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Relating trophic structure to mercury distribution in a Gulf of St.Lawrence food web : from zooplankton to colonial seabirds using stable nitrogen ($\sigma^{15}N$) and carbon ($\sigma^{15}C$) isotope analysis

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Relating trophic structure to mercury distribution in a Gulf of St. Lawrence food web: from zooplankton to colonial seabirds using stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope analysis

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ABSTRACT

Even at very low concentrations in the environment, mercury (Hg) has the potential to biomagnify in food chains reaching levels of concern in apex predators such as fish-eating seabirds. The aim of this study was to determine the trophic structure and the transfer of total mercury (THg) and methylmercury (MeHg) in a Gulf of St. Lawrence food web using stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope analysis. Since food is the main exposure route to Hg in organisms, I wanted to characterize and compare the diet of top level predator seabirds using traditional and novel techniques. I found that body weight, trophic level and lipid content of organisms were the best predictors of THg and MeHg in this food web. Although the influence of habitat on the overall Hg distribution in the food web was low, I was able to demonstrate an effect within taxonomical groups which indicates that sediments are a source of THg and MeHg in this ecosystem. Several organisms at the base of the food chain were above the MeHg threshold level for the protection of wildlife suggesting a potential threat for upper trophic level predators. Diet composition of seabirds determined using traditional and novel techniques led to similar and complementary results illustrating that these methods can be combined for future studies to monitor prey availability and to predict the exposure of consumers to Hg.

Résumé

Même à de très faibles concentrations dans l'environnement, le mercure (Hg) a le potentiel de bioamplifier dans les chaînes alimentaires atteignant des niveaux préoccupants chez les prédateurs apicaux tels que les oiseaux marins piscivores. Le but de cette étude était de déterminer la structure trophique ainsi que le transfert du mercure total (THg) et du méthylmercure (MeHg) dans un réseau alimentaire du Golfe du Saint-Laurent à l'aide de la composition en isotopes stables de l'azote ($\delta^{15}N$) et du carbone ($\delta^{13}C$). Puisque l'ingestion de nourriture est la principale voie d'exposition au Hg, j'ai voulu caractériser et comparer le régime alimentaire de prédateurs supérieurs comme les oiseaux marins à l'aide de techniques traditionnelles et novatrices. J'ai découvert que la masse corporelle, le niveau trophique et le contenu lipidique des organismes constituaient les meilleurs prédicteurs du THg et du MeHg dans ce réseau alimentaire. Bien que l'influence de l'habitat sur la distribution du Hg dans l'ensemble du réseau alimentaire soit minime, j'ai démontré un effet à l'intérieur des groupes taxonomiques ce qui indique que la couche de sédiment est une source de THg et de MeHg dans cet écosystème. Plusieurs organismes à la base de la chaîne alimentaire étaient au-dessus du seuil pour la protection de la faune ce qui suggère une menace potentielle pour les prédateurs de niveaux supérieurs. Le régime alimentaire des oiseaux marins déterminé à l'aide de techniques traditionnelles et novatrices a conduit à des résultats similaires et complémentaires illustrant que ces méthodes peuvent être combinées pour des études ultérieures dans le but de suivre la disponibilité des proies et de prédire l'exposition des consommateurs au Hg.

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CHAPTER 1

General introduction

1.1 THESIS PRESENTATION

The first chapter of this thesis will review the literature about the general concepts regarding mercury and the available tools to monitor and track it in the environment. The second chapter will provide original data on the distribution of total and methylmercury in a Gulf of St. Lawrence food web. The third chapter will focus on the feeding ecology of seabirds as the top level predators of this food web. The second and the third chapters are written as scientific papers. The fourth chapter provides the required synthesis and conclusions as an overall summary of the findings of this research project.

1.2 PROBLEM IDENTIFICATION

Methylmercury (MeHg) is a ubiquitous toxic chemical that is found in the environment. Although extensively studied in many aquatic ecosystems, there is little known about the transfer of MeHg in the biota of estuaries. Due to its potential to bioaccumulate in animals and to biomagnify in food chains, MeHg can reach levels of concern in upper trophic level consumers and potentially cause adverse health effects. The diet is the main route of exposure for mercury. The objective of this study is to determine the trophic structure and the transfer of mercury from the base of the food chain to the top. I also want to compare the diet of five species of seabirds using two different techniques.

1.3 MERCURY CYCLING AND UPTAKE FROM BIOTA

Mercury (Hg) is released in the environment by natural and anthropogenic sources with a higher proportion for the latter source (UNEP 2002). The main anthropogenic source of Hg in the atmosphere is the combustion of fossil fuels which accounts for about 65% of the global anthropogenic emissions (Pacyna et al. 2006). Gold production using Hg recovery methods is also an important sector which accounts for more than 10%. Asian countries are by far the most important contributors to the overall man-made emissions with more than half of the production. Fortunately, anthropogenic emissions are decreasing in some countries or the rate of increase is slowing down in others (Pacyna et al. 2003, Pacyna et al. 2006). Consequently, global anthropogenic emissions declined between 1995 and 2000 (Pacyna et al. 2006). The great majority of mercury released in the atmosphere is in the form of gaseous elemental Hg (Hg^0; Mason et al. 1994, Schroeder and Munthe 1998). Hg^0 has a residence time in the atmosphere of about 1 year (Slemr et al. 1985, Mason et al. 1994). It has a high vapour pressure and therefore a great tendency to escape from water to air (Schroeder and Munthe 1998). These features allow Hg^0 to be volatile and to be transported to remote areas by atmospheric winds (Fitzgerald et al. 1998). After being oxidized to mercuric ion (Hg^{2+}), it is deposited by precipitation (Muir et al. 1999, Macdonald et al. 2000), gas absorption and dry deposition (Muir et al. 1999). Some sites in north-eastern North America represent hot spots for Hg deposition (Travnikov and Ryaboshapko 2002, Miller et al. 2005) and for exposure to wildlife (Evers et al. 2007). This is mainly due to fossil fuel combustion in lower latitudes in north-eastern North America (Travnikov and Ryaboshapko 2002, Pacyna et al. 2003, Evers et al. 2007).

Although global anthropogenic emissions have recently diminished (Pacyna et al. 2006), there is evidence that Hg has increased in the environment during the last century. Mason et al. (1994) found that anthropogenic emissions have tripled the concentrations of Hg in the atmosphere and at the surface of the ocean over the last century. It has also been shown that Hg levels have increased in wildlife during the last decades (Burgess and Braune 2004, Braune et al. 2005, Braune 2007) and over the last century (Thompson et al. 1992, Monteiro and Furness 1997, Outridge et al. 2002). Monteiro and Furness (1997) found that methylmercury (MeHg) increased in a food chain by about 1.1 to 4.8 % per year between 1885 and 1994. This is in agreement with other studies showing an increase in the atmosphere of approximately 1 % per year for periods of over 150 years (Mason et al. 1994), 100 years (Fitzgerald 1995) and 20 years (Slemr and Langer 1992). Historical records of peat bog cores also suggest an increase in Hg deposition in Europe since preindustrial times (Madsen 1981).

Mercury becomes of greater concern when it is transformed into methylmercury (MeHg, for review, see Ullrich et al. 2001), which is believed to be the most toxic form of Hg (Scheuhammer 1987, Scheuhammer et al. 2007). The principal methylation process is thought to occur under reducing conditions in the anoxic zone of freshwater and estuarine sediments with sulphate-reducing bacteria (SRB) playing an important role (Compeau and Bartha 1985, Gilmour et al. 1992). However, SRB are not effective when the sulphate concentration is too high, which is the case in marine ecosystems (Gilmour and Henry 1992, Weber 1993). Consequently, the proportion of MeHg over THg is usually lower in marine environments (Wiener et al. 2003). Loseto et al. (2004) found that methylation occurred in Arctic wetlands without the action of SRB. This gives reason to believe that abiotic or other

biological mechanisms are contributing to the methylation process (Weber 1993, Loseto et al. 2004, Celo et al. 2006). Weber (1993) suggested that humic matter may contribute to abiotic methylation. Solar radiation was also found to be a significant factor for MeHg production in the oxic epilimnion of freshwater lakes (Siciliano et al. 2005). A low pH provides suitable conditions for methylation of Hg (Grieb et al. 1990, Spry and Wiener 1991) and organisms living in lower pH lakes display higher Hg concentrations (Burgess and Meyer 2008). Moreover, wetlands are known to provide favourable conditions for MeHg formation and transport due to high concentrations of organic matter (Wiener et al. 2003).

MeHg is the form of Hg that is biomagnified throughout food chains (Watras and Bloom 1992, Watras et al. 1998, Bowles et al. 2001, Campbell et al. 2005, Riget et al. 2007). This compound has a biological half-life of approximately 44 to 65 days in the blood of Cory's shearwaters (*Calonectris diomedea,* Monteiro and Furness 2001). MeHg binds to water-soluble thiol (i.e., sulphydryl: -SH) groups of amino acids such as cysteine and methionine and to polypeptides such as glutathione (Wolfe et al. 1998, Simmons-Willis et al. 2002). MeHg is readily absorbed by the intestine (> 90 %; Wolfe et al., 1998) and is distributed throughout the body by the blood stream (Monteiro and Furness 1995, Oliveira et al. 1999). MeHg binds to the amino acid cysteine and forms a molecule that is structurally similar to methionine, which allows MeHg to cross the blood-brain barrier by molecular mimicry (Ballatori 2002, Simmons-Willis et al. 2002). It also crosses the blood-placenta membrane resulting in a higher contamination of the foetus than the mother (Wolfe et al. 1998). Most common toxicological endpoints of MeHg include adverse effects on behaviour, reproductive mechanisms (Wolfe et al. 1998), reproductive success (Burgess and

Meyer 2008, Evers et al. 2008) and damage to the central nervous system (Wolfe et al. 1998).

Industrial discharge into waters has been shown to cause high concentrations of MeHg in aquatic wildlife and subsequently in humans. Minamata disease is a good example of massive contamination. A company producing acetaldehyde released Hg in wastewater in the Minamata Bay for several years (UNEP 2002). Highly polluted fish were eaten by the population reaching fatal toxicological levels. Several people died, others showed permanent nervous system disorders and children were born with malformations and dysfunctions. Another event occurred in Iraq where similar effects to that for Minamata Bay were reported after people consumed seeds treated with mercurial fungicides (Wiener et al. 2003).

MeHg is known to bioaccumulate in most organisms during their lifetime (Hall et al. 1997) and to biomagnify in food chains (for a review of biomagnification studies in aquatic ecosystems, see table 2.4). The mechanism by which MeHg enters the food chain is still poorly understood, but SRB as well as microplankton are believed to play an important role in the transfer between the biotope and the biota (Watras et al. 1998). MeHg is concentrated in the phytoplankton at the base of the food chain (Mason et al. 1996). Uptake of MeHg in aquatic organisms occurs through body surface, respiratory organs (e.g., gills) and food consumption (CCME 2003, Hammerschmidt and Fitzgerald 2006). Food consumption is known to be the most important source for MeHg uptake in higher trophic level animals (Hall et al. 1997). In air-breathing organisms such as mammals and birds, diet is the main route of exposure (CCME 2003, Wiener et al. 2003). Since the transfer efficiency of MeHg is greater than the biomass transfer efficiency in aquatic ecosystems (for review, see table 2.4), the concentration in top predators can reach levels that are several orders of magnitude

higher compared to the water in which they live. The augmentation of MeHg concentration from an organism relative to the water (bioaccumulation factor; BAF) increases with increasing trophic level (Watras et al. 1998, Bowles et al. 2001, Hammerschmidt and Fitzgerald 2006). However, the highest BAFs are observed between water and seston and between water and phytoplankton (Watras et al. 1998, Bowles et al. 2001, Hammerschmidt and Fitzgerald 2006). BAF of MeHg in seston ranges from about 5.0×10^4 to 7.9×10^5 times, whereas the increase in BAF for subsequent trophic levels is only 2 to 4 times higher (Watras et al. 1998, Bowles et al. 2001, Hammerschmidt and Fitzgerald 2006). This suggests that the microplankton is an important component for the uptake of MeHg in food chains. Consequently, fish can reach levels that are over 6.7×10^6 times more contaminated than the water in which they live (Bowles et al. 2001). Levels of MeHg are known to increase with age of fish (Hammerschmidt and Fitzgerald 2006) and marine mammals (Outridge et al. 2002), but this is not the case in birds (Honda et al. 1986, Thompson et al. 1991, Thompson et al. 1993). Hg in feathers does not increase with the age of adult birds (Thompson et al. 1991, Thompson et al. 1993). The excretion of MeHg via feathers is sufficient to prevent the accumulation throughout their lifetime (Honda et al. 1986).

Several hypotheses have been proposed to explain the variability in biomagnification intensity among food webs. A hypothesis was suggested that the size of an ecosystem is an indicator of biomagnification potential since small (usually freshwater) ecosystems are characterized by a limited choice of prey for predators, whereas more open (usually marine) ecosystems tend to display a broad range of prey species with highly variable concentrations of contaminants (Isaacs 1973). Moreover, it has been shown that THg concentrations in fish are negatively correlated with lake size (Bodaly et al. 1993). Since Hg concentration

increases exponentially with trophic level (for review, see table 2.4), a slight difference in trophic level at which a predator will feed will result in a large difference in Hg exposure. To test this theory, a study conducted in a large and diverse ecosystem (Miramichi River Estuary) failed to determine significant biomagnification of THg (Pastershank 2001). It was also suggested that the length of the food chain can explain the biomagnification potential of Hg (Rasmussen et al. 1990, Cabana and Rasmussen 1994, Cabana et al. 1994). Hg transfer efficiency being greater than the energy transfer efficiency, additional trophic steps should lead to higher Hg levels in upper predators. Temperature and therefore latitude have been suggested as other variables that could influence the ability of Hg to biomagnify. Organisms are known to grow faster in warmer regions and therefore to dilute the amount of Hg contained in their body (Pauly 1998, Campbell et al. 2003). Hg concentration at the base of the food web or in the water could also influence the biomagnification of this compound. Higher uptake at the base of the food chain would enhance Hg biomagnification.

Although trophic level is believed to be one of the most important variables driving the level of contamination in aquatic organisms, the source of organic matter and therefore, the feeding habitat is also a major component. Cossa and Gobeil (2000) showed that benthic organisms or organisms that were connected to the benthic habitat had higher THg concentrations in muscle tissues than pelagic organisms in the St. Lawrence Gulf and Estuary. Studies in Long Island Sound, a coastal ecosystem, found that the production of MeHg from the sediments was 70% of the total input of MeHg (Balcom et al. 2004, Hammerschmidt et al. 2004). Also, it has been observed that MeHg concentration is higher in feathers of seabirds eating mesopelagic prey compared with those eating epipelagic prey (Thompson et al. 1998). THg concentrations have been found to be higher in mesopelagic

fish (> 300 m) compared to epipelagic fish (< 200 m) and to be positively correlated with median daytime depth of 8 fish species (Monteiro et al. 1996). This could be due to the higher mercury methylation occurring in deep waters where the oxygen concentration is lower (Mason and Fitzgerald 1990, 1993, Monteiro et al. 1996, Monteiro and Furness 1997, Thompson et al. 1998, Cossa and Gobeil 2000).

Relatively low THg and MeHg levels at the base of the food chain can potentially be harmful for the health of top predators such as seabirds and marine mammals. For this reason, the Canadian Council of Ministers of the Environment (CCME 2000) suggested a tissue residue guideline (TRG) in whole prey of 0.033 μg MeHg/g wet weight (ww) judged safe for the protection of wildlife. Another study suggested that a MeHg level exceeding 0.02 μg/g ww in whole prey could possibly be harmful for fish-eating wild birds (Yeardley et al. 1998). THg levels in whole fish were found to reduce (0.21 μg/g ww) and inhibit (0.41 μg/g ww) the reproduction of common loon (*Gavia immer*), a wild piscivorous freshwater bird (Burgess and Meyer 2008). A comprehensive study for over 18 years on *G. immer* suggested a threshold value for THg of 3.0 μg/g ww in whole blood for reproductive, neurological and morphological adverse effects (Evers et al. 2008). Preliminary no and low observable adverse effect levels (NOAEL and LOAEL, respectively) of MeHg were estimated at 0.08 and 0.4 μg/g ww, respectively in the food of dosed chicks *G. immer* (Kenow et al. 2003, Meyer 2006). Further research is needed in order to reach a consensus regarding valid threshold values.

1.4 SEABIRDS

As long-living piscivorous top predators, seabirds have been recognized as good bioindicators and biomonitors of the environment (Kushlan 1993, Monteiro and Furness 1995, Becker and Chapdelaine 2003). The health of seabirds (from suborganism level to population level) reflects the health of the environment in which they live and feed (Kushlan 1993). Scarcity of food supply and availability, high concentration of pollutants in the environment, anthropogenic disturbance and change in climate conditions can have adverse effects on their reproduction and survival (Kushlan 1993, Becker and Chapdelaine 2003).

Seabirds are sensitive to bottom-up effects (Becker and Chapdelaine 2003) and this has prompted interest in the evaluation of their diet (for review, see Barrett et al. 2007). When prey species are less abundant, seabirds integrate alternative dietary items that can potentially contain lower caloric values. Rail and Chapdelaine (2000) showed that when the availability of capelin (*Mallotus villosus*) was limited, a higher proportion of human refuse was incorporated in the diet of herring gull (*Larus argentatus*) chicks. Populations of black-legged kittiwake (*Rissa tridactyla*) were found to fluctuate negatively with industrial fisheries which acted as competitors for lesser sandeels (*Ammodytes marinus*) in the North Sea from 1986 to 2002 (Frederiksen et al. 2004). On the other hand, populations of herring gull (*Larus argentatus*) were found to be linked positively with Atlantic cod (*Gadus morhua*) fisheries in the Gulf of St. Lawrence between 1925 and 1993 (Chapdelaine and Rail 1997). The latter study found that the availability of discarded fish from commercial fishing boats was correlated with the population size of *L. argentatus*.

Climate is known to influence seabird populations and survival. Populations of seabirds (*Uria lomvia* and *U. aalge*) were found to fluctuate with sea surface temperature

oscillation in the Arctic (Irons et al. 2008). The authors found that the intensity rather than the direction of the oscillation triggered the change in these populations. Distribution of underlying food items associated with the temperature would most likely explain the variations (Irons et al. 2008). Increase in sea surface temperature during winter in the North Sea was also found to reduce the recruitment of *A. marinus* the principal prey of *R. tridactyla.* This negatively affected the breeding success of the latter seabird and eventually the survival of the adults and the population growth rate (Frederiksen et al. 2004).

Colonial seabirds have been used as bioindicators and biomonitors of metals and persistent organic pollutants (POPs) contamination on spatial and/or temporal scale in environments such as the Arctic (Braune et al. 2001, Braune et al. 2002, Braune et al. 2005, Braune 2007), the Great Lakes (Mineau et al. 1984, Fox et al. 1998, Hebert et al. 1999a, Hebert et al. 1999b, Hebert et al. 2000, Hebert and Weseloh 2006, Norstrom and Hebert 2006) and the St. Lawrence River (Chapdelaine et al. 1987, Elliott et al. 1988, Comba et al. 1993). The population and productivity of northern gannet (*Sula bassanus*) in the Gulf of St. Lawrence between 1968 and 1984 were found to be affected by the presence of dichloro-diphenyl-trichloroethane (DDT, Chapdelaine et al. 1987, Elliott et al. 1988). Reduction of the eggshell thickness was observed in eggs containing higher levels of p,p'-dichloro-diphenyl-dichloroethylene (DDE; $r = -0.69$) and unhatched eggs had no outer calcified layer (Elliott et al. 1988). Seabirds are known to show high concentrations of pollutants due to biomagnification of chemical compounds throughout food chains. This trophodynamic pattern has been observed at several sites for THg, MeHg and POPs in the Arctic (Atwell et al. 1998, Fisk et al. 2001, Hobson et al. 2002, Campbell et al. 2005, Riget et al. 2007) and in the

Gulf of Farallones, CA, USA (Jarman et al. 1996). Seabirds can reach contaminant levels known to induce toxicological effects (Burgess and Meyer 2008, Evers et al. 2008).

1.5 STABLE ISOTOPE ANALYSIS

The application of naturally occurring stable isotope analysis (SIA) has increased over the last decades as a tool to test hypotheses in aquatic ecology. Stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes have been used to demystify predator-prey interactions (Phillips and Gregg 2003, Hammill et al. 2005), trophic structures and assemblages (Hobson and Welch 1992, Hobson 1993, Lesage et al. 2001), as well as distribution of Hg (Cabana and Rasmussen 1994, Jarman et al. 1996, Atwell et al. 1998, Reinfelder et al. 1998, Power et al. 2002, Campbell et al. 2005) and POPs (Rasmussen et al. 1990, Comba et al. 1993, Borga et al. 2001, Fisk et al. 2001, Hobson et al. 2002) in aquatic food webs.

The principle of SIA in ecology is based on the assumption that the naturally occurring stable isotope ratio of a consumer (mixture) reflects in a predictive way the average of the stable isotope ratios of its dietary items (sources, Deniro and Epstein 1978, 1981). When there is a switch in the diet of a predator, its isotopic signature will progress gradually towards the most recent dietary item. The biochemical turnover rate corresponds to the time when food items were assimilated and more precisely when tissues were synthesized (Tieszen et al. 1983). The turnover rate varies depending on the tissues or organs taken into consideration (Tieszen et al. 1983). Fast turnover rate tissues such as plasma and liver give an estimation of meals assimilated on a short-term period, whereas slow turnover rate tissues such as red blood cells, muscle and bone collagen give estimations over a longer period of time (Tieszen et al. 1983, Hobson and Clark 1992a, 1993, Dalerum

and Angerbjorn 2005). The turnover rate of a given organ is positively correlated to its metabolic rate (Tieszen et al. 1983). This allows doing retrospective dietary analysis depending on the tissue used. Other tissues reveal information about the diet assimilated at the time they developed such as feathers, hair, teeth and claws. These types of tissues are metabolically inert after growth and remain unaltered (Dalerum and Angerbjorn 2005). Extinct and past preserved animals in museums can therefore reveal important information about historical dietary shifts (Hilderbrand et al. 1996) and contaminant levels (Thompson et al. 1992, Monteiro and Furness 1997, Thompson et al. 1998).

Stable nitrogen isotope ($\delta^{15}N$) values are known to increase by approximately 3 to 5 ‰ (Deniro and Epstein 1981, Peterson and Fry 1987) with an average of 3.4 ‰ ± 1 SD per trophic level for most animals (Minagawa and Wada 1984, Cabana and Rasmussen 1994, Post 2002). A captive rearing study on an adult cormorant fed a known and constant mackerel diet for over 23 years revealed a lower trophic enrichment factor of 2.4‰ between the diet and muscle tissue (Mizutani et al. 1991). This has been acknowledged as a suitable trophic enrichment factor for birds in general (Hobson and Welch 1992). Bearhop et al. (2002) found a similar result of 2.6 ‰ between whole sprats (*Sprattus sprattus*) and whole blood of great skuas (*Catharacta skua*). The difference in fractionation between birds and other animals is believed to be caused by production of uric acid in the case of birds instead of urea (Hobson and Clark 1992b, Hobson and Welch 1992). The enrichment of ^{15}N over ^{14}N is due to the preferential excretion of the lighter nitrogen isotope in urine inducing a predictable enrichment of ^{15}N in a consumer compared to its diet (Peterson and Fry 1987). Consequently, $\delta^{15}N$ can be used as a reliable indicator to calculate the trophic level of a given organism.

Stable carbon isotope ($\delta^{13}C$) values show a slight increase with trophic level of approximately 1 ‰ (Deniro and Epstein 1978, Bearhop et al. 2002) although a lot of intertissue and interspecific variability exists (Dalerum and Angerbjorn 2005). $\delta^{13}C$ is mainly used for information on the source of carbon (Peterson and Fry, 1987). The carbon signature is mostly defined at the primary producer level and then it is conserved within the food chain. Primary producers from terrestrial and freshwater ecosystems show lower (or more negative) $\delta^{13}C$ values compared to producers from marine ecosystems (Fry and Sherr 1984, Peterson and Fry 1987). Within marine ecosystems, benthic primary production and benthic organisms show higher (or less negative) $\delta^{13}C$ values compared to pelagic primary production or pelagic organisms (Hobson 1993, France 1995, Hobson et al. 1995, France et al. 1998, Lesage et al. 2001, Hobson et al. 2002), although that hypothesis has been recently contested (Nadon and Himmelman 2006). France (1995) found that planktonic algae are 5‰ lower in $\delta^{13}C$ values compared to benthic algae and this difference was then reflected higher in the food chain. This would be due to the higher thickness of diffusive boundary layers of organisms living in low turbulence waters, such as benthic algae (Keeley and Sandquist 1992, France 1995). Inversely, pelagic algae exposed to higher turbulence have a reduced boundary layer and therefore a lower diffusion resistance. This results in a higher ^{13}C discrimination of CO_2 and HCO_3^- through the cell walls leading to a decrease in $\delta^{13}C$ values (Keeley and Sandquist 1992, France 1995).

1.6 DIET COMPOSITION

Since $\delta^{15}N$ and $\delta^{13}C$ are known to increase in a predictable fashion with trophic level and the source of primary production, the relative contribution of different food items

(sources) for a given consumer (mixture) can be assessed accurately when corrected for trophic enrichment factors. Simple mixing models have been used to estimate food sources of animals with SIA (Vander Zanden and Rasmussen 2001). Recently, more accurate mixing models have been published and allow estimating the error related to each proportion of prey in a diet using 1 or 2 isotopes (n) and $n + 1$ sources (Phillips and Gregg 2001). Phillips and Koch (2002) subsequently included organic nitrogen and carbon concentrations in their model since the omission these concentrations can lead to biased results when estimating the diet. Another model has been recently published for cases where animals have a very diverse diet and for which the number of prey items are exceeding ($> n + 1$) the amount of isotopic tracers available (Phillips and Gregg 2003). This latter model looks at all possible combinations of source proportions by iteration. The model provides the distribution of feasible contributions of each source to the mixture rather than giving a definitive solution.

Instead of the traditional stomach content dietary analysis that only provides a "snapshot" of what an organism is eating, SIA provides information about assimilated rather than just ingested diet (Vander Zanden et al. 1997). The isotopic signature is incorporated into macromolecules (i.e., proteins, lipids, carbohydrates and nucleic acids) that constitute tissues and organs (Tieszen et al. 1983). This also gives an average estimation of meals assimilated over a longer time period rather than a static and potentially inaccurate evaluation (Vander Zanden et al. 1997). However, SIA does not have the ability to reveal the representative species of the diet. Moreover, the evaluation of the diet gets more complex as the diversity increases (Phillips and Gregg 2003). For those reasons, it is important to combine traditional and novel techniques.

Traditional techniques used to evaluate seabird diet are usually restricted to the breeding period and to the breeding ground which is when and where birds are accessible (Barrett et al. 2007). These techniques are diverse (for review, see Barrett et al. 2007) and include identification of food items from feeding behaviour (Guillemette et al. 1992), stomach content analysis of living (Wilson 1984) or dead birds (Guillemette et al. 1992, Gaston and Bradstreet 1993), regurgitations (Rail and Chapdelaine 1998, Rail and Chapdelaine 2000), regurgitated pellets (Hebert et al. 1999c, Kubetzki and Garthe 2003) and feces (Kubetzki and Garthe 2003). Assessment of diet composition can also be accomplished by observation of fish-carrying species (Chapdelaine and Brousseau 1996) or by sampling at the feeding sites (Guillemette et al. 1992). Some of the aforementioned techniques present different stages of digestion of prey items; from completely fresh prey items (fish carried by adults to their chicks), slightly altered (stomach content, regurgitation) to highly digested (pellets, feces). Even if the items are highly digested, the size of food items can be reconstructed using recognizable hard parts that are known to reflect the original size (e.g., bones, exoskeletons, shells, Hammill et al. 2005). Alternatively, they can inform about the occurrence of a given item in the diet not identified by other methods. Most of these techniques are non-invasive and raise fewer ethical issues and therefore allow the gathering of good sample sizes. Although some techniques are more rewarding, time-efficient and informative than others, they are all complementary and enhance the accuracy of the assessment of diet composition. The confidence increases especially when these techniques are combined concurrently with SIA (Hobson et al. 1997, Hammill et al. 2005, Tierney et al. 2008).

1.7 GULF OF ST. LAWRENCE

The Gulf of St. Lawrence is one of the world's largest estuaries with 1.95 x 10^5 km^2 of body of water and a basin which includes the Laurentian Great Lakes, covering more than 1.6 x 10^6 km^2 (St. Lawrence Centre 1996). The depth of the Gulf of St. Lawrence ranges from 100 to 400 m but it can reach 500 m (St. Lawrence Centre 1996). There are about 15.5 million people living in the watershed area. This Gulf has a very well defined stratification with an input from the ocean of dense cold and salt water in the bottom (marine signal; less negative $\delta^{13}C$ signal), a brackish warmer water layer at the top coming from the discharge of the Great Lakes and other large rivers such as the Saguenay River and the Ottawa River (terrestrial signal; more negative $\delta^{13}C$ signal) and an intermediate layer in between resulting in the mixing of adjacent layers (intermediate signal). In the winter, the upper layers merge into one layer (Bugden 1981). The input of freshwater into the Gulf of St. Lawrence is estimated at more than 1.68 x 10^4 m^3/s with the main tributaries being the Ottawa (mean annual discharge: 1.9 x 10^3 m^3/s), the Saguenay (1.8 x 10^3 m^3/s) and the Manicouagan (1.0 x 10^3 m^3/s) rivers (St. Lawrence Centre 1996). Vertical fluxes of nutrients from lower layers are possible due to the physico-chemical properties of each of the water layers, the bathymetry and the currents of the Gulf. Therefore, these fluxes supply the surface water with a high concentration of nutrients (Yeats 1988b). Moreover, the Gulf of St. Lawrence's tributary rivers contain high concentrations of nutrients that are supplied to the surface water of the Gulf (Yeats 1988b). Consequently, primary production can reach levels as high as 385 g of carbon/m^2/yr (Pocklington 1988) which results in high biological diversity and abundance. This ecosystem is able to support organisms that have elevated energy requirements such as marine mammals and seabirds.

The Great Lakes-St. Lawrence watershed is a hot spot of THg deposition with some areas receiving more than 30 $\mu g/m^2$/year (Travnikov and Ryaboshapko 2002, Miller et al. 2005). Moreover, it has been estimated that about 170 tons of anthropogenic Hg have been released in the St. Lawrence from the 1940's to 1995 (Cossa and Gobeil 2000). Such high input is a potential threat for organisms living in this ecosystem. Cossa and Gobeil (2000) estimated that Hg in the Lower St. Lawrence Estuary is presently almost in balance between 1) input of Hg from riverine sources and sequestration in the estuarine sediments; 2) exchange between the Lower St. Lawrence Estuary and the Gulf and; 3) atmospheric Hg deposition and evasion.

1.8 General objectives

Mercury is affected by several variables that determine its concentration in the environment and its tendency to accumulate in biota. Estuarine ecosystems complicate the interpretation due to their multiple sources and fluxes of nutrients, contaminants and organisms. The Gulf of St. Lawrence food web has received very little attention in this regard. The objective of this study is to determine the community structure and the transfer of total and methylmercury in a Gulf of St. Lawrence ecosystem. In addition, I want to assess and compare the diet of seabirds having distinct ecological niches using traditional and novel techniques. This study will help to identify the mechanisms involved in Hg distribution as a function of trophic structure. To my knowledge, this is the first study to bridge the distribution of mercury species with trophic organization using stable isotope analysis in the Gulf of St. Lawrence.

CHAPTER 2

Trophic structure and mercury distribution in a Gulf of St. Lawrence food web using stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope analysis

2.1 INTRODUCTION

2.1.1 Problem identification

Even at low concentrations in the environment, mercury (Hg) can reach elevated levels in apex predators. Feeding ecology is believed to be a key variable for accumulation of Hg in organisms. The overall objective of this study is to link total mercury (THg) and methylmercury (MeHg) concentrations with the trophic structure of a Gulf of St. Lawrence food web using stable isotope analysis.

2.1.2 Mercury cycling and uptake from biota

Mercury (Hg) is a potent chemical released in the environment by natural and anthropogenic sources (UNEP 2002). The elemental form of Hg (Hg^0) is volatile, can travel long distances in the atmosphere and can be deposited at a different location than the point of origin (Fitzgerald et al. 1998). There are several indications that Hg concentrations have increased in the environment since pre-industrial times (Madsen 1981, Slemr and Langer 1992, Thompson et al. 1992, Mason et al. 1994, Fitzgerald 1995, Monteiro and Furness 1997, Outridge et al. 2002, Burgess and Braune 2004, Braune et al. 2005, Braune 2007). Hg becomes of greater concern when it is methylated in the form of methylmercury (MeHg, for review, see Ullrich et al. 2001). Methylation of Hg is a process that is believed to be greatly influenced by the action of sulphate-reducing bacteria (SRB) taking place in the anoxic zone

of aquatic sediments (Compeau and Bartha 1985, Gilmour et al. 1992). MeHg binds to proteins (Wolfe et al. 1998, Simmons-Willis et al. 2002), which allows this compound to bioaccumulate in organisms and to biomagnify in food chains (Watras and Bloom 1992, Hall et al. 1997, Watras et al. 1998, Bowles et al. 2001, Campbell et al. 2005, Riget et al. 2007). Food intake is thought to be the main route of exposure in higher trophic level organisms (Hall et al. 1997). High levels can be reached by upper trophic level consumers in food chains and potentially cause adverse effects (Wolfe et al. 1998, Burgess and Meyer 2008, Evers et al. 2008). Piscivorous seabirds have been acknowledged as being good Hg bioindicators of their environment due to their high position in food chains (Monteiro and Furness 1995).

2.1.3 Stable isotope analysis

Stable isotope analysis (SIA) can be used as an ecological tracer of the assimilated food sources of consumers. Stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes are known to undergo predictable stepwise enrichments between the diet and the consumer (Deniro and Epstein 1978, 1981). $\delta^{15}N$ is enriched in a consumer compared to its diet (Deniro and Epstein 1978, 1981) and primary production exhibits different $\delta^{13}C$ signatures in freshwater vs marine (Fry and Sherr 1984, Peterson and Fry 1987) and pelagic vs benthic habitats (Hobson 1993, France 1995, Hobson et al. 1995, France et al. 1998, Lesage et al. 2001, Hobson et al. 2002). The trophic position of organisms can therefore be estimated using $\delta^{15}N$ to calculate trophic level and $\delta^{13}C$ as an indicator of the source of organic matter (Peterson and Fry 1987). Mixing models allow inferences regarding the diet composition of a particular consumer when corrected for isotopic trophic enrichment (Phillips and Gregg

2001, Vander Zanden and Rasmussen 2001, Phillips and Koch 2002, Phillips and Gregg 2003).

2.1.4 Gulf of St. Lawrence

The Gulf of St. Lawrence is a vast ecosystem that receives inputs of freshwater and marine water. The exchange of nutrients from deep to surface waters triggers an important emergence of primary production increasing the diversity and the abundance of organisms.

Several variables determine the concentration of mercury in the environment and its propensity to accumulate in biota. Estuarine ecosystems complicate the interpretation due to their multiple sources and fluxes of nutrients, contaminants and organisms. The Gulf of St. Lawrence food web has received very little attention regarding the mechanisms involved in Hg distribution in relation to trophic structure. To my knowledge, this is the first study to relate the distribution of mercury with trophic structure using stable isotope analysis in the Gulf of St. Lawrence.

2.1.5 Objectives, hypotheses and predictions

The overall objective of this study is to relate mercury concentrations to the trophic structure of a Gulf of St. Lawrence food web using stable isotope analysis.

More specifically, I want to determine the main variables that are predicting the contamination levels of THg and MeHg in this food web using *i)* trophic level ($\delta^{15}N$), *ii)* level of connectivity with the benthos ($\delta^{13}C$), *iii)* total body weight and *iv)* lipid content of the organisms. I hypothesize that THg and MeHg concentrations are influenced by the feeding ecology and the physical characteristics of organisms. Then, THg and MeHg

concentrations will increase with trophic level, level of connectivity with the benthos and organism size, whereas concentrations will decrease with lipid content.

I want to determine whether the concentrations of THg and MeHg are influenced by depth in this ecosystem. Sediments are a source of THg and MeHg in the Gulf of St. Lawrence. I predict that Hg concentrations in water will be positively correlated with depth.

I want to determine which aspects of an ecosystem influence the rate of biomagnification of THg and MeHg. I hypothesize that ecosystem biodiversity, water temperature, food chain length and Hg concentration at the base of the food chain alter the rate of biomagnification. Consequently, the rate of biomagnification of THg and MeHg will be lower in marine ecosystems compared to freshwater ecosystems. Moreover, the rate of biomagnification will increase with latitude, the number of trophic levels and the concentration of THg and MeHg at the base of the food chain.

I want to examine the biomagnification of THg and MeHg in a Gulf of St. Lawrence food web from primary consumers to top level predators using $\delta^{15}N$ to calculate trophic level. If transfer efficiencies of THg and MeHg are greater than the biomass transfer efficiency in this aquatic ecosystem, then THg and MeHg concentrations in animals will increase with their trophic level.

