THE BIOTIC AND ABIOTIC INTERACTIONS INFLUENCING ORGANOCHLORINE CONTAMINANTS IN TEMPORAL TRENDS (1992-2003) OF THREE YUKON LAKES: FOCUS ON LAKE LABERGE

BY

MICHAEL J. RYAN

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Department of Soil Science
University of Manitoba
Winnipeg, Manitoba

ABSTRACT

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Periodic monitoring of contaminant levels in fish from the Yukon Territory indicated that organochlorine (OC) contaminants had rapidly declined since the early 1990s. This study examined OC concentrations, including chlordane (ΣCHL), ΣDDT, hexachlorocyclohexane (ΣHCH), toxaphene (ΣCHB), ΣPCB and chlorinated benzenes (ΣCBz) in sentinel fish (species of consistent annual observation and collection) from two Yukon lakes (Kusawa, Quiet), and from the aquatic food web of a focus lake (Lake Laberge) across several temporal points between 1993 and 2003. OC analysis and phytoplankton counts from dated sediment cores as well as climate data were also collected. Population, morphological (length, weight, age), biochemical (lipid content, $\delta^{13}C$, $\delta^{15}N$) and OC contaminant data for fish and invertebrates (zooplankton, snails, clams) were reviewed to elucidate the primary causes for these OC declines. Although some spatial differences in contaminant levels exist between the Yukon lakes, OC concentrations were declining for lake trout in all three lakes, with declines also noted for burbot from Lake Laberge. Several other fish species as well as zooplankton from Lake Laberge exhibited decreases in contaminant levels except northern pike, which registered consistently higher levels from 1993 to 2001. There was no evidence to support the hypotheses of changes in fish trophic levels or food sources with the exception of burbot, which marginally decreased, and northern pike, which climbed a half trophic level. Through OC flux analysis in dated sediments, the hypothesis that declines in abiotic deposition affected the contaminant levels in the food web was also negated. The closure of the Lake Laberge commercial fishery resulted in faster fish growth and larger fish populations, which are contributing to biomass dilution of OC concentrations, higher OC biomagnification factors for some species and likely changes in predator-prey interactions as resource competition increases. The large ratio of OC decreases in the lower vs. higher trophic levels of Lake Laberge have increased food web magnification factors (FWMF) for all six OC groups. It is also suspected that above-average temperatures and below-average precipitation in the lower Yukon region over the 1990s may have contributed towards an increase in lake primary production resulting in biomass dilution of contaminants in zooplankton for all three study lakes. Concurrently, shifts in the Lake Laberge zooplankton community, from climate fluctuations or increased fish predation, have gone from an abundance of *Cyclops scutifer* in 1993 to dominance by *Diaptomus pribilofensis* in 2001, although sample sites were limited. Characteristics specific to each species (e.g. body size, composition and metabolism) likely play a role in the significant OC declines measured in zooplankton. Fluctuations in population dynamics, species characteristics and OC contaminant concentrations in the Lake Laberge ecosystem may continue for

several years to come. Sentinel species such as lake trout, burbot, whitefish, cisco and plankton should continue to be monitored in all three Yukon lakes for future temporal correlations with contaminants or climate change.

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To my brother, mother and father, and to my 12-year, multi-university canine companion Chessy, I dedicate this thesis.

To my loving, patient and enduring wife Anna and our beautiful daughter Ailis, I now dedicate my unfettered time.

FOREWORD

As outlined in the Guide to Thesis Preparation for Graduate Students in the Department of Soil Science, November 1996, the manuscript style has been selected as the format for this thesis. Chapters 3 through 5 have been submitted or will be submitted for publication with the co-authors having provided some parts of research funding or sample analysis and methodology. The CPUE fish population data were kindly provided by A. Foos and S. Thompson of the Yukon Territorial Government. This data has never been formally published. The 1992-93 Lake Laberge data were provided by K. Kidd. Some of this data will be presented as part of the Lake Laberge study ecosystem study in Chapter 3 and 4; however, results and graphs from all chapters constitute my own conclusions and writing. The publications include the following:

Chapter 3. Ryan, M.J., G.A. Stern, M.V. Croft, K. Kidd, M. Diamond and P. Roach. 2005. Temporal Trends of Organochlorine Contaminants in Burbot and Lake trout from Three Selected Yukon Lakes. Sci. Tot. Environ. 351-352: 501-522.

Chapter 4. Ryan, M.J., G.A. Stern, M.V. Croft, K. Kidd, S. Gewurtz, M. Diamond, and P. Roach. 2005. Biotic Interactions in Temporal Trends (1992-2003) of Organochlorine Contaminants in Lake Laberge, Yukon Territory. Sci. Total. Environ. (in draft)

Chapter 5. Ryan, M.J., G.A. Stern, D. Armstrong, H.J. Kling, M. Diamond and P. Roach. 2005. Nutrient Levels, Historical Primary Production and Temporal Trends of Organochlorine Pesticides and PCB from Lake Laberge, Yukon Territory: Sediment Core Analysis. Arch. Environ. Toxicol. Chem. (in draft)

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1. INTRODUCTION

Organochlorine pesticides (OC) and polychlorinated biphenyls (PCB) are agricultural and industrial chemicals that were used for decades prior to the discovery of their detrimental environmental impacts (Lockhart et al. 1992). Most countries have since banned these chemicals from general use; however, several industrialized and developing countries still use some of these materials (Lockhart et al. 1992; UNEP Chemicals 2003) because cheaper and more effective pesticide alternatives do not yet exist. These chemicals are known to be resistant to environmental degradation and can be transported from areas of use to cooler climates through long-range atmospheric transport and deposition (Bidleman 1988; Bidleman 1999), a phenomena known as 'global distillation' (Goldberg 1975) or the 'grasshopper effect' (Wania and Mackay 1996). For this reason, OC pesticides and PCB continue to be present in biotic and abiotic samples in remote northern regions (CACAR 2003c).

Detailed and continuous temporal data for surveys in Arctic and sub-Arctic regions have been limited and sporadic over the past decades with few whole food web studies on OC pesticides and PCB (Muir et al. 1992; CACAR 2003a). Temporal studies are necessary to understand long-term trends in levels of these pollutants and such research is required to predict future trends of these contaminants as they are often found in human foods (Kinloch et al. 1992). However, observations that include only a few variables (e.g. one contaminant, one organism, one medium such as air or water) do not provide an adequate assessment of whole ecosystems, and many assumptions must be made with respect to the state of contaminants in the area or in organisms measured. Temporal surveys must include a broader analysis of the factors that affect these contaminants in the environment such that more detailed review of the ecosystem is required.

A research project in the early 1990s provided the basis for a temporal trend ecosystem study in lakes in the southern Yukon Territory (Kidd et al. 1993; Kidd et al. 1995b; Kidd 1996; Kidd et al. 1998). The research focused on five OC pesticide groups (chlordane, hexachlorocyclohexane, DDT, toxaphene, chlorinated benzenes) and polychlorinated biphenyl (PCB), in the aquatic biota of three Yukon lakes. The study analysed samples from both the biotic and abiotic compartments to elucidate the influences of biotic physiological variables on contaminants within the lake food webs and to investigate abnormally high OC levels in fish from Lake Laberge. The main objective of this study was to research the use of stable isotopes as predictors of trophic levels and contaminant burdens in aquatic biota across the study lakes (Kidd et al. 1995a). The research was not a temporal study. Since the 1992-1993 study concluded, periodic OC analyses (1996-1999) revealed a large decline in these contaminants in fish, but no detailed data were available to explain the occurrence.

Several hypotheses were reviewed to explain how biotic interactions of fish populations, morphological or biochemical measurements, or abiotic contaminant estimates (lake sediment) can support explanations for temporal changes in contaminants. The objective of my research was to elucidate the most prominent factors affecting contaminants in the Lake Laberge food web (abiotic or biotic) and to relate those factors to trends in sentinel species from other regional lakes through several means:

1. Measure current OC contaminant levels in sentinel fish species in three Yukon lakes and examine temporal trends using data from previous studies.
2. Review morphological changes between species across regional lakes to consider ecosystem-specific factors that have affected OC levels in the Yukon biota.
3. Collect food web samples from Lake Laberge, including fish and invertebrate species, and conduct temporal trend analyses of OC contaminants for Lake Laberge. The analysis incorporated correlations of population and biochemical characteristics of each species to determine the underlying factors affecting temporal trends in this food web.
4. Examine the food web structure of Lake Laberge using stable nitrogen and carbon isotopes to determine whether trophic level or food source changes have affected OC levels in the biota of Lake Laberge.
5. Analyze regional climate data with phytoplankton enumeration from sediment cores to investigate the hypothesis that general climate influences may affect primary production with subsequent impacts on contaminants in the food web of Lake Laberge.
6. Examine temporal nutrient concentrations in Lake Laberge and compare

to zooplankton community composition and phytoplankton enumeration from sediment cores to investigate the hypothesis that changes in nutrient levels may affect primary and secondary production in Lake Laberge.

7. Determine recent and historical OC fluxes to Lake Laberge through radioisotope dating and OC analyses of sediment core slices to investigate the hypothesis that abiotic deposition (runoff or atmospheric outfall) of contaminants to Lake Laberge has affected OC levels in biota.

2. LITERATURE REVIEW

2.1 Persistent Organic Pollutants: Organochlorines

The use of pesticides has been documented since the time of Homer in 1200 BC when sulfur was used for fumigation purposes (Ware 2000). Since that time, the evolution of chemistry brought about pesticide materials that were increasingly complex and hazardous to the environment. Through the 20th century, many organochlorine (OC) products were synthesized in large quantities (Ware 2000) for use as pesticides in crop protection and disease prevention, or as industrial lubricants and transformer oils, as in the case of polychlorinated biphenyls (PCB). Most of these OC chemicals have since been banned by industrialized nations (UNEP Chemicals 2003). Many of these materials represent health hazards to either wildlife or humans (Faroon and Olson 2000; Kajiwara et al. 2002; CACAR 2003b; CACAR 2003c), and the parent chemicals, as well as toxic degradation products, remain intact over long periods in ecosystems due to their slow environmental degradation.

The chemical and physical properties of the compounds within each of the six major organochlorine groups (Table 2.1) studied herein generally dictate how far the compounds can be transported by the atmosphere or in water (Van Pul et al. 1999; Bidleman 1999). These properties also define their fate and half-life in the environment as well as the propensity of the chemicals to accumulate in living tissues. The affinity for these chemicals to partition into lipids is best defined by their K_{ow} values (octanol-water partition coefficient). A higher K_{ow}, the greater the affinity the chemical has for lipids rather than water when at equilibrium between the two solutions. The phenyl or benzene ring structure of most organochlorines makes these compounds stable in the environment and resistant to degradation. Further information on physical-chemical parameters of organochlorine contaminants is extensively reviewed in CACAR (2003c).

Monsanto Corporation introduced PCB in 1929 for use as industrial lubricants and heat exchange materials in electric transformers. They had enormous international usage over six decades until the last known production of a PCB compound (Socol) was in Russia in 1993 (Barrie et al. 1992; Stern et al. 2005). PCB are chlorinated, dual-phenyl ring structures, usually in a mixture of up to 209 different congeners, that are thermally stable and highly resistant to degradation by biological decomposition with long environmental half-lives (Barrie et al. 1992; Diamond et al. 2005). PCB were banned in the U.S. in 1977 after approximately 600,000 tons were produced, 31% of which is estimated to have entered the global environment (Barrie et al. 1992). PCB have known effects on human (Faroon and Olson 2000) and aquatic animal health (Aulerich et al. 1973; Black et al. 1998).

DDT is one of the most infamous insecticides of the 20th century. Originally discovered in 1873 and forgotten, its rediscovery in 1939 by Dr. Paul Muller

Table 2.1 Chemical and physical characteristics of major organochlorine contaminants. Ranges are provided as values vary with degree of chlorination, temperature and methods. Summated from Mackay et al. (1992) and Mackay et al. (1997).

Compound	**Henry's Law Constant ($Pa/m^3/mol$)**	**Log K_{ow}**	**Water Solubility (g/m^3)**
CBz	263-451	2.13-3.0	0.5×10^{-2} - 7.8
HCH			
a-HCH	0.215-2.16	3.81-4.44	0.02-21.6
b-HCH	0.05-0.12	3.78-4.15	0.13-69.5
g-HCH	0.01-0.08	4.1-4.14	8.0-109.0
Chlordane	0.248-112	2.78-6.21	0.2×10^{-2} - 1.85
DDT	0.16-7.29	3.98-7.35	0.2×10^{-3} - 0.467
Toxaphene	0.06-6382	3.23-5.5	0.3-3.0
PCB			
Mono-	42.5-79.3	4.3-4.73	0.06-9.5
Di-	17-153.6	4.9-5.3	0.06-2.26
Tri-	77-102	5.5-5.9	0.015-1.09
Tetra-	1.72-94	5.6-6.35	0.8×10^{-3} - 0.38
Penta-	12.2-151.4	6.4-6.85	0.4×10^{-2} - 0.11
Hexa-	6.7-86	6.7-6.8	0.4×10^{-3} - 0.021
Hepta-	5.4-100 (est.)	6.7-7.1	0.45×10^{-3} - 1.8×10^{-2}
Octa-	38-100 (est.)	7.1-8.5	0.2×10^{-3} – 0.02
Nona-	100 (est.)	7.2-9.14	1.8×10^{-5} - 0.2×10^{-2}
Deca-	n/a	n/a	n/a

resulted in his receiving a Nobel Prize in medicine for the invention of a highly effective, low cost insecticide to combat diseases (e.g. malaria) carried by insects such as mosquitoes (Ware 2000). Similar to PCB, DDT is also a chlorinated biphenyl structure. DDT was introduced in North America in 1940s and since that point, an estimated 18.1 millon tonnes have been used worldwide (Ware 2000). Although DDT was banned in the U.S. in 1972-1973 and in Canada in the early 1980s, it is still used by many counties in Asia, South America and Africa. India is currently (as of 2000) the largest consumer using approximately 7 tonnes per year (Barrie et al. 1992; CACAR 2003c). The total global usage of DDT between 1950-1993 was estimated at 3,590,000 tonnes (Voldner and Li 1995). DDT is highly water insoluble, is extremely slow to degrade and has a long and well-publicized history of detrimental effects on organisms, most notably on birds (Willson 1978; Newton 1988; Thomas et al. 1992; Carson 1962).

Hexachlorocyclohexane (HCH) originated in 1825, but much like DDT, its insecticidal properties were not discovered until 1940 by French and British entomologists (Ware 2000). It is also known as benzenehexachloride (BHC) as its structure consists of a chlorinated benzene ring without the double bonds (hence the nomenclature of cyclohexane in HCH). Two forms of HCH pesticide exist. Technical HCH is a combination of the α– (55-70%), β– (5-14%), γ– (10-18%), δ– (6-10%) isomers, while the pure form of HCH pesticide, also known as lindane, is almost entirely the γ– isomer (Barrie et al. 1992; Ware 2000). Only lindane has insecticidal properties and is one of the few chemicals in this study that is still in use and production in industrialized countries (CACAR 2003c).

Technical HCH was banned in Canada in 1971 and in the U.S. in 1978 (Barrie et al. 1992). The total cumulative global usage of technical HCH and lindane that is accounted for was estimated at 550,000 tonnes and 720,000 tonnes, respectively (Voldner and Li 1995). However, a more recent quote of the total production of HCH was estimated at approximately 4.46 million tonnes from 1952 until its ban in 1983 with China noted as the biggest producer and consumer of this pesticide (Li et al. 1998). HCH acts as a neurotoxin producing similar symptoms to those caused by DDT (Ware 2000). Combined with its ability to bioaccumulate, and the environmentally resilient isomers from the technical mixture, HCH remains an environmental concern.

Chlorinated benzenes (CBz) were manufactured for use as industrial solvents and they were also created as by-products during the synthesis of organochlorine pesticides (US-EPA 1980). Chlorinated benzenes are structurally similar to HCH with variations in the numbers of chlorine substitutions. Water solubility of CBz compounds decreases while vapour pressure increases with increasing chlorine substitution, giving it a wide range of physical properties (Mackay et al. 1992). Hexachlorobenzene (HCB) was used as a seed dressing for the prevention of fungus growth between 1948 and 1972 and 318 tonnes were produced in the US in 1973 alone (Environment Canada 1993). Total production of CBz from 1973 to 1975 in the US was estimated at 160,000 tonnes (US-EPA 1980). There are known toxic effects of CBz on mammals and humans (Environment Canada 1993).

Chlordane is a mixture of compounds belonging to a group known as cyclodienes, which includes the common pesticides aldrin, dieldrin, heptachlor, mirex and endosulfan (Ware 2000). Chlordane was patented for use in 1945 and because of its three dimensional structure, it has many stereoisomers making it a mixture of over 120 compounds. This complex structure is also what makes chlordane stable in soils and resistant to UV light degradation (Ware 2000). The US EPA phased out agricultural uses of chlordane between 1975 and 1980 with an approximated 70,000 tons produced historically (Barrie et al. 1992). However, restricted use as a termiticide was allowed in both Canada and the US until about 1988 (Dearth and Hites 1991; Ware 2000) but this was followed by a worldwide withdrawal of the chemical in 1997 (Bidleman et al. 2004). The potential for biological effects of chlordane remains significant to mammals and it is also a frequent cause for EPA fish advisories in the US (TMDL Team 2001; CACAR 2003b).

Toxaphene, or chlorinated bornanes (CHB), was possibly the most highly used insecticide in North America since its development in 1946 (Barrie et al. 1992). It is created by chlorinating camphene (a derivative of pine tar) which results in a complex structure that has a theoretical number of congeners (mostly isomers) ranging in the tens of thousands (Vetter 1993). However, only a couple of hundred congeners exist in the technical mixture and even fewer in environmental samples. It is best separated by high resolution gas chromatography making it difficult to analyze (Barrie et al. 1992; CACAR 2003b). There was estimated world usage of 1,330,000 tonnes of toxaphene between

1950 and 1993 (Voldner and Li 1995). Although it was banned in the US in 1983, it is still listed as severely restricted or in-use by 41 other countries (CACAR 2003c). Toxaphene is highly toxic to fish (Barrie et al. 1992; Fahraeus-Van Ree and Payne 1997; Ware 2000; Fahraeus-Van Ree and Spurrell 2000; Scott et al. 2002) and is known to bioaccumulate in organisms (Kidd et al. 1995b).

2.2 Organochlorine Chemical Use in the Yukon

In 1993, an extensive survey on the historical use of contaminants in the Yukon was compiled (Nordin et al. 1993). The compounds that had some use in the region, either industrially, experimentally or through spraying programs, included DDT, lindane, aldrin, dieldrin, CBz, toxaphene and PCB. The following is a description of their use in the Yukon Territory.

Several experimental tests were done to determine the most effective insecticide for the Yukon area. These experiments involved the application of DDT, parathion, HCB, aldrin, dieldrin and CBz into the Watson River near Carcross. The most prominent chemical used for the control of mosquitoes and blackflies was DDT, although some forms (Tossit capsules) also included lindane in the mixture. From 1949 to 1969 an estimated 15 817 kg powdered and 20 511 litres of liquid DDT were used for aerial spraying and these values do not include amounts of DDT used for ground fogging practices. The insect control occurred primarily around the City of Whitehorse although several other communities were noted as being part of the DDT-insect control program including Keno City and Elsa, both several hundred kilometers north of Whitehorse. Although the historical report indicates that aerial spraying started around 1949 (Nordin et al. 1993), DDT in local lake sediments showed that it was used several years earlier, coinciding with the construction of the Alaska Highway during World War II (Rawn et al. 2001). Since the ban on DDT, municipal disposal of unused drums of DDT into local dumpsites may have occurred. The practices of the Royal Canadian Air Force (RCAF) and their use and disposal of unused drums of pesticides are unknown and could have contributed to local sources of the material over the past decades (Nordin et al. 1993).

The only recorded use of toxaphene in the Yukon occurred around 1963 on Hanson Lake. Toxaphene was used by the RCAF to eradicate unwanted fish species from the lake (Nordin et al. 1993; Vetter et al. 1999). Hanson Lake lies north of Keno City.

PCB were likely used in the Yukon region in electrical transformers. The transport of PCB through the Yukon Territory has also been reported (Nordin et al. 1993) and it has been suggested that PCB may have been distributed in the Yukon in contaminated petroleum products. Several oil compounds were used in large quantities not only for the dissolution of DDT for aerial spraying, but also on the ice surfaces of lakes. An example of this occurred on Lake Laberge where ‘lamp black’ was commonly poured on the ice surface to accelerate spring melting to open a channel for shipping (Seigel and McEwen 1984) along the Yukon River, north to Dawson City. Empty and unlabeled barrels are often found washed ashore in Lake Laberge to this day (P. Roach, pers. comm.). Diamond et al. (2005) has provided evidence of point source pollution of Lake Laberge by both PCB and DDT and it is suspected that the sources are from materials buried in a dumpsite near the edge of the Yukon River, upstream from the lake.

2.3 Organochlorine Atmospheric Transport and Historical Sediment Monitoring

Even though decades have passed since the ban of most organochlorine chemicals in Canada and the U.S., other developing countries continue to use them (UNEP Chemicals 2003). Some of these contaminants have never been used in sub-Arctic or Arctic regions and yet the chemicals have been measured at significant concentrations in aquatic biota (Bidleman et al. 1989; de March et al. 1998; CACAR 2003a). These contaminants migrate into remote areas of the world through a process known as global distillation (Goldberg 1975) or the grasshopper effect (Wania and Mackay 1996). Global distillation is the movement of chemicals from lower to higher altitudes and latitudes through the evaporation from warmer climates and deposition in colder regions. The process can undergo seasonal cycles, and a single contaminant may volatilize and condense several times before it reaches a climate cold enough to prevent re-volatilization (hence the term ‘grasshopper effect’). Extensive reviews on the phenomenon exist in other publications (Datta et al. 1998; Bard 1999; Van Pul et al. 1999; Van Dijk and Guicherit 1999; Bidleman 1999).

The source of contaminants in Arctic areas is believed to be from long range transport and deposition (Kidd et al. 1998; CACAR 2003c). Because the chemicals volatilize from warmer areas to colder regions, a latitudinal gradient in contaminant concentrations has been observed from lower to higher latitudes. As one example, HCH was measured in tree bark from 90 sites worldwide and concentrations were higher in Arctic regions (where HCH has never been used) compared to equatorial areas (Simonich and Hites 1995). A similar situation exists for locations of high altitude. These areas are more likely to have deposition of contaminated particulates from the atmosphere because of the colder climates at higher altitudes (Blais et al. 1998; Van Dijk and Guicherit 1999; Bidleman 1999; Carrera et al. 2002). This leads to pollution of mountain glaciers creating a potentially long-term source of contamination for receiving rivers and streams (Donald et al. 1999).

To determine the importance of atmospheric deposition to local burdens, studies have traced the sources of pollution through techniques such as back trajectories of atmospheric currents and comparisons of contaminant variations (e.g. isomers, enantiomers) to known composition values specific to a contaminant or a region of use (Bard 1999; Bidleman 1999; Bidleman et al. 2002; Bidleman et al. 2003). An unusual precipitation event was observed in 1988 where brown snow accumulated in a Canadian Arctic area (Welch et al. 1991). PCB, HCH, DDT and other contaminants were found in the snow and using atmospheric trajectories, it was suggested that the source of the material came from China. Another study observed seasonal differences in HCH concentrations in air samples over a 5-year period that coincided with atmospheric currents from areas in Western or Central Europe of known HCH use resulting in the conclusion that HCH congener profiles were significantly correlated with the origin of the air mass (Haugen et al. 1998).

Recent air monitoring in the Arctic has shown patterns of OC levels varying by season but coinciding with decreasing temporal trends in HCH, chlordanes and DDT concentrations over long term temporal trends (Bidleman 1999; Bidleman et al. 2002; Hung et al. 2002; Bidleman et al. 2004). However, concentrations of some metabolites of the parent OC compounds (notably DDE) were observed to increase or remained constant in air samples over the 5-year survey (Hung et al. 2002). Some OC pesticides, such as chlordane, are believed to be recycled to the atmosphere and eventually depositied to the northern regions from treated soils in southern regions (Bidleman et al. 2004). The effects of these trends on contaminant levels in other abiotic and biotic compartments are still being reviewed.

Surveys of historical OC fluxes in Arctic, sub-Arctic and the Great Lakes regions have been examined using lake sediments (Oliver et al. 1989; Vetter et al. 1999; Rawn et al. 2001; Stern et al. 2005). Sediment cores collected from the Arctic and the Yukon have shown decreasing levels of legacy pesticides and PCB since their height of production (Rawn et al. 2001; Stern et al. 2005). Results of a sediment survey from the Great Lakes also observed that a decline in atmospheric OC concentrations were reflected in the OC levels in lake sediment from that region (Oliver et al. 1989). Although declines in legacy OC have been observed in air and sediments, a few increases in OC occurred in some aquatic systems in the late 1980s (Rawn et al. 2001; Stern et al. 2005).

One study attributed the trend to biological events (algal production) related to climate changes (Stern et al. 2005).

Declines in OC levels in biota have occurred since the ban of legacy OC pesticides and PCB (Muir et al. 1992), but air and sediment measurements may not provide a clear assessment of these contaminant trends in ecosystems. Biota in aquatic ecosystems need to be monitored directly because local biological and abiotic conditions (e.g. regional climate fluctuations, biota population changes) can have dramatic effects on contaminant concentrations. OC concentrations in sediments and biota from two high Arctic lakes on Bear Island (Norway), had distinctly different levels of OC because of both higher inputs of OC from the guano of seabirds and the greater input of atmospheric pollutants to the higher altitude lake (Evenset et al. 2004). Macdonald et al. (1993) concluded that lake maximum depth is an important predictor of the bioaccumulation of PCB from sediment to biota. Biota in shallower lakes also had higher proportions of higher (over lesser) chlorinated PCB congeners suggesting one cannot predict PCB in biota based on sediment concentrations alone. Larsson et al. (1998) observed concentrations of PCB in plankton that were actually lower than the concentrations in sedimenting particulates collected from oligotrophic and eutrophic lakes. Conversely, PCB in zooplankton samples have also been measured with considerably higher contaminant levels than collected sediments on a dry weight basis (Macdonald and Metcalfe 1989). Ultimately, areas with elevated atmospheric deposition of OC pollutants will still be reflected in the contaminant loads in biota. This was evident from samples of mammalian herbivores and predators as well as insects from two study regions in Scandinavia receiving different atmospheric loads of OC contaminants (Larsson et al. 1990). Atmospheric sources, rather than sediment, were determined to be the most likely source of PCB to the Lake Michigan food web (Stapleton et al. 2001). In addition, it is estimated that regional fluctuations in weather patterns or increased global temperatures due to climate change will impact northern ecosystems and contaminant movements, and affect the transport of OC from abiotic to biotic compartments (Macdonald et al. 2003; CACAR 2003c).

2.4 Fishery History of Lake Laberge

Monitoring studies have examined various abiotic and biotic samples from Lake Laberge over the past 25 years and beginning in the 1990s, the Yukon Territorial Government started conducting fish population surveys to assess the health of the ecosystem following the closure of a commercial fishery (Brown et al. 1976; Lindsey et al. 1981; Ball 1983; Kirkland and Gray 1986; Connor and Sparling 1996; Foos 2001). The first recorded date for fishing on Lake Laberge was 1898 when a commercial fishing operation used approximately 3519 yards (3.2km) of nets for harvesting (Seigel and McEwen 1984). The first officially recorded statistic for Lake Laberge was logged in 1902 with harvest estimates as high as 20.4 tonnes that year (Seigel and McEwen 1984). During the period of 1908-1917, an estimated 64.1 tonnes of fish were taken from the lake, which includes only the known or reported amounts during a period when catch reporting regulations were unenforced (Seigel and McEwen 1984). Over-fishing was already becoming a concern for Lake Laberge in 1902 because catches of smaller-than-average whitefish were observed by the local fishery officer (Seigel and McEwen 1984). Until the early 1990s, Lake Laberge also sustained both sport and subsistence fisheries for tourists, local inhabitants and a band of First Nations people, the Ta'an Kwach'an (Seigel and McEwen 1984; Thompson 1996). Commercial fisheries also harvested fish on Kusawa and Quiet Lakes with closures in 1968 and 1989 respectively (Seigel and McEwen 1984; Foos 1998). However, the numbers of fish collected from the Laberge fishery far exceeds the amount of fish taken from Kusawa and Quiet Lakes combined. Fish harvests between 1972 and 1983 for Quiet Lake and Lake Laberge totalled 5964 and 18.7 tonnes, respectively. Of that catch, 53% (Quiet Lake) and 71% (Laberge) were lake trout (Seigel and McEwen 1984). A 1991 federal government human health advisory led to the closure of the Laberge commercial fisheries and all commercial licenses expired by 1993 (Connor and Sparling 1996; Thompson 1996). Records showed that the domestic and First Nations harvests were also greatly reduced in the early 1990s with only 89 kg (196 pounds) of lake trout reported harvested between 1990 and 1994 (Thompson 1996).

Although fish catch statistics have been monitored on Yukon lakes for over a century (Seigel and McEwen 1984; Cox 1999), OC contaminant surveys in Yukon lakes only began in the 1990s (e.g. Connor and Sparling 1996). Lake Laberge in particular became a focus of contaminant research in 1992 because of abnormally high levels of OC concentrations found in these fish compared to the same species from other regional lakes (Kidd et al. 1993; Kidd et al. 1998).

Contaminants are known to cause declines in the populations of various fish species (Newton 1988; Forbes and Calow 1999). In this case, it was suggested that the high concentrations in Lake Laberge fish may have a link to the exploitation of the population by the long standing commercial fishery (de Graff and Mychasiw 1994; Kidd 1996) by selectively reducing the number of top predators and allowing for a greater food base for the remaining individuals thus creating a 'longer' food chain.

2.5 Historical Contaminant Monitoring in Lake Laberge and Other Regional Lakes

Since the early 1990s, sentinel fish species (herein defined as species of consistent annual observation and collection) from Quiet, Kusawa and Laberge lakes have been periodically monitored for OC contaminants by the Department of Fisheries and Oceans (DFO) and the Department of Indian Affairs and Northern Development (DIAND). Concerns about the human health effects from OC contaminants in northern traditional foods (Kinloch et al. 1992; CACAR 2003b) arose after high concentrations of OC were measured in Lake Laberge fish. Burbot livers are a common delicacy for First Nations people. The high levels of contaminants detected in Lake Laberge samples resulted in a consumption advisory for burbot livers and lake trout flesh (Figure 2.1). Mean concentrations of PCB, DDT and CHB in Lake Laberge burbot were higher than samples from Fox Lake by factors of 26, 37 and 32, respectively (Kidd et al. 1998). Mean concentrations of PCB, DDT and CHB in Lake Laberge lake trout exceeded the concentrations in samples from Kusawa Lake by factors of 5, 12 and 3, respectively (Kidd et al. 1998). Since the closure of the Laberge commercial fisheries and the reduction of subsistence fishing, fish populations in Lake Laberge appear to have increased (Foos 2001) and coincidental declines in the high OC contaminant concentrations were observed (G. Stern, pers.comm.).

The assessment of Lake Laberge in 1992-1994 (Kidd et al. 1995a; Kidd et al. 1995b; Kidd 1996; Kidd et al. 1998) was done to determine the underlying cause of the high levels of contaminants in Lake Laberge relative to other regional lakes. It examined the food web structure of Lake Laberge, along with other regional reference systems, and contrasted biomagnification (generally defined as the increase in pollutant concentrations in animal tissues in successive members of a food chain (Moriarty 1988)) of OC among food webs. Another objective was to determine the morphological and biochemical factors that affected OC, which could be used as predictors of contaminant levels in lacustrine biota. The results of that study concluded that the longer than normal food chain for Lake Laberge was the primary contributor to the higher levels of contaminants in fish due to the effects of biomagnification (Kidd et al. 1995b; Kidd 1996; Kidd et al. 1998). However any effects the fishery had on contaminant loads was unknown. The long food chain may be more indicative of the size or diversity of the Lake Laberge ecosystem (Post et al. 2000) rather than the effects of the fishery.

Figure 2.1 Health advisory sign at Lake Laberge, YT. (Photo by M. Ryan)

2.6 Stable Isotopes and Contaminant Research

Perhaps the most important conclusion from the previously mentioned research was the correlation between contaminant concentrations and trophic levels as measured by the ratio of the stable nitrogen isotopes ^{14}N and ^{15}N. The ^{15}N isotope is preferentially retained over ^{14}N such that organisms higher in the food chain have a greater ratio of ^{15}N to ^{14}N, expressed as $\delta^{15}N$ (DeNiro and Epstein 1981; Peterson and Fry 1987) measured in parts per mil (‰). Predators are enriched in ^{15}N by approximately 3-5‰ per trophic level when compared to their prey (DeNiro and Epstein 1981; Minagawa and Wada 1984). Several researchers (Rolff et al. 1993; Cabana and Rasmussen 1994; Vander Zanden and Rasmussen 1996) have also presented similar methods for quantifying the biomagnification of persistent pollutants and have concluded that organisms higher in the food chain (as measured by $\delta^{15}N$ in fish tissues) have higher levels of contaminants. For example, each trophic level contributed an estimated 3.5 fold biomagnification factor for PCB concentrations in lake trout sampled across 83 Ontario lakes (Rasmussen et al. 1990). About 85% of the between-lake variability in PCB concentrations in lake trout was explained by their trophic positioning (Vander Zanden and Rasmussen 1996). Another study estimated that 99% of PCB levels in Lake Erie lake trout were derived from the food chain (Epplett et al. 2000). Along with fish lipid content, the trophic level of individual fish as well as the trophic structure of the food chain accounted for 59% and 72% of the concentrations of DDT and PCB in 25 species of fish from the Great Lakes, respectively (Rowan and Rasmussen 1992). If a species shifts its trophic position due to a change in diet (Vander Zanden et al. 1997; McCutchan et al. 2003), population interactions or migratory patterns (Jardine et al. 2003), the alteration could change the contaminant biomagnification potential for that species. Correlations between food chain length and biomagnification of OC contaminants have been shown in many other studies (Rolff et al. 1993; Cabana and Rasmussen 1994; Kiruluk et al. 1995; Kidd et al. 1995b; Zaranko et al. 1997; Camusso et al. 1998; Kidd et al. 1998; Stapleton et al. 2001; Evenset et al. 2004). . It should be noted that $\delta^{15}N$ only represents an average trophic position in any individual, which can be attained through the consumpation of many combinations of food items. Another method of determining the more precise food types assimilated must be used to assess this condition.

Trophic levels of biota can be measured by $\delta^{15}N$, but food sources are assessed through ^{12}C and ^{13}C stable isotope measurements (Peterson and Fry 1987; Hesslein et al. 1993). The ^{13}C to ^{12}C ratio (expressed as $\delta^{13}C$) of a predator will be closely related to the $\delta^{13}C$ of its prey (DeNiro and Epstein 1978; Peterson et al. 1985). With muscle-stable isotope turnover times estimated at more than one year for slower growing fish (Hesslein et al. 1993), and other stable isotope elements (e.g. sulphur) having functional relationships relating to different aquatic habitats (France and Peters 1997; Vander Zanden and Rasmussen 1999), the $\delta^{13}C$ is a practical parameter for monitoring predator-prey relationships (food sources) over time. For example, significant variations in $\delta^{13}C$ and metal contaminants between two albacore tuna populations collected from the same bay in the North Atlantic showed they actually inhabit distinctly different ecological niches (Das et al. 2000). These methods were also used to explain the high levels of contaminants in pelagic-feeding lake trout compared to the littoral-feeding mountain whitefish and lake trout, measured at a similar trophic level, in a sub-alpine lake (Campbell et al. 2000). Using $\delta^{13}C$ and stomach content analyses, it was determined that the lake trout with higher OC levels were feeding on pelagic copepods, which had significantly higher organochlorine concentrations compared to littoral prey. All of these research examples demonstrate that $\delta^{15}N$ and $\delta^{13}C$ are important tools for predicting contaminant concentrations in food webs.

2.7 Fish Population Characteristics, Contaminants and Ecosystem Dynamics

Many abiotic factors including climate, lake morphology and hydrology can affect the biotic components and contaminants in a lake ecosystem (Barrie et al. 1992; Edmundson et al. 2003). These factors will ultimately influence biotic growth, species diversity, densities and even annual survival as well as contaminant concentrations in each compartment of an ecosystem model (Figure 2.2). A hypothetical example of fish growth and contaminant transfers, as affected by a change in one abiotic factor, can be illustrated using a simple linear ecosystem model. Organic contaminants are deposited into a remote lake through atmospheric processes but the concentrations in the water are delineated by many factors such as presence of organics, volatilization, inflows and deposition rates (Barrie et al. 1992; Van Pul et al. 1999). Particles containing nutrients and contamaints may be washed out to other water bodies and equally, particles may be washed in from water bodies upstream changing some of the abiotic parameters across time. During a year of elevated sun exposure in the area, there is a subsequent stimulation of the growth of phytoplankton through the addition of light and heat; assuming available nutrients are not a limiting factor (Wetzel 2001). Contaminants are taken up from the water during normal phytoplankton growth but are now being assimilated at a different rate compared to previous years because of the elevated growth rate (Swackhamer and Skoglund 1993). As phytoplankton biomass increases, a larger food base for zooplankton is formed elevating zooplantion growth. Larger zooplankton form a bigger food base for planktivorous fish, and then piscivours fish, and so forth up the food chain. Contaminants are transferred from trophic level to trophic level in a ratio varied by growth amongst other factors. As these organisms die off, their remains fall to the sediments where benthic organisms now gain in growth and contaminants from the inputs from above (Figure 2.2). Since the ecosystem is a cyclical system, a combination of measures from both abiotic and biotic areas needs to be studied to derive conclusions about the state of biota and contaminants in a lake ecosystem.

In order to evaluate contaminant trends in a post- exploited lake, an assessment of the overall trophic status, fish populations and general ecosystem health is required. Fish population characteristics (e.g. age, fecundity, condition factors, populations, growth rates) have been put forth as an approach to assessing the overall health of an ecosystem and its inhabitants. For example,

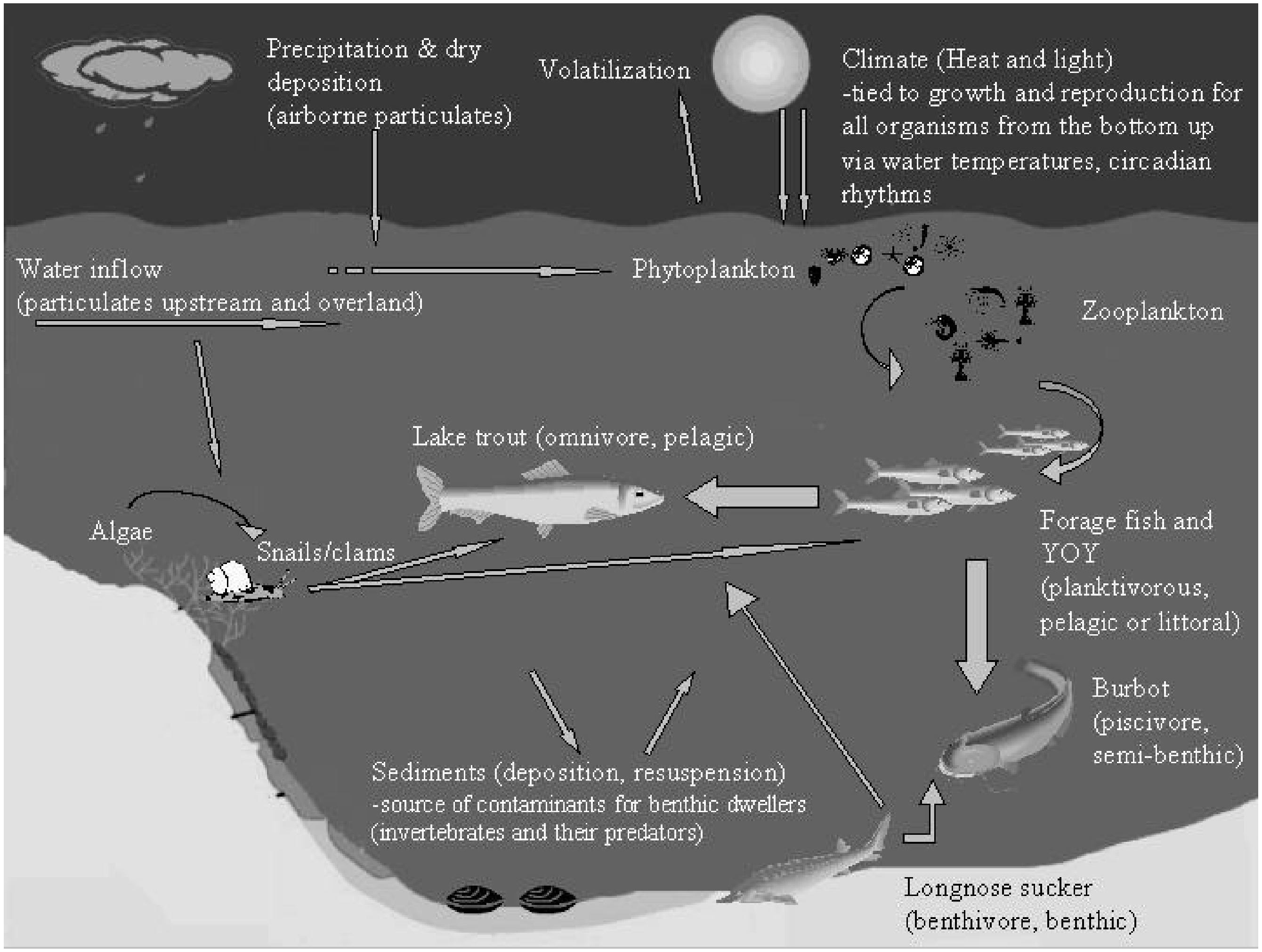


Figure 2.2 A simple lake-ecosystem model of biotic and abiotic interactions and contaminant transfers through the system.

increases in mean age of a fish species could be the result of recruitment failure, while a decrease in fecundity and condition factors may indicate an ecosystem limitation of food or habitat availability (Munkittrick and Dixon 1989). These population characteristics can be modified by changes in aquatic species populations in the food web either through natural ecosystem changes or anthropogenic influences (Mcdonald and Hershey 1989; Ferreri 1995; Hebert et al. 1997; O'Connor 2001; Mazumder and Edmundson 2002) with each change having a unique impact on the ecosystem dynamics. For example, shifts in zooplankton community composition towards larger bodied organisms may result in larger predatory fish as prey sizes influence growth and subsequently the body size of predators (Kerr 1971b; Matuszek et al. 1990; Trippel and Beamish 1993; Pazzia et al. 2002). The recruitment success of salmon in successive spawning years in Alaskan lakes has been linked to plankton biomass (Mazumder and Edmundson 2002; Edmundson et al. 2003), while establishment of a zooplankton species (*Mysis relicta*) in a Montana lake resulted in dramatic increases in lake trout populations (Stafford et al. 2002). Additionally, decreased lake trout growth has been associated with reductions in food availability thought to be caused by either increased trout abundances and competition for resources, or environmental conditions (Fabrizio et al. 2001) and fish production has long been correlated to annual phytoplankton production (Oglesby 1977; Downing et al. 1990). Fish stock exploitation typically results in higher fish growth rates as shown during the recovery of whitefish populations in the Northwest Territories (Healey 1975). The same response has been observed for other fish populations such as lake trout (Healey 1978; Ferreri 1995) and burbot (Hewson 1955). In order to account for shifts in OC concentrations in individual fish species from Lake Laberge, these influential population characteristics need to be considered in temporal trend surveys to identify underlying biotic principles for contaminant changes in aquatic animals.

The 1992-1994 research on Lake Laberge demonstrated that fish morphological, biochemical and population characteristics, other than stable isotopes (trophic levels), are significant predictors of OC concentrations (Kidd et al. 1995a; Kidd et al. 1998). These studies concur with other research that has also found correlations between OC contaminant concentrations and body size (Jensen et al. 1982; Smith 1998), age (Thomann and Connolly 1984; Larsson et al. 1991; Hammar et al. 1993) and lipid content (Larsson et al. 1991; Larsson et al. 1993; Bignert et al. 1993; Kucklick et al. 1996; Campbell et al. 2000). Other research has expanded on these correlations to include physicochemical characteristics of each chemical (Ruus et al. 2002). Additional factors affecting OC levels in fish includes metabolism of OC through food absorption or assimilation-excretion efficiencies (Jensen et al. 1982; Thomann 1989; Connolly 1991; Clark and Mackay 1991; Hammar et al. 1993; Gobas et al. 1999; Olsson et al. 2000; Fisk et al. 2001), biotransformation of OC (Sijm et al. 1992; Petersen and Kristensen 1998; Scott et al. 2002), reproductive loss through the parental transfer of OC to young (Sijm et al. 1992; Larsson et al. 1993; Jarman et al. 1996; Delorme et al. 1999), life stage (Sijm et al. 1992; Petersen and Kristensen 1998; Olsson et al. 2000) and growth (Jensen et al. 1982; Thomann and

Connolly 1984; Thomann 1989; Larsson et al. 1991; Sijm et al. 1992; Hammar et al. 1993).

Growth dilution is the reduction of contaminant concentrations in an organism through the proportional increase in tissue mass during periods of elevated growth (Figure 2.2). Biomass dilution is related to growth dilution but is defined more generally as reduction in contaminants through the increased mass from elevated growth within a community or population as a whole. These theories opposes the process of bioconcentration/bioaccumulation where bioconcentration is specifically defined as the "increase of pollutant concentrations from water when passing directly into aquatic species" while bioaccumulation includes a combined food, water or other sources of contaminant intake for this process (Moriarty 1988). Although growth may result in higher contaminant bioaccumulation through greater food consumption, evidence for the theory of growth dilution is supported by several studies. A model including growth rates and uptake efficiencies were better predictors of reductions in bioconcentration factors (by 2-5 fold) compared to simple lipid partitioning models in field studies of plankton (Thomann 1989). Fugacity ratios between plankton and sediment samples from Lake Erie showed that algal growth rates exceeding chemical kinetic rates, was an important factor in regulating contaminant exposure dynamics in the pelagic food web (Epplett et al. 2000). In addition, inverse relationships between OC concentrations and plankton biomass from 33 Southern Ontario lakes (Taylor et al. 1991), and Great Lakes algae (Lederman and Rhee 1982) were reported. Swackhamer and

Skoglund (1993) concluded that bioconcentration of PCB in plankton via physical kinetics is slow relative to phytoplankton growth. Increasing primary productivity was cited as the reason for lower levels of OC pollutants in northern pike (Larsson et al. 1992). Hammar et al. (1993) concluded that individual fish growth rates in late spring was the most important factor explaining the variation in DDT and PCB levels between two populations of Arctic char sampled from a lake in northern Sweden, while growth dilution was the only factor important in the uptake of higher chlorinated PCB in juvenile guppies (Sijm et al. 1992). Growth dilution may not solely be a factor of increased body mass but also related to body metabolism through a reduction in assimilated food that contain OC or increased efficiency in OC excretion as an organism has more access to food resources.

The ecology of each species will play an equally important role in delineating fish exposure to contaminants through the previously discussed routes. For example, the two primary sentinel species in Lake Laberge include burbot and lake trout. They each maintain a similar crossover in ecological niche, but species-specific characteristics set them apart. For example, it was observed that burbot produced 15 200- 224 000 eggs per pound of female fish, while lake trout have been reported to produce 400-1200 eggs per pound, providing a potential difference in energy utilization and contaminant loss through reproductive acitivites between these two species (Scott and Crossman 1973; Delorme et al. 1999). Burbot and trout are both piscivorous consumers, however, lake trout are more prone to being opportunistic omnivorous in consuming both fish and a variety of aquatic or terrestrial invertebrates (McPhail and Lindsey 1970; Scott and Crossman 1973). Lake trout also exist in a more pelagic niche while burbot are more benthic in their habitat preference. As such, burbot are exposed more to lake bottom processes (e.g. near sediment zones, detritus).

Using the correlations between the fish population characteristics and OC contaminants, researchers can make more accurate conclusions regarding the fate of contaminants influenced by changes in an aquatic ecosystem.

2.8 Climate Effects

Climate affects the biotic and abiotic processes in an aquatic ecosystem and may in turn impact the fate of OC in aquatic systems. Climate oscillations have been linked to significant biological processes such as salmon productivity (Mantua et al. 1997) and seasonal dynamics of plankton (Scheffer et al. 2001) through alterations in limnological variables such as thermocline depth, euphotic zone depth, water transparency and nutrient inflows (Schindler et al. 1996; Edmundson and Mazumder 2002). Primary productivity, and hence the potential for biomass dilution of contaminants, is highly controlled by climate factors like temperature and light (Northcote 1988; Straile and Geller 1998; Wetzel 2001) and by nutrients (Northcote 1988; Wetzel 2001). Warmer temperatures and more light result in higher primary productivity (Schindler et al. 1996; Findlay et al. 2001) although increased exposure to UV light can be harmful to aquatic plankton (Rautio 2001). Earlier spring melts and warmer temperatures broaden the phytoplankton growth season and leads to greater community diversity, composition, geographic distribution and food web structure changes (Schindler et al. 1996; Stemberger et al. 1996; Hairston 1996; Findlay et al. 2001; Weyhenmeyer 2001; Edmundson and Mazumder 2002; ACIA 2004) which may affect the movement of contaminants through the biotic compartment. Stewart et al. (2003) concluded that major changes in OC levels in Lake Winnipeg fish, after the 1997 flood event, were due to changes in the partitioning of OC at the base of the food web as the plankton community shifted towards a heavier percentage of calanoid copepods. Elevated melting of glaciers from warmer climate conditions in northern regions may provide more runoff and glacial silt carrying OC deposited at higher altitudes down into lower regions (Blais et al. 1998; Donald et al. 1999; ACIA 2004). Elevated levels of glacial silt can also reduce the euphotic zone depth resulting in a reduction of primary production in receiving waters (Edmundson et al. 2003) offsetting fast phytoplankton growth. Results from research on Toolik Lake (Alaska) concluded that young-of-the-year lake trout exposed to warmer climates, would need to consume eight times more food to attain the same end-of-year size as previous cohorts (Mcdonald et al. 1996). This change in ecological processes may increase or decrease OC concentrations in fish through elevated assimilation of contaminants or biomass dilution, respectively. Changes in precipitation may modify the amount of OC contaminants being depositied into ecosystems, affecting their bioavailability and uptake through dilution or changes in concentrations of dissolved humic substances (Larsson et al. 1992; ACIA 2004). Conversely, sustained warmer climates could reduce water levels through evaporation and concentrate contaminants into smaller lake volumes (ACIA 2004). It is believed that fluctuations in climatic conditions will have an effect on contaminant levels in both abiotic and biotic compartments, through various processes, in northern regions. Due to its potential impact, climate change needs to be considered in any long-term study of contaminants in Arctic and sub-Arctic regions (CACAR 2003c).

2.9 Temporal Trend Studies of Organochlorine Contaminants

Long-term temporal trend studies are necessary to account for annual variation in sample collections that may lead to inaccurate conclusions from analyzing short-term temporal data (Bignert et al. 1993). For example, Arnott et al. (1999) determined that zooplankton biodiversity and biomass were underestimated in a review of short-term studies on eight Canadian Shield lakes. Another study concluded that the interannual variation in spring zooplankton biomass in Lake Constance (Germany) was equal in magnitude to the response of the zooplankton community to long-term eutrophication in the ecosystem (Straile and Geller 1998); a conclusion that would not be observed with a short term study . Interannual and within-year variation were considerable factors when eight years of data on PCB in Baltic Sea fish were analysed (Bignert et al. 1993). Long-term trends were more readily apparent when a 17-year span of data were reviewed. This study concluded that interannual variation must be minimized by employing more frequent and long-term collections while continuing to isolate influential temporal factors affecting interannual variability (e.g. change in age, habitat, body size) in a contaminant dataset (Hebert and Haffner 1991; Bignert et al. 1993). It is important to include in these studies an assessment of the biotic parameters that can affect contaminant levels in abiotic and biotic compartments, to provide a thorough assessment of pollutant trends.

Long-term surveys of OC contaminants exist for several regions and animals including Great Lakes fish (Borgmann and Whittle 1991; Miller et al. 1992; Suns et al. 1993; De Vault et al. 1996; Madenjian et al. 1999), freshwater fish in the U.S. (Schmitt et al. 1999), Arctic seals, whales, polar bears (CACAR 2003a), birds (Carrera et al. 2002; Manosa et al. 2003), and marine fish in Norway (Skare et al. 1985) and the Baltic Sea (Bignert et al. 1993) although the information is somewhat limited. Usually only a few species (sometimes just one) from the food web were included in the surveys, studies were short or discontinuous, and few have observed correlations of contaminant shifts with climate or other factors previously discussed. Miller et al. (1992) observed a general decline in OC levels in lake trout in Lake Michigan and Lake Superior from 1975-1990, but only lipid content and ages were measured and used as factors to account for spatial and bioaccumulation differences between sub-species. A second study from Lake Ontario also observed reductions in overall OC levels in lake trout over an eleven year (1977-1988) period (Borgmann and Whittle 1991). The model developed in the study showed that alewife (the primary prey of lake trout) annual growth rates were closely related to mean PCB concentrations in trout but age, body size and reductions in lipid content were also reviewed as parameters influencing long-term temporal OC levels in trout. Trends for other species in the food web were not reviewed. A temporal trend study of PCB in Pottersburg Creek (Ontario) included collections of water, sediments, fish and invertebrates but the study only covered 5 years (Zaranko et al. 1997). Although the researchers concluded that PCB concentrations varied seasonally in biota, this may be explained as an artifact from interannual variation and that a longer study with more frequent sampling is required before drawing such a conclusion (Bignert et al. 1993). Stewart et al. (2003) did a thorough survey of Lake Winnipeg biota, water and sediments following a 1997 flood event and concluded that short-term variability in OC concentrations is more readily apparent in lower trophic level consumers. However, no further updates beyond 1995-2000 data on contaminant temporal trends for this lake have yet been published to corroborate the short term OC variability with data several years after the flood. The U.S. national contaminant biomonitoring program did an extensive survey of 117 locations around the country and collected 49 taxa of fish from 1976-1986 (Schmitt et al. 1999). The survey observed significant decreases of legacy OC contaminants (DDT, toxaphene, PCB, chlordane) in fish in the past two decades, which concurs with other research (Borgmann and Whittle 1991; De Vault et al. 1996). Although the study encompassed a wide geographic area and a long time span, the survey did not sample each site for each successive year. Only three fish types (one predatory and two benthic) were collected while other biota in the food webs, as well as the effects of ecological variables, abiotic inputs or climate influences went unobserved.

