	F	light gray	344	-	-	10
Beluga	F,Mu,L,S, I, B,M	DL2000-22	Nastapoka R	6-Aug-00	F	white	330	-	-	20
Beluga	F,Mu,L,S, I, B,M	DL2000-25	Nastapoka R	6-Aug-00	F	white	330	-	-	20
Beluga	F	DL2000-31	Umiujaq		F	light gray			5	5
Beluga	F,Mu,L,S, I, B	DLN01-03	Nastapoka R	1-Aug-01	F	adult	344	-	-	26
Beluga	F	DL2000-33			F	adult	340		-	25
Beluga	F,Mu,L,S, I, B	DLN01-10	Nastapoka R	4-Aug-01	F	white	350	204	-	31
FEMALES 2-15 years *n* = 6										
FEMALES 15-35 years *n* = 12										
TOTAL FEMALE BELUGA *n* = 18										
Male Beluga										
Beluga	F	DL99-02	Nastapoka R	-	M	-	-	-	<5	3
Beluga	F	DL99-03	Nastapoka R	-	M	-	-	-	27	27
Beluga	F	DL99-04	Nastapoka R	-	M	-	-	-	28	28
Beluga	F	DL99-05	Nastapoka R	-	M	-	-	-	20	20
Beluga	F	DL99-06	Nastapoka R	-	M	-	-	-	6	6

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
Beluga	F	DL99-08	Nastapoka R	-	M	-	-	-	18	18
Beluga	F	DL99-09	Nastapoka R	-	M	-	-	-	26	26
Beluga	F,Mu,L,S, I, B	DL2000-04	Nastapoka R	3-Aug-00	M	white	-	-	21	21
Beluga	F,Mu,L,S, I, B	DL2000-09	Nastapoka R	3-Aug-00	M	white	-	-	14	14
Beluga	F,Mu,L,S, I, B	DL99-17	Nastapoka R	3-Aug-00	M	gray	-	-	16	16
Beluga	F,Mu,L,S, I, B	DL99-26	Nastapoka R	3-Aug-00	M	gray	-	-	-	
Beluga	F,Mu,L,S, I, B	DL2000-23	Nastapoka R	6-Aug-00	M	white	330	-	-	20
Beluga	F,Mu,L,S, I, B	DL2000-24	Nastapoka R	6-Aug-00	M	white	370	-	-	34
Beluga	F	DL2000-32	Nastapoka R		M	adult				35
Beluga	F	DL2000-34	Nastapoka R		M	gray				12
Beluga	F	DL2000-35	Nastapoka R		M	gray				12
Beluga	F,Mu,L,S, I, B	DLN01-01	Nastapoka R	1-Aug-01	M	adult	350	-	-	>15
Beluga	F,Mu,L,S, I, B	DLN01-02	Nastapoka R	1-Aug-01	M	adult	370	-	-	>15
Beluga	F,Mu,L,S, I, B	DLN01-04	Nastapoka R	3-Aug-01	M	light gray	-	-	-	10
Beluga	F,Mu,L,S, I, B	DLN01-05	Nastapoka R	3-Aug-01	M	white	-	-	-	>15
Beluga	F,Mu,L,S, I, B	DLN01-06	Nastapoka R	3-Aug-01	M	light gray	-	-	-	8
Beluga	F,Mu,L,S, I, B	DLN01-07	Nastapoka R	3-Aug-01	M	white	-	-	-	>15
Beluga	F,Mu,L,S, I, B	DLN01-08	Nastapoka R	3-Aug-01	M	white	-	-	-	>15
Beluga	F,Mu,L,S, I, B	DLN01-09	Nastapoka R	3-Aug-01	M	l. gray	365	-	-	11
Beluga	F,Mu,L,S, I, B	DLN01-12	Nastapoka R	7-Aug-01	M	adult	390	240	-	30
Beluga	F,Mu,L,S, I, B	DLN01-13	Nastapoka R	7-Aug-01	M	white	400	218	-	34
Beluga)	F,Mu,L,S, I, B	DLN01-14	Nastapoka R	7-Aug-01	M	white	383	276	-	30
Beluga	F	DL2000-30	Nastapoka R	6-Aug-00	M	white	388	-	-	35
MALES 2-15 years *n* ▫ 9										
MALES 15-35 years *n* = 19										
TOTAL MALE BELUGA *n* = 28										

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
Beluga Calves										
Beluga	F,Mu,L,S, I, B	DL2000-02	Nastapoka R	2-Aug-00	M	calf	170	-	-	<1
Beluga	F,Mu,L,S, I, B	DL2000-10	Nastapoka R	3-Aug-00	F	calf	-	-	1	<1
Beluga	F	DL2000-26	Nastapoka R	6-Aug-00	M	calf	-	-	-	<1
Beluga	F	DL2000-27	Nastapoka R	6-Aug-00	F	calf	-	-	-	<1
Beluga	F	DL2000-28	Nastapoka R	6-Aug-00	M	calf	-	-	-	<1
Beluga	F	DL2000-29	Nastapoka R	6-Aug-00	X	calf	-	-	-	<1
Beluga	F,Mu,L,S, I, B	DLN01-11	Nastapoka R	4-Aug-01	F	calf	198	130	-	<1
TOTAL CALVES $n = 7$										
Beluga (Unknown Sex)										
Beluga	F,Mu,L,S, I, B	DL2000-06	Nastapoka R	3-Aug-00	TBD	white	-	-	-	>15
Beluga	F,Mu,L,S, I, B	DL2000-08	Nastapoka R	3-Aug-00	TBD	light gray	-	-	-	<10
Beluga	F,Mu,L,S, I, B	DL2000-11	Nastapoka R	3-Aug-00	TBD	adult	-	-	-	>15
TOTAL Unknown Sex $n = 3$										
Beluga Fetus										
Beluga	WB	DL2000-05 (fetus)	Nastapoka R	3-Aug-00	X	-	-	-	-	-
Beluga	WB	DL2000-16 (fetus)	Nastapoka R	6-Aug-00	X	-	-	-	-	-
Beluga	WB	DL2000-20 (fetus)	Nastapoka R	6-Aug-00	X	-	-	-	-	-
Beluga	WB	DLN01-10 (fetus)	Nastapoka R	7-Aug-00	X	-	-	-	-	-
TOTAL FETUS $n = 4$										

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
Beluga Milk										
Beluga	milk	DL00122 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	-	20
Beluga	milk	DL2000-05 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	34	34
Beluga	milk	DLN01-10 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	-	31
Beluga	milk	DL2000-01 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	-	21
Beluga	milk	DL99-25 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	15	15
Beluga	milk	DL00125 (milk)	Nastapoka R	6-Aug-00	F	-	-	-	-	15
Beluga	milk	DL00125 (milk)	Nastapoka R	7-Aug-00	F	-	-	-	-	34
Beluga	milk	DL00125 (milk)	Nastapoka R	8-Aug-00	F	-	-	-	-	20
TOTAL BELUGA MILK *n* = 8										
Seaducks (provided by M.Kwan NvRC)										
Common Eider Duck	L,F,K	CE-01	EHB	2001		-	-	-	-	-
Common Eider Duck	L,F,K	CE-02	EHB	2001		-	-	-	-	-
Common Eider Duck	L,F,K	CE-03	EHB	2001		-	-	-	-	-
Common Eider Duck	L,F,K	CE-04	EHB	2001		-	-	-	-	-
Common Eider Duck	L,F,K	CE-05	EHB	2001		-	-	-	-	-
Common Eider Duck	L,F,K	CE-06	EHB	2001		-	-	-	-	-
TOTAL Eider Ducks *n* = 6							-	-	-	
White winged Scoter	L,F,K	WWS-01	EHB	2001		-	-	-	-	-
White winged Scoter	L,F,K	WWS-02	EHB	2001		-	-	-	-	-
White winged Scoter	L,F,K	WWS-03	EHB	2001		-	-	-	-	-

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
White winged Scoter	L,F,K	WWS-04	EHB	2001		-	-	-	-	-
White winged Scoter	L,F,K	WWS-05	EHB	2001		-	-	-	-	-
TOTAL WW Scoters *n* = 5										
Ringed Seals (provided by D.C.G. Muir, NWRI)										
Ringed seal	F	M21	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	M22	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	M23	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	M28	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	M34	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	M35	Makkovik	-	M	-	-	-	-	-
Ringed seal	F	N7	Nain	-	M	-	-	-	-	-
Ringed seal	F	Q46	Quaqtaq	-	M	-	-	-	-	-
Ringed seal	F	Q59	Quaqtaq	-	M	-	-	-	-	-
Ringed seal	F	S5	Salluit	-	M	-	-	-	-	-
TOTAL MALE RINGED SEALS *n* = 10										
Ringed seal	F	N1	Nain	-	F	-	-	-	-	-
Ringed seal	F	N2	Nain	-	F	-	-	-	-	-
Ringed seal	F	N11	Nain	-	F	-	-	-	-	-
Ringed seal	F	Q48	Quaqtaq	-	F	-	-	-	-	-
Ringed seal	F	Q57	Quaqtaq	-	F	-	-	-	-	-
Ringed seal	F	Q58	Quaqtaq	-	F	-	-	-	-	-

Species	Tissue/ Viscera[a]	ID#	Location	Date Collected	Sex	Age class/ colour	Length (cm)	Girth (cm)	Age (years) Measured (tooth) [b]	Age (years) Estimated Allometric relationship [c]
Ringed seal	F	S3	Salluit	-	F	-	-	-	-	-
Ringed seal	F	S7	Salluit	-	F	-	-	-	-	-
Ringed seal	F	S10	Salluit	-	F	-	-	-	-	-
Ringed seal	F	M26	Makkovik	-	F	-	-	-	-	-

TOTAL FEMALE RINGED SEALS *n* = 10

[a] Tissue type legend: F= fat (adipose tissue/blubber), L=liver, Mu = Muscle, S = stomach contents, I= intestinal tissue, M = milk, B = whole blood, K= kidney. [b] Age determination by tooth ring analysis by NvRC (see Doidge et al., 1990). [c] Age determination by allometric relationship of age versus length from Doidge et al. (1990)

Appendix 2 Physical chemical properties of several organic chemicals.

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Chlorinated Biphenyls (CBs)											
Cl_2 (CB 7/9) 2,4 / 2,5	$C_{12}H_8Cl_2$	33284-50-3/34883-39-1	Group 3	MC-type CYP1A	5.15	7.09	35.20	223.1	226.4	1.40E+06	0.175
Cl_2 (CB 6) 2,3'	$C_{12}H_8Cl_2$	25569-80-6	Group 3	MC-type CYP1A	5.00	7.19	23.30	223.1	226.4	1.60E+06	0.147
Cl_2 (CB 8/5) 2,4' / 2,3	$C_{12}H_8Cl_2$	34883-43-7/16605-91-7	Group 3	MC-type CYP1A	5.24	7.38	31.31	223.10	226.40	6.20E+05	0.104
Cl_2 (CB 4/10) 2,2' / 2,6	$C_{12}H_8Cl_2$	13029-08-8/33146-45-1	Group 4	PB-type CYP2B	4.98	6.93	47.61	223.10	226.40	1.60E+06	0.366
Cl_3 (CB 23/34) 2,3,5 / 2',3,5	$C_{12}H_7Cl_3$	55720-44-0/37680-68-5	Group 3	MC-type CYP1A	5.50	8.13	21.30	257.54	247.30	1.69E+05	0.0269
Cl_3 (CB 29) 2,4,5	$C_{12}H_7Cl_3$	15862-07-4	Group 3	MC-type CYP1A	5.48	7.91	20.27	257.54	247.30	1.49E+05	0.0400
Cl_3 (CB 26) 2,3'5	$C_{12}H_7Cl_3$	38444-81-4	Group 3	MC-type CYP1A	5.65	8.03	32.93	257.54	247.30	1.87E+05	0.0320
Cl_3 (CB 25) 2,3',4	$C_{12}H_7Cl_3$	55712-37-3	Group 3	MC-type CYP1A	5.65	8.03	32.93	257.54	247.30	1.87E+05	0.0320
Cl_3 (CB 31) 2,4',5	$C_{12}H_7Cl_3$	16606-02-3	Group 3	MC-type CYP1A	5.67	8.45	20.27	257.54	247.30	1.78E+05	0.0150
Cl_3 (CB 28) 2,4,4'	$C_{12}H_7Cl_3$	7012-37-5	Group 3	MC-type CYP1A	5.00	8.14	32.02	257.54	247.30	1.52E+05	0.0260
Cl_3 (CB 21) 2,3,4	$C_{12}H_7Cl_3$	55702-46-0	Group 3	MC-type CYP1A	5.49	8.13	21.30	257.54	247.30	1.69E+05	0.0269
Cl_3 (CB 33/20) 2',3,4 / 2,3,3'	$C_{12}H_7Cl_3$	38444-86-9/38444-84-7	Group 3	MC-type CYP1A	5.71	8.32	22.70	257.54	247.30	1.33E+05	0.0190
Cl_3 (CB 19) 2,2',6	$C_{12}H_7Cl_3$	38444-73-4	Group 4	PB-type CYP2B	5.20	7.44	25.33	257.54	247.30	1.00E+05	0.117

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_3 (CB30) 2,4,6	$C_{12}H_7Cl_3$	35693-92-6	Group 4	PB-type CYP2B	5.60	8.04	22.70	257.54	247.30	2.00E+05	0.0300
Cl_3 (CB 18) 2,2',5	$C_{12}H_7Cl_3$	37680-65-2	Group 4	PB-type CYP2B	5.24	7.42	25.33	257.54	247.30	1.10E+05	0.1200
Cl_3 (CB 17) 2,2',4	$C_{12}H_7Cl_3$	37680-66-3	Group 4	PB-type CYP2B	5.20	7.63	25.33	257.54	247.30	1.10E+05	0.0760
Cl_3 (CB 27/24) 2,3',6 / 2,3,6	$C_{12}H_7Cl_3$	38444-76-7/55702-45-9	Group 4	PB-type CYP2B	5.60	8.10	20.27	257.54	247.30	1.60E+05	0.0260
Cl_3 (CB 16/32) 2,2',3 / 2,4',6	$C_{12}H_7Cl_3$	38444-78-9/38444-77-8	Group 4	PB-type CYP2B	5.71	8.12	22.70	257.54	247.30	1.33E+05	0.0250
Cl_3 (CB 22) 2,3,4'	$C_{12}H_7Cl_3$	38444-85-8	Group 3	MC-type CYP1A	5.86	8.17	21.38	257.54	247.30	1.60E+05	0.0250
Cl_4 (CB 54) 2,2',6,6'	$C_{12}H_6Cl_4$	15968-05-5	Group 5	-	5.93	7.90	55.73	292.00	268.20	1.80E+05	0.0410
Cl_4 (CB 50) 2,2',4,6	$C_{12}H_6Cl_4$	62796-65-0	Group 5	-	5.87	7.86	58.57	292.00	268.20	3.40E+04	0.0450
Cl_4 (CB 53) 2,2',5,6'	$C_{12}H_6Cl_4$	41464-41-9	Group 4	PB-type CYP2B	5.87	7.86	41.14	292.00	268.20	4.60E+04	0.0450
Cl_4 (CB 51) 2,2',4,6'	$C_{12}H_6Cl_4$	68194-04-7	Group 5	-	5.63	8.02	49.05	292.00	268.20	6.50E+04	0.0310
Cl_4 (CB 45) 2,2',3,6	$C_{12}H_6Cl_4$	70362-45-7	Group 5	-	5.75	8.40	32.83	292.00	268.20	4.50E+04	0.0130
Cl_4 (CB 46) 2,2',3,6'	$C_{12}H_6Cl_4$	41464-47-5	Group 5	-	5.70	8.40	32.83	292.00	268.20	4.50E+04	0.0130
Cl_4 (CB 73/52) 2,3',5',6 / 2,2',5,5'	$C_{12}H_6Cl_4$	74338-23-1/35693-99-3	Group 4	PB-type CYP2B	5.90	8.39	34.65	292.00	268.20	4.00E+04	0.0140
Cl_4 (CB 69) 2,3',4,6	$C_{12}H_6Cl_4$	60233-24-1	Group 4	PB-type CYP2B	6.10	9.22	19.10	292.00	268.20	5.90E+04	0.00206
Cl_4 (CB 49) 2,2',4,5'	$C_{12}H_6Cl_4$	41464-40-8	Group 4	PB-type CYP2B	5.95	8.29	27.96	292.00	268.20	4.00E+04	0.0170
Cl_4 (CB 43) 2,2',3,5	$C_{12}H_6Cl_4$	70362-46-8	Group 4	PB-type CYP2B	5.75	8.42	25.40	292.00	268.20	4.30E+04	0.0125
Cl_4 (CB 47/75/48) 2,2',4,4' / 2,4,4',6 / 2,2',4,5	$C_{12}H_6Cl_4$	2437-79-8//70362-47-9	Group 4	PB-type CYP2B	5.94	8.17	27.70	292.00	268.20	4.30E+04	0.0220

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_4 (CB 65) 2,3,5,6	$C_{12}H_6Cl_4$	33284-54-7	Group 4	PB-type CYP2B	5.98	9.21	25.84	292.00	268.20	6.80E+04	0.0021
Cl_4 (CB 62) 2,3,4,6	$C_{12}H_6Cl_4$	54230-22-7	Group 4	PB-type CYP2B	6.10	8.78	17.56	292.00	268.20	1.50E+04	0.0056
Cl_4 (CB 44) 2,2',3,5'	$C_{12}H_6Cl_4$	41464-39-5	Group 4	PB-type CYP2B	5.85	9.23	35.78	292.00	268.20	4.40E+04	0.0020
Cl_4 (CB 59/42) 2,3,3',6 / 2,2',3,4'	$C_{12}H_6Cl_4$	74472-33-6/36559-22-5	Group 4	PB-type CYP2B	5.75	8.34	23.30	292.00	268.20	3.60E+04	0.0152
Cl_4 (CB 72) 2,3,5,5'	$C_{12}H_6Cl_4$	41464-42-0	Group 3	MC-type CYP1A	6.03	8.35	42.25	292.00	268.20	1.60E+04	0.0180
Cl_4 (CB 71/41/64) 2,3',4',6 / 2,2',3,4 / 2,3,4',6	$C_{12}H_6Cl_4$	41464-46-4/ 52663-59-9 /52663-58-8	Group 4	PB-type CYP2B	5.76	8.26	25.90	292.00	268.20	1.60E+04	0.0180
Cl_4 (CB 68) 2,3',4,5'	$C_{12}H_6Cl_4$	73575-52-7	Group 3	MC-type CYP1A	6.03	8.35	42.25	292.00	268.20	1.60E+04	0.0180
Cl_4 (CB 40) 2,2',3,3'	$C_{12}H_6Cl_4$	38444-93-8	Group 4	PB-type CYP2B	5.95	9.23	20.47	292.00	268.20	4.40E+04	0.0020
Cl_4 (CB 57) 2,3,3',5	$C_{12}H_6Cl_4$	70424-67-8	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.0021
Cl_4 (CB 67) 2,3',4,5	$C_{12}H_6Cl_4$	73575-53-8	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.0021
Cl_4 (CB 58) 2,3,3',5'	$C_{12}H_6Cl_4$	41464-49-7	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.0021
Cl_4 (CB 63) 2,3,4',5	$C_{12}H_6Cl_4$	74472-34-7	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.0021
Cl_4 (CB 61/74) 2,3,4,6 / 2,4,4',5	$C_{12}H_6Cl_4$	33284-53-6/32690-93-0	Group 3	MC-type CYP1A	6.03	8.35	42.25	292.00	268.20	1.60E+04	0.018
Cl_4 (CB 70/76) 2,3',4',5 / 2',3,4,5	$C_{12}H_6Cl_4$	32598-11-1/70362-48-0	Group 3	MC-type CYP1A	6.03	8.35	42.25	292.00	268.20	1.60E+04	0.018
Cl_4 (CB 66) 2,3,4',4	$C_{12}H_6Cl_4$	32598-10-0	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.00210
Cl_4 (CB 55) 2,3,3',4	$C_{12}H_6Cl_4$	74338-24-2	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.00210

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_4 (CB 60/56) 2,3,4,4' / 2,3,3',4'	$C_{12}H_6Cl_4$	33025-41-1/41464-43-1	Group 3	MC-type CYP1A	5.98	9.53	25.84	292.00	268.20	6.80E+04	0.00210
Cl_5(CB 104) 2,2',4,6,6'	$C_{12}H_5Cl_5$	56558-16-8	Group 5	-	6.10	8.92	90.90	326.43	289.10	2.00E+04	0.00400
Cl_5 (CB 96) 2,2',3,6,6'	$C_{12}H_5Cl_5$	73575-54-9	Group 5	-	6.13	8.79	29.38	326.43	289.10	2.10E+04	0.00537
Cl_5 (CB 103) 2,2',4,5',6	$C_{12}H_5Cl_5$	60145-21-3	Group 5	-	6.20	8.92	35.00	326.43	289.10	2.00E+04	0.00400
Cl_5(CB 100) 2,2',4,4',6	$C_{12}H_5Cl_5$	39485-83-1	Group 2	-	6.23	8.63	62.62	326.43	289.10	3.00E+04	0.00780
Cl_5 (CB 94) 2,2',3,5,6'	$C_{12}H_5Cl_5$	73575-55-0	Group 5	-	6.26	8.98	24.87	326.43	289.10	2.10E+04	0.00350
Cl_5 (CB 95) 2,2',3,5',6	$C_{12}H_5Cl_5$	38379-99-6	Group 5	-	6.26	8.98	24.87	326.43	289.10	2.10E+04	0.00350
Cl_5 (CB 102/93) 2,2',4,5,6' / 2,2',3,5,6	$C_{12}H_5Cl_5$	68194-06-9/73575-56-1	Group 5	-	6.23	8.63	62.62	326.43	289.10	3.00E+04	0.00780
Cl_5(CB 98) 2,2',3',4,5	$C_{12}H_5Cl_5$	60233-25-2	Group 4	-	6.23	8.63	62.62	326.43	289.10	3.00E+04	0.00780
Cl_5 (CB 88) 2,2',3,4,6	$C_{12}H_5Cl_5$	55215-17-3	Group 5	-	6.48	8.31	34.65	326.43	289.10	1.20E+04	0.01610
Cl_5 (CB 91) 2,2',3,4',6	$C_{12}H_5Cl_5$	68194-05-8	Group 5	-	6.50	8.31	24.00	326.43	289.10	1.20E+04	0.01610
Cl_5 (CB 121) 2,3',4,5',6	$C_{12}H_5Cl_5$	56558-18-0	Group 1	-	6.48	8.63	28.60	326.43	289.10	3.00E+04	0.00780
Cl_5 (CB 92/84) 2,2',3,5,5' / 2,2',3,3',6	$C_{12}H_5Cl_5$	52663-61-3/52663-60-2	Group 5	-	6.26	8.98	24.87	326.43	289.10	2.10E+04	0.00350
Cl_5 CB (101/90) 2,2',4,5,5' / 2,2',3,4',5	$C_{12}H_5Cl_5$	35693-92-6/68194-07-0	Group 4	PB-type CYP2B	6.38	9.11	16.70	326.43	289.10	1.10E+04	0.00260
Cl_5 (CB 89) 2,2',3,4,6'	$C_{12}H_5Cl_5$	73575-57-2	Group 5	-	6.40	9.36	18.99	326.43	289.10	3.70E+03	0.00150
Cl_5 (CB 99) 2,2',4,4',5	$C_{12}H_5Cl_5$	38380-01-7	Group 2	-	6.38	9.36	21.68	326.43	289.10	3.70E+03	0.00150
Cl_5 (CB 113) 2,3,3',5',6	$C_{12}H_5Cl_5$	68194-10-5	Group 4	PB-type CYP2B	6.75	8.99	23.41	326.43	289.10	5.50E+03	0.00341

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_5 (CB 119) 2,3',4,4',6	$C_{12}H_5Cl_5$	56558-17-9	Group 2		6.74	8.24	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 112) 2,3,3',5,6	$C_{12}H_5Cl_5$	74472-36-9	Group 4	PB-type CYP2B	6.75	8.99	23.41	326.43	289.10	5.50E+03	0.00341
Cl_5 (CB 109/83) 2,3,3',4,6 / 2,2',3,3',5	$C_{12}H_5Cl_5$	74472-35-8/60145-20-2	Group 4	PB-type CYP2B	6.75	8.99	23.41	326.43	289.10	5.50E+03	0.00341
Cl_5 (CB 97/86) 2,2',3',4,5 / 2,2',3,4,5	$C_{12}H_5Cl_5$	41464-51-1/55312-69-1	Group 4	PB-type CYP2B	6.75	8.99	23.41	326.43	289.10	5.50E+03	0.00341
Cl_5 (CB 116/125/117) 2,3,4,5,6 / 2',3,4,5,6' / 2,3,4',5,6	$C_{12}H_5Cl_5$	18259-05-7/74472-39-2/68194-11-6	Group 4	PB-type CYP2B	6.74	8.24	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 115/87) 2,3,4',4,6 / 2,2',3,4,5'	$C_{12}H_5Cl_5$	74472-38-1/38380-02-8	Group 2	-	6.74	8.24	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 111) 2,3,3',5,5'	$C_{12}H_5Cl_5$	39635-32-0	Group 3	MC-type CYP1A	6.70	9.23	19.15	326.43	289.10	7.70E+03	0.00199
Cl_5 (CB 85) 2,2',3,4,4'	$C_{12}H_5Cl_5$	65510-45-4	Group 2	-	6.30	8.79	17.60	326.43	289.10	2.60E+04	0.00540
Cl_5 (CB 120) 2,3',4,5,5'	$C_{12}H_5Cl_5$	68194-12-7	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 110) 2,3,3',4',6	$C_{12}H_5Cl_5$	38380-03-9	Group 4	PB-type CYP2B	6.70	9.23	19.15	326.43	289.10	7.70E+03	0.00199
Cl_5 (CB 82) 2,2',3,3',4	$C_{12}H_5Cl_5$	52663-62-4	Group 4	PB-type CYP2B	6.26	8.79	26.65	326.43	289.10	2.60E+04	0.00540
Cl_5 (CB 124) 2,3,4,5,5'	$C_{12}H_5Cl_5$	70424-70-3	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 108/107) 2,3,3',4,5' / 2,3,3',4',5	$C_{12}H_5Cl_5$	70362-41-3/70424-68-9	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 123) 2',3,4,4',5	$C_{12}H_5Cl_5$	65510-44-3	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 106/118) 2,3,3',4,5 / 2,3',4,4',5	$C_{12}H_5Cl_5$	70424-69-0/31508-00-6	Group 3	MC-type CYP1A	6.74	8.24	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 114) 2,3,4,4',5	$C_{12}H_5Cl_5$	74472-37-0	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_5 (CB 122) 2',3,3',4,5	$C_{12}H_5Cl_5$	76842-07-4	Group 3	MC-type CYP1A	6.74	8.32	23.10	326.43	289.10	5.60E+03	0.01900
Cl_5 (CB 105) 2',3,3',4,4'	$C_{12}H_5Cl_5$	32598-14-4	Group 3	MC-type CYP1A	5.81	9.56	90.98	326.43	289.10	7.70E+03	0.00199
Cl_6 (CB 155) 2,2',4,4',6,6'	$C_{12}H_4Cl_6$	33979-03-2	Group 1	-	7.24	10.07	76.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 150) 2,2',3,4',6,6'	$C_{12}H_4Cl_6$	68194-08-1	Group 5	-	7.46	10.07	8.80	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 152) 2,2',3,5,6,6'	$C_{12}H_4Cl_6$	68194-09-2	Group 5	-	7.62	10.07	13.23	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 145) 2,2',3,4',5,5'	$C_{12}H_4Cl_6$	74472-40-5	Group 5	-	7.65	10.07	14.21	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 148) 2,2',3,4',5,6'	$C_{12}H_4Cl_6$	74472-41-6	Group 1	-	7.56	10.07	12.00	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 136) 2,2',3,3',6,6'	$C_{12}H_4Cl_6$	38411-22-2	Group 5	-	7.80	10.07	16.00	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 154) 2,2',4,4',5,6'	$C_{12}H_4Cl_6$	60145-22-4	Group 1	-	7.24	10.07	76.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 151) 2,2',3,5,5',6	$C_{12}H_4Cl_6$	52663-63-5	Group 5	-	7.46	10.07	8.80	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 135/144) 2,2',3,3',5,6' / 2,2',3,4,5',6	$C_{12}H_4Cl_6$	52744-13-5/68194-14-9	Group 5	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 147) 2,2',3,4',5,6	$C_{12}H_4Cl_6$	68194-13-8	Group 2	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 149) 2,2',3,4',5',6	$C_{12}H_4Cl_6$	38380-04-0	Group 5	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 139/140) 2,2',3,4,4',6 / 2,2',3,4,4',6'	$C_{12}H_4Cl_6$	56030-56-9/59291-64-4	Group 2	-	6.83	10.02	10.84	361.00	310.00	1.20E+03	0.00033
Cl_6 CB-143/1342,2',3,4,5,6' / 2,2',3,3',5,6	$C_{12}H_4Cl_6$	68194-15-0/52704-70-8	Group 5	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 142/131) 2,2',3,4,5,6 / 2,2',3,3',4,6	$C_{12}H_4Cl_6$	41411-61-4/61798-70-7	Group 5	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 133) 2,2',3,3',5,5'	$C_{12}H_4Cl_6$	35694-04-3	Group 1	-	6.64	9.99	9.93	361.00	310.00	8.10E+03	0.00036
Cl_6 (CB 146/161) 2,2',3,4',5,5' / 2,,3,3',4,5',6	$C_{12}H_4Cl_6$	51908-16-8/74472-43-8	Group 1	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 165) 2,3,3',5,5',6	$C_{12}H_4Cl_6$	74472-46-1	Group 1	-	7.55	10.83	5.98	361.00	310.00	5.00E+02	0.00005
Cl_6 (CB 132/153) 2,2',3,3',4,6'/ 2,2',4,4',5,5'	$C_{12}H_4Cl_6$	38380-05-1/35065-27-1	Group I	-	6.90	9.79	12.40	361.00	310.00	1.20E+03	0.00056
Cl_6 (CB 168) 2,3,4,4',5',6	$C_{12}H_4Cl_6$	59291-65-5	Group 1	-	7.55	10.83	5.98	361.00	310.00	5.00E+02	0.00005
Cl_6 (CB 141) 2,2',3,4,5,5'	$C_{12}H_4Cl_6$	52712-04-6	Group 4	PB-type CYP2B	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 137) 2,2',3,4,4',5	$C_{12}H_4Cl_6$	35694-06-5	Group 2	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030
Cl_6 (CB 130) 2,2',3,3',4,5'	$C_{12}H_4Cl_6$	52663-66-8	Group 2	-	6.67	10.07	9.50	361.00	310.00	2.70E+03	0.00030

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Cl_6 (CB 160/163/164/138) 2,3,3',4,5,6 / 2,3,3',4',5,6 / 2,3,3',4',5',6 / 2,2',3,4,4',5'	$C_{12}H_4Cl_6$	41411-62-5/74472-44-9/35065-28-2	Group 2	-	6.83	10.02	10.84	361.00	310.00	1.20E+03	0.00033
Cl_6 (CB 158) 2,3,3',4,4',6	$C_{12}H_4Cl_6$	74472-42-7	Group 2		6.83	10.02	10.84	361.00	310.00	1.20E+03	0.00033
Cl_6 (CB 129) 2,2',3,3',4,5	$C_{12}H_4Cl_6$	55215-18-4	Group 4	PB-type CYP2B	7.00	9.96	3.04	361.00	310.00	2.40E+03	0.00038
Cl_6 (CB 166) 2,3,4,4',5,6	$C_{12}H_4Cl_6$	41411-63-6	Group 2		7.40	10.82	5.98	361.00	310.00	5.20E+02	0.00005
Cl_6 (CB 159) 2,3,3',4,5,5'	$C_{12}H_4Cl_6$	39635-35-3	Group 3	MC-type CYP1A	7.40	11.55	5.98	361.00	310.00	5.20E+02	0.00005
Cl_6 (CB 162) 2,3,3',4',5,5'	$C_{12}H_4Cl_6$	39635-34-2	Group 4	PB-type CYP2B	7.40	11.55	5.98	361.00	310.00	5.20E+02	0.00005
Cl_6 (CB 128) 2,2,3,3',4,4'	$C_{12}H_4Cl_6$	38380-07-3	Group 2	-	7.00	9.96	3.04	361.00	310.00	2.40E+03	0.00038
Cl_6 (CB 167) 2,3',4,4',5,5'	$C_{12}H_4Cl_6$	52663-72-6	Group 3	MC-type CYP1A	7.40	11.55	5.98	361.00	310.00	5.20E+02	0.00005
Cl_6 (CB 156) 2,3,3',4,4',5	$C_{12}H_4Cl_6$	38380-08-4	Group 3	MC-type CYP1A	7.40	11.55	5.98	361.00	310.00	5.20E+02	0.00005
Cl_6 (CB 157) 2,3,3',4,4',5'	$C_{12}H_4Cl_6$	69782-90-7	Group 3	MC-type CYP1A	7.40	11.55	5.98	361.00	310.00	5.20E+02	0.00005
Cl7 (CB 188) 2,2',3,4',5,6,6'	$C_{12}H_3Cl_7$	74487-85-7	Group 1	-	7.20	10.58	8.41	395.32	330.90	4.51E+03	0.00009
Cl_7 (CB 184) 2,2',3,4,4',6,6'	$C_{12}H_3Cl_7$	74472-48-3	Group 1	-	7.11	9.56	1.62	395.32	330.90	4.51E+02	0.00094
Cl_7 (CB 179) 2,2',3,3',5,6,6'	$C_{12}H_3Cl_7$	52663-64-6	Group 5	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 176) 2,2',3,3',4,6,6'	$C_{12}H_3Cl_7$	52663-65-7	Group 5	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 186) 2,2',3,4,5,6,6'	$C_{12}H_3Cl_7$	74472-49-4	Group 5	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 178) 2,2',3,3',5,5',6	$C_{12}H_3Cl_7$	52663-67-9	Group 1	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 175) 2,2',3,3',4,5',6	$C_{12}H_3Cl_7$	40186-70-7	Group 1	-	7.14	10.11	5.40	395.32	330.90	4.12E+03	0.00027
Cl_7 (CB 187/182) 2,2',3,4',5,5',6 / 2,2',3,4,4',5,6'	$C_{12}H_3Cl_7$	52663-68-0/60145-23-5	Group I	-	7.20	10.58	8.41	395.32	330.90	4.51E+03	0.00009

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Cl_7 (CB 183) 2,2',3,4,4',5',6	$C_{12}H_3Cl_7$	52663-69-1	Group 1	-	7.00	9.86	1.62	395.32	330.90	7.00E+02	0.00048
Cl_7 (CB 185) 2,2',3,4,5,5',6	$C_{12}H_3Cl_7$	52712-05-7	Group 5	-	7.00	9.86	1.62	395.32	330.90	7.00E+02	0.00048
Cl_7 (CB 174/181) 2,2',3,3',4,5,6' / 2,2',3,4,4',5,6	$C_{12}H_3Cl_7$	38411-25-5/74472-47-2	Group 5	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 177) 2,2',3,3',4',5,6	$C_{12}H_3Cl_7$	52663-70-4	Group 2	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 171) 2,2',3,3',4,4',6	$C_{12}H_3Cl_7$	52663-71-5	Group 2	-	7.14	10.11	5.40	395.32	330.90	4.12E+03	0.00027
Cl_7 (CB 173) 2,2',3,3',4,5,6	$C_{12}H_3Cl_7$	68194-16-1	Group 2	-	7.14	10.11	5.40	395.32	330.90	4.12E+03	0.00027
Cl_7 (CB 192/172) 2,3,3',4,5,5',6 / 2,2',3,3',4,5,5'	$C_{12}H_3Cl_7$	74472-51-8/52663-74-8	Group 1	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 180) 2,2',3,4,4',5,5'	$C_{12}H_3Cl_7$	35065-29-3	Group 1	-	7.49	9.83	1.02	395.32	330.90	3.10E+02	0.00051
Cl_7 (CB 193) 2,3,3',4',5,5',6	$C_{12}H_3Cl_7$	69782-91-8	Group 1	-	7.46	10.61	12.15	395.32	330.90	4.00E+02	0.00009
Cl_7 (CB 191) 2,3,3',4,4',5',6	$C_{12}H_3Cl_7$	74472-50-7	Group 1	-	7.46	10.61	12.15	395.32	330.90	4.00E+02	0.00009
Cl_7 (CB 170/190) 2,2',3,3',4,4',5 / 2,3,3',4,4',5,6	$C_{12}H_3Cl_7$	35065-30-6/41411-64-7	Group 2	-	7.46	10.61	12.15	395.32	330.90	4.00E+02	0.00009
Cl_7 (CB 189) 2,3,3',4,4',5,5'	$C_{12}H_3Cl_7$	39635-31-9	Group 3	MC-type CYP1A	7.20	11.24	8.41	395.32	330.90	4.51E+03	0.00009
Cl_8 (CB 202) 2,2',3,3',5,5',6,6'	$C_{12}H_2Cl_8$	2136-99-4	Group 1	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 200) 2,2',3,3',4,5',6,6'	$C_{12}H_2Cl_8$	52663-73-7	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 204) 2,2',3,4,4',5,6,6'	$C_{12}H_2Cl_8$	74472-52-9	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 197) 2,2',3,3',4,4',6,6'	$C_{12}H_2Cl_8$	33091-17-7	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003

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Cl_8 (CB 199) 2,2',3,3',4,5,6,6'	$C_{12}H_2Cl_8$	52663-75-9	Group 5	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 198) 2,2',3,3',4,5,5',6'	$C_{12}H_2Cl_8$	68194-17-2	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 201) 2,2',3,3',4,5,5',6	$C_{12}H_2Cl_8$	40186-71-8	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 203/196) 2,2',3,4,4',5,5',6 / 2,2',3,3',4,4,5,6'	$C_{12}H_2Cl_8$	52663-76-0/42740-50-1	Group I	-	8.42	11.14	38.08	429.77	351.80	2.70E+02	0.00003
Cl_8 (CB 195) 2,2',3,3',4,4',5,6	$C_{12}H_2Cl_8$	52663-78-2	Group 2	-	7.80	11.24	10.13	429.77	351.80	2.70E+02	0.00002
Cl_8 (CB 194) 2,2',3,3',4,4',5,5'	$C_{12}H_2Cl_8$	35694-08-7	Group I	-	7.80	11.24	10.13	429.77	351.80	2.70E+02	0.00002
Cl_8 (CB 205) 2,3,3',4,4',5,5',6	$C_{12}H_2Cl_8$	74472-53-0	Group I	-	8.12	11.36	27.66	464.22	372.70	3.80E+01	0.00002
Cl_9 (CB 208) 2,2',3,3',4,5,5',6,6'	$C_{12}HCl_9$	52663-77-1	Group 1	-	8.16	11.36	32.50	464.22	372.70	3.80E+01	0.00002
Cl_9 (CB 207) 2,2',3,3',4,4',5,6,6'	$C_{12}HCl_9$	52663-79-3	Group I	-	8.12	11.36	27.66	464.22	372.70	3.80E+01	0.00002
Cl_9 (CB 206) 2,2',3,3',4,4',5,5',6	$C_{12}HCl_9$	40186-72-9	Group I	-	8.12	11.36	27.66	464.22	372.70	3.80E+01	0.00002
Cl_{10} (CB 209) 2,2',3,3',4,4',5,5',6,6'	$C_{12}Cl_{10}$	2051-24-3	Group I	-	8.40	11.87	20.84	498.66	393.60	2.10E+01	0.00001
Chlorobenzenes (CBz)											
1,3,5 TriCBz	$C_6H_3Cl_3$	108-70-3	-	-	3.80	5.84	192.50	181.45	138.90	8.46E+06	30.23000
1,2,4 TriCBz	$C_6H_3Cl_3$	120-82-1	-	-	4.70	5.81	145.00	181.45	137.90	3.33E+07	28.50000
1,2,3 TriCBz	$C_6H_3Cl_3$	81-61-6	-	-	4.46	6.50	147.00	181.45	146.10	1.93E+07	11.20000
1,2,3,5/1,2,4,5 TeCBz	$C_6H_2Cl_4$	95-94-3	-	-	4.50	8.17	122.00	247.00	179.60	5.60E+05	0.72000
1,2,3,4 TeCBz	$C_6H_2Cl_4$	634-66-2	-	-	4.50	8.17	62.00	247.00	179.60	5.92E+06	6.25000
PeCBz	C_6HCl_5	608-93-5	-	-	5.03	8.17	139.00	247.00	200.50	3.85E+05	0.21000
HCB	C_6Cl_6	118-74-1	-	-	5.50	7.11	131.00	285.00	182.50	5.02E+03	0.00150

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Hexachloro-cyclohexanes (HCHs)				-							
alpha-HCH	$C_6H_6Cl_6$	319-84-6	-	-	3.89	10.53	0.87	290.85	243.60	1.01E+07	0.87000
beta-HCH	$C_6H_6Cl_6$	319-85-7	-	-	3.81	10.53	0.07	290.85	243.60	2.40E+05	0.67000
gamma-HCH	$C_6H_6Cl_6$	319-86-8	-	-	4.14	10.53	0.08	290.85	243.60	2.13E+07	0.03100
				-							
p,p-DDT	$C_{14}H_9Cl_5$	50-29-3	-	-	6.91	10.75	1.31	354.50	333.50	1.00E-03	0.00002
o,p-DDT	$C_{14}H_9Cl_5$	789-02-7	-	-	6.76	10.60	1.31	354.50	333.50	1.00E-03	0.00002
p,p-DDE	$C_{14}H_8Cl_4$	72-55-9	-	-	6.96	9.44	7.95	318.04	305.20	1.74E+05	0.04440
o,p-DDE	$C_{14}H_8Cl_4$	3424-82-6	-	-	6.94	9.42	7.95	318.04	305.20	1.74E+05	0.00087
p,p-DDD	$C_{14}H_{10}Cl_4$	72-54-8	-	-	6.50	10.34	0.35	320.04	312.60	5.00E+04	0.00010
o,p-DDD	$C_{14}H_{10}Cl_4$	53-10-0	-	-	6.23	10.07	0.35	320.04	312.60	5.00E+04	0.00010
				-							
Cyclodienes											
aldrin	$C_{12}H_8Cl_6$	309-00-2	-	-	6.50	10.53	4.46	364.91	316.80	1.70E+04	0.00090
heptachlor	$C_{10}H_5Cl_7$	76-44-8	-	-	6.10	10.53	233.00	373.32	308.20	1.00E+05	0.05330
heptachlor epoxide	$C_{10}H_5Cl_7O$	1024-57-3	-	-	5.40	10.53	65.50	391.00	317.20	1.00E+05	0.00970
trans-chlordane	$C_{10}H_6Cl_8$	5103-74-2	-	-	6.22	10.10	0.26	409.78	336.50	5.00E+04	0.00013
cis-chlordane	$C_{10}H_6Cl_8$	5103-71-2	-	-	6.10	10.10	0.34	409.78	336.50	5.00E+04	0.00013
trans-nonachlor	$C_{10}H_5Cl_9$	39765-80-5	-	-	6.35	10.00	1.12	444.23	336.50	5.00E+04	0.00013
cis-nonachlor	$C_{10}H_5Cl_9$	5103-73-1	-	-	6.08	8.38	12.00	444.23	336.50	5.00E+04	0.00013
oxychlordane				-	6.02	10.53	6.60	389.00			
endosulfanI	$C_9H_6Cl_6O_3S$	115-29-7	-	-	3.40	10.29	1.31	391.00	312.80	5.30E+05	0.00002
endosulfanII	$C_9H_6Cl_6O_3S$	115-29-7	-	-	3.40	10.29	1.31	391.00	312.80	5.30E+05	0.00002
endosulfan sulfate				-	3.20	5.18	16.00	391.00		1.00E-03	0.00002
dieldrin	$C_{12}H_8Cl_6O$	60-57-1	-	-	5.40	8.73	1.12	380.91	318.20	1.40E+05	0.06000
methoxychlor	$C_{16}H_{15}Cl_3O_2$	72-43-5	-	-	3.20		1.00	345.65	354.30	1.00E+05	0.00019
mirex	$C_{10}Cl_{12}$	2385-85-5	-	-	7.50	7.96	839.40	545.50	403.20	1.30E+00	0.00022
octachlorostyrene					6.29	10.53	15.00	379.00	-	-	0.00022

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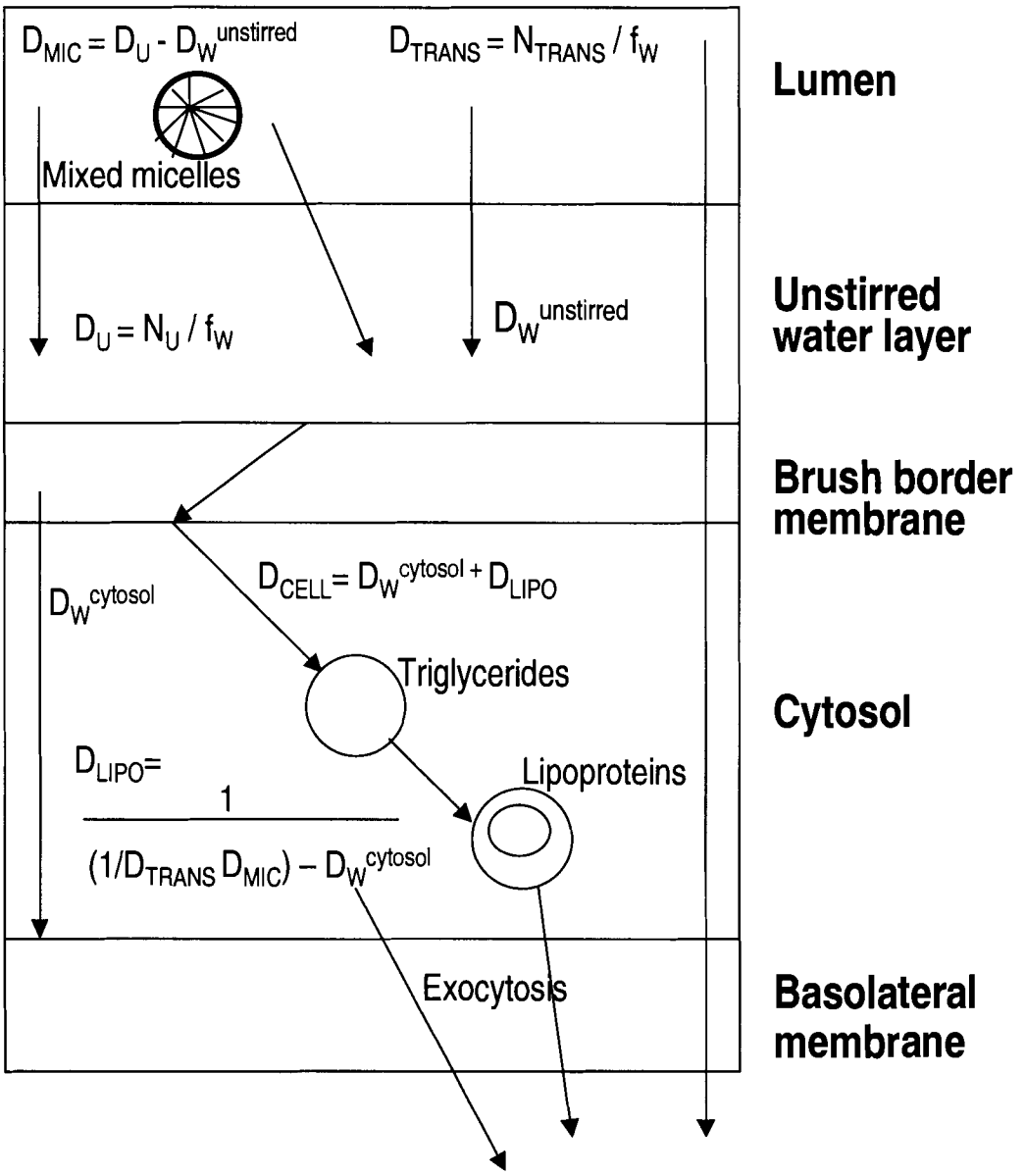
Dialkyl Phthalte Esters											
Dimethyl phtlate (DMP)	$C_{10}H_{10}O_4$	131-11-3	-	-	1.61	7.01	0.01	220.00	206.40	5.22E+09	0.26300
Diethyl phthalte (DEP)	$C_{12}H_{14}O_4$	84-66-2	-	-	2.54	7.55	0.03	266.00	254.00	5.91E+08	0.06500
Di-iso-butyl phthalte (DIBP)	$C_{16}H_{22}O_4$	84-69-5	-	-	4.27	8.54	0.13	278.00	342.80	9.90E+06	0.00100
Di n butyl pthalte (DBP)	$C_{16}H_{22}O_4$	84-74-2	-	-	4.27	8.54	0.13	287.00	342.80	9.90E+06	0.00100
Benzylbutyl phthalte (BBP)	$C_{19}H_{20}O_4$	85-68-7	-	-	4.70	8.78	0.21	295.00	364.80	3.80E+06	0.00250
Di (2-ethyl hexyl) phthalate (DEHP)	$C_{24}H_{38}O_4$	117-81-7	-	-	7.73	10.53	3.95	358.00	520.40	2.49E+03	0.00003
Di n-octyl pthalate (DnOP)	$C_{24}H_{38}O_4$	11-78-4	-	-	7.73	10.53	3.95	395.00	520.40	2.49E+03	0.00003
Di n-nonyl phthalate (DnNP)	$C_{26}H_{42}O_4$	84-76-4	-	-	8.60	11.03	9.26	450.00	564.80	3.08E+02	0.00001
Monoalkyl Phthalate Esters											
MMP	$C_9O_4H_8$	4376-18-5	-	-	1.17	-	-	180.16	-	1.54E+10	-
MEP	$C_{10}O_4H_{10}$	2306-33-4	-	-	1.67	-	-	194.19	-	4.86E+09	-
MBuP	$C_{12}O_4H_{14}$	131-70-4	-	-	2.53	-	-	222.24	-	6.50E+08	-
M C6-iso-mix	$C_{14}O_4H_{18}$	-	-	-		-	-	250.29	-		-
MBzP	$C_{15}O_4H_{12}$	2528-16-7	-	-	2.95	-	-	256.26	-	2.59E+08	-
M C7-iso-mix	$C_{15}O_4H_{20}$	-	-	-		-	-	264.32	-		-
MEHP+MnOP (M C8-iso mix)	$C_{16}O_4H_{22}$	4376-20-9	-	-	4.25	-	-	278.35	-	1.11E+07	-
M C9-iso-mix	$C_{17}O_4H_{24}$	-	-	-	4.68	-	-	292.37	-	3.99E+06	-
M-C10-iso-mix	$C_{18}O_4H_{26}$	-	-	-	5.11	-	-	306.40	-	1.43E+06	-
Brominated diphenyl ethers											
Br_1 (BDE 1) 2,	$C_{12}H_9BrO$	-	-	-	5.53	9.30	-	249.30	-	-	-
Br_1 (BDE 2) 3,	$C_{12}H_9BrO$	-	-	-	5.53	9.30	-	249.30	-	-	-
Br_1 (BDE 3) 4,	$C_{12}H_9BrO$	101-55-3	-	-	5.53	9.30	-	249.30	-	-	-