2.2 METHODS

2.2.1 Sample collections

2.2.1.1 Fish and invertebrates

Fish and some invertebrate samples were collected by the personnel of the Department of Fisheries and Oceans Canada (DFO) onboard the *CCGS Teleost* as part of ongoing research sampling in the Gulf of St. Lawrence. Samples were gathered in the region of Sept-Îles within a 60 km radius of Corossol Island, QC, Canada (50°10'N to 49°44'N and 66°59'W to 66°26'W; voyage number: IML-2006-043 (TE-676-677); figure 2.1) in August 2006. Whole specimens of Atlantic herring (*Clupea harengus*; ATHE), witch flounder (*Glyptocephalus cynoglossus*; WIFL), American plaice (*Hippoglossoides platessoides*; AMPL), northern shrimp (*Pandalus borealis*; PABO), striped shrimp (*P. montagui*; PMON), pink glass shrimp (*Pasiphaea multidentata*; PMUL) and snow crab (*Chionoecetes oplio*; SNCR; table 2.2) were sampled at five stations by horizontal trawling using a Campelen 1800 shrimp bottom trawl (mesh size 80/60/44 mm, codend liner 12.7 mm) for 15 minutes at 5 knots at different depths (112 to 282 m). American sandlance (*Ammodytes americanus*; SAND) were caught using a nylon fyke net VX-04, with an opening of 0.75 m and two wings of 15.2 m length, mesh size of 6.25 mm displayed at the entrance of Clet Stream in Sept-Îles Bay. Capelin (*Mallotus villosus*; CAPE) were sampled by hand while spawning on the beaches of Sept-Îles. All samples were kept individually in polyethylene bags except shrimp for which ten individuals were kept in each bag. An additional sampling was done in May 2007 for zooplankton 50 km southwest of Corossol Island (49°41'N and 66°44'W) by double oblique trawling using a Bongo net in nytex with an opening of 60 cm diameter and a mesh size of 333 μm for 24 minutes (downward oblique: 10 min, upward oblique: 14 min) at

2-3 knots between 0 and 250 m. The depth of the Bongo was recorded in real time using a hydrophone via a transmitter positioned on the net. Zooplankton (*Calanus finmarchicus*; CFIN, *C. hyperboreus*; CHYP and northern krill; *Meganyctiphanes norvegica*; MENO) was sorted by size using a stainless steel sieve with a 2 mm mesh size and then pooled by size. Each pool was kept in a polyethylene bag. Samples were preserved in a freezer at -20°C after collection prior to further treatment and analysis.

Littoral and benthic macroinvertebrates were collected under federal permits (permit numbers: CN15-2006 and CN03-2007 issued by Fisheries and Oceans Canada) on Corossol Island beaches in 2006-07 (Figure 2.2). Blue mussel (*Mytilus edulis*; BLMU), waved whelk (*Buccinum undatum*; BUCC), common tortoiseshell limpet (*Tectura testudinalis*; PATE), common periwinkle (*Littorina littorea*; BIGO), green sea urchin (*Strongylocentrotus droebachiensis*; URCH) and gammarid sp. (*Gammarellus sp.*; GAMA) were collected by hand. They were sorted by species to avoid cross-contamination and kept alive in high density polyethylene (HDPE) buckets for 24h at 4°C in order to allow their gut content to empty. The water was changed 2-3 times during this process to avoid reassimilation of excrement. The water in which organisms were kept was from the sites where they were collected and the temperature in the buckets corresponded to the sites of origin. Organisms were frozen after 24h and kept in a freezer at -20°C until they were shipped by mail on dry ice to the University of Ottawa (Ottawa, ON) where they were then kept in a freezer at -20°C until further treatment and analysis.

2.2.1.2 Seabirds

Twenty adult seabirds per species (21 for Rissa tridactyla; BLKI) were caught during the incubation period between May and June 2006-07 at Corossol Island (figure 2.2) using a drop trap, a noose pole or a hand net under federal government permit (permit number: 10711 issued by Environment Canada) and approved by the animal care committee of the University of Ottawa (BL-205). Herring gull (Larus argentatus; HERG) and great black-backed gull (Larus marinus; GBBG) were caught using a drop trap set above the nest. The trigger mechanism was activated automatically as soon as the adult was sitting on its nest. Razorbills (Alca torda; RAZO) were caught using a net on the end of a pole while birds were flying. Some individuals hiding in rock crevices were caught by the leg using a metal rod with a blunt hook. BLKI and common eider (Somateria mollissima; COEI) were caught using a net on the end of a pole while on their nest prior to or while taking flight. All birds were marked with stainless steel or aluminum bands provided by the Canadian Wildlife Service (CWS). Measurements of the weight and the wing were taken on seabirds. Blood and regurgitations were sampled simultaneously for the same adult. Regurgitations were limited to GBBG, HERG and BLKI in this study and were opportunistically collected. Blood was sampled via venipuncture of the ulnar vein. Rubbing (isopropyl) alcohol was applied to clean the surface of the ulnar vein and a sterile 25-gauge sized needle was used for sample collection. Approximately 3 mL of blood was retrieved with a 5 cc. heparinised syringe representing less than 1% of the weight of the animal (Gaunt and Oring 1997). Blood samples were transferred into mercury-free polyethylene vials and stored in a cool icebox then centrifuged in the field within 12 hours, using a VWR Galaxy® Mini microcentrifuge at 6 000 rpm for 5 minutes. Plasma and red blood cells were separated using

pipetters with polypropylene pipette tips and transferred into mercury-free vials and then frozen within 12 hours of collection. In 2006, Cryovials® were stored in liquid nitrogen at -190°C and in 2007 Eppendorf® tubes were stored in the freezer at -20°C. The samples were shipped on dry ice to the University of Ottawa where they were then stored in a freezer at -20°C prior to further treatment and analysis. In the field, a drop of blood was blotted within the pre-printed circle on Schleicher & Schuell® #903 blood collection paper. At the National Wildlife Research Centre (Ottawa, ON), DNA was extracted from blood spots and polymerase chain reaction (PCR) was used to determine the sex of the seabirds. A single set of primers, P2 and P8, was used to amplify homologous sections of two conserved chromo-helicase-DNA-binding (CHD) genes located on the sex chromosomes (Griffiths et al. 1998). Gel electrophoresis revealed a single CHD-Z band in males and a second distinctive CHD-W band in females. The validity of the method was assessed using measurements (wing length and total weight) taken on GBBG, HERG and BLKI, assuming that males were larger and heavier than females (Baird 1994, Pierotti and Good 1994, Good 1998). Sexual dimorphism was assessed for each species using one-tailed *t*-tests ($p < 0.05$). Moreover, COEI were sexed based on plumage (Goudie et al. 2000) to confirm the results of the PCR method.

2.2.1.3 Water and particulate organic matter

Sampling of water was done within 10 km of the south shore of Corossol Island using a 2 L VanDorn® bottle in opaque polyvinyl chloride at 0, 85 and 170 m depths in 2007. Depths were considered representative of the layers of the Gulf of St. Lawrence (St. Lawrence Centre 1996). The water samples were collected in duplicate in previously

hydrochloric acid-cleaned 1 L HDPE bottles. The bottles were conditioned three times with the water sampled before being filled. For mercury analysis, 5 mL of trace metal grade hydrochloric acid (HCl) was added to the sample bottles (0.5 %) immediately in the field in order to avoid any degradation of MeHg. For the carbon concentration and stable isotope analysis of dissolved/total organic/inorganic carbon (DIC/DOC and TIC/TOC), 1 L HDPE bottles were filled to the top without any treatment. All bottles were sealed in 2 polyethylene bags and kept in a dark and cool room at 4°C until further treatment and analysis.

Particulate organic matter (POM) was kindly provided by V. Lesage (DFO, pers. comm.). POM was sampled in 2001 by DFO personnel within a 75 km radius of Corossol Island at the depth of maximum chlorophyll production for each station (10 to 24 m) using a 4L Niskin® bottle (figure 2.1). Water samples were filtered through precombusted Whatman® GF/C glass-fiber filters (see Lesage et al. 2001 for methods).

2.2.2 Sample preparation

Organisms were measured using stainless steel calipers (± 0.01 mm) or a ruler (± 0.5 mm). In the field, the weight was recorded using a Pesola® balance (500 ± 2.5 g to 2500 ± 10 g) whereas a Metler Toledo® electronic balance (± 0.1 g) was used in the laboratory. The weight of the zooplankton was estimated using length in the regression equation:

$$M = aL^b \qquad (2.1)$$

where M is body mass, L is length, a is the intercept of the equation and b the slope. The weight of MENO was estimated using the Boysen and Buchholz (1984) equations for

May and June since zooplankton in my study were sampled during that period. Madsen et al. (2001) and Thor et al. (2005) modified an equation from Hirche and Mumm (1992) that considers a mass of carbon for CFIN and CHYP, respectively. The modified equation is:

$$M = (aL^b) * 100 / [C] \quad (2.2)$$

where [C] represents the percentage of elemental carbon contained in the samples which is measured using a CE 1110 Elemental Analyser (see *2.2.5 Stable isotope analysis*).

Organisms were rinsed with deionised water (DIW), cut into pieces and submerged in a polystyrene beaker containing liquid nitrogen. The frozen pieces were ground to a fine powder in stainless steel analytical mills (IKA® M 20 Universal mill or Retsch® Mixer Mill MM 301). Whole fish, shrimp, gammarids and zooplankton were homogenized whereas only the soft inner tissue was homogenized for molluscs, echinoderms and crabs. Approximately 1 g of dorsal muscle tissue was taken from each fish for THg analysis prior to homogenisation of the whole fish to do inter-tissue comparison (dorsal muscle vs whole). Small organisms such as zooplankton and gammarids were frozen using liquid nitrogen and ground using a mortar and pestle. The cellular fraction of seabird blood (hereinafter called red blood cells; RBCs) was analysed after centrifugation. Stainless steel dissecting tools and mercury-free plastic or glass cutting boards were used. All instruments and tools were acid-rinsed with 10% nitric acid (HNO_3), rinsed with ethanol and wiped with Kimwipes® between each manipulation to avoid cross-contamination. Sample tissues were placed into polypropylene microcentrifuge tubes.

2.2.3 Total mercury (THg) analysis

2.2.3.1 THg in animal tissues

Total mercury (THg) analysis was conducted at the Center for Advanced Research in Environmental Genomics, University of Ottawa by cold vapour atomic absorbance spectrometry (CVAAS) through gold amalgamation and thermal decomposition using a high temperature combustion instrument SP-3D Mercury analyser Nippon Instrument Corporation® (Osaka, Japan) following U.S. Environment Protection Agency (USEPA) method 7473 (USEPA 1998). Samples (between 0.01 and 0.2 g) were accurately weighed on a Sartorius® model BP121S or a Metler® model AB104-S analytical balance (± 0.1 mg) and were placed in a ceramic boat with successive layers of two additive mixtures (aluminum oxide and a 1:1 calcium hydroxide: sodium carbonate mixture). The boat was then placed in the decomposition chamber. Each sample was run in duplicate (or more) until a coefficient of variation below 15% was reached. In order to ensure analytical accuracy and reproducibility, blanks and standards from a stock of Mercury Reference Solution (certified 1 000 µg/g ± 1%; Fisher Scientific CSM114-100) brought to a concentration of 50 ng/g were tested in duplicate every 5-7 samples. Also, two standard reference material (DORM-2 and TORT-2) from the Canadian National Research Council were tested in duplicate every 15 samples. The recoveries of the reference materials were 97.2 ± 1.0 % (mean ± SE); $n = 53$ and 97.5 ± 2.2 %; $n = 6$ for DORM-2 and TORT-2, respectively. Replicate results were within the accepted range of 85 to 115% except 2 results with values of 115.8 and 84.1 %. The average of duplicate samples was used for statistical analysis. The concentrations of the samples are expressed in nanogram per gram dry weight (ng/g dw) but the analyses were done on fresh (wet) tissue. Moisture content was generated for each sample in order to

convert the Hg concentrations from wet to dry weight. To measure the moisture content, approximately 0.1 g of fresh homogenate was weighed on a Sartorius® model BP121S or a Metler® model AB104-S analytical balance (± 0.1 mg) before and after being freeze-dried at -150°C for 48 hours in a Labconco® model 4451F freeze-dryer. All instruments and tools used in this method were acid-rinsed with 10% HNO_3, rinsed with ethanol and wiped with Kimwipes® between each manipulation to avoid cross-contamination. The instrument detection limit for THg analysis was 0.01 ng total mercury (method 7473, USEPA 1998).

2.2.3.1 THg in water samples

THg analysis in water was done on unfiltered samples by oxidation, purge and trap, desorption and cold vapour atomic fluorescence spectrometry (CVAFS) using a Series 2600 Tekran® Mercury Analysis System (Knoxville, USA) following USEPA method 1631 (USEPA 2002). Briefly, all Hg (Hg^0, Hg^{1+}, MeHg, $(Me)^2Hg$, EtHg) was oxidized into Hg^{2+} using 0.4 mL of bromine monochloride (BrCl). A volume of 0.1 mL of hydroxylamine hydrochloride (NH_2OH•HCL) was added to the samples to neutralise any excess of BrCl and prevent free halogens from getting into the system. All Hg^{2+} was then reduced to volatile Hg^0 with stannous chloride ($SnCl_2$). Hg^0 was separated from solution by purging using argon as carrier gas by gas/liquid separation, passed though a soda lime trap and collected by a first gold trap, then desorbed thermally and captured by a second analytical gold trap. Hg was measured by CVAFS. Field and laboratory blanks (in duplicate) were passed through the whole procedure to ensure that the sampling and the method did not alter the Hg concentration. Results of both blank types were below the minimum level of quantitation (ML) of 0.5 ng/L (USEPA 2002). Blanks and standards from a stock of

Mercury Reference Solution brought to a concentration of 0.88 ng/L were tested every 3 samples. Two samples were spiked to calculate the recovery of the method. Each sample was run in duplicate and coefficients of variation were below 15%. The average of replicates was used for statistical analysis. The recovery of the spiked samples was 102.7 ± 0.65 % (mean ± SE; n = 2) and the recovery of the standards was 97.3 ± 2.06 % (mean ± SE; n = 7) which falls within the level of quality control acceptance criteria for this method (USEPA 2002). Protocol was followed in a mercury-free clean room. Shoe covers and Tyvek® lab coats laminated with saranex were worn at all times. The method detection limit was 0.05 ng/L (USEPA 2002).

2.2.4 Methylmercury (MeHg) analysis

Methylmercury (MeHg) analysis was done by capillary gas chromatography coupled with atomic fluorescence spectrometry (GC-AFS) using a Hewlett Packard® GC system model HP 6890 with a HP 7683 injector. Protocols were modified from Cai et al. (1997) and Cai et al. (1996) in animal tissue and water samples, respectively. MeHg analyses were conducted at the Center for Advanced Research in Environmental Genomics at the University of Ottawa. In this study, inorganic mercury refers to the difference between THg and MeHg and will be noted as IHg. The latter assumption is believed to be valid since most organic mercury is in the form of MeHg (Kim et al. 1996).

2.2.4.1 MeHg in animal tissues

Tissue samples (between 0.1 and 0.6 g) were accurately weighed on a Sartorius® model BP121S or a Metler® model AB104-S analytical balance (± 0.1 mg), then 2 mL of

potassium hydroxide (6 N) was added and the samples were shaken for 4 hours at 330 rpm on a VWR® orbital shaker model DS-500 followed by addition of HCl (6 N) until a pH between 1.0 and 3.0 was reached using HCl (20%) and potassium hydroxide (KOH; 20%) solutions. A mixture of acidic (HNO_3) potassium bromide and copper sulphate (KBr:$CuSO_4$ - 3:1) was added. An exact volume of methylene chloride (CH_2Cl_2) recorded by weight (± 0.1 mg) was added and the samples were shaken overnight at 330 rpm and then centrifuged at 2 500 rpm for 10 minutes in a Beckman® refrigerated centrifuge model J2-MC at 10°C. An exact volume of CH_2Cl_2 was transferred and 1 to 2 mL of sodium thiosulfate ($Na_2S_2O_3$; 0.01 M) was then added to the samples. The samples were shaken at 330 rpm for 20 minutes, vortexed and centrifuged at 2 500 rpm for 10 minutes. The $Na_2S_2O_3$ was transferred and acidic KBr:$CuSO_4$ as well as CH_2Cl_2 were added. The samples were shaken at 330 rpm for 15 minutes, vortexed and centrifuged at 5 000 rpm for 30 seconds in an Eppendorf® centrifuge model 5301. The CH_2Cl_2 was then transferred in a glass insert through a layer of anhydrous sodium sulphate (Na_2SO_4) to remove any trace of water. Borosilicate glass scintillation vials with Teflon® polytetrafluoroethylene caps and polypropylene microcentrifuge vials were used for this method. All instruments and tools used were acid-rinsed with 10% HNO_3 and wiped with ethanol between each manipulation to avoid cross-contamination. Each sample was run in duplicate and coefficients of variation were below 20 %. One blank and three standards reference materials (DORM-2, TORT-2 or DOLT-3) from the Canadian National Research Council were tested every 5-7 samples to ensure analytical accuracy and reproducibility. The recoveries of the reference materials were 100.6 ± 1.6 % (mean ± SE); n = 17, 96.1 ± 1.4 %; n = 27 and 105.4 ± 2.0 %; n = 4 for DORM-2, TORT-2 and DOLT-3, respectively. Replicate results were within the accepted range of 85

to 115%. The average of duplicate samples was used for statistical analysis. The concentration of the samples is expressed in nanogram per gram dry weight (ng/g dw) but the analyses were done on wet tissue (see *Total mercury analysis*). The absolute detection limit was 20 pg as Hg (Cai et al. 1997). THg and MeHg were analysed from the same sample to ensure a valid percentage of MeHg.

2.2.4.2 MeHg in water samples

For this method, 8 mL of KOH (6N) were added to the samples and the pH was adjusted to 3.00 (± 0.01) using HCl (20%) and KOH (20%) solutions, then 10 mL of pH 3.0 buffer (acetic acid mixed with sodium acetate) was added. A tubing system consisting of Teflon® tubes, silicone caps and 1 mL polypropylene pipette tips was washed with 500 mL of 1 % HCl with a peristaltic pump. Sulfydryl-cotton fiber was packed in 5 mL screening columns which were added to the system to be purged with 500 mL of DIW with a peristaltic pump. Samples were passed through the system and 15 mL of DIW followed by 6 mL of acidic KBr:$CuSO_4$ (2:1) mixture were added directly on the surface of the screening column to collect the eluate in borosilicate glass scintillation vials with Teflon® polytetrafluoroethylene caps. An exact volume of CH_2Cl_2 was added to the samples and to the standards (see below). Samples and standards were shaken at 330 rpm for 15 minutes, vortexed and centrifuged at 2 500 rpm for 10 minutes in a refrigerated centrifuge at 10°C. An exact volume of CH_2Cl_2 was then transferred in a glass insert through a layer of anhydrous Na_2SO_4. The standards consisted of acidic KBr:$CuSO_4$ mixture with water spiked with MeHgCl Standard Solution (certified 1 000 ug/g; Alfa aesar® stock no. 33553) yielded to a concentration of 5 µg/L through serial dilution. Five calibration standards were prepared

in duplicate samples and CH_2Cl_2 was added. The standards were subjected to the same extraction procedure used for samples. A matrix spike sample was prepared in duplicate using 0.1 mL of working solutions of MeHgCl in DIW and was subjected to the entire analytical procedure. Field and laboratory blanks (in duplicate) were passed through the whole process to ensure that the sampling and the method did not alter the Hg concentration. Samples, blanks, matrices and standards were subjected to GC-AFS and were run in duplicate. Coefficients of variation of duplicate samples were below 15 %. The average of duplicates was used for statistical analysis. The recovery of the spike samples was 102.3 ± 0.72 % (mean ± SE; n = 2). The concentrations of the samples are expressed in nanograms per litre wet weight (ng/L ww). The detection limit for this method was 0.01 ng/L (Cai et al. 1997). THg and MeHg were analysed from the same sample to ensure a valid percentage of MeHg.

2.2.5 Stable isotope analysis (SIA)

2.2.5.1 SIA in animal tissues

Samples were freeze-dried at -150°C for at least 48 hours and then homogenised into a fine powder. An exact mass (around 1 mg) of homogenised material was weighed using a microbalance (± 0.001 mg) and loaded into tin capsules. The isotopic composition of organic carbon and nitrogen were determined by flash combustion at 1800°C on a CE 1110 Elemental Analyser (Milan, Italy) connected to a Delta$^{Plus®}$ Advantage isotope ratio mass spectrometer (IRMS – Bremen, Germany) coupled with a ConFlo® III at the G.G. Hatch Stable Isotope Laboratory, University of Ottawa. Stable isotope ratios were quantified as the

deviation from standard reference material in parts per thousand (‰) according to the following equation:

$$\delta X = [\ (R_{sample} / R_{standard}) - 1\ ] * 1000 \qquad (2.3)$$

Where X is ^{13}C or ^{15}N and R is the corresponding ratio $^{13}C/^{12}C$ or $^{15}N/^{14}N$. The $R_{standard}$ values were based on the PeeDee Belemnite (PDB) for ^{13}C and atmospheric N^2 (AIR) for ^{15}N. The analytical precision was 0.20‰ for $\delta^{13}C$ and $\delta^{15}N$.

2.2.5.2 SIA in water and particulate organic matter samples

Carbon concentration and stable isotopes were measured using the method described by St-Jean (2003). Carbon concentration of dissolved/total organic/inorganic carbon (DIC/DOC and TIC/TOC) were determined using a modified OI Analytical wet oxidation Total Organic Carbon (TOC) analyser (College Station, USA) model 1010 at the G.G. Hatch Stable Isotope Laboratory. Samples were acidified using 5% phosphoric acid (H_3PO_4) to transform DIC and TIC to CO_2 which is believed to be proportional to DIC and TIC, respectively. The gases were purged using helium as the carrier gas and CO_2 was measured by a nondispersive infrared (NDIR) detector. After the completion of the reaction, persulfate reactant was added to the samples to oxidize the DOC and TOC at 100°C to form CO_2. The resultant CO_2 was then measured by NDIR. After passing through the NDIR, CO_2 was collected at the vent and sent through water and halogen traps. It was then passed through a packed nitrogen, carbon and sulphur (NCS) GC column and captured by a ConFlo® III continuous-flow interface and carbon isotope ratio ($\delta^{13}C$) was measured by a CE 1110 Elemental Analyser (EA) and a Finnegan MAT Delta$^{Plus®}$ Isotope Ratio Mass Spectrometer (IRMS).

Samples for dissolved carbon analysis (DIC/DOC) were filtered through 0.45 μm pore size Millipore® Millex®-HV Syringe Driven Filter Unit. Acid-washed (20% HCl) and DIW-rinsed TraceClean® 40 mL amber borosilicate vials with 6.3 mm silicone/Teflon® septa were used for this method. The 2 sigma analytical precision was 2 ppbC or 2%, whichever was higher for the concentration and 0.2‰ for the isotope.

The isotopic composition of POM samples was determined on an isochrom CF-IRMS connected to a Carla Erba EA (CHNS-O EA1108) at the Environmental Isotope Laboratory, University of Waterloo (Waterloo, ON). The analytical precision was 0.20‰ for $\delta^{13}C$ and $\delta^{15}N$ (see Lesage et al. 2001 for methods).

2.2.6 Physico-chemistry of water

Determination of pH was done using a VWR symphony SP21 pH meter. The calibration of the instrument was done using Thermo Scientific Orion® buffer solutions (pH = 4.01, 7.00 and 10.01) which gave a coefficient of determination (R^2) of 0.999. The precision of the instrument was 0.01. The salinity of the samples was determined using a Combo pH-conductimetre, model HI 98129 by Hanna instruments®. The calibration of the instrument was done using DIW and two conductivity/total dissolved solids Standards (100 and 500 μS/cm) from Ricca Chemical Company® and a R^2 of 0.999 was associated. The precision of the instrument is 1 μS/cm.

2.2.8 Statistical analysis and calculations

2.2.8.1 Trophic level and Hg

Trophic level (TL) was determined relative to *T. testudinalis* (PATE) as baseline ($TL_{T.\ testudinalis}$) which I assumed to occupy trophic level 2. This is a semi-sedentary primary

consumer species that has a relatively long lifespan and these conditions are believed to minimise spatiotemporal isotopic variations (Cabana and Rasmussen 1996, Vander Zanden and Rasmussen 1999, Post 2002). Trophic levels of most species were assessed using a modification of Hobson and Welch (1992) equation:

$$TL_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{T.\ testudinalis}) / 3.4 \quad (2.4)$$

where $TL_{consumer}$ and $\delta^{15}N_{consumer}$ are the trophic level and $\delta^{15}N$ of the organism of interest, respectively, $\delta^{15}N_{T.\ testudinalis}$ is the $\delta^{15}N$ of *T. testudinalis* (6.89 ± 0.04 ‰; mean ± SE; n = 10), and 3.4 ‰ is the isotopic enrichment factor for most organisms (Minagawa and Wada 1984, Post 2002). A long term captive-rearing study on great skua (*Catharacta skua*), a fish-eating bird fed on a known and constant diet of sprat (*Sprattus sprattus*) for 18 months suggests a trophic enrichment factor of 2.6 ‰ (n = 9) between whole fish (with lipid) and the whole blood of the bird (Bearhop et al. 2002). Several authors used a similar trophic enrichment factor of 2.4 ‰ between diet and muscle for seabirds in general (Hobson and Welch 1992, Hobson et al. 1995, Fisk et al. 2001, Hobson et al. 2002, Campbell et al. 2005, Riget et al. 2007). The equation 2.4 becomes:

$$TL_{bird} = 3 + (\delta^{15}N_{bird} - (\delta^{15}N_{T.\ testudinalis} + 2.6)) / 3.4 \quad (2.5)$$

Several calculations were used in the past regarding the way to report the transfer of contaminants in the environment and they were assigned diverse names and calculations. In this thesis, I will refer to the following definitions and calculations for the trophic transfer of

Hg. Biomagnification power of Hg throughout the food web under study was assessed using the slope (b) of the following simple linear regression (SLR):

$$\text{Log}_{10}\,[\text{Hg}] = b\,(\delta^{15}\text{N}) + a \qquad (2.6)$$

where Hg is THg or MeHg and a is the intercept. The latter equation is analogous to the equation developed by Broman et al. (1992) which used untransformed data. In order to compare the slopes of the compounds together, a SLR was done on inorganic Hg (IHg; difference between THg and MeHg) against δ^{15}N. Analyses of covariance (ANCOVA) could not be used to compare the slopes because THg and MeHg values were not independent (Zar 1999). In fact, the analyses were deliberately done on the same individuals to ensure a proper percentage of MeHg. Food web magnification factors (FWMFs) were calculated using equation 2.6 with TL instead of δ^{15}N:

$$\text{Log}_{10}\,[\text{Hg}] = b\,(\text{TL}) + a \qquad (2.7)$$

with b being used as follow (Fisk et al. 2001):

$$\text{FWMF} = 10^{\,b} \qquad (2.8)$$

This measurement represents the biomagnification potential over the entire food web taking into account the different trophic enrichment factors (equations 2.4 and 2.5). Another

method to measure the biomagnification factor (BMF) is applied to predator-prey interactions using the TL derived from $\delta^{15}N$ (Fisk et al. 2001):

$$BMF_{TLC} = ([Hg]_{predator} / [Hg]_{prey}) / (TL_{predator} / TL_{prey}) \quad (2.9)$$

This latter equation corrects for trophic level and is only valid with the assumption that a predator consumes a single prey type. This is rather unlikely as most species feed upon more than one prey type. I therefore used 2 or more sources mixing models (Phillips and Gregg 2001, Phillips and Koch 2002, Phillips and Gregg 2003) for SIA to quantify the proportion of sources that were likely to be found in the diet of the predators (see *3.2.3 Statistical analyses and calculations*; Chapter 3 for details about mixing models). I applied these proportions to the above equation and developed the following equation:

$$BMF_{SIA} = ([Hg]_{predator} / ([Hg]_{prey\ A} * f_A + [Hg]_{prey\ B} * f_B)) / (TL_{predator} / (TL_{prey\ A} * f_A + TL_{prey\ B} * f_B)) \quad (2.10)$$

where fA and fB are the proportions of prey A and prey B, respectively in the diet of the predator.

Bioaccumulation factors (BAFs) were calculated to assess the magnitude of Hg increase in an organism compared to water (Bowles et al. 2001):

$$BAF = [Hg]_{organism} / [Hg]_{water} \quad (2.11)$$

where $[Hg]_{organism}$ is the concentration of THg or MeHg in ng/g ww of a given organism and $[Hg]_{water}$ is the average concentration of THg or MeHg in ng/g ww over the entire water column. Water THg and MeHg concentrations used for the calculations were 0.90 and 0.12 pg/g ww, respectively. Values are reported in Log_{10} (BAF).

2.2.8.2 Physical characteristics and Hg

SLRs using THg and MeHg against stable carbon isotope ($\delta^{13}C$) values, lipid content or total weight as independent variables were applied. Multiple linear regressions (MLRs) using standardized THg, MeHg and IHg concentrations as well as the percentage of MeHg and the percentage of IHg as dependant variables and $\delta^{15}N$, $\delta^{13}C$, lipid content and total weight as independent variables were performed to assess the relative importance of the variables that are driving Hg levels throughout this food web. Values were standardized (0 ± 1; mean ± SD) for this test in order to compare variables that have different scales. Lipid content was calculated using the equations presented by McConnaughey and McRoy (1979):

$$L = 93 / [1 + (0.246 * C{:}N - 0.775)^{-1}] \qquad (2.12)$$

where $C{:}N$ is the ratio of the concentration of organic carbon over the concentration of organic nitrogen contained in the tissue analysed. To test the same hypothesis as for the MLR, a principal components analysis was used with standardized independent variables to determine which ones explain the most variability in this food web. The first principal component (PC1) was used as independent variable to predict Hg levels using SLRs.

2.2.8.3 Connectivity with benthos and Hg

To test if the connectivity to the benthos was a good predictor of Hg concentrations, THg and MeHg were normalized for the variable which was believed to be the most important confounding variable. $\delta^{15}N$ was used for seabirds and invertebrates, whereas body length was used for fish. Length was used instead of $\delta^{15}N$ for fish because the fit of the data was better. Statistical analyses were done separately on the species groups (seabirds, fish and invertebrates) and on individual species. Analyses of variance (ANOVAs) were used to test whether Hg concentrations were different between organisms exploiting pelagic habitat, benthic habitat or a mixture between the 2 habitats. ANOVAs were also used to determine if there was a significant difference between individual species within groups. If significant, a multiple comparison analysis (Tukey test) was used to determine which levels of a factor were significantly different. SLRs were done on individual groups and an analysis of covariance (ANCOVA) was done when applicable, to determine if a difference existed between the slopes of groups. When SLRs did not meet the assumptions of normality and/or homoscedasticity, polynomial regressions were executed progressively towards the most complex models until the fit of the data was not significantly better.

2.2.8.4 General considerations

In general, a Tukey test was used to assess which levels of a factor showed significant differences, where applicable. The homogeneity of the variances was tested on absolute values of residuals (Levene test) and the normality of the residuals was tested using the Kolmogorov-Smirnov test. When assumptions of parametric tests were not met and no transformation would satisfy the requirements, non-parametric tests were used. However,

considering the large sample size in this study (n = 272 for THg and n = 139 for MeHg) and consequently the high power, the assumptions of normality and homoscedasticity were often violated. In these cases, non-parametric tests were done to confirm the conclusions of the parametric tests but only the results of parametric tests are reported. Statistical analyses were performed using S-Plus (version 8.0) and an α significance threshold of 0.05 was used unless otherwise stated. A Bonferroni correction was applied to α when more than one comparison was done with the same statistical analysis in order to avoid inflating type I error (Zar 1999). In this document total mercury and methylmercury will be referred to as THg and MeHg, respectively.

2.3 RESULTS

In this chapter, I will first discuss the physical environment of a Gulf of St. Lawrence food web. I will then provide a description of the community structure and link it to food web mercury contamination.

2.3.1 Water physico-chemistry

Total mercury (THg) concentrations increased with depth ($F_{0.05(2)2,3} = 50.5$; $R^2 = 0.97$; $p = 0.005$) in a curvilinear fashion, whereas methylmercury (MeHg) did not show any trend with values consistently low throughout the water column ($F_{0.05(2)1,4} = 1.41$; $R^2 = 0.26$; $p = 0.30$). This resulted in a decrease of MeHg percentage in a linear trend ($F_{0.05(2)1,4} = 9.12$; $R^2 = 0.70$; $p = 0.039$) reaching 6.4 ± 0.2 % at 170 m depth compared to 27.6 ± 5.2 % for the surface water and 22.2 ± 3.5 % at 85 m depth (table 2.1; figure 2.3).

Water pH decreased ($F_{0.05(2)2.3} = 365$; $R^2 = 0.996$; $p < 0.001$) and salinity increased ($F_{0.05(2)2.3} = 80.5$; $R^2 = 0.98$; $p = 0.002$) with depth in curvilinear fashions (table 2.1; figure 2.4). Dissolved inorganic carbon (DIC) and total inorganic carbon (TIC) concentrations increased non-linearly ($F_{0.05(2)2.3} = 80.5$; $R^2 = 0.98$; $p = 0.002$ and $F_{0.05(2)2.3} = 189$; $R^2=0.99$; $p = 0.001$, respectively) reaching a plateau between 85 and 170 m for the latter (table 2.1; figure 2.5). Dissolved organic carbon (DOC) and total organic carbon (TOC) concentrations both seemed to decrease with depth, but only the latter was significant ($F_{0.05(2)2.3} = 13.3$; $R^2 = 0.77$; $p = 0.022$; table 2.1). Lower values for TOC and TIC compared to DOC and DIC are likely due to analytical precision or sample preparation.

$\delta^{13}C$ signatures decreased with depth at a similar rate for DIC and TIC ($F_{0.05(2)2.3}$ = 188; $R^2 = 0.99$; $p = 0.001$ and $F_{0.05(2)2.3} = 566$; R^2=0.997; $p < 0.001$, respectively; Table 2.1, Figure 2.6).

2.3.2 Trophic structure

A wide range of values was observed in this study for trophic level (calculated using $\delta^{15}N$), whereas $\delta^{13}C$ range was relatively narrow (table 2.2). Trophic level ranged from 1.20 ± 0.08 for particulate organic matter (POM; Lesage, pers. comm.) to 4.68 ± 0.02 for RAZO (table 2.2; figure 2.7). Trophic level was calculated using PATE as baseline instead of POM since this type of mollusc is believed to minimize the spatio-temporal variation of the isotopic signature (Cabana and Rasmussen 1996, Vander Zanden and Rasmussen 1999, Post 2002).

$\delta^{13}C$ values ranged from -17.26 ± 0.11 for BIGO to -23.15 ± 0.58 for POM. The source of energy in this food web originated mainly from pelagic sources such as POM and zooplankton with predatory species such as BLKI and RAZO enriched relative to these latter carbon sources. COEI was feeding more extensively on benthic organisms such as molluscs and URCH, whereas HERG and GBBG were obtaining their energy from both pelagic and benthic food sources. Terrestrial sources are also possible for both HERG and GBBG (see Chapter 3). Considering only organisms that were obtaining their energy from pelagic sources (within the dashed lines; figure 2.7), a positive correlation was found between trophic level and $\delta^{13}C$ ($t_{(0.05)172} = 18.5$; $r^2 = 0.67$; $p < 0.001$). GBBG and HERG were included in this latter analysis since they are known to feed extensively on pelagic fish (Rail and Chapdelaine 2000). A correlation between trophic level and $\delta^{13}C$ for the whole food

web resulted in a poor fit of the data ($r^2 = 0.03$), but was nonetheless significant ($t_{(0.05)274} = 2.67$; $p = 0.01$).

Organisms were generally enriched in ^{13}C as their connection with the benthos increased (Figure 2.8). This trend was valid for fish and invertebrates for both interspecific comparisons ($F_{(0.05)4,51} = 83.93$; $p < 0.001$ and $F_{(0.05)10,103} = 93.56$; $p < 0.001$, respectively) and among habitat comparisons ($F_{(0.05)2,53} = 84.06$; $p < 0.001$ and $F_{(0.05)2,111} = 174.72$; $p < 0.001$). Seabirds did not display any significant interspecific ($F_{(0.05)4,96} = 1.24$; $p = 0.298$) or among habitat ($F_{(0.05)2,98} = 1.51$; $p = 0.23$) differences in $\delta^{13}C$.

2.3.3 Mercury in fish and invertebrates

A high range of THg and MeHg concentrations can be observed in species of this food web (Figure 2.9a). THg levels in fish were well below the guideline for human fish consumption which is set to 500 ng/g ww (Health Canada 2007; 2000 ng/g dw assuming a moisture content of 75% in fish) with a highest value of 423 ng/g dw for AMPL (appendix A).

For MeHg a preliminary lowest observable adverse effect level (LOAEL) on the diet of Common loon chicks (*Gavia immer* fed fish *ad libitum* from hatch to 105 days) was estimated at 400 ng/g ww (1600 ng/g dw) as methylmercury chloride (MeHgCl) and a preliminary no observable adverse effect level (NOAEL) in the diet was estimated at 80 ng/g ww (320 ng/g dw; Kenow et al. 2003, Meyer 2006). Although the LOAEL was above the concentrations measured for the prey animals in my study, the NOAEL was closer to the levels observed (figure 2.9a). However, the NOAEL was not reached by any fish or

invertebrates in this study with the highest MeHg concentration being 223 ng/g dw for WIFL (appendix A).

A different picture emerges when looking at the tissue residue guideline (TRG) of 33 ng/g ww (132 ng/g dw) provided by the Canadian Council of Ministers of the Environment (CCME, 2000; Figure 2.9a). This guideline was calculated using the tolerable daily intake (TDI = 31.2 ng/g of body weight per day) of wild mallard ducks (*Anas platyrhynchos*) fed for three generations on a known contaminated diet (Heinz 1979) and taking into account the food intake by body weight ratio (FI:bw = 0.94) of Wilson's storm petrel (*Oceanites oceanicus*). A total of 10.3 % of fish and no invertebrates were above this limit. Similar results were obtained when looking at the MeHg guideline considered safe for the protection of fish-eating birds estimated at 20 ng/g ww (80 ng/g dw; Yeardley et al. 1998). If this guideline was used then, 30.1 % of fish and 10.8 % of invertebrates were above this limit (appendix A).