The ban of many OC chemicals has changed the focus of these long-term studies from global distillation-deposition influences to ecosystem processes

(Smith 1995; Stow et al. 1995; CACAR 2003a). A 20-year review of PCB concentrations in seven species of Lake Michigan fish showed a significant decrease followed by a stabilization of PCB levels through the mid-1980s, with some recent increases for Chinook and Coho salmon since the late 1980s. The increases were attributed to changes in growth dynamics in the mid trophic levels of the food web rather than to changes in atmospheric inputs or sediment sources (Stow 1995; Stow et al. 1995). A few studies have also reviewed the influence of climate on OC trends in Great lakes biota. Hebert et al. (1997) examined PCB in alewife and its primary predator, herring gulls, collected from Lake Ontario from 1974-1996. They concluded that PCB in gull eggs were correlated with seasonal climate fluctuations (as measured by heating degree days) and alewife biomass, suggesting that the seasonal growth of alewives is controlled by climatic factors ultimately influencing OC transferred into the herring gull diet and offspring. This was attributed to growth dilution of PCB in a fast growing alewife stock following years of depressed populations. Smith (1995) observed some OC chemicals behaving synchronously in biota across the Great Lakes and derived eight hypotheses to explain the phenomenon. The study examined historical data for herring gull eggs from the Great Lakes (1974-1992) and suggested that the synchronous contaminant trends were due to warmer spring weather, which is more conducive to phytoplankton growth. This resulted in biomass dilution of OC thereby producing less contaminated food for predatory herring gulls. With the observed increase in global temperatures (Macdonald et al. 2003; ACIA 2004), the effect of climate on abiotic and biotic movements of contaminants could be considerable. Both abiotic and biotic samples as well as annual climate conditions should be monitored in future temporal trend contaminant surveys.

3. TEMPORAL TRENDS OF ORGANOCHLORINE CONTAMINANTS IN BURBOT AND LAKE TROUT FROM THREE SELECTED YUKON LAKES

3.1 Abstract

Historical studies have demonstrated that organochlorine (OC) concentrations in top predators can vary considerably from lake to lake within a small geographic region but temporal trends of these contaminants have rarely been monitored in a sub-Arctic area for a long period of time. This study examined OC concentrations, including chlordane (CHL), DDT, hexachlorocyclohexane (HCH), toxaphene (CHB), PCB and chlorinated benzenes (CBz) in lake trout and burbot, from three Yukon lakes (Laberge, Kusawa, Quiet), over a span of 10 years (1993-2003). Temporal and spatial differences continue to exist in the OC concentrations of burbot and lake trout between these lakes. There is strong evidence that these contaminants are declining at various rates in lake trout (*Salveninus namaycush*) in Laberge, Kusawa and Quiet lakes. For example, ΣDDT concentrations have decreased 39%, 85% and 84% in Kusawa, Quiet and Laberge lakes respectively. Conversely, no consistent trends were observed in OC concentrations for burbot (*Lota lota*). For example, there is no evidence of a decline in toxaphene concentrations of Kusawa burbot yet a 58% decrease was observed in Laberge samples. Increases were also observed in the ΣHCH levels of Kusawa Lake

burbot, as well as increases in all OC groups (except ΣHCH) for the Quiet Lake burbot samples. Decreases were evident in ΣHCH and ΣCHB concentrations for Lake Laberge burbot and in ΣCHL for the Kusawa Lake samples. Spatial variations in OC levels are quite evident as Lake Laberge trout and burbot continued to maintain the highest levels over the 11-year period from 1992 to 2003 followed by Kusawa Lake and then Quiet Lake. These differences are related to a variety of factors especially the species morphological characteristics such as fish age, mass and lipid content. A decreasing trend in Quiet and Laberge lake trout lipid content, coupled with fluctuating condition factors and increases in body masses, suggest biotic changes may be occurring within the food webs due to fish population variations related to the cessation of commercial fishing or potentially an increase in lake plankton production related to annual climate variation. It is suspected that biotic factors rather than atmospheric inputs are the primary factors affecting the contaminant concentrations in lake trout and burbot in the study lakes.

3.2 Introduction

The Arctic was once considered a pristine environment; however, over the last 20 years, significant levels of organochlorine (OC) pesticides such as toxaphene (chlorobornanes) and hexachlorocyclohexane (HCH) and industrial chemicals such as polychlorinated biphenyls (PCB) have been found in its ecosystem and in the traditional foods eaten by local people (Muir et al. 1992; Lockhart et al. 1992; CACAR 2003b). It is well documented that long range atmospheric transport is the primary source of these contaminants to the northern regions (Bidleman 1999) and that once there, these chemicals bioaccumulate and biomagnify to upper trophic levels of the food web (Barrie et al. 1992); (Bidleman et al. 1989). Many monitoring projects have begun to assess the temporal levels of these contaminants in aquatic and terrestrial animals. Since atmospheric levels of some OC (lower chlorinated PCB, HCH, chlordanes and DDT) seem to have decreased in the Arctic over the past decade (CACAR 2003c; Stern et al. 2005), questions remain as to whether the levels of these compounds are also decreasing in northern aquatic biota.

Temporal studies of OC pesticides in aquatic biota are important in determining the time it takes for abiotic changes in contaminant levels to manifest into changes within the living parts of an ecosystem (Manosa et al. 2003) . However, changes in aquatic biota OC concentrations cannot be easily or directly predicted from atmospheric values or geographical location because of a variety of factors that influence tissue OC concentrations in organisms (Larsson et al. 1993; Kidd et al. 1993; Ruus et al. 2002). Population, morphology and biochemical characteristics can be used as predictors of organochlorine contaminant trends in fish and to assess overall ecosystem health. A few of these factors include lake productivity (Larsson et al. 1992), food web trophic structure (Kidd et al. 1995a; Vander Zanden and Rasmussen 1996), growth rates (Hammar et al. 1993; Olsson et al. 2000), diet (Manosa et al. 2003) and reproductive transfer/loss (Rasmussen et al. 1990; Larsson et al. 1993). In order to determine the trend of contaminants in fish, biological parameters such as lipid content, size and age must also be assessed (Larsson et al., 1993). Some are significant predictors of OC levels in biota and hence better predictors of OC trends within a specific ecosystem (*e.g.* a lake) than environmental concentrations. With an increasing number of temporal data points and information on species ecology and morphology, more precise conclusions can be drawn about OC trends and the changes in an ecosystem over time (Bignert et al. 1993).

Kidd et al. (1998) conducted a food web study on Lake Laberge, a sub-Arctic lake in the Yukon Territory. Fish from this lake were found to have considerably higher levels of OC contaminants compared to fish in other regional lakes such as Kusawa and Fox Lakes. It was found that lake trout, burbot and even lake whitefish in Lake Laberge were more piscivorous than their counterpoints in the other regional lakes (Kidd et al. 1995a; Kidd et al. 1998). This was attributed to the longer food chain length in Lake Laberge and the higher lipid content in the fish. Since 1992, tissues from lake trout (*Salvelinus namaycush*) and burbot (*Lota lota*) have been intermittently measured for OC levels in Laberge, Kusawa and Quiet lakes (Figure 3.1). Lake trout were until recently a commercially exploited fish, while local First Nations people prize burbot livers as a delicacy. The lake trout and burbot in these three Yukon lakes remain the focus of ongoing research to study the long-term temporal trends of contaminants in northern biota in the lower Yukon region.

In this study I report on the temporal trends of 6 major OC contaminant groups including chlorinated benzenes (CBz), chlorinated bornanes (CHB),

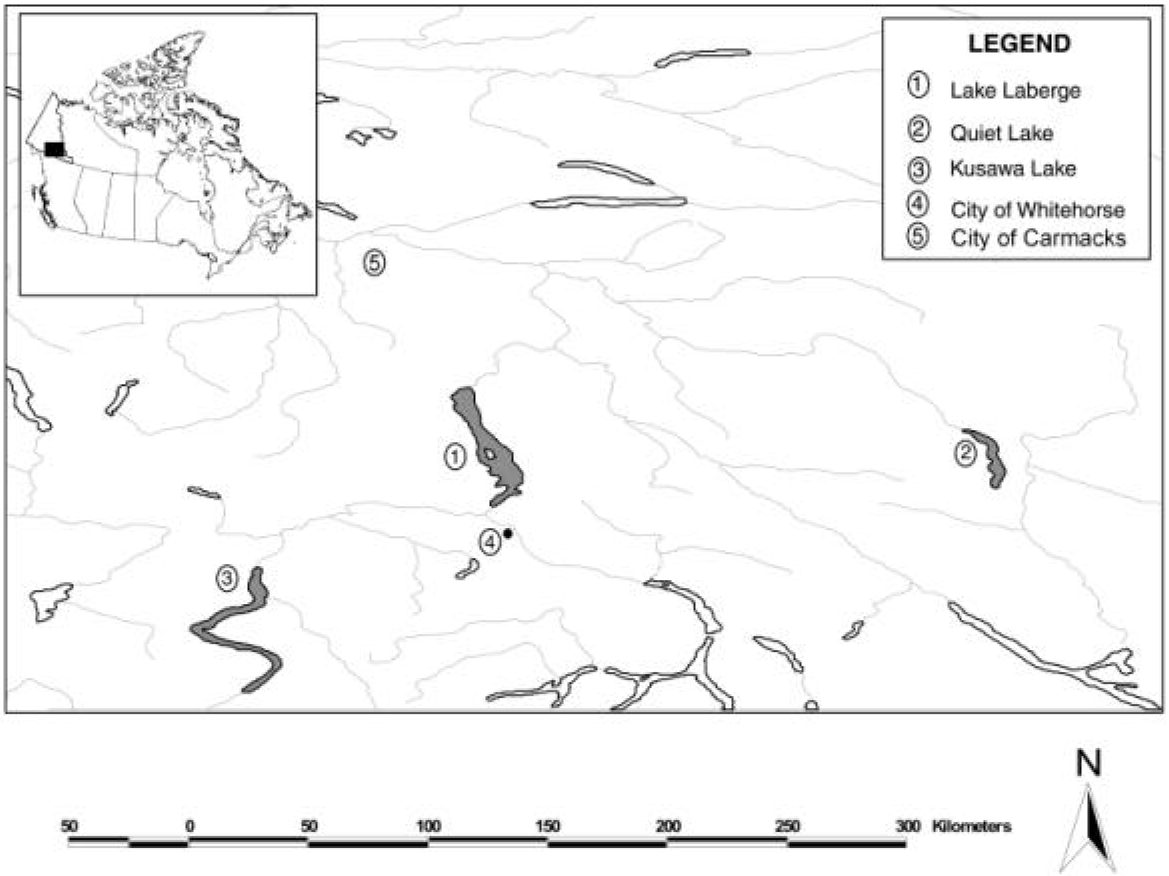


Figure 3.1. Map of Lake Laberge, Kusawa, and Quiet Lake in the Yukon Territory, Canada.

chlordanes (CHL), DDT, HCH and PCB in Quiet, Kusawa and Lake Laberge, over a span of 11 years (1992-2003). Lake trout and burbot were selected because other their importance to local sport fisherman and First Nations bands subsistence fisheries and the availability of archived samples.

3.3 Materials & Methods

3.3.1 Sample Collections

Lake trout (*Salvelinus namaycush*) and burbot (*Lota lota*) were collected from three Yukon lakes (Table 3.1) over multiple years. Lake trout collection occurred during the summers of 1993, 1999, 2001, 2002 for Quiet Lake, in 1993, 1999, 2001 and 2002 for Kusawa Lake and 1993, 1996, 2000, 2001 and 2002 for Lake Laberge. Burbot were collected in the spring and summers of 1994, 1997 and 1999 for Quiet Lake, in 1993 and 1999 for Kusawa Lake and in 1993, 1996, 1999, 2000 and 2001 for Lake Laberge. The primary collection years were 1993 and 2001. All other years maintained smaller sampling sizes due to logistical constraints.

Burbot were caught using long line angling while lake trout were captured using small mesh index nets (Thompson 1996; Foos 1998). Netting techniques, mesh sizes and capture locations were kept as similar as possible over time. Yukon Territorial Government fishing quotas limited capture of lake trout to 15 or less for some years. Morphological measurements included sex, fork lengths and weights (Table 3.2). Otoliths were removed for age analysis. Morphological measurements and tissue sampling were also done at the Indian and Northern Affairs lab in Whitehorse. Condition factors (K) were calculated using [weight (g) x 10^5 / (length (mm))^3]. Samples were wrapped in baked, and solvent-washed foil and shipped frozen to the Freshwater Institute in Winnipeg for analysis. Sections of trout dorsal muscle and burbot livers were frozen and kept at –20 ^{o}C prior to organochlorine analysis.

In addition, the Yukon Territorial Government (Renewable Resources) conducted population assessments of the fish stocks in 1993 for Kusawa Lake, 1993 and 1999 for Lake Laberge and in 1994 for Quiet Lake (Thompson 1996; Foos 1998; Foos 2001). Limnological data for the lakes was collected several times over the past two decades (Shortreed and Stockner 1983; Jack et al. 1983; Kirkland and Gray 1986; Kidd 1996; Thompson 1996; Foos 1998). The three lakes are all within a 125 km radius of the city of Whitehorse, possessing similar elevations, maximum depths and other parameters (Table 3.1), but Quiet Lake is notably smaller in overall volume and surface area.

3.3.2 Organochlorine Analysis

Methods for organochlorine extractions followed the general methods listed in Kidd et al. (1998). Fish muscle (10-15g) was homogenized with dry ice and mixed with 10g baked anhydrous sodium sulphate (600 ^{o}C for 6 hours). Each sample was spiked with 10 μL of PCB30 and octachloronapthalene (OCN) as internal standards. The material was then mixed with 10g of baked hydromatrix (600 ^{o}C for 6 hours) in an extraction cell and topped up with baked Ottawa sand (600 ^{o}C for 6hrs). A Dionex ASE300 was used with pesticide grade dichloromethane (DCM) and pesticide grade hexane (mixed 50:50) as the solvent. The solution was then mixed with 10g of sodium sulphate and allowed to settle for three hours. The extract was then reduced in a Rotovap and filtered through millipore filters (0.2 µm pore size - Type FG) followed by a DCM-hexane rinse bringing the extract up to 10mL in volume. Aluminum dishes were weighed and 1mL of the extract was removed to the dish to calculate tissue lipid (%) content following drying. The remaining 9mL was processed with rinses of 50:50 DCM-hexane through manual gel permeation chromatography (GPC), containing Envirobeads (S-X3). The eluent was concentrated in a Rotovap and evaporated further under a stream of nitrogen to 1mL for fractionation on a 1.2%-deactivated Florisil column. Recoveries of the internal standard PCB30 and OCN were 74.8% ± 13.6% and 79.4% ± 13.5% for lake trout, respectively.

Burbot livers were extracted using the Ball Mill method. Approximately 2.2g of liver were added to a ball mill tube with 10g of baked sodium sulphate, 25mL of hexane, 10µL of the internal standard (PCB30-OCN) and a steel ball. The tubes were sealed and shaken for 15 minutes then allowed to stand for 4 hours. After settling, the tubes were centrifuged for 10 minutes at 2000 rpm. The extract was decanted and another 25mL of hexane was added to each ball mill tube and re-shaken for 15 minutes. All 3 rinse extracts (75mL) were combined and reduced in a Rotovap. Samples were brought up 11mL in a graduated cylinder with hexane. Exactly 1mL was removed to weighed aluminum dishes for lipid content calculation following drying. Exactly 0.50mL of the remaining 10mL sample was used for fractionation on a 1.2%-deactivated Florisil column.

Table 3.1 Physical and chemical characteristics of Kusawa, Laberge and Quiet Lakes, Yukon Territory. All values are historical means (ranges) from various sources. Total dissolved solids (TDS), dissolved organic carbon (DOC), total phosphorous (TP), total dissolved phosphorous (TDP), total dissolved nitrogen (TDN).

	Laberge	Kusawa	Quiet
Max. length (km)	58 (48) [c (d)]	57 [c]	32 [c]
Max. width (km)	6 (4.2) [c (d)]	2.5 [c]	3.2 [c]
Latitude	61°11'N [c, d]	60° 20'N [f]	61° 05'N [a]
Longitude	135° 12'W [c, d]	136° 22'W [f]	133° 05'W [a]
Elevation (m)	628 [c]	672 [c]	787 (802) [b (c)]
Surface area (km^2)	213 (201) [c (d)]	142 [c]	37.8 [b]
Volume (km^3)	10.8 [d]	7.72 [f]	1.24 [b]
Maximum depth (m)	146 [d]	140 [f]	170 [b]
Mean depth (m)	54 [d]	54 [f]	32.9 [b]
pH	7.9	7 [c]	7.4 [a, b]
Drainage area (km^2)	25 385 [e]	4070 [e]	-
Flushing rate (yr^{-1})	0.93 [d]	-	-
	1.06 [e]		
Conductivity (μS/cm)	113 (56-370) [e (g)]	40 [c]	55 [b]
		74 (41-309) [e]	80 [c]
TDS (mg/L)	62 (54-75) [e]	40 [c]	57 [a]
		25 (19-36) [e]	35 (80) [b (c)]
DOC (mg/L)	2.61 (1.55-6.12) [e]	3.37 (1.70-9.36) [e]	
TP (μg/L)	5.1 (3.5-8.1) [e]	3.6 (2.2-5.2) [e]	4 [a]
	3.8-6.7 [d]		
TDP (μg/L)	2.5 (1.4-6.1) [e]	1.8 (0.9-4.0) [e]	1 [a]
	1.3-2.5 [d]		
TDN (μg/L)	66.2 (61.4-75.2) [e]	82.5 (72.1-92.6) [e]	177 [a]
	57-118 [d]		
NO_3 (μg/L)	16 (3.8-30) [e]	25.1 (18.3-28.8) [e]	15.5 [a]
NH_3 (μg/L)	15.2 (6.4-20.6) [e]	12 (2.2-19.6) [e]	
Secchi depth (m)	N/A	6.5 [c]	7.1-9.5 [a, c]
Alkalinity (mg/L $CaCO_3$)		15 [c]	38.3-44.0 [a, c]

[a] (Shortreed and Stockner 1983)
[b] (Foos 1998)
[c] (Lindsey et al. 1981)
[d] (Kirkland and Gray 1986)
[e] (Kidd 1996)
[f] (Thompson 1996)
[g] (Godin and Jack 1984)

Recoveries of the internal standard PCB30 and OCN were 80.5% ± 28.2% and 87.7% ± 17.4% for burbot, respectively.

All fish muscle and burbot liver extracts following were injected onto a Varian Star 3400 GC equipped with an electron capture detector (Ni^{63} electron source) and an 8100 auto-sampler. The column used was a J&W Scientific (Agilent Tech) DB-5, 60m, 250 μm ID with TMP as the solvent, Hydrogen Ultra High Purity (UHP) carrier gas (at ~2 mL/min), nitrogen UHP make-up gas (at ~50mL/min). Injection and temperature programs conditions were as follows: Injector temperature 80°C, detector temperature 300°C, initial temperature 100°C (hold time 2min), 1st temperature program increasing to 150°C at 15°C/min, 2nd temperature program increasing to 265°C at 3°C/min. Aldrin was used as a volume corrector prior to GC analysis.

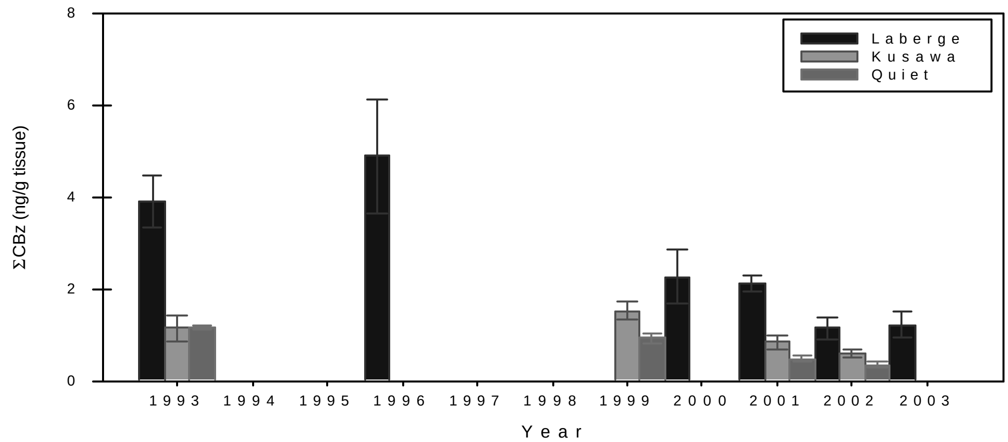
3.3.3 Statistical Analysis

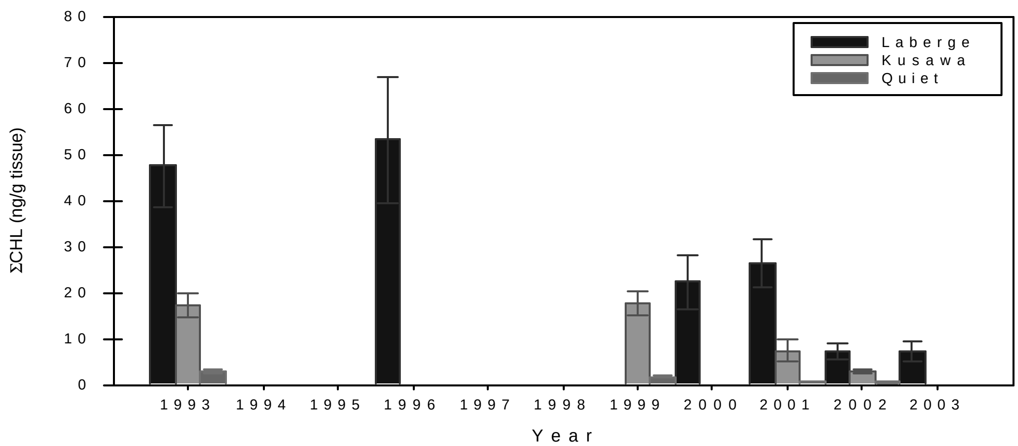
Statistical analyses were conducted using Systat 7.0® for Windows. Analyses include ANOVA on major OC categories grouped by fish species and by lake tested for significant differences across years. OC data were $\log_{10}$ transformed prior to statistical analyses to normalize the distribution of the data. For each fish species within each lake, temporal differences in OC, condition factors and lipid content were tested using ANOVA (p =0.05) followed by Tukey's post-hoc test. OC concentrations were not adjusted using analysis of covariance (ANCOVA) because of heterogeneity of regression slopes indicating significant interactions of both log weights and lipid content with OC over time (across years). Relationships between OC concentrations and biological parameters including tissue lipid (%), $\log_{10}$ weights, $\log_{10}$ lengths and $\log_{10}$ ages were analysed using general linear multiple regressions (GLMR). Based on research from Kidd (1996), models for lake trout included log weight and tissue lipid (%) while the models for burbot included weight, tissue lipid (%) and log age (where possible). Log length was removed from the models due to preliminary testing indicating a highly insignificant correlation with OC levels compared to either lipid or log weight. Regressions were run to determine the most influential factors on OC concentrations by fish species by lake within each year. Results are reported for the points where a factor other than lipid is a primary predictor of OC. Results are also reported with *p* values for the whole model and factors within the model format.

3.4 Results

3.4.1 Kusawa Lake

Kusawa lake trout exhibited significant decreases in most OC from 1993 to 2002. This included a drop of 47% in ΣCBz (p=0.01), 39% in ΣDDT (p<<0.05), 48% in ΣHCH (p=0.006), 82% in chlordanes (p<<0.05), 62% in ΣPCB (p<<0.05) and 64% in ΣCHB (p=0.004) when comparing 2002 to 1993 OC levels (Figure 3.2, Table 3.3). The significant decline in ΣPCB concentrations can be generally attributed to the decreases in the hepta-, hexa- and octa- PCB homologue groups (Table 3.3). No among-year differences were observed in the ratios of *p,p'*-DDT/ΣDDT in Kusawa except in one year, 1993, when the *p,p'*-DDE level





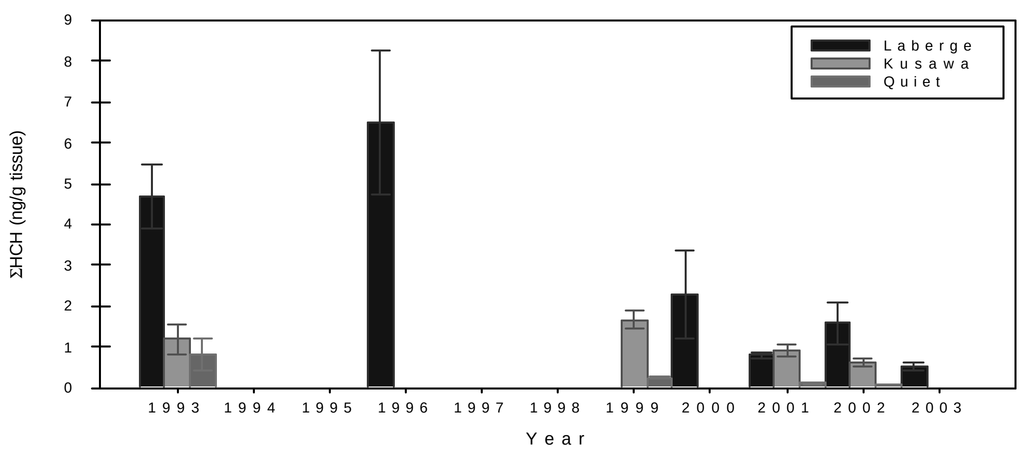


Figure 3.2. Mean concentrations (± SE) of ΣCBz, ΣCHL, ΣHCH, ΣDDT, ΣPCB and Toxaphene (ΣCHB) in lake trout muscle tissue from Laberge, Kusawa, and Quiet Lake (1993-2003).

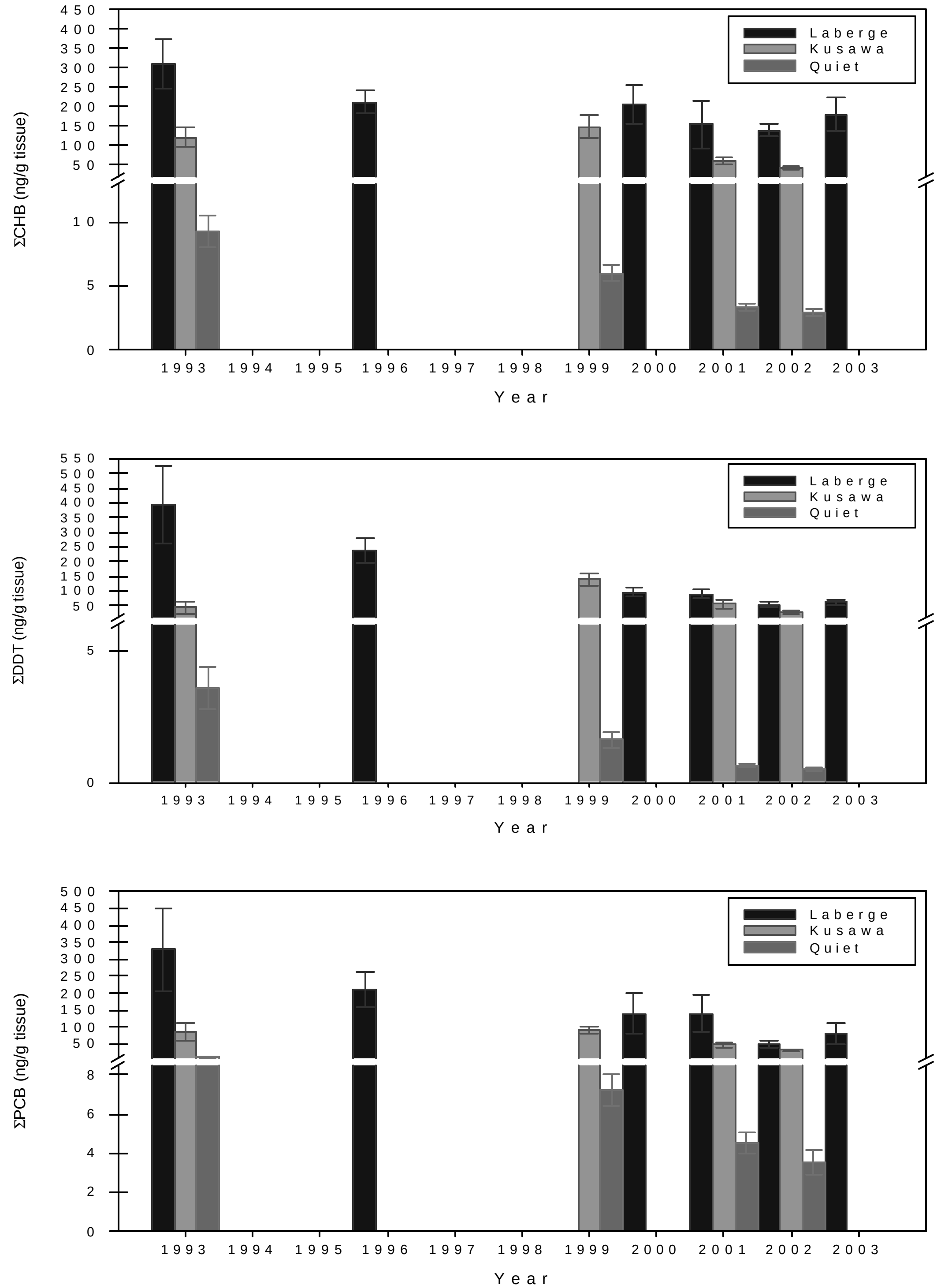


Figure 3.2 (cont'd). Mean concentrations (± SE) of ΣCBz, ΣCHL, ΣHCH, ΣDDT, ΣPCB and Toxaphene (ΣCHB) in lake trout muscle tissue from Laberge, Kusawa, and Quiet Lake (1993-2003).

constituted a greater proportion of ΣDDT compared to any other year. While ΣCHL levels and those of major chlordane related compounds and metabolites, such as *trans*-nonachlor and oxychlordane, declined significantly over this 11 year time period, there were no apparent trends in chlordane congener proportions. The ΣHCH concentrations declined steadily over the 11-year time period (~50%), as did the proportion of α-HCH relative to γ-HCH (Table 3.3).

The data from Kusawa Lake have revealed moderate differences in trout morphological parameters over the 9 years (four time points) between 1993-2002. Of the Kusawa lake trout sampled, mean ages decreased from 19 years (SE ±2 y) in 1993 to 12 years (SE ±1 y) in 2002 (Table 3.2). The results of the ANOVA and Tukey's tests showed a significant difference in the condition factor (K) over time in Kusawa trout (p=0.003) mostly due to a peak in the 1999 average that is statistically higher compared to all other years. Both the 1993 and 2002 Kusawa lake trout had a mean K of 1.0 (Table 3.2). A significant difference was also evident in lipid content across years in Kusawa lake trout (p=0.001). This is due to the significantly larger mean for fish sampled in 1999, which measured 2.2% higher than the next highest year (2001). There is no significant difference between 1993 and 2002 lipid averages, which measured 1.8% and 1.4% respectively (Table 3.2). The trout collected from Kusawa Lake in 1999 were higher in fat and larger in girth compared to fish from the other collection years but the lake trout have not changed significantly in morphology when comparing the 1993 fish to those collected in 2002.

Regressions of the organochlorine groups across years (using log weight, % lipid) were used to determine which factors significantly predicted OC concentrations. Age was also used to examine correlations with OC due to the significant change in mean age for lake trout from Kusawa Lake. Tissue lipid (%), and log weight both accounted for the majority of variability in the OC concentrations in trout except for 1993 ΣCBz (p =0.07), ΣCHL (*p*=0.32), ΣPCB (*p*=0.13) and ΣCHB (*p*=0.35). The model for 1993 ΣDDT was significant (Table 3.5) although neither log weight or lipid content were considered highly significant factors. Log weight was a significant predictor in 2001 as the regression model accounted for 91% to 98% of the within-lake variability for ΣHCH, ΣPCB, ΣDDT and ΣCHB in trout muscle samples (Table 3.5). Tissue lipid (%) was otherwise the most significant variable in OC concentrations for 2001. Age was not significantly correlated to any OC group for any year.

The Kusawa burbot showed no statistical evidence of changes in ΣDDT, ΣPCB, ΣCHB and ΣCBz concentrations over 7 years (only two data points) measured (*p*=0.09, 0.44, 0.11 and 0.63 respectively; Table 3.4, Figure 3.3). However, as was observed in the lake trout, there was a decrease in ΣCHL (38%, *p*=0.02) as well as a significant increase in γ–HCH concentrations (138%, *p*<0.05).

Analyses of Kusawa burbot livers and morphometrics data have shown different results compared to the lake trout. Kusawa burbot ages could not be analysed for temporal trends based on a single datum (Table 3.2). No significant differences were measured (using ANOVA) between the 1993 and 1999 Kusawa

Table 3.2 Morphological data (means ± SE) for lake trout and burbot from Laberge, Kusawa and Quiet lakes from 1993-2003.

	Lake	Year	N (*)	Age	Length (mm)	Weight (g)	K	% lipid
Burbot	**Kusawa**	**1993**	4		737.5 ± 48.9	2837.5 ± 725.8	0.7	27.9 ± 3.0
		1999	11 (10)	10 ± 1	489.1 ± 22.5	795.5 ± 108.4	0.6	23.8 ± 2.6
	Laberge	**1993**	13 (12)	13 ± 2	592.9 ± 22.6	1427.7 ± 158.6	0.7	42.6 ± 2.8
		1996	15		485.3 ± 26.6	921.7 ± 145	0.7	36.8 ± 2.3
		1999	11	17 ± 1	647.7 ± 21.9	1518.2 ± 153	0.5	53.8 ± 2.0
		2000	17	15 ± 1	623.2 ± 25.7	1371.2 ± 140.1	0.5	28 ± 2.3
		2001	30	16 ± 1	624.5 ± 17.7	1347.7 ± 112.1	0.5	23.1 ± 2.6
	Quiet	**1994**	5		477 ± 47.3	660 ± 137.3	0.6	20.6 ± 2.7
		1997	6	13 ± 2	606.7 ± 37.9	1500 ± 290.5	0.6	29.4 ± 5.5
		1999	3	14 ± 2	625 ± 44.4	1883.3 ± 518.3	0.7	18 ± 2.9
Lake trout	**Kusawa**	**1993**	10	19 ± 2	525 ± 28.6	1693.2 ± 335.9	1.0	1.8 ± 0.5
		1999	14 (12)	18 ± 1	514.6 ± 28.3	1839.3 ± 417.4	1.2	4.6 ± 0.8
		2001	9 (8)	12 ± 1	550.6 ± 39.3	2033.3 ± 433	1.1	2.4 ± 0.5
		2002	10 (8)	12 ± 1	500 ± 24.7	1344 ± 213.9	1.0	1.4 ± 0.3
	Laberge	**1993**	24 (8)	15 ± 2	479.7 ± 15.7	1522.7 ± 191.1	1.2	7.9 ± 0.9
		1996	13 (9)	22 ± 5	491.2 ± 27.3	1867.3 ± 415	1.4	9.6 ± 1.4
		2000	6	12 ± 2	590 ± 44	2753.3 ± 812.1	1.2	3.7 ± 0.8
		2001	16	14 ± 2	648.4 ± 26.5	3366.3 ± 410.3	1.2	5 ± 0.5

Lake trout	**Kusawa**	**2002**	5 (2)	12 ± 4	616 ± 17.1	2874 ± 336.7	1.2	4.2 ± 0.9
		2003	8		592.5 ± 34.7	2485 ± 427.1	1.1	4.7 ± 0.8
	Quiet	**1992**	5		537 ± 68.2	1816 ± 578.6	1.1	5.6 ± 0.8
		1994	97 [b]	14 ± 1	498 -	1408 -	1.1	-
		1999	8 (7)	20 ± 2	561.3 ± 17	2087.5 ± 187.7	1.2	2.4 ± 0.3
		2001	9 (7)	13 ± 2	486.7 ± 65.7	1524.4 ± 394.3	0.9	0.9 ± 0.2
		2002	9 (5)	11 ± 2	595 ± 58	2557.8 ± 717	1.0	0.7 ± 0.2

[a] Number of samples actually used for ageing.
[b] data from (Foos 1998)

Table 3.3 Mean (±SE) congener concentrations (ng/g ww) for major OC groups in lake trout muscle from Laberge, Kusawa and Quiet lakes from 1993-2003.

	Kusawa				**Laberge**						**Quiet**			
	1993	**1999**	**2001**	**2002**	**1993**	**1996**	**2000**	**2001**	**2002**	**2003**	**1992**	**1999**	**2001**	**2002**
å **PCB**	85.62	91.09	48.55	32.45	328.28	209.32	138.95	139.71	48.60	81.01	11.60	7.24	4.51	3.51
	(26.07)	(11.85)	(7.91)	(3.66)	(121.49)	(52.08)	(60.89)	(53.75)	(8.81)	(29.83)	(1.96)	(0.80)	(0.55)	(0.62)
Mono-di-triCB	2.37	6.01	3.65	2.46	11.43	16.84	3.83	6.60	3.66	2.10	0.63	0.70	0.47	0.37
	(0.53)	(0.74)	(0.59)	(0.24)	(2.34)	(5.48)	(1.37)	(2.25)	(0.86)	(0.54)	(0.16)	(0.09)	(0.06)	(0.08)
tetraCB	2.35	5.86	3.55	2.40	11.20	16.36	3.52	6.29	3.51	1.98	0.63	0.62	0.43	0.34
	(0.53)	(0.71)	(0.57)	(0.23)	(2.30)	(5.30)	(1.29)	(2.26)	(0.80)	(0.52)	(0.16)	(0.08)	(0.06)	(0.07)
pentaCB	16.30	25.31	12.52	8.25	71.99	56.51	26.51	24.44	12.42	17.60	3.15	1.66	0.97	0.79
	(3.81)	(3.25)	(2.13)	(0.82)	(23.32)	(15.65)	(14.92)	(10.38)	(2.18)	(8.08)	(0.77)	(0.19)	(0.11)	(0.13)
hexaCB	43.66	39.39	23.80	17.08	150.61	76.53	69.68	52.89	23.06	39.76	5.72	3.29	1.93	1.45
	(14.79)	(4.51)	(3.36)	(1.95)	(60.95)	(17.19)	(27.87)	(12.95)	(4.23)	(12.23)	(1.02)	(0.36)	(0.23)	(0.24)
heptaCB	19.40	16.30	6.66	3.67	73.62	45.58	31.99	44.93	7.24	17.43	1.22	1.15	0.80	0.66
	(5.98)	(2.78)	(1.42)	(0.50)	(27.92)	(10.33)	(13.14)	(22.79)	(1.04)	(7.36)	(0.23)	(0.13)	(0.09)	(0.11)
octaCB	3.52	2.70	1.18	0.61	17.76	9.01	6.21	8.60	1.29	3.53	0.08	0.01	<0.01	<0.01
	(1.03)	(0.45)	(0.29)	(0.11)	(7.82)	(2.10)	(3.87)	(4.93)	(0.28)	(1.90)	(0.03)	(<0.01)	(<0.01)	(<0.01)
å **DDT**	44.16	139.16	56.58	26.66	391.54	236.51	96.46	89.46	54.50	61.48	3.59	1.62	0.66	0.53
	(21.50)	(19.72)	(15.30)	(4.15)	(132.69)	(41.39)	(14.21)	(14.04)	(11.58)	(8.55)	(0.80)	(0.28)	(0.08)	(0.09)
p,p'-DDT	4.82	32.65	15.35	9.09	86.49	48.09	23.83	21.57	6.29	16.41	0.32	0.14	0.08	0.07
	(2.47)	(3.81)	(3.14)	(1.21)	(37.03)	(7.15)	(4.56)	(2.64)	(1.70)	(1.83)	(0.06)	(0.02)	(0.01)	(0.01)
p,p'-DDE	32.62	55.21	22.11	11.19	191.32	96.70	27.01	32.32	18.23	17.94	2.03	1.09	0.38	0.32
	(16.42)	(8.36)	(5.82)	(1.71)	(71.58)	(17.42)	(2.37)	(5.27)	(3.18)	(2.52)	(0.38)	(0.20)	(0.04)	(0.05)
p,p'-DDD	3.41	34.57	12.25	3.23	83.06	60.75	32.12	21.20	19.04	21.46	0.20	0.03	0.03	0.02
	(2.27)	(5.56)	(4.71)	(0.72)	(22.26)	(10.55)	(4.12)	(1.77)	(3.14)	(2.66)	(0.04)	(0.01)	(0.01)	(<0.01)
å **CBz**	1.15	1.52	0.84	0.61	3.92	4.90	2.26	2.11	1.15	1.21	1.15	0.93	0.47	0.35
	(0.28)	(0.20)	(0.14)	(0.09)	(0.57)	(1.24)	(0.59)	(0.17)	(0.25)	(0.28)	(0.04)	(0.11)	(0.07)	(0.07)
å **CHB**	120.80	148.38	61.03	43.47	310.96	212.23	207.33	154.20	139.23	179.31	9.29	5.99	3.30	2.89
	(24.94)	(29.29)	(8.55)	(5.02)	(62.36)	(28.31)	(49.90)	(60.46)	(16.88)	(42.79)	(1.25)	(0.67)	(0.27)	(0.31)

	Kusawa				Laberge						Quiet			
	1993	**1999**	**2001**	**2002**	**1993**	**1996**	**2000**	**2001**	**2002**	**2003**	**1992**	**1999**	**2001**	**2002**
å **CHL**	17.33	17.82	7.45	3.01	47.60	53.38	22.36	26.37	7.26	7.44	3.01	1.66	0.79	0.62
	(2.78)	(2.74)	(2.35)	(0.48)	(8.84)	(13.74)	(5.84)	(5.14)	(1.59)	(2.24)	(0.43)	(0.26)	(0.09)	(0.12)
oxychlor	0.82	1.17	0.48	0.24	3.02	3.98	0.44	0.62	0.98	0.19	0.32	0.15	0.09	0.06
	(0.21)	(0.18)	(0.14)	(0.03)	(0.69)	(1.28)	(0.10)	(0.22)	(0.22)	(0.06)	(0.11)	(0.02)	(0.01)	(0.01)
cis-	1.63	1.14	0.51	0.26	4.32	5.30	2.97	2.10	0.55	1.86	0.15	0.25	0.08	0.06
chlordane	(0.34)	(0.16)	(0.15)	(0.05)	(0.75)	(1.55)	(0.94)	(0.47)	(0.11)	(0.66)	(0.06)	(0.06)	(0.01)	(0.01)
trans-	0.27	1.40	0.54	0.24	3.60	5.56	0.27	0.54	0.43	0.18	0.20	0.13	0.06	0.05
chlordane	(0.06)	(0.21)	(0.17)	(0.04)	(0.69)	(1.48)	(0.06)	(0.11)	(0.10)	(0.05)	(0.02)	(0.02)	(0.01)	(0.01)
trans-	6.87	6.73	2.70	0.80	16.69	15.48	7.94	8.91	1.93	2.51	1.27	0.62	0.29	0.22
nonachlor	(1.14)	(1.09)	(1.01)	(0.11)	(3.60)	(3.30)	(2.87)	(3.03)	(0.39)	(0.85)	(0.24)	(0.10)	(0.03)	(0.04)
cis-	5.60	4.52	1.87	0.60	11.87	11.24	9.22	5.22	0.90	2.13	0.39	0.16	0.10	0.08
nonachlor	(0.93)	(0.74)	(0.65)	(0.12)	(2.43)	(2.42)	(1.80)	(1.05)	(0.19)	(0.60)	(0.07)	(0.03)	(0.01)	(0.02)
å **HCH**	1.21	1.68	0.91	0.62	4.69	6.50	2.30	0.80	1.58	0.54	0.82	0.25	0.11	0.08
	(0.36)	(0.23)	(0.14)	(0.08)	(0.78)	(1.79)	(1.08)	(0.07)	(0.50)	(0.10)	(0.37)	(0.04)	(0.02)	(0.02)
a-HCH	1.08	1.36	0.75	0.52	3.70	4.71	0.89	0.44	1.24	0.40	0.67	0.20	0.08	0.06
	(0.33)	(0.17)	(0.11)	(0.07)	(0.55)	(1.17)	(0.21)	(0.04)	(0.40)	(0.08)	(0.36)	(0.03)	(0.02)	(0.02)
β-HCH	0.01	0.07	0.04	0.03	0.16	0.34	1.27	0.22	0.09	0.05	0.05	<0.01	<0.01	<0.01
	(<0.01)	(0.01)	(0.01)	(0.01)	(0.05)	(0.11)	(0.89)	(0.04)	(0.03)	(0.04)	(0.01)	(<0.01)	(<0.01)	(<0.01)
g-HCH	0.12	0.25	0.12	0.07	0.82	1.45	0.14	0.14	0.26	0.09	0.10	0.04	0.03	0.02
	(0.03)	(0.05)	(0.02)	(0.01)	(0.20)	(0.52)	(0.03)	(0.01)	(0.07)	(0.02)	(0.02)	(0.01)	(<0.01)	(<0.01)

ΣDDT= *p,p*'-DDT, *p,p'*-DDE, *p p*'-DDD, *o,p*'-DDT, *o,p*'-DDE and *o,p*'-DDD; ΣHCH= α- β- and γ-HCH; ΣCHL= sum of all technical chlordane compounds and related metabolites, including heptachlor epoxide; ΣCBz = 1245-TCB, 1234-TCB, P5CBz and HCBz; ΣPCB = CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114,131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209

Table 3.4 Mean (±SE) congener concentrations (ng/g ww) for major OC groups in burbot liver from Laberge, Kusawa, Quiet Lakes from 1993-2001

	Kusawa		**Laberge**					**Quiet**		
	1993	**1999**	**1993**	**1996**	**1999**	**2000**	**2001**	**1994**	**1997**	**1999**
å **PCB**	104.02	129.78	1381.86	1598.31	1625.39	1172.08	1997.14	26.66	48.14	266.23
	(21.49)	(15.55)	(226.21)	(292.05)	(219.22)	(109.01)	(266.81)	(4.24)	(8.52)	(33.07)
Mono-di-/triCB	3.25	2.35	14.47	17.67	17.91	6.43	25.86	0.97	1.97	3.62
	(0.51)	(0.59)	(2.04)	(3.59)	(2.35)	(0.86)	(14.85)	(0.09)	(0.92)	(0.68)
tetraCB	8.27	7.73	73.24	90.11	89.41	57.13	130.88	2.20	3.32	14.54
	(1.47)	(1.24)	(11.59)	(17.01)	(12.26)	(6.86)	(24.27)	(0.55)	(0.87)	(3.03)
pentaCB	18.20	33.94	314.02	384.55	384.47	250.36	305.27	7.13	11.52	69.61
	(4.66)	(4.05)	(51.11)	(70.71)	(50.26)	(23.95)	(48.56)	(1.09)	(1.82)	(8.11)
hexaCB	54.90	63.90	577.55	717.34	697.71	581.01	832.95	10.90	20.07	146.92
	(10.76)	(7.65)	(96.22)	(134.26)	(99.65)	(53.26)	(120.26)	(1.78)	(3.11)	(18.12)
heptaCB	13.98	18.76	320.41	297.98	345.90	218.86	501.56	4.60	8.95	27.94
	(3.27)	(2.15)	(53.03)	(51.17)	(43.27)	(18.55)	(65.37)	(0.76)	(1.96)	(2.93)
octaCB	4.94	3.08	74.00	80.42	81.20	52.13	153.54	0.61	1.83	3.60
	(0.87)	(0.53)	(12.29)	(14.59)	(11.35)	(5.07)	(21.03)	(0.17)	(0.42)	(0.43)
å **DDT**	28.67	56.16	2229.09	2675.37	2849.84	2069.57	2452.63	39.65	89.84	84.01
	(3.30)	(9.38)	(358.20)	(475.56)	(361.70)	(216.53)	(431.04)	(4.14)	(16.06)	(9.59)
p,p'-DDT	2.32	14.24	692.89	877.15	892.61	447.30	249.23	5.62	12.97	17.13
	(0.19)	(2.65)	(125.83)	(163.20)	(121.01)	(64.48)	(45.58)	(0.79)	(3.20)	(1.30)
p,p'-DDE	19.66	27.38	836.44	958.18	1053.90	735.97	1360.42	27.47	63.54	45.68
	(2.81)	(4.31)	(111.12)	(159.53)	(123.28)	(66.57)	(254.12)	(3.73)	(10.79)	(5.42)
p,p'-DDD	2.10	8.94	580.17	659.83	725.64	746.91	755.74	5.56	10.98	10.40
	(0.13)	(2.11)	(104.66)	(116.52)	(94.06)	(76.67)	(145.22)	(0.56)	(1.71)	(1.24)
å **CBz**	5.07	5.28	33.51	40.11	41.58	26.31	33.35	4.68	6.68	8.42
	(1.58)	(0.63)	(4.93)	(7.11)	(5.31)	(3.40)	(2.80)	(0.56)	(0.67)	(0.82)
å **CHB**	279.40	208.30	2710.61	2659.19	3053.46	1281.08	1142.42	43.80	74.40	149.85
	(43.34)	(20.81)	(478.63)	(442.96)	(364.65)	(111.90)	(145.96)	(5.15)	(11.66)	(15.88)

	Kusawa		Laberge					Quiet		
	1993	**1999**	**1993**	**1996**	**1999**	**2000**	**2001**	**1994**	**1997**	**1999**
å **CHL**	58.17	35.61	203.08	270.22	247.76	219.01	280.43	12.83	21.94	39.48
	(7.42)	(5.07)	(34.47)	(52.10)	(36.73)	(24.86)	(30.87)	(1.58)	(3.44)	(4.13)
oxychlor	15.88	5.16	24.80	37.47	33.65	21.29	19.74	2.45	4.10	5.64
	(2.64)	(1.70)	(4.65)	(7.65)	(4.95)	(2.16)	(2.05)	(0.27)	(0.64)	(0.68)
cis-chlordane	1.22	1.79	12.03	10.60	11.35	11.66	12.30	1.42	1.72	2.58
	(0.07)	(0.09)	(2.97)	(2.10)	(1.76)	(1.09)	(3.12)	(0.13)	(0.10)	(0.29)
trans-chlordane	0.22	0.51	4.52	6.25	5.36	4.98	7.16	0.19	0.60	0.62
	(0.03)	(0.15)	(1.19)	(1.30)	(1.05)	(0.69)	(1.26)	(0.03)	(0.19)	(0.07)
trans-nonachlor	24.60	15.77	88.38	118.79	104.15	93.35	105.49	3.81	6.64	14.26
	(2.61)	(1.73)	(15.49)	(22.88)	(16.63)	(11.93)	(14.24)	(0.56)	(1.24)	(1.35)
cis-nonachlor	11.45	7.03	54.83	70.79	69.23	63.53	72.02	2.25	3.98	9.41
	(0.95)	(0.74)	(9.35)	(13.35)	(9.46)	(6.70)	(9.24)	(0.32)	(0.67)	(0.44)
å **HCH**	1.89	5.14	39.71	36.53	43.98	19.23	8.17	11.34	14.61	9.50
	(0.17)	(0.59)	(6.42)	(6.58)	(6.48)	(2.73)	(0.76)	(1.00)	(2.09)	(1.62)
a-HCH	1.27	1.26	27.57	29.96	36.29	15.97	6.75	9.41	12.02	4.28
	(0.11)	(0.09)	(4.53)	(5.33)	(5.13)	(2.27)	(0.62)	(0.77)	(1.73)	(0.59)
β-HCH	<0.01	0.49	0.74	1.10	1.15	0.72	0.12	0.36	0.51	0.69
	(<0.01)	(0.06)	(0.19)	(0.27)	(0.23)	(0.14)	(0.08)	(0.04)	(0.06)	(0.10)
g-HCH	0.62	3.39	5.79	5.47	6.54	2.54	1.29	1.57	2.09	4.54
	(0.19)	(0.48)	(0.97)	(1.03)	(1.21)	(0.33)	(0.19)	(0.21)	(0.32)	(0.93)

ΣDDT = *p,p*'-DDT, *p,p*'-DDE, *p p*'-DDD, *o,p*'-DDT, *o,p*'-DDE and *o,p*'-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = sum of all technical chlordane compounds and related metabolites, including heptachlor epoxide; ΣCBz = 1245TCB, 1234TCB, P5CBz and HCBz; ΣPCB = CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114,131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209

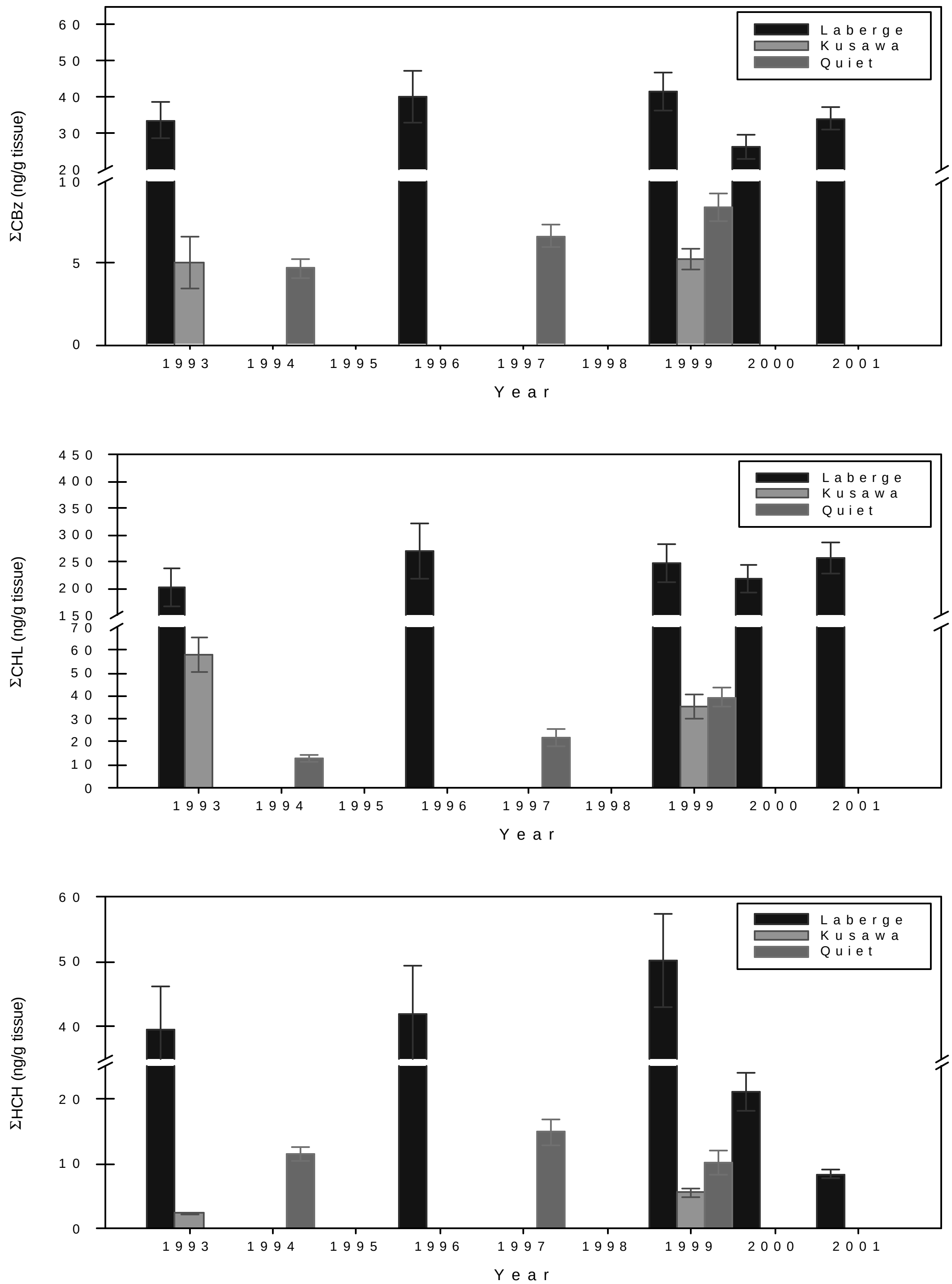


Figure 3.3 Mean concentrations (± SE) of ΣCBz, ΣCHL, ΣHCH, ΣDDT, ΣPCB and Toxaphene (ΣCHB) in burbot liver from Laberge, Kusawa, and Quiet Lake (1993-2001).