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Br_2 (BDE 10) 2,6	$C_{12}H_8Br_2O$	-	-	-	5.53	9.30	-	327.90	-	-	-
Br_2 (BDE 7) 2,4	$C_{12}H_8Br_2O$	-	-	-	5.53	9.30	-	327.90	-	-	-
Br_2 (UI DiBDE 1)	$C_{12}H_8Br_2O$	-	-	-	5.53	9.30	-	327.90	-	-	-
Br_2 (BDE 8/11) 2,4'/3,3'	$C_{12}H_8Br_2O$	-	-	-	5.53	9.30	-	327.90	-	-	-
Br_2 (BDE 12) 3,4	$C_{12}H_8Br_2O$	-	-	-	5.53	9.30	-	327.90	-	-	-
Br_2 (BDE 13) 3,4'	$C_{12}H_8Br_2O$	-	-	-	6.84	9.30	-	327.90	-	-	-
Br_2 (BDE 15) 4,4'	$C_{12}H_8Br_2O$	2050-47-7	-	-	6.84	9.30	-	327.90	-	-	-
Br_3 (BDE 30) 2,4,6	$C_{12}H_7Br_3O$	-	-	-	6.84	9.30	-	406.80	-	-	-
Br_3 (UI TriBDE 1)	$C_{12}H_7Br_3O$	-	-	-	6.84	9.30	-	406.80	-	-	-
Br_3 (BDE 32) 2,4',6	$C_{12}H_7Br_3O$	-	-	-	6.84	9.30	-	406.80	-	-	-
Br_3 (BDE 17) 2,2',4	$C_{12}H_7Br_3O$	-	-	-	6.84	9.30	-	406.80	-	-	-
Br_3 (BDE 25) 2,3',4	$C_{12}H_7Br_3O$	-	-	-	6.84	9.50	-	406.80	-	-	-
Br_3 (UI TriBDE 2)	$C_{12}H_7Br_3O$	-	-	-	6.84	9.50	-	406.80	-	-	-
Br_3 (BDE 28/33) 2,4,4'/2',3,4	$C_{12}H_7Br_3O$	49690-94-0	-	-	6.84	9.50	-	406.80	-	-	-
Br_3 (BDE 35) 3,3',4	$C_{12}H_7Br_3O$	-	-	-	6.84	9.50	-	406.80	-	-	-
Br_3 (BDE 37) 3,4,4'	$C_{12}H_7Br_3O$	-	-	-	6.84	9.50	-	406.80	-	-	-
Br_4 (BDE 75) 2,4,4',6	$C_{12}H_6Br_4O$	-	-	-	7.66	10.53	-	485.80	-	-	-
Br_4 (BDE 49) 2,2',4,5'	$C_{12}H_6Br_4O$	-	-	-	7.66	10.53	-	485.80	-	-	-
Br_4 (BDE 71) 2,3',4',6	$C_{12}H_6Br_4O$	-	-	-	7.66	10.53	-	485.80	-	-	-
Br_4 (BDE 47) 2,2',4,4'	$C_{12}H_6Br_4O$	40088-47-9	-	-	7.66	10.53	0.40	485.80	-	-	0.00052
Br_4 (BDE 66) 2,3',4,4'	$C_{12}H_6Br_4O$	-	-	-	7.66	10.82	-	485.80	-	-	-
Br_4 (BDE 77) 3,3',4,4'	$C_{12}H_6Br_4O$	-	-	-	7.00	10.87	-	485.80	-	-	-
Br_5 (UI PeBDE 1)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (UI PeBDE 2)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (UI PeBDE 3)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (UI PeBDE 4)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (UI PeBDE 5)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (UI PeBDE 6)	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (BDE 100) 2,2',4,4',6	$C_{12}H_5Br_5O$	-	-	-	7.00	11.13	-	564.70	-	-	-
Br_5 (BDE 101) 2,2',4,5,5'	$C_{12}H_5Br_5O$	-	-	-	7.00	11.31	-	564.70	-	-	-
Br_5 (BDE 119) 2,3',4,4',6	$C_{12}H_5Br_5O$	-	-	-	7.00	11.31	-	564.70	-	-	-
Br_5 (UI PeBDE 7)	$C_{12}H_5Br_5O$	-	-	-	8.10	11.31	-	564.70	-	-	-
Br_5 (UI PeBDE 8)	$C_{12}H_5Br_5O$	-	-	-	8.10	11.31	-	564.70	-	-	-

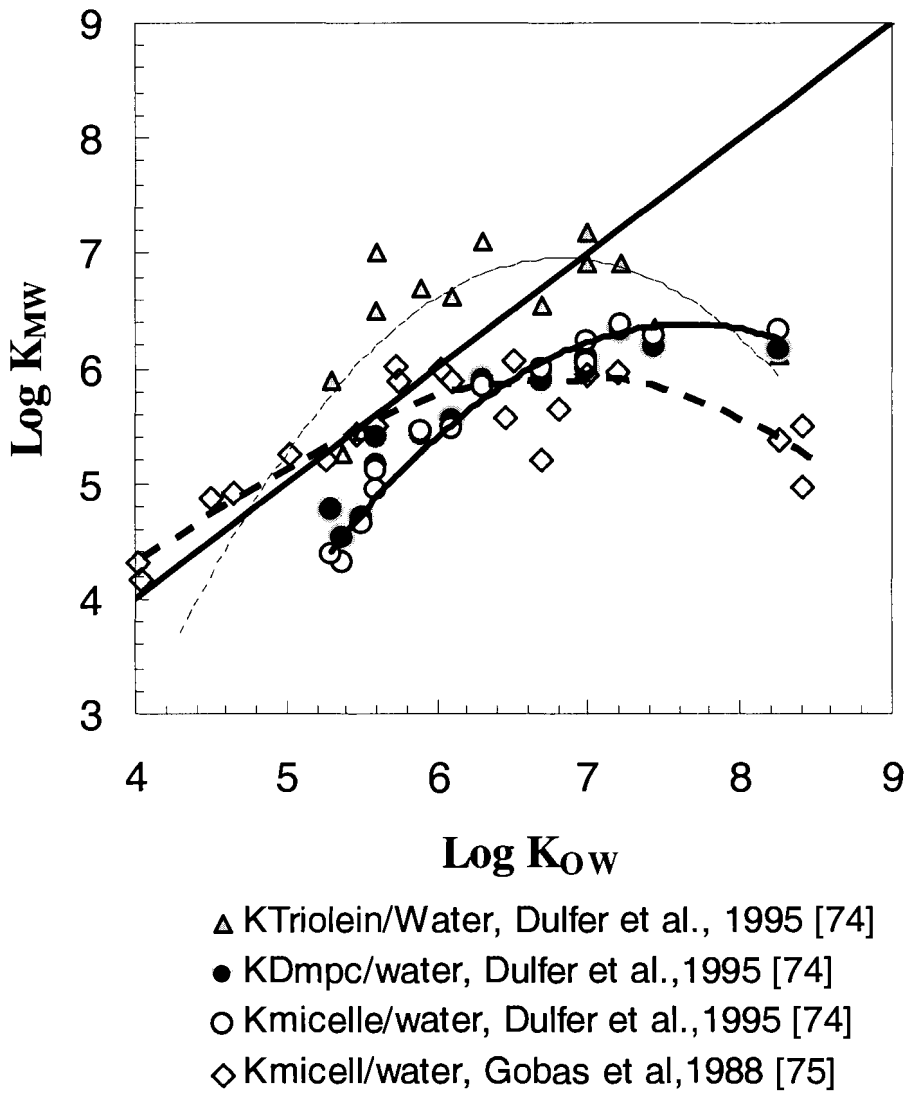
Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Br_5 (BDE 99) 2,2',4,4',5	$C_{12}H_5Br_5O$	32534-81-9	-	-	8.10	11.31	0.12	564.70	-	-	0.00004
Br_5 (BDE 116) 2,3,4,5,6	$C_{12}H_5Br_5O$	-	-	-	8.10	11.66	-	564.70	-	-	-
Br_5 (BDE 118) 2,3',4,4',5	$C_{12}H_5Br_5O$	-	-	-	8.10	11.66	-	564.70	-	-	-
Br_5 (BDE 85) 2,2',4,4',5	$C_{12}H_5Br_5O$	-	-	-	8.10	11.66	-	564.70	-	-	-
Br_5 (BDE 126) 3,3',4,4',5	$C_{12}H_5Br_5O$	-	-	-	8.10	11.97	-	564.70	-	-	-
Br_5 (BDE 105) 2,3,3',4,4'	$C_{12}H_5Br_5O$	-	-	-	8.10	11.97	-	564.70	-	-	-
Br_5 (BDE 155) 2,2',4,4',6,6'	$C_{12}H_5Br_5O$	-	-	-	8.10	11.97	-	564.70	-	-	-
Br_5 (BDE 154) 2,2',4,4',5,6'	$C_{12}H_5Br_5O$	-	-	-	8.10	11.92	-	564.70	-	-	-
Br_6 (UI HxBDE 1)	$C_{12}H_4Br_6O$	-	-	-	8.71	11.92	-	643.60	-	-	-
Br_6 (UI HxBDE 2)	$C_{12}H_4Br_6O$	-	-	-	8.71	11.92	-	643.60	-	-	
Br_6 (BDE 153) 2,2',4,4',5,5'	$C_{12}H_4Br_6O$	36483-60-0	-	-	8.71	11.82	-	643.60	-	-	-
Br_6 (BDE 140) 2,2',3,4,4',6'	$C_{12}H_4Br_6O$	-	-	-	8.71	11.82	-	643.60	-	-	-
Br_6 (BDE 138/166) 2,2',3,4,4',5'/2,3,4,4',5,6	$C_{12}H_4Br_6O$	-	-	-	8.71	11.50	-	643.60	-	-	-
Br_7 (BDE 183) 2,2',3,4,4',5',6	$C_{12}H_2Br_8O$	-	-	-	8.71	11.96	-	801.40	-	-	-
Br_7 (BDE 181) 2,2',3,4,4',5,6	$C_{12}H_2Br_8O$	-	-	-	8.71	11.96	-	801.40	-	-	-
Br_7 (BDE 190) 2,3,3',4,4',5,6	$C_{12}H_2Br_8O$	32536-52-0	-	-	8.71	11.96	0.01	801.40	-	-	0.00001
Br_{10} (BDE 209) 2,2',3,3',4,4',5,5',6,6'	$C_{12}Br_{10}O$	1163-19-5	-	-	9.97	-	0.00	959.20	-	-	0.00040
Synthetic Musks											
Cashmeran-Me (DPMI-)	$C_{14}H_{22}O$	33704-61-9	-	-	4.90	7.29	9.90	206.30	-	1.70E+05	5.20000
Celestolide-Me (ADBI-)	$C_{17}H_{24}O$	13171-00-1	-	-	6.60	6.73	1801.00	244.30	-	1.50E+04	0.02000

Chemical Name	Formula	CAS #	Group	Enzyme Type	Log K_{OW}	log K_{OA}	HLC ($mol \cdot m^3$ Pa)	MW ($g \cdot mol^{-1}$)	Le Bas Molar.VoL ($cm^3 \cdot mol^{-1}$)	Water Solubility ($ng \cdot L^{-1}$)	Vapor Pressure (Pa)
Phantolide-Me (AHMI-)	$C_{17}H_{24}O$	15323-35-0	-	-	6.70	7.27	646.00	244.30	-	2.70E+04	0.02400
Musk Ambrette	$C_{12}H_{16}N_2O_5$	83-66-9	-	-	4.50	9.88	0.01	295.00	-		0.00004
Traseolide (ATII)	$C_{18}H_{26}O$	68140-48-7	-	-	8.10	9.55	85.10	258.40	-	8.50E+04	1.20000
Galaxolide (HHCB)	$C_{18}H_{26}O$	1222-05-5	-	-	5.90	8.23	11.30	258.40	-	1.75E+06	0.07300
Tonalide (AHTN)	$C_{18}H_{26}O$	1506-02-1	-	-	5.70	7.99	12.50	258.40	-	1.25E+06	0.06800
Musk Xylene	$C_{12}H_{15}N_3O_6$	81-15-2	-	-	4.90	10.03	0.02	297.20	-	4.90E+05	0.00003
Musk Moskene	$C_{14}H_{18}N_2O_4$	116-66-5	-	-	4.90	10.03	0.02	297.20	-		0.00003
Musk Tibetene	$C_{13}H_{18}N_2O_4$	145-39-1	-	-	5.00	-	0.02	266.30	-	-	-
Musk Ketone	$C_{14}H_{18}N_2O_5$	81-14-1	-	-	4.30	9.90	0.01	294.30	-	1.90E+06	0.00004

Appendix 3 Illustration of proposed micelle mediated uptake model outlined in Dulfer et al. (*109*). The model is in fugacity format and D values are represented as chemical conductivity parameters, including $D_W^{UNSTIRRED}$, $D_W^{CYTOSOL}$, D_{MIC}, D_{LIPO} and D_{CELL} describing conductivity in unstirred water layer, cytosol, mixed micelles, lipoproteins, and enterocytes, respectively. D_U represents conductivity of uptake into the enterocyte and D_{TRANS} represents conductivity for transport over the enterocyte.



Appendix 4 Membrane-water partition coefficients K_{MW} vs. chemical log K_{OW} (ranging from 4 to 9), for Triolein ($K_{TRIOLEIN}$), DMPC vesicles (K_{DMPC}) and *n*-octanol.



Appendix 5 Di (Cl_2) to Deca (Cl_{10}) chlorinated biphenyl (PCB) congener concentrations in lichens and macro-algae (ng·g^{-1} lipid equivalent wt.), sediment (ng·g^{-1} OC wt) and tissues' of various marine biota (ng·g^{-1} lipid wt.) collected from E. Hudson's Bay during May and September 1999-2002.

		LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		**MACRO ALGAE** (*F. gardneri*) **(tissue)** (*n* =11)		**SEDIMENTS** (***n* ponar grabs)** (*n* =12)		**CAPELIN** (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_2 (PCB 7/9)	III	0.04	*0.014-0.13*	0.28	*0.11-0.69*	1.16	*0.37-3.47*	-	-
Cl_2 (PCB 6)	III	0.06	*0.021-0.17*	0.42	*0.17-1.04*	1.79	*0.59-5.48*	-	-
Cl_2 (PCB 8/5)	III	0.28	*0.10-0.78*	0.95	*0.21-4.31*	7.08	*1.97-25.4*	1.25	*0.56-2.81*
Cl_2 (PCB 4/10)	IV	0.16	*0.060-0.43*	0.55	*0.12-2.51*	4.65	*1.50-14.4*	0.32	*0.16-0.65*
Cl_3 (PCB 23/34)	III	-	-	-	-	-	-	-	-
Cl_3 (PCB 29)	III	-	-	-	-	-	-	-	-
Cl_3 (PCB 26)	III	0.03	*0.013-0.082*	0.16	*0.070-0.38*	0.94	*0.31-2.86*	0.83	*0.37-1.88*
Cl_3 (CB 25)	III	0.02	*0.006-0.055*	0.08	*0.034-0.20*	0.44	*0.14-1.34*	0.31	*0.13-0.74*
Cl_3 (CB 31)	III	0.17	*0.064-0.456*	0.87	*0.30-2.53*	4.61	*1.39-15.25*	5.87	*2.55-13.56*
Cl_3 (CB 28)	III	0.26	*0.11-0.62*	0.75	*0.26-2.16*	4.95	*1.65-14.9*	5.82	*2.66-12.76*
Cl_3 (CB 21)	III	-	-	-	-	-	-	-	-
Cl_3 (CB 33/20)	III	-	-	-	-	-	-	2.04	*0.87-4.75*
Cl_3 (CB 19)	IV	0.04		0.28	*0.12-0.67*	-	-	-	-
Cl_3 (CB30)	IV	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	IV	0.25	*0.098-0.63*	1.05	*0.27-4.16*	7.16	*2.12-24.19*	0.72	*0.33-1.60*
Cl_3 (CB 17)	IV	0.10	*0.037-0.25*	0.64	*0.25-1.61*	2.89	*0.87-9.62*	3.23	*1.55-6.74*
Cl_3 (CB 27/24)	IV	0.05	*0.021-0.10*	-	-			0.44	*0.089-2.18*
Cl_3 (CB 16/32)	IV	0.22	*0.089-0.56*	1.49	*0.58-3.83*	6.22	*1.99-19.50*	1.72	*0.76-3.89*
Cl_3 (CB 22)	III	-	-	-	-	-	-	1.62	*0.67-3.94*

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 54)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 53)	IV	-	-	-	-	-	-	0.35	*0.14-0.88*
Cl_4 (CB 51)	V	-	-	-	-	-	-	0.05	
Cl_4 (CB 45)	V	-	-	-	-	-	-	0.34	*0.13-0.88*
Cl_4 (CB 46)	V	-	-	-	-	-	-	11.04	*4.52-27.01*
Cl_4 (CB 73/52)	IV	0.12	*0.047-0.31*	0.38	*0.11-1.25*	3.01	*0.99-9.08*	1.83	*0.74-4.55*
Cl_4 (CB 69)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	IV	0.08	*0.029-0.21*	0.30	*0.10-0.87*	1.70	*0.54-5.34*	-	-
Cl_4 (CB 43)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	IV	0.04	*0.018-0.093*	-	-	-	-	-	-
Cl_4 (CB 65)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	IV	0.09	*0.035-0.24*	0.39	*0.14-1.13*	2.45	*0.76-7.84*	0.74	*0.34-1.64*
Cl_4 (CB 59/42)	IV	0.05	*0.021-0.14*	0.24	*0.089-0.65*	0.98	-	0.07	*0.027-0.19*
Cl_4 (CB 72)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	IV	0.15	*0.056-0.40*	0.57	*0.23-1.42*	2.97	*0.94-9.42*	0.21	*0.051-0.82*
Cl_4 (CB 68)	III	-	-	-	-	-	-	0.39	*0.048-3.14*
Cl_4 (CB 40)	IV	-	-	-	-	-	-	0.25	*0.11-0.56*
Cl_4 (CB 57)	III	-	-	-	-	-	-	0.05	
Cl_4 (CB 67)	III	-	-	-	-	-	-	0.11	*0.044-0.29*
Cl_4 (CB 58)	III	-	-	-	-	-	-	-	-

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 63)	III	-	-	-	-	-	-	0.25	*0.10-0.59*
Cl_4 (CB 61/74)	III	-	-	-	-	-	-	2.42	*0.99-5.92*
Cl_4 (CB 70/76)	III	-	-	0.13	-	-	-	4.07	*1.62-10.22*
Cl_4 (CB 66)	III	-	-	-	-	-	-	2.84	*1.15-7.03*
Cl_4 (CB 55)	III	-	-	-	-	-	-	0.05	
Cl_4 (CB 60/56)	III	-	-	-	-	-	-	0.80	*0.30-2.12*
Cl_5 (CB 104)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 103)	IV	-	-	-	-	-	-	0.11	*0.050-0.24*
Cl_5 (CB 100)	II	-	-	-	-	-	-	0.06	
Cl_5 (CB 94)	V	-	-	-	-	-	-	0.12	*0.053-0.29*
Cl_5 (CB 95)	V	0.12	*0.041-0.37*	0.37	*0.12-1.14*	1.89	*0.61-5.88*	8.23	*3.24-20.9*
Cl_5 (CB 102/93)	V	-	-	-	-	-	-	-	-
Cl_5 (CB 98)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 88)	V	-	-	-	-	-	-	0.10	*0.037-0.25*
Cl_5 (CB 91)	V	-	-	-	-	-	-	0.58	*0.23-1.45*
Cl_5 (CB 121)	I	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	V	0.06		-	-	-	-	-	-
Cl_5 CB (101/90)	IV	0.14	*0.041-0.49*	0.42	*0.16-1.10*	1.63	*0.53-4.95*	12.28	*5.26-28.7*
Cl_5 (CB 89)	V	0.06	*0.026-0.14*	-	-	-	-	1.67	*0.76-3.66*
Cl_5 (CB 99)	II	0.09	*0.028-0.26*	0.30	*0.14 -0.65*	0.95		9.01	*4.05 -20.03*
Cl_5 (CB 113)	IV	-	-	-	-	-	-	-	-

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 119)	II	-	-	-	-	-	-	0.57	*0.25-1.31*
Cl_5 (CB 112)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 109/83)	IV	-	-	-	-	-	-	0.44	*0.20-0.97*
I_5 (CB 97/86)	IV	0.06	*0.019-0.21*	-	-	-	-	2.43	*1.06-5.56*
Cl_5 (CB 116/125/117)	IV			-	-	-	-	-	-
Cl_5 (CB 115/87)	II	0.09	*0.024-0.32*	-	-	-	-	4.85	*2.16-10.91*
Cl_5 (CB 111)	III			-	-	-	-	-	-
Cl_5 (CB 85)	II	0.06	*0.021-0.18*	-	-	-	-	2.52	*1.13-5.60*
Cl_5 (CB 120)	III			-	-	-	-	-	-
Cl_5 (CB 110)	IV	0.22	*0.066-0.77*	0.30	*0.12 -0.74*	1.73	*0.79-3.83*	4.89	*2.11-11.33*
Cl_5 (CB 82)	IV	0.06		-	-	-	-	0.84	*0.375-1.88*
Cl_5 (CB 124)	III	-	-	-	-	-	-	0.26	*0.11-0.59*
Cl_5 (CB 108/107)	III	-	-	-	-	-	-	1.01	*0.45 -2.29*
Cl_5 (CB 123)	III	-	-	-	-	-	-	9.93	*4.39-22.5*
Cl_5 (CB 106/118)	III	0.19	*0.058-0.62*	0.39	*0.151-1.01*	1.94	*0.66-5.72*	-	-
Cl_5 (CB 114)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 122)	III	-	-	-	-	-	-	0.27	*0.14-0.54*
Cl_5 (CB 105)	III	-	-	-	-	-	-	2.60	*1.16-5.85*
Cl_6 (CB 155)	I	-	-	-	-	-	-	-	-
Cl_6 (CB 150)	V	-	-	-	-	-	-	-	-
Cl_6 (CB 152)	V	-	-	-	-	-	-	-	-
Cl_6 (CB 145)	V	-	-	-	-	-	-	-	

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 148)	I	-	-	-	-	-	-	1.59	*0.73-3.51*
Cl_6 (CB 136)	V	0.06	*0.015-0.26*	-	-	-	-	1.73	
Cl_6 (CB 154)	I	-	-	-	-	-	-	0.40	*0.16-0.99*
Cl_6 (CB 151)	V	0.05	*0.020-0.13*	0.29	*0.12-0.71*	0.81		4.66	*2.06-10.6*
Cl_6 (CB 135/144)	V	0.06	*0.015-0.27*	-	-	-	-	2.35	*0.98-5.64*
Cl_6 (CB 147)	II	-	-	-	-	-	-	0.30	*0.13-0.72*
Cl_6 (CB 149)	V	0.16	*0.036-0.71*	0.40	*0.12-1.35*	2.38	*0.82-6.92*	6.86	*2.61-18.0*
Cl_6 (CB 139/140)	II	-	-	-	-	-	-	0.09	
Cl_6 CB-143/134	V	-	-	-	-	-	-	0.52	*0.23-1.20*
Cl_6 (CB 142/131)	V	-	-	-	-	-	-	0.34	*0.14-0.82*
Cl_6 (CB 133)	I	-	-	-	-	-	-	0.10	
Cl_6 (CB 146/161)	I	0.06	*0.015-0.21*	0.18	-	-	-	2.89	*1.17-7.16*
Cl_6 (CB 165)	I	-	-	-	-	-	-	-	-
Cl_6 (CB 132/153)	I	0.24	*0.067-0.89*	0.56	*0.16-2.01*	3.19	*1.02-10.1*	18.04	*7.14-45.6*
Cl_6 (CB 168)	I	0.05	*0.015-0.19*	-	-	-	-	1.25	*0.53-2.93*
Cl_6 (CB 141)	IV	0.07	*0.016-0.27*	-	-	-	-	1.60	*0.69-3.65*
Cl_6 (CB 137)	II	-	-	-	-	-	-	1.31	*0.59-2.92*
Cl_6 (CB 160/163/164/138)	II	0.24	*0.066-0.90*	0.51	*0.18-1.51*	2.69	*0.85-8.54*	15.80	*7.18-34.8*
Cl_6 (CB 130)	II	-	-	-	-	-	-	0.51	*0.22-1.18*
Cl_6 (CB 158)	II	0.07	*0.021-0.19*	-	-	-	-	0.74	*0.33-1.66*
Cl_6 (CB 129)	IV	-	-	-	-	-	-	0.24	*0.12-0.49*
Cl_6 (CB 166)	II	-	-	-	-	-	-	-	

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 159)	III	-	-	-	-	-	-	0.19	*0.085-0.44*
Cl_6 (CB 162)	IV	-	-	0.04	-	-	-	0.38	*0.043-3.34*
Cl_6 (CB 128)	II	0.06	*0.021-0.19*	-	-	-	-	1.50	*0.66-3.40*
Cl_6 (CB 167)	III	-	-	-	-	-	-	0.34	*0.15-0.77*
Cl_6 (CB 156)	III	-	-	-	-	-	-	0.44	*0.19-1.00*
Cl_6 (CB 157)	III	-	-	-	-	-	-	0.24	*0.11-0.52*
Cl7 (CB 188)	I	-	-	-	-	-	-	0.21	*0.068-0.64*
Cl_7 (CB 184)	I	-	-	-	-	-	-	0.14	*0.044-0.46*
Cl_7 (CB 179)	V	0.07	*0.017-0.29*	-	-	-	-	1.02	*0.44-2.38*
Cl_7 (CB 176)	V	-	-	-	-	-	-	0.30	*0.13-0.71*
Cl_7 (CB 186)	V	-	-	-	-	-	-	-	
Cl_7 (CB 178)	I	0.05		-	-	-	-	0.87	*0.38-1.99*
Cl_7 (CB 175)	I			-	-	-	-	0.16	*0.067-0.40*
Cl_7 (CB 187/182)	I	0.12	*0.032-0.42*	0.38	*0.14-1.04*	2.57	*0.78-8.41*	4.62	*2.02-10.5*
Cl_7 (CB 183)	I	0.06	*0.017-0.19*	-	-	-	-	1.27	*0.54-2.98*
Cl_7 (CB 185)	V	-	-	-	-	-	-	0.19	*0.071-0.52*
Cl_7 (CB 174/181)	V	0.09	*0.025-0.30*	0.24	*0.088-0.65*	1.40	*0.42-4.70*	1.60	*0.59-4.34*
Cl_7 (CB 177)	II	0.09		-	-	-	-	1.07	*0.43-2.62*
Cl_7 (CB 171)	II	0.05		-	-	-	-	0.37	*0.16-0.90*
Cl_7 (CB 173)	II	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	I	0.04		-	-			0.31	*0.13-0.73*
Cl_7 (CB 180)	I	0.13	*0.035-0.51*	0.34	*0.12-0.96*	2.08	*0.66-6.51*	3.21	*1.27-8.09*

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_7 (CB 193)	I	-	-	-	-	-	-	0.24	*0.10-0.57*
Cl_7 (CB 191)	I	-	-	-	-	-	-	0.11	*0.041-0.30*
Cl_7 (CB 170/190)	II	0.08	*0.019-0.30*	-	-	-	-	1.10	*0.44-2.78*
Cl_7 (CB 189)	III	-	-	-	-	-	-	0.27	*0.12-0.61*
Cl_8 (CB 202)	I	0.04		-	-	-	-	0.70	*0.29-1.66*
Cl_8 (CB 200)	I	-	-	-	-	-	-	0.66	*0.27-1.65*
Cl_8 (CB 204)	I	-	-	-	-	-	-	-	
Cl_8 (CB 197)	I	-	-	-	-	-	-	0.17	*0.066-0.45*
Cl_8 (CB 199)	V	-	-	-	-	-	-	0.12	*0.035-0.39*
Cl_8 (CB 198)	I	-	-	-	-	-	-	0.80	*0.31-2.06*
Cl_8 (CB 201)	I	0.06	*0.019-0.22*	-	-	-	-	0.81	*0.30-2.19*
Cl_8 (CB 203/196)	I	0.04	*0.015-0.12*	-	-	-	-	0.80	*0.28-2.31*
Cl_8 (CB 195)	II	-	-	-	-	-	-	0.14	*0.043-0.47*
Cl_8 (CB 194)	I	0.10		-	-	-	-	0.44	*0.16-1.22*
Cl_8 (CB 205)	I	-	-	-	-	-	-	0.04	
Cl_9 (CB 208)	I	-	-	-	-	-	-	0.13	*0.050-0.32*
Cl_9 (CB 207)	I	-	-	-	-	-	-	0.13	*0.054-0.32*
Cl_9 (CB 206)	I	-	-	-	-	-	-	0.23	*0.098-0.55*
Cl_{10} (CB 209)	I	0.03		-	-	1.32	*0.609-2.88*	0.13	*0.050-0.34*
$\sum Cl_2$		0.54	*0.19-1.48*	1.46	*0.26-8.13*	11.9	*2.74-51.3*	1.35	*0.58-3.15*
$\sum Cl_3$		1.06	*0.42-2.69*	3.03	*0.55-16.8*	24.9	*7.71-80.9*	18.4	*8.28-40.9*
$\sum Cl_4$		0.50	*0.18-1.36*	0.82	*0.14-4.68*	9.88	*3.28-29.7*	19.7	*6.29-61.5*

		LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*		-		-		0.18 ± 0.10		-	
% Lipid ± *SD*		0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{EQ}) ± *SD*		2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
$\sum Cl_5$		0.85	*0.23-3.13*	0.95	*0.22-4.01*	4.30	*1.48-12.6*	62.5	*27.7-141*
$\sum Cl_6$		0.67	*0.15-2.93*	1.32	*0.32-5.47*	6.46	*1.85-22.6*	57.9	*25.4-132*
$\sum Cl_7$		0.42	*0.096-1.82*	0.90	*0.34-2.39*	3.92	*1.07-14.4*	17.1	*7.17-40.6*
$\sum Cl_8$		0.09	*0.016-0.55*	-	-	-	-	3.77	*1.48-9.58*
$\sum Cl_9$		-	-	-	-	-	-	0.47	*0.19-1.15*
$\sum Cl_{10}$		0.03		-	-	1.32	*0.61-2.88*	0.13	*0.050-0.34*
∑Diortho PCBs		3.12	*0.89-11.0*	4.12	*0.67-25.1*	42.4	*14.9-120*	140	*61.0- 322*
∑Mono ortho PCBs		1.06	*0.40-2.81*	1.83	*0.28-11 9*	20.4	*6.07-68.5*	42.5	*18.7-96.9*
∑PCBs		4.22	*1.29-13.8*	5.97	*0.96-37.1*	63.2	*21.6-186*	183	*80.5-416*

Appendix 5 continued.

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_2 (PCB 7/9)	III	0.16	*0.064-0.39*	0.15	*0.065-0.34*			0.03	*0.010-0.066*
Cl_2 (PCB 6)	III	0.22	*0.085-0.55*	0.21	*0.093-0.48*			0.05	*0.020-0.10*
Cl_2 (PCB 8/5)	III	0.81	*0.27-2.44*	0.92	*0.32-2.67*	0.19	*0.078-0.46*	0.37	*0.097-1.43*
Cl_2 (PCB 4/10)	IV	0.37	*0.11-1.28*	0.49	*0.14-1.73*	0.09	*0.033-0.22*	0.20	*0.055-0.69*
Cl_3 (PCB 23/34)	III	-	-	-	-	-	-	-	-
Cl_3 (PCB 29)	III	-	-	-	-	-	-	-	-
Cl_3 (PCB 26)	III	0.11	*0.049-0.27*	0.11	*0.048-0.27*	0.19	*0.064-0.56*	0.03	
Cl_3 (CB 25)	III	0.05	*0.020-0.13*	0.04	*0.017-0.11*	0.09	*0.040-0.19*	-	-
Cl_3 (CB 31)	III	0.72	*0.27-1.89*	0.54	*0.19-1.49*	1.38	*0.458-4.14*	0.34	*0.087-1.29*
Cl_3 (CB 28)	III	0.81	*0.34-1.93*	0.84	*0.28-2.51*	1.67	*0.54-5.15*	0.41	*0.10-1.70*
Cl_3 (CB 21)	III	-	-	-	-	-	-	0.12	*0.051-0.27*
Cl_3 (CB 33/20)	III	0.33	*0.13-0.88*	0.39	*0.13-1.18*	0.42	*0.15-1.18*	0.38	*0.14-1.10*
Cl_3 (CB 19)	IV	0.13	*0.051-0.32*	0.11	*0.047-0.25*	-	-	0.02	-
Cl_3 (CB30)	IV	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	IV	0.70	*0.24-2.08*	0.76	*0.26-2.27*	0.39	*0.11-1.37*	0.23	*0.095-0.58*
Cl_3 (CB 17)	IV	0.29	*0.10-0.85*	0.33	*0.13-0.87*	0.11	*0.032-0.39*	0.09	*0.032-0.25*
Cl_3 (CB 27/24)	IV	0.12	*0.050-0.28*	0.10	*0.041-0.23*	0.09	*0.013-0.58*	0.22	*0.025-1.90*

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_3 (CB 16/32)	IV	0.71	*0.25-1.99*	0.70	*0.26-1.93*	0.28	*0.10-0.81*	0.11	*0.011-1.24*
Cl_3 (CB 22)	III	0.23	*0.092-0.56*	0.20	*0.068-0.61*	0.33	*0.096-1.16*	0.10	*0.026-0.369*
Cl_4 (CB 54)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	V	-	-	-	-	-	-	0.01	
Cl_4 (CB 53)	IV	0.08	*0.034-0.20*	0.07	*0.032-0.16*	0.11	*0.038-0.31*	0.06	*0.023-0.16*
Cl_4 (CB 51)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 45)	V	0.10	*0.040-0.26*	0.08	*0.036-0.19*	0.08	*0.028-0.22*	0.03	*0.009-0.14*
Cl_4 (CB 46)	V	-	-	-	-	3.29	-	0.35	*0.017-6.91*
Cl_4 (CB 73/52)	IV	1.23	*0.43-3.52*	1.05	*0.21-5.35*	2.06	*0.30-14.2*	0.18	*0.063-0.51*
Cl_4 (CB 69)	IV	-	-	-	-	0.01	-	-	-
Cl_4 (CB 49)	IV	0.56	*0.21-1.48*	0.24	*0.073-0.78*	1.47	*0.34-6.311*	0.09	*0.036-0.23*
Cl_4 (CB 43)	IV	-	-	-	-	-	-		
Cl_4 (CB 47/75/48)	IV	0.34	*0.13-0.91*	0.28	*0.089-0.92*	0.63	*0.14-2.835*	0.03	*0.014-0.089*
Cl_4 (CB 65)	IV	-	-	-	-	-	-	0.08	*0.027-0.22*
Cl_4 (CB 62)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	IV	0.42	*0.16-1.09*	0.26	*0.091-0.75*	1.45	*0.38 -5.53*	0.12	*0.048-0.32*
Cl_4 (CB 59/42)	IV	0.19	*0.067-0.51*	0.10	*0.041-0.25*	0.17	*0.020-1.50*	0.05	*0.019-0.12*
Cl_4 (CB 72)	III	-	-	-	-	-	-	0.04	*0.017-0.085*
Cl_4 (CB 71/41/64)	IV	0.52	*0.113-2.346*	0.26	*0.066-1.03*	0.06	*0.018-0.22*	0.10	*0.048-0.22*

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 68)	III	0.06	-	0.15	-	0.21	*0.036-1.23*	-	-
Cl_4 (CB 40)	IV	0.06	*0.023-0.18*	-	-	0.10	*0.031-0.30*	0.01	-
Cl_4 (CB 57)	III	-	-	-	-	0.01	-	-	-
Cl_4 (CB 67)	III	-	-	-	-	0.05	*0.014-0.16*	-	-
Cl_4 (CB 58)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	III	0.16	*0.048-0.54*	0.09	*0.042-0.19*	0.15	*0.039-0.54*	0.10	*0.042-0.26*
Cl_4 (CB 61/74)	III	0.86	*0.28-2.62*	0.68	*0.202-2.26*	2.09	*0.48-9.18*	0.18	*0.049-0.66*
Cl_4 (CB 70/76)	III	0.57	*0.180-1.837*	0.35	*0.121.03*	2.48	*0.66-9.31*	0.29	*0.083-1.05*
Cl_4 (CB 66)	III	0.82	*0.26-2.56*	0.61	*0.18-2.06*	1.86	*0.50-6.91*	0.22	*0.067-0.72*
Cl_4 (CB 55)	III	-	-			0.02	*0.004-0.091*	0.01	
Cl_4 (CB 60/56)	III	0.29	*0.095-0.90*	0.24	*0.082-0.71*	0.43	*0.19-0.97*	0.10	*0.030-0.31*
Cl_5 (CB 104)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 103)	IV	0.09	*0.039-0.20*	0.10		0.06	*0.014-0.25*	0.00	
Cl_5 (CB 100)	II	0.09	*0.039-0.19*			0.05	*0.009-0.26*	0.00	
Cl_5 (CB 94)	V	0.25	*0.089-0.73*	0.14	*0.063-0.33*	0.05	*0.020-0.128*	0.08	*0.033-0.20*
Cl_5 (CB 95)	V	0.80	*0.27-2.33*	0.31	*0.080-1.20*	4.05	*0.93-17.6*	0.37	*0.082-1.63*
Cl_5 (CB 102/93)	V	-	-	-	-	0.05	*0.013-0.16*	0.02	-
Cl_5 (CB 98)	IV	-	-	-	-	-	-	0.00	-

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 88)	V	0.12	*0.060-0.25*	0.12	-	0.07	*0.015-0.33*	0.01	-
Cl_5 (CB 91)	V	0.17	*0.061-0.49*	0.25	-	0.38	*0.081-1.77*	0.04	*0.005-0.24*
Cl_5 (CB 121)	I	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	V	0.57	*0.181-1.80*	0.22	*0.037-1.28*	-	-	-	-
Cl_5 CB (101/90)	IV	2.47	*0.74-8.26*	0.52	*0.098-2.77*	8.05	*1.96-33.11*	0.66	*0.13-3.26*
Cl_5 (CB 89)	V	0.14	*0.052-0.38*	-	-	0.50	*0.15-1.70*	0.04	-
Cl_5 (CB 99)	II	3.44	*1.06-11.22*	2.99	*0.71-12.57*	7.70	*1.80-33.46*	0.46	*0.079-2.66*
Cl_5 (CB 113)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 119)	II	0.22	*0.072-0.67*	0.16	*0.045-0.59*	0.32	*0.081-1.26*	0.01	
Cl_5 (CB 112)	IV			-	-	-	-	-	-
Cl_5 (CB 109/83)	IV			-	-	0.16	*0.046-0.55*	0.00	
Cl_5 (CB 97/86)	IV	0.21	*0.081-0.53*	0.18	*0.042-0.76*	0.80	*0.22-2.84*	0.14	*0.030-0.67*
Cl_5 (CB 116/125/117)	IV	0.13	*0.044-0.37*	-	-	-	-	-	-
Cl_5 (CB 115/87)	II	0.78	*0.23-2.62*	0.30	*0.070-1.24*	1.99	*0.53-7.46*	0.18	*0.030-1.08*
Cl_5 (CB 111)	III	-	-	-	-	-	-	0.02	
Cl_5 (CB 85)	II	0.83	*0.25-2.71*	0.62	*0.17-2.27*	1.35	*0.35-5.18*	0.18	*0.037-0.90*
Cl_5 (CB 120)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 110)	IV	1.14	*0.34-3.81*	0.25	*0.045-1.40*	1.92	*0.50-7.36*	0.30	*0.060-1.54*

		COD (*B. saida*) **(muscle)** (*n* = 12)		SCULPIN (*M. scorpioides*) **(muscle)** (*n* = 12)		SALMON (*Salmo sp.*) **(muscle)** (*n* = 7)		BIVALVES (*M. edulis*) **(muscle)** (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 82)	IV	0.09	*0.037-0.23*	-	-	0.22	*0.070-0.695*	0.01	-
Cl_5 (CB 124)	III	0.13	*0.053-0.30*	-	-	0.11	*0.032-0.42*	-	-
Cl_5 (CB 108/107)	III	0.26	*0.080-0.830*	0.15		0.53	*0.14-2.05*	0.02	*0.008-0.043*
Cl_5 (CB 123)	III	0.86	*0.073-9.99*	2.57	*0.49-13.55*	0.84	*0.048-14.6*	0.28	*0.015-5.31*
Cl_5 (CB 106/118)	III	2.93	*0.96-8.95*	2.78	*0.78-9.85*	8.21	*1.42-47.62*	0.66	*0.13-3.42*
Cl_5 (CB 114)	III	0.16	*0.064-0.43*	-	-	0.15	*0.035-0.66*	0.01	-
Cl_5 (CB 122)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 105)	III	1.07	*0.32-3.58*	0.75	*0.21-2.67*	2.21	*0.52-9.35*	0.23	*0.054-1.02*
Cl_6 (CB 155)	I	0.23	*0.070-0.77*	0.24	*0.089-0.62*	-	-	-	-
Cl_6 (CB 150)	V	-	-	-	-	0.03	*0.007-0.16*		-
Cl_6 (CB 152)	V	-	-	-	-	-	-	-	-
Cl_6 (CB 145)	V	-	-	-	-	-	-	-	-
Cl_6 (CB 148)	I	0.17	*0.076-0.36*	0.16		0.38		0.04	
Cl_6 (CB 136)	V	0.13	*0.053-0.305*	0.11	*0.038-0.30*	0.60	*0.14-2.51*	0.10	*0.023-0.47*
Cl_6 (CB 154)	I	0.25	*0.070-0.88*	0.14	*0.060-0.33*	0.29	*0.071-1.21*	0.05	*0.008-0.29*
Cl_6 (CB 151)	V	0.97	*0.33-2.86*	0.39	*0.10-1.52*	2.77	*0.67-11.4*	0.27	*0.055-1.31*
Cl_6 (CB 135/144)	V	0.31	*0.11-0.86*	0.19	*0.046-0.77*	1.10	*0.29-4.12*	0.20	*0.043-0.91*
Cl_6 (CB 147)	II	0.08	*0.031-0.21*	-	-	0.18	*0.043-0.76*	0.01	*0.004-0.053*

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 149)	V	1.35	*0.47-3.87*	0.39	*0.094-1.63*	3.93	*1.02-15.1*	0.66	*0.13-3.31*
Cl_6 (CB 139/140)	II	0.08	*0.035-0.18*	-	-	0.03	*0.006-0.18*	-	-
Cl_6 CB-143/134	V	0.17	*0.056-0.50*	0.14	-	0.21	*0.043-0.99*	0.03	*0.007-0.159*
Cl_6 (CB 142/131)	V	0.26	*0.085-0.77*	0.14	*0.063-0.32*	0.06	-	-	-
Cl_6 (CB 133)	I	0.12	*0.051-0.29*	0.06	-	0.02	-	-	-
Cl_6 (CB 146/161)	I	1.22	*0.38-3.98*	0.30	*0.056-1.61*	2.46	*0.54-11.17*	0.32	*0.064-1.57*
Cl_6 (CB 165)	I	0.26	*0.030-2.23*	0.08	*0.039-0.18*	0.03			
Cl_6 (CB 132/153)	I	10.85	*3.14-37.5*	11.48	*2.97-44.4*	18.33	*4.33-77.6*	2.30	*0.45-11.6*
Cl_6 (CB 168)	I	0.27	*0.096-0.77*	0.14	*0.037-0.550*	0.52	*0.16-1.72*	0.18	*0.071-0.48*
Cl_6 (CB 141)	IV	0.48	*0.16-1.45*	0.17	*0.037-0.74*	0.89	*0.21-3.79*	0.08	
Cl_6 (CB 137)	II	0.27	*0.072-1.00*	0.33	*0.082-1.31*	0.91	*0.22-3.73*	0.26	*0.10-0.66*
Cl_6 (CB 130)	II	0.24	*0.070-0.86*	0.21	*0.058-0.77*	0.54	*0.12-2.48*	0.05	
Cl_6 (CB 160/163/164/138)	II	5.81	*1.34-25.27*	5.47	*1.39-21.53*	15.44	*3.59-66.44*	1.55	*0.29-8.15*
Cl_6 (CB 158)	II	0.40	*0.11-1.44*	0.36	*0.087-1.49*	0.71	*0.16-3.18*	0.08	*0.014-0.44*
Cl_6 (CB 129)	IV	0.06	*0.009-0.44*	-	-	-	-	-	-
Cl_6 (CB 166)	II	0.05	*0.015-0.18*	-	-	0.09	*0.032-0.26*	-	-
Cl_6 (CB 159)	III	0.07	*0.027-0.19*	-	-	0.15	*0.035-0.65*	0.01	

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 162)	IV	0.51	*0.11-2.39*	0.42	*0.11-1.64*	0.10	*0.024-0.43*	0.05	*0.006-0.38*
Cl_6 (CB 128)	II	0.71	*0.22-2.36*	0.67	*0.18-2.46*	1.30	*0.30-5.67*	0.16	*0.029-0.895*
Cl_6 (CB 167)	III	0.20	*0.055-0.69*	0.21	*0.10-0.44*	0.39	*0.084-1.78*	0.08	*0.015-0.38*
Cl_6 (CB 156)	III	0.31	*0.091-1.04*	0.22	*0.059-0.83*	0.54	*0.12-2.50*	0.08	*0.016-0.38*
Cl_6 (CB 157)	III	0.23	*0.083-0.63*	0.17	*0.063-0.44*	0.22	*0.051-0.92*	0.09	*0.033-0.24*
Cl7 (CB 188)	I	0.18	*0.054-0.61*	0.08	*0.022-0.29*	0.14	*0.027-0.67*	0.06	*0.024-0.17*
Cl_7 (CB 184)	I	0.16	*0.046-0.55*	0.15	*0.045-0.50*	0.11	*0.027-0.435*	0.06	-
Cl_7 (CB 179)	V	0.14	*0.048-0.42*	0.11	*0.035-0.35*	0.54	*0.14-2.09*	0.11	*0.027-0.48*
Cl_7 (CB 176)	V	0.08	-	-	-	0.14	*0.038-0.51*	0.05	*0.007-0.29*
Cl_7 (CB 186)	V	-	-	-	-	-	-	-	-
Cl_7 (CB 178)	I	0.44	*0.13-1.46*	0.40	*0.14-1.19*	0.80	*0.18-3.50*	0.11	*0.025-0.52*
Cl_7 (CB 175)	I	0.12	*0.040-0.34*			0.15	*0.034-0.64*	0.06	-
Cl_7 (CB 187/182)	I	1.46	*0.382-5.61*	0.42	*0.12-1.52*	4.45	*1.01-19.55*	0.57	*0.12-2.78*
Cl_7 (CB 183)	I	0.88	*0.27-2.89*	0.68	*0.19-2.40*	1.37	*0.23-6.29*	0.22	*0.044-1.08*
Cl_7 (CB 185)	V	0.09	*0.040-0.22*	-	-	0.12	*0.031-0.48*	-	-
Cl_7 (CB 174/181)	V	0.31	*0.11-0.93*	0.15	*0.051-0.47*	0.77	*0.201-2.96*	0.07	*0.010-0.49*
Cl_7 (CB 177)	II	0.12	*0.044-0.33*	0.11	*0.037-0.32*	0.73	*0.18-3.05*	0.15	*0.036-0.63*
Cl_7 (CB 171)	II	0.23	*0.073-0.75*	0.18	*0.057-0.57*	0.30	*0.067-1.33*	0.10	*0.022-0.44*

		COD (*B. saida*) **(muscle)** (*n* = 12)		SCULPIN (*M. scorpioides*) **(muscle)** (*n* = 12)		SALMON (*Salmo sp.*) **(muscle)** (*n* = 7)		BIVALVES (*M. edulis*) **(muscle)** (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_7 (CB 173)	II	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	I	0.22	*0.070-0.67*	0.11	*0.050-0.25*	0.34	*0.075-1.52*	-	-
Cl_7 (CB 180)	I	2.28	*0.66-7.90*	2.41	*0.64-9.11*	3.76	*0.79-17.79*	0.23	*0.047-1.15*
Cl_7 (CB 193)	I	0.16	*0.047-0.55*	0.09	*0.028-0.31*	0.26	*0.057-1.17*	-	-
Cl_7 (CB 191)	I	0.10	*0.044-0.22*	-	-	0.08	*0.017-0.33*	-	-
Cl_7 (CB 170/190)	II	0.94	*0.27-3.26*	0.90	*0.22-3.60*	1.32	*0.28-6.25*	0.06	*0.009-0.38*
Cl_7 (CB 189)	III	0.04	-	-	-	0.08	*0.021-0.34*	0.003	-
Cl_8 (CB 202)	I	0.33	*0.11-1.06*	0.43	*0.14-1.33*	0.42	*0.11-1.68*	0.13	*0.031-0.57*
Cl_8 (CB 200)	I	0.28	*0.084-0.91*	0.13	*0.038-0.45*	0.44	*0.11-1.77*	0.14	*0.035-0.56*
Cl_8 (CB 204)	I	-	-	-	-	0.04	-	-	-
Cl_8 (CB 197)	I	0.14	*0.040-0.459*	0.14	*0.040-0.51*	0.13	*0.031-0.53*	0.05	*0.014-0.18*
Cl_8 (CB 199)	V	-	-	-	-	0.04	*0.016-0.11*	0.002	-
Cl_8 (CB 198)	I	0.38	*0.13-1.10*	0.24	*0.079-0.74*	0.62	*0.15-2.62*	0.05	-
Cl_8 (CB 201)	I	0.40	*0.13-1.26*	0.19	*0.053-0.71*	-	-	0.004	-
Cl_8 (CB 203/196)	I	0.54	*0.17-1.77*	0.49	*0.15-1.58*	0.72	*0.16-3.21*	0.05	*0.008-0.324*
Cl_8 (CB 195)	II	0.10	*0.046-0.23*	-	-	0.08	*0.018-0.36*	0.001	-
Cl_8 (CB 194)	I	0.31	*0.10-0.98*	0.31	*0.10-0.93*	0.38	*0.077-1.86*	0.02	*0.005-0.133*
Cl_8 (CB 205)	I	0.04	*0.016-0.12*	-	-	0.04	*0.015-0.12*	0.0001	-
Cl_9 (CB 208)	I	0.21	*0.013-3.28*	0.09	*0.038-0.22*	0.14	*0.044-0.43*	-	-

		COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*		1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_9 (CB 207)	I	0.15	*0.046-0.49*	0.20	*0.054-0.75*	0.14	*0.041-0.45*	-	-
Cl_9 (CB 206)	I	0.23	*0.073-0.74*	0.44	*0.21-0.94*	0.28	*0.092-0.84*	-	-
Cl_{10} (CB 209)	I	0.27	*0.052-1.42*	0.47	*0.070-3.19*	0.06	*0.016-0.24*	0.03	*0.001-1.22*
$\sum Cl_2$		1.35	*0.39-4.62*	1.60	*0.50-5.13*	0.28	*0.113-0.67*	0.62	*0.18-2.10*
$\sum Cl_3$		3.67	*1.44-9.39*	3.61	*1.32-9.85*	4.82	*1.65-14.1*	1.73	*0.53-5.68*
$\sum Cl_4$		4.38	*1.56-12.2*	3.05	*0.80-11.6*	11.4	*2.69-48.2*	1.73	*0.55-5.47*
$\sum Cl_5$		14.2	*4.69-42.9*	8.37	*2.23-31.4*	40.2	*10.2-158*	3.11	*0.61-15.8*
$\sum Cl_6$		23.8	*7.16-79.3*	19.9	*5.19-77.0*	52.1	*12.57-215*	5.73	*1.11-29.5*
$\sum Cl_7$		7.27	*2.19-24.0*	5.53	*1.59-19.2*	15.5	*3.49-68.7*	1.46	*0.29-7.22*
$\sum Cl_8$		1.72	*0.49-6.06*	1.26	*0.32-4.97*	2.84	*0.654-12.4*	0.26	*0.050-1.40*
$\sum Cl_9$		0.54	*0.063-4.58*	0.27	*0.044-1.72*	0.55	*0.18-1.72*	-	-
$\sum Cl_{10}$		0.27	*0.052-1.42*	0.47	*0.070-3.19*	0.06	*0.016-0.24*	0.03	*0.001-1.22*
∑Diortho PCBs		52.2	*15.7-173*	38.3	*10.6-138*	106	*26.46-424*	11.4	*2.44-53.1*
∑Mono ortho PCBs		7.81	*2.57-23.8*	6.75	*2.09-21.7*	23.0	*6.22-85.0*	3.49	*0.89-13.7*
∑PCBs		60.7	*19.0-194*	45.8	*13.3-159*	129	*32.8-508*	14.9	*3.33-67.3*

Appendix 5 continued.