Percentage of MeHg constituting THg levels for whole fish was between 46.9 ± 7.1 for WIFL to 64.8 ± 2.6% for SAND (mean ± SE). Invertebrates displayed a variable percentage of MeHg with the zooplankton CHYP having the lowest average (6.4 ± 2.0 %) and BUCC having the highest average (56.9 ± 11.6 %; mean ± SE; table 2.3; figure 2.9b). The lowest value recorded for invertebrates was 0.57% for MENO and the highest was 80.7% for SNCR. In general, crustaceans other than zooplankton seemed to contain a high proportion of MeHg with 33.0 ± 10.3 for GAMA, 45.2 ± 14.7 for SNCR and 51.0 ± 7.8 % for SHRI (mean ± SE; table 2.3; figure 2.9b).

A strong positive linear relation was obtained for fish between THg concentration in whole body and in muscle ($Log_{10}[THg_{whole\ body}] = 0.840\ (Log_{10}[THg_{muscle}]) + 0.184$; R^2 =

0.81; $p < 0.001$; figure 2.10). Converting Health Canada's guideline based on muscle tissue (500 ng/g ww) into a value for the whole fish gave an estimate of 282 ng/g ww (1128 ng/g dw).

2.3.4 Mercury in seabirds

THg levels increased throughout the food chain with seabirds ranging from 640 ± 36.1 for COEI to 1789 ± 98.0 ng/g dw (mean ± SE; table 2.3, figure 2.9a) for RAZO. The lowest THg value was 308 ng/g dw for a female COEI and the highest was 3625 ng/g dw for a female GBBG (appendix A). The proportion of THg comprised by MeHg was high and consistent among all of the seabird species in this study with mean values ranging from 97.5 ± 0.83 for COEI to 99.8 ± 0.06 % for GBBG (mean ± SE; table 2.3, figure 2.9b). The lowest value was measured for BLKI with 90.3 %, whereas the highest value was measured in several individuals with 100 % of MeHg (appendix A). GBBG and RAZO showed significantly higher proportions of MeHg compared to BLKI and COEI, whereas HERG was not significantly different than any seabird species ($F_{(0.05)4,30} = 14.5$; $p < 0.001$).

2.3.5 Physical characteristics and mercury

THg concentration increased significantly with organism body weight with a slope of 0.337 ($F_{(0.05)1,270} = 494$; $R^2 = 0.65$; $p < 0.001$). The relationship was even steeper ($F_{(0.05)1,137} = 20.6$; $p < 0.001$) for MeHg with a slope of 0.487 ($F_{(0.05)1,137} = 276$; $R^2 = 0.67$; $p < 0.001$; figure 2.11).

THg level decreased with the lipid content of the different species with a negative slope of -3.01 ($F_{(0.05)1,270} = 347$; $R^2 = 0.56$; $p < 0.001$; figure 2.12). This is more apparent for

MeHg ($F_{(0.05)1,137}$ = 167; R^2 = 0.55; $p < 0.001$) which presents a significantly steeper negative slope of -4.14 ($F_{(0.05)1,137}$ = 9.21; p = 0.003).

2.3.6 Trophic level and mercury

THg was shown to biomagnify in this food web ($F_{(0.05)1,270}$ = 272; R^2=0.50; $p <$ 0.001; figure 2.13) with a biomagnification power (slope of the model) of 0.170. Although MeHg shows a significant relationship with δ^{15}N ($F_{(0.05)1,137}$ = 145; R^2=0.51; $p < 0.001$) with a slope of 0.235, it was not significantly higher than for THg ($F_{(0.05)1,137}$ = 2.53; p = 0.114) meaning that the biomagnification power was not higher for MeHg. There was a difference in Hg concentrations between organisms that are located higher in the food chain. Fish and decapods were lower and seabirds were higher than predicted based on their trophic status (figure 2.13).

When the biomagnification power of this system (b = 0.170) was compared to other ecosystems around the world (table 2.4), this study fell on the lower end of the range. There was a subtle difference between freshwater (small) and marine (large) ecosystems for THg biomagnification power with the former showing a higher average (fresh: b = 0.203 ± 0.013; mean ± SE) than the latter (marine: b = 0.174 ± 0.031; mean ± SE), albeit not statistically significant ($t_{(0.05)17}$ = 0.88; p = 0.202). The number of studies that measured biomagnification power of MeHg was too low to draw conclusions about the effect of the size of an ecosystem.

Hg concentrations in water or in primary producers were not available for all studies. It has been previously suggested that the intercept may be a good indicator of the contamination level at the base of the food chain (Jardine et al. 2006). Therefore, in order to

assess whether the baseline level of Hg could predict the biomagnification potential of a given ecosystem, the intercepts (a) of the regression models were correlated with their biomagnification power (b). The slopes of the models were not correlated significantly with their intercepts for either THg ($t_{(0.017)18} = 0.280$; $r^2 = 0.005$; $p = 0.783$) or MeHg ($t_{(0.05)4} = 0.821$; $r^2 = 0.25$; $p = 0.498$).

The length of the food chain to modify biomagnification potential was not verified in this overview as biomagnification slopes were not correlated significantly with inferred trophic level for either THg ($t_{(0.05)16} = -0.935$; $r^2 = -0.05$; $p = 0.364$) or MeHg ($t_{(0.05)4} = 0.347$; $r^2 = 0.05$; $p = 0.762$).

I tested whether biomagnification potential could be linked to temperature and therefore to latitude. Slopes of THg were not significantly correlated with latitude ($t_{(0.05)18} = -0.228$; $r^2 = -0.003$; $p = 0.822$), whereas intercepts were negatively correlated ($t_{(0.05)18} = -5.561$; $r^2 = -0.66$; $p < 0.001$). These results suggest that the concentration of THg at the base of the food chain decreases towards the North. No correlations were found for MeHg due to the small sample size ($p > 0.017$).

Relationships between Hg concentrations and trophic level were significantly positive for both THg ($F_{(0.05)1,270} = 388$; $R^2 = 0.59$; $p < 0.001$) and MeHg ($F_{(0.05)1,137} = 196$; $R^2 = 0.59$; $p < 0.001$) with slopes of 0.581 and 0.810, respectively (figure 2.14) revealing a high biomagnification potential for these compounds. Contrary to the relationship with $\delta^{15}N$ ($Log_{10}[Hg] = b * \delta^{15}N + a$), the slope for MeHg was steeper than for THg ($F_{(0.05)1,137} = 5.24$; $p = 0.024$) meaning that the biomagnification power was higher for MeHg when taking into account trophic level. Food web magnification factors (FWMFs) showed values of 3.81 for THg and 6.46 for MeHg. THg and MeHg concentrations for most seabirds and some

molluscs were higher than predicted according to their trophic level (figure 2.14). Most fish and some zooplankton contained less THg and MeHg than predicted based on their trophic level, whereas most decapods were lower than predicted for THg only.

When looking at the biomagnification factor corrected for trophic level and using complex predator-prey interactions assessed using stable isotope analysis (SIA) in mixing models (BMF_{SIA}), all poïkilotherms displayed lower mean values than the FWMF for THg, whereas all seabirds (homeotherms) were higher (table 2.5; figure 2.15). BMF_{SIA} were different for MeHg with SHRI and ATHE being markedly higher and BUCC and SAND being slightly higher than the FWMF (table 2.5; figure 2.15). Again, all BMF_{SIA} in seabirds were higher than the MeHg FWMF. When looking at the BMFs involving simple predator-prey interactions (BMF_{TLC}) the conclusions are different for some combinations of species, especially the ones involving CFIN and CAPE as prey which tend to amplify the BMF_{SIA} due to their low Hg levels compared to their predators (table 2.5).

Bioaccumulation factors (BAFs) for THg ranged from 3.47 ± 0.02 log units (mean ± SE) for the zooplankton CFIN to 5.76 ± 0.11 for RAZO (table 2.6). BAF was increasing by an average of 0.78 log units in typical predator-prey interactions (see table 2.5 for predator-prey interactions) ranging from -0.18 log units between CAPE and MENO to 1.84 log units between RAZO and CAPE. A small difference was found between CAPE and CFIN (0.45 log units) and negative values were found with CHYP (-0.16) and MENO (-0.18) meaning that CAPE would feed mainly on CFIN contrary to the proportion values that were found using SIA in mixing models (table 2.5). Differences of BAFs in predator-prey interactions involving seabirds consisted mostly of values greater than 1 log unit.

BAFs for MeHg were higher than for THg ranging from 3.63 ± 0.10 log units (mean ± SE) for the zooplankton CFIN to 6.67 ± 0.03 for RAZO. The differences in BAFs in predator-prey interactions were also higher than for THg with an average of 1.23 log units ranging from -0.30 log units between SNCR and SHRI to 2.75 log units between GBBG and MENO (table 2.6). All BAF predator-prey differences involving seabirds were greater than 1 log unit. MeHg has the potential to increase 4.68 x 10^6 times from water to the top of the food chain while THg has the potential to increase 5.75 x 10^5 times.

2.3.7 Connectivity with benthos and mercury

Positive linear relationships were significant but weak between Hg concentration and $\delta^{13}C$ for the overall models for THg ($F_{(0.05)1,269} = 38.6$; $R^2 = 0.13$; $p < 0.001$) and MeHg ($F_{(0.05)1,137} = 14.2$; $R^2 = 0.09$; $p < 0.001$) with slopes of 0.164 and 0.187, respectively. The slopes were not significantly different ($F_{(0.05)1,137} = 0.38$; $p = 0.538$). The seabirds being highly contaminated and intermediate in terms of connection with benthos (figure 2.16a, c), 5th and 3rd order polynomials best fitted THg ($F_{(0.05)5,265} = 36.9$; $R^2 = 0.41$; $p < 0.001$) and MeHg data ($F_{(0.05)3,135} = 24.9$; $R^2 = 0.36$; $p < 0.001$), respectively. When looking at individual groups, positive linear relationships with $\delta^{13}C$ could be established for fish and invertebrates for MeHg concentrations ($p < 0.05$; figure 2.16c) whereas a 2nd order polynomial relationship was found for invertebrates to best fit the THg data ($F_{(0.05)3,110} = 29.7$; $R^2 = 0.45$; $p < 0.001$; figure 2.16a). No significant difference was found between the slopes of fish and invertebrates for MeHg ($F_{(0.05)1,100} = 0.16$; $p = 0.690$). No significant relationships were found for seabirds for both THg ($F_{(0.05)1,99} = 0.05$; $R^2 < 0.001$; $p = 0.82$) and MeHg ($F_{(0.05)1,99} = 0.07$; $R^2 = 0.002$; $p = 0.790$).

Because I wanted to know if organisms' connectivity with the benthos could predict Hg concentrations, data were normalized to the variable known to best predict Hg, that is to say total length for fish and $\delta^{15}N$ for seabirds and invertebrates. Total length instead of $\delta^{15}N$ was chosen for fish because the fit of the data was better for the former. After normalizing for these variables, relationships became weaker or no longer statistically significant (figure 2.16) meaning that these confounding variables were important to predict Hg concentrations. Positive linear relationships were significant between Hg concentration and $\delta^{13}C$ for the overall models for $THg_{normalized}$ ($F_{(0.05)1,269} = 42.7$; $R^2 = 0.14$; $p < 0.001$) and $MeHg_{normalized}$ ($F_{(0.05)1,137} = 8.49$; $R^2 = 0.06$; $p = 0.004$) with slopes of 0.164 and 0.130, respectively. Although $MeHg_{normalized}$ slope was weaker than for $THg_{normalized}$, it was not significantly different ($F_{(0.05)1,137} < 0.001$; $p = 0.995$) meaning that Hg concentrations in benthic species were not determined exclusively by MeHg. Like the non-normalized data, 5th and 3rd order polynomials best fitted $THg_{normalized}$ ($F_{(0.05)5,265} = 35.5$; $R^2 = 0.40$; $p < 0.001$) and $MeHg_{normalized}$ data ($F_{(0.05)3,135} = 17.9$; $R^2 = 0.28$; $p < 0.001$), respectively. When looking at the individual groups, no significant difference was found between the slopes of fish and invertebrates for $THg_{normalized}$ ($F_{(0.05)1,166} = 1.18$; $p = 0.278$; figure 2.16c). No relation was found for seabirds in both $THg_{normalized}$ and $MeHg_{normalized}$ and for fish in $MeHg_{normalized}$ ($p > 0.05$).

After length and $\delta^{15}N$ normalization, THg and MeHg concentrations were higher in benthic habitat and/or in benthopelagic habitat (mixture) compared to pelagic habitat for all taxonomical groups taken individually ($p < 0.05$; figure 2.17). The omnivore GBBG and the diver RAZO were the most contaminated seabirds followed by the benthivore COEI, the omnivore HERG and the least contaminated surface-feeder BLKI for both $THg_{normalized}$

($F_{(0.05)4,96} = 27.4$; $p < 0.001$) and $MeHg_{normalized}$ ($F_{(0.05)4,30} = 11.6$; $p < 0.001$; Figure 2.17). The multiple comparison analysis for birds gave different results than when Hg data were not normalized (figure 3.5; Chapter 3). The transformed data for lower trophic level birds such as GBBG, HERG and COEI produced higher $THg_{normalized}$ and $MeHg_{normalized}$ concentrations than the non-transformed data, whereas the higher trophic level birds (BLKI and RAZO) produced lower values. Since there was no significant difference in the $\delta^{13}C$ signature (figure 2.8), further interpretation of Hg based on this isotope is not possible.

Among fish, benthic AMPL and WIFL as well as the benthopelagic SAND showed higher $THg_{normalized}$ concentrations than the pelagic CAPE and ATHE ($F_{(0.05)4,51} = 9.98$; $p < 0.001$). A different conclusion is drawn when looking at $MeHg_{normalized}$ concentrations where fish were not significantly different ($F_{(0.05)4,34} = 2.05$; $p = 0.110$) even though SAND seemed more contaminated than other fish.

Benthopelagic invertebrates had higher $THg_{normalized}$ levels than benthic and pelagic invertebrates ($F_{(0.05)2,111} = 46.9$; $p < 0.001$), while benthic and benthopelagic invertebrates had similar high $MeHg_{normalized}$ values followed by pelagic invertebrates having the lowest values ($F_{(0.05)2,62} = 8.86$; $p < 0.001$). Interspecific variations were very different between $THg_{normalized}$ and $MeHg_{normalized}$ (figure 2.17). This is due to the high variability in percentage of MeHg in invertebrates (figure 2.9b).

2.3.8 Trophic position and physical characteristics to predict mercury concentration

Organism weight was the most important predictor term in the THg model ($b' = -0.405$; $p < 0.001$) followed closely by lipid content ($b' = -0.327$; $p < 0.001$) and finally by trophic level ($b' = 0.253$; $p < 0.001$; Table 2.7). The general model explained 74.5% of the

variability of THg concentration ($p < 0.001$). The model was slightly different for MeHg showing the opposite order of importance of the last two terms with weight as the most important predictor in the model ($b' = 0.412$; $p < 0.001$) followed by trophic level ($b' = 0.305$; $p < 0.001$) then lipid content ($b' = -0.301$; $p < 0.001$). The general model explained 77.8% of the variation of MeHg in this food web ($p < 0.001$). Organisms' connectivity with the benthos was not an important predictor for THg ($t_{(0.05)268} = -1.50$; $p = 0.134$) or MeHg concentrations ($t_{(0.05)135} = 0.84$; $p = 0.360$) since $\delta^{13}C$ had been excluded from both multiple linear regression models ($p > 0.05$). However, in the model using IHg as the dependant variable, $\delta^{13}C$ was the second best predictor term ($b' = 0.292$; $p = 0.001$) after weight ($b' = -0.360$; $p = 0.001$) with a poor model explaining only 17.1% ($p < 0.001$), suggesting a weak association of IHg with the benthos. When using the percentage of MeHg as the dependant variable, weight was the most important predictor term ($b' = 0.489$; $p < 0.001$) followed by lipid content ($b' = -0.323$; $p < 0.001$), benthic connection ($b' = -0.258$; $p < 0.001$) and trophic level ($b' = 0.489$; $p = 0.001$) with a model explaining 68.9% of the variation ($p < 0.001$). The order of importance for the percentage of IHg was similar to the MeHg concentration model with weight ($b' = -0.453$; $p < 0.001$), trophic level ($b' = -0.297$; $p < 0.001$) and lipid content ($b' = 0.280$; $p < 0.001$) as the three most important predictors with the addition of benthic connection ($b' = 0.208$; $p = 0.001$) as the least important. This model explained 68.2% of the variation of IHg percentage ($p < 0.001$). None of the standardized partial coefficients within a model were significantly different ($t_{\alpha(n)}$; $p > 0.05$).

A principal components analysis (PCA) using total weight, lipid content, trophic level and benthic connection revealed that the first component explained 63.7% of the variation of the data throughout the entire food web (figure 2.18). The loadings for

component 1 were lipid content (-0.565), weight (0.562), trophic level (0.460) and benthic connection (0.392), in order of importance. This order was similar to the one obtained from the MLRs. The second component explained 23.8% of the variation. The loadings for component 2 were benthic connection (0.756), trophic level (-0.619), weight (-0.161) and lipid content (-0.140).

The relationship between Hg concentrations and component 1 offered a good fit of the data for both THg ($F_{(0.05)1,270} = 628$; $R^2 = 0.70$; $p < 0.01$) and MeHg ($F_{(0.05)1,137} = 326$; $R^2 = 0.70$; $p < 0.01$) with a significant difference between the slopes ($F_{(0.05)1,137} = 13.0$; $p < 0.01$; figure 2.19). This corroborates the results obtained with the MLR and the PCA showing that 70 to 80% of the variation of THg and MeHg are explained by the models and that total weight, lipid content and trophic level are the best predictors and benthic connection is the least important predictor of Hg concentration in an organism.

2.4 DISCUSSION

2.4.1 Water physico-chemistry

Water THg values in my study were well below the interim water quality guidelines for protection of wildlife in marine environments of 16 ng/L (Environment Canada 2003). No guideline was available for MeHg in marine environments, but a level of 4 ng/L is suggested for freshwater ecosystems (Environment Canada 2003). However, these guidelines do not guarantee the protection of predatory species that are at the top of the food chain (CCME 2003). The CCME (2003) thus suggests a MeHg level of 0.007 ng/L for the protection of wildlife. The USEPA recommends a MeHg water concentration of less than 0.05 ng/L and a THg water concentration of less than 0.641 ng/L for protecting birds from consuming more than the tolerable daily intake (USEPA 1997). My MeHg data exceed the CCME and the USEPA levels by 16.6 and 2.3 times on average, respectively. My THg values are 1.4 times higher than the USEPA guideline (USEPA 1997). Predatory species in this food web may therefore be at risk.

THg concentrations in water increased with depth as predicted, whereas MeHg concentrations did not show the expected positive trend with depth; on the contrary, it decreased severely. The range of my THg values in the water column is in agreement with Cossa and Gobeil (2000) in the Lower St. Lawrence Estuary who found THg concentrations between 0.36 and 1.6 ng/L. Contrary to my results however, they found higher values and variability of THg concentrations in surface water with a sharp decrease in values and variability with depth. Interestingly, they found high MeHg concentrations in sediment pore water (1.0 to 2.9 ng Hg/L).

According to my results, deep waters did not provide the ideal conditions for Hg methylation. The pH was expected to decrease with depth and the DOC concentration was expected to increase, providing the ideal conditions for Hg methylation and for sustaining the pool of MeHg. Although pH followed the predicted pattern, it was not low enough to determine the potential of methylation. A pH lower than 6 would have provided the proper conditions for methylation (Grieb et al. 1990, Spry and Wiener 1991). It has been shown that THg levels in fish increased with a decrease in pH in 120 lakes in Wisconsin ($R^2 = 0.69$; $p < 0.001$; Burgess and Meyer, 2008). DOC concentrations did not display any trend with depth. Suspended particulate matter is known to provide a good binding site for MeHg (Fitzgerald et al. 2007). Suspended particulate matter is found in greater concentrations at the surface waters of the Gulf of St. Lawrence (Yeats 1988a) and could therefore explain the higher percentage of MeHg in surface waters (Cossa and Gobeil 2000). The methylation process known to occur in anoxic sediments could possibly have been inhibited by the high concentration of sulphate in the water (Gilmour and Henry 1992, Weber 1993). A concentration higher than 5 mM of sulphate ions is known to stop the methylation potential by SRB and the sulphate concentration in oceans is estimated at 28 mM (Gilmour and Henry 1992).

I also expected to observe an increase in $\delta^{13}C$ values with depth which would have corroborated my confidence in ^{13}C enrichment in benthic organisms. Instead, I found that $\delta^{13}C$ decreased with depth. Tan and Strain (1988) also found more negative $\delta^{13}C$ values for total inorganic carbon (TIC) with depth in the Gulf of St. Lawrence with an average of 2‰ difference between surface and bottom water. They explained the increase in the surface waters by a high exchange with atmospheric carbon as well as fractionation due to

photosynthesis, whereas lower values at the bottom may have been due to a limited access to atmospheric carbon and to decomposition of organic matter.

2.4.2 Trophic structure

I found in this study that the source of energy was mostly derived from POM and zooplankton meaning that the majority of the organisms in this food web feed or depend upon these pelagic sources. The low $\delta^{13}C$ values in POM suggest a contribution of allochtonous source of primary production coming from freshwater systems (Peterson and Fry 1987). Littoral and benthic organisms were important in the diet of COEI that are known to feed extensively on BLMU and URCH (Guillemette et al. 1992). The generalists GBBG and HERG acquired their energy from both littoral and benthic sources in my study. It is also possible that GBBG and HERG were feeding on terrestrial sources (see Chapter 3) Similar patterns were observed in the North Water of northern Baffin Bay where a positive correlation was obtained between $\delta^{15}N$ and $\delta^{13}C$ for pelagic organisms ($r^2 = 0.79$) but no pattern was apparent for benthic organisms (Hobson et al. 2002). The range of isotopic values was similar to those of studies conducted elsewhere (when considering only comparable species) with pelagic component being low in $\delta^{13}C$ compared to the benthic component (McConnaughey and McRoy 1979, Hobson et al. 1995, Hobson et al. 2002, Riget et al. 2007) and predatory species being high in $\delta^{15}N$ compared to primary consumers (Hobson and Welch 1992, Atwell et al. 1998, Fisk et al. 2001, Hobson et al. 2002, Riget et al. 2007). Moreover, the ranges of my $\delta^{15}N$ and $\delta^{13}C$ values were similar to some Arctic (Hobson and Welch 1992, Hobson et al. 2002, Campbell et al. 2005, Dehn et al. 2006, Riget et al. 2007) and East Pacific (Jarman et al. 1996) food webs (when considering only

comparable species). My data showed lower $\delta^{13}C$ values compared to the Gulf of Oman (Al-Reasi et al. 2007) and higher compared to freshwater systems (Bowles et al. 2001, Power et al. 2002, Kidd et al. 2004). My isotopic data were similar to those found by Lesage et al. (2001) in the St. Lawrence Gulf and Estuary.

Benthic organisms had higher $\delta^{13}C$ values compared to organisms that inhabited either pelagic or both benthic and pelagic environments. This was particularly apparent for WIFL and AMPL among fish and most molluscs and URCH among invertebrates. In an extensive review on the St. Lawrence, Tan and Strain (1988) showed that $\delta^{13}C$ values in sediment (-22.4 ‰) were higher compared to those in particulate organic carbon in the water column (-24.9 ‰). Moreover, these authors found that $\delta^{13}C$ values in particulate organic carbon were slightly higher in deep (> 50 m) waters (-22.7 ‰) compared to shallow (< 50 m) waters (-23.9 ‰). On the other hand, they found lower $\delta^{13}C$ values for total inorganic carbon (TIC) in deeper water compared to the surface. When looking at the species level, GAMA did not show any difference with the benthopelagic organisms (SHRI, SNCR and BLMU). GAMA is known to be a scavenger and the inclusion of pelagic sources in its diet cannot be ruled out.

Differences in $\delta^{13}C$ signatures were expected among seabirds since they exploit different habitats and obtain their food in different ways (see Chapter 3 of this thesis). For instance, BLKI is more or less an obligate surface-feeder (Bayer 1983, Baird 1994), whereas GBBG and HERG have access to food sources other than those of pelagic origin (Pierotti and Good 1994, Chapdelaine and Rail 1997, Good 1998, Rail and Chapdelaine 2000). RAZO was considered in both pelagic and benthic environments in this study since it feeds almost exclusively on CAPE (pelagic fish) and SAND (benthopelagic fish) in the Gulf of St.

Lawrence (Chapdelaine and Brousseau 1996). RAZO can dive to depths of 150 m (Piatt and Nettleship 1985) and therefore may have access to fish that are more connected to the benthos. Finally, most of COEI's diet is composed of benthic prey (Cantin et al. 1974, Guillemette et al. 1992). The reason to explain the lack of difference in $\delta^{13}C$ values among seabirds in spite of their distinct feeding ecology is still unknown.

2.4.3 Mercury in fish and invertebrates

THg values for fish and invertebrates in my study were below Health Canada's guideline for human food consumption (Health Canada 2007). Values in my study were calculated using whole fish and therefore underestimated Hg levels in the edible portion of the fish (i.e., muscle) which was used in the development of abovementioned guideline (Health Canada 2007). THg concentrations were analysed in whole fish and muscle tissue in the same individuals in order to predict THg concentration in whole fish using muscle tissue. I found a value of 282 ng/g ww (1128 ng/g dw) for whole fish as a guideline for human food consumption. This latter value was closer to the THg levels in fish and invertebrates in this food web, but not alarming.

Hg levels found in this study were below the MeHg LOAEL and NOAEL suggested for a wild freshwater piscivorous bird (Kenow et al. 2003, Meyer 2006). However, these estimations were done on chicks and the authors suggest that birds may be more vulnerable after fledgling since Hg excretion via feathers is interrupted. These thresholds may eventually be reviewed and reduced for adult birds. Similar thresholds were found by Burgess and Meyer (2008) who observed significant reproductive impairment in breeding *G.*

immer feeding on prey averaging 0.21 μg/g ww and reproductive failure at concentrations over 0.41 μg/g ww.

A certain proportion of fish and invertebrates in my study exceeded THg values intended for the protection of piscivorous wildlife (Yeardley et al. 1998, CCME 2000). Predatory species located at the top of this food chain and feeding on prey exceeding suggested values could potentially be threatened and exhibit toxicological effects. However, caution should be taken in the interpretation of these thresholds. The seabird used by the CCME for the calculation of the tissue residue guideline (TRG) probably overestimates the value since *O. oceanicus* eats almost its complete body weight on a daily basis (FI:bw = 0.94; CCME, 2000). Seabirds in my study consume a lower proportion of their body weight according to the CCME (2000). As a consequence, the TRG value would increase. Additionally, Yeardley et al (1998) measured THg in whole fish and assumed a percentage of MeHg of 100 %. As seen in my study, percentage of MeHg over THg in fish is variable and averages 58.2 ± 2.61 %.

Percentage of MeHg over THg for whole fish in my study (58.2 ± 2.61 %) was much lower than reported for the edible portion of fish (i.e., muscle) with an estimate of over 95 % (Grieb et al. 1990, Bloom 1992). This divergence is due to the difference in percentage of MeHg in different organs. Liver and kidney are known to have a lower proportion of MeHg than muscle tissue in birds (Kim et al. 1996). This reduces the overall proportion of MeHg in the whole fish. Zooplankton showed values similar to the literature which indicated a mean proportion of 10 % (range: 1-19%) for whole organisms (Al-Reasi et al. 2007). The higher proportion of MeHg in some molluscs such as BUCC and some crustaceans such as GAMA, SHRI and SNCR could be explained by their feeding behaviour involving benthic

prey items (Himmelman and Hamel 1993, Lovrich and Sainte-Marie 1997, Savenkoff et al. 2006). Organisms connected to the benthos are believed to display elevated concentrations of MeHg due to greater access to the MeHg pool in the sediment layer (Cossa and Gobeil 2000). The methylation process via sulphate-reducing bacteria taking place in the anoxic zone of the sediment (Compeau and Bartha 1985, Gilmour et al. 1992) could influence MeHg uptake for organisms living and/or feeding in the benthos. However, percentage of MeHg increases with trophic level (tables 2.2 and 2.3) and SNCR, GAMA and BUCC are known to feed on carrion (Himmelman and Hamel 1993, Lovrich and Sainte-Marie 1997, Savenkoff et al. 2006) and therefore on prey items located potentially high in the food chain. This could have increased the exposure of these organisms to MeHg.

2.4.4 Mercury in seabirds

MeHg levels in all seabirds in my study were well below the adverse effect threshold of 3 000 ng/g ww in whole blood (10 000 ng/g dw assuming 70% of water in blood; appendix A) of *G. immer* advised by Evers et al. (2008). Adverse reproductive, neurological and morphological effects have been shown to occur above that limit for *G. immer* (Evers et al. 2008). The authors identified 3 risk categories: < 1 μg/g ww as low; between 1 and 3 μg/g ww as moderate and > 3 μg/g ww as high. The vast majority of the seabirds in my study were considered low according to that classification. Only 1 female GBBG fell into the moderate risk category. Burgess and Meyer (2008) estimated a 50% reduction in productivity of breeding loons when THg levels in blood of females were at 4.3 μg/g ww and a complete failure when levels were over 8.6 μg/g ww. The latter levels in females corresponded to THg concentrations in whole prey as low as 0.21 and 0.41 μg/g ww,

respectively (Burgess and Meyer 2008). On the other hand, THg concentrations over 95 μg/g ww were found in healthy black-footed albatross (*Diomedea nigripes*; Honda et al., 1990) suggesting that seabirds can accumulate high concentration of Hg without showing apparent adverse effects. It has been shown that the presence of seleno-DL-methionine decreased the toxicity of MeHg in adult *A. platyrhynchos* (Heinz and Hoffman 1998). The authors found that birds dosed with both seleno-DL-methionine and MeHg showed lower adverse neurological effects than birds dosed with MeHg only. The occurrence of selenium in nature could decrease the potency of MeHg allowing birds to tolerate high levels of MeHg. Those conclusions differ from what one would predict from the guideline for the protection of fish-eating birds (Yeardley et al. 1998) and from the TRG for wildlife (CCME 2000). Percentage of MeHg around 100% in blood was consistent with other studies (Wolfe et al. 1998).

2.4.5 Physical characteristics and mercury

THg and MeHg concentrations increased as organism size increased and lipid content decreased. Organisms generally occupy a higher trophic level as they increase in size (Deudero et al. 2004). This trend is generally observed among species (Jennings et al. 2001), but there is also evidence of intraspecific ontogenic shifts for some fish species (Deudero et al. 2004). Deudero et al (2004) found that fish show an increase of $\delta^{15}N$ values with length within species. The size of an organism determines the range of prey sizes it can consume (Cohen et al. 1993). When reaching an appropriate size, fish are able to feed on larger and higher trophic level prey items (Cohen et al. 1993) and consequently increase their Hg exposure (Desta et al. 2008). The variation in my study was probably explained in

large part by interspecific effect i.e., bigger organisms such as seabird species showed higher trophic levels and higher THg levels than smaller organisms such as zooplankton. Hg concentrations decreased with an increase in lipid content. Lipid content in the tissue used for seabirds (RBCs) was low and showed high Hg concentrations, whereas zooplankton contained high lipid content and low Hg levels. MeHg binds to water-soluble thiol groups of amino acids such as cysteine and methionine (Wolfe et al. 1998, Simmons-Willis et al. 2002). A recent study found a negative correlation between THg and lipid content in southern bluefin tuna (*Thunnus maccoyii*, Balshaw et al. 2008). Organisms containing a higher proportion of lipids in their tissues are therefore more likely to have lower Hg concentrations.

2.4.6 Trophic level and mercury

The relationship with $\delta^{15}N$ indicated that THg and MeHg biomagnified at similar rates in this food web, whereas the relationship with trophic level revealed that MeHg biomagnified at a higher rate than THg. The former result was surprising considering that MeHg is known to be the species that biomagnifies (Watras and Bloom 1992, Watras et al. 1998, Bowles et al. 2001, Campbell et al. 2005, Riget et al. 2007). The higher affinity of MeHg for proteins was expected to generate a higher biomagnification potential than for THg. Consistent with this prediction, the percentage of MeHg over THg increased substantially with trophic level (tables 2.2 and 2.3).

Higher and lower than expected concentrations based on stable isotope results were found for several organisms and this discrepancy could be due to differences in energy requirements of these organisms. Seabirds were the only homeotherms represented in this

study and have high energy requirements to sustain their metabolism, while poïkilotherms require less energy and are therefore less exposed to Hg accumulation via food intake (Fisk et al. 2001). Furthermore, the turnover rate of the contaminant is different than the turnover rate of $\delta^{15}N$ in the tissue selected (Fisk et al. 2001). For example, in whole blood of *C. diomedea* MeHg has a half-life of 44 to 65 days (Monteiro and Furness 2001) and $\delta^{15}N$ has a half-life of 14.4 days (Bearhop et al. 2002). A higher MeHg exposure of the birds to higher trophic level prey items two months before sampling blood would be reflected in the MeHg concentrations but not in the $\delta^{15}N$ signature. Also, Hg analyses were done on RBCs in seabirds. RBCs are high in haemoglobin and glutathione which regulate MeHg distribution in the blood (Doi and Tagawa 1983). The use of this tissue could therefore overestimate the Hg concentrations in the overall body compared to other studies that used seabird eggs (Jarman et al. 1996) or muscle tissue (Atwell et al. 1998, Campbell et al. 2005, Riget et al. 2007). Similarly, my Hg analyses were done on whole fish which gave a lower estimate of Hg concentration compared to other studies that used muscle tissue (Atwell et al. 1998, Campbell et al. 2005, Al-Reasi et al. 2007, Riget et al. 2007). It has been shown that THg levels were higher in muscle tissues than whole fish (Peterson et al. 2005, Peterson et al. 2007, this study). The choice of whole fish as a proxy was based on the assumption that this component was the most likely to be assimilated by the following trophic level. Seabirds and fish usually ingest the entire prey. Muscle tissue is a logical choice for studies intended to be applied to organisms being consumed by humans, but whole organisms are more relevant to food web studies conducted in the field and applied to wildlife.

The biomagnification of THg and MeHg in my study were similar to other studies around the world. The overview on biomagnification power done in this study did not allow

me to validate the ecosystem size-driven biomagnification hypothesis for THg. The lack of significant effect is probably due to some exceptions showing lower (Power et al. 2002, Ikemoto et al. 2008) and higher (Jarman et al. 1996) than expected values (table 2.4). Also, freshwater lakes were assumed to be small in my calculation which is inaccurate for some of the large and diverse lakes in my review (e.g., Lake Victoria, Campbell et al. 2003). Complex predator-prey interactions could diminish the exposure of THg for species that consume a high range of trophic level prey items. In effect, the exposure of a predator is reduced when it feeds on lower trophic level prey since an exponential relationship between THg and trophic level is observed (for review, see table 2.4). Studies considering MeHg were not sufficient to draw conclusions about ecosystem size as a predictor of biomagnification power. Moreover, studies that measured MeHg were conducted in large ecosystems, except Bowles et al (2001) which revealed the highest MeHg biomagnification power. The latter study was conducted in Lake Murray (Papua New Guinea) where conditions for methylation of Hg were suitable (a shallow lake with wetlands, high perimeter/area ratio, fluctuation of the level of water, low pH (Bowles et al. 2001)). Surprisingly, the authors reported low water concentrations of THg and MeHg with 1.42 ng/L and 0.067, respectively (table 2.4.).

Because the Hg-trophic level relationship is exponential, the higher level of THg at the base of a food chain is expected to enhance biomagnification. Biomagnification powers of the studies under investigation were not correlated with the THg level at the base of the food chain. However, calculations were done using the intercept of statistical models. Model intercepts could be erroneous estimates of the true level of THg at the base of the food chain. THg concentrations in water or in primary producers (e.g., phytoplankton) are thus

required to further test the hypothesis of greater THg biomagnification with greater levels at the base of the food chain.

I was unable to corroborate the food chain length-driven hypothesis. However, the inferred trophic level was an estimation based on the assumption that the baseline organism (see equation 2.4) occupied either a primary consumer or a secondary consumer position. This assumption might be incorrect, although comparisons between my inferred trophic level and those reported in the literature are fairly similar (table 2.4).

I was able to show that even though the THg baseline level was higher at lower latitudes, the biomagnification power remained unchanged. This suggests that the dilution process of Hg in tropical fish would control the biomagnification power even though the baseline level is higher. Organisms are known to grow faster in warmer regions (Pauly 1998) and therefore to dilute the amount of Hg contained in their body (Campbell et al. 2003). Methylation rate was positively correlated with temperature in six remote Canadian lakes (Bodaly et al. 1993) which would explain the increase of Hg baseline level in ecosystems closer to the equator. Inversely, it has been observed that as water temperature increases, the metabolic rate of fish increases which in turn augments food consumption and Hg exposure (Harris and Bodaly 1998). My overview does not corroborate the latter hypothesis.

Influence of other parameters such as the productivity, the diversity of species, pH and concentration of DOC on biomagnification power could not be established due to a lack of information in the different studies. Further information is needed on different ecosystems in order to draw conclusions. Authors should systematically report the aforementioned results when possible.

Food web magnification factor (FWMF) was higher for MeHg than for THg. The values were comparable with highly bioaccumulative lipophilic persistent organic pollutants (POPs), such as dieldrin, dichloro-diphenyl-dichloroethane (*p,p'*-DDD), dichloro-diphenyl-trichloroethane (*p,p'*-DDT) and most polychlorinated biphenyls (PCBs, Fisk et al. 2001). These compounds are known to biomagnify due to their high $\log_{10}$ octanol-water partition coefficient (K_{ow}) and therefore their high affinity for lipids (Fisk et al. 2001). Biomagnification factors of THg and MeHg using stable isotopes (BMF_{SIA}) were also comparable with POPs (Fisk et al. 2001). Values were higher and lower than predicted for most homeotherms (seabirds) and poïkilotherms (fish and invertebrates), respectively. As stated above, this is likely due to the higher energy requirement of warm blooded organisms to sustain their metabolism. Likewise, higher and lower than expected values were found in seabirds and fish, respectively for both PCB 180 (Fisk et al. 2001) and PCB 153 (Hobson et al. 2002). BMF_{TLC} and BMF_{SIA} showed different results. However, BMF assessment using complex interactions (BMF_{SIA}) seems a better choice than using simple predator-prey interactions since this calculation integrates a weighted average of potential prey items likely to constitute a realistic diet of the predators. Therefore, the assimilation of Hg via food intake is more plausible.