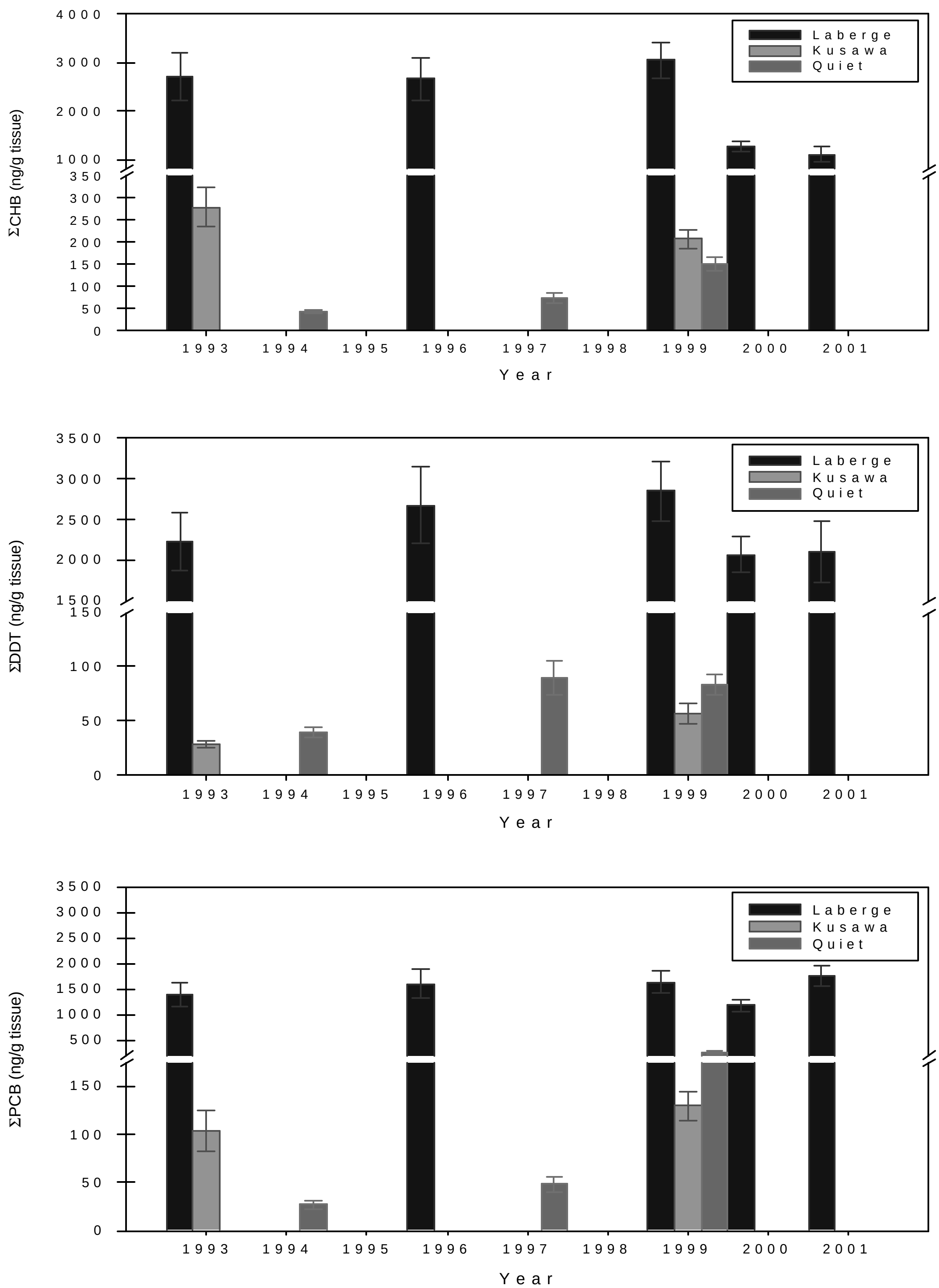


Figure 3.3 (cont'd). Mean concentrations (± SE) of ΣCBz, ΣCHL, ΣHCH, ΣDDT, ΣPCB and Toxaphene (ΣCHB) in burbot liver from Laberge, Kusawa, and Quiet Lake (1993-2001).

Table 3.5 Selected years of GLMR results for burbot and lake trout from Kusawa, Laberge and Quiet lakes.

Lake	**Species**	**Year**	**OC**	***p* (model)**	**r^2**	**variable**	***p* (2 tail)**
Kusawa	LT	1993	ΣCBz	0.07	0.52	% lipid	0.10
						log weight	0.79
			ΣHCH	0.02	0.71	% lipid	0.02
						log weight	0.79
			ΣCHL	0.32	0.27	% lipid	0.32
						log weight	0.85
			ΣDDT	0.03	0.60	% lipid	0.32
						log weight	0.16
			ΣPCB	0.13	0.43	% lipid	0.69
						log weight	0.20
			ΣCHB	0.35	0.25	% lipid	0.28
						log weight	0.96
Kusawa	LT	2001	ΣCBz	<<0.01	0.90	% lipid	<<0.01
						log weight	0.12
			ΣHCH	<<0.01	0.93	% lipid	<<0.01
						log weight	0.02
			ΣCHL	<<0.01	0.98	% lipid	<<0.01
						log weight	0.18
			ΣDDT	<<0.01	0.99	% lipid	<<0.01
						log weight	0.02
			ΣPCB	<<0.01	0.98	% lipid	<<0.01
						log weight	<<0.01
			ΣCHB	<<0.01	0.98	% lipid	<<0.01
						log weight	<<0.01

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Kusawa	BB	1993	ΣCBz	0.25	0.93	% lipid	0.86
						weight	0.23
			ΣHCH	0.78	0.38	% lipid	0.79
						weight	0.60
			ΣCHL	0.46	0.78	% lipid	0.92
						weight	0.41
			ΣDDT	0.36	0.86	% lipid	0.88
						weight	0.38
			ΣPCB	0.21	0.95	% lipid	0.70
						weight	0.24
			ΣCHB	0.29	0.91	% lipid	0.65
						weight	0.35
Kusawa	BB	1999	ΣCBz	0.05	0.70	% lipid	0.01
						weight	0.49
						log age	0.74
			ΣHCH	0.06	0.68	% lipid	0.02
						weight	0.28
						log age	0.74
			ΣCHL	0.11	0.61	% lipid	0.04
						weight	0.50
						log age	0.73
			ΣDDT	0.05	0.71	% lipid	0.01
						weight	0.40
						log age	0.80
			ΣPCB	0.08	0.66	% lipid	0.02
						weight	0.39

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Kusawa	BB	1999				log age	0.87
			ΣCHB	0.11	0.61	% lipid	0.03
						weight	0.87
						log age	0.31
Laberge	LT	1993	ΣCBz	<<0.05	0.81	% lipid	<<0.05
						log weight	0.33
			ΣHCH	<<0.05	0.75	% lipid	<<0.05
						log weight	0.33
			ΣCHL	<<0.05	0.62	% lipid	<<0.05
						log weight	0.46
			ΣDDT	<<0.05	0.42	% lipid	<<0.05
						log weight	0.19
			ΣPCB	0.02	0.33	% lipid	0.01
						log weight	0.44
			ΣCHB	0.03	0.30	% lipid	0.02
						log weight	0.30
Laberge	LT	1996	ΣCBz	<<0.05	0.93	% lipid	<<0.05
						log weight	0.01
			ΣHCH	<<0.05	0.90	% lipid	<<0.05
						log weight	<<0.05
			ΣCHL	<<0.05	0.90	% lipid	<<0.05
						log weight	0.02
			ΣDDT	0.01	0.64	% lipid	<<0.05
						log weight	0.09
			ΣPCB	<<0.05	0.81	% lipid	<<0.05
						log weight	0.02

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Laberge	LT	1996	ΣCHB	<<0.05	0.79	% lipid	<<0.05
						log weight	0.01
Laberge	LT	2001	ΣCBz	0.15	0.25	% lipid	0.06
						log weight	0.67
			ΣHCH	0.01	0.55	% lipid	<<0.05
						log weight	0.91
			ΣCHL	<<0.05	0.58	% lipid	0.44
						log weight	<<0.05
			ΣDDT	<<0.05	0.73	% lipid	0.20
						log weight	<<0.05
			ΣPCB	<<0.05	0.66	% lipid	0.91
						log weight	<<0.05
			ΣCHB	<<0.05	0.62	% lipid	0.41
						log weight	<<0.05
Laberge	BB	1993	ΣCBz	0.43	0.28	% lipid	0.18
						weight	0.50
						log age	0.22
			ΣHCH	0.36	0.32	% lipid	0.15
						weight	0.44
						log age	0.17
			ΣCHL	0.47	0.26	% lipid	0.38
						weight	0.93
						log age	0.38
			ΣDDT	0.47	0.26	% lipid	0.50
						weight	0.97
						log age	0.42

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Laberge	BB	1993	ΣPCB	0.40	0.30	% lipid	0.62
						weight	0.94
						log age	0.39
			ΣCHB	0.49	0.25	% lipid	0.41
						weight	0.99
						log age	0.42
Laberge	BB	2001	ΣCBz	<<0.05	0.50	% lipid	<<0.05
						weight	0.13
						log age	0.46
			ΣHCH	<<0.05	0.58	% lipid	<<0.05
						weight	0.65
						log age	0.56
			ΣCHL	<<0.05	0.54	% lipid	<<0.05
						weight	0.03
						log age	<<0.05
			ΣDDT	<<0.05	0.51	% lipid	0.01
						weight	0.22
						log age	0.01
			ΣPCB	0.01	0.40	% lipid	0.09
						weight	0.06
						log age	0.01
			ΣCHB	0.01	0.42	% lipid	0.01
						weight	0.40
						log age	0.04
Quiet	LT	1992	ΣCBz	0.04	0.97	% lipid	0.05
						log weight	0.02

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Quiet	LT	1992	ΣHCH	0.60	0.40	% lipid	0.36
						log weight	0.42
			ΣCHL	0.14	0.86	% lipid	0.28
						log weight	0.78
			ΣDDT	0.78	0.22	% lipid	0.89
						log weight	0.67
			ΣPCB	0.23	0.77	% lipid	0.92
						log weight	0.36
			ΣCHB	0.26	0.74	% lipid	0.24
						log weight	0.63
Quiet	LT	2001	ΣCBz	<<0.05	0.93	% lipid	<<0.05
						log weight	0.52
			ΣHCH	0.02	0.75	% lipid	0.01
						log weight	0.85
			ΣCHL	<<0.05	0.85	% lipid	<<0.05
						log weight	0.53
			ΣDDT	<<0.05	0.94	% lipid	<<0.05
						log weight	0.22
			ΣPCB	<<0.05	0.94	% lipid	<<0.05
						log weight	0.24
			ΣCHB	<<0.05	0.95	% lipid	<<0.05
						log weight	0.15
Quiet	BB	1994	ΣCBz	0.01	0.99	% lipid	<<0.05
						weight	0.04
			ΣHCH	0.38	0.61	% lipid	0.21
						weight	0.39

Lake	Species	Year	OC	*p* (model)	r^2	variable	*p* (2 tail)
Quiet	BB	1994	ΣCHL	0.19	0.81	% lipid	0.11
						weight	0.64
			ΣDDT	0.72	0.28	% lipid	0.48
						weight	0.60
			ΣPCB	0.52	0.48	% lipid	0.34
						weight	0.37
			ΣCHB	0.82	0.18	% lipid	0.65
						weight	0.93
Quiet	BB	1997	ΣCBz	0.02	0.71	% lipid	<<0.05
						weight	0.16
						log age	0.11
			ΣHCH	0.03	0.66	% lipid	0.18
						weight	0.01
						log age	0.68
			ΣCHL	0.01	0.73	% lipid	<<0.05
						weight	0.92
						log age	0.49
			ΣDDT	0.02	0.70	% lipid	<<0.05
						weight	0.61
						log age	0.42
			ΣPCB	0.03	0.66	% lipid	0.01
						weight	0.85
						log age	0.76
			ΣCHB	0.07	0.56	% lipid	0.02
						weight	0.82
						log age	0.62

burbot condition factors (p=0.8) or in liver lipid content (p=0.33). Overall, Kusawa burbot have not changed in morphology for the period of time studied.

Of all of the variables examined, only tissue lipid (%) was a mildly significant factor affecting the variability in the prediction of OC in 1999 burbot samples (ΣCBz 2 tail p=0.01, ΣHCH 2 tail p=0.01, ΣCHL 2 tail p=0.03, ΣDDT 2 tail p=0.01, ΣPCB 2 tail p=0.02, ΣCHB 2 tail p=0.03; Table 3.5) as determined by GLMR. Age data were not available for 1993 Kusawa samples and therefore log age was excluded from the GLMR model for that year. However, using tissue lipid (%) and weight as sole variables for 1993 resulted in no significant values indicating an influence on OC concentrations although some of the r^2 correlations were high. This is due to the small sample number (n=4) for Kusawa burbot from 1993. Statistically the numbers of fish do not support a significant model although the available data points (OC concentrations, weight and lipid) do correlate.

3.4.2 Lake Laberge

Laberge lake trout have shown a decrease of 69% in ΣCBz (p<<0.05), 88% in ΣHCH (p<<0.05), 84% in ΣCHL (p<<0.05), 75% in ΣPCB (p=0.02), 84% in ΣDDT (p<<0.05) and 42% in ΣCHB (p=0.008) when comparing 2003 concentrations to 1993 levels (Figure 3.2, Table 3.3). All PCB homologue group concentrations decreased in Lake Laberge trout over the ten-year time period. In addition, the relative proportions of ΣPeCB decreased from 24 to 17% of ΣPCB (p<0.05) while the ΣHpCB increased from 22 to 26% of ΣPCB (p=0.02). The relative proportions for the remaining PCB homologue groups did not differ significantly over time. No trends in the *p,p'*-DDT/ΣDDT or α-/γ-HCH isomer ratios or in the proportions of the *trans*- and *cis*-chlordane congeners were observed (Table 3.3).

Lake Laberge trout had small but significant changes in some morphological parameters. There were no significant differences between mean annual ages in Laberge trout among years (p=0.11) averaging 15 years of age in 1993 (±2 SE) to 14 years of age in 2001 (±2 SE; Table 3.2). However, there was a significant change in Laberge trout condition factors across years (p=0.02) with a peak in the 1996 K values (1.4) that was 0.2 units higher than any other year. Both the mean weights and lengths have increased significantly (p=0.002 and p<<0.05) from 1993 to 2003 (Table 3.2). Average weights increased from 1523 g (±191 g SE) in 1993 to 2485 g (±427 g SE) in 2003 (Table 3.2). Average lengths increased from 480 mm (±15 mm SE) in 1993 to 593 mm (±34 mm SE) in 2003 (Table 3.2). There was also a significant 40% decrease in Laberge trout lipid content from 1993 to 2003 (p=0.001,Table 3.2). In general the Laberge trout have become leaner in lipid content and larger (length and weight) over the study period, but there were no changes in condition factor.

GLMR models of Laberge lake trout showed both log weight and lipid content as the major predictors of OC concentrations with a trend towards log weight being a more significant factor than tissue lipid (%) in more recent years (Table 3.5). Log weight was the primary factor for all 2001 OC concentrations except for HCH (the model was insignificant (p=0.15) for the CBz group in 2001). Both tissue lipid (%) and log weight were strong factors in 1996 lake trout samples while lipid was the sole significant factor in 1993 accounting for the majority of variability in OC concentrations. The 2000, 2002 and 2003 samples produced mixed results partly due to smaller sample numbers for those years but the data still support both lipid and log weight as significant predictors of OC concentrations.

Laberge burbot exhibited no significant changes in ΣDDT, ΣHCH, ΣCBz, ΣCHL or ΣPCB across the study period (p=0.37, 0.001, 0.29, 0.62, and 0.28 respectively; Figure 3.3, Table 3.4). There were decreases of 58% in ΣCHB and 80% in ΣHCH levels after 1999 (both p<<0.05). The year 1999 was a notable peak in all OC concentrations for Laberge burbot. There was no change in overall proportions of OC congeners except for an increasing proportion of *p, p'*-DDD (12-15%) compared to *p, p'*-DDE (Table 3.4).

Burbot from Laberge showed some differences across years with respect to fish size and tissue lipid (%) of livers. No significant trends were measured in ages of Laberge burbot, which had a mean of 13 years (±2 SE) in 1993 and a mean of 16 years (±1 SE) in 2001 (Table 3.2). However, burbot did have a highly significant decrease in K (p<<0.05) and liver lipid content (p<<0.05) over time, which is different from what was observed in the Kusawa and Quiet Lake burbot samples. Condition factors decreased significantly from 0.7 to 0.5 (p<<0.05) while lipid contents decreased 45% from 1993 to 2001 (Table 3.2). There was a significant increase in burbot lengths (p<<0.05) and no significant increase in weights (marginal); however, the mean for 1996 samples is a mild outlier and is low compared to 1993, 1999, 2000 or 2001. Overall the fish have become leaner in girth and maintain a lesser fat content compared to earlier years.

Regression analyses for Laberge burbot showed that the GLMR model was not significant for 1993 contaminant data and log age, weight and tissue lipid (%) had very low correlations with OC concentrations from burbot livers sampled that year. Tissue lipid (%) and log age were the most significant factors correlating to OC concentrations in 1996, 1999, 2000 and 2001 (Table 3.5). It was noted for 1996 that weight was also significant predictor of all OC concentrations (2 tail $p<0.01$).

3.4.3 Quiet Lake

Significant decreases in all 6 OC groups were observed in Quiet Lake trout over the time period of this study. There were measured declines of 69% in ΣCBz ($p<<0.05$), 90% in ΣHCH ($p<<0.05$), 79% in chlordanes ($p<<0.05$), 69% in ΣPCB ($p<<0.05$), 85% in ΣDDT ($p<<0.05$) and 69% in ΣCHB ($p<<0.05$) when comparing 2002 OC to 1992 values (Table 3.3). There were no changes in the ratios of DDT, chlordane or in HCH congeners in Quiet Lake trout (Table 3.3).

Quiet Lake trout have shown considerable changes in morphometrics between 1992 and 2002. Lake trout ages from the fish sampled were significantly different across years ($p=0.009$) with a mean of 14 years (±1 SE) in 1994, 20 years (± 2 SE) in 1999 and 11 years (±2 SE) in 2002. This was accompanied by a significant change in K across years ($p<<0.05$) as 1999 K values were significantly higher compared to either of the 2001 or 2002 averages (K=1.2, 0.9 and 1.0 respectively; Table 3.2). There was a strong and consistent decrease in

% lipid across years in Quiet Lake trout (p<<0.05) falling from 5.6% in 1992 to 0.7% in 2002. Generally, the trout have increased then decreased in girth and decreased in lipid content over the years sampled.

The weight-tissue lipid (%) model was not significant for any 1992 OC groups except CBz (p=0.04, r^2 =0.97). GLMR showed tissue lipid (%) as the most significant predictor in all OC groups for 1999-2002 Quiet Lake trout (Table 3.5).

Unlike the lake trout, Quiet Lake burbot have shown an increase of 80% in ΣCBz (p=0.01), 207% in chlordanes (p=0.001), 898% in ΣPCB (p<<0.05), 111% in ΣDDT (p=0.007) and 242% in ΣCHB (p=0.001) when comparing 1999 to 1994 OC values. It should be noted that there was a low number of samples for this analysis (n=3 for 1999 Quiet Lake burbot; Table 3.2). ΣHCH were not significantly different across years (p=0.17) but there was a slight peak in 1997. As was observed in lake trout, there was no change in the ratios of DDT, CHL or HCH isomers in Quiet Lake burbot.

Analyses of Quiet Lake burbot showed few changes in the morphology measurements. There were no significant temporal trends in ages (p=0.06), liver lipid content (p=0.51) or fish condition factors (p=0.06). However there was a peak year in 1997 in which liver lipid contents were over 9% higher than in 1994 or 1999. No significant change in K was observed from 1994 to 1997; however, K increased marginally significantly (p=0.05) from 1997 (0.6) to 1999 (0.7).

The GLMR (log age, tissue lipid (%), weight model) run for Quiet Lake burbot could not be included in the model due to the low number of samples from 1999. As such, only 1994 and 1997 years could be analysed for OC concentration and morphometric comparison. Age values were absent for 1994 and also had to be left out of the regression models for that year. The model was insignificant for all 1994 OC except for CBz where both lipid content and weight were significant predictors of contaminant concentrations. The regression model accounted for 56%-73% of the within lake variability in OC concentrations in 1997. The GLMR model was significant for all OC groups in 1997, except CHB, with tissue lipid (%) being the most significant individual factor (Table 3.5).

3.5 Discussion

Organochlorine concentrations varied over time and location as well as between fish species in the study of Laberge, Quiet and Kusawa lakes. Lake trout were generally decreasing in OC concentrations over time, while burbot have shown little or no change across the study period. Lake trout from Laberge and Quiet Lakes had larger declines in OC levels compared to those in Kusawa Lake. The reasons for these differences are likely due to variations in the individual lake ecosystems such as species compositions (fish and invertebrates), population sizes, primary producer production and possibly abiotic factors (turbidity, nutrients, etc.).

The environmental monitoring of the three Yukon lakes attempted to assess the interactions between some fish morphometrics (length, weight, age) and biochemical characteristics (lipid content) with the contaminant data, to provide a hypothesis for changes in OC levels in regards to the lake biotic structures.

Previous studies have shown that lipid content is a major factor influencing the levels of OC in fish (Larsson et al. 1993; Ruus et al. 2002). The present study also observed that lipid was a significant predictor of OC concentrations within lakes for many of the years sampled. From 1996 onward, OC levels for both Lake Laberge and Quiet Lake trout decreased significantly and was accompanied by concurrent decreases in lipid content (Table 3.3). No significant change in lipid content was measured in the Kusawa fish and thus only marginal decreases in OC levels were observed. This presents Kusawa Lake as the best candidate for monitoring changes in atmospheric OC concentrations in conjunction with temporal trends in contaminants in fish. Laberge fish continued to maintain a significantly higher level of OC concentrations compared to Kusawa or Quiet Lake, which was attributed to both the higher lipid contents and higher trophic status for Laberge lake trout in earlier research (Kidd et al. 1998).

Although lipid content is one factor affecting OC levels in fish, age, growth and size are other parameters that need to be assessed (Kidd 1996; Larsson et al. 1992; Ruus et al. 2002) which are in turn affected by changes within the fish food web (Larsson et al. 1992; Ruus et al. 2002). Kilgour et al. (2005) and Munkittrick and Dixon (1989) proposed a method of measuring such morphometric and fish population characteristics as an assessment of environmental health. These parameters could be also be used to ascertain occurrences such as exploitation, recruitment failure, food limitation and niche shifts (adaptation of new behaviours by two or more populations in a community to reduce interspecies competition) within an ecosystem. Kusawa and Quiet Lake trout had significant decreases in mean ages over the past decade (Table 3.2) along with fluctuations in condition factors. Decreasing ages along with increased condition factors can be representative of an exploitation response (Kilgour et al. 2005). Although the mean condition factors of Laberge and Quiet Lake fish have not indicated consistent decreases over the period, the K values have fluctuated significantly and fish have become larger (based on weight and length data). Length and weight data have clearly shown an increase in Quiet trout weights while Laberge fish are increasing in both length and weight indicating the lake trout are getting larger overall regardless of the K value interpretation. A response from exploitation may be true especially for Lake Laberge and Quiet Lake trout based on their historical presence of commercial fisheries and a recent closure of such practices (Thompson 1996; Foos 1998). Each lake maintained a commercial fishery until 1989 for Quiet, 1991 for Laberge and 1968 for Kusawa Lake. Laberge trout ages increased and decreased from samples between 1993 and 1996 and condition factors also peaked during this same time frame (Table 3.2). Laberge trout also maintain a consistently higher condition factor in comparison to trout from Kusawa and Quiet Lake. Quiet Lake trout are seen to have a lower average lipid content and condition factors with a higher variability, but increasing in weight and length ranges, which may be indicative of a food limitation or niche shift (Kilgour et al. 2005) perhaps in competition with resident burbot. Burbot may be infringing on the lake trout habitat following the closure of the Quiet Lake commercial fishery in 1989 although they still constitute a minority species of the fish population (Foos 1998). An expected increase in the number of young-of-the-year lake trout as a food source for burbot may account for the recent significant increase in burbot girth. The decreasing average ages for Quiet

Lake trout do not conform to the idea of food limitation or niche shifts, but unfortunately, the relatively low number of age samples (<9) for the years collected may be misleading as a representative population sample. This prevents an exact conclusion on age effects with OC trends and population responses to stressors. However, regression results did show that age was inconsistently a minor but statistically significant factor accounting for variance in burbot OC concentrations. This coincides with the research by Kidd (1996), which described log age as being a factor for burbot ΣCBz, ΣCHL, ΣCHB concentrations although not a significant factor for any OC contaminant group in lake trout.

Since growth rates could not be calculated with the limited number of fish samples collected for these three lakes, GLMR analyses were run with weights and lengths. Log weight was a more significant factor as a predictor of OC concentrations surpassing lipid content as the most significant source of variation in ΣCHL, ΣDDT, ΣPCB and ΣCHB for Laberge trout from 2001 onward. This is an indication that a change in fish growth may be occurring in this lake affecting the contaminant levels in lake trout. The weights and lengths for burbot and lake trout from the other lakes in this study were considered insignificant factors in their effects on OC concentrations in tissues although lake trout from Quiet Lake are leaner and have increased in average body masses from early to more recent years. Kilgour et al. (2005) and Munkittrick and Dixon (1989) described a model of generalized response patterns of fish populations in which recruitment may result in increased energy usage and decreased energy storage by adults. This pattern generally follows the results seen in Laberge trout which have significantly less lipid content from previous years and a higher average weight and length suggesting increased growth rates (age averages remained unchanged).

It is notable that differences in most of the condition factors for burbot from all three lakes were statistically insignificant while K values for lake trout dropped in two out of the three lakes. Along with a slight increase in ages, these two parameters could suggest multiple stressors are having an effect on the Laberge burbot population, which may be associated with food supply problems or habitat decline. The food supply issue is a strong possibility based on recent population studies that reported a 2-3-fold increase in trout populations since 1992 (Foos 2001) creating competition for resources.

The Quiet Lake burbot are superficially increasing in size (but decreasing in conditions factors) as previously mentioned, and decreasing in lipid content suggesting a similar pattern as Laberge trout. However, the low number of samples (n<6) and general lack of food web data negates a reliable statistical assessment on the biotic structure of Quiet Lake at this time.

Kusawa fish have shown very few changes in comparison to Quiet or Laberge Lakes and these primarily insignificant trends are also reflected in the small changes in fish OC concentrations. Since some fundamental factors such as fecundity were not measured following the initial fish captures in 2000, and no other population characteristics are available, further conclusions on Quiet and Kusawa Lake fish population characteristics and OC or ecosystem changes cannot be drawn at this time.

The holistic environmental assessment models described by Kilgour et al. (2005) and Munkittrick and Dixon (1989) were designed for use with point source or non-point source pollution and sentinel fish species monitoring. The model recommends that "differences of 25% for liver size, gonad size, weight at age, and 10% difference for condition factor may be appropriate" as 'warning -level' criteria for site management. The weights and condition factors for Laberge and Quiet Lake trout fit these warning level criteria, although no point source pollution is suspected to have effected such a change. It is still indicative of a major biological shift affecting lake trout most likely due to natural environmental fluctuations (e.g. climate variation, water temperatures, seasonal ice, primary productivity growth). Based on its historical use as a commercial fisheries lake (Thompson 1996) and a recent increase in some fish populations (Foos 2001), the hypothesis of growth dilution of contaminants in Lake Laberge trout is being investigated along with correlations in trophic level changes using nitrogen and carbon stable isotope analyses (Chapter 4).

Shifts in lake production due to climate variation over the past 12 years must be considered in assessing the changes in OC concentrations and fish morphology. Annual climate trends may contribute to the differences in plankton productivity increasing or decreasing the food available for young, growing fish. Changes in lake productivity "follows closely the annual cycle of incident solar radiation in temperate lakes" (Wetzel 2001)and incident solar radiation can be partly measured by degree-days. The higher number of sunny days resulting in warmer weather creates a longer growing season for smaller algae with faster turnover rates (Wetzel 2001). Annual variations across regional lake ecosystems may provide a significantly longer growing season and hence an increase in lake primary productivity resulting in a shift in plankton community composition (Wetzel 2001). Climate data from Environment Canada (2002) from both Carmacks and Whitehorse (140 km north and 40km south of Lake Laberge respectively) has shown a general trend of decreasing annual rainfall (275mm in 1990 to 109mm in 1998; Chapter 5) and higher than historical average degree-days (above 5 ^{o}C) for the years 1990-1997 (1066 vs. 895 historical; Chapter 5) suggesting that the lakes were exposed to a warmer and drier growing season for primary producers. Larsson et al. (1992) demonstrated that as lake productivity increased, pollutants in fish decreased suggesting that plankton biomass is related to OC concentrations. Lakes with a higher plankton biomass also showed lower OC concentrations in the plankton demonstrating a negative correlation of OC with plankton proliferation (Taylor et al. 1991) while Stewart et al. (2003) reported changes in OC in fish following a flood event were most likely related to shifts within the plankton community and not linked to the transport of new compounds into the lake. Changes in the plankton community of Lake Laberge are reviewed in Chapter 4. The years of potentially higher plankton production from warmer and more dry climate conditions in the southern Yukon during the 1990s may have contributed to faster fish growth resulting in both larger average body masses for Laberge and Quiet Lake trout and modifying OC concentrations in fish. Although the pattern in annual average rainfall and degree days is quite notable between the Carmacks and Whitehorse climate stations (Environment Canada 2002), there is an expected loss in accuracy of predicting climate effects on each of the three lakes without exact weather data for each location.

Although a warmer climate may modify effects on biotic growth and OC bioaccumulation, the overall atmospheric concentrations of some organochlorines are declining (Hung et al. 2002) and will eventually impact on concentrations found in lake biota. Changes in atmospheric $\alpha-$ and $\beta-$HCH levels have occurred due to a decrease in worldwide usage of technical HCH (60-80% $\alpha-/\beta-$HCH, 10-15% $\gamma-$HCH; (Li et al. 1998; Hung et al. 2002). The, $\alpha-/\gamma-$ HCH ratios have also declined in trout from Quiet Lake, as well as in Kusawa and Quiet Lake burbot although this has not been directly linked to the aforementioned changes in atmospheric concentrations. Chlordane and DDT levels in Arctic air have declined and the profiles are consistent with "old" or "recycled" rather than new sources as no significant changes in the *p,p'*-DDE/ΣDDT ratio or in the proportions of *trans*- vs. *cis*-chlordane congeners were evident in fish from the three study lakes (Bidleman et al. 2002; Hung et al. 2002). Numerous biotic factors (previously discussed) could mask any changes from the abiotic level that affect contaminant concentrations in lake biota (Bignert et al. 1993; Larsson et al. 1998; Stewart et al. 2003).

Rawn et al. (2001) noted potential differences in OC concentrations among small, higher elevation lakes and larger, lower elevation lakes in the Yukon drainage based on a survey of six Yukon lake sediment cores. Higher elevation lakes may be influenced to a greater extent by atmospheric OC sources while the lower elevation lakes are more affected by sources from runoff and river inflows. However, the average elevation of the three lakes is within 110 m so the significance of such a relatively small difference is questionable. Unfortunately recent sediment cores were not available for OC analysis for these lakes, but it is more likely that the biotic influence over the OC contaminants in fish was more significant than the abiotic changes affecting Laberge, Kusawa and Quiet lakes over the study period. This is hypothesized as there are no common abiotic influences to explain the variation in contaminant concentation changes in fish across all three lakes.

3.6 Summary and Conclusions

The study has shown that contaminant levels in aquatic biota are changing temporally and continue to vary geographically among Yukon lakes, which in this study and previously (Kidd et al. 1993) has been shown to be due to differences in the aquatic food web dynamics. More recent temporal OC decreases have occurred in lake trout from all three lakes with the highest declines observed in Lake Laberge and Quiet Lake fish. OC levels were correlated with lake trout body size and lipids in Laberge and Quiet Lake. In contrast, no declines in lipids were observed for Kusawa lake trout and concurrently, only marginal declines in OC were measured. Inconsistent OC decreases and increases in burbot were evident in all three lakes with most declines in the Laberge samples. These OC changes are likely related to a combination of variables including closures of commercial fisheries on Laberge and Quiet Lake (in 1991 and 1989 respectively), and/or changes in lake production as affected by warmer and drier than historical average regional climate in the 1990s. A change in primary production could have direct and indirect effects on lake trout and burbot contaminant burdens through processes such as growth dilution as these species are primary predators of planktivorous forage fish. Changes in fish populations, post-fishery exploitation, may also create shifts in niche habitats (e.g. feeding or foraging behaviours) as levels of interspecies competition increases, ultimately modifying the structure of the food web. Shifts in the food web structure of these lakes can be determined with future analysis of stable isotopes and trophic levels (Chapter 4). Lake Laberge continues to maintain a higher level of OC in comparison to Kusawa or Quiet Lake due to its longer food chain.

The OC levels in biota from Yukon lakes vary primarily because of food chain lengths or food web dynamics, which outweigh atmospheric sources as a potential cause for contaminant declines. This is based on the observation of the lack of patterns or correlation of OC changes across regional lakes. Declines in atmospheric OC concentrations, as a rationale for OC changes, can be reviewed from historical abiotic inputs using sediment core analyses and is a parameter that must be reviewed in future research (Chapter 5).

4. BIOTIC INTERACTIONS IN TEMPORAL TRENDS (1992-2003) OF ORGANOCHLORINE CONTAMINANTS IN LAKE LABERGE, YUKON TERRITORY

4.1 Abstract

Recent declines in six organochlorine (OC) contaminant groups, chlordane (CHL), DDT, hexachlorocyclohexane (HCH), toxaphene (CHB), PCB and chlorinated benzenes (CBz) were measured in a sub-Arctic lake (Lake Laberge, YT) following the closure of a commercial fishery. This study examined morphological (length, weight, age), biochemical (lipid content, $\delta^{13}C$, $\delta^{15}N$), population and OC contaminant data for fish (nine species) and invertebrates (zooplankton, snails, clams) across several temporal points between 1993 and 2003 to elucidate the primary causes for these OC declines. It was determined that growth dilution and declines in prey OC were the major factors influencing the decrease of OC in lake trout, round whitefish and possibly zooplankton. A broad decline in tissue lipid contents for most fish species has also contributed to the decline of contaminants although no such change was evident for zooplankton. It is suspected that increases in fish populations or climate changes over the 1990s may have contributed towards an increase in lake primary production as well as a shift in plankton community composition. An increase in populations of seven fish species in Lake Laberge from 1991 to 1998 was

observed by catch per unit effort (CPUE) data. It was noted that CPUE declined for round and lake whitefish during this period but increased for all other species. Concurrently, the zooplankton community shifted from an abundance of *Cyclops scutifer* in 1993 to dominance by *Diaptomus pribilofensis* in 2001. Biomagnification factors (BMF) have increased for piscivorous fish yet remained stable for forage fish species over time indicating within-species differences in OC assimilation or elimination. The overall food web magnification factors (FWMF) have increased for all 6 OC groups while the intercepts of these regression relationships were lower suggesting there is a less contaminated food base for the Lake Laberge food chain. Based on $\delta^{15}N$ and $\delta^{13}C$ values, food web structure has not changed over time with the exception of burbot and possibly northern pike. A half trophic level shift upward was evident for northern pike, while burbot showed a slight decline. Lake whitefish exhibited a notable change in their food source indicating a shift towards a more benthic diet. Considering the generally slow recovery of exploited lake trout, fluctuations in population dynamics for several species and unique species ecology, the changes in OC concentrations in the Lake Laberge ecosystem may continue for several years to come. Sentinel species (herein defined as species of consistent annual observation and collection) such as lake trout, burbot, whitefish, cisco and plankton should continue to be monitored for future temporal correlations with contaminants or climate changes.

4.2 Introduction

It has now been well established that a wide range of chlorinated pesticides (e.g. toxaphene (CHB), chlordanes (CHL), hexachlorocyclohexanes (HCH)) and industrial chemicals such as polychlorinated biphenyls (PCB) undergo long-range atmospheric transport to the sub- and high Arctic regions where they then bioaccumulate and biomagnify up aquatic food chains (Bidleman et al. 1989; Welch et al. 1991; Lockhart et al. 1992; Barrie et al. 1992; Muir et al. 1992; Suedel et al. 1994; Van Dijk and Guicherit 1999; CACAR 2003a; CACAR 2003c). Lake Laberge in the Yukon Territory was studied intensively in the early 1990s because of abnormally high levels of OC contaminants in these fish relative to surrounding lakes. It was found that Lake Laberge had a longer food chain (Kidd et al. 1998) and, as a result, its biotic constituents have higher-than-average contaminant concentrations when compared to other regional lakes (Kidd et al. 1995b). The long food chain was attributed to the effects of a commercial fishery that existed on the lake for over 100 years, and was closed in 1991-92 because of the reported high levels of contaminants. Health Canada placed a health advisory on the consumption of fish from Lake Laberge that remains to this day (Figure 2.1).

Since the first study of contaminants in Lake Laberge in 1991 (Connor and Sparling 1996), concentrations of contaminants have dropped dramatically in fish (Chapter 3). The closure of a commercial fishery in 1991 is hypothesized as having an indirect effect on this phenomenon. Lake Laberge was commercially exploited since the summer of 1898 primarily for lake trout and whitefish although incidental catches of other species such as Arctic grayling and burbot were also kept for sale or consumption (Seigel and McEwen 1984; Thompson 1996). Increasing fish populations have been observed for seven out of ten of the known species in Lake Laberge from surveys in 1991 and 1999 (Foos 2001). Laberge lake trout in particular appear to have increased growth rates based on previous population surveys (Thompson 1996) and more recently measured temporal differences in body size (Chapter 3). An increased rate of growth, and recruitment, in fish following exploitation is an often-observed response (Hewson 1955; Healey 1975; Healey 1978; Mills et al. 2000) but the effect of cessation of a commercial fishery on a whole food web contaminant levels from a sub-arctic lake has rarely if ever been studied. Since the decline of contaminant levels in Lake Laberge fish was discovered, several hypotheses have been put forth to explain the phenomenon (Chapter 3). The hypotheses include biomass/growth dilution, shifts in trophic levels of predator and prey, or events unrelated to the cessation of the fishery, such as declines in atmospheric contaminant concentrations over the same span of time.

Several key morphological and biochemical parameters can be used to examine the first two of these hypotheses. Body size characteristics (length, weights) have been linked to contaminant concentrations in fish (Larsson et al. 1993; Kidd 1996, Chapter 3) and it has been previously demonstrated that growth rates provide a major influence (inverse relationship) on contaminant levels in biota (Thomann 1989; Larsson et al. 1991; Sijm et al. 1992). Growth rates influence contaminant loads in organisms through biomass or growth dilution (Sijm et al. 1992). As a species increases its rate or size of prey consumption, they increase their growth rate (Matuszek et al. 1990; Pazzia et al. 2002). This higher rate of growth results in a larger biomass as well as potential changes in bioenergetics of assimilation, excretion or food conversion efficiencies (Persson and Greenberg 1991; Pazzia et al. 2002). The body size/metabolism of the organism may increase in relation to contaminant intake or contaminant storage in lipids thus ‘diluting’ the contaminants within the body (Thomann 1989; Clark and Mackay 1991; Sijm et al. 1992; Hebert et al. 1997). Variability in tissue lipid content during a state of growth fluctuation may also cause changes in contaminant concentrations in organisms and there is significant evidence supporting the link between OC biomagnification and lipids (Thomann 1989; Larsson et al. 1991; Larsson et al. 1993).

Although fishing exploitation may increase growth rates, it also has the capacity to restructure a food web by eliminating predators or prey and changing the balance of the ecosystem (Mills et al. 2000). Levels of contaminants are significantly related to the trophic position of aquatic animals within a food web (Kidd et al. 1995a; Kidd et al. 1998; Fisk et al. 2001). The higher the trophic level of the species, the higher its contaminant levels (of biomagnifying compounds) that are measured (Rasmussen et al. 1990; Kidd et al. 1995a; Zaranko et al. 1997). A change in prey availability or predator populations within an ecosystem may result in changes in trophic positioning and subsequent OC accumulation. Since fish populations have changed over time in Lake Laberge (Foos 2001), it is possible that the changes within the trophic level structure are due to variations in prey and predator balances. Such a hypothesis may be evaluated using carbon and nitrogen stable isotopes because they are often used as indicators of changes in food sources or trophic level shifting (e.g. Vander Zanden and Rasmussen 1996). Trophic levels are measured using relative stable nitrogen isotope abundances ($\delta^{15}N$) which are strong predictors of lipophilic OC concentrations up a food chain (Vander Zanden and Rasmussen 1996; Kidd et al. 1998). Carbon isotope ratios ($\delta^{13}C$) can be used to determine shifts in sources of food (e.g. benthic vs. pelagic) over time and hence the origin of contaminants from consumed material (Das et al. 2000; Campbell et al. 2000). Both of these measures can provide insight into the dietary habits of any organism in the food web and provide an explanation for changes in OC levels.

Although research studies on OC concentrations often analyse a few key species, far fewer studies have monitored entire lake ecosystems for changes in abiotic OC inputs, productivity, food web structure, diet changes and fish populations. There are no other known studies on the effects of the closure of a commercial fishery on the contaminant loads in the food web of a sub-Arctic lake. This research focuses on temporal changes in various organochlorine contaminant concentrations in a sub-arctic lake food web and the relation of biotic parameters with these pollutants over the span of a decade. The main objective of this research is to determine the most probable causes of the recent declines observed in OC levels in fish through a temporal analysis of morphological (length, weight, age), biochemical (lipid content, $\delta^{13}C$, $\delta^{15}N$), population and OC contaminant data for fish (nine species) and invertebrates (zooplankton, snails, clams) in the food web in Lake Laberge.

4.3 Materials and Methods

4.3.1 Sample Collections

Lake Laberge is approximately 40km north of the city of Whitehorse in the Yukon Territory (61^{o}11'N, 135 ^{o} 12'W). Limnological data for the lake were collected several times over the past 2 decades as reported in Chapter 3 (Shortreed and Stockner 1983; Jack et al. 1983; Kirkland and Gray 1986; Kidd 1996; Thompson 1996; Foos 1998).

Nine fish species were collected from Lake Laberge including lake trout (*Salvelinus namaycush*), burbot (*Lota lota*), inconnu (*Stenodus leucichthys*), northern pike (*Esox lucius*), lake whitefish (*Coregonus clupeaformis*), least cisco (*Coregonus sardinella*), arctic grayling (*Thymallus arcticus*), longnose sucker (*Catostomus catostomus*), round whitefish (*Prosopium cylindraceum*) and broad whitefish (*Coregonus nasus*) over multiple years (Table 4.1). Lake trout collections occurred during the summers of 1992-1993, 1996, 2000-2003. Burbot were collected in the spring and summers of 1992-1993, 1996, 1999, 2000 and 2001. The primary collection years for all other fish were 1992-1993, 2000 and 2001 with a few samples from 1998. All 1992-1993 data were compiled as a single year and are subsequently referred to only as "1993" data. Only a single Arctic grayling was captured in 2001 with no samples available from previous years, so they were excluded from any analyses beyond reporting that the species was found in the lake. The presence of only small numbers of grayling was also reported by (Foos 2001). Half of the 1993 lake trout and burbot data were obtained from Kidd (1996).

Burbot were caught during spring months (March, April) using long line angling while all other species were captured during summer months (July, August) using small mesh index nets as described in (Thompson 1996) and (Foos 1998). This included two gangs of nets each with three panels 23 m long, 2.4 m depth using mesh sizes of 3.8 cm, 6.4 cm and 7.6 cm. Net set times, and capture locations were kept as similar as possible over time. Yukon Territorial Government fishing quotas limited capture of lake trout to 15 or less for some years. Morphological measurements and tissue sampling were done at the Indian and Northern Affairs lab in Whitehorse. Samples were wrapped in baked and solvent washed foil and shipped frozen to Fisheries and Oceans Canada's Freshwater Institute in Winnipeg for analysis. Sections of dorsal muscle and burbot livers were frozen and kept at –20 ^{o}C until organochlorine analysis. Otoliths (or cleithra for pike) were removed for age analysis. Morphological measurements included sex, fork length and mass from a weigh scale (Table 4.1). Condition factors (K) were calculated using [mass (g) x 10^5 / (length (mm))^3]. In addition, the Yukon Territorial Government (Renewable Resources) conducted population assessments of the fish stocks in 1991 and 1999 for Lake Laberge (Thompson 1996; Foos 1998; Foos 2001). Some of these data will be presented here. Changes in catch per unit effort (CPUE) were calculated as CPUE factors according to the following equation:

$$\textbf{CPUE factor = CPUE (\# in gillnet/hour)}_{1999} \textbf{ / CPUE (\# in gillnet/hour)}_{1991}$$

Table 4.1 Morphological data (means ± SE) for fish species collected in Lake Laberge from 1993 to 2003.

	Year	N*	Age	Length	Weight	K	% lipid
Laketrout	**1993**	24 (8)	15 ± 2	479.7 ± 15.7	1522.7 ± 191.1	1.25±0.05	7.9 ± 0.9
	1996	13 (9)	22 ± 5	491.2 ± 27.3	1867.3 ± 415.0	1.36±0.04	9.6 ± 1.4
	2000	6	12 ± 2	590.0 ± 44.0	2753.3 ± 812.1	1.20±0.05	3.7 ± 0.8
	2001	17	14 ± 2	646.2 ± 25.0	3322.9 ± 387.9	1.16±0.03	4.9 ± 0.5
	2002	5 (2)	12 ± 4	616.0 ± 17.1	2874.0 ± 336.7	1.21±0.04	4.2 ± 0.9
	2003	8	-	592.5 ± 34.7	2485.0 ± 427.1	1.13±0.04	4.7 ± 0.8
Burbot	**1993**	29 (27)	13 ± 1	568.6 ± 21.0	1395.0 ± 129.1	0.70±0.02	41.8 ± 2.1
	1996	15	-	485.3 ± 26.6	921.7 ± 145.0	0.71±0.02	36.8 ± 2.3
	1999	11	17 ± 1	647.7 ± 21.9	1518.2 ± 153.0	0.55±0.03	53.8 ± 2.0
	2000	18	15 ± 1	626.1 ± 24.4	1399.4 ± 135.1	0.55±0.02	27.9 ± 2.2
	2001	30	17 ± 1	634.8 ± 18.6	1383.0 ± 111.1	0.53±0.01	23.5 ± 2.5
Inconnu	**2000**	2	7 ± 0	540.0 ± 20.0	1675.0 ± 175.0	1.06±0.01	1.5 ± 0.2
	2001	6 (5)	13 ± 1	680.8 ± 35.6	2258.3 ± 657.2	0.77±0.21	7.7 ± 1.8
Northern pike	**1993**	8	7 ± 1	633.8 ± 27.4	1825.0 ± 257.2	0.69±0.02	0.4 ± 0.0
	1998	1	10	1005.0	7500.0	0.74	0.9
	2000	5 (4)	7 ± 1	686.0 ± 74.5	2428.0 ± 843.2	0.67±0.02	0.3 ± 0.0
	2001	6	-	777.5 ± 25.3	3170.0 ± 303.1	0.67±0.04	0.3 ± 0.1

	Year	N*	Age	Length	Weight	K	% lipid
Lake whitefish	**1993**	30 (26)	10 ± 1	318.0 ± 14.9	432.2 ± 45.8	1.11±0.03	2.8 ± 0.3
	1998	5	9 ± 2	335.0 ± 16.4	428.0 ± 78.9	1.09±0.04	1.4 ± 0.1
	2000	14 (13)	9 ± 1	319.3 ± 7.6	330.7 ± 16.0	1.03±0.06	0.5 ± 0.1
	2001	14	-	331.8 ± 10.1	380.7 ± 43.7	0.99±0.03	0.6 ± 0.1
Cisco	**1993**	7 (3)	4 ± 0	172.4 ± 18.8	58.0 ± 15.4	0.90±0.05	5.0 ± 1.1
	2000	22 (20)	4 ± 1	210.9 ± 5.4	86.4 ± 5.6	0.89±0.02	0.7 ± 0.1
	2001	2	5 ± 2	232.5 ± 17.5	85.0 ± 15.0	0.67±0.03	0.9 ± 0.5
Longnose sucker	**1993**	12 (3)	24 ± 4	432.9 ± 6.0	852.1 ± 32.2	1.05±0.02	1.0 ± 0.2
	2000	14	16 ± 1	470.7 ± 8.0	958.6 ± 40.2	0.92±0.03	0.7 ± 0.2
	2001	6	14 ± 4	323.3 ± 57.4	428.3 ± 162.2	0.87±0.08	0.6 ± 0.1
Round whitefish	**1993**	7 (1)	15	221.7 ± 43.4	167.6 ± 106.8	0.82±0.03	2.1 ± 0.2
	2000	5 (4)	9 ± 1	314.0 ± 20.1	290.0 ± 58.1	0.92±0.04	0.4 ± 0.1
	2001	38 (20)	5 ± 1	268.2 ± 9.7	170 ± 20.7	0.80±0.02	0.6 ± 0.0
Broad whitefish	**2000**	8	11 ± 1	440.6 ± 13.6	982.5 ± 79.6	1.15±0.07	1.5 ± 0.6

* Numbers of samples in brackets applies to mean ages

Invertebrates were collected with benthic dredges and shoreline sampling during the months of June and July of 2000 and 2001. The taxa were sorted, identified and then samples were frozen for stable isotope analysis. Zooplankton samples were collected in late July and early August in 1993, 2000, 2001, by horizontal tows using a 250 μm mesh, 40cm diameter net. Vertical tows (15m and 30m) were also taken in 1993 and 2002 using the same net and at the same locations. The 1993 collection involved 6 hauls at 15m in 2 locations, which were combined into one pooled sample (K.Kidd, unpublished data) while the 2002 data involved 6 hauls at 15m (2 samples) and one haul at 30m (not included in count totals). All zooplankton samples were preserved in buffered formalin for identification and enumeration (A. Salki, DFO, Winnipeg and North South Consultants, Winnipeg).

4.3.2 Organochlorine Analysis

Methods for organochlorine extractions followed the same procedures reported in Chapter 3 and Kidd et al. (1998). In brief, fish muscle was homogenized, mixed with anhydrous sodium sulphate and spiked with PCB30-OCN as an internal standard. The material was processed in a Dionex ASE300 with dichloromethane (DCM) and hexane (mixed 50:50). The extract was reduced, filtered and measured for percentage of lipids. Aluminum dishes were weighed and 1mL of the extract was dried in the dish to calculate tissue lipid. The remaining extract was processed through manual gel permeation chromatography (GPC) followed by fractionation on a 1.2%-deactivated Florisil column. Recoveries of the internal standard PCB30 and OCN were 64% ± 25% and 74% ± 25% for all fish samples combined (n=229), respectively.

Burbot livers and invertebrates were extracted using the Ball Mill method (see Chapter 3), spiked with the internal standard (PCB30-OCN), diluted and sub-sampled to determine tissue lipid (%). A portion of the remaining sample was fractionated on a 1.2%-deactivated Florisil column.

All extracts were injected onto a Varian Star 3400 GC equipped with an electron capture detector (Ni^{63} electron source) and an 8100 auto-sampler. The column used was a J&W Scientific (Agilent Tech) DB-5, 60m, 250 μm ID with trimethylpentane as the solvent, Hydrogen Ultra High Purity (UHP) carrier gas (at ~2 mL/min), nitrogen UHP make-up gas (at ~50mL/min). Injection and temperature programs conditions were as follows:

Injector temperature 80°C, detector temperature 300°C, initial temperature 100°C (hold time 2min), 1st temperature program increasing to 150°C at 15°C/min, 2nd temperature program increasing to 265°C at 3°C/min.

Aldrin was used as a volume corrector prior to GC analysis.

4.3.3 Biomagnification Factors

Biomagnification factors (BMF) were calculated for the 1993 and combined 2000-2001 data sets. The calculation used zooplankton from each time period as a common divisor for all species to normalize the differences in the base OC zooplankton concentrations across the two temporal points. BMF were calculated using wet weight concentrations (see Section 4.3.5).

e.g.

$$\mathbf{BMF_{year}\ (fish) = [OC\ in\ fish]_{year} / [OC\ in\ zooplankton]_{year}}$$

4.3.4 Stable Isotope Analyses and Trophic Levels

One mg samples of oven dried and powdered tissue (skinless muscle for fish, whole plankton and soft body parts of other aquatic invertebrates) were sent for stable isotopes analysis at the University of Waterloo, Earth Sciences Department. Samples were run for nitrogen and carbon on an Isochrom Continuous Flow Stable Isotope Mass Spectrometer (Micromass) coupled to a Carlo Erba Elemental Analyzer. Results were generally corrected to laboratory nitrogen standards IAEA-N1 and IAEA-N2 (both Ammonium Sulphate) and carbon standards IAEA-CH6 (sugar), EIL-72 (cellulose) and EIL-32 (graphite). EIL-70b is a lipid extracted/ball-milled fish material, and along with several NIST organic materials, is often used as a monitoring standard. The error for clean ball-milled standard material is ± 0.2‰ for carbon and ± 0.3‰ for nitrogen. Standards were placed throughout each run at a range of weights to allow for an additional linearity correction, when necessary, due to machine fluctuations or to samples of varying signal peak areas. Nitrogen and carbon compositions are calculated based on Carlo Erba Elemental Standards B2005, B2035 and B2036 with an error of ± 1%.