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_2 (PCB 7/9)	III	0.06	*0.023-0.14*	0.05	*0.020-0.15*	-	-	0.09	-
Cl_2 (PCB 6)	III	0.08	*0.037-0.18*	0.09	*0.033-0.23*	-	-	0.13	-
Cl_2 (PCB 8/5)	III	0.31	*0.099-0.95*	0.33	*0.10-1.09*	0.34	*0.149-0.79*	0.30	*0.078-1.13*
Cl_2 (PCB 4/10)	IV	0.10	*0.035-0.31*	0.10	*0.028-0.30*	0.09	*0.036-0.24*	0.08	*0.021-0.31*
Cl_3 (PCB 23/34)	III	0.02	*0.005-0.074*	0.01	*0.003-0.035*	0.01	*0.004-0.027*	0.02	*0.004-0.075*
Cl_3 (PCB 29)	III	0.01	*0.004-0.027*	0.01	*0.003-0.033*	0.01	-	0.02	*0.007-0.061*
Cl_3 (PCB 26)	III	0.30	*0.099-0.93*	0.24	*0.073-0.78*	0.26	*0.11-0.595*	0.32	*0.080-1.25*
Cl_3 (CB 25)	III	0.04	*0.014-0.12*	0.04	*0.012-0.12*	0.03	*0.013-0.092*	0.03	*0.008-0.11*
Cl_3 (CB 31)	III	5.06	*1.42-18.0*	2.63	*0.59-11.6*	2.37	*0.88-6.39*	4.38	*0.82-23.3*
Cl_3 (CB 28)	III	2.96	*0.94-9.29*	2.45	*0.86-6.93*	2.66	*1.12-6.31*	4.32	*1.46-12.7*
Cl_3 (CB 21)	III	0.0023	-	0.84	*0.22-3.26*	-	-	0.98	-
Cl_3 (CB 33/20)	III	0.71	*0.22-2.27*	0.49	*0.13-1.84*	0.57	*0.208-1.536*	0.94	*0.136-6.51*
Cl_3 (CB 19)	IV	0.25	*0.10-0.64*	0.05	*0.003-0.99*	-	-	0.14	-
Cl_3 (CB30)	IV	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	IV	4.39	*1.47-13.1*	1.52	*0.37-6.29*	1.35	*0.55-3.34*	2.99	*0.783-11.40*
Cl_3 (CB 17)	IV	1.32	*0.42-4.14*	0.56	*0.14-2.19*	0.48	*0.19-1.18*	0.98	*0.267-3.63*
Cl_3 (CB 27/24)	IV	0.19	*0.051-0.72*	0.11	*0.034-0.37*	0.11	*0.048-0.26*	0.16	*0.043-0.55*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_3 (CB 16/32)	IV	1.58	*0.49-5.02*	0.88	*0.24-3.24*	0.85	*0.36-2.02*	1.49	*0.39-5.64*
Cl_3 (CB 22)	III	0.06	*0.007-0.44*	0.20	*0.036-1.12*	0.16	*0.045-0.54*	0.15	*0.010-2.12*
Cl_4 (CB 54)	V	0.06	*0.026-0.13*	0.01	*0.004-0.046*	-	-	0.01	-
Cl_4 (CB 50)	V	0.06	*0.020-0.15*	0.02	*0.004-0.12*	0.01	*0.003-0.018*	0.07	-
Cl_4 (CB 53)	IV	2.72	*0.95-7.76*	0.26	*0.029-2.28*	0.43	*0.15-1.28*	0.94	*0.132-6.76*
Cl_4 (CB 51)	V	0.57	*0.212-1.54*	0.11	*0.025-0.51*	0.06	*0.020-0.20*	0.59	-
Cl_4 (CB 45)	V	2.68	*0.94-7.66*	0.54	*0.11-2.61*	0.40	*0.14-1.16*	1.22	*0.25-5.97*
Cl_4 (CB 46)	V	0.42	*0.15-1.18*	0.13	*0.031-0.53*	0.11	*0.040-0.28*	0.28	*0.064-1.22*
Cl_4 (CB 73/52)	IV	148	*49.7-437*	19.2	*3.20-115*	10.6	*3.23-35.0*	37.8	*5.65-252*
Cl_4 (CB 69)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	IV	38.9	*12.4-121*	6.39	*1.08-37.6*	3.59	*1.14-11.3*	10.9	*2.09-57.3*
Cl_4 (CB 43)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	IV	19.4	*6.62-56.9*	2.98	*0.53-16.9*	1.59	*0.49-5.17*	5.40	*0.88-33.3*
Cl_4 (CB 65)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	IV	31.2	*10.4-92.6*	6.40	*1.26-32.3*	4.04	*1.34-12.1*	13.1	*2.76-62.4*
Cl_4 (CB 59/42)	IV	5.72	*1.63-20.0*	1.71	*0.34-8.66*	0.99	*0.33-2.95*	2.78	*0.66-11.6*
Cl_4 (CB 72)	III	-	-	0.98	*0.33-2.88*	-	-	0.60	-

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 71/41/64)	IV	0.64	*0.076-5.44*	0.37	*0.026-5.34*	0.19	*0.032-1.10*	0.58	*0.060-5.58*
Cl_4 (CB 68)	III	1.02	*0.025-41.1*	1.16	*0.41-3.25*	0.09	-	2.14	*0.806-5.68*
Cl_4 (CB 40)	IV	1.33	*0.45-3.94*	0.40	*0.093-1.72*	0.29	*0.107-0.79*	0.80	*0.189-3.38*
Cl_4 (CB 57)	III	0.02	*0.008-0.072*	0.01	*0.004-0.052*	0.02	*0.006-0.060*	0.02	*0.006-0.097*
Cl_4 (CB 67)	III	0.06	*0.021-0.17*	0.03	*0.009-0.13*	0.02	*0.006-0.079*	0.03	*0.007-0.13*
Cl_4 (CB 58)	III	0.01	*0.004-0.018*	0.01	*0.004-0.044*	-	-	0.06	-
Cl_4 (CB 63)	III	0.22	*0.055-0.86*	0.17	*0.042-0.71*	0.16	*0.049-0.49*	0.21	*0.027-1.69*
Cl_4 (CB 61/74)	III	36.7	*12.14-111*	6.97	*1.48-32.9*	4.31	*1.31-14.1*	12.9	*2.31-72.0*
Cl_4 (CB 70/76)	III	5.83	*1.79-18.8*	3.36	*0.90-12.5*	3.46	*1.31-9.08*	5.90	*1.28-27.2*
Cl_4 (CB 66)	III	20.8	*6.74-63.8*	6.07	*1.52-24.3*	4.07	*1.35-12.2*	10.8	*2.42-48.6*
Cl_4 (CB 55)	III	0.84	*0.27-2.61*	0.15	*0.023-0.90*	0.07	*0.022-0.251*	0.22	*0.029-1.66*
Cl_4 (CB 60/56)	III	1.63	*0.477-5.56*	1.13	*0.28-4.54*	1.00	*0.37-2.68*	1.48	*0.31-7.21*
Cl_5 (CB 104)	IV	0.08	*0.020-0.29*	0.02	*0.005-0.082*	0.01	*0.004-0.049*	0.03	*0.005-0.17*
Cl_5 (CB 96)	IV	0.61	*0.19-1.85*	0.10	*0.018-0.51*	0.06	*0.017-0.18*	0.18	*0.028-1.12*
Cl_5 (CB 103)	IV	1.87	*0.59-5.86*	0.28	*0.052-1.49*	0.13	*0.037-0.46*	0.41	*0.059-2.84*
Cl_5 (CB 100)	II	1.07	*0.33-3.44*	0.18	*0.038-0.80*	0.08	*0.024-0.29*	0.25	*0.038-1.63*
Cl_5 (CB 94)	V	0.50	*0.17-1.48*	0.16	*0.042-0.59*	0.15	*0.053-0.43*	0.23	*0.052-0.97*
Cl_5 (CB 95)	V	160	*51.9-491*	20.4	*3.65-115*	10.9	*3.15-37.7*	36.8	*5.39-250*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 102/93)	V	2.54	*0.81-7.98*	0.28	*0.055-1.39*	0.20	*0.055-0.69*	0.73	*0.11-4.97*
Cl_5 (CB 98)	IV	-	-	0.23	-	-	-	0.23	-
Cl_5 (CB 88)	V	1.38	*0.41-4.61*	0.25	*0.042-1.41*	0.14	*0.042-0.47*	0.46	*0.074-2.87*
Cl_5 (CB 91)	V	19.0	*6.18-58.1*	2.45	*0.43-14.1*	1.22	*0.35-4.29*	3.87	*0.54-27.4*
Cl_5 (CB 121)	I	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	V	80.4	*33.2-194*	6.04	*2.44-14.9*	-	-	-	-
Cl_5 CB (101/90)	IV	245	*83.33-721*	30.4	*5.06-182*	16.5	*4.80-56.9*	44.6	*6.74-295*
Cl_5 (CB 89)	V	15.4	*5.45-43.42*	2.25	*0.37-13.6*	1.46	*0.45-4.68*	4.11	*0.700-24.1*
Cl_5 (CB 99)	II	191	*64.21-571*	24.9	*4.31-144*	12.5	*3.66-42.6*	32.7	*4.48-238*
Cl_5 (CB 113)	IV	0.15	*0.050-0.43*	0.04	*0.005-0.28*	0.02	*0.005-0.10*	0.06	*0.007-0.50*
Cl_5 (CB 119)	II	9.70	*3.23-29.1*	1.34	*0.21-8.36*	0.70	*0.20-2.42*	2.15	*0.292-15.8*
Cl_5 (CB 112)	IV	0.17	*0.043-0.659*	0.04	*0.009-0.23*	0.04	*0.012-0.12*	0.07	*0.012-0.40*
Cl_5 (CB 109/83)	IV	4.83	*1.71-13.6*	0.63	*0.10-3.71*	0.39	*0.11-1.34*	1.05	*0.15-6.98*
Cl_5 (CB 97/86)	IV	24.3	*6.98-84.3*	4.78	*0.86-26.4*	2.75	*0.86-8.76*	6.88	*1.34-35.3*
Cl_5 (CB 116/125/117)	IV	0.23	*0.075-0.71*	0.05	*0.007-0.37*	0.03	*0.009-0.091*	0.11	*0.031-0.377*
Cl_5 (CB 115/87)	II	59.7	*19.1-186*	9.58	*1.64-54.45*	4.93	*1.47-16.4*	13.1	*2.27-75.4*
Cl_5 (CB 111)	III	-	-	-	-	-	-	-	-

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 85)	II	37.7	*12.1-116*	6.04	*1.10-32.9*	3.00	*0.89-10.0*	7.79	*1.29-46.9*
Cl_5 (CB 120)	III	0.33	*0.099-1.08*	0.04	*0.005-0.31*	0.02	*0.005-0.10*	0.07	*0.009-0.58*
Cl_5 (CB 110)	IV	48.0	*12.7-181*	10.3	*1.82-58.3*	6.71	*2.24-19.6*	16.2	*3.42-76.5*
Cl_5 (CB 82)	IV	5.64	*1.70-18.7*	1.16	*0.21-6.39*	0.70	*0.21-2.25*	1.75	*0.34-8.77*
Cl_5 (CB 124)	III	1.99	*0.64-6.16*	0.25	*0.032-2.01*	0.11	*0.028-0.44*	0.30	*0.046-1.95*
Cl_5 (CB 108/107)	III	2.77	*0.77-9.89*	1.42	*0.31-6.29*	0.88	*0.30-2.53*	1.52	*0.27-8.44*
Cl_5 (CB 123)	III	8.92	*1.88-42.3*	1.26	*0.21-7.48*	0.64	*0.18-2.24*	1.39	*0.19-9.91*
Cl_5 (CB 106/118)	III	171	*55.70-522*	24.2	*4.06-144*	11.9	*3.42-41.1*	34.5	*5.25-226*
Cl_5 (CB 114)	III	0.39	*0.11-1.29*	0.14	*0.012-1.73*	0.06	*0.015-0.26*	0.14	*0.020-0.99*
Cl_5 (CB 122)	III	4.18	*1.38-12.63*	0.53	*0.093-3.06*	0.34	*0.096-1.18*	0.95	*0.13-6.82*
Cl_5 (CB 105)	III	42.9	*14.08-130*	7.79	*1.31-46.1*	3.50	*1.01-12.1*	9.97	*1.743-57.0*
Cl_6 (CB 155)	I	3.21	*1.44-7.17*	-	-	-	-	-	-
Cl_6 (CB 150)	V	0.83	*0.282-2.470*	0.15	*0.029-0.730*	0.07	*0.019-0.258*	0.16	*0.023-1.15*
Cl_6 (CB 152)	V	0.34	*0.116-1.010*	0.06	*0.011-0.355*	0.03	*0.007-0.102*	0.07	*0.009-0.49*
Cl_6 (CB 145)	V	0.12	*0.041-0.364*	0.03	*0.005-0.137*	0.01	*0.002-0.045*	0.02	*0.003-0.17*
Cl_6 (CB 148)	I	8.62	*1.09-68.00*	-	-	-	-	-	-
Cl_6 (CB 136)	V	31.5	*11.1-89.12*	4.10	*0.68-24.62*	2.14	*0.576-7.99*	5.11	*0.73-35.5*
Cl_6 (CB 154)	I	8.80	*2.97-26.04*	1.46	*0.29-7.24*	0.67	*0.18-2.41*	1.31	*0.17-9.56*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 151)	V	106	*37.9-300*	15.2	*2.59-89.0*	6.79	*1.83-25.0*	15.3	*2.09-111*
Cl_6 (CB 135/144)	V	44.1	*16.1-120*	5.95	*0.98-36.07*	2.85	*0.75-10.78*	6.83	*0.94-49.40*
Cl_6 (CB 147)	II	6.52	*2.31-18.38*	1.00	*0.17-5.67*	0.42	*0.11-1.542*	0.94	*0.12-7.04*
Cl_6 (CB 149)	V	217	-	29.0	*4.82-174*	14.4	*3.92-52.8*	32.2	*4.40-235*
Cl_6 (CB 139/140)	II	0.58	*0.15-2.11*	0.19	*0.039-0.88*	0.12	*0.034-0.42*	0.11	*0.015-0.86*
Cl_6 CB-143/134	V	7.66	*2.61-22.3*	1.16	*0.19-6.84*	0.55	*0.15-1.97*	1.26	*0.18-8.52*
Cl_6 (CB 142/131)	V	4.40	*0.69-27.7*	0.48	*0.049-4.73*	0.68	*0.19-2.40*	0.67	*0.15-2.89*
Cl_6 (CB 133)	I	0.92	*0.29-2.83*	0.34	*0.033-3.51*	0.10	*0.031-0.30*	0.22	*0.033-1.46*
Cl_6 (CB 146/161)	I	79.1	*27.4-227*	13.3	*2.41-73.5*	5.58	*1.52-20.3*	11.4	*1.58-82.6*
Cl_6 (CB 165)	I	0.56	*0.11-2.66*	0.36	*0.13-0.96*	0.04		0.21	*0.094-0.47*
Cl_6 (CB 132/153)	I	518	*172.8-1,550*	82.9	*14.6-468*	32.3	*9.18-113*	75.4	*10.3-548*
Cl_6 (CB 168)	I	35.3	*12.0-103*	4.12	*0.655-25.8*	1.96	*0.51-7.49*	5.03	*0.70-35.9*
Cl_6 (CB 141)	IV	14.1	*3.48-56.8*	3.64	*0.69-19.1*	1.65	*0.51-5.21*	3.50	*0.59-20.7*
Cl_6 (CB 137)	II	25.5	*8.86-73.2*	3.59	*0.70-18.2*	1.63	*0.43-6.09*	4.03	*0.46-34.6*
Cl_6 (CB 130)	II	16.4	*4.84-55.7*	2.91	*0.46-17.9*	0.87	*0.23-3.21*	2.59	*0.34-19.7*
Cl_6 (CB 160/163/164/138)	II	384	*125-1,170*	62.6	*11.4-343*	23.8	*6.65-85.0*	61.8	*8.85-431*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 158)	II	19.7	*6.37-60.6*	3.20	*0.55-18.4*	1.06	*0.27-4.14*	3.00	*0.41-21.62*
Cl_6 (CB 129)	IV	2.29	*0.70-7.47*	0.61	*0.11-3.35*	0.19	*0.071-0.51*	0.41	*0.079-2.10*
Cl_6 (CB 166)	II	1.18	*0.37-3.69*	0.25	*0.047-1.30*	0.09	*0.027-0.32*	0.22	*0.029-1.65*
Cl_6 (CB 159)	III	4.05	*0.81-20.0*	1.25	*0.25-6.13*	0.39	*0.10-1.49*	0.67	*0.082-5.50*
Cl_6 (CB 162)	IV	1.48	*0.32-6.75*	0.57	*0.12-2.69*	0.18	*0.052-0.63*	0.46	*0.052-4.00*
Cl_6 (CB 128)	II	38.6	*11.9-124*	6.75	*1.24-36.6*	2.38	*0.66-8.50*	6.81	*0.90-51.0*
Cl_6 (CB 167)	III	6.51	*1.58-26.7*	1.24	*0.22-6.82*	0.60	*0.12-2.80*	1.21	*0.18-8.03*
Cl_6 (CB 156)	III	10.1	*2.91-34.7*	2.48	*0.48-12.7*	0.78	*0.21-2.80*	1.71	*0.28-10.32*
Cl_6 (CB 157)	III	3.85	*1.16-12.7*	0.91	*0.15-5.26*	0.33	*0.086-1.25*	0.71	*0.12-4.20*
Cl7 (CB 188)	I	2.45	*0.47-12.6*	0.76	*0.20-2.90*	0.28	*0.075-1.05*	0.47	*0.070-3.16*
Cl_7 (CB 184)	I	1.49	*0.29-7.64*	0.50	*0.12-1.91*	0.17	*0.049-0.616*	0.30	*0.044-1.99*
Cl_7 (CB 179)	V	21.3	*5.17-87.3*	4.11	*0.77-21.8*	1.64	*0.389-6.93*	2.96	*0.41-21.2*
Cl_7 (CB 176)	V	6.15	*1.47-25.6*	1.14	*0.20-6.18*	0.36	*0.094-1.42*	0.78	*0.110-5.47*
Cl_7 (CB 186)	V	0.04	*0.013-0.10*	0.01	*0.004-0.042*	0.01		0.01	*0.002-0.059*
Cl_7 (CB 178)	I	21.6	*5.29-88.3*	4.89	*1.00-23.6*	1.44	*0.38-5.4*	2.98	*0.40-21.65*
Cl_7 (CB 175)	I	3.17	*0.73-13.67*	0.70	*0.15-3.21*	0.23	*0.061-0.87*	0.46	*0.064-3.27*
Cl_7 (CB 187/182)	I	112	*25.4-500*	27.8	*5.91-130*	7.79	*2.02-29.8*	15.7	*2.185-112*
Cl_7 (CB 183)	I	37.4	*8.55-163.8*	8.64	*1.75-42.3*	2.33	*0.57-9.4*	4.58	*0.62-33.4*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_7 (CB 185)	V	3.44	*0.77-15.3*	0.80	*0.16-3.83*	0.24	*0.063-0.90*	0.46	*0.068-3.14*
Cl_7 (CB 174/181)	V	29.6	*7.61-115*	5.10	*0.97-26.6*	1.88	*0.49-7.19*	4.06	*0.57-28.44*
Cl_7 (CB 177)	II	20.1	*5.99-67.4*	3.99	*0.76-20.7*	1.16	*0.28-4.66*	2.49	*0.35-17.59*
Cl_7 (CB 171)	II	10.7	*3.05-37.6*	2.14	*0.42-10.8*	0.60	*0.15-2.29*	1.24	*0.17-8.90*
Cl_7 (CB 173)	II	0.14	*0.035-0.57*	0.04	*0.01-0.14*	0.01	*0.003-0.05*	0.03	*0.005-0.21*
Cl_7 (CB 192/172)	I	9.02	*2.54-31.9*	2.29	*0.49-10.5*	0.56	*0.13-2.25*	1.06	*0.15-7.36*
Cl_7 (CB 180)	I	104	*28.8-375*	23.2	*4.73-113*	5.93	*1.46-23.4*	11.5	*1.66-78.8*
Cl_7 (CB 193)	I	6.66	*1.92-23.0*	1.53	*0.30-7.62*	0.41	*0.10-1.57*	0.88	*0.12-6.19*
Cl_7 (CB 191)	I	1.66	*0.502-5.46*	0.37	*0.07-1.85*	0.11	*0.030-0.40*	0.29	*0.038-2.25*
Cl_7 (CB 170/190)	II	41.9	*11.3-154*	8.89	*1.77-44.4*	2.06	*0.51-8.24*	4.42	*0.63-30.76*
Cl_7 (CB 189)	III	1.09	*0.31-3.75*	0.36	*0.085-1.49*	0.08	*0.022-0.31*	0.15	*0.025-0.92*
Cl_8 (CB 202)	I	14.4	*4.12-50.4*	4.14	*0.98-17.32*	1.02	*0.24-4.21*	1.49	*0.19-11.46*
Cl_8 (CB 200)	I	8.60	*2.22-33.2*	3.03	*0.76-11.97*	0.79	*0.18-3.33*	1.04	*0.14-7.66*
Cl_8 (CB 204)	I	0.24	*0.067-0.83*	0.07	*0.023-0.243*	0.03	*0.008-0.119*	0.04	*0.007-0.207*
Cl_8 (CB 197)	I	2.00	*0.478-8.33*	0.76	*0.194-3.011*	0.21	*0.050-0.926*	0.25	*0.033-1.83*
Cl_8 (CB 199)	V	0.69	*0.188-2.50*	0.23	*0.060-0.901*	0.05	*0.013-0.220*	0.08	*0.013-0.552*
Cl_8 (CB 198)	I	10.8	*1.72-68.3*	2.10	*0.214-20.56*	1.24	*0.284-5.36*	1.21	*0.089-16.5*
Cl_8 (CB 201)	I	20.2	*3.44-118*	5.43	*0.969-30.3*	-	-	2.41	*0.421-13.7*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_8 (CB 203/196)	I	20.2	*5.69-71.4*	7.08	*1.61-31.09*	1.25	*0.280-5.61*	1.22	*0.158-9.41*
Cl_8 (CB 195)	II	1.83	*0.499-6.71*	0.69	*0.155-3.07*	0.18	*0.044-0.720*	0.20	*0.029-1.34*
Cl_8 (CB 194)	I	11.3	*2.96-43.2*	4.32	*0.933-19.9*	0.79	*0.169-3.67*	0.94	*0.129-6.798*
Cl_8 (CB 205)	I	0.42	*0.124-1.44*	0.18	*0.042-0.749*	0.04	*0.011-0.177*	0.09	*0.015-0.552*
Cl_9 (CB 208)	I	6.85	*0.163-287*	1.44	*0.100-20.5*	9.36	*0.18-485*	0.13	*0.019-0.830*
Cl_9 (CB 207)	I	1.29	*0.225-7.34*	0.33	*0.043-2.51*	0.26	-	0.12	*0.018-0.776*
Cl_9 (CB 206)	I	2.77	*0.477-16.0*	1.39	*0.238-8.15*	0.50	-	0.23	*0.036-1.41*
Cl_{10} (CB 209)	I	1.13	*0.268-4.79*	0.80	*0.195-3.26*	0.12	*0.031-0.499*	0.05	*0.006-0.425*
$\sum Cl_2$		0.42	*0.134-1.33*	0.45	*0.130-1.57*	0.44	*0.185-1.02*	0.39	*0.099-1.55*
$\sum Cl_3$		16.4	*5.61-47.7*	9.49	*2.73-32.8*	8.93	*3.69-21.6*	14.7	*3.704-58.4*
$\sum Cl_4$		318	*111-907*	60.0	*11.7-305*	35.9	*11.7-109*	114	*21.12-615*
$\sum Cl_5$		1080	*365-3,170*	153	*26.7-879*	80.3	*23.9-268*	226	*35.1-1,450*
$\sum Cl_6$		1600	*552-4,640*	249	*44.1-1,400*	102	*28.4-366*	241	*33.8-1,720*
$\sum Cl_7$		436	*115-1,650*	97.9	*20.16-475*	26.5	*6.7-105*	54.7	*7.74-386*
$\sum Cl_8$		74.5	*18.7-296.1*	25.7	*6.0-108*	5.54	*1.26-24.30*	7.72	*1.05-56.6*
$\sum Cl_9$		19.2	*0.814-450*	3.51	*0.287-42.9*	14.7	*0.587-368*	0.40	*0.061-2.64*
$\sum Cl_{10}$		1.13	*0.268-4.77*	0.80	*0.195-3.26*	0.12	*0.031-0.499*	0.05	*0.006-0.42*
∑Diortho PCBs		3360	*1,1204-10,020*	587	*117.5-2,930*	244	*67.8-881*	562	*84.4-3,736*
∑Mono ortho PCBs		314	*104-944*	71.2	*14.89-340.27*	39.6	*12.92-121.6*	105	*20.2-541*

		MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{EQ}) ± *SD*		89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
∑PCBs		3690	*1,250-10,900*	661	*134-3,260*	286	*81.8-999*	669	*104-4,280*

Appendix 5 continued.

		MALE BELUGA BLOOD (*D. leucas*) (n = 7)		MALE BELUGA LIVER (*D. leucas*) (n = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (n = 7)		FEMALE BELUGA LIVER (*D. leucas*) (n = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_2 (PCB 7/9)	III	-	-	1.2	*0.188-8.02*	-	-		
Cl_2 (PCB 6)	III	-	-	1.7	*0.262-10.7*	-	-		
Cl_2 (PCB 8/5)	III	1.7	-	5.8	*0.821-40.5*	0.6	*0.229-1.71*	1.3	*0.631-2.86*
Cl_2 (PCB 4/10)	IV	-	-	5.0	*1.16-21.4*	-	-	-	-
Cl_3 (PCB 23/34)	III	-	-	-	-	-	-	-	-
Cl_3 (PCB 29)	III	1.1	-	-	-	-	-	-	-
Cl_3 (PCB 26)	III	-	-	0.9	*0.150-5.36*	-	-	0.5	*0.167-1.31*
Cl_3 (CB 25)	III	28.0	-	0.3	*0.033-1.93*	2.4	*0.594-9.45*	0.1	*0.048-0.231*
Cl_3 (CB 31)	III	16.5	*0.956-283*	8.8	*1.35-57.2*	1.6	*0.563-4.52*	-	-
Cl_3 (CB 28)	III	1.7	*0.44-6.61*	5.9	*0.924-37.68*	1.3	*0.446-3.50*	3.9	*1.05-14.3*
Cl_3 (CB 21)	III	-	-	-	-	-	-	-	-
Cl_3 (CB 33/20)	III	-	-	-	-	-	-	-	-
Cl_3 (CB 19)	IV	-	-	0.9	*0.138-5.64*	-	-	0.2	*0.055-0.450*
Cl_3 (CB30)	IV	16.2	-	-	-	1.9	*0.652-5.32*	-	-
Cl_3 (CB 18)	IV	4.0	*1.817-8.78*	12.7	*3.69-43.9*	0.6	-	-	-
Cl_3 (CB 17)	IV	1.1		2.7	*0.319-22.6*	-	-	0.2	*0.058-0.528*
Cl_3 (CB 27/24)	IV	1.8	*0.300-10.3*	1.3	*0.466-3.76*	1.4	*0.416-4.88*	1.1	*0.425-2.86*
Cl_3 (CB 16/32)	IV	1.5	*0.505-4.62*	10.1	*3.046-33.4*	-	-	-	-

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_3 (CB 22)	III	-	-		-	-	-	-	-
Cl_4 (CB 54)	V	-	-	0.2	-	-	-	-	-
Cl_4 (CB 50)	V	1120	*404-3,080*	0.2	-	22.8	*0.02-28,583*	-	-
Cl_4 (CB 53)	IV	1.1	*0.406-3.11*	1.7	*0.188-14.5*	-	-	0.1	*0.036-0.31*
Cl_4 (CB 51)	V	1.0		0.8	*0.286-2.48*	-	-		
Cl_4 (CB 45)	V	2.9	*0.530-15.9*	2.9	*0.576-14.3*	0.6	*0.223-1.47*	0.2	
Cl_4 (CB 46)	V	14.1	*1.13-174*	1.6	*0.212-11.4*	6.4		19.4	*3.62-103*
Cl_4 (CB 73/52)	IV	22.6	*1.80-282*	65.0	*7.08-597*	7.8	*1.506-39.9*	4.2	*0.789-21.9*
Cl_4 (CB 69)	IV	40.4	-	-	-	-	-	-	-
Cl_4 (CB 49)	IV	41.3	*5.19-327*	36.1	*10.1-128*	4.8	*2.02-11.54*	-	-
Cl_4 (CB 43)	IV	8.1	*3.24-19.9*	4.4	*1.02-18.5*	1.7		5.4	*1.07-27.3*
Cl_4 (CB 47/75/48)	IV	19.4	*2.73-137*	19.7	*5.69-67.8*	2.0	*0.824-4.90*	-	-
Cl_4 (CB 65)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	IV	45.6	*7.06-294*	33.3	*9.91-111*	6.1	*1.77-20.8*	-	-
Cl_4 (CB 59/42)	IV	5.0	*1.01-25.1*	4.9	*0.802-29.3*	1.6	*0.651-3.78*	0.4	-
Cl_4 (CB 72)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	IV	4.1	*0.561-29.5*	11.9	*2.21-64.0*	1.9	*0.639-5.69*	0.7	-
Cl_4 (CB 68)	III	-	-	-	-	-	-	-	-

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 40)	IV	8.1	*0.265-248*	1.9	*0.545-6.48*	4.6	*1.54-13.4*	-	-
Cl_4 (CB 57)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 67)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 58)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 61/74)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 70/76)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 66)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 55)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 60/56)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 104)	IV	-	-	-	-	-	-		
Cl_5 (CB 96)	IV	2.2	*0.713-6.77*	0.8	*0.320-1.94*	1.0	*0.206-4.941*	-	-
Cl_5 (CB 103)	IV	0.9	*0.440-1.94*	1.6	*0.378-7.01*	1.1	-	0.5	*0.190-1.46*
Cl_5 (CB 100)	II	-	-	1.2	*0.465-3.00*	-	-	-	-
Cl_5 (CB 94)	V	0.9	-	0.5	*0.223-1.22*	-	-	-	-
Cl_5 (CB 95)	V	27.4	*2.25-333*	98.7	*17.7-547*	7.7	-	19.0	*4.434-81.16*
Cl_5 (CB 102/93)	V	55.4	-	2.8	*0.726-10.7*	3.7	*1.05-13.17*	-	-
Cl_5 (CB 98)	IV	2.2	*0.367-13.68*	0.3	*0.115-0.969*	3.0	*1.12-8.20*	0.4	*0.118-1.17*
Cl_5 (CB 88)	V	2.2	*0.164-30.4*	2.1	*0.650-6.54*	4.1	*1.32-12.6*	2.3	*0.491-10.9*

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 91)	V	4.6	*1.17-17.6*	18.1	*4.98-65.6*	0.9		-	-
Cl_5 (CB 121)	I	4.3	-	-	-	1.1	*0.398-2.960*	-	-
Cl_5 (CB 92/84)	V	27.4	*12.1-61.7*	43.4	*12.52-150*	2.3	-	-	-
Cl_5 CB (101/90)	IV	38.5	*2.78-533*	157	*27.82-888*	10.7	-	32.6	*5.96-177*
Cl_5 (CB 89)	V	10.8	*2.40-48.5*	11.7	*2.383-57.9*	1.3	-	1.4	-
Cl_5 (CB 99)	II	29.1	*3.08-274*	115	*20.3-648*	7.6	-	26.3	*4.08-169*
Cl_5 (CB 113)	IV	1.2	-	0.6	-	-	-	-	-
Cl_5 (CB 119)	II	10.4	*0.768-140*	6.8	*1.27-36.4*	8.8	*2.43-31.5*	1.5	*0.259-8.19*
Cl_5 (CB 112)	IV	3.7	*1.39-9.82*	0.5	*0.133-1.82*	0.3	*0.126-0.749*	0.6	*0.122-3.10*
Cl_5 (CB 109/83)	IV	28.8	*13.4-61.6*	6.4	*2.05-20.0*	48.3		13.2	*6.34-27.5*
Cl_5 (CB 97/86)	IV	453	*13.9-14,650*	23.0	*5.87-89.7*	5.1	*0.01-2,205*	-	-
Cl_5 (CB 116/125/117)	IV	34.9	-	0.7	*0.270-1.59*	1.0	*0.277-3.62*	-	-
Cl_5 (CB 115/87)	II	22.7	*8.34-61.8*	41.4	*7.94-215*	2.2		11.0	*2.08-57.6*
Cl_5 (CB 111)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 85)	II	13.8	*3.84-49.9*	24.7	*4.73-128*	1.2	*0.255-5.26*	6.4	*1.17-35.3*
Cl_5 (CB 120)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 110)	IV	54.5	*7.97-371*	42.7	*8.27-220*	12.4	*3.45-44.5*	14.5	*2.95-70.8*
Cl_5 (CB 82)	IV	2.6	*1.14-6.07*	4.0	*0.813-19.7*	2.2	-	1.4	*0.339-5.94*

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 124)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 108/107)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 123)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 106/118)	III	318	-	96.7	*5.39-1,740*	11.3	*2.89-44.2*	-	-
Cl_5 (CB 114)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 122)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 105)	III	-	-	-	-	-	-	-	-
Cl_6 (CB 155)	I	-	-	1.4	*0.278-6.78*	-	-	-	-
Cl_6 (CB 150)	V	22.2		0.7	*0.205-2.44*	1.4	-	-	-
Cl_6 (CB 152)	V	2300	*100-5,270*	0.4	*0.126-1.09*	4050	-	-	-
Cl_6 (CB 145)	V	11.1		0.2	*0.070-0.343*	-	-	-	-
Cl_6 (CB 148)	I	14.1	*2.69-74.1*	1.6	*0.451-5.56*	2.9	*0.882-9.69*	-	-
Cl_6 (CB 136)	V	-	-	23.7	*6.93-80.9*	-	-	-	-
Cl_6 (CB 154)	I	153	-	7.1	*2.15-23.9*	5.0	*1.39-17.5*	-	-
Cl_6 (CB 151)	V	33.2	*13.9-78.6*	50.6	*9.58-267*	5.1	-	13.0	*2.24-74.9*
Cl_6 (CB 135/144)	V	21.3	*5.41-83.6*	29.6	*8.09-108*	2.8	*0.813-9.31*	9.5	
Cl_6 (CB 147)	II	82.2	*33.3-202*	7.5	*1.90-29.3*	11.3	-	26.8	*6.27-114*
Cl_6 (CB 149)	V	241	-	178	*51.8-610*	9.9	*2.57-38.3*	-	-

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 139/140)	II	18.0	-	1.1	*0.327-3.56*	1.2	-	-	-
Cl_6 CB-143/134	V	3.2	*1.19-8.47*	3.6	*0.75-16.7*		-	1.5	*0.225-9.47*
Cl_6 (CB 142/131)	V	4.9	*1.22-19.2*	0.7	*0.207-2.19*	1.2	-	-	-
Cl_6 (CB 133)	I	16.8	-	0.1	*0.054-0.216*	-	-	0.1	-
Cl_6 (CB 146/161)	I	39.4	*6.39-242*	42.5	*8.56-210*	10.2	*2.43-42.1*	14.6	*2.12-100*
Cl_6 (CB 165)	I	8.1	-	1.0	*0.253-3.74*	-	-	-	-
Cl_6 (CB 132/153)	I	187	*72.1-485*	327	*63.8-1,670*	32.8	-	101	*14.6-692*
Cl_6 (CB 168)	I	110	-	30.5	*8.90-104*	4.6	*1.27-16.53*	-	-
Cl_6 (CB 141)	IV	5.4	*2.26-12.7*	9.5	*1.84-49.0*	1.8	-	3.7	*0.503-27.45*
Cl_6 (CB 137)	II	4.0	*1.21-13.3*	8.8	*1.749-43.847*	1.1	-	2.7	*0.430-16.48*
Cl_6 (CB 130)	II	8.8	*3.54-21.6*	14.0	*2.69-72.7*	0.5	-	3.9	*0.644-23.6*
Cl_6 (CB 160/163/164/138)	II	137	*53.8-345*	230	*44.5-1,190*	20.5	-	71.6	*10.4-491*
Cl_6 (CB 158)	II	6.7	*2.68-16.9*	13.2	*2.36-74.0*	1.0	-	3.2	*0.488-20.3*
Cl_6 (CB 129)	IV	48.8		1.8	*0.429-7.51*	1.1	*0.357-3.44*	-	-
Cl_6 (CB 166)	II	5.0	*0.600-42.1*	1.0	*0.278-3.31*	0.5	*0.149-1.46*	0.5	-
Cl_6 (CB 159)	III	-	-	-	-	-	-	-	-
Cl_6 (CB 162)	IV	-	-	-	-	-	-	-	-

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{EQ}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 128)	II	20.7	*3.01-142*	24.0	*4.59-124*	4.4	*1.18-16.10*	6.6	*1.15-37.3*
Cl_6 (CB 167)	III	-	-	-	-	-	-	-	-
Cl_6 (CB 156)	III	-	-	-	-	-	-	-	-
Cl_6 (CB 157)	III	-	-	-	-	-	-	-	-
Cl7 (CB 188)	I	79.7	-	3.1	*1.13-8.58*	2.3	*0.717-7.08*	-	-
Cl_7 (CB 184)	I	11.1	-	2.1	*0.456-9.74*	-	-	-	-
Cl_7 (CB 179)	V	14.3	*1.10-183*	14.0	*2.64-74.0*	6.8	*1.35-34.26*	2.9	*0.565-15.2*
Cl_7 (CB 176)	V	1.6	*0.635-3.86*	5.0	*1.06-23.5*	-	-	0.7	
Cl_7 (CB 186)	V	1.6	*0.165-14.8*	1.1	*0.071-16.9*	0.9	*0.328-2.69*	0.4	*0.161-0.898*
Cl_7 (CB 178)	I	9.0	*2.99-27.0*	16.2	*3.68-71.5*	1.2	*0.388-3.81*	5.9	*1.20-29.4*
Cl_7 (CB 175)	I	1.3	*0.464-3.76*	3.9	*0.615-25.0*	0.9	-	1.0	*0.255-3.55*
Cl_7 (CB 187/182)	I	44.0	*11.2-171*	95.2	*22.8-397*	5.5	*1.54-19.33*	32.5	*7.49-141*
Cl_7 (CB 183)	I	10.0	*3.62-27.5*	27.2	*5.68-129*	2.8	-	10.2	*2.17-48.3*
Cl_7 (CB 185)	V	3.7	*0.085-163*	3.5	*0.661-18.39*	32.1	*8.929-115.51*	0.9	*0.233-3.150*
Cl_7 (CB 174/181)	V	19.5	*4.96-76.3*	26.7	*5.523-129.50*	1.9	*0.586-6.168*	5.4	*1.603-18.455*
Cl_7 (CB 177)	II	8.9	*1.61-49.07*	14.2	*3.184-62.941*	1.7	*0.521-5.863*	3.8	*0.874-16.565*
Cl_7 (CB 171)	II	4.6	*0.867-24.7*	8.6	*1.937-38.103*	1.0	*0.291-3.705*	2.4	*0.543-10.973*
Cl_7 (CB 173)	II	45.6	-	0.3	*0.024-4.235*	2.2	*0.867-5.411*	0.1	-

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_7 (CB 192/172)	I	7.5	*0.290-193*	8.1	*1.517-42.98*	13.8	*1.25-152.58*	2.4	*0.555-10.799*
Cl_7 (CB 180)	I	32.1	*11.7-87*	74.3	*13.38-412.02*	2.0	*0.372-10.298*	24.4	*3.808-156.763*
Cl_7 (CB 193)	I	3.6	*1.1-11.9*	5.7	*1.276-25.083*	0.6	-	1.9	*0.411-8.963*
Cl_7 (CB 191)	I	0.9	*0.383-1.9*	2.7	*0.533-13.890*	-	-	0.9	
Cl_7 (CB 170/190)	II	9.1	*3.37-24.5*	30.6	*6.751-138.92*	0.9	*0.159-4.845*	8.7	*1.932-39.069*
Cl_7 (CB 189)	III	-	-	-	-	-	-	-	-
Cl_8 (CB 202)	I	3.8	*1.78-7.9*	11.0	*3.000-40.396*	1.5	*0.343-6.266*	2.9	
Cl_8 (CB 200)	I	6.1	*0.572-64.113*	7.7	*2.297-25.575*	2.8	*0.912-8.646*	5.0	*0.970-25.399*
Cl_8 (CB 204)	I	13.4	-	0.3	*0.049-1.511*	0.7	*0.255-1.925*	0.2	*0.044-0.759*
Cl_8 (CB 197)	I	1.7	*0.147-19.331*	1.9	*0.562-6.571*	1.0	*0.282-3.394*	1.4	*0.280-6.839*
Cl_8 (CB 199)	V	8.8	-	0.5	*0.137-1.962*	-	-	0.3	*0.064-1.038*
Cl_8 (CB 198)	I	5.2	*2.067-13.062*	1.8	*0.234-14.380*	0.3	*0.114-0.899*	7.3	*1.289-41.161*
Cl_8 (CB 201)	I	3.7	-	18.3	*6.81-49.055*	0.3	*0.079-0.834*	-	
Cl_8 (CB 203/196)	I	34.3	-	12.7	*2.96-54.120*	1.2	*0.455-3.231*	8.8	*1.479-52.815*
Cl_8 (CB 195)	II	2.7	*0.202-35.661*	1.7	*0.39-7.414*	0.4	*0.165-0.832*	1.0	*0.157-5.833*
Cl_8 (CB 194)	I	4.0	*1.584-9.956*	7.2	*1.55-33.5*	4.2	-	6.0	*1.330-27.502*
Cl_8 (CB 205)	I	38.0	-	0.5	*0.112-2.51*	1.9	*0.532-6.996*	0.3	*0.072-1.596*
Cl_9 (CB 208)	I	1.5	*0.222-10.038*	3.0	*1.017-8.98*	0.4	*0.120-1.343*	2.6	*0.683-10.126*
Cl_9 (CB 207)	I	4.7	*0.091-243.395*	3.0	*0.955-9.62*	8.2	*2.626-25.633*	2.1	*0.645-6.622*

		MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*		0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_9 (CB 206)	I	4.9	*0.282-83.576*	5.0	*1.640-15.041*	2.3	*0.647-8.331*	4.3	*1.168-15.509*
Cl_{10} (CB 209)	I	1.2	*0.184-7.221*	3.5	*1.164-10.734*	0.4	*0.097-1.755*	4.5	*1.480-13.549*
$\sum Cl_2$		1.7	-	10.2	*1.483-70.382*	0.6	*0.229-1.711*	1.3	*0.631-2.869*
$\sum Cl_3$		12.0	*1.065-134.590*	34.8	*6.538-184.72*	5.6	*1.476-21.59*	5.9	*1.806-19.586*
$\sum Cl_4$		674.9	*122.830-3,707.894*	162.4	*27.90-944.99*	62.1	*3.76-1,024.5*	29.7	*5.710-154.936*
$\sum Cl_5$		1791.0	*749.343-4,280.519*	688.7	*129.6-3,658.3*	106.1	*8.22-1,369.1*	134.6	*28.748-630.54*
$\sum Cl_6$		2427.6	*840.428-7,012.343*	913.7	*173.5-4,810.0*	112.0	*6.9-1,799.7*	251.8	*38.79-1,633.7*
$\sum Cl_7$		238.6	*31.252-1,821.75*	367.8	*84.5-1,600.26*	71.6	*18.62-275.03*	104.8	*21.973-499.98*
$\sum Cl_8$		16.1	*1.634-157.786*	60.4	*17.67-206.47*	7.6	*2.115-27.235*	33.2	*7.026-156.586*
$\sum Cl_9$		13.0	*0.476-354.737*	8.4	*2.113-33.409*	7.8	*1.615-37.709*	9.0	*2.493-32.221*
$\sum Cl_{10}$		1.2	*0.184-7.221*	3.5	*1.164-10.734*	0.4	*0.097-1.755*	4.5	*1.480-13.549*
∑Diortho PCBs		5876.8	*2,605.12-13,257.30*	2212.3	*463.2-10,565.7*	457.2	*40.1-5,212.3*	573.3	*105.94-3,102.44*
∑Mono ortho PCBs		9.1	*0.302-275.258*	79.5	*5.933-1,065.19*	10.2	*1.60-64.65*	5.9	*1.88-18.49*
∑PCBs		5960.3	*2,623.82-13,539.54*	2342.4	*485.50-11,301.46*	484.4	*43.83-5,352.85*	579.6	*107.72-3,118.93*

Appendix 5 continued.

		FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 11)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_2 (PCB 7/9)	III	-	-	0.091	-	-	-	-	-
Cl_2 (PCB 6)	III	0.061	*0.029-0.126*	0.098	*0.036-0.268*	-	-	0.03	-
Cl_2 (PCB 8/5)	III	0.29	*0.106-0.790*	0.33	*0.111-0.973*	0.22	*0.032-1.460*	0.21	*0.078-0.562*
Cl_2 (PCB 4/10)	IV	0.11	*0.038-0.294*	0.12	*0.036-0.420*	0.23	-	0.03	-
Cl_3 (PCB 23/34)	III	0.006	-	0.008	*0.003-0.023*	-	-	-	-
Cl_3 (PCB 29)	III	-	-	0.003	*0.002-0.007*	-	-	-	-
Cl_3 (PCB 26)	III	0.27	*0.088-0.816*	0.23	*0.054-0.943*	0.25	-	0.06	-
Cl_3 (CB 25)	III	0.055	*0.024-0.125*	0.046	*0.014-0.152*	-	-	0.03	-
Cl_3 (CB 31)	III	3.87	*0.823-18.180*	2.26	*0.688-7.455*	-	-	-	-
Cl_3 (CB 28)	III	5.79	*1.522-22.022*	7.69	*2.709-21.806*	3.40	*0.755-15.321*	11.10	*2.183-56.479*
Cl_3 (CB 21)	III	0.24	*0.114-0.491*	0.35	*0.163-0.742*	-	-	-	-
Cl_3 (CB 33/20)	III	0.17	*0.077-0.387*	0.18	*0.058-0.552*	-	-	-	-
Cl_3 (CB 19)	IV	0.025	-	0.061	-	-	-	-	-
Cl_3 (CB30)	IV	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	IV	0.22	*0.080-0.625*	0.26	*0.084-0.832*	0.16	*0.032-0.799*	0.22	*0.092-0.531*
Cl_3 (CB 17)	IV	0.085	*0.032-0.225*	0.096	*0.030-0.304*	0.10	*0.014-0.645*	0.10	*0.037-0.243*
Cl_3 (CB 27/24)	IV	0.031	*0.014-0.071*	0.033	*0.010-0.111*	-	-	0.03	-
Cl_3 (CB 16/32)	IV	0.24	*0.095-0.630*	0.29	*0.094-0.909*	0.30	*0.044-2.025*	0.26	*0.106-0.629*

		FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 11)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_3 (CB 22)	III	0.22	*0.068-0.679*	0.19	*0.050-0.745*	-	-	-	-
Cl_4 (CB 54)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	V	0.047	-	-	-	-	-	-	-
Cl_4 (CB 53)	IV	0.021	*0.009-0.054*	0.017	*0.008-0.036*	-	-	-	-
Cl_4 (CB 51)	V	-	-	-	-	-	-	-	-
Cl_4 (CB 45)	V	0.017	*0.005-0.054*	0.014	*0.005-0.038*	0.13	-	0.08	-
Cl_4 (CB 46)	V	0.009	*0.004-0.018*	0.018	*0.006-0.055*	-	-	-	-
Cl_4 (CB 73/52)	IV	7.43	*1.765-31.291*	10.09	*3.641-27.984*	0.67	*0.140-3.189*	1.26	*0.271-5.904*
Cl_4 (CB 69)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	IV	1.66	*0.436-6.351*	2.24	*0.792-6.356*	0.34	*0.081-1.388*	0.98	*0.242-3.980*
Cl_4 (CB 43)	IV					1.01	*0.148-6.818*	10.65	*5.275-21.520*
Cl_4 (CB 47/75/48)	IV	1.73	*0.380-7.889*	2.38	*0.924-6.140*	0.65	*0.217-1.968*	1.77	*0.599-5.251*
Cl_4 (CB 65)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	IV	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	IV	1.08	*0.361-3.244*	1.35	*0.445-4.122*	0.43	*0.106-1.722*	1.46	*0.135-15.640*
Cl_4 (CB 59/42)	IV	0.127	*0.043-0.369*	0.13	*0.046-0.366*	0.24	*0.045-1.304*	1.45	*0.135-15.610*
Cl_4 (CB 72)	III	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	IV	0.30	*0.040-2.245*	0.26	*0.027-2.544*	0.79	*0.221-2.824*	6.87	*1.700-27.758*
Cl_4 (CB 68)	III	0.20	*0.066-0.607*	0.22	-		-	-	-

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{EQ}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_4 (CB 40)	IV	0.028	*0.007-0.103*	0.044	*0.021-0.094*	0.18	-	1.04	*0.069-15.695*
Cl_4 (CB 57)	III	0.012	*0.004-0.039*	0.015	-	-	-	-	-
Cl_4 (CB 67)	III	0.034	*0.014-0.082*	0.028	*0.009-0.086*	-	-	-	-
Cl_4 (CB 58)	III	0.005	*0.001-0.020*	0.007	-	-	-	-	-
Cl_4 (CB 63)	III	0.44	*0.066-2.938*	0.27	*0.031-2.257*	-	-	-	-
Cl_4 (CB 61/74)	III	12.47	*2.438-63.747*	18.38	*6.649-50.784*	-	-	-	-
Cl_4 (CB 70/76)	III	2.89	*1.112-7.527*	2.98	*1.046-8.462*	-	-	-	-
Cl_4 (CB 66)	III	8.23	*1.866-36.324*	7.42	*1.566-35.195*	-	-	-	-
Cl_4 (CB 55)	III	0.009	*0.004-0.021*	0.017	*0.006-0.054*	-	-	-	-
Cl_4 (CB 60/56)	III	2.94	*0.741-11.640*	2.53	*0.629-10.174*	-	-	-	-
Cl_5 (CB 104)	IV	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	IV	0.004	-	-	-	-	-	0.05	-
Cl_5 (CB 103)	IV	0.022	*0.006-0.077*	0.018	*0.004-0.076*	-	-	0.08	*0.016-0.352*
Cl_5 (CB 100)	II	0.013	*0.005-0.034*	-	-	-	-	0.04	-
Cl_5 (CB 94)	V	0.11	*0.044-0.270*	0.096	*0.042-0.219*	-	-	-	-
Cl_5 (CB 95)	V	1.61	*0.344-7.508*	1.82	*0.563-5.909*	0.77	*0.170-3.470*	5.64	*1.431-22.211*
Cl_5 (CB 102/93)	V	-	-	0.008	-	-	-	-	-
Cl_5 (CB 98)	IV	-	-	-	-	-	-	0.46	-
Cl_5 (CB 88)	V	0.19	*0.033-1.154*	0.22	*0.069-0.689*	0.24	*0.068-0.843*	0.38	*0.110-1.299*

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (n = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (n = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (n =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (n = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 91)	V	0.26	*0.063-1.096*	0.36	*0.139-0.906*	0.10	*0.024-0.412*	0.72	*0.106-4.933*
Cl_5 (CB 121)	I	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	V	-	-	-	-	-	-	-	-
Cl_5 CB (101/90)	IV	26.84	*5.521-130.500*	36.22	*14.20-92.37*	1.90	*0.453-7.965*	21.33	*6.832-66.562*
Cl_5 (CB 89)	V	0.019	*0.008-0.046*	0.022	*0.006-0.072*	0.12	*0.023-0.606*	2.21	*0.181-27.012*
Cl_5 (CB 99)	II	33.49	*5.678-197.566*	42.10	*15.50-114.30*	17.54	*4.44-69.29*	119.10	*18.92-749.59*
Cl_5 (CB 113)	IV	0.013	-	-	-	-	-	-	-
Cl_5 (CB 119)	II	1.19	*0.231-6.191*	1.42	*0.525-3.846*	0.19	*0.047-0.765*	1.65	*0.441-6.176*
Cl_5 (CB 112)	IV	0.008	-	-	-	0.08	-	0.10	*0.039-0.281*
Cl_5 (CB 109/83)	IV	0.070	*0.023-0.215*	0.061	*0.020-0.184*	-	-	1.60	
Cl_5 (CB 97/86)	IV	0.53	*0.191-1.484*	0.53	*0.180-1.579*	0.11	*0.039-0.319*	1.16	*0.064-21.039*
Cl_5 (CB 116/125/117)	IV	3.02	*0.755-12.087*	3.63	-	2.08	-	4.55	*0.095-217.230*
Cl_5 (CB 115/87)	II	4.45	*1.139-17.399*	6.33	*2.937-13.652*	2.73	*0.425-17.49*	9.12	*1.708-48.654*
Cl_5 (CB 111)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 85)	II	3.36	*0.659-17.098*	4.34	*1.585-11.882*	1.52	*0.374-6.199*	14.58	*4.378-48.582*
Cl_5 (CB 120)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 110)	IV	5.47	*1.640-18.227*	5.60	*1.840-17.031*	1.06	*0.324-3.470*	4.63	*0.447-48.065*
Cl_5 (CB 82)	IV	0.062	*0.024-0.156*	0.059	*0.020-0.170*	0.02	*0.001-0.353*	2.08	*0.210-20.726*
Cl_5 (CB 124)	III	0.058	*0.019-0.178*	0.077	*0.021-0.285*	-	-	-	-

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_5 (CB 108/107)	III	2.37	*0.486-11.551*	2.16	*0.551-8.493*	-	-	-	-
Cl_5 (CB 123)	III	0.983	*0.108-8.930*	4.49	*0.17-116.14*	-	-	-	-
Cl_5 (CB 106/118)	III	51.39	*11.421-231.19*	47.81	*13.81-165.46*	-	-	-	-
Cl_5 (CB 114)	III	0.92	*0.154-5.452*	1.04	*0.374-2.882*	-	-	-	-
Cl_5 (CB 122)	III	-	-	-	-	-	-	-	-
Cl_5 (CB 105)	III	11.40	*1.971-65.979*	13.13	*4.565-37.767*	-	-	-	-
Cl_6 (CB 155)	I	-	-	-	-	-	-	-	-
Cl_6 (CB 150)	V	0.003	-	-	-	-	-	-	-
Cl_6 (CB 152)	V	0.063	-	0.054	-	0.16	-	0.03	-
Cl_6 (CB 145)	V	-	-	-	-		-	-	-
Cl_6 (CB 148)	I	-	-	0.15	-	0.15	*0.033-0.637*	2.17	*0.343-13.677*
Cl_6 (CB 136)	V	0.11	*0.037-0.314*	0.084	*0.027-0.262*	-	-	-	-
Cl_6 (CB 154)	I	0.67	*0.140-3.218*	0.64	*0.181-2.230*	-	-	-	-
Cl_6 (CB 151)	V	1.14	*0.270-4.780*	1.46	*0.493-4.331*	0.44	*0.105-1.858*	3.23	*0.977-10.712*
Cl_6 (CB 135/144)	V	0.76	*0.185-3.072*	0.82	*0.331-2.052*	0.31	*0.080-1.230*	2.60	*0.889-7.595*
Cl_6 (CB 147)	II	0.24	*0.044-1.245*	0.26	*0.092-0.737*	1.18	*0.378-3.691*	12.67	*1.025-156.581*
Cl_6 (CB 149)	V	7.31	*1.704-31.384*	7.74	*3.147-19.051*	4.01	*1.15-13.99*	31.91	*10.391-98.012*
Cl_6 (CB 139/140)	II	0.005	-	0.008	-	-	-	-	-
Cl_6 CB-143/134	V	2.25	-	1.91	*0.929-3.916*	-	-	-	-

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_6 (CB 142/131)	V	0.96	*0.188-4.871*	1.47	*0.508-4.220*	0.49	*0.127-1.861*	7.24	*1.028-51.011*
Cl_6 (CB 133)	I	0.49	*0.022-10.890*	0.32	*0.057-1.761*	0.15	*0.043-0.511*	0.59	*0.062-5.574*
Cl_6 (CB 146/161)	I	16.0	*2.477-103.277*	19.35	*7.559-49.538*	13.49	*3.783-48.115*	124.86	*16.705-933.291*
Cl_6 (CB 165)	I	0.13	-	-	-	-	-	0.19	
Cl_6 (CB 132/153)	I	126.71	*19.42-826.64*	153.46	*60.12-391.64*	102.77	*27.35-386.0*	841.44	*123.5-5,731.1*
Cl_6 (CB 168)	I	0.50	*0.104-2.439*	0.68	*0.268-1.725*	-	-	-	-
Cl_6 (CB 141)	IV	1.87	*0.365-9.591*	2.13	*0.689-6.597*	0.25	*0.064-1.013*	1.80	*0.488-6.636*
Cl_6 (CB 137)	II	3.09	*0.487-19.643*	3.55	*1.117-11.291*	1.31	*0.262-6.584*	7.65	*1.767-33.161*
Cl_6 (CB 130)	II	3.18	*0.458-22.093*	3.60	*1.462-8.883*	3.23	*1.022-10.17*	95.66	*23.522-389.05*
Cl_6 (CB 160/163/164/138)	II	61.72	*9.494-401.177*	82.95	*37.47-183.62*	63.79	*17.43-233.4*	508.52	*65.97-3,919.3*
Cl_6 (CB 158)	II	1.76	*0.291-10.634*	2.51	*1.046-6.020*	1.89	*0.536-6.675*	15.13	*2.432-94.073*
Cl_6 (CB 129)	IV	0.25	*0.036-1.711*	0.30	*0.118-0.780*	0.14	*0.045-0.439*	1.13	*0.253-5.021*
Cl_6 (CB 166)	II	0.23	*0.031-1.722*	0.20	*0.082-0.493*	0.25	*0.070-0.909*	1.00	*0.185-5.429*
Cl_6 (CB 159)	III	0.42	*0.062-2.894*	0.57	*0.230-1.414*	-	-	-	-
Cl_6 (CB 162)	IV	0.43	*0.065-2.837*	0.43	*0.162-1.147*	-	-	-	-
Cl_6 (CB 128)	II	1.15	*0.181-7.247*	1.74	*0.601-5.057*	8.01	*2.05-31.20*	71.76	*8.306-619.951*
Cl_6 (CB 167)	III	0.74	*0.131-4.210*	0.56	*0.120-2.647*	-	-	-	-
Cl_6 (CB 156)	III	2.10	*0.329-13.391*	2.64	*1.080-6.466*	-	-	-	-
Cl_6 (CB 157)	III	0.89	*0.149-5.303*	1.02	*0.456-2.292*	-	-	-	-

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl7 (CB 188)	I			0.003		2.18	*0.911-5.215*	1.18	*0.220-6.352*
Cl_7 (CB 184)	I	0.074	*0.014-0.392*	0.037	*0.010-0.147*	0.40	*0.108-1.453*	0.34	*0.122-0.952*
Cl_7 (CB 179)	V	0.067	*0.029-0.159*	0.067	*0.023-0.196*	0.20	*0.049-0.771*	0.89	*0.199-3.936*
Cl_7 (CB 176)	V	0.042	*0.017-0.100*	0.037	*0.014-0.094*	0.09	*0.026-0.300*	0.50	*0.149-1.657*
Cl_7 (CB 186)	V	-	-	-	-	-	-	-	-
Cl_7 (CB 178)	I	3.24	*0.435-24.210*	3.44	*1.23-9.60*	1.18	*0.287-4.834*	20.60	*3.348-126.796*
Cl_7 (CB 175)	I	0.058	*0.012-0.286*	0.087	*0.036-0.208*	1.13	*0.359-3.549*	4.16	*0.752-22.982*
Cl_7 (CB 187/182)	I	9.22	*1.330-63.932*	13.76	*5.468-34.631*	31.18	*8.71-111.5*	237.48	*36.4-1,549.1*
Cl_7 (CB 183)	I	4.91	*0.701-34.424*	7.02	*2.616-18.841*	9.27	*2.417-35.52*	71.11	*12.09-418.28*
Cl_7 (CB 185)	V	0.079	*0.018-0.352*	0.089	*0.027-0.295*	0.07	*0.019-0.25*	0.31	*0.113-0.845*
Cl_7 (CB 174/181)	V	0.62	*0.127-3.040*	0.48	*0.118-1.946*	0.71	*0.196-2.594*	3.79	*1.378-10.433*
Cl_7 (CB 177)	II	0.57	*0.092-3.528*	1.13	*0.356-3.613*	4.98	*1.47-16.83*	45.68	*6.181-337.59*
Cl_7 (CB 171)	II	0.81	*0.122-5.356*	1.18	*0.406-3.445*	2.15	*0.556-8.321*	21.84	*3.665-130.083*
Cl_7 (CB 173)	II	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	I	1.46	*0.180-11.791*	1.77	*0.594-5.277*	1.47	*0.332-6.534*	7.79	*1.333-45.480*
Cl_7 (CB 180)	I	19.06	*2.55-142.260*	26.16	*8.598-79.566*	24.76	*5.39-113.59*	148.33	*27.89-788.67*
Cl_7 (CB 193)	I	1.25	*0.168-9.281*	1.76	*0.610-5.062*	1.84	*0.446-7.607*	11.10	*1.582-77.937*
Cl_7 (CB 191)	I	0.33	*0.048-2.271*	0.44	*0.154-1.236*	0.42	*0.102-1.708*	2.24	*0.389-12.865*
Cl_7 (CB 170/190)	II	6.30	*0.836-47.423*	9.23	*3.031-28.103*	7.19	*1.482-34.872*	52.91	*8.062-347.204*

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Cl_7 (CB 189)	III	0.14	*0.027-0.707*	0.17	*0.061-0.449*				
Cl_8 (CB 202)	I	1.87	*0.249-14.024*	1.85	*0.684-5.029*	2.45	*0.672-8.914*	27.11	*3.828-191.941*
Cl_8 (CB 200)	I	0.023	*0.009-0.059*	0.019	*0.007-0.053*	3.90	*1.340-11.33*	13.63	*2.569-72.272*
Cl_8 (CB 204)	I	0.022		0.007	*0.001-0.030*	0.04	*0.012-0.129*	0.04	
Cl_8 (CB 197)	I	0.044	*0.007-0.291*	0.054	*0.018-0.164*	2.18	*0.620-7.677*	6.09	*1.344-27.563*
Cl_8 (CB 199)	V	0.011	*0.005-0.026*	0.009	*0.002-0.037*	0.04	*0.012-0.141*	0.18	*0.065-0.507*
Cl_8 (CB 198)	I	0.59	*0.039-9.127*	1.11	*0.073-16.853*	0.22	*0.056-0.866*	0.62	*0.129-3.036*
Cl_8 (CB 201)	I	3.12	*0.351-27.699*	1.50	*0.648-3.460*	5.60	*1.391-22.57*	33.33	*4.905-226.477*
Cl_8 (CB 203/196)	I	2.73	*0.266-28.104*	2.92	*0.956-8.935*	6.74	*1.48-30.67*	39.27	*7.443-207.239*
Cl_8 (CB 195)	II	0.363	*0.046-2.865*	0.48	*0.136-1.692*	1.07	*0.235-4.838*	7.62	*1.369-42.404*
Cl_8 (CB 194)	I	1.86	*0.221-15.628*	2.44	*0.704-8.454*	4.61	*0.875-24.23*	29.89	*4.893-182.627*
Cl_8 (CB 205)	I	0.20	*0.049-0.832*	0.12	*0.035-0.438*	0.33	*0.075-1.483*	1.46	*0.251-8.474*
Cl_9 (CB 208)	I	0.22	*0.028-1.703*	0.20	*0.062-0.616*	2.78	*0.59-13.018*	10.56	*1.673-66.612*
Cl_9 (CB 207)	I	0.032	*0.004-0.231*	0.062	*0.021-0.181*	2.84	*0.58-13.87*	5.47	*1.194-25.035*
Cl_9 (CB 206)	I	0.71	*0.076-6.544*	0.74	*0.206-2.656*	5.11	*0.87-29.76*	21.80	*3.701-128.419*
Cl_{10} (CB 209)	I	0.21	*0.024-1.779*	0.22	*0.076-0.650*	5.43	*0.994-29.71*	15.07	*2.496-90.971*
ΣCl_2		0.42	*0.150-1.162*	0.49	*0.142-1.652*	0.23	*0.030-1.81*	0.24	*0.105-0.525*
ΣCl_3		8.78	*3.023-25.470*	11.46	*3.89-33.64*	3.71	*0.81-16.97*	11.85	*2.520-55.671*
ΣCl_4		41.92	*10.262-171.20*	50.55	*17.17-148.76*	3.18	*0.780-12.96*	19.07	*5.075-71.634*
ΣCl_5		135.09	*26.550-687.29*	159.89	*57.9-441.5*	27.64	*7.25-105.35*	247.22	*76.9-794.3*

		FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 11)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{EQ}) ± *SD*		71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
Congener	**Group**	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
ΣCl_6		234.59	*37.06-1,484.84*	289.77	*120.3-697.8*	201.93	*55.21-738.4*	1698.89	*248.1-11,631*
ΣCl_7		48.65	*6.812-347.444*	67.23	*23.703-190.7*	89.39	*22.2-358.3*	640.56	*107.2-3,825.4*
ΣCl_8		8.41	*0.991-71.357*	10.32	*3.414-31.1*	27.64	*6.676-114.4*	163.85	*27.94-960.775*
ΣCl_9		0.97	*0.112-8.295*	0.96	*0.270-3.430*	10.77	*2.037-56.985*	37.99	*6.5-220.1*
ΣCl_{10}		0.21	*0.024-1.7*	0.22	*0.076-0.6*	5.43	*0.994-29.71*	15.07	*2.49-90.97*
ΣDiortho PCBs		390.64	*66.108-2,308.3*	486.25	*206.5-1,144.8*	372.74	*97.1-1,429.5*	2953.04	*517.1-16,86*
ΣMono ortho PCBs		97.78	*20.825-459.0*	110.34	*36.6-332.38*				
ΣPCBs		492.74	*87.853-2,763.6*	602.32	*252.3-1,437.6*	372.74	*97.186-1,429.58*	2953.04	*517.12-16,863.34*

Appendix 6 Organochlorine pesticide concentrations in lichens and macro-algae (ng·g^{-1} lipid equivalent wt.), sediment (ng·g $^{-1}$ OC wt) and tissues' of various marine biota (ng·g^{-1} lipid wt.) collected from E. Hudson's Bay during May and September 1999-2002.

	LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		**MACRO ALGAE** (*F. gardneri*) **(tissue)** (*n* =11)		**SEDIMENTS** (***n* ponar grabs)** (*n* =12)		**CAPELIN** (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Chlorobenzenes (CBz)								
1,3,5 TriCBz	0.09	*0.025-0.328*	1.07	*0.384-3.010*	0.44	*0.045-4.314*	-	-
1,2,4 TriCBz	0.08	*0.023-0.304*	3.73	*1.144-12.196*	79.40	*20.119-313.388*	-	-
1,2,3 TriCBz	0.09	*0.025-0.330*	1.53	*0.496-4.738*	1.84	*0.328-10.282*	-	-
1,2,3,5/1,2,4,5 TeCBz	0.24	*0.060-0.985*	0.67	*0.236-1.906*	1.15	*0.199-6.601*	-	-
1,2,3,4 TeCBz	1.00	*0.250-4.009*	0.98		0.78	*0.141-4.355*	-	-
PeCBz	2.57	*1.069-6.164*	0.44	*0.094-2.062*	1.17	*0.263-5.190*	-	-
HCB	27.20	*9.374-78.930*	0.29	*0.059-1.432*	3.33	*0.964-11.520*	-	-
∑CBz	32.28	*11.780-88.471*	8.41	*3.525-20.049*	97.49	*27.118-350.460*	-	-
Hexachlorocyclohexanes (HCHs)								
α-HCH	9.71	*2.164-43.593*	21.72	*7.576-62.246*	7.61	*2.436-23.747*	-	-
β-HCH	-	-	3.66	*1.455-9.206*	0.59	*0.196-1.748*	-	-
γ-HCH	4.08	*0.997-16.669*	4.76	-	1.23	*0.340-4.469*	-	-
∑HCHs	13.84	*3.160-60.644*	27.21	*10.114-73.225*	9.12	*2.934-28.357*	-	-

	LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		MACRO ALGAE (*F. gardneri*) **(tissue)** (*n* =11)		SEDIMENTS **(*n* ponar grabs)** (*n* =12)		CAPELIN (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Dichlordiphenyl trichloroethanes (DDTs)								
p,p-DDT	0.63	*0.151-2.607*	-	-	2.15	*0.383-12.049*	-	-
o,p-DDT	0.51	*0.142-1.850*	-	-	1.65	*0.215-12.612*	-	-
p,p-DDE	0.46	*0.131-1.637*	0.93	*0.116-7.454*	1.99	*0.367-10.842*	-	-
o,p-DDE	0.04	*0.012-0.122*	2.19	-	0.46	*0.033-6.335*	-	-
p,p-DDD	0.24	*0.059-0.951*	0.22	-	0.71	*0.158-3.233*	-	-
o,p-DDD	0.14	*0.035-0.563*	-	-	1.21	-	-	-
SUM DDTs	1.69	*0.414-6.866*	1.31	*0.101-16.906*	3.41	*0.563-20.629*	-	-
Cyclodienes								
aldrin	-	-	2.91	-	25.25	-	-	-
heptachlor	0.01	*0.003-0.011*	0.02	-	0.20	-	-	-
heptachlor epoxide	1.46	*0.582-3.676*	0.61	*0.200-1.892*	-	-	-	-
octachlorostyrene	0.09	*0.025-0.310*	-	-	0.08	*0.017-0.423*	-	-
trans-chlordane	-	-	3.22	-	37.47	-	-	-
cis-chlordane	0.23	*0.064-0.797*	-	-	1.03	*0.317-3.377*	-	-
trans-nonachlor	0.39	*0.089-1.745*	0.53	*0.018-15.573*	1.88	*0.297-11.911*	-	-

	LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		MACRO ALGAE (*F. gardneri*) **(tissue)** (*n* =11)		SEDIMENTS **(*n* ponar grabs)** (*n* =12)		CAPELIN (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
cis-nonachlor	0.24	*0.068-0.822*	0.04	-	0.37	*0.096-1.451*	-	-
oxychlordane	-	-	1.92	-	9.38	-	-	-
α-endosulfan	-	-	-	-	0.16	-	-	-
β-endosulfan	0.03	-	-	-	0.33	-	-	-
endosulfan sulfate	-	-	-	-	0.16	-	-	-
dieldrin	1.00	*0.321-3.117*	0.77	*0.254-2.323*	1.94	*0.702-5.362*	-	-
methoxychlor	-	-	-	-	-	-	-	-
mirex	0.03	*0.007-0.088*	0.39		0.32	*0.042-2.492*	-	-
∑Chlordanes	0.86	*0.208-3.542*	0.83	*0.021-32.549*	3.16	*0.501-19.943*	-	-

Appendix 6 continued.

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Chlorobenzenes (CBz)								
1,3,5 TriCBz	0.20	*0.054-0.747*	0.36	-	0.16	*0.016-1.579*	0.15	*0.060-0.378*
1,2,4 TriCBz	3.08	*1.205-7.858*	4.83	*1.274-18.300*	2.35	*0.601-9.188*	5.53	*2.227-13.718*
1,2,3 TriCBz	0.52	*0.184-1.486*	0.44	*0.071-2.757*	0.13	*0.021-0.842*	0.11	*0.036-0.344*
1,2,3,5/1,2,4,5 TeCBz	0.48	*0.193-1.197*	1.16	*0.167-8.059*	0.10	*0.025-0.439*	0.10	*0.027-0.351*
1,2,3,4 TeCBz	0.65	*0.259-1.627*	0.20	*0.032-1.243*	0.26	*0.064-1.028*	0.26	*0.074-0.885*
PeCBz	0.89	*0.400-2.001*	2.62	*0.543-12.635*	1.47	*0.424-5.130*	0.37	*0.114-1.204*
HCB	22.32	*8.280-60.164*	13.96	*5.107-38.173*	35.43	*11.870-105.754*	1.06	*0.408-2.749*
∑CBz	27.64	*11.296-67.623*	25.38	*10.471-61.513*	39.73	*13.449-117.368*	7.53	*3.077-18.418*
Hexachlorocyclohexanes (HCHs)								
α-HCH	14.46	*4.675-44.724*	18.2	*8.095-40.853*	14.91	*4.803-46.306*	5.647	*2.348-13.583*
β-HCH	0.84	*0.214-3.267*	1.25	*0.545-2.882*	0.47	*0.127-1.762*	3.660	-
γ-HCH	2.47	*0.800-7.601*	3.41	*1.490-7.814*	3.24	*1.000-10.473*	1.041	-
∑ HCHs	9.84	*1.532-63.151*	12.23	*1.704-87.702*	18.54	*5.976-57.540*	5.835	*2.273-14.980*
Dichlordiphenyltrichloroethanes (DDTs)								
p,p-DDT	2.84	*0.693-11.647*	2.54	*0.987-6.535*	6.86	*2.230-21.096*	-	
o,p-DDT	0.86	*0.218-3.37*	-	-	4.66	*1.460-14.864*	-	
p,p-DDE	42.14	*10.045-176.8*	24.63	*6.252-97.051*	51.12	*16.718-156.30*	1.110	*0.461-2.675*
o,p-DDE	0.15	*0.049-0.438*	-	-	0.13	*0.018-0.932*	-	-

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
p,p-DDD	4.39	*1.047-18.426*	1.04	*0.420-2.563*	8.05	*2.599-24.934*	-	-
o,p-DDD	0.92	*0.325-2.583*	0.52	-	2.68	*0.922-7.800*	-	-
∑DDTs	50.08	*12.1-206.2*	26.6836	*6.461-110.204*	73.92	*24.458-223.398*	1.110	*0.461-2.675*
Cyclodienes								
aldrin	-	-	-	-	0.07	*0.026-0.198*	-	-
heptachlor	0.02	-	-	-	0.03	-	-	-
heptachlor epoxide	4.85	*1.745-13.502*	3.89	*1.220-12.376*	-	-	-	-
octachlorostyrene	2.00	*0.448-8.946*	0.46	*0.184-1.170*	1.51	*0.469-4.865*	-	-
trans-chlordane	-	-	-	-	-	-	-	-
cis-chlordane	5.84	*1.703-20.050*	4.82	*1.631-14.244*	22.63	*6.317-81.030*	0.313	-
trans-nonachlor	12.80	*3.792-43.187*	24.42	*10.249-58.176*	53.60	*14.500-198.116*	1.678	*0.720-3.914*
cis-nonachlor	8.08	*2.018-32.371*	6.12	*1.761-21.243*	15.13	*5.108-44.841*	-	-
oxychlordane	8.58	*2.216-33.177*	5.74	*0.738-44.645*	-	-	0.168	*0.072-0.391*
α-endosulfan	-	-	-	-	0.41	-	-	-
β-endosulfan	2.91	*1.266-6.692*	-	-	0.85	-	-	-
endosulfan sulfate	-	-	-	-	0.18	-	-	-
dieldrin	8.74	*3.331-22.929*	8.45	*2.662-26.802*	15.92	*4.873-51.993*	2.303	*0.895-5.923*
methoxychlor	-	-	-	-	-	-	-	-

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
mirex	0.95	*0.240-3.757*	1.02	*0.376-2.759*	0.84	*0.276-2.559*	0.196	*0.085-0.450*
∑Chlordanes	21.48	*5.837-79.034*	20.32	*2.927-141.026*	29.30	*5.587-153.688*	1.764	*0.794-3.917*

Appendix 6 continued.

	MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (~1 year) (*D. leucas*) (Blubber) (*n* = 9))	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Chlorobenzenes (CBz)								
1,3,5 TriCBz	0.20	*0.041-1.017*	0.18	*0.042-0.802*	0.16	*0.018-1.443*	0.24	*0.056-1.031*
1,2,4 TriCBz	4.56	*1.201-17.310*	6.58	*2.007-21.603*	6.85	*1.130-41.572*	12.01	*3.377-42.711*
1,2,3 TriCBz	0.21	*0.054-0.808*	0.24	*0.071-0.789*	0.37	*0.095-1.452*	0.26	*0.076-0.910*
1,2,3,5/1,2,4,5 TeCBz	2.69	*0.953-7.594*	1.23	*0.343-4.401*	1.88	*0.582-6.053*	4.67	*0.997-21.927*
1,2,3,4 TeCBz	2.67	*0.846-8.403*	2.18	*0.582-8.180*	3.81	*1.421-10.190*	3.43	*0.788-14.903*
PeCBz	17.50	*5.604-54.619*	7.74	*1.993-30.084*	12.71	*4.271-37.844*	25.32	*6.151-104.252*
HCB	345.52	*133.31-895.55*	93.29	*24.34-357.55*	113.51	*39.344-327.512*	430.93	*76.26-2,434.99*
∑CBz	376.97	*148.62-956.15*	111.70	*30.11-414.32*	141.67	*49.315-406.99*	486.92	*90.18-2,629.03*
Hexachlorocyclohexanes (HCHs)								
α-HCH	34.11	*4.571-254.515*	57.50	*15.75-209.81*	76.01	*23.931-241.413*	186.56	*56.290-618.304*
β-HCH	42.26	*14.82-120.50*	15.55	*4.75-50.821*	12.86	*4.203-39.317*	49.08	*8.777-274.463*
γ-HCH	30.75	*8.921-105.962*	16.42	*5.29-50.93*	16.93	*5.542-51.718*	62.67	*13.329-294.682*
∑HCHs	119.32	*32.53-437.57*	95.20	*33.31-272.00*	106.03	*33.952-331.122*	307.10	*78.712-1,198.186*
Dichlordiphenyltrichloroethanes (DDTs)								
p,p-DDT	428.16	*97.11-1,887.7*	75.45	*17.32-328.55*	39.75	*10.491-150.65*	88.44	*9.836-795.11*

	MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (~1 year) (*D. leucas*) (Blubber) (*n* = 9))	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
o,p-DDT	468.05	*121.3-1,805.9*	65.01	*14.31-295.16*	29.88	*6.870-129.967*	178.78	*14.37-2,224.18*
p,p-DDE	1702.04	*430.2-6,733.7*	305.71	*65.57-1,425.2*	171.55	*44.373-663.203*	505.86	*56.625-4,519.13*
o,p-DDE	3.68	*0.512-26.38*	0.60	*0.048-7.534*	5.46	*1.635-18.262*	3.22	*0.103-100.634*
p,p-DDD	294.42	*66.41-1,305.2*	54.12	*14.292-204.94*	31.44	*10.078-98.056*	114.09	*16.261-800.424*
o,p-DDD	69.09	*11.874-401.98*	10.91	*3.069-38.790*	10.27	*3.040-34.690*	31.81	*3.978-254.329*
∑DDTs	2521.53	*695.0-9,147.8*	519.39	*119.5-2,255.9*	281.53	*74.101-1,069.651*	1032.15	*137.8-7,728.104*
Cyclodienes								
aldrin	0.34	*0.041-2.809*	0.08	*0.010-0.613*	0.08	*0.028-0.220*	1.06	*0.291-3.874*
heptachlor	1.00	*0.214-4.720*	0.07	*0.019-0.269*	0.07		0.50	*0.075-3.375*
heptachlor epoxide	200.45	*70.58-569.21*	-	-	-	-	203.47	-
octachlorostyrene	4.42	*1.499-13.059*	2.43	*0.728-8.127*	1.99	*0.744-5.313*	5.12	*1.192-21.955*
trans-chlordane	7.75	-	11.55	*3.202-41.639*	-	-	13.73	*6.461-29.163*
cis-chlordane	168.92	*50.719-562.56*	57.87	*16.113-207.85*	-	-	48.46	*22.420-104.727*
trans-nonachlor	871.84	*289.7-2,623.7*	306.91	*82.6-1,139.91*	166.93	*47.0592.4*	216.37	*96.227-486.537*
cis-nonachlor	119.30	*39.1-363.7*	56.45	*17.9-177.6*	38.25	*12.2-119.6*	48.84	*19.508-122.298*
oxychlordane	732.37	*281.0-1,908.7*	-	-	-	-	334.84	-
α-endosulfan	-	-	-	-	-	-	-	-
β-endosulfan	12.56	*4.504-35.053*	4.87	*1.232-19.213*	2.75	*0.651-11.655*	4.13	*1.882-9.048*

	MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (~1 year) (*D. leucas*) (Blubber) (*n* = 9))	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
endosulfan sulfate	0.86	*0.212-3.514*	0.58	*0.113-3.000*	0.31	*0.127-0.735*	0.35	*0.035-3.595*
dieldrin	17.01	*0.862-335.570*	23.98	*0.596-964.243*	110.75	*34.995-350.513*	288.36	*44.6-1,862.5*
methoxychlor	-	-	0.16	-	-	-	-	-
mirex	23.79	*6.519-86.831*	10.30	*2.323-45.676*	2.37	*0.586-9.575*	4.86	*0.54-43.661*
∑Chlordanes	234.35	*10.0-5,472.2*	407.73	*114.9-1,446.8*	207.77	*60.2-716.7*	84.44	*7.05-1,011.1*

Appendix 6 continued.

	MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Chlorobenzenes (CBz)								
1,3,5 TriCBz	1.74	-	0.82	*0.236-2.836*	0.19	-	-	-
1,2,4 TriCBz	36.03	-	16.31	*3.816-69.706*	18.92	*6.547-54.685*	10.88	*4.993-23.701*
1,2,3 TriCBz	1.33	-	2.03	*0.429-9.628*	0.73	*0.182-2.931*	2.34	*0.723-7.548*
1,2,3,5/1,2,4,5 TeCBz	5.96	*1.725-20.608*	4.86	*1.552-15.242*	1.75	*0.394-7.812*	2.25	*0.751-6.745*
1,2,3,4 TeCBz	5.91	*1.254-27.872*	5.71	*1.714-19.017*	3.92	*0.628-24.506*	6.91	*3.172-15.056*
PeCBz	36.66	*9.922-135.431*	23.81	*8.031-70.580*	15.01	*5.117-44.043*	18.82	*8.059-43.927*
HCB	697.60	*170.8-2,849.1*	537.51	*186.5-1,548.5*	207.83	*70.688-611.021*	395.36	*186.300-839.005*
∑CBz	755.46	*188.1-3,033.4*	600.90	*208.7-1,729.6*	184.00	*44.362-763.162*	436.76	*203.91-935.48*
HCHs								
α-HCH	58.19	*7.382-458.613*	78.25	*24.1-253.8*	36.54	*9.999-133.56*	105.19	*29.604-373.76*
β-HCH	33.85	*4.915-233.18*	35.07	*10.29-119.4*	8.34	*2.366-29.41*	13.09	*4.722-36.27*
γ-HCH	17.29	*2.260-132.2*	33.11	*5.560-197.1*	10.55	*3.266-34.07*	4.50	*1.466-13.847*
∑HCHs	92.89	*12.021-717.7*	163.85	*57.44-467.3*	53.88	*17.492-165.9*	122.98	*35.732-423.2*
DDTs								
p,p-DDT	54.53	*3.352-887.189*	21.63	*3.511-133.285*	11.54	*4.263-31.250*	-	-
o,p-DDT	19.87	*3.049-129.519*	28.34	*3.789-212.016*	10.63	-	-	-
p,p-DDE	1029.81	*266.9-3,973.1*	1113.02	*208.2-5,948.2*	125.51	*30.731-512.600*	201.90	*38.881-1,048.419*
o,p-DDE	1.75	*0.295-10.425*	10.88	*0.801-147.758*	0.91	-	0.44	*0.071-2.672*
p,p-DDD	95.93	*23.036-399.4*	142.58	*41.505-489.7*	19.57	*4.306-88.979*	70.50	*13.323-373.047*
o,p-DDD	54.52	*17.329-171.5*	113.09	*27.34-467.7*	7.29	*1.826-29.1*	33.33	*8.189-135.6*

	MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
∑DDTs	1227.27	*306.9-4,906.8*	1439.12	*292.3-7,085.2*	165.95	*43.064-639.471*	306.63	*60.36-1,557.5*
Cyclodienes								
aldrin	-	-	0.88	*0.24-3.1*	-	-	1.32	-
heptachlor	-	-	0.81	*0.21-3.6*	-	-	0.31	-
heptachlor epoxide	-	-	180.97	*67.10-488.0*	-	-	-	-
octachlorostyrene	3.11	*0.565-17.127*	3.45	*1.33-8.9*	1.76	*0.739-4.184*	5.09	*1.764-14.661*
trans-chlordane			5.30	*2.20-12.7*	-	-	-	-
cis-chlordane	42.19	*15.494-114.8*	118.93	*51.57-274.2*	-	-	-	-
trans-nonachlor	298.85	*128.190-696.6*	558.13	*243.7-1,278.0*				
cis-nonachlor	57.31	*10.318-318.2*	68.87	*24.90-190.49*	21.60	*6.715-69.452*	60.16	*13.237-273.382*
oxychlordane	-	-	378.79	*171.1-838.24*	-	-	-	-
α-endosulfan	-	-	-	-	-	-	-	-
β-endosulfan	-	-	-	-	-	-	-	-
endosulfan sulfate	-	-	0.59	*0.24-1.3*	-	-	-	-
dieldrin	371.02	*76.51-1,798.2*	383.76	*21.39-6,884.1*	121.51	*41.473-356.021*	-	-
methoxychlor	-	-	-	-	-	-	-	-
mirex	8.80	*1.683-45.981*	16.20	*5.74-45.68*	3.41	*0.939-12.363*	15.71	*3.396-72.656*
∑Chlordanes	120.93	*15.84-923.0*	396.57	*63.2-2,485.4*	13.37	*1.877-95.295*	65.57	*14.929-288.023*

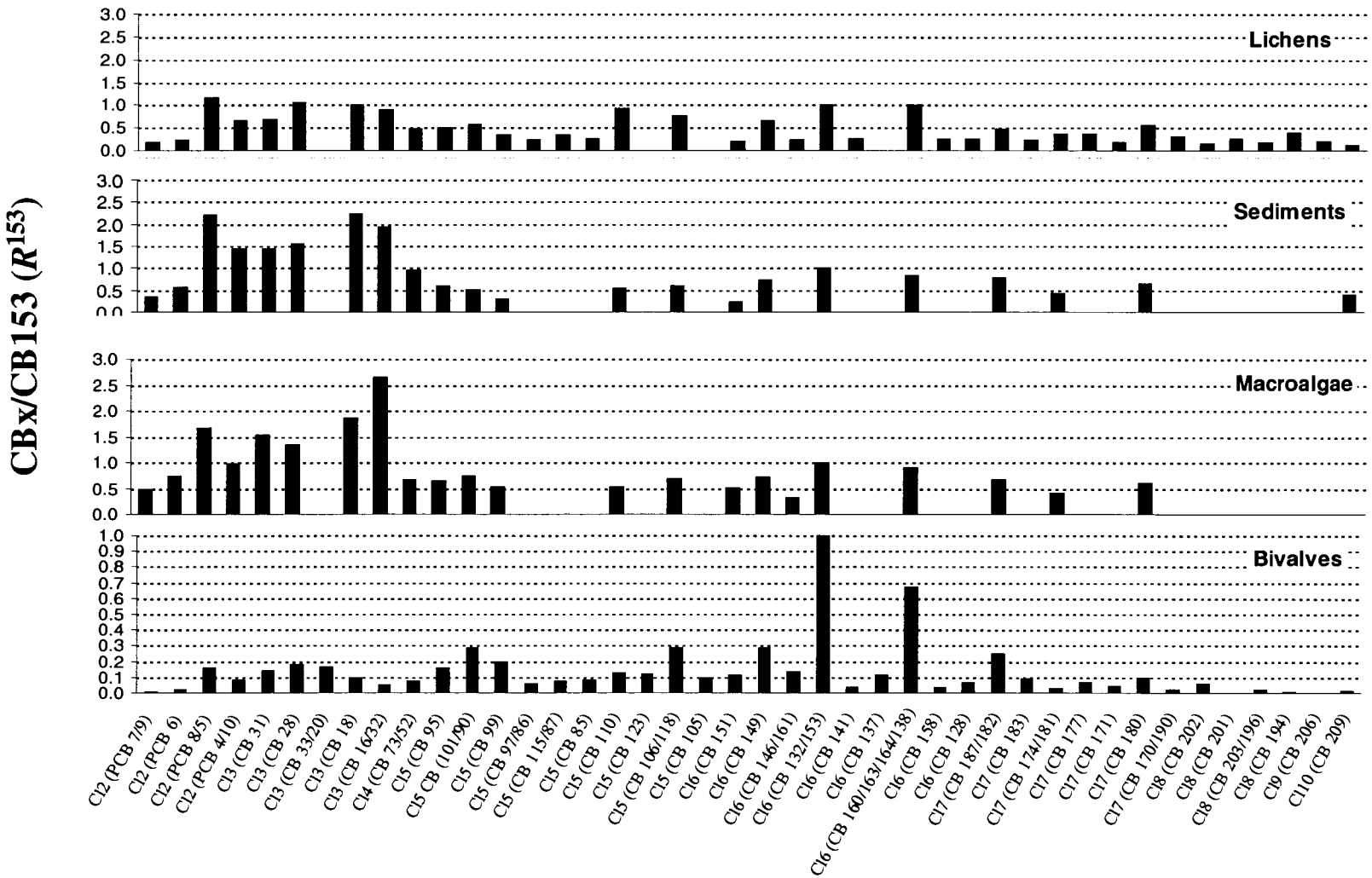
Appendix 6 continued.

	FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
Chlorobenzenes (CBz)								
1,3,5 TriCBz	0.17	*0.065-0.437*	0.20	*0.031-1.286*	-	-	-	-
1,2,4 TriCBz	10.20	*5.020-20.723*	6.75	*1.219-37.319*	-	-	-	-
1,2,3 TriCBz	0.21	*0.085-0.530*	0.22	*0.037-1.306*	17.47	-	-	-
1,2,3,5/1,2,4,5 TeCBz	8.05	*1.876-34.571*	6.10	*0.882-42.189*	4.09	*1.321-12.670*	1.04	*0.395-2.717*
1,2,3,4 TeCBz	0.91	*0.380-2.193*	1.65	*0.393-6.909*	13.27	*4.085-43.081*	0.96	*0.302-3.032*
PeCBz	6.12	*2.219-16.899*	8.60	*2.451-30.189*	1.65	*0.404-6.763*	2.82	*0.495-16.057*
HCB	14.18	*5.123-39.266*	31.02	*3.450-278.939*	98.53	*25.353-382.905*	79.59	*30.777-205.812*
∑CBz	*40.58*	*14.76-111.5*	78.11	*17.23-354.0*	62.09	*11.858-325.148*	83.89	*31.971-220.113*
Hexachlorocyclohexanes (HCHs)								
α-HCH	182.82	*48.08-695.0*	81.96	*6.50-1,032.62*				
β-HCH	12.88	*2.741-60.542*	21.88	*5.613-85.278*	19.11	*4.828-75.674*	10.16	*3.716-27.764*
γ-HCH	8.47	*2.380-30.137*	9.97	*1.845-53.857*	-	-	-	-
∑HCHs	204.32	*53.17-785.12*	145.33	*24.46-863.51*	19.11	*4.828-75.674*	10.16	*3.716-27.764*
Dichlordiphenyltrichloroethanes (DDTs)								
p,p-DDT	48.56	*4.943-477.102*	44.83	*9.574-209.945*	3.56	*1.357-9.357*	4.53	*1.953-10.527*
o,p-DDT	0.24	-	6.80	*0.03-1,558.21*	5.48	-	2.40	

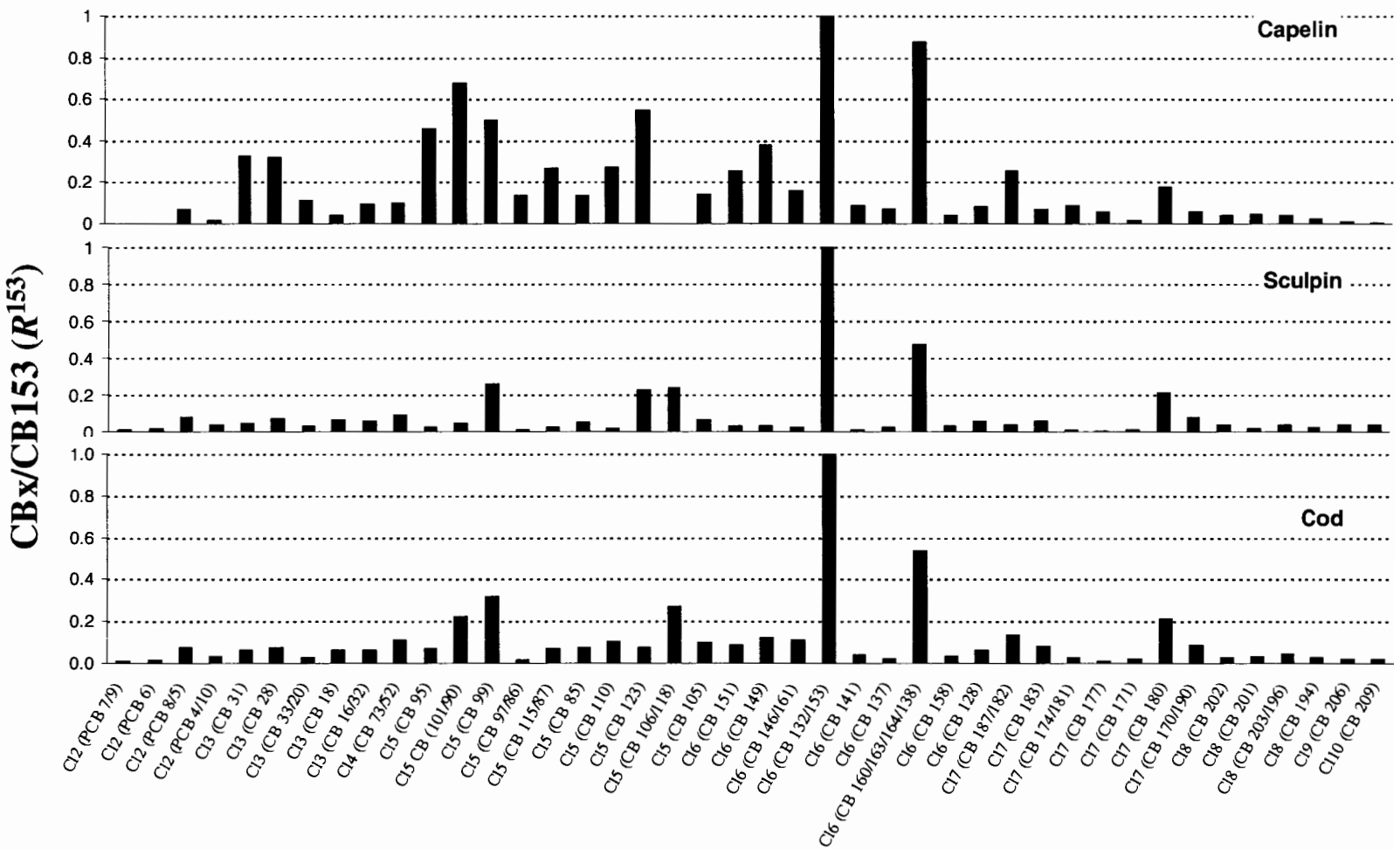
	FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
p,p-DDE	426.16	*37.9-4,789.5*	347.91	*147.1-822.4*	224.21	*55.325-908.639*	645.66	*126.979-3,283.008*
o,p-DDE	0.05	*0.015-0.169*	0.21	*0.010-4.505*	0.60	-	-	-
p,p-DDD	6.40	*1.155-35.423*	12.53	*1.430-109.737*	7.69	*1.673-35.339*	9.90	*3.022-32.464*
o,p-DDD	-	-	3.38	*0.114-100.514*	-	-	1.66	*0.681-4.065*
∑DDTs	482.55	*44.0-5,291.2*	413.31	*174.121-981.0*	233.31	*57.048-954.173*	667.80	*135.7-3,285.5*
Cyclodienes								
aldrin	-	-	0.18	*0.022-1.496*	0.47	*0.236-0.953*	0.70	-
heptachlor	0.38	*0.050-2.938*	0.36	*0.103-1.275*	0.07	-	-	-
heptachlor epoxide	-	-	90.03	-	-	-	-	-
octachlorostyrene	2.43	*0.398-14.867*	0.39	*0.006-23.527*	4.68	*1.425-15.383*	2.03	*0.690-5.996*
trans-chlordane	10.21	*2.100-49.632*	6.90	*2.267-20.988*	-	-	-	-
cis-chlordane	36.28	*3.318-396.737*	23.93	*8.928-64.130*	3.75	-	3.82	-
trans-nonachlor	84.29	*9.177-774.178*	84.52	*18.33-389.4*	33.09	*15.050-72.748*	20.10	*7.552-53.517*
cis-nonachlor	10.26	*1.581-66.582*	9.37	*1.916-45.775*	15.24	*3.051-76.159*	7.01	*2.337-21.056*
oxychlordane			268.36	-	173.34	*76.081-394.917*	693.35	*304 -1,579*
α-endosulfan			0.30	*0.101-0.874*	2.32	-	-	-
β-endosulfan	3.02	*0.269-33.913*	2.26	*0.624-8.179*	-	-	-	-
endosulfan sulfate	0.19		0.32	*0.100-1.011*	-	-	0.87	-
dieldrin	125.84	-	54.64	*15.06-198.15*	194.36	*71.098-531.33*	109.38	*42.42-281.96*

	FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6)		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***	**GM**	***(95% CL)***
methoxychlor	0.06	-	-	-	-	-		
mirex	4.45	*0.406-48.715*	3.46	*0.773-15.518*	18.95	*4.264-84.190*	11.02	*3.424-35.473*
∑Chlordanes	145.55	*16.40-1,291.3*	69.82	*7.058-690.644*	25.02	*4.089-153.030*	7.68	*0.723-81.542*

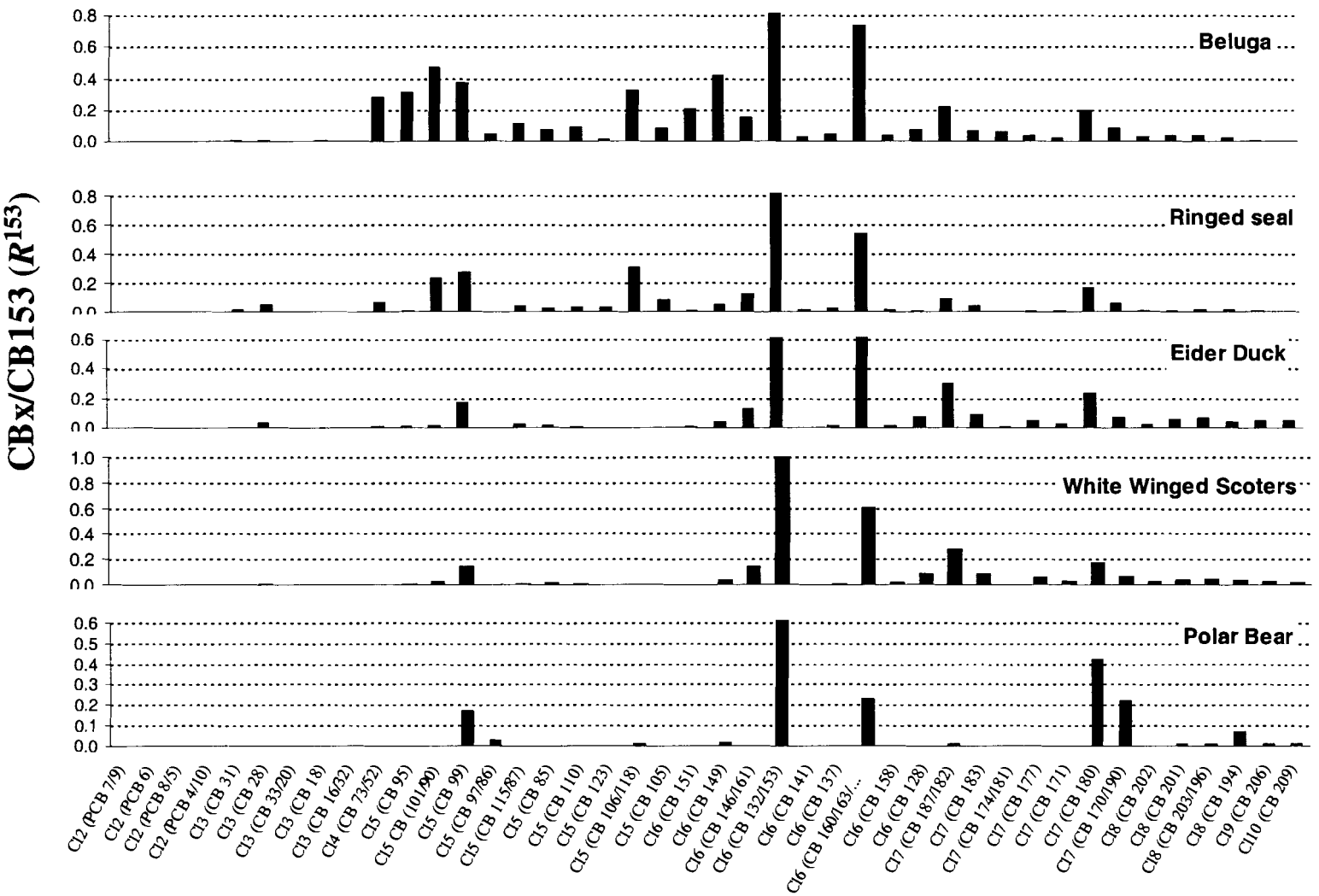
Appendix 7 Plots of relative ratios of CBx/CB153 (R^{153}) values of selected CB congeners in various biota from E. Hudson Bay.



Appendix 7 continued



Appendix 7 continued



Appendix 8 Calculated Biomagnification factors (BMFs), Metabolic Index (MI) and Biodilution factors (BDFs) for POPs in various organisms of the E Hudson Bay Marine Food web.

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_2 (PCB 7/9)	Group 3	0.29	1.11	12.8	0.31	1.05	11.3	-	-	-
Cl_2 (PCB 6)	Group 3	0.27	1.14	13.7	0.28	1.10	12.6	-	-	-
Cl_2 (PCB 8/5)	Group 3	0.54	0.84	6.92	0.47	0.87	7.40	0.58	2.26	183.7
Cl_2 (PCB 4/10)	Group 4	0.49	0.87	7.49	0.38	0.97	9.25	1.19	1.95	89.3
Cl_3 (PCB 23/34)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (PCB 29)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (PCB 26)	Group 3	0.38	0.99	9.80	0.38	0.96	9.15	9.31	1.06	11.4
Cl_3 (CB 25)	Group 3	0.28	1.13	13.4	0.32	1.04	11.1	-	-	-
Cl_3 (CB 31)	Group 3	0.34	1.04	10.9	0.45	0.89	7.82	-	-	-
Cl_3 (CB 28)	Group 3	0.60	0.79	6.13	0.59	0.78	5.98	8.20	1.11	12.9
Cl_3 (CB 21)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (CB 33/20)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (CB 19)	Group 4	0.21	1.25	17.67	0.24	1.16	14.3	-	-	-
Cl_3 (CB30)	Group 4	-	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	Group 4	0.40	0.97	9.37	0.36	0.98	9.62	0.69	2.19	154
Cl_3 (CB 17)	Group 4	0.28	1.12	13.19	0.25	1.15	13.9	1.08	1.99	97.9
Cl_3 (CB 27/24)	Group 4	-	-	-	-	-	-	-	-	-
Cl_3 (CB 16/32)	Group 4	0.26	1.16	14.5	0.26	1.13	13.61	2.61	1.61	40.6
Cl_3 (CB 22)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 54)	Group 5	-	-	-	-	-	-	-	-	-

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 50)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 53)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 51)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 45)	Group 5	-	-	-	-	-	-	3.85	1.44	27. 6
Cl_4 (CB 46)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 73/52)	Group 4	1.52	0.39	2.45	1.78	0.29	1.97	3.72	1.45	28.5
Cl_4 (CB 69)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	Group 4	0.42	0.94	8.79	0.99	0.55	3.53	3.65	1.46	29.0
Cl_4 (CB 43)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	Group 4	-	-	-	-	-	-	18.8	0.75	5.65
Cl_4 (CB 65)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	Group 4	0.36	1.01	10.3	0.58	0.78	6.04	3.46	1.49	30.6
Cl_4 (CB 59/42)	Group 4	0.22	1.22	16.5	0.42	0.92	8.36	5.04	1.32	21.0
Cl_4 (CB 72)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	Group 4	0.25	1.18	14.9	0.49	0.85	7.14	7.74	1.14	13.7
Cl_4 (CB 68)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 40)	Group 4	-	-	-	-	-	-	13.9	0.88	7.61
Cl_4 (CB 57)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 67)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 58)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	Group 3	-	-	-	-	-	-	-	-	-

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 61/74)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 70/76)	Group 3	1.30	0.45	2.85	2.13	0.22	1.64	-	-	-
Cl_4 (CB 66)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 55)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 60/56)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 104)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 103)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 100)	Group 2	-	-	-	-	-	-	-	-	-
Cl_5 (CB 94)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 95)	Group 5	0.46	0.91	8.09	1.18	0.47	2.97	2.10	1.70	50.53
Cl_5 (CB 102/93)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 98)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 88)	Group 5	-	-	-	-	-	-	46.8	0.36	2.26
Cl_5 (CB 91)	Group 5	-	-	-	-	-	-	2.73	1.59	38.9
Cl_5 (CB 121)	Group 1	-	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 CB (101/90)	Group 4	0.68	0.74	5.47	3.20	0.04	1.09	2.88	1.57	36.8
Cl_5 (CB 89)	Group 5	-	-	-	-	-	-	2.64	1.60	40.2
Cl_5 (CB 99)	Group 2	5.11	-0.14	0.73	5.88	-0.23	0.60	38.3	0.44	2.76
Cl_5 (CB 113)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 119)	Group 2	-	-	-	-	-	-	15.1	0.85	7.01

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_5 (CB 112)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 109/83)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 97/86)	Group 4	-	-	-	-	-	-	0.79	2.13	133
Cl_5 (CB 116/125/117)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 115/87)	Group 2	-	-	-	-	-	-	15.1	0.85	7.03
Cl_5 (CB 111)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 85)	Group 2	-	-	-	-	-	-	8.31	1.11	12.8
Cl_5 (CB 120)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 110)	Group 4	0.44	0.93	8.44	2.00	0.24	1.75	3.50	1.48	30.3
Cl_5 (CB 82)	Group 4	-	-	-	-	-	-	2.74	1.59	38.6
Cl_5 (CB 124)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 108/107)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 123)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 106/118)	Group 3	4.09	-0.04	0.91	4.31	-0.09	0.81	-	-	-
Cl_5 (CB 114)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 122)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 105)	Group 3	-	-	-	-	-	-	-	-	-
Cl_6 (CB 155)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 150)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 152)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 145)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 148)	Group 1	-	-	-	-	-	-	3.45	1.49	30.7

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 136)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 154)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 151)	Group 5	0.76	0.69	4.90	1.88	0.27	1.86	1.64	1.81	64.5
Cl_6 (CB 135/144)	Group 5	-	-	-	-	-	-	1.57	1.83	67.4
Cl_6 (CB 147)	Group 2	-	-	-	-	-	-	82.2	0.11	1.28
Cl_6 (CB 149)	Group 5	0.54	0.84	6.87	1.85	0.28	1.89	6.04	1.24	17.6
Cl_6 (CB 139/140)	Group 2	-	-	-	-	-	-	-	-	-
Cl_6 CB-143/134	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 142/131)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 133)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 146/161)	Group 1	0.93	0.60	3.99	3.79	-0.03	0.92	42.76	0.39	2.47
Cl_6 (CB 165)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 132/153)	Group I	11.2	-0.48	0.33	10.60	-0.48	0.33	44.7	0.37	2.36
Cl_6 (CB 168)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 141)	Group 4	-	-	-	-	-	-	3.20	1.52	33.1
Cl_6 (CB 137)	Group 2	-	-	-	-	-	-	5.02	1.32	21.1
Cl_6 (CB 130)	Group 2	-	-	-	-	-	-	64.2	0.22	1.65
Cl_6 (CB 160/163/164/138)	Group 2	5.82	-0.20	0.64	6.18	-0.25	0.57	41.2	0.41	2.57
Cl_6 (CB 158)	Group 2	-	-	-	-	-	-	23.7	0.65	4.47
Cl_6 (CB 129)	Group 4	-	-	-	-	-	-	-	-	-
Cl_6 (CB 166)	Group 2	-	-	-	-	-	-	-	-	-
Cl_6 (CB 159)	Group 3	-	-	-	-	-	-	-	-	-

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 162)	Group 4	6.57	-0.25	0.56	7.84	-0.35	0.45	-	-	-
Cl_6 (CB 128)	Group 2	-	-	-	-	-	-	49.7	0.33	2.13
Cl_6 (CB 167)	Group 3	-	-	-	-	-	-	-	-	-
Cl_6 (CB 156)	Group 3	-	-	-	-	-	-	-	-	-
Cl_6 (CB 157)	Group 3	-	-	-	-	-	-	-	-	-
Cl7 (CB 188)	Group 1	-	-	-	-	-	-	34.0	0.49	3.11
Cl_7 (CB 184)	Group 1	-	-	-	-	-	-	6.50	1.21	16.3
Cl_7 (CB 179)	Group 5	-	-	-	-	-	-	1.71	1.79	61.8
Cl_7 (CB 176)	Group 5	-	-	-	-	-	-	1.90	1.75	55.7
Cl_7 (CB 186)	Group 5	-	-	-	-	-	-	-	-	-
Cl_7 (CB 178)	Group 1	-	-	-	-	-	-	10.39	1.01	10.2
Cl_7 (CB 175)	Group 1	-	-	-	-	-	-	20.4	0.72	5.21
Cl_7 (CB 187/182)	Group I	0.60	0.79	6.20	2.06	0.23	1.70	54.5	0.29	1.94
Cl_7 (CB 183)	Group 1	-	-	-	-	-	-	42.6	0.40	2.48
Cl_7 (CB 185)	Group 5	-	-	-	-	-	-	-	-	-
Cl_7 (CB 174/181)	Group 5	0.33	1.05	11.2	0.67	0.72	5.22	10.1	1.02	10.5
Cl_7 (CB 177)	Group 2	-	-	-	-	-	-	33.42	0.50	3.17
Cl_7 (CB 171)	Group 2	-	-	-	-	-	-	22.06	0.68	4.80
Cl_7 (CB 173)	Group 2	-	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	Group 1	-	-	-	-	-	-	-	-	-
Cl_7 (CB 180)	Group 1	3.71	0.00	1.00	3.50	-	1.00	106	-	-
Cl_7 (CB 193)	Group 1	0.21	1.24	17.53	0.37	0.98	9.57	478	-0.65	0.22

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_7 (CB 191)	Group 1	-	-	-	0.22	1.19	15.6	-	-	-
Cl_7 (CB 170/190)	Group 2	2.04	0.26	1.82	2.14	0.21	1.64	120	-0.05	0.88
Cl_7 (CB 189)	Group 3	-	-	-	0.10	1.54	34.5	-	-	-
Cl_8 (CB 202)	Group 1	0.97	0.58	3.82	0.76	0.67	4.62	18.3	0.76	5.78
Cl_8 (CB 200)	Group I	0.30	1.10	12.48	0.63	0.75	5.58	27.8	0.58	3.81
Cl_8 (CB 204)	Group I	-	-	-	-	-	-	-	-	-
Cl_8 (CB 197)	Group I	0.33	1.05	11.34	0.31	1.06	11.42	43.3	0.39	2.45
Cl_8 (CB 199)	Group 5	-	-	-	-	-	-	17.9	0.77	5.91
Cl_8 (CB 198)	Group I	0.55	0.83	6.77	0.87	0.61	4.03	4.43	1.38	23.9
Cl_8 (CB 201)	Group I	0.44	0.92	8.40	0.90	0.59	3.89	1280	-1.08	0.082
Cl_8 (CB 203/196)	Group I	1.11	0.52	3.34	1.23	0.45	2.84	134	-0.10	0.79
Cl_8 (CB 195)	Group 2	-	-	-	0.23	1.18	14.9	-	-	-
Cl_8 (CB 194)	Group I	0.70	0.72	5.29	0.71	0.69	4.90	185	-0.24	0.57
Cl_8 (CB 205)	Group I	-	-	-	0.10	1.55	35.1	-	-	-
Cl_9 (CB 208)	Group 1	0.21	1.25	17.60	0.47	0.87	7.49	-	-	-
Cl_9 (CB 207)	Group I	0.46	0.91	8.09	0.34	1.01	10.2	-	-	-
Cl_9 (CB 206)	Group I	1.01	0.56	3.67	0.53	0.82	6.63	-	-	-
Cl_{10} (CB 209)	Group I	1.07	0.54	3.46	0.62	0.75	5.68	202	-0.28	0.52

	Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
	BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Chlorobenzenes (CBz)									
1,3,5 TriCBz	-	-	-	-	-	-	-	-	-
1,2,4 TriCBz	-	-	-	-	-	-	-	-	-
1,2,3 TriCBz	-	-	-	-	-	-	-	-	-
1,2,3,5/1,2,4,5 TeCBz	-	-	-	-	-	-	42.4	0.40	2.50
1,2,3,4 TeCBz	-	-	-	-	-	-	51.7	0.31	2.05
PeCBz	-	-	-	-	-	-	4.47	1.38	23.7
HCB	1.75	0.33	2.12	2.79	0.10	1.25	92.9	0.06	1.14
Hexachlorocyclohexanes (HCHs)									
α-HCH	0.24	1.18	15.3	0.19	1.26	18.15	0.29	2.56	365
β-HCH	0.18	1.32	20.7	0.12	1.47	29.3	24.0	0.64	4.41
γ-HCH	0.49	0.88	7.60	0.35	1.00	9.94	0.79	2.13	134
Dichlordiphenyltrichloroethanes (DDTs)									
p,p-DDT	0.08	1.67	46.7	0.09	1.60	39.41	-	-	-
o,p-DDT	-	-	-	-	-	-	-	-	-
p,p-DDE	6.16	-0.22	0.60	10.5	-0.48	0.33	202	-0.28	0.53
o,p-DDE	-	-	-	0.04	1.98	95.4	-	-	-
p,p-DDD	-	-	-	-	-	-	-	-	-
o,p-DDD	-	-	-	-	-	-	-	-	-

		Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
		BMF_{MAX} =	3.7		BMF_{MAX} =	3.5		BMF_{MAX} =	106	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cyclodienes										
aldrin		-	-	-	-	-	-	-	-	-
heptachlor		-	-	-	-	3.17	1470	-	-	-
heptachlor epoxide		-	-	-	-	-	-	-	-	-
***trans*-chlordane**		-	-	-	-	-	-	-	-	-
***cis*-chlordane**		0.25	1.16	14.6	0.31	1.06	11.38	12.0	0.95	8.83
***trans*-nonachlor**		-	-	-	-	-	-	19.7	0.73	5.38
***cis*-nonachlor**		0.47	0.90	7.87	0.62	0.75	5.63	-	-	-
oxychlordane		0.96	0.59	3.87	1.43	0.39	2.45	-	-	-
α-endosulfan		-	-	-	-	-	-	-	-	-
β-endosulfan		-	-	-	-	-	-	-	-	-
endosulfan sulfate		-	-	-	-	-	-	-	-	-
dieldrin		0.65	0.76	5.70	0.67	0.72	5.20	84.4	0.10	1.26
methoxychlor		-	-	-				-	-	-
mirex		2.04	0.26	1.82	1.90	0.27	1.84	-	-	-
octachlorostyrene		-	-	-	-	-	-	-	-	-

Appendix 8 continued.

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_2 (PCB 7/9)	Group 3	-	-	-	0.3	1.5	29.2	0.4	2.1	127
Cl_2 (PCB 6)	Group 3	0.70	2.96	906	0.4	1.4	25.6	0.4	2.1	122
Cl_2 (PCB 8/5)	Group 3	0.56	3.06	1130	0.4	1.4	24.8	0.4	2.1	122
Cl_2 (PCB 4/10)	Group 4	0.16	3.60	3950	0.3	1.6	39.6	0.3	2.2	164
Cl_3 (PCB 23/34)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (PCB 29)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (PCB 26)	Group 3	2.23	2.45	284	2.1	0.7	4.9	2.7	1.2	17.1
Cl_3 (CB 25)	Group 3				0.8	1.1	13.4	0.8	1.7	55.4
Cl_3 (CB 31)	Group 3				3.7	0.4	2.8	7.1	0.8	6.5
Cl_3 (CB 28)	Group 3	26.8	1.38	23.7	3.0	0.5	3.4	-	-	-
Cl_3 (CB 21)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (CB 33/20)	Group 3	-	-	-	1.5	0.8	6.9	2.1	1.3	21.3
Cl_3 (CB 19)	Group 4	-	-	-	0.4	1.4	25.3	2.0	1.4	22.9
Cl_3 (CB30)	Group 4	-	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	Group 4	0.94	2.83	673	2.2	0.7	4.7	6.3	0.9	7.3
Cl_3 (CB 17)	Group 4	1.07	2.77	591	1.9	0.7	5.4	4.5	1.0	10.2
Cl_3 (CB 27/24)	Group 4	0.12	3.73	5330	0.9	1.0	10.8	1.6	1.4	28.1
Cl_3 (CB 16/32)	Group 4	2.26	2.45	281	1.2	0.9	8.2	2.2	1.3	20.4
Cl_3 (CB 22)	Group 3	-	-	-	0.9	1.1	11.5	0.3	2.3	183
Cl_4 (CB 54)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	Group 5	-	-	-	-	-	-	-	-	-

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 53)	Group 4	-	-	-	3.1	0.5	3.3	32.7	0.1	1.4
Cl_4 (CB 51)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 45)	Group 5	2.30	2.44	275	5.2	0.3	1.9	26.1	0.2	1.7
Cl_4 (CB 46)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 73/52)	Group 4	7.04	1.96	90.2	15.6	-0.2	0.7	120	-0.4	0.4
Cl_4 (CB 69)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	Group 4	10.7	1.77	59.4	11.4	0.0	0.9	69.3	-0.2	0.7
Cl_4 (CB 43)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	Group 4	50.9	1.10	12.5	8.8	0.1	1.2	57.6	-0.1	0.8
Cl_4 (CB 65)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	Group 4	11.8	1.73	53.9	15.2	-0.2	0.7	74.1	-0.2	0.6
Cl_4 (CB 59/42)	Group 4	30.3	1.32	20.9	9.2	0.0	1.1	30.8	0.2	1.5
Cl_4 (CB 72)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	Group 4	67.3	0.98	9.44	0.7	1.2	14.1	1.3	1.6	36.5
Cl_4 (CB 68)	Group 3	-	-	-	18.4	-0.3	0.6	16.2	0.5	2.8
Cl_4 (CB 40)	Group 4	82.9	0.88	7.66	6.2	0.2	1.6	20.4	0.4	2.2
Cl_4 (CB 57)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 67)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 58)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	Group 3	-	-	-	1.1	1.0	9.5	1.3	1.5	34.0
Cl_4 (CB 61/74)	Group 3	-	-	-	8.1	0.1	1.3	42.7	0.0	1.1

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 70/76)	Group 3	-	-	-	5.8	0.2	1.7	10.1	0.7	4.5
Cl_4 (CB 66)	Group 3	-	-	-	7.4	0.1	1.4	25.2	0.3	1.8
Cl_4 (CB 55)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 60/56)	Group 3	-	-	-	3.9	0.4	2.6	5.6	0.9	8.2
Cl_5 (CB 104)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 103)	Group 5	15.9	1.60	39.8	3.2	0.5	3.2	21.1	0.3	2.2
Cl_5 (CB 100)	Group 2	20.8	1.49	30.6	2.1	0.7	4.8	12.3	0.6	3.7
Cl_5 (CB 94)	Group 5	-	-	-	0.6	1.2	16.5	2.0	1.4	23.2
Cl_5 (CB 95)	Group 5	15.41	1.62	41.2	25.7	-0.4	0.4	201	-0.6	0.2
Cl_5 (CB 102/93)	Group 5	-	-	-	-	-	-			
Cl_5 (CB 98)	Group 4	115	0.74	5.51	-	-	-			
Cl_5 (CB 88)	Group 5	73.9	0.93	8.58	2.0	0.7	5.1	11.3	0.6	4.1
Cl_5 (CB 91)	Group 5	19.9	1.50	31.8	14.1	-0.1	0.7	109.1	-0.4	0.4
Cl_5 (CB 121)	Group 1	-	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	Group 5	-	-	-	10.6	0.0	1.0	141	-0.5	0.3
Cl_5 CB (101/90)	Group 4	32.3	1.29	19.6	12.3	-0.1	0.8	99.4	-0.3	0.5
Cl_5 (CB 89)	Group 5	49.6	1.11	12.8	16.1	-0.2	0.6	110	-0.4	0.4
Cl_5 (CB 99)	Group 2	260	0.39	2.44	7.3	0.1	1.4	55.7	-0.1	0.8
Cl_5 (CB 113)	Group 4									
Cl_5 (CB 119)	Group 2	132	0.68	4.80	6.1	0.2	1.7	44.1	0.0	1.0
Cl_5 (CB 112)	Group 4	-	-	-	-	-	-	-	-	-

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_5 (CB 109/83)	Group 4	367	0.24	1.72	-	-	-	-	-	-
Cl_5 (CB 97/86)	Group 4	8.25	1.89	76.9	23.2	-0.4	0.4	118	-0.4	0.4
Cl_5 (CB 116/125/117)	Group 4	-	-	-	0.4	1.4	26.3	1.8	1.4	25.3
Cl_5 (CB 115/87)	Group 2	50.4	1.10	12.6	12.3	-0.1	0.8	76.7	-0.2	0.6
Cl_5 (CB 111)	Group 3									
Cl_5 (CB 85)	Group 2	79.6	0.90	7.97	7.3	0.1	1.4	45.5	0.0	1.0
Cl_5 (CB 120)	Group 3									
Cl_5 (CB 110)	Group 4	15.3	1.62	41.5	9.0	0.1	1.1	42.1	0.0	1.1
Cl_5 (CB 82)	Group 4	270	0.37	2.35	12.6	-0.1	0.8	60.9	-0.1	0.7
Cl_5 (CB 124)	Group 3	-	-	-	2.0	0.7	5.1	15.8	0.5	2.9
Cl_5 (CB 108/107)	Group 3	-	-	-	5.5	0.3	1.8	10.8	0.6	4.2
Cl_5 (CB 123)	Group 3	-	-	-	1.5	0.8	6.9	10.4	0.6	4.4
Cl_5 (CB 106/118)	Group 3	-	-	-	8.3	0.1	1.2	58.3	-0.1	0.8
Cl_5 (CB 114)	Group 3	-	-	-	0.9	1.1	11.8	2.4	1.3	19.4
Cl_5 (CB 122)	Group 3	-	-	-						
Cl_5 (CB 105)	Group 3	-	-	-	7.3	0.1	1.4	40.1	0.1	1.1
Cl_6 (CB 155)	Group 1	-	-	-	-	-	-	13.9	0.5	3.3
Cl_6 (CB 150)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 152)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 145)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 148)	Group 1	51.5	1.09	12.3	-	-	-	52.1	-0.1	0.9
Cl_6 (CB 136)	Group 5	-	-	-	32.3	-0.5	0.3	248.5	-0.7	0.2

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 154)	Group 1	-	-	-	5.9	0.2	1.7	35.5	0.1	1.3
Cl_6 (CB 151)	Group 5	12.0	1.72	52.7	15.6	-0.2	0.7	110	-0.4	0.4
Cl_6 (CB 135/144)	Group 5	13.1	1.69	48.6	19.2	-0.3	0.5	142	-0.5	0.3
Cl_6 (CB 147)	Group 2	882	-0.14	0.71	12.6	-0.1	0.8	82.2	-0.3	0.6
Cl_6 (CB 149)	Group 5	48.0	1.12	13.2	21.5	-0.3	0.5	161	-0.5	0.3
Cl_6 (CB 139/140)	Group 2	-	-	-	2.4	0.6	4.3	7.4	0.8	6.2
Cl_6 CB-143/134	Group 5	-	-	-	6.9	0.2	1.5	45.6	0.0	1.0
Cl_6 (CB 142/131)	Group 5	-	-	-	1.9	0.7	5.4	17.2	0.4	2.6
Cl_6 (CB 133)	Group 1	-	-	-	2.8	0.6	3.6	7.5	0.8	6.1
Cl_6 (CB 146/161)	Group 1	396	0.21	1.60	10.9	0.0	0.9	64.8	-0.2	0.7
Cl_6 (CB 165)	Group 1	-	-	-	1.4	0.9	7.3	2.2	1.3	20.9
Cl_6 (CB 132/153)	Group I	366	0.24	1.73	7.6	0.1	1.3	47.7	0.0	1.0
Cl_6 (CB 168)	Group 1	-	-	-	15.1	-0.2	0.7	130	-0.5	0.4
Cl_6 (CB 141)	Group 4	22.7	1.45	28.0	7.6	0.1	1.3	29.3	0.2	1.6
Cl_6 (CB 137)	Group 2	29.3	1.34	21.69	13.4	-0.1	0.8	95.3	-0.3	0.5
Cl_6 (CB 130)	Group 2	1900	-0.48	0.33	11.9	-0.1	0.9	67.2	-0.2	0.7
Cl_6 (CB 160/163/164/138)	Group 2	328	0.29	1.93	10.8	0.0	0.9	66.1	-0.2	0.7
Cl_6 (CB 158)	Group 2	190	0.53	3.34	7.9	0.1	1.3	48.6	0.0	0.9
Cl_6 (CB 129)	Group 4	-	-	-	9.5	0.0	1.1	35.7	0.1	1.3
Cl_6 (CB 166)	Group 2	-	-	-	4.8	0.3	2.1	22.8	0.3	2.0
Cl_6 (CB 159)	Group 3	-	-	-	17.1	-0.2	0.6	55.6	-0.1	0.8
Cl_6 (CB 162)	Group 4	-	-	-	1.1	1.0	9.0	2.9	1.2	15.7

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 128)	Group 2	445	0.15	1.42	9.5	0.0	1.1	54.2	-0.1	0.8
Cl_6 (CB 167)	Group 3	-	-	-	6.3	0.2	1.6	33.3	0.1	1.4
Cl_6 (CB 156)	Group 3	-	-	-	8.0	0.1	1.3	32.6	0.1	1.4
Cl_6 (CB 157)	Group 3	-	-	-	4.0	0.4	2.6	16.8	0.4	2.7
Cl7 (CB 188)	Group 1	18.4	1.54	34.4	4.2	0.4	2.4	13.5	0.5	3.4
Cl_7 (CB 184)	Group 1	5.58	2.06	114	3.1	0.5	3.2	9.5	0.7	4.8
Cl_7 (CB 179)	Group 5	7.78	1.91	81.6	29.0	-0.5	0.4	150	-0.5	0.3
Cl_7 (CB 176)	Group 5	10.7	1.77	59.3	13.5	-0.1	0.8	73.1	-0.2	0.6
Cl_7 (CB 186)	Group 5	-	-	-	-	-	-	-	-	-
Cl_7 (CB 178)	Group 1	181	0.54	3.49	11.2	0.0	0.9	49.5	0.0	0.9
Cl_7 (CB 175)	Group 1	74.9	0.93	8.47	6.0	0.2	1.7	27.2	0.2	1.7
Cl_7 (CB 187/182)	Group I	415	0.19	1.53	19.0	-0.3	0.5	77.0	-0.2	0.6
Cl_7 (CB 183)	Group 1	327	0.29	1.94	9.8	0.0	1.0	42.6	0.0	1.1
Cl_7 (CB 185)	Group 5	-	-	-	8.6	0.1	1.2	36.8	0.1	1.2
Cl_7 (CB 174/181)	Group 5	53.7	1.07	11.8	16.2	-0.2	0.6	94.3	-0.3	0.5
Cl_7 (CB 177)	Group 2	307	0.32	2.07	33.4	-0.5	0.3	168	-0.6	0.3
Cl_7 (CB 171)	Group 2	224	0.45	2.83	9.2	0.0	1.1	46.0	0.0	1.0
Cl_7 (CB 173)	Group 2	-	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	Group 1	-	-	-	10.6	0.0	1.0	41.9	0.0	1.1
Cl_7 (CB 180)	Group 1	635	-	-	10.2	-	-	45.7	-	-
Cl_7 (CB 193)	Group 1	2880	-0.66	0.22	9.5	0.0	1.1	41.3	0.0	1.1
Cl_7 (CB 191)	Group 1	-	-	-	3.7	0.4	2.7	16.7	0.4	2.7

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_7 (CB 170/190)	Group 2	885	-0.14	0.71	9.4	0.0	1.1	44.5	0.0	1.0
Cl_7 (CB 189)	Group 3				8.0	0.1	1.3	24.3	0.3	1.9
Cl_8 (CB 202)	Group 1	203	0.50	3.12	12.4	-0.1	0.8	43.2	0.0	1.1
Cl_8 (CB 200)	Group I	97.1	0.82	6.54	10.9	0.0	0.9	31.1	0.2	1.5
Cl_8 (CB 204)	Group I	-	-	-	-	-	-	-	-	-
Cl_8 (CB 197)	Group I	121	0.72	5.26	5.7	0.3	1.8	14.8	0.5	3.1
Cl_8 (CB 199)	Group 5	79.7	0.90	7.96						
Cl_8 (CB 198)	Group I	12.52	1.71	50.7	5.5	0.3	1.9	28.3	0.2	1.6
Cl_8 (CB 201)	Group I	7630	-1.08	0.083	13.7	-0.1	0.7	51.1	0.0	0.9
Cl_8 (CB 203/196)	Group I	779	-0.09	0.81	13.0	-0.1	0.8	37.1	0.1	1.2
Cl_8 (CB 195)	Group 2	-	-	-	6.7	0.2	1.5	17.8	0.4	2.6
Cl_8 (CB 194)	Group I	1200	-0.28	0.52	13.7	-0.1	0.7	36.0	0.1	1.3
Cl_8 (CB 205)	Group I	-	-	-	4.1	0.4	2.5	9.7	0.7	4.7
Cl_9 (CB 208)	Group 1	-	-	-	7.0	0.2	1.5	33.3	0.1	1.4
Cl_9 (CB 207)	Group I	-	-	-	2.2	0.7	4.7	8.5	0.7	5.4
Cl_9 (CB 206)	Group I	-	-	-	6.0	0.2	1.7	11.9	0.6	3.8
Cl_{10} (CB 209)	Group I	562	0.05	1.13	2.9	0.5	3.5	4.2	1.0	11.0
Chlorobenzenes (CBz)										
1,3,5 TriCBz		-	-	-	0.9	1.0	11.1	1.0	1.7	45.0
1,2,4 TriCBz		-	-	-	2.1	0.7	4.8	1.5	1.5	30.8
1,2,3 TriCBz		-	-	-	0.5	1.4	22.5	0.4	2.1	114
1,2,3,5/1,2,4,5 TeCBz		10.7	1.8	59.2	2.6	0.6	4.0	5.6	0.9	8.2

	White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
	BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
1,2,3,4 TeCBz	3.7	2.2	170	3.4	0.5	3.0	4.1	1.0	11.1
PeCBz	7.6	1.9	83.4	8.7	0.1	1.2	19.6	0.4	2.3
HCB	75.1	0.9	8.5	4.2	0.4	2.4	15.5	0.5	3.0
Hexachlorocyclohexanes (HCHs)									
α-HCH	-	-	-	4.0	0.4	2.6	2.4	1.3	19.4
β-HCH	2.8	2.4	228.9	18.6	-0.3	0.5	50.5	0.0	0.9
γ-HCH	-	-	-	6.7	0.2	1.5	12.5	0.6	3.7
Dichlordiphenyltrichloroethanes (DDTs)									
p,p-DDT	-	-	-	26.6	-0.4	0.4	151	-0.5	0.3
o,p-DDT	-	-	-	75.8	-0.9	0.1	0.0	-1.1	0.1
p,p-DDE	581.5	0.0	1.1	7.3	0.1	1.4	40.4	0.1	1.1
o,p-DDE	-	-	-	4.1	0.4	2.5	25.1	0.3	1.8
p,p-DDD	-	-	-	12.3	-0.1	0.8	67.0	-0.2	0.7
o,p-DDD	-	-	-	11.9	-0.1	0.9	75.4	-0.2	0.6
Cyclodienes									
aldrin	-	-	-						
heptachlor	-	-	-	4.63	0.34	2.20	64.9	-0.15	0.70
heptachlor epoxide	-	-	-	-	-	-	41.3	0.04	1.11
***trans*-chlordane**	-	-	-	-	-	-	-	-	-
***cis*-chlordane**	12.2	1.72	51.9	9.90	0.01	1.03	28.9	0.20	1.58
***trans*-nonachlor**	11.9	1.72	53.0	23.9	-0.37	0.42	68.1	-0.17	0.67
***cis*-nonachlor**	-	-	-	6.98	0.16	1.46	14.8	0.49	3.10
oxychlordane	4130	-0.81	0.15	-	-	-	85.4	-0.27	0.54
α-endosulfan	-	-	-	-	-	-	-	-	-

		White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
		BMF_{MAX} =	635		BMF_{MAX} =	10.2		BMF_{MAX} =	45.7	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
β-endosulfan		-	-	-	1.67	0.78	6.09	4.32	1.02	10.6
endosulfan sulfate		-	-	-	-	-	-	-	-	-
dieldrin		47.5	1.13	13.4	2.74	0.57	3.71	1.95	1.37	23.5
methoxychlor		-	-	-	-	-	-	-	-	-
mirex		56.33	1.05	11.28	10.9	-0.03	0.94	25.1	0.26	1.82
octachlorostyrene		-	-	-	1.21	0.92	8.38	2.21	1.32	20.7

Appendix 8 continued.