High bioaccumulation factor (BAF) values were found in RAZO for THg and MeHg with a potential to increase 5.75×10^5 and 4.68×10^6 times from water to the top of the food chain, respectively. Higher BAF values for MeHg compared to THg are due to the higher potential of MeHg to biomagnify compared to inorganic Hg.

2.4.7 Connectivity with benthos and mercury

$THg_{normalized}$ and $MeHg_{normalized}$ levels in fish and invertebrates were found to increase with $\delta^{13}C$ (except $MeHg_{normalized}$ for fish), whereas seabirds did not show any trend. Benthic connection has been used to explain interspecific differences in Hg when removing the preponderant effects of trophic level (seabirds and invertebrates) and total length (fish). $THg_{normalized}$ and $MeHg_{normalized}$ concentrations were higher for benthic species and species exploiting both pelagic and benthic environments compared to pelagic species.

The generally higher concentrations of Hg among organisms feeding on benthos compared to organisms feeding on pelagos in this study are consistent with the literature (Mason and Fitzgerald 1990, 1993, Monteiro et al. 1996, Monteiro and Furness 1997, Thompson et al. 1998, Cossa and Gobeil 2000). This relationship is believed to result from the methylation of mercury taking place in hypoxic or anoxic deep waters (Mason and Fitzgerald 1990, 1993). Sulphate-reducing bacteria are known to be an important cause of Hg methylation taking place in the anoxic zone of sediments (Compeau and Bartha 1985, Gilmour et al. 1992). Cossa and Gobeil (2000) found that the sediment layer in the Lower St. Lawrence Estuary was a source of Hg for wildlife. The generally higher concentration (although not always significant) for organisms exploiting both pelagic and benthic environments compared to organisms exploiting either pelagic or benthic environments is difficult to interpret. Abiotic factors such as sunlight radiation were shown to produce MeHg in surface water of freshwater lakes (Siciliano et al. 2005). According to the authors, photoproduction may be linked to dissolved organic matter (DOM) concentration. Kidd et al. (2003) showed that for normalized weight and trophic position, THg concentrations were significantly higher in pelagic fish (with low $\delta^{13}C$ values) compared to benthic fish (with

high $\delta^{13}C$ values). They suggest that the most plausible explanations for this pattern are the difference between habitats of Hg input at the base of the food chain or the different growth rates of the fish. However, if such processes would take place at appreciable levels in this ecosystem, pelagic organisms would display higher levels than I found.

2.4.8 Trophic position and physical characteristics to predict mercury concentration

When looking at the variables mentioned above that are believed to explain Hg variability in this food web, I found that mercury can be predicted using foraging strategies such as trophic level ($\delta^{15}N$) and benthic connection ($\delta^{13}C$) as well as physical characteristics such as total weight and lipid content. In all cases, total weight explained more of the variation in Hg levels followed by trophic level or lipid content. $\delta^{13}C$ was not a good predictor term for either THg or MeHg. However, benthic connection was significant for inorganic Hg concentration and percentage as well as MeHg percentage. Similar results were obtained by Kidd et al. (2003) who found that weight, $\delta^{15}N$ and $\delta^{13}C$ explained 74% of the variation of THg in fish ($p < 0.001$).

2.5 CONCLUSIONS

THg increased in the water column with depth, whereas there was no trend in MeHg concentration. The sediment layer is believed to be a source for inorganic Hg in the Gulf of St. Lawrence which corroborates my hypothesis. However, the conditions for methylation in the profundal zone of the water column were insufficient to increase the pool of MeHg. Concentrations of THg and MeHg were greater among organisms living or feeding in the

benthic environment than those living in the pelagic environment. This suggests high concentrations of MeHg in the sediments that are readily assimilated by benthic organisms.

Although $\delta^{13}C$ values in total and dissolved organic and inorganic carbon decreased with depth, $\delta^{13}C$ was still a good indicator of benthic connection since benthic species displayed high $\delta^{13}C$ values relative to pelagic species.

THg and MeHg were found to biomagnify with values in upper trophic level predators being several orders of magnitude greater than the environment in which they lived or fed. Biomagnification potentials of THg and MeHg in my study were comparable to those found in other food webs and to those of persistent organic pollutants. I was unable to corroborate hypotheses based on the influence of food chain length, ecosystem size, baseline contamination level or temperature on the magnitude of biomagnification. However, baseline Hg level was negatively correlated with latitude suggesting that temperature affects the uptake of Hg by organisms.

In this food web, mass, trophic level and lipid content were found to be the best predictors of THg and MeHg. Connectivity with benthos was generally a poor predictor of Hg concentrations, but it did explain variation in some cases. It was shown that benthic species and species that are exploiting both pelagic and benthic habitats were more contaminated than pelagic species. Further studies are needed to explain the latter finding.

Table 2.1. Mean (± SE) THg and MeHg (ng/L), percentage of MeHg (%), pH, salinity, $\delta^{13}C$ (‰) and concentration of carbon (mg/L) of TIC/TOC, DIC/DOC measurements in water samples in relation to depth (0, 85 and 170 m). The significance of the overall models and the slopes are given for every measurement. Slopes are in bold when significantly different than zero.

Measurements		Depth			p	R^2	b	b_2
		Surface (n=2)	85 m (n=2)	170 m (n=2)				
[THg]		0.458 ± 0.126	0.531 ± 0.052	1.670 ± 0.022	0.005 [a]	0.971	**-0.004**	**0.000**
[MeHg]		0.117 ± 0.001	0.124 ± 0.005	0.107 ± 0.002	0.301	0.260	-0.000	
MeHg (%)		27.6 ± 5.2	22.2 ± 3.5	6.4 ± 0.2	0.039 [b]	0.695	**-0.001**	
pH		8.01 ± 0.01	7.76 ± 0.01	7.69 ± 0.01	<0.001 [a]	0.996	**-0.004**	**0.000**
Salinity		39.4 ± 0.35	42.8 ± 0.15	43.00 ± 0.10	0.002 [a]	0.982	**0.059**	**-0.000**
DIC	[C]	27.4 ± 0.43	30.8 ± 0.18	31.6 ± 0.18	0.002 [a]	0.983	**0.055**	**-0.000**
	$\delta^{13}C$	1.38 ± 0.04	0.42 ± 0.04	0.20 ± 0.08	0.001 [a]	0.992	**-0.016**	**0.000**
DOC	[C]	0.77 ± 0.02	0.66 ± 0.01	0.66 ± 0.04	0.055	0.643	-0.001	
	$\delta^{13}C$ [c]	--	--	--				
TIC	[C]	27.5 ± 0.00	30.2 ± 0.25	30.5 ± 0.06	0.001 [a]	0.992	**0.046**	**<0.001**
	$\delta^{13}C$ [c]	1.51 ± 0.03	0.49 ± 0.01	0.26 ± 0.05	<0.001 [a]	0.997	**-0.017**	**<0.001**
TOC	[C]	0.64 ± 0.04	0.57 ± 0.01	0.55 ± 0.01	0.022	0.768	**-0.001**	
	$\delta^{13}C$ [c]	--	--	--				

[a] Significance of the quadratic polynomial model

[b] Values were arcsine-transformed for the statistical analysis

[c] Values below the detection limit

Table 2.2 Mean (± SE) stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope values and trophic level for all species collected in a Gulf of St. Lawrence food web, Québec, Canada in 2006-07. Feeding habitats and feeding modes are shown. Codes in this table are used throughout the thesis.

Taxa	Code	Feeding [a]	*n*	$\delta^{15}N$ (‰)	Trophic level [b]	$\delta^{13}C$ (‰)
Particulate organic matter [c]						
	POM		4	4.16 ± 0.28	1.20 ± 0.08	-23.15 ± 0.58
Invertebrata						
Crustacea						
Zooplankton						
Calanus finmarchicus [d]	CFIN	PL, GR	10	8.33 ± 0.13	2.42 ± 0.04	-22.71 ± 0.05
Calanus hyperboreus [d]	CHYP	PL, GR	5	8.02 ± 0.12	2.33 ± 0.04	-22.44 ± 0.05
Meganyctiphanes norvegica [d]	MENO	PL, PR	6	9.19 ± 0.18	2.68 ± 0.05	-21.09 ± 0.05
Amphipoda						
Gammarellus sp. [d]	GAMA	LI/BE, PR/SC	10	8.13 ± 0.07	2.37 ± 0.02	-19.57 ± 0.17
Decapoda						
Shrimps	SHRI		10	12.44 ± 0.11	3.63 ± 0.03	-19.55 ± 0.22
Pandalus borealis [d]	PABO	BE/PE, PR/SC	6	12.42 ± 0.13	3.63 ± 0.04	-19.65 ± 0.22
P. montagui [d]	PMON	BE/PE, PR/SC	2	12.56 ± 0.55	3.66 ± 0.16	-18.59 ± 0.19
Pasiphaea multidentata [d]	PMUL	BE/PE, PR/SC	2	12.38 ± 0.05	3.61 ± 0.01	-20.24 ± 0.16
Chionoecetes oplio	SNCR	BE, PR/SC	12	13.15 ± 0.24	3.83 ± 0.07	-19.93 ± 0.12
Mollusca						
Gastropoda						
Littorina littorea [d]	BIGO	LI, GR/DF	10	7.88 ± 0.09	2.29 ± 0.03	-17.26 ± 0.11
Buccinum undatum [d]	BUCC	LI/BE, PR/SC	10	9.70 ± 0.11	2.83 ± 0.03	-17.83 ± 0.12
Tectura testudinalis [d]	PATE	LI, GR	10	6.89 ± 0.04	2.00 ± 0.01	-18.60 ± 0.16
Bivalvia						
Mytilus edulis [d]	BLMU	LI/BE, FF	21	7.18 ± 0.12	2.09 ± 0.03	-19.53 ± 0.17
Echinodermata						
Strongylocentrotus droebachiensis	URCH	BE, DF/OM	10	5.85 ± 0.05	1.69 ± 0.01	-17.70 ± 0.24
Vertebrata						
Osteichtyes						
Mallotus villosus	CAPE	PE, PR	12	13.21 ± 0.14	3.86 ± 0.04	-20.87 ± 0.10
Ammodytes americanus	SAND	BE/PE, PR	13	11.58 ± 0.09	3.38 ± 0.03	-20.51 ± 0.17
Hippoglossoides platessoides	AMPL	BE, PR	10	14.43 ± 0.07	4.22 ± 0.02	-19.18 ± 0.22
Glyptocephalus cynoglossus	WIFL	BE, PR	10	14.45 ± 0.17	4.22 ± 0.05	-17.78 ± 0.13
Clupea harengus	ATHE	PE, PR	11	12.24 ± 0.19	3.57 ± 0.06	-21.34 ± 0.11
Aves						
Rissa tridactyla	BLKI	SF, PR/SC	21	15.15 ± 0.03	4.67 ± 0.01	-18.89 ± 0.02
Alca torda	RAZO	PE, PR	20	15.21 ± 0.05	4.68 ± 0.02	-19.26 ± 0.03
Somateria mollissima	COEI	BE, PR	20	11.10 ± 0.10	3.47 ± 0.03	-18.92 ± 0.21
Larus argentatus	HERG	SF/LI, PR/SC	20	12.98 ± 0.27	4.03 ± 0.08	-18.92 ± 0.14
Larus marinus	GBBG	SF/LI, PR/SC	20	12.82 ± 0.42	3.98 ± 0.12	-19.04 ± 0.17

[a] Feeding habitats and feeding modes: Feeding habitat: PL=plankton; BE=benthic; LI=littoral; PE=pelagic; SF=surface. Feeding mode: FF=filter-feeder; PR=predator; SC=scavenger; OM=omnivore; DF=detritus-feeder; GR=grazer.

[b] Trophic levels were calculated using *Tectura testudinalis* as the baseline and a trophic enrichment value of 3.4‰ for most organisms (Minagawa and Wada, 1984; Post, 2002) and 2.6‰ for birds (Bearhop *et al.*, 2002).

[c] From Lesage, Fisheries and Oceans Canada, pers. comm.

[d] Several organisms were pooled (2-300) and the associated sample sizes (*n*) represent the number of pools composed of several individuals

Table 2.3 Mean (± SE) THg, MeHg (ng/g dw), percentage of MeHg (%) and moisture content values (%) for all species collected in a Gulf of St. Lawrence food web. Latin names are provided in table 2.2.

Taxa Code	*n*	*n* [a]	THg (ng/g dw)	MeHg (ng/g dw)	MeHg (%)	H_2O (%)
Particulate organic matter [b]						
POM	4		--	--	--	--
Invertebrata						
Crustacea						
Zooplankton						
CFIN [c]	10	5	12.6 ± 0.63	2.55 ± 0.55	21.4 ± 5.5	78.2 ± 0.8
CHYP [c]	5	5	64.7 ± 7.47	3.89 ± 1.08	6.4 ± 2.0	82.5 ± 0.9
MENO [c]	6	5	60.2 ± 4.51	6.49 ± 2.62	10.0 ± 3.6	80.6 ± 1.2
Amphipoda						
GAMA [c]	10	7	39.2 ± 3.06	14.7 ± 4.53	33.0 ± 10.3	75.9 ± 0.8
Decapoda						
SHRI	10	5	138 ± 11.2	75.2 ± 12.9	51.0 ± 7.8	71.8 ± 0.5
PABO [c]	6	5	139 ± 14.7	75.2 ± 12.9	51.0 ± 7.8	71.3 ± 0.7
PMON [c]	2	0	167 ± 14.6	--	--	72.4 ± 0.4
PMUL [c]	2	0	108 ± 22.9	--	--	72.5 ± 1.2
SNCR	12	5	231 ± 18.7	103 ± 38.0	45.2 ± 14.7	84.6 ± 1.4
Mollusca						
Gastropoda						
BIGO [c]	10	5	50.5 ± 1.75	12.8 ± 1.34	25.4 ± 2.1	81.3 ± 0.5
BUCC [c]	10	5	127 ± 8.06	84.7 ± 19.1	56.9 ± 11.6	80.0 ± 0.5
PATE [c]	10	5	50.8 ± 1.34	9.32 ± 0.42	19.1 ± 1.1	82.2 ± 0.3
Bivalvia						
BLMU [c]	21	13	104 ± 5.95	24.3 ± 2.54	24.8 ± 2.9	88.7 ± 0.5
Echinodermata						
URCH	10	5	42.0 ± 3.59	5.31 ± 1.04	12.5 ± 2.0	88.0 ± 0.6
Vertebrata						
Osteichtyes						
CAPE	12	8	28.1 ± 2.18	17.5 ± 3.00	57.8 ± 7.6	72.1 ± 0.4
SAND	13	8	71.2 ± 16.2	54.7 ± 20.1	64.8 ± 2.6	69.5 ± 1.3
AMPL	10	8	146 ± 38.2	77.4 ± 18.8	56.1 ± 5.5	77.0 ± 0.8
WIFL	10	7	179 ± 21.9	100 ± 27.1	46.9 ± 7.1	78.5 ± 0.8
ATHE	11	8	104 ± 11.0	69.2 ± 6.11	63.7 ± 4.5	65.0 ± 1.0
Aves						
BLKI	21	7	805 ± 52.3	688 ± 80.5	95.0 ± 1.2	66.5 ± 0.5
RAZO	20	7	1789 ± 98.0	1777 ± 137	99.8 ± 0.1	69.9 ± 0.8
COEI	20	7	640 ± 36.1	565 ± 53.3	97.5 ± 0.8	68.9 ± 0.6
HERG	20	7	724 ± 69.3	624 ± 135	99.4 ± 0.2	66.5 ± 0.4
GBBG	20	7	1540 ± 212	1611 ± 427	99.8 ± 0.1	67.4 ± 0.6

[a] Sample size (*n*) used for MeHg analysis

[b] From Lesage, Fisheries and Oceans Canada, pers. comm.

[c] Several organisms were pooled (2-300) and the associated sample sizes (*n*) represent the number of pools composed of several individuals

Table 2.4. Biomagnification power of THg and MeHg, trophic level (TL) of the top predator and water Hg concentrations in various ecosystems throughout the world. Biomagnification power was calculated using the slope (*b*) of a simple linear regression (unless otherwise stated) of mercury concentration in relation to stable nitrogen isotope ($\delta^{15}N$) values. The intercepts of the models (*a*) are also shown.

Compound / Area	Moist	Slope (*b*)	Intercept (*a*)	Infered TL [h]	TL [i]	Hg in water (ng/L) THg	Hg in water (ng/L) MeHg	Author
Total mercury (THg)								
Gulf of Oman, Asia	ww	0.07	-2.62	3.93				Al-Reasi et al., 2007
	ww	0.13 [e]	-3.57					
Davis Strait, West Greenland	dw	0.079 [d]		4.29	4.01			Riget et al., 2007
	ww [a]	0.096 [d]						
	dw	0.11 [c]						
Mekong Delta, South Viet Nam	dw	0.101	-2.496	3.00	3			Ikemoto et al., 2008
	ww [a]	0.114	-3.303					
Gulf of Mexico, USA	ww	0.163	-2.40					Cai et al., 2007
Gulf of St Lawrence, Canada	dw [b]	0.134	0.739	4.75	4.68	0.90	0.12	This study
	ww	0.170	-0.293					
Lake Victoria, Africa	ww [a]	0.163	0.479	2.82		1.7-5.8	0.2-1	Campbell et al., 2003
	ww [a]	0.163 [g1]	-0.033					
	ww [a]	0.165 [g2]	-0.276	3.70		2.9-4.5		
Stewart Lake, Canada	ww	0.192	-2.615	3.24				Power et al., 2002
North Water Polynya, Canada	ww	0.197	-3.407	4.84	4.9			Campbell et al., 2005
Lancaster Sound region, Canada	dw	0.20 [c]	-3.3	4.17				Atwell et al., 1998
Fly estuary, Papua New Guinea	ww	0.205	-0.709	5.53				Yoshinaga et al., 1992
Lake Chad, Africa	ww	0.21	-0.37	3.35				Kidd et al., 2004
Lake Malawi, Africa	ww [a]	0.20	-0.15	3.62				Kidd et al., 2003
	ww [a]	0.23 [f1]	-0.30					
	ww [a]	0.25 [f2]	-0.90					
Lake Victoria, Africa	ww [a]	0.280	-0.808	3.68				Campbell et al., 2004
5 temperate lakes, Canada	ww	0.17 - 0.29	-2.82 - -2.17	2.97 - 3.38				Kidd et al., 1995
Gulf of Farallones, U.S.A.	dw	0.32 [d]		3.53				Jarman et al., 1996
Methyl mercury (MeHg)								
Gulf of Oman, Asia	ww	0.14 [e]	-3.90	3.93				Al-Reasi et al., 2007
Davis Strait, West Greenland	dw	0.147 [d]		4.29	4.01			Riget et al., 2007
	ww [a]	0.181 [d]						
Gulf of St Lawrence, Canada	dw [b]	0.201	-0.507	4.75	4.68	0.90	0.12	This study
	ww	0.235	-1.526					
North Water Polynya, Canada	ww [a]	0.223	-3.882					Campbell et al., 2005
Lake Murray, Papua New Guinea	ww	0.28	-2.88	4.00		1.42	0.067	Bowles et al., 2001

[a] Hg analysis done on dried tissue and converted into a wet weight basis using moisture content data from the literature or from the data generated during the experiment

[b] Hg analysis done on wet tissue and converted into a dry weight basis using moisture content data generated during the experiment

[c] Slope of a reduced major axis: $\log_{10}[Hg]=b(\delta^{15}N)+a$

[d] Slope of a simple linear regression: $\ln[Hg]=b(\delta^{15}N)+a$ calculated into: $\log_{10}[Hg]=b(\delta^{15}N)+a$

[e] Zooplankton were excluded from their model

[f] Only pelagic (1) and benthic (2) were used for our statistical analysis

[g] Only Napoleon Gulf adjusted to Winam Gulf (1) and Winam Gulf (2) were used our statistical analysis

[h] Infered TL were calculated using $(\delta^{15}N_{top\ predator}-\delta^{15}N_{baseline})/3.4$. This represents an estimation of the top predator in the study. Only animals believed to represent a primary or a secondary consumer were chosen for the baseline.

[i] Trophic level of the top predator (average) reported in the studies

Table 2.5. Biomagnification factors for simple (BMF_{TLC}) and complex (BMF_{SIA}) predator-prey interactions and food web magnification factors (FWMFs) for THg and MeHg in a Gulf of St. Lawrence food web. Hg concentrations on a wet weight basis were used for the calculations in order to facilitate comparisons with other studies.

Predator	Prey	diff TL [a]	THg [b]		MeHg [b]		Proportions [b]
			BMF_{TLC}	BMF_{SIA}	BMF_{TLC}	BMF_{SIA}	
BUCC	GAMA	0.46	2.24	2.20	3.89	7.24	
	BLMU	0.74	1.59		4.21		0.380
	URCH	1.14	3.11		16.0		0.620
SHRI	MENO	0.95	2.47	2.71	11.9	13.8	0.688
	CHYP	1.30	2.18		20.7		0.313 [d]
	CFIN	1.21	9.59		25.3		
	GAMA	1.26	2.68		3.95		--
SNCR	Zooplankton [c]	1.35	2.56	1.47	11.4	1.48	0.368 [d]
	GAMA	1.46	2.22		2.64		0.369
	SHRI	0.20	0.83		0.67		0.264
CAPE	MENO	1.18	0.46	0.54	2.55	3.31	0.492
	CHYP	1.53	0.41		4.44		0.508 [d]
	CFIN	1.44	1.80		5.43		
SAND	MENO	0.70	1.42	1.84	9.56	6.70	--
	CHYP	1.05	1.25		16.6		0.635 [d]
	CFIN	0.96	5.51		20.3		
	GAMA	1.01	1.54		3.18		0.365
AMPL	SAND	0.84	1.21	1.21	0.85	0.97	0.498 [e]
	CAPE	0.36	3.70		3.20		
	BUCC	1.39	0.83		0.70		0.502
WIFL	SAND	0.84	1.37	1.37	1.03	1.16	0.492 [e]
	CAPE	0.36	4.21		3.85		
	BUCC	1.39	0.94		0.84		0.508
ATHE	MENO	0.89	2.33	3.00	13.8	20.9	0.270
	CHYP	1.24	2.06		24.0		0.730 [d]
	CFIN	1.15	9.06		29.4		
BLKI	SAND	1.29	9.37	12.4	9.92	15.4	0.509
	CAPE	0.81	28.8		37.2		0.331
	SHRI	1.04	5.39		7.97		0.160
RAZO	SAND	1.30	18.6	21.6	25.2	29.8	0.810
	CAPE	0.82	56.9		94.4		0.190
COEI	BLMU	1.38	10.1	9.00	37.6	23.5	0.727 [f]
	URCH	1.78	19.7		143		
	SNCR	-0.36	6.40		13.2		0.273
HERG	FISH	0.41	15.3	9.02	18.1	16.4	0.201 [e]
	DECA	0.30	6.17		10.7		0.268 [g]
	MENO	1.35	13.9		105		0.244
	BLMU	1.94	10.7		38.0		0.163
	BUCC	1.20	6.73		9.02		0.123
GBBG	FISH	0.36	31.6	18.6	43.9	39.7	0.201 [e]
	DECA	0.25	12.8		25.9		0.268 [g]
	MENO	1.30	28.7		255		0.244
	BLMU	1.89	22.1		91.9		0.163
	BUCC	1.15	13.9		21.8		0.123
FWMF			3.81		6.46		

[a] Trophic levels (TL) were calculated using $\delta^{15}N$ after the equations 2.4 or 2.5 depending on the taxa.

[b] BMF_{TLC} was calculated for simple predator-prey interactions and corrected for TL using the equation 2.9 after Fisk *et al.* (2001) and BMF_{SIA} was calculated for more complex predator-prey interactions (2 or more preys) after the equation 2.10 using SIA in mixing models (Phillips & Gregg, 2001, Phillips and Koch, 2002, Phillips & Gregg, 2003).

[c] Include MENO, CHYP and CFIN; [d]CHYP and CFIN; [e]CAPE and SAND; [f]BLMU and URCH; [g]SHRI and SNCR

Table 2.6. Log_{10} Bioaccumulation factors (BAFs) for THg and MeHg for all species collected in a Gulf of St. Lawrence food web (mean ± SE). Mean water concentrations used for the calculations were 9.03×10^{-4} and 1.16×10^{-4} ng/g for THg and MeHg, respectively.

Code	Log_{10} BAF THg	Log_{10} BAF MeHg
CFIN	3.47 ± 0.02	3.63 ± 0.10
CHYP	4.09 ± 0.06	3.66 ± 0.15
MENO	4.10 ± 0.04	3.80 ± 0.31
GAMA	4.01 ± 0.04	4.19 ± 0.27
BIGO	4.02 ± 0.01	4.32 ± 0.04
BUCC	4.44 ± 0.03	5.08 ± 0.14
PATE	4.00 ± 0.01	4.15 ± 0.02
BLMU	4.09 ± 0.03	4.35 ± 0.06
URCH	3.73 ± 0.02	3.70 ± 0.08
SHRI	4.62 ± 0.04	5.23 ± 0.09
SNCR	4.55 ± 0.04	4.94 ± 0.23
CAPE	3.92 ± 0.03	4.53 ± 0.14
SAND	4.29 ± 0.07	5.02 ± 0.11
AMPL	4.47 ± 0.08	5.12 ± 0.07
WIFL	4.60 ± 0.04	5.16 ± 0.11
ATHE	4.58 ± 0.05	5.31 ± 0.04
BLKI	5.45 ± 0.03	6.25 ± 0.05
RAZO	5.76 ± 0.02	6.67 ± 0.03
COEI	5.33 ± 0.02	6.17 ± 0.05
HERG	5.39 ± 0.04	6.21 ± 0.08
GBBG	5.67 ± 0.06	6.54 ± 0.12

Table 2.7. Multiple linear regression models for THg, MeHg, IHg (ng/g dw), percentage of MeHg and IHg (%) as dependant variables and total weight (g), trophic level (TL), lipid content (%), and benthic connection ($\delta^{13}C$) as independent variables. The variables were standardised (0 ± 1; mean ± SD). The variables in percentage were arcsine transformed whereas the other variables were log transformed. Standardized partial coefficients within a model were not significantly different ($t_{\alpha(n)}$; $p > 0.05$).

Terms	Standardized partial coefficients (b')	SE	p	$F_{(0.05)k,n}$ [a]	Adjusted MLR coefficient (R_a^2)	p
[THg]						
Weight	0.405	0.049	<0.001	266	0.745	<0.001
Lipid %	-0.327	0.044	<0.001			
TL	0.253	0.041	<0.001			
[MeHg]						
Weight	0.412	0.061	<0.001	165	0.778	<0.001
TL	0.305	0.054	<0.001			
Lipid %	-0.301	0.053	<0.001			
MeHg %						
Weight	0.489	0.074	<0.001	77.6	0.689	<0.001
Lipid %	-0.323	0.059	<0.001			
$\delta^{13}C$	-0.258	0.074	<0.001			
TL	0.234	0.070	0.001			
[IHg]						
Weight	-0.360	0.103	0.001	10.9	0.171	<0.001
$\delta^{13}C$	0.292	0.088	0.001			
IHg %						
Weight	-0.453	0.075	<0.001	75.3	0.682	<0.001
TL	-0.297	0.071	<0.001			
Lipid %	0.280	0.075	<0.001			
$\delta^{13}C$	0.208	0.060	0.001			

[a] df=3, 268 for [THg]; 3, 135 for [MeHg] and [IHg]; and 4, 134 for % MeHg and % IHg

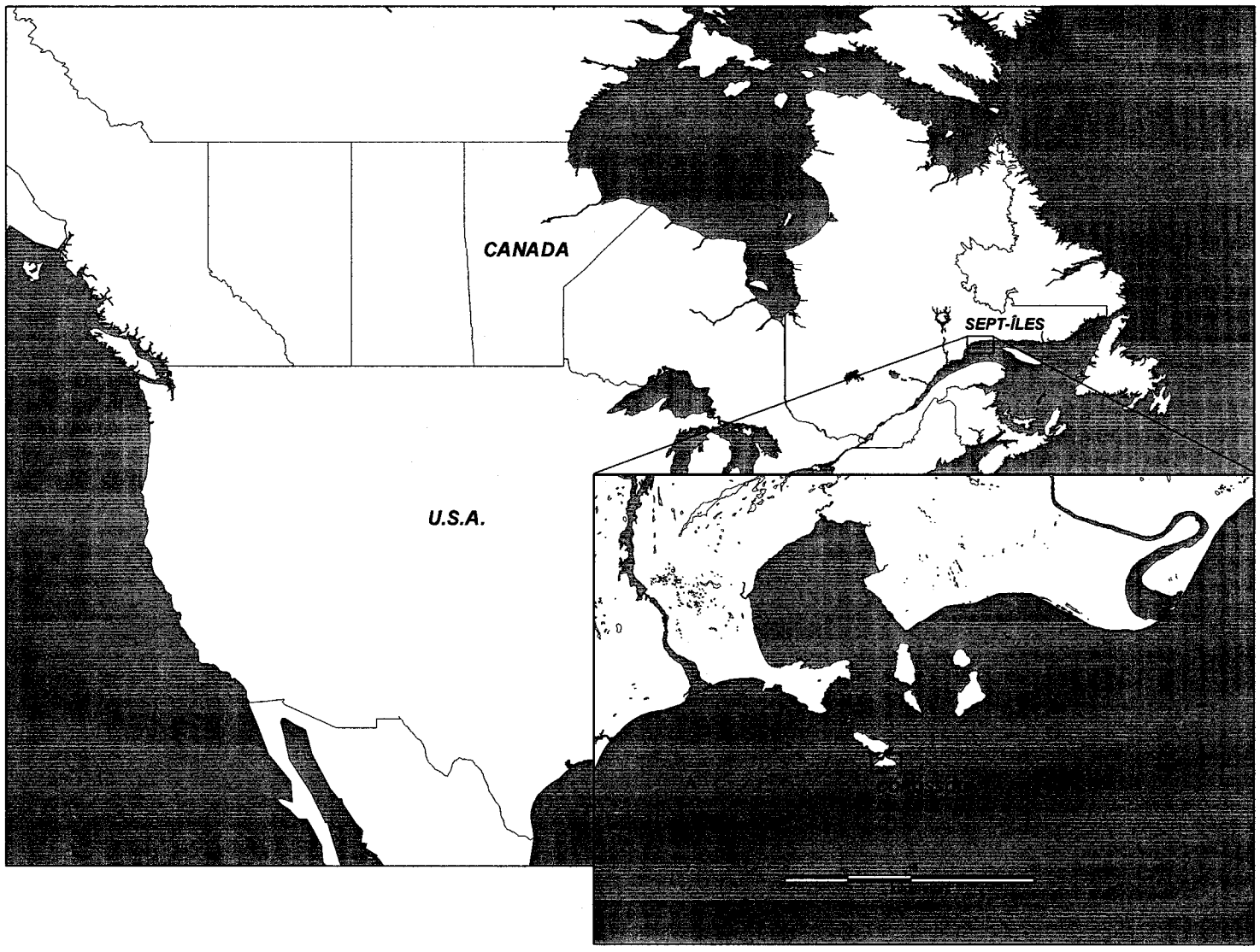

Figure 2.1. Sampling area in the Gulf of St. Lawrence, Québec, Canada.

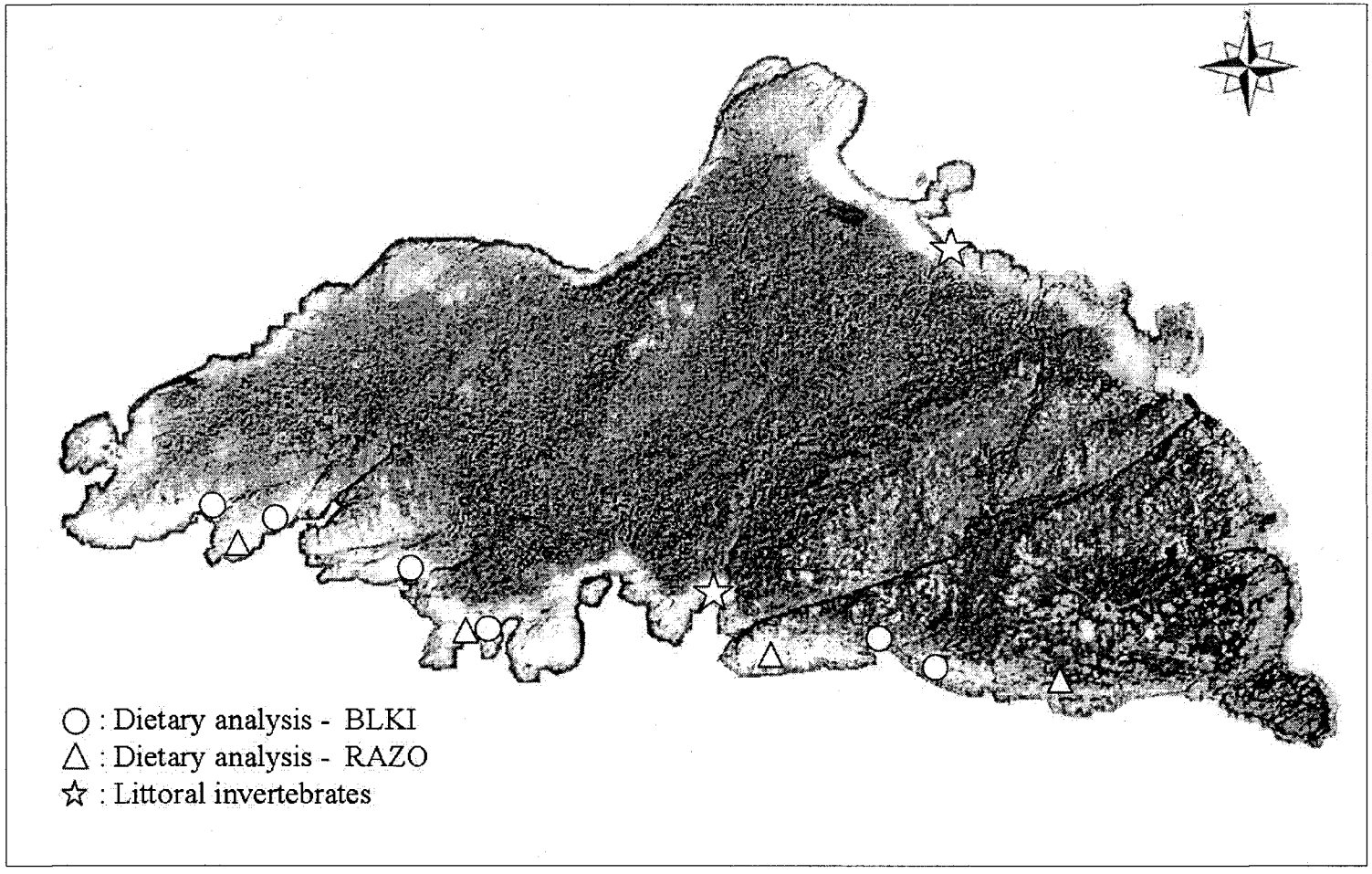

Figure 2.2 Study site and localization of the sampling stations on Corossol Island in 2006-07.

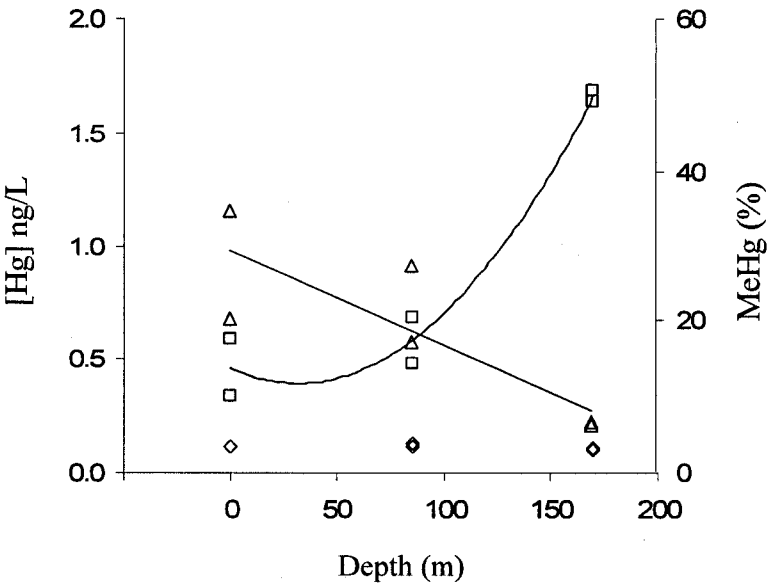

Figure 2.3. THg (squares), MeHg (diamonds) concentrations (ng/L) and percentage of MeHg (triangles; %) in water at 3 depths (0, 85 and 170 m). The significance of the overall models and the slopes are given in table 2.1.