All samples were analysed twice for reproducibility. A material of known isotopic composition (Pharmamedium) was run with every 10 samples to blind test the accuracy of the lab results.

Nitrogen and carbon isotopes ratios are reported in ‘delta (δ)’ notation in parts per thousand, or per mil (‰) according to the following equation:

$$\delta^{15}N \text{ or } \delta^{13}C = ((Ratio_{sample} / Ratio_{standard}) - 1) \times 1000$$

where Ratio= $^{15}N / ^{14}N$ or $^{13}C / ^{12}C$.

Trophic levels of fish species were calculated using the following equation:

$$TL\ fish_{year} = (\delta^{15}N\ fish_{year} - \delta^{15}N\ snails_{year}) / 3.4‰ + 2$$

to adjust for temporal differences in ^{15}N at the base of the food chain (Post et al. 2000) in Lake Laberge prior to statistical analyses.

4.3.5 Statistical Analysis

Statistical analyses were conducted using Systat 10.2® for Windows. For each fish species, temporal differences in morphological parameters (length, weight, age, condition factor) as well as tissue lipid (%) content, were tested using analysis of variance (ANOVA); p =0.05) followed by Tukey’s post-hoc analysis to test for differences between individual years. Log_{10} values were used if it normalized the distribution of the data. Due to low N for ages in some years, some year categories were removed prior to the ANOVA of mean ages over time. The 2000 and 2001 datasets were also combined for some species analyses (stable isotopes when n<5, BMFs and general linear multiple regressions) to improve sample sizes for comparisons.

Major OC categories grouped by fish species were tested for significant differences across years using ANOVA. OC concentrations were not adjusted using analysis of covariance (ANCOVA) with lipid as a covariate because of heterogeneity of regression slopes indicating significant interactions of lipid content with OC concentrations over time (across years). In addition, regressions showed that lipid did not exhibit a consistent linear relationship with each OC group for each species for each year of samples. OC concentrations were adjusted using ANCOVA with age as a covariate for burbot data, but only for the OC groups that showed a linear relationship (and no year interaction) between age and OC as determined by general linear models (GLM). Wet weight OC concentrations were used in the statistical analyses and BMF calculations, and all values are reported in wet weights unless otherwise indicated. Since tissue lipid (%) was not a primary predictor of OC in all fish in all years based on multiple regression analyses and because of the previously mentioned year*lipid interaction, there was no normalizing the BMF for each species for this parameter. OC data were $\log_{10}$ transformed prior to statistical analyses to normalize the distribution of the data.

Relationships between OC concentrations and biological parameters including tissue lipid (%), $\log_{10}$ weights and $\log_{10}$ lengths were analysed using general linear multiple regressions (GLMR) for fish that exhibited significant temporal, morphological and OC concentration differences. This included least cisco, round whitefish, longnose sucker, lake whitefish, lake trout and burbot. Inconnu and broad whitefish were excluded due to their low sample numbers while northern pike were excluded due to a large leverage in two points of data, along with one outlier and low sample numbers. Based on research from Kidd (1996) and from the results of preliminary single regressions and stepwise regressions, models for longnose sucker included log length and tissue lipid (%) while the models for round whitefish included weight and tissue lipid (%). Cisco and lake whitefish regression models included log weight and tissue lipid (%) as variables. For this study, regressions were run to determine the most influential factors on OC concentrations for each fish species and within each year. Results for lake trout and burbot have already been reported in Chapter 3. However, some results for burbot (Section 3.3.3) were re-analysed due to a recent addition of 16 points of 1993 data (Kidd, 1996).

The slope of the OC-$\delta^{15}N$ regression or food web magnification factors (FWMF), provides an estimate of the rates of contaminant biomagnification through the food web (Kidd et al. 1995b; Kiruluk et al. 1995). General linear models were used, with $\delta^{15}N$ and year as factors (and a $\delta^{15}N$ * year interaction term) to compare the changes in slopes of OC concentrations from biota sampled between 1993 and 2000-2001 data. The OC-$\delta^{15}N$ relationship for 2000 and 2001 datasets were also tested to ensure homogeneity of slopes prior to pooling the data for these two years.

Differences in trophic levels for each fish species between 1993 and 2000-2001 data were tested using ANOVA. Lipid content and weights were also plotted with $\delta^{15}N$ to observe correlations between trophic level, fish weights and body fat levels.

4.4 Results

4.4.1 Contaminants and Morphology Trends

4.4.1.1 Lake trout. The results of the lake trout OC concentrations from 1993 to 2001 are reported in Chapter 3 (see Chapter 3 Results section and Table 4.2). All 6 OC contaminant groups have shown significant temporal decreases in concentrations from 1993 to 2003 (Table 4.3).

Changes in ages, lengths, weights and tissue lipid (%) are described in Chapter 3. In brief, significant increases have occurred in both mean lengths and weights along with significant decreases in lipid content for this species (Table 4.1, Table 4.4).

4.4.1.2 Burbot. The results of the burbot OC concentration trends from 1993 to 2001 have changed slightly from Chapter 3. The additional data (see Methods) and use of the ANCOVA procedure has allowed for the age adjusting of 3 categories including chlordane, DDT and PCB as the 3 groups were shown to have a linear relationship with age (ΣCHL p=0.01, r^2=0.09; ΣDDT p=0.05, r^2=0.05; ΣPCB p<<0.05, r^2=0.16) and no significant interaction term (age*year, p>0.05). Of these three OC groups, only the ΣDDT concentrations showed any change (a significant decrease, p=0.03) when age was used as a covariate (Figure 4.1). The ANOVA results on the remaining OC groups have shown ΣHCH is significantly lower in 2001 compared to 1993 (Table 4.3, Table 4.2; Figure 4.1). This was originally reported in Chapter 3 as having remained unchanged, although Chapter 3 data did have a significant decrease between 1999 and 2000-

2001 samples. The other OC groups remained unchanged from the earlier report (Chapter 3 results section).

In contrast to data used in Chapter 3, mean burbot ages herein displayed a significant temporal increase from 1993 to 2001 (p=0.01;Table 4.1, Table 4.4, Figure 4.2). Mean burbot fork lengths also had a significant increase from 1993 to 2001 (p<0.01; Figure 4.2), while mean weights had no significant change from 1993 to 2001 (p=0.09) and liver lipid contents had a significant decrease from 1993 to 2001 (p<0.01) as reported in Chapter 3. Condition factor (K) results still indicate a significant decrease in values from 1993 to 2001 (p<0.01; Figure 4.2) from 0.7 to 0.5 as reported in Chapter 3.

GLMR analysis for the revised 1993 dataset showed that the model was significant for each OC group (Table 4.5) compared to the results from Chapter 3 (Table 3.5) that showed the model was not significant for the contaminants. Tissue lipid (%) and log age were the most consistent, significant factors correlating with OC concentrations in burbot in both the 1993 and 2000-2001 data.

Table 4.2 Mean OC concentrations (ng/g ww) by organochlorine group (±SE), species and year for fish and zooplankton from Lake Laberge collected from 1993 to 2003.

		1993	1996	1998	1999	2000	2001	2002	2003
Lake trout	**N**	24	13			6	16	5	8
	å **CBz**	3.92	4.90			2.26	2.11	1.15	1.21
		(0.57)	(1.24)			(0.59)	(0.17)	(0.25)	(0.28)
	å **HCH**	4.69	6.50			2.30	0.80	1.58	0.54
		(0.78)	(1.79)			(1.08)	(0.07)	(0.50)	(0.10)
	å **CHL**	47.60	53.38			22.36	26.37	7.26	7.44
		(8.84)	(13.74)			(5.84)	(5.14)	(1.59)	(2.24)
	å **DDT**	391.54	236.51			96.46	89.46	54.50	61.48
		(132.69)	(41.39)			(14.21)	(14.04)	(11.58)	(8.55)
	å **PCB**	328.28	209.32			138.95	139.71	48.60	81.01
		(121.49)	(52.08)			(60.89)	(53.75)	(8.81)	(29.83)
	å **CHB**	310.96	212.23			207.33	154.20	139.23	179.31
		(62.36)	(28.31)			(49.90)	(60.46)	(16.88)	(42.79)
Burbot	**N**	29	15		11	17	27		
	å **CBz**	25.97	40.11		41.58	26.31	33.98		
		(2.75)	(7.11)		(5.31)	(3.40)	(3.05)		
	å **HCH**	32.20	42.06		50.25	21.09	8.43		
		(3.22)	(7.50)		(7.15)	(3.04)	(0.79)		
	å **CHL**	235.27	270.22		247.76	219.01	258.73		
		(31.71)	(52.10)		(36.73)	(24.86)	(28.98)		
	å **DDT**	2393.46	2675.37		2849.84	2069.57	2101.94		
		(314.79)	(475.56)		(361.70)	(216.53)	(381.02)		
	å **PCB**	1287.44	1598.31		1625.39	1172.08	1753.41		
		(161.07)	(292.05)		(219.22)	(109.01)	(211.19)		
	å **CHB**	2413.99	2659.19		3053.46	1281.08	1106.47		
		(264.70)	(442.96)		(364.65)	(111.90)	(150.14)		

		1993	1996	1998	1999	2000	2001	2002	2003
Inconnu	**N**					2	6		
	å **CBz**					0.53	3.83		
						(0.06)	(0.96)		
	å **HCH**					0.32	1.17		
						(0.03)	(0.41)		
	å **CHL**					1.19	14.77		
						(0.09)	(3.95)		
	å **DDT**					20.50	75.07		
						(1.92)	(12.78)		
	å **PCB**					6.69	64.15		
						(0.06)	(15.70)		
	å **CHB**					10.95	81.33		
						(2.37)	(30.43)		
Northern pike	**N**	8		1		5	5		
	å **CBz**	0.18				0.13	0.61		
		(0.02)		0.06		(0.01)	(0.33)		
	å **HCH**	0.12				0.06	0.08		
		(0.01)		0.18		(0.01)	(0.01)		
	å **CHL**	0.99				0.47	7.95		
		(0.11)		0.56		(0.12)	(3.05)		
	å **DDT**	8.35				13.68	13.81		
		(0.97)		3.32		(3.86)	(3.41)		
	å **PCB**	7.47				8.08	7.12		
		(1.10)		3.19		(2.94)	(1.85)		
	å **CHB**	9.69				1.80	4.62		
		(1.37)		1.58		(0.44)	(1.11)		

		1993	1996	1998	1999	2000	2001	2002	2003
Lake whitefish	**N**	30		5		14	14		
	å **CBz**	1.16		0.37		0.27	0.31		
		(0.11)		(0.04)		(0.04)	(0.06)		
	å **HCH**	1.58		0.31		0.13	0.12		
		(0.18)		(0.06)		(0.01)	(0.02)		
	å **CHL**	8.94		2.95		0.56	0.98		
		(1.03)		(0.34)		(0.09)	(0.39)		
	å **DDT**	82.53		24.59		15.73	17.11		
		(11.60)		(2.36)		(1.81)	(3.75)		
	å **PCB**	66.56		5.13		9.34	20.40		
		(10.42)		(0.66)		(2.69)	(6.04)		
	å **CHB**	62.13		42.75		2.49	4.83		
		(8.54)		(7.68)		(0.57)	(2.05)		
Cisco	**N**	7				21	2		
	å **CBz**	1.38				0.23	0.75		
		(0.33)				(0.05)	(0.63)		
	å **HCH**	1.62				0.18	0.17		
		(0.43)				(0.04)	(0.09)		
	å **CHL**	6.37				0.85	0.67		
		(1.44)				(0.37)	(0.17)		
	å **DDT**	31.77				6.98	8.57		
		(5.94)				(1.65)	(5.07)		
	å **PCB**	21.21				3.70	5.16		
		(3.33)				(0.67)	(0.13)		
	å **CHB**	68.28				3.71	2.16		
		(13.69)				(0.61)	(1.62)		

		1993	1996	1998	1999	2000	2001	2002	2003
Longnose sucker	**N**	12				12	3		
	å **CBz**	0.27				0.20	0.20		
		(0.04)				(0.04)	(0.04)		
	å **HCH**	0.38				0.11	0.15		
		(0.05)				(0.02)	(0.02)		
	å **CHL**	2.15				0.52	1.29		
		(0.39)				(0.17)	(0.57)		
	å **DDT**	16.12				15.89	34.63		
		(2.24)				(3.23)	(12.74)		
	å **PCB**	13.61				12.45	31.79		
		(2.13)				(5.06)	(7.91)		
	å **CHB**	17.45				1.40	4.77		
		(3.49)				(0.50)	(3.02)		
Round whitefish	**N**	7				4	20		
	å **CBz**	0.79				0.45	0.44		
		(0.18)				(0.16)	(0.10)		
	å **HCH**	0.93				0.13	0.17		
		(0.17)				(0.03)	(0.02)		
	å **CHL**	3.27				0.34	3.11		
		(0.76)				(0.06)	(1.03)		
	å **DDT**	22.58				11.72	4.50		
		(4.16)				(2.74)	(1.77)		
	å **PCB**	17.37				5.00	3.16		
		(3.68)				(0.53)	(0.84)		
	å **CHB**	25.76				0.98	1.44		
		(4.49)				(0.23)	(0.31)		

		1993	1996	1998	1999	2000	2001	2002	2003
Broad whitefish	**N**					8			
	å **CBz**					0.52			
						(0.07)			
	å **HCH**					0.25			
						(0.11)			
	å **CHL**					0.28			
						(0.09)			
	å **DDT**					8.18			
						(3.12)			
	å **PCB**					1.57			
						(0.40)			
	å **CHB**					0.37			
						(0.15)			
Zooplankton	**N**	4				7	4		
	å **CBz**	0.54				0.09	0.08		
		(0.08)				(0.01)	(0.03)		
	å **HCH**	1.26				0.27	0.20		
		(0.44)				(0.04)	(0.03)		
	å **CHL**	1.35				0.18	0.14		
		(0.46)				(0.05)	(0.03)		
	å **DDT**	4.09				1.16	1.05		
		(0.77)				(0.21)	(0.25)		
	å **PCB**	5.33				2.07	0.68		
		(0.38)				(1.31)	(0.30)		
	å **CHB**	15.04				2.92	2.56		
		(3.87)				(0.73)	(0.62)		

Table 4.3 Statistical results from temporal trend analysis of OC in fish over time for Lake Laberge biota from 1993 to 2001 and a list of probable attributing causes for changes in OC concentrations.

Fish	**# temporal points**	**ΣCBz**	**ΣHCH**	**ΣCHL**	**ΣDDT**	**ΣPCB**	**ΣCHB**	**Possible cause for OC concentration trend**
Lake trout	6	d	d	d	d	d	d	Decreased lipid, growth dilution, decrease in prey OC
Burbot	5	nc	d	nc	d	nc	d	Decreased lipid, older fish, decrease in prey OC, lower trophic level
Inconnu	2	-	-	-	-	-	-	
Northern pike	4	i	d	i	nc	nc	d	More piscivorous, growth dilution, decrease in prey OC
Lake whitefish	4	d	d	d	d	d	d	Decreased lipid, decrease in prey OC
Cisco	3	d	d	d	d	d	d	Decrease in prey OC, some growth dilution
Longnose sucker	3	nc	d	d	nc	i (m)	d	Marginally younger and smaller fish, possible decreasing lipid trend
Round whitefish	3	nc	d	nc	d	d	d	Decreased lipid, growth dilution, decrease in prey OC
Broad whitefish	1	-	-	-	-	-	-	
Zooplankton	3	d	d	d	d	d	d	Possible growth dilution (higher densities), community species composition change

i = increase significantly (*p*<0.05)
d = decrease significantly (*p* <0.05)
nc = no change (*p*> 0.05)
m = marginal significance (*p* =0.04 - 0.06)

Table 4.4 Significant temporal trends in morphological parameters and tissue lipid (%) over time in aquatic biota from Lake Laberge collected from 1993 to 2001.

Fish	**# temporal points**	**tissue lipid (%)**	**Length**	**Weight**	**Age**	**K**
Lake trout	6	d	i	i	nc	nc
Burbot	5	d	i	nc	i	d
Inconnu	2	-	-	-	-	-
Northern pike	4	nc	i*	i*	nc	nc
Lake whitefish	4	d	nc	nc	nc	nc
Cisco	3	d	i	i (m)	nc	d
Longnose sucker	3	nc	d*	d*	d (m)*	d
Round whitefish	3	d	i	i	nc*	nc
Broad whitefish	1	-	-	-	-	-
plankton	3	nc	-	-	-	-

* See results texts for caveats

i = increase significantly (p<0.05)

d = decrease significantly (p <0.05)

nc = no change (p> 0.05)

m = marginal significance (p =0.04 - 0.06)

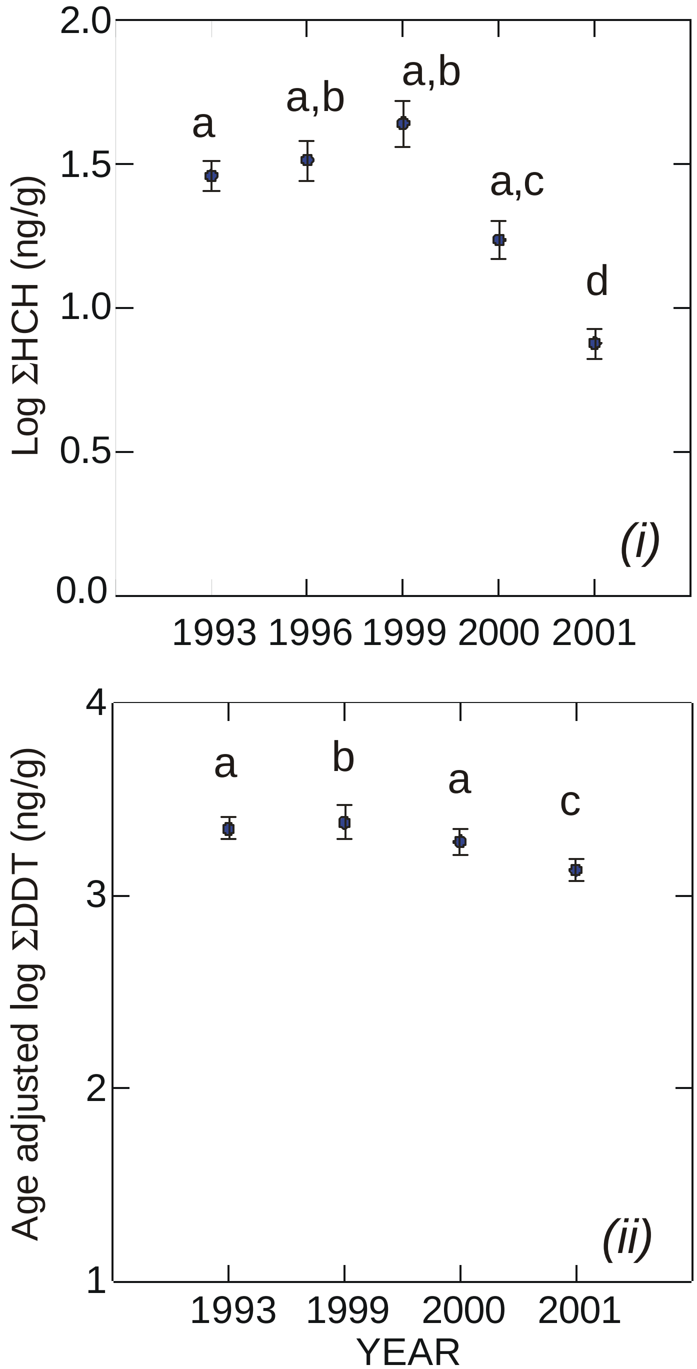


Figure 4.1 Least squares means (and SE) of (i) log ΣHCH and (ii) age adjusted log ΣDDT (ng/g ww) concentrations in burbot from Lake Laberge from 1993 to 2001. Note there were no 1996 ages for burbot available. Significant differences (p<0.05) between years are shown by different letters.

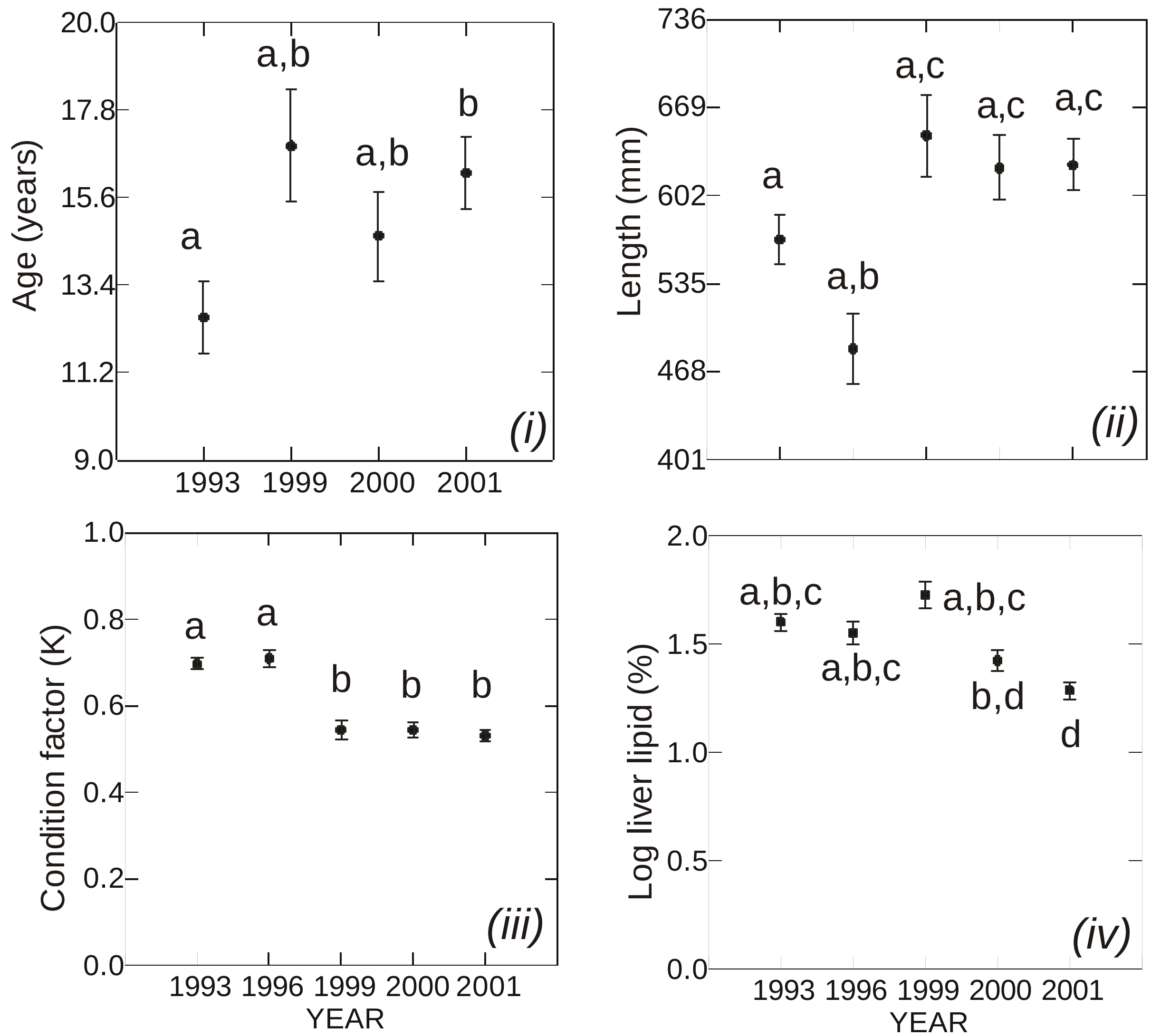


Figure 4.2 Least squares means (and SE) of (i) ages, (ii) lengths, (iii) condition factors and (iv) tissue lipid (%) content in burbot from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

Table 4.5 General linear multiple regression results for burbot from Lake Laberge for samples collected in 1993 and 2000-2001 (pooled) and analysed for OC groups.

Year	**OC**	***p* (model)**	**r^2**	**Variable**	***p (2 tail)***
1993	ΣCBz	0.02	0.34	% lipid	0.01
				weight	0.41
				log age	0.03
	ΣHCH	0.10	0.25	% lipid	0.04
				weight	0.80
				log age	0.15
	ΣCHL	<<0.05	0.48	% lipid	0.24
				weight	0.26
				log age	0.19
	ΣDDT	<<0.05	0.51	% lipid	0.18
				weight	0.31
				log age	0.13
	ΣPCB	<<0.05	0.45	% lipid	0.12
				weight	0.94
				log age	0.04
	ΣCHB	<<0.05	0.47	% lipid	0.03
				weight	0.70
				log age	0.05
2000-2001	ΣCBz	<<0.05	0.50	% lipid	<<0.05
				weight	0.13
				log age	0.46
	ΣHCH	<<0.05	0.58	% lipid	<<0.05
				weight	0.65
				log age	0.56
	ΣCHL	<<0.05	0.54	% lipid	<<0.05
				weight	0.03
				log age	<<0.05
	ΣDDT	<<0.05	0.51	% lipid	0.01
				weight	0.22
				log age	0.01
	ΣPCB	0.01	0.40	% lipid	0.09
				weight	0.06
				log age	0.01
	ΣCHB	0.01	0.42	% lipid	0.01
				weight	0.40
				log age	0.04

4.4.1.3 Inconnu. There were too few samples for a comparison of morphological characteristics (N=0 for 1993, N=2 for 2000) or OC concentrations (Table 4.2) over time. Mean values for morphological parameters and OC levels are summarized in Table 4.1 and Table 4.2, respectively.

4.4.1.4 Northern Pike. Mean concentrations of OC in northern pike exhibited some changes from 1993 to 2001 (Tables 4.2 and 4.3). The ΣCBz concentrations showed a marginally significant increase over time (p=0.04, Figure 4.3) although there were no statistically significant differences between any two years. A similar trend was noted for ΣDDT but there was no significant change (p=0.30) in concentrations from 1993 to 2001 (Figure 4.3). The ΣHCH levels decreased significantly over time (p=0.003; Figure 4.3). Both ΣCHL and ΣCHB showed significant fluctuations over time (both p=0.01) but no consistent trend was apparent (Figure 4.3), while ΣPCB concentrations had no significant change from 1993 to 2001 (p=0.74).

Morphological parameters of northern pike also displayed significant changes over time but with some variability in 1998 data (Table 4.1, Table 4.4). There was no significant change in mean age over time (p=0.15). There were significant increasing trends in length and weights (both p=0.01) but the spike in 1998 samples have had a large influence on the ANOVA result (Figure 4.4). The differences in mean length and weight between 1993 and 2001 pike are not

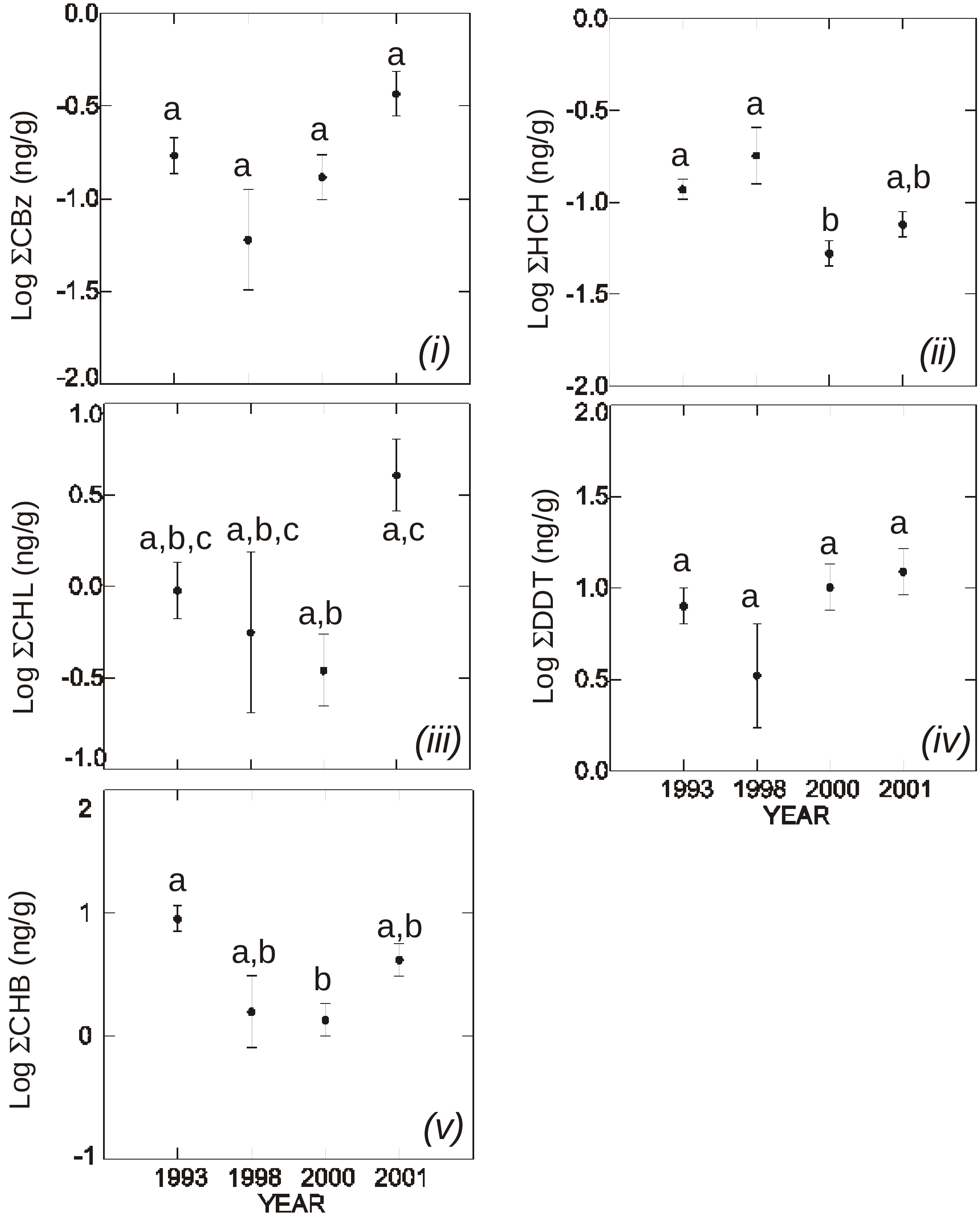


Figure 4.3 Least squares means (and SE) of (i) log ΣCBz, (ii) log ΣHCH, (iii) log ΣCHL, (iv) log ΣDDTand (v) log ΣCHB (ng/g ww) concentrations in northern pike from Lake Laberge from 1993 to 2001. Significant differences (*p*<0.05) between years are shown by different letters on the graph.

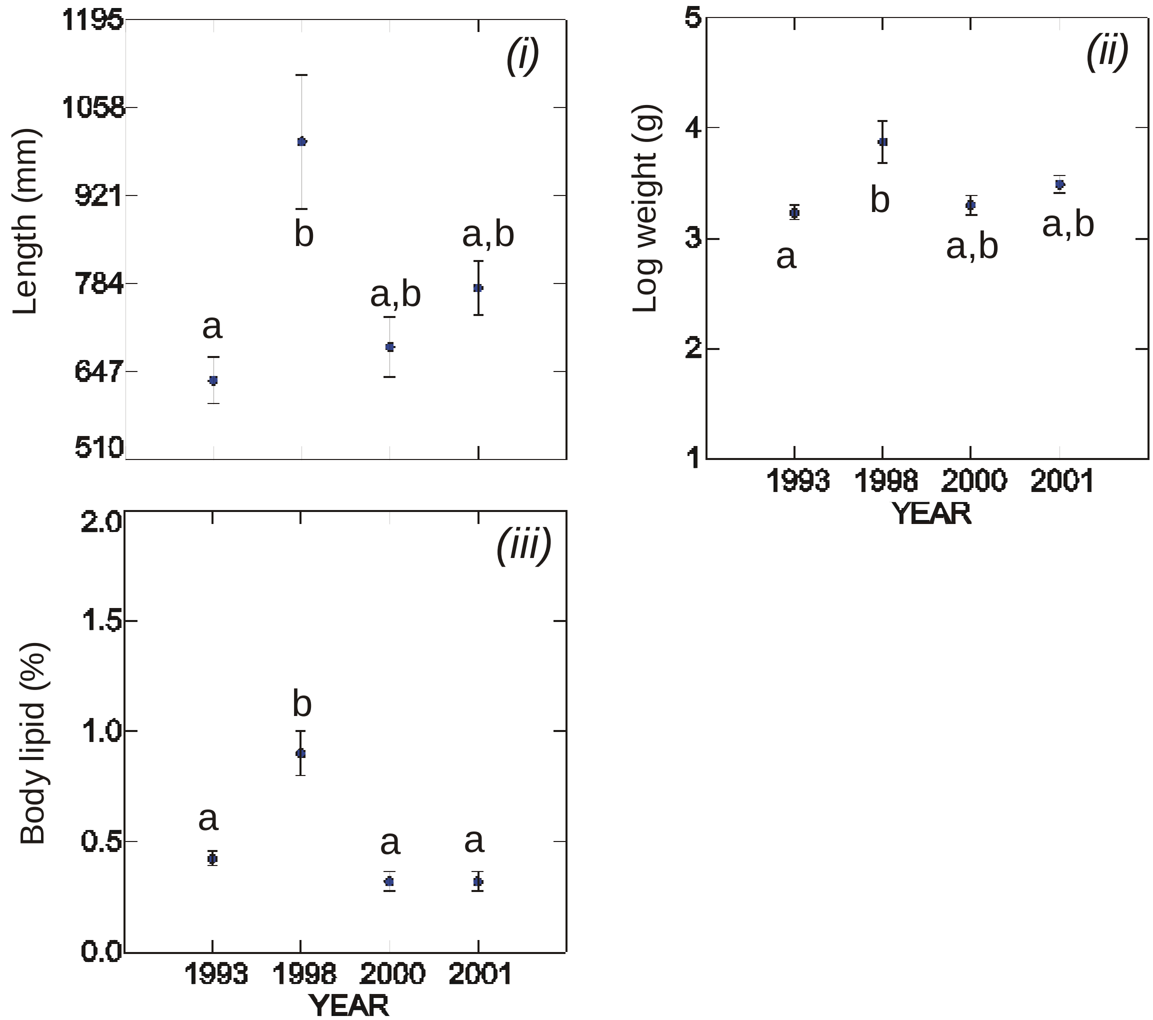


Figure 4.4 Least squares means (and SE) of (i) fork length, (ii) log weight and (iii) tissue lipid (%) content in northern pike from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

significant (*p*=0.08 and 0.09, respectively). Tissue lipid (%) had a similar pattern compared to the other morphological parameters with a significant change in values over time (*p*=0.001) but also with a spike in 1998 samples. The difference in lipid content between 1993 and 2001 pike was not statistically significant (*p*=0.29) and there was no evidence of a trend in the mean (Figure 4.4). Condition factors showed no significant change in pike over time (*p*=0.41).

4.4.1.5 Lake Whitefish. The OC concentrations in lake whitefish (Tables 4.2 and 4.3) significantly decreased from 1993 to 2001 (*p*<<0.01 for all 6 OC groups; Figure 4.5). The ΣPCB levels appear to have increased slightly since 1998, but it should be noted there were few samples for this year (N=5). There was no significant difference between 1998 and 2001 ΣPCB concentrations (*p*=0.07).

There was a significant decrease in lipid content (*p*<<0.01, Figure 4.6) but no significant changes in age (*p*=0.55), length (*p*=0.87), or weight (*p*=0.45) in lake whitefish over time (Table 4.1, Table 4.4). Although not statistically significant (*p*=0.1), condition factors do exhibit a slight decreasing trend over time (Figure 4.6).

Results of the GLMR showed a correlation of OC concentrations with both lipid and log weight in 1993 samples, although lipid content was not statistically significant within the model for ΣDDT and ΣPCB while log weight was not significant for ΣHCH (Table 4.6) in this year. There was a change in the lipid content-OC relationship for 2000-2001 samples as lipid content became more significant within the model than log weight for 5 out of the 6 OC groups.

Table 4.6 General linear multiple regression results for lake whitefish from Lake Laberge for samples collected in 1993 and 2000-2001 (pooled) and analysed for OC groups.

Year	**OC**	***p* (model)**	**r^2**	**Variable**	***p* (2 tail)**
1993	S**CBz**	<<0.05	0.61	% lipid	<<0.05
				log weight	0.04
	S**HCH**	<<0.05	0.60	% lipid	<<0.05
				log weight	0.08
	S**CHL**	<<0.05	0.46	% lipid	0.01
				log weight	<<0.05
	S**DDT**	0.01	0.28	% lipid	0.36
				log weight	0.01
	S**PCB**	0.01	0.30	% lipid	0.49
				log weight	<<0.05
	S**CHB**	<<0.05	0.44	% lipid	<<0.05
				log weight	0.02
2000-2001	S**CBz**	0.01	0.34	% lipid	<<0.05
				log weight	0.84
	S**HCH**	<<0.05	0.52	% lipid	<<0.05
				log weight	0.03
	S**CHL**	<<0.05	0.50	% lipid	<<0.05
				log weight	0.62
	S**DDT**	<<0.05	0.37	% lipid	<<0.05
				log weight	0.72
	S**PCB**	0.16	0.36	% lipid	0.06
				log weight	0.49
	S**CHB**	0.03	0.24	% lipid	0.01
				log weight	0.75

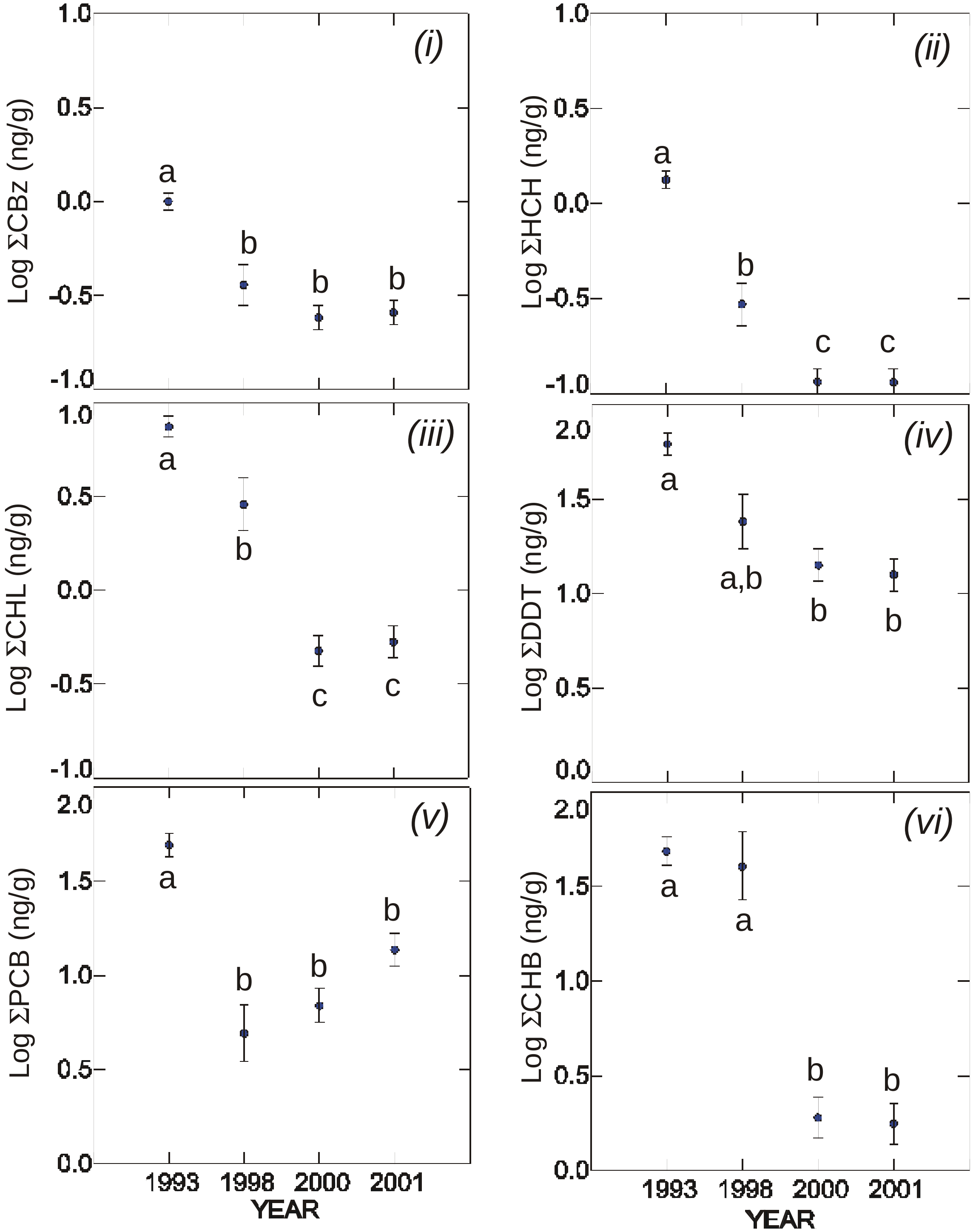


Figure 4.5 Least squares means (and SE) of (i) log ΣCBz, (ii) log ΣHCH, (iii) log ΣCHL, (iv) log ΣDDT, (v) log ΣPCB and (vi) log ΣCHB ww concentrations in lake whitefish from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

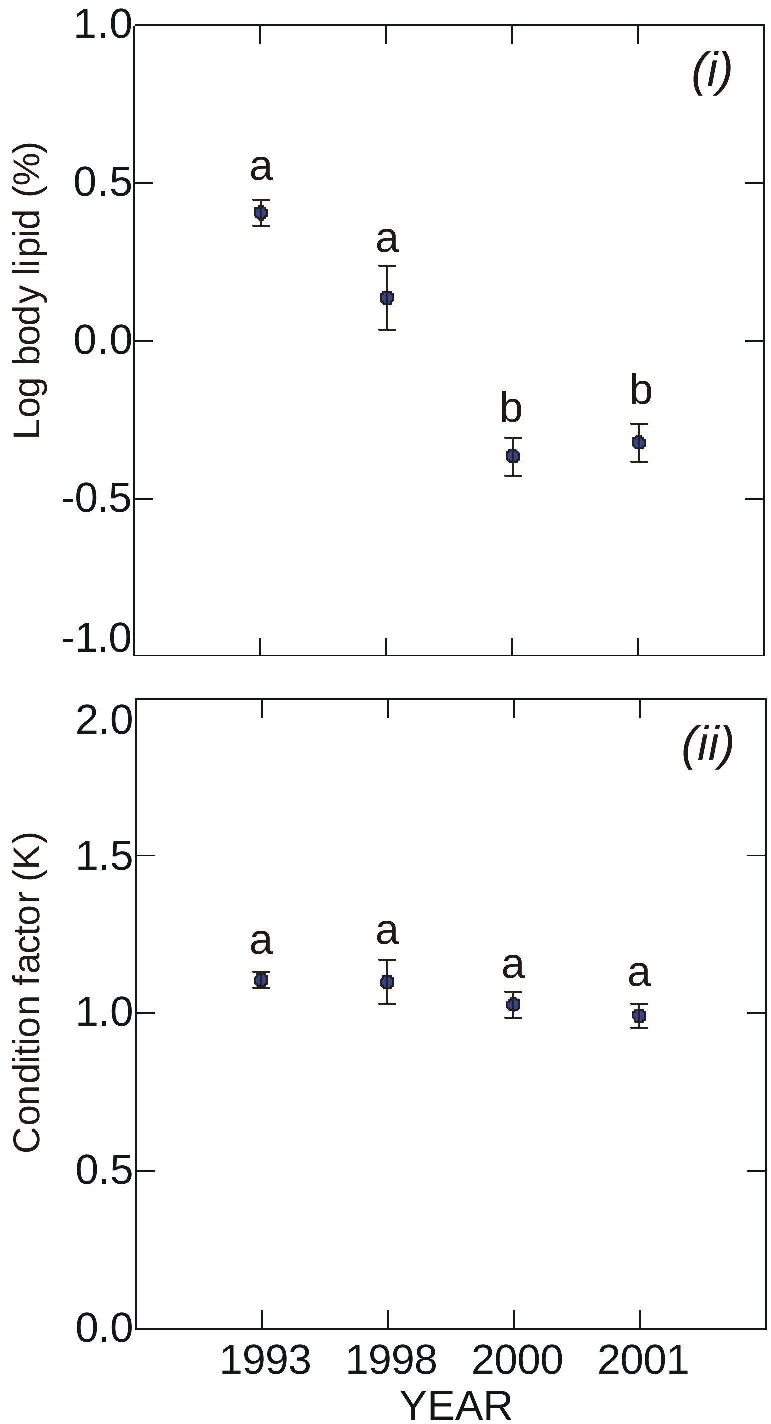


Figure 4.6 Least squares means (and SE) (i) log lipid content and (ii) condition factor in lake whitefish from Lake Laberge from 1993 to 2001. Significant differences (*p*<0.05) between years are shown by different letters on the graph.

4.4.1.6 Least Cisco. Analyses of OC concentrations in least cisco showed significant decreases (*p*<<0.01) for all 6 OC groups from 1993 to 2001 (Table 4.2 and 4.3; Figure 4.7). Means for 2001 had high variance due to the low sample numbers for that year (n=2) but Tukey's test was used for comparison of values between 1993 and 2000 samples to confirm the results of the ANOVA.

Morphological parameters of least cisco showed significant changes over time (Table 4.1, Table 4.4). Although mean age was not significantly different from 1993 to 2001 (*p*=0.56), lengths showed a significant increase (*p*<0.02) over the same period (Figure 4.8). Mean weight had a marginally significant increase from 1993 to 2001 (*p*<0.04), which was more significant (*p*=0.03) when the 2001 data (n=2) were removed (Figure 4.8). Lipid content had a highly significant decrease from 1993 to 2001 (*p*<0.01; Figure 4.8). Condition factors displayed a significant decrease (*p*=0.01) from 1993 to 2001, although there were no significant differences when the 2001 data were removed (*p*=0.98; Figure 4.8).

Results of the GLMR showed a significant correlation of most OC concentrations with the log weight and tissue lipid (%) model in 1993 samples, although tissue lipid (%) was not as significant as log weight within the model except for ΣHCH. There was a change in the tissue lipid (%)-log weight-OC relationship in 2000-2001 as GLMR showed the model was only significant for ΣCBz and ΣCHB with tissue lipid (%) being the only significant variable within the model for these two contaminant groups (Table 4.7). The model accounted for much less of the variance in OC concentrations compared to 1993 data.

Table 4.7 General linear multiple regression results for cisco from Lake Laberge for samples collected in 1993 and 2000-2001 (pooled) and analysed for OC groups.

Year	OC	*p* (model)	r^2	Variable	*p* (2 tail)
1993	S**CBz**	0.01	0.89	% lipid	0.13
				log weight	0.12
	S**HCH**	<<0.05	0.94	% lipid	0.05
				log weight	0.06
	S**CHL**	0.01	0.93	% lipid	0.10
				log weight	0.06
	S**DDT**	<<0.05	0.95	% lipid	0.16
				log weight	0.01
	S**PCB**	<<0.05	0.94	% lipid	0.20
				log weight	0.02
	S**CHB**	<<0.05	0.95	% lipid	0.53
				log weight	0.01
2000-2001	S**CBz**	0.02	0.34	% lipid	0.01
				log weight	0.25
	S**HCH**	0.51	0.06	% lipid	0.39
				log weight	0.54
	S**CHL**	0.20	0.15	% lipid	0.15
				log weight	0.39
	S**DDT**	0.21	0.14	% lipid	0.22
				log weight	0.28
	S**PCB**	0.95	0.01	% lipid	0.79
				log weight	0.91
	S**CHB**	0.05	0.26	% lipid	0.02
				log weight	0.87

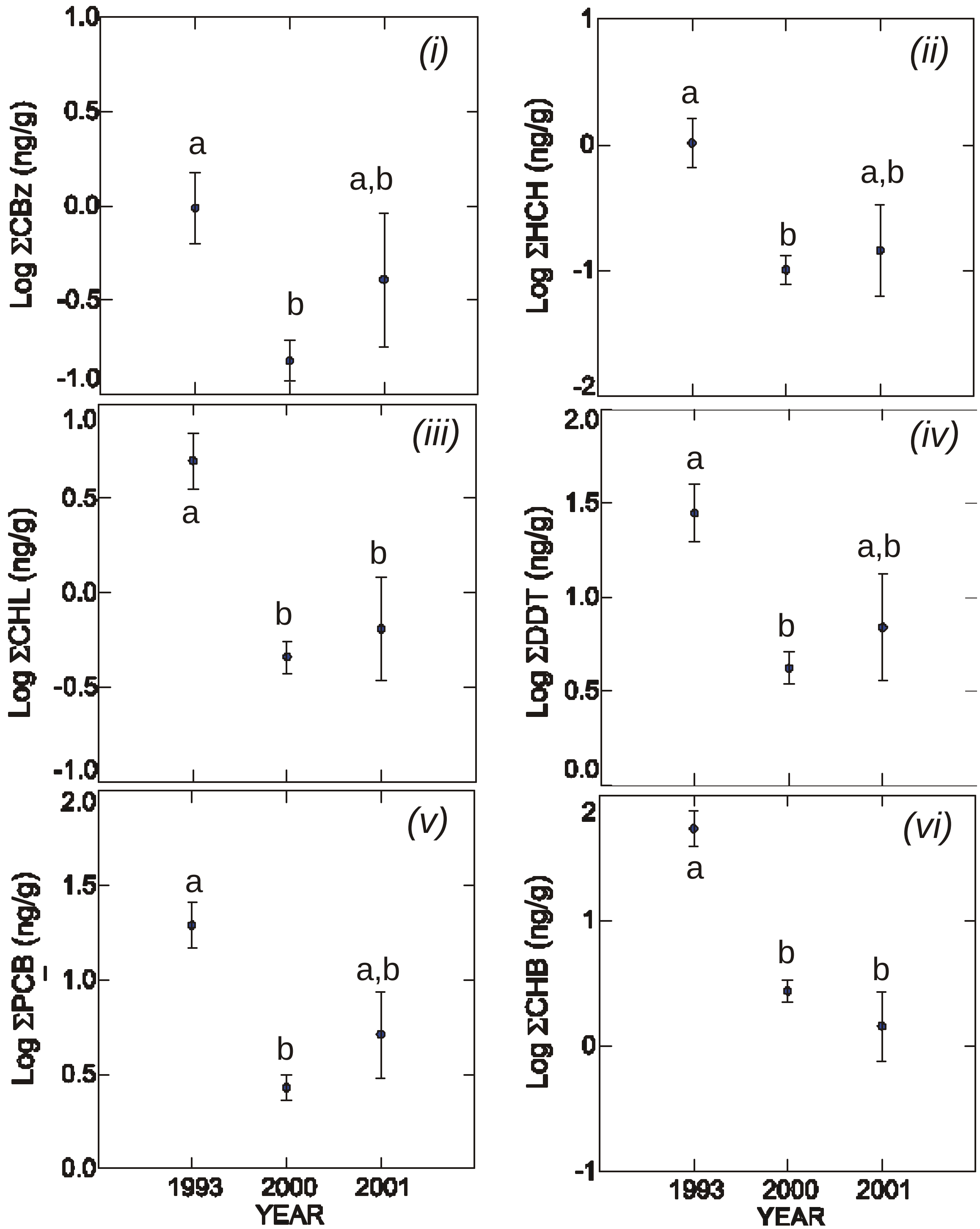


Figure 4.7 Least squares means (and SE) of (i) log ΣCBz, (ii) log ΣHCH, (iii) log ΣCHL, (iv) log ΣDDT, (v) log ΣPCB and (vi) log ΣCHB (ng/g ww) concentrations in least cisco from Lake Laberge from 1993 to 2001. Significant differences (*p*<0.05) between years are shown by different letters on the graph.

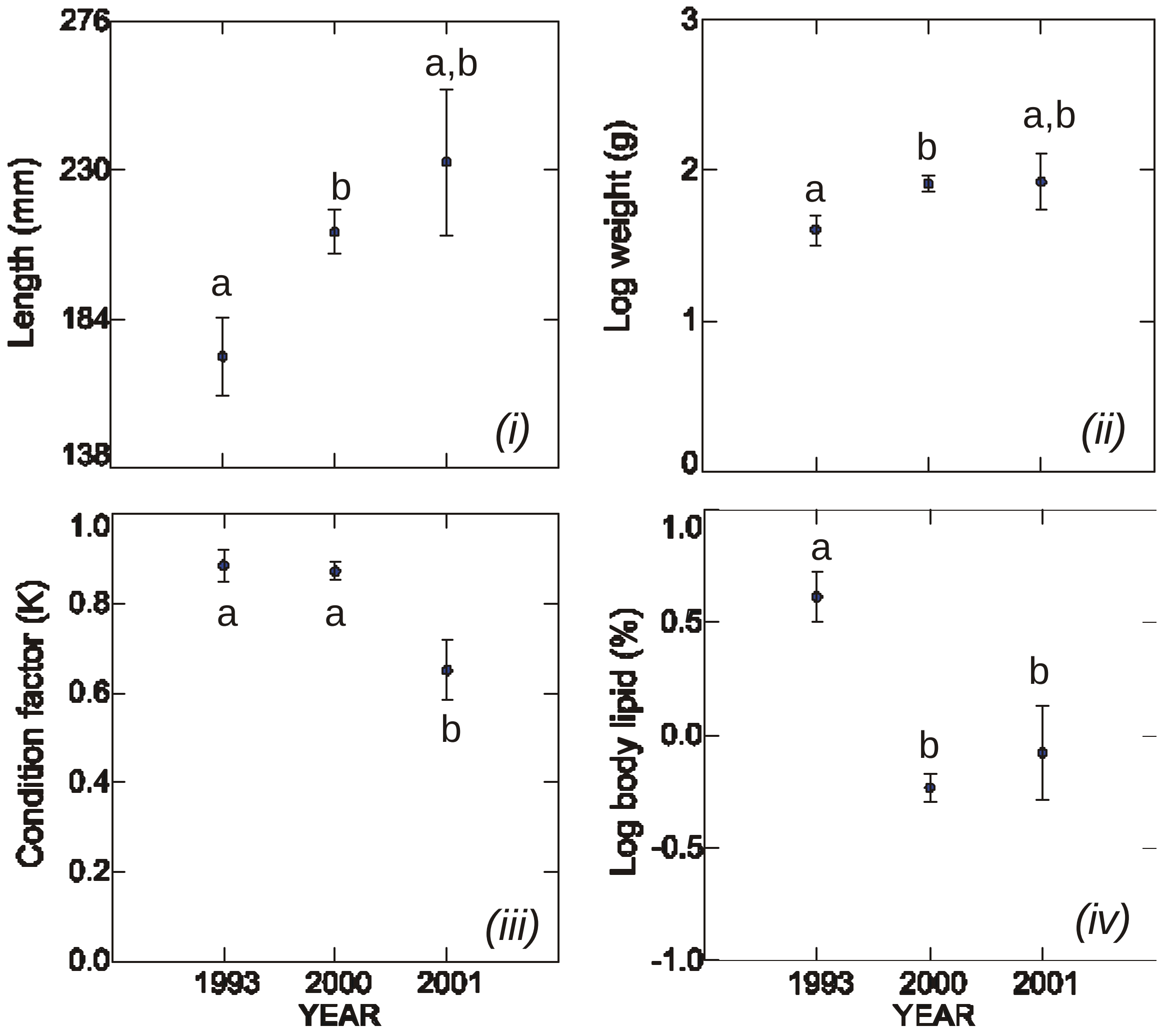


Figure 4.8 Least squares means (and SE) of (i) fork length, (ii) log weight, (iii) condition factors and (iv) log lipid content in least cisco from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

4.4.1.7 Longnose Sucker. No clear temporal trends were evident for half of the OC groups (Table 4.3) and data were more variable for 2001 likely due a lower number of samples analysed for that year (Table 4.2). There were no significant changes in ΣCBz (p=0.34), ΣDDT (p=0.19) or ΣPCB (p=0.07) concentrations. Significant decreases were noted for ΣHCH, ΣCHL and ΣCHB concentrations (all p<<0.01; Figure 4.9).