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_2 (PCB 7/9)	Group 3	-	-	-	-	-	-	0.58	1.30	19.95
Cl_2 (PCB 6)	Group 3	-	-	-	0.28	1.48	29.86	0.45	1.41	25.49
Cl_2 (PCB 8/5)	Group 3	0.10	1.58	1.25	0.36	1.37	23.54	0.40	1.45	28.46
Cl_2 (PCB 4/10)	Group 4	0.16	1.36	1.45	0.28	1.47	29.74	0.33	1.54	34.92
Cl_3 (PCB 23/34)	Group 3	-0.32	4.10	0.48	-	-	-	-	-	-
Cl_3 (PCB 29)	Group 3	-0.12	2.63	0.75	-	-	-	-	-	-
Cl_3 (PCB 26)	Group 3	-0.05	2.22	0.89	2.36	0.55	3.55	1.99	0.76	5.78
Cl_3 (CB 25)	Group 3	0.13	1.48	1.34	1.10	0.88	7.60	0.92	1.10	12.54
Cl_3 (CB 31)	Group 3	-0.25	3.49	0.57	5.39	0.19	1.55	3.16	0.56	3.64
Cl_3 (CB 28)	Group 3	0.03	1.85	1.07	7.14	0.07	1.17	9.48	0.08	1.21
Cl_3 (CB 21)	Group 3	-	-	-	-	-	-	-	-	-
Cl_3 (CB 33/20)	Group 3	-0.30	3.97	0.50	0.52	1.21	16.11	0.54	1.33	21.35
Cl_3 (CB 19)	Group 4	-	-	-	0.20	1.62	41.84	0.48	1.38	23.86
Cl_3 (CB30)	Group 4	-	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	Group 4	-0.17	2.94	0.68	0.32	1.42	26.23	0.38	1.48	30.42
Cl_3 (CB 17)	Group 4	-0.15	2.82	0.70	0.29	1.46	29.01	0.33	1.55	35.26
Cl_3 (CB 27/24)	Group 4	-0.07	2.33	0.85	0.26	1.50	31.84	0.28	1.61	40.73
Cl_3 (CB 16/32)	Group 4	-0.16	2.84	0.70	0.35	1.38	24.11	0.41	1.44	27.76
Cl_3 (CB 22)	Group 3	-0.09	2.44	0.81	0.95	0.94	8.78	0.85	1.13	13.45
Cl_4 (CB 54)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	Group 5	-0.65	8.82	0.22	-	-	-	-	-	-

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 53)	Group 4	0.08	1.65	1.20	0.26	1.51	32.52	0.21	1.74	55.54
Cl_4 (CB 51)	Group 5	-0.67	9.22	0.22	-	-	-	-	-	-
Cl_4 (CB 45)	Group 5	-0.22	3.29	0.60	0.16	1.71	51.16	0.13	1.93	86.08
Cl_4 (CB 46)	Group 5	-0.21	3.23	0.61	-	-	-	-	-	-
Cl_4 (CB 73/52)	Group 4	-0.23	3.35	0.59	6.04	0.14	1.38	8.20	0.15	1.40
Cl_4 (CB 69)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	Group 4	-0.23	3.36	0.59	2.96	0.45	2.83	3.99	0.46	2.88
Cl_4 (CB 43)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	Group 4	-0.23	3.33	0.60	5.13	0.21	1.63	7.06	0.21	1.63
Cl_4 (CB 65)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	Group 4	-0.26	3.57	0.56	2.57	0.51	3.25	3.22	0.55	3.57
Cl_4 (CB 59/42)	Group 4	-0.30	3.92	0.51	0.68	1.09	12.27	0.69	1.22	16.55
Cl_4 (CB 72)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	Group 4	-0.44	5.46	0.36	0.58	1.16	14.39	0.51	1.35	22.61
Cl_4 (CB 68)	Group 3	-	-	-	3.19	0.42	2.62	3.49	0.52	3.29
Cl_4 (CB 40)	Group 4	-0.23	3.37	0.59	0.43	1.29	19.57	0.68	1.23	16.81
Cl_4 (CB 57)	Group 3	-0.13	2.69	0.74	-	-	-	-	-	-
Cl_4 (CB 67)	Group 3	0.08	1.63	1.21	-	-	-	-	-	-
Cl_4 (CB 58)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	Group 3	-0.12	2.64	0.75	2.74	0.48	3.05	1.65	0.84	6.97
Cl_4 (CB 61/74)	Group 3	-0.16	2.86	0.69	14.49	-0.24	0.58	21.37	-0.27	0.54

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 70/76)	Group 3	-0.18	3.03	0.65	5.03	0.22	1.66	5.18	0.35	2.22
Cl_4 (CB 66)	Group 3	-0.16	2.83	0.70	10.00	-0.08	0.84	9.02	0.10	1.27
Cl_4 (CB 55)	Group 3	-0.14	2.72	0.73	-	-	-	-	-	-
Cl_4 (CB 60/56)	Group 3	-0.16	2.86	0.69	10.05	-0.08	0.83	8.66	0.12	1.33
Cl_5 (CB 104)	Group 5	-0.07	2.31	0.86	-	-	-	-	-	-
Cl_5 (CB 96)	Group 5	-0.20	3.11	0.64	-	-	-	-	-	-
Cl_5 (CB 103)	Group 5	-0.18	2.99	0.66	0.25	1.53	33.76	0.21	1.74	54.98
Cl_5 (CB 100)	Group 2	-0.15	2.78	0.71	0.15	1.74	55.26	-	-	-
Cl_5 (CB 94)	Group 5	0.09	1.60	1.24	0.43	1.29	19.60	0.38	1.48	30.41
Cl_5 (CB 95)	Group 5	-0.20	3.12	0.63	2.02	0.62	4.14	2.29	0.70	5.01
Cl_5 (CB 102/93)	Group 5	-0.39	4.83	0.41	-	-	-	-	-	-
Cl_5 (CB 98)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 88)	Group 5	-0.32	4.12	0.48	1.59	0.72	5.27	1.78	0.81	6.44
Cl_5 (CB 91)	Group 5	-0.16	2.85	0.70	1.51	0.74	5.53	2.04	0.75	5.62
Cl_5 (CB 121)	Group 1	-	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 CB (101/90)	Group 4	-0.16	2.88	0.69	10.88	-0.11	0.77	14.68	-0.11	0.78
Cl_5 (CB 89)	Group 5	-0.18	3.02	0.66	0.14	1.79	61.30	0.15	1.87	74.10
Cl_5 (CB 99)	Group 2	-0.12	2.62	0.76	9.74	-0.07	0.86	12.24	-0.03	0.94
Cl_5 (CB 113)	Group 4	-0.38	4.78	0.41						
Cl_5 (CB 119)	Group 2	-0.31	4.00	0.50	5.44	0.19	1.54	6.46	0.25	1.78
Cl_5 (CB 112)	Group 4	-0.23	3.38	0.59	-	-	-	-	-	-

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_5 (CB 109/83)	Group 4	-0.14	2.75	0.72	-	-	-	-	-	-
Cl_5 (CB 97/86)	Group 4	-0.19	3.05	0.65	2.58	0.51	3.25	2.58	0.65	4.45
Cl_5 (CB 116/125/117)	Group 4	-0.08	2.40	0.83	23.59	-0.45	0.35	28.37	-0.39	0.40
Cl_5 (CB 115/87)	Group 2	-0.17	2.93	0.68	5.72	0.17	1.46	8.13	0.15	1.41
Cl_5 (CB 111)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 85)	Group 2	-0.16	2.83	0.70	4.05	0.31	2.06	5.24	0.34	2.19
Cl_5 (CB 120)	Group 3	0.29	1.01	1.96	-	-	-	-	-	-
Cl_5 (CB 110)	Group 4	-0.22	3.27	0.61	4.79	0.24	1.74	4.91	0.37	2.34
Cl_5 (CB 82)	Group 4	-0.18	3.02	0.66	0.67	1.10	12.57	0.63	1.26	18.09
Cl_5 (CB 124)	Group 3	-0.05	2.24	0.89	0.46	1.26	18.16	0.61	1.27	18.82
Cl_5 (CB 108/107)	Group 3	-0.19	3.07	0.65	9.21	-0.04	0.91	8.41	0.14	1.37
Cl_5 (CB 123)	Group 3	-0.05	2.24	0.89	1.15	0.86	7.28	5.25	0.34	2.19
Cl_5 (CB 106/118)	Group 3	-0.18	2.99	0.66	17.56	-0.32	0.48	16.33	-0.15	0.70
Cl_5 (CB 114)	Group 3	-0.43	5.34	0.37	5.56	0.18	1.50	6.30	0.26	1.82
Cl_5 (CB 122)	Group 3	-0.63	8.36	0.24	-	-	-	-	-	-
Cl_5 (CB 105)	Group 3	-0.21	3.19	0.62	10.67	-0.11	0.78	12.29	-0.03	0.93
Cl_6 (CB 155)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 150)	Group 5	-0.22	3.27	0.61	-	-	-	-	-	-
Cl_6 (CB 152)	Group 5	-0.23	3.33	0.59	-	-	-	-	-	-
Cl_6 (CB 145)	Group 5	-0.21	3.18	0.62	-	-	-	-	-	-
Cl_6 (CB 148)	Group 1	-	-	-	-	-	-	0.89	1.11	12.92
Cl_6 (CB 136)	Group 5	-0.11	2.55	0.78	0.85	0.99	9.79	0.66	1.24	17.37

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 154)	Group 1	-0.04	2.17	0.91	2.71	0.49	3.09	2.56	0.65	4.48
Cl_6 (CB 151)	Group 5	-0.10	2.47	0.80	1.17	0.85	7.15	1.50	0.88	7.63
Cl_6 (CB 135/144)	Group 5	-0.12	2.60	0.76	2.44	0.54	3.43	2.66	0.63	4.31
Cl_6 (CB 147)	Group 2	-0.10	2.51	0.79	2.96	0.45	2.82	3.28	0.54	3.49
Cl_6 (CB 149)	Group 5	-0.09	2.42	0.82	5.42	0.19	1.54	5.74	0.30	2.00
Cl_6 (CB 139/140)	Group 2	-0.05	2.21	0.90	0.06	2.15	140.75	0.10	2.07	117.56
Cl_6 CB-143/134	Group 5	-0.11	2.55	0.78	13.42	-0.21	0.62	11.36	0.00	1.01
Cl_6 (CB 142/131)	Group 5	0.26	1.09	1.82	3.75	0.35	2.23	5.74	0.30	2.00
Cl_6 (CB 133)	Group 1	-0.30	3.92	0.51	4.01	0.32	2.08	2.60	0.64	4.41
Cl_6 (CB 146/161)	Group 1	-0.05	2.25	0.88	13.09	-0.19	0.64	15.84	-0.14	0.72
Cl_6 (CB 165)	Group 1	-0.43	5.37	0.37	0.49	1.23	17.05			
Cl_6 (CB 132/153)	Group I	-0.10	2.52	0.79	11.68	-0.15	0.72	14.15	-0.09	0.81
Cl_6 (CB 168)	Group 1	-0.11	2.55	0.78	1.84	0.66	4.53	2.50	0.66	4.60
Cl_6 (CB 141)	Group 4	-0.15	2.79	0.71	3.90	0.33	2.15	4.44	0.41	2.58
Cl_6 (CB 137)	Group 2	-0.03	2.12	0.94	11.55	-0.14	0.72	13.27	-0.06	0.86
Cl_6 (CB 130)	Group 2	-0.22	3.26	0.61	13.01	-0.19	0.64	14.74	-0.11	0.78
Cl_6 (CB 160/163/164/138)	Group 2	-0.14	2.71	0.73	10.62	-0.10	0.79	14.27	-0.09	0.80
Cl_6 (CB 158)	Group 2	-0.17	2.91	0.68	4.34	0.28	1.92	6.20	0.27	1.85
Cl_6 (CB 129)	Group 4	-0.05	2.21	0.90	3.87	0.33	2.16	4.73	0.38	2.42
Cl_6 (CB 166)	Group 2	-0.19	3.05	0.65	4.49	0.27	1.86	3.88	0.47	2.95
Cl_6 (CB 159)	Group 3	0.06	1.72	1.15	5.80	0.16	1.44	7.82	0.17	1.47
Cl_6 (CB 162)	Group 4	-0.02	2.09	0.95	0.85	0.99	9.86	0.85	1.13	13.47

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 128)	Group 2	-0.14	2.74	0.72	1.61	0.72	5.20	2.45	0.67	4.69
Cl_6 (CB 167)	Group 3	-0.07	2.32	0.86	3.80	0.34	2.20	2.88	0.60	3.99
Cl_6 (CB 156)	Group 3	-0.11	2.52	0.79	6.81	0.09	1.23	8.58	0.13	1.34
Cl_6 (CB 157)	Group 3	-0.09	2.46	0.81	3.88	0.33	2.15	4.47	0.41	2.57
Cl7 (CB 188)	Group 1	-0.04	2.18	0.91				0.02	2.80	628.56
Cl_7 (CB 184)	Group 1	-0.07	2.32	0.85	0.47	1.25	17.81	0.24	1.68	48.38
Cl_7 (CB 179)	Group 5	0.02	1.90	1.04	0.48	1.24	17.56	0.47	1.39	24.31
Cl_7 (CB 176)	Group 5	-0.06	2.27	0.87	0.50	1.23	16.83	0.44	1.42	26.27
Cl_7 (CB 186)	Group 5	-0.42	5.22	0.38	-	-	-	-	-	-
Cl_7 (CB 178)	Group 1	-0.04	2.16	0.92	7.43	0.05	1.13	7.88	0.16	1.46
Cl_7 (CB 175)	Group 1	-0.03	2.14	0.92	0.50	1.22	16.78	0.75	1.19	15.40
Cl_7 (CB 187/182)	Group I	-0.03	2.12	0.94	6.30	0.12	1.33	9.40	0.09	1.22
Cl_7 (CB 183)	Group 1	-0.01	2.01	0.98	5.60	0.17	1.49	8.00	0.16	1.44
Cl_7 (CB 185)	Group 5	-0.03	2.13	0.93	0.85	0.99	9.85	0.95	1.08	12.10
Cl_7 (CB 174/181)	Group 5	-0.06	2.26	0.88	1.98	0.63	4.23	1.53	0.88	7.52
Cl_7 (CB 177)	Group 2	-0.04	2.19	0.90	4.75	0.25	1.76	9.47	0.08	1.21
Cl_7 (CB 171)	Group 2	-0.04	2.16	0.92	3.46	0.38	2.42	5.07	0.36	2.26
Cl_7 (CB 173)	Group 2	-0.84	13.64	0.15	-	-	-	-	-	-
Cl_7 (CB 192/172)	Group 1	-0.01	2.02	0.98	6.76	0.09	1.24	8.21	0.15	1.40
Cl_7 (CB 180)	Group 1	0.00	1.98	1.00	8.36	0.00	1.00	11.48	0.00	1.00
Cl_7 (CB 193)	Group 1	-0.05	2.23	0.89	7.74	0.03	1.08	10.90	0.02	1.05
Cl_7 (CB 191)	Group 1	-0.62	8.22	0.24	3.32	0.40	2.52	4.40	0.42	2.61

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_7 (CB 170/190)	Group 2	-0.04	2.16	0.92	6.68	0.10	1.25	9.80	0.07	1.17
Cl_7 (CB 189)	Group 3	0.01	1.95	1.01	3.07	0.44	2.73	3.69	0.49	3.11
Cl_8 (CB 202)	Group 1	0.07	1.69	1.17	5.60	0.17	1.49	5.56	0.31	2.06
Cl_8 (CB 200)	Group I	0.08	1.67	1.19	0.08	2.01	102.60	0.07	2.23	171.03
Cl_8 (CB 204)	Group I	0.07	1.69	1.17						
Cl_8 (CB 197)	Group I	0.11	1.53	1.30	0.33	1.41	25.53	0.40	1.46	28.69
Cl_8 (CB 199)	Group 5	-0.02	2.08	0.95						
Cl_8 (CB 198)	Group I	0.51	0.61	3.24	1.55	0.73	5.39	2.90	0.60	3.95
Cl_8 (CB 201)	Group I				7.86	0.03	1.06	3.78	0.48	3.04
Cl_8 (CB 203/196)	Group I	0.10	1.59	1.24	5.03	0.22	1.66	5.38	0.33	2.13
Cl_8 (CB 195)	Group 2	0.18	1.32	1.51	3.53	0.37	2.37	4.66	0.39	2.46
Cl_8 (CB 194)	Group I	0.19	1.29	1.54	5.91	0.15	1.42	7.75	0.17	1.48
Cl_8 (CB 205)	Group I	-0.07	2.35	0.84	4.61	0.26	1.81	2.83	0.61	4.06
Cl_9 (CB 208)	Group 1	2.37	0.01	236.08	1.07	0.89	7.85	0.95	1.08	12.13
Cl_9 (CB 207)	Group I	0.88	0.26	7.66	0.21	1.60	39.93	0.41	1.45	27.92
Cl_9 (CB 206)	Group I	0.81	0.31	6.46	3.03	0.44	2.76	3.18	0.56	3.61
Cl_{10} (CB 209)	Group I	-	-	-	0.76	1.04	11.01	0.82	1.15	14.01

Group	Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
	BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Chlorobenzenes (CBz)									
1,3,5 TriCBz	1.2	0.6	4.2	0.8	1.0	10.0	1.0	1.1	11.6
1,2,4 TriCBz	3.9	0.1	1.3	3.3	0.4	2.5	2.2	0.7	5.2
1,2,3 TriCBz	0.5	1.0	10.0	0.4	1.3	20.6	0.4	1.4	27.2
1,2,3,5/1,2,4,5 TeCBz	9.7	-0.3	0.5	16.8	-0.3	0.5	12.7	0.0	0.9
1,2,3,4 TeCBz	5.3	0.0	1.0	1.4	0.8	6.0	2.5	0.7	4.5
PeCBz	28.3	-0.8	0.2	6.8	0.1	1.2	9.6	0.1	1.2
HCB	19.3	-0.6	0.3	0.6	1.1	13.2	1.4	0.9	8.3
HCHs									
α-HCH	12.9	-0.4	0.4	12.6	-0.2	0.7	5.7	0.3	2.0
β-HCH	58.7	-1.1	0.1	15.4	-0.3	0.5	26.2	-0.4	0.4
γ-HCH	25.4	-0.7	0.2	3.4	0.4	2.4	4.0	0.5	2.8
DDTs									
p,p-DDT	31.1	-0.8	0.2	17.1	-0.3	0.5	15.8	-0.1	0.7
o,p-DDT	208.4	-1.6	0.0	0.3	1.5	30.5	7.9	0.2	1.4
p,p-DDE	12.0	-0.4	0.4	10.1	-0.1	0.8	8.3	0.1	1.4
o,p-DDE	21.9	-0.6	0.2	0.3	1.4	24.2	1.4	0.9	8.1
p,p-DDD	26.0	-0.7	0.2	1.5	0.8	5.7	2.9	0.6	4.0
o,p-DDD	34.7	-0.8	0.1	-	-	-	3.7	0.5	3.1
Cyclodienes									
aldrin	-	-	-	-	-	-	-	-	-
heptachlor	32.6	-0.8	0.2	24.8	-0.5	0.3	23.4	-0.3	0.5
heptachlor epoxide	41.9	-0.9	0.1	-	-	-	-	-	-
trans-chlordane	-	-	-	-	-	-	-	-	-

		Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
		BMF_{MAX} =	1.9		BMF_{MAX} =	8.4		BMF_{MAX} =	11.5	
	Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
***cis*-chlordane**		8.3	-0.2	0.6	6.2	0.1	1.3	4.1	0.4	2.8
***trans*-nonachlor**		16.9	-0.5	0.3	6.6	0.1	1.3	6.6	0.2	1.7
***cis*-nonachlor**		6.0	-0.1	0.8	1.3	0.8	6.6	1.2	1.0	9.9
oxychlordane		39.0	-0.9	0.1	-	-	-	-	-	-
α-endosulfan		-	-	-	-	-	-			
β-endosulfan		1.4	0.5	3.5	1.0	0.9	8.1	0.8	1.2	14.8
endosulfan sulfate		-	-	-	-	-	-	-	-	-
dieldrin		33.0	-0.8	0.2	14.4	-0.2	0.6	6.3	0.3	1.8
methoxychlor		-	-	-	-	-	-	-	-	-
mirex		5.1	0.0	1.0	4.7	0.3	1.8	3.6	0.5	3.1
octachlorostyrene		2.6	0.3	2.0	1.2	0.8	6.9	0.2	1.8	59.1

Appendix 8 continued.

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_2 (PCB 7/9)	Group 3	2.02	2.16	142.83	-	-	-	-	-	-
Cl_2 (PCB 6)	Group 3	1.61	2.25	179.63	-	-	-	-	-	-
Cl_2 (PCB 8/5)	Group 3	2.32	2.10	124.63	-	-	-	-	-	-
Cl_2 (PCB 4/10)	Group 4	1.66	2.24	174.02	-	-	-	-	-	-
Cl_3 (PCB 23/34)	Group 3				-	-	-	-	-	-
Cl_3 (PCB 29)	Group 3	2.89	2.00	99.88	-	-	-	-	-	-
Cl_3 (PCB 26)	Group 3	22.36	1.11	12.90	-	-	-	-	-	-
Cl_3 (CB 25)	Group 3				-	-	-	-	-	-
Cl_3 (CB 31)	Group 3	17.79	1.21	16.22	-	-	-	-	-	-
Cl_3 (CB 28)	Group 3	48.79	0.77	5.91	-	-	-	-	-	-
Cl_3 (CB 21)	Group 3	7.75	1.57	37.24	-	-	-	-	-	-
Cl_3 (CB 33/20)	Group 3	1.22	2.37	235.65	-	-	-	-	-	-
Cl_3 (CB 19)	Group 4	7.95	1.56	36.30	-	-	-	-	-	-
Cl_3 (CB30)	Group 4	-	-	-	-	-	-	-	-	-
Cl_3 (CB 18)	Group 4	51.74	0.75	5.58	-	-	-	-	-	-
Cl_3 (CB 17)	Group 4	2.85	2.01	101.31	-	-	-	-	-	-
Cl_3 (CB 27/24)	Group 4	0.40	2.86	715.84	-	-	-	-	-	-
Cl_3 (CB 16/32)	Group 4	6.72	1.63	42.95	-	-	-	-	-	-
Cl_3 (CB 22)	Group 3	5.23	1.74	55.16	-	-	-	-	-	-
Cl_4 (CB 54)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 50)	Group 5	-	-	-	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 53)	Group 4	0.76	2.58	377.85	-	-	-	-	-	-
Cl_4 (CB 51)	Group 5	-	-	-	-	-	-	-	-	-
Cl_4 (CB 45)	Group 5	1.05	2.44	274.92	-	-	-	-	-	-
Cl_4 (CB 46)	Group 5	0.13	3.33	2159.88	-	-	-	-	-	-
Cl_4 (CB 73/52)	Group 4	148.00	0.29	1.95	6.5	1.2	14.4	1.81	1.14	13.85
Cl_4 (CB 69)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 49)	Group 4	64.37	0.65	4.48	-	-	-	-	-	-
Cl_4 (CB 43)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 47/75/48)	Group 4	180.17	0.20	1.60	22.4	0.6	4.2	-	-	-
Cl_4 (CB 65)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 62)	Group 4	-	-	-	-	-	-	-	-	-
Cl_4 (CB 44)	Group 4	28.85	1.00	10.00	-	-	-	-	-	-
Cl_4 (CB 59/42)	Group 4	7.09	1.61	40.71	-	-	-	-	-	-
Cl_4 (CB 72)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 71/41/64)	Group 4	6.75	1.63	42.77	-	-	-	-	-	-
Cl_4 (CB 68)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 40)	Group 4	9.29	1.49	31.07	-	-	-	-	-	-
Cl_4 (CB 57)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 67)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 58)	Group 3	-	-	-	-	-	-	-	-	-
Cl_4 (CB 63)	Group 3	6.67	1.64	43.25	-	-	-	-	-	-
Cl_4 (CB 61/74)	Group 3	267.99	0.03	1.08	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_4 (CB 70/76)	Group 3	26.64	1.04	10.83	-	-	-	-	-	-
Cl_4 (CB 66)	Group 3	89.25	0.51	3.23	-	-	-	-	-	-
Cl_4 (CB 55)	Group 3	8.09	1.55	35.68	-	-	-	-	-	-
Cl_4 (CB 60/56)	Group 3	69.65	0.62	4.14	3.3	1.5	28.5	-	-	-
Cl_5 (CB 104)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 96)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 (CB 103)	Group 5	10.32	1.45	27.96	-	-	-	-	-	-
Cl_5 (CB 100)	Group 2	-	-	-	-	-	-	-	-	-
Cl_5 (CB 94)	Group 5	3.12	1.97	92.42	-	-	-	-	-	-
Cl_5 (CB 95)	Group 5	13.13	1.34	21.97	-	-	-	-	-	-
Cl_5 (CB 102/93)	Group 5	1.03	2.45	279.31	-	-	-	-	-	-
Cl_5 (CB 98)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 88)	Group 5	112.14	0.41	2.57	-	-	-	-	-	-
Cl_5 (CB 91)	Group 5	25.85	1.05	11.16	-	-	-	-	-	-
Cl_5 (CB 121)	Group 1	-	-	-	-	-	-	-	-	-
Cl_5 (CB 92/84)	Group 5	-	-	-	-	-	-	-	-	-
Cl_5 CB (101/90)	Group 4	144.67	0.30	1.99	-	-	-	1.19	1.32	20.95
Cl_5 (CB 89)	Group 5	1.28	2.35	225.75	-	-	-	-	-	-
Cl_5 (CB 99)	Group 2	242.35	0.08	1.19	24.0	0.6	3.9	9.05	0.44	2.77
Cl_5 (CB 113)	Group 4				-	-	-	-	-	-
Cl_5 (CB 119)	Group 2	299.87	-0.02	0.96	-	-	-	-	-	-
Cl_5 (CB 112)	Group 4	-	-	-	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_5 (CB 109/83)	Group 4	36.92	0.89	7.81	-	-	-	-	-	-
Cl_5 (CB 97/86)	Group 4	9.96	1.46	28.98	-	-	-	-	-	-
Cl_5 (CB 116/125/117)	Group 4	-	-	-	-	-	-	-	-	-
Cl_5 (CB 115/87)	Group 2	92.16	0.50	3.13	-	-	-	-	-	-
Cl_5 (CB 111)	Group 3				-	-	-	-	-	-
Cl_5 (CB 85)	Group 2	62.40	0.67	4.62	-	-	-	-	-	-
Cl_5 (CB 120)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 110)	Group 4	48.70	0.77	5.92	-	-	-	-	-	-
Cl_5 (CB 82)	Group 4	20.07	1.16	14.37	-	-	-	-	-	-
Cl_5 (CB 124)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 108/107)	Group 3	299.56	-0.02	0.96	-	-	-	-	-	-
Cl_5 (CB 123)	Group 3	41.91	0.84	6.88	-	-	-	-	-	-
Cl_5 (CB 106/118)	Group 3	189.46	0.18	1.52	1.9	1.7	49.9	5.52	0.65	4.54
Cl_5 (CB 114)	Group 3	467.85	-0.21	0.62	-	-	-	-	-	-
Cl_5 (CB 122)	Group 3	-	-	-	-	-	-	-	-	-
Cl_5 (CB 105)	Group 3	147.26	0.29	1.96	-	-	-	3.38	0.87	7.41
Cl_6 (CB 155)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 150)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 152)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 145)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 148)	Group 1	9.21	1.50	31.34	-	-	-	-	-	-
Cl_6 (CB 136)	Group 5	2.11	2.14	136.87	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 154)	Group 1	33.63	0.93	8.58	-	-	-	-	-	-
Cl_6 (CB 151)	Group 5	14.29	1.31	20.18	-	-	-	-	-	-
Cl_6 (CB 135/144)	Group 5	10.92	1.42	26.42	-	-	-	-	-	-
Cl_6 (CB 147)	Group 2	47.81	0.78	6.03	-	-	-	-	-	-
Cl_6 (CB 149)	Group 5	30.69	0.97	9.40	1.3	1.9	71.0	-	-	-
Cl_6 (CB 139/140)	Group 2	-	-	-	-	-	-	-	-	-
Cl_6 CB-143/134	Group 5	155.05	0.27	1.86	-	-	-	-	-	-
Cl_6 (CB 142/131)	Group 5	-	-	-	-	-	-	-	-	-
Cl_6 (CB 133)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 146/161)	Group 1	161.55	0.25	1.79	4.8	1.3	19.4	-	-	-
Cl_6 (CB 165)	Group 1	-	-	-	-	-	-	-	-	-
Cl_6 (CB 132/153)	Group I	175.95	0.22	1.64	35.2	0.4	2.7	15.11	0.22	1.66
Cl_6 (CB 168)	Group 1	9.71	1.47	29.71	-	-	-	-	-	-
Cl_6 (CB 141)	Group 4	70.74	0.61	4.08	-	-	-	-	-	-
Cl_6 (CB 137)	Group 2	35.78	0.91	8.06	18.8	0.7	5.0	-	-	-
Cl_6 (CB 130)	Group 2	188.92	0.18	1.53	-	-	-	-	-	-
Cl_6 (CB 160/163/164/138)	Group 2	141.05	0.31	2.05	13.3	0.8	7.0	12.47	0.30	2.01
Cl_6 (CB 158)	Group 2	82.86	0.54	3.48	-	-	-	-	-	-
Cl_6 (CB 129)	Group 4	-	-	-	-	-	-	-	-	-
Cl_6 (CB 166)	Group 2	-	-	-	-	-	-	-	-	-
Cl_6 (CB 159)	Group 3	257.02	0.05	1.12	-	-	-	-	-	-
Cl_6 (CB 162)	Group 4	24.29	1.08	11.88	-	-	-	-	-	-

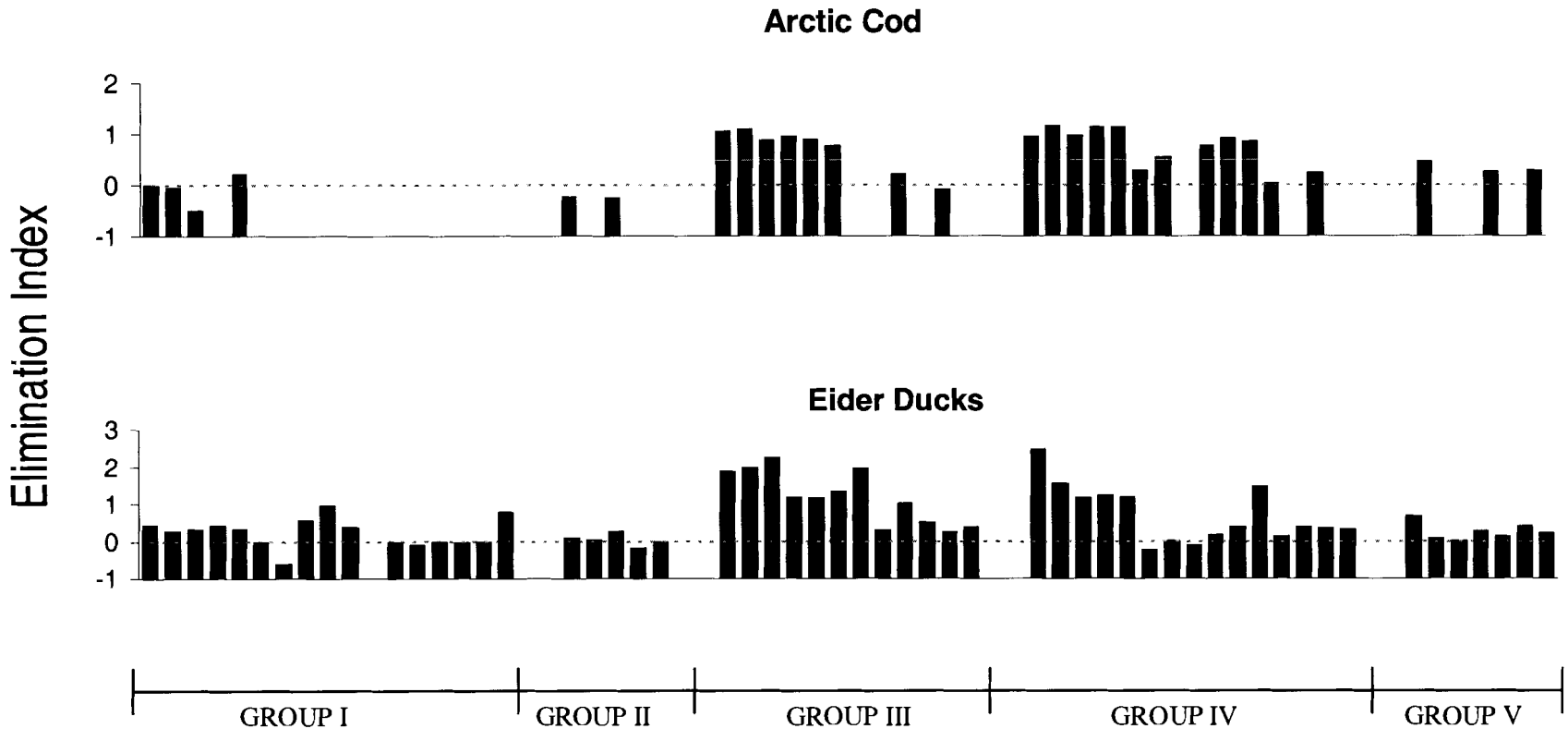
		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_6 (CB 128)	Group 2	28.49	1.01	10.13	-	-	-	-	-	-
Cl_6 (CB 167)	Group 3	19.74	1.17	14.61	-	-	-	-	-	-
Cl_6 (CB 156)	Group 3	89.67	0.51	3.22	50.7	0.3	1.9	28.48	-0.055	0.88
Cl_6 (CB 157)	Group 3	30.50	0.98	9.46	76.3	0.1	1.2	-	-	-
Cl7 (CB 188)	Group 1	0.14	3.33	2114.52	-	-	-	-	-	-
Cl_7 (CB 184)	Group 1	1.61	2.25	178.83	-	-	-	-	-	-
Cl_7 (CB 179)	Group 5	1.55	2.27	186.25	-	-	-	-	-	-
Cl_7 (CB 176)	Group 5	2.09	2.14	138.39	-	-	-	-	-	-
Cl_7 (CB 186)	Group 5	-	-	-	-	-	-	-	-	-
Cl_7 (CB 178)	Group 1	79.93	0.56	3.61	-	-	-	-	-	-
Cl_7 (CB 175)	Group 1	4.13	1.84	69.88	-	-	-	-	-	-
Cl_7 (CB 187/182)	Group I	63.30	0.66	4.56	5.3	1.3	17.9	11.39	0.34	2.20
Cl_7 (CB 183)	Group 1	85.08	0.53	3.39	15.8	0.8	6.0	11.39	0.34	2.20
Cl_7 (CB 185)	Group 5	-	-	-	-	-	-	-	-	-
Cl_7 (CB 174/181)	Group 5	17.88	1.21	16.13	-	-	-	-	-	-
Cl_7 (CB 177)	Group 2	20.05	1.16	14.39	-	-	-	-	-	-
Cl_7 (CB 171)	Group 2	31.96	0.96	9.03	-	-	-	-	-	-
Cl_7 (CB 173)	Group 2	-	-	-	-	-	-	-	-	-
Cl_7 (CB 192/172)	Group 1	-	-	-	-	-	-	-	-	-
Cl_7 (CB 180)	Group 1	288.47	-	-	94.0	-	-	25.10	-	-
Cl_7 (CB 193)	Group 1	1200.17	-0.62	0.24	-	-	-	-	-	-
Cl_7 (CB 191)	Group 1	-	-	-	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Cl_7 (CB 170/190)	Group 2	406.63	-0.15	0.71	132.3	-0.1	0.7	23.82	0.023	1.054
Cl_7 (CB 189)	Group 3	129.79	0.35	2.22	-	-	-	-	-	-
Cl_8 (CB 202)	Group 1	36.59	0.90	7.89	-	-	-	-	-	-
Cl_8 (CB 200)	Group I	0.35	2.92	828.08	-	-	-	-	-	-
Cl_8 (CB 204)	Group I	-	-	-	-	-	-	-	-	-
Cl_8 (CB 197)	Group I	2.82	2.01	102.20	-	-	-	-	-	-
Cl_8 (CB 199)	Group 5	10.77	1.43	26.79	-	-	-	-	-	-
Cl_8 (CB 198)	Group I	58.68	0.69	4.92	-	-	-	-	-	-
Cl_8 (CB 201)	Group I	903.35	-0.50	0.32	-	-	-	-	-	-
Cl_8 (CB 203/196)	Group I	152.67	0.28	1.89	-	-	-	-	-	-
Cl_8 (CB 195)	Group 2	1593.53	-0.74	0.18	-	-	-	-	-	-
Cl_8 (CB 194)	Group I	258.76	0.05	1.12	448.1	-0.7	0.2	-	-	-
Cl_8 (CB 205)	Group I	5758.65	-1.30	0.05	-	-	-	-	-	-
Cl_9 (CB 208)	Group 1	-	-	-	-	-	-	-	-	-
Cl_9 (CB 207)	Group I	-	-	-	-	-	-	-	-	-
Cl_9 (CB 206)	Group I	-	-	-	-	-	-	-	-	-
Cl_{10} (CB 209)	Group I	21.83	1.12	13.22	-	-	-	-	-	-
Chlorobenzenes (CBz)										
1,3,5 TriCBz		0.98	2.47	294.44	-	-	-	-	-	-
1,2,4 TriCBz		0.94	2.49	306.83	-	-	-	-	-	-
1,2,3 TriCBz		1.54	2.27	187.64	-	-	-	-	-	-
1,2,3,5/1,2,4,5 TeCBz		35.67	0.91	8.09	-	-	-	-	-	-

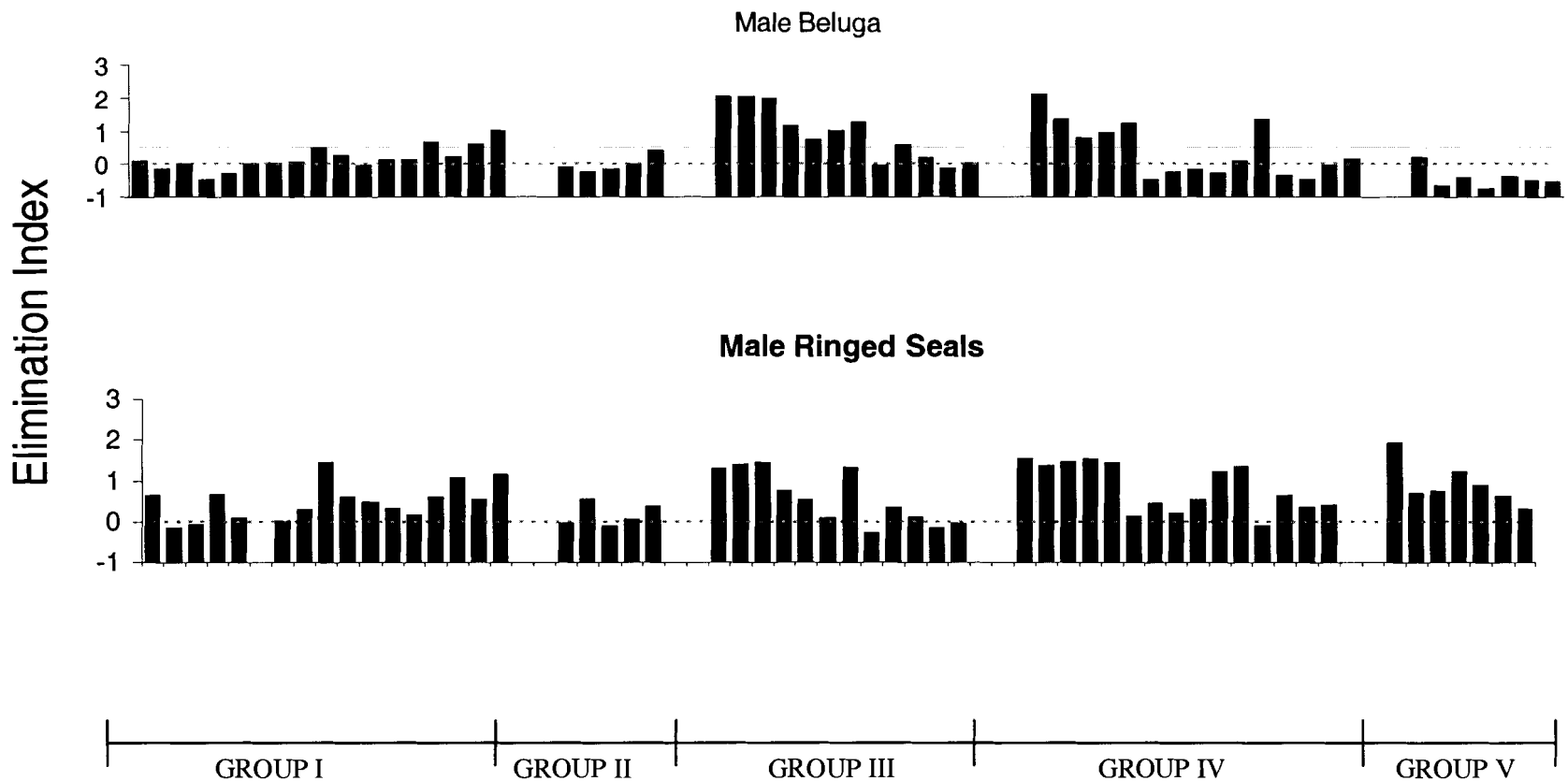
	Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
	BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
1,2,3,4 TeCBz	13.86	1.32	20.81	-	-	-	-	-	-
PeCBz	82.84	0.54	3.48	-	-	-	-	-	-
HCB	0.50	2.76	577.98	1.01	1.97	93.11	7.75	0.51	3.24
Hexachlorocyclohexanes (HCHs)									
α-HCH	10.90	1.42	26.47	12.24	0.89	7.68	11.17	0.35	2.25
β-HCH	17.92	1.21	16.10	4.17	1.35	22.53	-	-	-
γ-HCH	11.92	1.38	24.19	-	-	-	27.58	-0.04	0.91
Dichlordiphenyltrichloroethanes (DDTs)									
p,p-DDT	-	-	-	-	-	-	6.68	0.58	3.76
o,p-DDT	-	-	-	-	-	-	-	-	-
p,p-DDE	548.92	-0.28	0.53	2.06	1.66	45.66	69.96	-0.45	0.36
o,p-DDE	-	-	-	-	-	-	-	-	-
p,p-DDD	-	-	-	-	-	-	-	-	-
o,p-DDD	-	-	-	-	-	-	-	-	-
Cyclodienes									
aldrin	-	-	-	-	-	-	-	-	-
heptachlor	-	-	-	-	-	-	-	-	-
heptachlor epoxide	-	-	-	-	-	-	-	-	-
***trans*-chlordane**	-	-	-	-	-	-	-	-	-
***cis*-chlordane**	262.14	0.04	1.10	-	-	-	-	-	-
***trans*-nonachlor**	364.40	-0.10	0.79	1.81	1.72	51.92	9.70	0.41	2.59
***cis*-nonachlor**	-	-	-	-	-	-	13.76	0.26	1.82
oxychlordane	-	-	-	-	-	-	5.48	0.66	4.58
α-endosulfan	-	-	-	-	-	-	-	-	-

		Walrus / Mussels			Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
		BMF_{MAX}	289		BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	CB Group	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
β-endosulfan		-	-	-	-	-	-	-	-	-
endosulfan sulfate		-	-	-	-	-	-	-	-	-
dieldrin		130.80	0.34	2.21	2.99	1.50	31.46	-	-	-
methoxychlor		-	-	-	-	-	-	-	-	-
mirex		107.51	0.43	2.68				9.64	0.42	2.60
octachlorostyrene		-	-	-	-	-	-	-	-	-

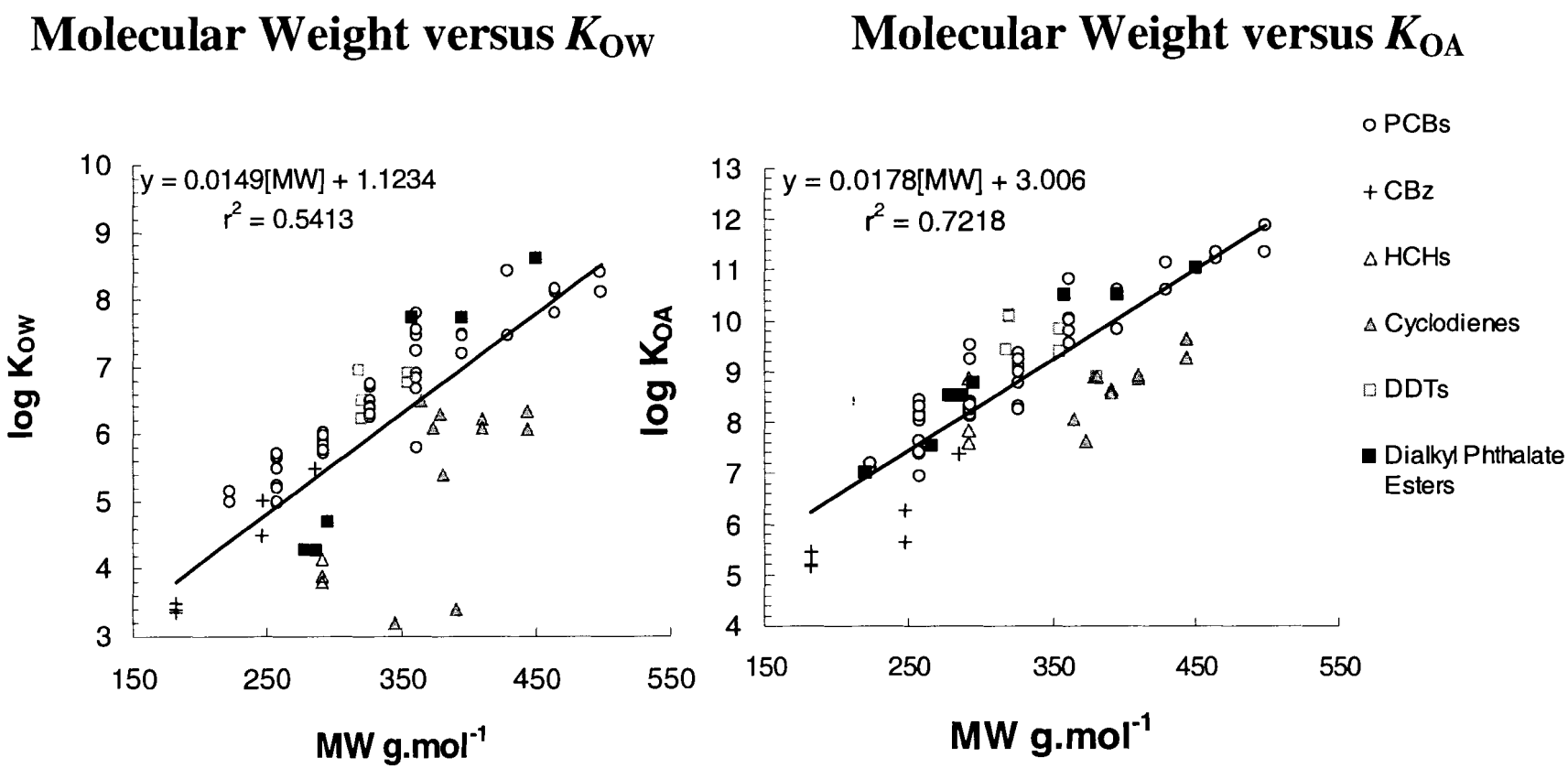
Appendix 9 Elimination index values for Group I to V PCB congeners in various organisms of the E. Hudson Bay marine food web.



Appendix 9 continued



Appendix 10 Relationship between molecular weight (MW g.mol^{-1}) and chemical log K_{OW} and log K_{OA} for several classes of organic contaminants.



Appendix 11 Results of log-linear regression analysis for log [POP] versus trophic level (TL) and corresponding food web magnification factors (FWMFs) for water-ventilating ectotherms, air-breathing endotherms and for the overall food web.