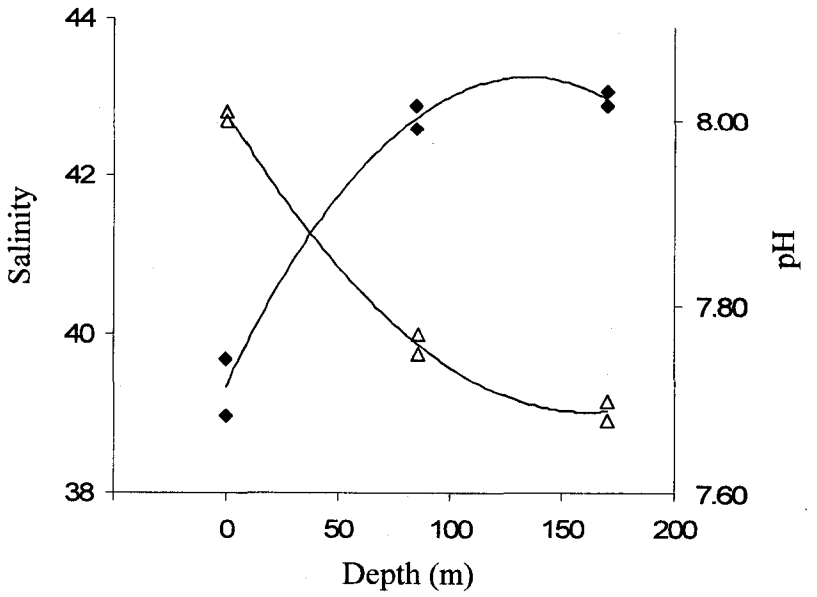

Figure 2.4. Salinity (filled diamonds) and pH (triangles) in water at 3 depths (0, 85 and 170 m). The significance of the overall models and the slopes are given in table 2.1.

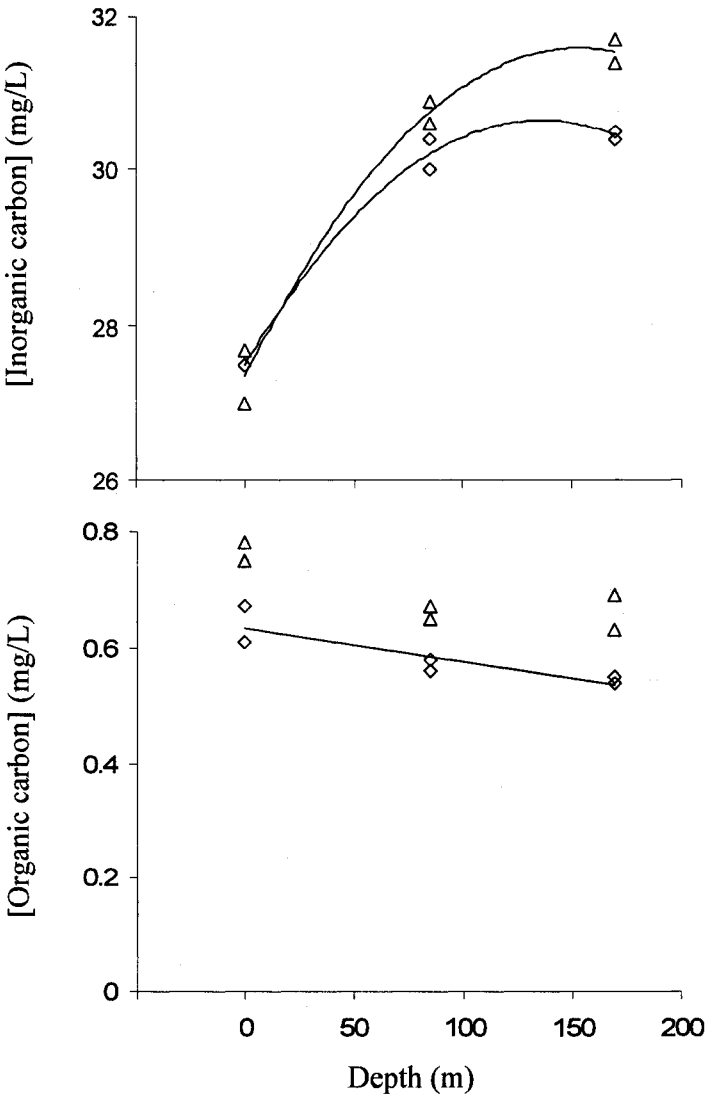

Figure 2.5. Concentrations of carbon (mg/L) in total (diamonds) and dissolved (triangle) inorganic (upper panel) and organic (lower panel) carbon in water at 3 depths (0, 85 and 170 m). The significance of the overall models and the slopes are given in table 2.1.

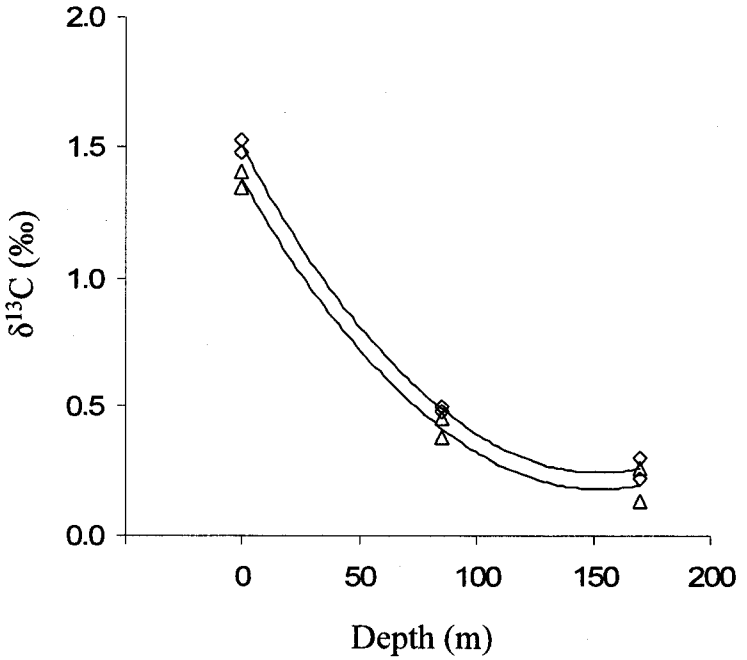

Figure 2.6. Stable carbon isotope $\delta^{13}C$ (‰) in total (diamonds) and dissolved (triangle) inorganic carbon in water at 3 depths (0, 85 and 170 m). The significance of the overall models and the slopes are given in table 2.1.

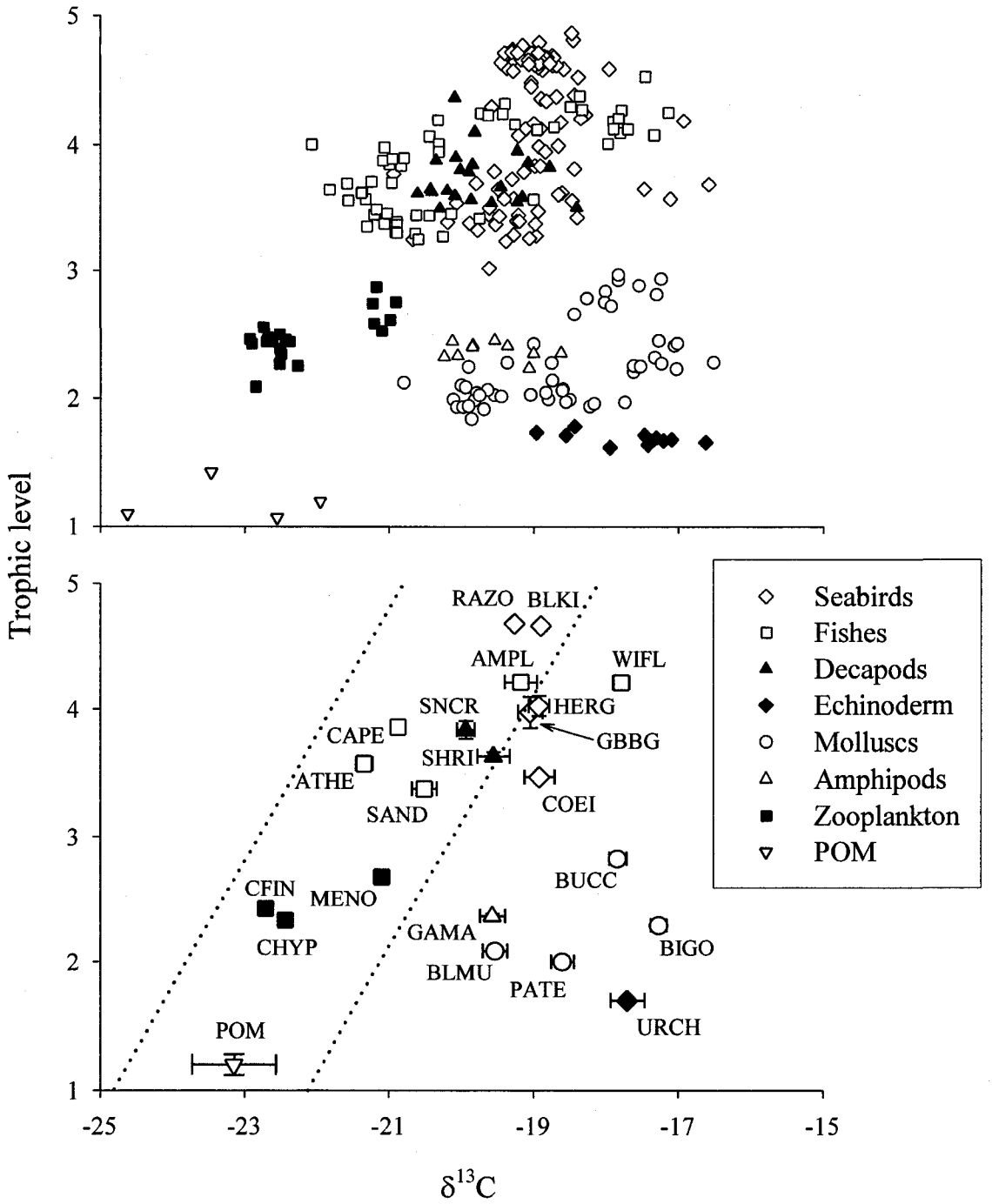

Figure 2.7. Relationship between trophic level and stable carbon isotope ($\delta^{13}C$) values for all species collected in a Gulf of St. Lawrence food web. Upper panel shows all data points for each group and lower panel shows mean (± SE) values for each species. Dashed lines represent the boundaries of a theoretical food web derived from particulate organic matter (POM). POM values were kindly contributed by Lesage (DFO, pers. comm).

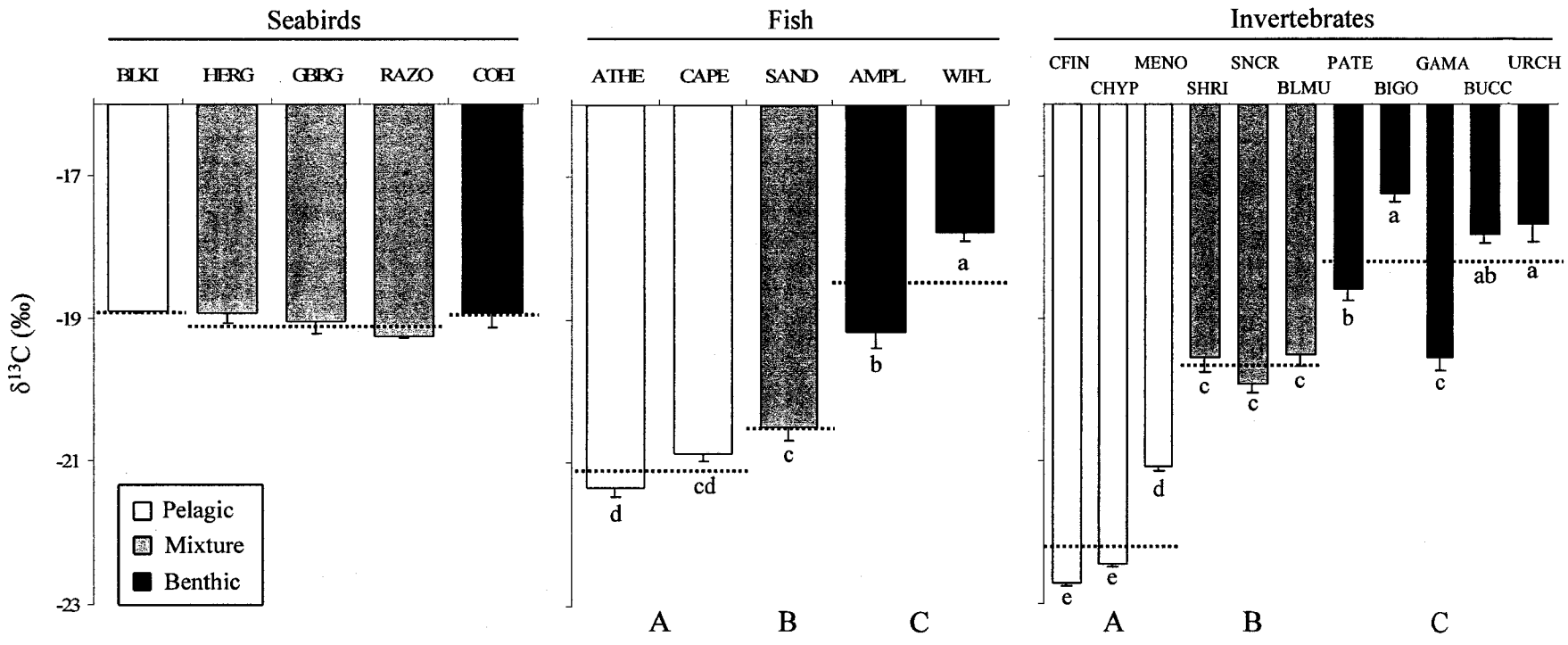

Figure 2.8. Stable carbon isotope ($\delta^{13}C$) values for all species collected in a Gulf of St. Lawrence food web. Organisms were classified considering their feeding habitat as pelagic (white), benthic (black) or intermediate between the two (mixture, shown in gray) and the dashed lines represent the average for these habitats within a taxonomical group. Lines sharing a common capital letter do not vary significantly. Individuals sharing common lower case letters do not vary significantly within a taxonomical group. Statistical analyses were done separately for each taxonomical group.

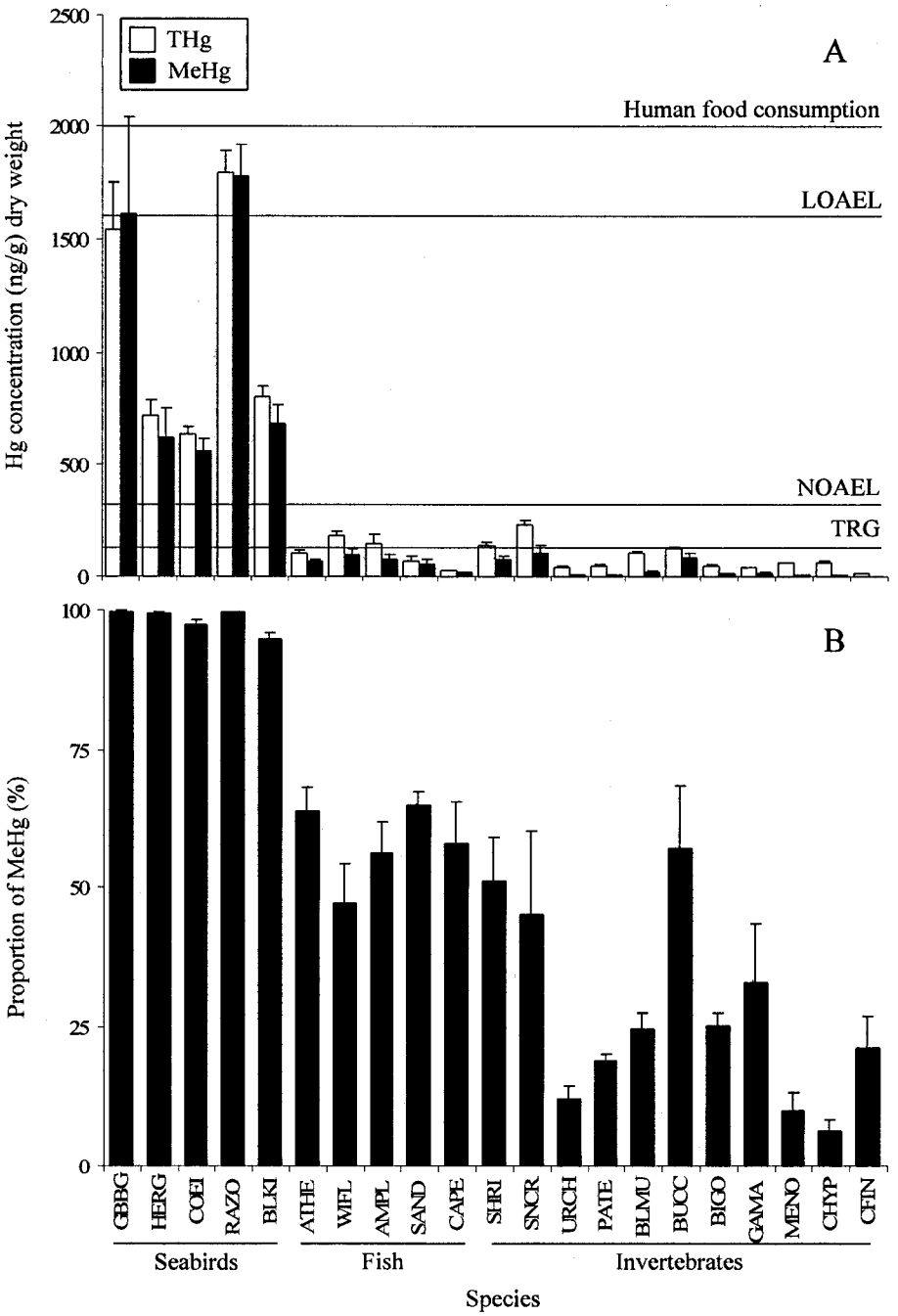

Figure 2.9. a) THg (white bars) and MeHg (black bars) concentrations (ng/g dw; mean ± SE) and b) proportion of MeHg over THg (% ± SE) for all species collected in a Gulf of St. Lawrence food web. Health Canada's guideline for fish consumption (Health Canada 2007), a preliminary lowest observable adverse effect level (LOAEL), a preliminary no observable adverse effect level (NOAEL, Kenow et al. 2003, Meyer 2006) as well as a tissue residue guideline (TRG) for the protection of fish-eating birds (CCME 2000) are presented. The guidelines are usually reported on a wet weight basis, but it was converted into a dry weight basis assuming a moisture content of 75% (mean for this study = 74.7%).

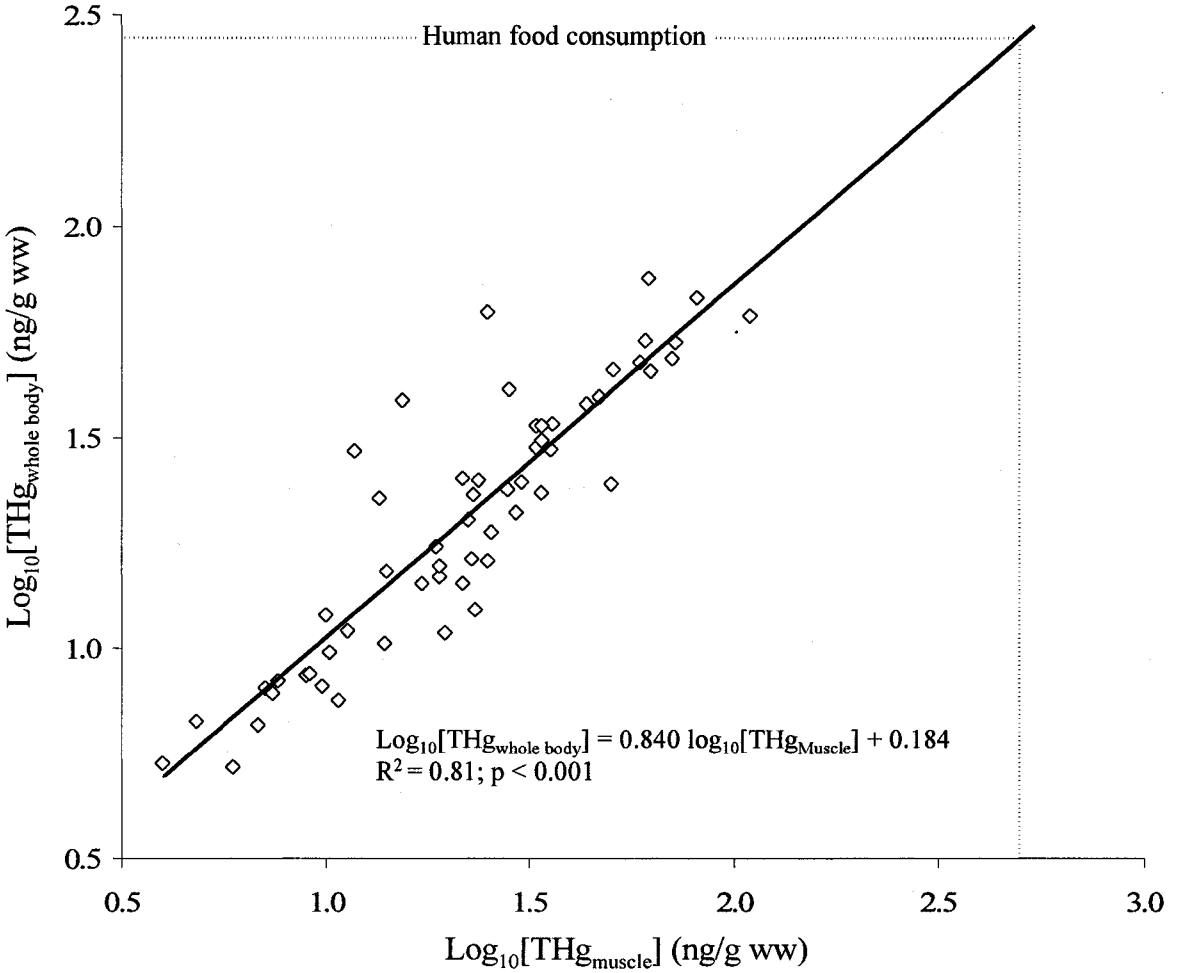

Figure 2.10. Log_{10} transformed THg concentration in whole organisms against dorsal muscle in five species of fish (n = 57) in a Gulf of St. Lawrence food web. The equation of the model is shown on the graph. The dashed line represents Health Canada's guideline for human food consumption for fish muscle and its conversion into whole body according to the model.

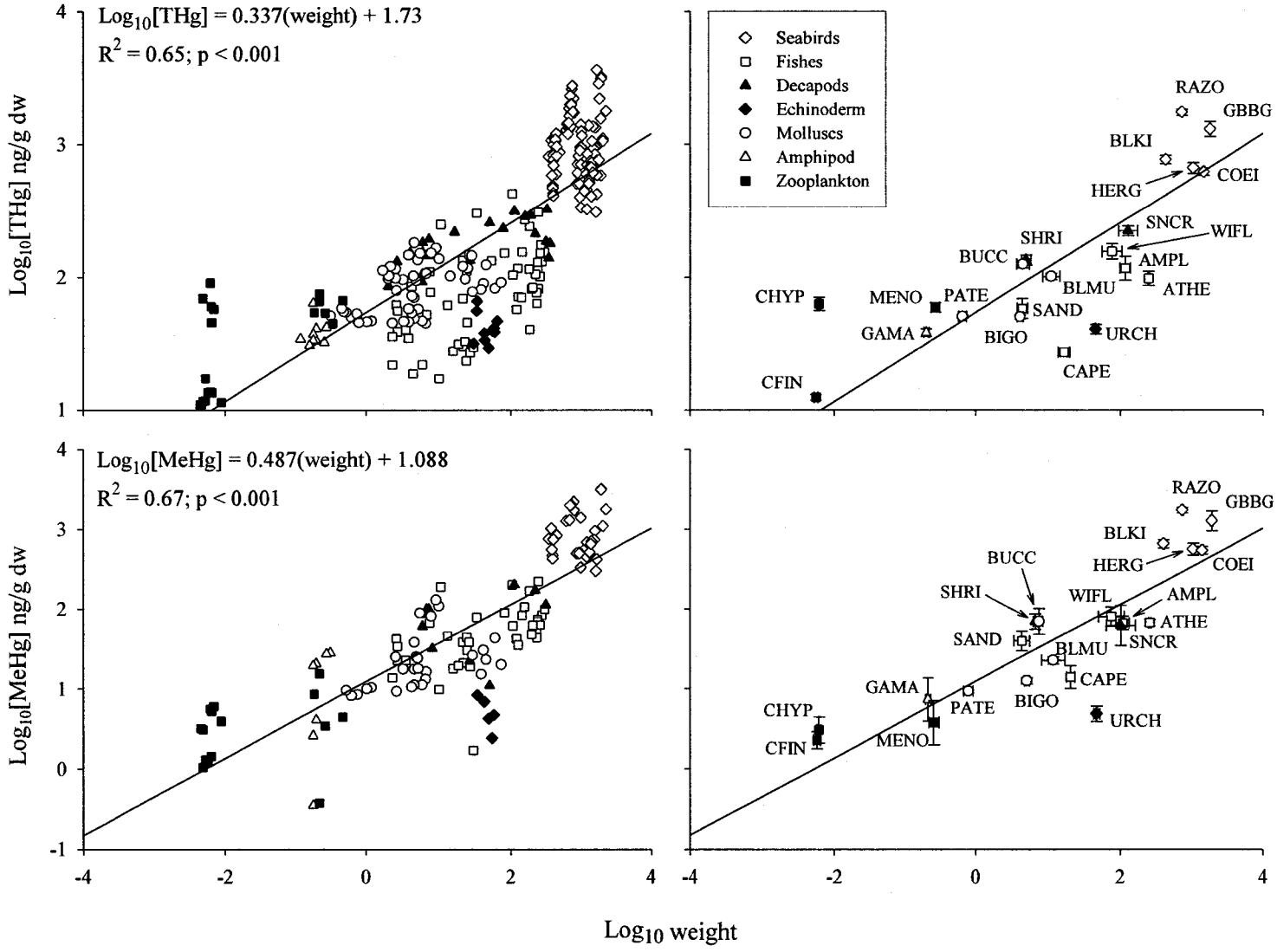

Figure 2.11. THg (above) and MeHg (below) concentrations (ng/g dw) as a function of total weight (g) for all species collected in a Gulf of St. Lawrence food web. Logarithmic transformations were applied.

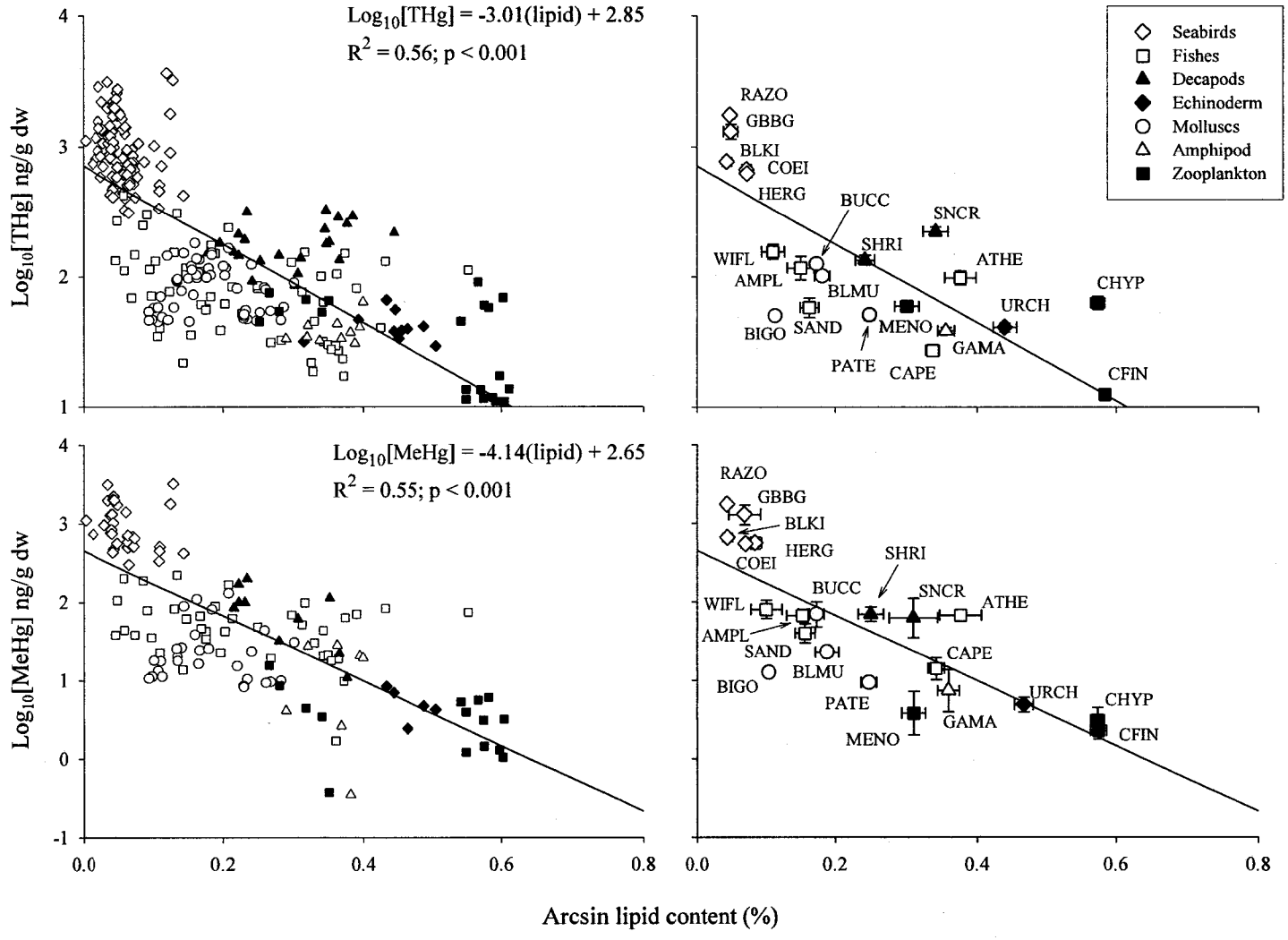

Figure 2.12. THg (above) and MeHg (below) concentrations (ng/g dw) in relation to lipid content (%) for all species collected in a Gulf of St. Lawrence food web. Logarithmic transformations were applied to THg and MeHg whereas arcsine transformations were applied to lipid content.

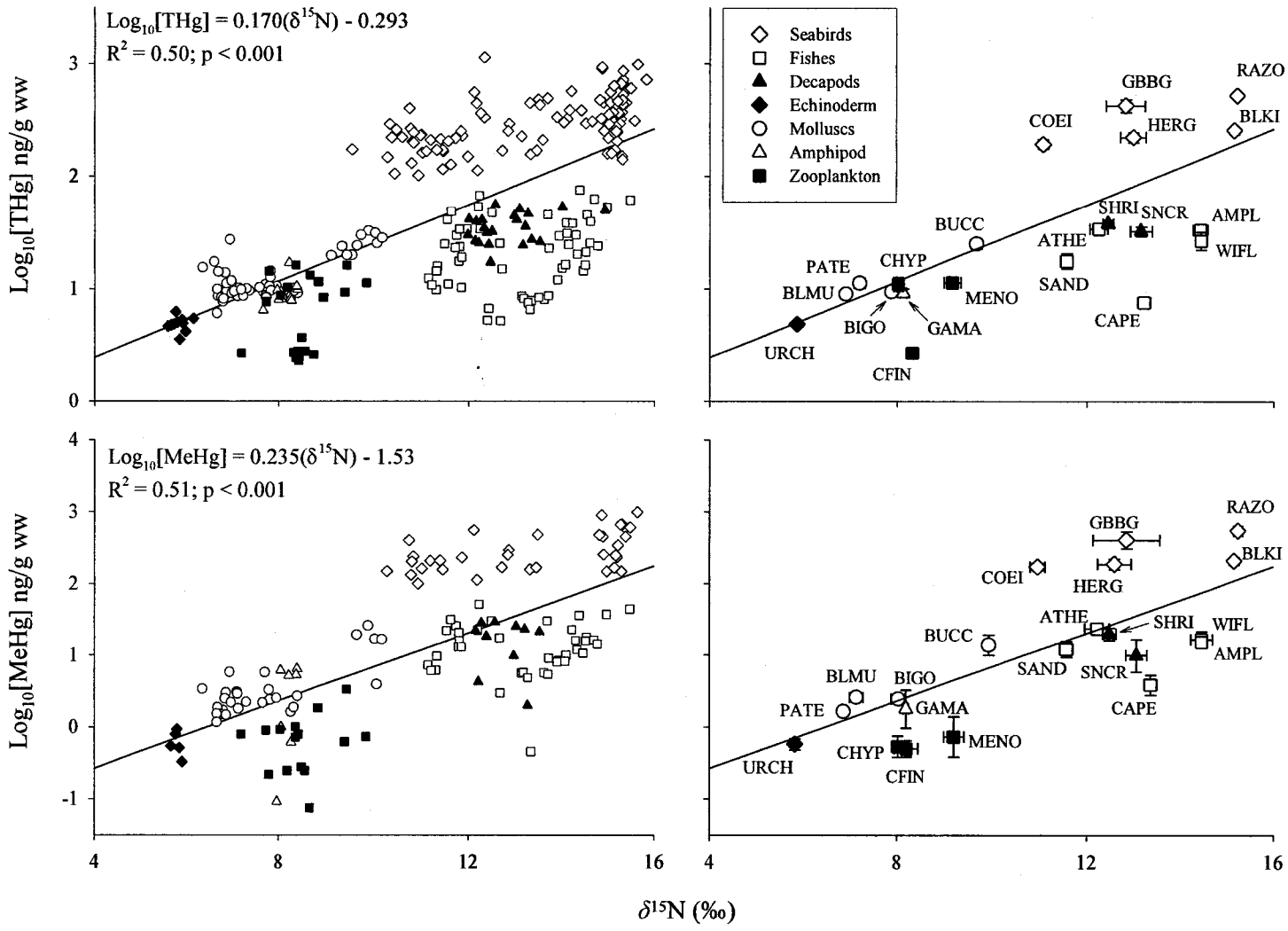

Figure 2.13. THg (above) and MeHg (below) concentrations (ng/g ww) in relation to stable nitrogen isotope ($\delta^{15}N$) for all species collected in a Gulf of St. Lawrence food web. Logarithmic transformations were applied to dependant variables. Hg concentrations are presented on wet weight basis on this graph in order to compare biomagnification power (*b*) with other studies.

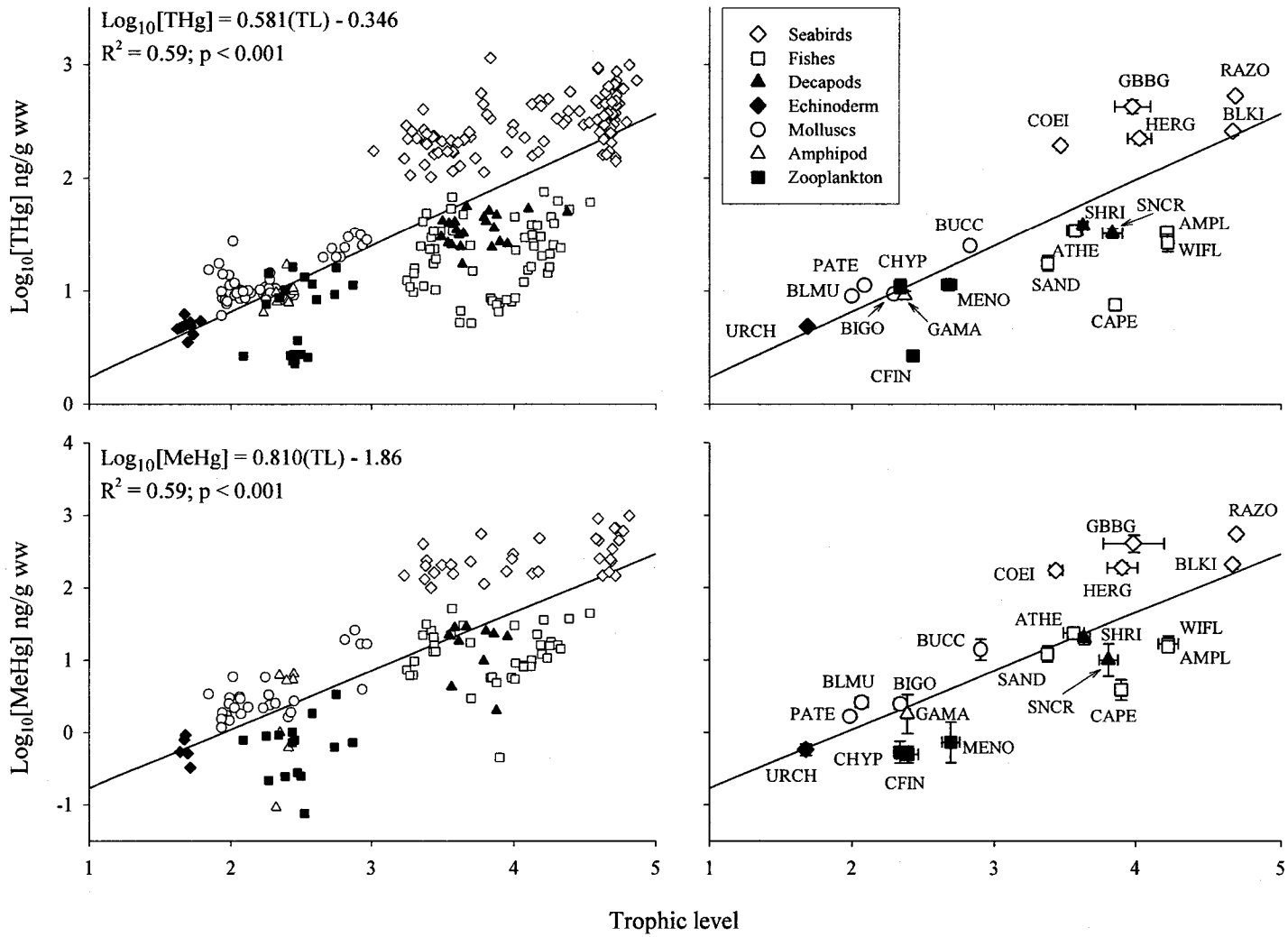

Figure 2.14. THg (above) and MeHg (below) concentrations (ng/g ww) in relation to trophic level for all species collected in a Gulf of St. Lawrence food web. Logarithmic transformations were applied to dependant variables. Mercury concentrations are presented on a wet weight basis in order to compare the food web magnification factor (FWMF) with other studies. Trophic levels were assessed using stable nitrogen isotope (δ^{15}N).