Analyses of the morphological parameters showed that mean age in suckers did not change in the whole model from 1993 to 2001 (p=0.06) but there is evidence of a decline as 1993 is significantly different when compared to 2001 (Figure 4.10). However, only 3 fish were aged in 1993 in the comparison (Tables 4.1 and 4.4). Both length and weight had significant decreases (p>>0.01) but only for 2001 (N=6) compared to both other years (Figure 4.10). There is no significant change in lengths or weights between the 1993 (N=12) and 2000 (N=14) samples (p=0.30 and 0.40 respectively) as determined by Tukey's test. Tissue lipid (%) had no significant change over time (p=0.14) but there is evidence of a decreasing trend (Figure 4.10). Condition factors have also shown a significant decrease (p<<0.05) from 1993 to 2001 (Figure 4.10).

Results of the GLMR showed an inconsistent pattern of OC correlation with lipid content that was more prevalent in samples from 1993, specifically for ΣCBz, ΣHCH and ΣCHB. Otherwise the regression models were not statistically significant with log lengths or tissue lipid (%) for 2000-2001 data (Table 4.8).

Table 4.8 General linear multiple regression results for longnose sucker from Lake Laberge for samples collected in 1993 and 2000-2001 (pooled) and analysed for OC groups.

Year	**OC**	***p* (model)**	**r^2**	**Variable**	***p* (2 tail)**
1993	S**CBz**	**0.01**	0.67	% lipid	0.01
				log length	0.15
	S**HCH**	0.05	0.48	% lipid	0.04
				log length	0.24
	S**CHL**	0.04	0.52	% lipid	0.09
				log length	0.06
	S**DDT**	0.22	0.28	% lipid	0.35
				log length	0.19
	S**PCB**	0.22	0.28	% lipid	0.56
				log length	0.13
	S**CHB**	0.01	0.64	% lipid	0.03
				log length	0.03
2000-2001	S**CBz**	0.09	0.36	% lipid	0.04
				log length	0.79
	S**HCH**	0.20	0.26	% lipid	0.08
				log length	0.67
	S**CHL**	0.30	0.20	% lipid	0.13
				log length	0.64
	S**DDT**	0.28	0.21	% lipid	0.16
				log length	0.30
	S**PCB**	0.53	0.11	% lipid	0.29
				log length	0.57
	S**CHB**	0.35	0.17	% lipid	0.19
				log length	0.86

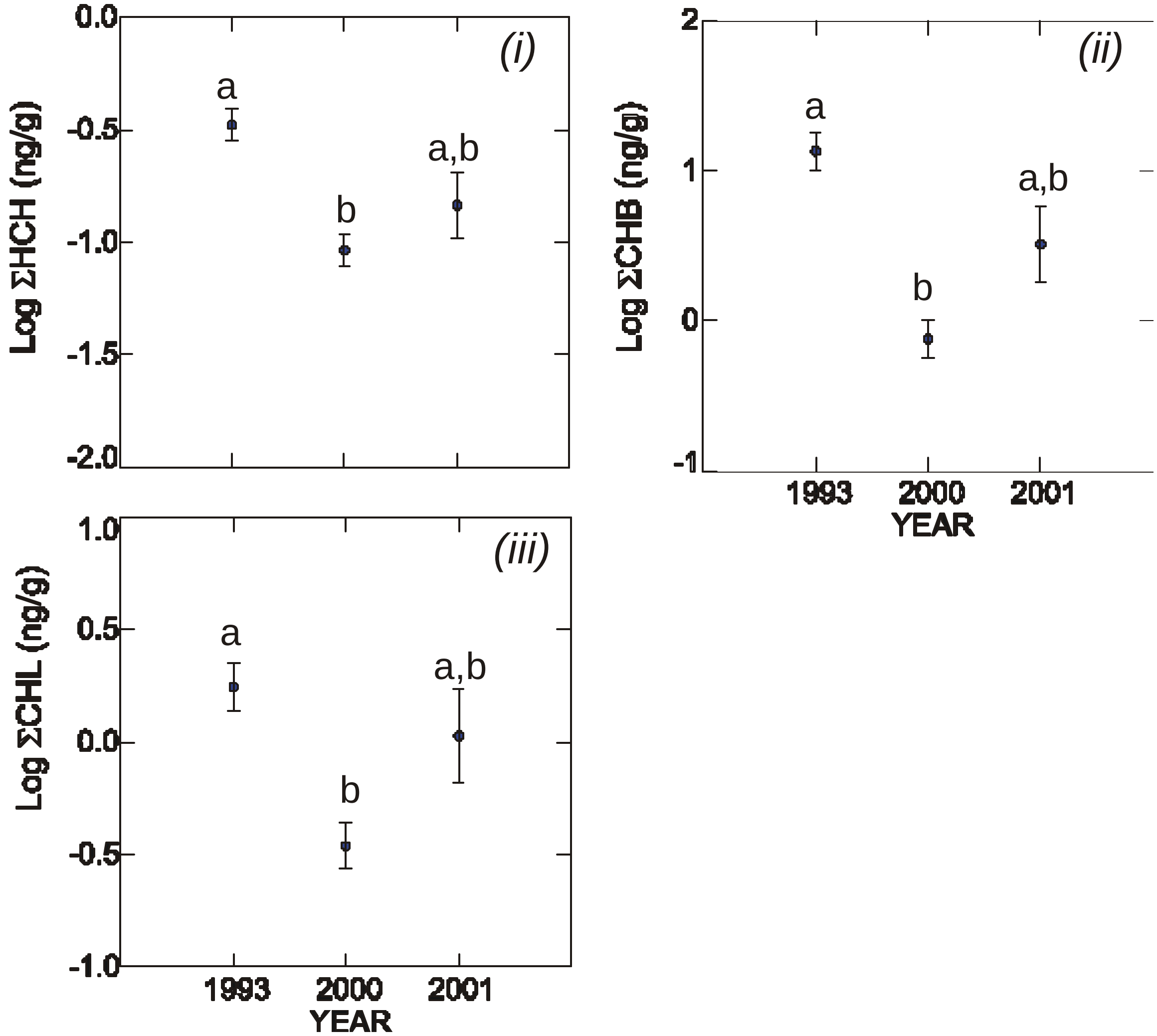


Figure 4.9 Least squares means (and SE) of (i) log ΣHCH, (ii) log ΣCHB and (iii) log ΣCHL (ng/g ww) concentrations in longnose sucker from Lake Laberge from 1993 to 2001. Significant differences ($p<0.05$) between years are shown by different letters on the graph.

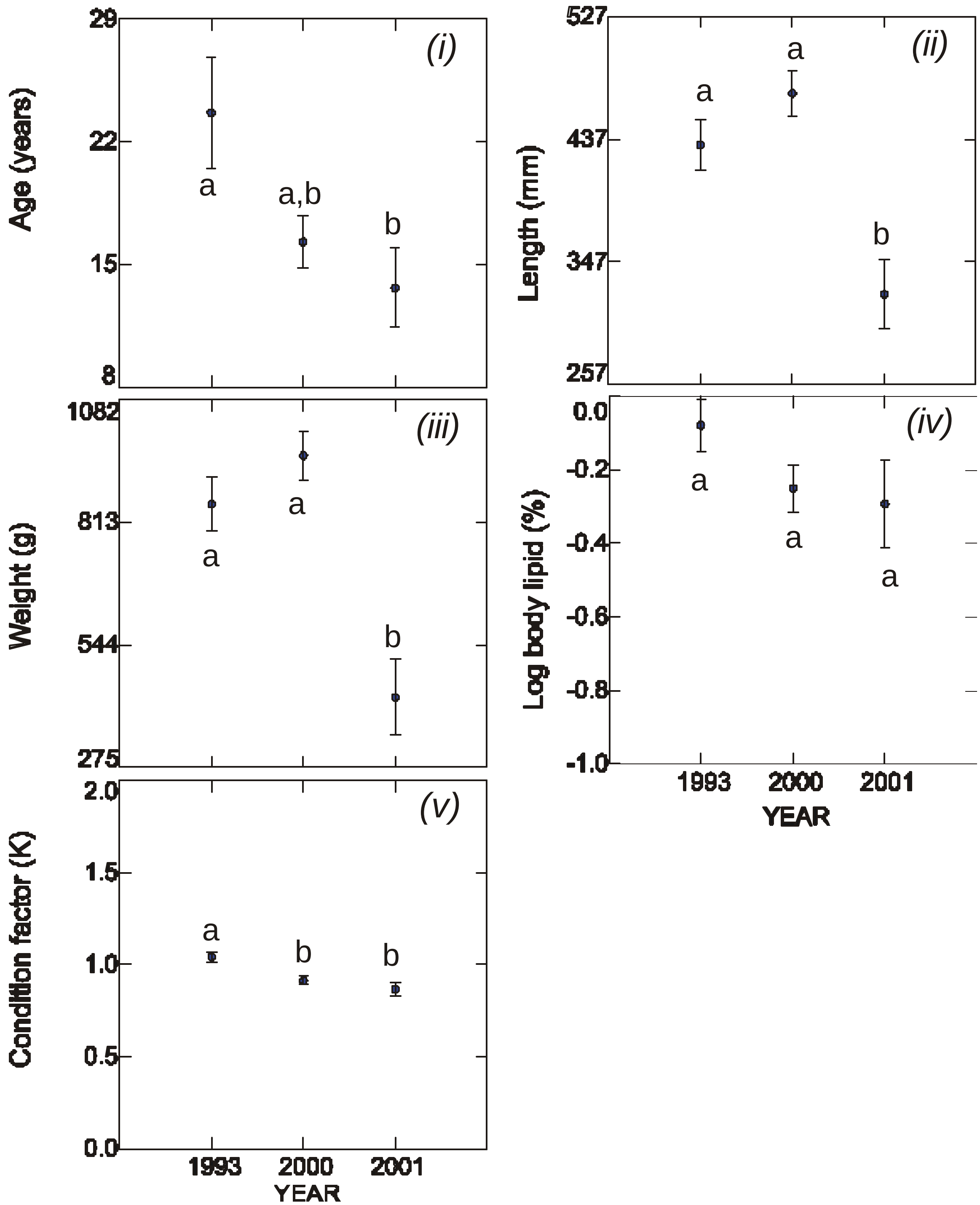


Figure 4.10 Least squares means (and SE) of (i) age, (ii) fork length, (iii) weight, (iv) log lipid content and (v) condition factors in longnose sucker from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

4.4.1.8 Round Whitefish. Results of the ANOVA for round whitefish demonstrated several significant decreases in contaminants (Table 4.3) although with increased variability in concentrations likely due to the lower number of samples for 2000 (N=4, Table 4.2). The ΣCBz and ΣCHL concentrations had no significant changes (p=0.10 and 0.08, respectively) but both exhibited decreasing trends (Figure 4.11). Significant decreases in concentrations (all p<0.01) were noted for the remaining 4 OC groups, ΣHCH, ΣDDT, ΣPCB and ΣCHB (Figure 4.11).

A temporal comparison of round whitefish ages was not done due to the low number of samples from 1993 (Table 4.1, Table 4.4). Round whitefish mean lengths and weights both had significant increases from 1993 to 2001 (both p<0.01; Figure 4.12). Tissue lipid (%) displayed a significant decrease (p>0.01; Figure 4.12) while condition factors showed no significant difference from 1993 to 2001 (p=0.31).

Results of the GLMR showed a distinct pattern of OC correlation with both lipid and log weight in 2000-2001 samples, except for ΣHCH and ΣCHB. The model was not significant for any contaminant group for 1993 fish (Table 4.9).

4.4.1.9 Broad Whitefish. Samples of broad whitefish were only available for the year 2000 such that no temporal analysis was possible. Mean values for morphological parameters and OC levels are summarized in Tables 4.1 and 4.2, respectively.

Table 4.9 General linear multiple regression results for round whitefish from Lake Laberge for samples collected in 1993 and 2000-2001 (pooled) and analysed for OC groups.

Year	OC	*p* (model)	r^2	Variable	*p* (2 tail)
1993	ΣCBz	0.49	0.30	% lipid	0.26
				log weight	0.77
	ΣHCH	0.90	0.05	% lipid	0.72
				log weight	0.74
	ΣCHL	0.36	0.40	% lipid	0.28
				log weight	0.25
	ΣDDT	0.50	0.29	% lipid	0.31
				log weight	0.46
	ΣPCB	0.61	0.22	% lipid	0.35
				log weight	0.84
	ΣCHB	0.23	0.52	% lipid	0.44
				log weight	0.19
2000-2001	ΣCBz	<<0.05	0.51	% lipid	<<0.05
				log weight	0.07
	ΣHCH	0.60	0.05	% lipid	0.73
				log weight	0.32
	ΣCHL	0.01	0.34	% lipid	0.08
				log weight	0.04
	ΣDDT	0.04	0.27	% lipid	0.03
				log weight	0.07
	ΣPCB	0.01	0.36	% lipid	0.11
				log weight	<<0.05
	ΣCHB	0.19	0.15	% lipid	0.11
				log weight	0.06

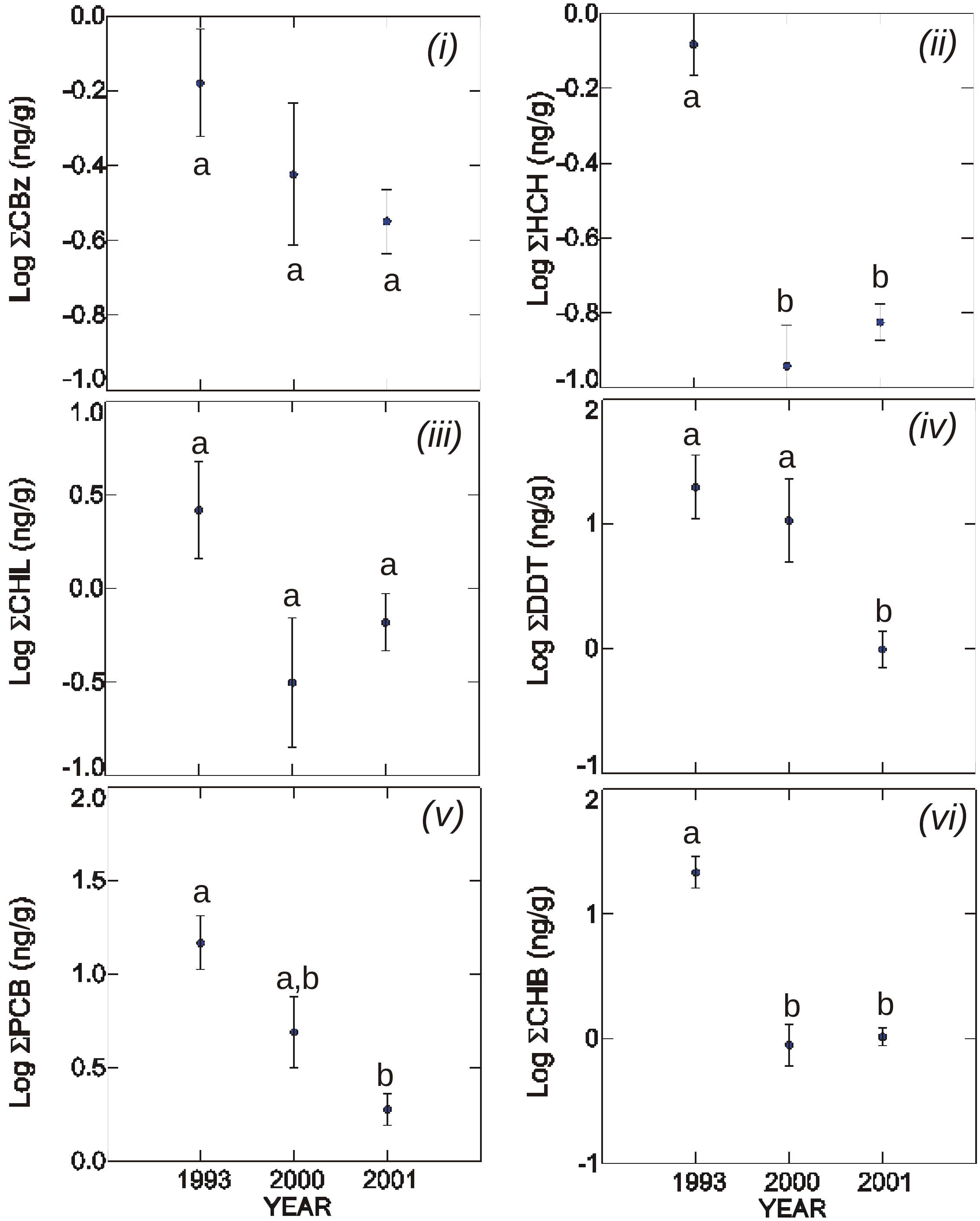


Figure 4.11 Least squares means (and SE) of (i) log ΣCBz, (ii) log ΣHCH, (iii) log ΣCHL, (iv) log ΣDDT, (v) log ΣPCB and (vi) log ΣCHB (ng/g ww) concentrations in round whitefish from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

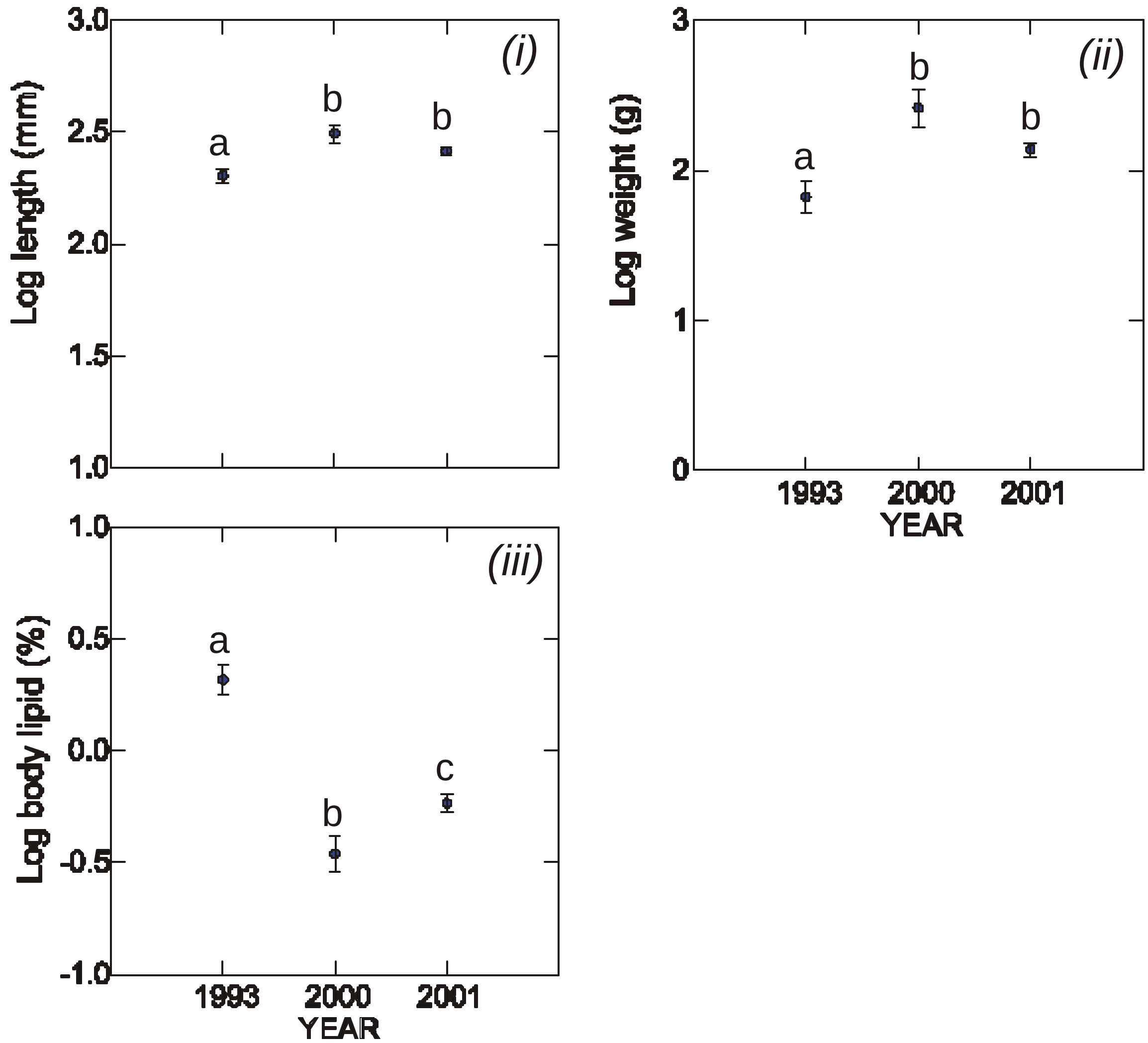


Figure 4.12 Least squares means (and SE) of (i) log length, (ii) log weight and (iii) log lipid content in round whitefish from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

4.4.1.10 Zooplankton. OC concentrations in plankton have shown considerable changes from 1993 to 2001 (Tables 4.2 and 4.3). Significant decreases (p<0.01 except for ΣPCB where p=0.02) were noted for all 6 OC groups (Figure 4.13). Levels of ΣCBz, ΣHCH, ΣCHL, ΣDDT and ΣCHB in 2001 were 14%, 16%, 10%, 26%, 13% and 17% of 1993 concentrations, respectively.

Analysis of the tissue lipid (%) content for zooplankton showed there was no significant change (p=0.41) in this parameter from 1993 (1.5%) to 2001 (1.6%). Linear regressions showed no correlation between any of the 6 OC groups and tissue lipid in plankton (p>0.05 for all groups).

4.4.2 Zooplankton Populations and Community Composition

Examination of the community composition numbers (Table 4.10) showed there was evidence of an increase in zooplankton densities, as well as a change in species composition, when comparing 1982, 1993 and 2002 vertical hauls. In the 2002 collection year, results showed the calanoid copepod *Cyclops scutifer* had decreased in abundance to account for less than half of the 1982 and 1993 community composition, while the cyclopoid copepod *Diaptomus pribilofensis* had increased in abundance and became the dominant zooplankton in Lake Laberge. Zooplankton densities were also higher in 2002 compared to 1993 by 42%. However, due to the low number of samples and lack of raw data for the 1982 collections (Section 4.3.1, Table 4.10), a direct statistical analysis was not possible. Although the sampling and enumeration methodologies were comparable for 1993 and 2002 data, sampling was not extensive so these data should be interpreted with caution.

4.4.3 Biomagnification Factors (BMF)

Biomagnification factors primarily increased in all OC categories and in most fish between 1993 and 2000-2001 (Table 4.11). The major exceptions were for lake whitefish, which only had a BMF increase in ΣCBz while the remaining 5 OC groups had decreases, and cisco, which showed only slight increases in ΣCBz and chlordane BMF but otherwise had decreases for the remaining 4 OC groups. Round whitefish BMFs were noted as having increases (ΣCBz, ΣCHL), decreases (ΣPCB, ΣCHB) and no changes (ΣHCH, ΣDDT) in BMFs across the 6 OC groups. The most consistent increases occurred in higher trophic level species including lake trout, burbot and pike.

4.4.4 Stable Isotopes and Trophic Levels

The $\delta^{15}N$ values indicated that none of the Lake Laberge species have shifted trophic levels as defined by a difference of 3-5 ‰ (Minagawa and Wada 1984; Jardine et al. 2003). The most notable variance was for northern pike (Table 4.12), which increased in $\delta^{15}N$ by 1.47 ‰, almost a half trophic level. Lake whitefish also increased by 1.16‰ and longnose sucker by 1.05‰ from 1993 to 2001, however, the small numbers of samples for some years undermined an accurate temporal trend assessment of the $\delta^{15}N$ means. The $\delta^{15}N$ appeared to increase for snails and zooplankton from 2000 to 2001, however this change was not considered significant. It is suspected the difference was caused by some variation in sample locations for snails and in processing of zooplankton (size sieving vs. non-sieved samples).

Calculated trophic levels showed a statistically significant decrease for burbot (Table 4.13) while all other species remained unchanged from 1993 to 2000-2001 (pooled).

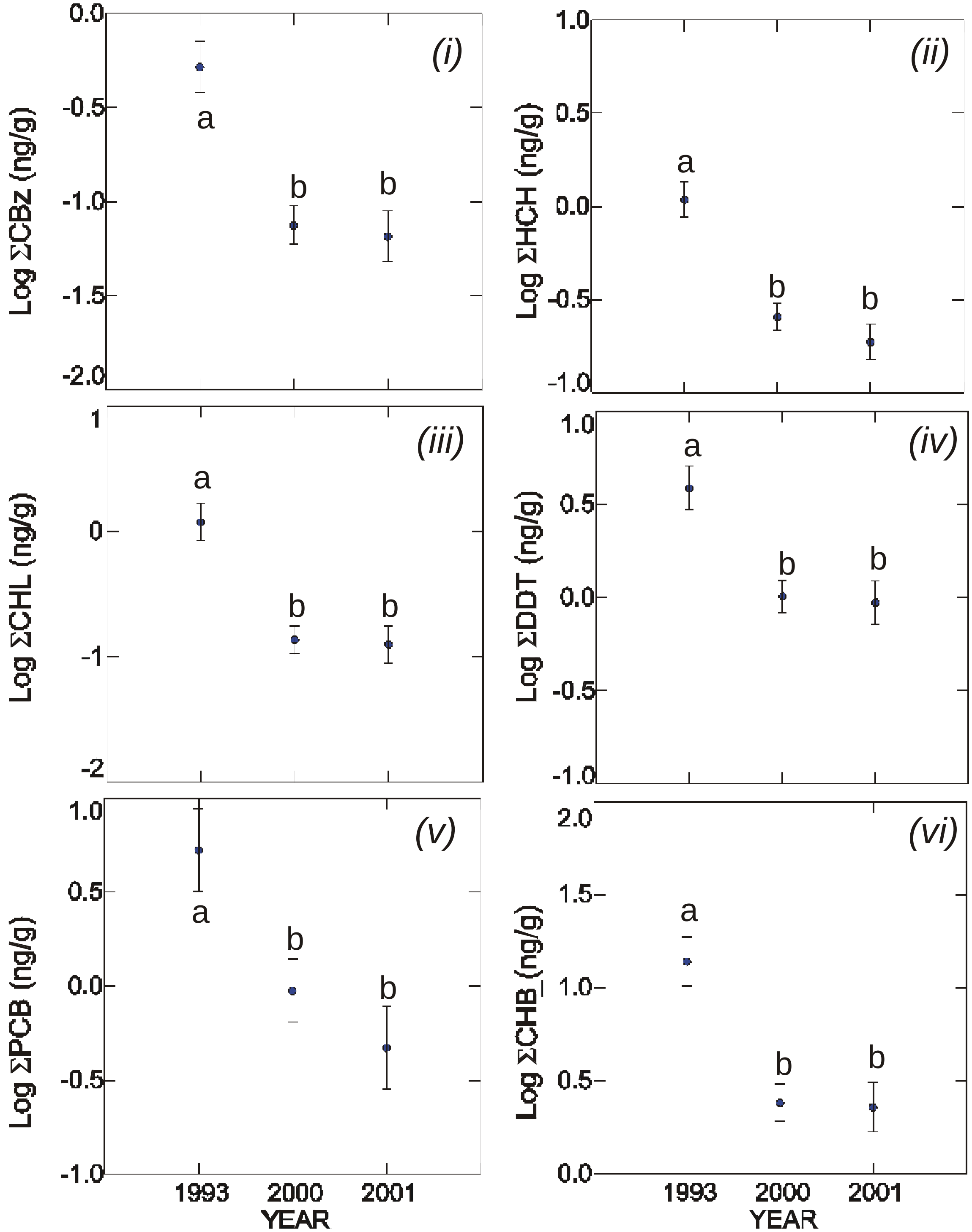


Figure 4.13 Least squares means (and SE) of (i) log ΣCBz, (ii) log ΣHCH, (iii) log ΣCHL, (iv) log ΣDDT, (v) log ΣPCB and (vi) log ΣCHB (ng/g ww) concentrations in zooplankton from Lake Laberge from 1993 to 2001. Significant differences (p<0.05) between years are shown by different letters on the graph.

Table 4.10 Total densities and community composition (as a percent of total counts) of zooplankton collected in Lake Laberge in 1982, 1993 and 2002.

Species	Body length (mm)	% of community 1982[b]	1993[c] (n=6x1)[d]	2002 (n=6x2)[d]
Cyclops scutifer	1.1-1.9[a] (0.9-1.2)[b]	84.6	80.0	35.3
Cyclopoid nauplii		0.0	0.4	0.1
Diaptomus pribilofensis	1.0-1.8[a] (0.9)[b]	9.6	0.9	46.6
Senecella calanoides	2.3[b]	0.1	0.0	0.5
Daphnia middendorffiana (also *D. pulex*)	1.3-3.0[a]	<0.1	0.0	7.4
Daphnia longiremis	0.8-1.2	4.0	18.0	6.9
Daphnia spp. (no ephippia)			0.0	2.8
Daphnia rosea		1.5		
Eubosmina longispina (also *E. coregoni*)		0.2	0.0	0.6
Total crustaceans per litre		n/a	9.73	13.89

[a] from Ward, Whipple, and Edmondson (1959) and Pennak (1989)
[b] from Lake Laberge samples (Kirkland and Gray 1986)
[c] from K. Kidd (pers. comm.)
[d] see section 4.3.1

Table 4.11 Biomagnification factors for all Lake Laberge fish for 1993 and 2000-2001 combined data. BMFs were calculated using zooplankton as the reference base for all species.

Species	OC group	1993	2000-2001
Lake trout	å **CBz**	7.3	25.7
	å **HCH**	3.7	5.0
	å **CHL**	35.2	155.3
	å **DDT**	95.8	81.7
	å **PCB**	61.6	89.2
	å **CHB**	20.7	60.4
Burbot	å **CBz**	48.1	370.9
	å **HCH**	25.7	54.9
	å **CHL**	174.0	1495.7
	å **DDT**	585.6	1868.6
	å **PCB**	241.7	977.2
	å **CHB**	160.5	420.5
Inconnu	å **CBz**		40.2
	å **HCH**		4.3
	å **CHL**		68.7
	å **DDT**		56.3
	å **PCB**		26.4
	å **CHB**		24.0
Northern pike	å **CBz**	0.3	4.5
	å **HCH**	0.1	0.3
	å **CHL**	0.7	25.9
	å **DDT**	2.1	12.3
	å **PCB**	1.4	4.9
	å **CHB**	0.7	1.2
Lake whitefish	å **CBz**	2.1	3.5
	å **HCH**	1.3	0.5
	å **CHL**	6.6	4.7
	å **DDT**	20.2	14.7
	å **PCB**	12.5	9.5
	å **CHB**	4.1	1.3

Table 4.11 cont'd Biomagnification factors for all Lake Laberge fish for 1993 and 2000-2001 combined data. BMFs were calculated using zooplankton as the reference base for all species.

Species	OC group	1993	2000-2001
Cisco	å **CBz**	2.5	3.3
	å **HCH**	1.3	0.7
	å **CHL**	4.7	5.1
	å **DDT**	7.8	6.4
	å **PCB**	4.0	2.4
	å **CHB**	4.5	1.3
Longnose sucker	å **CBz**	0.5	2.3
	å **HCH**	0.3	0.5
	å **CHL**	1.6	3.4
	å **DDT**	3.9	15.0
	å **PCB**	2.6	9.0
	å **CHB**	1.2	0.5
Round whitefish	å **CBz**	1.5	5.3
	å **HCH**	0.7	0.7
	å **CHL**	2.4	16.3
	å **DDT**	5.5	5.1
	å **PCB**	3.3	2.2
	å **CHB**	1.7	0.5
Broad whitefish	å **CBz**		0.8
	å **HCH**		0.4
	å **CHL**		0.5
	å **DDT**		2.8
	å **PCB**		0.3
	å **CHB**		0.1

Table 4.12 Stable nitrogen isotope ratios ($\delta^{15}N$, ‰) by species or invertebrate family over the sampling period.

	1993*			2000			2001		
Species	**n**	**Mean**	**SD**	**n**	**Mean**	**SD**	**n**	**Mean**	**SD**
Lake trout	34	12.38	0.84	6	13.49	0.33	17	13.18	0.39
Burbot	32	12.06	0.59	18	12.34	0.41	30	12.41	0.41
Inconnu				2	11.48	0.35	6	11.73	0.33
Northern pike	10	9.62	0.75	5	10.83	0.52	5	11.09	0.21
Lake whitefish	36	8.22	1.15	15	9.27	0.45	13	9.38	0.71
Cisco	14	7.87	0.47	22	8.68	0.39	2	8.99	0.61
Arctic grayling							1	8.97	
Longnose sucker	12	7.57	0.92	14	8.15	0.56	6	8.62	0.75
Round whitefish	8	7.16	0.76	6	8.05	0.87	20	7.65	0.85
Broad whitefish	3	6.55	1.10	7	7.85	0.44			
Tricoptera	9	2.13	0.91				3	5.18	1.02
Chironomids	8	4.02	1.14				1	4.20	
Snails (Lymnaeidae)	6	2.05	1.01	17	1.96	1.36	18	3.21	0.56
Snails (Planorbidae & Valvatidae)	4	1.26	1.39						
Clams (Sphaeridae)	2	3.60	1.99	1	3.11		1	2.48	
Dipterans (other than chironomids)							3	2.18	0.38
Ephemeroptera							1	1.65	
Zooplankton	6	4.94	1.22	9	4.95	0.69	5	5.97	0.11

* some data included from Kidd (1996)

Table 4.13 Calculated trophic levels for fish from Lake Laberge collected in 1993 and 2000-2001 and ANOVA result.

	1993*			2000-2001			
Species	**n**	**Mean**	**SE**	**n**	**Mean**	**SE**	***p***
Lake trout	44	5.20	0.04	23	5.13	0.02	0.32
Burbot	32	5.07	0.03	48	4.88	0.02	<0.05
Inconnu				8	4.67	0.04	
Northern pike	10	4.32	0.07	10	4.47	0.04	0.08
Lake whitefish	36	3.91	0.06	28	3.98	0.03	0.35
Cisco	13	3.81	0.04	24	3.80	0.02	0.76
Longnose sucker	12	3.72	0.08	20	3.64	0.04	0.61
Round whitefish	8	3.60	0.08	26	3.51	0.05	0.4
Broad whitefish	3	3.42	0.19	7	3.54	0.05	0.39

Regression plots of the log concentration of the 6 OC groups with $\delta^{15}N$ for all biotic samples demonstrated an increase in the slope (rate of food web magnification (FWMF)) for OC categories for 2000-2001 pooled data in comparison with 1993 samples (Figure 4.14, Table 4.14). GLM results show that all regressions of OC with $\delta^{15}N$ for both 1993 and 2000-2001 were highly significant (p<<0.01) while the results of the ANCOVA shows that slopes of the regressions lines are significantly different (all results p<0.01 except for HCH where p=0.01; Table 4.14). No significant differences in slopes or intercepts were found between 2000 and 2001 biota data prior to pooling.

Tissue $\delta^{13}C$ displayed a relatively high amount of variance between years but only 2 species had significant trends over time (Table 4.15). Burbot were slightly more depleted in ^{13}C (p=0.003) with a difference of 0.52‰ from 1993 to 2001. Lake whitefish were more enriched in ^{13}C with a significant increase of 1.73‰ (p=0.001) between 1993 and 2001. Longnose sucker also appeared marginally less depleted (0.85‰ difference), but this was not significant (p=0.11). Differences in the $\delta^{13}C$ of snails were likely due to a variation in the collection area between 2000 and 2001 (B. Burns, pers. comm.).

Plots of $\delta^{15}N$ and tissue lipid, as well as $\delta^{15}N$ and weight for all fish species and temporal points combined, demonstrated there was no correlation between trophic level and these two parameters (data not shown, p<0.01, r^2=0.37 and 0.31, respectively).

4.4.5 Fish Populations

Results of fish population surveys in 1991 and 1999 showed a distinct increase in the catch per unit effort (CPUE) of 6 out of 9 species including lake trout, broad whitefish, northern pike, inconnu, longnose sucker and least cisco (Figure 4.15 and 4.16). Two notable changes include increased CPUE factors for lake trout (top predator) and least cisco (primary prey of piscivores) of 2.1 and 3.8 fold, respectively. It should be noted that burbot were excluded from these CPUE analyses as the small mesh netting method was not designed for the capture of this species and results may be highly variable (A. Foos, pers. comm.). Both of the CPUE values (# and kg caught per gillnet hour) increased for each of the previously mentioned species.

Decreases in CPUE values (# and kg per gillnet hour) occurred for both arctic grayling (data not shown) and round whitefish, while the # caught per gillnet/hour for lake whitefish decreased and kg/gillnet hour increased. This indicated that fewer fish were captured in 1999 compared to 1991, however their total weight in 1999 was greater than the total weight captured in 1991. These data were standardized for fishing effort (A. Foos. pers. comm.). In contrast with the results in Table 4.4, the morphological data obtained by the Yukon Territorial Government (YTG) indicates that mean weights of lake whitefish may have increased from 371 g to 436 g (n=255 and 446 for 1991 and 1999, respectively) although YTG has not analysed this data statistically and these averages fall well within the ranges presented in Table 4.1 (Foos 2001)

Table 4.14 Slopes and intercepts (± SE) from OC concentrations (ng/g ww) and $\delta^{15}N$ regression models for 1993 and 2000-2001 (pooled) biotic samples from Lake Laberge.

	1993			2000-2001			
	Slope (m)	**Intercept (b)**	**r^2**	**Slope (m)**	**Intercept (b)**	**r^2**	***p***
å **CBz**	0.17 (0.02)	-1.52 (0.22)	0.40	0.30 (0.02)	-3.07 (0.20)	0.56	<0.01
å **HCH**	0.16 (0.02)	-1.31 (0.24)	0.31	0.25 (0.02)	-2.81 (0.21)	0.44	0.01
å **CHL**	0.22 (0.02)	-1.11 (0.24)	0.47	0.43 (0.02)	-3.80 (0.23)	0.67	<0.01
å **DDT**	0.23 (0.03)	-0.31 (0.26)	0.45	0.42 (0.02)	-2.73 (0.22)	0.68	<0.01
å **PCB**	0.21 (0.02)	-0.23 (0.24)	0.44	0.41 (0.02)	-2.77 (0.21)	0.68	<0.01
å **CHB**	0.23 (0.02)	-0.27 (0.24)	0.48	0.44 (0.02)	-3.32 (0.25)	0.64	<0.01

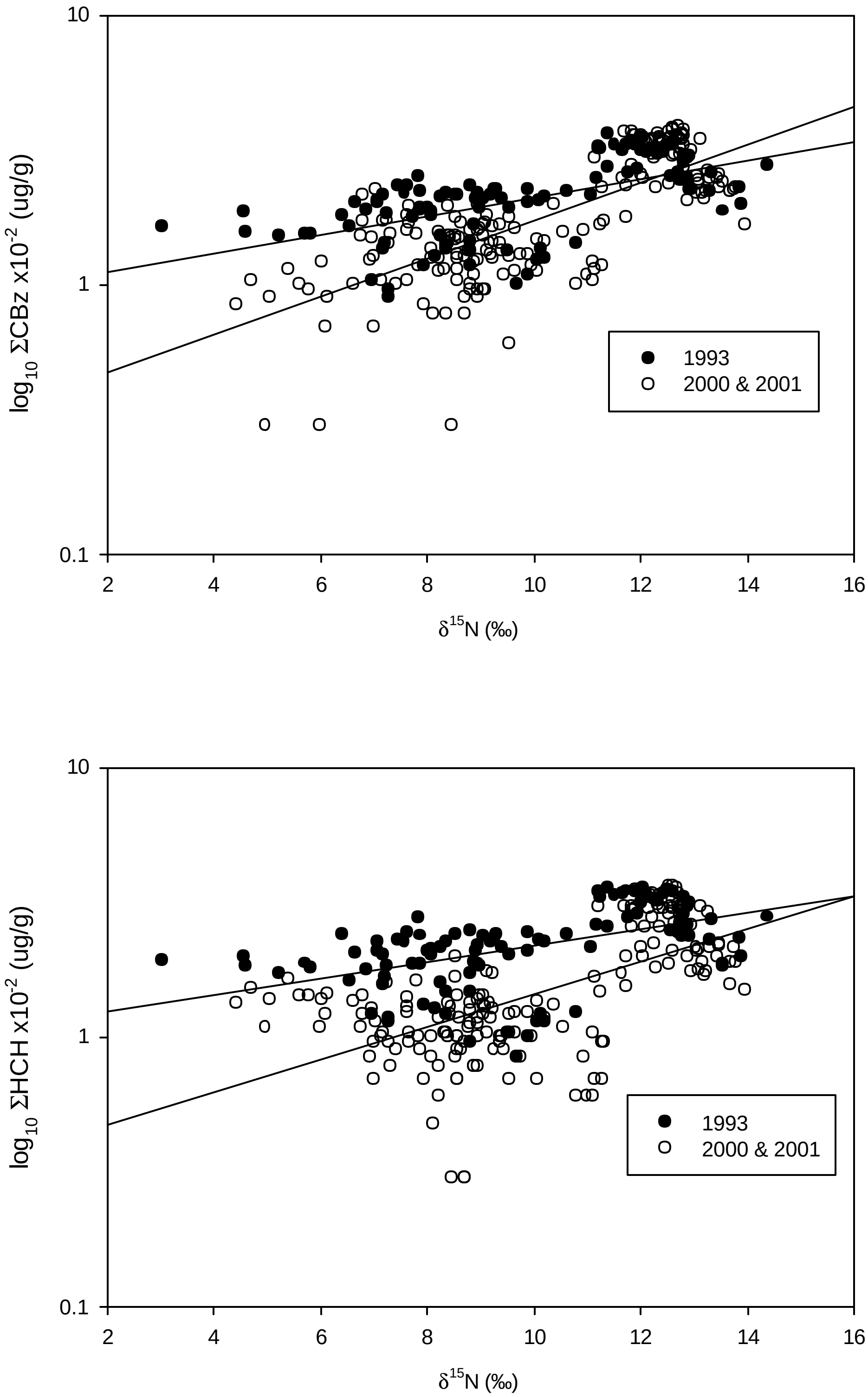
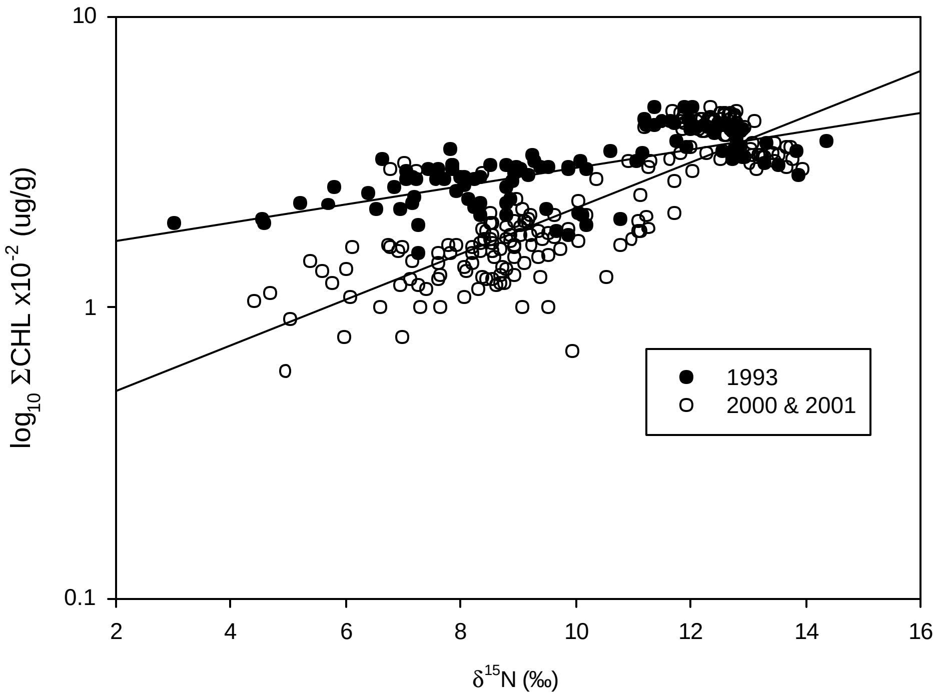


Figure 4.14. Regressions of organochlorine concentrations (μg/g ww) versus δ^{15}N (‰) for all biotic samples from Lake Laberge for 1993 and 2000-2001.



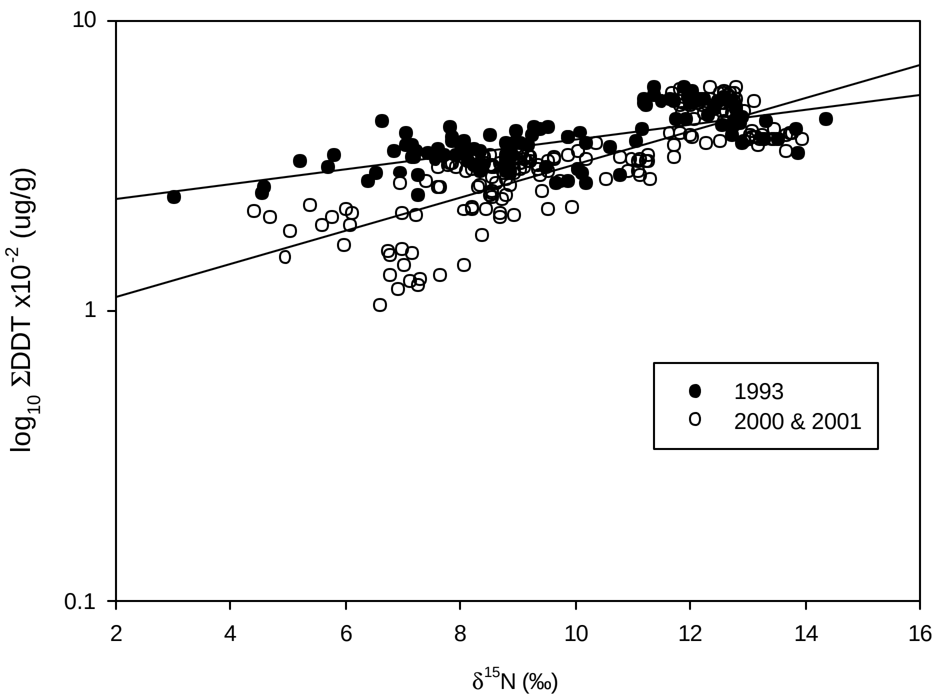
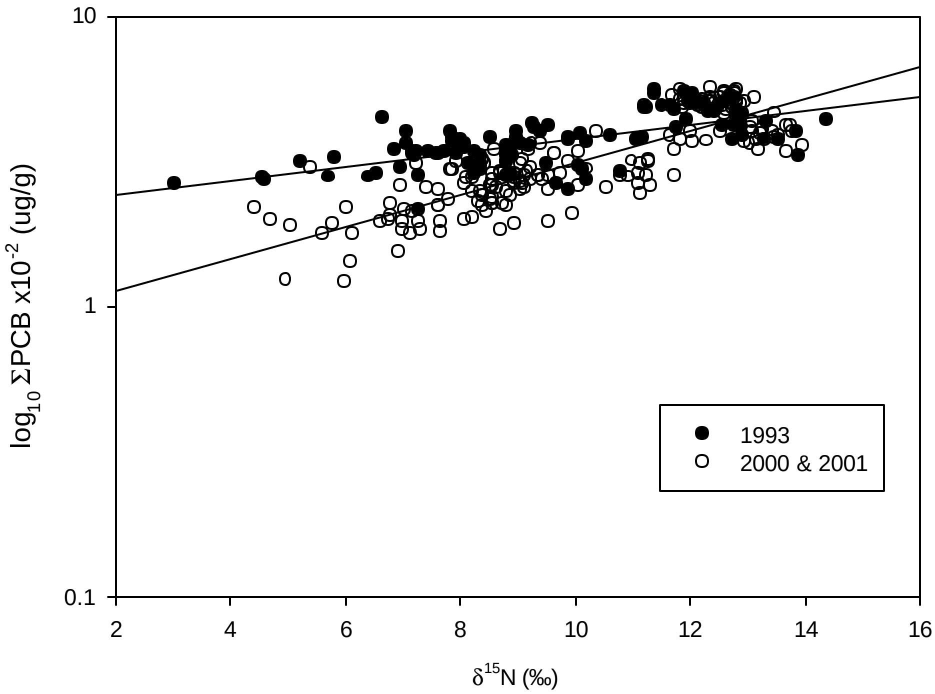


Figure 4.14 cont'd. Regressions of organochlorine concentrations (μg/g ww) versus δ^{15}N (‰) for all biotic samples from Lake Laberge for 1993 and 2000-2001.



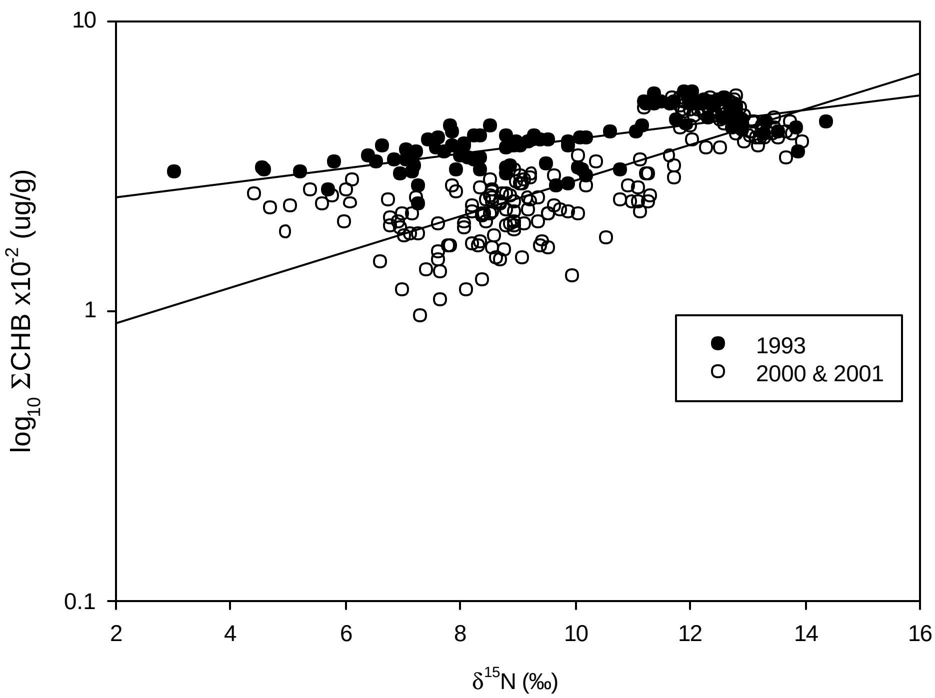


Figure 4.14 cont'd. Regressions of organochlorine concentrations (μg/g ww) versus δ^{15}N (‰) for all biotic samples from Lake Laberge for 1993 and 2000-2001.

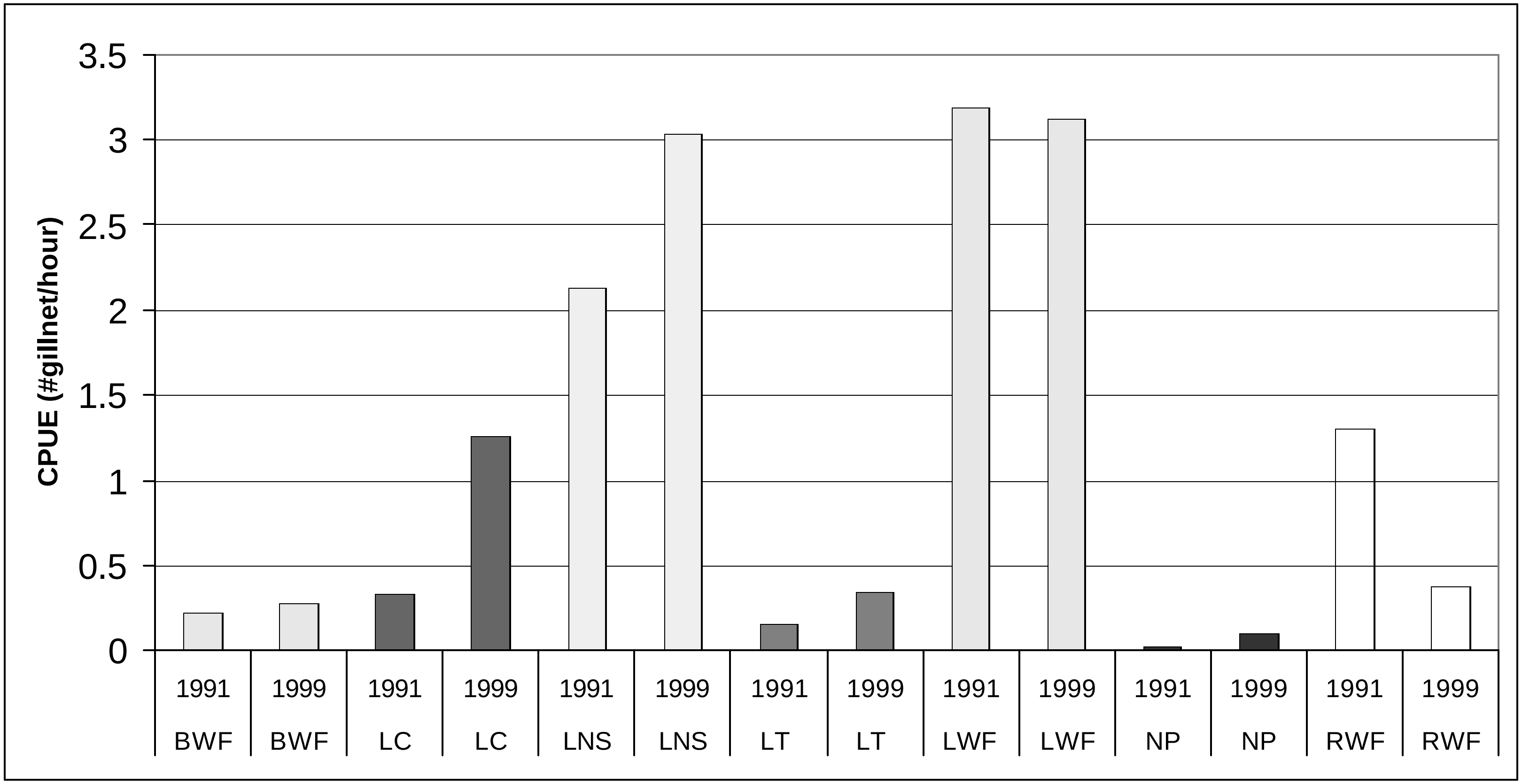


Figure 4.15. Catch per unit effort (# in gillnet per hour) for selected fish from Lake Laberge netted in 1991 and 1999 small mesh net surveys (data courtesy of A. Foos, Yukon Territorial Government). BWF- broad whitefish, LC- least cisco, LNS- longnose sucker, LT- lake trout, LWF- lake whitefish, NP- northern pike, RWF- round whitefish.