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)
1,3,5 TriCBz	0.02·(TP) -0.76 r^2 =-0.141	1.04 (0.70-1.55)	-0.06·(TP) -0.44 r^2 =-0.070	0.88 (0.55-1.40)	-0.05·(TP) - 0.50 r^2 =-0.043	0.89 (0.61-1.30)
1,2,4 TriCBz	0.16·(TP) + 0.001 r^2 =0.151	1.44 (1.21-1.70)	0.26·(TP) -0.25 r^2 =0.205	1.84 (1.44-2.35)	0.20·(TP) - 0.06 r^2 =0.132	1.60 (1.33-1.91)
1,2,3 TriCBz	0.11·(TP) -0.95 r^2 =-0.001	1.30 (1.00-1.68)	0.12·(TP) -0.78 r^2 =-0.017	1.33 (0.94-1.89)	0.16·(TP) - 1.01 r^2 =0.028	1.45 (1.12-1.88)
1,2,3,5/1,2,4,5 TeCBz	0.15·(TP) -1.05 r^2 =0.030	1.41 (1.07-1.85)	0.36·(TP) -1.09 r^2 =0.443	2.30 (1.88-2.82)	0.44·(TP) - 1.64 r^2 =0.335	2.74 (2.17-3.44)
1,2,3,4 TeCBz	0.16·(TP) -0.84 r^2 =-0.013	1.45 (0.95-2.23)	0.31·(TP) -0.77 r^2 =0.187	2.03 (1.49-2.77)	0.46·(TP) - 0.57 r^2 =0.250	2.86 (2.14-3.82)
PeCBz	0.43·(TP) -1.37 r^2 =0.546	2.68 (2.18-3.29)	0.55·(TP) -1.07 r^2 =0.457	3.54 (2.58-4.85)	0.78·(TP) - 2.24 r^2 =0.589	5.98 (4.68-7.66)
HCB	1.01·(TP) -2.23 r^2 =0.914	10.34 (8.84-12.09)	1.01·(TP) -2.14 r^2 =0.862	10.23 (7.52-13.91)	1.01·(TP) - 2.20 r^2 =0.856	10.30 (8.59-12.3)
α-HCH	0.23·(TP) + 0.37 r^2 =0.541	1.69 (1.53-1.86)	0.25·(TP) + 0.51 r^2 =0.206	1.78 (1.41-2.24)	0.29·(TP) + 0.25 r^2 =0.299	1.96 (1.68-2.29)
β-HCH	- 0.05·(TP) -0.07 r^2 =0.025	0.89 (0.80-0.99)	0.48·(TP) -0.58 r^2 =0.809	3.03 (2.72-3.38)	0.44·(TP) - 0.71 r^2 =0.437	2.78 (2.26-3.42)
γ-HCH	0.18·(TP) -0.17 r^2 =0.291	1.50 (1.30-1.74)	0.32·(TP) + 0.03 r^2 =0.144	2.09 (1.45-3.01)	0.56·(TP) - 1.18 r^2 =0.348	3.62 (2.71-4.84)
p,p-DDT	0.25·(TP) + -0.29 r^2 =0.668	1.77 (1.62-1.94)	0.58·(TP) -0.59 r^2=0.728	3.79 (3.13-4.58)	0.51·(TP) -0.74 r^2=0.505	3.26 (2.66-3.99)

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	**FWMF (95%CL)**	**$\log[C_B] = mx + b$**	**FWMF (95%CL)**	**$\log[C_B] = mx + b$**	**FWMF (95%CL)**
p,p-DDE	0.96·(TP) -1.82 r^2 =0.775	9.20 (7.14-11.9)	1.04·(TP) -1.61 r^2 =0.526	10.84 (6.35-18.5)	1.15·(TP) -2.25 r^2 =0.613	14.06 (10.0-19.7)
heptachlor epoxide	0.60·(TP)-1.43 r^2 =0.913	3.96 (3.28-4.78)	0.97·(TP) -1.78 r^2 =0.970	9.29 (8.37-10.3)	1.02·(TP) -2.15 r^2 =0.850	10.46 (8.30-13.19)
trans-chlordane	0.22·(TP) -0.44 r^2 =0.540	1.65 (1.31-2.10)	-	-	0.33·(TP) -0.66 r^2 =0.821	2.14 (1.92-2.38)
cis-chlordane	0.34·(TP) -0.41 r^2 =0.643	2.17 (1.88-2.50)	-	-	0.60·(TP) -0.82 r^2 =0.734	3.98 (3.39-4.68)
trans-nonachlor	0.94·(TP) -1.93 r^2 =0.854	8.66 (6.83-10.98)	1.07·(TP) -1.86 r^2 =0.834	11.84 (8.92-15.7)	1.17·(TP) -2.42 r^2 =0.852	14.84 (12.0-18.3)
dieldrin	0.70·(TP) -1.39 r^2 =0.959	4.98 (4.67-5.31)	0.82·(TP) -1.47 r^2 =0.541	6.65 (4.58-9.66)	0.80·(TP) -1.54 r^2 =0.542	6.25 (4.83-8.09)
mirex	0.36·(TP) -1.29 r^2 =0.405	2.29 (1.80-2.92)	0.64·(TP) -1.54 r^2 =0.625	4.37 (3.36-5.70)	0.84·(TP) -2.59 r^2 =0.556	6.87 (5.13-9.20)
Group I PCBs						
Cl_6 (CB 132/153)	0.84·(TP) - 1.68 r^2 =0.866	6.84 (6.20-7.56)	1.05·(TP) - 1.72 r^2 =0.864	11.33 (10.08-12.73)	1.04·(TP) - 2.10 r^2 =0.842	11.02 (9.93-12.22)
Cl_7 (CB 180)	0.67·(TP) - 1.86 r^2 =0.742	4.67 (4.12-5.28)	0.86·(TP) - 1.63 r^2 =0.836	7.19 (6.28-8.22)	0.91·(TP) - 2.23 r^2 =0.658	8.10 (6.90-9.49)
Cl_7 (CB 187/182)	0.45·(TP) - 1.37 r^2 =0.411	2.84 (2.40-3.36)	0.83·(TP) - 1.53 r^2 =0.778	6.80 (5.81-7.96)	0.83·(TP) - 2.05 r^2 =0.491	6.72 (5.48-8.25)
Cl_8 (CB 194)	0.53·(TP) - 2.26 r^2 =0.210	3.41 (2.33-5.00)	0.31·(TP) - 0.45 r^2 =0.203	2.06 (1.64-2.59)	0.83·(TP) - 2.82 r^2 =0.400	6.75 (5.13-8.89)
Cl_9 (CB 206)	0.22·(TP) - 1.31 r^2 =0.031	1.65 (1.13-2.43)	0.24·(TP) - 0.80 r^2 =0.115	1.74 (1.33-2.28)	0.44·(TP) - 1.77 r^2 =0.269	2.76 (2.18-3.48)

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)
Cl_{10} (CB 209)	0.28·(TP) - 1.58 r^2 =0.170	1.90 (1.55-2.34)	0.33·(TP) - 1.49 r^2 =0.528	2.14 (1.91-2.40)	0.35·(TP) - 1.67 r^2 =0.293	2.23 (1.94-2.57)
Group II PCBs						
Cl_5 (CB 99)	0.73·(TP) - 1.84 r^2 =0.595	5.31 (4.37-6.46)	0.83·(TP) - 1.29 r^2 =0.747	6.77 (5.66-8.10)	0.98·(TP) - 2.28 r^2 =0.593	9.64 (7.87-11.81)
Cl_6 (CB 160/163/164/138)	0.74·(TP) - 1.61 r^2 =0.764	5.49 (4.84-6.23)	0.91·(TP) - 1.73 r^2 =0.724	8.08 (7.25-9.00)	0.96·(TP) - 1.92 r^2 =0.700	9.18 (7.90-10.68)
Cl_7 (CB 170/190)	0.56·(TP) - 1.90 r^2 =0.646	3.59 (3.14-4.11)	0.79·(TP) - 1.76 r^2 =0.826	6.11 (5.38-6.94)	0.83·(TP) - 2.35 r^2 =0.599	6.76 (5.71-8.01)
Cl_8 (CB 195)	0.76·(TP) - 3.57 r^2 =0.159	5.73 (2.18-15.05)	0.31·(TP) - 1.19 r^2 =0.233	2.03 (1.65-2.49)	0.62·(TP)-2.58 r^2 =0.334	4.19 (3.14-5.57)
Group III PCBs						
Cl_3 (CB 28)	0.34·(TP) - 1.07 r^2 =0.429	2.17 (1.93-2.45)	0.43·(TP) - 1.18 r^2 =0.883	2.69 (2.54-2.85)	0.41·(TP) - 1.21 r^2 =0.598	2.57 (2.36-2.79)
Cl_4 (CB 66)	0.56·(TP) -1.81 r^2 =0.701	3.62 (3.14-4.18)	0.71·(TP) -1.70 r^2 =0.842	5.18 (4.62-5.82)	0.73·(TP) -2.00 r^2 =0.696	5.38 (4.70-6.17)
Cl_5 (CB 105)	0.59·(TP) -1.84 r^2 =0.736	3.87 (3.37-4.46)	0.80·(TP) -1.78 r^2 =0.865	6.27 (5.58-7.05)	0.81·(TP) -2.11 r^2 =0.706	6.49 (5.59-7.54)
Cl_5 (CB 106/118)	0.67·(TP) -1.75 r^2 =0.834	4.64 (4.13-5.21)	0.92·(TP) -1.58 r^2 =0.856	8.25 (7.17-9.49)	0.96·(TP) -2.15 r^2 =0.651	9.08 (7.48-11.02)
Group IV PCBs						
Cl_3 (CB 18)	0.21·(TP) -0.88 r^2 =0.405	1.62 (1.50-1.75)	0.30·(TP) -0.86 r^2 =0.415	2.01 (1.78-2.27)	0.31·(TP) -1.04 r^2 =0.357	2.04 (1.85-2.25)

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)
Cl_4 (CB 44)	0.29·(TP) -1.38, r^2 =0.549	1.94 (1.77-2.11)	0.65·(TP) -1.42 r^2 =0.656	4.45 (3.78-5.24)	0.66·(TP) -2.01 r^2 =0.416	4.61 (3.79-5.62)
Cl_4 (CB 73/52)	0.51·(TP) -1.64 r^2 =0.712	3.23 (2.93-3.57)	0.90·(TP) -1.73 r^2 =0.820	7.90 (6.86-9.10)	0.87·(TP) -2.18 r^2 =0.575	7.33 (6.13-8.76)
Cl_5 (CB 95)	0.39·(TP) -1.32 r^2 =0.228	2.47 (1.96-3.12)	0.74·(TP) -1.22 r^2 =0.499	5.49 (4.21-7.16)	0.81·(TP) -2.06 r^2 =0.347	6.45 (4.89-8.50)
Cl_5 CB (101/90)	0.52·(TP) -1.39 r^2 =0.378	3.31 (2.68-4.09)	0.92·(TP) -1.57 r^2 =0.822	8.36 (7.18-9.73)	.92·(TP) -2.10 r^2 =0.515	8.31 (6.67-10.34)
Cl_5 (CB 110)	0.40·(TP) -1.35 r^2 =0.213	2.50 (1.95-3.20)	0.69·(TP) -1.34 r^2 =0.644	4.93 (4.09-5.96)	0.77·(TP) -2.06 r^2 =0.416	5.83 (4.65-7.32)
Cl_6 (CB 149)	0.43·(TP) -1.39 r^2 =0.484	2.70 (2.35-3.09)	0.91·(TP) -1.72 r^2 =0.793	8.22 (7.04-9.59)	0.82·(TP) -1.98 r^2 =0.501	6.65 (5.46-8.11)
Group V PCBs						
Cl_7 (CB 179)	0.22·(TP) -1.42 r^2 =0.181	1.65 (1.43-1.91)	0.57·(TP) -1.47 r^2 =0.381	3.70 (2.84-4.82)	0.56·(TP) -1.98 r^2 =0.245	3.61 (2.82-4.63)
Cl_7 (CB 176)	0.22·(TP) -1.47 r^2 =0.380	1.64 (1.42-1.89)	0.46·(TP) -1.48 r^2 =0.346	2.88 (2.31-3.60)	0.47·(TP) -1.69 r^2 =0.312	2.96 (2.38-3.68)
Cl_7 (CB 185)	0.15·(TP) -1.38 r^2 =0.420	1.41 (1.30-1.54)	0.43·(TP) -1.54 r^2 =0.453	2.72 (2.30-3.22)	0.43·(TP) -1.73 r^2 =0.351	2.72 (2.27-3.25)
Cl_7 (CB 174/181)	0.31·(TP) -1.48 r^2 =0.255	2.06 (1.70-2.48)	0.61·(TP) -1.41 r^2 =0.456	4.10 (3.22-5.21)	0.66·(TP) -2.07 r^2 =0.319	4.61 (3.58-5.93)

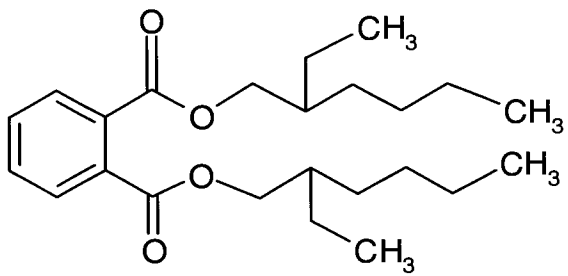
Appendix 12 Logarithms of POPs Bioaccumulation Factors (log BAF) for organisms of the E. Hudson's Bay marine food web, along with previously reported values of gas-phase air concentrations (C_{AG}, pg m^{-3}) and freely dissolved surface seawater concentrations (C_{WD} ng L^{-1}).

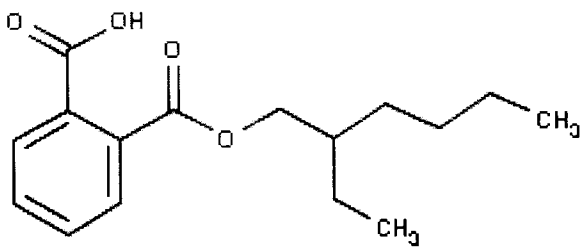
	C_{AG} (pg m^{-3})	C_{WD} (ng L^{-1})	Log BAF Cod/ Water	Log BAF Bivalves/ Water	Log BAF Eider Duck/ Air	Log BAF Male Beluga/ Air	Log BAF Male Ringed Seals/ Air	Log BAF Polar Bear/Air	Log BAF Humans/ Air
Chlorobenzenes (CBz)									
1,3,5 TriCBz	-		4.9	4.8		5.7	5.7		
1,2,4 TriCBz	-		5.7	5.9	6.0	7.4	7.6		
1,2,3 TriCBz	-		5.0	4.4	6.4	5.7	5.8		
1,2,3,5/1,2,4,5 TeCBz	4.7		5.7	5.0	6.3	6.9	7.3		
1,2,3,4 TeCBz	22.8		5.4	5.0	6.6	6.3	6.1		
PeCBz	67.2		6.8	6.5		7.4	7.1		
HCB	63.0	1.4	9.5	8.2	8.0	9.7	8.7	8.2	8.9
Hexachlorocyclohexanes (HCHs)									
α-HCH	61.0	2.20	6.6	6.2	7.4	8.7	9.1		
β-HCH	0.19	0.03	5.2	5.2	11.4	11.4	11.1	12.5	12.9
γ-HCH	7.60	0.28	5.4	5.0	7.5	9.6	9.1		
Dichlordiphenyltrichloroethanes (DDTs)									
p,p-DDT	0.15	0.002			11.1	12.5	11.5		12.0
o,p-DDT	0.21					12.3	10.5		
p,p-DDE	0.36	0.001	10.3	8.7	11.7	12.7	12.0	12.2	13.3
o,p-DDE	0.14		6.5			10.4	9.2		
p,p-DDD	0.055		10.5		11.6	12.7	11.4		
o,p-DDD	0.04					12.2	10.9		
Cyclodienes									
aldrin									
heptachlor	0.037		7.9			*10.4*	*10.0*		

	C_{AG} (pg m^{-3})	C_{WD} (ng L^{-1})	Log BAF Cod/ Water	Log BAF Bivalves/ Water	Log BAF Eider Duck/ Air	Log BAF Male Beluga/ Air	Log BAF Male Ringed Seals/ Air	Log BAF Polar Bear/Air	Log BAF Humans/ Air
heptachlor epoxide	1.1	8.40×10^{-3}	11.3			11.3	10.9		
octachlorostyrene									
trans-chlordane	0.31	9.40×10^{-4}				10.4	10.3		
cis-chlordane	0.95	8.80×10^{-4}			10.5	11.2	10.4		
trans-nonachlor	0.78	$7.0 \times 10\text{-}4$	8.2	7.3		12.0	11.0		
cis-nonachlor	0.11	3.0×10^{-4}	9.7		11.1	12.0	10.9		12.4
oxychlordane	0.61		10.2			12.1	11.6		
α-endosulfan		3.60×10^{-3}							
β-endosulfan	3.60	4.10×10^{-3}			9.2	9.5	8.8		
endosulfan sulfate									
dieldrin	1.10	1.20×10^{-2}	9.0	8.4	10.7	10.2	10.7	11.4	
methoxychlor	0.17								
mirex	0.072		6.7		10.4	11.5	10.7		11.9
Group I PCBs									
Cl_6 (CB 132/153)	0.68	1.7×10^{-4}	9.7	9.0	12.4	11.9	12.6	14.1	13.7
Cl_7 (CB 180)	0.58	3.0×10^{-5}	8.8	7.8	12.1	11.3	12.1	14.0	13.6
Cl_7 (CB 187/182)	0.40		10.2	9.8	12.9	11.4	12.7	13.2	14.1
Cl_8 (CB 194)	0.07		10.5	9.4	12.7	11.2	12.6	15.2	
Cl_9 (CB 206)	0.05		10.9		12.6	10.7	12.5		
Cl_{10} (CB 209)	0.06		11.4	10.4	13.0	10.3	12.7		
Group II PCBs									
Cl_5 (CB 99)	0.31			9.0	12.0	11.8	12.0	13.3	13.1
Cl_6 (CB 160/163/164/138)	0.34	1.6×10^{-4}	10.2	9.7	12.5	12.1	12.6	13.6	13.8

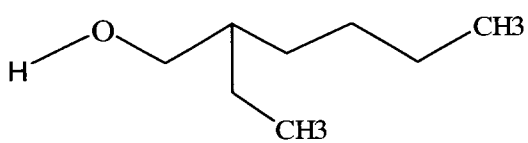
	C_{AG} (pg m^{-3})	C_{WD} (ng L^{-1})	Log BAF Cod/ Water	Log BAF Bivalves/ Water	Log BAF Eider Duck/ Air	Log BAF Male Beluga/ Air	Log BAF Male Ringed Seals/ Air	Log BAF Polar Bear/Air	Log BAF Humans/ Air
Cl_7 (CB 170/190)	0.16		10.4	9.2	12.7	11.4	12.7	14.6	14.2
Cl_8 (CB 195)	0.06		10.0	7.9	12.3	10.5	12.2		
Group III PCBs									
Cl_3 (CB 28)	1.16	1.8×10^{-4}	7.8	7.5	9.0	9.4	9.6		
Cl_4 (CB 66)	0.06					11.5			
Cl_5 (CB 105)	0.06	4.0×10^{-5}				11.9			
Cl_5 (CB 106/118)	0.53	1.0×10^{-4}	8.6	8.0	10.5	11.5	10.6	11.0	11.5
Group IV PCBs									
Cl_3 (CB 18)	5.16			6.3	8.2	8.9	7.5		
Cl_4 (CB 44)	0.67			8.4	11.2	10.7	10.4		
Cl_4 (CB 73/52)	1.55	1.7×10^{-4}	8.6	7.7	10.8	11.0	10.3	11.0	11.2
Cl_5 (CB 95)	1.11			8.5	11.3	11.2	10.2		
Cl_5 CB (101/90)	0.64	2.50× 10-4	9.2	8.7	11.6	11.6	11.5		11.8
Cl_5 (CB 110)	0.41	1.3× 10-4	9.2	8.6	11.0	11.1	10.6		
Cl_6 (CB 149)	0.70	1.80× 10-4	9.7	9.4	12.4	11.5	11.8	11.8	
Group V PCBs									
Cl_7 (CB 179)	0.12				11.6	11.2	9.8		
Cl_7 (CB 176)	0.09					10.8			
Cl_7 (CB 185)	0.09				11.1	10.6	10.1		
Cl_7 (CB 174/181)	0.21		8.1	7.4	11.6	11.1	10.6		

Appendix 13 Metabolic pathway of dialkyl phthalate ester (DPE) hydrolytic de-esterification.

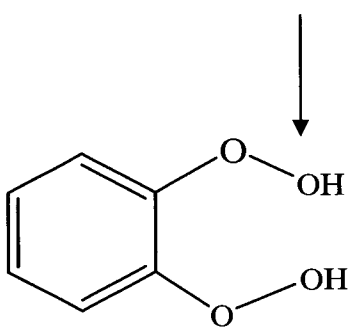




Mono (2-ethylhexyl) phthalate (MEHP)



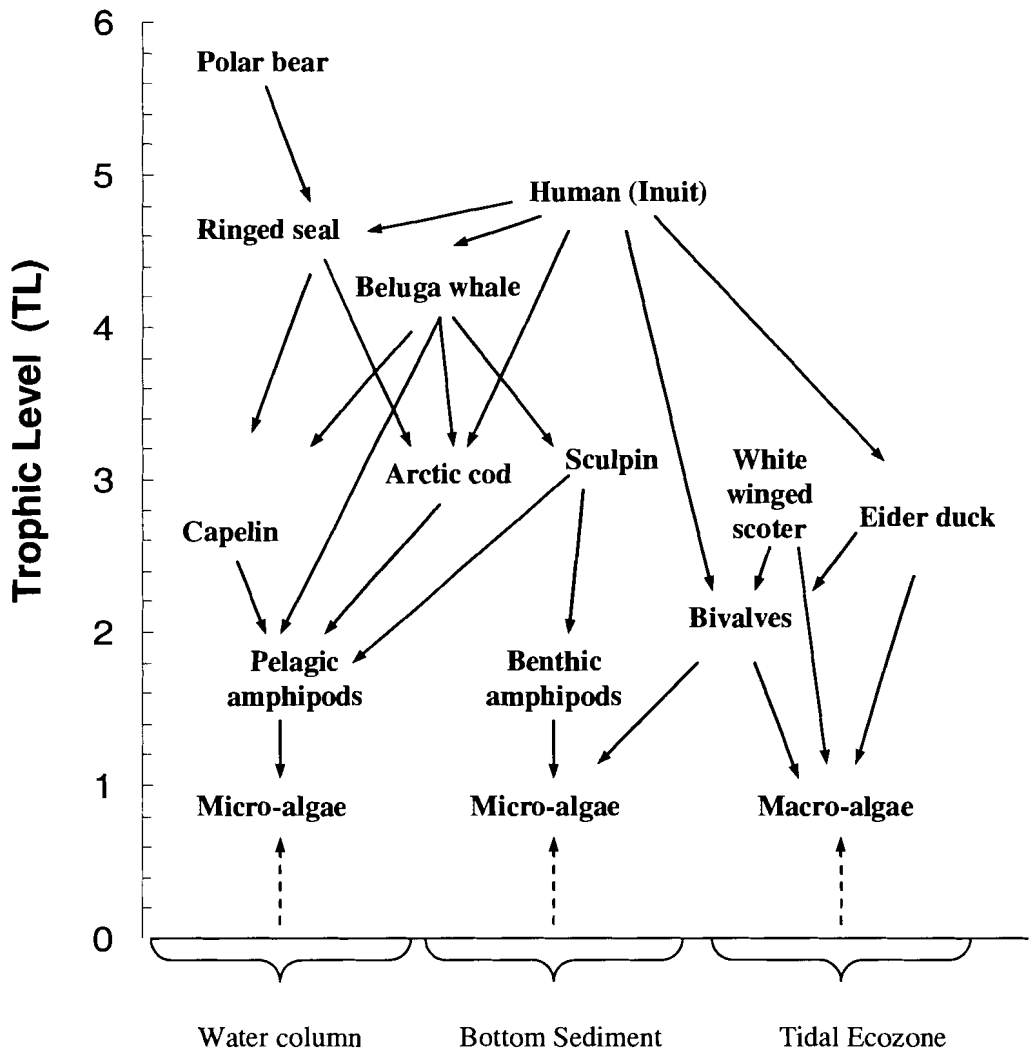
2-ethylhexanol (2-EH)



ω and ω-1 oxidation

Phthalic acid

Appendix 14 Conceptual illustration of E. Hudson Bay marine food web organisms and assigned trophic levels (TL).



Appendix 15 Dialkyl phthalate ester (DPE) concentrations in lichens and macro-algae (ng·g^{-1} lipid equivalent wt.), sediment (ng·g^{-1} OC wt) and tissues' of various marine biota (ng·g^{-1} lipid wt.) collected from E. Hudson's Bay during May and September 1999-2002.

	LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		**MACRO ALGAE** (*F. gardneri*) **(tissue)** (*n* =11)		**SEDIMENTS** (***n* ponar grabs)** (*n* =12)		**CAPELIN** (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10			
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Dialkyl phthalate esters								
DMP	4.87	*1.8-13.2*	38.68	*15.0-99.9*	115.5	*44.1-302.1*	16.1	*3.9-66.4*
DEP	77.79	*32.9-183.5*	288.90	*119.1-700.6*	9590.3	*1,492.3-61,631.6*	130.0	*45.5-371.1*
DIBP	35.61	*11.1-114.2*	103.60	*41.1-261.3*	258.3	*86.2-774.0*	18.4	*4.394-76.8*
DBP	196.77	*66.5-582.3*	1459.11	*171.3-12,430.2*	2569.5	*860.8-7,669.7*	201.7	*43.065-944.5*
BBP	33.77	*11.6-98.3*	306.17	*114.2-820.7*	557.0	*133.8-2,318.9*	30.2	*5.2-176.6*
DEHP	2980.2	*727.6-12,206.9*	5452.87	*1,347.2-22,07.*	1736.7	*304.3-9,910.9*	2457.0	*645.0-9,360.1*
DnOP	32.38	*9.6-109.3*	83.60	*22.8-306.5*	40.6	*8.2-201.4*	25.6	*8.857-74.2*
DnNP	43.77	*15.4-124.8*	129.31	*22.6-741.2*	30.8	*3.7-258.8*	34.9	*11.262-107.9*
∑DPEs	3561.7	*976.9-12,985.5*	9226.96	*1,962.2-43,386*	18,221	*3,853.7-86,143.5*	2989.0	*820.7-10,886.1*

	COD (*B. saida*) (muscle)		SCULPIN (*M. scorpioides*) (muscle)		MALE BELUGA (*D. leucas*) (Blubber)		MALE BELUGA (*D. leucas*) (Liver)	
	(*n* = 12)		(*n* = 12)		(*n* = 21)		(*n* = 14)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		89.4 ± 0.53		2.1 ± 0.44	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		89.4 ± 0.53		2.1 ± 0.44	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Dialkyl phthalate esters								
DMP	31.56	*6.19-160.9*	27.64	*6.83-111.8*	3.35	*1.068-10.53*	47.3	*14.6-153.6*
DEP	187.23	*54.95-637.9*	190.63	*62.98576.91*	221.14	*66.8-731.9*	541.4	*149.6-1,959*
DIBP	72.83	*17.45-303.8*	28.13	*7.53-105.1*	26.97	*8.2-88.1*	122.3	*25.82-579.3*
DBP	528.35	*105.46-2,646.8*	288.39	*74.06-1,123.1*	112.97	*29.5-432.1*	1594.7	*469.1-5,420*
BBP	139.12	*43.71-442.8*	55.34	*11.31-270.77*	44.47	*8.613-229.56*	281.6	*70.64-1,122*
DEHP	2241.04	*646.9-7,763.2*	2973.62	*650.4-13,595.4*	1553.72	*162.6-14,843.4*	1313.3	*71.8-24,009*
DnOP	35.06	*5.58-220.0*	42.42	*5.8-307.3*	85.96	-	53.2	*8.608-329.3*
DnNP	71.51	*10.939-467.4*	98.70	*16.1-604.8*	-	-	61.5	*8.05-469.9*
∑DPEs	3105.81	*815.34-11,830.36*	3914.27	*971.7-15,767.6*	2368.93	*356.4-15,742.6*	6510.0	*999 -42,430*

Appendix 16 Regression results and FWMFs for DPEs in water-ventilating and air-breathing organisms and overall food web.

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)
Dialkyl Phthalate Esters						
DMP	0.29·(TP) + 0.51, r^2 =0.587	1.95 (1.74-2.17)	-0.24·(TP) + 1.05, r^2 =0.579	0.58 (0.52-0.64)	-0.07·(TP) + 1.08, r^2 =-0.005	0.84 (0.70-1.02)
DEP	0.19·(TP) + 1.67, r^2 =0.602	1.53 (1.44-1.63)	-0.01·(TP) + 1.87, r^2 =-0.052	0.99 (0.89-1.10)	0.06·(TP) + 1.89, r^2 =0.030	1.16 (1.05-1.28)
DIBP	0.10·(TP) + 1.38, r^2 =0.096	1.25 (1.12-1.40)	-0.16·(TP) + 1.64, r^2 =0.372	0.69 (0.62-0.76)	-0.07·(TP) + 1.68, r^2 =0.017	0.85 (0.75-0.96)
DBP	0.09·(TP) + 2.31, r^2 =0.039	1.24 (1.07-1.43)	-0.26·(TP) + 2.66, r^2 =0.446	0.55 (0.48-0.64)	-0.13·(TP) + 2.70, r^2 =0.064	0.74 (0.63-0.87)
BBP	0.12·(TP) + 1.57, r^2 =0.143	1.32 (1.18-1.48)	-0.16·(TP) + 1.84, r^2 =0.291	0.69 (0.61-0.78)	-0.06·(TP) + 1.89, r^2 =0.002	0.87 (0.76-1.00)
DEHP	0.05·(TP) + 3.24, r^2 =0.000	1.12 (1.00-1.26)	-0.18·(TP) + 3.49, r^2 =0.213	0.65 (0.55-0.77)	-0.10·(TP) + 3.50, r^2 =0.041	0.80 (0.69-0.92)
DnOP	0.05·(TP) + 1.35, r^2 =-0.009	1.13 (0.98-1.29)	0.02·(TP) + 1.38, r^2 =-0.072	1.04 (0.89-1.22)	0.04·(TP) + 1.37, r^2 =-0.014	1.10 (0.97-1.25)
DnNP	0.11·(TP) + 1.44, r^2 =0.083	1.30 (1.12-1.52)	-0.24·(TP) + 1.05, r^2 =0.579	0.58 (0.52-0.64)	-0.07·(TP) + 1.08, r^2 =-0.005	1.30 (1.12-1.52)

Appendix 17 Polybrominated diphenyl ether (PBDE) congener concentrations lichens and macro-algae (ng·g^{-1} lipid equivalent wt.), sediment (ng·g $^{-1}$ OC wt) and tissues' of various marine biota (ng·g^{-1} lipid wt.) collected from E. Hudson Bay during May and September 1999-2002.

	LICHEN (*C. rangiferina*) **(tissue)** (*n* =11)		**MACRO ALGAE** (*F. gardneri*) **(tissue)** (*n* =11)		**SEDIMENTS** (***n* ponar grabs**) (*n* =12)		**CAPELIN** (*M. villosus*) **(whole body)** (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_1 (BDE 1)	-	-	-	-	-	-	-	-
Br_1 (BDE 2)	-	-	-	-	-	-	-	-
Br_1 (BDE 3)	-	-	-	-	-	-	-	-
Br_2 (BDE 10)	-	-	-	-	-	-	-	-
Br_2 (BDE 7)	-	-	-	-	-	-	-	-
Br_2 (UI DiBDE 1)	-	-	-	-	-	-	-	-
Br_2 (BDE 8/11)	-	-	-	-	-	-	-	-
Br_2 (BDE 12)	-	-	-	-	-	-	-	-
Br_2 (BDE 13)	-	-	-	-	-	-	-	-
Br_2 (BDE 15)	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 1)	-	-	-	-	-	-	-	-
Br_3 (BDE 32)	-	-	-	-	-	-	-	-
Br_3 (BDE 17)	-	-	-	-	-	-	-	-
Br_3 (BDE 25)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 2)	-	-	-	-	-	-	-	-
Br_3 (BDE 28/33)	-	-	2.32	*0.80-6.69*	-	-	-	-
Br_3 (BDE 35)	-	-	-	-	-	-	-	-

	LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_3 (BDE 37)	-	-	-	-	-	-	-	-
Br_4 (BDE 75)	-	-	-	-	-	-	-	-
Br_4 (BDE 49)	0.36	-	7.90	*2.42-25.79*	7.64	*1.43-40.83*	0.70	*0.32-1.50*
Br_4 (BDE 71)					-	-	-	-
Br_4 (BDE 47)	3.59	*1.14-11.28*	68.13	*8.79-527.60*	30.50	*4.31-215.82*	7.25	*3.02-17.41*
Br_4 (BDE 66)	-	-	6.08	*1.94-19.09*	4.78	*0.73-31.07*	-	-
Br_4 (BDE 77)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 1)	-	-	-	-	-	-	0.31	
Br_5 (UI PeBDE 2)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 3)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 4)	-	-	-	-	-	-	2.53	*0.91-7.02*
Br_5 (UI PeBDE 5)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 6)	-	-	-	-	-	-	-	-
Br_5 (BDE 100)	0.84	*0.26-2.74*	29.77	*3.19-278.01*	17.19	*2.87-102.99*	1.70	*0.72-3.99*
Br_5 (BDE 101)	-	-	3.34	*1.17-9.55*	-	-	-	-
Br_5 (BDE 119)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 7)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 8)	-	-	1.42	-	-	-	-	-
Br_5 (BDE 99)	4.07	*1.31-12.68*	159.89	*16.35-1,563.2*	67.62	*9.49-481.54*	5.16	*2.20-12.11*
Br_5 (BDE 116)	-	-	-	-	-	-	-	-
Br_5 (BDE 118)	-	-	2.39	-	-	-	-	-

	LICHEN (*C. rangiferina*) (tissue) (*n* =11)		MACRO ALGAE (*F. gardneri*) (tissue) (*n* =11)		SEDIMENTS (*n* ponar grabs) (*n* =12)		CAPELIN (*M. villosus*) (whole body) (*n* =8)	
% OC ± *SD*	-		-		0.18 ± 0.10		-	
% Lipid ± *SD*	0.525 ± 0.065		0.845 ± 0.21		-		1.41 ± 0.15	
% Lipid Equivalent (L_{Eq}) ± *SD*	2.30 ± 0.01		1.63 ± 0.20		0.06 ± 0.04		2.81 ± 0.15	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_5 (BDE 85)	-	-	14.38	*4.32-47.83*	11.14	*1.51-81.98*	-	-
Br_5 (BDE 126)	-	-	-	-	-	-	-	-
Br_5 (BDE 105)	-	-	-	-	-	-	-	-
Br_5 (BDE 155)			2.29	-	-	-	-	-
Br_5 (BDE 154)	0.41	*0.13-1.37*	33.31	*10.83-102.50*	10.79	*1.87-62.16*	2.43	*0.85-6.90*
Br_6 (UI HxBDE 1)	-	-	-	-	-	-	-	-
Br_6 (UI HxBDE 2)	0.90	-	9.14	*0.43-195.52*	9.31	*1.56-55.47*	0.44	-
Br_6 (BDE 153)	0.49	*0.14-1.62*	21.71	*2.26-208.83*	11.23	*1.89-66.83*	-	-
Br_6 (BDE 140)	-	-	2.89	*0.92-9.03*	7.42		-	-
Br_6 (BDE 138/166)	-	-	1.92	*0.63-5.86*	5.01		-	-
Br_7 (BDE 183)	-	-	3.14	-	-	-	-	-
Br_7 (BDE 181)	-	-	-	-	-	-	-	-
Br_7 (BDE 190)	-	-	-	-	-	-	-	-
$\sum Br_3$-BDEs	-	-	2.32	*0.80-6.69*	-	-	-	-
$\sum Br_4$-BDEs	3.61	*1.13-11.54*	75.44	*9.33-610.05*	31.86	*4.31-235.3*	7.65	*3.23-18.11*
$\sum Br_5$-BDEs	4.76	*1.52-14.91*	198.15	*20.03-1,960.2*	81.74	*10.94-610.6*	9.52	*4.09-22.14*
$\sum Br_6$-BDEs	0.84	*0.20-3.49*	47.88	*4.62-495.84*	27.88	*4.54-171.02*	2.63	*1.05-6.56*
$\sum Br_7$-BDEs	-	-	0.56	*0.025-12.79*	-	-	-	-
$\sum$BDEs	9.26	*2.87-29.81*	323.89	*34.38-3,050.6*	81.80	*7.50-891.57*	18.06	*7.72-42.2*

Appendix 17 continued.

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_1 (BDE 1)	-	-	-	-	-	-	-	-
Br_1 (BDE 2)	-	-	-	-	-	-	-	-
Br_1 (BDE 3)	-	-	-	-	-	-	-	-
Br_2 (BDE 10)	-	-	-	-	-	-	-	-
Br_2 (BDE 7)	-	-	-	-	-	-	-	-
Br_2 (UI DiBDE 1)	-	-	-	-	-	-	-	-
Br_2 (BDE 8/11)	-	-	-	-	-	-	-	-
Br_2 (BDE 12)	-	-	-	-	-	-	-	-
Br_2 (BDE 13)	-	-	-	-	-	-	-	-
Br_2 (BDE 15)	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 1)	-	-	-	-	-	-	-	-
Br_3 (BDE 32)	-	-	-	-	-	-	-	-
Br_3 (BDE 17)	-	-	0.66	-	-	-	-	-
Br_3 (BDE 25)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 2)	-	-	-	-	-	-	-	-
Br_3 (BDE 28/33)	0.30	*0.073-1.24*	1.07	*0.24-4.86*	0.19	*0.068-0.54*	-	-
Br_3 (BDE 35)	-	-	-	-	-	-	-	-
Br_3 (BDE 37)	-	-	-	-	-	-	-	-
Br_4 (BDE 75)	-	-	0.32		0.09		-	-
Br_4 (BDE 49)	0.61	*0.14-2.60*	3.10	*0.66-14.62*	0.43	*0.16-1.19*	0.34	*0.12-0.96*

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_4 (BDE 71)	-	-	0.38	*0.10-1.42*	-	-	-	-
Br_4 (BDE 47)	5.28	*1.53-18.27*	25.96	*3.04-221.8*	3.55	*1.15-10.97*	2.89	*0.74-11.35*
Br_4 (BDE 66)	1.04	*0.43-2.54*	2.38	*0.44-13.01*	0.25	*0.10-0.65*	0.29	*0.073-1.12*
Br_4 (BDE 77)	-	-	0.01		-	-	-	-
Br_5 (UI PeBDE 1)	-	-	0.23		0.17	*0.058-0.47*	-	-
Br_5 (UI PeBDE 2)	-	-	0.53		0.07	*0.030-0.18*	-	-
Br_5 (UI PeBDE 3)	-	-					-	-
Br_5 (UI PeBDE 4)	0.19	-	0.97		0.59	*0.20-1.73*	-	-
Br_5 (UI PeBDE 5)	-	-	0.31	*0.035-2.72*	0.03	*0.004-0.15*	-	-
Br_5 (UI PeBDE 6)	-	-					-	-
Br_5 (BDE 100)	1.29	*0.35-4.76*	8.71	*0.99-76.01*	1.01	*0.32-3.21*	0.94	*0.227-3.886*
Br_5 (BDE 101)	-	-	0.69	*0.16-3.01*	0.17	*0.058-0.48*	-	-
Br_5 (BDE 119)	-	-	-	-	0.06	*0.018-0.18*	-	-
Br_5 (UI PeBDE 7)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 8)	-	-	0.65	-	-	-	-	-
Br_5 (BDE 99)	2.10	*0.55-8.02*	27.79	*1.98-390.4*	2.07	*0.69-6.28*	2.64	*0.40-17.29*
Br_5 (BDE 116)					-	-	-	-
Br_5 (BDE 118)			0.61	*0.27-1.39*	-	-	-	-
Br_5 (BDE 85)	0.27	*0.067-1.08*	3.89	*1.10-13.77*	-	-	0.53	*0.15-1.85*
Br_5 (BDE 126)	-	-	-	-	-	-	-	-
Br_5 (BDE 105)	-	-	-	-	-	-	-	-
Br_5 (BDE 155)	-	-	0.95		-	-	-	-

	COD (*B. saida*) **(muscle)** (*n* = 12)		SCULPIN (*M. scorpioides*) **(muscle)** (*n* = 12)		SALMON (*Salmo sp.*) **(muscle)** (*n* = 7)		BIVALVES (*M. edulis*) **(muscle)** (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
Br_5 (BDE 154)	0.39	*0.095-1.59*	6.47	*1.55-27.075*	0.71	*0.16-3.12*	0.22	*0.057-0.83*
Br_6 (UI HxBDE 1)	-	-	-	-	-	-	-	-
Br_6 (UI HxBDE 2)	0.16	*0.049-0.52*	8.63	*2.92-25.47*	0.26	*0.079-0.86*	0.40	*0.13-1.22*
Br_6 (BDE 153)	0.24	*0.060-0.94*	8.08	*2.16-30.16*	0.36	*0.12-1.04*	0.46	*0.16-1.28*
Br_6 (BDE 140)	-	-	0.50	*0.22-1.161*	-	-	-	-
Br_6 (BDE 138/166)	-	-	0.63	*0.16-2.56*	-	-	-	-
Br_7 (BDE 183)	-	-	-	-	-	-	-	-
Br_7 (BDE 181)	-	-	-	-	-	-	-	-
Br_7 (BDE 190)	-	-	-	-	-	-	-	-
$\sum Br_3$-BDEs	0.30	*0.073-1.243*	1.11	*0.235-5.27*	0.19	*0.068-0.514*		
$\sum Br_4$-BDEs	5.80	*1.62-20.845*	28.54	*3.23-252.24*	4.13	*1.36-12.51*	3.10	*0.77-12.39*
$\sum Br_5$-BDEs	3.47	*0.93-12.967*	36.11	*2.69-484.91*	3.95	*1.32-11.75*	3.41	*0.52-22.32*
$\sum Br_6$-BDEs	0.46	*0.099-2.15*	14.73	*2.52-86.07*	1.09	*0.30-3.96*	0.47	*0.070-3.19*
$\sum Br_7$-BDEs	-	-	-	-	-	-	-	-
$\sum$BDEs	9.78	*2.62-35.79*	72.82	*6.44-823.4*	9.28	*3.032-28.4*	5.38	*0.86-33.46*

Appendix 17 continued.

	MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		**FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)**		**FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)**		**BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)**	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_1 (BDE 1)	-	-	-	-	-	-	-	-
Br_1 (BDE 2)	-	-	0.024	-	-	-	-	-
Br_1 (BDE 3)	-	-	0.029	-	-	-	-	-
Br_2 (BDE 10)	-	-	-	-	-	-	-	-
Br_2 (BDE 7)	-	-	-	-	-	-	-	-
Br_2 (UI DiBDE 1)	0.01	*0.003-0.034*	0.004	*0.001-0.009*	-	-	-	-
Br_2 (BDE 8/11)	-	-	-	-	-	-	-	-
Br_2 (BDE 12)	-	-	-	-	-	-	-	-
Br_2 (BDE 13)	-	-	0.001	-	-	-	-	-
Br_2 (BDE 15)	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	0.01	-	0.004	-	-	-	-	-
Br_3 (UI TriBDE 1)	-	-	-	-	-	-	0.01	-
Br_3 (BDE 32)	0.01	*0.006-0.033*	-	-	0.04	-	0.01	-
Br_3 (BDE 17)	0.06	*0.020-0.16*	0.05	*0.018-0.16*	0.05	*0.016-0.15*	0.10	*0.031-0.29*
Br_3 (BDE 25)	0.01	*0.006-0.025*	0.01	*0.004-0.042*	-	-	0.02	*0.006-0.041*
Br_3 (UI TriBDE 2)	0.04	*0.014-0.12*	0.04	*0.010-0.12*	0.03	-	0.06	*0.013-0.241*
Br_3 (BDE 28/33)	0.48	*0.17-1.38*	0.24	*0.077-0.76*	0.19	*0.072-0.516*	0.51	*0.142-1.811*
Br_3 (BDE 35)	0.12	*0.040-0.35*	0.05	-	-	-	0.18	-
Br_3 (BDE 37)	0.02	-	-	-	-	-	-	-
Br_4 (BDE 75)	0.14	*0.061-0.34*	0.09	*0.033-0.26*	0.16	*0.047-0.56*	0.19	*0.029-1.21*

	MALE BELUGA (Age 16-35) (*D. leucas*) (Blubber) (*n* = 21)		FEMALE BELUGA (Age 5-35) (*D. leucas*) (Blubber) (*n* = 14)		FEMALE BELUGA (*D. leucas*) (Milk) (*n* = 8)		BELUGA CALVES (*D. leucas*) (Blubber) (*n* = 9)	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_4 (BDE 49)	1.53	*0.34-6.93*	1.05	*0.18-6.15*	1.01	*0.28-3.64*	2.16	*0.41-11.39*
Br_4 (BDE 71)	-	-	-	-	-	-	-	-
Br_4 (BDE 47)	15.16	*5.54-41.52*	6.59	*1.68-25.83*	3.90	*1.27-11.91*	11.31	*2.42-52.79*
Br_4 (BDE 66)	0.34	*0.12-0.95*	0.21	*0.071-0.62*	0.17	*0.057-0.49*	0.37	*0.098-1.42*
Br_4 (BDE 77)	0.02	*0.006-0.064*	0.01	*0.003-0.084*	0.01	*0.004-0.047*	0.05	*0.010-0.21*
Br_5 (UI PeBDE 1)	1.10	*0.33-3.69*	0.49	*0.13-1.79*	0.32	*0.11-0.99*	0.95	*0.18-4.95*
Br_5 (UI PeBDE 2)	0.33	*0.099-1.12*	0.22	*0.047-1.02*	0.08	*0.029-0.20*	0.19	*0.032-1.12*
Br_5 (UI PeBDE 3)	0.98	*0.37-2.56*	0.15	*0.031-0.68*	0.05	*0.010-0.21*	0.27	*0.026-2.85*
Br_5 (UI PeBDE 4)	5.63	*1.81-17.46*	1.88	*0.48-7.28*	1.03	*0.35-3.02*	3.91	*0.75-20.54*
Br_5 (UI PeBDE 5)	0.05	*0.011-0.219*	0.05	*0.008-0.273*	0.06	*0.018-0.19*	0.11	*0.025-0.466*
Br_5 (UI PeBDE 6)	0.18	*0.069-0.450*	0.10	*0.027-0.359*	-	-	0.21	*0.042-1.056*
Br_5 (BDE 100)	3.07	*1.04-9.09*	1.59	*0.436-5.799*	0.91	*0.28-2.99*	2.23	*0.43-11.74*
Br_5 (BDE 101)	0.39	*0.096-1.57*	0.28	*0.060-1.307*	0.20	*0.051-0.81*	0.59	*0.081-4.338*
Br_5 (BDE 119)	0.13	*0.042-0.38*	0.06	*0.016-0.220*	0.05	*0.015-0.19*	0.13	*0.017-0.922*
Br_5 (UI PeBDE 7)	0.09	*0.026-0.34*	0.03	*0.006-0.111*	0.05	*0.016-0.16*	0.24	*0.084-0.706*
Br_5 (UI PeBDE 8)	0.03	*0.006-0.15*	0.05	*0.013-0.177*	0.03	-	0.07	*0.012-0.392*
Br_5 (BDE 99)	2.34	*0.79-6.9*	1.54	*0.372-6.369*	1.03	*0.29-3.64*	2.02	*0.425-9.59*
Br_5 (BDE 116)	-	-	0.01	*0.001-0.037*	-	-	0.04	-
Br_5 (BDE 118)	0.09	*0.027-0.28*	0.04	*0.010-0.204*	0.02	-	0.07	*0.010-0.44*
Br_5 (BDE 85)	-	-	0.02	*0.003-0.086*	-	-	0.06	*0.029-0.13*
Br_5 (BDE 126)	0.25	*0.076-0.79*	0.11	*0.021-0.55*	-	-	0.19	*0.017-2.00*

	MALE BELUGA (Age 16-35) (*D. leucas*) **(Blubber)** (*n* = 21)		**FEMALE BELUGA (Age 5-35)** (*D. leucas*) **(Blubber)** (*n* = 14)		**FEMALE BELUGA** (*D. leucas*) **(Milk)** (*n* = 8)		**BELUGA CALVES** (*D. leucas*) **(Blubber)** (*n* = 9)	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_5 (BDE 105)	0.02	-	-	-	-	-	-	-
Br_5 (BDE 155)	0.15	*0.017-1.224*	0.06	*0.003-1.235*	0.09	*0.026-0.323*	0.03	*0.001-0.885*
Br_5 (BDE 154)	1.59	*0.479-5.277*	0.98	*0.249-3.869*	0.42	*0.117-1.494*	0.76	*0.122-4.747*
Br_6 (UI HxBDE 1)	0.06	*0.017-0.194*	0.02	*0.002-0.268*	0.03	*0.010-0.073*	0.03	*0.003-0.343*
Br_6 (UI HxBDE 2)	0.07	*0.004-1.307*	0.10	*0.005-1.923*	0.18	*0.050-0.656*	0.06	*0.003-0.903*
Br_6 (BDE 153)	0.79	*0.213-2.950*	0.50	*0.132-1.918*	0.21	*0.060-0.764*	0.42	*0.074-2.408*
Br_6 (BDE 140)	0.01	*0.002-0.040*	0.01	*0.002-0.035*	-	-	0.01	*0.002-0.071*
Br_6 (BDE 138/166)	0.003	*0.001-0.008*	0.01	*0.002-0.023*	0.01	-	0.00	*0.000-0.022*
Br_7 (BDE 183)	0.08	*0.022-0.265*	0.07	*0.019-0.225*	0.05	*0.019-0.120*	0.07	*0.015-0.284*
Br_7 (BDE 181)	-	-	0.01	*0.003-0.024*	0.01		0.01	*0.002-0.063*
Br_7 (BDE 190)	0.01	-	-	-	-	-	0.05	-
$\sum Br_3$-BDEs	0.63	*0.23-1.72*	0.30	*0.092-0.96*	0.23	*0.086-0.60*	0.67	*0.18-2.37*
$\sum Br_4$-BDEs	16.03	*5.75-44.68*	7.62	*2.03-28.57*	4.96	*1.59-15.44*	13.99	*3.04-64.34*
$\sum Br_5$-BDEs	14.05	*4.84-40.82*	6.25	*1.62-24.05*	3.70	*1.19-11.48*	10.36	*2.02-53.13*
$\sum Br_6$-BDEs	2.99	*0.86-10.40*	1.93	*0.496-7.50*	0.87	*0.24-3.12*	1.51	*0.272-8.34*
$\sum Br_7$-BDEs	0.08	*0.022-0.26*	0.07	*0.020-0.23*	0.04	*0.010-0.14*	0.07	*0.015-0.35*
$\sum$BDEs	34.09	*12.08-96.13*	16.09	*4.42-58.61*	9.86	*3.18-30.50*	26.76	*5.57-128.45*

Appendix 17 continued.