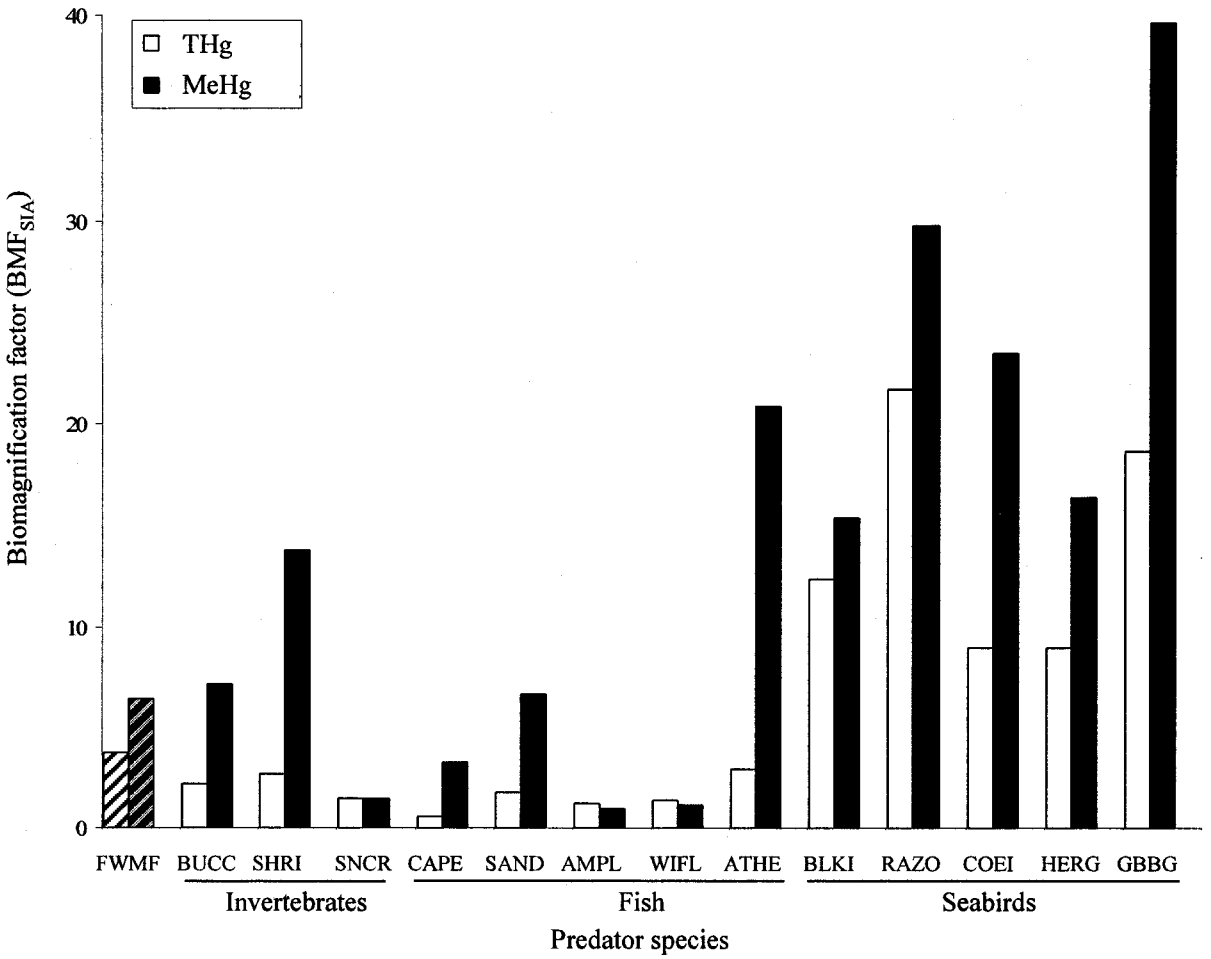

Figure 2.15. Biomagnification factors (BMF_{SIA}) and food web magnification factors (FWMFs) of THg (white bars) and MeHg (black bars) in a Gulf of St. Lawrence food web. BMF_{SIA} were calculated using complex predator-prey interactions (two or more prey items) and corrected for trophic level. The proportions of different prey items in the diet of the predators were based on mixing models using SIA (Phillips and Gregg 2001, Phillips and Koch 2002, Phillips and Gregg 2003). The prey items for each predator and the related proportions used for the BMF calculations are presented in table 2.5.

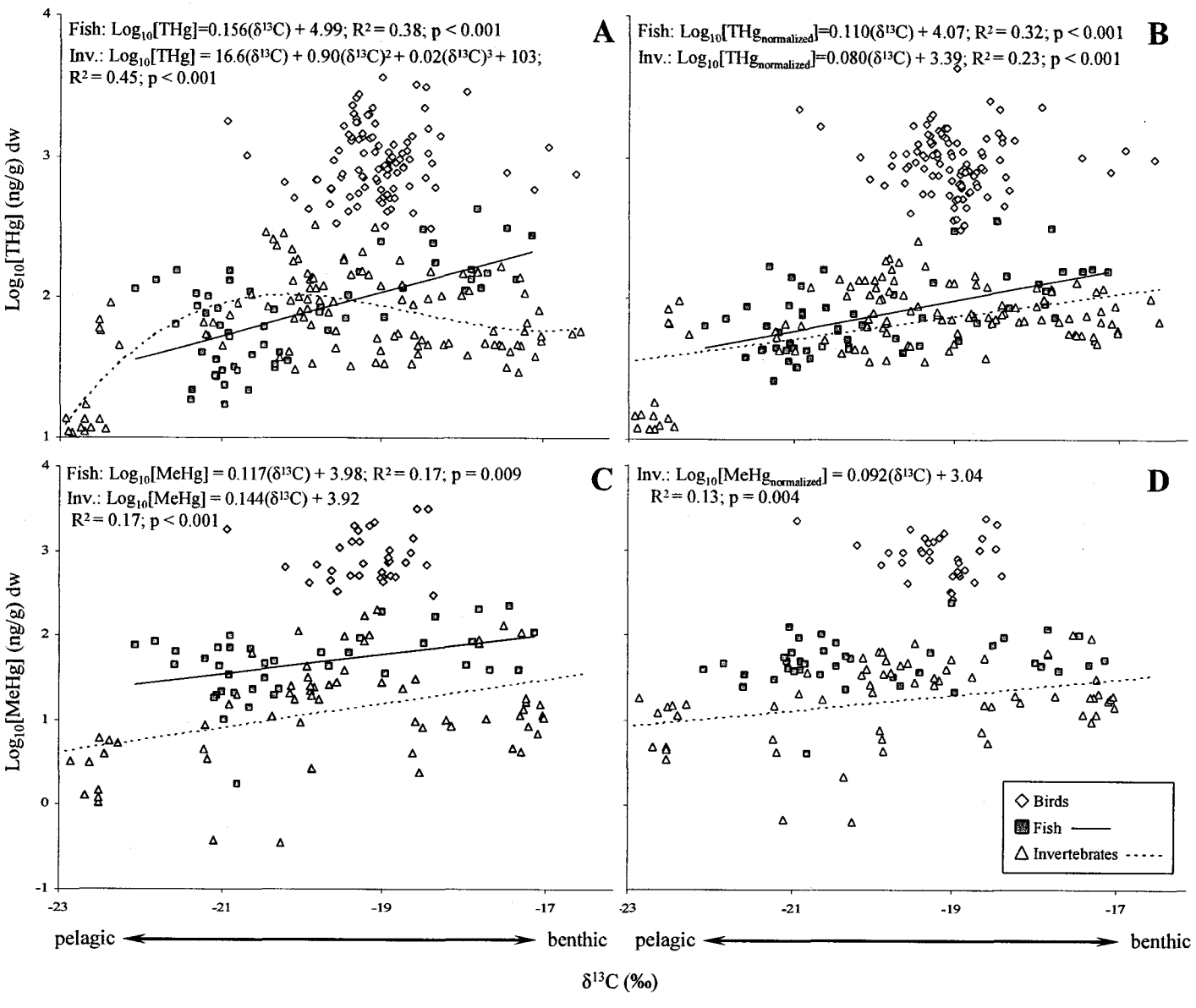

Figure 2.16. THg and MeHg concentrations (ng/g dw) in relation to stable carbon isotope ($\delta^{13}C$) values for all species collected in a Gulf of St. Lawrence food web. Raw (A, C) and normalized (B, D) data are presented for THg (top panels) and MeHg (bottom panels). Birds (diamonds) and invertebrates (triangles) were normalized for the nitrogen isotope ($\delta^{15}N$) values and fish (gray squares) were normalized for length. Equations of the models are given when a significant relationship was found within a group. An arrow is presented to show the increase of $\delta^{13}C$ with the level of connectivity of organisms to the benthos.

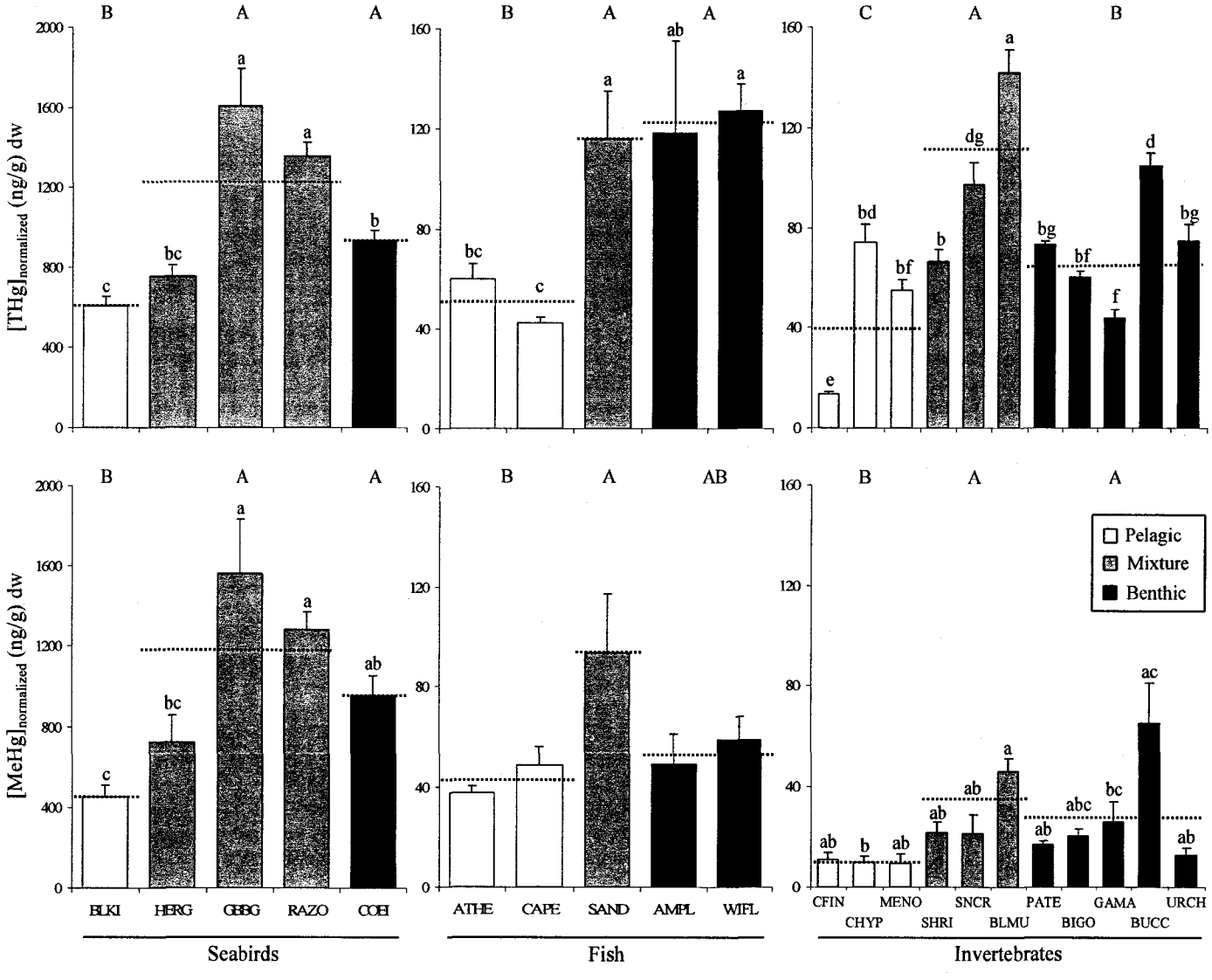

Figure 2.17. Mean (± SE) normalized THg (top panels) and MeHg (bottom panels) concentrations. Hg concentrations for birds and invertebrates were normalized for nitrogen isotope ($\delta^{15}N$) value and fish were normalized for length. Statistical analysis was done separately on taxonomical groups. Organisms were classified considering their feeding habitat: pelagic (white), benthic (black) or a mixture between the two (gray). The dashed lines represent the average for the habitats. Lines sharing common capital letters do not vary significantly. Individuals that share lower case letters do not vary significantly within a group.

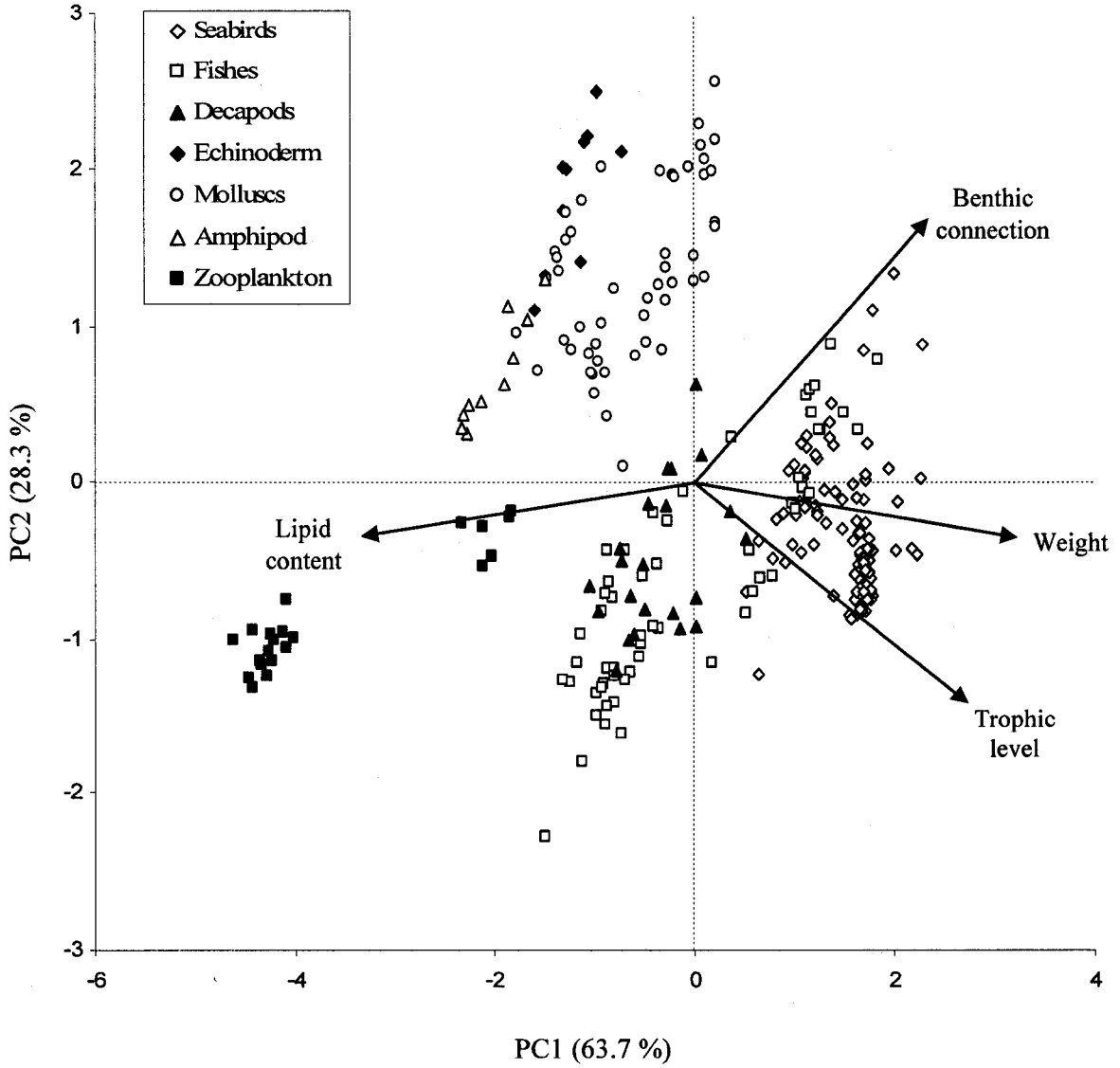

Figure 2.18. Principal components analysis (PCA) using weight, lipid content, trophic level and benthic connection ($\delta^{13}C$) for all species collected in a Gulf of St. Lawrence food web. Weight was log-transformed and lipid content was arcsine-transformed. Variables were standardized (0 ± 1; mean ± SD). Axes for all variables are shown on the graph.

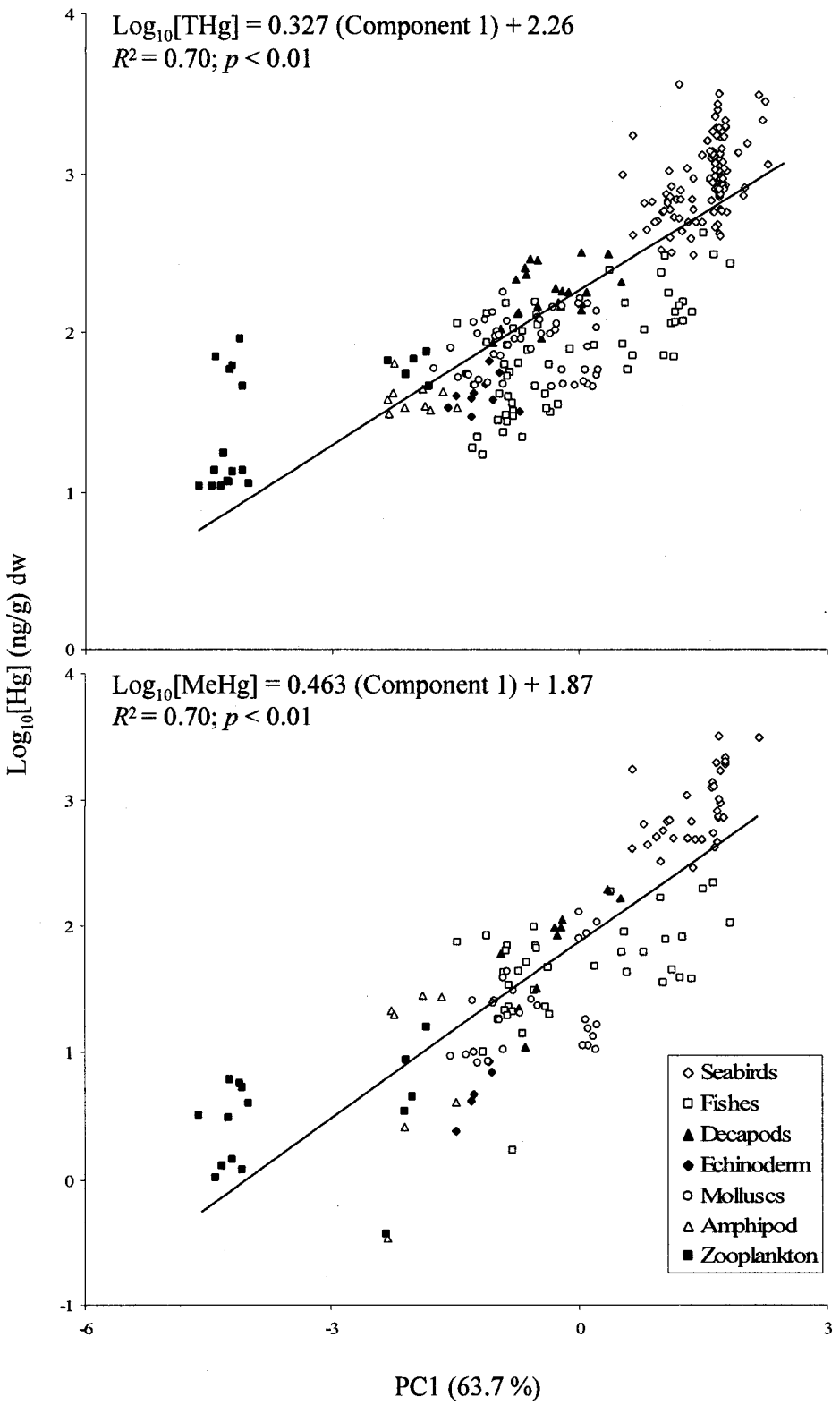

Figure 2.19. THg (above) and MeHg (below) concentrations (ng/g ww) in relation to component 1 calculated by a PCA (see figure 2.18) for all species collected in a Gulf of St. Lawrence food web. Logarithmic transformations were applied to mercury concentrations.

CHAPTER 3

Diet composition of five seabird species in a Gulf of St. Lawrence food web: linking stable isotope analysis (δ^{15}N and δ^{13}C) with traditional dietary techniques

3.1 INTRODUCTION

3.1.1 Problem identification

Seabirds are top predators and serve as useful bioindicators of the environment in which they live and feed. An assessment of their diet is important to establish the species that are consumed and therefore available in the ecosystem. Monitoring fluctuations in abundance of these prey species can help making management decisions and law enforcement in marine ecosystems regarding fisheries, anthropogenic disturbance and pollution. However, traditional techniques to determine the diet of seabirds are often laborious and confined to the chick-rearing period. As such, it may not necessarily reflect the actual diet of the adults. The application of stable isotope analysis in dietary studies can provide an integration over space and time that is complementary to traditional techniques. The aim of this study is to determine the merit of using stable isotope techniques to assess the diet of seabirds in the Gulf of St. Lawrence.

3.1.2 Seabirds

Seabirds have been recognized as good bioindicators and biomonitors of the environment due to their long lifespan and their high level in food chains (Kushlan 1993, Monteiro and Furness 1995, Becker and Chapdelaine 2003). The health of seabirds reflects the health of the environment in which they live and feed (Kushlan 1993). Scarcity of food

supply, high concentrations of pollutants in the environment, anthropogenic disturbance and change in climatic conditions can have adverse effects on their reproductive success and adult survival (Kushlan 1993, Becker and Chapdelaine 2003). Seabirds are sensitive to food availability and abundance (Becker and Chapdelaine 2003) and this has prompted interest in the evaluation of their diet (for review, see Barrett et al. 2007). During periods of food shortage, seabirds can substitute their usual diet with alternative items with lower energy density (Rail and Chapdelaine 2000). Populations of seabirds can be negatively (Frederiksen et al. 2004) or positively impacted (Chapdelaine and Rail 1997) by industrial fisheries.

3.1.3 Diet composition

In ecology, stable isotope analysis (SIA) has emerged as a powerful tool to evaluate predator-prey interactions (Phillips and Gregg 2003, Hammill et al. 2005). Stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes are known to undergo predictable stepwise enrichments between the diet and consumers. $\delta^{15}N$ is higher in a consumer compared to its diet (Deniro and Epstein 1978, 1981) while $\delta^{13}C$ is mainly used to establish the source of carbon (Peterson and Fry, 1987). The carbon signature is defined at the primary producer level and it is then conserved in successive trophic levels. Only a small trophic fractionation has been shown to occur in $\delta^{13}C$ values (Deniro and Epstein 1978, Bearhop et al. 2002). The trophic position of consumers can therefore be identified using $\delta^{15}N$ to calculate the trophic level and using $\delta^{13}C$ as an indicator of the source of organic matter (Peterson and Fry 1987).

Given the predictable trophic enrichments of $\delta^{15}N$ and $\delta^{13}C$, the relative contribution of different food items (sources) for a particular consumer (mixture) can be assessed using mixing models (Phillips and Gregg 2001, Vander Zanden and Rasmussen 2001, Phillips and

Koch 2002, Phillips and Gregg 2003). SIA has the advantage of supplying information about assimilated rather than just ingested diet contrary to traditional stomach content dietary analysis that provides a "snapshot" of what an organism is eating at a particular time (Vander Zanden et al. 1997). However, SIA has not the ability to reveal the representative species of the diet. Moreover, the evaluation of the diet gets more complex as the diversity increases (Phillips and Gregg 2003). It is therefore important to combine traditional and novel techniques.

In seabirds, diet studies using traditional techniques are usually restricted to the breeding period and to the breeding ground, which is when and where the birds are readily accessible (Barrett et al. 2007). The techniques are diverse (for review, see Barrett et al. 2007) and include the identification of food items from: feeding behaviour (Guillemette et al. 1992), stomach content analysis of living (Wilson 1984) or dead birds (Guillemette et al. 1992, Gaston and Bradstreet 1993), regurgitations (Rail and Chapdelaine 1998, Rail and Chapdelaine 2000), regurgitated pellets (Hebert et al. 1999c, Kubetzki and Garthe 2003), feces (Kubetzki and Garthe 2003), observation of fish-carrying species (Chapdelaine and Brousseau 1996) and sampling at the feeding sites (Guillemette et al. 1992). Although some techniques are more rewarding, time-efficient and informative than others, they are all complementary and enhance the accuracy of the assessment of diet composition. Confidence further increases when these techniques are combined with SIA (Hobson et al. 1997, Hammill et al. 2005, Tierney et al. 2008).

3.1.4 Gulf of St. Lawrence

The Gulf of St. Lawrence is one of the world's largest estuaries and its watershed includes the Laurentian Great Lakes as well as other large rivers (St. Lawrence Centre 1996). Vertical fluxes from deep waters and input from rivers supply surface waters with a high amount of nutrients (Yeats 1988b). Consequently, primary production can reach elevated levels (Pocklington 1988) and this results in high biological diversity and abundance. This ecosystem is able to support organisms that have high energy requirements such as marine mammals and seabirds.

Seabirds in the Gulf of St. Lawrence are very diverse and abundant with 20 marine bird species and over 7.0×10^5 breeding individuals (Cotter and Rail 2007, Rail and Cotter 2007, Rail, pers. comm.). Seabird populations in the Gulf of St. Lawrence seem to fluctuate with the intensity of law enforcement (Rail and Chapdelaine 2004), fisheries activities (Chapdelaine and Rail 1997) and availability of small prey fish (Chapdelaine and Brousseau 1991, Chapdelaine 1995, Chapdelaine and Brousseau 1996, Rail et al. 1996).

As bioindicators of ecosystems, seabirds can provide valuable information about the state of their ecosystem. The assessment of their diet can reveal prey availability in the ecosystem and therefore the potential source of eventual fluctuations of populations. Dietary studies can be laborious and costly and provide information about the breeding season only, whereas SIA can potentially reveal information throughout the entire annual cycle.

3.1.6 Objectives, hypotheses and predictions

The aim of this study is to compare diet composition estimates using stable isotope signatures ($\delta^{15}N$ and $\delta^{13}C$) in red blood cells of four seabird species with traditional dietary

methods (identification based on observations, regurgitations and regurgitated pellets). I hypothesized that stable isotope analysis is an accurate tool for diet composition assessment. Therefore, the assessment of diet composition using stable isotope analysis techniques will lead to similar results to traditional dietary studies.

3.2 METHODS

Detailed description of the methodologies regarding the study site, sample collection in the field, laboratory methods (sample preparation and chemical analyses) as well as the statistical analyses and calculations are provided in Chapter 2 of this thesis. Latin names and codes are provided for each species in table 2.2 (Chapter 2).

3.2.1 Sample collections

Five species of seabirds were caught on Corossol Island (figures 2.1 and 2.2; Chapter 2). Razorbill (*Alca torda*; RAZO), black-legged kittiwake (*Rissa tridactyla*; BLKI) were caught on the southern part of the island and common eider (*Somateria mollissima*; COEI) on the western part of the island. Great black-backed gull (*Larus marinus*; GBBG) and herring gull (*Larus argentatus*; HERG) were caught on the edges of the island.

Some seabirds (HERG, GBBG and BLKI in this study) may regurgitate spontaneously the content of their crop when they are disturbed (Rail and Chapdelaine 2000, Barrett et al. 2007). Regurgitations of adult and young BLKI were opportunistically collected throughout the 2007 breeding season and accurately identified to species in the field using published keys (Scott and Scott 1988, Nozères and Bérubé 2003, Fontaine 2006). Each prey item was weighed and in the case of weakly digested specimens, length was also recorded. Mass was used to estimate caloric content (kJ/g) of each prey type (Steimle and Terranova 1985, Birkhead and Nettleship 1987, Martensson et al. 1994, Martensson et al. 1996, Lawson et al. 1998, table 3.1). Regurgitations of adult and young HERG and GBBG were opportunistically collected throughout the 2006 season and were treated together. The reasons were that *i)* these seabird species have similar diets (Pierotti and Good 1994, Good

1998) and *ii)* the isotopic signature of the two species did not differ significantly for either nitrogen or carbon ($t_{0.025(39)}$= -0.330; p = 0.744 and $t_{0.025(39)}$= -0.542; p = 0.591 for δ^{15}N and δ^{13}C, respectively; Chapter 2). Prey items were identified to the lowest taxonomic level possible using published keys (Perron 1985, Scott and Scott 1988, Squires 1990, Nozères and Bérubé 2003, Fontaine 2006). Regurgitations were analysed as previously described for BLKI and caloric values were estimated for each prey item (Cummins and Wuycheck 1971, Steimle and Terranova 1985, Birkhead and Nettleship 1987, Guillemette et al. 1992, Martensson et al. 1994, Martensson et al. 1996, Lawson et al. 1998, Ciancio et al. 2007, USDA 2007, table 3.1). Regurgitated pellets were also opportunistically sampled throughout the breeding season. Prey items were identified to the lowest taxonomic level possible but no attempt was made to reconstruct the size of the original prey and therefore, only the frequency of occurrence was calculated for these dietary samples.

During the chick rearing period, adult RAZO carry food items to their chicks by holding the items crosswise in their bill and it is possible to identify the species comprising each food load. Identification of food items was done visually from a distance using binoculars or a telescope during the summer 2007. The length of the prey was estimated by two observers in proportion to the length of the bird's bill (59.3 ± 1.8 mm; mean ± SD; Chapdelaine and Brousseau 1996) and a consensus was reached before recording the value. The observations were recorded systematically when a bird was seen with a prey item. Greater effort was carried out using a blind on the western part of the island (figure 2.2; Chapter 2) with watches of four hours between 6:00 am and 12:00 pm. Further calculations (Koehler et al. 1969, Lambert and Bernier 1989) gave estimates of the mass of sandlance (*Ammodytes americanus*; SAND) and capelin (*Mallotus villosus*; CAPE), both being the

predominant prey items for RAZO in this region and at that time (Chapdelaine and Brousseau 1996). This was done according to the following equations:

$$\text{Mass of SAND} = (209 * 10^{-6}) * \text{Length}^{2.11} \quad (3.1)$$

$$\text{Mass of CAPE} = (2.7 * 10^{-5}) * \text{Length}^{2.76} \quad (3.2)$$

Caloric values were then calculated for each prey (table 3.1) using the mass. Pictures of RAZOs holding a prey in their bill were taken throughout the breeding season to validate the visual (*in situ*) estimation of the length of the prey items. A two-tailed paired *t*-test revealed that there was no significant difference between the *in situ* estimation and the estimation done with photographs ($t_{0.025(2)8} = -1.77$; $p = 0.115$). The confidence regarding the *in situ* estimation method was good considering the high power of this statistical analysis (Zar 1999). A scare-off technique was also performed throughout the breeding season to collect and measure (mass and length) prey items dropped by adults carrying food back to the colony. None of the fish length measurements and estimations using the methods mentioned above were significantly different from the length of the items used for the food web analysis ($F_{(3,43)} = 2.69$; $p = 0.058$; Chapter 2) using SAND as proxy.

3.2.3 Statistical analyses and calculations

Mixing models for 2 sources (Phillips and Gregg 2001), 3 sources (Phillips and Koch 2002) and multiple sources (Phillips and Gregg 2003) were used to determine the relative contribution of each prey or prey item to the diet of a seabird using the stable isotope approach. In the case of the 2 sources model (IsoError), the isotope with the highest

difference between the isotopic signatures of the sources was used ($\delta^{15}N$) to minimise the proportional standard error. For the 3 sources model (IsoConc), values of 2.6 and 1 % were applied for $\delta^{15}N$ and $\delta^{13}C$, respectively to account for the trophic enrichment between whole prey and the blood of the seabirds (Bearhop et al. 2002). Moreover, the concentration of organic nitrogen and carbon was used in this model to compensate for potential differences in elemental composition of the sources (Phillips and Koch 2002). $\delta^{13}C$ values were lipid-normalized in the case of BLKI using the equations presented in McConnaughey and McRoy (1979) to calculate lipid-normalized $\delta^{13}C$ ($\delta^{13}C'$):

$$\delta^{13}C' = \delta^{13}C + D\,(-0.207 + 3.90 / (1 + 287 / L)) \qquad (3.3)$$

where D is the ^{13}C-depletion of lipids compared to protein (D = 6 ‰). Lipid content (L) was calculated using equation 2.12 (Chapter 2). $\delta^{13}C$ data were only normalized for BLKI because several individuals were otherwise falling outside the mixing polygon.

Multiple food items (> 3) were found in the diet of GBBG and HERG (see *Sample collection*). The food items were arbitrarily grouped into categories based on aspects of their type, phylogeny, size, feeding behaviour, habitat and/or their isotopic signature. The five categories that were most represented in the regurgitations of GBBG and HERG based on the frequency of occurrence, numerical abundance, relative mass and relative energy were included in the computer program (IsoSource). However, more emphasis was put on the relative caloric value of prey items to establish the predominant categories. Food items found in regurgitated pellets were also taken into account to determine the categories. The choice was also supported by the literature (Baird 1994, Pierotti and Good 1994, Good 1998,

Goudie et al. 2000, Hipfner and Chapdelaine 2002) and by behavioural observations in the field between 2005 and 2007.

In this thesis, overall diet composition refers to estimations over the entire breeding season. The calculation considers all the regurgitations, regurgitated pellets or food load in a single season. The meal specific diet composition refers to the average of the proportion of a prey item in each regurgitation, regurgitated pellet or food load. The meal specific diet composition was used in the comparison with the diet composition assessment using stable isotope analysis.

In order to determine if the diet composition assessed using mixing models reflected the one assessed in the field, Student *t*-tests were used to compare each prey or food item categories between the two methods. Data from the two methods were from different individuals, so they were treated as two independent groups. Bonferroni corrections were applied with $\alpha = 0.025$, 0.017 and 0.01 for 2, 3 and 5 food items (or food categories), respectively. Statistical analyses were performed using S-Plus (version 8.0) and an α of 0.05 was used to determine the significance level unless otherwise stated.

3.3 RESULTS

In this chapter, I will assess the diet of five species of seabirds which have distinct feeding ecology using 1) traditional techniques consisting of examination of food items in the field and 2) stable isotope analysis of blood of these species. I will then determine whether the methods lead to similar results.

3.3.1 Black-legged kittiwake

The diet of BLKI essentially consisted of CAPE, SAND and shrimp in order of importance of contribution to the diet for all estimations used (table 3.2). CAPE was identified in 82.0 % of the regurgitations and this proportion dropped to 51.0 % based upon numerical abundance. The relative mass and energy of CAPE to overall diet composition over the entire breeding season was 76.4 and 81.4 %, respectively. These percentages were close to the ones reported when looking at the relative meal specific diet composition of BLKI with 76.6 ± 5.5 and 77.7 ± 5.4 (%; mean ± SE) for mass and energy, respectively.

The relative contribution of prey species to the diet of BLKI estimated using a published three-source, dual-isotope mixing model (IsoConc, Phillips and Koch 2002) was mostly CAPE (50.9 %) and SAND (33.1%), whereas the proportion of shrimp was less important with 16 % (figure 3.1). The diet composition assessed using the mixing model was similar to the diet assessed in the field. The proportion of CAPE in the diet of BLKI in the field was 77.7 % followed by 16.6 % of SAND and 5.6 % for shrimp. Only CAPE ($t_{0.025(63)} = 6.86$; $p < 0.001$; figure 3.1) differed significantly between the two methods.

3.3.2 Great black-backed gull and herring gull

The diet of GBBG and HERG (*Larus* spp.) was very diverse (table 3.3). CAPE was the most important contributor to the diet of *Larus* spp. based on the energy provided (28.9 %) and this value increased when grouping all the fish together (40.5 %). Human refuse provided a considerable amount of energy at 22.3 %. However, the latter value should be considered with great caution since this item (cheese) was found in only one regurgitation (frequency of occurrence 2.5 %; table 3.3), and since it was heavy (113.5 g) and dense in terms of energy (16.8 kJ/g), its relative contribution to the diet was overestimated. Moreover, this item would probably not have been assimilated by the bird since it was wrapped in plastic. It was therefore excluded from the comparison between the two methods (see figure 3.2). Farmed meat was also an important proportion of the diet when looking at relative energy (8.8 %), but again, this item was found in two regurgitations only (frequency of occurrence: 5 %). Molluscs provided 8.5 % of energy to the diet with BUCC being most important (7.5 %). Although insects were important in the diet when looking at the frequency of occurrence (47.5 %) or at the numerical abundance (67.9 %), the contribution decreased when looking at the relative energy provided (3 %). Fruits and BLMU were only important in the diet when taking into account the frequency of occurrence in the regurgitated pellets with 13.7 and 16.2 %, respectively. Crabs, mammals, urchins and plants could only be identified in the pellets (table 3.3).