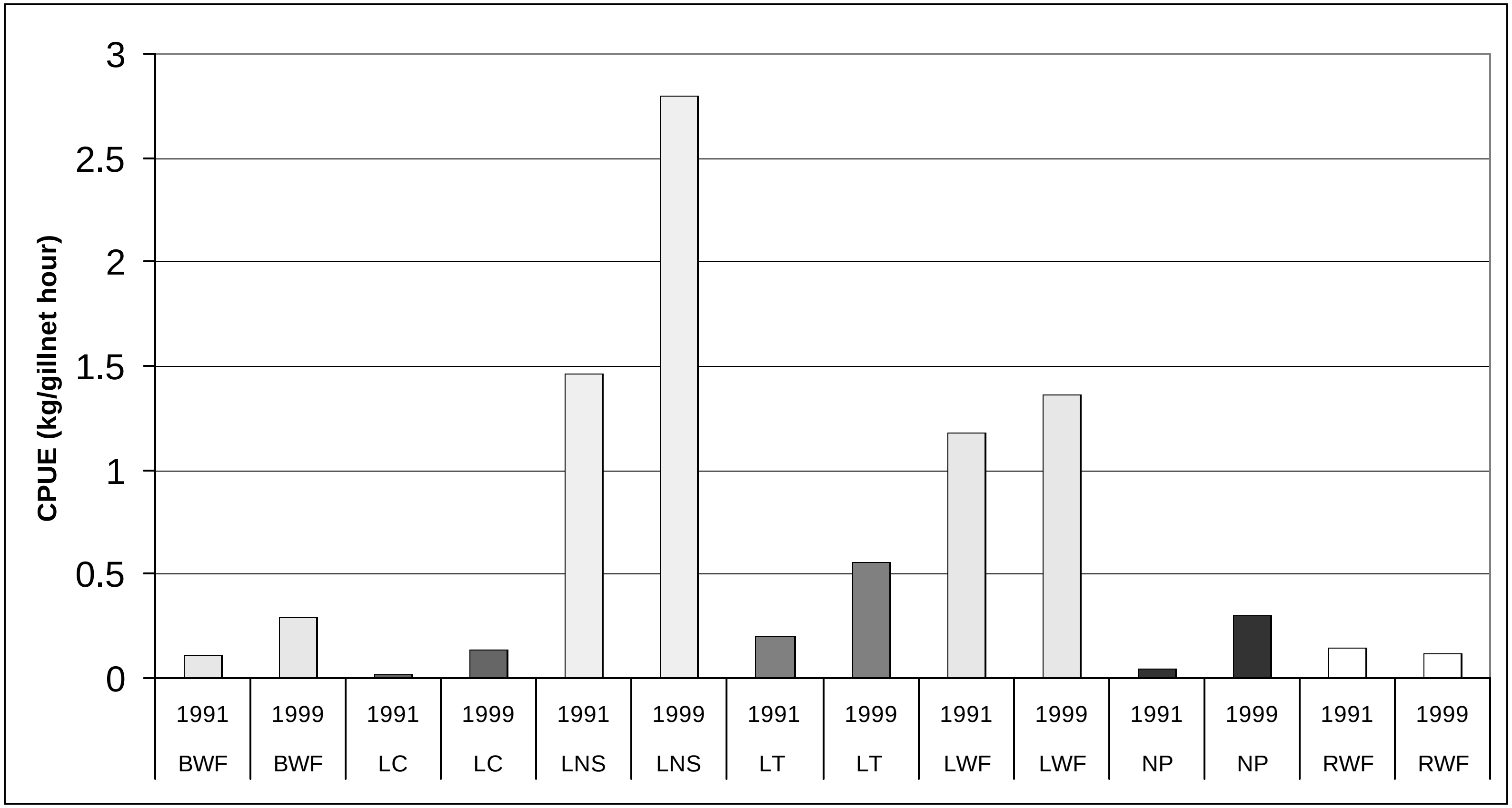


Figure 4.16. Catch per unit effort (kgs fish caught in gillnet per hour) for selected fish from Lake Laberge netted in 1991 and 1999 small mesh net surveys (data courtesy of A. Foos, Yukon Territorial Government). BWF- broad whitefish, LC- least cisco, LNS- longnose sucker, LT- lake trout, LWF- lake whitefish, NP- northern pike, RWF- round whitefish.

Table 4.15 Stable carbon isotope ratios ($\delta^{13}C$, ‰) by species or invertebrate family over the sampling period.

	1993*			2000			2001		
Species	**n**	**Mean**	**SD**	**n**	**Mean**	**SD**	**n**	**Mean**	**SD**
Lake trout	32	-29.03	1.75	6	-29.51	2.36	17	-28.21	1.39
Burbot	32	-26.89	0.73	18	-27.45	0.80	30	-27.41	0.47
Inconnu				2	-26.07	0.09	6	-27.40	0.61
Northern pike	10	-24.64	0.45	5	-24.71	0.71	5	-25.34	0.73
Lake whitefish	36	-26.42	1.56	15	-25.04	1.37	13	-24.69	1.41
Cisco	12	-29.29	3.02	22	-29.65	0.37	2	-29.95	0.29
Arctic grayling							1	-23.23	n/a
Longnose sucker	12	-22.03	1.08	14	-21.39	0.83	6	-21.18	0.54
Round whitefish	9	-23.81	2.03	6	-24.36	2.64	20	-25.25	3.17
Broad whitefish	3	-25.14	3.92	7	-21.75	1.17			
Tricoptera	9	-24.37	0.93				3	-32.61	3.29
Chironomids	8	-22.85	0.63				1	-22.54	
Snails (Lymnaeidae)	7	-23.63	0.67	17	-20.38	2.78	18	-24.27	1.69
Snails (Planorbidae & Valvatidae)	4	-26.30	2.99						
Clams (Sphaeridae)	3	-22.61	4.53	1	-27.69		1	-28.32	
Dipterans (other than chironomids)							3	-25.20	2.94
Ephemeroptera							1	-29.58	
Zooplankton	6	-33.31	0.93	9	-33.03	1.26	5	-33.17	0.22

* some data included from Kidd (1996)

4.5 Discussion

The concentrations of six organochlorine groups, chlordane (CHL), DDT, hexachlorocyclohexane (HCH), toxaphene (CHB), PCB and chlorinated benzenes (CBz), have decreased significantly in zooplankton and six species of Lake Laberge fish from 1993 to 2003. This occurrence contrasts recent research on organochlorine levels in Great Lakes fish which showed contaminant concentrations had stabilized and were no longer undergoing rapid declines as seen in the 1970s and 80s (Borgmann and Whittle 1991; Stow et al. 1995). The purpose of our research was to investigate the causes behind the temporal decline of contaminants in the Lake Laberge ecosystem. This chapter has focused on specific morphological and population variables as well as food web dynamics that typically affect contaminant concentrations in fish.

4.5.1 Growth Dilution and OC Contaminants

Organochlorine concentrations in the food chain of Lake Laberge have shown considerable decreases from 1993 to 2001 (up to 2003 for lake trout). Major changes in the morphology of some species have coincided with this occurrence and have likely affected part of this change in OC concentrations. The results in Chapter 3 showed that there were correlations of body weight with contaminant concentrations for lake trout, while the results of Chapter 4 also demonstrated a similar correlation exists with some forage fish lower in the food chain (cisco, lake whitefish and round whitefish). Other researchers have also concluded that growth rates have a major influence on contaminants, describing an increase in body size with decreasing contaminant levels as ‘growth dilution’ (Borgmann and Whittle 1991; Sijm et al. 1992; Larsson et al. 1992; Hammar et al. 1993; Hebert et al. 1997; Kidd et al. 1998).

Models of growth dilution using field data demonstrated that the effect of growth rate on bioaccumulation was greater than the lipid-contaminant relationship indicating that the concentration of bioaccumulating contaminants in a fish body was affected by more than just a lipid-partitioning mechanism (Thomann 1989; Borgmann and Whittle 1991). This conclusion is supported by other research (Larsson et al. 1991; Hammar et al. 1993; Stow et al. 1995). Contaminant burdens were related to the bioenergetics of the organisms (food conversion efficiency; Sijm et al. 1992) as well as the assimilation efficiency and excretion rate of organochlorine chemicals in fish (Olsson et al. 2000; Ruus et al. 2002). Clark and Mackay (1991) noted that an increase in the concentration of chlorinated hydrocarbons in the food created a proportional increase in the concentrations of the contaminants in the test fish while an increase in feeding rates did not create a similar proportional contaminant increase. The rationale is that although a higher food intake results in more food material ingested, the material spends less time being digested. Hence the consumed food moves faster through the body resulting in decreased uptake of OC from that material (Sijm et al. 1992). Alternatively, any increase in the assimilation of nutrients (possibly due to higher food intake) during the life cycle of a fish, may provide a greater capacity for biotransformation and hence elimination of contaminants (Sijm et al. 1992). OC concentration trends in Great Lakes lake trout were tied to fluctuations in the growth of alewives, and their varying diet, suggesting that food web interactions regarding food intake play a strong role in contaminant regulation (Borgmann and Whittle 1991; Madenjian et al. 1999).

Based on the morphological changes (mass, lengths, K) observed over time, growth dilution may be occurring with Lake Laberge lake trout, burbot, longnose sucker, cisco and round whitefish. Results from a YTG fish population survey for Lake Laberge (Foos 2001) has provided evidence of recent increases in fish populations for most species. A higher abundance of prey should result in a higher food intake for predatory species in Lake Laberge such as lake trout and burbot.

Research has shown that an increase in fish growth rates is elicited by an increase in prey size although not by increases in prey numbers (Kerr 1971b; Matuszek et al. 1990; Trippel and Beamish 1993; Pazzia et al. 2002). These studies suggest that growth dilution may be occurring for lake trout because their main prey has increased in size. The diet of Laberge lake trout consists primarily of least cisco (Kidd 1996). Cisco have increased in body size and numbers concurrently with lake trout from 1993 to 2001. Coinciding with this increase in lake trout prey size is a decrease in OC concentrations in both cisco and lake trout. There is also evidence of a strong inverse relationship of OC concentrations with lake trout mass (Chapter 3). Hence growth dilution may be occurring in the lake trout if they are growing at faster rates in more recent years compared to the earlier sampled fish. An examination of several lake trout otoliths from Lake Laberge revealed that the fish had no indications of abnormal growth patterns in 1993 versus 2001 samples (K. Mills, pers.comm.; data not shown). There were no indications of "growth stanzas", which are characterized by periods of slow growth followed by accelerated growth indicating that something changed in the food consumption of fish during their life times. However, the more recent fish (2000-2001) had a normal pattern of growth with a faster first year compared to 1993 samples, which could indicate the increased growth rates of lake trout begins at the young-of-the-year (YOY), planktivorous stage.

Although growth rates were not calculated (for lake trout or other fish), similar inferences about growth dilution of contaminants can also be made for round whitefish based on their changes in size to larger animals from the 1993 to 2001 samples and on the GLMR correlations of body weight with OC concentrations. Least cisco, although increasing in lengths, had only a marginal increase in weight while GLMR showed that neither lipid nor weight was a significant factor affecting the OC concentrations in this fish for 2000-2001. Both of these variables were significant predictors of OC concentrations for 1993 samples indicating a change in the influential parameters on contaminants had occurred over time for this species. Cisco OC concentrations have decreased more likely due to the decrease in contaminant concentrations of their prey, zooplankton.

Burbot have had some OC decreases (ΣDDT, ΣCHB, ΣHCH) over time, while other OC levels have not significantly changed. Burbot have shown increases in their mean age that may partly explain the contaminant trends. The increase in mean ages may be offsetting some of the decreased trends for the same OC categories seen in other species. Age is a variable known to correlate to OC concentrations in burbot (Kidd 1996). It is more probable that decreased lipid content, rather than growth dilution, is the primary cause for the various OC declines in this species.

Longnose suckers from Lake Laberge also showed evidence of declines in some OC concentrations (ΣCHB, ΣHCH, ΣCHL) over time. The 2000-2001 samples were observed to be younger in age and marginally smaller in size compared to 1993 fish. This may account for some of the general decreases in OC concentrations since smaller, younger fish typically have lower OC levels (Sijm et al. 1992). Longnose suckers are also benthic feeders (McPhail and Lindsey 1970) and without further analysis of their feeding rates or enumerations of their primary benthic prey, conclusions about sucker growth rates are limited. There are no data available on changes in the benthic invertebrates, and limited data for zooplankton, so it is only speculation that the food abundances have increased for insectivorous or planktivorous fishes in Lake Laberge.

The interspecies differences in contaminants (concentrations and trends) are expected as contaminant accumulation relates to the varying habitat and feeding behaviours specific to each species (Hebert and Haffner 1991). There is no evidence to support the hypothesis of growth dilution for burbot, cisco, lake whitefish or longnose sucker. Other rationales, such as lower OC concentrations in their prey, will also influence the trends in contaminants and will be reviewed. It is suspected that the theory of growth dilution is occurring primarily for lake trout and round whitefish in Lake Laberge.

OC concentrations in zooplankton collected in 2001 from Lake Laberge have declined to less than 30% of the 1993 levels. Lower OC concentrations in zooplankton are likely due to decreased contaminant levels in primary producers (phytoplankton). This decrease may also be caused by growth dilution. Research has shown that OC concentrations and bioconcentration factors in plankton have an inverse relationship with plankton biomass and cell densities (Lederman and Rhee 1982; Taylor et al. 1991). Although zooplankton OC levels do correlate with water OC concentrations, the process of equilibration is not particularly fast, and seasonal variables, such as light and water temperatures, are highly influential on growth (Swackhamer and Skoglund 1993; Smith 1995). The rate of zooplankton biomass growth and death may exceed the rates of contaminant equilibration with the environment such that plankton communities maintain diluted OC concentrations per weight of mass (Swackhamer and Skoglund 1993; Smith 1995) and OC concentrations in water become a weak predictor of levels in plankton. This dilution effect provides planktivorous predators with a food base that is less contaminated (Smith 1995). An assessment of zooplankton from our study has shown an increase in populations with counts higher in 2001 samples compared to 1993. Higher zooplankton numbers suggest either an increased forage base or decreased predation. In this study, we did not specifically examine changes in phytoplankton, although it may be possible to do this through the analysis of sediment cores (Gajewski et al. 1997; Stern et al. 2005; Chapter 5).

4.5.2 Lipids and Biomagnification Factors (BMF)

Lipid content is known to be a major predictor of OC in fish (Kidd et al. 1998) as OC concentrations are positively related to lipid content in the tissues

(Rasmussen et al. 1990; Larsson et al. 1991; Rowan and Rasmussen 1992; Kucklick et al. 1996). Lake Laberge lake whitefish exhibited no temporal changes in morphology but still registered a significant decrease in OC that was correlated with changes in lipid content (Results 4.4.1.5). Decreases in lipid body contents of lake trout, burbot, round whitefish, and longnose sucker have also been correlated to decreases in OC concentrations over the study period (Chapter 3 and Section 4.4.1).

The decline in tissue lipid contents for fish in Lake Laberge may be due to changes in foraging behaviour (e.g. intensity), dietary habits, prey lipid and increased reproduction. Considering the recent increases in fish populations and zooplankton densities, foraging behaviours may have been altered from previous years. For example, Laberge lake trout may spend less energy and time foraging as more cisco are in the lake, whereas elevated fish foraging behaviour decreases growth rates from escalated energy use due to the lengthier time spent searching for prey (Kerr 1971a; Hebert et al. 1997; Pazzia et al. 2002). Since the lake trout prey (cisco) in Lake Laberge were also larger, fewer cisco need to be ingested to maintain a higher rate of growth (Kerr 1971c; Matuszek et al. 1990), but because the primary prey selection has not changed, deviations in $\delta^{15}N$ would not be expected, as observed in Lake Laberge fish. Population data have shown higher abundances of top predators and some forage fish. The lipid content in prey or forage fish may decrease due to an increase energy utilization in the higher numbers of predators. Lipid levels may also be affected with an increase in intraspecific competition for spawning and the effects of reproduction or related behaviour on OC concentrations is specific to each species (Larsson et al. 1993). Alternatively, since OC can be passed along to young via reproductive routes (Larsson et al. 1993; Fahraeus-Van Ree and Payne 1997; Delorme et al. 1999), highly successful reproducing fish will lose OC, as well as lipids, to offspring if reproduction is occurring more often or more intensively, causing a decrease in temporal OC concentrations as observed in Lake Laberge. Reproductive loss and foraging energetics, both of which affect lipid body contents, are likely influencing contaminant loads in all fish in Lake Laberge at varying magnitudes. Least cisco is the only species for which GLMR have shown that both body size and lipid are not significant factors affecting the temporal trends of OC as previously mentioned. It is suspected that the decreased OC levels in cisco must be due to the decreases in OC concentrations measured in zooplankton, its primary prey. Tissue lipid content may not account for the entire decrease in OC concentrations in some Lake Laberge biota, but it is a more likely factor for fish that are not exhibiting patterns typical of growth dilution (e.g. lake whitefish).

The lipid contents measured in zooplankton from 1993 to 2001 remained unchanged, yet the OC concentrations dropped by more than 70% for all 6 OC groups in this period. Since the decrease in OC levels cannot be linked to the lipid content of zooplankton, other factors must be responsible such as a community composition change or a reduction in prey OC concentrations.

Biomagnification and bioaccumulation are affected by trophic level, diet and tissue lipid content (Rasmussen et al. 1990; Kidd et al. 1995b; Zaranko et al. 1997; Kidd et al. 1998). Both tissue lipid and trophic level are positively related to OC concentrations, as biomagnification is greater for fish higher in the food chain. With the general decrease in lipids for most fish sampled from Lake Laberge from 1993 to 2001, OC levels were expected to decline and hence the BMF of contaminants should follow a similar pattern, but lipid is not the only factor affecting biomagnification (Kidd et al. 1995b). BMF increased for lake trout, burbot and pike despite decreases in tissue lipids. The analysis of trophic levels failed to show any evidence to support the increases in BMF and the calculated trophic level for burbot even had indications of a decline. Part of the higher BMF was likely due to the greater magnitude of declines in zooplankton OC concentrations in comparison to the declines in lake trout, burbot and pike from 1993 to 2001 (e.g. faster kinetics of declines in zooplankton). In contrast, the forage fish (lake whitefish, round whitefish, cisco and longnose sucker) all have inconsistent trends or decreasing trends in BMF. There is a greater degree of OC decline (when related to concentrations in zooplankton) in these species from 1993 to 2001 compared to the higher trophic level fish. BMFs have been noted to be higher in larger bodied animals (Fisk et al. 2001). The effects from decreases in lipid levels and changes in feeding behaviour on BMF are also applicable to species of fish lower in the food chain.

The OC-δ^{15}N regression graphs demonstrated that the food web magnification (FWMF) had increased significantly between 1993 and 2000-2001 for all OC groups in aquatic biota from Lake Laberge. The FWMF were less than values reported in marine systems but these values are not directly comparable because of differences in calculations (Jarman et al. 1996; Fisk et al. 2001). The

FWMF in Lake Laberge doubled for all OC (except HCH which increased but did not double), indicating there was a faster rate of OC magnification along the food chain for 2000-2001 data. The lower FMWF for ΣHCH was expected due its lower lipophilicity (K_{ow}) compared to the other five OC groups (Kidd et al. 1995a). The FWMF increases may be caused by factors specific to the physiology or behaviour of each species or because of the lower contaminant levels found in zooplankton or forage fish. Feeding behaviour and diet can have a significant effect on bioaccumulation rates. For example, two groups of lake trout from Lake Michigan had distinctly different bioaccumulation rates of PCB (Miller et al. 1992). The lower PCB bioaccumulation rate in deep water trout was attributed to their consumption of larger, higher caloric content fish (Miller et al. 1992). As prey size increases, predators may also receive the offsetting benefit of growth dilution. Increases in FWMF in Lake Laberge biota may be due to the more populous prey items available for pike, burbot and lake trout. As fish feeding rates increase, their BMFs are expected to increase linearly which could elevate the FWMF; however this only goes to a point where OC uptake efficiency declines with increased food consumption (Sijm et al. 1992). The comparisons of FWMF between 1993 and 2000-2001 have shown increases for all 6 OC groups. The increased BMF for piscivores and greater magnitude of OC declines in organisms lower in the food chain are the likely factors influencing this trend.

4.5.3 Populations and Food Web Dynamics

Exploitation, addition or removal of a fish species has had historical impacts on fish growth rates (Hewson 1955; Healey 1975; Healey 1978; Mills et al. 2000), body size (Healey 1975), fecundity (Munkittrick and Dixon 1989), maturation ages (Healey 1975; Healey 1978; Munkittrick and Dixon 1989) and prey structure (Mills and Forney 1983; Trippel and Beamish 1993; McNaught et al. 1999) in aquatic ecosystems. It was expected that changes in such population characteristics would occur following the closure of the century long commercial fishery on Lake Laberge. Changes to the status of lake trout are known to have significant impacts on the food web community structure due to changes in fish predation (Trippel and Beamish 1993; Northcote 1988; Merrick et al. 1992; Wetzel 2001) although one study has shown that changes in lake trout abundance had no detectable effect on burbot numbers (Carl 1992). Trout stocking of an alpine lake did result in a change in the plankton community towards a heavier dominance by a smaller *Diaptomus* and smaller phytoplankton species over larger crustaceans (McNaught et al. 1999). Lake trout have also influenced the size characteristics of slimy sculpin as an increase in sculpin size coincided with the decline in lake trout predation (Mcdonald and Hershey 1992). Both lake trout and round whitefish have been shown to affect the distribution, size and density patterns of molluscs in an Alaskan lake through size selective predation (Merrick et al. 1992). The recent fluctuation in fish populations in Lake Laberge is likely influencing the body size characteristics of some prey fish and zooplankton thus shifting the community composition or species characteristics observed in Lake Laberge.

The zooplankton structure in Lake Laberge has changed over time likely due to a shift in production, community composition or in the timing of seasonal succession. Densities of zooplankton have increased and there is a shift from the dominance of *Cyclops scutifer* in 1982 (85%) and 1993 (80%) towards dominance by *Diaptomus pribilofensis* in 2002 (47%). The shift could be indicative of a long-term change caused by several dry and above average temperature summers (1981-1984, 1994-1997; Chapter 5) in the Yukon region (Environment Canada 2002). Temporal cycles of *Cyclops* populations have been noted for Alaskan lakes, while *Diaptomus* did not exhibit similar patterns over the same period (Edmundson et al. 2003). Therefore it is possible that the observations of community shifts in Lake Laberge may just be part of the natural life cycle for these zooplankton species and are unrelated to climate changes. Climate change effects on plankton were also studied in the Experimental Lakes Area in Ontario, Canada. A rise in temperatures (2 ^{o}C) in 1980s compared to other decades, resulted in both increased phytoplankton biomass and number of species, along with a shift in the community species composition, during drought years (Findlay et al. 2001).

Zooplankton and phytoplankton communities change over the course of a season in the process known as 'succession', which ultimately affects other organisms in an aquatic food web. Succession occurs due to shifts in water nutrients, temperatures and light periods, and as such, is largely moderated by climate (Wetzel 2001). Abnormal variations in climate can easily offset a pattern in succession as shown by Edmundson et al. (2003), who concluded that the depth of the euphotic zone in an Alaskan lake decreased due to an increase in glacial runoff and silt loadings during dry, hot years, resulting in a significant decrease in planktivorous copepod biomass and subsequent increases in numbers of primary producers. Changes in a seasonal pattern over extended periods of time may result in shifts of the succession times and ultimately a shift in the habits or growth potential of planktivorous predators. Fish production is known to closely correlate with primary production (Downing et al. 1990) and even non-planktivorous species such as lake trout have been related to zooplankton densities (O'Brien et al. 2004). For example, an increase in the availability of *Diaptomus pribilofensis* was shown to account for growth differential in YOY lake trout from inshore and offshore habitats, between two Arctic lakes (Mcdonald et al. 1992) and it was suggested that *Diaptomus* might be an important supplemental food resource for salmonids in years when cyclopoid copepods such as *Cyclops scutifer* are in lesser abundance (Edmundson et al. 2003). The addition of *Mysis relicta* to a Montana lake resulted in dramatic increases in the lake trout populations through improved survival of YOY (Stafford et al. 2002). Other research concluded that copepod biomass in spring, along with the abundance of fish spawners, is directly related to the fall fry salmonid production and strongly influences recruitment success in the next generation (Edmundson et al. 2003). Both fry weight and abundance were related to the biomass populated by *Cyclops* and *Diaptomus* species in that research. With the increasing shift towards *Diaptomus pribilofensis* dominance in the zooplankton community in Lake Laberge, it is speculated that an increased growth rate, and ultimately growth dilution of contaminants, is beginning at the YOY stage. An increased level of planktivory by YOY fish may only further change the structure of the plankton community (Mazumder and Edmundson 2002) as plankton diapause stages have often been related to fish predation as a method of avoidance adaptation (Northcote 1988; Wetzel 2001). Changes in the zooplankton community within Lake Laberge as previously noted might directly account for the increased growth in some fish, including lake trout, although no direct correlations were studied in this project. Factors determining the size spectrum and species community composition of plankton are important determinants of the structure of the pelagic food web and ultimately the partitioning of contaminants.

The OC concentrations in zooplankton from Lake Laberge declined over 70% between 1993 and 2001. These declines may be effected by a change in the plankton community composition due to the unique differences in physiology that could account for the temporal declines in OC levels. Differences in physiology that could affect contaminant concentrations include body size (Fisk et al. 2001) and hence surface area that is in contact with the water, which is a primary source of OC contaminants for plankton (Swackhamer and Skoglund 1993). A shift from *Cyclops scutifer*, to a smaller and more herbivorous *Diaptomus pribilofensis* (noted as having a smaller maximum body size by length (Table 4.10; Kirkland and Gray 1986), may account for some of the overall decrease in OC levels in zooplankton from Lake Laberge. The shift towards *D. pribilofensis* dominance may not show in $\delta^{15}N$ analyses because of two main factors, which could not be examined; the amount of herbivory vs. carnivorous activity compared to *C.scutifer,* and the amount of community shift necessary to demonstrate a change in trophic status from mass samples of plankton. Other species differences include varying elimination rates (Swackhamer and Skoglund 1993) or different body composition (e.g. lipids, proteins; Parsons et al. 1961), potentially affecting contaminant partitioning rates (Lederman and Rhee 1982). Such a change in the community composition will eventually relate to changes in contaminants higher in the food chain through trophic cascade (Smith 1995; Stewart et al. 2003). Stewart et al. (2003) concluded that major changes in fish OC levels following the Red River flood of 1997 were not linked to the transport of new chemicals into the ecosystem, but rather were due to species shifts within the plankton community. These studies provide information on the importance of plankton community composition interactions to upper trophic levels, which may be influencing OC contaminant levels in Lake Laberge.

It was hypothesized that the closure of the commercial fishery would shorten the food chain length in Lake Laberge due to increased intraspecies competition resulting in more individual organisms shifting foraging efforts towards lower trophic levels (Kidd, 1996). Since higher trophic level feeding results in higher contaminant concentrations in fish (Rasmussen et al. 1990), feeding at a lower level will subsequently produce OC declines in that species. Stable isotope ratios ($\delta^{15}N$ and $\delta^{13}C$) indicated no changes within the trophic levels or food sources for Lake Laberge biota with the exception of burbot and possibly northern pike. Studies have shown that dietary habits have a large impact on contaminant levels between species within an ecosystem. For example, a large disparity between OC levels in pelagic lake trout and littoral-feeding whitefish and lake trout was attributed to differences in food sources as measured by $\delta^{13}C$ data (Campbell et al. 2000). Lake trout were feeding on a pelagic copepod that was much higher in fat content compared to littoral zooplankton species and this accounted for the difference in contaminant levels. Burbot from Lake Laberge have not changed significantly in $\delta^{13}C$, but given the trend towards a more depleted $\delta^{13}C$ in 2001 compared to 1993 and knowing the burbot are generally piscivorous, it is possible they were consuming more cisco in their diet. Considering the marginally lower trophic level for burbot, it is more likely that the decrease in lipid content and OC concentrations in prey was responsible for the declines in some OC concentrations in this species.

Northern pike may have increased a half trophic level from 1993 to 2001 based on $\delta^{15}N$ analyses indicating a shift in their prey items towards a more piscivorous diet. Pike are known piscivores often inhabiting the shallows, weed beds and river mouths (Scott and Crossman 1973), which are primary habitat locations for young, small fish. Given the fact that fish populations have increased in Lake Laberge, it is suspected that pike have begun ingesting more, and possibly larger, YOY fish in their diet thus increasing their $\delta^{15}N$. Although their temporal $\delta^{13}C$ values had remained unchanged based on a definition of enrichment roughly 0.8-1.3‰±1.1‰ relative to the diet; (DeNiro and Epstein 1978; McCutchan et al. 2003; Jardine et al. 2003), the $\delta^{13}C$ values have become slightly more depleted since 1993, providing evidence that northern pike may have a higher intake of more pelagic prey (e.g. least cisco) in their diet. Since high trophic level fish correlate with higher levels of OC (Kidd et al. 1995a; Kiruluk et al. 1995; Zaranko et al. 1997; Kidd et al. 1998), the increased $\delta^{15}N$ of pike combined with larger body size and decreased lipids may account for some of the increased (or stabilized rather than decreasing) levels of contaminants observed in this species. It should be noted that pike have a strong association with littoral habitats that may explain the lack of correlation in temporal OC levels compared to the other benthic and pelagic species in this study.

Lake whitefish have become more enriched in $\delta^{13}C$ suggesting that this fish has increased its consumption of benthic prey (Jardine et al. 2003). This includes snails and chironomids ($\delta^{13}C$ range of –20‰ to –24‰ and –22‰, respectively), which were listed as important prey for lake whitefish in earlier studies (Kidd 1996). It has also been noted that copepods, especially *Diaptomus,* were important in the initial stages of growth for young lake whitefish (Scott and Crossman 1973), which provides an explanation for the decline in whitefish OC similar to other pelagic dwelling fish in Lake Laberge.

Although the fish populations of Lake Laberge have changed since the closure of the commercial fishery in 1991, any resulting shifts in the food web through foraging competition or food abundance has not dramatically altered the structure or hierarchy of the chain. Lake Laberge continues to maintain a relatively long food chain in comparison with other regional lakes. However, a general observation can made that the more pelagic species (zooplankton, cisco, lake whitefish, lake trout) have demonstrated the most consistent changes in OC concentrations compared to predominantly benthic organisms (burbot, longnose suckers) which has likely been caused by differences in the food web interactions beginning at the plankton trophic level.

4.6 Summary and Conclusions

This study has found that OC contaminant concentrations have decreased significantly in biota sampled from Lake Laberge from 1993 to 2003. Over the same period, fish populations have increased for most species following the closure of the commercial fishery in the early 1990s. Coinciding with this population increase were significant changes to the morphology and tissue lipid contents for most fish. Zooplankton OC concentrations also decreased over time, however, the lipid content of those organisms remained unchanged. A notable change did occur in the densities and community composition of zooplankton between 1982, 1993 to 2002. Stable isotope data remained consistent over the study period for all fish except northern pike (partial increase in $\delta^{15}N$), burbot (decrease in calculated trophic level) and lake whitefish (more enriched in $\delta^{13}C$). Biomagnification factors were higher for 2001 piscivorous fish and relatively unchanged for forage fish species while the overall FWMF were elevated when comparing 1993 to 2000-2001 samples for all 6 OC groups.

Changes in fish morphology and tissue lipid contents were likely related to recent fluctuations in the fish and zooplankton populations of Lake Laberge. Growth dilution and decreased OC in prey are suspected as the primary influences on OC levels in lake trout and round whitefish. An increase in fish populations provided a larger food base for piscivores resulting in growth dilution of contaminants within lake trout. Non-piscivorous species undergoing growth dilution suggests an increase in plankton assemblages (the food base), which coincides with the zooplankton density changes observed. Growth dilution at the bottom of the food web provides an added factor in reducing the contaminant load available for predatory consumption thus diminishing the base contaminant concentrations that are ultimately biomagnified. Recent declines in fish lipid levels were likely related to changes in intraspecies competition for resources, growth at YOY stages, or shifts in the community composition of plankton, the primary prey for YOY fish. The declines in lipid content are suspected as having large effects on OC in lake whitefish and burbot, but were likely contributing to the overall decreased contaminants for all fish.

Research has suggested that climate can indirectly modify the transfer of chemicals in food chains through effects on fish growth (Smith 1995). Since fish production generally increases with primary production (Downing et al. 1990; Rowan and Rasmussen 1992) and climate is a major factor directing phases of plankton succession, then weather can indirectly affect contaminant concentrations in aquatic biota through primary productivity (Larsson et al. 1992). It is speculated that climate variations may be affecting primary production in Lake Laberge causing growth dilution of OC at the plankton trophic level. Shifts in plankton community composition due to climate change or increased fish predation (from higher populations of fish in Lake Laberge), may result in decreased contaminant levels in these organisms as the contaminant loads relate to the specific characteristics unique to each species (physiology, behaviour and metabolic characteristics). Growth dilution of plankton or a change in community composition results in lower levels of bioavailable contaminants for predators by setting a lower base value for contaminant concentrations in primary prey. Although biomagnification still occurs, the initial OC concentrations are less and hence a decrease in concentrations in upper trophic levels is measured (trophic cascade). Without a change in primary production in Lake Laberge, a change in species composition, at the plankton trophic level, towards the smaller-bodied *D. pribilofensis*, as opposed to the larger-bodied *C. scutifer,* could account for lower base OC values as each plankton species differs in cell size (water to surface area ratios), morphology, metabolism and chemical composition. This change in OC at the lowest trophic level is expected to have a significant influence (trophic cascade) on OC concentrations upwards in the food web as observed with round whitefish.

Skare et al. (1985) concluded that fluxes of OC in and out of a fjord ecosystem were related to the area (temperatures, currents and precipitation), but that variance in OC concentrations in individuals is affected by differences in growth, lipids, age, migration habit and metabolic capacities. All of these factors may influence contaminants in biota and many of them have had temporal variations in the Lake Laberge system, although it seems evident that the predominantly pelagic species have more consistent trends in OC compared to the more benthic animals. Overall the decrease in OC in Lake Laberge biota is directly related to natural responses of the biota in the ecosystem to such stimulus and stressors following exploitation and regional climate variations. The Lake Laberge ecosystem may continue to show fluctuations in populations, species characteristics and OC concentrations for several years to come, and as such, sentinel species including lake trout, burbot, whitefish, cisco and plankton should continue to be monitored for future temporal correlations.

5. NUTRIENT LEVELS, HISTORICAL PRIMARY PRODUCTION AND TEMPORAL TRENDS OF ORGANOCHLORINE PESTICIDES AND PCB FROM LAKE LABERGE, YUKON TERRITORY: SEDIMENT CORE ANALYSIS

5.1 Abstract

The recent temporal trends of contaminants and historical primary production in Lake Laberge were measured using radioisotope dating, organochlorine (OC) analyses and phytoplankton enumeration methods from collected sediment cores. The fluxes of OC pesticides and PCB in sediments through the 1990s remained relatively stable with slight increases for all groups except chlordane in more recent years. All 6 groups, DDT, HCH, PCB, chlorinated benzenes (CBz), chlordane (CHL), toxaphene (CHB) had average fluxes (1992-1999) less than historical peaks except for HCH in 1999. Elevated levels of β-HCH in the core slice dated to 1999 (surface) coincides with a year of abnormally high precipitation for the lower Yukon region. During this decade, phytoplankton assemblages climbed to an all time 51 year high (primarily in 1995) as measured by diatoms, chrysophytes and cyanophytes enumerated from sediment slices. The population increases, also noted in 1982, generally correlate with warm, dry periods (droughts) that exceed 2 years in length. No correlations of sediment OC levels were found with these changes in phytoplankton populations. It is suspected that growth dilution of contaminants in the 1990s that occurred in most Lake Laberge fish coincides with the dramatic growth of phytoplankton during the same period

providing evidence for growth dilution of contaminants from the base of the food web. We postulated that the regional climate change in the mid-1990s (warm, low precipitation) may be responsible for creating a concurrent change in both recent OC sediment flux and phytoplankton growth in Lake Laberge through related mechanisms (heat, light, terrestrial runoff, varying sedimentation, atmospheric deposition).

5.2 Introduction

Organochlorine pesticides (OC) and PCB have long been monitored in the Arctic and sub-Arctic aquatic environment even though these materials have had little use in these areas for decades (Barrie et al 1992). The 'grasshopper effect' theory explains how some of these contaminants continue to settle into the northern regions (Bidleman et al. 1989) while other sources remain due to the slow environmental degradation of material from past regional use (Barrie et al. 1992; Van Dijk and Guicherit 1999; Bidleman 1999; Rawn et al. 2001; CACAR 2003c).

Monitoring the levels of contaminants atmospherically transported or deposited into terrestrial and aquatic areas is required for the analysis of temporal OC trends. The use of dated sediment cores has become common as an accurate method of determining contaminant flux to the northern lakes and rivers since sediments act as sinks for OC and PCB (Muir et al. 1996; Gajewski et al. 1997; Rawn et al. 2001; Stapleton et al. 2001; Outridge et al. 2005; Stern et al. 2005). Historical concentrations of contaminants can be measured from sediment cores by combining gas chromatography OC analysis of sediment material with radioisotope dating techniques using ^{137}Cs and ^{210}Pb (Oldfield and Appleby 1984; Wilkinson and Simpson 2003).

Sediment cores not only maintain layers of deposited soil, but they also contain materials such as biotic remnants including siliceous phytoplankton skeletons. For example, the hard shell skeletons of chrysophytes (Chrysophyta) and diatoms (Bacillariophyta) which settle into the sediments can be used as general indicators of historical primary productivity levels through time (Gajewski et al. 1997; Wolfe 2000; Outridge et al. 2005; Stern et al. 2005). A measure of temporal primary production provides information on periods of high phytoplankton growth periods within an aquatic food web. It may also assist in the prediction of shifts in contaminants through biota as plankton populations, hence biomass, relates to environmental OC concentrations (Taylor et al. 1991; Swackhamer and Skoglund 1993).

Phytoplankton contaminant burdens are affected by water and suspended sediment concentrations as well as by growth rate (Lederman and Rhee 1982; Taylor et al. 1991; Swackhamer and Skoglund 1993). Although environmental OC levels can be an important factor affecting contaminants in plankton (through bioconcentration or ingestion), growth rates can supersede the influence of this factor as seen in OC concentrations in algae during high growth and low growth periods (Swackhamer and Skoglund 1993). Several primary limitations including temperature, light exposure and nutrients determine seasonal plankton growth rates (Wetzel 2001). If these factors become less restrictive, the potential for increased plankton growth and hence biomass dilution of OC contaminants within plankton is possible. Therefore, the analysis of OC trends in phytoplankton needs to be assessed in correlation with changes in available nutrients and regional climate over time. A history of climate and nutrient data and plankton counts in dated sediment cores are often overlooked or unmeasured parameters but can affect primary and secondary production and hence bioavailable OC in aquatic systems.

The primary objective of this research was to quantify the recent OC pesticides and PCB contaminant fluxes to Lake Laberge to investigate the hypothesis that temporal abiotic contaminant inputs (atmospheric or runoff) or changes associated with climate variations are contributing to the decreases seen in Lake Laberge biota (Chapter 4). A history of contaminants in sediment cores from Lake Laberge, 1992 and prior, was recently published elsewhere (Rawn et al. 2001), but a second objective of our research was to update and quantify more recent OC sediment fluxes to correlate with the temporal trend food web study (Chapter 4) from 1992 to 2001. The third objective was to observe any significant variations in nutrient levels in Lake Laberge water samples compared to previous reports, along with regional climate shifts, that may account for changes in primary production. The last objective was to assess patterns in historical primary production through the enumeration of phytoplankton from the sediment core slices. A correlation of production with contaminant flux provides evidence for the hypothesis of biomass dilution at the base of the Lake Laberge food web, which is also suspected as a large contributor to OC declines in higher trophic levels (Swackhamer and Skoglund 1993; Stewart et al. 2003). The analyses of these abiotic and climate factors (annual precipitation and growing degree-days), should provide insight into the historical contaminant inputs and influences on primary production, and ultimately an explanation for the recent decline in contaminants in the biotic constituents of Lake Laberge.

5.3 Materials and Methods

5.3.1 Sample Collections

The south tip of Lake Laberge is approximately 45km north of the city of Whitehorse (61^{o}11'N, 135^{o} 12'W) in the Yukon Territory and is part of the Yukon River system. Limnological data on the lake has previously been reported (Table 3.1, Figure 3.1).

Unfiltered water samples were taken at 6 locations on Lake Laberge at depths of 0-1m (Figure 5.1) during the last two weeks of July and first week of August (2000 and 2001) using solvent washed steel cans (for OC) or polyethylene jars and analysed for nutrients using the methods of Stainton et al. (1977).

Sediment core samples were taken in the deepest sections of the lake, two at W3, and one at both W5 and W6 (Figure 5.1) using a 10 cm diameter, 50 cm length KB corer (March, 2000). Cores were sliced into 0.5 cm (surface layer up to 13 cm) and 1 cm sections (after 13 cm), weighed and stored frozen (0-10 ^{o}C) in Whirlpak® bags prior to analyses. All samples were shipped to the Freshwater Institute, Winnipeg (Fisheries and Oceans, Canada).

Daily climate information (growing degree days and annual precipitation data) was obtained or calculated from Environment Canada (2002) from weather stations located north and south of Lake Laberge in the cities of Carmacks and Whitehorse (approximately 100 km and 45 km, respectively). Climate data from the city of Carmacks was used in conjunction with Whitehorse as the Whitehorse dataset had periodic gaps in annual information. The Carmacks climate data should be considered the more accurate of the two, but the Whitehorse data may be slightly more indicative of weather as the station is geographically closer to Lake Laberge. Annual averages for Whitehorse data may be underestimated as annual means were discarded only if one or more months of data were missing for any given year. Missing data from individual days may also provide an underestimate of mean GDD and precipitation.

5.3.2 Sediment Core Analysis

Four cores were collected and dated and one was analysed for OC content and microfossils (W3, Figure 5.1). Wet sediment slices were freeze-dried and stored at 5^{o}C in the dark until analysed for ^{210}Pb, ^{137}Cs, OC concentrations, total and inorganic carbon. Organic carbon was determined by combustion of dried sediment in an oxygen-helium atmosphere at 950-975°C and quantification of CO_2 using a CE 240-XA Elemental Analyser (Exeter Analytical Inc., North Chelmsford, MA).

The ^{210}Pb and ^{137}Cs activity profiles in core slices were obtained by direct counts on an α/γ spectrometer (with Ge-Li semiconductor detector), which were used to calculate age as a function of core depth (Oldfield and Appleby 1984; Wilkinson and Simpson 2003) according to the detailed methods in

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Figure 5.1 Map of Lake Laberge with water and sediment sample locations (W1-W6).

Muir et al. (1996). Sediment focusing factors were used to areal normalize contaminant fluxes for physical processes such as sediment resuspension and basin deposition.

The factors were calculated using the ratio of non-disturbed (where possible) sediment ^{210}Pb flux to the regional atmospheric ^{210}Pb flux (Muir et al. 1996; Wilkinson and Simpson 2003; US-EPA 2004).

Sedimentation rates were calculated using both linear and constant rate of supply (CRS) models (Oldfield and Appleby 1984; Wilkinson and Simpson 2003). The CRS model was used to calculate fluxes of OC but both models returned similar sedimentation rates within 5% of each other. Sediment OC fluxes (ng/m^2/y) were corrected for sedimentation rates among slices and differences in organic carbon according to the following calculation:

$$Flux = \frac{C_{dw} \cdot S_{dw}}{f_{oc} \cdot F}$$

where C_{dw} is the dry-weight OC concentration in each slice (ng/g), S_{dw} is the sedimentation rate in each slice (g/m^2/y), f_{oc} is the fraction of organic carbon in each slice and F is the focusing factor of the core. Dry-weight contaminant fluxes and OC inventories (ng/m^2) were also determined to allow future comparisons with literature values for other lakes:

$$Inventory = \sum \frac{C_{dw} \cdot S_{dw} \cdot I}{F}$$

where I is the slice interval in years (Muir et al., 1996).

5.3.3 Organochlorine Analysis

The core (W3) was analysed for the same PCB and OC pesticides as in previous chapters (Chapter 3, Chapter 4). Sediment slices were extracted using the methods listed in Stern et al. (2005). In brief, freeze-dried sediments were combined with anhydrous sodium sulphate, internal standards (PCB30, OCN) and extracted in an accelerated solvent extractor (ASE 200, Dionex Canada Ltd., Oakville, ON). Sulphur was removed by treatment of the extracts with activated copper powder to reduce chromatogram noise and peak artefacts (Muir et al. 1996). Extracts were reduced in volume and fractionated on 1.2% deactivated Florisil (Chapter 3 and 4 Methods). Extracts were then injected onto a Varian Star 3400 GC equipped with an electron capture detector (Ni^{63} electron source) and an 8100 auto-sampler. The column used was a J&W Scientific (Agilent Tech) DB-5, 60m, 250 μm ID with TMP as the solvent, Hydrogen Ultra High Purity (UHP) carrier gas (at ~2 mL/min), nitrogen UHP make-up gas (at ~50mL/min). Injection and temperature programs conditions were as follows:

Injector temperature 80°C, detector temperature 300°C, initial temperature 100°C (hold time 2min), 1st temperature program increasing to 150°C at 15°C/min, 2nd temperature program increasing to 265°C at 3°C/min

Chemicals were quantified by comparing chemical peak areas (identified by retention time) to those of commercially available standards of known concentration. Recoveries of the internal standard PCB30 and OCN were 60% ± 8% and 61% ± 10% for sediment sections, respectively.

5.3.4 Phytoplankton Enumeration

Sediment sections from the W3 core were also enumerated for phytoplankton densities. Sub-sample slices were rehydrated in 20 ml of distilled water, sonicated, and 1 ml aliquots diluted 8-64x. Concentrations of microfossils were analysed using an Utermohl settling chamber and a Wild M40 inverted microscope following the methods in Findlay et al (1998) and Kling (1998). Samples were analysed for non-siliceous and siliceous microfossils such as blue-green akinetes, blue-green sheaths, green algae remains, zooplankton exoskeletons, diatoms frustules, chrysophyte resting cysts and dinoflagellate remains, amongst other abiotic and organic particles (H. Kling, pers. comm.). Microfossils were enumerated to the genus and major groups or to species taxonomy where possible. The major groups observed included Bacillariophyta (diatoms), Cyanophyta (filamentous blue green algae) and Chrysophyta (silica resting cysts). The microfossil data was plotted against years as determined by the core dating and OC fluxes in sediment slices to interpret trends in phytoplankton that may indicate the occurrence of short or long term events in the watershed (Gajewski et al. 1997; Outridge et al. 2005) affecting OC concentration patterns in the food web through general estimates of phytoplankton production in Lake Laberge.

5.3.5 Growing Degree-Days (GDD)

Growing degree-days were calculated by subtracting a base temperature ($5^{o}C$) from the mean temperature for the day in each month. If the resultant number was negative, the number of GDD for that day is zero. GDD totals were then summed by year. Years that had more than one month of absent data between April and October were not used in the review. The base temperature of 5^{o}C represents a general, minimal temperature required for diatom photosynthesis and hence growth (Wetzel, 2001). GDD were used to compare annual temperature change and plankton abundance as was done by other research (Findlay et al. 2001; Environment Canada-Yukon, pers. comm.).

5.4 Results

5.4.1 Sedimentation and Core Dating

Results of the sedimentation rate calculations and core dating were in good agreement with other research. The mean sedimentation rate in this study was 1244 g m^{-2} y^{-1} (Figure 5.2) which was slightly higher than the 999 g/m^{2}/yr calculated by Rawn et al. (2001), although the range from all 4 cores was 756-1404 g/m^{2}/yr with the highest sedimentation rate across 25 years noted for 1999. Focus factors ranged from 1.1 to 1.7, which also agrees with the 1.4 figure recently calculated by Rawn et al. (2001). The ^{210}Pb and ^{137}Cs activities were in good correlation noting that the 1966 ^{137}Cs peak corresponds with the 1966 ^{210}Pb value (Figure 5.3). Organic carbon content exceeded 98.5% of the total carbon content (range 98.5-99.2%) for each slice analysed thus negating any corrections for OC fluxes based on carbon differences in the core.

5.4.2 Organochlorine Trends and Fluxes

The ΣCBz fluxes ranged from 269.2 to 4185.5 ng/m^2/yr, starting with increases through the 1940s and 1950s with the peak value occurring in 1975 (Table 5.1). Fluxes have since decreased to pre-1975 levels with a slight rise by the end of 1999.

The ΣHCH showed slow increases up to the late 1970s with distinguished peaks occurring in recent slices from 1997-1999 (Table 5.1). The α and γ-HCH isomers were dominant until the 1990s when the β isomer became most prevalent (Figure 5.4 and Figure 5.5). Recent increases in the flux of β-HCH correlate with an increase in annual regional precipitation both north and south of Lake Laberge, which was abnormally high for the Carmacks station (Figure 5.5).

Chlordane fluxes were generally highest through the late 1960s and 1970s with slow and steady declines up to 1999 (Table 5.1). The *cis*-, *trans*-chlordane ratio which has been shown to be declining in other areas (Bidleman et al. 2002), has remained relatively consistent in the Lake Laberge core with a sharp decline from 1995-1997 followed by a spike in 1999 again (Figure 5.6) coinciding with a year of abnormally high precipitation as measured at Carmacks and an elevated precipitation year as measured at Whitehorse.

The ΣDDT fluxes showed large increases starting around the mid 1950s peaking in 1964 and declining significantly until 1997. A small peak was evident for the 1999 core slice similar to ΣHCH and ΣCBz (Table 5.1). Although ΣDDT has decreased over time, the proportion of the microbial degradation product *p,p'*-DDD to the parent compound *p,p'*-DDT decreased towards surface layers (Figure 5.7)

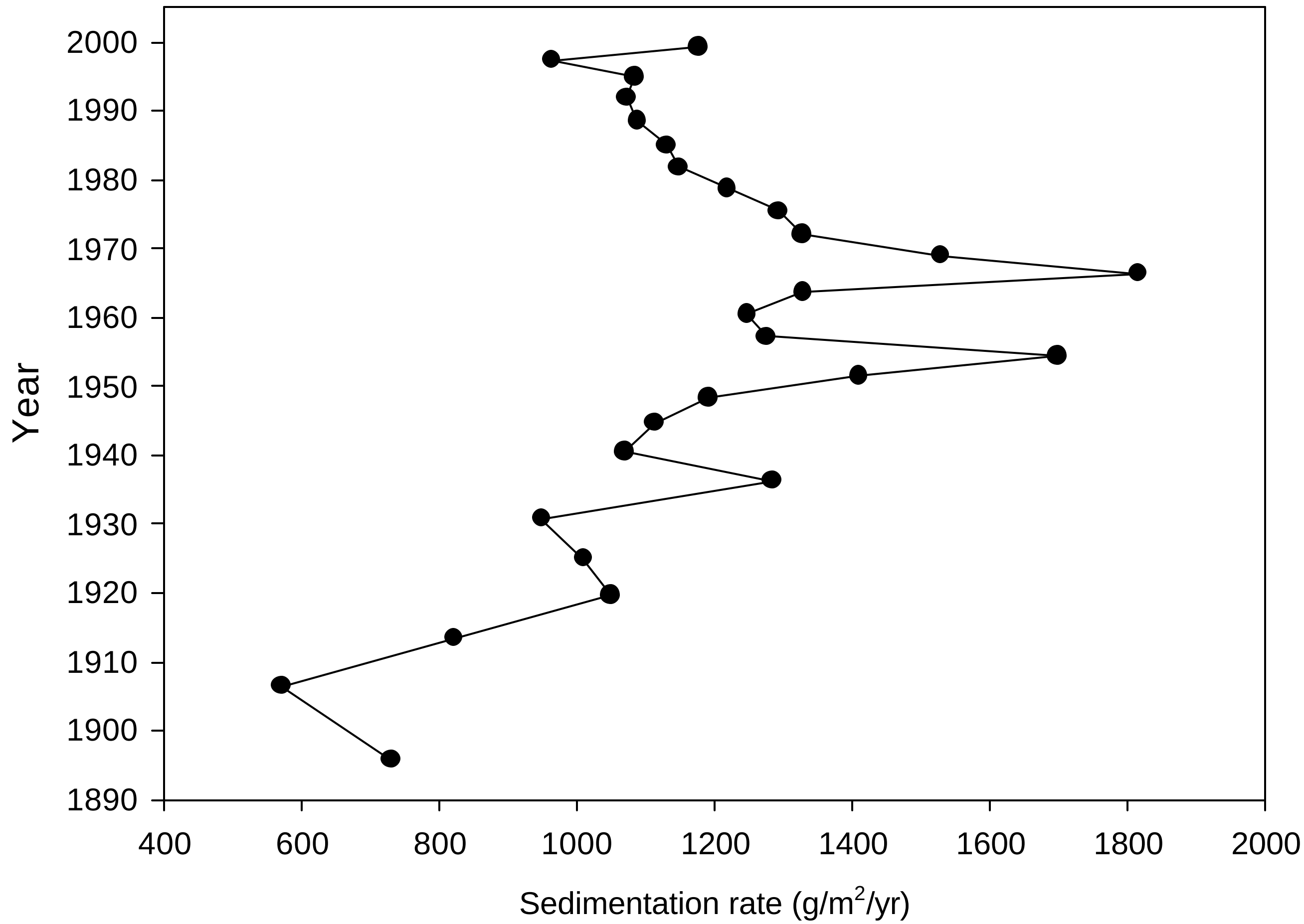

Figure 5.2 Sedimentation rate (g/m^2/yr) of the W3 core collected from Lake Laberge, YT in March, 2000.