	MALE BELUGA BLOOD (*D. leucas*)		MALE BELUGA LIVER (*D. leucas*)		FEMALE BELUGA BLOOD (*D. leucas*)		FEMALE BELUGA LIVER (*D. leucas*)	
	(*n* = 7)		(*n* = 16)		(*n* = 7)		(*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_1 (BDE 1)	-	-	-	-	-	-	-	-
Br_1 (BDE 2)	-	-	-	-	-	-	-	-
Br_1 (BDE 3)	-	-	-	-	-	-	-	-
Br_2 (BDE 10)	-	-	-	-	-	-	-	-
Br_2 (BDE 7)	-	-	-	-	-	-	-	-
Br_2 (UI DiBDE 1)	-	-	-	-	-	-	-	-
Br_2 (BDE 8/11)	-	-	-	-	-	-	-	-
Br_2 (BDE 12)	-	-	-	-	-	-	-	-
Br_2 (BDE 13)	-	-	-	-	-	-	-	-
Br_2 (BDE 15)	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 1)	-	-	-	-	-	-	-	-
Br_3 (BDE 32)	-	-	0.5	*0.17-1.48*	-	-	-	-
Br_3 (BDE 17)	-	-	-	-	-	-	-	-
Br_3 (BDE 25)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 2)	-	-	-	-	-	-	-	-
Br_3 (BDE 28/33)	1.7	-	0.4	*0.16-1.23*	-	-	-	-
Br_3 (BDE 35)	-	-	-	-	-	-	-	-
Br_3 (BDE 37)	-	-	-	-	-	-	-	-
Br_4 (BDE 75)	-	-	1.4	-	-	-	-	-

	MALE BELUGA BLOOD (*D. leucas*)		MALE BELUGA LIVER (*D. leucas*)		FEMALE BELUGA BLOOD (*D. leucas*)		FEMALE BELUGA LIVER (*D. leucas*)	
	(*n* = 7)		(*n* = 16)		(*n* = 7)		(*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_4 (BDE 49)	-	-	1.2	*0.25-5.29*	-	-	2.1	-
Br_4 (BDE 71)	-	-			-	-	-	-
Br_4 (BDE 47)	10.1	*1.81-56.04*	15.4	*5.0-47.48*	9.6	-	9.6	*2.93-31.14*
Br_4 (BDE 66)	-	-	-	-	-	-	-	-
Br_4 (BDE 77)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 1)	2.5	-	0.7	*0.27-2.04*	-	-	0.6	-
Br_5 (UI PeBDE 2)	-	-	0.3	*0.14-0.83*	-	-	0.4	-
Br_5 (UI PeBDE 3)	-	-	-	-	-	-		-
Br_5 (UI PeBDE 4)	11.2	-	1.8	*0.45-6.88*	0.9		1.0	*0.39-2.28*
Br_5 (UI PeBDE 5)	-	-	0.3	*0.10-0.63*	-	-		
Br_5 (UI PeBDE 6)	-	-	0.2		-	-		
Br_5 (BDE 100)	2.5	*0.41-15.43*	3.8	*1.35-10.48*	1.9	*0.77-4.55*	2.6	*0.86-8.01*
Br_5 (BDE 101)	2.0	-	0.6	*0.19-1.83*	-	-	-	-
Br_5 (BDE 119)	-	-	0.6	-	-	-	-	-
Br_5 (UI PeBDE 7)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 8)	-	-	0.2	*0.10-0.46*	-	-	-	-
Br_5 (BDE 99)	3.8	*0.58-24.37*	4.3	*1.36-13.61*	4.6	*1.91-11.21*	3.4	*1.11-10.18*
Br_5 (BDE 116)	-	-	-	-	-	-	-	-
Br_5 (BDE 118)	-	-	0.2	-	-	-	-	-
Br_5 (BDE 85)	3.4	-	-	-	0.3	-	-	-
Br_5 (BDE 126)	-	-	-	-	-	-	-	-

	MALE BELUGA BLOOD (*D. leucas*)		MALE BELUGA LIVER (*D. leucas*)		FEMALE BELUGA BLOOD (*D. leucas*)		FEMALE BELUGA LIVER (*D. leucas*)	
	(*n* = 7)		(*n* = 16)		(*n* = 7)		(*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_5 (BDE 105)	-	-	-	-	-	-	-	-
Br_5 (BDE 155)	-	-	0.5	*0.18-1.36*	-	-	-	-
Br_5 (BDE 154)	6.4	*0.7-59.53*	2.2	*0.82-5.76*	1.3	-	2.2	*0.97-5.09*
Br_6 (UI HxBDE 1)	-	-	0.1	*0.053-0.31*	-	-	-	-
Br_6 (UI HxBDE 2)	4.8	-	0.9	*0.28-3.18*	0.6	-	0.4	*0.18-1.06*
Br_6 (BDE 153)	5.3	-	0.7	*0.19-2.19*	0.7	-	0.5	*0.23-0.95*
Br_6 (BDE 140)	-	-	-	-	-	-	-	-
Br_6 (BDE 138/166)	-	-	-	-	-	-	-	-
Br_7 (BDE 183)	1.3	*0.51-3.18*	0.2					
Br_7 (BDE 181)	-	-	-	-	-	-	-	-
Br_7 (BDE 190)	-	-	-	-	-	-	-	-
$\sum Br_3$-BDEs	1.7	-	0.6	*0.19-2.01*	-	-	-	-
$\sum Br_4$-BDEs	10.1	*1.81-56.04*	13.4	*3.35-53.89*	9.6	-	11.0	*4.07-29.42*
$\sum Br_5$-BDEs	5.9	*0.79-43.28*	10.6	*3.51-31.93*	7.0	*2.592-19.06*	7.3	*2.32-23.02*
$\sum Br_6$-BDEs	8.0	*0.64-99.62*	3.5	*1.20-10.06*	2.6	-	3.1	*1.38-7.10*
$\sum Br_7$-BDEs	1.3	*0.51-3.16*	0.2	-	-	-	-	-
$\sum$BDEs	18.1	*2.85-114.95*	27.0	*8.25-88.42*	10.9	*2.16-55.01*	21.5	*7.75-59.38*

Appendix 17 continued.

	FEMALE RINGED SEALS (P. hispida) (Blubber) (*n* = 7)		**MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)**		**EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6**		**WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)**	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_1 (BDE 1)	-	-	-	-	-	-	-	-
Br_1 (BDE 2)	-	-	-	-	-	-	-	-
Br_1 (BDE 3)	-	-	-	-	-	-	-	-
Br_2 (BDE 10)	-	-	-	-	-	-	-	-
Br_2 (BDE 7)	-	-	-	-	-	-	-	-
Br_2 (UI DiBDE 1)	-	-	-	-	-	-	-	-
Br_2 (BDE 8/11)	-	-	-	-	-	-	-	-
Br_2 (BDE 12)	-	-	-	-	-	-	-	-
Br_2 (BDE 13)	-	-	-	-	-	-	-	-
Br_2 (BDE 15)	0.004	-	0.008	*0.002-0.034*	0.95	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 1)	-	-	-	-	-	-	-	-
Br_3 (BDE 32)	-	-	-	-	-	-	-	-
Br_3 (BDE 17)	0.012	-	0.017	*0.007-0.041*	-	-	-	-
Br_3 (BDE 25)	-	-	-	-	-	-	-	-
Br_3 (UI TriBDE 2)	0.017	*0.004-0.065*	0.031	-	-	-	-	-
Br_3 (BDE 28/33)	0.24	*0.050-1.17*	0.28	*0.11-0.71*	0.11	*0.027-0.47*	0.084	*0.022-0.32*
Br_3 (BDE 35)	-	-	-	-	-	-	-	-
Br_3 (BDE 37)	-	-	-	-	-	-	-	-
Br_4 (BDE 75)	-	-	0.044	-	-	-	-	-

	FEMALE RINGED SEALS (P. hispida) (Blubber) (n = 7)		MALE RINGED SEALS (P. hispida) (Blubber) (n = 7)		EIDER DUCKS (S. mollissima) (Liver) (n =6		WHITE WINGED SCOTERS (M. fusca) (Liver) (n = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_4 (BDE 49)	0.15	*0.018-1.32*	0.12	*0.034-0.44*	-	-	1.19	*0.47-3.03*
Br_4 (BDE 71)	-	-	-	-	-	-	-	-
Br_4 (BDE 47)	6.43	*0.94-44.28*	9.23	*3.39-25.08*	3.66	*0.88-15.19*	14.66	*4.53-47.43*
Br_4 (BDE 66)	0.12	*0.022-0.59*	0.13	*0.038-0.42*	0.18			
Br_4 (BDE 77)	0.028	*0.002-0.33*	0.023	*0.008-0.070*	-	-	-	-
Br_5 (UI PeBDE 1)	0.029		0.038	*0.017-0.086*	-	-	-	-
Br_5 (UI PeBDE 2)	-	-	0.014	-	-	-	0.16	
Br_5 (UI PeBDE 3)	-	-	-	-	-	-	-	-
Br_5 (UI PeBDE 4)	0.19	*0.039-0.90*	0.11	*0.035-0.367*	-	-	-	-
Br_5 (UI PeBDE 5)	-	-	0.007	-	-	-	-	-
Br_5 (UI PeBDE 6)	-	-	-	-	-	-	-	-
Br_5 (BDE 100)	0.88	*0.13-6.14*	1.01	*0.35-2.91*	3.48	*0.92-13.19*	18.26	*5.63-59.28*
Br_5 (BDE 101)	0.023	*0.002-0.23*	0.020	*0.008-0.046*				
Br_5 (BDE 119)	0.042	*0.008-0.22*	0.027	*0.008-0.088*	0.21	*0.052-0.82*	0.27	*0.10-0.70*
Br_5 (UI PeBDE 7)	0.046	*0.013-0.16*	0.015		-	-	-	-
Br_5 (UI PeBDE 8)	0.020				-	-	-	-
Br_5 (BDE 99)	1.64	*0.24-11.69*	1.99	*0.75-5.30*	3.63	*0.98-13.43*	5.88	*2.01-17.15*
Br_5 (BDE 116)	-	-	-	-	-	-	-	-
Br_5 (BDE 118)	0.031	*0.002-0.46*	0.044	*0.015-0.13*	-	-	-	-
Br_5 (BDE 85)	0.025	*0.005-0.13*	0.031	*0.010-0.091*	-	-	-	-
Br_5 (BDE 126)	0.12	-	-	-	-	-	0.61	-

	FEMALE RINGED SEALS (P. hispida) (Blubber) (*n* = 7)		**MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)**		**EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6**		**WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)**	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{EQ}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
Br_5 (BDE 105)	-	-	-	-	-	-	-	-
Br_5 (BDE 155)	0.075	*0.015-0.37*	0.038	*0.008-0.186*	0.15	*0.046-0.48*	0.404	*0.088-1.85*
Br_5 (BDE 154)	0.14	*0.018-1.08*	0.12	*0.035-0.429*	3.33	*0.79-14.04*	14.02	*4.30-45.66*
Br_6 (UI HxBDE 1)	0.018	*0.005-0.065*	0.001	*0.0009-0.011*	0.27	*0.075-0.977*	0.73	*0.20-2.64*
Br_6 (UI HxBDE 2)	0.032	*0.002-0.63*	0.12	*0.003-4.065*	0.12	*0.030-0.50*	0.25	*0.098-0.62*
Br_6 (BDE 153)	0.41	*0.030-5.47*	0.32	*0.125-0.827*	3.66	*0.83-16.12*	13.02	*4.91-34.53*
Br_6 (BDE 140)	0.018	*0.004-0.079*	-	-	0.15	-	0.07	-
Br_6 (BDE 138/166)	0.015	-	0.006	*0.001-0.028*	0.17	*0.033-0.91*	0.21	*0.081-0.56*
Br_7 (BDE 183)	0.046	*0.01-0.19*	0.043	*0.018-0.102*	1.49	*0.325-6.82*	2.92	*0.76-11.19*
Br_7 (BDE 181)	0.004	-	-	-	0.23	*0.038-1.33*	0.26	*0.095-0.70*
Br_7 (BDE 190)	-	-	-	-	-	-	-	-
$\sum Br_3$-BDEs	0.26	*0.055-1.20*	0.29	*0.11-0.76*	0.11	*0.027-0.47*	0.084	*0.022-0.32*
$\sum Br_4$-BDEs	6.7	*0.98-45.94*	9.37	*3.44-25.53*	3.68	*0.88-15.38*	15.56	*5.01-48.38*
$\sum Br_5$-BDEs	2.8	*0.395-19.68*	3.18	*1.19-8.47*	7.23	*1.95-26.73*	25.32	*9.06-70.79*
$\sum Br_6$-BDEs	0.66	*0.067-6.43*	0.69	*0.20-2.36*	7.63	*1.80-32.37*	28.8	*10.11-82.29*
$\sum Br_7$-BDEs	0.047	*0.011-0.19*	0.043	*0.018-0.10*	1.61	*0.35-7.43*	3.05	*0.77-12.11*
$\sum$BDEs	10.6	*1.54-72.95*	13.6	*5.04-36.74*	19.65	*4.94-78.22*	71.29	*28.72-176.9*

Appendix 18 Regression results and FWMFs for PBDEs and PCBs in water-ventilating and air-breathing organisms and overall food web.

	WATER-VENTILATING ORGANISMS		AIR-BREATHING ORGANISMS		FOOD WEB (ALL ORGANISMS)	
	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)	$\log[C_B] = mx + b$	FWMF (95%CL)
Br_2 (BDE 15)	0.11·(TP) + -1.09, r^2 =1.000	1.29 (1.29-1.30)	-0.29·(TP) + -0.57, r^2 =0.414	0.51 (0.40-0.65)	-0.29·(TP) + -0.57, r^2 =0.414	0.51 (0.40-0.65)
Br_3 (BDE 17)	0.29·(TP) + -0.72, r^2 =0.855	1.93 (1.77-2.10)	-0.30·(TP) + -0.11, r^2 =0.852	0.50 (0.48-0.54)	-0.25·(TP) + -0.02, r^2 =0.174	0.57 (0.46-0.70)
Br_3 (BDE 28/33)	0.01·(TP) + -0.31, r^2 =-0.076	1.03 (0.76-1.40)	0.10·(TP) + -0.83, r^2 =0.063	1.25 (1.11-1.42)	0.07·(TP) + -0.66, r^2 =0.002	1.17 (1.01-1.36)
Br_4 (BDE 49)	-0.03·(TP) + 0.01, r^2 =-0.023	0.94 (0.78-1.12)	-0.17·(TP) + 0.45, r^2 =0.187	0.68 (0.58-0.81)	-0.08·(TP) + 0.16, r^2 =0.011	0.83 (0.71-0.96)
Br_4 (BDE 47)	-0.03·(TP) + 0.92, r^2 =-0.016	0.94 (0.78-1.13)	0.01·(TP) + 1.06, r^2 =-0.024	1.02 (0.93-1.12)	0.08·(TP) + 0.65, r^2 =0.020	1.21 (1.08-1.36)
Br_4 (BDE 66)	0.09·(TP) + -0.34, r^2 =0.018	1.23 (1.04-1.47)	-0.09·(TP) + -0.16, r2 =0.180	0.81 (0.75-0.87)	-0.05·(TP) + -0.13, r^2 =0.002	0.88 (0.79-0.99)
Br_4 (BDE 77)	0.24·(TP) + -0.83, r^2 =1.000	1.73 (1.72-1.73)	-0.33·(TP) + -0.27, r^2 =0.910	0.46 (0.44-0.49)	-0.23·(TP) + -0.27, r^2 =0.170	0.58 (0.46-0.75)
Br_5 (BDE 100)	-0.12·(TP) + 0.65, r^2 =0.015	0.76 (0.63-0.93)	-0.17·(TP) + 1.13, r^2 =0.210	0.67 (0.60-0.75)	-0.08·(TP) + 0.66, r^2 =0.012	0.83 (0.73-0.94)
Br_5 (BDE 99)	-0.23·(TP) + 1.41, r^2 =0.058	0.59 (0.46-0.76)	-0.35·(TP) + 1.78, r^2 =0.640	0.45 (0.41-0.49)	-0.26·(TP) + 1.46, r^2 =0.152	0.55 (0.48-0.64)
Br_5 (BDE 118)	0.04·(TP) + -0.33, r^2 =0.250	1.08 (1.02-1.15)	-0.27·(TP) + 0.01, r^2 =0.603	0.53 (0.47-0.61)	-0.33·(TP) + 0.32, r^2 =0.408	0.47 (0.37-0.59)
Br_5 (BDE 154)	-0.22·(TP) + 0.76, r^2 =0.075	0.60 (0.47-0.78)	-0.36·(TP) + 1.49, r^2 =0.442	0.44 (0.38-0.51)	-0.26·(TP) + 1.03, r^2 =0.135	0.55 (0.46-0.65)
Br_6 (UI HxBDE #2)	-0.08·(TP) + 0.19, r^2 =-0.045	0.84 (0.55-1.27)	=-0.33·(TP) + 0.32, r^2 =0.099	0.47 (0.34-0.65)	-0.33·(TP) + 0.53, r^2 =0.079	0.47 (0.34-0.64)
Br_6 (BDE 153)	-0.19·(TP) + 0.73, r^2 =0.040	0.65 (0.47-0.89)	-0.32·(TP) + 1.26, r2 =0.436	0.47 (0.42-0.54)	-0.28·(TP) + 1.08, r^2 =0.218	0.52 (0.45-0.61)
Br_7 (BDE 183)	-0.01·(TP) + -0.17, r^2 =1.000	0.97 (0.97-0.97)	-0.73·(TP) + 1.96, r^2 =0.686	0.19 (0.15-0.22)	-0.73·(TP) + 1.99, r^2 =0.645	0.19 (0.15-0.23)

Appendix 19 Calculated Biomagnification factors (BMFs), Metabolic Index (MI) and Biodilution factors (BDFs) for PBDEs in various organisms of the E Hudson Bay Marine Food web.

	Sculpin / Amphipod			Arctic Cod / Amphipod			Eider Duck / Mussels		
	BMF_{MAX} = BMF	3.7 EI	BDF	BMF_{MAX} = BMF	3.5 EI	BDF	BMF_{MAX} = BMF	106 EI	BDF
Br_2 (BDE 15)	-	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-	-
Br_3 (BDE 17)	-	-	-	-	-	-	-	-	-
Br_3 (BDE28/33)	-	-	-	-	-	-	-	-	-
Br_4 (BDE 49)	-	-	-	-	-	-	-	-	-
Br_4 (BDE 47)	-	-	-	-	-	-	1.27	1.92	83.77
Br_4 (BDE 66)	-	-	-	-	-	-	0.64	2.22	166.32
Br_4 (BDE 77)	-	-	-	-	-	-	-	-	-
Br_5 (BDE 100)	-	-	-	-	-	-	3.71	1.46	28.61
Br_5 (BDE 99)	-	-	-	-	-	-	1.38	1.89	76.98
Br_5 (BDE 118)	-	-	-	-	-	-	-	-	-
Br_5 (BDE 154)	-	-	-	-	-	-	15.34	0.84	6.91
Br_6 (BDE 153)	-	-	-	-	-	-	7.94	1.13	13.35
Br_7 (BDE 183)	-	-	-	-	-	-	-	-	-
$\sum Br_3$-BDEs	-	-	-	-	-	-	-	-	-
$\sum Br_4$-BDEs	-	-	-	-	-	-	1.19	1.95	89.32
$\sum Br_5$-BDEs	-	-	-	-	-	-	2.12	1.70	50.10
$\sum Br_6$-BDEs	-	-	-	-	-	-	16.15	0.82	6.56
$\sum Br_7$-BDEs	-	-	-	-	-	-	-	-	-
$\sum$BDEs	-	-	-	-	-	-	3.65	1.46	29.04

Appendix 19 continued.

	White winged Scoter/ Mussels			Female Beluga/ Cod			Male Beluga/ Cod		
	BMF_{MAX} =	635		BMF_{MAX} =	11.2		BMF_{MAX} =	45.7	
	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Br_2 (BDE 15)	-	-	-	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-	-	-	-
Br_3 (BDE 17)	-	-	-	-	-	-	-	-	-
Br_3 (BDE28/33)	-	-	-	0.8	1.1	12.7	1.6	1.5	28.7
Br_4 (BDE 49)	3.5	2.3	179.9	1.7	0.8	6.0	2.5	1.3	18.3
Br_4 (BDE 47)	5.1	2.1	125.4	1.2	0.9	8.2	2.9	1.2	15.9
Br_4 (BDE 66)	-	-	-	0.2	1.7	50.5	0.3	2.2	142.2
Br_4 (BDE 77)	-	-	-	-	-	-	-	-	-
Br_5 (BDE 100)	19.4	1.5	32.7	1.2	0.9	8.2	2.4	1.3	19.2
Br_5 (BDE 99)	2.2	2.5	285.2	0.7	1.1	13.9	1.1	1.6	41.0
Br_5 (BDE 118)	-	-	-	-	-	-	-	-	-
Br_5 (BDE 154)	64.6	1.0	9.8	2.5	0.6	4.0	4.1	1.0	11.1
Br_6 (BDE 153)	28.2	1.4	22.5	2.1	0.7	4.8	3.3	1.1	13.7
Br_7 (BDE 183)									
$\sum Br_3$-BDEs	-	-	-	1.0	1.0	10.3	2.1	1.3	21.8
$\sum Br_4$-BDEs	5.0	2.1	126.4	1.3	0.9	7.7	2.8	1.2	16.5
$\sum Br_5$-BDEs	7.4	1.9	85.7	1.8	0.8	5.7	4.0	1.1	11.3
$\sum Br_6$-BDEs	61.1	1.0	10.4	4.2	0.4	2.4	6.5	0.8	7.1
$\sum Br_7$-BDEs	-	-	-	-	-	-	-	-	-
$\sum$BDEs	13.2	1.7	48.0	1.6	0.8	6.2	3.5	1.1	13.1

Appendix 19 continued

	Beluga Calves/ Milk			Female Ringed Seals/ Cod			Male Ringed Seals/ Cod		
	BMF_{MAX} = 1.9			BMF_{MAX} = 8.4			BMF_{MAX} = 11.5		
	BMF	EI	BDF	BMF	EI	BDF	BMF	EI	BDF
Br_2 (BDE 15)	2.0	0.0	1.0	-	-	-	-	-	-
Br_3 (BDE 30)	2.6	-0.1	0.7	-	-	-	-	-	-
Br_3 (BDE 17)	2.2	0.0	0.9	-	-	-	-	-	-
Br_3 (BDE28/33)	2.9	-0.2	0.7	0.8	1.0	10.4	0.9	1.1	12.5
Br_4 (BDE 49)	2.2	-0.1	0.9	0.2	1.5	33.5	0.2	1.8	58.2
Br_4 (BDE 47)	3.5	-0.3	0.6	1.2	0.8	6.9	1.7	0.8	6.6
Br_4 (BDE 66)	2.4	-0.1	0.8	0.1	1.9	76.0	0.1	2.0	94.4
Br_4 (BDE 77)	2.0	0.0	1.0	-	-	-	-	-	-
Br_5 (BDE 100)	4.1	-0.3	0.5	0.7	1.1	12.2	0.8	1.2	14.7
Br_5 (BDE 99)	1.8	0.0	1.1	0.8	1.0	10.4	1.0	1.1	12.1
Br_5 (BDE 118)	2.0	0.0	1.0	-	-	-	-	-	-
Br_5 (BDE 154)	-	-	-	0.4	1.4	23.2	0.3	1.6	36.3
Br_6 (BDE 153)	-	-	-	1.7	0.7	4.9	1.4	0.9	8.5
Br_7 (BDE 183)	2.9	-0.2	0.7	-	-	-	-	-	-
$\sum Br_3$-BDEs	2.8	-0.2	0.7	0.9	1.0	9.8	1.0	1.1	11.9
$\sum Br_4$-BDEs	1.7	0.0	1.1	1.2	0.9	7.2	1.6	0.9	7.1
$\sum Br_5$-BDEs	2.0	0.0	1.0	0.8	1.0	10.4	0.9	1.1	12.5
$\sum Br_6$-BDEs	2.7	-0.1	0.7	1.4	0.8	5.9	1.5	0.9	7.6
$\sum Br_7$-BDEs	2.0	0.0	1.0	-	-	-	-	-	-
$\sum$BDEs	2.6	-0.1	0.7	1.1	0.9	7.7	1.4	0.9	8.3

Appendix 19 continued.

	Polar Bear / Ringed Seal			Inuit Women / Traditional Diet [a]		
	BMF_{MAX}	94.0		BMF_{MAX}	25.1	
	BMF	**EI**	**BDF**	**BMF**	**EI**	**BDF**
Br_2 (BDE 15)	-	-	-	-	-	-
Br_3 (BDE 30)	-	-	-	-	-	-
Br_3 (BDE 17)	-	-	-	-	-	-
Br_3 (BDE28/33)	-	-	-	-	-	-
Br_4 (BDE 49)	-	-	-	-	-	-
Br_4 (BDE 47)	1.62	1.76	57.87	1.59	1.20	15.77
Br_4 (BDE 66)	-	-	-	-	-	-
Br_4 (BDE 77)	-	-	-	-	-	-
Br_5 (BDE 100)	-	-	-	-	-	-
Br_5 (BDE 99)	-	-	-	0.77	1.51	32.51
Br_5 (BDE 118)	-	-	-	-	-	-
Br_5 (BDE 154)	-	-	-	0.07	2.58	379.05
Br_6 (BDE 153)	-	-	-	0.48	1.72	52.33
Br_7 (BDE 183)	-	-	-	1.59	1.20	15.77
$\sum Br_3$-BDEs	-	-	-	-	-	-
$\sum Br_4$-BDEs	-	-	-	-	-	-
$\sum Br_5$-BDEs	-	-	-	-	-	-
$\sum Br_6$-BDEs	-	-	-	-	-	-
$\sum Br_7$-BDEs	-	-	-	-	-	-
$\sum$BDEs	-	-	-	-	-	-

Appendix 20 Concentrations of polybrominated diphenyl ether (PBDE) and their hydroxylated (OH-) and methoxylated (MeO-) derivatives in biota (ng·g^{-1} lipid wt.) collected from E. Hudson Bay during May and September 1999-2002.

	COD (*B. saida*) **(muscle)** (*n* = 12)		**SCULPIN** (*M. scorpioides*) **(muscle)** (*n* = 12)		**SALMON** (*Salmo sp.*) **(muscle)** (*n* = 7)		**BIVALVES** (*M. edulis*) **(muscle)** (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{EQ}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
PBDEs	-	-	-	-	-	-	-	-
BDE-28/33	0.30	0.073-1.24	1.07	0.236-4.86	0.19	0.068-0.514	-	-
BDE-47	5.28	1.52-18.27	25.96	3.03-221.83	3.55	1.14-10.974	2.89	0.73-11.34
BDE-100	1.29	0.34-4.76	8.71	0.99-76.01	1.01	0.32-3.20	0.94	0.22-3.88
BDE-99	2.10	0.54-8.01	27.79	1.97-390.42	2.07	0.68-6.28	2.64	0.40-17.28
BDE-153	0.24	0.060-0.94	8.08	2.16-30.16	0.36	0.12-1.04	0.46	0.16-1.28
BDE-154	0.39	0.095-1.59	6.47	1.54-27.07	0.71	0.16-3.11	0.22	0.057-0.83
BDE-183	-	-	-	-	-	-	-	-
∑BDEs	9.78	2.67-35.79	72.82	6.44-823.42	9.28	3.03-28.39	5.38	0.86-33.46
OH-BDEs								
6'-OH-BDE17	-	-	-	-	-	-	-	-
4'-OH-BDE30	-	-	-	-	-	-	-	-
2'-OH-BDE28	-	-	-	-	-	-	-	-
3'-OH-BDE28	-	-	-	-	-	-	-	-
4'-OH-BDE17	-	-	-	-	-	-	-	-
6'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE68	-	-	-	-	-	-	-	-
2'-OH-BDE75	-	-	-	-	-	-	-	-
6-OH-BDE47	-	-	-	-	-	-	-	-

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
4'-OH-BDE69	-	-	-	-	-	-	-	-
3-OH-BDE47	-	-	-	-	-	-	-	-
2'-OH-BDE66	-	-	-	-	-	-	-	-
5-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE74	-	-	-	-	-	-	-	-
6'-OH-BDE66	-	-	-	-	-	-	-	-
4-OH-BDE42	-	-	-	-	-	-	-	-
4'-OH-BDE121	-	-	-	-	-	-	-	-
6-OH-BDE90	-	-	-	-	-	-	-	-
6-OH-BDE99	-	-	-	-	-	-	-	-
4-OH-BDE90	-	-	-	-	-	-	-	-
2-OH-BDE123	-	-	-	-	-	-	-	-
6-OH-BDE85	-	-	-	-	-	-	-	-
∑OH-BDEs								
MeO-BDEs								
6'- MeO -BDE17	-	-	-	-	-	-	-	-
4'- MeO -BDE30	-	-	-	-	-	-	-	-
2'- MeO -BDE28	-	-	-	-	0.17	*0.066-0.41*	0.17	*0.054-0.51*
4'- MeO -BDE17	0.12	-	-	-	0.08	*0.027-0.24*	1.26	*0.23-6.94*
3'- MeO -BDE28	-	-	-	-	-	-	-	-
6'- MeO -BDE49	-	-	-	-	0.50	*0.14-1.81*	0.42	*0.12-1.388*

	COD (*B. saida*) (muscle) (*n* = 12)		SCULPIN (*M. scorpioides*) (muscle) (*n* = 12)		SALMON (*Salmo sp.*) (muscle) (*n* = 7)		BIVALVES (*M. edulis*) (muscle) (*n* = 11)	
% Lipid ± *SD*	0.26 ± 0.05		0.37 ± 0.16		5.41 ± 0.27		0.6 ± 0.12	
% Lipid Equivalent (L_{Eq}) ± *SD*	1.12 ± 0.05		1.24 ± 0.16		5.41 ± 0.27		1.8 ± 0.12	
	GM	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***	**GM**	***95% CL***
2'- MeO -BDE68	2.30	*0.73-7.23*	0.6	*0.28-1.38*	6.12	*1.65-22.53*	2.84	*0.81-9.92*
2'- MeO -BDE75	-	-	-	-	0.28	-	-	-
6- MeO -BDE47	4.96	*1.51-16.24*	1.4	*0.36-5.10*	34.27	*9.78-119.99*	8.84	*2.31-33.82*
4'- MeO -BDE69	-	-	-	-	-	-	-	-
2'- MeO -BDE74	2.22	*0.76-6.42*	2.3	*0.87-5.97*	-	-	-	-
3- MeO -BDE47	-	-	-	-	-	-	-	-
2'- MeO -BDE66	0.22	*0.092-0.54*	-	-	-	-	0.27	*0.10-0.67*
5- MeO -BDE47	-	-	-	-	-	-	-	-
6'- MeO -BDE66	0.22	*0.092-0.54*	-	-	0.36	*0.091-1.450*	0.26	*0.12-0.54*
4'- MeO -BDE49	-	-	-	-	-	-	-	-
4- MeO -BDE42	-	-	-	-	-	-	-	-
4'- MeO -BDE121	-	-	-	-	-	-	-	-
6- MeO -BDE90	-	-	-	-	-	-	-	-
6- MeO -BDE99	-	-	-	-	-	-	-	-
4- MeO -BDE90	-	-	-	-	-	-	-	-
2- MeO -BDE123	-	-	-	-	-	-	-	-
6-MeO-BDE85	-	-	-	-	-	-	-	-
∑MeO-BDEs	9.98	*3.30-30.14*	2.99	*0.72-12.33*	41.59	*11.78-146.82*	13.72	*3.43-54.83*
∑OH-BDEs/∑BDEs	-	-	-	-	-	-	-	-
∑MeO-BDEs/∑BDEs	1.02	-	0.041	-	4.48	-	2.55	-

Appendix 20 continued.

	MALE BELUGA (Age 16-35) (*D. leucas*) **(Blubber)** (*n* = 21)		**FEMALE BELUGA (Age 5-35)** (*D. leucas*) **(Blubber)** (*n* = 14)		**FEMALE BELUGA** (*D. leucas*) **(Milk)** (*n* = 8)		**BELUGA CALVES** (*D. leucas*) **(Blubber)** (*n* = 9)	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
PBDEs								
BDE-28/33	0.48	0.16-1.37	0.24	0.077-0.75	0.19	0.072-0.51	0.51	0.14-1.81
BDE-47	15.16	5.53-41.51	6.59	1.68-25.83	3.90	1.27-11.91	11.31	2.42-52.79
BDE-100	3.07	1.03-9.08	1.59	0.43-5.79	0.91	0.27-2.99	2.23	0.42-11.73
BDE-99	2.34	0.79-6.90	1.54	0.37-6.36	1.03	0.29-3.64	2.02	0.42-9.59
BDE-153	0.79	0.21-2.95	0.50	0.13-1.91	0.21	0.060-0.76	0.42	0.074-2.40
BDE-154	1.59	0.47-5.27	0.98	0.24-3.86	0.42	0.11-1.49	0.76	0.12-4.74
BDE-183	-	-	-	-	-	-	-	-
∑BDEs	34.09	12.08-96.13	16.09	4.41-58.60	9.86	3.18-30.50	26.76	5.57-128.45
OH-BDEs								
6'-OH-BDE17	-	-	-	-	-	-	-	-
4'-OH-BDE30	-	-	-	-	-	-	-	-
2'-OH-BDE28	0.020	*0.004-0.095*	0.0037	-	-	-	-	-
3'-OH-BDE28	-	-	-	-	-	-	0.021	-
4'-OH-BDE17	-	-	0.012	-	-	-	-	-
6'-OH-BDE49	0.085	*0.026-0.27*	0.024	*0.009-0.062*	0.024	*0.007-0.083*	0.11	*0.022-0.50*
2'-OH-BDE68	0.045	*0.012-0.17*	0.019	*0.008-0.047*	0.020	*0.005-0.08*	0.12	*0.029-0.47*
2'-OH-BDE75	0.023	-	-	-	-	-	-	-
6-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE69	-	-	-	-	-	-	-	-

	MALE BELUGA (Age 16-35) (*D. leucas*) **(Blubber)** (*n* = 21)		**FEMALE BELUGA (Age 5-35)** (*D. leucas*) **(Blubber)** (*n* = 14)		**FEMALE BELUGA** (*D. leucas*) **(Milk)** (*n* = 8)		**BELUGA CALVES** (*D. leucas*) **(Blubber)** (*n* = 9)	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
3-OH-BDE47	0.012	*0.003-0.049*	0.017	*0.008-0.040*	-	-	0.027	-
2'-OH-BDE66	-	-	-	-	-	-	-	-
5-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE74	-	-	-	-	-	-	0.012	-
6'-OH-BDE66	-	-	0.0025	-	-	-	-	-
4-OH-BDE42	0.27	*0.091-0.797*	0.16	*0.064-0.394*	0.119	*0.042-0.339*	0.23	*0.077-0.704*
4'-OH-BDE121	-	-	-	-	-	-	-	-
6-OH-BDE90	0.023	*0.008-0.069*	0.017	*0.008-0.040*	-	-	0.027	-
6-OH-BDE99	-	-	-	-	-	-	-	-
4-OH-BDE90	-	-	-	-	-	-	-	-
2-OH-BDE123	-	-	-	-	-	-	-	-
6-OH-BDE85	-	-	-	-	-	-	-	-
∑OH-BDEs	0.43	*0.15-1.19*	0.22	*0.091-0.52*	0.15	*0.053-0.39*	0.48	*0.13-1.75*
MeO-BDEs								
6'- MeO -BDE17	0.17	*0.071-0.40*	0.12	*0.056-0.27*	0.11	*0.040-0.29*	0.27	*0.12-0.62*
4'- MeO -BDE30	0.18	*0.063-0.49*	-	-	0.16	-	-	-
2'- MeO -BDE28	0.27	*0.094-0.77*	0.28	*0.13-0.60*	0.26	*0.093-0.72*	0.32	*0.10-0.95*
4'- MeO -BDE17	-	-	0.012	*0.005-0.027*	-	-	0.008	*0.003-0.019*
3'- MeO -BDE28	-	-	0.0061	*0.002-0.020*	-	-	0.007	*0.003-0.015*
6'- MeO -BDE49	2.8	*0.85-9.80*	0.58	*0.220-1.540*	0.54	*0.17-1.67*	2.8	*0.46-16.80*

	MALE BELUGA (Age 16-35) (*D. leucas*) **(Blubber)** (*n* = 21)		**FEMALE BELUGA (Age 5-35)** (*D. leucas*) **(Blubber)** (*n* = 14)		**FEMALE BELUGA** (*D. leucas*) **(Milk)** (*n* = 8)		**BELUGA CALVES** (*D. leucas*) **(Blubber)** (*n* = 9)	
% Lipid ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
% Lipid Equivalent (L_{Eq}) ± *SD*	89.4 ± 0.53		89.7 ± 0.17		37.1 ± 8.6		90.0 ± 1.8	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
2'- MeO -BDE68	57.6	*18.00-184.47*	12.23	*4.757-31.437*	12.1	*3.78-38.77*	52.5	*9.72-283.39*
2'- MeO -BDE75	-	-	-	-	-	-	-	-
6- MeO -BDE47	237.2	*72.43-777.03*	48.2	*19.831-116.945*	51.1	*16.45-158.38*	251.4	*43.7-1,443.3*
4'- MeO -BDE69	-	-	-	-	-	-	-	-
2'- MeO -BDE74	-	-	-	-	-	-	-	-
3- MeO -BDE47	0.16	*0.070-0.395*	0.095	*0.036-0.25*	0.11	*0.040-0.28*	0.21	*0.071-0.629*
2'- MeO -BDE66	0.10	*0.034-0.317*	0.062	*0.015-0.25*	0.071	*0.020-0.25*	0.15	*0.049-0.459*
5- MeO -BDE47	-	-	-	-	0.052	-	-	-
6'- MeO -BDE66	0.17	*0.065-0.485*	0.10	*0.044-0.24*	0.13	*0.038-0.42*	0.26	*0.077-0.894*
4'- MeO -BDE49	-	-	-	-	-	-	0.17	-
4- MeO -BDE42	-	-	-	-	-	-	-	-
4'- MeO -BDE121	0.27	*0.091-0.797*	0.16	*0.064-0.39*	0.12	*0.042-0.33*	0.23	*0.077-0.70*
6- MeO -BDE90	1.62	*0.450-5.811*	0.14	*0.052-0.40*	0.14	*0.035-0.52*	0.74	*0.057-9.68*
6- MeO -BDE99	1.09	*0.258-4.617*	0.13	*0.049-0.37*	0.074	*0.023-0.24*	0.65	*0.045-9.44*
4- MeO -BDE90	-	-	-	-	-	-	0.0025	-
2- MeO -BDE123	-	-	-	-	-	-	-	-
6-MeO-BDE85	-	-	-	-	-	-	-	-
∑MeO-BDEs	299.4	*92.14-972.92*	61.8	*25.56-149.53*	64.48	*20.75-200.3*	308.8	*54.73-1,742.22*
∑OH-BDEs/∑BDEs	0.013	-	0.014	-	0.015	-	0.018	-
∑MeO-BDEs/∑BDEs	8.78	-	3.84	-	6.54	-	11.54	-

Appendix 20 continued.

	MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		**MALE BELUGA LIVER** (*D. leucas*) (*n* = 16)		**FEMALE BELUGA BLOOD** (*D. leucas*) (*n* = 7)		**FEMALE BELUGA LIVER** (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
PBDEs								
BDE-28/33	1.65	-	0.44	*0.16-1.23*	-	-	-	-
BDE-47	10.07	*1.81-56.04*	15.40	*4.99-47.47*	9.62	-	9.55	*2.99-31.14*
BDE-100	2.52	*0.41-15.42*	3.77	*1.35-10.48*	1.88	*0.77-4.55*	2.63	*0.86-8.01*
BDE-99	3.77	*0.58-24.37*	4.31	*1.36-13.61*	4.62	*1.90-11.20*	3.36	*1.10-10.18*
BDE-153	5.33	-	0.66	*0.19-2.19*	0.70	-	0.47	*0.22-0.95*
BDE-154	6.45	*0.69-59.53*	2.17	*0.81-5.76*	1.29	-	2.23	*0.97-5.09*
BDE-183	-	-	-	-	-	-	-	-
∑BDEs	18.10	*2.85-114.94*	27.01	*8.24-88.42*	10.90	*2.16-55.01*	21.46	*7.75-59.38*
OH-BDEs								
6'-OH-BDE17	-	-	-	-	-	-	-	-
4'-OH-BDE30	-	-	-	-	-	-	-	-
2'-OH-BDE28	-	-	-	-	-	-	-	-
3'-OH-BDE28	-	-	-	-	-	-	-	-
4'-OH-BDE17	-	-	-	-	-	-	-	-
6'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE68	-	-	-	-	2.23	-	-	-
2'-OH-BDE75	-	-	-	-	-	-	-	-
6-OH-BDE47	-	-	-	-	9.91	-	-	-
4'-OH-BDE69	-	-	-	-	-	-	-	-
3-OH-BDE47	-	-	-	-	-	-	-	-

	MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
2'-OH-BDE66	-	-	-	-	-	-	-	-
5-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE74	-	-	-	-	-	-	-	-
6'-OH-BDE66	-	-	-	-	-	-	-	-
4-OH-BDE42	-	-	-	-	-	-	-	-
4'-OH-BDE121	-	-	-	-	-	-	-	-
6-OH-BDE90	-	-	-	-	-	-	-	-
6-OH-BDE99	-	-	-	-	-	-	-	-
4-OH-BDE90	-	-	-	-	-	-	-	-
2-OH-BDE123	-	-	-	-	-	-	-	-
6-OH-BDE85	-	-	-	-	-	-	-	-
∑OH-BDEs	-	-	-	-	12.1	-	-	-
MeO-BDEs								
6'- MeO -BDE17	0.81	*0.21-3.02*	0.42	*0.08-2.18*	0.55	*0.26-1.12*	-	-
4'- MeO -BDE30	-	-	-	-	-	-	-	-
2'- MeO -BDE28	1.8	-	0.70	*0.14-3.42*	0.43	*0.18-0.96*	-	-
4'- MeO -BDE17	-	-	-	-	-	-	0.12	*0.500-0.027*
3'- MeO -BDE28	-	-	-	-	-	-	-	-
6'- MeO -BDE49	1.41	*0.16-11.83*	4.19	*0.78-22.37*	0.42	-	-	-
2'- MeO -BDE68	12.86	*2.11-78.06*	75.9	*13.75-419.20*	6.682	*2.52-17.68*	1.417	*0.17-11.29*
2'- MeO -BDE75	-	-	2.08	*0.542-7.99*	-	-	-	-

	MALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		MALE BELUGA LIVER (*D. leucas*) (*n* = 16)		FEMALE BELUGA BLOOD (*D. leucas*) (*n* = 7)		FEMALE BELUGA LIVER (*D. leucas*) (*n* = 3)	
% Lipid ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
% Lipid Equivalent (L_{Eq}) ± *SD*	0.59 ± 0.08		2.10 ± 0.44		0.56 ± 0.10		1.74 ± 0.55	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
6- MeO -BDE47	65.85	*10.36-418.27*	368.7	*59.81-2,270.7*	24.365	*6.79-87.43*	3.144	*0.17-57.35*
4'- MeO -BDE69	-	-	-	-	-	-	-	-
2'- MeO -BDE74	3.24	*1.37-7.64*	0.59	-	-	-	1.319	*0.21-8.156*
3- MeO -BDE47	-	-	0.43	*0.11-1.54*	-	-	-	-
2'- MeO -BDE66	-	-	0.15	-	-	-	0.179	*0.34-0.093*
5- MeO -BDE47	-	-	-	-	-	-	-	-
6'- MeO -BDE66	0.47	*0.21-1.03*	0.54	*0.16-1.82*	-	-	0.179	*0.34-0.093*
4'- MeO -BDE49	-	-	-	-	-	-	-	-
4- MeO -BDE42	-	-	-	-	-	-	-	-
4'- MeO -BDE121	-	-	-	-	-	-	-	-
6- MeO -BDE90	-	-	0.56	*0.210-1.481*	-	-	-	-
6- MeO -BDE99	-	-	0.30		-	-	-	-
4- MeO -BDE90	-	-	0.48	*0.157-1.480*	-	-	-	-
2- MeO -BDE123	-	-	-	-	-	-	-	-
6-MeO-BDE85	-	-	-	-	-	-		
∑MeO-BDEs	81.37	*12.84-515.67*	450.7	*74.7-2,719.5*	29.0	*7.4-113.61*	6.26	*0.20-193.2*
∑OH-BDEs/∑BDEs	-	-	-	-	1.11	-	-	-
∑MeO-BDEs/∑BDEs	4.50	-	16.69	-	2.66	-	0.29	-

Appendix 20 continued.

	FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		**MALE RINGED SEALS** (*P. hispida*) **(Blubber)** (*n* = 7)		**EIDER DUCKS** (*S. mollissima*) **(Liver)** (*n* =6		**WHITE WINGED SCOTERS** (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
PBDEs	-	-	-	-	-	-	-	-
BDE-28/33	0.24	0.050-1.17	0.28	0.11-0.70	0.11	0.027-0.47	0.08	0.022-0.32
BDE-47	6.43	0.93-44.28	9.23	3.39-25.08	3.66	0.88-15.19	14.66	4.53-47.43
BDE-100	0.88	0.127-6.14	1.01	0.35-2.90	3.48	0.92-13.19	18.26	5.62-59.27
BDE-99	1.68	0.24-11.69	1.99	0.75-5.30	3.63	0.98-13.43	5.88	2.01-17.15
BDE-153	0.41	0.030-5.46	0.32	0.12-0.82	3.66	0.83-16.11	13.02	4.91-34.52
BDE-154	0.14	0.018-1.08	0.12	0.035-0.42	3.33	0.78-14.03	14.02	4.30-45.66
BDE-183	-	-	-	-	-	-	-	-
∑BDEs	10.58	1.53-72.95	13.60	5.03-36.74	19.65	4.93-78.22	71.29	28.71-176.95
OH-BDEs								
6'-OH-BDE17	-	-	-	-	-	-	-	-
4'-OH-BDE30	-	-	-	-	-	-	-	-
2'-OH-BDE28	-	-	-	-	-	-	-	-
3'-OH-BDE28	-	-	-	-	-	-	-	-
4'-OH-BDE17	-	-	-	-	-	-	-	-
6'-OH-BDE49	-	-	0.004	*0.001-0.014*	-	-	-	-
2'-OH-BDE68	-	-	-	-	-	-	-	-
2'-OH-BDE75	-	-	-	-	-	-	-	-
6-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE69	-	-	-	-	-	-	-	-
3-OH-BDE47	-	-	-	-	-	-	-	-

	FEMALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		MALE RINGED SEALS (*P. hispida*) **(Blubber)** (*n* = 7)		EIDER DUCKS (*S. mollissima*) **(Liver)** (*n* =6		WHITE WINGED SCOTERS (*M. fusca*) **(Liver)** (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{Eq}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
2'-OH-BDE66	-	-	-	-	-	-	-	-
5-OH-BDE47	-	-	-	-	-	-	-	-
4'-OH-BDE49	-	-	-	-	-	-	-	-
2'-OH-BDE74	-	-	-	-	-	-	-	-
6'-OH-BDE66	-	-	-	-	-	-	-	-
4-OH-BDE42	0.023	-	0.022	*0.007-0.065*	-	-	-	-
4'-OH-BDE121	-	-	-	-	-	-	-	-
6-OH-BDE90	-	-	-	-	-	-	-	-
6-OH-BDE99	-	-	-	-	-	-	-	-
4-OH-BDE90	-	-	-	-	-	-	-	-
2-OH-BDE123	-	-	-	-	-	-	-	-
6-OH-BDE85	-	-	-	-	-	-	-	-
∑OH-BDEs	0.023	-	0.024	*0.010-0.063*	-	-	-	-
MeO-BDEs								
6'- MeO -BDE17	0.011	*0.002-0.052*	0.009	*0.004-0.022*	-	-	-	-
4'- MeO -BDE30			0.015		-	-	-	-
2'- MeO -BDE28	0.050	*0.018-0.140*	0.033	*0.006-0.186*	-	-	-	-
4'- MeO -BDE17	0.034	*0.015-0.078*	0.020	*0.006-0.062*	-	-	-	-
3'- MeO -BDE28	-	-	-	-	-	-	-	-
6'- MeO -BDE49	0.073	*0.013-0.41*	0.045	*0.010-0.20*	-	-	0.11	*0.041-0.27*
2'- MeO -BDE68	0.80	*0.173-3.70*	1.79	*0.52-6.14*	0.29	*0.053-1.55*	0.22	*0.089-0.54*
2'- MeO -BDE75	-	-	-	-	-	-	-	-

	FEMALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		MALE RINGED SEALS (*P. hispida*) (Blubber) (*n* = 7)		EIDER DUCKS (*S. mollissima*) (Liver) (*n* =6		WHITE WINGED SCOTERS (*M. fusca*) (Liver) (*n* = 5)	
% Lipid ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
% Lipid Equivalent (L_{EQ}) ± *SD*	71.2 ± 2.81		73.4 ± 4.63		3.47 ± 0.81		5.65 ± 1.25	
	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*	GM	*95% CL*
6- MeO -BDE47	3.79	*0.61-23.45*	4.54	*1.06-19.34*	0.86	*0.18-3.95*	2.04	*0.33-12.31*
4'- MeO -BDE69	-	-	0.031	-	-	-	-	-
2'- MeO -BDE74	-	-	-	-	-	-	-	-
3- MeO -BDE47	-	-	0.006	*0.002-0.022*	-	-	-	-
2'- MeO -BDE66	-	-	0.014	*0.002-0.12*	-	-	-	-
5- MeO -BDE47	-	-			-	-	-	-
6'- MeO -BDE66	-	-	0.012	*0.003-0.046*	-	-	-	-
4'- MeO -BDE49	-	-	0.001		-	-	-	-
4- MeO -BDE42	-	-			-	-	-	-
4'- MeO -BDE121	0.023	-	0.022	*0.007-0.065*	-	-	-	-
6- MeO -BDE90	0.020	*0.007-0.053*	0.013	*0.003-0.057*	0.27	*0.060-1.26*	-	-
6- MeO -BDE99	0.017	*0.006-0.044*	0.012	*0.002-0.055*	0.18	-	-	-
4- MeO -BDE90	-	-	-	-	-	-	-	-
2- MeO -BDE123	-	-	-	-	-	-	-	-
6-MeO-BDE85	-	-	-	-	-	-	-	-
∑MeO-BDEs	4.73	*0.80-27.71*	6.61	*1.70-25.68*	1.05	*0.20-5.41*	2.12	*0.34-13.24*
∑OH-BDEs/∑BDEs	0.0022	-	0.0018	-	-	-	-	-
∑MeO-BDEs/∑BDEs	0.45	-	0.4860	-	0.053	-	0.030	-