When the dietary items were summarized into general categories (table 3.4), fish and human food were by far the most important items in the overall diet of GBBG and HERG with 40.5 and 31.1 % of relative energy, respectively. Unknown vertebrate and BUCC were present in the diet with 7.5 % contribution to the energy budgets of these species followed

by MENO (3.6 %) and seastar (3.5 %). Although insects, decapods, plants and BLMU were below 3% of relative energy, these prey items were important either when looking at the frequency of occurrence in the regurgitations (insect: 47.5 %) or in the pellets (15.8, 19.7 and 18.4 % for decapods, plant and BLMU, respectively). When looking at the meal specific diet composition assessed in the field, fish were the most important contributor with 44.8 % followed by DECA and BLMU with 17.2 % each. MENO and BUCC were the least important contributors in the diet with 10.3% for both prey items (figure 3.2a; table 3.4).

The diet composition estimated using the mixing model IsoSource (Phillips and Gregg 2003) was similar for all dietary items with mean values between 10.3 and 26.8 % (figure 3.2a). DECA was the most important contributor with a mean value of 26.8 % (1-99th percentile: 1-45 %) followed by MENO with a mean of 24.4 % (9-42 %) and by fish with a mean of 20.1 % (7-48 %). BLMU and BUCC were the least important contributors with means of 16.3 % (3-31 %) and 12.3% (2-25 %), respectively (figure 3.2).

The only significant difference between the diet composition methods was found for MENO which was more than two fold higher using the mixing model compared to the field ($t_{(0.025)57} = -3.20$; $p = 0.003$). Surprisingly, the methods were not significantly different for fish even though there was a two fold difference between them ($t_{(0.025)57} = 1.71$; $p = 0.097$). Proportions of DECA, BLMU and BUCC were similar between the methods and did not exhibit any significant difference ($p > 0.05$).

3.3.3 Razorbill

SAND dominated the diet when looking at the frequency of occurrence and numerical abundance with 92.3 and 94.0 %, respectively. CAPE constituted the remainder

of the diet. No other prey species were identified throughout the entire breeding season. The contribution of CAPE to the diet was more important when accounting for mass (20.8 %) and energy content (25.6 %) than when using other estimations. For both dietary assessment methods, SAND was by far the most important prey in the diet of RAZO with 81.0 and 92.3 % for the diet composition assessment using the mixing model and meal specific diet composition, respectively (table 3.5; figure 3.3). Methods showed similar proportions for both prey species ($t_{0.025(94)}$; $p > 0.025$; figure 3.3).

3.3.4 Common eider

Diet composition of COEI was constituted of 72.7 % of an aggregation of URCH and BLMU. SNCR constituted the remainder of the diet with 27.3 % (figure 3.4). No dietary assessment was done in the field for this species and comparison between methods could therefore not be done.

3.4 DISCUSSION

Seabird muscle tissue has been used in other studies to determine predator-prey interactions but this requires lethal sampling. For this reason, blood was used in this study. The isotopic turnover rate of whole blood (11.4 days) is similar to muscle tissues (12.4 days) but a statistically significant difference between these tissues was observed ($F_{4,53} = 4.5$; $p < 0.01$; Hobson and Clark 1992a). Trophic enrichment factors for $\delta^{15}N$ were found to be similar among whole blood, plasma, RBCs and breast muscle of captive dunlin (*Calidris alpina pacifica*) maintained on a constant diet for 54 days (Evans Ogden et al. 2004). Blood is easy to collect, it has the advantage of being non-lethal and it is useful to assess the diet of seabirds (Tierney et al. 2008). Because RBCs have a relatively fast turnover, they provide information regarding feeding ecology within a short-term period (Peterson and Fry 1987).

3.4.1 Black-legged kittiwake

Contrary to *Larus* spp., the specialized diet of BLKI and RAZO allowed a simple comparison between the methods. BLKI is a piscivorous surface-feeder that may reach 0.5 to 1 m when diving (Bayer 1983) mostly feeding on schooling fish and small invertebrates caught at the surface. In the Gulf of St. Lawrence, its diet is known to consist almost entirely of CAPE and SAND during the breeding season (Chapdelaine and Brousseau 1989, Rail, pers. comm.) with a generally higher frequency of occurrence and relative mass for SAND (Rail, pers. comm.). Interannual variability in the importance of shrimp, ATHE, sculpins and other types of fish has been reported (Rail, pers. comm.). My data suggest the opposite for the major prey items with CAPE being more important than SAND. $\delta^{13}C$ data were

normalized for lipid content for BLKI only because several individuals were otherwise falling outside the mixing polygon.

3.4.2 Great black-backed gull and herring gull

The high diversity of the diet of GBBG and HERG provided a diffuse diet composition using mixing models. This complicated the identification of preponderant sources and the comparison between the methods. The individuals were scattered throughout the mixing polygon (figure 3.2b) and the identification of predominant sources was problematic.

GBBG is a generalist predator that consumes fish, aquatic and terrestrial invertebrates, mammals, birds, carrion, discarded fishery items and human refuse. Kleptoparasitism was also reported for this species (Good 1998) meaning that GBBG can consume prey items that are usually out of their reach. This makes the prediction and the interpretation of their diet even more difficult. Some cases of cannibalism were also identified (Good 1998).

HERG is a generalist predator with a similar diet and feeding behaviour to that of GBBG, although the latter feeds higher in the food chain (Pierotti and Good 1994, Good 1998). Capelin was found to be the most important prey item in the diet of HERG in the St. Lawrence Gulf and Estuary in 1994-97 with 62.5 % of contribution by mass in the total diet (Rail and Chapdelaine 2000). Other types of fish represented 22.7 % of the diet and the remainder was composed of crustaceans, annelids, human refuse and other types of food (Rail and Chapdelaine 2000).

3.4.3 Razorbill

RAZO is a pursuit diver usually foraging at depths greater than 30 m (Wanless et al. 1990), but can reach over 100 m (Piatt and Nettleship 1985). The numerical abundance of SAND was 64 to 78% of the prey brought by adults to their chicks in the Gulf of St. Lawrence between 1990 and 1992, whereas CAPE constituted the vast majority (if not the totality) of the remaining proportion (Chapdelaine and Brousseau 1996). Chapdelaine and Brousseau (1996) found that CAPE was more important in the diet when looking at the relative mass with 53 to 69 % followed by SAND. My study differs in that it suggests a dominant contribution of SAND in both numerical abundance and relative mass. A trophic enrichment factor of 3.4 ‰ (Minagawa and Wada 1984, Post 2002) was used instead of 2.6‰ (Bearhop et al. 2002) because several individual RAZO were otherwise falling outside the mixing polygon. The former trophic enrichment factor was therefore considered more suitable for this species.

3.4.4 Common eider

COEI is a benthivorous diver in shallow waters that derives its energy mostly from BLMU (44.4 to 83.8%), URCH (20.4 to 60.0%) and spider crab (*Hyas araneus*; 0 to 42.3%) during the winter, but can also feed on other type of molluscs and echinoderms (Guillemette et al. 1992). *Littorina* spp, BLMU and *Gammarus oceanicus* were found to constitute 95% of their diet during the summer (Cantin et al. 1974). According to the latter authors, *Nereis virens*, ATHE's eggs, algae, URCH, insects and other types of molluscs can also be included in the diet before and during the breeding period. The large variance in the $\delta^{13}C$ signature in this study (table 2.2; figure 2.7; Chapter 2) is thus not surprising considering the diversity of

prey items consumed by COEI. Moreover, some COEI had low $\delta^{13}C$ values indicating a pelagic carbon source. Although ATHE eggs were not included in the analysis, low $\delta^{13}C$ values can be expected compared to the adult ATHE (figure 2.7; Chapter 2) which could have decreased COEI $\delta^{13}C$ signatures in some individuals.

3.4.5 Comparison of methods

The data found in the literature were consistent with the results of this field study for most species, except for COEI for which data were not generated in the field. Therefore, discrepancies between the methods reported here would not be the result of improperly identifying field diet items. Possible reasons explaining differences between the methods can be diverse, but are summarized as follows.

The methods did not reflect the same period of time during which seabirds were feeding. The time-lag between the moment when the birds were caught and when the dietary studies were conducted is an important aspect to take into account. The blood was sampled at the beginning of the breeding season while the adults were incubating, whereas the diet in the field was assessed mostly afterwards. Diet is known to vary seasonally for several seabird species (Cantin et al. 1974, Hobson 1993). The turnover rate of blood for $\delta^{15}N$ and $\delta^{13}C$ is approximately 14.4 and 15.7 days, respectively (Bearhop et al. 2002) and therefore, the isotopic signature of blood reflected food assimilated two weeks before the birds were caught. An ideal option would be to do the dietary evaluation in the field first, taking into consideration the turnover rate of the tissue of interest (RBC in this case) and then sample the tissue for SIA that corresponds to the turnover rate of the tissue. However, an important constraint would emerge since it is sometimes not possible to assess the diet of a species

before the breeding season. Some birds are offshore before arriving to the breeding site and it can be laborious to identify accurately their diet composition with a sufficient sample size. Moreover, catching these seabirds and sampling tissues in a non-lethal way before the incubation period is difficult (Barrett et al. 2007). For logistical reasons and in order to maximize the sample size in a non-lethal way, birds were caught during the incubation period (mid-incubation) to avoid nest desertion by the adults. Dietary analyses in the field were conducted afterwards throughout the breeding season.

The trophic enrichment factor is another important assumption that differs substantially between and within species (Vanderklift and Ponsard 2003, Jardine et al. 2006). δ^{15}N is known to increase by approximately 3 to 5 ‰ (Deniro and Epstein 1981, Peterson and Fry 1987) with an average of 3.4 ‰ per trophic level for most animals (Minagawa and Wada 1984, Cabana and Rasmussen 1994, Post 2002) and 2.6 ‰ for birds (Bearhop et al. 2002). Differences in degree of fractionation between birds and other animals is believed to result from the production of uric acid in birds instead of urea (Hobson and Clark 1992b, Hobson and Welch 1992). A meta-analysis conducted on 134 estimations of predator-prey trophic enrichment factors for δ^{15}N suggested high variability among and within taxonomic classes (Vanderklift and Ponsard 2003). The difference within a class is mostly due to the tissue used. For instance, muscle and feathers lead to substantial intraspecific differences for both δ^{13}C and δ^{15}N trophic enrichment factors (Bearhop et al. 2002, Cherel et al. 2005, Dalerum and Angerbjorn 2005). Trophic enrichment factors were also found to increase with the concentrations of nitrogen and carbon in the diet within the same species (Pearson et al. 2003). Nutritional stress was also found to alter the trophic fractionation factor of δ^{15}N within the same species due to preferential excretion of the

lighter ^{14}N (Hobson et al. 1993). Lipid concentration in the tissue used for SIA is also known to affect $\delta^{13}C$ significantly (Bearhop et al. 2002) resulting from the fractionation of the heavier ^{13}C during the oxidation of pyruvate to acetyl-CoA during lipid synthesis (Deniro and Epstein 1977). To avoid this issue, lipids can be removed from the tissue of interest or normalized using the carbon:nitrogen ratio (C:N) routinely provided with the stable isotope results (McConnaughey and McRoy 1979). To avoid the abovementioned sources of uncertainty, it has been suggested that trophic enrichment factors should be generated under controlled laboratory experiments for each species of interest prior to conducting dietary studies (Jardine et al. 2006). However, such an approach is time-consuming, costly and not feasible for all wildlife species. In this study, the trophic enrichment factor used for RAZO was different (3.4 ‰) than for BLKI, GBBG and HERG (2.6 ‰). Different trophic enrichment factors could lead to different conclusions.

In this study, the diet composition assessed in the field mostly represented the diet of the chicks. It has been shown that it may not necessarily constitute an exact concordance with the adults' diet (Gaston and Bradstreet 1993, Hobson 1993, Barrett et al. 2007). Adults might bring prey with higher energy density and keep the prey with lower caloric values or lower trophic level prey items for themselves (Hobson 1993, Barrett et al. 2007). On the other hand, adults are able to eat larger prey items than nestlings (Shealer 1998). Carrying large prey items back to the breeding ground can be energetically costly (Barrett et al. 2007). However, food availability is probably an important variable determining the type of prey brought to the young. The diet of the chicks is therefore likely to be similar to that of adults.

3.5 CONCLUSIONS

The assessment of diet using stable isotope analysis led to similar results to assessments done in the field, although significant differences existed for some species. Seabirds that had specialized diets were easier to assess than species with diverse diets. Discrepancies between methods can be explained by several variables that may or may not be controlled by the investigator. Timing is believed to be important in order to compare methods since seasonal shifts exist for several seabird species and food availability can vary according to the season. Also, the trophic enrichment factor between the predator and its diet can vary substantially depending on the tissue used and the nutritional value of the prey. Finally, the concordance between the adults' and the chicks' diet has to be established prior to conducting a study for which the diet of the nestlings is chosen as a proxy.

More controlled studies need to be conducted on species that are acknowledged as being suitable bioindicators and biomonitors of the environment in order to establish valid trophic enrichment factors. This will help to identify more accurately the importance of keystone prey species thereby facilitating assessments of food availability in ecosystems. Moreover, this could allow predicting the exposure of top level predators to environmental bioaccumulative and biomagnifying contaminants present at the base of the food chain.

Table 3.1. Energy density (kJ/g) for dietary items used to calculate the diet composition of seabirds. Surrogate dietary items were used when actual dietary items could not be found in the literature. Codes are provided in table 2.2.

Dietary item	Surrogate dietary item	Energy density (kJ/g ww)	References
Unknown vertebrate	Vertebrates	7.8	Cummins & Wuycheck, 1971
Unknown Bird	HERG	8.1	Cummins & Wuycheck, 1971
ATHE		10.6	Steimle & Terranova, 1985
WIFL		3.6	Steimle & Terranova, 1985
AMPL		4.1	Steimle & Terranova, 1985
CAPE		7.9	Birkhead & Nettleship, 1987; Lawson et al 1998; Martensson et al 1996; Martensson et al 1994
SAND		6.0	Steimle & Terranova, 1985; Birkhead & Nettleship, 1987; Lawson et al 1998
Unknown fish	Mean of 57 species of fish	5.5	Steimle & Terranova, 1985
C. oplio	*Cancer irroratus* (Common crab)	3.7	Steimle & Terranova, 1985
PABO, PMUL and PMON	*P. borealis*	4.8	Lawson et al 1998
Gammarellus sp.	Gammaridae	3.4	Cummins & Wuycheck, 1971
MENO		3.4	Steimle & Terranova, 1985
URCH		1.3	Steimle & Terranova, 1985; Guillemette et al 1992; Cummins & Wuycheck, 1971
Asterias rubens and *Leptasterias polaris*	Mean of 4 species of sea star	2.9	Steimle & Terranova, 1985
BLMU		2.0	Ciancio etal 2007; Guillemette et al 1992
BUCC		7.7	Steimle & Terranova, 1985
Insects in general	Insecta	3.2	Cummins & Wuycheck, 1971
Coleoptera		5.8	Ciancio etal 2007
Berries	Blueberries, frozen, unsweetened (NDB No. 09054)	2.1	USDA, 2007
Farmed meat	Pork, center cut chops, fresh pork (NDB No. 10855)	7.0	USDA, 2007
Human refuse	Cheese, cheddar (NDB No. 01009)	16.8	USDA, 2007

Table 3.2. Diet composition of adult or young BLKI from regurgitations (n = 50) collected on Corossol Island. The absolute and relative frequency of occurrence, numerical abundance, mass and energy values are shown for each prey item and the data represent a mean for overall diet composition over the entire breeding season. Relative mass and energy (%; mean ± SE) provided by each prey within each meal (meal specific diet composition) are also presented. Shrimp were identified to species where possible, but were grouped together as shrimp (SHRI) in the statistical analyses. Caloric values of prey items are provided in table 3.1.

	Overall diet composition								Meal specific diet composition		
	Frequency of occurrence		Numerical abundance		Mass		Energy			Based on mass	Based on energy [a]
	n	(%) [b]	*n*	(%)	(g)	(%)	(kJ)	(%)	*n*	(%)	(%)
CAPE	41	82.0	49	51.0	367	76.4	2880	81.4	41	76.6 ± 5.5	77.7 ± 5.4
SAND	11	22.0	32	33.3	94.0	19.6	566.4	16.0	11	17.1 ± 5.0	16.6 ± 5.0
SHRI	7	14.0	15	15.6	19.0	4.0	91.2	2.6	7	6.3 ± 3.0	5.6 ± 2.9
Total	59	118	96	100	480	100	3540	100	59	100	100

[a] Diet composition used to compare with mixing model using IsoConc (Phillips and Koch 2002)

[b] Calculated using the number of regurgitations (n = 50)

Table 3.3. Detailed diet composition of adult or young GBBG and HERG from pellets (n = 76) and regurgitations (n = 29) collected on Corossol Island. The absolute and relative frequency of occurrence, numerical abundance, mass and energy values are shown for each dietary item and the data represent a mean for overall diet composition over the entire breeding season.

	Pellets		Regurgitations							
	Frequency of occurrence		Frequency of occurrence		Numerical abundance		Mass		Energy	
	nb	(%) [a]	nb	(%) [b]	nb	(%)	(g)	(%)	(kJ)	(%)
CAPE	0	0.0	7	17.5	25	1.4	315	24.5	2482	28.9
Human refuse	0	0.0	1	2.5	1	0.1	114	8.8	1911	22.3
Farmed meat	0	0.0	2	5.0	2	0.1	108	8.3	752.1	8.8
BUCC	1	1.3	3	7.5	17	1.0	83.7	6.5	644	7.5
SAND	0	0.0	4	10.0	17	1.0	88.4	6.9	533	6.2
Bird	1	1.3	1	2.5	1	0.1	65	5.0	527	6.1
Fish	2	2.6	3	7.5	3	0.2	83	6.4	456	5.3
MENO	0	0.0	3	7.5	266	15.4	89.8	7.0	305	3.6
Sea star	2	2.6	2	5.0	2	0.1	103	8.0	299	3.5
Lepidoptera	0	0.0	6	15.0	613	35.5	49.9	3.9	159	1.8
PABO	0	0.0	2	5.0	5	0.3	20.2	1.6	97.0	1.1
Fruit	14	18.4	2	5.0	100	5.8	43.1	3.3	91.8	1.1
BLMU	14	18.4	5	12.5	110	6.4	44.8	3.5	88.4	1.0
Vertebrate	0	0.0	1	2.5	1	0.1	8.6	0.7	66.7	0.8
Mammal or bird	0	0.0	1	2.5	1	0.1	6.6	0.5	51.2	0.6
Coleoptera	0	0.0	5	12.5	80	4.6	5.7	0.4	33.0	0.4
Hymenoptera	0	0.0	2	5.0	147	8.5	6.8	0.5	21.6	0.3
Shrimp	0	0.0	3	7.5	3	0.2	3.8	0.3	18.2	0.2
Insect	1	1.3	2	5.0	200	11.6	4.9	0.4	15.6	0.2
Diptera	0	0.0	2	5.0	102	5.9	4.3	0.3	13.7	0.2
PMUL	0	0.0	1	2.5	1	0.1	1.5	0.1	7.2	0.1
Hemiptera	0	0.0	1	2.5	30	1.7	0.9	0.1	5.2	0.1
Orthoptera	0	0.0	1	2.5	2	0.1	0.7	0.1	2.2	0.0
Debris	35	46.1	4	10.0	0	0.0	37.9	2.9	0.0	0.0
Crab	10	13.2								
Mammal	4	5.3								
URCH	4	5.3								
Decapod	2	2.6								
Plant	1	1.3								
Total	91	119.7	64	160	1729	100	1289	100	8579	100

[a] Calculated using the number of regurgitated pellets (n = 76)

[b] Calculated using the number of regurgitations (n = 40)

Table 3.4. Summary of the diet composition of adult or young GBBG and HERG from pellets (n = 76) and regurgitations (n = 40) collected on Corossol Island. The absolute and relative frequency of occurrence, numerical abundance, mass and energy values are shown for each dietary item and the data represent a mean for overall diet composition over the entire breeding season. Relative mass and energy (%; mean ± SE) provided by each prey within each meal (meal specific diet composition) are also presented (n = 29).

	Overall diet composition										Meal specific diet composition
	Pellets		Regurgitations								Regurgitations
	Frequency of occurrence		Frequency of occurrence		Numerical abundance		Mass		Energy		Mass or energy [a]
	nb	(%) [b]	nb	(%) [c]	nb	(%)	(g)	(%)	(kJ)	(%)	(%)
Fish	2	2.6	14	35.0	45	2.6	487	37.8	3470	40.5	44.8 ± 9.4
Human food	0	0.0	3	7.5	3	0.2	221	17.2	2663	31.1	
Unknown vertebrate	5	6.6	3	7.5	3	0.2	80.2	6.2	645	7.5	
BUCC	1	1.3	3	7.5	17	1.0	83.7	6.5	644	7.5	10.3 ± 5.8
MENO	0	0.0	3	7.5	266	15.4	89.8	7.0	305	3.6	10.3 ± 5.8
Seastar	6	7.9	2	5.0	2	0.1	103	8.0	299	3.5	
Insect	1	1.3	19	47.5	1174	67.9	73.2	5.7	250	2.9	
DECA	12	15.8	6	15.0	9	0.5	25.5	2.0	122	1.4	17.2 ± 7.1
Plant	15	19.7	2	5.0	100	5.8	43.1	3.3	91.8	1.1	
BLMU	14	18.4	5	12.5	110	6.4	44.8	3.5	88.4	1.0	17.2 ± 7.1
Debris	35	46.1	4	10.0	0	0.0	37.9	2.9	0	0.0	
Total	91	120	64	160	1729	100	1289	100	8579	100	100

[a] Diet composition used to compare with mixing models using IsoSource (Phillips and Gregg 2003). A sample size of 29 regurgitations was used for the calculation

[b] Calculated using the number of regurgitated pellets (n = 76)

[c] Calculated using the number of regurgitations (n = 40)

Table 3.5. Diet composition of adult RAZO from prey loads (n = 78) collected on Corossol Island. The absolute and relative frequency of occurrence, numerical abundance, mass and energy values are shown for each prey and the data represent a mean for overall diet composition over the entire breeding season. Relative mass and energy (%; mean ± SE) provided by each prey within each meal (meal specific diet composition) are also presented.

	Overall diet composition								Meal specific diet composition		
	Frequency of occurrence		Numerical abundance		Mass		Energy			Based on mass	Based on energy [a]
	n	(%) [b]	n	(%)	(g)	(%)	(kJ)	(%)	n	(%)	(%)
CAPE	6	7.7	7	6.0	160	20.8	1260	25.6	6	7.7 ± 3.0	7.7 ± 3.0
SAND	72	92.3	110	94.0	607	79.2	3650	74.4	72	92.3 ± 3.0	92.3 ± 3.0
Total	78	100	117	100	766	100	4910	100	78	100	100

[a] Diet composition used to compare with mixing model using IsoError (Phillips and Gregg 2001)
[b] Calculated using the number of regurgitations (n = 78)

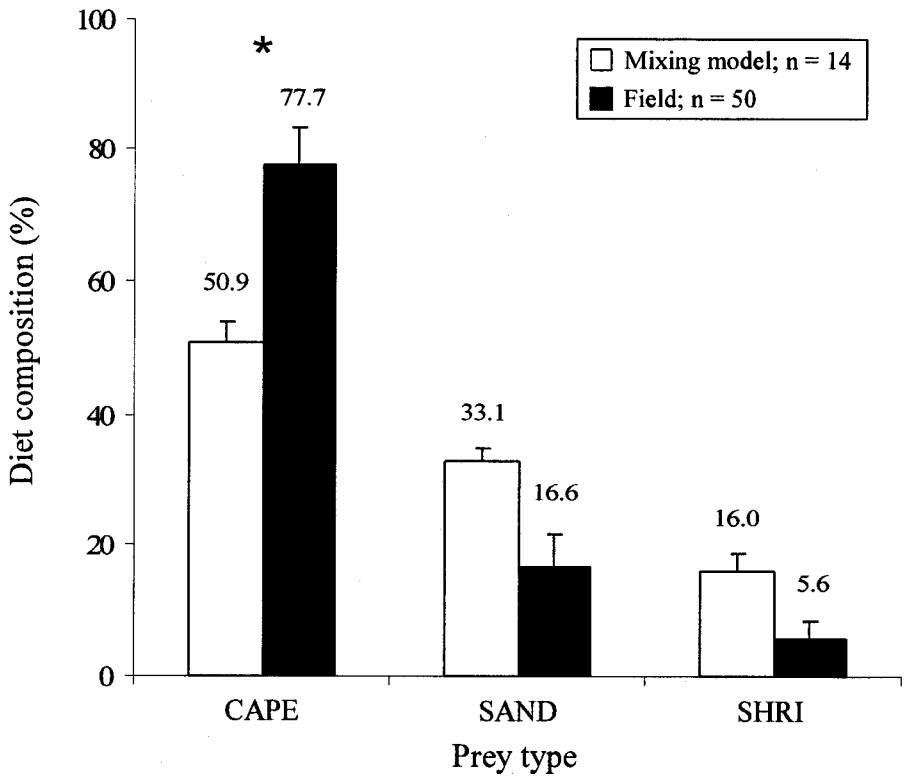

Figure 3.1. Comparison of the diet composition (%; mean ± SE) of BLKI assessed *i)* using a three-source dual-isotope mixing model (IsoConc; Phillips and Koch 2002) with stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes in the red blood cells of adults (white bars; n = 14) and *ii)* using the energy density of different species contained in the regurgitations of adults and young collected on Corossol Island (black bars; n = 50). Seven birds were excluded from the mixing model analysis since they were outside the mixing polygon. Asterisks (*) are shown for sources that differ between the methods. Diet composition using dietary analysis was estimated as the relative energy (%; mean ± SE) provided by each prey within each meal (meal specific diet composition).

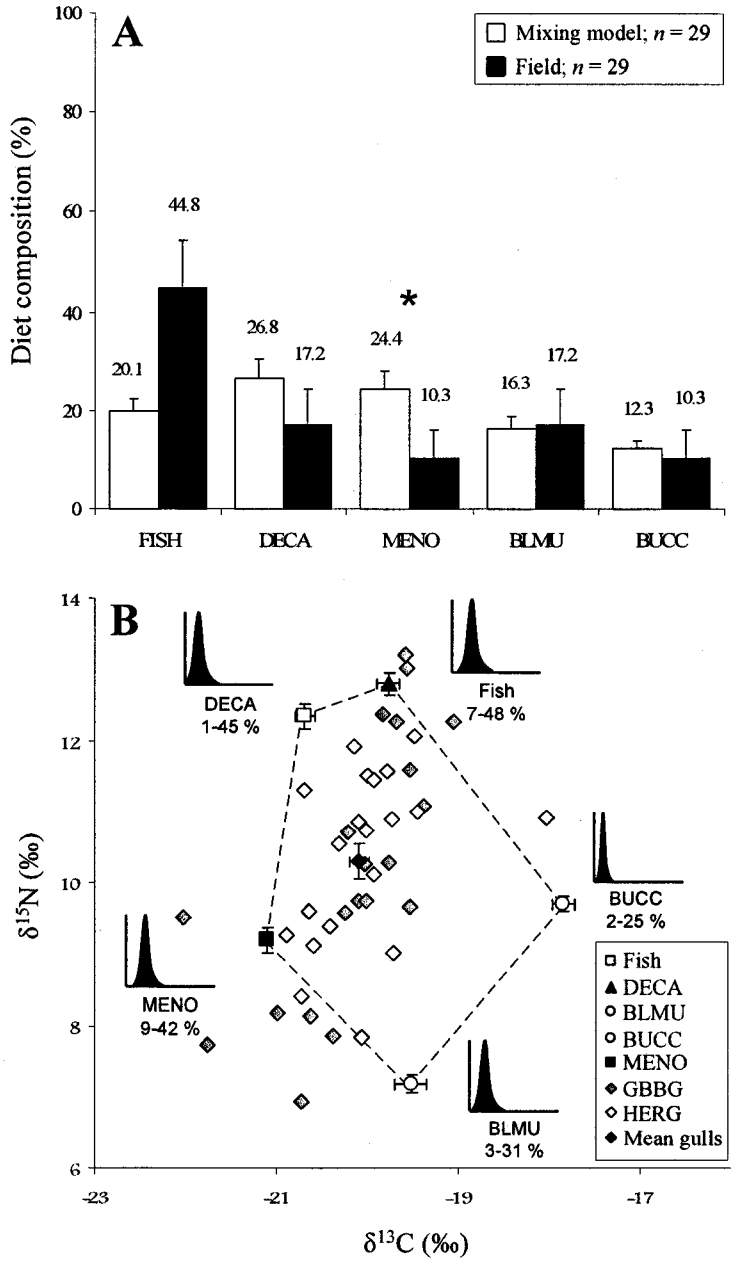

Figure 3.2. A) Comparison of the diet composition (%; mean ± SE) of GBBG and HERG assessed using *i)* a multiple-source dual-isotope mixing model (IsoSource; Phillips and Gregg 2003) with stable nitrogen (δ^{15}N) and carbon (δ^{13}C) isotopes in the red blood cells of adults (white bars; n = 29) and *ii)* the energy density of different dietary items contained in the regurgitations of adults or young collected on Corossol Island (black bars; n = 29). Diet composition using dietary analysis was estimated as the relative energy (%; mean ± SE) provided by each dietary item within each meal (meal specific diet composition). Asterisks (*) are shown for sources that differ between the methods. B) Mixing polygon for δ^{15}N and δ^{13}C with histograms showing the distribution of feasible contributions of five sources to the gulls' diet. Values shown under dietary items represent the 1-99 percentile ranges of the feasible distributions. Individual HERG and GBBG as well as the mean for pooled species are shown after correcting for trophic enrichment factors. Eleven individuals falling outside the mixing polygon were not considered for the analysis.

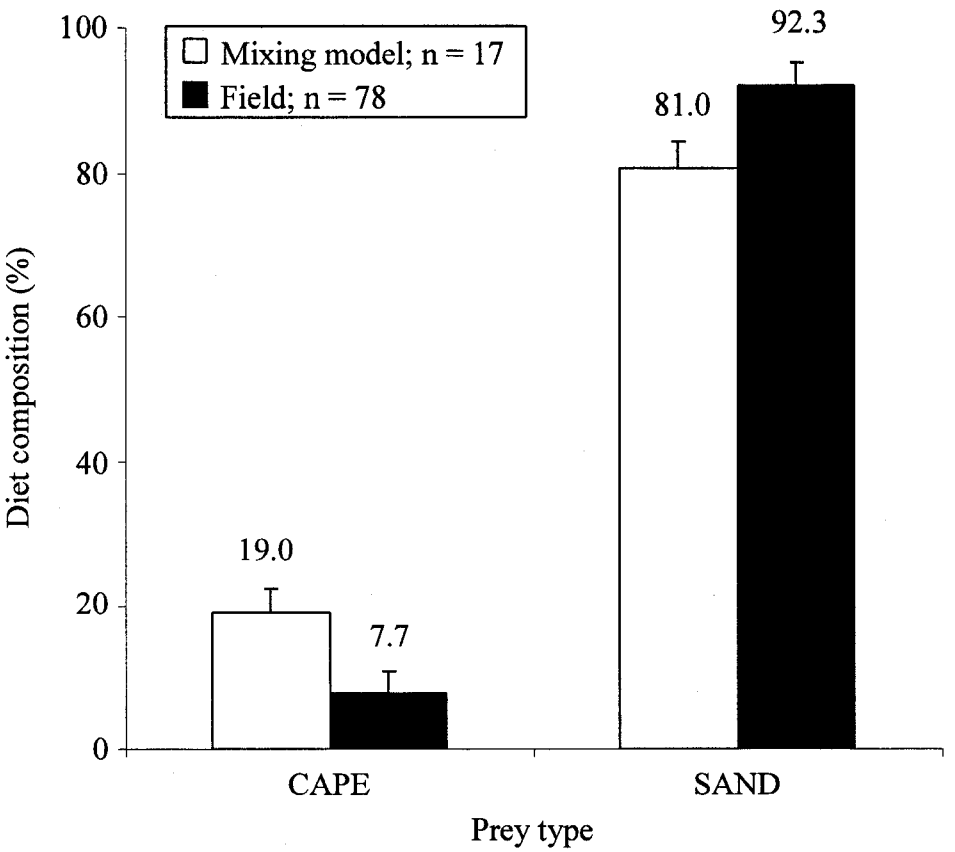

Figure 3.3. Comparison of the diet composition (%; mean ± SE) of RAZO assessed using *i)* a published two-source single isotope mixing model (IsoError; Phillips and Gregg 2001) with stable nitrogen isotope ($\delta^{15}N$) in the red blood cells of adults (white bars; $n = 17$) and *ii)* using the energy density of different species contained in prey loads that adults brought to their chicks on Corossol Island in 2007 (black bars; $n = 78$). Diet composition using dietary analysis was estimated as the relative energy (%; mean ± SE) provided by each dietary item within each meal (meal specific diet composition). Asterisks (*) are shown for sources that differ between the methods.

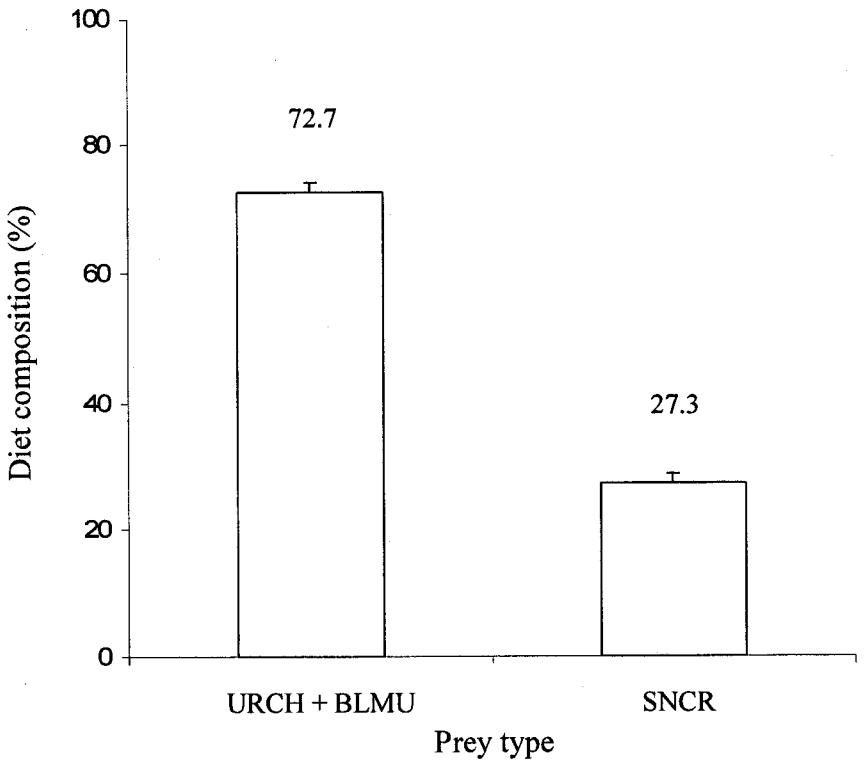

Figure 3.4. Diet composition (%; mean ± SE) of COEI assessed using a published two-source single isotope mixing model (IsoError; Phillips and Gregg 2001) with stable nitrogen isotope ($\delta^{15}N$) in the red blood cells of female adults ($n = 20$) on Corossol Island.

CHAPTER 4

GENERAL CONCLUSIONS

This study illustrates that even though THg concentrations increased with depth of the water column, MeHg levels remained constant. As a result, the fraction of MeHg decreased with depth. Species living or feeding in the benthos were more contaminated with THg and MeHg than species living and feeding strictly in the pelagic zone. This corroborated the hypothesis that sediments are a source of Hg in the Gulf of St. Lawrence. Surprisingly, species that were exploiting both the surface and the bottom of this ecosystem were more contaminated in some cases than the species exploiting only the benthos. Further studies need to be conducted regarding the possible explanations for this finding.

The energy of the upper trophic level species was mostly derived from pelagic primary production with particulate organic matter (POM) at the base of the food chain. This study suggests that this food web is supported by pelagic zooplankton which derives its energy from POM. Low stable carbon isotope ($\delta^{13}C$) values also suggest an allochtonous source of primary production. POM would be derived from freshwater ecosystems as well as from local sources. More extensive studies need to be done to determine whether primary production is produced *in situ* or imported from freshwater systems.

Hg was found to biomagnify substantially in this food web. The results obtained for THg and MeHg were comparable with other food webs around the world. MeHg biomagnification differed significantly from THg biomagnification only when looking at the relationship with trophic level. THg and especially MeHg were found to biomagnify at levels comparable to persistent organic pollutants (POPs) reaching values in seabirds that

were several orders of magnitude higher than values found in the water. Although Hg levels in top level predators were not considered as being high, levels of several organisms at the base of the food chain exceeded levels considered safe for the protection of wildlife. This suggests that 1) top predator species not covered in this study could potentially be at risk and/or 2) adverse health effects could be observed (e.g., at the molecular level) in top level predators in this study. More studies need to be conducted on these two topics.

I failed to obtain evidence to support the hypotheses regarding the influence of the size of the ecosystem, the length of the food chain, the Hg baseline level and the temperature to explain variations in biomagnification power of THg. However, I found that latitude was negatively correlated with THg concentration at the base of the food chain. Nevertheless, this needs further investigation since it has been recognized that Hg levels are high in the Arctic (Macdonald et al. 2000). The scarcity of studies that have measured MeHg did not allow me to compare my work with other food webs. As seen in this study, estimating MeHg in whole fish or invertebrates based on the assumption that all THg is in the form of MeHg could lead to erroneous results. This study suggests that further research should systematically include MeHg measurements when reporting THg results, especially for species occupying positions lower in the food chain.