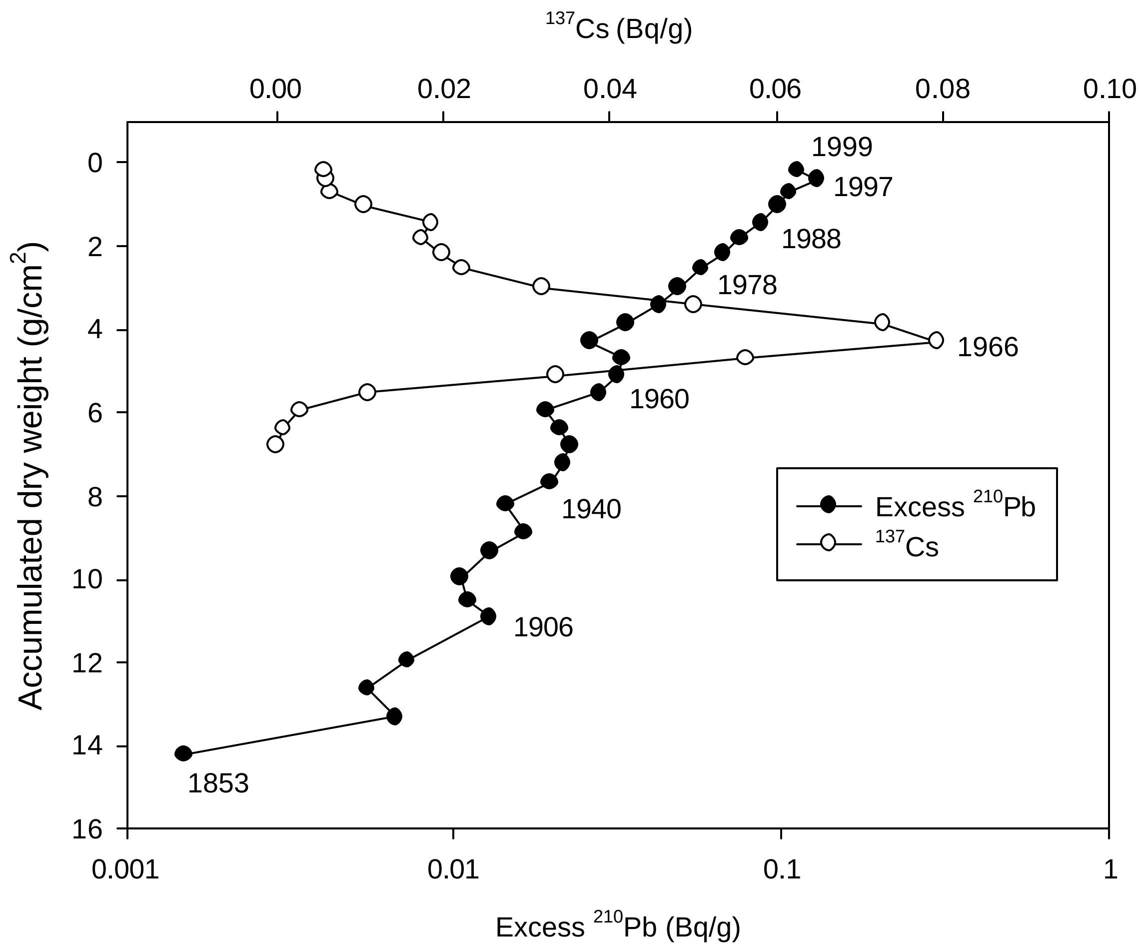

Figure 5.3 Activity (Bq/g) of the radioisotopes ^{210}Pb and ^{137}Cs and estimated dates of the W3 core collected from Lake Laberge, YT in March, 2000.

Table 5.1 Percent organic carbon content, OC fluxes and inventories (sediment rate corrected). Inventories include core slices from 1948 to 1999 (51 years) for OC and from 1951 to 1999 for PCB from a core collected from Lake Laberge, YT in March, 2000.

		Flux (ng/m^2/yr)					
Year	**% org C**	S**CBZ**	S**HCH**	S**CHL**	S**DDT**	S**PCB**	S**CHB**
1999	99.2	2043.2	1033.5	176.6	917.5	8542.7	1852.5
1997	99.1	1508.5	607.9	283.5	655.3	8997.3	1839.5
1995	99.0	1896.6	188.6	258.0	960.7	6009.9	1633.2
1992	99.0	723.5	228.4	213.1	1106.8	4636.3	1448.5
1989	98.9	1168.6	172.9	203.8	1183.9	4002.7	1860.4
1985	99.0	1655.1	289.5	297.5	1725.0	6204.0	2726.7
1982	99.0	1868.5	335.9	271.1	2186.2	6113.3	1993.8
1979	98.7	2253.6	353.9	338.1	3254.8	7857.8	1867.2
1975	98.7	4185.5	362.3	325.1	7275.8	9000.9	1360.7
1972	98.7	1449.3	233.1	272.8	19333.2	4416.9	945.1
1969	98.5	1676.3	197.5	373.0	21233.9	7483.3	1411.3
1966	98.5	1441.7	206.4	366.5	35587.0	9629.0	2064.6
1964	98.7	1710.6	205.8	267.6	70402.0	4728.7	609.6
1961	98.5	685.8	453.0	274.4	43774.6	6068.1	1706.9
1957	98.5	510.4	145.9	159.0	30011.9	4053.1	724.1
1954	98.5	396.3	287.8	270.5	10483.6	5712.9	6671.0
1952	98.5	489.1	153.0	152.5	2358.2	2585.4	746.3
1948	98.5	269.2	0.0	40.7	354.9		1411.3
Average (1948-1999)		**1440.7**	**303.1**	**252.4**	**14044.7**	**6093.7**	**1826.3**
Average (1992-99)		**1543.0**	**514.6**	**232.8**	**910.1**	**7046.6**	**1693.4**
		Inventory (μg/m^2)					
		S**CBZ**	S**HCH**	S**CHL**	S**DDT**	S**PCB**	S**CHB**
		26.8	5.2	4.7	263.3	115.2	34.4

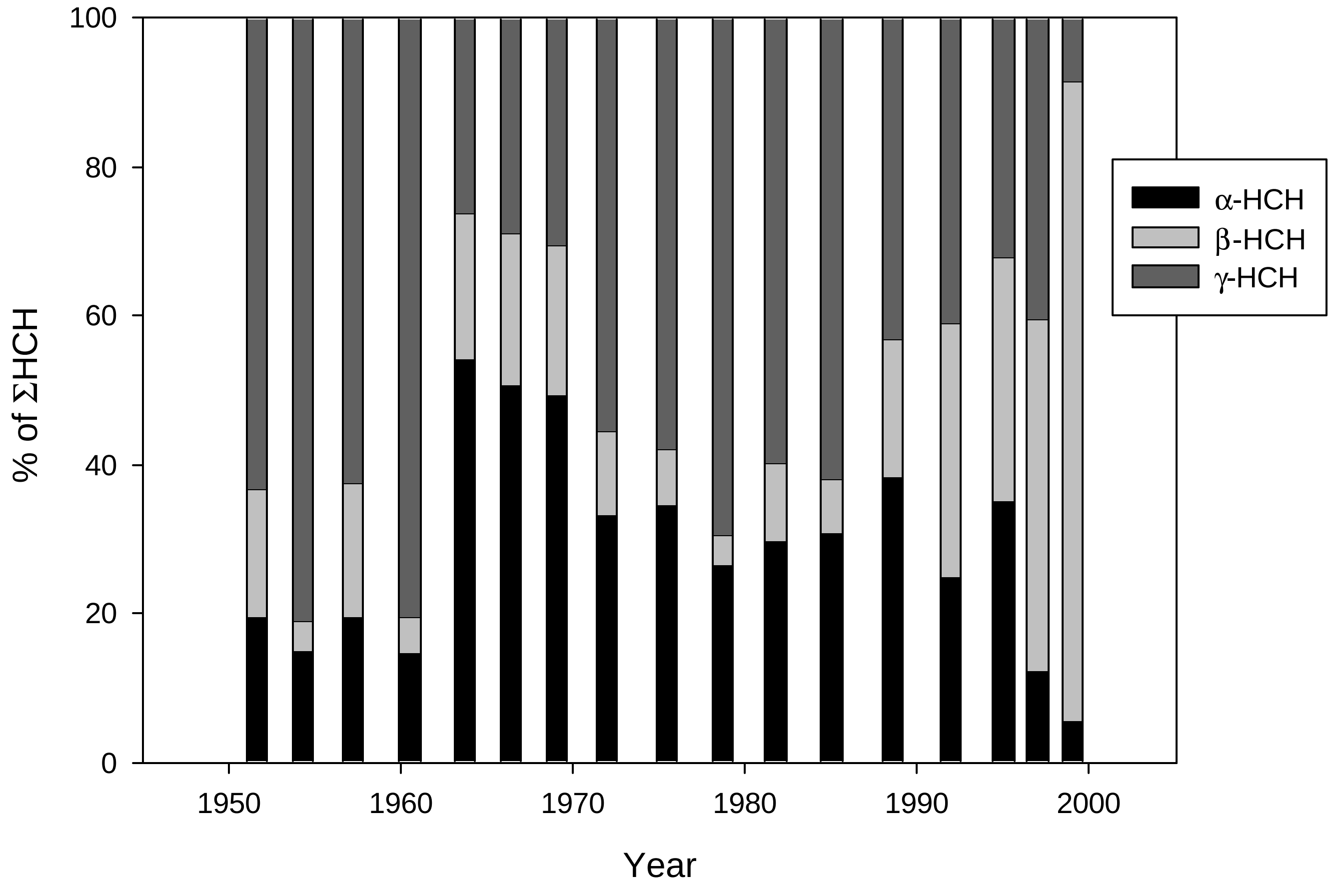

Figure 5.4 Percentage of HCH isomers over time in a core collected from Lake Laberge, YT in March, 2000.

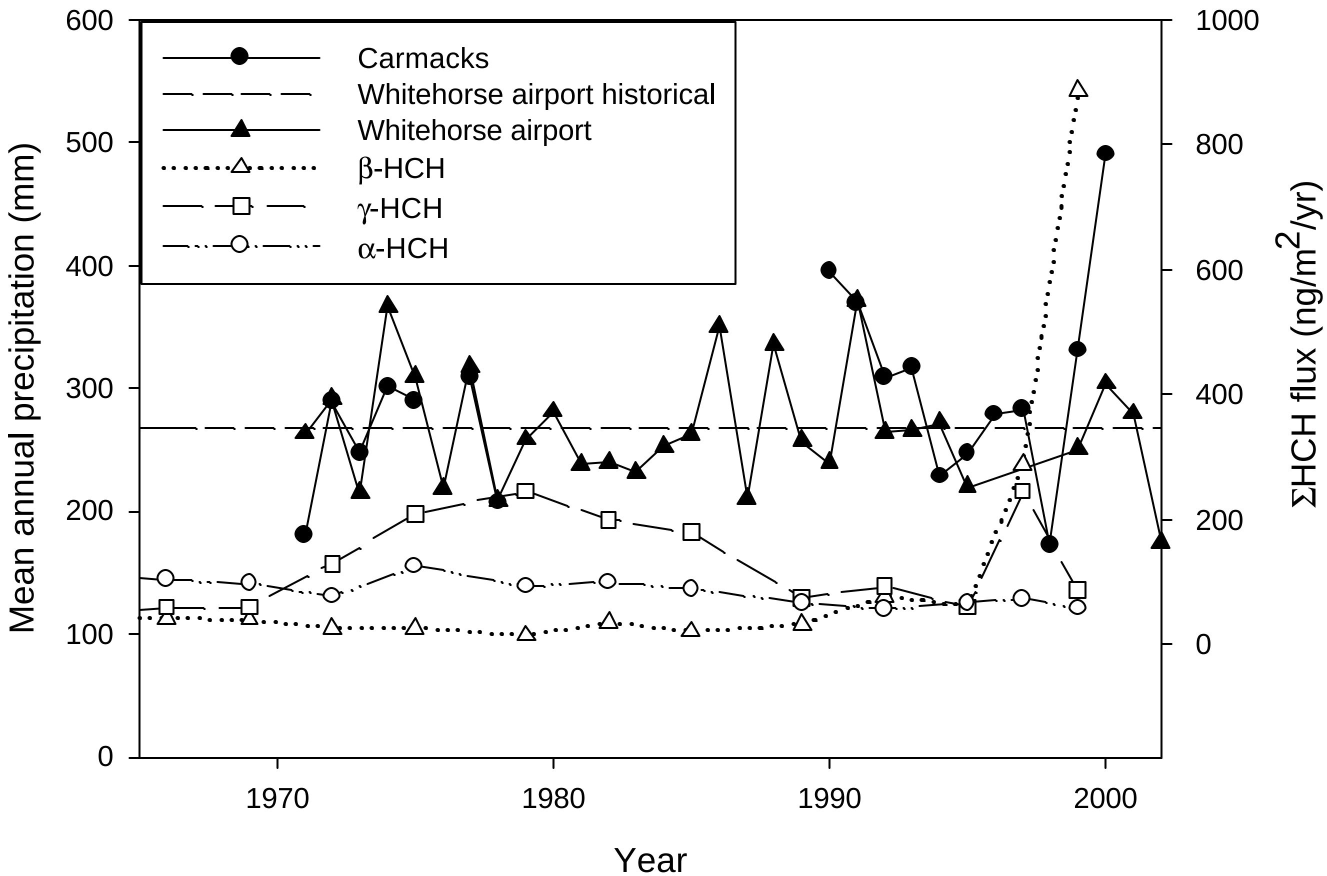

Figure 5.5 Mean annual precipitation (mm) north (Carmacks) and south (Whitehorse airport) of Lake Laberge and HCH sediment fluxes (ng/m^2/yr). The Whitehorse airport 30-year (1971-2001) mean precipitation is included.

likely due to longer microbial degradation in older slices. A ratio of ΣDDT, *p,p'*-DDT and *p,p'*-DDD (Figure 5.8) provides an estimate of DDT half-life (Rawn et al. 2001). The DDT half-life was calculated as 26.2 years, which is comparable to other values (21, 22 and 32 years) for northern lakes (Rawn et al. 2001; Stern et al. 2005).

The ΣPCB fluxes rose steadily from the early 1950s, peaking in 1966 at 9629 ng/m^2/yr before declining to half that level by 1989 (Table 5.1). A recent resurgence in ΣPCB flux was measured in the 1990s and correlates with detected increases from other northern lakes (Rawn et al. 2001; Stern et al. 2005). The ΣPCB composition was dominated by pentaCB until the 1980s when proportions of mono/di/tri-, tetra-, penta- and hexaCB were relatively similar until 1995 to 1999 when mono/di/triCB percentages were elevated. This coincided with a period of depressed precipitation and above average temperatures for the region (Environment Canada 2002). Regression lines show only the mono/di/triCB percentages were related to time but were heavily influenced by three high concentrations in the mid to late 1990s (Figure 5.9). Otherwise the composition of ΣPCB has not significantly changed over time.

Toxaphene (ΣCHB) fluxes increased through the 1960s peaking in 1966 followed by a decrease and another peak again in 1985. Since that point, fluxes have remained relatively constant with a slight increase in 1997-99 compared to 1992 values (Table 5.1).

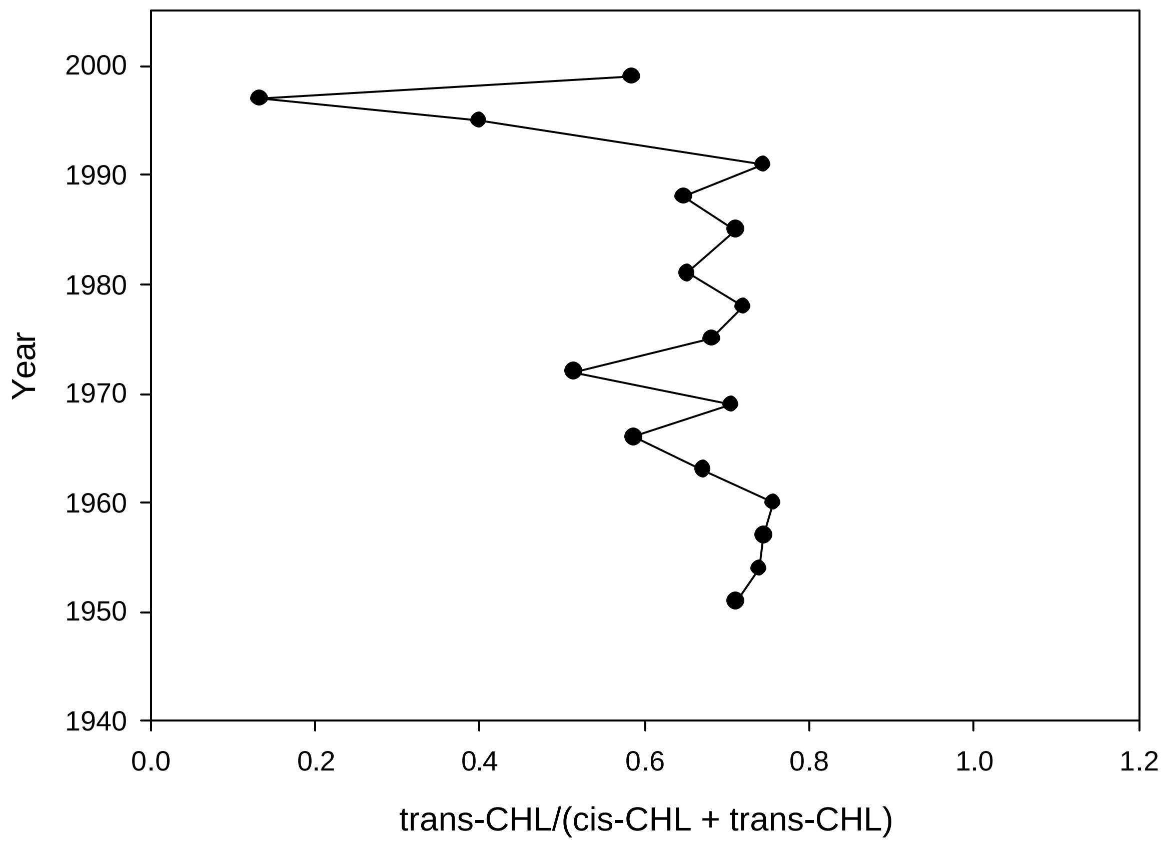

Figure 5.6 *Cis* and *trans*-chlordane enantiomer ratios over time in a core collected from Lake Laberge, YT in March, 2000.

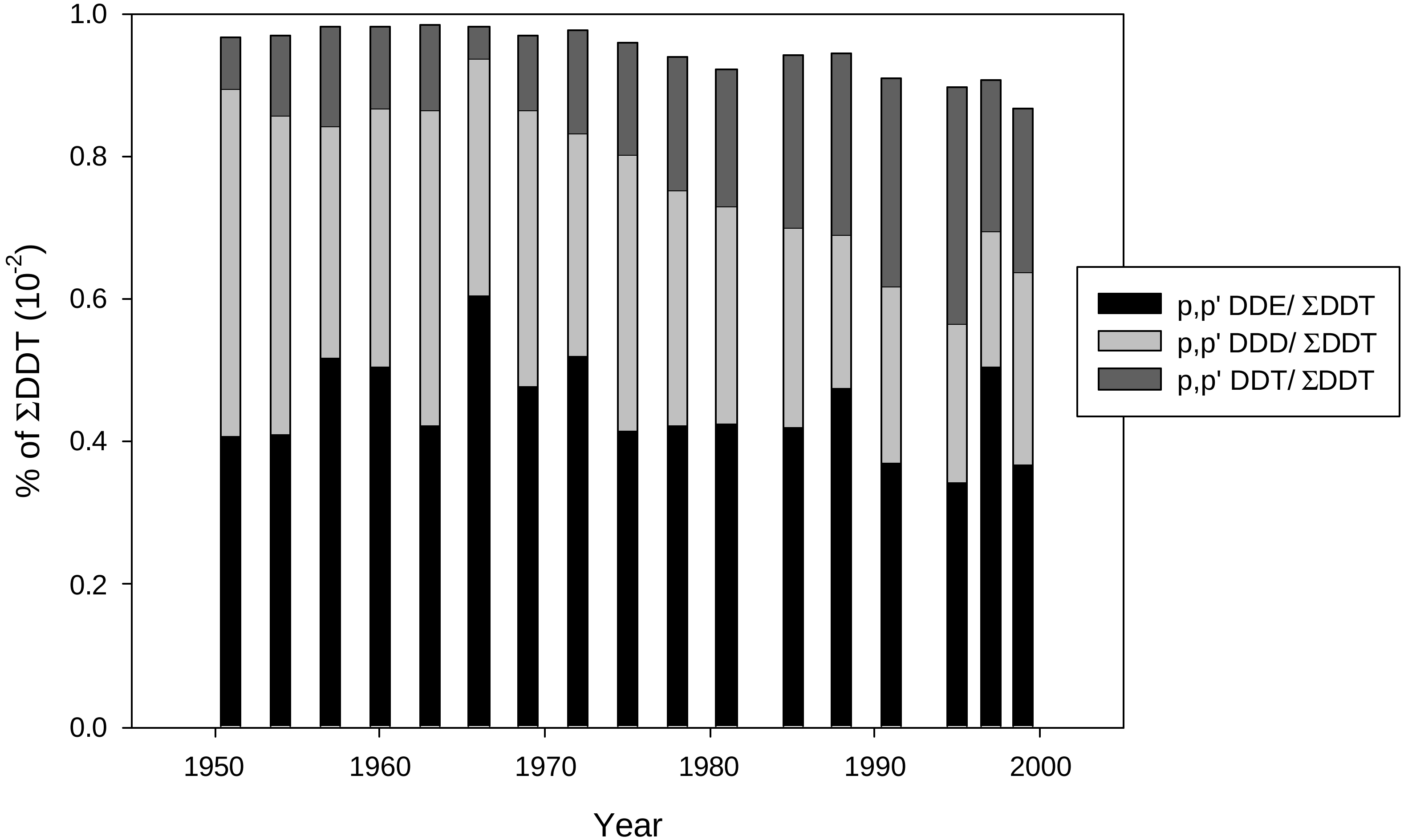

Figure 5.7 Percentage of *p,p'*DDD, *p,p'*DDE and *p,p'*DDT to ΣDDT over time in a core collected from Lake Laberge, YT in March, 2000.

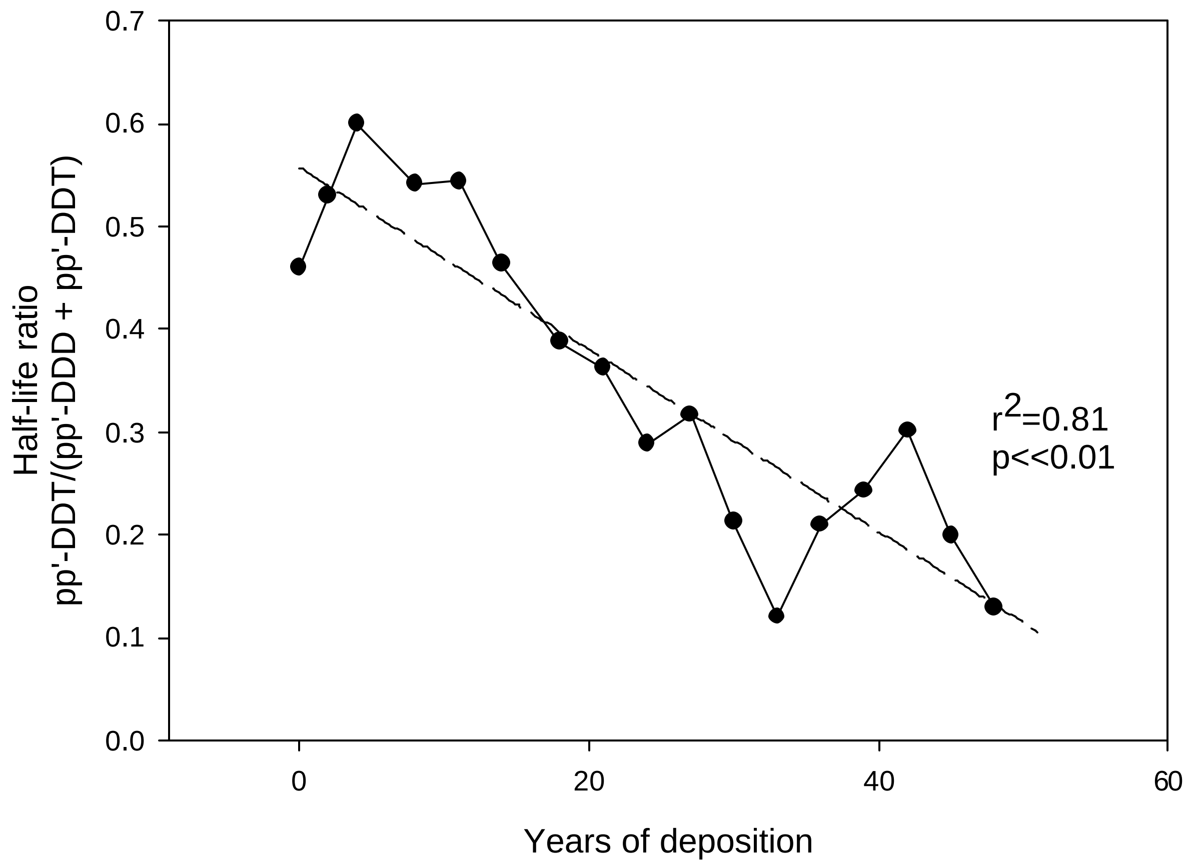

Figure 5.8 Ratio of *p,p*'-DDT to primary degradation product *p,p*'-DDD over time in the W3 sediment core from Lake Laberge. Slopes along the regression line calculate a half-life of *p,p*'-DDT at 26.2 years.

5.4.3 Organochlorine Trends and Phytoplankton Counts

OC fluxes were plotted with phytoplankton counts (Chrysophytes and diatoms) to elucidate patterns in plankton populations and OC deposition in sediments over time (Figures 5.10- 5.16). There is no evidence to indicate that patterns of OC fluxes with phytoplankton counts were anything more than coincidental occurrences. However, it can be noted that since 1980, chlordane and phytoplankton levels seemed to follow a similar pattern for increases and decreases, and from 1965, ΣPCB, ΣCHB and phytoplankton levels also generally followed similar patterns, although at some points were slightly offset in years. The ΣDDT fluxes from 1980 to 2000 were plotted separately to evaluate trends overshadowed by high historical concentrations (Figure 5.14), which showed a continuously decreasing trend in ΣDDT flux with a small increase in 1999.

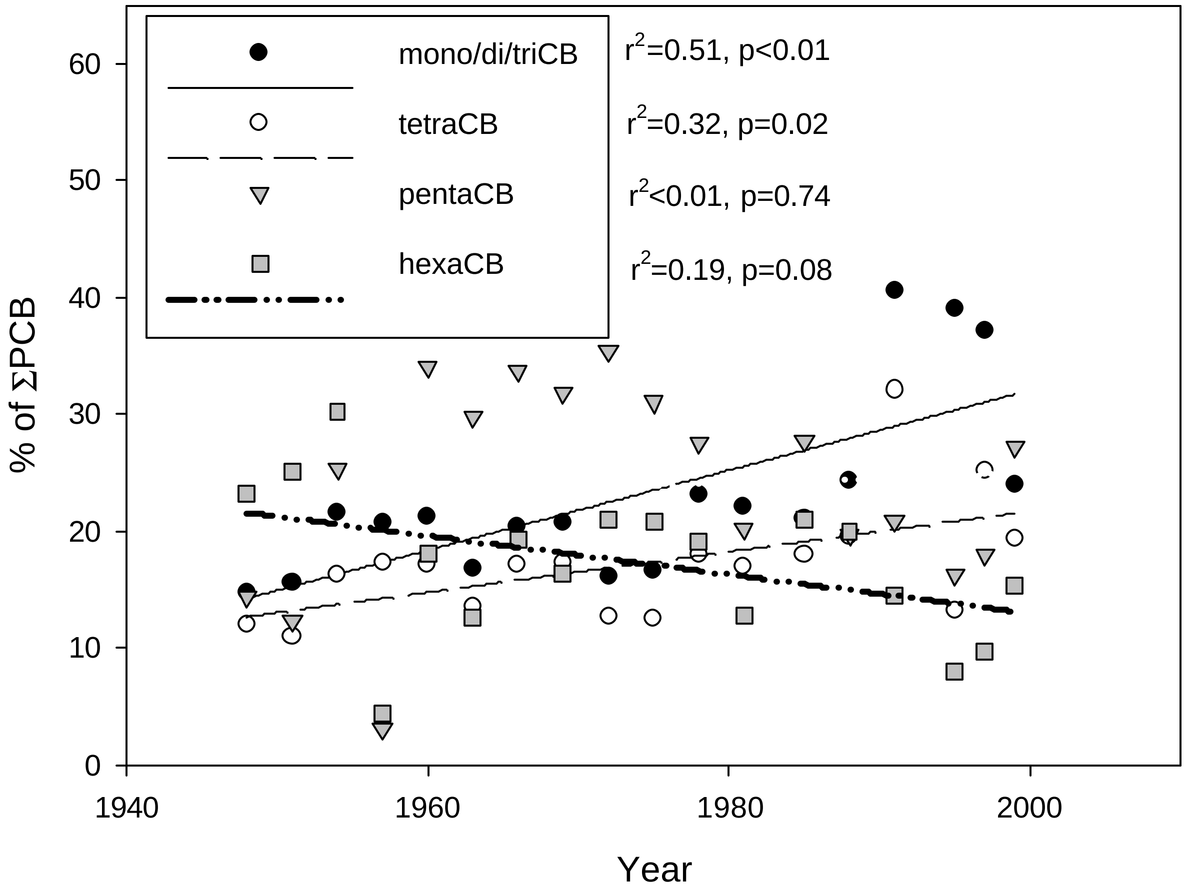

Figure 5.9 PCB congener groups over time in the W3 sediment core from Lake Laberge.

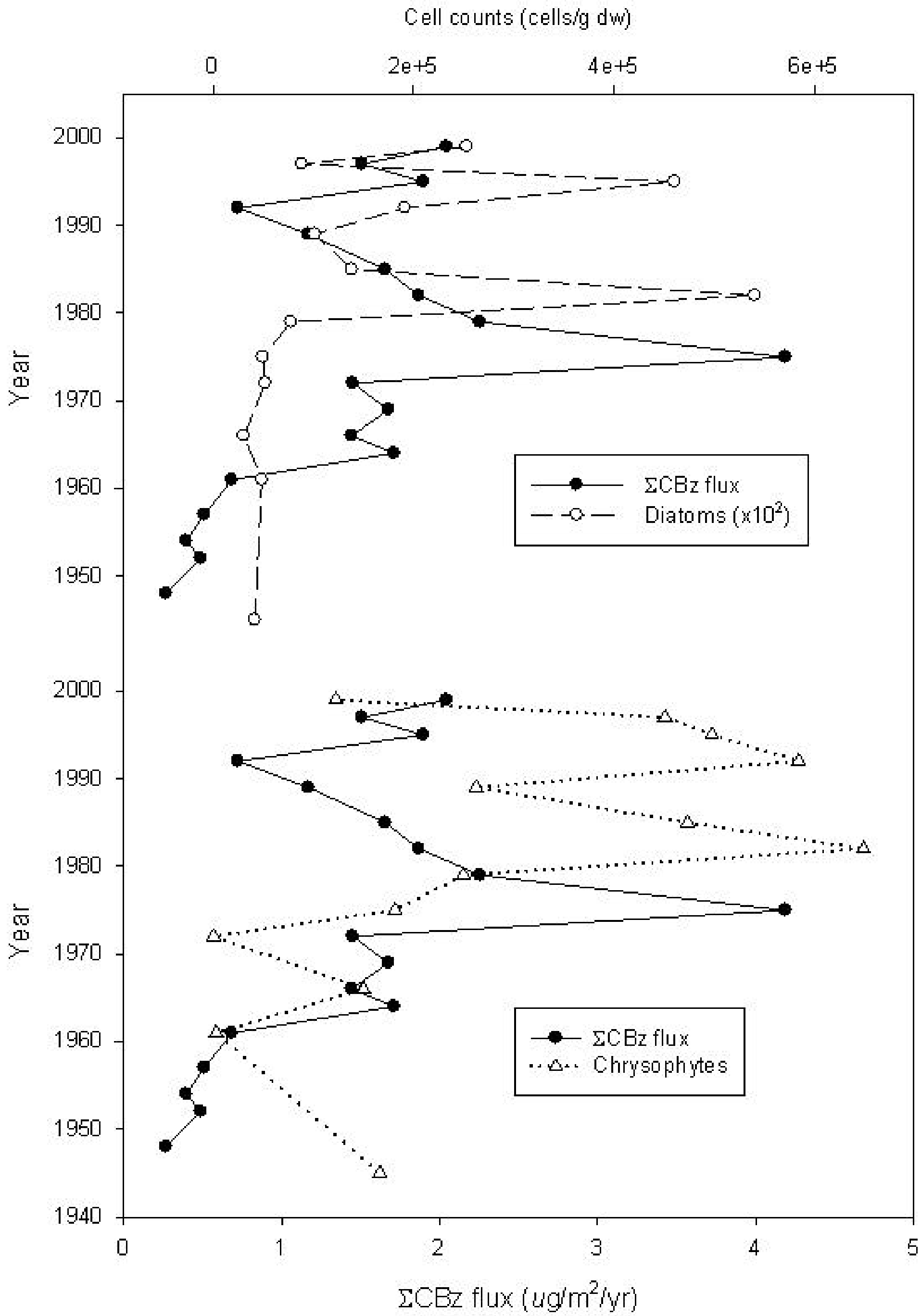

Figure 5.10 Phytoplankton cell counts (cells/g dw) over time vs. ΣCBz flux (μg/m^2/yr) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

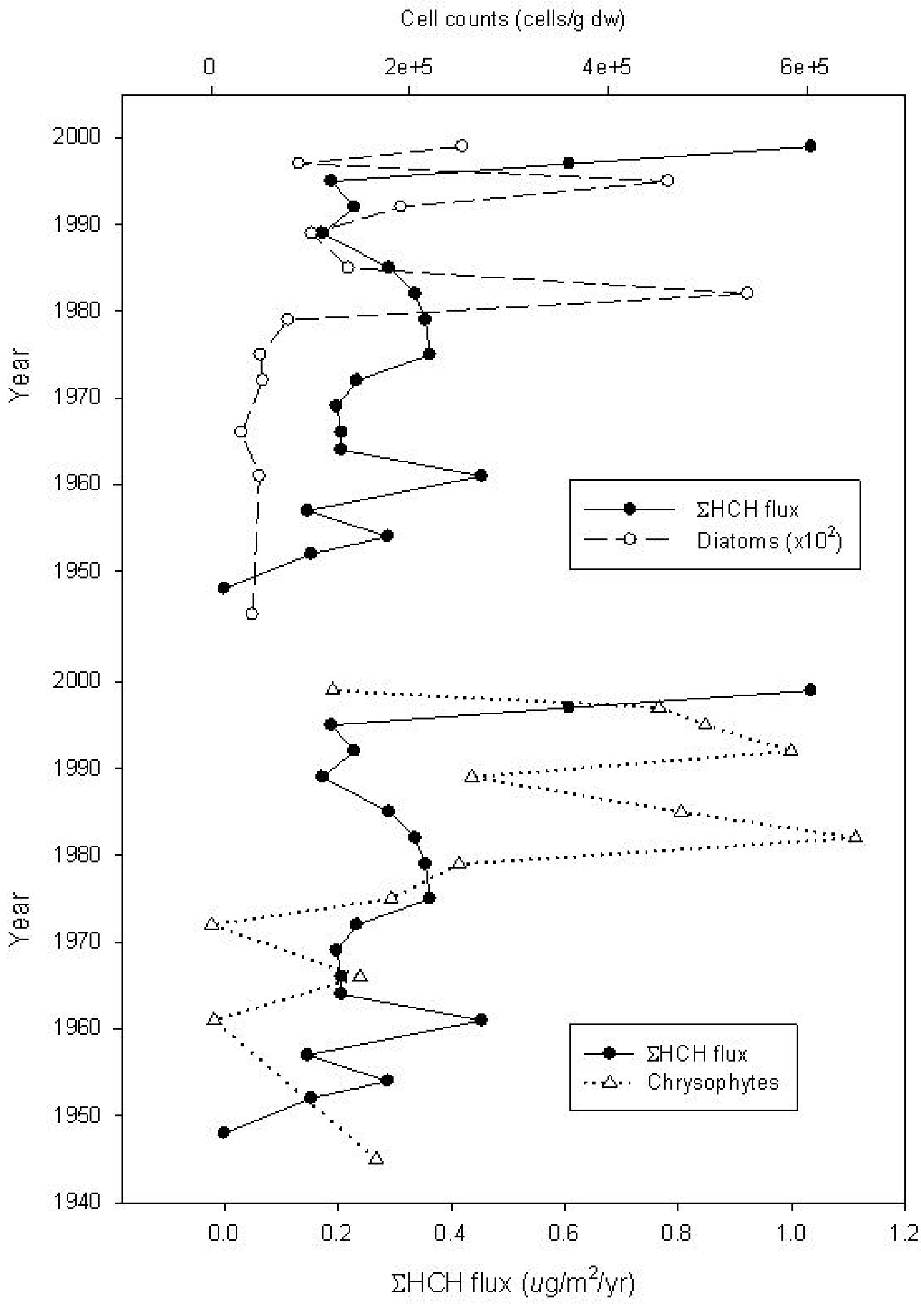

Figure 5.11 Phytoplankton cell counts (cells/g dw) over time vs. ΣHCH flux ($\mu g/m^2/yr$) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

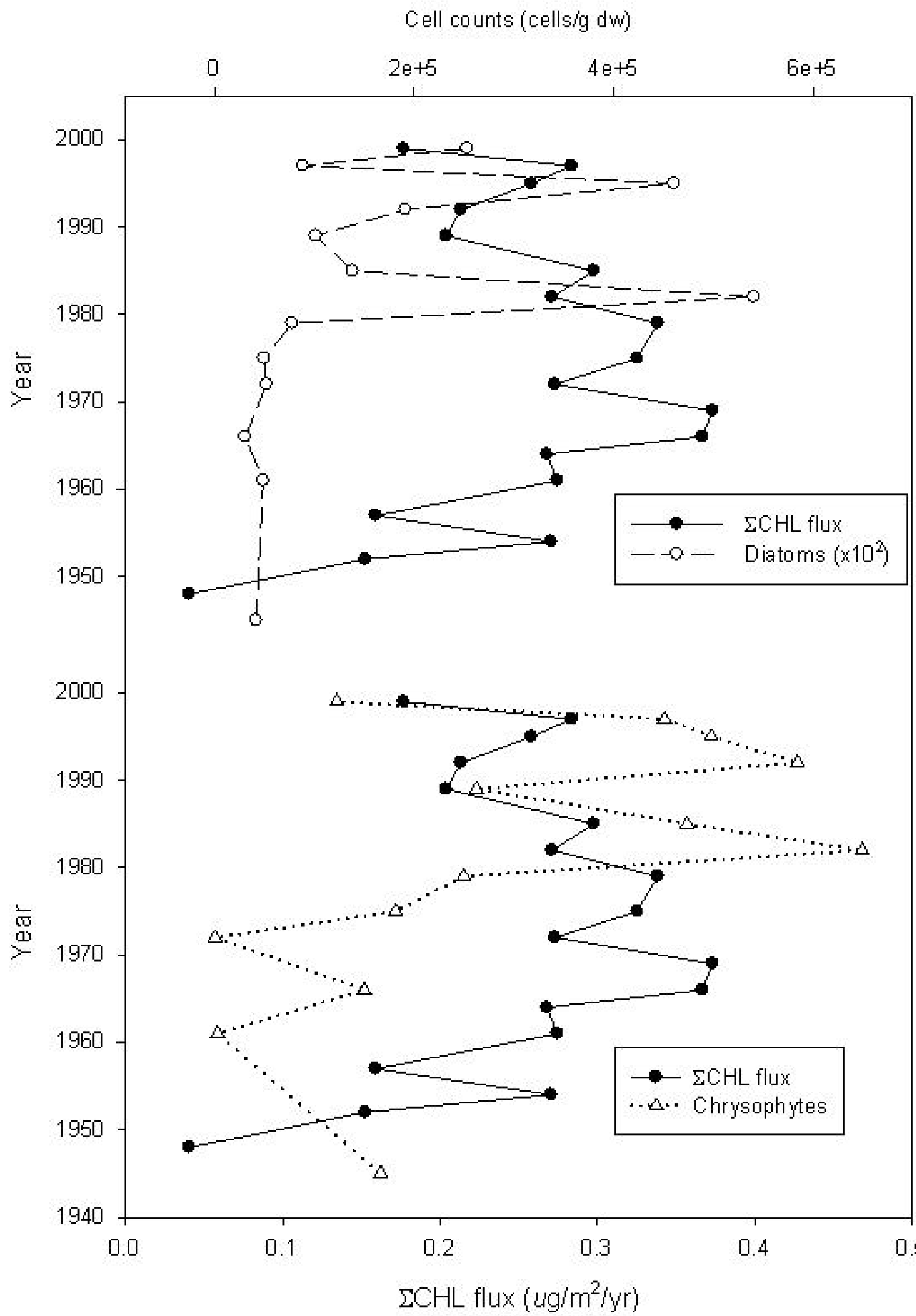

Figure 5.12 Phytoplankton cell counts (cells/g dw) over time vs. ΣCHL flux ($\mu g/m^2/yr$) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

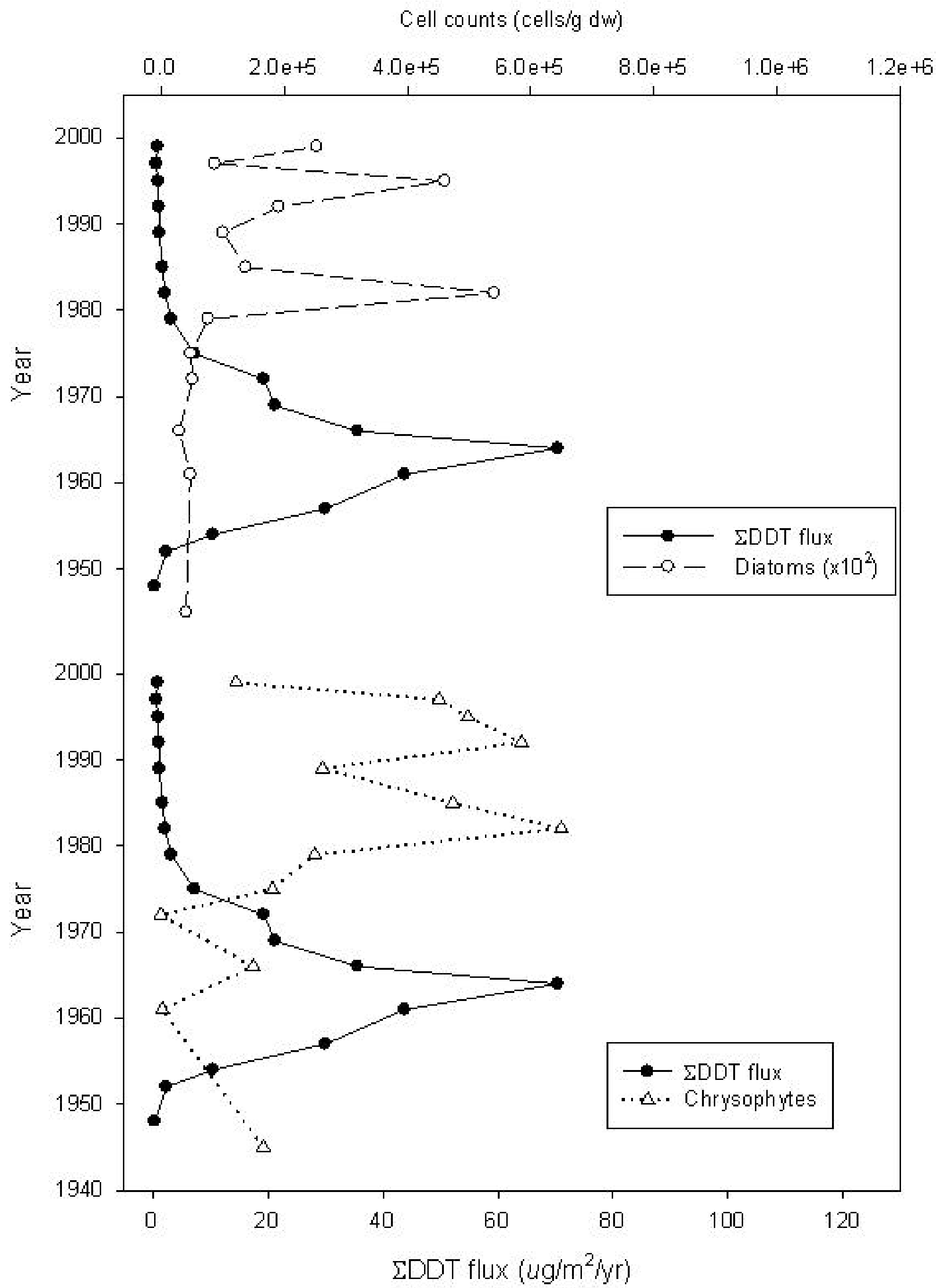

Figure 5.13 Phytoplankton cell counts (cells/g dw) over time vs. ΣDDT flux (μg/m^2/yr) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

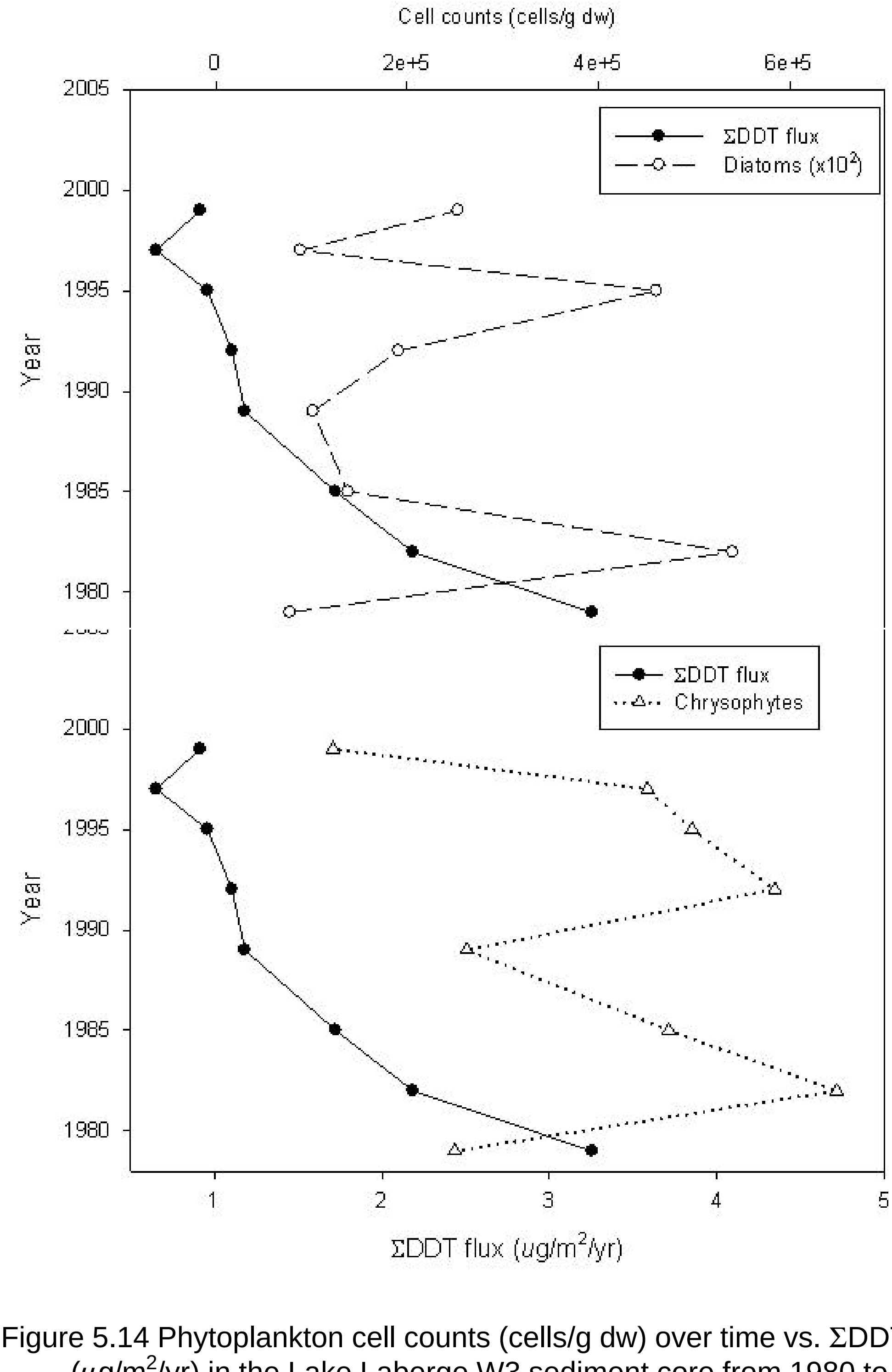

Figure 5.14 Phytoplankton cell counts (cells/g dw) over time vs. ΣDDT flux (μg/m^2/yr) in the Lake Laberge W3 sediment core from 1980 to 2000. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

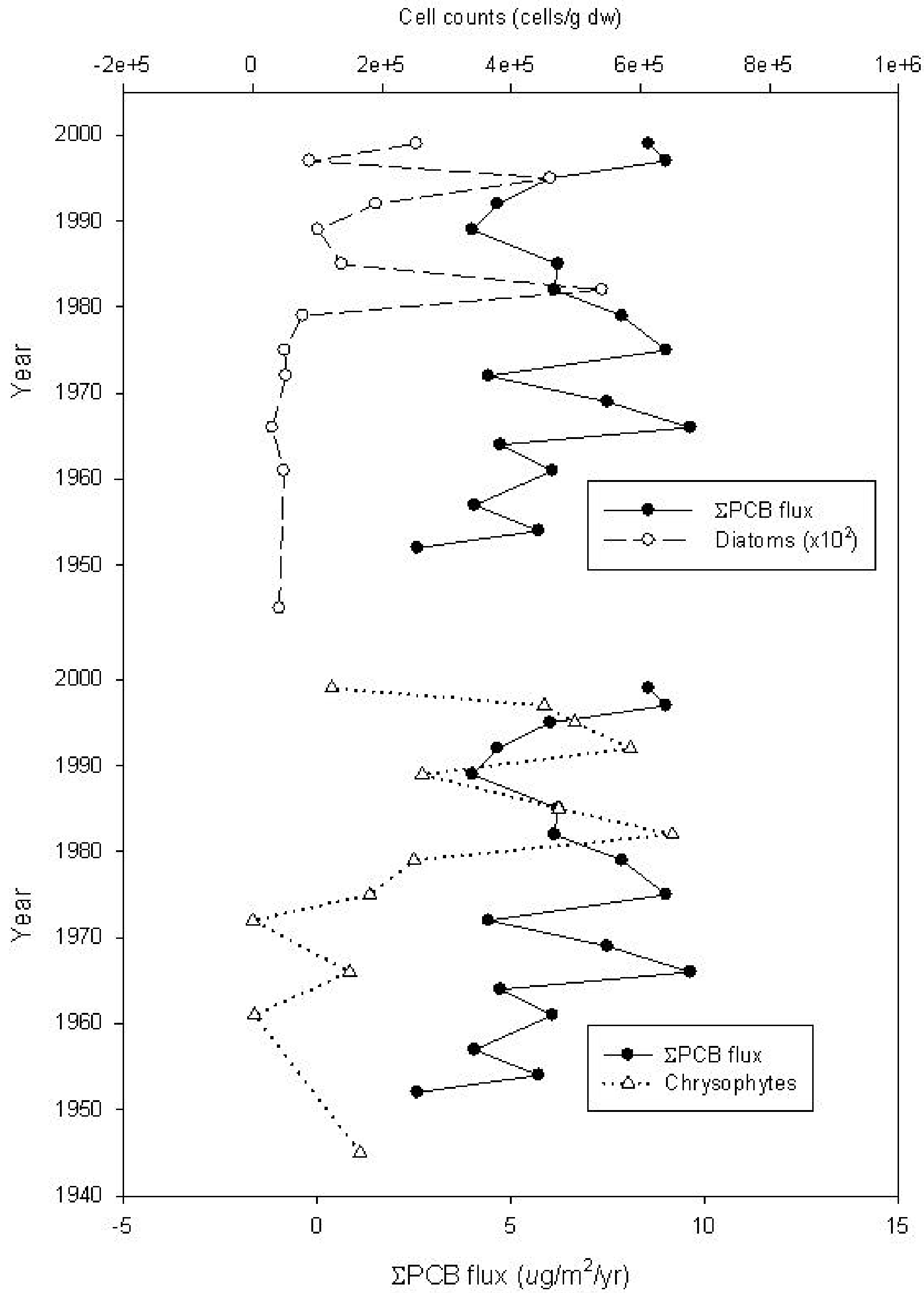

Figure 5.15 Phytoplankton cell counts (cells/g dw) over time vs. ΣPCB flux (μg/m^2/yr) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

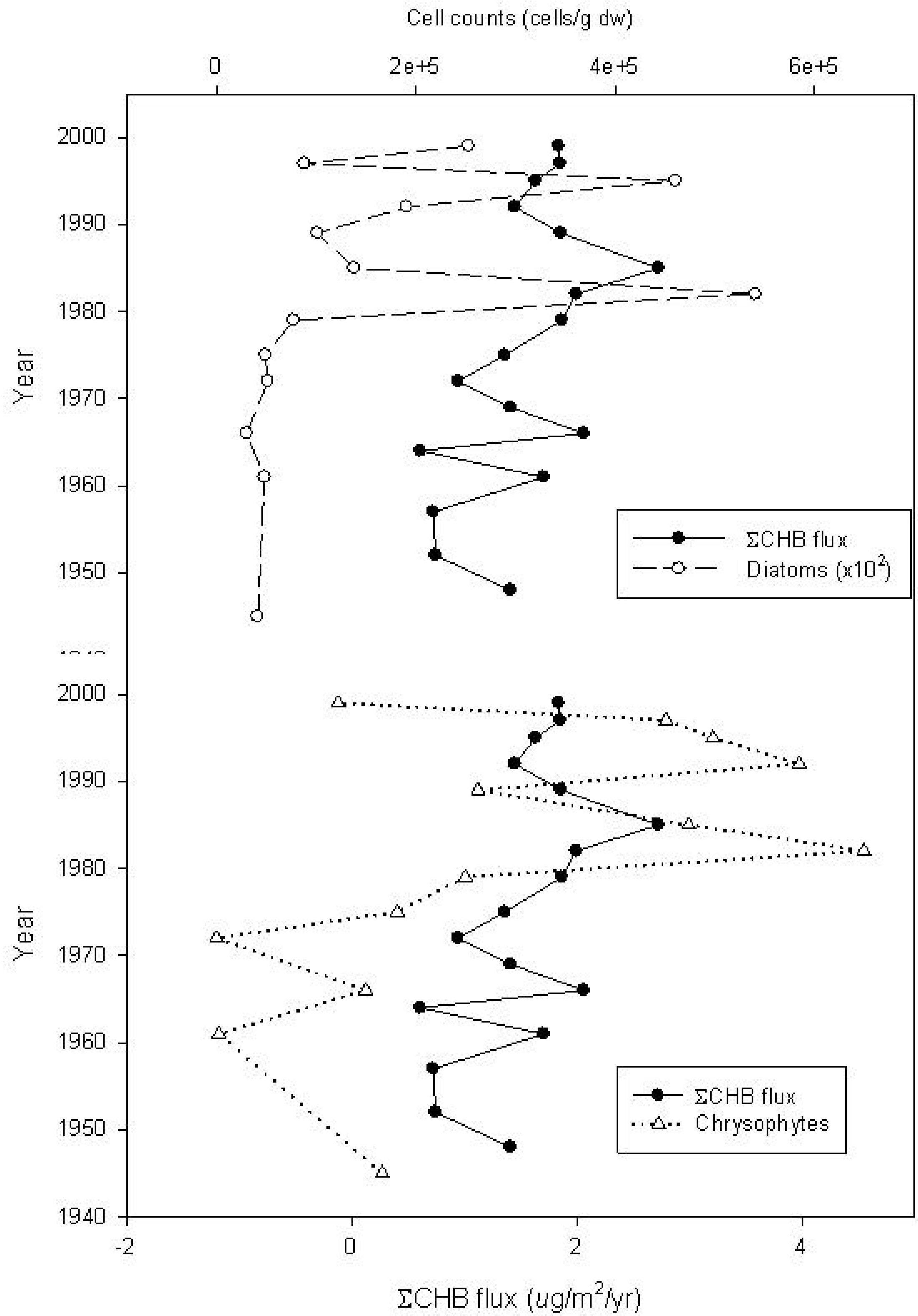

Figure 5.16 Phytoplankton cell counts (cells/g dw) over time vs. ΣCHB flux (μg/m^2/yr) in the Lake Laberge W3 sediment core. Phytoplankton counts included diatoms and chrysophytes (see Methods for taxa).

5.4.4 Phytoplankton Enumeration, Climate Trends and Nutrient Levels

Phytoplankton plotted against 30 years of growing degree-days (GDD) and annual precipitation data (1971-2001) have shown population peaks during periods of drought (defined as 2 or more years of warmer, more dry weather compared to previous annual conditions and historical averages). Phytoplankton assemblages showed major peaks in the following years: 1982 for both diatoms (Figure 5.17, Figure 5.18) and chrysophytes (Figure 5.19), 1992 for chrysophytes, and 1995 for all 3 major groups including cyanophytes (Figure 5.20) providing strong indications of increased overall primary production in Lake Laberge during the 1990s. Over the same period, drought conditions were evident. Growing degree-days peaked in the early 1980s for Carmacks while GDD for Whitehorse was on a steady increase since 1972. GDD peaked again in 1990 for both locations and for a third time between 1992 and 1996 registering the highest GDD in the 30-year data set. Mean annual rainfall remained consistent for 2 or more years between 1980-1984 and again from 1993-1997 before a 30-year record high was set in 1999 for the Carmacks region (Figure 5.18).

Although an increase in phytoplankton assemblages was evident, there were no apparent long-term temporal changes in lake nutrients compared to historical ranges (Table 5.2). Lake Laberge is still classified as oligotrophic.

Table 5.2 Historical and current water quality data from Lake Laberge surface water samples. TDS-total dissolved solids, DIC/DOC- dissolved inorganic/organic carbon, TP- total phosphorous, TDP-total dissolved phosphorous, TDN-total dissolved nitrogen. Single digits are mean values otherwise ranges are presented.

	1978-1980 [a]	1982 [b]	1993 [c]	2000
Conductivity (μS/cm)	56-370	85-105	113	73-106
TDS (mg/L)	46-75		62 (54-75)	
DIC (mg/L)				10.2 (9.6-10.6)
DOC (mg/L)			2.61 (1.55-6.12)	1.44 (1.3-1.8)
TP (μg/L)		3.8-6.7	5.1 (3.5-8.1)	6.2 (4.0-9.9)
TDP (μg/L)		1.3-2.5	2.5 (1.4-6.1)	1.8 (1.0-2.1)
TDN (μg/L)		57-118	66.2 (61.4-75.2)	88.0 (75-105)
NO_3^- (μg/L)	<10-38		16 (3.8-30)	13.4 (11-15)
NO_2^- (μg/L)	<5			1.0 (0-1)
NH_3 (μg/L)			15.2 (6.4-20.6)	
NH_4^+ (μg/L)	<5-32			20.0 (15-30)
Suspended N (μg/L)		5-10		15.4 (15-17)
Suspended P (μg/L)		2.0-5.4		4.4 (3-8)
Suspended C (μg/L)				170

[a] (Godin and Jack 1984)
[b] (Kirkland and Gray 1986)
[c] (Kidd 1996)

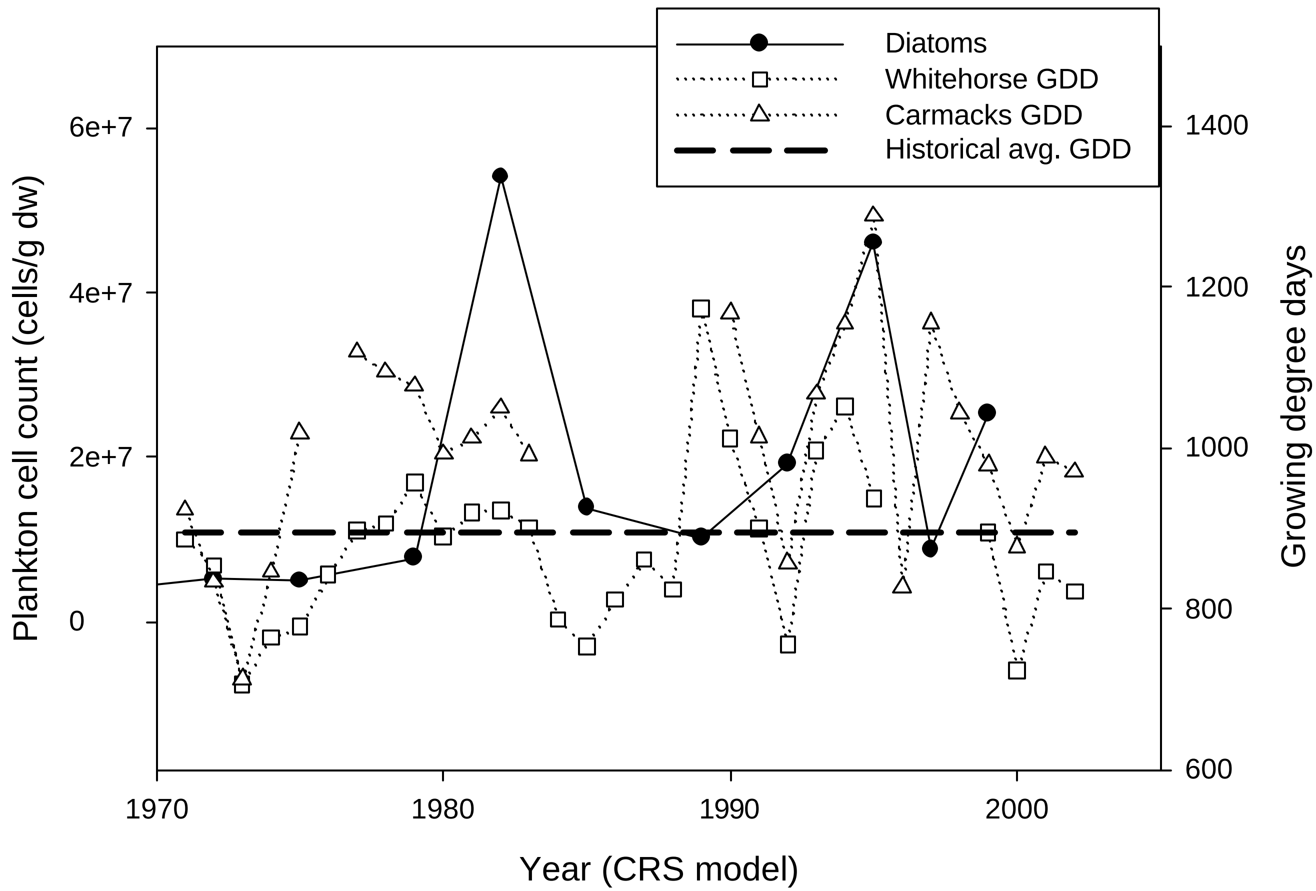

Figure 5.17 Growing degree-days above 5^{o}C north (Carmacks) and south (Whitehorse airport) of Lake Laberge and diatom concentrations (cells/g dw) over the past three decades. The Whitehorse airport 30-year (1971-2001) GDD mean is included.

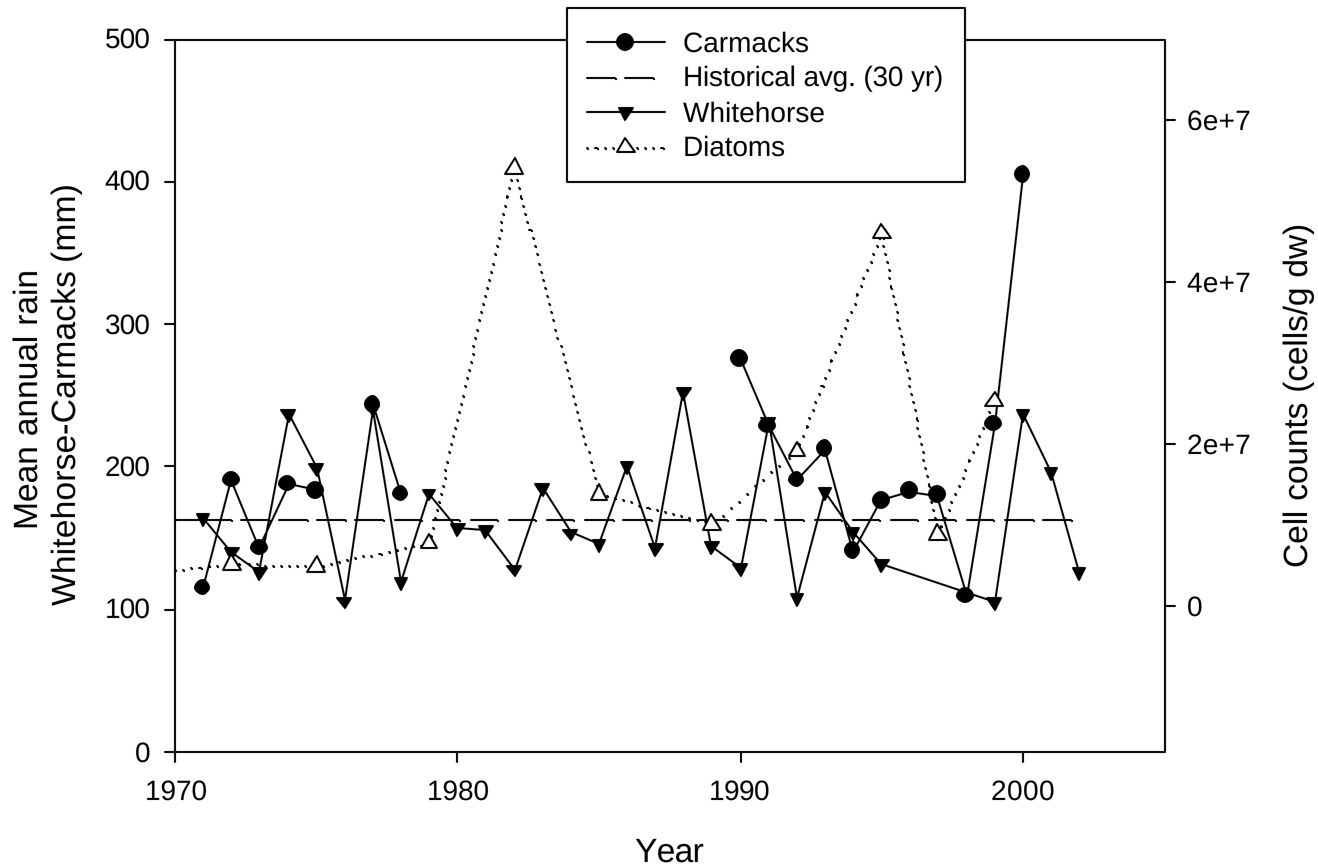

Figure 5.18 Mean annual rainfall (mm) north (Carmacks) and south (Whitehorse airport) of Lake Laberge and diatom concentrations (cells/g dw). The Whitehorse airport 30-year (1971-2001) rainfall mean is included.

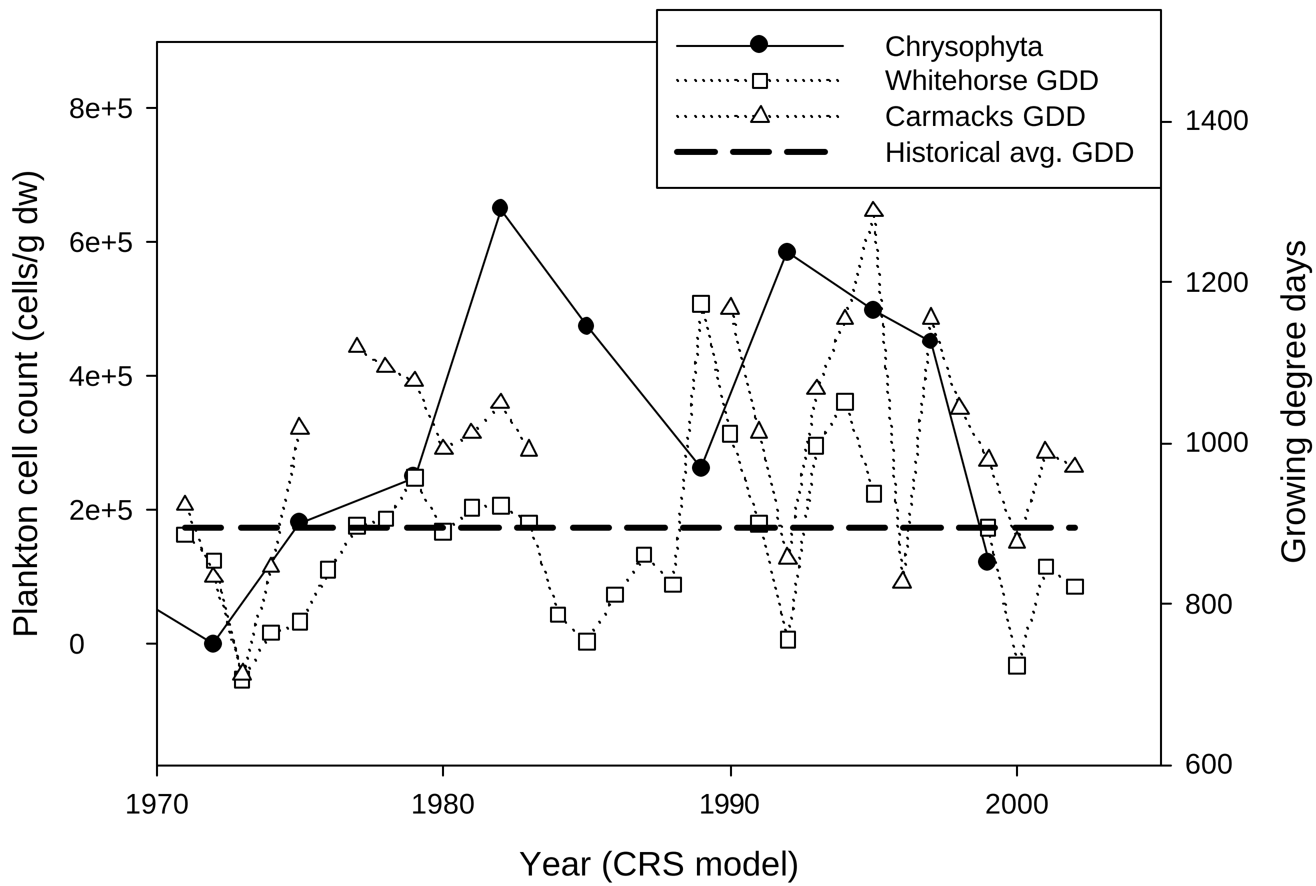

Figure 5.19 Growing degree-days above 5°C north (Carmacks) and south (Whitehorse airport) of Lake Laberge and chrysophyte concentrations (cells/g dw) over the past three decades. The Whitehorse airport 30-year (1971-2001) GDD mean is included.

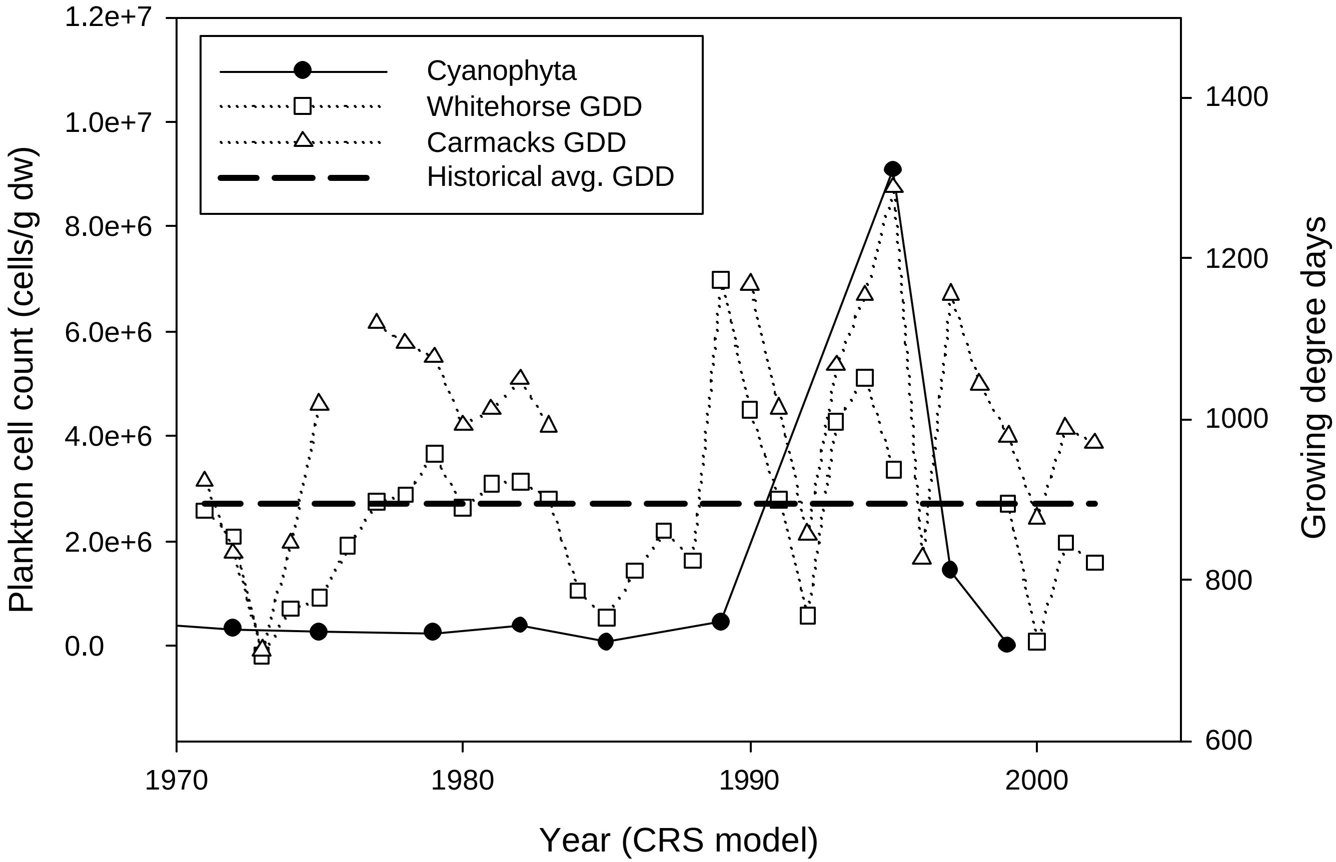

Figure 5.20 Growing degree-days above 5^{o}C north (Carmacks) and south (Whitehorse airport) of Lake Laberge and cyanophyte concentrations (cells/g dw) over the past three decades. The Whitehorse airport 30-year (1971-2001) GDD mean is included

5.5 Discussion

The sedimentation rates, focus factors and general OC trends in Lake Laberge core samples correlated with other analyses of Yukon lake sediment cores (Rawn et al. 2001). Sedimentation rates increased from the early 1900s with a peak around the mid-1950s, which coincides with the building of a dam upstream near Whitehorse (R. Gee, Yukon Energy Corp. pers. comm.) but there is little historical information to account for the remaining trends. The pre-1992 OC flux values conform to trends found in 7 other Yukon Territory lakes which was previously described by other researchers (Rawn et al. 2001). In general, fluxes of the OC in Lake Laberge decreased following international bans of pesticides and PCB chemicals and levels were similar to those found in Lake Winnipeg, a temperate climate lake (Rawn et al. 2001). However, the results of the sediment core survey from Lake Laberge in 2000 indicated that some OC groups (e.g. ΣHCH, ΣCBz, ΣPCB) were higher on average (1992-1999) compared to the historical average (1948-1999; Table 5.1). This partly conflicts with other reports on temporal trends in pesticides measured in northern atmospheric samples which observed general declines in α- and γ-HCH, although temporal trends for chlordanes and *p*, *p*'-DDE concentration were similar to the OC flux trends measured in Lake Laberge (Hung et al. 2002). Parallel increases in HCH, chlordane, toxaphene, PCB, DDT fluxes were observed in recent sediment samples from more remote Arctic areas and were attributed to variations in ice cover, primary productivity and precipitation, as influenced by climate changes (Macdonald et al. 2005; Stern et al. 2005). The recent increases in Lake Laberge sediment core contaminant fluxes for some OC may also be related to influences of the regional climate conditions and less directly related to atmospheric concentrations of these contaminants.

Annual variations in precipitation (snowfall and rainfall) could have significant impacts on the OC influx to a lake through atmospheric deposition as OCs follow the global distillation effect (Section 2.3; Goldberg 1975; Bidleman et al. 1989; Lockhart et al. 1992; Van Dijk and Guicherit 1999; Bidleman 1999). A large precipitation event (or wet seasons) would increase deposition of OC contaminants into both the terrestrial and aquatic zones (Van Pul et al. 1999; Van Dijk and Guicherit 1999). Such contaminant inputs to the lake sediment could occur through water inflow (Kirchner et al. 2000) or particulate sedimentation either from the atmospheric fallout (e.g. snow) or from the increase in terrestrial soil runoff from high flow events. The 1990s had above average temperatures and ended with a significant rise in annual precipitation, which was the second highest amount on record in 10 years (since 1990) for the region of Carmacks (north of Lake Laberge). The warm temperatures could affect glacial melting rates upstream or the elevated precipitation may have resulted in increased terrestrial runoff and higher input flow into Lake Laberge from the Takhini River increasing overall sedimentation rates, which was observed in the 1999 Laberge core slice. The 1999-dated slice had the highest sedimentation rate in 25 years coinciding with above average precipitation that year. Greater deposition of inflow particulates to the Lake Laberge sediments could result in the elevation in OC fluxes (as noted for the sampled surface slices) although the source of particulates could also be detritus fallout from the elevated phytoplankton populations noted in the lake (Figure 5.17 Figure 5.19 Figure 5.20). Heavy rain events would especially affect contaminants with higher water solubility such as ΣHCH. Li et al. (2002) observed that β-HCH, which has a lower Henry's law constant compared to the other HCH isomers, is more readily removed from atmosphere in precipitation than α or γ compounds. This may partly explain the recent rise in ΣHCH flux in 1999 as it related to a year of abnormally high precipitation in lower Yukon region.

Recent increases in contaminant fluxes may be related not only to precipitation events but also to the origin of the atmospheric particulates from areas such as India, eastern Asia and Russia (Welch et al. 1991; Barrie et al. 1992; Van Pul et al. 1999; Bidleman 1999). Enantiomer ratios of a fresh technical chlordane mixture can be used to trace pesticide emissions compared to the weathered compounds usually found in the environment (Bidleman et al. 2002). Recent increases in the *trans-: cis-*chlordane ratio in the core from Lake Laberge (Figure 5.6) coincide with the intermittent observations of increases in *trans-cis* chlordane ratios in air samples from Alert, NWT from 1995-1997 indicating that a recent use (early to mid 1990s) of fresh chlordane may have occurred (Hung et al. 2002). The source of chlordane in the Arctic air samples was unknown but back trajectories indicated the air masses originated from sectors of Eastern and Western Russia (32.6%) and Europe/North Atlantic (28%) rather than from North America. The historical chlordane ratios in Arctic sediment cores have supported the source-trend data (new or weathered material) observed in air concentrations (Bidleman et al. 2003; Bidleman et al. 2004) while atmospheric loadings have also been related to sediment deposition in other areas (Carrera et al. 2002). A future analysis of back trajectories of air currents may reveal the source of particulates in Yukon precipitation in the late 1990s and provide evidence for the hypothesis that Lake Laberge OC fluxes are related to OC atmospheric trends.

This hypothesis may also explain some of the rationale behind changes in other OC compounds. The chlordane isomer ratios resulted in a large rebound towards equal proportions of *cis*- and *trans*-chlordane from 1997 to 1999. This ratio has been declining in other areas due to the preferential degradation of the *trans*-chlordane isomer by soil microbes and possibly photochemical reactions (Bidleman 1999; Bidleman et al. 2002; Bidleman et al. 2003; Bidleman et al. 2004). In addition, the isomer ratios are not spatially consistent across North America and some locations show a high degree of deviation in temporal trends. Bidleman et al. (2004) noted that the relative contributions of chlordane from soil derived ‘primary’ emissions and from previously deposited and revolatilized ‘secondary’ emissions could not yet be determined. The recent resurgence of the *trans*-chlordane compound in the core from Lake Laberge may be influenced by the atypical year of high precipitation late in the 1990s by bringing greater amounts of fresh chlordane into the Yukon watershed. Welch et al. (1991) reported on fluxes of pollutants (PAH, ΣDDT) in Arctic snow samples collected from a single fallout event demonstrating that a one-time atmospheric deposition of contaminants in an area could account for a significant percentage of the total annual accumulation. Proportions of parent *p,p'*-DDT and its subsequent degradation product, *p,p'*-DDD, are also declining in sediments in Lake Laberge.

This is likely due to the length of microbial degradation over time producing a larger amount of degraded product in older sediments such that the ratio of *p, p'*-DDT to *p,p'*-DDD increases towards surface slices. Changes in the ratio could also be due to either a slowdown of the degradation of the parent DDT compound or a recent increase in DDT to the Lake Laberge area. It is unknown whether regional climate shifts may have increased microbial degradation or whether atmospheric inputs may have increased in recent years.

The effects of the high precipitation in the late 1990s can also be extended to both PCB and toxaphene in the same manner as previous contaminants. Evidence of this hypothesis in Lake Laberge is shown in the elevated levels of mono/di/triCBs from sediment slices (1995 to 1999; Figure 5.9) compared to other more chlorinated, and less water soluble PCB (Table 2.1). PCB and toxaphene compounds are also bound to particulates in the atmosphere which can be deposited into aquatic basins during years of high precipitation (Barrie et al. 1992). However, the toxaphene flux to sediments in Lake Laberge only mildly increased between 1997 and 1999. These values were still lower than mid 1980 values, which were elevated for several years immediately following the ban of toxaphene in the U.S. in 1983 (Ware 2000). None of the contaminants had greater increases in fluxes than the most water soluble compound (β-HCH) providing evidence that the abnormally high precipitation event of 1999 likely contributed to the recent OC flux patterns observed in the Lake Laberge core samples.

The 1990-1999 Yukon weather records showed some of the highest consistent annual temperatures north and south of Lake Laberge, as measured by growing degree-days above 5^{o}C (Figure 5.17-5.20), when compared to the 30-year history. Changes in worldwide climates through elevated temperatures as noted in the Arctic Climate Impact Assessment (ACIA 2004) may result in higher atmospheric loadings to colder regions via global distillation (Goldberg 1975; Van Dijk and Guicherit 1999). PCB and chlorinated pesticides have seasonal and even diurnal cycles in ambient air in response to fluctuations in air temperatures and even humidity (Bidleman 1999). Warmer climates also have an effect on sub-Arctic, glacially fed lakes. Lake Laberge exhibits riverine characteristics due to its intermediate residence time, above average turbidity and its long, relatively narrow body (4 km at the widest), which carries a strong current (Kirkland and Gray 1986; Wetzel 2001). Its water is supplied 19% by Takhini River and during periods of high flow, Lake Laberge receives glacially deposited sediments from the Takhini's eroding banks. This often results in turbidity in Lake Laberge created by sediment-laden water carried by a strong current down the eastern side due to the lake morphology and inflow from the Yukon River (Kirkland and Gray 1986). It has been shown that glacial sediments retain OC compounds from high altitude deposition (Blais et al. 1998) and, as such, warmer summers may contribute to elevated glacial melt and hence increased OC laden particulates into Lake Laberge. However, an increased particulate load may also provide a mechanism for OC removal from water depending on the organic content of the particulates thus reducing bioavailability of contaminants by sequestering OC to the sediment.

Research on Great Lakes sediment has shown that overall the sedimentation rates, suspended sediment concentrations and sediment organic content are ineffective in predicting PCB and DDT concentrations in fish, although they are important in determining bioavailability of material (Rowan and Rasmussen 1992). It should be noted that none of the OC levels measured in Lake Laberge sediment slices through the 1990s had any significant decreases to suggest that abiotic OC concentration trends were responsible for the recent contaminant declines observed in Lake Laberge biota (Chapter 4).

Both abiotic and biotic factors influence the deposition of OC to sediments; the latter affects fluxes because contaminants partition into phytoplankton or bacteria before being deposited on lakebeds through processes such as excretion or death (Rowan and Rasmussen 1992; Larsson et al. 1998). Examining the phytoplankton concentrations in the sediment core can provide insights to the growth patterns of these plants over time and whether their fluctuation in populations have any correlation with OC that are deposited into lake sediments by acting as sinks (removing contaminants from the water compartment and into the sediment). Studies have concluded that adsorption of PCB onto suspended solids, which would include plankton detritus, followed by deposition of these particles to the lake bottom is a major mechanism for transport into sediments (Formica et al. 1988), although other researchers suggest the process is relatively inefficient (Macdonald and Metcalfe 1989). Sediments from Clear and Scugog Lakes (Ontario) that were previously analysed for ΣPCB, were shown as having a different ratio of congeners compared to biota such as zooplankton (Macdonald and Metcalfe 1989) suggesting that a correlation between plankton populations and sediment OC concentrations would be unlikely. The results of the Lake Laberge samples showed that phytoplankton assemblages did indeed increase to higher levels in the 1990s, specifically 1995 for cyanophytes and diatoms, while chrysophytes maintained elevated populations from about 1992 to 1995. Correlations of these population increases with recent increases in OC fluxes in sediment samples were inconsistent, with fluxes being slightly higher for ΣHCH, ΣCHL, ΣPCB and ΣCHB after the growth event around 1997 (Figures 5.10-5.16). During the lone comparable event in 30 years (approximately 1982) that recorded an equivalent increase in phytoplankton populations in Lake Laberge, sediment OC fluxes had measured only mild increases in ΣCHL and ΣPCB (9% and 1% respectively) and a larger increase (36%) in ΣCHB three years following the growth event (1985). The phytoplankton populations were temporally correlated with some fluctuations in OC levels over time, although no consistent patterns occurred over the entire 51-year span. The few correlations in this core might be associated with periods of high production and usage (Stern et al. 2005) combined with as of yet unknown effects from climate variation and abiotic (e.g. atmospheric, terrestrial runoff) inputs. For example, the 1982-year of phytoplankton population increases correlated with a period of maximum toxaphene flux to Lake Laberge.

The theory of contaminant biomass dilution in Lake Laberge biota (presented in Chapter 4) can be supported with evidence of primary production increases during periods of contaminant declines in higher trophic levels. Such evidence was obtained by measuring phytoplankton remains from sediment cores. A significant increase in 1982 phytoplankton assemblages, compared to historical numbers (Figures 5.17-5.20), occurred in Lake Laberge, coinciding with a period of drought (low mean precipitation and higher than average GDD) from 1979-1983. A similar drought period from 1992-1995 also coincided with a significant increase in phytoplankton assemblages. Although there were no collections of fish for OC levels in 1982, previous results (Chapter 4) have shown significant declines in fish and zooplankton OC levels (Chapter 4) from 1992 to 2001, after the elevation in phytoplankton populations were observed in the sediment core. This finding was similar to other research from the Great Lakes. Herring gull eggs analysed for contaminants across several Great Lakes had significantly non-random, temporal fluctuations in OC concentrations, which were not linked to variations in the contaminant levels of abiotic samples (Smith 1995). Instead, the study suggested that the temporal variations were most consistent with the mechanism of biomass dilution. Warm spring weather, which was conducive to phytoplankton growth, provided a food base of less contaminated phytoplankton resulting in decreased OC concentrations in fish and further up the food web to predatory herring gulls and their eggs (Smith 1995). The concept of biomass dilution in phytoplankton is supported by other research (Lederman and Rhee 1982; Swackhamer and Skoglund 1993; Larsson et al. 1998). Research on phytoplankton population fluctuations as affected by changes in climate (Gajewski et al. 1997; Findlay et al. 2001) may provide some insight into the link between contaminant fluxes in lake biota in future years.

Dry seasons (low precipitation) will decrease lake turbidity resulting in warmer surface water temperatures, clarification of the water column allowing for more light penetration forming a deeper euphotic zone (Schindler et al. 1996; Schindler 2001; Edmundson and Mazumder 2002). Increases in phytoplankton biomass under such conditions have also been observed (Schindler et al. 1996; Findlay et al. 2001). Kirkland and Gray (1986) estimated that if flushing rates were rapid enough, then algal biomass accumulation would likely be reduced in Lake Laberge. Therefore in contrast, a period of lower flushing rates in Lake Laberge during drought years, may provide the expectation for algal accumulation (higher primary production).

Higher plankton production and biomass relates to a larger food base for predators higher in the food chain equating to greater fish yields (Oglesby 1977; Shortreed and Stockner 1983; Downing et al. 1990). Edmundson et al. (2003) concluded that the potential for decreased fish stock production is greater when elevated particulate loads reduce light penetration and primary productivity during periods of warmer climates and increased runoff in glacially fed sub-Arctic lakes. Conversely, the potential for increased fish stock production should be greater when low particulate loads increase light penetration and primary productivity during periods of cool climates and decreased runoff in glacially fed sub-Arctic lakes. In addition, prolonged warm, dry conditions causing lower flows during spring melt in temperate lakes have been observed to reduce some nutrient inputs (dissolved organic carbon, silica and phosphorous) into receiving water bodies (Schindler et al. 1996) which would affect primary productivity and ultimately fish production. Hence the effect of climate on primary and fish productivity must continue to be examined.

Although Laberge is classified as an oligotrophic lake (Kirkland and Gray 1986), it is hypothesized that its spring and early summer phytoplankton growth is more limited by temperatures and light than by nutrients as observed in other sub-Arctic glacial lake studies (Edmundson et al. 2003). Wetzel (2001) reported increases of plankton molting rates and brood production that occurred with increasing temperatures from various studies. Findlay et al. (2001) observed that a drought in the 1980s (decreased precipitation and above average air temperatures measured by HDD or heating degree days) coincided with an increase in plankton biomass and diversity despite decreased nutrient inputs. In addition, the authors reported a shift in species composition to a greater proportion of chrysophytes, an observation also made for Lake Laberge in 1982 and 1992-95. Analyses of nutrients in Lake Laberge have shown no evidence of changes as of 2000-2001 compared to historical measurements (Table 5.2) to account for the increases seen in phytoplankton growth. Total phosphorous levels, which are linked to phosphate concentrations (Hudson et al. 2000), had no relationship with chlorophyll concentrations (Kirkland and Gray 1986) giving an indication that phosphorous in the form of phosphate, which is an important limiting factor for phytoplankton growth (Hudson et al. 2000; Wetzel 2001), is not the primary limiting factor in Lake Laberge. Previous observations showed that phosphorous levels were higher on average in Lake Laberge compared to Kusawa and Quiet Lakes (Table 3.1) and that Lake Laberge was the most productive lake out of three other Yukon lakes surveyed (Kirkland and Gray

1986). It could be expected that with future drought periods, phytoplankton may resume above average growth leading to further biomass dilution of OC contaminants in Lake Laberge.

The effects of temperature and precipitation on particulate contaminant loads to the Yukon region are unknown, but greater water clarity, greater light penetration (deeper euphotic zone) and warmer temperatures in Lake Laberge are the likely causes of high production during the 1990s. Hence biomass or growth dilution and species diversity, partly through closure of the commercial fishery and climate variations, are the most probable causes for the declines in contaminants in Lake Laberge biota (Chapter 4) compared to any influence provided by atmospheric deposition or contaminated sediment loads from glacial runoff. The conclusion that OC concentration changes in a lake food web are highly influenced by phytoplankton growth, as affected by annual climatic conditions, rather than point source (aquatic or terrestrial) or atmospheric influx of contaminants, is also supported by other research (Lederman and Rhee 1982; Smith 1995).

5.6 Summary and Conclusions

The study has shown that OC pesticides and PCB levels in a sediment core from Lake Laberge have decreased since the North American ban of these chemicals, yet concentrations in the past decade remain relatively stable and have even shown small increases in fluxes. These general trends conform to data reviewed from other arctic and sub-arctic sediment cores (Rawn et al. 2001; Stern et al. 2005). Effects of recent climate variations in the Lake Laberge area are likely responsible for part of the fluctuations seen in sediment contaminant levels. Above average precipitation events in the late 1990s could contribute to increased OC loading to the lake through elevated atmospheric deposition and the presence of OC bearing particulates from terrestrial runoff. In addition, warmer climates through the 1990s may result in elevations in glacial runoff due to melting further contributing to OC from the high altitude areas.

Conversely, the years of low rain and higher annual temperatures have likely contributed to the increased growth of phytoplankton in the mid-1990s. During years of reduced precipitation there is an expected decrease in the amount of particulate matter influx into the lakes thereby increasing water clarity and phytoplankton growth (Schindler et al. 1996). Above average annual temperatures, as measured by growing degree-days in Lake Laberge, are expected to coincide with a greater occurrence of incident solar radiation (Wetzel 2001) resulting in more heat and light available to primary producers for growth. These elevated levels of phytoplankton seem unimpeded by the levels of nutrients in the oligotrophic lake.

Numerous physical, limnological and biological factors combine in various magnitudes to affect the movements of OC through the Lake Laberge food web; however, the dramatic rise in phytoplankton populations during the mid-1990s is thought to be the most significant factor in determining OC concentrations in these organisms and their predators.

6. GENERAL DISCUSSION

Data on temporal changes in organochlorine pesticides and PCB in northern ecosystems has long been lacking (CACAR 2003a). Temporal trend and spatial contaminant studies are well documented for a few other areas, such as the Great Lakes (Borgmann and Whittle 1991); however, whole aquatic food web studies combined with abiotic samples and climate analysis are rare. The purpose of this study was to gather biotic and abiotic samples from sub-Arctic lakes to begin a temporal trend assessment of contaminant concentrations. In addition, a study in 1992-1994, Kidd (1996) setup a baseline for a 10-year comparison between contaminant levels in a whole lake food web. Kidd's study examined OC concentrations in the food web of Lake Laberge as they related to trophic levels, stable isotopes and various morphological and biochemical variables (length, weight, age, lipid content) of the biota. Recent analyses of the sentinel species, lake trout and burbot, indicated that significant declines in OC levels had occurred in these Lake Laberge fish over a 10-year period. In order to determine whether OC changes were specific to Lake Laberge or whether changes in OC levels were occurring in tandem with other lakes in the region, the contaminant levels in sentinel species from two other Yukon lakes, Kusawa and Quiet Lakes, were reviewed in comparison. The food web of Lake Laberge was reviewed in further depth to elucidate probable causes of the significant decline in OC concentrations in fish. Concurrent measurements of climate trends and sediment cores (phytoplankton assemblages, OC levels) from the focus lake supplemented the data.

Several hypotheses were examined to explain these recent declines in OC concentrations in fish. This included trophic level changes, biomass or growth dilution, lake specific ecosystem factors, historical deposition trends and climate impacts.

Chapter 3 examined the temporal trends of OC concentrations in lake trout and burbot from three Yukon lakes to determine if the declines in OC from Laberge fish were specific to that lake or whether the decreases were occurring across the region. Although contaminant concentrations generally decreased in the six major OC groups (ΣCHL, ΣDDT, ΣHCH, ΣCHB, ΣPCB and ΣCBz) in the sentinel fish species (species of consistent annual observation and collection), the changes in Kusawa and Quiet Lakes were of lower magnitude when compared to Lake Laberge. This suggests that additional factors, such as post-commercial fishery effects, likely caused the larger temporal declines in contaminants in the Lake Laberge fish.

Trends in OC varied among fish species indicating that the factors affecting OC concentrations in these animals were not the same across species. Decreases in contaminant concentrations in lake trout were evident in all 3 lakes, yet similar patterns did not occur in burbot. There was no evidence from Kusawa or Quiet Lake to support a hypothesis that OC concentrations have declined in burbot. Significant contaminant trends may be masked by the potential interannual variation (Bignert et al. 1993) especially for Kusawa and Quiet burbot due to the limited samples for this species from some years. Based on larger sample sizes in Lake Laberge, burbot showed a decrease in some OC contaminants over the 10-year period, but this may have been caused by factors specific to the Laberge food web or due to the ecology differences (benthic vs. pelagic, piscivorous vs. omnivorous) between burbot and lake trout (Chapter 4).

In contrast, the temporal declines in lake trout contaminant levels in these Yukon lakes were significant in all lakes. OC concentrations showed positive correlations with lake trout body size for recent years and positive correlations with lipids (for most years) in Lake Laberge and Quiet Lake while none of the parameters examined (age, length, weight, condition factors, tissue lipid (%)) had any correlation with decreases in Kusawa Lake fish. Although some significant temporal changes in these lake trout parameters have been observed, the OC declines were only partly explained by changes in these morphological variables for Lake Laberge trout. Since changes in OC levels occurred in lake trout across all three Yukon lakes, and those contaminant declines were only partly explained by fish population and correlating morphological characteristics, it is probable that a broader regional variable such as climate influence on components specific to each lake ecosystem (e.g. differences in dominant and diversity of plankton species), affected these temporal trends in OC concentrations for this fish species.

The effects of variation in annual climate was hypothesized as causing a change within the food webs through primary production fluctuations, and ultimately OC biomagnification potential, within lakes in the Yukon region. This phenomenon has been noted in other studies (Smith 1995; Hebert et al. 1997).

Above average annual temperatures and below average precipitation, from approximately 1992 to 1997, has coincided with declines in zooplankton OC levels and increases in phytoplankton populations (Lake Laberge samples; Chapters 4 and 5). This provides evidence towards the hypothesis that climate conditions during the 1990s have played a role in the OC concentration changes in Yukon lakes. Although recent climate variation was likely responsible for the changes in lake trout OC concentrations for Quiet and Kusawa Lake through plankton biomass dilution or changes in plankton community composition, further review of sediment core samples, other fish species and phytoplankton assemblages for these lakes is needed to review this hypothesis.

Contrasting lake trout OC trends in the three Yukon lakes, it appears less likely that climate fluctuations are having as large an influence on OC concentrations in the more benthic dwelling species such as burbot. It is more probable that observed contaminant trend differences between the lake trout and burbot are due to species-specific ecology differences and/or lake specific reasons (e.g. burbot and lake trout food preferences and foraging habitats-pelagic vs. benthic, percentage composition of fish and invertebrate prey consumed, interspecies competition, plankton community compositions between lakes). Concentrations of contaminants in Laberge samples decreased more than the levels in trout from Quiet or Kusawa, which coincided with significant changes in fish population and morphological characteristics. This supports the hypothesis that factors other than plankton biomass dilution, which are specific to the Lake Laberge food web, may be compounding the observed OC declines from biota in this lake.

Chapter 4 reviewed a more in-depth analysis of the Lake Laberge OC contaminant trends by examining the whole aquatic food web. It was hypothesized that shifts in the trophic status or food sources for biota in Lake Laberge occurred following the closure of the commercial fishery in 1991 causing predatory fish to feed at lower trophic levels due to increased competition, subsequently reducing biomagnification of OC contaminants (de Graff and Mychasiw 1994; Kidd 1996). However, no trophic level changes or changes in fish food sources were observed with stable carbon and nitrogen isotope analyses for fish from Lake Laberge with two exceptions. Burbot marginally decreased in its calculated trophic level (TL) while northern pike was more enriched in $\delta^{15}N$ (using the defintion of a trophic level (3-5‰)) by a half level when comparing 1993 to 2001 data, partly explaining the small decreases in burbot and increases in northern pike OC levels. Five of seven species of fish in Lake Laberge have increased in abundance, whereas two species, lake whitefish and round whitefish, have notably declined in populations (Foos 2001), possibly through predation or increased resource competition. The body size of some species (lake trout round whitefish and least cisco) has increased over the 10-year period and body sizes have been linked to changes in contaminant levels (Chapter 4). This suggests that growth rates are influencing the contaminant burdens for some fish in Lake Laberge.

It was hypothesized that changes in climate affecting zooplankton growth was the most probable cause for declines in OC levels through biomass dilution and that concurrent changes in the zooplankton community composition could be influencing the movement of contaminants in the biotic compartment to predators. Under this hypothesis, declining levels of OC contaminants in Lake Laberge zooplankton, a primary food source for forage fish, would be expected to have an impact on OC concentrations in predators higher in the food chain (Rasmussen et al. 1990). Whether the zooplankton community composition has changed from climate factors or elevations in fish predation from larger fish populations, the change in dominance to *Diaptomus pribilofensis* over *Cyclops scutifer* from 1992 to 2002 could have a significant influence on the movement of contaminants through the food web through physiological factors (e.g. metabolism, OC uptake/elimination or clearance, growth rates) unique to each species (Chapter 4); however this hypothesis requires further research at the plankton level. The change in species dominance may also be an important factor influencing the growth and body size changes noted for some Laberge fish. The complex food web interactions between predator and prey in Lake Laberge could not be directly assessed; however, through the morphological parameter measurements, fish population and zooplankton community changes observed, proof of the OC contaminant declines in Lake Laberge as influenced by the closure of the fishery or climate are more evident.

Chapter 5 examined OC levels and phytoplankton counts in dated sediment cores, coupled with Environment Canada climate data to provide supporting evidence for the OC trends reviewed in Chapter 3 and 4 and to determine whether there have been changes in OC deposition to Lake Laberge. The sediment core results showed that OC levels deposited to the sediments have declined since the ban of these substances, a result which has been shown in other studies (Rawn et al. 2001). However, there were no indications of recent declines in deposition to account for the changes in fish contaminant concentrations and, in contrast, some OC fluxes increased in the late 1990s. Thus, there was no evidence to support the hypothesis that a change in atmospheric or runoff deposition was responsible for the decreases in OC levels in the lake biota.

Climate data have already shown correlations of below average precipitation and above average temperature years coinciding with spikes in phytoplankton populations and hence, primary production. The hypothesis of biomass dilution of contaminants for plankton is supported by this data as the reduction in measured zooplankton OC followed a period of high phytoplankton production.

Nutrient concentrations were measured in conjunction with plankton sampling to examine any hypotheses regarding human influences on Lake Laberge primary production through sewage dumping either locally or upstream from the city of Whitehorse. The results of the nutrient analysis showed that concentrations were well within historical measurements negating the aforementioned hypothesis of sewage dumping or other hypotheses that involve significant fluctuations in nutrients as a major variable.

The organochlorine pesticides and PCB have high affinities for partitioning in living organisms and as such, are subject to biomagnification. This process is highly influenced by the contaminant movements at the base of the food web. Strong influences on biomagnification of OC in a food web comes from individual species characteristics such as physiological differences (fat content, body size), behavioural adaptations (feeding preferences, responses to changes in prey or predator populations), changes in ecosystem parameters (increased fish populations resulting in competition for resources, changing age structure of species) all which are indirectly or directly controlled through climatic conditions (primary production, nutrients, biomass dilution) and anthropogenic influences (commercial fishing). The recent declines in OC contaminants for three Yukon lakes are coupled to annual climate fluctuations (e.g. temperatures, precipitation), influences specific to each lake (e.g. plankton community composition) and influences specific to each species (e.g. biomass dilution). Lake Laberge in particular has been highly affected by a recent closure of a commercial fishery allowing for increases in fish populations and growth. The combinations of the aforementioned variables are the most probable reasons for the recent declines in OC levels in biota from the Yukon study lakes. As succinctly stated by Hebert and Haffner (1991), "Although chemical and physiological parameters may determine which contaminants have the potential to bioaccumulate, it is the regulation of exposure through ecological processes that will determine the degree to which that potential is realized".

7. SUMMARY AND CONCLUSIONS

Temporal shifts, primarily declines, in organochlorine contaminants are evident in biota from three Yukon study lakes. The weight of evidence from this study shows the factors responsible for causing these changes include lake and species-specific biological conditions (Chapter 3 and 4) as well as wider ranging regional effects like climate fluctuations (Chapter 5). Variations in regional climates will have significant impacts on the primary production of these northern lakes. Increased amounts of light and heat during periods of drought likely increased the growth rates of primary producing plankton, as seen in historical phytoplankton populations (Chapter 5), thereby causing a growth or biomass dilution effect in contaminate concentrations at the base of the aquatic food webs. With a lower base of contaminant concentrations in the food web, the effect will become evident at higher trophic levels over time. Regional climate shifts may have caused the community composition changes in plankton species such that unique plankton characteristics (mucilaginous sheaths, fat contents, body sizes), which are influential on OC concentrations, may become variable within and between lakes. Such parameters can directly affect the water-body partitioning of contaminants and modify bioavailability of OC compounds for predators. It is suspected that such shifts in the primary production of Quiet, Kusawa and Lake Laberge are responsible for some of the recent declines seen in OC in fish, as the decreases cannot be completely accounted for by other factors. There is no evidence to support the hypothesis that these declines are due to decreases in atmospheric or other abiotic inputs of OC to the lakes (Chapter 5). Further research on the food webs and sediment cores of Quiet and Kusawa lake, with special attention to the plankton dynamics, would be of benefit in understanding the biological interactions specific in each of these lakes for future contaminant trend studies.

Lake Laberge is unique among Quiet and Kusawa lakes due to its closure of the commercial fishery in 1991. Although Quiet Lake also maintained a fishery until 1989, Lake Laberge historically sustained a much larger fishing pressure. The closure of the fishery has since allowed fish populations to increase in size for seven out of ten species monitored (Chapter 4). There has been an increase in body sizes for some species along with changes in lipid contents and age structures, most likely due to the increased competition for resources in the lake. The growth dilution effect was evident for OC contaminants in several fish species. These changes in morphological parameters, population characteristics, species interactions and lower OC at the base of the food chain have all contributed towards the highly significant decline in OC for most Lake Laberge species. There is no evidence to support the hypothesis that contaminants declined due to changes in the trophic levels of individual fish species (Chapter 4). Future research for Lake Laberge should continue to include periodic collections of fish and invertebrates that coincide with the four-year population surveys performed by the Yukon Territorial Government. More observations of the plankton dynamics (densities, community composition) should occur on a seasonal basis. Concurrent air and water analyses need to be integrated with plankton studies to confirm that OC in the aquatic environment have remained stable and are a minor influence on contaminant shifts in mid- to upper trophic level biota.

8. CONTRIBUTION TO KNOWLEDGE

1. The study has started a temporal trend project providing information on short-term environmental contaminant changes as it relates to regional climate patterns in the Yukon region. This data can be used in future Yukon region assessments as global climate changes continue.
2. The research has produced the first temporal trend data (10 years) for contaminants in the aquatic food web of a sub-Arctic lake. Few studies exist on the temporal trends of legacy pesticides in aquatic, sub-Arctic food webs. The data can be used to track the progress of contaminant degradation (or gains) in Kusawa, Quiet and Lake Laberge.
3. This research has produced the first information on the changes in contaminants in the aquatic food web of a sub-Arctic lake following the cessation of a commercial fishery. Observations of anthropogenic impacts on aquatic ecosystems are hard to determine because of the large effort required to pre and post monitor the site. The Lake Laberge system was monitored just before and for some time after a major anthropogenic influence (fishing industry) was discontinued thus allowing for a comparison of the effect that commercial fishing cessation has had on fish populations, trophic structures and subsequently OC contaminants in the food web.

4. This work demonstrated that OC contaminant levels can be highly variable among lake ecosystems, even within close geographical proximity. It also supports the theory that changes in the biotic structure of a food web is a more significant influence on contaminant concentrations in the aquatic food chain compared to the concentrations of abiotic depositions of these chemicals.
5. The research is Important for the local Whitehorse community for tourism and native fisheries since these data may be used to reassess the Health Canada fish consumption advisories that currently exist on Lake Laberge. With the decrease in contaminants noted, the health advisories may be lifted if contaminant levels fall under the criteria for safe human consumption of fish tissues and help to promote sport or subsistence fishing and tourism commerce.

9. FUTURE RESEARCH

1. Initiate detailed interannual phytoplankton characterization (seasonal dynamics, community composition, biomass, OC concentrations) and relations to interannual climate changes. Broaden the research on changes in species dominance.
2. Research changes in the photic zone and relations to phytoplankton growth. Add Secchi depths to future limnological analyses to review the hypothesis of light (and related heat) limitation vs. nutrient limitation for plankton growth.
3. Review changes in contaminant concentrations in the water column and relate to interannual fluctuations in OC concentrations in plankton.
4. Relate trends in OC concentrations in phytoplankton to zooplankton through tandem sampling and analysis.
5. Continue monitoring sentinel species of fish in Lake Laberge and broaden samples for monitoring in Kusawa and Quiet Lake to include lower trophic level fish and plankton.
6. Initiate cooperative research with Yukon Territorial Government during years of population assessments to gather larger sample numbers to calculate growth rates of lake trout and analyze for correlations with contaminant levels as well as confirm the hypothesis of growth rate changes over the 1990s.
7. Research more young-of-the-year growth dynamics relating to food preference, growth and contaminant levels at this stage of the food chain.

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