In the food web under study, total weight was the best predictor for THg and MeHg concentrations and was usually followed by trophic level and lipid content. Benthic connection was a poor predictor of Hg concentration at the food web scale, but provided insights within taxonomic classes after normalizing for trophic level or length of the organisms. The overall model explained a high proportion of the variability of Hg.

Stable isotope analysis (SIA) was found to be a good predictor of the diet for some specialized species. Generalist species were problematic and therefore the utility of stable isotope analysis modelling for diet assessment may be limited to specialist seabirds. Several variables can confound the interpretation of the diet assessment using SIA and further research is needed. Captive rearing studies need to be performed on suitable bioindicators species to validate assumptions regarding isotope trophic fractionation in different tissues. The simultaneous use of SIA and traditional techniques can provide an exhaustive and accurate evaluation of short- and long-term diets. A reliable evaluation of diet could help maintain or implement management measures on areas believed to be important for the conservation of wildlife. An accurate assessment of diet could identify important routes of contaminant transfer that could be useful in determining the sources of contamination. Moreover, predictions of the exposure of a consumer to contaminants using the values of the prey species could be possible.

In general, this study provided new knowledge on the trophic structure and the distribution of mercury (Hg) in a Gulf of St. Lawrence ecosystem. The high diversity and complex assemblages of species make difficult the interpretation of the possible variables that are driving energy and contaminant fluxes. Fortunately, the interest in this type of problematic has been acknowledged and the literature on the topic is growing. However, an urgent need regarding the standardization of concepts and methodologies has to be recognized in order to compare studies together and draw broader conclusions on a worldwide scale.

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APPENDIX A – RAW DATA

Table A. Attributes (name, code, taxonomical group, length, weight and sex), total mercury (THg) and methylmercury (MeHg) concentrations (ng/g ww), moisture content (%), stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes (‰), percentage of carbon (C) and nitrogen (N) and trophic level (TL) of all the species used for the Gulf of St. Lawrence food web under study.

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Great black-backed gull	GBBG	Bird		1700	F	131	131	68.80	-19.88	62.4	10.79	16.0	3.38
Great black-backed gull	GBBG	Bird		2300	M	554	552	68.81	-20.93	60.7	12.12	16.0	3.77
Great black-backed gull	GBBG	Bird		1975	M	899	897	71.86	-18.58	60.1	14.89	15.7	4.59
Great black-backed gull	GBBG	Bird		1850	M	289		71.37	-20.66	58.1	10.34	15.8	3.25
Great black-backed gull	GBBG	Bird		1700	F	1131		68.80	-18.98	60.6	12.34	16.1	3.84
Great black-backed gull	GBBG	Bird		1955	M	172		70.36	-19.61	46.1	9.55	14.1	3.02
Great black-backed gull	GBBG	Bird		1940	M	569		63.55	-18.43	46.6	14.22	14.3	4.39
Great black-backed gull	GBBG	Bird		1540	F	257		67.79	-19.27	46.2	10.48	14.3	3.29
Great black-backed gull	GBBG	Bird		1630	F	444		66.55	-19.13	46.9	12.17	14.3	3.79
Great black-backed gull	GBBG	Bird		1960	M	984	984	68.41	-18.45	47.4	15.65	14.3	4.81
Great black-backed gull	GBBG	Bird		1610	F	292	291	69.45	-18.66	46.7	12.88	14.2	4.00
Great black-backed gull	GBBG	Bird		1570	F	251	250	65.90	-18.92	46.1	12.85	14.3	3.99
Great black-backed gull	GBBG	Bird		2090	M	400	399	63.76	-19.52	45.5	10.75	14.4	3.37
Great black-backed gull	GBBG	Bird		1500	F	444		67.27	-19.11	46.7	13.32	14.2	4.13
Great black-backed gull	GBBG	Bird		1510	F	331		64.31	-18.90	46.6	12.34	14.3	3.84
Great black-backed gull	GBBG	Bird		2090	M	360		65.98	-18.43	46.6	12.26	14.2	3.81
Great black-backed gull	GBBG	Bird		1850	M	715		67.35	-18.47	46.7	15.83	14.3	4.86
Great black-backed gull	GBBG	Bird		1310	M	492		64.47	-18.28	46.9	13.70	14.4	4.24
Great black-backed gull	GBBG	Bird		1600	F	294		64.94	-18.73	47.0	14.99	14.4	4.62
Great black-backed gull	GBBG	Bird		1800	M	930		67.36	-17.95	46.6	14.89	14.3	4.59
Herring gull	HERG	Bird		1000	M	112	112	66.37	-19.54	59.7	12.19	16.1	3.79
Herring gull	HERG	Bird		940	F	156	156	69.11	-18.90	59.5	13.33	16.1	4.13
Herring gull	HERG	Bird		1215	M	161	160	64.52	-19.62	59.9	11.02	16.2	3.45

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Herring gull	HERG	Bird		890	F	226		68.11	-19.21	59.7	13.15	16.0	4.08
Herring gull	HERG	Bird		980	F	322		66.03	-18.89	56.0	14.13	16.0	4.36
Herring gull	HERG	Bird		1140	M	299		66.60	-18.82	55.7	14.06	16.1	4.34
Herring gull	HERG	Bird		1010	F	332		64.80	-19.58	55.7	13.91	16.1	4.30
Herring gull	HERG	Bird		1000	F	478	477	65.88	-18.62	55.8	13.50	16.2	4.18
Herring gull	HERG	Bird		1020	F	384		69.23	-18.68	54.9	14.18	16.0	4.38
Herring gull	HERG	Bird		970	F	127		68.14	-18.60	55.7	11.62	16.2	3.63
Herring gull	HERG	Bird		870	F	168	167	66.53	-18.83	56.1	12.72	16.2	3.95
Herring gull	HERG	Bird		1250	M	229	228	66.63	-19.79	55.7	11.87	16.1	3.70
Herring gull	HERG	Bird		950	F	169	166	66.55	-18.99	56.3	13.47	16.3	4.17
Herring gull	HERG	Bird		1130	M	149		66.22	-19.30	55.7	11.99	16.2	3.73
Herring gull	HERG	Bird		930	F	293		63.98	-19.03	55.8	14.54	16.3	4.49
Herring gull	HERG	Bird		890	F	210		65.53	-18.34	55.3	13.60	16.1	4.21
Herring gull	HERG	Bird		970	F	305		66.00	-18.38	61.2	14.68	16.1	4.53
Herring gull	HERG	Bird		1220	M	105		67.33	-18.96	55.5	10.44	16.1	3.28
Herring gull	HERG	Bird		1040	M	429		63.05	-16.92	46.4	13.54	14.3	4.19
Herring gull	HERG	Bird		1000	F	216		69.44	-19.49	45.9	11.71	14.0	3.65
Common eider	COEI	Bird		1200	F	208	205	64.94	-19.61	56.4	11.20	16.3	3.50
Common eider	COEI	Bird		1230	F	213	208	69.64	-18.46	56.1	11.41	16.0	3.56
Common eider	COEI	Bird		1350	F	167	155	69.48	-19.27	55.6	11.46	15.9	3.58
Common eider	COEI	Bird		1240	F	264		68.78	-18.97	56.4	10.77	16.1	3.38
Common eider	COEI	Bird		1650	F	102	99	67.07	-18.39	54.1	10.94	15.6	3.43
Common eider	COEI	Bird		1450	F	233		77.87	-19.21	54.0	11.01	15.2	3.45
Common eider	COEI	Bird		1710	F	229		69.58	-19.47	55.0	10.98	15.7	3.44
Common eider	COEI	Bird		1380	F	251		66.43	-16.57	54.9	11.84	15.7	3.69
Common eider	COEI	Bird		1420	F	115		71.63	-19.40	54.5	11.45	15.5	3.58
Common eider	COEI	Bird		1430	F	243	237	66.49	-19.23	54.9	10.84	15.9	3.40

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Common eider	COEI	Bird		1350	F	147	147	71.52	-19.38	54.8	10.29	15.6	3.24
Common eider	COEI	Bird		1410	F	203	200	69.30	-20.18	55.0	10.81	15.6	3.39
Common eider	COEI	Bird		1540	F	198		67.42	-19.19	54.8	10.84	15.7	3.40
Common eider	COEI	Bird		1830	F	217		71.52	-17.46	54.1	11.72	15.1	3.66
Common eider	COEI	Bird		1720	F	171		70.71	-17.10	54.2	11.45	15.3	3.58
Common eider	COEI	Bird		1350	F	222		67.17	-19.77	54.4	10.60	15.6	3.33
Common eider	COEI	Bird		1420	F	220		67.16	-19.05	54.4	10.38	15.6	3.26
Common eider	COEI	Bird		1490	F	204		66.64	-18.65	54.6	11.57	15.8	3.61
Common eider	COEI	Bird		1310	F	167		68.62	-18.93	54.3	11.11	15.6	3.48
Common eider	COEI	Bird		1310	F	171		66.31	-20.06	54.5	11.33	15.7	3.54
Razorbill	RAZO	Bird		720	M	590	590	69.88	-19.15	51.5	15.39	15.5	4.74
Razorbill	RAZO	Bird		805	F	447	446	74.08	-19.29	51.2	15.40	15.1	4.74
Razorbill	RAZO	Bird		790	F	668	666	69.77	-19.10	51.9	15.33	15.5	4.72
Razorbill	RAZO	Bird		740	F	715		72.23	-19.31	51.4	15.34	15.2	4.72
Razorbill	RAZO	Bird		715	F	527		69.25	-19.08	52.0	15.34	15.5	4.72
Razorbill	RAZO	Bird		700	M	661		71.41	-19.35	51.9	15.27	15.4	4.70
Razorbill	RAZO	Bird		750	M	855		68.77	-19.28	52.8	15.34	15.6	4.72
Razorbill	RAZO	Bird		680	M	560		69.78	-19.31	52.3	15.07	15.4	4.64
Razorbill	RAZO	Bird		690	M	438		67.65	-19.22	52.5	15.30	15.7	4.71
Razorbill	RAZO	Bird		730	M	603	602	70.04	-19.15	52.5	15.50	15.6	4.77
Razorbill	RAZO	Bird		700	M	663	660	66.53	-19.34	52.4	15.29	15.6	4.71
Razorbill	RAZO	Bird		615	F	451	449	64.74	-19.37	52.7	14.91	15.8	4.59
Razorbill	RAZO	Bird		690	F	474	473	63.59	-19.28	52.3	14.84	15.6	4.57
Razorbill	RAZO	Bird		720	F	323		76.19	-19.37	51.9	15.29	15.2	4.71
Razorbill	RAZO	Bird		725	M	633		67.94	-19.17	52.3	15.14	15.5	4.66
Razorbill	RAZO	Bird		650	F	384		67.86	-19.03	52.1	14.44	15.7	4.46
Razorbill	RAZO	Bird		685	M	447		72.53	-19.45	52.0	15.06	15.2	4.64

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Razorbill	RAZO	Bird		635	F	369		73.94	-19.40	51.8	15.31	15.1	4.71
Razorbill	RAZO	Bird		765	M	432		75.32	-19.30	51.6	15.33	15.1	4.72
Razorbill	RAZO	Bird		670	F	474		67.23	-19.22	51.7	15.32	15.5	4.71
Black-legged kittiwake	BLKI	Bird		375.0	M	356	341	66.64	-18.91	52.9	15.24	15.7	4.69
Black-legged kittiwake	BLKI	Bird		345.0	F	246	232	69.53	-18.91	51.7	15.22	15.5	4.69
Black-legged kittiwake	BLKI	Bird		395.0	M	151	146	68.97	-19.02	52.2	15.31	15.5	4.71
Black-legged kittiwake	BLKI	Bird		440.0	M	204		65.64	-18.85	52.2	15.21	15.6	4.68
Black-legged kittiwake	BLKI	Bird		420.0	F	309		64.56	-18.92	52.4	15.58	15.7	4.79
Black-legged kittiwake	BLKI	Bird		432.5	M	363		65.53	-18.74	52.6	15.24	15.8	4.69
Black-legged kittiwake	BLKI	Bird		420.0	M	332		69.96	-18.68	52.2	14.99	15.6	4.62
Black-legged kittiwake	BLKI	Bird		405.0	M	323		67.17	-18.87	52.5	14.87	15.6	4.58
Black-legged kittiwake	BLKI	Bird		450.0	F	252	251	69.87	-18.93	52.1	14.93	15.6	4.60
Black-legged kittiwake	BLKI	Bird		495.0	M	294		66.18	-18.95	52.1	15.27	15.6	4.70
Black-legged kittiwake	BLKI	Bird		392.5	F	158	146	66.05	-18.98	52.6	15.00	15.7	4.62
Black-legged kittiwake	BLKI	Bird		402.5	F	273	246	66.90	-18.72	52.3	15.19	15.6	4.68
Black-legged kittiwake	BLKI	Bird		385.0	F	171	164	70.49	-19.00	52.3	15.16	15.5	4.67
Black-legged kittiwake	BLKI	Bird		405.0	F	249		67.43	-18.81	52.5	15.19	15.6	4.68
Black-legged kittiwake	BLKI	Bird		442.5	M	461		61.68	-18.85	52.5	15.22	15.7	4.69
Black-legged kittiwake	BLKI	Bird		400.0	F	160		62.44	-18.88	52.5	15.09	15.7	4.65
Black-legged kittiwake	BLKI	Bird		447.5	M	290		64.69	-18.92	52.4	15.04	15.7	4.63
Black-legged kittiwake	BLKI	Bird		430.0	F	368		65.91	-19.07	52.4	15.12	15.6	4.66
Black-legged kittiwake	BLKI	Bird		400.0	F	140		65.55	-18.93	52.4	15.32	15.6	4.71
Black-legged kittiwake	BLKI	Bird		405.0	F	218		68.70	-19.06	52.1	15.02	15.4	4.63
Black-legged kittiwake	BLKI	Bird		447.5	M	352		63.25	-18.77	52.0	15.03	15.5	4.63
Atlantic Herring	ATHE	Fish	295.0	245.9	M	29.9	25.6	63.48	-21.04	46.9	11.74	7.9	3.43
Atlantic Herring	ATHE	Fish	296.0	229.7	M	23.7	16.0	69.10	-21.20	52.2	11.81	10.1	3.45
Atlantic Herring	ATHE	Fish	308.5	274.3	F	47.5	30.2	63.72	-21.82	58.7	12.49	9.1	3.65

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Atlantic Herring	ATHE	Fish	320.0	301.6	M	48.7	31.1	68.48	-20.89	54.5	11.63	10.5	3.39
Atlantic Herring	ATHE	Fish	305.0	238.5	M	45.6	30.0	59.49	-22.07	58.6	13.70	7.0	4.00
Atlantic Herring	ATHE	Fish	295.0	236.7	M	41.2	22.0	68.17	-20.89	53.9	11.54	10.8	3.37
Atlantic Herring	ATHE	Fish	291.5	232.8	Un	25.3	17.3	60.35	-21.58	54.0	12.66	9.9	3.70
Atlantic Herring	ATHE	Fish	297.0	258.3		53.7	22.3	64.68	-21.56	55.3	12.20	9.6	3.56
Atlantic Herring	ATHE	Fish	301.5	256.6	M	34.0		66.31	-21.17	54.8	11.96	10.1	3.49
Atlantic Herring	ATHE	Fish	274.0	183.8	M	15.0		63.13	-21.24	56.7	12.71	8.9	3.71
Atlantic Herring	ATHE	Fish	289.5	222.3	M	34.0		67.71	-21.32	53.3	12.24	9.4	3.57
Witch Flounder	WIFL	Fish	292.0	142.8	M	30.1	16.1	80.52	-17.90	42.5	14.32	11.1	4.19
Witch Flounder	WIFL	Fish	170.5	21.4	J	29.6	8.4	77.88	-17.33	39.9	13.95	11.8	4.08
Witch Flounder	WIFL	Fish	179.0	24.4	J	22.9	9.1	79.51	-17.97	40.3	13.73	11.7	4.01
Witch Flounder	WIFL	Fish	289.0	182.8	M	53.0	37.1	77.90	-18.36	43.6	14.99	10.1	4.38
Witch Flounder	WIFL	Fish	330.0	244.2	M	61.2	44.2	80.16	-17.45	43.3	15.49	11.3	4.53
Witch Flounder	WIFL	Fish	173.5	21.6	U	15.3		77.88	-17.80	40.6	14.02	11.6	4.10
Witch Flounder	WIFL	Fish	188.5	26.4	J	38.6	10.0	73.89	-17.69	39.3	14.11	11.2	4.12
Witch Flounder	WIFL	Fish	298.0	155.1	M	45.5	17.8	83.19	-17.13	37.9	14.56	11.2	4.26
Witch Flounder	WIFL	Fish	281.5	124.1	M	25.6		78.05	-17.78	42.9	14.62	10.9	4.27
Witch Flounder	WIFL	Fish	338.0	265.1	M	39.6		77.40	-18.33	44.0	14.63	10.6	4.28
Witch Flounder	WIFL	Fish	221.0	53.9	U	31.2		76.46	-17.89	41.1	14.12	11.2	4.13
American Plaice	AMPL	Fish	219.5	81.7	M	38.2	22.7	74.75	-19.26	42.6	14.24	10.2	4.16
American Plaice	AMPL	Fish	246.0	104.4	M	75.5	35.8	82.17	-17.82	35.6	14.40	10.4	4.21
American Plaice	AMPL	Fish	282.0	194.5	F	20.3	12.0	75.11	-20.32	46.7	14.35	10.1	4.19
American Plaice	AMPL	Fish	177.0	33.4	M	62.6	16.2	79.39	-18.49	36.9	14.72	10.2	4.30
American Plaice	AMPL	Fish	255.0	107.1	M	24.1	14.3	76.77	-19.40	41.3	14.79	10.5	4.32
American Plaice	AMPL	Fish	282.0	203.4	F	21.3	15.8	74.60	-19.73	46.6	14.54	10.7	4.25
American Plaice	AMPL	Fish	252.0	120.4	F	14.4	10.6	74.88	-19.63	42.7	14.48	10.4	4.23
American Plaice	AMPL	Fish	259.0	126.5	M	16.4	8.2	76.97	-18.95	44.3	14.09	12.2	4.12

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
American Plaice	AMPL	Fish	270.5	140.7	M	16.3		76.92	-19.43	43.7	14.52	10.6	4.24
American Plaice	AMPL	Fish	292.0	178.9	F	24.7		78.44	-18.72	41.6	14.16	11.5	4.14
American Sandlance	SAND	Fish	204.0	13.2	U	17.5	13.1	71.56	-20.44	43.1	11.79	10.6	3.44
American Sandlance	SAND	Fish	149.0	7.5	M	33.9	20.6	69.20	-20.62	44.0	11.80	10.9	3.44
American Sandlance	SAND	Fish	170.5	10.7	F	67.6	51.3	72.86	-19.00	42.4	12.23	11.9	3.57
American Sandlance	SAND	Fish	113.5	2.3	J	9.81	6.16	55.19	-20.63	44.4	11.31	11.4	3.30
American Sandlance	SAND	Fish	106.5	2.6	J	19.1	13.1	69.10	-21.02	45.6	11.85	10.7	3.46
American Sandlance	SAND	Fish	109.0	2.7	M	12.4	7.2	68.11	-20.59	45.9	11.15	10.8	3.25
American Sandlance	SAND	Fish	108.5	2.7	U	15.9	9.6	71.60	-20.91	46.4	11.34	11.3	3.31
American Sandlance	SAND	Fish	120.5	3.5	U	10.9	6.1	73.07	-20.25	42.3	11.22	11.4	3.27
American Sandlance	SAND	Fish	155.5	7.7		23.3		69.78	-19.75	44.4	11.71	11.7	3.42
American Sandlance	SAND	Fish	98.5	2.3		11.1		69.05	-21.06	44.2	11.56	11.2	3.37
American Sandlance	SAND	Fish	123.5	3.8	F	10.3		70.55	-20.14	42.8	11.84	11.6	3.46
American Sandlance	SAND	Fish	145.0	6.8	F	14.4		72.47	-20.89	47.1	11.32	10.6	3.30
American Sandlance	SAND	Fish	117.5	4.2	F	24.9		70.81	-21.30	48.7	11.49	10.6	3.35
Capelin	CAPE	Fish	157.0	26.0	M	8.65	5.63	72.80	-20.83	52.6	13.14	9.7	3.84
Capelin	CAPE	Fish	146.5	21.8	M	8.18	5.76	72.85	-20.99	53.5	13.18	9.7	3.85
Capelin	CAPE	Fish	160.0	27.2	M	8.05	5.66	70.32	-21.06	51.8	13.63	9.1	3.98
Capelin	CAPE	Fish	158.0	24.7	M	12.1	8.0	73.57	-20.44	51.1	13.92	9.6	4.07
Capelin	CAPE	Fish	122.5	10.1	F	5.21	2.97	69.82	-20.96	54.9	12.68	9.5	3.70
Capelin	CAPE	Fish	167.0	30.3	M	7.86	0.45	73.49	-20.79	51.0	13.35	9.1	3.90
Capelin	CAPE	Fish	142.0	15.8	M	7.54	4.86	72.82	-21.08	53.0	13.28	9.5	3.88
Capelin	CAPE	Fish	149.0	18.7	M	8.69	5.42	72.26	-20.31	49.4	13.72	10.4	4.01
Capelin	CAPE	Fish	155.0	24.0	M	6.58		71.83	-20.95	54.2	13.32	9.4	3.89
Capelin	CAPE	Fish	159.5	23.0	M	8.38		74.45	-20.31	49.5	13.51	10.1	3.95
Capelin	CAPE	Fish	117.0	5.9	M	6.68		69.58	-21.37	51.1	12.43	9.7	3.63
Capelin	CAPE	Fish	105.5	4.4	M	5.28		71.78	-21.38	50.6	12.40	9.6	3.62

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Northern shrimp	PABO	Decapod	125.4	6.0		31.3	18.1	70.21	-20.60	46.7	12.39	9.2	3.62
Northern shrimp	PABO	Decapod	131.9	6.9		40.7	27.9	71.99	-19.15	42.2	12.29	9.6	3.59
Northern shrimp	PABO	Decapod	126.4	6.0		39.7	21.9	74.15	-19.22	41.1	12.16	9.4	3.55
Northern shrimp	PABO	Decapod	136.4	8.2		44.6	9.7	69.34	-19.89	45.6	12.98	9.4	3.79
Northern shrimp	PABO	Decapod	125.2	7.4		55.6	28.6	71.04	-19.45	42.9	12.57	9.6	3.67
Northern shrimp	PABO	Decapod	120.6	5.9		26.6		71.19	-19.58	44.7	12.14	9.8	3.54
Pink glass shrimp	PMUL	Decapod	111.3	2.1		24.5		71.34	-20.40	47.1	12.43	9.3	3.63
Pink glass shrimp	PMUL	Decapod	115.2	2.7		34.5		73.70	-20.08	46.2	12.33	10.0	3.60
Striped shrimp	PMON	Decapod	108.0	4.1		41.4		72.79	-18.40	40.7	12.01	9.9	3.51
Striped shrimp	PMON	Decapod	124.9	6.0		50.9		71.94	-18.77	41.9	13.10	9.9	3.83
Snow crab	SNCR	Decapod	91.1	315.5	M	41.0	24.9	77.83	-20.01	45.4	13.03	8.2	3.81
Snow crab	SNCR	Decapod	84.5	222.5	M	26.0	21.0	87.65	-19.22	41.6	13.54	9.4	3.96
Snow crab	SNCR	Decapod	70.0	113.0	M	35.8	22.8	88.58	-19.07	40.5	13.22	9.0	3.86
Snow crab	SNCR	Decapod	55.2	51.1	F	46.5	2.0	81.82	-20.34	47.0	13.28	8.1	3.88
Snow crab	SNCR	Decapod	47.3	27.2	F	25.3	4.2	81.07	-19.86	43.4	12.22	7.6	3.57
Snow crab	SNCR	Decapod	41.2	16.7	F	49.7		77.09	-20.09	46.8	14.95	7.0	4.37
Snow crab	SNCR	Decapod	95.6	327.1	M	53.2		83.57	-19.81	46.8	14.03	8.5	4.10
Snow crab	SNCR	Decapod	97.9	361.8	M	27.1		84.79	-20.07	47.2	13.36	8.6	3.90
Snow crab	SNCR	Decapod	89.0	158.4	M	17.2		93.99	-20.19	40.0	12.47	7.1	3.64
Snow crab	SNCR	Decapod	107.3	344.5	M	24.2		82.58	-19.84	39.9	13.17	7.8	3.85
Snow crab	SNCR	Decapod	84.8	192.2	M	32.3		88.91	-20.42	42.7	12.50	7.2	3.65
Snow crab	SNCR	Decapod	60.9	78.5	M	29.9		87.14	-20.29	43.1	11.98	7.9	3.50
Green Sea Urchin	URCH	Echinoderm	45	34.4		6.24	0.79	90.55	-17.20	39.2	5.78	6.0	1.67
Green Sea Urchin	URCH	Echinoderm	50	43.2		5.03	0.92	86.70	-17.09	42.2	5.81	6.3	1.68
Green Sea Urchin	URCH	Echinoderm	55	59.0		4.73	0.53	88.57	-17.41	43.6	5.68	6.0	1.64
Green Sea Urchin	URCH	Echinoderm	53	49.7		3.53	0.51	87.93	-17.30	41.8	5.86	5.5	1.70
Green Sea Urchin	URCH	Echinoderm	53	55.5		5.30	0.32	86.60	-18.55	43.6	5.92	6.3	1.71

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Green Sea Urchin	URCH	Echinoderm	45.9	33.8		4.83		91.30	-16.62	35.9	5.74	5.4	1.66
Green Sea Urchin	URCH	Echinoderm	51	43.9		4.14		87.66	-18.96	42.7	5.99	6.3	1.74
Green Sea Urchin	URCH	Echinoderm	44.3	30.7		4.92		84.45	-17.46	41.4	5.93	8.0	1.72
Green Sea Urchin	URCH	Echinoderm	58	65.1		5.41		88.39	-18.43	40.4	6.16	6.7	1.79
Green Sea Urchin	URCH	Echinoderm	55.5	60.1		4.61		88.04	-17.94	41.2	5.61	6.1	1.62
Common tortoiseshell limpet	PATE	Mollusc	20.5	1.2		7.67	1.69	83.66	-17.73	42.5	6.79	9.5	1.97
Common tortoiseshell limpet	PATE	Mollusc	22	1.0		8.63	1.86	81.36	-18.22	44.2	6.68	9.1	1.94
Common tortoiseshell limpet	PATE	Mollusc	18.5	0.8		8.39	1.50	82.39	-18.16	43.1	6.75	9.7	1.96
Common tortoiseshell limpet	PATE	Mollusc	18	0.6		8.88	1.46	82.25	-18.50	43.4	6.86	9.7	1.99
Common tortoiseshell limpet	PATE	Mollusc	16	0.5		10.1	1.78	81.37	-18.59	43.8	7.13	9.2	2.07
Common tortoiseshell limpet	PATE	Mollusc	16	0.5		9.60		82.54	-18.80	42.5	6.87	9.5	1.99
Common tortoiseshell limpet	PATE	Mollusc	18.5	0.8		8.10		82.27	-18.55	42.9	6.80	9.3	1.97
Common tortoiseshell limpet	PATE	Mollusc	18.5	0.8		8.95		83.13	-18.83	43.6	7.04	9.4	2.04
Common tortoiseshell limpet	PATE	Mollusc	15.5	0.5		11.1		80.88	-19.54	43.7	6.97	8.9	2.02
Common tortoiseshell limpet	PATE	Mollusc	15	0.3		9.13		82.16	-19.04	43.2	6.98	9.7	2.03
Blue mussel	BLMU	Mollusc	77.0	60.8		10.8	5.7	86.81	-19.90	38.7	7.71	8.3	2.24
Blue mussel	BLMU	Mollusc	61.0	41.9		8.94	3.04	90.03	-18.60	30.3	7.10	6.0	2.06
Blue mussel	BLMU	Mollusc	60.5	45.4		12.6	2.4	89.79	-18.75	33.3	7.82	7.3	2.27
Blue mussel	BLMU	Mollusc	59.0	38.8		8.51	1.61	89.45	-17.05	26.8	8.27	6.1	2.41
Blue mussel	BLMU	Mollusc	62.5	29.6		8.54	2.88	89.01	-19.64	35.0	7.11	9.0	2.06
Blue mussel	BLMU	Mollusc	51.0	28.9		14.4		90.06	-19.37	35.5	7.83	9.4	2.28
Blue mussel	BLMU	Mollusc	51.5	25.4		9.49		91.84	-19.79	33.3	7.03	7.8	2.04
Blue mussel	BLMU	Mollusc	53.5	23.5		9.57		90.11	-18.74	33.6	7.36	8.5	2.14
Blue mussel	BLMU	Mollusc	50.0	21.6		10.1		91.63	-19.00	34.0	8.32	8.0	2.42
Blue mussel	BLMU	Mollusc	47.5	14.4		8.62		91.52	-20.00	30.9	7.23	7.2	2.10
Blue mussel	BLMU	Mollusc	82.2	73.7		10.0	2.2	88.97	-20.79	37.7	7.30	9.8	2.12
Blue mussel	BLMU	Mollusc	36.0	4.9		11.2	3.0	88.14	-20.11	40.4	6.86	10.5	1.99

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Blue mussel	BLMU	Mollusc	51.0	4.6		27.3	5.8	84.95	-19.45	41.6	6.94	10.4	2.01
Blue mussel	BLMU	Mollusc	37.0	4.6		6.06	1.52	91.39	-20.06	33.8	6.66	9.1	1.93
Blue mussel	BLMU	Mollusc	35.0	5.1		13.9	2.5	89.72	-19.82	38.3	6.84	9.5	1.99
Blue mussel	BLMU	Mollusc	31.0	4.3		9.85		86.36	-19.94	43.7	7.18	11.0	2.09
Blue mussel	BLMU	Mollusc	31.0	3.2		11.8	2.2	87.69	-19.75	40.0	6.97	10.4	2.02
Blue mussel	BLMU	Mollusc	29.0	2.6		9.89	1.16	87.54	-19.98	42.2	6.66	9.0	1.93
Blue mussel	BLMU	Mollusc	29.0	2.6		15.4	3.4	86.69	-19.86	40.1	6.35	9.7	1.84
Blue mussel	BLMU	Mollusc	27.0	2.2		17.3		85.58	-19.69	40.4	6.60	10.2	1.91
Blue mussel	BLMU	Mollusc	26.5	2.0		14.1		85.51	-19.90	39.6	6.69	9.9	1.94
Waved whelk	BUCC	Mollusc	44.0	10.1		24.1	19.0	82.46	-17.29	42.2	9.65	10.5	2.81
Waved whelk	BUCC	Mollusc	40.0	9.2		32.7	25.6	80.21	-17.53	46.3	9.89	10.7	2.88
Waved whelk	BUCC	Mollusc	37.5	7.9		31.1	16.5	79.54	-17.82	44.6	10.03	10.7	2.92
Waved whelk	BUCC	Mollusc	37.5	6.7		25.3	3.9	76.24	-17.23	44.8	10.07	11.1	2.94
Waved whelk	BUCC	Mollusc	34.5	5.5		28.1	16.5	81.36	-17.82	43.1	10.18	11.1	2.97
Waved whelk	BUCC	Mollusc	32.5	4.5		19.8		80.69	-18.26	42.8	9.55	10.3	2.78
Waved whelk	BUCC	Mollusc	31.0	3.9		29.9		79.60	-18.00	43.0	9.73	10.4	2.84
Waved whelk	BUCC	Mollusc	26.0	2.6		19.6		79.96	-18.43	42.9	9.12	10.5	2.66
Waved whelk	BUCC	Mollusc	25.0	2.1		19.8		80.61	-18.01	43.1	9.44	10.7	2.75
Waved whelk	BUCC	Mollusc	23.5	1.7		23.6		78.94	-17.92	41.7	9.34	10.5	2.72
Common periwinkle	BIGO	Mollusc	27.0	6.6		9.16	2.69	79.64	-17.27	41.7	8.41	11.3	2.45
Common periwinkle	BIGO	Mollusc	25.5	5.9		10.5	2.5	77.63	-17.32	41.1	7.96	11.3	2.31
Common periwinkle	BIGO	Mollusc	24.0	5.2		10.5	3.2	81.79	-17.23	40.9	7.80	11.2	2.27
Common periwinkle	BIGO	Mollusc	24.5	4.6		9.39	2.15	80.73	-17.02	40.7	7.66	10.9	2.23
Common periwinkle	BIGO	Mollusc	23.0	4.2		9.51	1.86	82.28	-17.01	41.2	8.33	11.4	2.42
Common periwinkle	BIGO	Mollusc	23.0	3.9		10.2		82.27	-17.62	41.2	7.59	10.8	2.20
Common periwinkle	BIGO	Mollusc	22.5	3.9		8.67		81.24	-17.54	41.1	7.72	11.3	2.24
Common periwinkle	BIGO	Mollusc	22.5	3.9		9.58		83.42	-16.51	41.0	7.83	11.1	2.28

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Common periwinkle	BIGO	Mollusc	21.0	3.3		8.39		81.96	-17.61	41.2	7.74	10.9	2.25
Common periwinkle	BIGO	Mollusc	18.5	2.3		8.33		81.55	-17.52	41.6	7.73	10.5	2.25
Gammarid	GAMA	Mollusc	35	0.3		9.86	6.36	77.36	-19.54	41.7	8.41	7.4	2.45
Gammarid	GAMA	Mollusc	31	0.3		9.38	6.09	77.79	-19.00	38.5	8.06	7.4	2.34
Gammarid	GAMA	Mollusc	28.6	0.3		8.21		74.57	-19.36	40.2	8.26	7.5	2.40
Gammarid	GAMA	Mollusc	23	0.2		10.5	5.3	74.49	-20.12	42.7	8.39	7.1	2.44
Gammarid	GAMA	Mollusc	23	0.2		8.10	0.99	75.79	-18.62	38.5	8.07	7.8	2.35
Gammarid	GAMA	Mollusc	20.7	0.2		9.73	0.09	73.95	-20.24	41.5	7.98	7.1	2.32
Gammarid	GAMA	Mollusc	20.7	0.2		7.85	0.61	76.67	-19.84	38.9	8.28	6.8	2.41
Gammarid	GAMA	Mollusc	20.7	0.2		16.9	5.2	73.42	-19.85	39.2	8.23	6.5	2.39
Gammarid	GAMA	Mollusc	19	0.2		8.00		73.96	-20.05	40.7	8.00	6.9	2.33
Gammarid	GAMA	Mollusc	15	0.1		6.48		81.18	-19.06	36.7	7.67	7.0	2.23
Northern krill	MENO	Zooplankton	27.5	0.22		16.1	3.3	78.60	-20.90	43.9	9.44	9.3	2.75
Northern krill	MENO	Zooplankton	30.5	0.47		9.34	0.62	86.05	-21.22	47.2	9.40	9.1	2.74
Northern krill	MENO	Zooplankton	30	0.34		8.37		81.45	-20.98	46.2	8.96	10.0	2.61
Northern krill	MENO	Zooplankton	27	0.22		13.2	0.1	79.89	-21.10	49.7	8.67	9.0	2.52
Northern krill	MENO	Zooplankton	29	0.27		11.3	0.7	79.00	-21.17	49.9	9.84	9.2	2.87
Northern krill	MENO	Zooplankton	26	0.19		11.5	1.8	78.73	-21.20	46.6	8.85	9.6	2.58
Calanus hyperboreus	CHYP	Zooplankton	6	0.01		16.3	1.0	82.00	-22.38	54.8	8.37	6.3	2.44
Calanus hyperboreus	CHYP	Zooplankton	6	0.01		8.78	0.92	84.82	-22.50	57.9	8.04	6.4	2.34
Calanus hyperboreus	CHYP	Zooplankton	6	0.01		7.67	0.89	83.18	-22.27	56.2	7.73	6.8	2.25
Calanus hyperboreus	CHYP	Zooplankton	6	0.00		14.4	0.2	79.22	-22.52	60.5	7.79	6.4	2.26
Calanus hyperboreus	CHYP	Zooplankton	6	0.01		10.3	0.2	83.03	-22.52	56.2	8.19	6.3	2.38
Calanus finmarchicus	CFIN	Zooplankton	4.5	0.01		2.29	0.79	79.91	-22.44	58.2	8.43	7.0	2.45
Calanus finmarchicus	CFIN	Zooplankton	4	0.00		2.68	0.79	75.40	-22.85	60.2	7.19	6.3	2.09
Calanus finmarchicus	CFIN	Zooplankton	4	0.01		3.68	0.28	78.69	-22.68	60.2	8.49	6.4	2.47
Calanus finmarchicus	CFIN	Zooplankton	4	0.00		2.70	0.72	76.80	-22.62	59.4	8.37	6.7	2.44

Table A. continued

Common name	Code	Taxonomical group	Length (mm)	Weight (g)	Sex	THg (ng/g)	MeHg (ng/g)	Moisture content (%)	$\delta^{13}C$ (‰)	C (%)	$\delta^{15}N$ (‰)	N (%)	TL
Calanus finmarchicus	CFIN	Zooplankton	4	0.01		2.78	0.25	79.49	-22.52	57.4	8.57	6.9	2.49
Calanus finmarchicus	CFIN	Zooplankton	4	0.00		2.77		74.78	-22.69	60.2	8.40	6.5	2.44
Calanus finmarchicus	CFIN	Zooplankton	4	0.01		2.42		82.05	-22.70	57.9	8.37	6.6	2.44
Calanus finmarchicus	CFIN	Zooplankton	4	0.00		2.70		75.39	-22.90	59.4	8.32	6.2	2.42
Calanus finmarchicus	CFIN	Zooplankton	4	0.01		2.60		77.93	-22.74	58.5	8.75	6.4	2.55
Calanus finmarchicus	CFIN	Zooplankton	4	0.01		2.48		81.97	-22.93	60.2	8.44	6.2	2.46